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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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**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 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.

**Keywords:** Integrated assessment; contaminants; marine; monitoring; biota

## 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

# 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 amongst 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 24h. 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 benthic-demersal surveys using bottom trawling: CGFS 2018 (Channel Ground Fish Survey) for the English Channel and the eastern Celtic Sea (doi: [10.17600/18000517](https://doi.org/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](https://doi.org/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 to 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., 2021, 2016) and their different feeding habitats and thus prey sources. Common dolphins can be found in both coastal and offshore waters to more than 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 less than 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.

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 <sup>-1</sup> w.w. <sup>1</sup>		
				Number of samples ( <sup>2</sup> )		
				EC	CS	BoB
<b>Bivalves</b>	<b>Flesh</b> Pool of ≥ 50 individuals	Mussel	Suspension feeder	0.017/0.020 22 (22 t)	0.014/0.017 7 (5 t)	0.018/0.022 14 (9 t)
		<i>Mytilus</i> spp				
<b>Fish</b>	<b>Muscle</b> Individual sample or pool for anchovy	Atlantic mackerel	Mainly pelagic piscivore	0.051/0.059 15	0.022/0.028 20	0.078/0.085 25
		European anchovy	Pelagic planktivore			0.033/0.045 14
		European hake	Demersal piscivore		0.019/0.022 10	0.040/0.044 25
		European plaice	Benthic invertebrate feeder	0.085/0.121 8		
		Poor cod	Planktivorous			0.064/0.080 13
		Spotted dogfish	Demersal invertebrate feeder	0.568/0.677 17	0.251/0.316 16	0.335/0.396 23
		Whiting	Demersal piscivore	0.099/0.118 12		
		<i>Trisopterus minutus</i>				
		<i>Scyliorhinus canicula</i>				
		<i>Merlangius merlangus</i>				
<b>Birds</b>	<b>Blood</b> Individual sample	European shag	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	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	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	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	Coastal opportunist			0.147/0.163 8 (1 c)
		<i>Larus michahellis</i>				
		Northern gannet	Offshore piscivore		0.281/0.318 15 (1 c)	
		<i>Morus bassanus</i>				
		<i>Phalacrocorax aristotelis</i>				
<b>Mammals</b>	<b>Liver</b> Individual sample	Common dolphin	Pelagic consumer		10.5/11.4 259 (19 y)	
		Harbour porpoise	Demersal consumer	12.4/15.4 68 (16 y)		8.8/11.7 56 (16 y)
		<i>Phocoena phocoena</i>				

<sup>1</sup> Mean and upper one-sided interval confidence (IC) limits are calculated from log concentration of Hg.

<sup>2</sup> In brackets: number of temporal trend analyses for bivalves (t), number of colonies for birds (c), number of years monitored for mammals (y).

## 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% (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](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, (2014, 2013) . For the common dolphin, a single geographical assessment unit is currently recognised 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 ( $EQS_{sp} = 0.02 \text{ mg kg}^{-1} \text{ w.w.}$ , EC, 2013) and the Low Risk Concentration for birds (LRC =  $1 \text{ mg kg}^{-1} \text{ w.w.}$ ). 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 ( $EQS_{sp} = 0.02 \text{ mg kg}^{-1} \text{ w.w.}$ , EC, 2013). For mammals, the threshold of  $61 \text{ mg kg}^{-1} \text{ w.w.}$  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 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.

