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Evaluation of a marine mammal status and trends contaminants indicator for European waters

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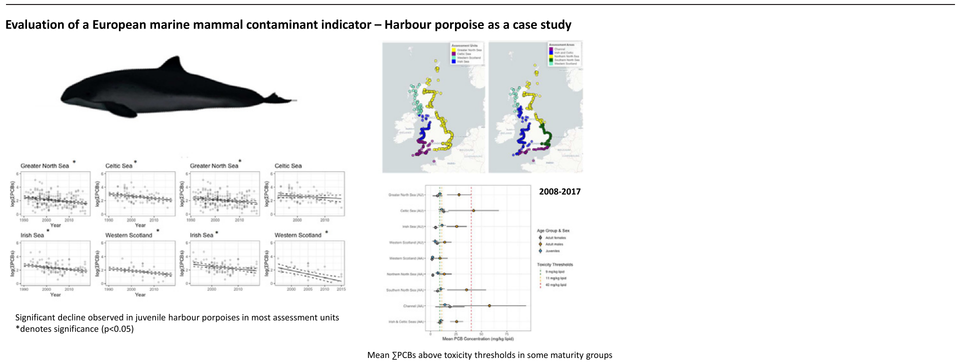
HIGHLIGHTS

- Trends and status of blubber ΣPCBs in European harbour porpoises assessed (1990–2017).
- Year, nutritional condition & latitude significant predictors of ΣPCBs in juveniles
- ΣPCBs declined significantly in juveniles, overtime, in most assessment units/areas.
- Animals exposed to levels of PCBs deemed a toxicology threat, among all maturity classes.
- Relative proportion above PCB thresholds declined during last 10 years of the study.

GRAPHICAL ABSTRACT



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ABSTRACT

Marine mammals are vulnerable to the bioaccumulation, biomagnification and lactational transfer of specific types of pollutants, such as industrial polychlorinated biphenyls (PCBs), due to their long-life spans, feeding at a high trophic level and unique fat stores that can serve as depots for these lipophilic contaminants. Currently, European countries are developing indicators for monitoring pollutants in the marine environment and assessing the state of biodiversity, requirements under both Regional Seas Conventions and European legislation. As sentinel species for marine ecosystem and human health, marine mammals can be employed to assess bioaccumulated contaminants otherwise below current analytical detection limits in water and lower trophic level marine biota. To aid the development of Regional Seas marine mammal contaminants indicators, as well as Member States obligations under descriptor 8 of the EU Marine Strategy Framework Directive, the current study aims to further develop appropriate methodological standards using data collected by the established UK marine mammal pollutant monitoring programme (1990 to 2017) to assess the trends and status of PCBs in harbour porpoises. Within this case study, temporal trends of PCB blubber concentration in juvenile harbour porpoises were analysed using multiple linear regression models and toxicity thresholds for the onset of physiological (reproductive and immunological) endpoints were applied to all sex-maturity groups. Mean PCB blubber concentrations were observed to decline in all harbour porpoise Assessment Units and OSPAR

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Assessment Areas in UK waters. However, a high proportion of animals were exposed to concentrations deemed to be a toxicological threat, though the relative proportion declined in most Assessment Units/Areas over the last 10 years of the assessment. Recommendations were made for improving the quality of the assessment going forward, including detailing monitoring requirements for the successful implementation of such an indicator.

1. Introduction

The EU Marine Strategy Framework Directive (2008/56/EC) establishes a 'framework within which Member States shall take the necessary measures to achieve or maintain good environmental status (GES) in the marine environment'. Marine strategies developed by Member States under the Directive should take an ecosystem-based approach to the management of human activities, and GES is assessed through defining a series of conservation targets/objectives under eleven descriptors, for which indicators should be developed to evaluate against (EU Commission Decision, 2010). While Member States have, for the most part, made use of existing monitoring for indicator development, the Commission outlined in 2017 that ecosystem components should be linked appropriately to anthropogenic pressures and impacts on the marine environment (EU Commission Decision, 2017), and thus current indicator development requires reconsideration. A key aspect of the Marine Strategy Framework Directive (MSFD) is that Member States are required to coordinate work on their marine strategies at the regional seas level rather than solely focusing on national waters, which is appropriate for transboundary marine species and where contaminants, for example, are transported, dispersed and deposited via the atmosphere and surface waters in the region. Within the North-east Atlantic, regional work is being coordinated by the Convention for the Protection of the Marine Environment of the North-East Atlantic (the OSPAR Convention). Member States can however decide on whether to report under the MSFD either at the scale of their regional seas or national waters (Murphy et al., 2021).

Contaminants are addressed under two descriptors of the MSFD, descriptor 8 'concentrations of contaminants give no effects' and descriptor 9 'contaminants in seafood are below safe levels'. OSPAR, in its 2017 intermediate assessment, evaluated eleven different pressure and impact contaminant related indicators, including status and trends in the concentrations of Polychlorinated Biphenyls (PCB), Polybrominated Diphenyl Ethers (PBDEs), Polycyclic Aromatic Hydrocarbons (PAHs), organotin and heavy metals (mercury, cadmium and lead) in various matrices such as fish, shellfish and sediment, depending on the indicator in question. Other indicators included, evaluating the status and trends in the levels of imposex in marine gastropods (i.e. assessing impacts of exposure to tributyltin compounds in shellfish), and inputs of mercury, cadmium and lead via water and air to the Greater North Sea.[1] Notably, higher-level marine organisms are not currently assessed through any indicator; these are species that due to elevated position in marine food webs can be used to assess bioaccumulated contaminants otherwise below current analytical detection limits in water and lower trophic level marine biota. Additionally, higher-level organism contaminant pressure and impact related indicators could be linked to state indicators, developed under descriptors 1 (biodiversity is maintained) and 4 (elements of food webs ensure long-term abundance and reproduction) of the MSFD, or OSPAR's common biodiversity state indicators for said species. Thus, facilitating future application of the DPSIR (driver, pressure, state, impact and response model of intervention) analytical framework approach (Patrício et al., 2016) for higher-level organisms as pressure related indicators are required to evaluate changes in population status, per se, and also to successfully implement a Programme of Measures to achieve GES (Murphy et al., 2021).

Marine mammals are useful sentinels for marine ecosystem and human health, as many species have long life spans (hence permit the expression of chronic diseases including neoplasia, abnormalities in growth and development, and reproductive failure), feed at a high trophic level (top/apex predators) and have unique fat stores that can serve as depots for lipophilic contaminants (Bossart, 2011; Reif, 2011; Ross, 2000). Additionally, several species are long-term coastal residents, and hence are representative of a reasonably defined local area. Marine mammals are vulnerable to the bioaccumulation, biomagnification and lactational transfer of specific types of lipophilic (and hydrophobic) persistent organic pollutants (POPs) such as organochlorine pesticides (e.g. DDTs and dieldrin) and industrial polychlorinated biphenyls (PCBs) (Loganathan and Kannan, 1994; Murphy et al., 2018; Ross, 2000). The Commission Decision (EU) 2017/848, laying down criteria and methodological standards for GES of marine waters and specifications and standardized methods for monitoring and assessment, and repealing Decision 2010/477/EU, was adopted in May 2017. Within the revised Commission Decision, it was outlined that additional scientific and technical progress was required to achieve the stated objectives of the Commission, including the evaluation of pressure indicators related to contaminants (as well as bycatch and marine litter) for relevant ecosystem elements. While the Decision notes that the species composition under descriptor 8 should be understood to refer to the lowest taxonomic level appropriate for the assessment, it further outlines in criteria 2 that Member States are to establish a list of species that are at risk from contaminants, and that the health of those species, as well as their relative abundance, are not adversely affected due to cumulative and synergetic effects from exposure to contaminants (EC Decision, 2017). The establishment of pollutant concentration thresholds that may have deleterious effects was discussed within the Commission Decision and it was noted that biomarkers or population demographic characteristics, such as fecundity rates, survival rates, mortality rates and reproductive capacity, are also relevant to the assessment of overall health effects.

In 2014, the International Council for the Exploration of the Sea (ICES) Working Group on Marine Mammal Ecology (WGMME) proposed the development of a marine mammal PCB toxicity threshold indicator under the MSFD (ICES WGMME, 2014). This was further developed by OSPAR's Marine Mammal Expert Group (OMMEG) in 2019, with the intent of developing an indicator on trends and status of PCBs in marine mammals that included a contaminant-effects based component. The proposed indicator was reviewed by both OSPAR's Biodiversity Committee (BDC) and Hazardous Substances & Eutrophication Committee (HASEC), and Contracting Parties expressed their support for the continued evaluation of said indicator, with the potential to broadening the indicator to 'persistent chemicals' such as mercury (ICES WGMME, 2020). It is anticipated that work on such an indicator will be included as a case study within OSPAR's 2023 Quality Status Report.

