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Thesis for degree of MSc by Research Ocean Sciences

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Key words: Pollutants, organochlorines, metals, Europe, conservation, harbour porpoise

I hereby declare that this thesis is the results of my own investigations, except where otherwise stated. All other sources are acknowledged by bibliographic references.

This work has not previously been accepted in substance for any degree and is not being concurrently submitted in candidature for any degree unless, as agreed by the University, for approved dual awards.

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Abstract

In European shelf seas, harbour porpoises are considered particularly vulnerable, with negative impacts from high levels of pollutants, which can harm many aquatic organisms. Higher trophic species, such as porpoises, are particularly at risk due to bioaccumulation of pollutants. Negative effects on reproduction and resistance to disease have been demonstrated for porpoises in the region. Identifying locations where high densities of porpoise and concentrations of pollutants co-occur would identify where mitigation is most needed. This study attempts to address this gap in our knowledge by producing maps of key pollutants and of porpoise abundance at a regional scale.

This study sets out to identify where in European waters harbour porpoises are at higher risk from Cadmium, Lead, Mercury, PAHs and PCBs – by producing maps of pollutants to compare with porpoise abundance, identifying overlap. Relationships between recorded concentrations of pollutants and relevant environmental variables were established and used to predict spatial variations in concentrations of sedimentary pollutants across European waters. These were then compared with the lethal dosages (LD_{50}) for mammals, and pollutant concentrations in sediment above the LD_{50} were plotted to show the regions of concern.

This study found that large areas of the North Sea pose a risk to harbour porpoises with sediment pollutant levels above the LD_{50} values for marine mammals around the Orkney and Shetland Isles (Cadmium), along the mainland European coastline (Mercury) and even across the entire North Sea region (Lead). No predicted PAH and PCB concentrations were above the LD_{50} values. When these predictions were overlapped with porpoise abundance, the southern North Sea was predicted to be a region of risk for harbour porpoise populations.

Further work is required to eliminate erroneous results and to improve the accuracy of the maps, and to incorporate data from the water column and within biota. However, this study provides a solid starting point in assessing risk from pollutants on a greater spatial scale than is possible by point sampling (which is logistically and financially impossible at the same scale).

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Introduction

The twenty-first century has seen the marine environment facing a wide variety of anthropogenic pressures, greater than in any previous century (Halpern et al., 2008, 2015). These are most evident in shelf seas and within the coastal zone where human activities tend to be concentrated (Ramesh et al., 2015).

Cetaceans form an important component of marine ecosystems across the planet. As top predators, they have a strong top-down influence on food chains (Bengston and Laws, 1985; Williams et al., 2004; Baum and Worm, 2009) and in some circumstances function as ecosystem engineers by influencing numbers of keystone species (Filatova et al., 2019) and through "trophic competition" (Barbraud and Cotté, 2008; Friedlaender et al., 2008). They bring carbon to the seabed and nutrients to the surface – aiding the fight against climate change – through "whale pumps" (Roman and McCarthy, 2010; Roman et al., 2014) and can even alter the chemistry of an ecosystem (Ainley et al., 2010). They are also important "ecosystem sentinels" (Avila et al., 2018; IJsseldijk, et al., 2021). Aside from their ecological roles, these animals have an economical and cultural importance, supporting rapidly increasing wildlife-watching industries (O'Connor et al., 2009; Highman et al., 2014; Hoyt and Parsons, 2014) and invoking a strong sense of connection to the natural world for humans (DeMares, 2015; Yerbury and Boyd, 2018; Yerbury and Weiler, 2020). By being a vital component of ecosystems and human society, it is important that we work towards protecting them from the multiple threats they face.

Of the 90 species of cetaceans currently recognised in the world, over one-third are considered to be somewhere between near-threatened and critically endangered on the IUCN Red List (IUCN – SSC Cetacean Specialist Group, 2022). Threats are numerous: whaling, by-catch in commercial fishing gear, prey depletion by commercial fishing, chemical pollution, collisions with vessels, underwater noise disturbance, habitat alterations associated with offshore construction, and climate change (Evans and Anderwald, 2016; Avila et al., 2018; Evans, 2020).

Pollutants are a well-known human threat, which has been linked to events impacting biodiversity (McCrink-Goode, 2014). Top predators, in particular, are vulnerable, and one such example is mammal-eating orcas (Jepson et al., 2016; Schnitzler et al., 2019). As a result of concerns for aquatic life under increased pressure from anthropogenic pollutants, the EU Marine Strategy Framework Directive (2008) listed pollution as a major descriptor for the assessment of Good Environmental Status. Following a review of the literature, five notable aquatic pollutants were identified by this study as of ecotoxicological concern: Cadmium, Lead, Mercury, PAH (Polycyclic Aromatic Hydrocarbons), and PCBs (Polychlorinated Biphenyls) due to their chronic negative impacts (OSPAR, 2010). In 2002, at the Stockholm Convention on Persistent Organic Pollutants (Stockholm Convention on POPs), PCBs were identified as one pollutant that member states must eliminate from future production lines by 2025 (Fiedler et al., 2019). As a result, regulations were put in place to remove these chemicals (OSPAR, 2010; European Union, 2019).

Unfortunately, organochlorine pollutants persist within the environment long after regulations have been implemented (Voldner and Li. 1995, Jepson and Law. 2016, Williams et al. 2019). Heavy metals – not regulated by the Stockholm Convention – have also been present in the European marine environment for a considerable period of time (Scherer et al., 2003; Radakovitch et al., 2008; García-Tarrasón et al., 2013) as well as further afield (Bloom, 1977; Naser, 2013). Their persistence means that they remain problematic despite legislative efforts and identifying scenarios where chemical pollutants could have population-level impacts that help focus conservation efforts and resources.

Table 1: Toxic effects on mammalian health of Cadmium, Lead, Mercury, PAHs, and PCBs.

Pollutant	Effect on mammalian health	Citation
Cadmium	Carcinogenic Endocrine-disruptive Reduce reproductive ability Reduce immune response	Das et al., 2004; Papers cited by Pan et al., 2010; Zhu et al., 2020
Lead	Behavioural changes Reduced reproductive ability	Munksgaard et al., 2010; Assi et al., 2016
Mercury	Neurotoxic Fatigue Reduced immune response	Das et al., 2004; Schuehammer et al., 2015; Aeseth et al., 2018
PAHs	Carcinogenic Damaged internal microbiome	Gu et al., 2017; Parajuli et al., 2017
PCBs	Reduced reproductive ability Reduce immune response	Jansen et al., 1993; Tanabe et al., 1994; Jepson et al., 1999; Hall et al., 2006; Pierce et al., 2008; Murphy et al., 2015; Fossi et al., 2018; Fair et al., 2020; Williams et al., 2020a

Several physiological and behavioural impacts from pollutants have been reported in marine mammals (Law, 1996; O'Shea, 1999; Reijnders et al., 1999; Hall, 2001; Das et al., 2003, 2004; Evans, 2014; Fossi and Panti, 2018). PCBs have been linked to reproductive failure and immunosuppression in harbour porpoises (Jepson et al. 1999, Jepson et al. 2005, Hall et al. 2006, Murphy et al. 2015). Furthermore, Cadmium, Mercury and PCBs have been shown to "supress lymphocyte proliferation" in marine and quasi-marine mammals at low concentrations; as low as "0.001 ppm" for PCBs, "0.002 ppm" for Mercury, and "0.1 ppm" for Cadmium (Desforges et al., 2016). This is in addition to other biochemical interactions as a result of pollutant exposure (Table 1).

Pollutants of anthropogenic origin are of particular concern for marine mammals, not only due to bioaccumulation (Pierce et al., 2008), but also to a lack of ability of individuals to remove pollutants from their bodies (Tanabe et al., 1994). Females can offload their POP burdens to their calf via the placenta and in their milk (Williams et al., 2020a), resulting in the first-born calf in particular being subjected to potentially toxic levels of contaminants even before they have had a chance to reproduce (Murphy et al., 2015). In addition to these effects on juvenile porpoise, Williams et al. (2021) also found exposure to PCBs to be associated with reduced testes weight in harbour porpoises which they considered will significantly impact the recruitment rate of a population.

Whilst impacts on individuals may be cryptic – due to being predominantly internal (Table 1), they can have population-level impacts. For example, a well-known pod of orcas ranging around the west coasts of Britain and Ireland has steadily declined in numbers over the last 30 years (Beck et al., 2014; Jourdain et al., 2019; Schnitzler et al., 2019). Although a direct link cannot be proven, PCB levels in UK orcas have been detected at very high concentrations (∑PCB concentration > 40 mg/kg lw; Jepson et al., 2016). Desforges et al. (2018) predicts that on a global scale, general declines in orca populations will be seen worldwide as a result of PCB burdens. The same trend is seen elsewhere in humpback dolphins (Guo et al., 2021) and finless porpoise (Xie et al., 2021).

It is therefore important that we understand the extent of the threat posed by these chemicals as well as the geographical areas which pose greatest risk to particular cetacean populations so that appropriate mitigation measures can be taken. This can be done by combining "area-based conservation" and "pressure-based conservation" approaches to identify regions in need of further protection, as suggested by Evans (2018). By knowing where there is an increased risk for cetaceans from pollutants, appropriate conservation measures can be implemented. An example of this approach would be to reduce or avoid (where possible) other pressures such as fishing in regions where cetaceans are likely to be most heavily impacted by pollutants. It will, therefore, be useful to determine where cetaceans might be impacted the most by specific pollutants. To do this, we need maps of both cetacean distribution and pollutant concentrations.

Many studies have generated Species Distribution Maps for a wide range of taxa across the animal kingdom. In recent years, these have extended to cetaceans, and examples from Europe include Gómez de Segura et al. (2008) showing the distribution of small cetaceans in the Spanish Mediterranean, Breen et al. (2017) modelling cetacean abundance around Ireland, and Waggitt et al. (2020) modelling the abundance of a variety of cetaceans in a major part of the OSPAR region. By contrast, there is a significant lack of maps which demonstrate spatial variation in the concentrations of pollutants that are necessary to implement effective conservation measures.

Since pollutant concentrations may correlate with specific environmental variables, we can predictively map these in the same way that we predictively map cetacean abundance. For example, areas with high levels of pollutants may be associated with particular seabed or water mass properties, including depth, sediment, water-column mixing of temperature and salinity. Determining the statistical relationships between known pollutant concentrations and environmental variables is especially important due to the patchiness of sampling of pollutant concentrations. Combining species distribution maps with those of pollutant levels can help conservation efforts in those regions where multiple pressures exist. Entanglement in fishing gear and ingestion of marine debris are considered major threats to populations for which risk maps are useful to identify hot spots (Breen et al., 2017, Currie et al., 2017; Gregorietti et al., 2021). However, similar approaches have yet to be performed for chemical pollutants.

The North Sea represents one of the most industrialised regions in the world, with major settlements around its coastlines (van Mil and Rutte, 2021). The southern sector is influenced by freshwater input from substantial river systems (Howarth, 2001) whereas the northern sector is influenced by prominent oceanic currents transporting water-masses from the wider Atlantic (Winther and Johannessen, 2006). This combination of urbanisation and multiple water-masses originating from different locations makes the North Sea vulnerable to chemical pollution.

Thirty cetacean species have been recorded in European waters (Evans, 2020), although only a few of them are regular within the North Sea (Reid et al., 2003; Hammond et al., 2013; Waggitt et al., 2020). The commonest and most widespread species is the harbour porpoise (Hammond et al., 2002, 2013, 2021; Waggitt et al., 2020; see also Fig. 1). Following overall declines in the second half of the last century, porpoises in the North Sea since the 1990s showed a southward range shift, becoming rarer in the northern North Sea and more abundant in the German Bight and Southern Bight (Hammond et al., 2013; Gilles et al., 2016; Bouveroux et al., 2020; Waggitt et al., 2020), where they often occur in proximity to areas of high urbanisation and riverine input. Moreover, unlike those in some other North Atlantic regions (e.g., Greenland), harbour porpoise in the North Sea exhibit relatively small ranges (Nielsen et al., 2018), meaning that animals may be chronically exposed to pollutants. Therefore, harbour porpoises in the North Sea could be considered vulnerable to chemical pollutants. Indeed, there have been declines of harbour porpoise in the Skagerrak and the German Bight (Hansen and Høgslund, 2021; Nachtsheim et al., 2021) coupled with numerous strandings of the species (Siebert et al., 2006; Peltier et al., 2013; Wright et al., 2013; IJsseldyk et al., 2020), highlighting the value of monitoring harbour porpoises in this region (Peschko et al., 2016). However, it is important to assess harbour porpoise numbers across the entirety of their European range in order to assess whether they are at increased risk in particular locations bearing in mind regional variations in background and biota pollutant concentrations.

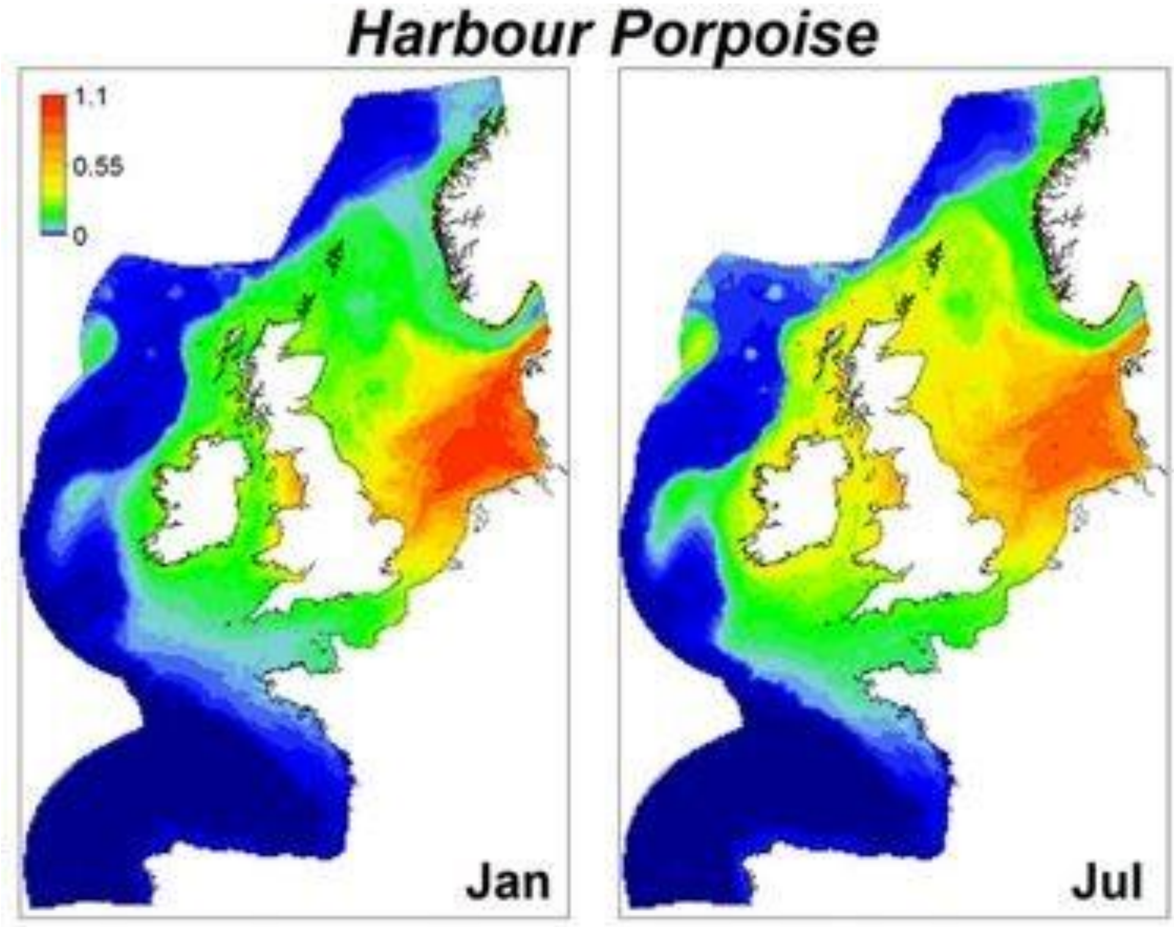

Figure 1: Distribution of harbour porpoises in January and July across West European Seas (Adapted from Waggitt et al., 2020).