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	Group of species	Step I	Step II	Step III	Assessment parameter
Number of years in dataset	Assessment unit	Contamination index for each species in each assessment unit	Contamination ratio/trend for each species in each assessment unit	Spatial aggregation Contamination ratio or trend for each species in each marine region	Species integration Contamination ratio/trend for each group of species in each marine region
<b>Absolute assessment</b> 1-2 years	<b>Fish</b> 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 <sup>-1</sup> w.w. (2013/39/UE, 2013)	$CR_{sp}$	$CS_{fish} = \frac{\sum CR_{sp}}{\sqrt{N_{sp}}}$ <1: GES reached ≥1: GES not reached
	<b>Birds</b> 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 <sup>-1</sup> w.w. (Ackerman <i>et al.</i> , 2016)	$CR_{sp} = \text{mean}$ $CR_{colony*sp}$	$CS_{birds} = \frac{\sum CR_{sp}}{\sqrt{N_{sp}}}$ <1: GES reached ≥1: GES not reached
<b>Absolute assessment</b> ≥ 3 years	<b>Bivalves</b> 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 <sup>-1</sup> w.w. (2013/39/UE, 2013)	$CR_{sp} = \text{mean}$ $CR_{station*sp}$	$CS_{bivalves} = CR_{sp}$ <1: GES reached ≥1: GES not reached
	<b>Mammals</b> 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 <sup>-1</sup> w.w. (Rawson <i>et al.</i> , 1993)	$CR_{sp} = CR_{unit*sp}$	$CS_{mammals} = \frac{\sum CR_{sp}}{\sqrt{N_{sp}}}$ <1: GES reached ≥1: GES not reached
<b>Relative assessment</b> ≥ 5 years	<b>Bivalves</b> 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 ( <i>T</i> ) 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
	<b>Mammals</b> ICES and OSPAR assessment unit	Dynamic Linear Model & WAIC selection: log[Hg] ~ Year + Age + Sex + δ <sup>13</sup> C or δ <sup>15</sup> N	Linear model: Slope ( <i>T</i> ) and statistical significance (P value)	-	$N_{species} \leq 2$ : $T_{GES} = T_{max}$ (One-Out-All-Out) $T_{GES*mammals}$ non-significant or decreases significantly: GES unknown $T_{GES*mammals}$ increases significantly: GES not reached

### 3 Results and Discussion

#### 3.1 Four complementary groups of species

The order of increasing Hg concentrations was bivalves < fish = birds < mammals ( $F = 105.96$ ,  $P\text{-value} < 0.001$ ) 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<sup>-1</sup> 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<sup>-1</sup> w.w. (Supplementary Material, Table S2). Pelagic fish species (*e.g.* European anchovy, Atlantic mackerel) 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<sup>-1</sup> 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<sup>-1</sup> 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 ( $F = 3.89$ ,  $P\text{-value} = 0.0287$ ). 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<sup>-1</sup> 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 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<sup>-1</sup> w.w. for birds and 61.1 mg.kg<sup>-1</sup> 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, Table S3 and Table 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<sup>-1</sup> 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<sup>-1</sup> 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  $61 \text{ mg kg}^{-1}$  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.

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 ( $p < 0.05$ ), <sup>NS</sup> non-significant.