Within the UK, the harbour porpoise (*Phocoena phocoena*) is used as a sentinel top predator species for monitoring long-term trends in chemical contaminant exposure in the marine environment, namely organochlorine pesticides, brominated flame retardants, and hexabromocyclododecane (HBCD). Accumulating levels of brominated flame retardants observed in UK-stranded porpoise blubber in the 1990s was partially responsible for the EU-wide ban of the commercial penta- and octa-mix polybrominated diphenyl ether (PBDE) products in 2004 (Law et al., 2012a). This was followed by a significant and consistent decline in observed concentrations of brominated diphenyl ethers (BDEs) in the marine sentinel species during the period 2008 to 2012 (Law et al., 2012a). Similar declines were also observed in HBCD, as well as organochlorine pesticides such as dichlorodiphenyltrichloroethane (DDT) and dieldrin, as well as tributyltin compounds in UK-stranded porpoise blubber for the same period (Law et al., 2012a; Law et al., 2012b). Despite these declines however, the toxic effects

[1] https://oap.ospar.org/en/ospar-assessments/intermediate-assessment-2017/pressures-human-activities/contaminants/.

of long-term exposure to multiple pollutants at low dose levels during sensitive development stages of the harbour porpoise is unknown (Murphy et al., 2015; Murphy et al., 2018). Declines in the measured concentration of several of the environmentally persistent and toxic PCBs was more muted and plateaued in UK harbour porpoises around 1998 (Jepson et al., 2016; Law et al., 2012a). Since 2007, levels of PCBs appear to have begun to decline again, though the overall rate of decline remains slow in comparison to other pollutants (Williams et al., 2020a). Given these are substances with environmental half-lives of years to decades (Hickie et al., 2007; Jonsson et al., 2003; Sinkkonen, 2000), it is not surprising there is persistence within UK harbour porpoises (Jepson et al., 2016; Law et al., 2012a; Stuart-Smith and Jepson, 2017). These results however also suggest continued environmental input of PCBs into the marine environment (Jepson et al., 2016; Stuart-Smith and Jepson, 2017).

Exposure to PCBs, has been suggested in harbour porpoises to induce immune-suppression (Hall et al., 2006; Yap et al., 2012), as well as impact thyroid function (Schnitzler et al., 2008) and foetal and newborn survival (Murphy et al., 2015). Further, during the period 1990 and 2008, harbour porpoise calves (i.e. animals less than one year old) in the southern North Sea showed an increase in some contaminant types, including PCBs, with this age class reported as the most vulnerable due to gestational and lactational transfer and/or a lower ability to eliminate these compounds (Weijs et al., 2010). Harbour porpoise calves have also been shown to be exposed to a more neurotoxic mixture of PCBs than adult females and males due to the selective maternal offloading of certain PCB congeners (Williams et al., 2020a). Further, Weijs et al. (2010) suggested that calves may be feeding at a higher trophic position than their mothers, essentially 'consuming the tissues of their mothers'. While more recent work on porpoises sampled in the southern North Sea suggested that calves of nutritionally stressed females had a greater potential for toxicity, as their mothers offloaded a higher pollutant load in their milk (van den Heuvel-Greve et al., 2021).

Decades of rigorous experimental and epidemiological studies have shown that PCBs have a range of dose-dependent toxic effects such as immunosuppression, endocrine disruption and reproductive impairment in mammalian species tested, including humans (Diamanti-Kandarakis et al., 2009; EEA Technical report, 2012; Kannan et al., 2000). In UK waters, mean ΣPCBs concentrations in adult male and female porpoises (sampled between 1990 and 2012) exceeded an established mammalian toxicity threshold of 9 mg/kg ΣPCBs for onset of physiological (immunological and reproductive) endpoints in marine mammals (Jepson et al., 2016; Kannan et al., 2000; Law et al., 2012a), and some individual porpoises exceeded the 41 mg/kg ΣPCBs threshold associated with profound reproductive impairment in Baltic ringed seals (*Pusa hispida*) (Helle et al., 1976; Jepson et al., 2016). More recent analysis modelling temporal trends has shown that the overall mean ΣPCBs concentrations fell below an established mammalian toxicity threshold for physiological endpoints (9 mg/kg lipid) around 2007, however they are still associated with increased rates of infectious disease mortality (Williams et al., 2020a).

Despite production of PCBs being stopped in Europe in 1980s and the banning of the main uses of PCBs in products in 1986, with disposal being targeted (OSPAR, 2010), there is a need for renewed steps to reduce PCB inputs into the marine environments in Europe (as addressed within the revised EU Regulation 2019/1021 on persistent organic pollutants), and the continued monitoring of their toxic effects on top predators (Jepson et al., 2016; Stuart-Smith and Jepson, 2017). To aid the development of OSPAR's marine mammal contaminants indicator and Member States obligations under the MSFD, the current study aims to further develop appropriate methodological standards using data collected by the established UK marine mammal pollutant monitoring programme to assess the trends and status of PCBs in harbour porpoises sampled in UK waters, as a case study. This paper will further delineate recommendations for improving the quality of the assessment going forward, both for the harbour porpoise and other potential marine mammal species and persistent chemicals, including detailing monitoring requirements for the successful implementation of such an indicator.

2. Methods

2.1. Sampling

This analysis was carried out using data provided by the UK Cetacean Strandings Investigation Programme (CSIP). The PCB blubber concentrations were determined in 662 harbour porpoises that stranded on the coast of the UK between 1990 and 2017, and necropsied according to standard post-mortem procedures for cetaceans (IJsseldijk et al., 2019). A standardized methodology was used to extract and preserve the blubber samples for PCB analysis (Law, 1994). In brief, full thickness blubber samples were taken from the left side of the body, proximal to either the cranial or caudal insertion of the dorsal fin, wrapped in food-grade foil and preserved at −20 °C (Law, 1994). The individuals selected for PCB analysis were prioritized by their state of decomposition using the scoring system set out by Law et al. (2006). To minimize the impact of changes in PCB tissue concentrations and distribution that are associated with decomposition, fresher carcasses were prioritized (Law et al., 2006). The individuals analysed were otherwise considered a representative sample of the strandings that occurred over the period.

2.2. PCB analysis

The concentrations of the sum of 25 individual chlorobiphenyl (CB) congeners (Σ25 CBs) (on a mg/kg wet weight basis) were determined at the Cefas laboratory (Lowestoft) using a method that was validated following participation in the QUASIMEME (Quality Assurance of Information for Marine Environmental Monitoring in Europe) laboratory proficiency scheme and followed the recommendations of the International Council for the Exploration of the Sea (ICES) (de Boer and Law, 2003; de Boer and Wells, 1997; ICES, 1998; Webster et al., 2013). In cases where the congener concentrations were below the limit of quantification (<0.0003 or <0.0004 mg/kg wet weight), the concentration was set at half the limit (Law et al., 2012a). The numbers of the International Union of Pure and Applied Chemistry CBs congeners analysed were: 18, 28, 31, 44, 47, 49, 52, 66, 101, 105, 110, 118, 128, 138, 141, 149, 151, 153, 156, 158, 170, 180, 183, 187, and 194. This selection was chosen to ensure incorporation of the seven PCBs prioritized for international monitoring by ICES (ΣICES7) and included those that are relatively abundant in commercial PCB mixtures with a broad range of chlorination. The sum of the 25 individual CB congener concentrations was calculated and normalized to a lipid basis (mg/kg lipid) by solvent extracting lipids from the blubber and calculating the hexane extractable lipid content (Webster et al., 2013).

2.3. Pathological analyses

As part of the pathological investigations, several biological and life-history attributes were determined. Body length and sexual maturity status were used to categorize individuals into age-maturity groups. Sexual maturity was assessed using gross gonadal size and appearance and in a representative subset, histological evidence of spermiogenesis in male testes (Murphy, 2008; Murphy et al., 2005). Female reproductive maturity was determined by identification of one or more ovarian corpora (lutea or albicantia) (Murphy et al., 2015). Neonates were defined as individuals with a body length <90 cm, juveniles as individuals with a body length >90 cm that were sexually immature, and adults as sexually mature individuals with a body length >90 cm (after Jepson, 2003). For the purposes of this study, the neonates and juveniles were grouped together and classed as juveniles. For smaller cetaceans like the harbour porpoise, a basic index of weight to length ratio is thought to be the most appropriate metric of body condition and is widely acknowledged as a good predictor of fitness in marine mammals (Beauplet and Guinet, 2007; Kershaw et al., 2017). The weight and length data variable for the individuals in this study followed a power relationship and so a power regression model was fitted to obtain a metric that could be used as a proxy for body condition. The residuals from the best-fit regression line were extracted and used for further modelling whereby, values above the model fit represented cases in good nutrition and individuals below the line represented cases in poor nutritional condition.

