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ABSTRACT

Chemical contaminant concentrations in wild organisms are used to assess environmental status under the European Marine Strategy Framework Directive. However, this approach is challenged by the complex intra- and inter-species variability, and the different regional features. In this study, concentrations in trace elements (As, Cd, Hg and Pb), polychlorinated biphenyls (PCBs), polychlorodibenzo-para-dioxines (PCDDs) and polychlorodibenzofuranes (PCDFs) were monitored in 8 fish species sampled on the continental shelf of three French regions: the Eastern English Channel (EEC) and Bay of Biscay (BoB) in the Northeast Atlantic Ocean, and the Gulf of Lions (GoL) in Western Mediterranean Sea. Our objectives were to identify species or regions more likely to be contaminated and to assess how to take this variability into account in environmental assessment. While concentrations were higher in benthic and demersal piscivores, PCB and PCDD/F concentrations (lipid-weight) were similar in most teleost species. For Cd, Hg and Pb, the trophic group accumulating the highest concentrations depended on the contaminant and region. Concentrations in Hg, PCBs and PCDD/Fs were higher in the EEC and/or GoL than in BoB. Cadmium and Pb concentrations were highest in the BoB. Lipid content accounted for 35% to 84% of organic contaminant variability. Lipid normalisation was employed to enhance robustness in the identification of spatial patterns. Contaminant patterns in chondrichthyans clearly differed from that in teleosts. In addition, trophic levels accounted for ≤1% and ≤33% of the contaminant variability in teleost fishes in the EEC and BoB, respectively. Therefore, developing taxa-specific thresholds might be a more practical way forward for environmental assessment than normalisation to trophic levels.

KEYWORDS

Metals, Persistent Organic Pollutants, Bioaccumulation, Trophic level, Lipid content, Monitoring

HIGHLIGHTS

- The trophic group most contaminated depended on the contaminant and region.

- Regional variability was linked with river inputs, trophic status and functioning and element cycling.

- Hg, Cd, PCBs and TEQ were correlated with trophic levels in the BoB, not in the EEC.

- Lipid normalisation enhances robustness in the identification of spatial patterns.

- Taxa-specific thresholds seemed more relevant than trophic adjustment for GES assessment.

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1. INTRODUCTION

Chemical contaminants are part of everyday life for many modern societies. They can reach the environment from multiple natural or anthropic sources as they may be 1) naturally occurring and used in industrial processes such as metal production, 2) unintentionally formed as by-products of natural and human-induced chemical processes such as dioxins and furans (polychlorodibenzo-para-dioxines (PCDD) and polychlorodibenzofurane (PCDF)), or 3) synthesised specifically for industrial processes and consumer products such as polychlorinated biphenyls (PCB) (OSPAR, 2010, OSPAR, in prep.).

Once in the environment, chemical contaminants may persist and/or affect wildlife. Amongst the known contaminants, arsenic (As), cadmium (Cd), lead (Pb) and mercury (Hg) are trace metals recognised for their toxicity even at low levels (e.g. Ishaque et al., 2006). Over the past few decades, after the tragic history of Minamata disease involving masspoisoning with methyl-mercury (MeHg), various countries attributed much more importance to the management of metal pollution that resulted in decreased Hg emission in the United States of America, Europe and Canada (Matsuo, 2000, Sun et al., 2020). However Streets et al. (2019) estimated that worldwide Hg emissions increased between 2010 and 2015 by 1.8%/y as a result of a Hg emission increase in Central America, Eastern Africa and Southeast Asia. Furthermore, PCBs and dioxins have been classified as Persistent Organic Pollutants (POP, http://www.pops.int/) in the Stockholm convention due to their toxicity and environmental persistence (UNEP, 2019). Previous regulations (*e.g.* PCBs have been banned in France since 1987) have resulted in a decline of PCB concentrations over the last decades, but this trend has slowed down. PCBs still persist locally in the marine environment above thresholds indicating a possible impact on marine organisms and/or humans, even in regions where PCBs are not produced (e.g. in the Arctic or Africa Gioia et al., 2008, Jepson and Law, 2016). This is mainly due to a combination of the long-range transport ability of PCBs and prevalence of open burning of waste, *e.g.* across Africa, particularly electronic waste, including that received from developed countries (White et al., 2021). Therefore, although these contaminants (i.e. metals, PCBs and dioxins) are subject to regulations or conventions at the global, regional (European, Regional Sea convention) or national levels, they are still of concern for environmental and for human health.

The fate of contaminants in the environment needs to be better understood in order to protect the marine ecosystem and biodiversity upon which human health and marine-related economic and social activities depend. Monitoring of these contaminants is required to assess good environmental status under Descriptor 8 of the European Marine Strategy Framework Directive (MSFD Directive 2008/56/EC). Marine organisms such as bivalves or fish accumulate persistent contaminants over time, making them useful bioindicators of long-term changes in environmental quality (Yancheva et al., 2018, Simonnet-Laprade et al., 2021, Constenla et al., 2022). In France, chemical contamination has been monitored in coastal bivalves since the late 1970’s. In order to extend the spatial coverage of contaminant monitoring to the French continental shelves, and to extend the monitoring to higher trophic level species that can be exposed to contaminants in different ways than bivalves, chemical contaminants have more recently been monitored (since 2014) in fishes sampled during fishery surveys along the metropolitan French coast (Baudrier et al., 2018).

However, intra- and inter-species variability in contaminant concentrations has to be taken into account when using fish contamination as an indicator of the environmental status. Levels of chemical contaminants in fish often differ among individuals and species (Ghosn et al., 2020). Both intra- and inter-species variability can be related to i) environmental contamination, ii) environmental conditions favouring bioaccumulation and biomagnification (e.g. depth, habitat, temperature, primary production; Cossa et al., 2022) and iii) biological drivers including body length, age, sex, growth rate, metabolism capacity, feeding guild or trophic level (Lavoie et al., 2013, Cresson et al., 2016, Burke et al., 2020, Lescord et al., 2020, Donadt et al., 2021, Cossa et al., 2022). Therefore, though intra- and inter-species variability in chemical contamination illustrates the natural variability, they also lead to increased complexity in terms of environmental assessment and regional comparison. In order to assess contamination patterns, *e.g.* identifying region or species particularly vulnerable to contamination, and to compensate for differences in sampling strategies, contaminant levels in fish might be normalised to lipid content for hydrophobic substances which tend to accumulate in non-polar compartments and/or to a common trophic level for compounds likely to biomagnify in the trophic network. It is however essential to verify and quantify the relationship between the potential normaliser (lipid content or trophic level) and the contaminant concentrations to avoid introducing more variability and misinterpreting the data (Hebert and Keenleyside, 1995).

One reliable approach to achieve a better understanding of the functioning of marine ecosystems and also to identify key indicators, is to compare ecosystems with different food web characteristics (Murawski et al., 2010, Safi et al., 2019, Sun et al., 2020). In the present study, we propose to compare fish contamination in three French metropolitan regions: two from the Northeast Atlantic coast, the Eastern English Channel (EEC) and Northern Bay of Biscay (BoB) and one in the Western Mediterranean Sea, the Gulf of Lions (GoL). These three regions have different sea- and land-based sources of contamination, trophic status, water depth and connection to the open ocean (Table SM 1).

In the present study, we compared fish contaminant concentrations among species within each region, as well as among regions within one species. The relationship with selected drivers that could be used as potential normalisers for environmental assessment (lipid content and trophic level) was determined. Our objectives were 1) to identify species or regions more likely to be contaminated, and 2) to assess whether the use of lipid content or trophic level as normalisers could support environmental assessment based on *in situ* fish monitoring.

2. MATERIAL AND METHODS

2.1. Sampling: joint surveys, spatial coverage, dissections

Fish were collected during four existing fisheries-related surveys, covering i) the Eastern English Channel (EEC, January 2015) through IBTS (International Bottom Trawl Survey, doi: 10.17600/15001500), ii) the Northern Bay of Biscay (BoB, fall 2014) through EVHOE (EValuation des ressources Halieutiques de l'Ouest Européen, doi: 10.17600/14002000) and iii) the Gulf of Lions (GoL, May 2015) through PELMED (PELagiques MEDiterranée, doi: 10.17600/15006400) and in June 2015 through MEDITS (MEDiterranean International Trawl Survey, doi: 10.17600/15006300, Fig. 1). Survey features were detailed in Baudrier et al. (2018).

The EEC is an epicontinental mesotrophic system characterised by low depths (<50 m in the studied area), making local food webs to be more likely based on benthic sources (Cresson et al., 2020). It is also an area with many potential sources of contaminants from urbanised and industrialised catchment in both France and Great Britain (including the Seine River), and intensive marine traffic (Tappin and Millward, 2015, Aksoyoglu et al., 2016). The BoB is an open system with local food webs likely to be mainly based on pelagic sources (Cresson et al., 2020), and with multiple catchment inputs e.g. the major rivers the Loire (Boutier et al., 1993, Couture et al., 2010) and the Garonne which ends in the Gironde estuary (Schäfer and Blanc, 2002, Schäfer et al., 2022) and several smaller ones (e.g. Adour River, Sharif et al., 2014, Mille et al., 2021). Finally, the GoL belongs to the semi-enclosed and oligotrophic Mediterranean Sea, whose particularity is to shelter smaller individuals with slower growth rates than in Atlantic systems for similar species (Cossa et al., 2012, Chouvelon et al., 2018, Cossa et al., 2022). Moreover, some chemical inputs from the Rhône River into the GoL may be reflected in fish from this area (e.g. PCB, Cresson et al., 2015). The main characteristics of the three studied regions are summarised Table SM 1.

Contaminants were analysed in a total of 233 individuals or groups of individuals (mackerel in the GoL were grouped by 2 to 4). For 189 of them, carbon (C) and nitrogen (N) stable isotopes were also analysed, and these data were partially published in Cresson et al. (2020).

