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Development of an integrated indicator to assess chemical contamination in different marine species: The case of mercury on the French Atlantic continental shelf

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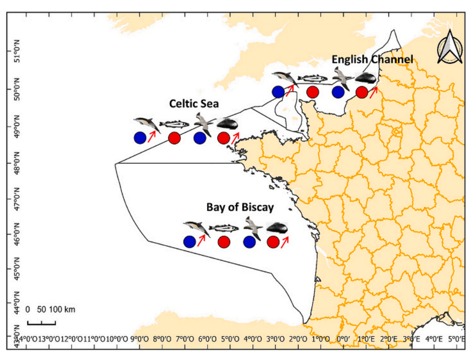
HIGHLIGHTS

- Monitoring different groups of species is needed to observe the Entire Exclusive Economic Zone.
- Hg concentrations in bivalves and fish were higher than environmental thresholds.
- Hg levels increased over time in bivalve and dolphin.
- Hg is at level possibly giving rise to pollution effects in French Atlantic coast.

GRAPHICAL ABSTRACT



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ABSTRACT

Good Environmental Status (GES) for Descriptor 8 (D8) of the Marine Strategy Framework Directive (MSFD) is considered to be achieved when concentrations of contaminants are at levels not giving rise to pollution effects. This study proposes a framework to assess GES in marine waters adjacent to France, including four groups of species (bivalves, fish, birds and mammals) living on the continental shelf and covering different dimensions of the marine environment. This framework is applied to mercury (Hg) in the three marine regions along the French Atlantic coast and includes two assessment types: i) an absolute assessment by comparing contamination levels with environmental thresholds, and ii) a relative assessment by comparing contamination levels over time, performed for bivalves and mammals that had long time-series available. Mercury concentrations were higher than environmental thresholds for bivalves and fish in all the three studied regions. Plus, they significantly increased since the 2000s for most bivalve stations and for the common dolphin *Delphinus delphis*. Our results

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therefore indicate that Hg concentrations have increased in marine waters and have reached levels possibly giving rise to pollution effects in biota from the three marine regions. The present study also highlighted the complementarity of monitoring Hg concentrations in each group of species and each type of assessment, making it possible to propose a conceptual framework for assessing the environmental pressure of bioaccumulated and biomagnified contaminants over the continental shelf.

1. Introduction

Biodiversity and the cleanliness, good health and productivity of oceans and seas are a precious heritage which should be preserved (Laffoley et al., 2020). The Marine Strategy Framework Directive (MSFD) is the main policy protecting the European Union marine waters and primarily aims at achieving a Good Environmental Status (GES) throughout the entire Exclusive Economic Zone, from coastal to offshore waters (EC, 2008). In order to achieve GES, the European Commission structured the MSFD on the basis of 11 qualitative descriptors. Among these 11 descriptors, Descriptor 8 (D8) concerns contaminants. GES for D8 is defined as “Concentrations of contaminants are at levels not giving rise to pollution effects”. Concentrations of both organic and metal contaminants should therefore be monitored in the environment.

Since 1979, the French ‘mussel watch’ long-term monitoring program (named “ROCCH”) provides information on biota contamination from Atlantic coasts, using bivalves as sentinel species. Bivalves were among the first organisms used as bioindicators, *i.e.* considered able to represent the environmental contamination from values measured in these filter-feeding organisms (Claisse, 1989). Due to contaminant bioaccumulation in their tissues, a balance between environmental and bivalve concentrations is observed, and their sedentary lifestyle means that they can indicate the local contamination pressure. In France, bivalve monitoring provides temporal (> 20 years of historical data) and spatial (> 100 monitored stations) information of the coastal environment contamination. However, this information is focused on coastal contamination, and potential chemical pressure on the rest of the continental shelf, which is an important part of the Exclusive Economic Zone to be assessed within the framework of the MSFD, remains to be monitored. Several contaminants such as mercury (Hg) are known to bioaccumulate with age and to biomagnify in food webs (Wang, 2002; Wang et al., 2021), resulting to concentrations higher in older organisms and increase along food webs. Therefore, long-lived and/or high-trophic level species present generally higher levels of contamination than bivalves. As a consequence, complementary to historical coastal bivalve monitoring, recent monitoring programs considering fish, birds and mammals have been implemented in France to assess the chemical contamination in biota living on the continental shelf. Each group of species is assumed to provide complementary information on the environmental contamination due to their specific contaminant exposure and accumulation capacity. Hence, bivalves report on local and coastal contamination at the base of food webs while fish, birds and mammals that are higher trophic level consumers provide contamination information on their specific distribution areas in terms of distance from the coast, depth and geographical extent.

Mercury is a non-essential heavy metal released in the environment from both natural and anthropogenic sources (*e.g.* volcanism and waste incineration) and is recognized as hazardous for organisms. For instance, it is one of the priority substances of the European Water Framework Directive (EC, 2013). Because feeding is the main pathway responsible for Hg transfer in most marine organisms (Hall et al., 1997), Hg concentrations are generally observed to increase with the trophic level of species, and therefore Hg is observed at high concentrations in piscivorous fish species (Cossa et al., 2012), long-lived species or marine top predators such as marine mammals (Aguilar et al., 1999; Caurant et al., 1994; Law, 1996) and seabirds (Ackerman et al., 2016; Monteiro and Furness, 1995). Top-predators are integrative and sensitive to environmental changes and are thus reliable tracers of environmental Hg pressure (Capelli et al., 2000). In addition, the Hg bioaccumulation pattern in marine organisms is a complex process resulting from a combined effect of both abiotic, *e.g.* habitats of species (Chouvelon et al., 2012), and biotic factors, *e.g.* age, sex, trophic functioning of ecosystems and feeding habitat (Chouvelon et al., 2018; Cossa et al., 2012; Cresson et al., 2014), which generates Hg content variability at both inter- and intraspecific levels. These factors should be understood or standardized to promote the use of indicators based on marine organisms' contamination.

This study presents the development of indicators related to Hg contamination in different groups of species including bivalves, fish, birds and mammals, to assess the chemical contamination in the three marine regions defined by the MSFD (*i.e.* English Channel, Celtic sea and Bay of Biscay) along the French Atlantic coast.

2. Material and methods

2.1. Sampling

Mussels (*Mytilus spp.*) were collected and prepared within the French “ROCCH” framework, as described in Claisse, 1989 (Table 1). A pool of at least fifty individuals of a limited size range (35–65 mm in shell length representing 2–3 years of growth) was constituted each year at each monitoring station in November until 2016 and then in February since 2017 (both periods are outside the main reproduction period). Individuals were sampled in natural or aquaculture beds. After collection, mussels were cleaned of epibiota and depurated for 24 h. They were individually measured, shucked from their shell and drained before being pooled into one sample. All samples were then freeze-dried, homogenized and ground for further analysis.

Fish were collected within the French monitoring program of chemical contamination of food webs (“CoRePh”), supported by the MSFD optimisation action of fisheries management surveys (Baudrier et al., 2018). Fish were sampled during the annual bentho-demersal surveys using bottom trawling: CGFS 2018 (Channel Ground Fish Survey) for the English Channel and the eastern Celtic Sea (doi: 10.17600/18000517) and EVHOE 2018 (EValuation Halieutique Ouest de l'Europe) for the western Celtic Sea and the Bay of Biscay (doi: 10.17600/18000518). Seven fish species were selected based on several criteria including: i) ubiquitous species, allowing a comparison between marine regions (*e.g.* European hake *Merluccius merluccius*, Atlantic mackerel *Scomber scombrus*, spotted dogfish *Scyliorhinus canicula*) and more local species for site specificities (*e.g.* European plaice *Pleuronectes platessa*, whiting *Merlangius merlangus*); ii) species with high biomass and abundance, allowing a good representativeness within each marine region; iii) species with commercial interest; iv) species belonging to different trophic levels, allowing a good representativeness of the food web, including demersal and benthic species with intermediate to high trophic levels and small pelagic species with lower trophic levels (Table 1). Individuals were caught within a limited range of fish size and during the non-breeding period in order to limit the effects of fish age and/or length and reproduction on Hg concentration variations (Supplementary Material, Table S2). For the European anchovy *Engraulis encrasicolus* (small fish *i.e.* 10–17 cm), individuals were grouped into pools to ensure sufficient matter for contaminant analysis. Individuals were then frozen at −20 °C on board. Back in the laboratory, individuals were defrosted and filets (muscle tissue) were dissected out. Muscle was chosen for fish to inform MSFD descriptors for both environmental and seafood contamination. All samples were then freeze-dried, homogenized and ground for further analysis.