The latitudes and longitudes of the stranding locations of each animal were collected and used to investigate spatial variation. Trends were assessed according to two different schemes for division of areas: (1) the harbour porpoise Assessment Units (AUs) within English, Welsh and Scottish waters agreed by the Joint IMR/NAMCCO international workshop on the status of harbour porpoises in the North Atlantic (NAMMCO-IMR, 2019); and (2) the OSPAR Contaminants Assessment Areas (AAs) (or OSPAR sub-regions) that are used to assess contaminants in fish and shellfish (OSPAR IA, 2017). The harbour porpoise-AUs in UK waters were: Greater North Sea, Celtic Sea, Irish Sea, and Western Scotland; and the OSPAR-AAs in UK waters were: Northern North Sea, Southern North Sea, the Channel, Irish Sea, Celtic Seas, and Western Scotland. For the analysis of the OSPAR Assessment Areas we combined the Irish and Celtic Seas to increase the statistical power to detect trends and to allow for further comparison, an approach also taken by the IMR/NAMMCO workshop when modelling population dynamics of harbour porpoises in those regions (Fig. 1).

2.4. Trends assessment

All analyses were carried out using the statistical software R (version 3.6.3) (R Core Team, 2016). To determine appropriate methodological standards for the trends assessment we evaluated two separate modelling approaches. First, we modelled temporal trends of PCB concentrations for the UK as a whole and used the spatial model coefficients (latitude and longitude) to estimate trends in each Assessment Unit/Area using the mean latitudes and longitude values for each unit/area (i.e. the central point within each pre-defined unit/area). As an alternative approach to estimate trends, we built separate models for each of the Assessment Units/Areas and used the spatial model coefficients (latitude and longitude) to estimate PCB concentrations in each year at the mean latitude and longitude for each unit/area. We also used the model results to estimate the yearly percentages change in PCB concentrations for each unit/area. We chose to derive yearly percentage changes as these are presented as part of the OSPAR trends assessment of PCBs in biota and sediment and so would allow for comparisons to be made between environmental compartments assessed by OSPAR (2021).

For both of the approaches, we modelled Σ25 CBs with covariates that have been shown to influence concentrations (e.g., nutritional condition) and may therefore confound temporal trends (Aguilar et al., 1999; Tanabe et al., 1981). Adult males and females were excluded from the analysis to reduce the impact of age-related heterogeneity caused by the capacity for females to offload contaminants through transplacental and lactational transfer (Aguilar et al., 1999; Tanabe et al., 1981), and bioaccumulation in mature males (Murphy et al., 2015). The sample sizes and standard errors in mean Σ25 CBs concentrations for each of the Assessment Units/Areas for juvenile porpoises are shown in Table 1.

Following data exploration, the relationship between Σ25 PCBs and other covariates was established to be linear. Using Σ25 CBs as the response variable, several multiple linear regression models were fitted. Variables included in the full model were year, nutritional condition (derived from body weight to length ratio), sex, latitude, and longitude. All possible variable combinations were tested to obtain several candidate models, which were ranked according to their AIC (Akaike's Information Criterion) values. Our final prediction was obtained by averaging the set of plausible models (ΔAIC < 2) from the candidate models (Burnham and Anderson, 2004). Model validation was performed by assessing the diagnostic plots and plotting the model residuals against selected variables to assess the variance.

2.5. Status assessment

To carry out the contaminant status assessment we determined mean summed PCB blubber concentrations, mean concentrations for the ICES 7 suite of congeners (CB congeners 28, 52, 101, 118, 138, 153, 180) and mean concentrations for the five congeners present in the highest concentrations for each of the Assessment Units/Areas. Separate values were determined for adult females, adult males and juveniles. To carry out status assessments in fish and shellfish, OSPAR compare concentrations against two sets of assessment values: Background Assessment Concentrations (BACs), which are assumed to represent concentrations at or close to background levels (i.e., zero for man-made substances) and Environmental Assessment Criteria (EACs), which are defined as concentrations below which no chronic effects are expected to occur (OSPAR, 2009). There are

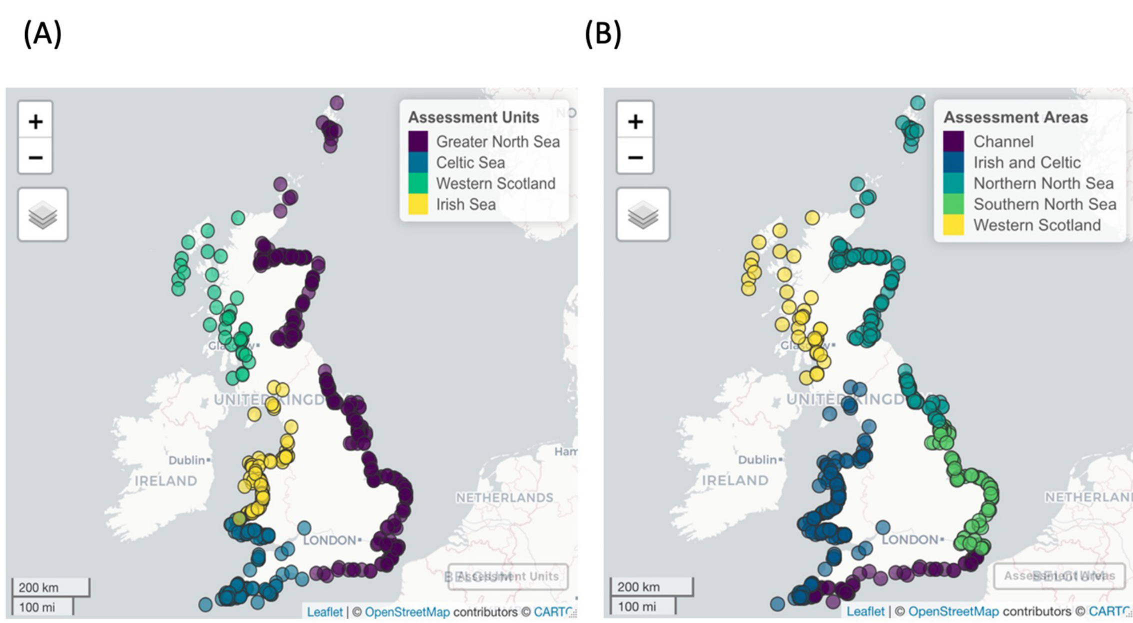

Fig. 1. Geographic locations and Assessment Unit/Area classifications of the individuals that stranded and were analysed to obtain blubber concentrations for the sum of 25 selected congeners of polychlorinated biphenyls (Σ25CBs). The colours of the dots represent the Assessment Unit/Area classification. (A) Harbour porpoise Assessment Units defined by the joint IMR/NAMMCO international workshop, (B) OSPAR contaminants Assessment Areas for fish and shellfish. The Irish & Celtic Seas Assessment Areas have been combined. PCB congeners analysed were: 18, 28, 31, 44, 47, 49, 52, 66, 101, 105, 110, 118, 128, 138, 141, 149, 151, 153, 156, 158, 170, 180, 183, 187, and 194.

Table 1
Sample sizes and standard deviation for each assessment area for the trend assessment of mean summed PCB blubber concentrations in juvenile harbour porpoises (n = 387). *PCB congeners analysed were: 18, 28, 31, 44, 47, 49, 52, 66, 101, 105, 110, 118, 128, 138, 141, 149, 151, 153, 156, 158, 170, 180, 183, 187, and 194.

	N	Mean Σ25CBs (mg/kg lipid)	Standard error
Harbour porpoise Assessment Unit			
Greater North Sea	205	12.7	17.3
Celtic Sea	73	19.5	19.5
Irish Sea	84	14.86	16.8
Western Scotland	25	9.23	8.88
OSPAR Assessment Area			
Northern North Sea	113	8.5	8.47
Southern North Sea	75	16.4	22.9
Channel	32	24.6	23.3
Irish & Celtic Seas	155	16.0	16.9
Western Scotland	12	4.6	3.1

no recommended EACs for PCBs in biota therefore EACs in fish and shellfish tissue are derived from EACs that exist for sediment using biota sediment accumulation factors (OSPAR, 2020). In the absence of EACs for marine mammals, we compared the mean summed PCB blubber concentrations against two widely used published toxicity thresholds in marine mammals, 9 mg/kg lipid weight (lw) ΣPCBs for onset of physiological (immunological and reproductive) endpoints (Jepson et al., 2016; Kannan et al., 2000; Law et al., 2012a), and 41 mg/kg lw ΣPCBs, one of the highest PCB thresholds associated with profound reproductive impairment in Baltic ringed seals (*Pusa hispida*) (Helle et al., 1976; Jepson et al., 2016). An additional threshold of 11 mg/kg lw ΣPCBs was applied to mature females to ascertain if they had successfully reproduced and offloaded some of their PCB burden (Murphy et al., 2015). We also calculated the proportion of animals above each of the thresholds.

