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Temporal Increases in Mercury Concentrations are Associated with Increased Risk of Death by Infectious Disease in Harbour Porpoises (*Phocoena Phocoena*)

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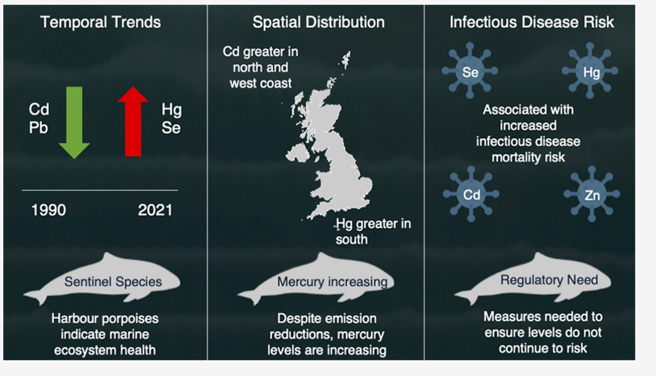

ABSTRACT: Trace elements, particularly heavy metals like mercury, pose significant risks to marine ecosystems due to their toxic and bioaccumulative properties. Concentrations in some marine species show concerning temporal increases, yet spatio-temporal trends in UK marine mammals have not been comprehensively assessed for nearly two decades. Marine mammals serve as sentinel species for ocean health due to their long lifespan and high trophic level, making them vulnerable to bioaccumulative pollutants. Monitoring trace elements, in highly exposed populations, is critical for assessing trends and indices of health, like infectious disease. Using liver tissue samples of 738 UK-stranded harbor porpoises (*Phocoena phocoena*) collected between 1990 and 2021, we found mercury and selenium concentrations have increased, in contrast to declines in cadmium and lead. Spatial analyses revealed that mercury concentrations are highest at lower latitudes, while cadmium increases northward. Levels of zinc, mercury, selenium, and cadmium are significantly associated with infectious disease mortality risk. Our findings highlight the importance of monitoring trace elements in sentinel species to inform conservation efforts and evaluate the effectiveness of pollution mitigation policies such as the Minamata Convention. Increasing mercury levels, despite emission reductions, underscores the urgent need for improved measures to protect marine biodiversity and ecosystem health.

KEYWORDS: *trace elements, marine mammals, disease, mercury, temporal trend, cetaceans*

1. INTRODUCTION

Trace elements, particularly heavy metals like mercury (Hg), pose a significant threat to marine ecosystems.[1] Apex predators such as marine mammals are particularly susceptible to elements that biomagnify and bioaccumulate because they are long-lived and feed at a high trophic level.[2] Trace elements are naturally occurring and have a wide range of toxicities and deleterious effects. While some, such as iron (Fe), cobalt (Co), copper (Cu), and zinc (Zn), are essential nutrients, they can become toxic in high concentrations. Others, like Hg, cadmium(Cd), and lead (Pb), are highly toxic even at low concentrations.[3] Although these metals are naturally occurring, anthropogenic activities (including the increasing production of electronic waste) have significantly elevated their environmental concentrations above baseline levels, resulting in serious ecological and health risks.[4]

Recent studies on trace element concentrations in freshwater and marine environments have revealed contrasting temporal trends. In UK rivers, there is evidence that metal concentrations have decreased over the past 30 years.[5] However, in the marine environment, trends vary between species, locations and trace elements with a high percentage of sites assessed by the Centre for Environment, Fisheries and Aquaculture Science (Cefas) exceeding environmental thresholds and upward trends observed in Pb, Cd and Zn in offshore

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fish.[6] Globally, similar variability exists, with regional and species-specific differences highlighting the need for more comprehensive monitoring.[7−11]

Marine mammals are an excellent group to study the impacts of pollutants as they are considered important sentinels of ocean health.[12] They are particularly vulnerable to heavy metal toxicity, especially from Hg, which is the primary biomagnifiable metal, due to their high trophic position and long lifespans, which facilitate the bioaccumulation and biomagnification of toxic elements.[12] Used alongside other species (e.g., fish or invertebrates), they offer complementary insights into long-term exposure and may serve as more relevant indicators for assessing risks to ecosystem and human health. Toothed whales are of particular concern as they are one of the most contaminated groups of marine mammals and Hg has been linked to several deleterious effects (e.g., immunotoxicity, neurotoxicity).[13] The majority of research into trace elements in marine mammals has primarily focused on mercury due to its high toxicity. Marine mammals are mainly exposed to mercury through their diet which they absorb as methylmercury (MeHg), a highly toxic form produced by microbial processes in aquatic systems.[14] Methylmercury accumulates in marine mammal tissues and is associated with serious health issues, including neurotoxicity, reproductive issues, liver disease and immunosuppression.[15−17] When assessing risk caused by Hg concentrations, it is crucial to also consider selenium (Se) concentrations as marine mammals have a detoxification mechanism that can mitigate the impact of high dietary Hg exposure, through the formation of nontoxic inert complex Hg−Se complex Tiammanite.[18] To account for this, studies typically determine the Hg:Se molar ratio to evaluate the potential risk of mercury. A ratio greater than 1 indicates a potential risk as it suggests there is excess Hg. While this mechanism can protect against exposure to Hg, Se is an essential nutrient for numerous biological functions therefore, deficiencies (caused by detoxification of Hg) can cause several negative effects, including oxidative damage and inflammation, contributing to the neurotoxic effects of MeHg.[13]

In the UK, the harbor porpoise is used as a sentinel top predator species for monitoring long-term trends in chemical pollutant exposures in the marine environment.[19] Declines have been observed in several pollutants, including brominated diphenyl ethers, Hexabromocyclododecane (HBCD), dichlorodiphenyltrichloroethane (DDT) and dieldrin between 1990 and 2000[18] as well as in some trace elements between 1990 and 2008.[20,21] However, trace element concentrations and trends have not been published in the scientific literature for almost two decades, a period roughly equivalent to three generations of harbor porpoises, whose generation length is estimated to be 5−6 years.[22,23] Moreover, the most recent analysis did not analyze temporal trends or investigate biological and spatial factors related to exposures.[20] Therefore, it is crucial to assess burdens and spatiotemporal trends to determine the current risk of exposure particularly as the harbor porpoise is listed by OSPAR (the Convention for the Protection of the Marine Environment of the North-East Atlantic) as a species under threat or in decline in the Greater North Sea and Celtic Sea, partly due to the harmful effects of pollutants, including mercury.[24,25] Additionally, monitoring concentrations in sentinel species is crucial for assessing marine ecosystem health and the effectiveness of mitigation plans such as the UK government's Environment Plan, which commits to substantially reducing the levels of harmful chemicals entering the environment,[26] and the Minamata Convention, a multilateral treaty that aims to reduce mercury pollution.