Six species of birds were monitored and chosen according to their diet, feeding areas, geographical distribution and conservation status in France. Three categories were defined: coastal piscivores, coastal opportunists and offshore piscivores (Table 1). Chicks (prior to fledging) were captured by hand in colonies. Blood (1.5 to 3 ml) was taken either from the brachial or tarsal vein, depending on the species. Ventral feathers were also sampled, but the present study focuses only on Hg results in blood, as Hg concentrations in chick feathers and blood are tightly correlated (Binkowski et al., 2021, data not shown). The sampling period was between March and October (depending on the species), and three fieldwork campaigns were undertaken (2019, 2020 and 2021). Bird captures were done in coordination with local partners managing the protected areas where the birds were captured, mostly during fieldwork already planned on their sites (counting, ringing) in order to minimize disturbance of the colonies.

Mammal samples came from individuals stranded between 2001 and 2019 on the French Atlantic coasts and collected by the French national stranding network "RNE". The common dolphin *Delphinus delphis* and the harbour porpoise *Phocoena phocoena* were selected for the chemical contamination monitoring program for their good representativeness of mammal species (*i.e.* common species frequently observed and stranded), their high stranding rate in the Atlantic (Meheust et al., 2021; Peltier et al., 2016, 2021) and their different feeding habitats and thus prey sources. Common dolphins can be found in both coastal and offshore waters to >1000-m depth, however, stranding carcasses are most likely to belong to individuals living at the continental shelf. Porpoises frequently visit shallow bays, estuaries and tidal channels with <200-m depth and the majority of sightings occur within 10 km of land (Savouré-Soubelet et al., 2016). Both mammal species mainly consumed small schooling fish (*e.g.* European sardine *Sardina pilchardus*, scads *Trachurus spp.*) but porpoises forage more on species living close to the seafloor (*e.g.* poor cod or whiting) (Santos et al., 2013; Santos and Pierce, 2003; Spitz et al., 2006). Each animal was examined by experienced members of the RNE. Then, animals were identified to species, measured and sexed by external observation of genital cavities. After dissection, a piece of the left lobe of the liver as well as a piece of the muscle near the dorsal fin were collected from all the individuals and stored at −20 °C before being freeze-dried, homogenized and ground to powder for analyses. Liver was selected for Hg analyses as being the main storage tissue for this chemical element and muscle for carbon and nitrogen stable isotopes analyses. Finally, teeth and gonads were also collected for age and sexual maturity determination.

2.2. Mercury analyses

Mercury concentrations were measured in the flesh (the whole soft tissues) for bivalves, the muscle for fish, the blood for birds and the liver for mammals. Methodologies for Hg analysis are detailed in Lebigre et al. (2022) for bivalves, Mauffret et al. (2023) for fish, Binkowski et al. (2021) for birds and Méndez-Fernandez et al. (2022) for mammals. A conversion of Hg concentrations to wet weight (w.w.) was achieved with an individual conversion factor obtained by weighing the mass difference before and after freeze-drying for fish samples. For birds, a wet mass percent of 79 % (Eagles-Smith et al., 2008) was used for conversion from dry weight (d.w.) to w.w. For bivalves and mammals, an average of dry mass percent was also used as a conversion factor and equaled 16.3

Table 1
Sampling and Hg concentrations (mean and 95 % upper one-sided interval confidence (IC) limit) in the three French Atlantic marine regions (EC: English Channel, CS: Celtic Sea and BoB: Bay of Biscay).

Group of species	Tissue	Species	Diet/feeding habitat	[Hg]: Mean/IC mg kg^{-1} w.w.[a] Number of samples[b]		
				EC	CS	BoB
Bivalves	Flesh Pool of ≥50 individuals	Mussel *Mytilus* spp	Suspension feeder	0.017/0.020 22 (22 t)	0.014/0.017 7 (5 t)	0.018/0.022 14 (9 t)
Fish	Muscle Individual sample or pool for anchovy	Atlantic mackerel *Scomber scombrus*	Mainly pelagic piscivore	0.051/0.059 15	0.022/0.028 20	0.078/0.085 25
		European anchovy *Engraulis encrasicolus*	Pelagic planktivore			0.033/0.045 14
		European hake *Merluccius merluccius*	Demersal piscivore		0.019/0.022 10	0.040/0.044 25
		European plaice *Pleuronectes platessa*	Benthic invertebrate feeder	0.085/0.121 8		
		Poor cod *Trisopterus minutus*	Planktivorous			0.064/0.080 13
		Spotted dogfish *Scyliorhinus canicula*	Demersal invertebrate feeder	0.568/0.677 17	0.251/0.316 16	0.335/0.396 23
		Whiting *Merlangius merlangus*	Demersal piscivore	0.099/0.118 12		
Birds	Blood Individual sample	European shag *Phalacrocorax aristotelis*	Coastal benthic piscivore	0.212/0.223 17 (1 c)	0.194/0.222 44 (3 c)	0.187/0.210 58 (4 c)
		Great black-backed gull *Larus marinus*	Coastal opportunist	0.388/0.453 33 (2 c)	0.551/0.686 12 (1 c)	0.284/0.332 59 (4 c)
		Lesser black-backed gull *Larus fuscus*	Coastal opportunist	0.095/0.124 23 (2 c)	0.088/0.118 14 (1 c)	0.137/0.152 30 (2 c)
		European herring gull *Larus argentatus*	Coastal opportunist	0.073/0.094 57 (4 c)	0.029/0.038 45 (3 c)	0.071/0.077 102 (7 c)
		Yellow-legged gull *Larus michahellis*	Coastal opportunist			0.147/0.163 8 (1 c)
		Northern gannet *Morus bassanus*	Offshore piscivore		0.281/0.318 15 (1 c)	
Mammals	Liver Individual sample	Common dolphin *Delphinus delphis*	Pelagic consumer	10.5/11.4 259 (19 y)		
		Harbour porpoise *Phocoena phocoena*	Demersal consumer	12.4/15.4 68 (16 y)	8.8/11.7 56 (16 y)	

[a] Mean and upper one-sided interval confidence (IC) limits are calculated from log concentration of Hg.
[b] In brackets: number of temporal trend analyses for bivalves (t), number of colonies for birds (c), number of years monitored for mammals (y).

% (OSPAR, 2022a) and 28.0 % (calculated from historical mammal data), respectively.

2.3. Indicator development: assessment types, statistical tool and assessment parameters

The contamination assessment of each group of species with regard to the Hg contamination was conducted by applying two combined assessment types (Table 2). The OSPAR convention in its hazardous substance assessment presented this distinction between absolute and relative assessment (https://dome.ices.dk/OHAT/trDocuments/2022/help_methods_biota_contaminants.html). An absolute assessment (comparison to environmental thresholds) was performed for the four groups of species, and a relative (temporal trend) assessment was conducted for bivalves and mammals as they have at least 5 years of data. Each group of species was assessed at a specific geographical assessment unit in accordance with their lifestyle, mobility and thus their Hg exposure. The geographical assessment scale was the station for the bivalves, the marine region for fish and the colony for birds. For mammals, ICES and OSPAR have defined five assessment units for European Atlantic waters for the harbour porpoise, including two for French waters (English Channel and Celtic sea & Bay of Biscay, IAMMWG, 2015; ICES (2013, 2014). For the common dolphin, a single geographical assessment unit is currently recognized in the European Atlantic waters (ICES, 2013).