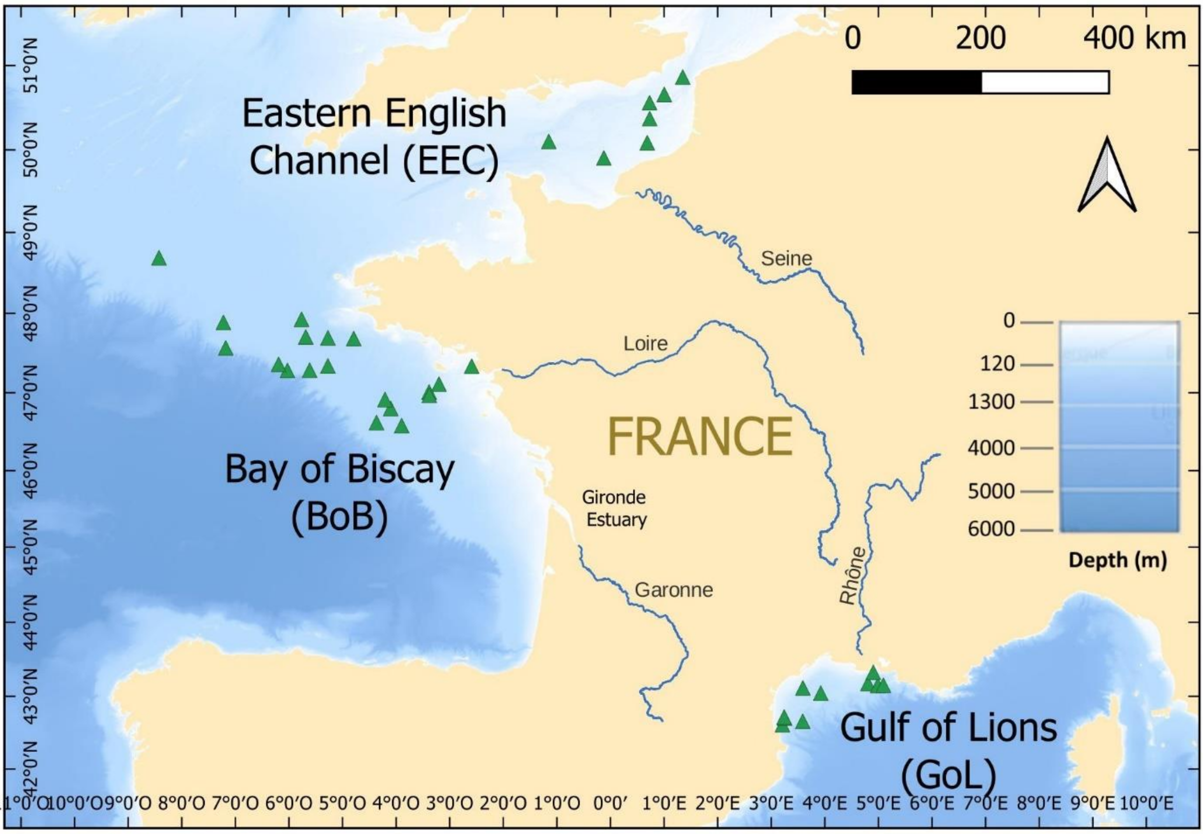


Fig. 1. Sampling stations (green triangles) in the three studied regions.

Eight fish species were collected, including seven teleost and one chondrichthyan species (Table 1). The sampling strategy was chosen according to regional context. In the EEC, individuals from 5 species were sampled: one pelagic piscivore (Atlantic mackerel *Scomber scombrus*), two demersal piscivores (whiting *Merlangius merlangus* and Atlantic cod *Gadus morhua*), one benthic invertebrate feeder (European plaice *Pleuronectes platessa*) and one demersal invertebrate feeder (lesser-spotted dogfish *Scyliorhinus canicula*). In the BoB, individuals from 5 species were sampled: two zooplankton feeders (blue whiting *Micromesistius poutassou* and European sardine *Sardina pilchardus*), two piscivores (one pelagic: the mackerel *S. scombrus* and one demersal: European hake *Merluccius merluccius*) and one demersal invertebrate feeder (lesser-spotted dogfish *S. canicula*). In the GoL, the sampling focused on two piscivores, one pelagic (mackerel *S. scombrus*) and one demersal (hake *M. merluccius*).

After sampling, fishes were directly stored at -20°C on board. In the laboratory, they were measured (total length to the nearest cm), weighed (total weight to the nearest g) and a large piece of fish dorsal muscle was collected under clean conditions, *i.e.* under fume hoods, using clean material (rinsed with ethanol) and in calcined glass (no plastics), then stored frozen, freeze-dried and ground into a fine powder. Three subsamples of the homogenised muscle were then shipped to 1) ANSES (Maisons Alfort, France) for trace element determination; 2) LABERCA (Nantes, France) for organic contaminants determination and to 3) LIENSs (La Rochelle, France) for C and N stable isotope measurements. In the present study, contaminant concentrations were measured in the fish muscle, which is the most consumed fish tissue (to mutualize analysis with human health assessment), and one of the most important fish tissues in quantity allowing quantification of various contaminants at trace and ultra-trace levels.

2.2. Trace element determination

Arsenic, Cd, Hg and Pb contents were measured in fish muscle samples using an ISO 17025 accredited method (French Accreditation Committee, COFRAC) described by Noel et al. (2005). The method is described in detail (including the accuracy and precision) in the Supplementary material. Briefly, 0.3 to 0.4 g of each sample was digested in a closed microwave system using 3mL of HNO_3. Sample digests were then diluted up to 50 mL with ultra-pure water and then analysed by inductively coupled plasma-mass spectrometry (ICP-MS) on the same day. Mean limits of detection and quantification (LOD/LOQ), calculated on a wet-weight basis (ww), were 1.2/4.1 µg/kg for both As and Hg, 0.2/0.8 µg/kg for Cd and 0.7/2.4 µg/kg for Pb.

2.3. Organic contaminant determination

Dioxin (17 PCDD/F congeners) and PCB (12 dioxin-like PCB (DL-PCBs) and 6 non-dioxin-like (NDL-PCB) congeners) were determined in fish muscle using an accredited method (ISO/IEC 17025:2005 standard) described by Vaccher et al. (2018). Briefly, extraction was carried out using a mixture of toluene and acetone (70:30, v/v). Purification and fractionation of PCDD/Fs and PCBs were carried-out on an automated system. Analysis of cleaned-up extracts was conducted by gas chromatography coupled to a double electromagnetic sector high resolution mass spectrometer. Lipid content was determined gravimetrically. For PCBs and dioxins, identification criteria and quantification fulfil the quality assurance and quality control (QA/QC) criteria recommended by EU (2017) and are described in the Supplementary Material with a detailed description of the method. LOD were between 0.003 and 0.017 ng/kg ww for PCDD/Fs from tetra- to octa-chlorinated substituted congeners, and 0.02 ng/kg ww for PCBs. Values were automatically corrected considering the recovery yield of the ^{13}C-labelled internal standards.

Toxic equivalent (TEQ) values for DL-PCBs and PCDD/Fs were calculated using toxic equivalent factors (TEF) attributed to the 17 dioxin congeners and 12 DL-PCB congeners according to their relative toxicity compared to the most toxic ones, *i.e.* TCDD (2,3,7,8-TCDD) and PeCDD (1,2,3,7,8-PeCDD) which are attributed a TEF=1 according to Van den Berg et al. (2006). In the present study, “NDL-PCBs” refers to the sum of these 6 main non dioxin-like congeners (CB28, CB52, CB101, CB138, CB153 and CB180) unless it is clearly indicated that it refers to congeners separately (*e.g.* “NDL-PCB congeners”).

2.4. Stable isotope analyses and trophic level calculation

Carbon and N stable isotopes were measured in fish muscle using a Flash EA 2000 elemental analyzer equipped with the Smart EA option (Thermo Scientific, Milan, Italy), coupled with a

Delta V Plus isotope ratio mass spectrometer with a Conflo IV interface (Thermo Scientific, Bremen, Germany). Individual stable isotopes data were used to assess individual trophic levels in fish (TL_{fish}) according to the following equation:

$$TL_{fish} = \frac{\delta^{15}N_{fish} - [(\alpha\delta^{15}N_{pelagic\ baseline}) + (1-\alpha)\delta^{15}N_{benthic\ baseline}]}{TDF} + TL_{baseline}\text{, where}$$

- $\delta^{15}N$: value of each individual;
- $\delta^{15}N_{pelagic\ baseline}$ and $\delta^{15}N_{benthic\ baseline}$: average $\delta^{15}N$ values for zooplankton and bivalves considered as baseline proxies of the pelagic and benthic trophic pathways, respectively;
- $TL_{baseline}$: trophic level of the baseline, set to 2 for both zooplankton and bivalves;
- TDF: trophic discrimination factors. Two TDF were applied: 3.4 for teleosts, and 2.3 for chondrichthyans, to account for their probable different N metabolism (Hussey et al., 2010, Logan and Lutcavage, 2010);
- α: the average pelagic contribution calculated for each studied species and region by Cresson et al. (2020) (Table 1).

2.5. Statistics

Data treatment was performed using R software (version 4.1.0, 2021-05-18, R Core Team 2021). Concentrations <LOD and <LOQ were substituted by LOD/2 and LOQ/2, respectively. Substitution of "less-than" values is generally considered more bias-prone than modern and theoretically sound approaches (*e.g.* maximum likelihood estimation, robust regression on order statistics, Helsel (2006)). However, this is not necessarily the case especially when percentage of "less-than" is moderate (e.g. George et al., 2021), and modelling approaches do not directly apply to mixtures such as TEQ. On a general basis, data distribution was visually verified to check for evident bias induced by data treatment. Individual contaminant concentrations are converted from dry-weight (dw) to wet-weight (ww) and from ww to lipid-weight (lp) with the sample percentage of humidity and lipid content, respectively. Contaminant concentrations and lipid contents were log-transformed prior to analysis to bring their distribution closer to normality. Normality and homoscedasticity conditions were assessed using Shapiro Wilks and Breusch – Pagan tests, respectively.

Trophic levels, body lengths, lipid contents, and contaminant concentrations were compared among species or regions using ANOVA and Tukey HSD as a post hoc test when normality and homoscedasticity conditions were met (lm() of the stats package (R basis) and tukey_hsd() of the rstatix package (Kassambara, 2021)). Otherwise, groups were compared using Kruskal Wallis and Dunn as post hoc test when more than 2 groups were compared, or Wilcoxon test when comparing hake and mackerel in the GoL (dunn_test() and wilcox.test() of the rstatix package, $p<0.05$).

Linear regressions and Pearson correlations (stat_cor() of the ggpubr package (Kassambara, 2020)) were assessed between i) trophic level or lipid content (for organic contaminants), and ii) contaminant concentrations, in each region by combining all the teleost individuals (multispecies approach). At similar trophic level or lipid content, contaminant concentrations were considered similar among regions if their 95% confidence intervals overlapped.

Relationships between individual contaminant concentrations (not as a sum) and biometric variables (body length, trophic level and lipid content) in the three regions for the three species sampled in more than one region (dogfish, hake and mackerel) were assessed by redundancy analysis ordination (rda() function of vegan package (Oksanen et al., 2020)), using contaminant concentrations in fish muscle as response variables and biometric variables as explanatory variables. Only congeners quantified in >70% of the samples were included in the analysis. Scaling 2 is represented so that the angles between response and explanatory variables in the biplot reflect their linear correlations.

3. RESULTS

3.1. Differences in trophic levels, lipid contents and contaminant concentrations among species in each region

Trophic levels and lipid contents. Trophic levels of the studied individuals ranged from 3.1 to 5.8 depending on the species and region (Table 1). In the EEC, individual trophic levels were similar for plaice and mackerel (mean: 3.6 and 3.8, respectively), and were both lower than for whiting, cod and dogfish (mean from 4.3 to 4.4). In the BoB, trophic levels of dogfish (4.8) were significantly higher than hake (3.9), which were also higher than sardine, mackerel and blue whiting (mean from 3.5 to 3.7). In the GoL, hake and mackerel had similar trophic levels (3.8, Table 1), as opposed to what was observed in the BoB. In each region dogfish was amongst the species with the highest trophic levels and mackerel among those with the lowest ones. These results are consistent with results of a previous study dedicated to trophic ecology of the three fish communities (Cresson et al., 2020). Trophic level could thus be used as a covariable, to compare and explain levels of biomagnifiable contaminants between ecosystems.

Lipid content in the studied individuals ranged from 0.3 to 19.2% depending on the species and region (Table 1). In each region, mackerel presented the highest lipid contents (Table 1), together with sardine in the BoB (no sardine sampled in EEC and GoL). Intra- and inter-species variability in lipid content is typical and reflects different energy storage strategies as discussed in the literature (e.g. Vollenweider et al., 2011). In the present study, lipid content is used as a covariable, to compare and explain levels of lipophilic contaminants between ecosystems.