Several studies have also focused upon threats to harbour porpoise from chemical pollution in and around the North Sea. Populations of porpoises and some other cetacean species in European waters are exposed to elevated levels of contaminants leading to health concerns (Aguilar et al., 2002; Das et al., 2004; Lahaye et al., 2007; Law et al., 2010; Law et al., 2012; Williams et al., 2020b).

Jepson et al. (2016) presented a map of PCB levels in porpoises around the coasts of the British Isles, indicating hotspots. However, risk assessments on a wider scale for the North Sea have yet to be conducted. Schwake et al. (2002) published a probabilistic risk-assessment to identify overlap between pollutant burdens and reproductive impacts in bottlenose dolphins off the east coast of United States, providing insights into which populations are likely to exhibit impaired reproductive function. A probabilistic risk-assessment approach has yet to be applied to the North Sea, although recommended by Jepson et al. (2009). The current study attempts to risk assess regions of the North Sea where porpoise population declines could occur based on pollutant concentrations using a probabilistic risk-assessment approach.

To achieve these aims, this project: (1) produces maps of predicted Cadmium [Cd], Lead [Pb], Mercury [Hg], Polycyclic Aromatic Hydrocarbons [PAHs], and Polychlorinated Biphenyls [PCBs] concentrations, (2) overlays maps of major contaminants with existing maps of harbour porpoise, and (3) identifies areas where high densities of harbour porpoise coincide with high concentrations of contaminants. The resulting risk-assessment maps will be useful for the Regional Agreement on the Conservation of Small Cetaceans of the Baltic, North-East Atlantic, Irish and North Seas (ASCOBANS) so that they can focus on targeted monitoring of pollutant levels, in addition to mitigating more easily controllable threats, such as noise disturbance and fisheries interactions (Cervin et al., 2020).

Methods

Study Area

Analyses focus on the North Sea between approximately -3.71° and 9.71° longitude and -51.00° and 60.28° latitude. This region encompasses several contrasting habitats within the North Sea, allowing broad-scale comparisons of contaminants in different regions; the deep (>100 m) Norwegian Trench in the north-west, the shallow (<50 m) and freshwater influenced waters of the German and Southern Bight in the south, and the moderately deep and Atlantic Ocean influenced waters around Shetland and Orkney in the north. By including areas with different habitat characteristics, this study region provides the comparisons of contaminant levels amongst contrasting habitats needed to develop predictive models.

General Modelling Approach

To identify areas where harbour porpoise may be exposed to damaging concentrations of contaminants, analyses were divided into several discrete components: (1) point contaminant measurements from previous or ongoing monitoring and research programmes were sourced, quantifying observed variations in contaminants across the study region, (2) continuous environmental measurements from oceanographic models and remote-sensing were collated, allowing environmental conditions at the location of point contaminant measurements to be ascertained, (3) statistical relationships between point contaminant measurements and continuous environmental conditions were estimated, and these statistical relationships used to predict levels of contamination across the study region, and (4) these predictions of contaminations were combined with existing predictions of porpoise densities across the study region, leading to risk assessments for harbour porpoise. The following sections describe these four discrete components in further detail.

Pollutant Data

Measurements of Cadmium, Lead, Mercury, PAHs and PCBs were primarily sourced from the ICES database (https://www.ices.dk/data/data-portals/Pages/DOME.aspx). To maximise sample sizes, these measurements were complemented by those sourced from individual scientists and/or organisations performing research into contaminants but who have not yet contributed data to the ICES database.

Nevertheless, these data were screened to ensure that measurements were comparable and relevant to study aims. Whilst measurements of contaminants were obtained from several media (sediment, water-column, animal tissue) within the ICES database, only measurements from the sediment were considered for analyses. This decision was made because: (1) Focusing on sediments ensured that measurements were broadly comparable. (2) Porpoises' general selection of prey near the seabed in the North Sea (Santos and Pierce, 2003) suggests that contaminants in sediment are more likely to be passed up to porpoises, and (3) there was an insufficient number of data points available from water samples and in tissue for accurate modelling to take place over the entire North Sea region. Amongst sediment samples, measurements of contaminants were collected using a variety of analytical techniques, which could produce different values. Therefore, only samples collected using SED2000 were used to further ensure compatibility, before units were standardized by converting all measurements into µg/g. These approaches provided measurements of contaminants across the North Sea that were considered comparable and relevant for the study aims. Extreme values in deeper waters were removed as they were found in areas where harbour porpoises do not frequent as these values would likely influence the end results to an extent where they exaggerated relationships between contaminants and environmental variables.

When the data collated from the ICES datasets between 1977 and 2021 were selected for the SED2000 analytical technique, dataset sizes were as follows: Cadmium = 5563; Lead = 6526; Mercury = 5387; PAHs = 93585; PCBs = 73456.

Data were collated from around Northern Europe as shown in Figure 2 below:

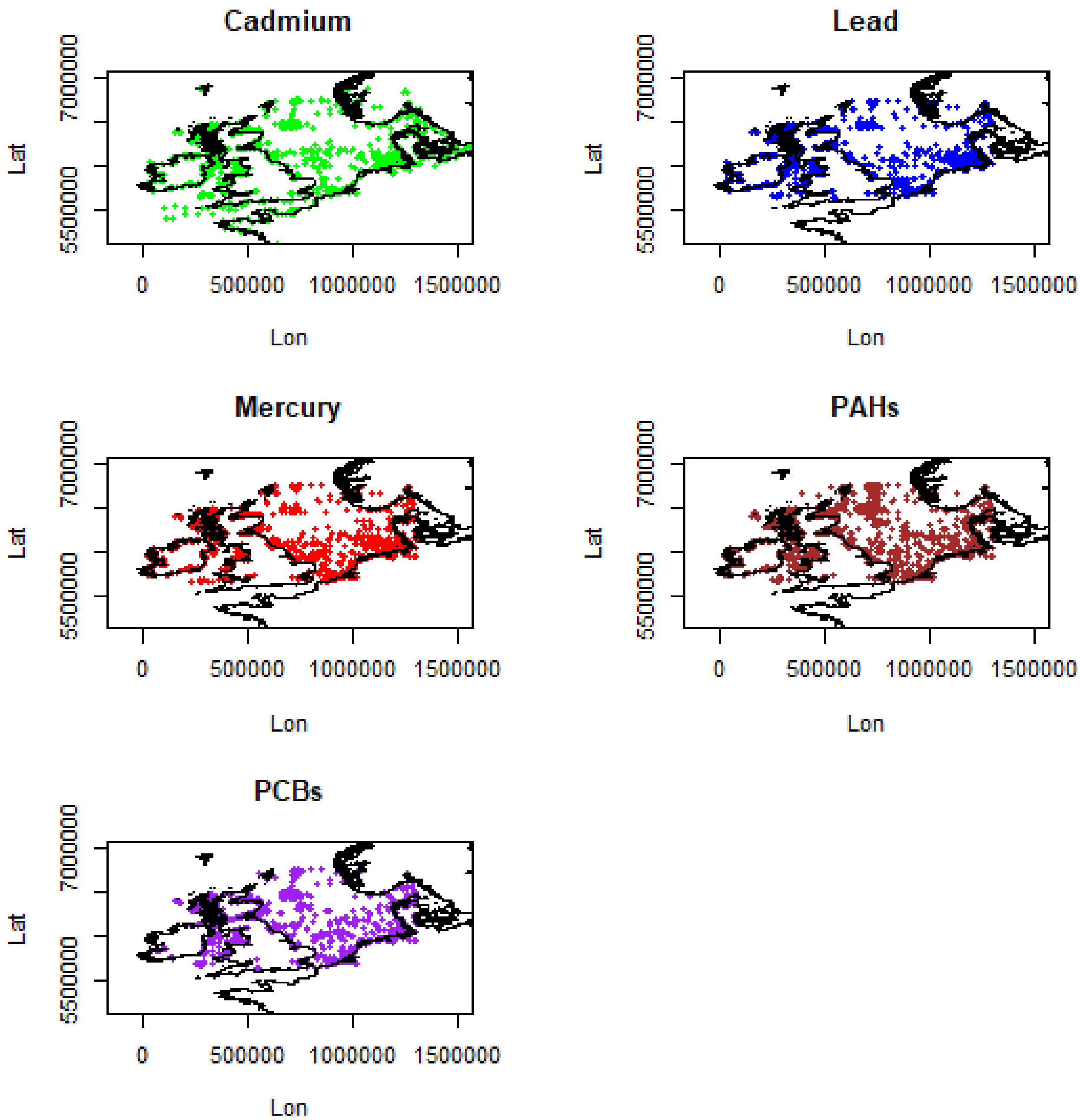

Figure 2: The locations of Cadmium, Lead, Mercury, PAH and PCB samples in the OSPAR region. Coordinates are projected in the UTM30 projection. Axis units of measurement is metres.

Environmental Data

Environmental descriptors were sourced from oceanographic models and remote sensing sources. Collectively, these variables describe persistently different habitats across the North Sea, facilitating the detection and quantification of differences in contaminant levels amongst habitats.

Values of seabed depth were provided at <100m resolution from the EMODnet Bathymetry archive (EMODnet Bathymetry - https://www.emodnet-bathymetry.eu/) and discriminate between shallow waters in the Southern and German Bights, deeper waters in the Norwegian Trench, and intermediate waters in the central North Sea.

Values of water temperature, water salinity and water currents were provided at a 1.5 km and monthly resolution from FOAM AMM15 oceanographic models on the Copernicus Marine Environmental Monitoring Service (Copernicus Marine Service - https://www.copernicus.eu/en). Only values from 2019 were used which allowed for the usage of higher-resolution output from the AMM15 model, which only provides outputs from 2019 onwards. This was not deemed an issue because relative variation in temperatures and salinities would remain similar amongst years.

Combining mean surface temperatures and salinities identify prominent water-masses in the North Sea, discriminating between warm saline Atlantic Ocean water and cooler less saline European shelf water. In isolation, salinity also identifies regions immediately adjacent to large estuaries in the southern North Sea. High variances in surface temperature and salinity amongst months identify ROFI (Regions of Freshwater Influence), linked to freshwater being considerable cooler than seawater in winter, and warmer than seawater in summer. In shelf-seas, tidal currents and water-column mixing are also prominent processes (Simpson and Sharples, 2012), with mean current speeds and the Simpson's Stratification Index (HU_3 = 8cpk E I 37TagQ (Simpson and Hunter, 1974)) collectively discriminating between areas characterised by dynamic turbulent and settled un-turbulent water columns.

Finally, values of sediment properties were sourced from predictive models developed by Wilson et al. (2018). Whilst % of different sediment types are provided, initial inspection showed that % mud helped discriminate amongst relevant habitats, being considerably higher in the Norwegian Trench and central North Sea.

A summary of environmental variables used in analyses is provided in Table 2.

Data Processing

Whilst contaminant concentrations undoubtedly differ seasonally and interannually (Wright and Mason, 1999; Evanset et al., 2016), inspection of the collation of contaminant measurements showed spatiotemporal heterogeneity across months and years, with very few sites being sampled at regular monthly and annual intervals. This heterogeneity prevented reliable analyses into spatiotemporal variation in contaminants across the North Sea, so analyses focused on estimating spatial variation instead.

Although, omitting information on spatiotemporal changes in contaminant concentrations and porpoise densities brings limitations to risk assessment, it seems likely that spatial variation in contaminant concentrations and porpoise densities exceeds temporal variation, with the latter occupying broadly similar habitats in the North Sea across the annual cycle (Gilles et al 2016, Waggitt et al.,2020), and the former likely concentrated around persistent sources of contaminants (i.e. estuaries, ocean currents).

All environmental variables were resampled at 2.5 km resolution. Whilst this is at a lower resolution than originally provided, 2.5 km resolution represented the maximum resolution / number of grid cells where efficient processing and analysis was possible on computers available at the time. To couple the contaminant concentrations with the environmental variables, where multiple concentrations had been sampled at a single location over time, the mean pollutant concentration across all samples and summaries of temperature and salinities within each grid cell was calculated, providing additional information on both in the study area.

Table 2: Environmental variables used in order to predict pollutant concentrations.

Variable	Short Name	Units	Source
Annual Temperature Mean	AnTPM	°C	Copernicus Marine Service
Annual Temperature Variance	AnTPV	°C	Copernicus Marine Service
Annual Salinity Mean	AnSAL	Ppt	Copernicus Marine Service
Annual Salinity Variance	AnSAV	Ppt	Copernicus Marine Service
Simpson Hunter	HU_3	Log10 (m-2 s3)	Copernicus Marine Service
Current Speed	CUR	m/s	Copernicus Marine Service
Distance to Land	LND	Metres	Indicated by coordinates of data
Depth	BAT	Metres	EMODnet
Sediment	MUD	% Mud	Wilson et al., 2018

Modelling Approaches

Model Setup: Generalised Linear Models (GLMs) were selected over commonly used Generalised Additive Models (GAM) owing to several properties of the dataset and inspection of predictions from exploratory GAMs. Specifically, GAMs may be susceptible to overfitting in data characterised by heterogeneous coverage. Here, for example, inspection of relationships from preliminary GAMs sometimes demonstrated illogical and complicated relationships between response and explanatory variables. In addition, there was evidence of oversensitivity to areas of intense sampling and/or extreme values. These overfitted and exaggerated relationships culminated in predictions that were considerably higher than the average values of contaminants in a very small number of cells, often at extremities of environmental conditions, in particular at increased depths. Subsequently, constraining relationships between response and explanatory variables to linear and quadratic terms in the GLM approach provided more logical relationships and reduced the likelihood of overfitting and oversensitivity to heterogeneous coverage. Salinity maps showed areas of estuarine influence compared to fully marine areas whilst sediment maps showed areas of fine sediment compared to areas of coarse sediment. Therefore, it is expected that these can be treated with a linear approach. However, temperature variance and mixing are treated as quadratics as different habitats are identified by moderate or stable salinities and mixed, frontal or stratified mixing. Models used Tweedie distributions due to high variance (high frequency of small values and low frequency of high values) and low means (leading to overdispersion). Explanatory variables were the environmental conditions, whilst the response variables were the corresponding pollutant concentrations.

Model Selection: The process of choosing environmental variables first involved tests for collinearity which showed no evidence of extreme values between the variables. The best-performing combination of environmental variables were chosen through multi modal inferences based on the approaches of Burnham and Anderson (1998) as the actual approaches can be complex, from the MuMIn package and dredge function (Bartón, 2022). This involved constructed models for all combinations of explanatory variables and extracting their AIC values. The AIC values were compared and the model with the lowest AIC value was deemed as optimal. Predictions across the North Sea were then made using the optimal model. Uncertainty in predictions, represented by standard error in model parameters and Deviance Explained, was also predicted.