Each assessment (absolute and relative) was carried out in three steps, and led to an assessment parameter (Table 2). The absolute assessment depended on the number of years of data:

- 1–2 years monitoring (fish and birds): an absolute assessment was realized by comparison to an environmental threshold. Step I, a contamination index for each species (C_{sp} for fish and $C_{colony*sp}$ for birds) in each assessment unit was defined as the upper one-side 95 % confidence interval of the mean value based on log Hg concentrations measured, all years combined. Step II, a contamination ratio (CR_{sp} for fish and $CR_{colony*sp}$ for birds) was calculated, informing on the distance to thresholds and calculated as the ratio between the contamination index and the environmental threshold. The Environmental Quality Standard for secondary poisoning has been chosen as the Hg environmental threshold for fish ($EQS_{sp} = 0.02$ mg kg^{-1} w.w., EC, 2013) and the Low Risk Concentration for birds (LRC = 1 mg kg^{-1} w.w.). A spatial integration was carried out for birds (integration of colonies) by calculating CR_{sp} as an average of $CR_{colony*sp}$. Step III, in order to integrate the different species, a contamination score (CS) inspired from the CHASE integration tool (HELCOM, 2017) was determined as the sum of CR_{sp} divided by the square root of the number of species. To achieve GES, the assessment parameter CS has to be lower than 1.
- $\geq$ 3 years monitoring (bivalves and mammals): Step I, an absolute assessment was carried out in each assessment unit with a contamination index ($C_{station*sp}$ for bivalves and $C_{unit*sp}$ for mammals) defined as the upper one-side 95 % confidence interval of the mean value, based on log Hg concentrations measured in the most recent monitoring year (OSPAR, 2022b). Step II, a contamination ratio ($CR_{station*sp}$ for bivalves and $CR_{unit*sp}$ for mammals) was calculated as the ratio between the contamination index and the environmental threshold. The EQS_{sp} has been chosen as the Hg environmental threshold for bivalves ($EQS_{sp} = 0.02$ mg kg^{-1} w.w., EC, 2013). For mammals, the threshold of 61 mg kg^{-1} w.w. was selected since liver abnormalities were associated with chronic Hg accumulation on bottlenose dolphins *Tursiops truncatus* stranded on the southwest coast of Florida (Rawson et al., 1993). A spatial integration is performed for bivalves (integration of stations) by calculating CR_{sp} as an average of $CR_{station*sp}$. Step III, the contamination score (CS) was determined by the sum of CR_{sp} divided by the square root of the

Table 2
Sequences of the three steps for both assessment types (absolute or relative) performed to develop an assessment parameter for each group of species.

Assessment type Number of years in dataset	Group of species Assessment unit	Step I Contamination index for each species in each assessment unit	Step II Contamination ratio/trend for each species in each assessment unit	Step III Spatial aggregation Contamination ratio or trend for each species in each marine region	Assessment parameter Species integration Contamination ratio/trend for each group of species in each marine region
Absolute assessment 1–2 years	Fish Marine region	C_{sp} = upper one-sided 95 % confidence limit on the mean log concentration including all the monitored years	$CR_{sp} = \frac{C_{sp}}{Threshold}$ EQS = 0.02 mg.kg^{-1} w.w. (2013/39/UE, 2013)	CR_{sp}	$CS_{fish} = \frac{\sum CR_{sp}}{\sqrt{N_{sp}}}$ <1: GES reached ≥1: GES not reached
	Birds Colony	$C_{colony*sp}$ = upper one-sided 95 % confidence limit on the mean log concentration including all the monitored years	$CR_{colony*sp} = \frac{C_{colony*sp}}{Threshold}$ LRC = 1 mg.kg^{-1} w.w. (Ackerman et al., 2016)	CR_{sp} = mean $CR_{colony*sp}$	$CS_{birds} = \frac{\sum CR_{sp}}{\sqrt{N_{sp}}}$ <1: GES reached ≥1: GES not reached
Absolute assessment ≥ 3 years	Bivalves Station	$C_{station*sp}$ = upper one-sided 95 % confidence limit on the mean log concentration in the most recent year monitored	$CR_{station*sp} = \frac{C_{station*sp}}{Threshold}$ EQS = 0.02 mg.kg^{-1} w.w. (2013/39/UE, 2013)	CR_{sp} = mean $CR_{station*sp}$	$CS_{bivalves} = CR_{sp}$ <1: GES reached ≥1: GES not reached
	Mammals ICES and OSPAR assessment unit	$C_{unit*sp}$ = upper one-sided 95 % confidence limit on the mean log concentration in the most recent monitored year	$CR_{unit*sp} = \frac{C_{unit*sp}}{Threshold}$ 61.1 mg.kg^{-1} w.w. (Rawson et al., 1993)	$CR_{sp} = CR_{unit*sp}$	$CS_{mammals} = \frac{\sum CR_{sp}}{\sqrt{N_{sp}}}$ <1: GES reached ≥1: GES not reached
Relative assessment ≥ 5 years	Bivalves Station	Linear model (5–6 years): log[Hg] ~ Year Linear Mixed Model (> 6 years): log[Hg] ~ Year (fixed) + Year (random) + Sample (random) + Analytical (random)	Linear model: Slope (T) and statistical significance (P value)	$N_{station} > 2$: $T_{GES*bivalves} = \underline{T}$	$T_{GES*bivalves}$ non-significant or decreases significantly: GES unknown $T_{GES*bivalves}$ increases significantly: GES not reached
	Mammals ICES and OSPAR assessment unit	Dynamic Linear Model & WAIC selection: log[Hg] ~ Year + Age + Sex + δ^{13}C or δ^{15}N	Linear model: Slope (T) and statistical significance (P value)	Not applicable	$N_{species} \leq 2$: $T_{GES} = T_{max}$ (One-Out-All-Out) $T_{GES*mammals}$ non-significant or decreases significantly: GES unknown $T_{GES*mammals}$ increases significantly: GES not reached

number of species. In this study, *Mytilus spp.* was the only species monitored therefore CS is equal to CR_{sp}. To achieve GES, the assessment parameter CS has to be lower than 1.

The relative assessment, based on Hg concentrations trend over time (T), was performed when at least 5 years of data were available (bivalves and mammals). Step I, a linear trend of log Hg concentrations over time was performed for each species in each assessment unit. For bivalves, a linear model (5–6 years) or a linear mixed model (> 6 years) was applied, according to the OSPAR assessment tool (https://dome.ices.dk/ohat/?assessmentperiod=2022, OSPAR, 2022b). For mammals, a dynamic linear model including several confounding factors (age, sex, carbon and nitrogen stable ratios) known to cause variation in Hg concentrations was applied. Step II, the slope T and the statistical significance of the linear model fitted were determined. Step III, a spatial integration for bivalves was performed calculating T_{GES} as an average of T at the different stations or different regions ($\underline{T}$). For mammals, both species were integrated according to the One-Out-All-Out method with T_{GES} equal to the highest value of T (T_{max}). Therefore, the GES is not achieved when the assessment parameter T_{GES} is positive (Hg concentrations increase) and significantly different to zero (non-parametric Kruskal-Wallis test) or, where the One-Out-All-Out method was applied, if at least one linear trend presents a significant increase of Hg concentrations over time.

2.4. Statistical analyses

To investigate variations in Hg concentrations, an analysis of variance (ANOVA) was performed on Hg concentrations, including the group of species and marine regions as factors. The significance of effects was tested by F-tests. A Box–Cox transformation (Box and Cox, 1964) on Hg concentrations was necessary to achieve normality.

3. Results and discussion

3.1. Four complementary groups of species

The order of increasing Hg concentrations was bivalves < fish = birds < mammals (F = 105.96, P-value <0.001) in the three marine regions, which was consistent with both their trophic position in the food web and their feeding habitats (Fig. 1). Bivalves presented the lowest contamination index (*i.e.* $C_{station*sp}$ for bivalves), varying from 0.01 to 0.13 mg kg^{-1} w.w. in the different marine regions (Supplementary Material, Table S1), which could be explained by their low trophic level compared to the three other groups studied.