Trace elements. Arsenic and Hg were quantified (*i.e.* > LOQ) in all the samples. Lead and Cd were quantified in 6 to 100% of the samples depending on the species. In both the EEC and BoB, trace element concentrations were higher in chondrichthyan than in teleost species, by up to 34 times for As (mean: 1.3 and 44.6 mg/kg ww in EEC mackerel and dogfish, respectively, Table 2, Fig. 2), 28 times for Cd (1.0 and 28.2 µg/kg ww in BoB hake and dogfish, respectively), and 8 times for Hg (80 and 364 µg/kg ww in BoB plaice and dogfish, respectively). Dogfish were also amongst the individuals with the highest Pb concentrations (Table 2). In the EEC, As concentrations in teleosts were *ca*. 2 to 3 times higher in benthic feeders (plaice: 15.4 mg/kg ww) than in demersal piscivores (cod: 8.6 mg/kg ww and whiting: 4.3 mg/kg ww), and *ca.* 12 times higher than in pelagic piscivores (mackerel: 1.3 mg/kg ww, Table 2). Cadmium and Hg concentrations were similar in the four studied species (cod, mackerel, plaice, whiting, Table 2) while Pb quantification level was higher in two of the benthic/demersal fishes (75% in plaice and 60% in cod) than in mackerel and whiting (<20%). In the BoB, As and Pb concentrations were higher in zooplankton feeders (blue whiting for As: 5.5 mg/kg ww, sardine for Pb: 25 µg/kg ww) than in piscivores (2.8 and 1.4 mg/kg ww for As in hake and mackerel, 1.8 and 2.9 µg/kg ww for Pb in hake and mackerel, respectively). In contrast, Hg concentrations were *ca*. twice higher in piscivores (24.3 and 24.6 mg/kg ww for hake and mackerel, respectively) than in zooplankton feeders (15.6 and 14.7 µg/kg ww in blue whiting and sardine, respectively). Cadmium concentrations were 3 to 5 times higher in pelagic species (from 3.0 to 5.5 µg/kg ww in blue whiting, mackerel, sardine) than in demersal ones (1.0 µg/kg ww in hake). In the GoL, As (3 times, 6.1 and 1.8 mg/kg ww in hake and mackerel, respectively) and Hg (5 times, 196 and 40 µg/kg ww) concentrations were higher in demersal (hake) than in pelagic piscivores (mackerel), while the opposite was observed for Cd (8 times, 0.2 and 1.6 µg/kg ww) and Pb (3 times, 1.3 and 3.4 µg/kg ww).

Organic contaminants. NDL-PCB congeners were quantified in all the samples. Quantification rate of each DL-PCB and PCDD/F congener is presented in the Supplementary Material. Mackerel and sardine, which showed the highest lipid content, also showed the highest NDL-PCB and TEQ concentrations on a wet-weight (ww) basis (Table 2, Fig. 2). When normalised to lipids, NDL-PCB and TEQ concentrations were similar in most species in the EEC and BoB, with the noticeable exceptions of dogfish and one zooplankton feeder (blue whiting in BoB) which showed lower concentrations. In the GoL, mackerel showed similar TEQ but lower NDL-PCB concentrations compared to hake.

Table 1. Trophic characteristics, total length and lipid content of the species sampled in the Eastern English Channel (EEC), Bay of Biscay (BoB) and Gulf of Lions (GoL) in 2014/2015. *Different grouping letters indicate significant differences among A: species within each region and B: regions for species sampled in more than one region (bold letters indicate groups with the highest levels).*

	Trophic group[1]	Pelagic contrib. [1]	n	**Trophic level**				**Total length (mm)**				**Lipid content (%)**			
				Mean	*Min*	*Max*		**Mean**	*Min*	*Max*		**Mean**	*Min*	*Max*	
A: Values and differences between fish species within each region															
Eastern English Channel (EEC, fishery campaign: IBTS)							[2]				[3]				[3]
Cod (*Gadus morhua*)	Demersal piscivore	0.40	5	**4.35**	*4.13*	*4.52*	**b**	**368**	*286*	*409*	bc	**0.67**	*0.46*	*0.86*	b
Dogfish (*Scyliorhinus canicular*)	Demersal invertebrate feeder	0.43	13	**4.43**	*4.07*	*5.29*	**b**	**526**	*389*	*634*	**c**	**1.57**	*1.23*	*2.01*	d
Mackerel (*Scomber scombrus*)	Pelagic piscivore	0.63	16	**3.80**	*3.24*	*4.41*	a	**333**	*288*	*387*	ab	**8.08**	*3.36*	*12.1*	**e**
Plaice (*Pleuronectes platessa*)	Benthic invertebrate feeder	0.34	12	**3.64**	*3.12*	*4.21*	a	**299**	*256*	*344*	a	**0.94**	*0.46*	*1.24*	c
Whiting (*Merlangius merlangus*)	Demersal piscivore	0.49	16	**4.34**	*4.05*	*4.57*	**b**	**314**	*257*	*382*	ab	**0.53**	*0.27*	*0.63*	a
Bay of Biscay (BoB, fishery campaign: EVHOE)							[2]				[3]				[3]
Blue whiting (*Micromesistius poutassou*)	Zooplankton feeders	0.96	33	**3.51**	*3.15*	*3.81*	a	**166**	*150*	*180*	b	**1.59**	*0.62*	*3.09*	b
Dogfish (*Scyliorhinus canicular*)	Demersal invertebrate feeder	0.89	26	**4.81**	*4.28*	*5.28*	**c**	**380**	*255*	*600*	**d**	**2.08**	*1.32*	*3.09*	c
Hake (*Merluccius merluccius*)	Demersal piscivore	0.80	39	**3.88**	*3.47*	*4.15*	b	**280**	*200*	*350*	c	**1.32**	*0.55*	*2.71*	a
Mackerel (*Scomber scombrus*)	Pelagic piscivore	0.80	20	**3.61**	*3.27*	*4.14*	a	**239**	*172*	*317*	c	**7.64**	*1.11*	*19.2*	**e**
Sardine (*Sardina pilchardus*)	Zooplankton feeders	0.82	12	**3.73**	*3.24*	*4.34*	ab	**111**	*90*	*143*	a	**3.76**	*1.53*	*6.92*	d
Gulf of Lions (GoL, fishery campaigns: MEDITS, PELMED)							[2]				[3]				[3]
Hake (*Merluccius merluccius*)	Demersal piscivore	0.95	24	**3.87**	*3.59*	*4.11*	a	**279**	*248*	*317*	**b**	**0.66**	*0.45*	*1.05*	a
Mackerel (*Scomber scombrus*)	Pelagic piscivore	0.96	17	**3.79**	*3.59*	*4.00*	a	**186**	*127*	*234*	a	**1.11**	*0.44*	*2.41*	**b**
B: Differences between regions for 3 fish species [3]															
				EEC	**BoB**	**GoL**		**EEC**	**BoB**	**GoL**		**EEC**	**BoB**	**GoL**	
Dogfish	Demersal invertebrate feeder		39	a	**b**			**b**	a			a	**b**		
Hake	Demersal piscivore		63		a	a			a	a			**b**	a	
Mackerel	Pelagic piscivore		53	a	a	a		**c**	b	a		**b**	**b**	a	

[1]: from Cresson et al 2020: the pelagic contribution is the proportion of pelagic source in one species network as estimated by the mixing model employed in Cresson et al 2020.
[2]: differences assessed by ANOVA and Tukey HSD as post hoc test ($p < 0.05$). [3]: differences assessed by Kruskal Wallis and Dunn as post hoc test or Wilcoxon test in GoL (2 species, $p < 0.05$). Boxplots corresponding to this panel (B) are given in Supplementary Material (Fig. SM 1) to visualise the data.

Table 2. Concentrations of contaminants in fish from the EEC, BoB and GoL sampled in 2014/2015. *Different grouping letters indicate significant differences among A: species within each region, B: regions for species sampled in more than one region (bold letters indicate the group with the highest concentrations). Organic contaminants are compared based on both wet-weight (ww) and lipid-weight (lp) basis in A and only in lp-basis in B.*

	n[1]	As (mg/kg ww) [2]				Cd (µg/kg ww)					Hg (µg/kg ww) [2]				Pb (µg/kg ww)					TEQ (pg TEQ/g ww)					NDL-PCB (ng/g ww)[2]				
A: Concentrations and differences between fish species within each region																													
		Mean	*Min*	*Max*		**Mean**	*Min*	*Max*	*%Q*[3]		**Mean**	*Min*	*Max*		**Mean**	*Min*	*Max*	*%Q*[3]		**Mean**	*Min*	*Max*	**ww**	**lp**	**Mean**	*Min*	*Max*	**ww**	**lp**
Eastern English Channel (EEC)					[4]					[5]				[5]					[5]				[4]	[4]				[4]	[4]
Cod	5	**8.6**	*1.6*	*27.6*	b	**0.5**	*0.1*	*1.7*	*20%*	a	**89.3**	*77.3*	*107.4*	a	**43**	*0.4*	*202*	*60%*	ab	**0.39**	*0.22*	*0.64*	c	**b**	**2.10**	*1.17*	*3.71*	a	**b**
Dogfish	13	**44.6**	*17.5*	*71.4*	**d**	**3.0**	*0.4*	*5.8*	*92%*	**b**	**364**	*259*	*666*	**b**	**3.5**	*0.4*	*9.0*	*62%*	ab	**0.05**	*0.03*	*0.07*	a	a	**1.06**	*0.41*	*2.07*	a	a
Mackerel	16	**1.34**	*0.13*	*3.82*	a	**0.4**	*0.1*	*1.4*	*19%*	a	**119**	*40*	*243*	a	**14**	*0.4*	*212*	*13%*	[6]	**3.73**	*1.48*	*9.05*	**d**	**b**	**30.6**	*8.0*	*88.9*	**b**	**b**
Plaice	12	**15.4**	*6.9*	*27.7*	c	**0.6**	*0.1*	*2.7*	*25%*	a	**89.6**	*31.4*	*259.3*	a	**8.6**	*1.2*	*27.9*	*75%*	**b**	**0.49**	*0.15*	*1.13*	c	**b**	**2.39**	*0.54*	*7.63*	a	**b**
Whiting	16	**4.83**	*1.98*	*8.85*	b	**0.3**	*0.1*	*3.5*	*6%*	a	**79.7**	*56.0*	*135.6*	a	**1.8**	*0.4*	*9.8*	*19%*	a	**0.17**	*0.10*	*0.31*	b	**b**	**1.15**	*0.44*	*2.80*	a	**b**
Bay of Biscay (BoB)					[4]					[5]				[5]					[5]				[4]	[4]				[4]	[4]
Blue whiting	33 (19/16)	**5.46**	*3.03*	*9.01*	c	**5.5**	*0.41*	*17.6*	*100%*	b	**15.6**	*10.0*	*44.6*	a	**6.4**	*1.2*	*35.9*	*53%*	b	**0.08**	*0.04*	*0.22*	a	a	**0.46**	*0.15*	*1.26*	a	a
Dogfish	26 (13/13)	**11.0**	*6.6*	*14.4*	**d**	**28.2**	*3.6*	*81.7*	*100%*	**c**	**117**	*75*	*189*	**d**	**21**	*1*	*54*	*92%*	**c**	**0.12**	*0.07*	*0.32*	a	a	**0.60**	*0.31*	*1.63*	ab	a
Hake	39 (19/20)	**2.80**	*2.02*	*4.64*	b	**1.0**	*0.1*	*2.8*	*42%*	a	**24.3**	*16.0*	*36.2*	c	**2.9**	*0.3*	*13.6*	*37%*	ab	**0.15**	*0.05*	*0.88*	a	ab	**1.67**	*0.31*	*11.38*	b	**b**
Mackerel	20	**1.43**	*0.80*	*2.52*	a	**3.0**	*0.4*	*9.9*	*95%*	b	**24.6**	*10.6*	*58.5*	bc	**1.8**	*0.4*	*9.4*	*30%*	a	**0.90**	*0.07*	*5.22*	**b**	**bc**	**7.16**	*0.51*	*61.66*	**c**	**b**
Sardine	12	**2.26**	*1.61*	*2.99*	b	**3.7**	*1.1*	*10.3*	*100%*	b	**14.7**	*7.5*	*23.5*	ab	**25**	*7*	*91*	*100%*	**c**	**0.63**	*0.28*	*1.08*	**b**	**c**	**5.41**	*1.50*	*10.68*	**c**	**b**
Gulf of Lions (GoL)					[4]					[5]				[4]					[5]				[4]	[4]				[4]	[4]
Hake	24	**6.14**	*3.01*	*8.53*	**b**	**0.2**	*0.1*	*0.9*	*8%*	a	**196**	*88*	*356*	**b**	**1.3**	*0.4*	*5.5*	*17%*	a	**0.32**	*0.11*	*0.75*	a	a	**9.05**	*2.79*	*19.20*	a	**b**
Mackerel	17	**1.81**	*1.18*	*2.80*	a	**1.6**	*0.1*	*8.7*	*53%*	**b**	**40.1**	*18.6*	*94.0*	a	**3.4**	*0.4*	*13.8*	*53%*	**b**	**0.59**	*0.31*	*0.94*	**b**	a	**8.19**	*4.56*	*15.37*	a	a