3. Results

3.1. Trends assessment

Mean concentrations were highest in the Celtic Sea harbour porpoise-AU and the Channel OSPAR-AA and lowest in the western Scotland harbour porpoise-AU and OSPAR-AA and the Northern North Sea OSPAR-AA (see Table 1). When the UK dataset was modelled as a single geographic unit, year was a significant predictor of PCB concentrations. This modelling approach detected statistically significant temporal trends in all of the Assessment Units/Areas (Table 2, Figs. 2A and 3A). The model for the whole of the UK dataset included nutritional condition and latitude as significant predictors (Table 2). Longitude and sex were also included as nonsignificant predictors. When the Assessment Units and Areas were modelled separately, we detected significant negative temporal trends in the Greater North Sea, Western Scotland and Irish Sea harbour porpoise-AUs, and Southern North Sea, Irish & Celtic Seas and western Scotland OSPAR-AAs. However, trends were not statistically significant in the Celtic Sea harbour porpoise-AU and the Channel and Northern North Sea OSPAR-AAs (Figs. 2B and 3B). This may be due to the limited power of the models to detect statistically significant trends because of inadequate sample sizes or because there was no significant change in PCB concentrations over the period. The full model results are shown in the Supporting Information Tables 1–9.

Table 2
Summary statistics of the linear regression model fitted to the data for the whole of the UK where Σ25CBs (mg/kg lipid) was the dependent variable. All continuous variables were centred and scaled. (The coefficient estimates were calculated in relation to a female). *PCB congeners analysed were: 18, 28, 31, 44, 47, 49, 52, 66, 101, 105, 110, 118, 128, 138, 141, 149, 151, 153, 156, 158, 170, 180, 183, 187, and 194.

log(Σ25CBs)				
Predictors	Estimates	CI		p
(Intercept)	76.21	52.12	−100.29	**<0.001**
Latitude	−0.15	−0.18	−0.11	**<0.001**
Rel body wt	−1.07	−1.53	−0.61	**<0.001**
Year	−0.03	−0.05	−0.02	**<0.001**
Sex [M]	−0.1	−0.26	−0.06	0.214
Longitude	0.02	−0.02	−0.06	0.299
Observations	387			
R^2	−0.456			

Both modelling approaches show that mean PCB blubber concentrations are declining in all of the Assessment Units/Areas. The separate modelling approaches produced similar trend estimates in terms of the estimated rates of decline and associated levels of uncertainty across all the Assessment Units/Areas, except for Western Scotland and the Channel (Figs. 2 and 3). When Western Scotland was modelled as a separate harbour porpoise-AU and OSPAR-AA, the rates of decline were steeper than when the trends were estimated using model coefficients derived for the whole of the UK, though confidence intervals were wider. When the Channel OSPAR-AA was modelled separately, PCB concentrations decreased at a slower rate than when the trend was estimated using model coefficients for the whole of the UK however, year was not statistically significant in the model (see Supplementary Information Table 7). The similarity in trend estimates produced from the two modelling approaches, for most of the Assessment Units/Areas, is also reflected in the estimates for yearly mean concentration change (see Fig. 4).

If we assume that modelling each Assessment Unit/Area separately is a more robust statistical approach then it appears that concentrations are declining at the slowest rate in the Northern North Sea followed by the Celtic Sea, the Channel, the Greater North Sea, the Southern North Sea, and Irish Sea and the Celtic Sea regions, with concentrations decreasing at the fastest rate off the coast of Western Scotland. Excluding regions where trends were not statistically significant (e.g. the Channel, Northern North Sea, and Celtic Sea), these data show that concentrations were declining most slowly in regions at lower latitudes (see Fig. 4).

3.2. Status assessment

3.2.1. Summed PCB concentrations

Comparison of the mean summed PCB blubber concentrations between Assessment Units/Areas shows an increasing gradient of contamination from north to south and from west to east, for the period 1990–2017 (Fig. 5). In all of the Assessment Units/Areas, adult males had the highest mean summed PCB concentrations, however, differences between adult males and adult females and juveniles were much greater in the Southern North Sea, the Channel and the Irish & Celtic Seas OSPAR-AAs, and the Irish Sea and Celtic Sea harbour porpoise-AUs than the other Assessment Units/Areas. Adult females had the lowest mean PCB concentrations in all the Assessment Units/Areas, except for the Channel OSPAR-AA and the Western Scotland harbour porpoise-AU, where mean concentrations were greater than juveniles. Given that adult males in the Northern North Sea had far lower concentrations than adult males in the Southern North Sea, the concentrations for the Greater North Sea must be primarily driven by the reported burdens in the Southern North Sea OSPAR-AA, demonstrating the importance of choosing appropriate geographic boundaries for assessment (Fig. 5) (see Table 10, Supplementary Material). These patterns were fairly consistent when these data were aggregated over the most recent ten-year period (2008–2017), as adult females had lower mean PCB concentrations in all the Assessment Units/Areas except for the Channel OSPAR-AA, Western Scotland harbour porpoise-AU and Celtic Sea harbour porpoise-AU (Fig. 6).

3.2.2. PCB congener concentrations

Similar to the results for summed PCB concentrations, adult males had the highest mean concentrations of the top five congeners by concentration, with PCB153 being the most abundant congener in all the Assessment

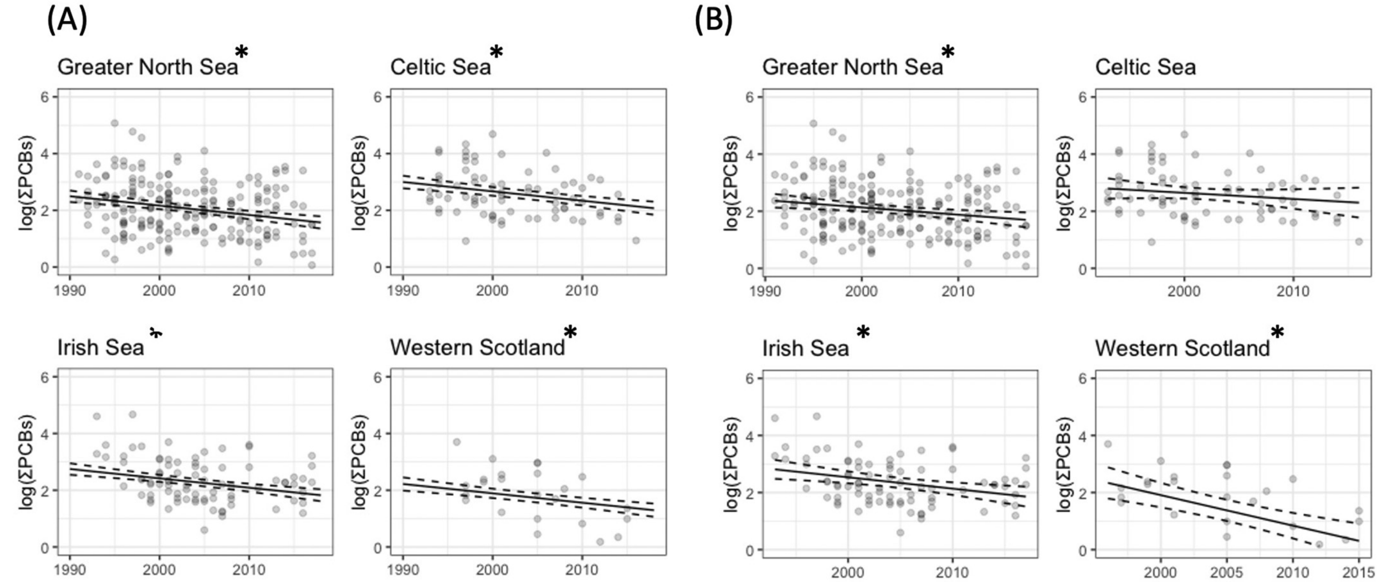

Fig. 2. Modelled temporal trend of the sum of 25 chlorobiphenyl congeners (Σ25CBs) (mg/kg lipid) concentrations in UK harbour porpoise. The solid lines represent the model predictions for each year and the dashed lines represent 95 % confidence intervals (1.96 times the standard error). *denotes significance ($p < 0.05$) (A) Trends derived by predicting mean PCB concentrations for each harbour porpoise Assessment Unit using model coefficients developed for the entire UK. (B) Trends derived by predicting mean PCB concentrations for each harbour porpoise Assessment Unit using model coefficients derived separately for each unit. Trends were not significant for the Celtic Sea Assessment Unit. PCB congeners analysed were: 18, 28, 31, 44, 47, 49, 52, 66, 101, 105, 110, 118, 128, 138, 141, 149, 151, 153, 156, 158, 170, 180, 183, 187, and 194.

Units/Areas among adult males. However, this pattern was not consistent for ICES 7 congeners as juveniles tended to have higher mean concentrations of the lower chlorinated congeners PCB101 and PCB118 (Supplementary Information Tables 11). In general, differences between adult males and adult females and juveniles were greatest for the more persistent, highly chlorinated, congeners (PCB138, PCB153, PCB180), as expected (Fig. 7, Supplementary Information Tables 11 and 12). In most Assessment Units/Areas for the period 1990 to 2017, mean concentrations are higher in juveniles than adult females however, in the Western Scotland harbour porpoise-AU and the Channel OSPAR-AA, adult females have greater mean concentrations of PCB138, PCB153, PCB180 than juveniles.