Given the reduction in monitoring trace element trends in UK harbor porpoises over recent years, understanding their spatiotemporal trends and investigating their associations with health indices such as infectious disease is vital to better understand the threat posed to marine mammals, and take necessary steps to minimize risk. To address this, we evaluated the hepatic concentrations of eight trace elements (Cr, nickel (Ni), Cu, Zn, Cd, Pb, Se and Hg), as well as the molar ratio between Hg and Se (Hg:Se) in harbor porpoises that stranded along the coast of Great Britain, using one of the largest marine mammal strandings data sets available globally, spanning over three decades. The aims of our study were to (i) determine and summarize trace element concentrations in 739 harbor porpoises (ii) investigate spatiotemporal trends in trace element concentrations and (iii) determine trends in causes of mortality and associations between trace element concentrations and infectious disease mortality.

2. METHODS

2.1. Sampling. Harbour porpoise necropsies (n = 2589) were carried out between 1990 and 2021, by the Cetacean Strandings Investigation Programme (CSIP) in England and Wales and by the Scottish Marine Animal Strandings Scheme (SMASS), according to standard necropsy protocols for marine mammals.[27,28] Trace element analyses of liver samples were carried out on a subset of these individuals (n = 738). Carcasses were prioritised for toxicological analysis according to their state of decomposition using a standardized classification system for marine mammals.[28] Carcasses were prioritised in this way to minimize possible changes in concentrations that may occur with decomposition.[29] Of the carcasses analyzed for trace elements, 90% were classified as extremely fresh or only slightly decomposed. We ensured that the individuals that underwent toxicological analysis were a representative sample of the strandings that occurred over the study period by testing for statistical differences in the proportions of sex-maturity classes (neonates, juveniles, adult females and adult males) between the toxicological data set and the complete strandings data set (chi-squared age and sex class $X^2 = 7.2116$, $p > 0.05$).

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 sex-maturity classes. Sexual maturity in males was assessed using gross gonadal size and appearance and, in a representative subset, histological evidence of spermiogenesis in male testes.[30] Female reproductive maturity was determined by identification of one or more ovarian corpora (*lutea or albicantia*).[30,31] Age class classification was based on body length and examination of gonadal tissues as follows: neonates (body length ≤ 90 cm); juveniles (sexually immature, body length >90 cm) and adults (sexually mature, body length >90 cm).[32]

2.2. Disease Diagnosis and Classification. Each examination had a supervising veterinarian who determined the cause of death by gross examination followed by ancillary testing (including parasitology, bacteriology, histology, virology and histology) as required. "Infectious disease" is a broad category used for analyses within the UK strandings program, consisting of a number of cause of death categories of infectious origin.[33] Within this broad category, we have provided further details on classification and diagnosis for the categories (SI Section 1.1) that contained the highest number of cases.

2.3. Trace Element Analyses. To conduct this analysis, we have combined data from previous studies ($n = 492$)[20] with newly generated, directly comparable unpublished data, all produced in the same laboratory using identical methods ($n = 247$). For each individual, cross-sectional samples of liver were collected, placed in a plastic zip lock bag, and preserved at −20 °C. Concentrations of Hg, Cr, Ni, Cu, Zn, Cd, Pb and Se were determined (on a mg/kg wet weight (ww) basis), by inductively coupled plasma-mass spectrometry (ICP-MS) at the Cefas laboratory (Lowestoft) following methods described in detail by ref 34. The method followed recommendations of the International Council for the Exploration of the Sea (ICES) and have been validated under the QUASIMEME laboratory proficiency scheme (further detail is provided in SI Section 1.2). In cases where concentrations were below the limit of quantification, the concentrations were set at the limit divided by the square root of 2.[35]

2.4. Statistical Analyses. All statistical analyses were carried out using the statistical computer program R (version 4.0).[36] To address the various aims of the study, we used multiple data sets, each with different sample sizes and variables (Figure S1).

To investigate differences due to body condition we used a basic index of weight-to-length ratio, which is considered an appropriate metric of body condition and is widely acknowledged as a good predictor of fitness in marine mammals.[37,38] To derive this metric we removed animals which had missing weight or length data which reduced the sample size to 700 for analysis including this metric. The weight and length data 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 modeling whereby values above the model fit represented cases in good nutrition and individuals below the line represented cases in poor nutritional condition. This method may slightly overestimate condition in pregnant females; however, based on published pregnancy rates of UK stranded harbor porpoises (29%; Murphy et al. 2015), we estimate that only 5% of individuals in our sample were pregnant, and thus unlikely to have significantly biased our findings. To investigate spatial variation, the latitudes and longitudes of the stranding locations of each animal were collected. A pilot assessment of pollutants in marine mammals was carried out in 2022 at the OSPAR region level;[39] therefore, the contaminant assessment areas defined by OSPAR to investigate pollutant trends in fish and shellfish were used for this analysis (Figure S2).[19,40] The OSPAR contaminant areas assessed were the Irish & Scottish West Coast, the Irish Sea, the Celtic Sea, the Channel, the Southern North Sea, and the Northern North Sea. Summary information containing mean body weight, length and sample size for each sex-maturity class and OSPAR area are included in Table S2.

2.4.1. Spatiotemporal Trends. To investigate spatiotemporal trends of the trace element concentrations, we fitted linear regression models to selected variables that could explain the variability in the data. For each model, the log-transformed concentration was the response variable. The predictor variables included in the full models were selected according to the biological rational that they could influence concentrations. These were, date of stranding, sex-maturity class, latitude, longitude, relative body weight, mean blubber thickness and body weight. As described previously all possible variable combinations were tested to obtain candidate models. The final predictions were then obtained by averaging the set of plausible models ($\Delta AIC < 2$) from the candidate models. Separate analyses were carried out to examine trends in OSPAR contaminants assessment areas whereby the latitude and longitude variables were replaced with the OSPAR area. For post hoc comparisons between OSPAR areas, we carried out a general linear hypothesis test using the *multicomp* package.[41] Posthoc pairwise comparisons of group means were then carried out using the Tukey's Honest Significant Difference (HSD) method to adjust for multiple comparisons. This method was chosen due to its flexibility in handling multiple comparisons across different group means, while controlling for Type I error. We only assessed risk against published hepatic thresholds for Hg as we were unable to find established thresholds for other trace elements. The thresholds applied were those defined by Dietz et al. (2022), which assign risk categories as follows: no risk (<16 mg/kg ww), low risk (16−64 mg/kg ww), moderate risk (64−83 mg/kg ww), high risk (83−123 mg/kg ww), and severe risk (>123 mg/kg ww). We also considered the threshold of 61.1 mg/kg ww from Rawson et al. (1993), which relates to toxic effects of mercury in the liver. While other trace elements are undoubtedly important, their inclusion, in this part of the analysis, was beyond the scope of this study due to the lack of comparable toxicity thresholds. We also investigated Hg concentrations in relation to a proposed framework to assess Good Environmental Status (GES) under Descriptor 8 of the Marine Strategy Framework Directive (MSFD). This includes an absolute assessment against environmental thresholds and a relative assessment in relation to time trends.[42]