The monitored fish species were mainly positioned from low to intermediate trophic levels (from average 2.9 to 4.3, personal communication) and presented intermediate Hg concentrations with C_{sp} ranging from 0.02 to 0.68 mg kg^{-1} w.w. (Supplementary Material, Table S2). Pelagic fish species (*e.g.* European anchovy, Atlantic mackerel)

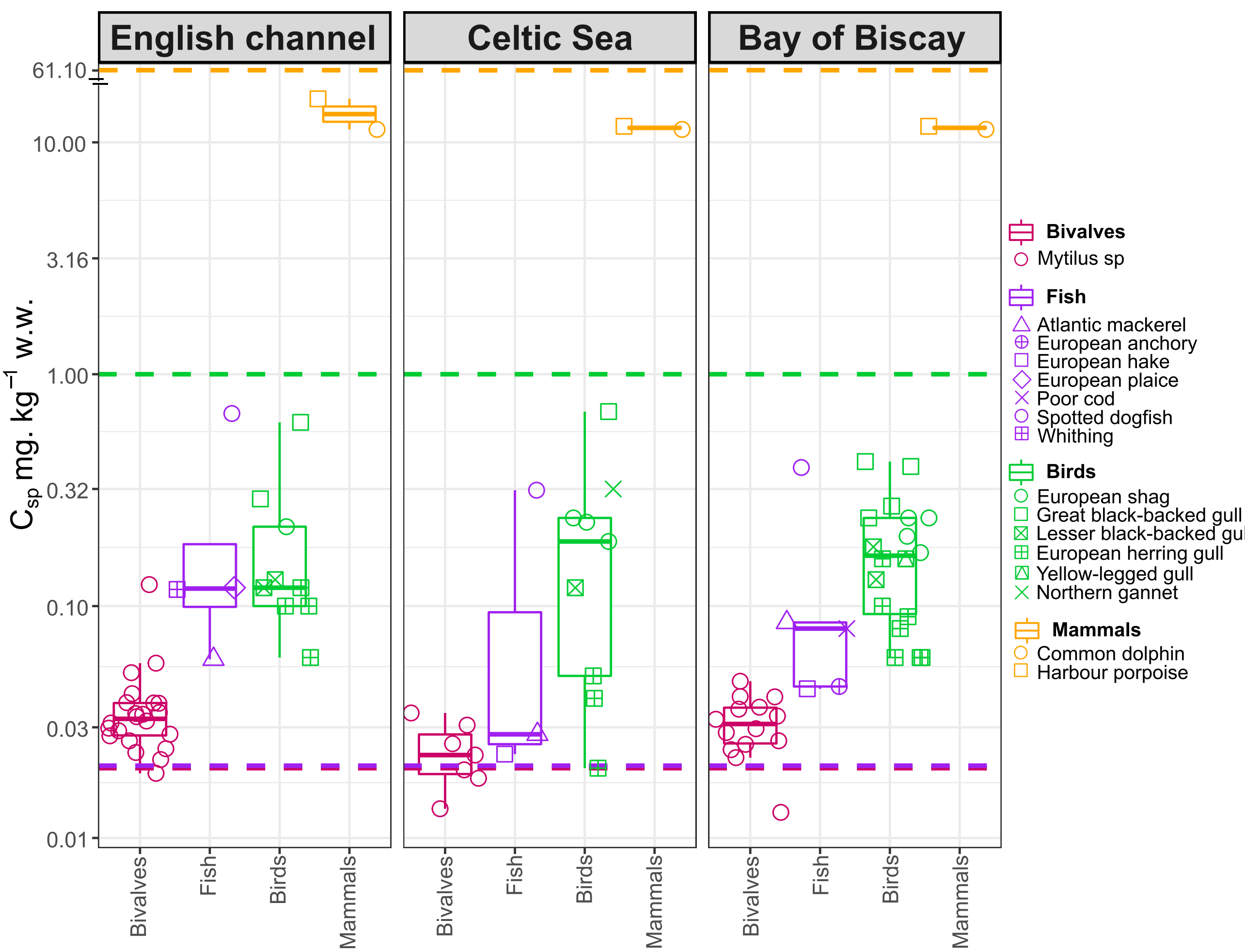

Fig. 1. Concentrations of Hg for four groups of species in the three marine regions. The bottom and top of the box are the first and the third quartiles of the C_{sp} distribution, the horizontal segment is the median, whiskers represent the most extreme data point within 1.5 interquartile range distribution. Horizontal hatched lines represent the threshold values used at 0.02 mg.kg^{-1} w.w. for bivalves and fish (EQS in pink and purple), 1.00 mg.kg^{-1} w.w. for birds (LRC in green) and 61.10 mg.kg^{-1} w.w. for mammals (in yellow).

presented lower Hg concentrations in muscle than the benthic species (*e.g.* European plaice, spotted dogfish), consistently with several studies (Chouvelon et al., 2012; Storelli, 2008).

Concerning birds, their contamination index (*i.e.* $C_{colony*sp}$) varied from 0.06 to 0.69 mg kg^{-1} w.w. (Supplementary Material, Table S3) and the differences observed in Hg contamination among species can be explained by trophic position and foraging ecology (terrestrial *versus* marine resources) of the species. The most contaminated bird was the great black-backed gull, which has the highest trophic level among the monitored species, and feeds mostly on marine resources (Binkowski et al., 2021). On the other hand, the herring gull showed the lowest Hg concentrations, which is consistent with the terrestrial and low trophic level diet of this species. Piscivorous species, such as the Northern gannet and the European shag, were more contaminated by Hg than generalist species, except for the great black-backed gull which has a high trophic level and sometimes forages on other seabird species.

The highest Hg concentrations were observed for marine mammals and varied from 6.81 to 15.43 mg kg^{-1} w.w. (Supplementary Material, Table S4). Despite their different diet (Méndez-Fernandez et al., 2022), Hg concentrations were similar between the two species. In most cases, chemical contaminants reach the marine environment from land-based sources (*i.e.* industrial activities, urban and riverine inputs), this is why monitoring programs were historically implemented in coastal environments. However, some of them are emitted directly (*i.e.* oil and gas exploitation and shipping) or re-mobilized in the marine environment itself (Tornero and Hanke, 2016), which also expose marine offshore species. In addition, some chemical contaminants such as Hg are highly volatile, *i.e.* can be transported long-distance from their source *via* the atmosphere (Cossa et al., 2022), and are known to bioaccumulate in marine organisms and to biomagnify through food webs resulting in higher concentrations for high-level species and top predators, which is confirmed in the present study. Furthermore, despite their offshore lifestyle, fish and mammals presented Hg concentrations measured at levels possibly giving rise to pollution effects.

This study also highlighted differences in Hg variation within groups of species related to their assessment unit. Recently, Rudershausen et al. (2023) also acknowledge that obtaining a comprehensive understanding of Hg trends in marine fauna requires examining multiple species across inshore, nearshore, and offshore environments. Each species is exposed to Hg through its diet/habitat, thus integrating contamination at a specific scale (Fig. 2). Bivalves are exposed to Hg at a very coastal and localized scale (sampling station), allowing observation of contamination differences at a regional scale, which is illustrated by the lowest Hg concentrations observed for samples from the Celtic Sea ($F = 3.89$, P-value $= 0.0287$). At a higher spatial scale, fish and birds presented the highest Hg variations related to differences in diet/habitat of the monitored species. Finally, the lowest variation of Hg concentrations at a regional scale was observed for mammal species. The assessment unit of both studied species included several marine regions, providing information on Hg pressure at a larger geographical scale. Therefore, each group of species provided specific information on environmental Hg pressure (Table 1 & Fig. 2). Bivalves informed on the environmental Hg contamination at local, coastal and shallow scales. Fish and birds informed on contamination at a regional scale (higher mobility), including pelagic and benthic habitats (variation in diet), further offshore and deeper than bivalves. Finally, mammals provided information on contamination at the highest regional, offshore and depth scales (Fig. 2). Hence, a framework based on four groups of species provides a comprehensive view representativeness of Hg exposure for biota living on the continental shelf, considering the three dimensions of marine environments, and thus, an integrated representation of the Entire Exclusive Economic Zone as required by the MSFD. A monitoring program including the four groups of species is worth pursuing in order to track the status and changes of chemical contamination over time and covering different dimensions of the marine environment.