Model Predictions**:** The final models were combined with continuous measurements to predict spatial variations in contaminants across the study region. To reduce anomalously high predictions in extreme habitats, i.e., within fjords and estuaries, environmental variables were constrained to 5% and 95% percentiles. Based on the environmental variables selected for the model, it is predicted that regions closer to industrialised or densely populated coastal settlements will be subjected to higher pollutant concentrations.

Spatial autocorrelation in residuals was inspected using variograms. This was important as autocorrelation violates the assumption of independence, shrinks confidence intervals, and produces misleading estimates of uncertainty in model predictions.

Software: R Studio was used to run the models and plot results using the following packages: raster (Hijmans, 2022), mgcv (Wood, 2011), plyr (Hadley Wickham, 2011), rgdal (Bivand et al., 2022), maptools (Biavnd and Lewin-Koh, 2022), automap (Hiesmestra et al., 2008), MuMIn (Bartoń, 2022), geoR (Ribeiro Jr et al., 2022), gstat (Pebesma, 2004) and sp (Pebesma and Bivand, 2005).

Probabilistic Risk Assessment

A probabilistic risk assessment was used to identify locations where damaging contaminant levels coincided with high densities of harbour porpoise.

Schwake et al. (2002) proposed the following equation when assessing the risk of PCBs affecting bottlenose dolphins off the US coast where x = a specific concentration, Rx = risk at a specific concentration, P(X) = probability of having tissue concentration X, and P(E|X) = probability of the adverse effect at concentration X (Schwake et al., 2002):

$$\mathbf{Rx = P(X)P(E|X)}$$

The same protocol was applied to the current study, predicting the risk of harbour porpoises being negatively affected by marine pollutants.

Rx = risk that the concentration of the pollutant in the sediment would have a lethal effect on porpoises.

P(X) = probability of a particular pollutant concentration at that location. As this study is aiming to determine the risk value of the pollutant concentration predicted at given coordinates, P(X) is 1 as it has been predicted that there will be that particular pollutant value at those coordinates.

P(E|X) = risk of the concentration of the pollutant having a lethal effect on harbour porpoises. This was calculated as the occurrence of a value equal to or greater than the LD50 – P(LD50) – (lethal dosage for 50% of the population [Merriam-Webster.com]).

How P(E|X) was calculated:

- P(E|X) was calculated by determining if the concentration was above or below the published level resulting in the onset of adverse effects and above or below the published level resulting in the mortality of individuals, as in table 3.
- When [Pollutant] ≥ LD50, P(E|X) = 1; and when [Pollutant] < onset level, P(E|X) = 0.

Table 3: Published LD_{50} values for Cadmium, Lead, Mercury, PAHs, and PCBs for mammals.

Pollutant	LD_{50} Value (µg/g)	Reference
Cadmium	50	Järup et al., 2000
Lead	n/a	n/a
Mercury	30	Shore et al., 2011
PAH	2.1	Sun et al., 2021
PCB	41	Jepson et al., 2016

Rx was calculated for all predicted pollutant concentrations by the GLMs, and then separate maps were plotted across the OSPAR region for both P(O) and P(LD50). As a result, this study was able to show locations in the OSPAR region where the pollutant concentrations were likely to cause both an onset of adverse effects and mortality.

Finally, this was compared to porpoise densities to determine where both the porpoise density was above the 25th, 50th and 75th percentile of the maximum density, and the pollutant concentration was that of the LD_{50} value. Densities of porpoise were sourced from updated outputs from Waggitt et al. (2020), focusing on the North Sea at 2.5 km resolution.

It is important to note that LD_{50} values are relate to contaminant burdens within the body of the mammal. As the maps in this study demonstrate pollutant levels in sediments, due to low number of data points within porpoise themselves, where biomagnification occurs, areas below the LD_{50} in the following maps may be concealing threats to porpoise. For future studies this can be amended with modelling porpoise concentrations, however, this study provides a useful starting point to doing this.

For Lead, it seems that there are no reported LD_{50} values. Flora et al. (2012) reports a chronic poisoning value of "40-60 micrograms per decilitre", however, this is not an LD_{50} value. It should be noted that it is unlikely that porpoises will display symptoms of lead poisoning unless they ingest lumps of lead like a did harbour seal as reported by Zabka et al. (2006). For lead only maps with predicted concentrations were produced.

Results

Cadmium

The model producing the lowest AIC value included which included AnTPV and HU3 (modelled as quadratics) and so was used predict Cadmium concentrations in the North Sea. The largest predicted value of Cadmium concentrations was 4760 µg/g, and the smallest predicted (non-zero) value was 1.4×10^{-11} µg/g. On average, the model, which used the AnTPV and HU3 environmental variables predicted a value of 17.6 µg/g. This model explained 18.00% of the deviance seen in the raw data (Table 4). Figure 7 demonstrates no evidence of autocorrelation with variance in values decreasing with distance apart.

Cadmium concentrations were predicted to be highest along the UK's eastern coast, from the Humber estuary to the islands of Orkney and Shetland. Cadmium can also be found at high concentrations north of the Dogger Bank and along the southern edge of the Norwegian Trench through to the Skagerrak. They also occur where the English Channel meets the Southern Bight and along the Norwegian coast.

Cadmium concentrations are above the LD_{50} value predominantly along the UK's east coast, for example in the Orkney Islands, along the Scottish coast between Edinburgh and Aberdeen, and along the Yorkshire coast south to Flamborough Head. They can also be found in concentrations above the LD_{50} in the Skagerrak.

Areas with a harbour porpoise density above the 25th percentile of the maximum porpoise density in the North Sea and [Cd] above the LD_{50} are solely from the Flamborough Head region out to the south of the Dogger Bank. There are no areas with a harbour porpoise density above the 50th percentile of the maximum porpoise density in the North Sea and [Cd] above the LD_{50} value (figure 6).

Table 4: Model to examine how environmental variables affect Cadmium concentrations in European sediments. Only the models with the 5 lowest AIC values are included in the table.

Model	df	AIC	Delta	Weight
$AnTPV^2$, $HU3^2$	7	546.6	0.00	0.523
MUD, $AnTPV^2$, $HU3^2$	8	548.4	1.85	0.207
AnSAL, $AnTPV^2$, $HU3^2$	8	548.6	2.00	0.193
AnSAL, MUD, $AnTPV^2$, $HU3^2$	9	550.4	3.83	0.077
$AnTPV^2$	5	561.3	14.73	0.000

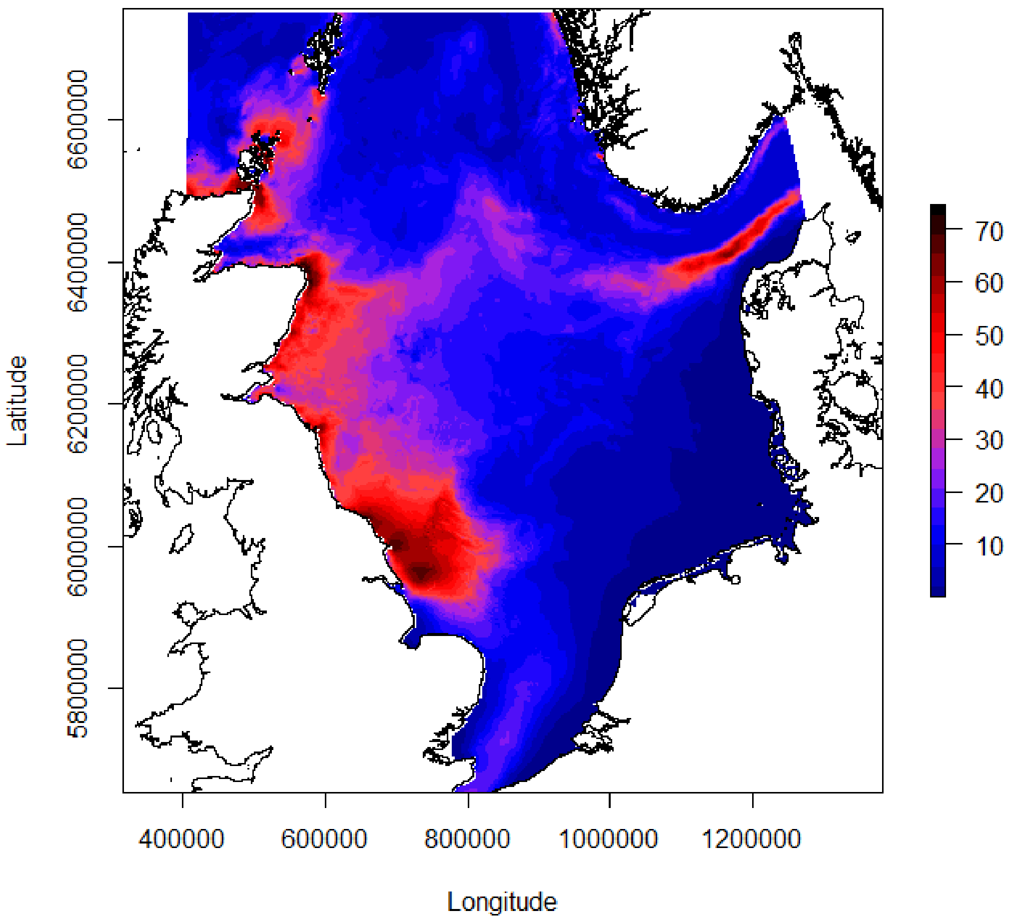

Figure 3: Cadmium concentrations (μg/g) in sediment across the North Sea region, using a UTM30N projection. Axis units of measurement is metres.

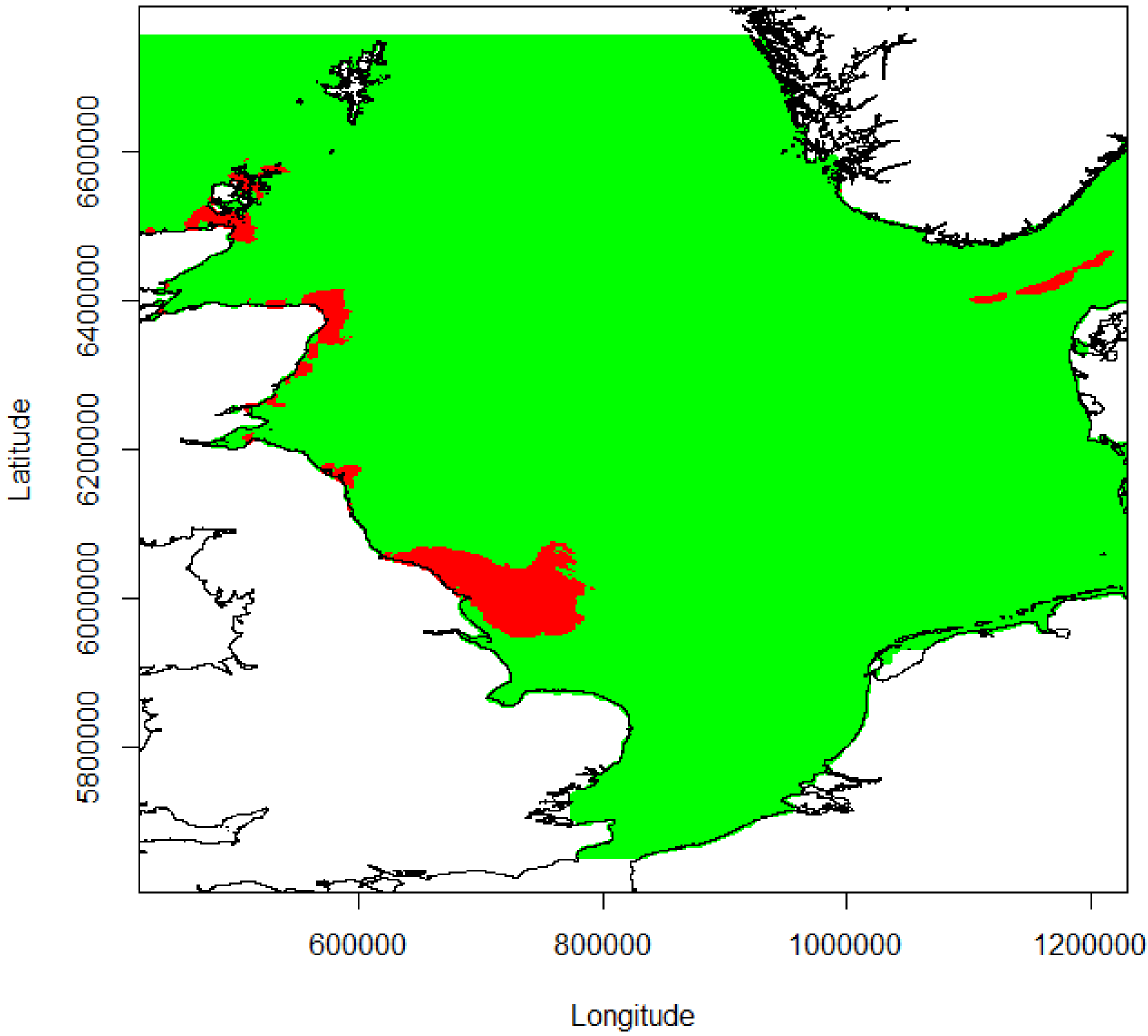

Figure 4: Risk assessment for Cadmium exposure for harbour porpoises across the North Sea region. Regions in red have [Cd] in the sediment above the LD_{50} for Cadmium levels from table 3. Coordinates were projected using the UTM30N projection. Axis units of measurement is metres.

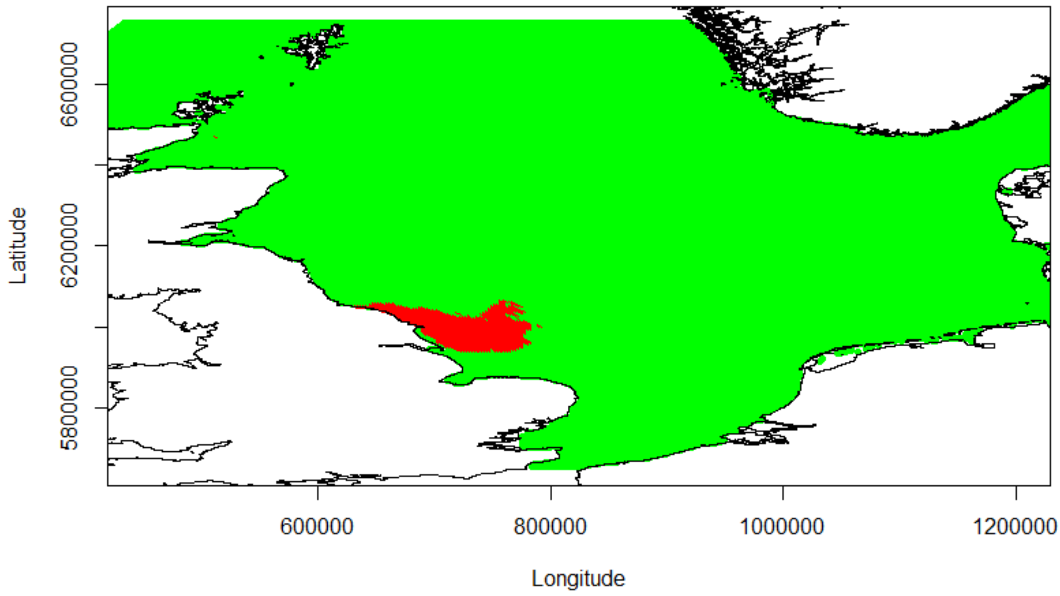

Figure 5: Regions (red) which demonstrate Cadmium concentrations in the sediment above the LD_{50} value for Cadmium and have harbour porpoise densities above the lowest 25th percentile in Waggitt et al. (2020). Axis units of measurement is metres.