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

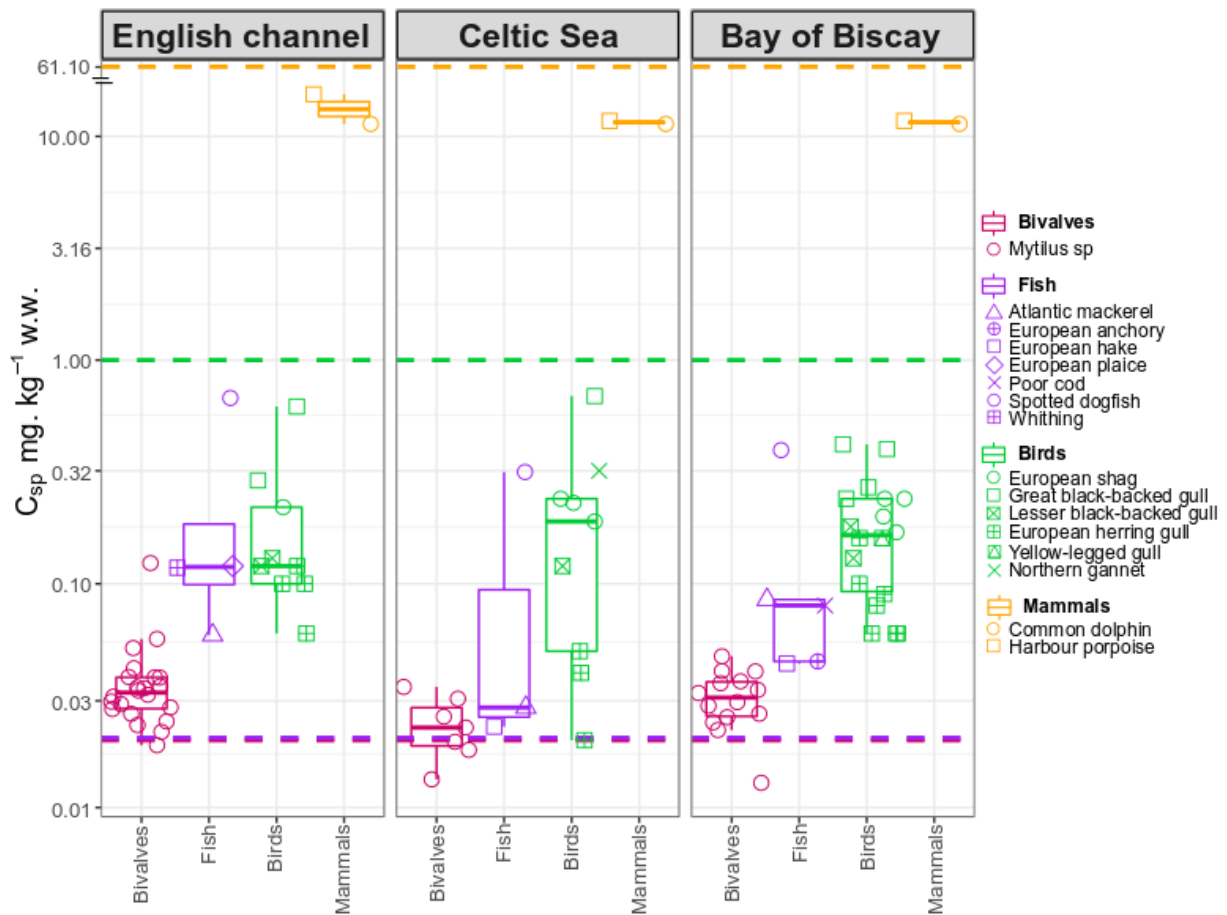


Fig. 1. Concentrations of Hg for four groups of species in the three marine regions. The bottom and top of the box are the first and the third quartiles of the  $C_{sp}$  distribution, the horizontal segment is the median, whiskers represent the most extreme data point within 1.5 interquartile range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

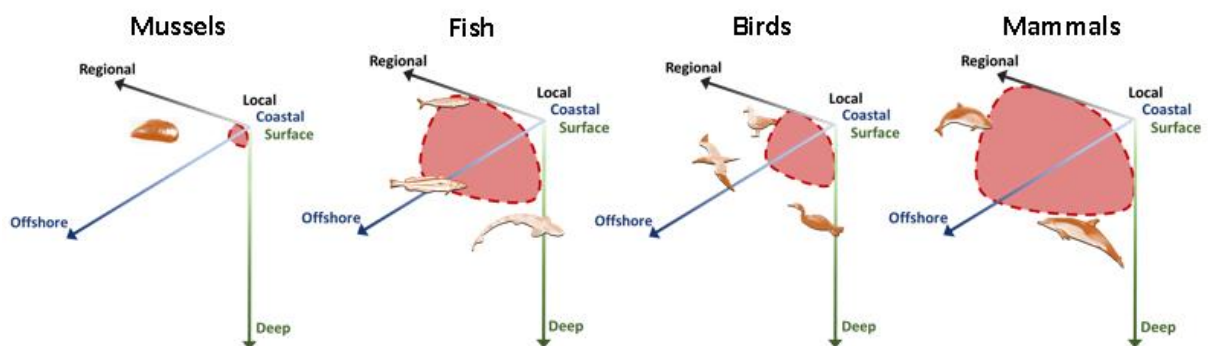


Fig. 2. A multi-matrix approach including four complementary groups of species to assess the environmental Hg contamination (geographic extent in black, distance from the coast in blue and depth in green).



## 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.

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## Supplementary material

Table S1. Absolute and relative assessments in mercury contamination for bivalves in the three marine regions along the French Atlantic coast.