3.2. Absolute assessment

An absolute assessment was performed for the four groups of species (Table 3). For bivalves, the Hg contamination index $C_{station*sp}$ was higher than the environmental threshold (EQS = 0.02 mg kg^{-1} w.w.) for most of the monitored stations, from 0.7 to 6.5 times higher in the three marine regions on the French Atlantic coast (Fig. 1). The assessment parameter $CS_{bivalves}$ was 1.3, 1.6 and 1.9 in the Celtic Sea, the Bay of Biscay and the English Channel, respectively. In the same way for fish, C_{sp} exceeded the EQS from 1.2 to 33.9 times higher for all monitored fish species. Consequently, CS_{fish} values were above the GES thresholds and were higher than those obtained for bivalves, and varied from 10.6 in the Celtic Sea to 24.4 in the English Channel. Accordingly, the GES for fish and bivalves was not achieved and indicated that Hg concentrations in the environment are at levels possibly giving rise to pollution effects on marine life. The EQS defined for Hg in biota based on secondary poisoning was used as the environmental threshold for fish and bivalve assessments. The Water Framework Directive (WFD) defines it as the maximum acceptable concentration of Hg in biota in order to protect both environmental and human health (EC, 2013). The EQS was the lowest no effect concentration (NOEC) among 17 studies reviewed in the EQS dossier for Hg secondary poisoning, it is related to a study on rhesus monkey growth in the WFD dossier for Hg and its compounds (EC, 2005). Its value is lower than the natural background levels for naturally occurring substances derived by OSPAR ($BAC_{fish\ muscle}$: 0.035 mg.kg^{-1} w.w.). As Hg EQS refers to the risk of secondary poisoning by top predators, it should be compared with fish whole-body concentration. In the present study, Hg concentrations in mussel whole body and fish muscle are used. Hg accumulates in tissues rich in proteins containing SH-groups like, *e.g.*, muscle (Eisler, 2007) and Hg concentrations are

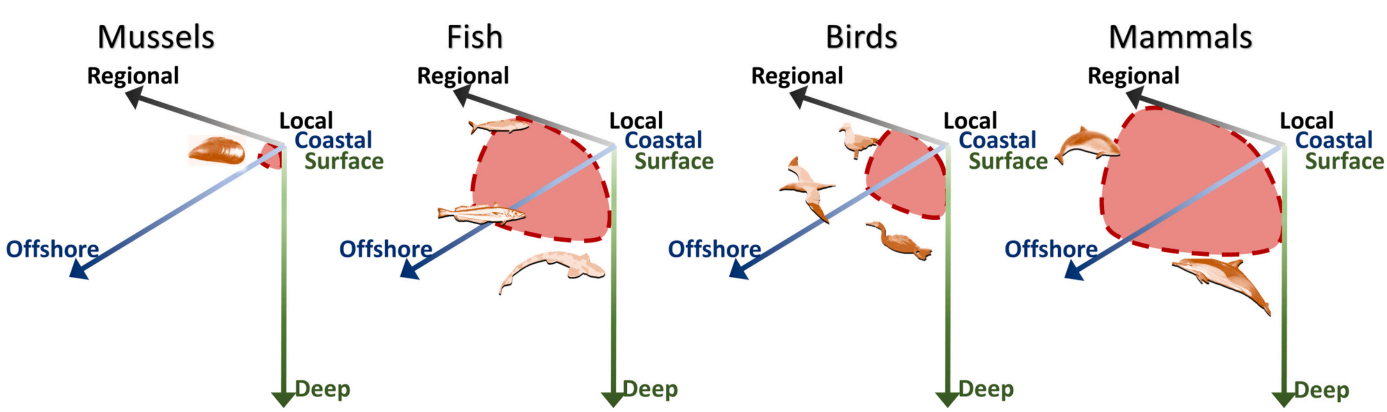

Fig. 2. A multi-matrix approach including four complementary groups of species to assess the environmental Hg contamination (geographic extent in black, distance from the coast in blue and depth in green).

Table 3
Values of assessment parameters (contamination score (CS) and temporal trend (T_{GES}) for absolute and relative assessments, respectively) and Good Environmental Status (GES) (reached in blue and not reached in red) according to the group of species in the three marine regions along the French Atlantic coast.*significant ($p < 0.05$), [NS] non-significant.

Marine region	Group of species	Assessment parameter: Absolute assessment (CS < 1 / CS > 1)	Relative assessment (T_{GES}* and < 0 or T_{GES}^{NS} / T_{GES}* and > 0)
English Channel	Bivalves	1.9	0.008 mg kg^{-1} w.w./year*
	Fish	24.4	-
	Birds	0.8	-
	Mammals	0.3	0.549 mg kg^{-1} w.w./year*
Celtic Sea	Bivalves	1.3	0.008 mg kg^{-1} w.w./year*
	Fish	10.6	-
	Birds	0.6	-
	Mammals	0.3	0.549 mg kg^{-1} w.w./year*
Bay of Biscay	Bivalves	1.6	0.001 mg kg^{-1} w.w./year*
	Fish	14.5	-
	Birds	0.8	-
	Mammals	0.3	0.549 mg kg^{-1} w.w./year*

typically similar or higher by *ca.* 1.4 times in w.w. (as determined in chub, bream, perch) in fish muscle than in whole fish (Fliedner et al., 2018). Consequently, fish muscles would be a suitable tissue for the purpose of conservative assessment relative to the EQS for Hg (OSPAR, 2016). However, this approximation is not possible for all chemicals. For instance, PFOS, another WFD priority substance which also bond to proteins, is typically highest in liver, kidneys and blood and the major fraction of PFOS is found in the carcasses of fish and not in fillets (mean whole fish-to-fillet ratios based on measured PFOS concentrations ranged between 2.5 and 3.1 in w.w., in Fliedner et al., 2018).

For birds and mammals, $C_{colony*sp}$ and $C_{unit*sp}$ were lower than the environmental thresholds (LRC = 1 mg.kg^{-1} w.w. for birds and 61.1 mg.kg^{-1} w.w. for mammals), from 0.1 to 0.7 and from 0.2 to 0.3 lower for birds and mammals, respectively, in the three marine regions (Supplementary Material, Tables S3 and S4). Consequently, the GES was achieved, with both assessment parameters CS_{birds} (ranging from 0.42 in the Bay of Biscay to 0.62 in Celtic Sea) and $CS_{mammals}$ (being equal to 0.3 in the three marine regions). LRC was derived based on extensive field observation reporting that at approximately 1.0 mg.kg^{-1} w.w. in bird blood, bird reproduction was altered (breeding and egg hatchability), enzymatic activity was modified, and behavior impaired in different bird species (Ackerman et al., 2016). The bird LRC has therefore been specifically developed to assess the risk for effects on reproduction and adverse effects on body condition and behavior from Hg exposure in adult birds (Chastel et al., 2022). Thus, the risk may be underestimated when Hg concentrations are measured in chicks, as was the case in the present study. Chicks were selected instead of adults as they provide information on the local contamination reflecting the contamination coming from the food brought by parents to the chicks, as prey are captured in the vicinity of the colony which is very important when working on migratory birds (Albert et al., 2019; Blévin et al., 2013). Hence, working on chicks offers a way to avoid measuring the contamination of adults' wintering sites.

Few studies have determined Hg threshold concentrations for health effects in marine mammals. Assessing the impact of Hg (or other contaminants) exposure and accumulation at the population-level for wildlife species (that are sea dependent) is challenging for obvious logistical reasons. Moreover, laboratory experiments with marine mammal species would require a permission that would not be granted by Animal Care Committees now that they are protected species. However, it was first reported that concentrations around 61 mg kg^{-1} w.w. of total Hg in the liver of marine mammals were damaging to hepatic processes (Law et al., 1992). This commonly used threshold for liver in mammals (Kershaw and Hall, 2019) is based on chronic Hg accumulation that has been associated with liver abnormalities in 9 bottlenose dolphins stranded in Florida by Rawson et al. (1993). They analyzed Hg concentrations in liver samples and examined samples for histologic studies. The results showed a four-fold increase in active liver disease in dolphins associated with liver Hg concentrations above 61 mg kg^{-1} w.w.

The use of thresholds for environmental health assessment appears as a powerful tool. They are widely used under regulatory frameworks but should be used with expert judgments and comprehensive knowledge of their derivation, including i) data types (field, laboratory) used to derive them and to which ones the threshold should be compared with, ii) uncertainty associated with the derived threshold value, iii) threshold protection objectives. The derivation and use of thresholds assume that both toxicity and detoxification pathways are understood. The most important form of Hg in the food chain is methylmercury (MeHg), which is absorbed in the intestine and crosses the blood brain barrier and placenta to exert toxic effects on the central nervous system of adults and foetuses (Alexander and Oskarsson, 2018; Syversen and Kaur, 2012). The MeHg:Hg ratio increases along the food chain and is usually dominated by the total Hg in bivalves and MeHg in fish and top predators, though interspecific differences are observed *e.g.* among bivalves (Pan and Wang, 2011) or top predator fish (Rudershausen et al., 2023). Therefore, a threshold related to MeHg might help reducing the uncertainty around the assessment. Also, selenium (Se) is thought to provide a buffer against the toxicity of MeHg. Zhang et al. (2014) proposed a new criterion for Se/Hg exposure assessment to help reducing the uncertainty around Hg absolute assessment. However, recent studies reported that more information is needed to provide a strong scientific basis for modifying current fish consumption advisories on the basis of Se:Hg ratios, especially on how Se may reduce MeHg toxicity in consumers, how it reduces Hg transfer in aquatic food webs and how Se inhibits Hg bioavailability to, and/or methylation by microbial communities (Gerson et al., 2020; Gochfeld and Burger, 2021). Finally, thresholds are developed to inform on the risk of adverse effects on the health. Protection objectives differ among thresholds *e.g.* for mercury, EQS is meant to protect ecosystem health by protecting the top predators from secondary poisoning, it is based on few laboratory experiments with laboratory species; LRC is meant to protect birds, it is based on rare field studies with birds; maximum permissible concentration in food is meant to protect human health.