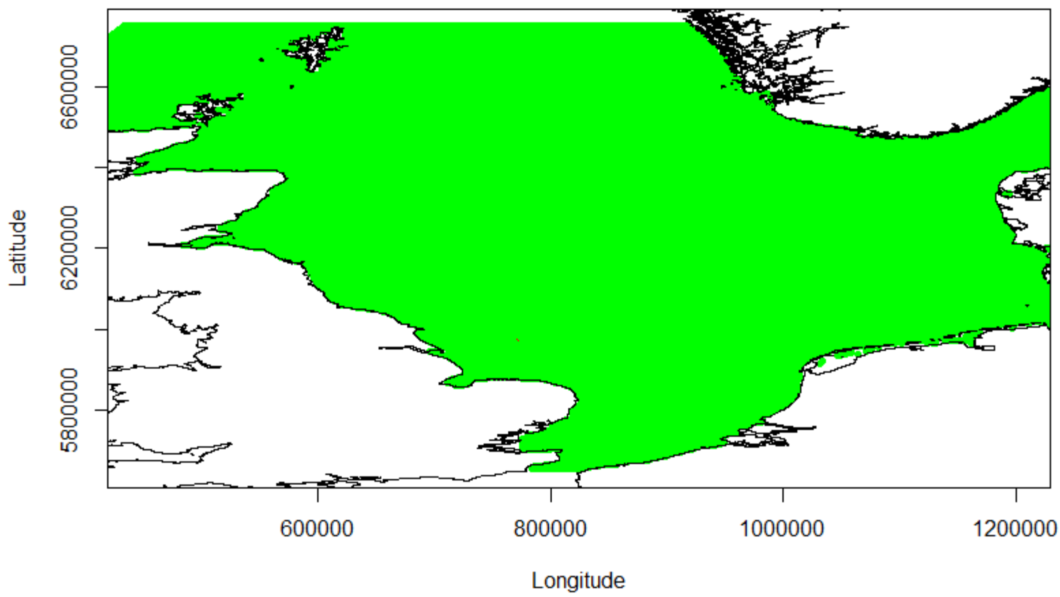

Figure 6: Regions (red) which demonstrate Cadmium concentrations in the sediment above the LD_{50} value for Cadmium and have harbour porpoise densities above the 50th percentile in Waggitt et al. (2020). Axis units of measurement is metres.

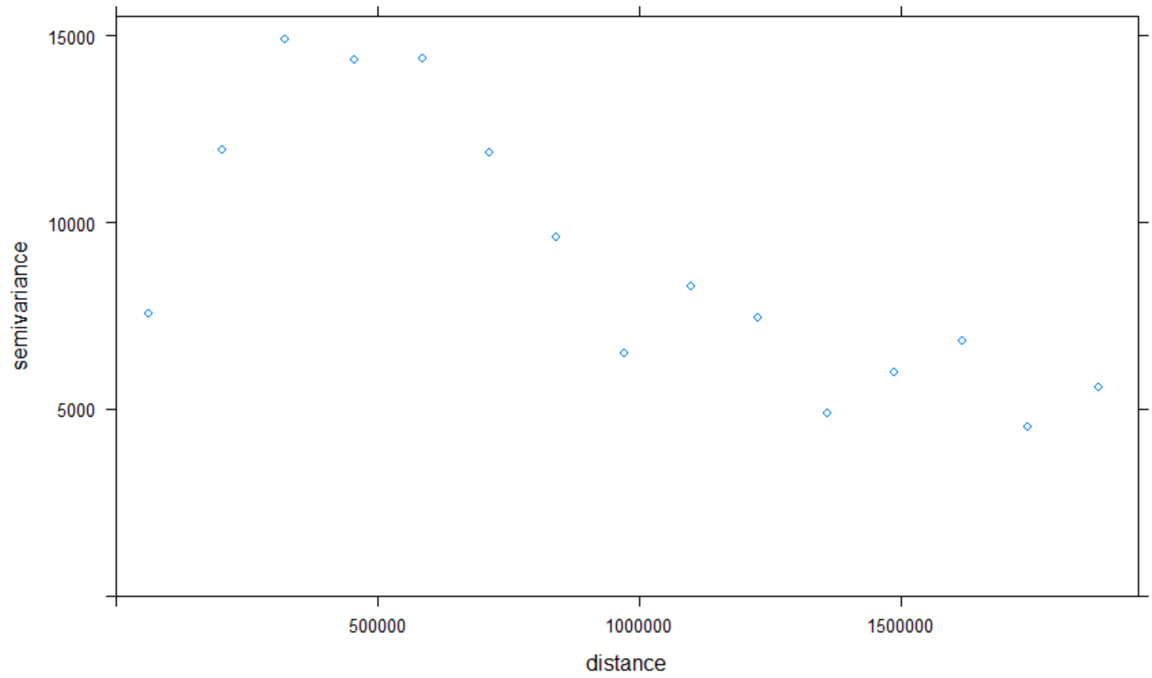

Figure 7: Variogram for the Cadmium model. Sill = 15,000; Nugget = 5,200; Range = 40,000.

<u>Lead</u>

The largest predicted value was 2.1×10^{11} µg/g and the smallest predicted (non-zero) value was 4.0×10^{6} µg/g. On average, the model, which includes the AnSAL, MUD, AnTPV, and HU3 environmental variables, predicted a value of 1.6×10^{9} µg/g. This model explains 5.47% of the deviance explained in the raw data (Table 5). Figure 9 demonstrates no evidence of autocorrelation.

Lead concentrations are predicted to be found at their highest of the west coast of Denmark, particularly in proximity to the Wadden Sea National Park, the north coast of Denmark, and within fjords along the Norwegian coastline such as those near Stavanger and Kristiansand.

Table 5: Model to examine how environmental variables affect Lead concentrations in European sediments. Only the models with the 5 lowest AIC values are included in the table. AnTPV and HU3 are modelled as quadratics. The model which included AnSAL, MUD, AnTPV and HU3 produced the lowest AIC value and so was used to predict Lead concentrations in the North Sea.

Model	df	AIC	Delta	Weight
AnSAL, MUD, $AnTPV^2$, $HU3^2$	9	3128.4	0.00	0.992
AnSAL, MUD, $HU3^2$	7	3138.9	10.48	0.005
MUD, $AnTPV^2$, $HU3^2$	8	3140.9	12.51	0.002
$AnTPV^2$, $HU3^2$	7	3144.3	15.84	0.000
AnSAL, $AnTPV^2$, $HU3^2$	8	3145.7	17.27	0.000

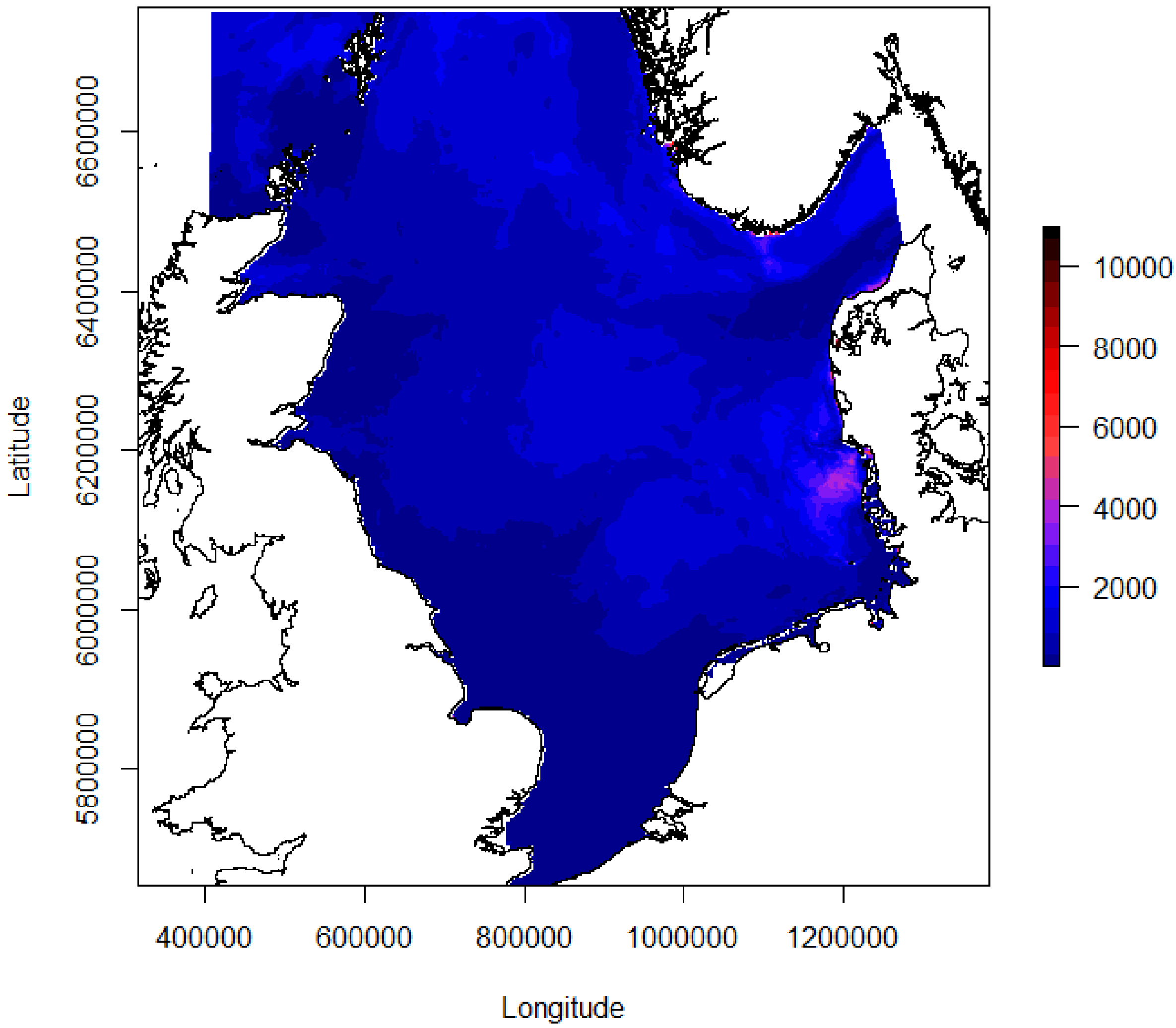

Figure 8: Lead concentrations (µg/g) in sediment across the North Sea region, using a UTM30N projection. Axis units of measurement is metres.

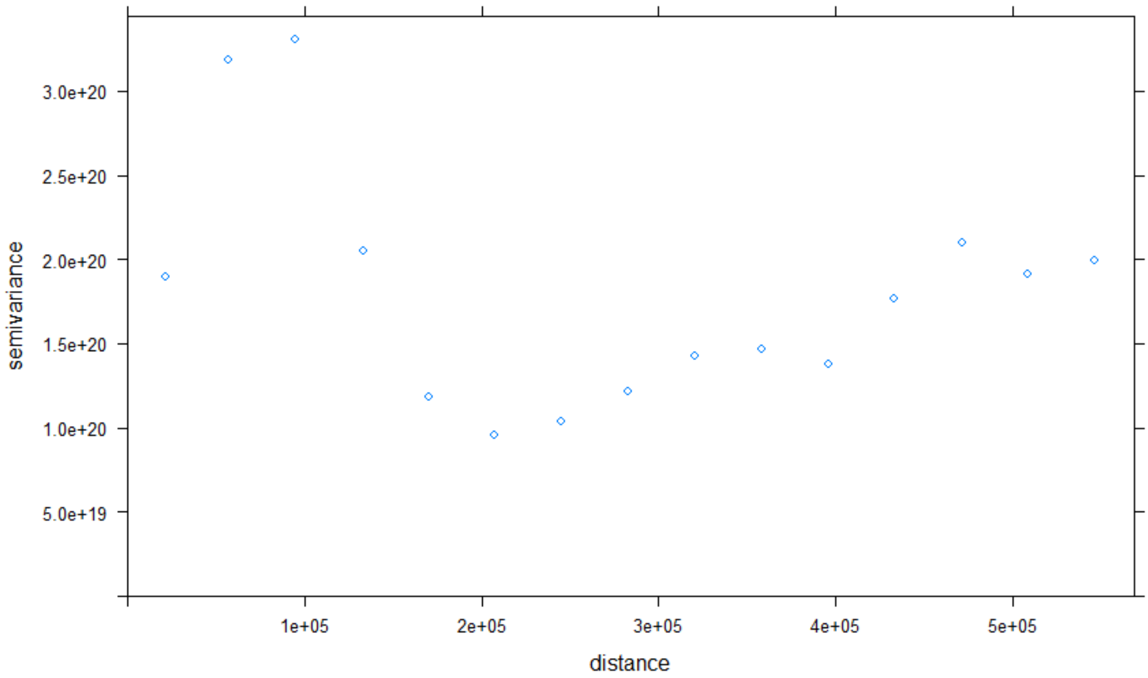

Figure 9: Variogram for Lead model. Sill = 3.2×10^{20}; nugget = 1.7×10^{20}; range = 1×10^{5}.

<u>Mercury</u>

The largest predicted value was 1.0×10^{7} µg/g and the smallest predicted (non-zero) value was 2.3×10^{7} µg/g. On average, the model, which used the AnSAL and AnTPV environmental variables, predicted a value of 3.1×10^{4} µg/g. This model explained 6.39% of the variance found in the raw data (Table 6). Figure 15 demonstrates no evidence of autocorrelation.

The highest concentrations of sedimentary Mercury can be found in the German Bight of the North Sea. Other high concentrations of Mercury occur across the mainland European coast, with progressively lower concentrations being found further out into the North Sea. Other high concentrations can be found in the Thames and Humber estuaries. Lowest concentrations are in the northernmost regions of the North Sea. Mercury concentrations are above the LD_{50} value along the European coastline as well as in the Thames and Humber estuaries, the Wash and the Firth of Forth, and along the Suffolk coast.

Areas with a harbour porpoise density above the 25th percentile of the maximum porpoise density in the North Sea and [Hg] above the LD_{50} are along the European mainland coast from Rotterdam to Denmark's northern coastline. Other regions include the southern banks of the Thames estuary, parts of the Suffolk coast, the Wash and the Humber Estuary. A similar pattern is observed for those areas with a harbour porpoise density above the 50th percentile, except that the north coast of Denmark and the Humber Estuary are no longer identified as of concern. Areas where the harbour porpoise density is above the 75th percentile and [Hg] are above the LD_{50} can be found off the European coast between Rotterdam (The Netherlands) and Bremerhaven (Germany). Off the UK, highlighted areas include where the Rivers Welland and Great Ouse meet the Wash and where the River Medway meets the Thames Estuary.

Table 6: Model to examine how environmental variables affect Mercury concentrations in European sediments. Only the models with the 5 lowest AIC values are included in the table. AnTPV and HU3 are modelled as quadratics. The model which included AnSAL and AnTPV produced the lowest AIC value and so was used to predict Mercury concentrations in the North Sea.

