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A large diversity of organohalogen contaminants reach the meso- and bathypelagic organisms in the Bay of Biscay (northeast Atlantic)

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Abstract :

Deep-sea ecosystems play a key role in the cycling and vertical transfer of matter and energy in oceans. Although the contamination of deep-sea demersal and benthic organisms by persistent organic pollutants has been proven, deep pelagic species have been far less studied. To fill these gaps, we studied the occurrence of a large variety of hydrophobic organic contaminants including polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs), legacy and alternative brominated flame retardants (BFRs) and per- and polyfluoroalkyl substances (PFASs) in crustaceans and fish species collected in the Bay of Biscay, northeast Atlantic. The results highlighted the global predominance of PCBs in fish, followed by OCPs, PFASs and PBDEs, with highly variable concentrations among species. Most of the chlorinated or brominated contaminants showed increasing concentrations with increasing δ15N values, while most PFASs showed inverse trends. The contaminant profiles and diagnostic ratios revealed species-specific metabolic capacities and peculiar contribution of highly-brominated BFRs.

Highlights

► Organohalogen contaminants were ubiquitous in deep pelagic crustaceans and fish. ► PCBs were predominant in fish followed by OCPs, PFASs and PBDEs. ► BFR profiles were peculiar with high contributions of BDE-209 and DBDPE. ► Specific ratios highlight species-specific metabolic capacities. ► Long-chain PFCAs showed biodilution with increasing $\delta^{15}N$ values.

Keywords : Deep-sea, Crustaceans, Fish, Persistent organic pollutants, Contaminants of emerging concern, Species -specific profiles

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1. Introduction

The deep-sea pelagic environment (< 200 m) is one of the largest ecosystems on Earth and supports a high diversity and abundance of marine species, especially in the bathyal horizon (< 2000 m) (Rogers, 2015). In particular, meso- (< 1000 m) and bathypelagic (1000–2000 m) communities represent essential components of oceanic biomass and important prey for higher trophic levels including large pelagic fish (tunas, sharks), marine mammals and seabirds. They play a key role in the cycling and vertical transfer of matter and energy in oceans (Bernal et al., 2015; Bianchi et al., 2013; Catul et al., 2011). As a consequence of their strong diel vertical migrations (DVM) during which some species (Chouvelon et al., 2022; Eduardo et al., 2021; Takahashi et al., 2000) feed in surface waters (epipelagic zone, 0–200m) at night and move back to meso- and bathypelagic zones during the day, meso- and bathypelagic organisms release particulates, organic matter and associated contaminants *via* faecal pellet egestion and may therefore increase transfers from surface waters to deeper horizons (Belcher et al., 2019; Bernal et al., 2015). Furthermore, their upward migration, during which they become available to surface predators, leads to the transfer of organic matter and contaminants accumulated in their tissues from deep horizons back to epipelagic layers. In addition to their essential role in biogeochemical cycles, mesopelagic organisms have also raised interest regarding their exploitation as new resources for human consumption as well as the fish meal and oil industry (Berntssen et al., 2021; Grimaldo et al., 2020).

Although deep-sea ecosystems have been studied for decades, most studies focused on deep demersal or benthic communities and rarely on deep pelagic organisms, especially those from the ocean "twilight zone" (200–1000 m), which thus remain the most understudied ones. Recent publications have therefore highlighted the urgent need to increase knowledge in various research areas for these deep pelagic ecosystems, including the fate and impact of organic contamination (Martin et al., 2020; Sanganyado et al., 2021). Indeed, oceans and deep waters in particular are final sinks for anthropogenic wastes, including chemicals produced from industrial, urban, domestic and agricultural uses (Froescheis et al., 2000; Looser et al., 2000; Zhang et al., 2019). Contaminant sources to oceans are

numerous and include mainly (80%) land-based but also sea-based activities. Atmospheric transport, continental inputs *via* rivers and runoff as well as direct discharges contribute to their widespread occurrence in oceans (Landrigan et al., 2020; Ramu et al., 2006; Tornero and Hanke, 2016). Although deep-sea ecosystems are remote from direct anthropogenic sources of pollutants, various studies have shown that persistent hydrophobic organic contaminants are transported to deep oceanic waters, including the hadal trenches (Cui et al., 2020; Dasgupta et al., 2018; Jamieson et al., 2017; Takahashi et al., 2010; Webster et al., 2014). Because of their high hydrophobicity, long half-lives and long-distance transport, persistent organic pollutants (POPs) and other substances showing similar properties are of particular concern in our global environment (Cousins et al., 2019; Jones, 2021).