B: Differences between regions for 3 fish species (metals in ww, organic in lp) [5]

	As EEC	As BoB	As GoL	Cd EEC	Cd BoB	Cd GoL	Hg EEC	Hg BoB	Hg GoL	Pb EEC	Pb BoB	Pb GoL	TEQ EEC	TEQ BoB	TEQ GoL	NDL-PCB EEC	NDL-PCB BoB	NDL-PCB GoL
Dogfish	**b**	a		a	**b**		**b**	a		a	**b**		a	**b**		**b**	a	
Hake		a	**b**		**b**	a		a	**b**		**b**	a		a	**b**		a	**b**
Mackerel	a	a	a	a	**c**	a	**c**	a	b	[6]	[6]	[6]	**b**	a	**b**	**c**	a	**b**

[1]: Total number of samples analysed for contaminants (trace elements/organics analysis are detailed when they differed).
[2]: As, Hg and NDL-PCB (*i.e.* Sum of 6 NDL-PCB congeners CB28, CB52, CB101, CB138, CB153 and CB180) are quantified in 100 % of the samples;
[3]: % quantified samples (>LOQ);
[4]: differences assessed by ANOVA and Tukey HSD as post hoc test ($p < 0.05$);
[5]: differences assessed by Kruskal Wallis and Dunn as post hoc test or Wilcoxon test in GoL (2 species, $p < 0.05$). Boxplots corresponding to this panel (B) are given in Supplementary Material (Fig. SM 1) to visualise the data;
[6]: Low quantification and one unexplained Pb outliers in the BoB hampered comparison between regions for mackerel.

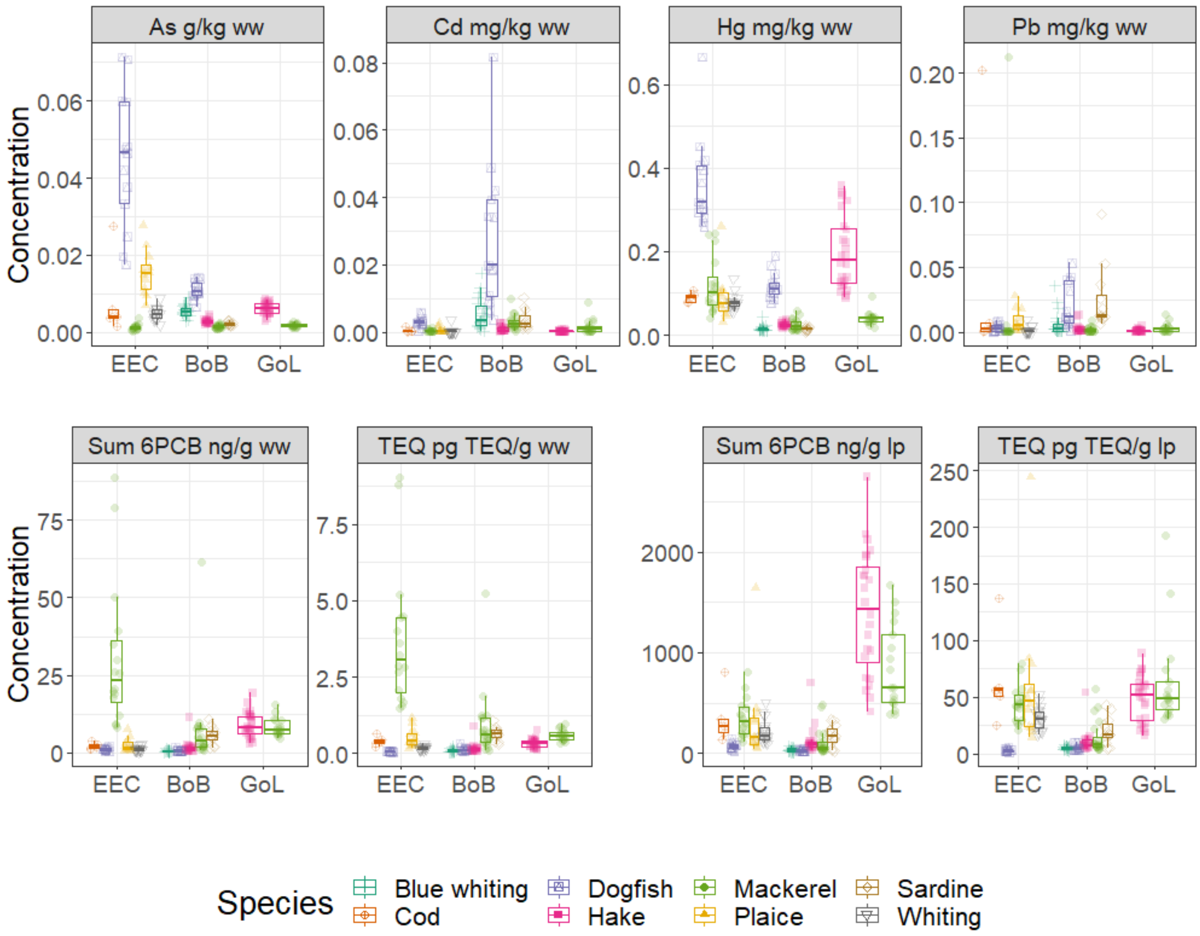


Fig. 2. Concentrations of contaminants in each species by region. *Significant differences between species and regions are given in Table 2.*

3.2. Contaminant relationships with fish trophic level or lipid content in each region

Data selection for the correlation analysis. Correlations between contaminant concentrations and trophic level or lipid content (for organic contaminants) were assessed and discussed in teleost species in both the EEC and BoB. The GoL dataset is presented in Fig. 3 to display differences in contaminant concentrations among regions for fishes at similar trophic level or lipid content. Correlations assessed in chondrichthyans are presented in Fig. SM 2 solely for information. However, datasets corresponding to teleosts in the GoL and to chondrichthyans in the three studied regions were not suited for robust correlation analysis as the trophic level and lipid content ranges were narrow (Table 1), potentially leading to a high risk of false non-significant correlations. Correlations for teleosts in the GoL and for chondrichthyans in each region are therefore not discussed in the present study.

Trace elements. Trophic levels of teleost individuals in the EEC and BoB varied by 1.4 and 1.2 TL, respectively (Table 1). Arsenic, Cd and Pb concentrations in teleosts varied by at least one order of magnitude in each region, and slightly less than one order of magnitude for Hg (Table 2). Fish concentrations in the four trace elements in the EEC, as well as of As and Pb in the BoB, were not correlated with fish trophic level ($p>0.05$, Fig. 3). In the BoB, Cd and Hg concentrations significantly decreased and increased with fish trophic level, respectively ($r^2 < 0.33$, Fig. 3).

Organic contaminants. NDL-PCB and TEQ concentrations in individuals varied by *ca.* 2 orders of magnitude in each region, and lipid contents by more than one order of magnitude (Table 1 and Table 2). NDL-PCB and TEQ concentrations were correlated with fish muscle lipid content, which explained 35 to 84% of the variability (Fig. 3). Once normalised to lipids, NDL-PCB and TEQ concentrations were not correlated with trophic levels in the EEC (p>0.05). In the BoB, NDL-PCB and TEQ concentrations increased with fish trophic levels which explained 26 and 32% of the NDL-PCB and TEQ variability, respectively (p<0.05).