Model	df	AIC	Delta	Weight
AnSAL, $AnTPV^2$	6	-109.6	0.00	0.580
AnSAL, MUD, $AnTPV^2$	7	-107.7	1.91	0.222
AnSAL, $AnTPV^2$, $HU3^2$	8	-106.5	3.07	0.125
AnSAL, MUD, $AnTPV^2$, $HU3^2$	9	-105.0	4.60	0.058
MUD, $AnTPV^2$, $HU3^2$	8	-99.5	10.08	0.004

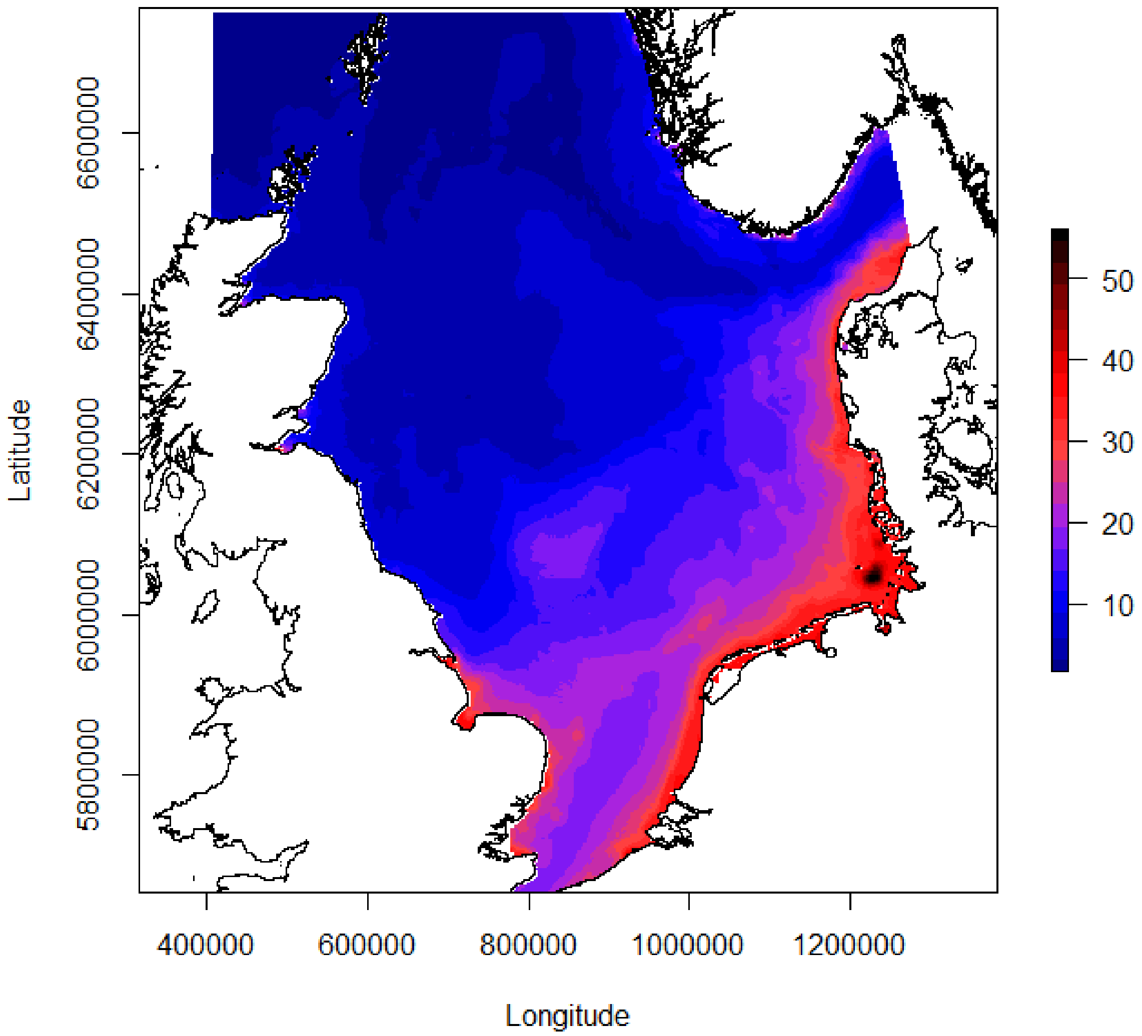

Figure 10: Mercury concentrations (µg/g) in sediment across the North Sea region, using a UTM30N projection. Axis units of measurement is metres.

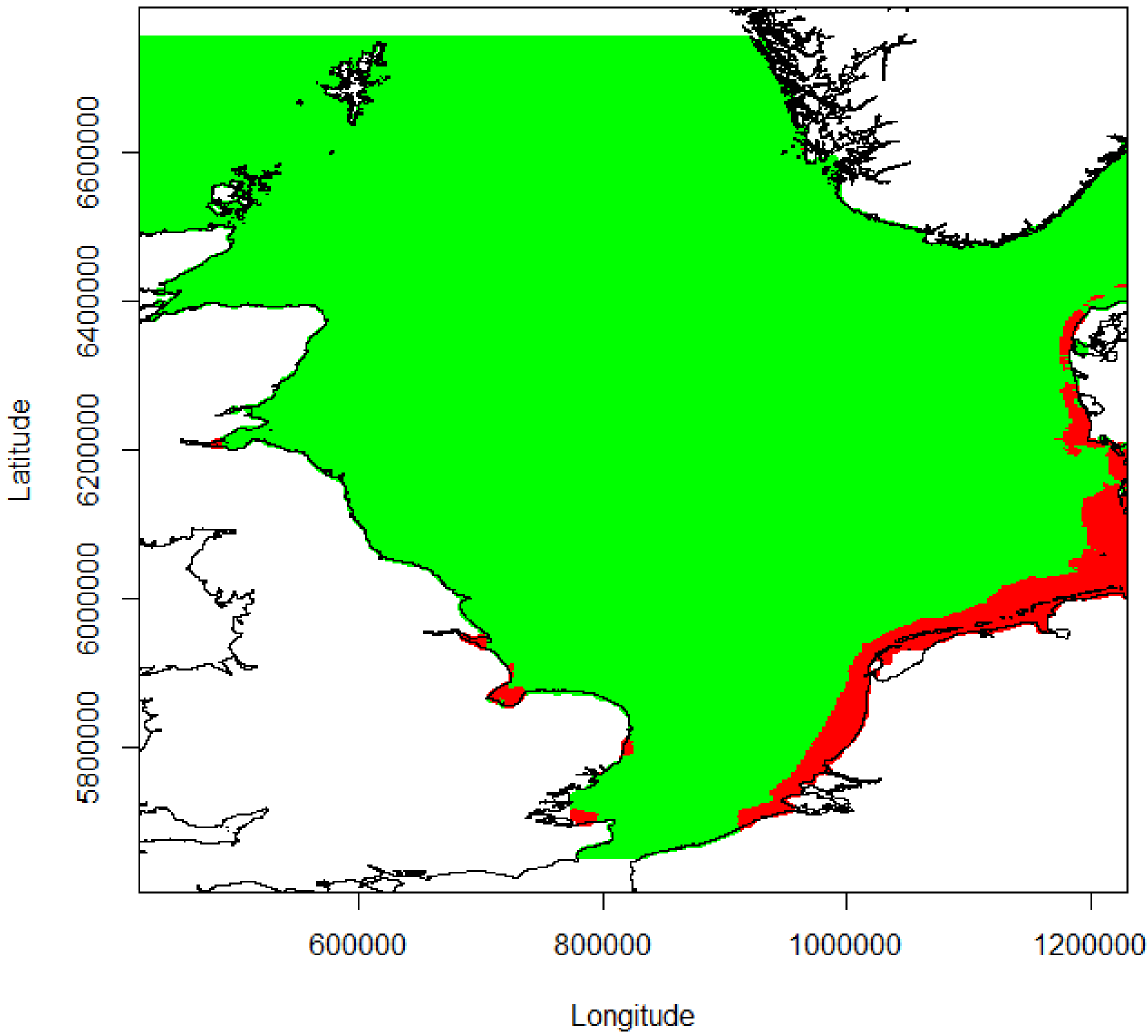

Figure 11: Risk assessment for Mercury exposure for harbour porpoises across the North Sea region. Regions in red have [Hg] in the sediment above the LD_{50} for Mercury values from table 3. Mercury concentrations were above the LD_{50} value in the Moray Firth, the Firth of Forth, the Thames estuary, between Rotterdam and the north of Denmark, and along Norway's south coast. Axis units of measurement is metres.

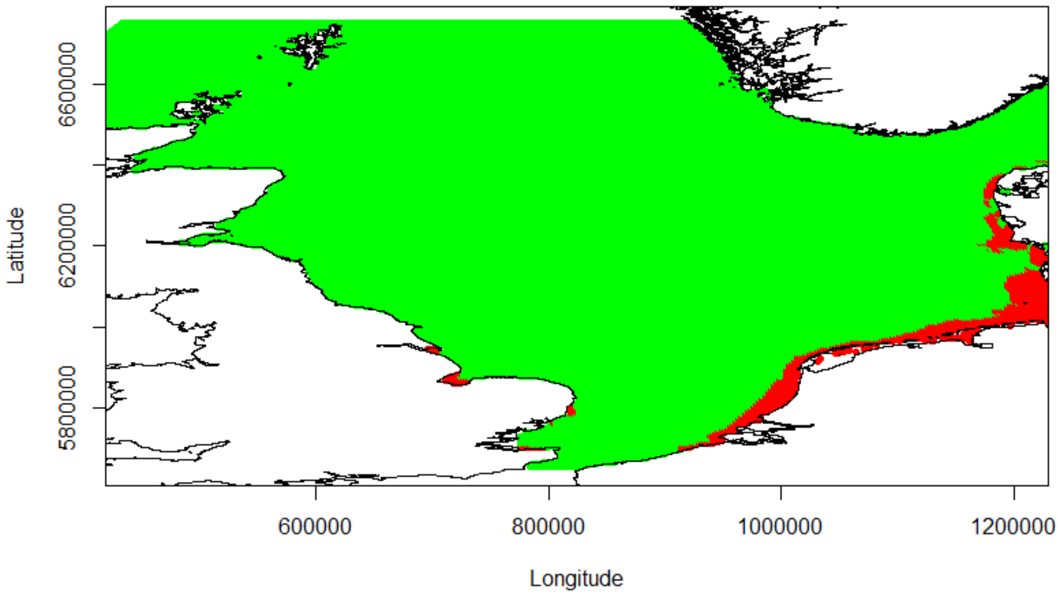

Figure 12: Regions (red) which demonstrate Mercury concentrations in the sediment above the LD_{50} value for Mercury and have harbour porpoise densities above the 25th percentile in Waggitt et al. (2020). Axis units of measurement is metres.

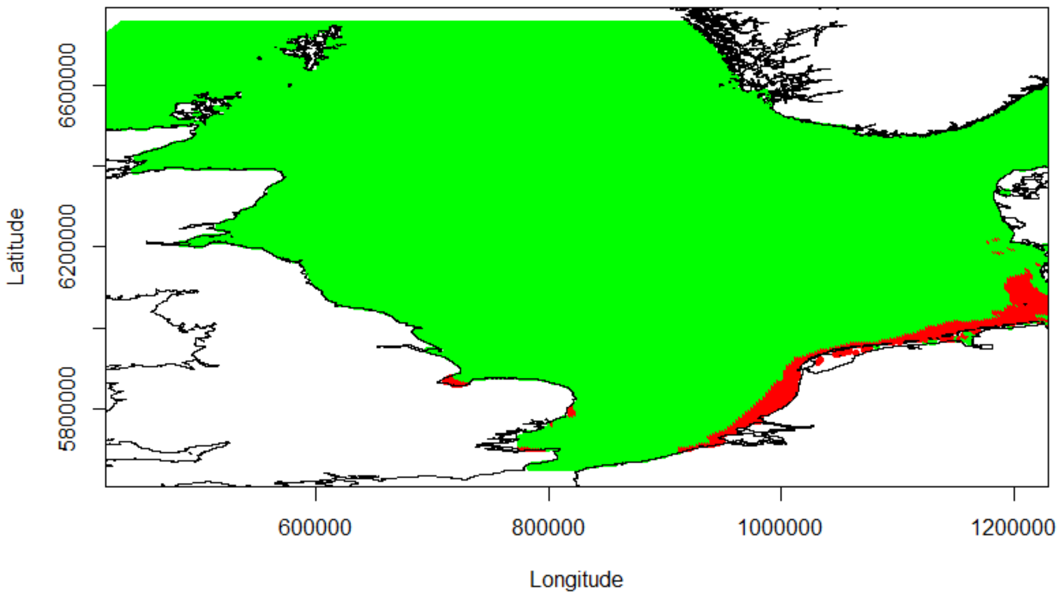

Figure 13: Regions (red) which demonstrate Mercury concentrations in the sediment above the LD_{50} value for Mercury and have harbour porpoise densities above the 50th percentile in Waggitt et al. (2020). Axis units of measurement is metres.

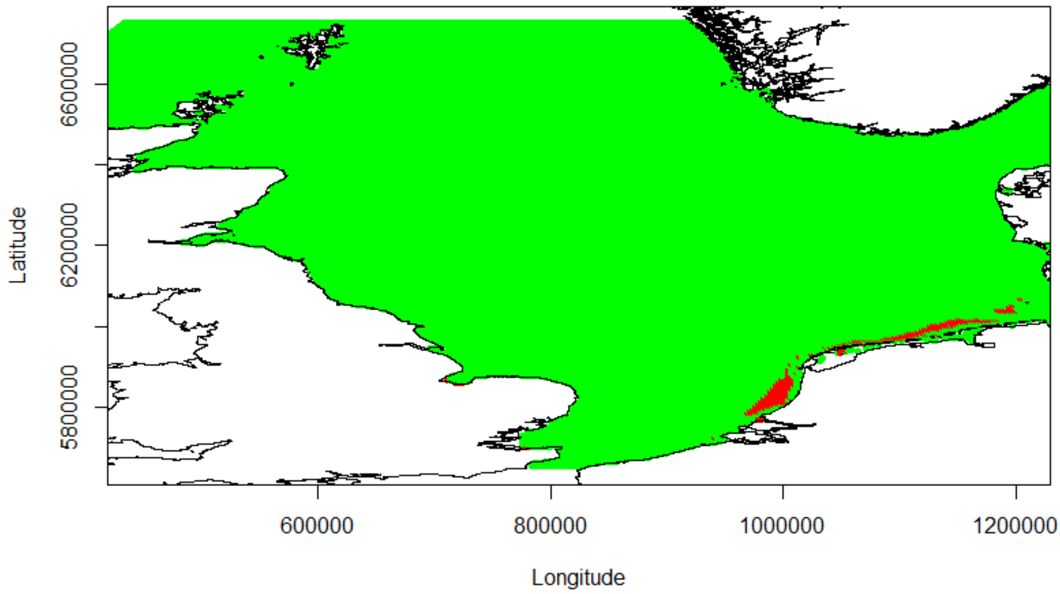

Figure 14: Regions (red) which demonstrate Mercury concentrations in the sediment above the LD_{50} value for Mercury and have harbour porpoise densities above the 75th percentile in Waggitt et al. (2020). Axis units of measurement is metres.