Despite their regulation for decades and the remoteness of deep ecosystems from direct sources, the transport of legacy POPs to the deep sea has been proven, and various deep-sea species have been shown to be prone to high exposure to these contaminants, leading to their bioaccumulation at higher levels than in shallower water organisms (Froescheis et al., 2000; Looser et al., 2000; Mormede and Davies, 2003; Ramu et al., 2006 and references therein) and making chemical contamination by POPs one of the anthropogenic pressures in the deep sea (Stemmler and Lammel, 2013). Among the legacy POPs, polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs) and polybrominated diphenyl ethers (PBDEs) are by far the mostly-studied organic contaminants in deep ecosystems (Covaci et al., 2008; Ramu et al., 2006; Romero-Romero et al., 2017; Storelli et al., 2009; Takahashi et al., 2010; Webster et al., 2014). However, most of these studies relate to demersal and benthic species living near or on the bottom floor of oceans (most of the references above), while true deep pelagic species are more seldom considered. Other substances with similar properties as POPs are of similar concern, as very few data on their occurrence and fate in deep-sea ecosystems are available so far. Among them are alternative flame-retardants such as those used in the replacement of legacy compounds, for example, BTBPE (1,2-bis(2,4,6-tribromophenoxy)ethane) and DBDPE (decabromodiphenylethane), which replaced penta- and deca-BDE respectively. Per- and polyfluoroalkyl substances (PFASs), including the regulated long-chain perfluorocarboxylic acids

(PFCAs) in particular, represent another under-studied class of compounds, although increasing concern has been recognised for this vast family of less hydrophobic compounds for whom oceans constitute the final reservoir (Armitage et al., 2009; Yamashita et al., 2008). Their transport to deep seas mainly *via* advection and settling particles has indeed been recently emphasised (Sanchez-Vidal et al., 2015; Sanganyado et al., 2021; Zhang et al., 2019).

Despite their importance for ocean health and services and their exposure to various anthropogenic threats, deep-sea ecosystems and deep pelagic ones, in particular, are still poorly studied, including the "twilight zone" towards which concerns have been recently raised regarding climate change and human exploitation (Martin et al., 2020). Specifically, information on organic contaminant occurrence and understanding of their bioaccumulation and biomagnification in deep pelagic food webs are necessary to better assess organic contaminants' ecological impacts (Sanganyado et al., 2021) and assess health risks potentially associated with human exploitation of these deep resources (Grimaldo et al., 2020; Wiech et al., 2020). In this context, this study aimed to investigate the accumulation of organohalogen contaminants (OCs) in meso- and bathypelagic species from the Bay of Biscay, northeast Atlantic, focusing on crustaceans and fish. The studied contaminants include a large diversity of organic contaminant families such as the legacy POPs listed in the Stockholm Convention (i.e. PCBs, OCPs, PBDEs and PFASs such as perfluorooctane sulfonate -PFOS and perfluorohexanoic acid -PFHxA) and other regulated contaminants (i.e. long-chain PFCAs). The contamination was notably studied using the stable carbon and nitrogen isotope compositions ($\delta^{13}C$ and $\delta^{15}N$ values) concomitantly analysed on organisms, as respective trophic tracers of organic matter sources sustaining them and of their trophic position within the deep pelagic community studied. The results are expected to serve as a benchmark in future studies and are of prime interest to characterise and evaluate the chemical exposure of various organisms having a central role in marine ecosystems. This study provides another light on the contamination of the species living in the twilight zone in addition to the previously published results on major and trace elements analysed in the same samples (Chouvelon et al., 2022).

2. Materials and methods

2.1. Sampling

Samples were collected on the French slope of the Bay of Biscay (NE Atlantic) during the annual EVHOE fishery survey, on board the Ifremer R/V Thalassa in October 2017 (https://doi.org/10.17600/17002300). Fish (11 species) and crustacean (3 species) samples were collected at night using a 25 m vertical opening pelagic trawl in the deep scattering layer (ca 800 m depth in the water column; 1330 m bottom floor). All samples were collected during the same haul of 60 min at a speed of approximately 4 knots. The selected fish individuals belonged to the most abundant species, including one species of Sternoptychidae and three species of Myctophidae, two of the most abundant mesopelagic fish families globally (Catul et al., 2011; Valinassab et al, 2007).