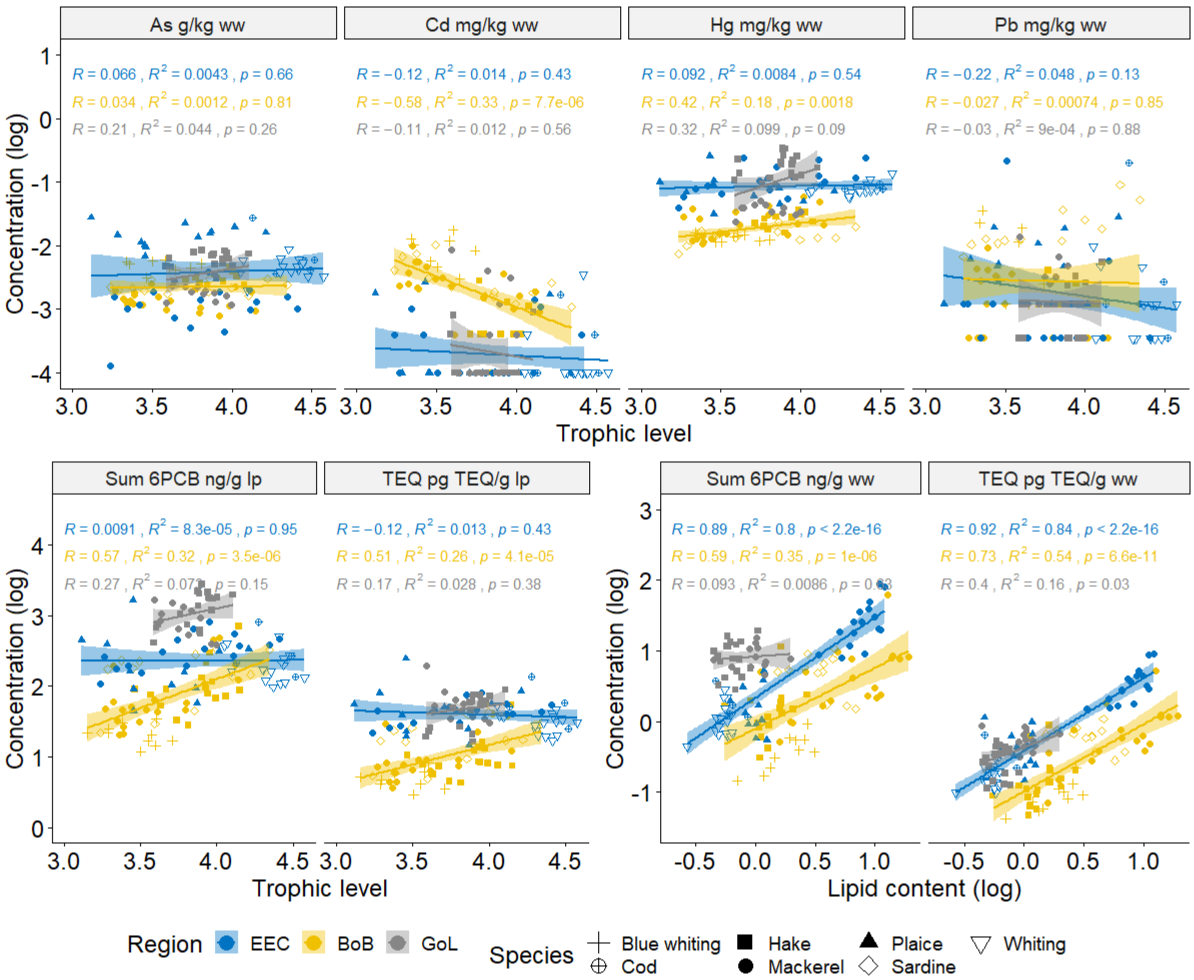


Fig. 3. Linear regressions (Pearson correlation (R), correlation coefficient (R^2), and p-value (p)) between contaminant concentrations and trophic levels or lipid contents (for organic contaminants) for teleost fish in each region.

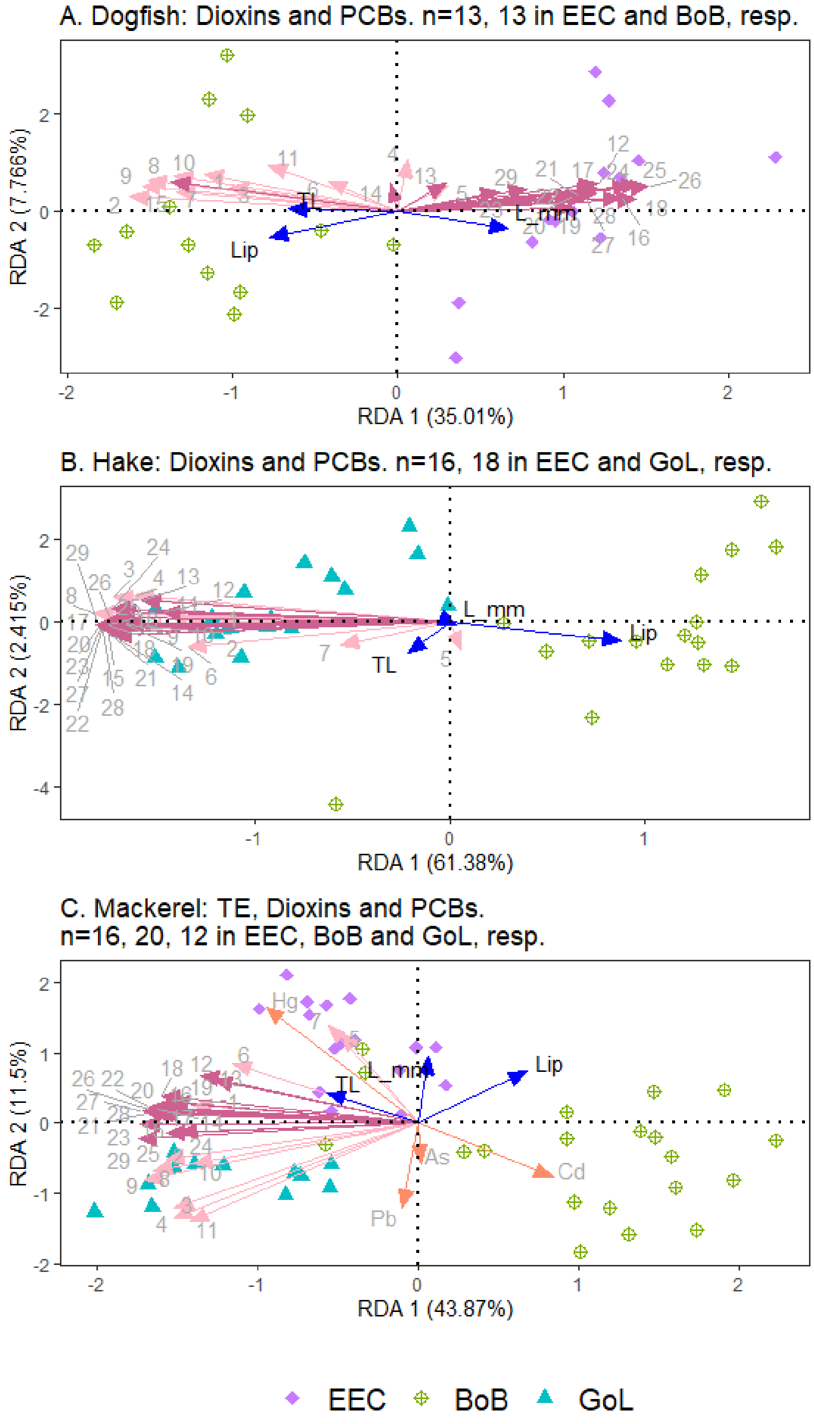
3.3. Differences among regions

Dogfish from the BoB had higher trophic levels and lipid contents but were smaller than the individuals sampled in EEC. Their concentrations in Cd and Pb were also higher in the BoB than in EEC (Cd: 9 times; 3 and 28 µg/kg ww in the EEC and BoB, respectively; Pb: 6 times; 3.5 and 21 µg/kg ww). In contrast, concentrations in As, Hg and NDL-PCB were higher in individuals from the EEC (As: 4 times; 45 and 11 mg/kg ww in the EEC and BoB, respectively; Hg: 3 times; 364 and 117 µg/kg ww; NDL-PCBs: 2 times both in ww- and lp-basis: 1.06 and 0.60 ng/g ww or 0.69 and 0.29 ng/g lp, Table 1). TEQ concentrations in dogfish were higher in the BoB than in EEC in ww (2 times: 0.05 and 0.12 pg TEQ/g ww in the EEC and BoB, respectively) but were higher in EEC in lp (2 times: 0.037 and 0.006 pg TEQ/g ww in the EEC and BoB, respectively). Concentrations in most DL- and NDL-PCB congeners (16 quantified in >70% of the samples out of 18) were higher in individuals from the EEC, while concentrations in 10 out of 11 PCDD/F congeners were higher in the BoB (Fig. 4).

Hake showed higher lipid contents in the BoB than in GoL, but they had similar trophic levels and lengths. Their Cd and Pb concentrations were also higher in the BoB (Cd: 5 times; 1 and 0.2 µg/kg ww in the BoB and GoL, respectively; Pb: 2 times; 2.9 and 1.3 µg/kg ww), while As, Hg, NDL-PCBs and TEQ in lp-basis concentrations were higher in individuals from the GoL (As: 2 times; 2.8 and 6.14 mg/kg ww; Hg: 8 times; 24 and 196 µg/kg ww; NDL-PCBs: 12 times; 1.2 and 14.0 ng/g lp; TEQ: 4 times; 0.11 and 0.48 pg/g lp, Table 1). Concentrations in PCB and TEQ in ww showed a similar pattern as in lp but with slightly lower differences between the two regions. At the congener level, hake concentrations in the 29 PCB and dioxin congeners were all highly correlated and higher in the GoL (lp-basis) while their lipid contents were twice twice as high in BoB (Fig. 4).

Mackerel had similar trophic levels in the three regions but they had lower lipid contents and were smaller in the GoL than in EEC and BoB (Table 1). Their As concentrations were similar in the three regions and ranged from 1.3 to 1.8 mg/kg ww (Table 2). Mercury concentrations in mackerel in the EEC were 5 and 3 times higher than in BoB and GoL, respectively (119, 25 and 40 µg/kg ww in the EEC, BoB and GoL, respectively Table 2). Their Cd concentrations in the BoB were 8 and 2 times higher than in EEC and GoL, respectively (0.4, 3.0 and 1.6 µg/kg ww in the EEC, BoB and GoL, respectively Table 2). The 18 PCB congener concentrations were higher in mackerel from both the GoL and EEC than those from BoB, and were anti-correlated with individual lipid contents at the inter-regional level (Fig. 4). Concentrations in some highly chlorinated PCDD/F congeners were markedly higher in the GoL (3 hepta- and octa-chlorinated congeners at the bottom left panel of the RDA, Fig. 4), and some slightly chlorinated congeners were markedly higher in the BoB (1 tetra- and 1 penta-PCDF on the top left panel of the RDA).

The general pattern observed at the species level (dogfish, hake or mackerel, Table 1, Table 2, Fig. SM 1) or at the multispecies level (Fig. 3) suggested that fishes in the BoB showed lower Hg, NDL-PCB and TEQ concentrations and higher Cd and Pb concentrations than in EEC and GoL. No clear pattern for As was observed (Fig. 3). In hake and to a lesser extent in mackerel, PCB and PCDD/F congeners were all correlated. In dogfish however, PCDD/F congener concentrations were higher in the BoB than EEC, while the opposite was observed for PCB congeners (EEC>BoB, Fig. 4). Finally, although organic contaminants were highly positively correlated with lipid content at the regional level (Fig. 3), they were anti-correlated with lipid content at the inter-regional level (Fig. 4).



PCB and Dioxins in the RDA are quantified in > 70% of the samples: 1: 1,2,3,7,8-PeCDD, 2: 1,2,3,6,7,8-HxCDD, 3: 1,2,3,4,6,7,8-HpCDD, 4: OCDD, 5: TCDF, 6: 1,2,3,7,8-PeCDF, 7: 2,3,4,7,8-PeCDF, 8: 1,2,3,4,7,8-HxCDF, 9: 1,2,3,6,7,8-HxCDF2, 10: 2,3,4,6,7,8-HxCDF, 11: 1,2,3,4,6,7,8-HpCDF, 12: CB77, 13: CB81, 14: CB126, 15: CB169, 16: CB105, 17: CB114, 18: CB118, 19: CB123, 20: CB156, 21: CB157 22: CB167, 23: CB189, 24: CB28, 25: CB52, 26: CB101, 27: CB138, 28: CB153, 29: CB180.