2.4.2. Temporal Trends in Causes of Mortality. To determine temporal trends in mortality, we first tested whether there was a significant trend in the overall number of reported strandings. We fitted a generalized additive model using the *mgcv* package with the number of strandings per year as the response variable and the year as a smooth predictor.[43] The model assumed a negative binomial distribution due to overdispersion in the count data. This analysis was carried out on the wider strandings data set ($n = 7171$), which does not include cause of death as post-mortem examinations are only carried out on a minority of stranded animals. We then tested whether there was a significant temporal trend in the causes of mortality using the data set of animals that had undergone necropsies. To standardize the data and limit the impact of potential biases associated with strandings data (including recording effort and carcase drift), counts were converted to proportions. Cause of death was categorized into three classes: infectious disease, trauma (e.g., bycaught, ship-strike, bottlenose dolphin attack) and others (the latter including starvation and live stranding) (see SI 2.2). To assess changes in the proportions of the causes of mortality, yearly frequencies were modeled against year of stranding, cause of death class, sex maturity class by fitting a generalized negative binomial model. We tested all possible variable combinations (Year × cause of death × sex-maturity class) to obtain several candidate models which were ranked according to their AIC (Akaike's Information Criterion) values. Our final prediction model was obtained by averaging the set of plausible models

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Table 1. Mean Concentration, % Change Per Year from Regression Models, Significant of Temporal Trend and Factors Included in Regression Models for Each Trace Element[a]

trace element	mean concentration (mg/kg ww)	change per year %	95% confidence interval	significant trend (Yes/No)	factors included in model (bold indicates $p < 0.05$)
Cd	0.26	−1.34	−2.10 to −0.60	Yes	**Sex_Maturity Class, Latitude, Relative Body wt., Lat*Long, Date Found, Mean Blubber thickness, Longitude**
Cr	0.25	−9.84	−10.55 to −9.14	Yes	**Latitude, Date Found**, Relative Body wt., Longitude, Lat*Long
Cu	14.73	+0.097	−0.32 to +0.52	No	**Sex_Maturity Class, Mean Blubber thickness, Relative Body wt.**, Date Found,Longitude, Latitude
Hg	20	+1.01	+0.34 to +1.68	Yes	**Sex_Maturity Class, Latitude, Relative Body wt., Mean Blubber thickness, Date Found**, Longitude, Lat*Long
Ni	0.15	−7.62	−8.39 to −6.85	Yes	**Date Found**, Longitude, Latitude, Lat*Long, Relative Body wt.
Pb	0.06	−5.54	−6.26 to −4.81	Yes	**Longitude, Sex_Maturity Class, Date Found, Lat*Long**, Latitude, Relative Body wt.
Se	10.44	+1.56	+0.95 to +2.18	Yes	**Sex_Maturity Class, Relative Body wt., Latitude, Mean Blubber thickness, Date Found**, Longitude
Zn	59.68	0.0095	−0.16 to +0.15	No	**Sex_Maturity Class, Mean Blubber thickness, Relative Body wt.**, Latitude, Longitude, Lat*Long, Date Found
Hg:Se mratio	0.66	−0.0045	−0.23 to +0.22	No	**Sex_Maturity Class, Latitude, Longitude, Relative Body wt.**, Lat*Long, Date Found

[a]Mean, Maximum, Minimum values and sample sizes for each variable measured, split by sex-maturity class, are shown in Table S3.

with $\Delta < 2$.[44,45] To validate the model, we plotted the residuals of the model against other variables to ensure there were no systematic relationships and assessed residual variance for patterns indicative of heteroscedasticity. Geographic variation was assessed in the same way with OSPAR area included as an additional variable.

2.4.3. Associations with Infectious Disease. To investigate possible associations between trace elements and infectious disease mortality we used an established case-controlled approach to compare animals that died of infectious disease (cases) with animals that died from trauma (controls).[21,46] We removed animals that died from "Other" causes of death such as starvation and live stranding, this led to a sample size of 584.

We carried out principal component analysis (PCA) to reduce the complexity of the data set by transforming the trace elements into a small number of principal components (PCs). These PCs capture the most significant variations in the data set and allow patterns and relationships with outcomes, such as infectious disease, to be identified. To carry out the analysis, each of the trace element concentrations were log transformed, zero centered and scaled to have unit variance. We fitted a logistic regression model with cause of death as the dependent variable and sex-maturity class, latitude, longitude and date of stranding and the PCs as the predictors. To determine which PCs should be included in the model, we carried out cross-validation and compared AICs of the models that included PC1 up to the total number of PCs that accounted for >80% variance. Once we had selected the most appropriate number of PCs to include, we carried out the same model averaging approach described previously.

To further investigate the relationships between trace elements and infectious disease, we fitted a weighted quantile sum (WQS) regression model. Using the *gWQS* package, we estimated a body burden index (the WQS index) to summarize the overall exposure to the mixture of trace elements.[47] WQS regression has been specifically designed to address the challenges that can arise in mixture analysis, particularly when exposures are highly correlated and there is a need to identify which components of a mixture contribute most to the outcome. The coefficients were constrained to be positive as we hypothesize that the trace element mixture will increase the risk of infectious disease mortality. We log-transformed the trace elements and grouped them into quartiles. As part of the WQS regression a weighted linear index was calculated, which represented the body burden index of all the trace elements. The WQS was then included in a logistic regression model with cause of death as the dependent variable and WQS, sex-maturity class, latitude, longitude, mean blubber thickness, relative body weight and body weight as predictor variables. Owing to collinearity between relative body weight, mean blubber thickness and body weight we either included relative body weight or mean blubber thickness and body weight based on which combination resulted in the lowest AIC. Due to the detoxification interaction between Hg and Se and their high correlation we fit three separate WQS regression models as it would be challenging to interpret results if Hg and Se were included in the same model. The first model included the Hg:Se molar ratio and excluded Se and Hg, the second model included Hg and excluded Se and the third model included Se and excluded Hg. For each analyses, we tested all possible variable combinations to obtain several candidate models, which were ranked according to their AIC values. The models with the lowest AIC were used in the analysis. The data were randomly split into two data sets (40% training set, 60% validation set). The training set was bootstrapped 1000 times to obtain the weighted index of each chemical and the model parameter estimates. The corresponding weight of each chemical was used to determine the extent to which that chemical contributed to the WQS index.[48] A sectioned threshold parameter (1/number of components) was derived to identify significant chemicals.[49] Further information about this approach is available in the Supporting Information Section 1.3.