Handling of samples was conducted on-board using rigorous protocols to avoid external contamination. Samples were stored at –20 °C until further processing in the laboratory. To obtain sufficient material for the quantification of OCs, the whole bodies of individuals belonging to the same species were pooled by specimens of similar sizes (Table 1). Considering the sizes reported in the literature, most fish except *Aphanopus carbo* were adult fish. A small piece of white muscle (< 3% of individual total weight) was also collected for the analysis of stable isotopes of carbon and nitrogen as trophic tracers. Whenever possible, fish individuals' sexes were determined and noted in the composition of each pool; all pools were made of both male and female individuals except for *Serrivomer beanii* (males only) and *Stomias boa* (females only). All fish stomachs were emptied from their major visible content; however, most stomachs were found to be empty, suggesting either a delay between the sampling and the last feeding or a fast stomach emptying. When large enough, fish were analysed individually; this was the case for 3 samples of *Serrivomer beanii,* 2 samples of *Stomias boa* and 3 samples of *Aphanopus carbo* (Table 1). After pooling, the samples were homogenised using a blender with stainless steel arms, freeze-dried and finely ground up with a ball mill MM400 (Retsch) using bowls and marbles with a zirconium oxide coating. Immediately after freeze-drying, the moisture

158 percentage was determined in each sample; they varied between 69% and 78% in crustaceans and
159 between 64% and 88% in fish (Table 1).

160 ***2.2. Chemical analyses***

161 Extractable organic matter, used as a proxy for total lipid content (TLC), was determined gravimetrically
162 using 500 mg of sample extracted with a mixture of hexane and acetone (80:20 v:v) using pressurised
163 liquid extraction (PLE). The extracts were evaporated to dryness and TLC was expressed in % of dry
164 weight (dw).

165 PCBs, OCPs and BFRs were determined as described by Munschy et al. (2020a). Briefly, 5–10 g of
166 samples were extracted by PLE with dichloromethane, followed by gel permeation chromatography, a
167 silica and alumina adsorption chromatography column and a two-dimensional HPLC system with two
168 columns coupled in series. Analyses were performed by gas chromatography (Agilent 6890, Palo Alto,
169 CA, USA) coupled to high-resolution mass spectrometry (AutoSpec Ultima, Waters Corp.). BDE-209,
170 DBDPE and BTBPE were analysed using an Agilent 7890B gas chromatograph coupled to a triple
171 quadrupole mass spectrometer Waters Xevo TQS-µ (Millford, US) using atmospheric pressure chemical
172 ionisation operated in the positive mode. The samples were analysed for 35 PCBs ranging from tri- to
173 decachlorinated congeners, including the 12 dioxin-like (dl-) PCBs (CB-77, -81, -105, -114, -118, -123, -
174 126, -156, -157, -167, -169, -189), the 6 indicator (i-) PCBs (CB-28, -52, -101, -138, -153, -180), various
175 OCPs (*p,p'*-DDT, *o,p'*-DDT, *o,p'*-DDD, *p,p'*-DDD, *p,p'*-DDE, dieldrin, aldrin, hexachlorocyclohexanes –
176 HCHs and hexachlorobenzene -HCB, referred to as ∑ OCPs later in the text) and BFRs including 36 PBDE
177 congeners from tri- to decabrominated ones (Table S5) and non-PBDE BFRs (HBB -hexabromobenzene,
178 BB-153 -2,2',4,4',5,5'-hexabromobiphenyl, BTBPE, DBDPE).