Overall, these results obtained with the absolute assessment highlight the fact that, with this type of assessment, thresholds play a prominent role since CR and CS are dependent on this value. Though absolute assessment is (highly) sensitive to the threshold used, it has been widely used as this assessment type is intended to inform on the risk for adverse health effects on organisms or ecosystems.

3.3. Relative assessment

Relative assessment was performed with bivalves and mammals for which up to 20 years of data were available (Table 3). For bivalves, among the 36 temporal trends (one per station) analyzed for the three marine regions, 26 showed a significant increase of Hg concentrations ($p > 0.05$, Supplementary Material, Table S1). For mammals, a significant increase of Hg concentrations was found for the common dolphin within its assessment unit including the three marine regions ($p > 0.05$, Supplementary Material, Fig. S1 and Table S4). The harbour porpoise showed non-significant trends in both assessment units (English Channel and Celtic Sea/Bay of Biscay) ($p < 0.05$, Supplementary Material, Fig. S1 and Table S4). A positive and significant assessment parameter at the regional level illustrates a significant increase of Hg concentration in the three marine regions ($p > 0.05$, Supplementary Material, Table S1 for bivalve and Fig. S1 for mammals).

Overall, Hg concentrations increased on average by up to 0.008 mg kg w.w./year in bivalves and by 0.549 mg kg w.w./year in dolphins over the last 20 years (Table 3). This could reflect the direct Hg increase at a global level (Streets et al., 2019) and/or an indirect higher exposure to Hg due to a change in individual diet under global change, toward prey with higher Hg contents. In both cases, Hg impregnation in these sentinel species increases so that individuals should continuously allocate energy for detoxification to adapt to this pressure, which could result in a lower ability to tackle new stresses.

The advantage to assess the GES for a group of species based on a relative assessment (here temporal trends) is that factors like age or sex of an individual (that have an effect on Hg concentrations) can be considered in a linear trend as done in the models developed for mammals (Méndez-Fernandez et al., 2022). Thus, relative assessment presents the advantage of being temporally integrated, it is not dependent on a threshold value, reveals the chemical contamination changes and improves the prevention of risks of degradation in the environment (water or food web). Therefore, the use of a combined assessment appears as a powerful tool to achieve a more global assessment of Hg pressure on biota.

4. Conclusions

This study proposes an assessment framework based on four groups of species (bivalves, fish, birds and mammals). The goal was to assess chemical contamination pressure of biota for Hg contamination in the three marine regions along the French Atlantic coast. The results showed the variability of Hg concentrations among and within groups of species related to their assessment units and feeding habitats, highlighting the importance of a framework based on four complementary groups of species to assess the environmental Hg pressure on biota living on the continental shelf. Complementary groups of species in terms of Hg exposure have a better representativeness with regard to the food web and allow coastal to offshore environments to be taken into account, as required by MSFD. Bivalves informed on the environmental Hg contamination at local, coastal and shallow scales, while fish and birds provided information on more regional Hg contamination, further offshore, and deeper than bivalves (due to their higher mobility, including pelagic and benthic habitats). Finally, mammals provided information on Hg contamination at the supra-regional and deeper scales. In addition, two assessment types (absolute and relative) were combined to assess Hg pressure and have proved to be complementary. For bivalves, Hg concentrations were higher than the environmental threshold (EQS) in the three marine regions. These results indicate that Hg concentrations were at levels possibly giving rise to pollution effects in coastal and sessile species. This was also observed when investigating more offshore and mobile groups of species, since Hg concentrations measured in fish were also above the environmental threshold. In addition, for bivalves and mammals, a significant increase of Hg concentrations over time (since the 2000s) was observed. An absolute assessment, based on threshold comparison, can inform on adverse contamination effects on biota but should be considered with expert judgment, and a relative assessment offers a reliable indication of change in Hg contamination over time in order to improve the prevention of risks of degradation. The framework developed here would be useful to assess the environmental pressure of other contaminants having the potential to accumulate in biota and biomagnify up food webs such as organic contaminants (*e.g.* polychlorinated biphenyl, polybrominated diphenyl ethers). Chemical contaminant pressure for other marine species living beyond the continental shelf, such as the mesopelagic fauna, could also be considered in an integrated framework.

CRediT authorship contribution statement

Tiphaine Mille: Conceptualization, Formal analysis, Investigation, Methodology, Visualization, Writing – original draft, Writing – review & editing. **Nathalie Wessel:** Conceptualization, Data curation, Methodology, Writing – review & editing, Funding acquisition. **Mélanie Brun:** Conceptualization, Data curation, Methodology, Writing – review & editing. **Paco Bustamante:** Conceptualization, Resources, Methodology, Writing – review & editing, Funding acquisition. **Tiphaine Chouvelon:** Conceptualization, Methodology, Writing – review & editing. **Paula Méndez-Fernandez:** Conceptualization, Resources, Investigation, Methodology, Writing – review & editing. **Gauthier Poiriez:** Conceptualization, Resources, Investigation, Methodology, Writing – review & editing. **Jérôme Spitz:** Conceptualization, Resources, Methodology, Writing – review & editing, Funding acquisition. **Aourell Mauffret:** Conceptualization, Methodology, Supervision, Project administration, Writing – review & editing, Funding acquisition.

Declaration of competing interest

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

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

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References

Ackerman, J.T., Eagles-Smith, C.A., Herzog, M.P., Hartman, C.A., Peterson, S.H., Evers, D.C., Jackson, A.K., Elliott, J.E., Vander Pol, S.S., Bryan, C.E., 2016. Avian mercury exposure and toxicological risk across western North America: a synthesis. Sci. Total Environ. 568, 749–769. https://doi.org/10.1016/j.scitotenv.2016.03.071.

Aguilar, A., Borrell, A., Pastor, T., 1999. Biological factors affecting variability of persistent pollutant levels in cetaceans. J. Cetacean Res. Manage. 83–116 https://doi.org/10.47536/jcrm.v1i1.264.

Albert, C., Renedo, M., Bustamante, P., Fort, J., 2019. Using blood and feathers to investigate large-scale Hg contamination in Arctic seabirds: a review. Environ. Res. 177, 108588 https://doi.org/10.1016/j.envres.2019.108588.

Alexander, J., Oskarsson, A., 2018. Toxic metals. In: Chemical Hazards in Foods of Animal Origin, ECVPH Food Safety Assurance. Wageningen Academic Publishers, pp. 157–180. https://doi.org/10.3920/978-90-8686-877-3_07.

Baudrier, J., Lefebvre, A., Galgani, F., Saraux, C., Doray, M., 2018. Optimising French fisheries surveys for marine strategy framework directive integrated ecosystem monitoring. Mar. Policy 94, 10–19. https://doi.org/10.1016/j.marpol.2018.04.024.

Binkowski, L.J., Fort, J., Brault-Favrou, M., Gallien, F., Le Guillou, G., Chastel, O., Bustamante, P., 2021. Foraging ecology drives mercury contamination in chick gulls from the English Channel. Chemosphere 267, 128622. https://doi.org/10.1016/j.chemosphere.2020.128622.

Blévin, P., Carravieri, A., Jaeger, A., Chastel, O., Bustamante, P., Cherel, Y., 2013. Wide range of mercury contamination in chicks of southern ocean seabirds. PLoS One 8, e54508. https://doi.org/10.1371/journal.pone.0054508.

Box, G.E.P., Cox, D.R., 1964. An analysis of transformations. J. R. Stat. Soc. 26, 211–252.