3. RESULTS

3.1. Trace Element Concentrations. Zn had the highest mean hepatic concentration (59.7 mg/kg ww) followed by Hg (Table 1). The other elements decreased in mean concentration from Cu > Se > Cd > Cr > Ni > Pb. The maximum concentrations followed a similar pattern, however, the maximum concentration of Cr was lower than Ni (Table S3). The mean Hg:Se molar ratios were below one across all

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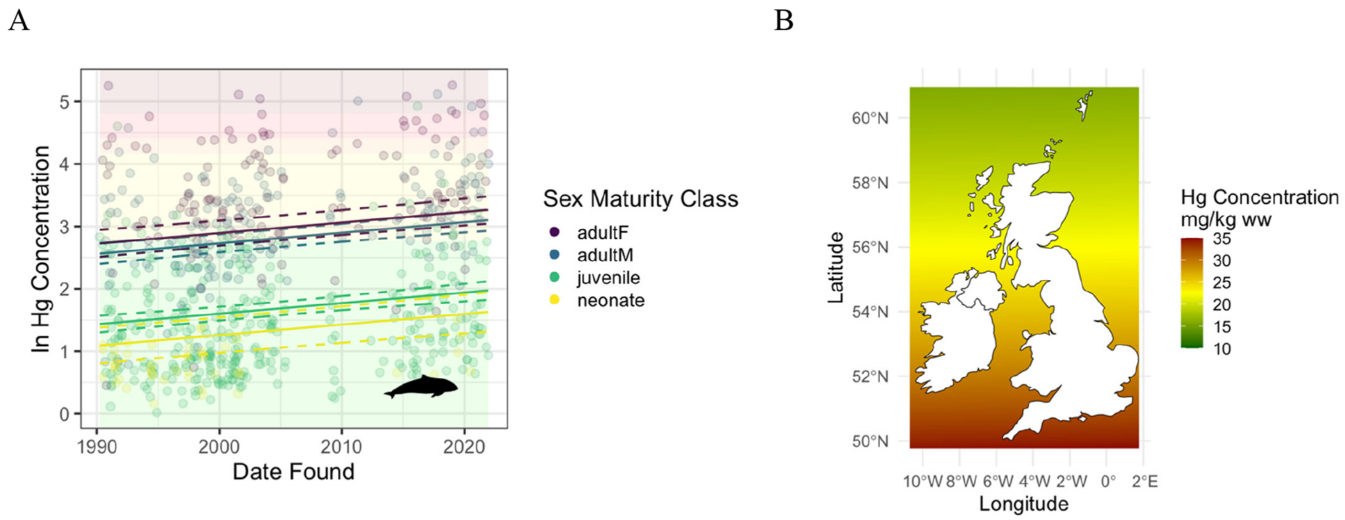

Figure 1. (A) Modeled temporal trend in log Mercury concentrations (mg/kg wet weight). The solid lines represent the model estimates for each year, and the dashed lines represent 95% confidence intervals (1.96 times the standard error). The dots show the measured pollutant concentrations. Trends lines were calculated using the model coefficients with all variables except for Mercury held constant at their mean values. The model coefficients are shown in Table S5. Colors denote risk intervals defined by refs 7,50 . (No risk (green), Low risk (yellow). Moderate risk (orange), High risk (red) and severe risk (dark red)) defined in Table S4. (B) Modeled spatial distribution of Hg concentrations. Estimates were derived from the model using 2021 as the reference year.

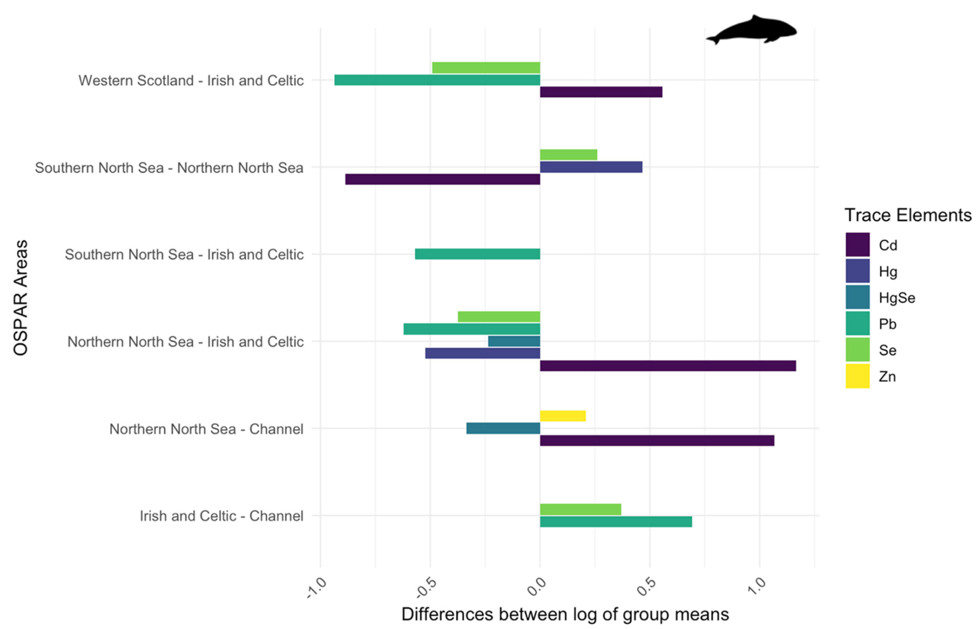

Figure 2. Differences in trace element concentrations (mg/kg ww) between OSPAR areas, based on pairwise comparisons using Tukey's posthoc test ($p < 0.05$). The differences shown represent the log-transformed mean differences between the first and second OSPAR area in each contrast (e.g., "Northern North Sea − Channel" = [Northern North Sea] minus [Channel]). for each trace element, obtained from separate averaged models for each element. The OSPAR area contrasts are in Tables S15−S19.

sex-maturity classes with the highest ratio in adult males (0.96), followed by adult females (0.72), juveniles (0.52) and neonates (0.41) (Table S3). Of the 717 animals for which Hg and Se concentrations were available, 12% (n = 84) had a ratio greater than one.