3.2.3. Toxicity thresholds

When we compared mean PCB blubber concentrations against thresholds for toxic effects and maternal offloading, we found that high proportions of animals are exposed to concentrations deemed to be a toxicological threat

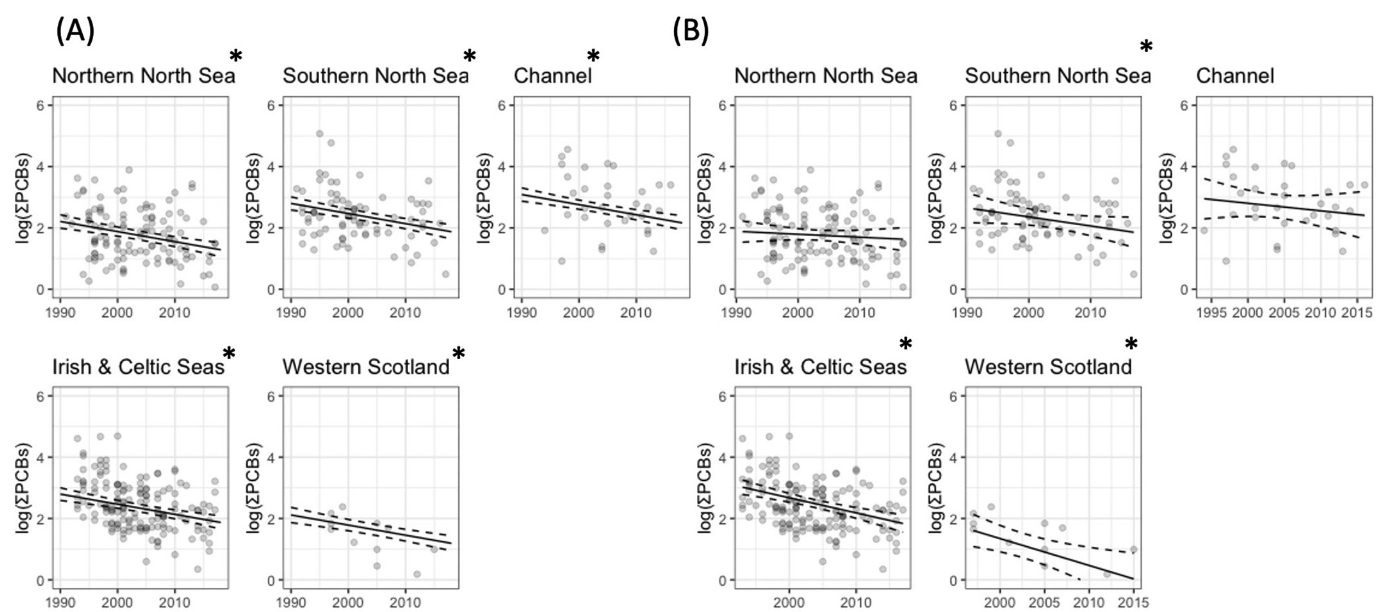

Fig. 3. Modelled temporal trend of the sum of 25 chlorobiphenyl congeners (Σ25CBs) (mg/kg lipid) concentrations in UK harbour porpoises. The solid lines represent the model predictions for each year and the dashed lines represent 95 % confidence intervals (1.96 times the standard error). *denotes significance ($p < 0.05$). (A) Trends derived by predicting mean PCB concentrations for each OSPAR Assessment Area using model coefficients developed for the entire UK. (B) Trends derived by predicting mean PCB concentrations for each OSPAR Assessment Area using model coefficients derived separately for each area. Trends were not significant for the Northern North Sea and the Channel Assessment Areas. PCB congeners analysed were: 18, 28, 31, 44, 47, 49, 52, 66, 101, 105, 110, 118, 128, 138, 141, 149, 151, 153, 156, 158, 170, 180, 183, 187, and 194.

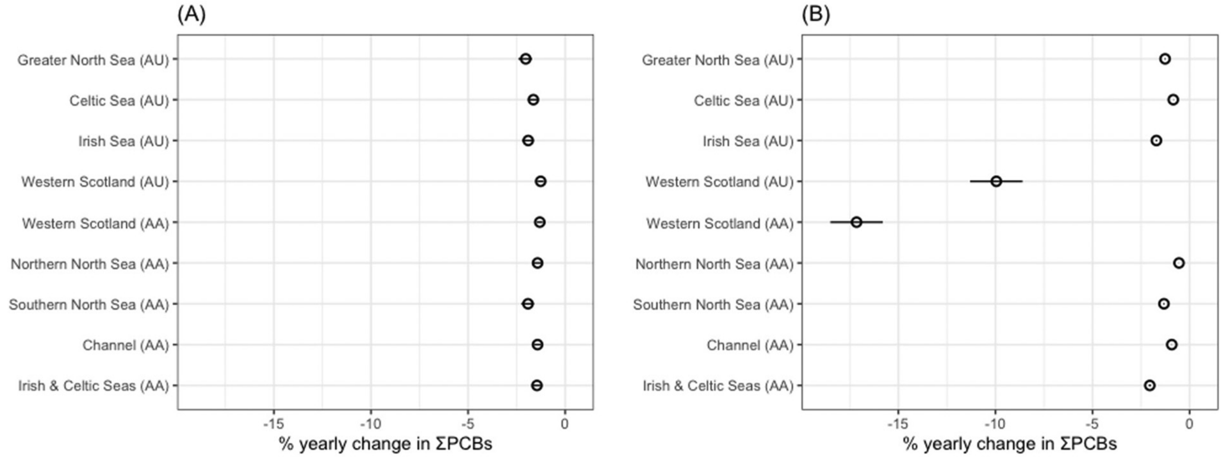

Fig. 4. Predictions from the models for yearly percentage change in PCB concentration in UK harbour porpoises. (A) Trends derived from model for the entire UK. (B) Trends derived by modelling each assessment separately therefore not all trends are significant. The lines represent 95 % confidence intervals (twice the standard error). Trends in (B) not significant for Celtic Sea harbour porpoise-AU and the Channel and Northern North Sea and Irish & Celtic Seas OSPAR-AAs. PCB congeners analysed were: 18, 28, 31, 44, 47, 49, 52, 66, 101, 105, 110, 118, 128, 138, 141, 149, 151, 153, 156, 158, 170, 180, 183, 187, and 194.

(Table 3). Notably, mean PCB concentrations in juveniles, adult males and adult females were above the lowest threshold for sub-lethal toxic effects (9 mg/kg) in all of the Assessment Units/Areas, except for the Western Scotland, and Northern North Sea OSPAR-AAs where mean concentrations in adult females and juveniles were below the threshold for the time period 1990 to 2017 (Fig. 5, Supplementary Information Table 10). In addition, mean concentrations in adult males were above the highest published threshold for toxic effects (41 mg/kg) in the Celtic Sea harbour porpoise-AU and Channel OSPAR-AA indicating likely adverse effects on mature male harbour porpoises in these regions (Fig. 5). More than 68 % of adult males had concentrations that exceeded the 9 mg/kg lipid threshold for sub-lethal effects over both of the time periods analysed (1990–2017 and 2008–2017). When we looked at the most recent ten years of data, we found that mean PCB concentrations in juveniles and adult males were above the lowest threshold for sub-lethal toxic effects in all geographical regions except for juveniles in the Western Scotland harbour porpoise-AU and OSPAR-AA and the Northern North Sea OSPAR-AA (Fig. 6, Supplementary Table 10). Whereas for adult females, who can offload their pollutant burden, mean concentrations were above the threshold in one of the four harbour porpoise Assessment Units (the Celtic Sea) and two of the five OSPAR Assessment Areas (the Channel and the Irish & Celtic Seas).

Comparing the proportions of animals above toxicological thresholds between both time periods it is apparent that the proportions of animals above thresholds have generally decreased however, 44 % still have concentrations above the 9 mg/kg lipid threshold for the period 2008–2017. For adult females, >29 % had PCB concentrations above the 11 mg/kg threshold reported for reproductive failure in the species (as evidenced due to a lack of offloading during gestational and/or lactational transfer) (Murphy et al., 2015) using the full dataset (1990–2017). The proportion was greatest in the Channel OSPAR-AA, as 67 % of females had PCB concentrations above this threshold in both time periods. Whereas within the Celtic Sea harbour porpoise-AU, the proportion increased from 40 to 56 % when only the most recent ten years of data were analysed.