Fig. 4. Redundancy analysis ordination diagram with contaminant concentrations in fish muscle as response variables, and biological drivers as explanatory variables for dogfish (A), hake (B) and mackerel (C). *Response variables are PCDD/Fs (in lp-basis, pink arrows), PCBs (DL and NDL, in lp-basis, hotpink arrows), trace elements (TE, in ww-basis, orange arrows) when analysed in the same individuals as the ones analysed for organic contaminants, i.e. for mackerel in C. Explanatory variables (blue arrows) are body length (L_mm), trophic level (TL) and lipid content (Lip). Percentage of variability explained by each axis is indicated in parenthesis.*

4. DISCUSSION

4.1. Are there species or regions more likely to be contaminated than others?

4.1.1. Teleost species or regions more likely to be contaminated by metals

Arsenic. In the EEC and GoL, As concentrations were higher in benthic and demersal fish than in pelagic piscivores, which is in line with As tendency to accumulate in sediment (Albuquerque et al., 2021). This pattern (benthic/demersal>pelagic) was only highlighted for piscivores (hake>mackerel) in the BoB. Arsenic concentrations did not increase with fish trophic level in the present study ($p>0.05$). They have been previously linked to trophic processes but the direction of this relationship is variable (Donadt et al., 2021). Arsenic biodilution or weak relationship with trophic descriptors were also previously reported, *e.g.* in US lakes (Revenga et al., 2012) or through a meta-analysis in the marine environment (Sun et al., 2020). In contrast, Lescord et al. (2020) suggested that As may biomagnify in freshwater food webs of boreal lakes and rivers. Zhang et al. (2016) reported that variations of As concentration among fish species could be attributed to 1) their prey type, composed of different proportions of inorganic As (the most toxic As species) and organic As (less toxic, which is also more bioavailable); and 2) fish ability to biotransform As to less toxic forms. Though As concentrations differed among regions for hake (EEC>BoB) and dogfish (GoL>BoB), it was similar among regions for mackerel and at the multispecies level. Therefore, no clear pattern as regards to the regional variability of As concentrations in fish was observed. Further explorations of As speciation and bioavailability are needed to assess general tendencies regarding its behaviour in marine fish.

Mercury. The preferential accumulation of Hg in benthic than in pelagic fish, its biomagnification in trophic networks and its generally higher concentrations in Mediterranean organisms than in their Atlantic counterparts (from sponges to fishes, marine mammals, and seabirds) has been reported previously (Cossa et al., 2012, Lavoie et al., 2013, Cresson et al., 2016, Chouvelon et al., 2018, Burke et al., 2020, Sun et al., 2020, Cossa et al., 2022). In the present study, these trends were also observed in some regions and for several species with some notable exceptions discussed below.

In the GoL, Hg concentrations were higher in demersal than pelagic piscivores. However, this pattern was not observed in the EEC and BoB (demersal=pelagic piscivores). In the BoB, Hg concentrations were higher in piscivores than in zooplankton feeders, and were positively correlated with trophic level, supporting its ability to biomagnify. However, in the EEC, Hg concentrations were similar among species and unrelated to fish trophic level. One of the EEC specificities is its limited depth, which might limit the possibility to of showing a clear distinction between benthic, demersal and pelagic networks that might all be connected (Cresson et al., 2020). In addition, the benthic contribution in the food web was higher in the EEC than in BoB or GoL. Yoshino et al. (2020) suggested that Hg bioaccumulation in fish was actually mainly an effect of the concentration of Hg at the base of the food web, and that the subsequent biomagnification was secondary, benthic food webs based on the microphytobenthos being the dominant pathway for Hg biomagnification; this would support the higher Hg concentration in the EEC. Madgett et al. (2021) showed on the coasts of Scotland that the relationship between Hg concentrations and trophic levels was significant only when sharks were considered. When only teleost and benthic invertebrates were considered, TMF even suggested a trophic dilution of Hg (TMF=0.9, $p<0.05$). One hypothesis might be that Hg concentrations, instead of linearly increasing (when log-transformed) in the trophic network, rather slowly increase or are stable for a limited range of trophic network sections and then suddenly jump to the next trophic level.

Mercury concentrations in hake were higher in the GoL than in BoB as previously reported (e.g. Cossa et al., 2012). Chouvelon et al. (2018) suggested that this would be mainly explained by the oligotrophic conditions of the Mediterranean Sea resulting in i) higher Hg bioavailability due to enhanced Hg methylation, ii) higher Hg concentrations at lower trophic levels (*e.g.* phytoplankton) because of their smaller size and less abundant density, and iii) lower biodilution of Hg body burden in organisms due to slower growth rate, as compared to mesotrophic environments. However, mackerel showed higher Hg concentrations in the EEC (supposedly mesotrophic) than in the GoL (more oligotrophic). There are few studies on the Hg contamination of fishes from the English Channel (e.g. Henry et al., 2017) and to the best of our knowledge, none compared the EEC with GoL or BoB. Since the benthic food web is a dominant pathway for Hg accumulation (Yoshino et al., 2020, Cossa et al., 2022), the higher benthic contribution for mackerel in the EEC than in GoL (37 versus 4%, Cresson et al. (2020), Table 1) might partially explain the higher Hg concentrations in mackerel from the EEC. One could suggest that mackerel sampled in the EEC might be older than those in the GoL because they are longer, and therefore would have accumulated higher Hg concentrations over their lifespan. However, because fish growth rate is expected to be slower in the GoL than in the Atlantic (Mellon-Duval et al., 2010, Cossa et al., 2012), mackerel age in both regions cannot be compared based on their individual length. In further studies, the age of the individuals should be recorded whenever possible.

Cadmium and Lead. Cadmium and Pb concentrations in fish were 1) higher in zooplankton feeders and decreased with trophic level in the BoB, 2) higher in individuals with the lowest trophic level (*i.e.* mackerel) in the GoL, in line with their biodilution in trophic network (Espejo et al., 2018, Gu et al., 2018, Madgett et al., 2021). Sun et al. (2020) however reported that Cd and Pb might biomagnify in the first levels of marine food web, especially from primary producers to primary consumers.

Cadmium and Pb concentrations were higher in the BoB than in both EEC and GoL. In the EEC, i) trophic status is more mesotrophic, ii) water depth is lower leading to higher chances for prey-predator interactions, iii) opportunity for terrestrial input to be diluted is lower as compared to an open system such as the BoB. Therefore, food resources could be expected to be higher in the EEC than BoB, resulting in higher growth rate and biodilution in EEC. However, the results were the opposite of what might be expected between the BoB and GoL when considering the mesotrophic status of the BoB, which would provide conditions for higher growth rate and biodilution than in the oligotrophic GoL. It appeared therefore that the trophic status of the system does not fully explain the spatial variability in Cd and Pb concentrations. Apart from the trophic status of the system, historical inland sources of Cd and Pb are known to be important. Over the BoB continental shelf, the hydrological structure has been shown to be mainly influenced by two main rivers, namely the Loire and Gironde (Planque et al., 2004, Puillat et al., 2004). One of the main inland sources of Cd may be the historical metallurgy-related Cd pollution of the Gironde estuary, with the major French open-pit coal mine located 400 km upstream on the Gironde River. In 1979, Cd concentrations in wild oysters at the Gironde estuary were *ca.* 100 mg/kg dw instead of 1–2 mg/kg dw in the nearby Arcachon lagoon (Boutier, 1981). An accidental metal spill occurred in 1986 led to the mine being closed, followed by over three decades of monitoring, clean-up and remediation actions (Schäfer and Blanc, 2002, Schäfer et al., 2022). The Loire estuary represents one of the main inland sources of Pb in the BoB, with the Octel-Kulhman chemical plant only 15 km upstream from the estuary that produced from 1938 to 1996 alkyl−lead added to gasoline used in France and other European countries (Boutier et al., 1993, Couture et al., 2010). The above cited references report that, even after decades, Cd and Pb contamination still impacts the chemical signature at the mouth of both rivers. More accurate models on contaminant fluxes from these rivers to offshore are needed to assess the actual potential Cd and Pb impact on the offshore community on the BoB continental shelf. On the offshore side of the BoB, Cd is enriched in the surface waters from upwelling regions near the shelf edge of the European continental margin relative to areas or periods of lower productivity (Cotté-Krief et al., 2002), and might also account for the higher Cd concentrations in fish from the BoB.

4.1.2. Teleost species or regions more likely to be contaminated by PCB and dioxins

PCBs and PCDD/Fs. Because of their hydrophobic properties, PCBs and PCDD/Fs are likely to accumulate in non-polar compartments. As a consequence, body burdens of PCBs and PCDD/Fs are higher in fat fish such as mackerel or sardine on a ww-basis. Lipid content indeed explained a large proportion of PCB and TEQ variability in the EEC and BoB. However, at the inter-regional level, PCB and PCDD/F congener concentrations were anticorrelated with the lipid content, mainly due to the fact that fish from the GoL show higher concentrations while being less fat than those from both the EEC and BoB.

Higher PCB concentrations in hake from the GoL compared to those from BoB have been previously reported (Bodiguel et al., 2009), which might be due to either i) the trophic status of the GoL providing less opportunity for biodilution as discussed above for Hg, Cd and Pb, or ii) higher environmental concentrations in Dioxins and PCBs in the GoL. In 2006, PCBs and especially DL-PCBs were found in high concentrations in fish from the Rhône River, resulting in a ban on fish consumption. Several major PCB sources have been identified along the Rhône River, *e.g.* 1) the PCB treatment facility upstream of Lyon city, which was authorised to release PCBs into the Rhône River, 2) the Lyon city and industrial corridor downstream of the city, 3) Rhône tributaries likely to be still releasing PCBs in the 2000s (Mourier et al., 2014). In the GoL, Rhône River outflows have been shown to play a major role in PCB contamination (Ruus et al., 2006) in sediments (Salvadó et al., 2013) and organisms (Harmelin-Vivien et al., 2012, Alekseenko et al., 2018). Atmospheric inputs and wastewater discharges, especially during periods of flooding, also play a role in PCB levels observed in fish from the GoL (Alekseenko et al., 2018).

Overall, in the EEC, our results suggested a limited influence of the fish habitat (pelagic, demersal, benthic), diet (zooplankton, piscivore) or trophic levels on NDL-PCB and TEQ concentrations (lp-basis). In the BoB, individual trophic levels explained up to 32% of the lipid-normalised NDL-PCB and TEQ variability. Previous studies reported PCB biomagnification in trophic networks (especially for the recalcitrant CB153 congener), *e.g.* in arctic networks (e.g. Fisk et al., 2001, Kelly et al., 2008), and in pelagic and demersal fish food webs in the BoB and GoL (Harmelin-Vivien et al., 2012, Romero-Romero et al., 2017, Castro-Jiménez et al., 2021). PCDD/Fs are however less prone to biomagnify. Higher hydrophobic PCDD/Fs have been shown to decline with increasing trophic level, which was explained by a reduced membrane permeability due to steric hindrance of larger molecular size compared to the lower chlorinated congeners (Ruus et al., 2006, Castro-Jiménez et al., 2021).

4.1.3. The specificity of dogfish

Dogfish showed i) higher trace element concentrations, ii) lower PCB and TEQ concentrations than teleost species, and iii) different PCB and PCDD/F profiles between the EEC and BoB, while PCB, PCDD/F congeners were all highly correlated in hake and mackerel. Higher trace element and PCB concentrations in chondrichthyans than in teleost species have been previously reported (Cresson et al., 2014, Cresson et al., 2016, Madgett et al., 2021). Jeffree et al. (2010) observed differences in radioactive trace element uptake and depuration rates between chondrichthyan and teleost taxa. They suggested that this was linked to differences in i) physiology and anatomy including dermal thickness, scale and skeletal structure, intestine morphology and function, osmoregulation, and growth rates (Helfman et al., 2009), and ii) phylogenetic and evolutionary divergence between chondrichthyans and teleosts, which occurred more than 500 million years ago.