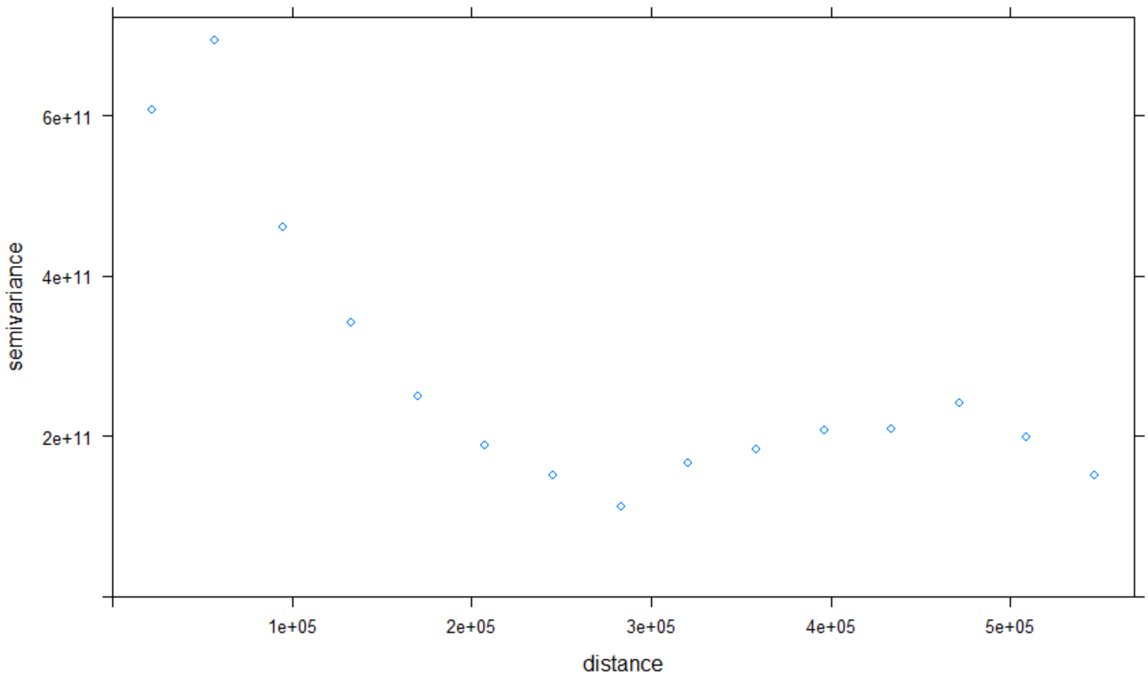

Figure 15: Variogram for Mercury model. Sill = 7×10^{11}; nugget = 5.8×10^{11}; range = 0.7×10^{5}.

PAHs

The largest predicted value was 8.1×10^{4} µg/g and the smallest predicted (non-zero) value was 4.0×10^{12} µg/g. On average, the model, using the AnSAL, MUD, AnTPV, and HU3 environmental variables, predicted a value of 3.6 µg/g. This model explains 20.80% of the deviance found in the raw data (Table 7). Figure 18 demonstrates some evidence of autocorrelation with lower variance with smaller distances apart.

Sedimentary PAH concentrations can be found at their highest along the Norwegian coastline, particularly around the fjords near Kristiansand, Stavanger, and Bergen. PAHs occur in relatively high concentrations on the Dogger Bank, in the Humber Estuary and the Firth of Forth. However, no regions exhibit PAH concentrations above the LD_{50} value across the North Sea.

Table 7: Model to examine how environmental variables affect PAH concentrations in European sediments. Only the models with the 5 lowest AIC values are included in the table. AnTPV and HU3 are modelled as quadratics. The model which included AnSAL, MUD, AnTPV and HU3 produced the lowest AIC value and so was used to predict PAH concentrations in the North Sea.

Model	df	AIC	Delta	Weight
AnSAL, MUD, $AnTPV^2$, $HU3^2$	9	-11202.9	0.00	0.616
AnSAL, $AnTPV^2$, $HU3^2$	8	-11202.0	0.95	0.384
$AnTPV^2$, $HU3^2$	7	-11139.4	63.57	0.000
AnSAL, MUD, $AnTPV^2$	7	-11139.1	63.82	0.000
MUD, $AnTPV^2$, $HU3^2$	8	-11138.1	64.86	0.000

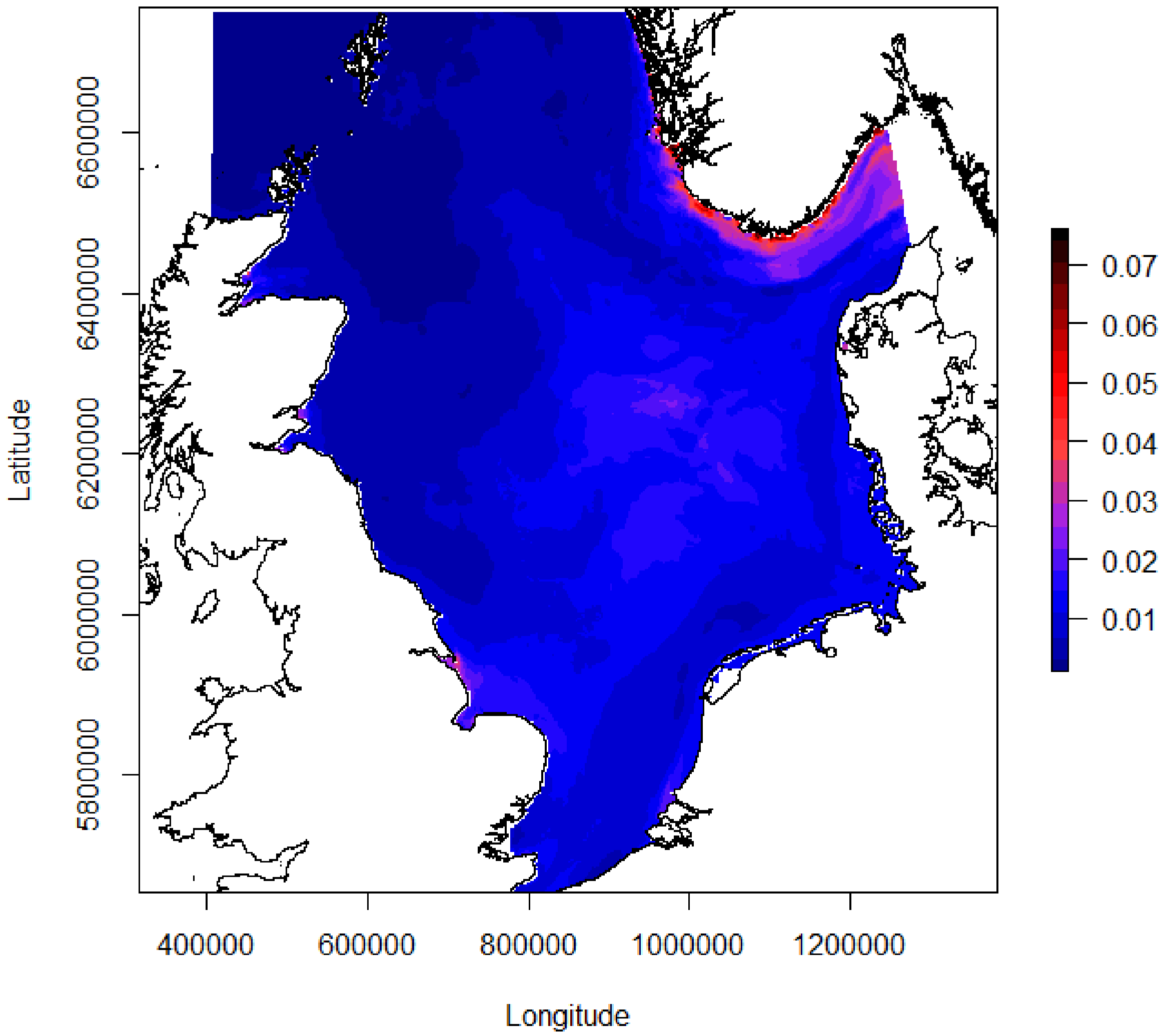

Figure 16: PAHs concentration (μg/g) in sediment across the North Sea region, using a UTM30N projection. Axis units of measurement is metres.

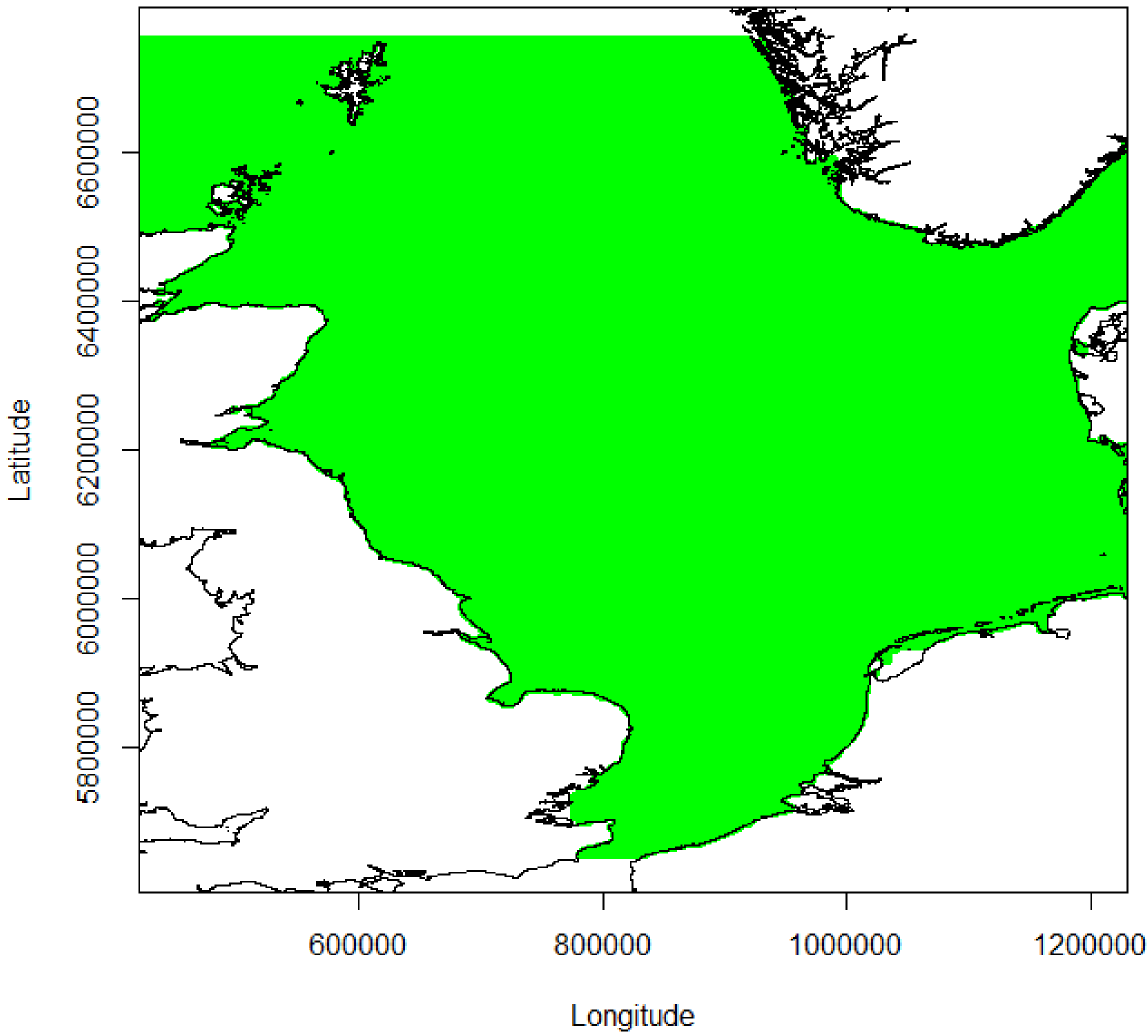

Figure 17: Risk assessment for PAHs exposure for harbour porpoises across the North Sea region. Regions in red have [PAHs] in the sediment above the LD_{50} for PAH values from table 3. No sub-regions of the North Sea had PAH concentrations in the sediment above the LD_{50} value. Axis units of measurement is metres.

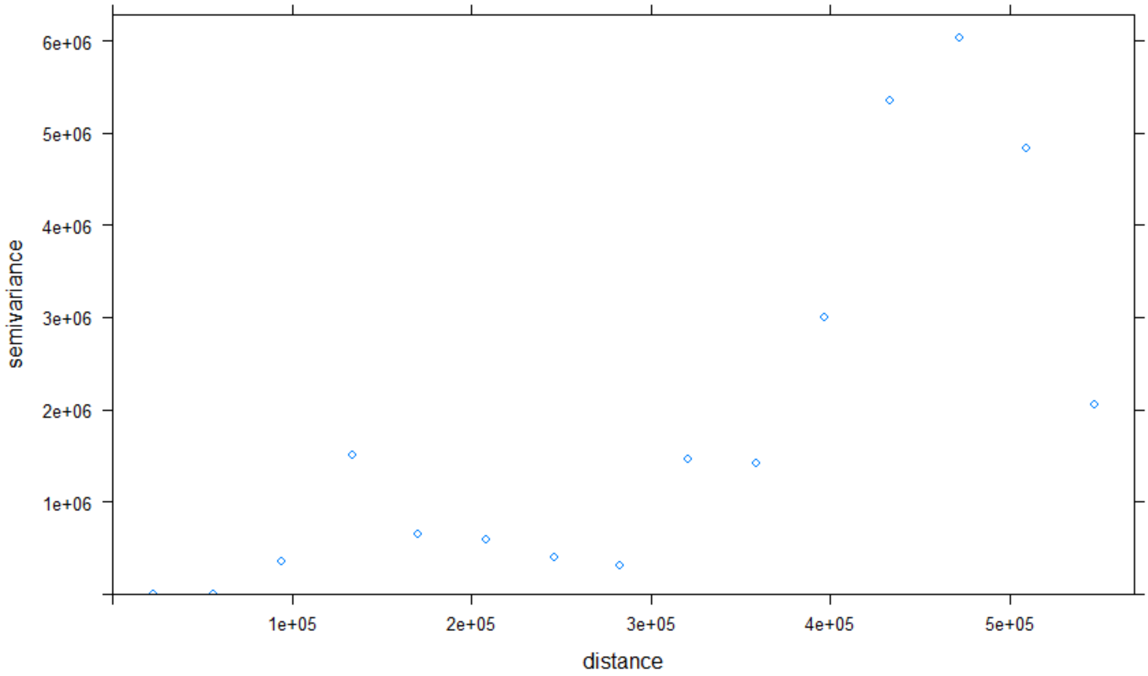

Figure 18: Variogram for PAH model. Sill = 1.4×10^{6}; nugget = 0; range = 1.3×10^{5}.

<u>PCBs</u>

The highest predicted value for PCBs was 2.2x10 µg/g and the smallest predicted (non-zero) value was 2.0×10^{-8} µg/g. On average, the model using the AnSAL, MUD, AnTPV and HU3 environmental variables predicted a value of 9.8×10^{-2} µg/g. This model explains 21.3% of the deviance found in the raw data (Table 8). Figure 21 demonstrates no evidence of autocorrelation.

Sedimentary PCB concentrations are highest off the Humber Estuary. They can also be found in relatively high concentrations near estuaries which are fed from rivers which pass by or through major cities, including London, Edinburgh, Rotterdam, Bremen and Hamburg via the River Thames, rivers flowing into the Firth of Forth, the Rhine/Hollands Diep, and Rivers Weser, and Elbe, respectively. PCB concentrations are predominantly found at highest values in the southern parts of the North Sea. However, similar to PAHs, no sedimentary PCB concentrations were predicted to be above the LD_{50} value for PCBs.