179 PFASs were determined according to Munschy et al. (2020a). Briefly, 1 g of sample was extracted using
180 liquid-solid extraction (LSE) with a blend of MeOH/KOH and purified onto two consecutive SPE
181 cartridges: an Oasis WAX weak anion exchange stationary phase and an Envicarb charcoal stationary
182 phase. Analysis was performed using an Acquity ultra-performance liquid chromatograph (UPLC®,
183 Waters Corp.) coupled to a triple quadrupole mass spectrometer (Xevo® TQ-S micro, Waters Corp.)

interfaced with an electrospray ionisation source Z-sprayTM (Waters Corp.). The mass spectrometer was operated in negative ionisation mode using multiple reaction monitoring (MRM) with argon as the collision gas. PFASs were analysed for five C_4- to C_{10}-perfluoroalkyl sulfonates (PFSAs) and nine C_6- to C_{14} perfluorocarboxylic acids (PFCAs), namely: perfluorobutane sulfonate (PFBS); perfluorohexane sulfonate (PFHxS); perfluoroheptane sulfonate (PFHpS); perfluorooctane sulfonate (PFOS); perfluorodecane sulfonate (PFDS); perfluorohexanoic acid (PFHxA); perfluoroheptanoic acid (PFHpA); perfluorooctanoic acid (PFOA); perfluorononanoic acid (PFNA); perfluorodecanoic acid (PFDA); perfluoroundecanoic acid (PFUnDA); perfluorododecanoic acid (PFDoDA); perfluorotridecanoic acid (PFTrDA) and perfluorotetradecanoic acid (PFTeDA).

Quality Assurance/Quality Control (QA/QC) procedures were carefully followed during the entire analytical protocol. This included quantification by isotopic dilution using ^{13}C-labeled compounds, five-to-six-point calibration curves in each sequence of samples to calculate relative response factors and check linearity, laboratory blank determination (whole analytical procedure), in-house quality control sample, participation in QUASIMEME (Quality Assurance of Information for Marine Environmental Monitoring in Europe) and intercomparison exercises with satisfactory Z scores. Detailed analytical parameters and QA/QC results are given in the SI.

2.3. Statistical analyses

All statistical analyses were performed using StatSoft Statistica software v 13.3 with a significance level (α) of < 0.05. Concentrations below the limits of quantification (LOQs) were assigned as missing values (i.e. counted as zero in the sums), and only compounds quantified in more than 50% of the samples were considered for statistical analyses. Data were tested for normality using the Shapiro-Wilk's test and parametric or non-parametric tests were performed depending on a normal distribution. Correlations (e.g. between total lipid content and POP concentrations and between contaminant concentrations) were tested using simple linear regression coefficients and Spearman's rank correlation test was used to evaluate the strength and direction of relationships. Data comparisons (biological parameters, POP concentrations and ratios) across groups were performed using non-

parametric tests (Mann–Whitney: MW- and one-way ANOVA Kruskal-Wallis's: KW -test to compare independent groups) for non-normally distributed data. Results were considered significant only when both tests gave significant results. Principal component analysis (PCA) was performed on normalised concentrations to avoid the concentration effect.

3. Results and discussion

3.1. Trophic markers

The detailed results obtained for trophic markers on this deep pelagic community were presented in Chouvelon et al. (2022). The values specifically obtained for the samples here analysed for organic contaminants (a selection of those analysed for chemical elements by Chouvelon et al., 2022) are summarised in Table 1. Briefly, $\delta^{13}C$ values ranged from –20.30‰ to –18.26‰, with no significant differences between crustaceans and fish. This relatively small variability for $\delta^{13}C$ values (~2‰ difference) indicates similar carbon sources sustaining the species (Chouvelon et al., 2022). The $\delta^{15}N$ values showed more variability, ranging from 9.24‰ to 12.23‰ (Table 1). This range suggests a difference of at least one trophic level (following the expected mean difference of 3.4‰ per trophic level, Post, 2002) among species of the studied food web. $\delta^{15}N$ values were significantly different (MW, $p = 0.0017$) between crustaceans (9.58 ± 0.28‰ on average, n = 5) and fish (10.76 ± 0.91‰, n = 28).