Marine region	Bivalve species	Station	Absolute assessment			Relative assessment		
			$C_{station}$ $\mu\text{g kg}^{-1}$ d.w.	$CR_{station*sp}$	$CR_{sp} =$ $CS_{bivalves}$ GES	Years	$T$ $\mu\text{g kg}^{-1}$ d.w./year	$T_{GES*bivalves}$ $(T)$ $\mu\text{g kg}^{-1}$ d.w./year GES
		Threshold	121.95 $\mu\text{g kg}^{-1}$ d.w.	1	1			
English Channel	<i>Mytilus</i> spp	Ambleteuse	257.09	2.11	1.87 Not reached	2005 - 2020	4.9*	5.06* Not reached
		Antifer - digue	316.69	2.6		2005 - 2020	6.5*	
		Baie St Michel est 6	148.87	1.22		2011 - 2020	10.7*	
		Bdv Grandcamp Ouest	171.91	1.41		2005 - 2020	2.4*	
		Berck Bellevue	160.99	1.32		2005 - 2020	5.9*	
		Bréville	213.26	1.75		2005 - 2020	8.1*	
		Cap de la Hève	169.13	1.39		2005 - 2017	0.4 <sup>NS</sup>	
		Chausey	235.21	1.93		2009 - 2020	11.3*	
		Goury	182.51	1.5		2011 - 2020	0.5 <sup>NS</sup>	
		Grande rade de Cherbourg	195.91	1.61		2005 - 2019	3.9*	
		Le Moulard	209.48	1.72		2005 - 2020	2.1 <sup>NS</sup>	
		Meuvaines ouest	204.64	1.68		2011 - 2020	6.8*	
		Ouistreham	211.03	1.73		2005 - 2020	4.2*	
		Oye plage	143.37	1.18		2005 - 2020	2.1 <sup>NS</sup>	
		Pirou Nord	133.65	1.1		2005 - 2020	3.8*	
		Pointe de St Quentin	177.92	1.46		2005 - 2020	5.4*	
		Port en Bessin	192.10	1.58		2005 - 2020	2.4 <sup>NS</sup>	
		St Germain de Varreville	116.55	0.96		2009 - 2020	4.4*	
		Varengeville	234.40	1.92		2005 - 2020	2.5 <sup>NS</sup>	
		Villers sur mer	233.74	1.92		2010 - 2020	13.1*	
Villerville	348.09	2.85	2005 - 2020	3.2*				
Yport	759.56	6.23	2011 - 2020	6.7 <sup>NS</sup>				
Celtic Sea	<i>Mytilus</i> spp	Baie de la Fresnaye	111.00	0.91	1.27 Not reached	2005 - 2020	5.0*	4.90 <sup>NS</sup>
		Baie de Lampaul	82.01	0.67		-	-	
		Kervel	120.66	0.99		2005 - 2020	5.2*	
		La Gauthier	156.41	1.49		-	-	
		Le Vivier sur mer	212.24	1.74		2005 - 2020	6.5*	
		Pointe du Roselier	139.77	1.33		2012 - 2016	3.8 <sup>NS</sup>	
		St Michel en Grève	188.08	1.79		2005 - 2016	4.0*	
Bay of Biscay	<i>Mytilus</i> spp	Barres de Pen Bron 1	79.12	0.65	1.57 Not reached	-	-	6.18*
		Capbreton ouest	199.43	1.64		2005 - 2020	2.9*	
		Ile d'Yeu est	249.13	2.04		-	-	
		Ile Dumet (a)	249.83	2.05		2012 - 2020	18.5*	
		La Jument	136.23	1.12		2006 - 2020	0.8 <sup>NS</sup>	
		La Mouclière	224.83	1.84		2005 - 2020	2.6*	
		La Pointe de la Roche	181.49	1.49		-	-	
		Le Halguen	147.43	1.21		2012 - 2020	17*	
		Pen Bé	205.76	1.69		2005 - 2020	7.1*	
		Pointe Castelli	220.52	1.81		-	-	
		Pointe de Chemoulin	174.81	1.43		2005 - 2020	3.3*	
		Pointe de Moustierlin	161.04	1.32		2005 - 2020	2.4*	
		Pointe er Fosse	155.46	1.27		2005 - 2020	1 <sup>NS</sup>	
		Saumonards Filières	290.89	2.39		-	-	

\*p<0.05, <sup>NS</sup> non-significant.  $C_{station}$ : Hg concentration in bivalves from one station. CR: Concentration ratio. CS: Contamination score. T: temporal trend. GES: Good Environmental Status. sp: species. Years refer to the period covered by the temporal trend assessment



Table S2. Absolute assessments in mercury contamination for fish in the three marine regions along the French Atlantic coast.