3.2. Spatiotemporal Trends And Environmental Status Assessments. *Mercury (Hg).* Hg concentrations have increased over the last three decades at a rate of 1 mg/kg ww per year (Tables 1, S5, Figure 1).The mean Hg concentration in the most recent year of the study (2021) was 35.36 (CI = 19.51−51.6) mg/kg ww. In the most recent ten years of the study (2012−2021), 9% (16/188) of individuals had Hg levels classed as high or severe risk (>83 mg/kg ww) while 90% (169/188) had Hg levels classed as low or no risk (<64 mg/kg ww) (according to marine mammal risk thresholds defined by Dietz et al.[7] and derived from harp seals[50]). When we carried out an absolute and relative assessment of Hg concentrations in relation to Good Environmental Status under the MSFD we found that despite mean levels being below the threshold of 61 mg/kg ww.,[17,20] GES was not achieved owing to the increasing concentrations in Hg.

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Sex-maturity class, latitude, body weight and mean blubber were significant predictors in the model. Concentrations were positively associated with body weight but negatively associated with mean blubber thickness. Adult females had the highest concentrations of Hg followed by adult males, juveniles and neonates. There was no interaction between sex-maturity class and the rate of decline. We also found spatial variation such that Hg concentrations were highest at lower latitudes.

When we included OSPAR area rather than latitude and longitude in the model, Hg concentrations were significantly higher in the Southern North Sea and the Irish and Celtic Seas when compared with the Northern North Sea (Figure 2). The full model coefficients are shown in Table S5.

Mercury:Selenium Molar Ratio (Hg:Se). There was no statistically significant temporal trend in the Hg:Se molar ratio (Table S7). However, we did find spatial variation such that the Hg:Se ratio was significantly higher at lower latitudes. We also found significant differences between sex-maturity classes with adult males having significantly higher ratios than adult females, juveniles and neonates. When we included the OSPAR areas in the model, we found the Hg:Se ratio was higher in the Irish and Celtic Seas compared to the Northern North. We also found Hg:Se ratios were greater in the Channel compared to the Irish and Celtic Sea (Figure 2).

Cadmium (Cd). Cd concentrations have significantly decreased over the last three decades (Table S8, Figure S3). Mean blubber and body weight were also significant predictors such that cadmium concentrations were negatively associated with mean blubber but positively associated with body weight. Sex-maturity class was also a significant predictor with highest concentrations in adult males followed by adult females, juveniles and neonates. All differences were significant except for between adult females and juveniles. We also found spatial variation such that (in contrast to Hg) Cd concentrations increased with latitude. Longitude was also a significant predictor with greater concentrations on the west coast than the east coast. When the OSPAR areas were included in the model, we found Cd concentrations were higher in the Northern North Sea than the Irish and Celtic Seas, the Channel and the Southern North Sea. We also found concentrations were higher in Western Scotland than the Irish and Celtic Sea (Figure 2).

Lead (Pb). Similar to Cd, Pb concentrations have decreased over the study period (Table S9). Sex-maturity class was also significant such that adult females have significantly higher concentrations than adult males, juveniles and neonates. Latitude was not a significant predictor, however, we did find significant longitudinal variation with higher concentrations on the west coast compared to the east. This was also demonstrated in the model that included OSPAR areas, as concentrations were higher in the Irish and Celtic Seas when compared to the Northern and Southern North Sea. Concentrations were also significantly higher in the Irish and Celtic Sea than Western Scotland indicating a north to south gradient on the west coast (Figure 2).

Selenium (Se). Similar to Hg, Se concentrations have increased over the study period (Table S10). Body weight and mean blubber thickness were also significant predictors in the model, such that concentrations were positively associated with body weight and negatively associated with blubber thickness. We also found that adult females had significantly higher concentrations than adult males, juveniles and neonates and animals that stranded at lower latitudes had higher concentrations. We also found significant differences between OSPAR areas as concentrations were higher in the Northern North Sea than the Southern North Sea and in the Irish and Celtic Seas compared with Scotland. Concentrations in the Irish and Celtic Sea were also higher than the Northern North Sea and the Channel (Figure 2).

Chromium (Cr). When we modeled Cr concentrations, date of stranding and latitude were the only significant predictors in the model (Table S11). Cr concentrations have steadily decreased over the last 30 years and exhibited a latitudinal gradient such that concentrations were higher at more southerly latitudes. There were no statistically significant differences between OSPAR areas.

Nickel (Ni). Date of stranding was the only significant predictor in the model indicating a steady decline in Ni concentrations between 1990–2021 (Table S12). Similar to Cr, there were no significant differences between the OSPAR areas.

Copper (Cu). There were no significant temporal or spatial trends in Cu concentrations (Table S13). However, sex-maturity class, body weight and mean blubber thickness were significant predictors. Concentrations of Cu were negatively associated with body weight but positively associated with mean blubber (the inverse of Hg and Se). Neonates had significantly higher concentrations of Cu than adult females, adult males and juveniles, which all had similar concentrations. There were no significant differences between OSPAR areas.

Zinc (Zn). There were no significant temporal trends in Zn concentrations (Table S14). Latitude and longitude were not significant predictors, however, when OSPAR area was included in the model we found concentrations were higher in the Channel compared to the Northern North Sea. Body weight and mean blubber were both negatively associated with Zn concentrations. We also found significant differences between sex-maturity classes such that, adult females had the highest Zn concentrations followed by adult males, juveniles and neonates.

3.3. Temporal Trends in Causes of Mortality and Associations between Trace Elements and Infectious Disease. Between 1990 and 2021 there was a significant increase in the overall number of strandings ($n = 7171$) in the UK (edf = 2.95, p-value <0.05) (Figure S4). When we investigated trends in causes of death, using the necropsy data set ($n = 2589$), we found a significant increase (1% per year) in the number of harbor porpoises that died from infectious disease (betareg, $p = < 0.05$) (Table S20), corresponding with a significant decline in deaths from trauma (−1% per year) and no significant change in other causes of death. We were unable to test trends in specific types of infectious disease due to the small sizes in each category. Visual inspection of the absolute number of cases and yearly frequencies in each category indicated that no single category drove the proportional increase in cases (Figure S5). When we incorporated the OSPAR areas into our analysis, we found regional differences in causes of mortality (Table S21). However, there were no temporal trends in these differences. Proportional deaths from infectious disease were highest in the Irish & Celtic Sea followed by the Northern North Sea, the Southern North Sea, Western Scotland and the Channel.