Capelli, R., Drava, G., De Pellegrini, R., Minganti, V., Poggi, R., 2000. Study of trace elements in organs and tissues of striped dolphins (Stenella coeruleoalba) found dead along the Ligurian coasts (Italy). Adv. Environ. Res. 4, 31–42. https://doi.org/10.1016/S1093-0191(00)00005-8.

Caurant, F., Amiard, J.-C., Amiard-Triquet, C., Sauriau, P.-G., 1994. Ecological and biological factors controlling the concentrations of trace elements (As, Cd, Cu, Hg, Se, Zn) in delpinids <I>*Globicephala melas*</I> from the North Atlantic Ocean. Mar. Ecol. Prog. Ser. 103, 207–219. https://doi.org/10.3354/meps103207.

Chastel, O., Fort, J., Ackerman, J.T., Albert, C., Angelier, F., Basu, N., Blévin, P., Brault-Favrou, M., Bustnes, J.O., Bustamante, P., Danielsen, J., Descamps, S., Dietz, R., Erikstad, K.E., Eulaers, I., Ezhov, A., Fleishman, A.B., Gabrielsen, G.W., Gavrilo, M., Gilchrist, G., Gilg, O., Gíslason, S., Golubova, E., Goutte, A., Grémillet, D., Hallgrimsson, G.T., Hansen, E.S., Hanssen, S.A., Hatch, S., Huffeldt, N.P., Jakubas, D., Jónsson, J.E., Kitaysky, A.S., Kolbeinsson, Y., Krasnov, Y., Letcher, R.J., Linnebjerg, J.F., Mallory, M., Merkel, F.R., Moe, B., Montevecchi, W.J., Mosbech, A., Olsen, B., Orben, R.A., Provencher, J.F., Ragnarsdottir, S.B., Reiertsen, T.K., Rojek, N., Romano, M., Søndergaard, J., Strøm, H., Takahashi, A., Tartu, S., Thórarinsson, T.L., Thiebot, J.-B., Will, A.P., Wilson, S., Wojczulanis-Jakubas, K., Yannic, G., 2022. Mercury contamination and potential health risks to Arctic seabirds and shorebirds. Sci. Total Environ. 844, 156944 https://doi.org/10.1016/j.scitotenv.2022.156944.

Chouvelon, T., Spitz, J., Caurant, F., Mèndez-Fernandez, P., Chappuis, A., Laugier, F., Le Goff, E., Bustamante, P., 2012. Revisiting the use of δ15N in meso-scale studies of marine food webs by considering spatio-temporal variations in stable isotopic signatures – the case of an open ecosystem: the Bay of Biscay (North-East Atlantic). Prog. Oceanogr. 101, 92–105. https://doi.org/10.1016/j.pocean.2012.01.004.

Chouvelon, T., Cresson, P., Bouchoucha, M., Brach-Papa, C., Bustamante, P., Crochet, S., Marco-Miralles, F., Thomas, B., Knoery, J., 2018. Oligotrophy as a major driver of mercury bioaccumulation in medium-to high-trophic level consumers: a marine ecosystem-comparative study. Environ. Pollut. 233, 844–854. https://doi.org/10.1016/j.envpol.2017.11.015.

Claisse, D., 1989. Chemical contamination of French coasts: the results of a ten years mussel watch. Mar. Pollut. Bull. 20, 523–528. https://doi.org/10.1016/0025-326X(89)90141-0.

Cossa, D., Harmelin-Vivien, M., Mellon-Duval, C., Loizeau, V., Averty, B., Crochet, S., Chou, L., Cadiou, J.-F., 2012. Influences of bioavailability, trophic position, and growth on methylmercury in hakes (*Merluccius merluccius*) from northwestern Mediterranean and northeastern Atlantic. Environ. Sci. Technol. 46, 4885–4893. https://doi.org/10.1021/es204269w.

Cossa, D., Knoery, J., Banaru, D., Harmelin, M., Sonke, J., Hedgecock, I., Bravo, A., Rosati, G., Canu, D., Horvat, M., Sprovieri, F., Pirrone, N., Heimbürger-Boavida, L.-E., 2022. Mediterranean mercury assessment 2022: an updated budget, health consequences, and research perspectives. Environ. Sci. Technol. 56 https://doi.org/10.1021/acs.est.1c03044.

Cresson, P., Fabri, M., Bouchoucha, M., Brach-papa, C., Chavanon, F., Jadaud, A., Knoery, J., Marco-miralles, F., Cossa, D., 2014. Mercury in Organisms From the Northwestern Mediterranean Slope: Importance of Food Sources. https://doi.org/10.1016/j.scitotenv.2014.07.069.

Eagles-Smith, C.A., Ackerman, J.T., Adelsbach, T.L., Takekawa, J.Y., Miles, A.K., Keister, R.A., 2008. Mercury correlations among six tissues for four waterbird species breeding in San Francisco Bay, California, USA. Environ. Toxicol. Chem. 27, 2136–2153. https://doi.org/10.1897/08-038.1.

EC, 2005. Common Implementation Strategy for the Water Framework Directive. Environmental Quality Standards (EQS) Substance Data Sheet. Priority Substance No. 21 Mercury and its Compounds CAS-No. 7439-97-6. Final version. Brussels, 15 January 2005.

EC, 2008. Directive 2008/56/EC of the European Parliament and the Council of 17 June 2008 Establishing a Framework for Community Action in the Field of Marine Environmental Policy (Marine Strategy Framework Directive). Off. J. Eur. Union.

EC, 2013. Directive 2013/39/EU of the European Parliament and of the Council of 12 August 2013 amending Directives 2000/60/EC and 2008/105/EC as regards priority substances in the field of water policy. Off. J. Eur. Union.

Eisler, R., 2007. Eisler's Encyclopedia of Environmentally Hazardous Priority Chemicals (eBook ISBN: 9780080547077 [WWW Document]. URL (accessed 7.16.23)).

Fliedner, A., Rüdel, H., Knopf, B., Lohmann, N., Paulus, M., Jud, M., Pirntke, U., Koschorreck, J., 2018. Assessment of seafood contamination under the marine strategy framework directive: contributions of the German environmental specimen bank. Environ. Sci. Pollut. Res. 25, 26939–26956. https://doi.org/10.1007/s11356-018-2728-1.

Gerson, J.R., Walters, D.M., Eagles-Smith, C.A., Bernhardt, E.S., Brandt, J.E., 2020. Do two wrongs make a right? Persistent uncertainties regarding environmental selenium–mercury interactions. Environ. Sci. Technol. 54, 9228–9234. https://doi.org/10.1021/acs.est.0c01894.

Gochfeld, M., Burger, J., 2021. Mercury interactions with selenium and sulfur and the relevance of the Se:Hg molar ratio to fish consumption advice. Environ. Sci. Pollut. Res. Int. 28, 18407–18420. https://doi.org/10.1007/s11356-021-12361-7.

Hall, B.D., Bodaly, R.A., Fudge, R.J.P., Rudd, J.W.M., Rosenberg, D.M., 1997. Food as the dominant pathway of methylmercury uptake by fish. Water Air Soil Pollut. 100, 13–24. https://doi.org/10.1023/A:1018071406537.

HELCOM, 2017. Downloads & Data – State of the Baltic Sea – Second HELCOM Holistic Assessment.

IAMMWG, 2015. Management Units for Cetaceans in UK Waters (January 2015). JNCC Report No. 547, JNCC Peterborough.

ICES, 2013. Report of the Working Group on Marine Mammal Ecology (WGMME), 4-7 February 2013. ICES, Paris, France. ICES CM 2013/ACOM:26. 117 pp.

ICES, 2014. Report of the Working Group on Marine Mammal Ecology (WGMME), 10-13 March 2014. ICES, Woods Hole, MA, USA. ICES CM 2014/ACOM:27. 234 pp.

Kershaw, J.L., Hall, A.J., 2019. Mercury in cetaceans: exposure, bioaccumulation and toxicity. Sci. Total Environ. 694, 133683 https://doi.org/10.1016/j.scitotenv.2019.133683.