Table 8: Model to examine how environmental variables affect PCB concentrations in European sediments. Only the models with the 5 lowest AIC values are included in the table. AnTPV and HU3 are modelled as quadratics. The model which included AnSAL, MUD, AnTPV and HU3 produced the lowest AIC value and so was used to predict PCB concentrations in the North Sea.

Model	df	AIC	Delta	Weight
AnSAL, MUD, $AnTPV^2$, $HU3^2$	9	-10670.0	0.00	0.672
AnSAL, $AnTPV^2$, $HU3^2$	8	-10668.6	1.45	0.326
$AnTPV^2$, $HU3^2$	7	-10657.2	12.81	0.001
MUD, $AnTPV^2$, $HU3^2$	8	-10655.9	14.12	0.001
AnSAL, MUD, $HU3^2$	7	-10648.0	21.99	0.000

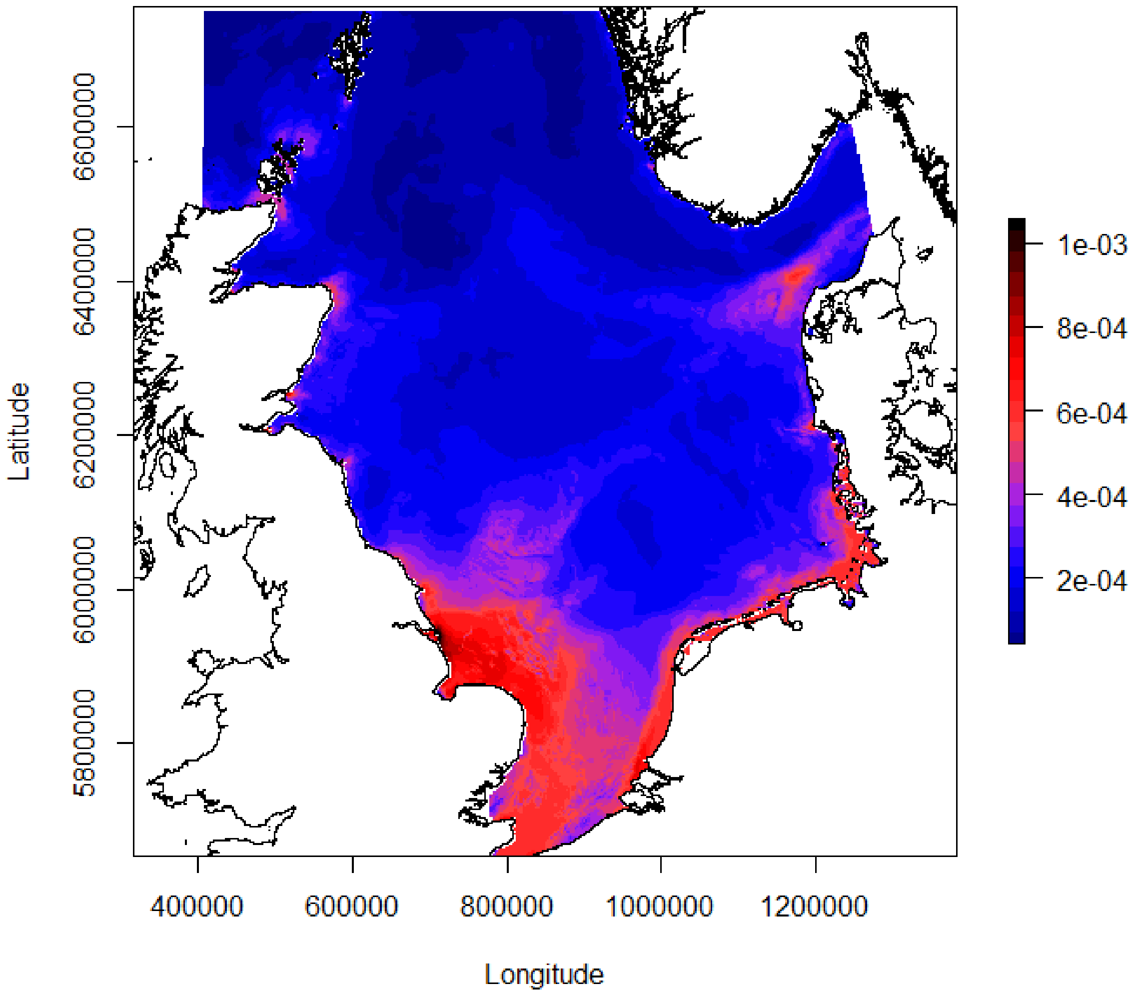

Figure 19: PCBs concentration (μg/g) in sediment across the North Sea region, using a UTM30N projection. Axis units of measurement is metres.

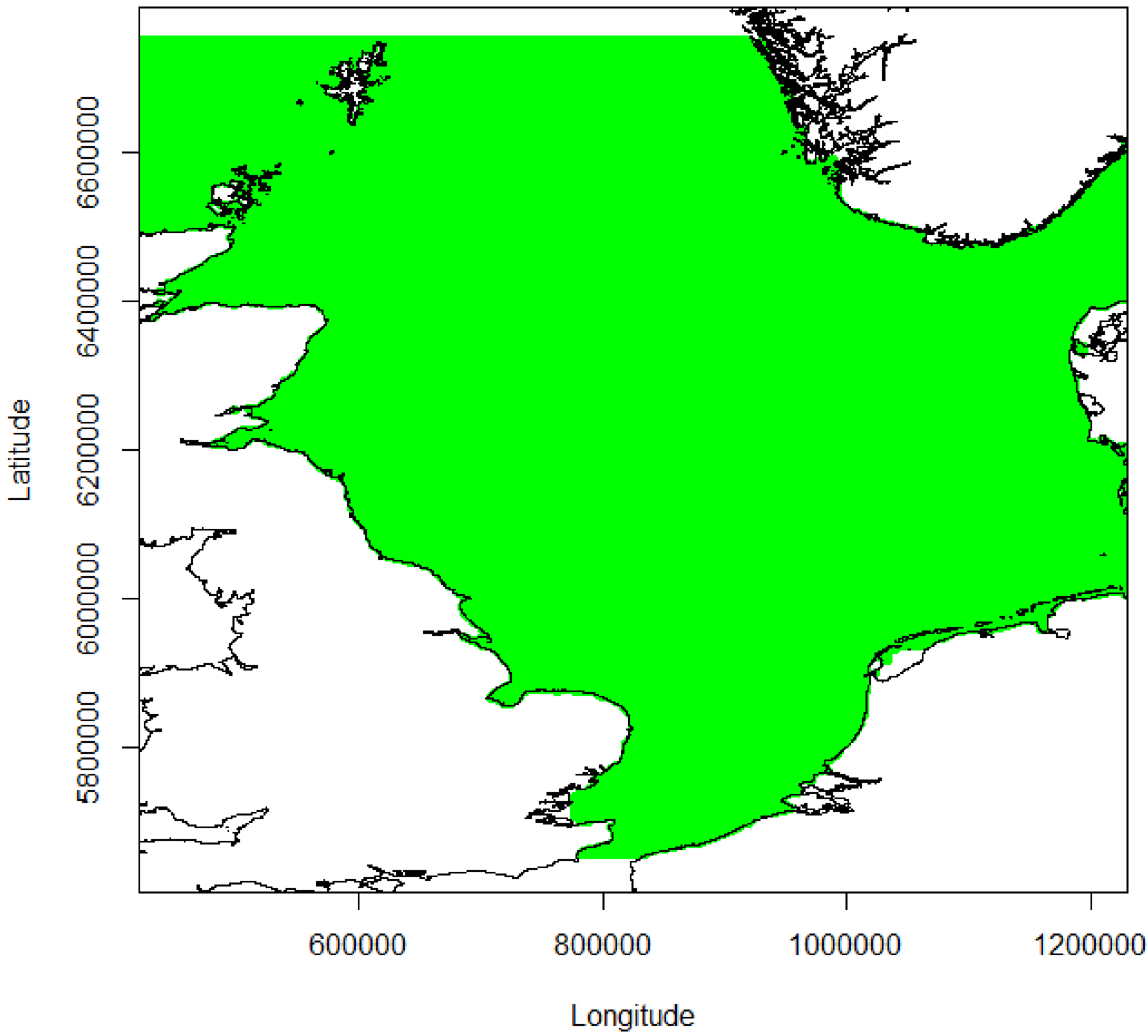

Figure 20: Risk assessment for PCBs exposure for harbour porpoises across the North Sea region. Regions in red have [PCBs] in the sediment above the LD_{50} for PCB values from table 3. No sub-regions of the North Sea had PCB concentrations in the sediment above the LD_{50} value. Axis units of measurement is metres.

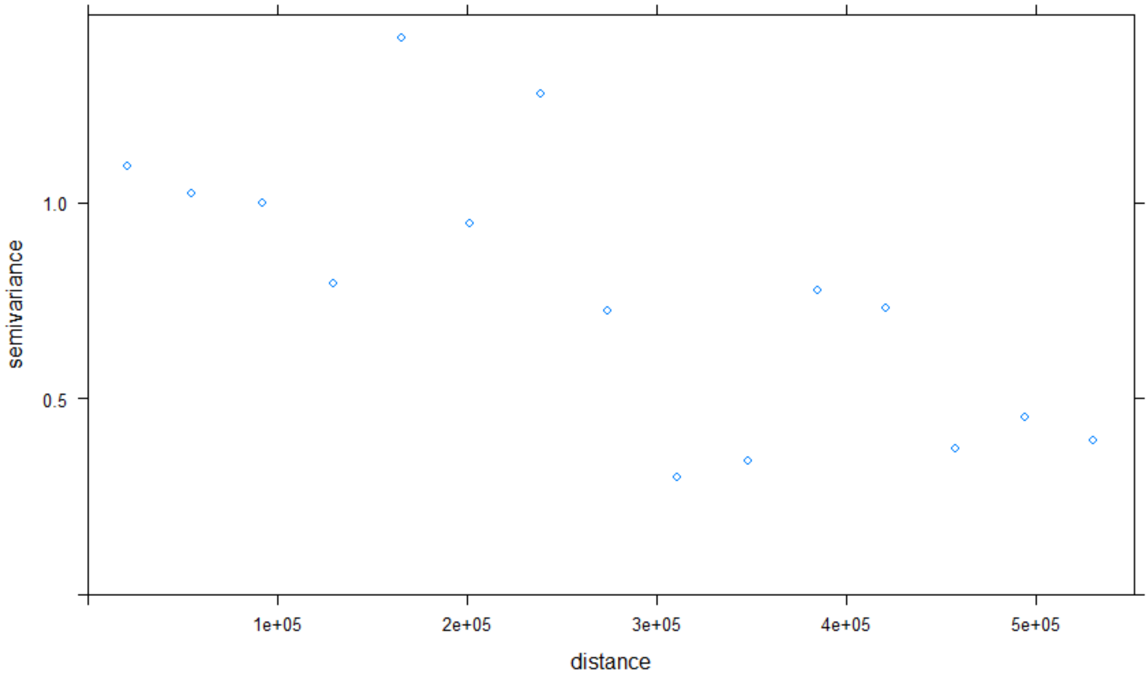

Figure 21: Variogram for PCB model. Sill = 1.3, nugget = 1.3, range = 0.

Model Performance

Based on Table 9 (below), model performance is varied. Overall, deviance explained is low, however, it seems that the models for Cadmium, PAHs and PCBs performed much better than the models for Lead and Mercury.

It is of concern that all models predicted lower maximum concentrations than what was sampled, however, table 9 does not take into account that extreme values were removed from dataset of sampled concentrations to avoid exaggeration of relationships between pollutant concentrations and environmental variables. Mean predicted concentration for Cadmium was the closest to the mean sampled concentration of all the pollutants selected for this study.

Table 9 suggests that Lead was the weakest model of this study with lowest percentage of deviance explained and the maximum, minimum, and mean predicted concentrations were several factors of 10 different to the sampled concentrations.

Table 9: Deviance explained by and R-squared for the model with the lowest AIC values for each pollutant. In addition, there is also a comparison of the largest, smallest and average concentrations of both pollutants sampled and included in the raw data and predicted concentrations in this study (non-zero values only).

Pollutant	**Deviance Explained (%)**	**Sampled Concentrations (µg/g)**			**Predicted Concentrations (µg/g)**			**R-squared**
		Maximum	Minimum	Mean	Maximum	Minimum	Mean	
Cadmium	18.00	4.76×10^{3}	1.40×10^{-11}	1.77×10^{1}	7.72x10	0.04×10^{-2}	1.53×10^{1}	0.0155
Lead	5.47	2.60×10^{11}	4.00×10^{-6}	1.58×10^{9}	2.86×10^{4}	6.33	7.47×10^{2}	-0.265
Mercury	6.39	1.00×10^{7}	2.28×10^{-7}	3.14×10^{4}	6.38x10	1.66	1.17x10	0.0889
PAHs	20.80	8.12×10^{4}	4.00×10^{-12}	3.63×10^{1}	2.3×10^{-1}	5.63×10^{-4}	1.11×10^{-2}	0.0036
PCBs	21.30	2.18×10^{1}	2.00×10^{-8}	9.78×10^{-2}	1.68×10^{-3}	3.99×10^{-5}	2.36×10^{-4}	0.107

Table 10: Standard error of models (µg/g). Also included are the 1st quartile, median and 3rd quartile of each of the models (µg/g).

Pollutant	**Standard Error**	**1st Quartile**	**Median**	**3rd Quartile**
Cadmium	7.59	5.76	1.08x10	2.04x10
Lead	2.99×10^{2}	2.34×10^{2}	5.97×10^{2}	9.58×10^{2}
Mercury	4.94	4.46	7.34	1.66x10
PAHs	3.4×10^{-3}	4.08×10^{-3}	8.59×10^{-3}	1.18×10^{-2}
PCBs	9.71×10^{-5}	1.20×10^{-4}	1.81×10^{-4}	2.63^{-4}

Discussion

This study set out to predict concentrations of five major contaminants (Cadmium, Lead, Mercury, PAHs and PCBs), across the North Sea, in order to assess which regions demonstrated pollutant concentrations which pose a threat to harbour porpoise.

The main findings of this study are: 1) Some contaminants need greater attention than others; 2) Several environmental variables were key across contaminants; 3) Areas of concern depend upon the contaminant; and 4) Model performance is mixed, and reliability depends on the pollutant.

Contaminant Levels

Lead is predicted by this study to be at high levels across the entire study area (figure 9) and shows particularly high levels off the Danish and Norwegian coasts, with relatively high concentrations also in areas of the eastern and northern parts of the North Sea (figure 8).

Hinrichs et al. (2002) suggest that there is anthropogenically sourced Lead from rivers flowing into the German Bight, suggesting the same trends as for Cadmium in the North Sea, with the Elbe and Weser being the main sources for high Lead concentrations in the German Bight. In addition, Rose et al. (2012) demonstrated mechanisms for Lead pollution in Scotland through erosion, which may be a cause of Lead input via the Firth of Forth and Moray Firth. These two estuaries are predicted by this study to have higher concentrations of Lead pollution in the sediments than the rest of the east coast of the United Kingdom. This indicates that large estuaries across the North Sea are the principal source of Lead pollution, and this may be the result of soil erosion from potential natural sources rather than from more direct anthropogenic activities.