4. Discussion

Here, using the UK harbour porpoise PCB dataset as a case study collected as part of the UK CSIP, we have shown that by employing appropriate methods, marine mammal contaminant data can be used to generate meaningful insights into the trends of contaminants in the marine environment. Moreover, we have shown that the indicator and the proposed methodological framework can be used by Member States to report under

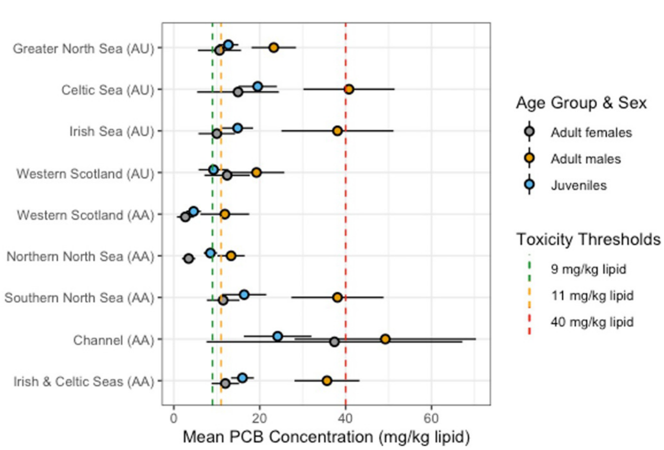

Fig. 5. Mean summed PCB blubber concentrations (mg/kg lipid) in the Assessment Units/Areas between 1990 and 2017 for juvenile, adult female and adult male harbour porpoises. PCB congeners analysed were: 18, 28, 31, 44, 47, 49, 52, 66, 101, 105, 110, 118, 128, 138, 141, 149, 151, 153, 156, 158, 170, 180, 183, 187, and 194.

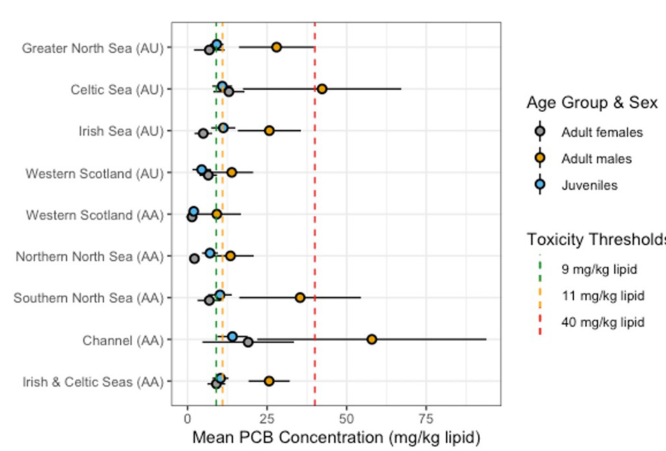

Fig. 6. Mean summed PCB blubber concentrations (mg/kg lipid) in the Assessment Units/Areas between 2008 and 2017 for juvenile, adult female and adult male harbour porpoises. PCB congeners analysed were: 18, 28, 31, 44, 47, 49, 52, 66, 101, 105, 110, 118, 128, 138, 141, 149, 151, 153, 156, 158, 170, 180, 183, 187, and 194.

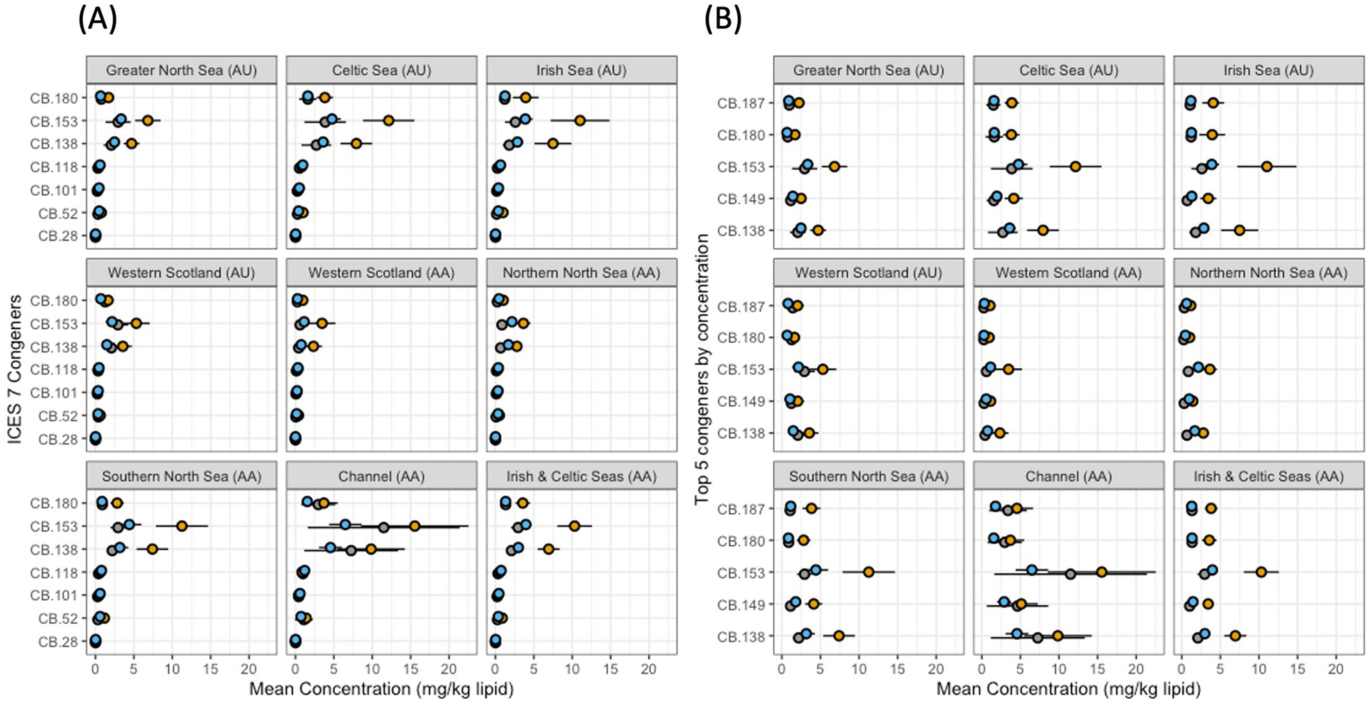

Fig. 7. Mean congener concentrations of each sex-maturity group in the Assessment Areas/Units between 1990 and 2017 of (A) 7 I.E. congeners. PCB congeners analysed were: 28, 52, 101, 118, 138, 153, and 180. (B) Top 5 congeners by concentration. PCB congeners analysed were: 138, 139, 153, 180, and 187. Juveniles = blue, adult females = grey, adult males = orange.

descriptor 8 of the MSFD 'Concentrations of contaminants are at levels not giving rise to pollution effects' in relation to marine mammals. We have developed methodological standards that can be used to assess the trends and status of PCBs and aid the implementation of a candidate marine mammal contaminant indicator within OSPAR. This includes the assessment of temporal trends within OSPAR's contaminants Assessment Areas and harbour porpoise Assessment Units and the comparison of mean summed PCB concentrations with toxicity thresholds, which aim to protect against PCB related effects in marine mammals'.

This paper provides the basis for a framework to assess contaminant monitoring in marine mammals. The framework comprises methods to assess temporal trends and the status of contamination allowing assessments to be made across species and regions if required. The methodology is simple and transparent and allows for different levels of data to be included in the models according to availability. This case study has demonstrated that the indicator can provide regional assessments of levels of PCB contamination in higher trophic level organisms in the marine environment. Nonetheless, there are still a number of issues that need to be considered to ensure our proposed methodology is scientifically robust and cost-effective for use by all.

4.1. Assessing temporal trends

The difficulties in assessing temporal trends of contaminants in marine mammals have been well documented as several confounding factors can influence blubber concentrations (Jepson et al., 2016; Williams et al., 2020a). For example, mature females that have successfully reproduced tend to have lower body burdens of persistent contaminants than mature males and juveniles as they are able to offload some of their pollutant burden via gestation and lactation (Aguilar et al., 1999). Therefore, if proportions of males and females are not consistent across the time period and this is not accounted for in the statistical analysis then any modelled trends are likely to be inaccurate. To overcome this problem, we propose that trend assessments follow the approach we have demonstrated here and exclude adult males and females from the analysis, or alternatively perform the analysis separately for different sex-maturity classes, i.e. juvenile, adult male and adult female. Degree of decomposition (if using samples taken at necropsy) has also been shown to influence contaminant concentrations in marine mammals (Law, 1994). To minimize the impact of changes in pollutant tissue dispersion and concentrations associated with decomposition we propose that trend and status assessments only include blubber samples taken from animals that have undergone minimal to moderate levels of decomposition, according to the condition scoring guide outlined in the ASCOBANS and ACCOBAMS post-mortem protocol (IJsseldijk et al., 2019). A further problem to overcome is the potential confounding effect of body condition on contaminant blubber concentrations as animals in poorer condition have less blubber and therefore, tend to have elevated blubber concentrations of PCBs (Hall et al., 2006). To account for the confounding influence of body condition we recommend that, as per our analysis, the weight and length of each individual is collected so that a proxy metric for body condition can be derived and included in the statistical models. Where animals are biopsied, this information would therefore not be available, limiting the assessment of this confounding factor.