Marine region	Fish species	N	Breeding season <sup>1</sup>	Size range cm	$C_{sp}$	$CR_{sp}$	$CS_{fish}$ GES
	Threshold				0.02 mg kg <sup>-1</sup> w.w.	1	1
English Channel	Whiting	12	Jan.-Sept.	27.0 - 31.8	0.118	5.9	24.36 Not reached
	Plaice	8	Dec.-March	24.5 - 28.6	0.12	6.05	
	Mackerel	15	April-July	26.0 - 32.1	0.059	2.95	
	Spotted dogfish	17	All year, more active in June-July	39.0 - 60.0	0.677	33.85	
Celtic Sea	Hake	10	Nov-May	26.8 - 35.0	0.023	1.15	10.58 Not reached
	Mackerel	20	April-July	15.5 - 31.1	0.028	1.4	
	Spotted dogfish	16	All year, more active in June-July	36.3 - 58.0	0.316	15.8	
Bay of Biscay	Anchovy	14	March-Aug	10.0 - 16.8	0.045	2.25	14.52 Not reached
	Hake	25	Nov-May	29.2 - 36.5	0.044	2.2	
	Mackerel	25	April-July	24.3 - 32.5	0.085	4.25	
	Spotted dogfish	23	All year, more active in June-July	30.3 - 49.5	0.396	19.8	
	Poor cod	13	Feb.-Avril.	15.1 - 21.5	0.080	4	

$C_{sp}$ : Hg concentration in one fish species in one marine region. CR: Concentration ratio. CS: Contamination score. GES: Good Environmental Status. sp: species

<sup>1</sup> Breeding season according to Fishbase, consulted in 2018

Table S3: Absolute assessments in mercury contamination for birds in the three marine regions along the French Atlantic coast.

Marine region	Bird species	Colony	N	$C_{colony*sp}$	$CR_{colony*sp}$	$CR_{sp}$	$CS_{birds}$ GES
		Threshold		1 mg kg <sup>-1</sup> w.w.	1	1	
English Channel	Great black-backed gull	Baie de Seine	15	0.29	0.29	0.46	0.46 Reached
		Chausey	18	0.62	0.62		
	European herring gull	Calais	10	0.06	0.06	0.10	
		Boulogne	15	0.12	0.12		
		Baie de Seine	15	0.10	0.10		
		Chausey	17	0.10	0.10		
	Lesser black-backed gull	Calais	12	0.12	0.12	0.13	
		Boulogne	11	0.13	0.13		
	European shag	Chausey	17	0.22	0.22	0.22	
Celtic Sea	Great black-backed gull	Rade de Brest	12	0.69	0.69	0.69	0.62 Reached
	European herring gull	Sept-îles	15	0.02	0.02	0.04	
		Morlaix	15	0.05	0.05		
		Abers	15	0.04	0.04		
	Lesser black-backed gull	Abers	14	0.12	0.12	0.12	
	European shag	Sept-îles	16	0.23	0.23	0.22	
		Rade de Brest	12	0.19	0.19		
		Ti Saozon	16	0.24	0.24		
Northern gannet	Sept-îles	15	0.32	0.32	0.32		
Bay of Biscay	European herring gull	Glénan	15	0.06	0.06	0.09	0.42 Reached
		Lorient	15	0.10	0.10		
		Méaban	12	0.08	0.08		
		Dumet	14	0.06	0.06		
		Bilho	15	0.06	0.06		
		Yeu	15	0.09	0.09		
		Arguin	15	0.16	0.16		
	Lesser black-backed gull	Belle-Ile	15	0.18	0.18	0.16	
		Yeu	15	0.13	0.13		
	Yellow-legged gull	Arguin	8	0.16	0.16	0.16	
	Great black-backed gull	Glénan	14	0.42	0.42	0.33	
		Houat	15	0.24	0.24		
		Bilho	15	0.27	0.27		
		Pilier	15	0.40	0.40		
	European shag	Glénan	15	0.17	0.17	0.21	
		Houat	15	0.24	0.24		
		Méaban	14	0.24	0.24		
		Pilier	13	0.20	0.20		

$C_{colony*sp}$ : Hg concentration in one bird species from one colony. CR: Concentration ratio. CS: Contamination score. GES: Good Environmental Status. sp: species

Table S4. Absolute and relative assessments in mercury contamination for mammals in the three marine regions along the French Atlantic coast.