4.2. Data normalisation for environmental status assessment under the MSFD (D8)

Normalisation. Lipid content explained a majority of NDL-PCB and TEQ variability in the present study, especially in the EEC and BoB. In biota, normalisation of lipophilic contaminant concentrations to lipids is typically used to enhance robustness in the identification of spatial or temporal trends in contamination. Fliedner et al. (2018) also suggested that normalisation to lipid can partly overcome discrepancies between contaminant concentrations in muscle and whole fish, which is especially important for comparison to tissue-specific thresholds. In our study, data were normalised to the total extractable lipid content. Normalisation to a more specific lipid fraction might be even more powerful. For instance, Kelly et al. (2008) normalised PCB and PBDE concentrations to lipid equivalent fraction. This approach recognizes that biological matrices with low lipid fractions (*e.g.* <1%) tend to store a significant fraction of lipophilic contaminant in a non-lipidic fraction of organic matter, but it requires knowing the protein and carbohydrate fractions of the sample.

Normalisation of contaminant concentrations that likely biomagnify (*e.g.* Hg and PCBs) to a common trophic level has been considered useful to harmonise data obtained in different species, and compensate for differences in sampling strategies. The European Water Framework Directive (WFD) recommends 4.5 to 5 (predatory fish) as a common trophic level so that the assessment would be sufficiently protective to top predators (EU, 2014). In our study, trophic levels explained ≤1% ($p>0.05$) and ≤33% ($p<0.05$) of the contaminant variability in teleost fish in the EEC and BoB, respectively. This suggested that normalisation to trophic level would have limited interest as each fish species already provided an equivalent level of protection close to the recommended one (individual trophic level from 3.1 to 5.2). Therefore, considering the difficulty of generalising TMF values for Hg and PCBs in marine systems (Walters et al., 2016, Fliedner et al., 2018, OSPAR, 2019) and the fact that one of the major sources of variation in one region was the taxon (*e.g.* present study), the development of taxa-specific thresholds might be a rather practical way forward for environmental assessment.

MSFD descriptor 8 assessment in 2018 (cycle 2). The present data was used to assess the good environmental status (GES) for D8 in 2018 in France (Mauffret et al., 2018) (Table SM 3). Percentile 95 of contaminant concentrations in each species from each region was compared to Environmental Assessment Criteria (EAC) developed by OSPAR for PCBs. Threshold exceedance was observed for CB118 in 6 out of the 8 species from all regions (mackerel, whiting, cod, plaice, hake and sardine, OSPAR EAC 25 µg/kg lp), as well as for several additional PCB congeners (CB52, CB101, CB138, CB180) in the GoL, in line with the high PCB concentrations in fish from this region. CB118 is the only one among OSPAR PCB common indicators with dioxin-like properties and therefore one of the most toxic.

An EQS (Environmental Quality Standard) for Hg (20 µg/kg ww) has been developed under the WFD. It is based on environmental risk linked to secondary poisoning and applies to whole fish. It was not used in the 2018 MSFD French assessment as it was still under revision by OSPAR prior to marine application. If conversion of Hg concentrations from muscle to whole fish and trophic adjustment are ignored in the absence of recognized conversion factors, the Hg EQS was exceeded in all the species and regions observed in the present survey (compared to percentile 95). This suggests that Hg is at levels giving rise to biological effects in French waters, as previously observed in freshwater (e.g. Fliedner et al., 2018) and marine environment (Madgett et al., 2021).

5. CONCLUSION

Within each of the three studied regions, As concentrations were higher in benthic and demersal piscivores in line with its tendency to accumulate in sediments. PCBs and PCDD/Fs (lipid-weight) concentrations were similar in most teleost species. In the EEC, Cd, Hg and Pb concentrations were similar in the four teleost species. In the BoB and GoL, the trophic group accumulating the highest Cd, Hg and Pb concentrations depended on the contaminant and the region.

Concentrations in Hg, PCBs and PCDD/Fs were higher in the GoL than in BoB in line with the oligotrophic status of the Mediterranean seas and inland sources of PCBs from the Rhône River. To the best of our knowledge, this is however the first study comparing fish contamination in the EEC with that in the GoL and BoB. Mercury concentrations were higher in mackerel from the EEC, where benthic contribution in the food web was higher, than from the GoL. Cadmium and Pb concentrations were highest in the BoB. A better comprehension of Cd and Pb geochemical cycles in the BoB is needed to confirm how local and oceanic sources could reach organisms from the BoB continental shelf. Further explorations of As speciation and bioavailability are needed to assess general tendencies regarding its behaviour in marine fish.

Lipid content explained from 35% to 84% of organic contaminant variability. Lipid normalisation was useful to enhance robustness in the identification of spatial patterns in contamination by lipophilic substances. Contamination patterns in chondrichthyan clearly differed from that in teleost fish. In the present study, individual trophic levels were significantly correlated to contaminant concentrations only for several substances, and only in the BoB. This does not suggest that trophic biomagnification or biodilution could not be observed with a larger dataset; however, it indicates that normalisation to a common trophic level might not be systemically relevant when the objective is to analyse spatial patterns based on the monitoring of one taxon (*e.g.* teleost fish). Development of taxa-specific thresholds might be a practical way forward to refine environmental assessment.

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Supplementary Material

Table SM 1. Characteristics and main contaminant sources of the three studied marine regions along the metropolitan French coast.

Marine region	Characteristics	Main contaminant sources
Eastern English Channel (EEC)	Epicontinental mesotrophic system Low depth Food webs more likely based on benthic sources	Urbanised and industrialised catchment in both France (e.g. Seine River) and Great Britain Intensive marine traffic
Northern Bay of Biscay (BoB)	System open to the Atlantic Ocean Food webs more likely based on pelagic sources	Multiple catchment inputs e.g. the major Loire and Gironde Rivers and several minor ones
Gulf of Lions (GoL)	Semi-enclosed and oligotrophic Mediterranean Sea Smaller individuals with slower growth rates than in Atlantic systems for similar species	Rhône River Marine traffic including touristic ferries

Trace element determination: detailed method, quality assurance and quality control (QA/QC) criteria.

Arsenic, Cd, Hg and Pb contents were measured in fish muscle samples using an ISO 17025 accredited method (French Accreditation Committee, COFRAC) described in Noel et al. (2005). Briefly, 0.3 to 0.4 g of each sample were precisely weighed in a quartz digestion vessel and digested with a mixture of 3 mL of 67% suprapur HNO_3 (VWR chemicals, Prolabo, France) and 3 mL of ultra-pure water in a closed microwave digestion system (Multiwave 3000, Anton-Paar, Courtaboeuf, France). Sample digests were then analysed by ICP-MS (7700x Agilent Technologies, Courtaboeuf, France). Mean limits of detection and quantification (LOD/LOQ), calculated based on wet-weight (ww), were 1.2/4.1 µg/kg for both As and Hg, 0.2/0.8 µg/kg for Cd and 0.7/2.4 µg/kg for Pb. Individual LOD and LOQ might slightly differ from the mean due to slight variation in sample weight and humidity.

Method accuracy and precision were assessed on a daily basis by the analysis of a certified reference material (CRM) ERM-CE278k (mussel tissue, Institute for Reference Materials and Measurements, Geel, Belgium, Table SM 2).

Table SM 2*. Results of trueness on certified reference material (CRM) ERM-CE278k (mussel tissue).*

Element	ERM-278k			
	Certified value (mg/kg)	Confidence interval (mg/kg)[1]	Mean ± SD measured value (n=10, mg/kg)	Min-max (mg/kg)
Pb	2.18	1.69 – 2.67	2.04 ± 0.04	1.99 - 2.13
Cd	0.336	0.260 - 0.412	0.292 ± 0.008	0.277 - 0.310
As	6.7	5.2 – 8.2	6.5 ± 0.2	6.1 - 6.9
Hg	0.071	0.055 – 0.087	0.075 ± 0.007	0.068 - 0.084

[1]: Calculated from the certified value (M) of the CRM as: $CI = M \pm \left[k \times \frac{CV_R \times M}{100}\right]$
with k = 3 (p = 99%); M the certified value and CVR, the intermediate precision coefficient of variation (CVR = 7.5 %).

Organic contaminant determination: detailed method and QA/QC.

Fish muscle samples were analysed for the seventeen 2,3,7,8-substituted PCDD/Fs and twelve DL-PCBs with toxic equivalency factors (TEFs) assigned by the World Health Organization (WHO, Van den Berg et al., 2006). The six NDL-PCBs included in the EU regulation were also determined (EU, 2017). PCDD/Fs and PCBs were determined according to a validated and accredited method (ISO/IEC 17025:2005 standard) fully described elsewhere (Vaccher et al., 2018).

Succinctly, samples were extracted automatically in a pressurised solvent extraction device (SpeedExtractor E-914; BÜCHI Sarl, France) through three successive static cycles using a mixture of toluene and acetone at 70:30 (v/v) as solvent, pressure set at 100 bar and temperature at 120 °C. Next, the solvent was evaporated and the lipid content of the sample was determined gravimetrically. Purification and fractionation of PCDD/Fs and PCBs were carried-out on an automated system (Autosym GO-4-HT; Miura-Shimadzu, Japan) in four sequential chromatographic steps involving a silver nitrate impregnated silica gel, a sulfuric acid impregnated silica gel, an active carbon and an activated alumina column.

Analysis of cleaned-up extracts was conducted by gas chromatography (7890A; Agilent Technologies, USA) coupled to a double electromagnetic sector high resolution mass spectrometer (JMS-700D and 800D; Jeol, Japan) set at a resolution of 10 000. Chromatographic separation of PCDD/F congeners was carried out on a DB-5MS column (60 m × 0.25 mm i.d., 0.25 µm film thickness; Agilent Technologies, USA), while for PCBs a HT8-PCB column (60 m × 0.25 mm i.d., 0.25 µm film thickness; SGE Analytical Science, UK) was used. Electron ionisation was set at 70 eV and source temperature at 280 °C. PCDD/F and PCB congeners identification criteria and quantification fulfil the conditions and guidelines of the EPA methods 1668 (EPA, 2008) and related European directives (EU, 2017).

The procedure integrated the quality assurance and quality control (QA/QC) criteria to fulfil the requirements of the European legislation laying down sampling procedures and the method of analysis for determination of PCDD/Fs and DL-PCBs (EU, 2017). Blanks were included in every series of samples to check for interference and cross-contamination. The chromatographic separation was checked (<25% peak to peak between 1,2,3,4,7,8-HxCDF and 1,2,2,6,7,8-HxCDF) and recoveries of individual congeners were within 30-140% as required by the European Commission 2017. The limits of detection (LOD) for fish muscle were between 0.003 and 0.017 pg g^{-1} of fresh weight for PCDD/Fs from tetra- to octa-chlorinated substituted congeners. All the DL-PCBs and NDL-PCBs were detected with a LOD fixed at 0.02 pg g^{-1} of fresh weight. All target compounds were quantified using the isotope-dilution method. PCDD/F and PCB values were automatically corrected considering the recovery rate of the ^{13}C-labelled internal standards. The associated measurement uncertainties for the values of PCDD/Fs-WHO-TEQ (2005), DL-PCBs-WHO-TEQ (2005), PCDD/Fs + DL-PCBs)-WHO-TEQ (2005) and sum of NDL-PCBs were 17%, 22%, 19% and 25% respectively.