To assess the temporal trends of PCBs in marine mammals we have presented results using two approaches. First, we assessed trends by building a model for the entire geographic range and using the spatial model coefficients to estimate the trends within each of the sub-regions. Second, we assessed trends by building separate models for each of the sub-regions and using those models to estimate trends. Our aim was to compare the results from the two approaches and determine which approach would be more suitable. We found the direction of the trends was consistent for both of the approaches and that the magnitudes of the trends were relatively consistent across most of the sub-regions, excluding the sub-regions with the lowest sample sizes (Western Scotland and the Channel). The main difference between the two approaches is the power to detect statistical significance. An advantage of using one model for the entire geographical region is that the model has greater power to detect trends. However, it should be noted that this approach assumes that spatiotemporal variation is constant across the entire region, which is an assumption that is unlikely to be met. In addition, this approach does not identify sub-regions where

Table 3
Percentage of animals above published toxicity thresholds for Σ25CBs blubber concentrations, split by Assessment Unit/Area, sex-maturity group, and time period: (1990–2017 and 2008–2017). Adult males and juveniles were not compared with the 11 mg/kg lipid threshold as this threshold only applies to reproductive failure in adult females. *PCB congeners analysed were: 18, 28, 31, 44, 47, 49, 52, 66, 101, 105, 110, 118, 128, 138, 141, 149, 151, 153, 156, 158, 170, 180, 183, 187, and 194.

Assessment	Sex-maturity group	N = (*for period 1990–2017)	Mean Σ25CBs Concentration (mg/kg lipid)* 1990–2017	Standard Error	% above toxicity thresholds (N =)					
					9 mg/kg lipid		11 mg/kg lipid		41 mg/kg lipid	
					1990–2017	2008–2017	1990–2017	2008–2017	1990–2017	2008–2017
Harbour porpoise Assessment Unit										
Greater North Sea	adultF	60	10.64	2.57	30 (18)	18 (4)	27 (16)	18 (4)	3 (2)	5 (1)
Greater North Sea	adultM	87	23.27	2.65	67 (58)	58 (15)	–	–	15 (13)	19 (5)
Greater North Sea	Juvenile	206	12.69	1.2	45 (93)	38 (23)	–	–	4 (8)	0 (0)
Celtic Sea	adultF	15	14.94	4.86	53 (8)	67 (6)	40 (6)	56 (5)	7 (1)	0 (0)
Celtic Sea	adultM	17	40.78	5.42	94 (16)	100 (4)	–	–	47 (8)	25 (1)
Celtic Sea	Juvenile	73	19.52	2.28	67 (49)	47 (8)	–	–	15 (11)	0 (0)
Irish Sea	adultF	33	10.01	2.18	39 (13)	17 (1)	24 (8)	0 (0)	6 (2)	0 (0)
Irish Sea	adultM	21	38.1	6.66	95 (20)	86 (6)	–	–	24 (5)	0 (0)
Irish Sea	Juvenile	84	14.86	1.83	56 (47)	52 (12)	–	–	2 (2)	0 (0)
Western Scotland	adultF	27	12.41	2.7	37 (10)	30 (3)	33 (9)	20 (2)	7 (2)	0 (0)
Western Scotland	adultM	14	19.24	3.32	71 (10)	75 (3)	–	–	7 (1)	0 (0)
Western Scotland	Juvenile	25	9.23	1.78	40 (10)	14 (1)	–	–	0 (0)	0 (0)
OSPAR Assessment Area										
Northern North Sea	adultF	28	3.45	0.79	7 (2)	0 (0)	4 (1)	0 (0)	0 (0)	0 (0)
Northern North Sea	adultM	54	13.33	1.62	52 (28)	31 (4)	–	–	4 (2)	0 (0)
Northern North Sea	Juvenile	113	8.51	0.8	34 (38)	26 (9)	–	–	1 (1)	0 (0)
Southern North Sea	adultF	29	11.49	1.95	45 (13)	25 (2)	41 (12)	25 (2)	0 (0)	0 (0)
Southern North Sea	adultM	26	38.1	5.48	92 (24)	88 (7)	–	–	35 (9)	38 (3)
Southern North Sea	Juvenile	75	16.37	2.64	56 (42)	44 (8)	–	–	7 (5)	0 (0)
Channel	adultF	9	37.4	15.21	67 (6)	67 (4)	67 (6)	67 (4)	33 (3)	17 (1)
Channel	adultM	12	49.24	10.77	92 (11)	83 (5)	–	–	42 (5)	50 (3)
Channel	Juvenile	33	24.17	4.02	73 (24)	69 (9)	–	–	18 (6)	0 (0)
Irish & Celtic	adultF	62	10.15	1.79	45 (28)	44 (8)	32 (2)	28 (5)	6 (4)	0 (0)
Irish & Celtic	adultM	39	36.86	4.49	95 (37)	92 (11)	–	–	28 (11)	0 (0)
Irish & Celtic	Juvenile	155	16.2	1.46	60 (93)	44 (18)	–	–	6 (9)	0 (0)
Western Scotland	adultF	7	12.41	2.7	0 (0)	0 (0)	0 (0)	0 (0)	0 (0)	0 (0)
Western Scotland	adultM	8	19.24	3.32	50 (4)	50 (1)	–	–	0 (0)	0 (0)
Western Scotland	Juvenile	12	9.23	1.78	8 (1)	0 (0)	–	–	0 (0)	0 (0)

sample sizes may be too low to accurately detect trends. Modelling each of the sub-regions separately is likely to be a more robust approach and provide a more accurate representation of temporal trends. In addition, by using this approach power analysis can be used to greater effect to identify areas where further sampling is required or where there is no detectable change in concentrations. However, by subsetting these data, statistical power is reduced, and this was apparent in our analysis as year was not a significant term for several of the sub-regions when assessed independently. To further examine the statistical power of our trend assessments this work will be followed up with analysis to determine appropriate sample sizes for the Assessment Units/Areas within OSPAR/MSFD reporting periods, see future work below. We will recommend sample sizes that are required to be able to detect a downward or upward trend if the trend is significant at the 5 % significance level, as per the methodology used for the OSPAR PCB contaminants indicator in fish.

The choice of spatial management unit over which to assess status and trends of contaminants is also an important consideration for the implementation of the indicator. Monitoring blubber PCB concentrations of cetaceans, as well as seals, can be considered as a key aspect in assessing GES according to the MSFD; by linking the pressure indicator to a state indicator, such as marine mammal abundance and distribution, within a DPSIR framework. Given the high mobility of marine mammals, and the distributional range of populations, assessments (of mean concentrations of Σ25PCBs lipid) should be made on a wide scale (range of population or Assessment Unit). Though, status assessments of blubber PCB concentrations can also be undertaken at the group/cohort/individual level, which may be relevant for small populations. It is apparent from our case study however, that choice of the spatial management unit for monitoring trends can have a substantial influence on the results. For example, when the North Sea was treated as two areas (Northern North Sea and Southern North Sea as per the OSPAR Assessment Area) rates of decline were slowest in the Northern North Sea in comparison to other areas and were almost zero when the second modelling approach was used. However, when the North Sea was treated as one area (Greater North Sea as per the harbour porpoise Assessment Unit) the rate of decline was less than that observed for the Southern North Sea OSPAR-AA. Therefore, it was apparent that the trend in the Northern North Sea influenced the overall trend for the Greater North Sea, driving the observed slow decline. Thought it should also be noted that the Greater North Sea harbour porpoise-AU encompasses waters of the eastern Channel. Within the model assessing OSPAR Assessment Areas, latitude was negatively correlated with PCB concentrations, and harbour porpoises inhabiting the Southern North Sea had higher burdens at the start of the time series, compared to porpoises in the Northern North Sea (Fig. 3). It is recommended for future work that both types of assessments (units and areas) are undertaken, as employing OSPAR contaminants Assessment Areas (i.e. OSPAR sub-regions) allows the indicator to be compared more easily with the other OSPAR contaminants indicators that exist for other environmental matrices (e.g., sediment, fish and shellfish). Further, as shown here in the case of the harbour porpoise, it provided further evidence on trends at a North Sea level.