3.3.1. Principal Component Analysis. Eight principal components (PCs) were required to describe the variance in trace element profiles, with the first 5 accounting for over 80%

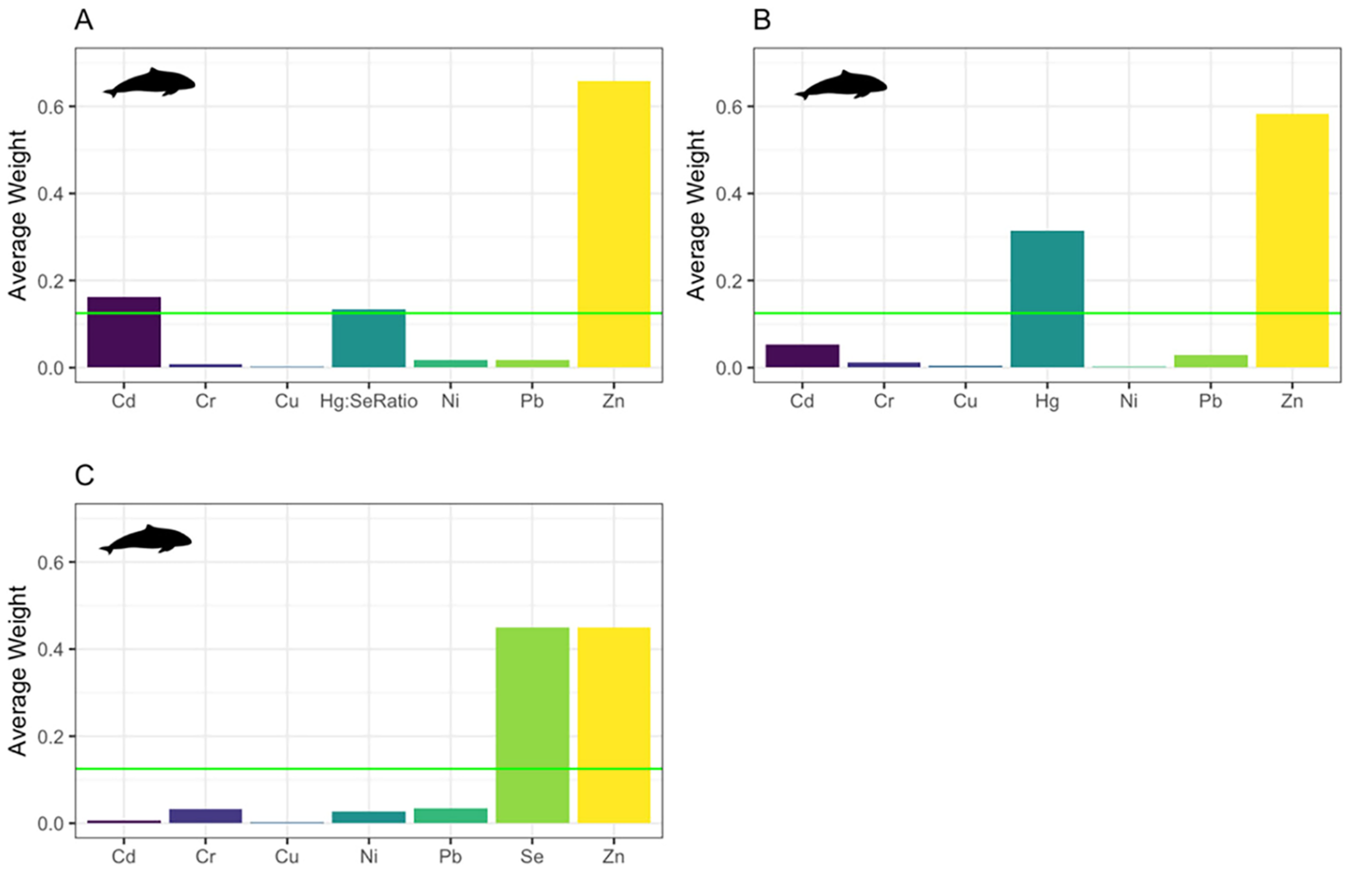

Figure 3. Weights from weighted quantile sum regression for trace element body burden index and risk of infectious disease mortality. The horizontal green line is the threshold for significant components in the index. Cause of death ~ WQS + Sex-maturity + Latitude + Mean Blubber Thickness. (A) Hg:Se molar ratio included, Hg and Se excluded (B) Hg included, Se excluded (C) Se included, Hg excluded.

Table 2. Summary of Weighted Quantile Sum (WQS) Regression Models Assessing the Association between Trace Element Mixtures and the Odds of Infectious Disease Mortality in Harbour Porpoises

WQS model setup	significant trace elements	WQS model coefficient (log-odds of infectious disease mortality)	std. error	factors included in model (bold indicates $p < 0.05$)
Hg:Se ratio included, Hg and Se excluded	Zn, Hg:Se mratio	0.98	0.22	**Relative Body wt., Sex-Maturity Class, WQS,** Latitude, Longitude
Hg included, Se and Hg:Se ratio excluded	Zn, Hg	1.371	2.973	**WQS, Relative Body wt., Sex-Maturity Class,** Latitude, Longitude
Se included, Hg and Hg:Se ratio excluded	Zn, Se, Cd	1.272	0.301	**WQS, Relative Body wt., Sex-Maturity Class, Longitude,** Latitude

of the total variance. The first two PCs accounted for 26.7% and 22.69% of the variance respectively and revealed clustering by cause of death (Figure S7A). Positive scores for Hg, Se, Cd and Zn were observed across both components (Figure S7B). When we modeled cause of death against the PCs, the best fitting model included the first five PCs, however, PC1 and PC3 were not significant predictors. PCs 2 and 5 were negatively associated with infectious disease mortality whereas PC4 was positively associated (Table S25), however, it should be noted that they only accounted for 20%, 10% and 12% of total variance, respectively. PCs 2 and 5 were characterized by positive loadings for Cu and Pb and negative loadings for Zn, Hg and Se, while PC4 was characterized by a large positive loading for Zn (Figure S6).

Sex-maturity class, longitude and relative body weight were also significantly associated with infectious disease mortality risk. Juveniles and neonates have significantly reduced risk than adult females, and animals with higher relative body weight are also less likely to have died from infectious disease. There was no latitudinal association, however, longitude was positively associated with risk of mortality such that animals that stranded along the east coast are more likely to have died from infectious disease than those on the west.

3.3.2. Weighted Quantile Sum Regression (WQS). For all three models (Hg:Se, Hg and Se), WQS score was positively associated with risk of infectious disease mortality (Hg:Se ratio, Hg, Se, ORs = 0.98, 1.27, 1.37,) (Tables 3, S22−24). Zn exceeded the threshold parameter and had the greatest impact on infectious disease mortality risk in all three models. Se, Hg and Cd, Hg:Se ratio also exceeded the threshold parameter in their respective model (Figure 3A) (Table 2). Similar to the results of the PCA, longitude, sex-maturity class and body condition were also significant predictors of infectious disease mortality (Table S25). Further information regarding the interpretation of this approach is available in the Supporting Information.