Quantification of DL-PCB and PCDD/F congeners.

DL-PCBs were quantified in nearly all the samples (94% for CB169 and >99% for the 11 other DL-PCBs). Mean concentrations for DL-PCBs were higher than PCDD/F concentrations, they ranged from 0.41 to 707 pg/g ww for CB81 and CB118, respectively. Both most toxic TEQ congeners, TCDD and PeCDD, were quantified in 64 and 76% of the samples (n = 184), respectively, at mean concentrations of 0.02 and 0.04 pg/g ww, respectively. For the other PCDD, percentage of quantification ranged from 48% to 83% with no clear trend with the chlorination level. For PCDF, percentage of quantification was lower for the highest chlorinated congeners as it ranged from 14% (1,2,3,4,7,8,9-HpCDF (hepta-chlorinated) and OCDF (octa-chlorinated)) to >90% (2,3,7,8-TCDF (TCDF, tetra-chlorinated), 2,3,4,7,8-PeCDF (penta-chlorinated), 1,2,3,4,7,8-HxCDF and 1,2,3,6,7,8-HxCDF (both hexa-chlorinated)). Mean concentrations per congeners ranged from ≤ 0.01 pg/g ww (0.007 pg/g ww for 1,2,3,7,8,9-HxCDF and 0.01 pg/g ww for 1,2,3,4,7,8,9-HpCDF) to 0.45 pg/g ww (TCDF). TCDF was attributed the third higher toxic equivalent value in TEQ calculation, i.e. 0.1. It was the dominant dioxin congener in fish from the present study, in terms of concentration and occurrence (% of quantification).

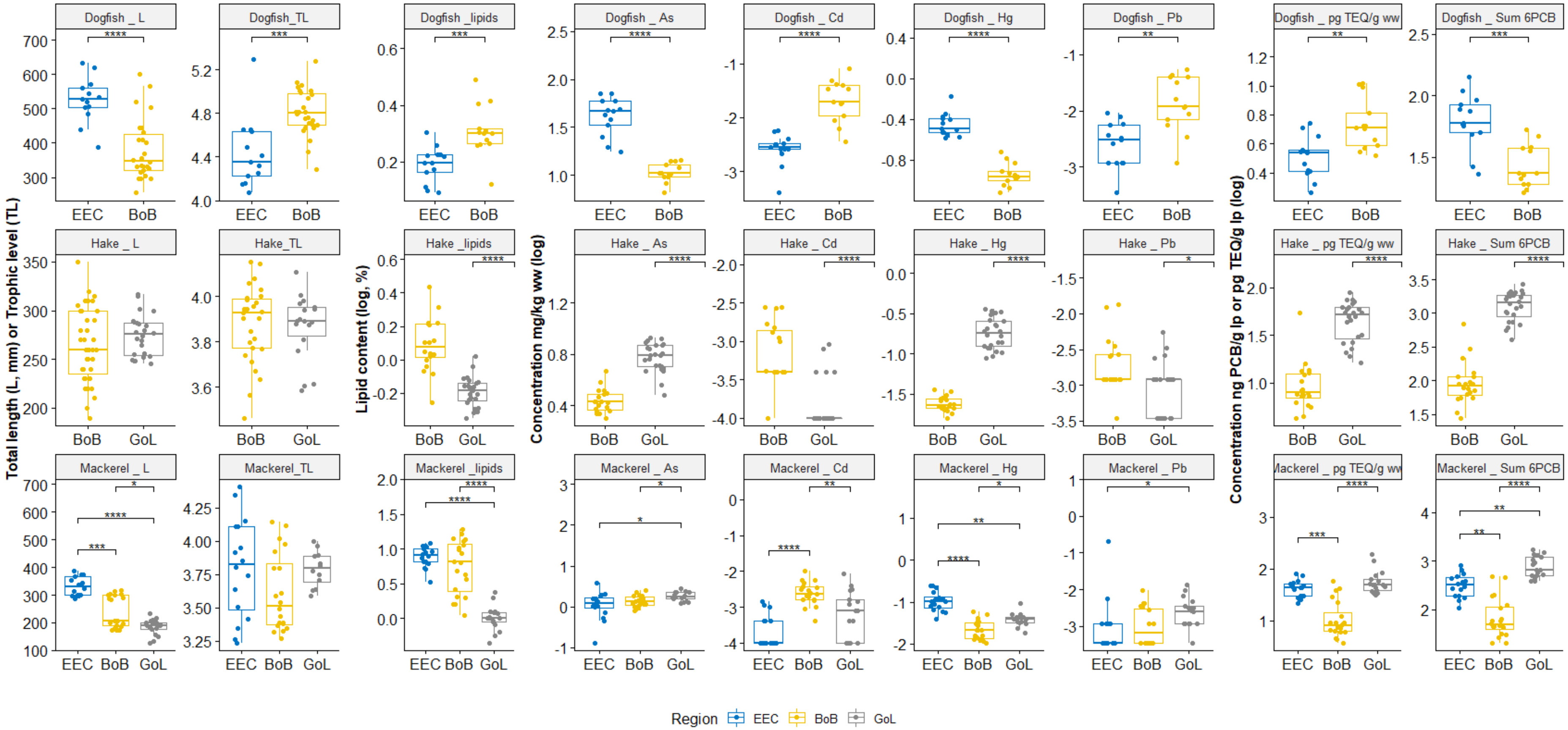

*Fig. SM 1. Comparison of biological parameters and contaminant levels in fish muscle among regions for dogfish, hake and mackerel (significant differences between regions according to Kruskal Wallis and Dunn as post hoc test or Wilcoxon test *<0.05, **<0.01, ***<0.001, ****<0.0001).*

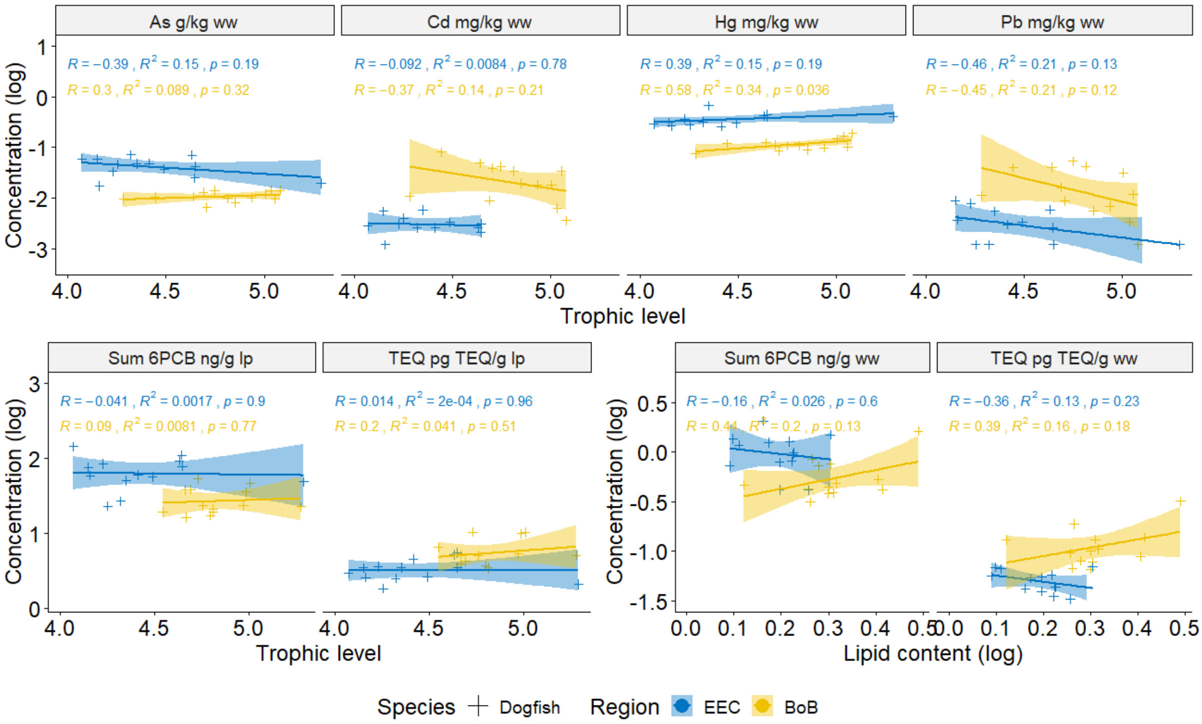

Fig. SM 2. Linear regressions (Pearson correlation (R), correlation coefficient (R²), and p-value (p)) between contaminant concentrations in chondrichthyans (dogfish), and trophic levels or lipid contents (for organic contaminants) in each region.

Table SM 3. Good environmental status of the French metropolitan continental shelf, according to fish contamination, as assessed in 2018 for the second MSFD cycle. *Number of species exceeding thresholds (compared to 95 percentile) / number of species monitored are indicated in blue cells when all the species complied with threshold and in red when at least one species exceeded threshold.*

Contam. family	Contam.	Threshold value	EEC Cod (*n*=5) Dogfish (*n*=13) Mackerel (*n*=16) Whiting (*n*=16) Plaice (*n*=12)	BoB Blue whiting (*n*=19/16[1]) Dogfish (*n*=13) Mackerel (*n*=20) Hake (*n*=19/20[1]) Sardine (*n*=12)	GoL Mackerel (*n*=17) Hake (*n*=24)
Trace elements	**Cd [2]**	MPC_{SAR} = 0.10 mg/kg ww MPC_{All} = 0.050 mg/kg ww	0/5	1/5 (Dogfish)	0/2
	Hg [3]	MPC_{Sharks} = 0.10 mg/kg ww MPC_{All} = 0.050 mg/kg ww	0/5	0/5	0/2
	Pb	EC = 0.3 mg/kg ww	0/5	0/5	0/2
PCB	**CB28**	EAC = 67 µg/kg lp	0/5	0/5	0/2
	CB52	EAC = 108 µg/kg lp	0/5	0/5	2/2
	CB101	EAC = 121 µg/kg lp	0/5	0/5	2/2
	CB138	EAC = 317 µg/kg lp	0/5	0/5	1/2 (Hake)
	CB153	EAC = 1 585 µg/kg lp	0/5	0/5	0/2
	CB180	EAC = 469 µg/kg lp	0/5	0/5	1/2 (Hake)
	CB118 (DL)	EAC = 25 µg/kg lp	4/5 (Mac., Whiting, Cod, Plaice)	2/5 (Mac., Hake, Sar.)	2/2
Dioxins DL-PCBs	**TEQ**	MPC = 6.5 µg TEQ/kg ww	1/5 (Mac.)	0/5	0/2

[1] Number of samples analysed for metals and organic contaminants, when they differ. [2] Maximum permissible concentration (MPC) for Cd depends on the species: EC_{SAR} (0.10 mg/kg ww) for sardine; EC_{All} (0.050 mg/kg ww) for the other monitored species (reg EC n°1881/2006). This threshold is used in the absence of environmental threshold but does not inform on GES according to D8. [3] MPC for Hg depends on the species: MPC_{Sharks} (1.0 mg/kg ww) for sharks (*i.e.* dogfish); MPC_{All}: (0.5 mg/kg ww) for the other monitored species (reg EC n°1881/2006). This threshold is used in the absence of environmental threshold but does not inform on GES according to D8.

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