4.2. Status assessment

Our status assessment has demonstrated that the indicator can be used to determine differences in contamination status at different life stages across the geographic range. For example, we found that adult females in the Western Scotland harbour porpoise-AU and the Channel OSPAR-AA had comparatively higher mean PCB concentrations in relation to juveniles than other AUs/AAs when using data from the whole sampling period (see Fig. 5). This may be the result of fewer adult females being able to either carry foetus to term, or newborns dying soon after birth (Murphy et al., 2015; Murphy et al., 2018; Murphy et al., 2010), demonstrating that the indicator can be used to inform better conservation management of animals in these regions. While juveniles had higher mean concentrations of the lower chlorinated congeners PCB101 and PCB118, which may be due to transplacental and lactational transfer being easier for lower chlorinated compounds (Borrell and Aguilar, 2007). Further comparison between the units/areas demonstrated that concentrations increased from north to south and from west to east, which is reflective of greater industrial activity and population density in these areas (e.g. Reis et al., 2018). Our comparison of concentrations of individual congeners found that throughout the entire sampling period, as expected, for the more persistent, highly chlorinated congeners (PCB138, PCB153 and PCB180), adult males had higher concentrations than adult females and juveniles, as a result of bioaccumulation. When we compared concentrations in juveniles and adult females, we found that in most of the units/areas, concentrations were higher in juveniles than adult females. However, this was not the case in the Western Scotland harbour porpoise-AU and the Channel OSPAR-AA as adult females had greater concentrations of PCB52, PCB138, PCB153 and PCB180, and over the entire sampling period, which may be further evidence of reproductive failure in these areas, and further demonstrates the potential of the indicator as a tool to inform on the health status in the species. It is important to note that our analysis of individual congeners demonstrated that of the top five congeners present by concentration, two are not listed in the ICES 7 congeners, PCB187 and PCB149. Therefore, assessing, at least, the full suite of 25 CB congeners is recommended. Moreover, the ICES 7 congeners have been shown to be poor predictors for totals PCBs in several environmental matrices (Megson et al., 2019).

As part of the status assessment, we compared PCB blubber concentrations against published thresholds for toxic effects in marine mammals to assess the significance of risk of pollution, defined by OSPAR as the status where chemicals are at a hazardous level. We found that high proportions of animals are exposed to concentrations deemed to be a toxicological threat. For the purposes of this case study, we aggregated the data over two separate time periods, one spanning the entire 30-year period and the other spanning the most recent 10 years of data. The proportion of animals exposed to concentrations above the 9 mg/kg lipid toxicity threshold for the onset of physiological effects (reproductive and immunological) was found to be lower in all units/areas using the most recent 10 years of data, with the exception of adult females in the Celtic Sea harbour porpoise-AU, and adult males in the Celtic Sea AU, the Western Scotland AU and Western Scotland OSPAR-AA (Table 3). This may be caused by differences in body condition or age, as they were not controlled for in the status assessment, or cases of reproductive failure/geographic variation in pregnancy rates in respect of females (Murphy et al., 2015; Murphy et al., 2020). Notably, 100 % of adult males (and 67 % of adult females) in the Celtic Sea AU were above the 9 mg/kg lipid toxicity threshold for the period 2008–2017. It is also interesting to note that the proportion of juveniles above the 9 mg/kg toxicity threshold was lower in period 2 in all units/areas, though only marginally so in some. Our results demonstrate that it is important to choose an appropriate time period over which to aggregate these data, as the proportion of animals exposed to PCB concentrations deemed to be a toxicological threat varied greatly depending on the time period chosen for the assessment.

4.3. Setting thresholds for status assessment

For the purposes of this case study, we have used three separate thresholds for PCB-mediated effects in marine mammals (9 mg/kg, 11 mg/kg, and 41 mg/kg). The setting of appropriate thresholds for the regional assessments will require further consideration beyond the scope of this work however, we have briefly discussed below some of the possible approaches. It is important to note that any agreed thresholds should account for differences in the effects of exposure to contaminants that occur at different life stages. Exposure to a low dose during a critical period, such as in utero and early development, may have more of a profound detrimental effect than higher exposure later in life (Diamanti-Kandarakis et al., 2009). For example, developmental effects of PCBs have been shown to occur at lower levels of exposure, therefore, it is likely that effects based thresholds focusing on development effects should be lower in juveniles than adults (Baumann et al., 1983; Hall et al., 2006; Vitalone et al., 2010; Williams et al., 2020b).

The wide range of thresholds that exist for marine mammals (Desforges et al., 2016; Helle et al., 1976; Kannan et al., 2000; Mos et al., 2010) highlights the uncertainties that are associated with current approaches and demonstrates that further work is required to improve the accuracy of risk assessments in these species. However, in the absence of laboratory toxicity testing data for marine mammal endpoints such as reproductive effects, a cautious use of thresholds developed for other species combined with epidemiological studies may improve risk assessments. Given the lack of toxicological information that is available for marine mammals, the use of Kannan et al.'s (2000) effects-based threshold of 9 mg/kg sum18–25 CB concentrations for the onset of physiological effects (Jepson et al., 2016) is appropriate to assess unit/area level risk and allows for comparisons to be made across different studies. However, it is important to consider the wide range of toxicity thresholds that exist and make use of new methodologies and approaches where possible. For instance, as demonstrated in this case study, the status assessment could compare unit/area level risk using a range of thresholds to generate best case and worst-case scenarios to provide more comprehensive risk assessments. Within the current assessment, we focused on available thresholds for reproductive effects in marine mammals, as any impairment to reproductive output at a unit/area level will have a direct effect on local population size (assessed via a marine mammal abundance indicator).

An alternative approach, which would facilitate the comparison of the marine mammal indicator with other OSPAR contaminants indicators, would be to compare PCB tissue concentrations against environmental assessment criteria (EACs) similar to the approach used by OSPAR to assess PCB contamination in fish (OSPAR IA, 2017). To carry out those assessments, EACs that have been derived for sediment are used to calculate concentrations of PCBs in fish liver in equilibrium. Hence, a similar approach could be used to convert EACs in fish liver to PCB blubber concentrations in marine mammals. An advantage of this approach is that status assessments would be consistent and comparable across all of the OSPAR PCB indicators for biota. However, biotransformation factors would need to be derived and there may be significant margins of error when converting between different matrices, though keeping assessments based on lipid weight may remove potential biases for the latter.

Comparing PCB blubber concentrations with effects-based thresholds, which could be used to assess health status and determine the probable impacts of PCB exposure, provides an integrated biological and chemical assessment of contaminant impacts. Using effects based thresholds would support the integrated monitoring of contaminants and their effects, provide a more holistic assessment of status and could be used to inform broader ecological assessments (Vethaak et al., 2017). This approach could also be adapted to incorporate exposure to mixtures of contaminants as well as other anthropogenic stressors (e.g., noise pollution) covered by OSPAR.

4.4. Future work

A time-series of pressure indicators are required to help interpret changes in the abundance and distribution of marine mammals, as well as successfully implement a Programme of Measures to achieve GES (Murphy et al., 2021). Pollution by hazardous substances, such as PCBs, is considered as one of the major anthropogenic threats to marine mammals. It is easy to understand and quantify, and there is a clear link with human activities. Analytical methods for contaminant concentrations in tissues are both highly sensitive and internationally standardized for comparison with, for example, tissue PCB levels in other regions. Chemical analysis was consistently undertaken by the same laboratory within the current study, employing the same methodology throughout the years. Although many factors can be controlled, the inclusion of additional datasets from other laboratories will inherently increase analytical variability.

Further work is required to fully implement a marine mammal contaminants related indicator in western European waters. This includes: (1) full agreement on the species/Assessment Units/Assessment Areas against which targets will be set. (2) Agreement on the (persistent) contaminants of concern to be addressed by the indicator. The target set for the status and trend assessment should indicate the level at which conservation objectives will be met. For example, returning anthropogenic chemicals to background levels for the trend assessment. (3) Ensure there is a standardized sample and data collection protocol for stranded and bycaught animals, and biopsy of free-living animals. (4) Development of a standardized reporting methodology, including the creation of a database of individual pollutant levels, based on national input, which contains the relevant information from which to make such assessments. (5) Agreement on, and further development of, thresholds to be employed for the contaminants of concern. Continued development of dose-response relationships between PCBs and reproductive impacts for cetacean populations is required. This includes separate risk assessments from exposure, in addition to dose-response relationships, for dl-PCBs and non-dl-PCBs. (6) Further assessment of confounding factors such age, body condition, reproductive activity, and health status on individual pollutant loads. (7) Specifically, for the summed PCB trend assessment, further work is required to estimate the statistical power to detect a significant trend between reporting periods. The ability to detect changes in summed PCB concentrations will largely depend on the variability of concentrations and the confounding factors, and future work will explore the ability to accurately detect trend at a range of temporal scales, using low, medium and high frequency sampling. Work should also further explore any limitations that may exist when only the ICES 7 congeners are employed, including an assessment of trends using only the ICES 7 congeners. Finally, (8) further work is also required to assess the effects from exposure to multiple pollutants, such as additive and synergistic effects of exposure to PCBs and other pollutants in western European waters, including new emerging pollutants.

CRediT authorship contribution statement

Conceptualization: SM, RSW, PDJ, AB. Investigation: RW, AB, AB, JLB, JB, NJD, RD, MtD, RP, MP, RW, PDJ, OL, SM. Formal analysis: RSW, SM, OL. Data curation: RSW, AB, AB, JLB, JB, NJD, RD, MtD, RP, MP, RW, PDJ, SM. Roles/Writing - original draft: RSW, SM. Writing - review & editing: AB, AB, JLB, JB, NJD, RD, MtD, MP, RW, OL.

Data availability

The authors do not have permission to share data.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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