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4. DISCUSSION

We show that hepatic concentrations of Zn, Hg, Cd and Se are significantly associated with increased rates of infectious disease mortality in UK-stranded harbor porpoises. However, Cd was only significant in the model that included the Hg:Se molar ratio and excluded Hg and Se as individual predictors (Figure 3A). This suggests that Cd's association with infection may be masked when the detoxifying role of Se is not captured via its balance with Hg. It is important to note, that Hg has no physiological role and is only toxic, meaning its relationship with infectious disease is clearer to derive. Our findings, coupled with the temporal increase in Hg concentrations, are concerning. Projections indicate methyl mercury concentrations will increase in some marine mammal populations under future climate scenarios and similar increases have been observed in other marine species both in the UK and worldwide.[7,51−53] This trend, when considered alongside the multitude of threats facing marine mammals (including other immunosuppressive pollutants (e.g., PCBs), bycatch, noise pollution, and prey depletion[21,54]) highlights[21,55] the need to reduce environmental Hg concentrations. Such efforts are not only crucial for the protection of marine mammals but are also critical to ensure the health of the broader marine ecosystem.

This is the first study in almost two decades to assess spatiotemporal trace elements in UK-stranded marine mammals. We found declines in Cd and Pb concentrations over time, while Zn showed no trend. In contrast, Hg and Se levels have risen, with the increasing Hg concentrations being particularly concerning due to Hg's high toxicity and persistence. The Hg:Se ratio plays a critical role in evaluating Hg toxicity owing to the detoxifying effects of Se.[13,18] Although no significant trend was observed in the Hg:Se ratio, the rise in Se levels may reflect a compensatory response to Hg exposure, potentially leading to Se deficiency and associated health impacts. Our findings align with regional and wider assessments that have reported similar increases in Hg in marine biota.[7,10,52,53] However, there are regional and species-specific variations. In UK waters, OSPAR reported increases in levels of Hg in fish and shellfish, while Defra's H4 indicator (which assesses the exposure and effects of chemicals on wildlife) reported stable concentrations in fish.[52,53] Globally, similar regional and species-specific variations have been documented. For example, while Hg levels have increased in some Arctic marine mammals (e.g., polar bears (*Ursus maritimus*) and pilot whales (*Globicephala melas*)), declines have been observed in ringed seals (*Pura hispada*) and belugas (*Delphinapterus leucas*) and a metanalysis of global Hg concentrations in cetaceans spanning 45 years found no overall trend.[7,10,13] A study on harbor porpoises and common dolphins (*Delphinus delphis*) that stranded along the French Atlantic Coast did not find any significant temporal trends in Hg and Pb in harbor porpoises but found increases in Hg and decreases in Pb concentrations in common dolphins. The rates of increase of Hg in our study were almost double those recorded in common dolphins in France.[56] Comparative studies indicate that Hg levels in our study are higher than those reported in West Greenland and Danish waters,[57] similar to those reported in the North Sea and lower than those reported in the Celtic and Irish seas.[56] Concentrations of Cu, Se and Zn were less than half those reported in the Southern North Sea and the Bay of Biscay.[58] Together, these findings highlight the regionally specific nature of contaminant exposure, underscoring the importance of local monitoring to detect emerging pollution trends and assess ecological risk.

Despite the overall reduction in European anthropogenic mercury emissions following reduction measures in the 1970s, Hg concentrations in the open ocean are thought to have tripled globally at depths of 100−1000 m.[59] Therefore, the observed increase in Hg may indicate an increased supply of this accumulated subsurface legacy Hg to subsurface and surface ocean food webs.[8] Additionally, the divergence between trends in emissions and biota could be linked to climate-mediated environmental changes that impact Hg deposition and bioaccumulation.[60,61] Analysis of stable isotopes and Hg concentrations in the Arctic shows that ocean currents transporting legacy Hg from Asia and North America may explain why atmospheric Hg deposition has been decreasing in the region while Hg levels have been increasing in many Arctic species.[11] An important next step would be to speciate mercury into elemental and methyl mercury to better assess the risk[62] and identify sources. Additionally, analysis of stable isotopes would help infer the impact of temporal changes in foraging on pollutant loads. Our findings demonstrate that continuous long-term monitoring of sentinel species is vital to assess the effectiveness of emissions reductions and the necessity of more stringent controls.[56]

Our analysis also revealed significant geographical variations in trace element concentrations. Cd exhibited a latitudinal gradient, increasing from south to north. This likely reflects regional dietary differences, as harbor porpoises in Scotland are thought to consume more oceanic cephalopods and other Cd-enriched species than those in England, owing to their greater abundance in Scottish waters.[63] In contrast, Hg concentrations were higher at lower latitudes (aligning with OSPAR's assessment in fish and shellfish[53]), while Pb concentrations demonstrated a longitudinal gradient, with higher concentrations on the west coast. These spatial gradients may be due to a number of factors including locations of historical industrial activities,[64,65] prevailing westerly winds carrying potential trans-Atlantic sources of lead,[66] greater rainfall causing increased atmospheric deposition,[67] and localized sources such as river discharge. These spatial patterns align with concentration gradients of POPs, such as PCBs, which are likely to have compounding impacts on health.[21,68] Understanding the spatial distribution of toxic trace elements is critical for targeting conservation efforts and emissions monitoring.

While trace elements were linked to infectious disease mortality, their relationship is complex, as several of the trace elements we analyzed (Cu, Se and Zn) are critical for physiological functions and maintaining health, with their levels regulated by homeostatic mechanisms.[69] An imbalance in concentrations, whether through high environmental exposures or detoxification-induced deficiencies, can lead to harmful effects. For example, Zn can be toxic in high concentrations while Se deficiency can lead to numerous negative health outcomes.[69] Zn (unlike Hg) plays an important physiological role and has been shown to be essential for immune system function in mammals.[69] For instance, infectious diseases in humans have been shown to elevate hepatic Zn concentrations, suggesting a complex interaction between Zn and the immune response.[70] Increased Zn concentrations have also previously been associated with disease occurrence in cetaceans, hence, elevated levels may be a response to poorer health status rather than a contributory

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cause.[62,71] Our results reflect this relationship as both the WQS regression and PCA demonstrated a strong association between Zn and infectious disease making it difficult to determine whether elevated Zn concentrations are a consequence rather than a cause of infection. In contrast, the Hg:Se ratio provides clearer insight into toxicity due to Hg's detrimental effects and Se's detoxifying role. Yet, this interaction does somewhat complicate the interpretation of their independent effects, as Se may mitigate Hg toxicity, potentially biasing the WQS regression by down-weighting Hg's influence. We attempted to mitigate this by building separate models so that Hg and Se were not included in the same model. The weights of Hg, Se and Hg:Se ratio demonstrate that the interaction between these variables is the most important determinant in relation to infectious disease. However, experimental data specific to marine mammals are needed to define toxic thresholds for Hg:Se imbalances. Additionally, our analysis revealed a negative association between Cr, Ni, and infectious disease, supporting prior research suggesting these elements may have protective effects in small quantities.[69] Although it should be noted that nearly half of Ni concentrations (48%) and around one-third of Cr and Pb concentrations were below the LOQ, which may limit the reliability of model estimates for these elements. Results for these elements should therefore, be interpreted with caution.