PAH concentrations were notably lower than the concentrations of the heavy metals found in this study and figure 17 suggests that the entire North Sea is below the lethal dosage for PAHs. It should be noted that when Law and Whinnett (1992) sampled PAH concentrations in the muscles of harbour porpoises, the highest concentrations were also below the LD_{50} value for PAHs, which supports the findings of this study that PAHs do not result in lethal effects for harbour porpoise in the North Sea, although this statement must be taken loosely due to the effects of biomagnification of this persistent chemical. Polycyclic aromatic hydrocarbons have been demonstrated to be biodegradable when exposed to microorganisms (Carmichael and Pfaender, 1996) and this may suggest that PAHs are being broken down to lower concentrations and could explain the lower predicted PAH concentrations if microorganisms are breaking down the pollutant before it enters the marine environment. It is also possible that PAHs are less ubiquitous than metals as they are predominantly associated with industrial processes, such as the disposal of petroleum-containing materials (Carmichael and Pfaender, 1996), as opposed to the more domestic uses of Lead and Mercury such as their well-known past usages in pipes and thermometers. Bases on the literature, with reduced anthropogenic usage, there will naturally be less input of PAHs into the marine environment.

PCB levels are predicted by this study to be highest along the coasts of south-east England and between northern France and northern Denmark. It is notable that there are a large number of industrialised cities along those stretches of coastline, including Hull, London, Antwerp, Rotterdam, Bremen and Hamburg. Voorspoels et al. (2004) found that with increased distance to land, there were reduced PCB concentrations, indicating that PCBs are transported to the marine environment from inland, and this was supported by the findings of Klamer et al. (2005). This spatial trend is also seen in other biota, with sea stars near the German coast exhibiting the highest PCB concentrations compared to further offshore in a study by den Besten et al. (2001). This trend both in sediment and biota suggests that estuaries in the southern North Sea are a major source of PCBs in the North Sea, and supports the findings of the present study. Jartun and Pettersen (2010) also had main concerns for contaminant levels in harbours of urban or industrial areas, whilst Lockyer and Kinze (2003) found increased PCB concentrations in harbour porpoises off Denmark in comparison to those off Norway.

For PCBs, unfortunately, the risk maps, do not provide a clear enough picture of which regions demonstrate the highest risk for harbour porpoise in European waters. A maximum sampled value of 21.8 μg/g (table 9) is still below the LD_{50} value for PCBs suggesting that sedimentary PCB concentrations are not of a lethal risk to harbour porpoises – before taking into account the likely effect of bioaccumulation. This has to be taken very cautiously as, in the German Bight, there has been a visible decrease in porpoise abundance (Nachtsheim et al., 2021) along with evidence of low fertility rates (Cervin et al., 2020), which may indicate that high levels of PCMs in this region are having `a biological effect. In addition, Williams et al. (2023) found that although PCB levels are dropping within porpoises, "a high proportion of animals were exposed to concentrations deemed to be a toxicological threat". Therefore, the results of this study must be taken cautiously, and with additional data points for harbour porpoise, it will be much easier to assess risk for porpoises from PCBs.

Large amounts of the North Sea are predicted to have contaminant levels in sediment above the chronic poisoning level for Lead predicted by Flora et al. (2012). Additional data sampling points would help confirm this. However, if true, it does suggest that large numbers of European harbour porpoises are at risk from the toxic effects that Lead exposure can bring (Table 1) if ingested. Further studies investigating LD_{50} values for Lead in harbour porpoise specifically are required. This may result in a change of results in this study or a hint at a level of degradation in the environment or detoxification in the animal depending on the number of stranded porpoises with the cause of death being as a result of exposure to Lead.

The areas of greatest concern are the Wadden Sea and German Bight (figure 8). Interestingly, sedimentary Lead was also found across the Baltic Sea region, in the Oslo Rift, Gotland Basin, Gdansk Basin, and Mecklenburg Strait (Bjørlykke et al., 1990; Zillén et al., 2012; Zaborska, 2014; Robinson et al., 2017). However, with relatively fewer porpoises along the Norwegian and Swedish coasts (Waggitt et al., 2020), conservation measures are less important here in relation to marine pollutants. Despite this study suggesting that Lead poisoning is a major issue for harbour porpoises, there are very few papers which suggest that Lead is a danger for harbour porpoises. Camphuysen and Siemensma (2015) reported that studies typically find Cadmium and Mercury to be more commonly found in the bodies of deceased porpoises. Furthermore, they went on to describe POPs being of a larger concern compared to other pressures (even non-pollution related pressures).

Regardless of any findings of the present study, measures to prevent any sub-lethal effects should be put in place to protect harbour porpoise (and other marine mammals) in the southern North Sea from the effects of PAHs (table 1). The findings of Acevedo-Whitehouse et al. (2018) support this study's previous conclusion that regions which exhibit PAH concentrations capable of inducing sub-lethal impacts on harbour porpoise need identifying. Acevedo-Whitehouse et al. (2018) found that DNA damage may be caused by exposure to PAH concentrations below the lethal dosage. To truly assess which areas pose a risk to harbour porpoises from PAHs, some risk mapping of sub-lethal levels should occur.

If correct, this study lends support to the idea that harbour porpoise who succumb to the negative effects of PCBs (Table 1) have been exposed to the sub-lethal impacts of these pollutants (such as those demonstrated by Williams et al. (2021)) for an extended period of time rather than being exposed to lethal, short-term, levels of these legacy contaminants. However, it must be noted that large areas of the North Sea coast are known for their high concentrations of PCBs in top predators, for example the otter off Sweden (Roos et al., 2001), and harbour porpoises around the UK coasts (Jepson et al., 2016). Further data are required to determine whether this study is correct in suggesting that porpoises are subject to prolonged periods of sub-lethal PCB exposure or a short period of lethal exposure as the literature deems otherwise.

Environmental Drivers

It is notable that of the environmental variables, AnTPV occurred in the model with the lowest AIC value for every pollutant. As mentioned previously, this variable indicates the presence of estuarine and freshwater input and land-based human activities. Furthermore, the fact that AnSAL was present for all contaminant models except Cadmium, supports the idea that estuaries and their proximity to anthropogenic activities on land, such as industry and urbanised areas, are a major source of contaminant input into the North Sea. Interestingly, Hunter's Stratification Index also occurred in multiple models, with the model for Mercury being the only one where HU_3 was not present. This indicates that the mixture of depth and current speed could be having a large influence on the spatial distribution of pollutants across the North Sea once those have entered the marine environment. The substrate, mud, appeared in the models as significant for Lead, PAHs and PCBs. This is interesting for the persistent organic pollutants since these chemicals are hydrophobic in nature (Cailleaud et al., 2007) and so may be found in greater concentrations in regions where the percentage of mud in the sediment is higher.

Potential Contaminant Hotspots

Cadmium is predicted by this study to be at its highest concentrations in sediments in the north-western North Sea (figure 3). For Mercury, the south-east coast of England, the Wadden Sea and the German Bight are regions that pose lethal risk to harbour porpoises, according to figure 11 (figures 12-14 also demonstrate that these areas contain a large proportion of porpoises exposed to lethal levels of Mercury). Both can be found in coastal regions but in very different habitats. Mercury has been predicted to be highest near large estuaries along the mainland coast of northern Europe. On the other hand, Cadmium does not seem to be associated with large estuaries and high levels occur principally along the UK coastline.

Cadmium concentrations are above the LD_{50} values for marine mammals off the north-east of England as well as around the Orkney and Shetland Islands. Vane et al. (2020) also found that areas around the UK are subject to relatively high concentrations of Cadmium in sediments. Furthermore, Berg et al. (2000) and Lahaye et al. (2007) found that there were higher concentrations of Cadmium in harbour porpoise off Scotland and Ireland than in the rest of European waters. When comparing the concentrations along the UK coastline to the coasts of mainland Europe, it should be noted that lower concentrations were also seen off Belgium's coastline and the Wadden Sea in the study by Le et al. (2021). However, as shown in figure 2, this study included few data points which may have influenced this result and so the link between the results of this study and those of Le et al. (2021) should be taken cautiously.

In other parts of the North Sea, this study concluded that Cadmium concentrations were relatively low in the German Bight, but higher north of Denmark and south of Norway and Sweden (figure 3). Figure 4 also shows these areas as being areas above the LD_{50} value for Cadmium. These results are also in contrast to the findings of Gao et al. (2013) who demonstrated that metal concentrations were higher in estuaries across Belgium and Germany than offshore in the North Sea. Since Kubier et al. (2019) also demonstrated higher concentrations in water near the city of Bremen, further analyses are needed to confirm this.

The north-east of England is known for larger numbers of harbour porpoises in relation to adjacent areas (Waggitt et al., 2020) that have Cadmium concentrations above the LD_{50} value and, therefore, this has been highlighted in figures 5 and 6 as a region of particular concern for porpoise populations. Das et al. (2006) demonstrated that Cadmium levels were lower in harbour porpoises in the southern North Sea than those found off Iceland. The present study suggests that Cadmium levels are higher in the deeper waters of the northern North Sea and so it is plausible that increased levels of Cadmium found in Icelandic harbour porpoises (Das et al., 2006) may apply also porpoises in Scotland and northern England.

The results of the present study also match the findings of Das et al. (2004), where porpoises in the southern North Sea, around the mainland European coast, had increased loads of Mercury by comparison to those found off Norway and in the Baltic. This further mirrors what is seen in this study where porpoises off the Norwegian coast are exposed to Mercury concentrations below the LD_{50} value for marine mammals. Furthermore, in 1991, a female harbour porpoise stranded in the Castricum municipality of the Netherlands was analysed by Kastelein and Lavaleije (1992) who found high concentrations of Mercury in its liver and kidney. Those concentrations reported by Kastelein and Lavaleije (1992) are below the LD_{50} value for Mercury in table 3, however, and Mercury was not deemed as the cause of mortality for this individual. Despite that, however, this stranding is still important as it highlights high concentrations of Mercury in an individual found stranded in a region indicated by the present study as one of concern.

Cadmium entering the marine environment can be as a result of both natural and anthropogenic processes (Suhani et al., 2021), which include aquaculture and wastewater treatment (Yuan et al., 2019), agricultural uses of phosphate fertilizers (Satarug et al. 2003) which leach into the ground and surface water (Ulrich, 2019), and accumulation in plant species (Satarug et al. 2003). The European Commission is attempting to minimise Cadmium content in fertilisers but so far with limited success (Ulrich, 2019). This could explain why in the present study, Cadmium is not associated with major estuaries near large cities and suggests that a major source of Cadmium input to the marine environment is through the agricultural sector, which is typically away from large cities.

Estuaries are a source of Mercury, especially during periods of flooding (Saniewska et al., 2014) and, with increased climate change, Mercury is more likely to be released from riverine sediments into the marine environment due to more acidic pH concentrations (Kammann et al., 2023). Within the region modelled by this study as having higher predicted Mercury concentrations, there is also a variety of large estuaries which are fed from rivers, such as the Rhine, Weser and Elbe. Panagos et al. (2021) compared the levels of Mercury in soils across Europe and showed that the Elbe estuary is a hotspot with increased levels in soils. Interestingly, the mouth of the Elbe was the region which was predicted by this study to have the highest concentrations of sedimentary Mercury.

Overall, this study suggests that one cannot necessarily assume that high-risk areas occur alongside urbanized estuaries. This will vary between contaminants. In the case of Mercury, the results of this study show a trend consistent with the initial hypothesis that urbanised estuaries are the principal source of this pollutant in the marine environment, whereas the source of Cadmium may be either natural or anthropogenic.

Model performance

Broad patterns have been seen across the North Sea which match the hypothesis that increased pollutant concentrations can be found in coastal regions which are near areas of increased industrial activity or increased population density compared with the centre of the North Sea. However, model performance varied. Although some performed reasonably well, others were poor in their explanatory power. This is for several reasons: 1) This study did not account for temporal variations in contaminants, both seasonally and annually. Future models could include this; 2) Extreme values were omitted because their inclusion resulted in unrealistic relationships and predictions. However, these are actual measurements and so do pose risk. Future models could investigate the means to better predict extremities in pollutant levels; 3) This study used coarse-scale environmental variables. Local-scale processes may be of greater importance. Nevertheless, predictions did make general sense, and highlighted areas and pollutants of concern and so serve as a strong starting point for further conservation measures.

Between 1995 and 2020, Mercury concentrations in sediment in the German Bight have decreased (Kammann et al., 2023). Although Mercury concentrations were only sampled in the sediment at three locations in this study rather than modelled across the region, it does highlight a drawback of this current study, namely that insufficient data were available to make North Sea-wide assessments of temporal variation in pollutant concentrations, the data having to be combined to make any assessments. This study may therefore be missing temporal trends as demonstrated by Mercury concentrations in sediments in the German Bight and, so risk assessments must be interpreted with care. On the other hand, Mercury concentrations were found to have increased in dab, *Limanda limanda* (Kammann et al., 2023).

Figure 18 demonstrates some autocorrelation for PAHs and so this must also be taken into account when assessing the performance of the models. Caution is need in interpretation as models may be overcomplicated. Future studies will need to work on improving this to reduce the effect that data points at close proximity have upon one another.

Conclusions and Future Recommendations

This study set out to determine which regions of European waters, with a focus on the North Sea, pose the greatest risk for harbour porpoise from marine pollutants. The study took a novel approach to assess pollutant levels across greater spatial scales than point sampling can offer, and, for this reason, the study provides the opportunity to better risk assess harbour porpoises across its habitat.

These include the inclusion of additional data, both spatially and temporally, and the inclusion of ocean current directions within the General Linear Models as this might allow a better understanding of how particular pollutants may be found in regions far away from industrial zones (Cappello et al., 2020). Cappello et al. (2020) used a complex form of kriging to determine the temporal profile of surface ocean currents, and a similar approach might be useful going forward. In addition, GIS could be used for calculating distance to multiple estuaries.

A major consideration to be taken into account is that this study only reports predicted concentrations in sediments rather than in the harbour porpoise themselves and so assessments into risk must be interpreted cautiously. This approach is at an early stage and when the approach is refined, and – importantly - additional data points have been collated to make modelling porpoise data feasible over very large areas, i.e., the North Sea, it would be interesting to begin to model additional pollutants to further assess risk for harbour porpoises. Furthermore, the risk maps produced by this study could be applied to other cetacean species and to other marine predators in the North Sea. Now that we have begun to establish a protocol to predict pollutant concentrations, marine top predators in the North Sea and further afield, could be assessed quantitatively for risk of population level impacts from marine pollutants.

The quantitative risk assessment used by this study represents a positive step forward in understanding the risk faced by porpoises depending on exposure to pollutant concentrations. In order to improve it further, the concentrations which induce negative effects but do not result in mortality can be included. This would produce maps which show where, in the North Sea or elsewhere, pollutant concentrations have no impact, where they have negative but sub-lethal effects, and where they result in actual mortality. This would allow for a greater assessment of risk to be performed.

In the meantime, this study has identified coastal regions as being areas of concern due to the proximity to estuaries and industrialised areas. It has predicted that no single coastline around the North Sea is likely to be the sole cause for a harbour porpoise decline. However, the German Bight is identified as a region of particular concern. It is recommended that measures are put in place to reduce the impact that pollutants may be having on harbour porpoises in this region of the North Sea, and to address cumulative effects from other stressors such as fisheries to halt local population declines that are currently being observed in this region.

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Which regions of European waters exhibit the highest risk for harbour porpoises from marine pollutants?

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