Marine region	Assessment unit	Mammal species	Absolute assessment			Relative assessment	
			$C_{unit*sp}$ mg kg <sup>-1</sup> w.w.	$CR_{unit*sp} = CR_{sp}$	$CS_{mammals}$ GES	$T$ mg kg <sup>-1</sup> d.w./year (years)	$T_{GES*mammals}$ ( $T_{max}$ ) mg kg <sup>-1</sup> d.w./year GES
		Threshold	61.1 mg kg <sup>-1</sup> w.w.	1	1		
English Channel	EC + CS + BoB	Common dolphin	15.43	0.25	0.30 Reached	1.96* (2001-2019)	1.96* Not reached
	EC	Harbour porpoise	10.65	0.17		0.36 <sup>NS</sup> (2004-2019)	
Celtic Sea + Bay of Biscay	EC + CS + BoB	Common dolphin	15.43	0.25	0.26 Reached	1.96* (2001-2019)	1.96* Not reached
	CS + BoB	Harbour porpoise	6.81	0.11		0.12 <sup>NS</sup> (2004-2019)	

\*p<0.05, <sup>NS</sup> non-significant.  $C_{unit*sp}$ : Hg concentration in one mammal species from one assessment unit. CR: Concentration ratio. CS: Contamination score. T: temporal trend. GES: Good Environmental Status. sp: species. Years refer to the period covered by the temporal trend assessment

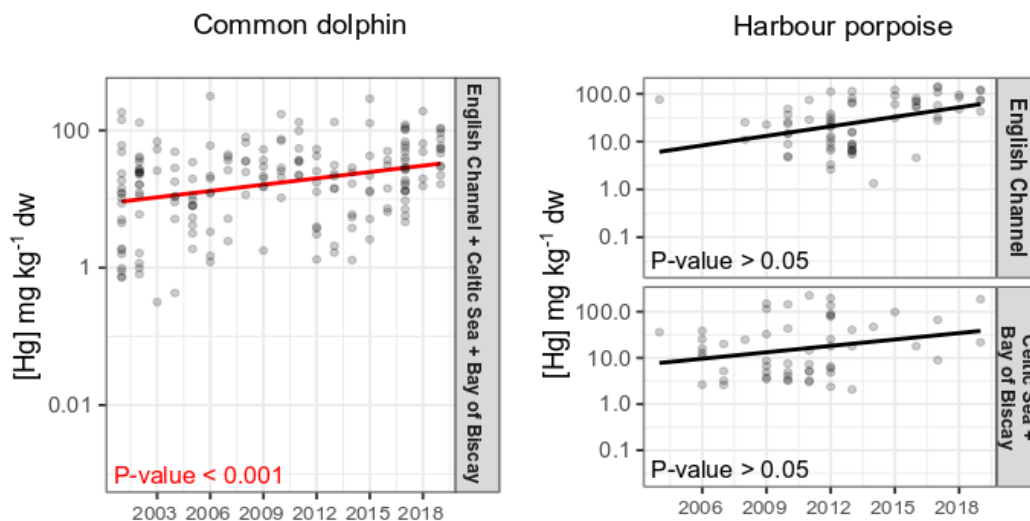


Fig. S1. Linear trend of Hg concentrations over time for the two monitored mammal species in the three marine regions along the French Atlantic coast. Grey points are Hg concentration (log transformed) measured in the liver. Lines represent trends obtained from a dynamic linear model (DLM) including age, sex and  $\delta_{13}C$  or  $\delta_{15}N$  as confounding factors known to cause Hg concentration variation (significant increase of Hg concentration in red, non-significant increase of Hg concentration in black).