Laffoley, D., Baxter, J.M., Amon, D.J., Currie, D.E.J., Downs, C.A., Hall-Spencer, J.M., Harden-Davies, H., Page, R., Reid, C.P., Roberts, C.M., Rogers, A., Thiele, T., Sheppard, C.R.C., Sumaila, R.U., Woodall, L.C., 2020. Eight urgent, fundamental and simultaneous steps needed to restore ocean health, and the consequences for humanity and the planet of inaction or delay. Aquat. Conserv. Mar. Freshwat. Ecosyst. 30, 194–208. https://doi.org/10.1002/aqc.3182.

Law, R.J., 1996. Metals in marine mammals. In: Beyer, W.N., Heinz, G.H., Redmond-Norwood, A.W. (Eds.), Environmental Contaminants in Wildlife: Interpreting Tissue Concentrations, SETAC Special Publications Series. Lewis Publishers.

Law, R.J., Jones, B.R., Baker, J.R., Kennedy, S., Milne, R., Morris, R.J., 1992. Trace metals in the livers of marine mammals from the Welsh coast and the Irish Sea. Mar. Pollut. Bull. 24, 296–304. https://doi.org/10.1016/0025-326X(92)90590-3.

Lebigre, C., Aminot, Y., Munschy, C., Drogou, M., Le Goff, R., Briant, N., Chouvelon, T., 2022. Trace metal elements and organic contaminants are differently related to the growth and body condition of wild European sea bass juveniles. Aquat. Toxicol. 248, 106207 https://doi.org/10.1016/j.aquatox.2022.106207.

Mauffret, A., Chouvelon, T., Wessel, N., Cresson, P., Bănaru, D., Baudrier, J., Bustamante, P., Chekri, R., Jitaru, P., Le Loc'h, F., Mialet, B., Vaccher, V., Harmelin-Vivien, M., 2023. Trace elements, dioxins and PCBs in different fish species and marine regions: importance of the taxon and regional features. Environ. Res. 216, 114624 https://doi.org/10.1016/j.envres.2022.114624.

Meheust, E., Dars, C., Dabin, W., Demaret, F., Méndez-Fernandez, P., Peltier, H., Spitz, J., Caurant, F., Van Canneyt, O., 2021. Les échouages de mammifères marins sur le littoral français en 2020. In: Rapport scientifique de l'Observatoire Pelagis. Université et CNRS, La Rochelle (43 pages).

Méndez-Fernandez, P., Spitz, J., Dars, C., Dabin, W., Mahfouz, C., André, J.-M., Chouvelon, T., Authier, M., Caurant, F., 2022. Two cetacean species reveal different long-term trends for toxic trace elements in European Atlantic French waters. Chemosphere 294, 133676. https://doi.org/10.1016/j.chemosphere.2022.133676.

Monteiro, L.R., Furness, R.W., 1995. Seabirds as monitors of mercury in the marine environment. In: Porcella, D.B., Huckabee, J.W., Wheatley, B. (Eds.), Mercury as a Global Pollutant: Proceedings of the Third International Conference Held in Whistler, British Columbia, July 10–14, 1994. Springer Netherlands, Dordrecht, pp. 851–870. https://doi.org/10.1007/978-94-011-0153-0_90.

OSPAR, 2016. Mercury Assessment in the Marine Environment Assessment Criteria Comparison (EAC/EQS) for Mercury.

OSPAR, 2022a. Factors for converting the basis of assessment concentrations in biota. https://dome.ices.dk/ohat/trDocuments/2022/help_ac_basis_conversion.html.

OSPAR, 2022b. Assessment methodology for contaminants in biota. https://dome.ices.dk/ohat/trDocuments/2022/help_methods_biota_contaminants.html.

Pan, K., Wang, W.-X., 2011. Mercury accumulation in marine bivalves: influences of biodynamics and feeding niche. In: Environmental Pollution, Nitrogen Deposition, Critical Loads and Biodiversity, 159, pp. 2500–2506. https://doi.org/10.1016/j.envpol.2011.06.029.

Peltier, H., Authier, M., Deaville, R., Dabin, W., Jepson, P.D., van Canneyt, O., Daniel, P., Ridoux, V., 2016. Small cetacean bycatch as estimated from stranding schemes: the common dolphin case in the northeast Atlantic. Environ. Sci. Pol. 63, 7–18. https://doi.org/10.1016/j.envsci.2016.05.004.

Peltier, H., Authier, M., Caurant, F., Dabin, W., Daniel, P., Dars, C., Demaret, F., Meheust, E., Van Canneyt, O., Spitz, J., Ridoux, V., 2021. In the wrong place at the wrong time: identifying spatiotemporal co-occurrence of bycaught common dolphins and fisheries in the Bay of Biscay (NE Atlantic) from 2010 to 2019. Front. Mar. Sci. 8.

Rawson, A.J., Patton, G.W., Hofmann, S., Pietra, G.G., Johns, L., 1993. Liver abnormalities associated with chronic mercury accumulation in stranded Atlantic bottlenose dolphins. Ecotoxicol. Environ. Saf. 25, 41–47. https://doi.org/10.1006/eesa.1993.1005.

Rudershausen, P.J., Cross, F.A., Runde, B.J., Evans, D.W., Cope, W.G., Buckel, J.A., 2023. Total mercury, methylmercury, and selenium concentrations in blue marlin Makaira nigricans from a long-term dataset in the western north Atlantic. Sci. Total Environ. 858, 159947 https://doi.org/10.1016/j.scitotenv.2022.159947.

Santos, M.B., Pierce, G.J., 2003. The diet of harbour porpoise (Phocoena phocoena) in the Northeast Atlantic. Oceanogr. Mar. Biol. 41, 355–390.

Santos, M.B., German, I., Correia, D., Read, F.L., Cedeira, J.M., Caldas, M., Lopez, A., Velasco, F., Pierce, G.J., 2013. Long-term variation in common dolphin diet in relation to prey abundance. Mar. Ecol. Prog. Ser. 481, 249–268. https://doi.org/10.3354/meps10233.

Savouré-Soubelet, A., Aulagnier, S., Haffner, P., Moutou, F., Van Canneyt, O., Charrassin, J.-B., Ridoux, V. (Eds.), 2016. Atlas des mammifères sauvages de France, volume 1: Mammifères marins. Muséum national d'Histoire naturelle, Paris; IRD, Marseille, 480 p. (Patrimoines naturels; 74). https://doi.org/10.5852/cpn74.

Spitz, J., Rousseau, Y., Ridoux, V., 2006. Diet overlap between harbour porpoise and bottlenose dolphin: an argument in favour of interference competition for food? Estuar. Coast. Shelf Sci. 70, 259.

Storelli, M.M., 2008. Potential human health risks from metals (Hg, Cd, and Pb) and polychlorinated biphenyls (PCBs) via seafood consumption: estimation of target hazard quotients (THQs) and toxic equivalents (TEQs). Food Chem. Toxicol. 46, 2782–2788. https://doi.org/10.1016/j.fct.2008.05.011.

Streets, D.G., Horowitz, H.M., Lu, Z., Levin, L., Thackray, C.P., Sunderland, E.M., 2019. Global and regional trends in mercury emissions and concentrations, 2010–2015. Atmos. Environ. 201, 417–427. https://doi.org/10.1016/j.atmosenv.2018.12.031.

Syversen, T., Kaur, P., 2012. The toxicology of mercury and its compounds. J. Trace Elem. Med. Biol. 26, 215–226. https://doi.org/10.1016/j.jtemb.2012.02.004.

Tornero, V., Hanke, G., 2016. Chemical contaminants entering the marine environment from sea-based sources: a review with a focus on European seas. Mar. Pollut. Bull. 112, 17–38. https://doi.org/10.1016/j.marpolbul.2016.06.091.

Wang, W.X., 2002. Interactions of trace metals and different marine food chains. Mar. Ecol. Prog. Ser. 243, 295–309. https://doi.org/10.3354/meps243295.

Wang, Z., Li, Y., Kong, F., Li, M., Xi, M., Yu, Z., 2021. How do trophic magnification factors (TMFs) and biomagnification factors (BMFs) perform on toxic pollutant bioaccumulation estimation in coastal and marine food webs. Reg. Stud. Mar. Sci. 44, 101797 https://doi.org/10.1016/j.rsma.2021.101797.

Zhang, H., Feng, X., Chan, H.M., Larssen, T., 2014. New insights into traditional health risk assessments of mercury exposure: implications of selenium. Environ. Sci. Technol. 48, 1206–1212. https://doi.org/10.1021/es4051082.