Maturity class and nutritional condition significantly influenced infectious disease risk. Adults had a higher risk of infectious disease, which may be due to adults being exposed to a greater number of pathogens and differences in prey selection, and juveniles and neonates being more vulnerable to other causes of mortality such as bycatch and starvation. Additionally, our model may not fully distinguish between the effects of maturity class and trace element concentrations, as toxic elements like the methyl form of Hg bioaccumulate, leading to higher concentrations in older individuals. Therefore, the elevated risk in adults may partly reflect higher concentrations of Hg rather than age alone. Nutritional condition had the largest effect on infectious disease mortality risk, but nutritional condition and infectious disease are intrinsically linked. Nutritional stress weakens the immune system, while infections can cause nutritional stress. Consequently, the extent to which nutritional stress directly contributes to infectious disease mortality likely varies between individuals, influenced by a combination of environmental and biological factors.

While our analysis used maturity class as a proxy for age, we acknowledge the absence of precise age data (e.g., from tooth layer counts) as a limitation. Harbour porpoises exhibit a wide age range, living up to ∼20 years, with sexual maturity reached at approximately 3−4 years of age.[72] Many trace elements, particularly mercury, are known to bioaccumulate with age. Therefore, maturity class may not fully capture the influence of age-related accumulation, especially among adults where age spans over a decade. The lack of finer-scale age resolution may have introduced additional variability into our models and potentially obscured more nuanced age-related patterns in trace element burdens. Future studies incorporating accurate age estimation would improve the ability to disentangle the effects of age, maturity, and toxicant exposure on health outcomes.

The observed relationship between trace element tissue burdens and infectious disease mortality is particularly concerning in the context of the significant increase in the number of animals dying from infectious disease over time as well as the increase in overall strandings numbers. It is, however, important to consider that an increase in strandings may not be the direct cause of reduced population health but can also be explained by increased reporting effort.[73] Additionally, trends in causes of death, such as the increase in disease cases, may be due to an increasing population, changes in local distribution and/or increased incidence of disease in the population but could also be influenced by biases when selecting individuals to examine.

Our findings make an important contribution to understanding trace element trends and their associations with infectious disease mortality in cetaceans. However, the scope of our study did not include whether risk varies across different pathogens or sublethal infections. Future research should include more in-depth pathological investigations in relation to specific infectious diseases.[74] We also cannot completely rule out that selection bias in the controls may have impacted our findings. While we attempted to select the cases and controls independently of trace element exposure, there is a possibility that animals that died of physical trauma had a higher or lower exposure than the general population. However, we suspect that this is likely to have had a minimal effect as there is no evidence to suggest that animals that die from physical trauma are not representative of the broader population in terms of trace element exposure. Therefore, we consider these individuals an appropriate reference group for comparison. Animal movement and carcass drift may also have affected our results, particularly the spatiotemporal analysis. Very little is known about the home range size of harbor porpoises in the UK. Therefore, large home ranges or the movement of carcasses in ocean currents may cause individuals to accrue contaminants in different locations to where they strand. However, spatial trends of persistent organic pollutants (POPs) in porpoises were found to correspond with historical use and production suggesting that stranded animals can be used to assess spatial contamination. Moreover, tagged porpoises exhibit relatively high site fidelity with seasonally shifting but broadly consistent spatial patterns, which supports our spatial approach.[75] Hence, we are confident that the OSPAR assessment units that we have used should be large enough to minimize the impact of animal movement on our results. Moreover, the effect of carcass drift should be minimized by selecting fresher carcasses because this increases the likelihood that an animal died close to where they were found.[76]

When comparing Hg hepatic concentrations against risk thresholds defined by Dietz et all.,[7] in the most recent ten years of the study, 9% of individuals were classified as at severe or high risk of health effects (>64 mg/kg ww). However, the studies on which these thresholds are based reflect relatively severe pathological effects and are far higher than those required to cause mild liver lesions in humans.[13] Moreover, these thresholds only account for liver-related impacts yet, our study highlighted associations between Hg concentrations and infectious disease. Additionally, a substantial body of work documents Hg's effects on other physiological systems including immune function.[13] Effects threshold concentrations for suppression of lymphocyte proliferation in cetaceans, derived from field and laboratory data, are significantly lower than those for hepatic damage, at 0.047 Hg mg/kg, which suggests a large proportion of the porpoises in our study

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experienced Hg-induced immunosuppression.[77] Further research is necessary to establish robust toxic thresholds for chronic exposures incorporating indirect effects of imbalances in other trace elements and the impacts of cumulative exposure to other pollutants.

This study highlights the enduring risks posed by legacy pollutants, like heavy metals. While global attention often shifts toward novel emerging contaminants, the continued impact of legacy pollutants on marine ecosystems serves as a stark reminder of their toxic legacy. The rise in mercury concentrations, raises concerns about the effectiveness of current measures under the Minamata Convention and highlights the need for strengthened global implementation and monitoring and the potential impacts of climate-mediated environmental change that may lead to increases in environmental concentrations despite reductions in emissions.

Moreover, nearly half of Hg emissions are attributed to the combustion of fossil fuels and biomass (24%) and industrial activities such as smelting and cement production (28%), with around 50% of total emissions originating in Asia.[78] Therefore, the reduction of Hg levels is only likely to be achieved if we can achieve global climate targets. Our findings not only inform the conservation management of marine mammals but offer insights into the health of marine ecosystems globally. Given the pervasive and persistent threat of chemical pollution, our findings are globally significant and demonstrate that more aggressive reduction targets are urgently required to halt the increase in concentrations of mercury,one of the most toxic trace elements.[79]

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.5c08346.

> Methods used for disease diagnosis and classification; trace element analysis and weighted quantile sum workflow and interpretation guide. Supplementary figures: schematic depicting the data sets used for the different parts of the analysis; geographic locations of the stranded animals and the OSPAR area classifications; plots of modeled temporal trends of additional trace elements, strandings per year, yearly causes of death; loadings for the PCA and a plot of PC1 vs PC2. Supplementary tables: percentage of nondetects of each trace element; mean weight, length and sample size for each sex-maturity class by OSPAR area; mean, min, mac values of each trace element split by sex-maturity class; mercury concentrations classified into risk levels; summary statistics of the models fitted to each trace element; summary statistics of the weighted quantile regression models (PDF)

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Notes

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