3.2. Total lipid contents (TLCs)

The TLCs exhibited high variations among taxa and species, ranging from 4.3 ± 0.9% dw (n = 3) in *Pasiphaea sivado* to 51% dw in *Ephyrina figueirai* (n = 1) for crustaceans and, for fish, from 6.1 ± 0.1% dw (n = 3) in *Xenodermichthys copei* to 41.9 ± 9.6% dw (n = 3) in *Notoscopelus kroeyeri* (Table 2). A high diversity in TLCs was found between the crustacean species (rsd 130%), while the fish species could be distinguished between low-TLC species (*Serrivomer beanii, Xenodermichthys copei, Lampanyctus crocodilus, Chauliodus sloani* and *Aphanopus carbo*) and the other species. High lipid contents (i.e. above 15% dw) were determined in the crustaceans *Sergia robusta* and *Ephyrina figueirai* as well as in the Papalepididae *Arctozenus risso,* the Sternoptychidae *Argyropelecus olfersii,* the Myctophidaes

Myctophum punctatum and *Notoscopelus kroeyeri*, the Stomiidae *Stomias boa* and the Platytroctidae *Searsia koefoedi*. These results agree with the energy densities reported by Spitz et al. (2010) in various forage fish species from the Bay of Biscay. Except in *Stomias boa* for which the replicate made from 3 individuals showed extremely different values compared to the two other replicates (made of 1 individual each), TLCs were fairly similar between replicate pool samples of the same species, with an average rsd of 24% (12-34% range excluding *Stomias boa*). The TLC values in *Stomias boa* replicates (all female individuals) showed a higher rsd (69%), with high TLC values of 17.6% dw and 19.5% dw for the two 35 cm and 32 cm individuals respectively, whilst the replicate made from 3 individuals of 27.7 ± 1.5 cm (on average) exhibited a much lower TLC value, i.e. 2.7% dw. The only noticeable difference noted during dissection was a slight difference between the maturity stages (based on gonad visual observation) with a less mature stage for the smaller individuals. Concomitantly to the maturity stage, ontogenic differences may also explain the difference observed in the lipid contents of *Stomias boa* samples, as energy reserves were shown to be positively correlated with size at the fish species level (Anthony et al., 2000; Cargnelli & Gross, 1997). However, at the scale of the fish community analysed here, TLCs showed a significant linear inverse relationship with fish lengths ($p = 0.014$), indicating a higher energy content for the smaller species. Classically, TLCs showed an inverse significant linear relationship ($p < 0.001$) with humidity percentages (Spitz et al., 2010).

TLC values were similar to those reported previously in the literature, even though different extraction methods were used (chloroform:methanol:water (1:2:1) in Sen Özdemir et al., 2019 and ether-ethyl in Spitz et al., 2010 versus hexane and acetone (80:20 v:v) for our samples).

3.3. Bioaccumulation of organohalogen contaminants in the deep pelagic ecosystem

3.3.1. Organic contaminant concentrations and relative contributions show high inter-species variabilities

The concentrations of the various OC families analysed are presented in Table 2 in both dw (all contaminants) and lw (for PCBs, OCPs and PBDEs) and details for each compound are given in Tables S3 to S6.

In both crustaceans and fish species, PCBs showed high detection frequencies. Most PCB congeners showed detection frequencies of 100%, except for CB-77 and CB-189 (97%), CB-126 and CB-169 (91% and 58% of the samples, respectively) (Table S3), whilst CB-81 was seldom detected above the LOQ (6% of the samples). CBs -29, -30, -112 and -114 were never detected. Among OCPs, dieldrin, endrin, and mirex were detected in all samples, similarly to the DDT isomer *p,p'*-DDE, whilst the other DDT isomers showed detection frequencies in the 88–94% range (Table S4). Among HCHs, β-HCH was detected in 100% of the samples, above γ-HCH (73%), α-HCH (58%) and δ-HCH (6%). HCB and PeCB were detected in 97% and 52% of the samples, respectively. The other OCPs, namely, aldrin, isodrin, α-endosulfan, β-endosulfan and endosulfan sulphate, were never detected. BDEs -28, -47, -49, -66, -99, -100, -119, -126, -153, -154 and -155 were the most frequently detected (> 70% of the samples), while BDEs -77, -183, -184, -202, -205, -207 and -209 showed intermediate detection frequencies (27–55%). The other PBDE congeners, i.e. BDEs 30, -71, -85, -138, -171 and -204 were below LOQs in all samples. The non-PBDE BFRs HBB, BB-153 and DBDPE were detected in 61, 73 and 76% of the samples, respectively. BTBPE was only detected in 7 samples (21%). Among PFASs, PFOS, PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA and PFTeDA were detected in 100% of the samples. PFOA was detected in 45% of the samples, PFDS in 27% and PFHpA was seldom detected (12%) (Table S4). PFHpS and PFHxA were below LOQs in all samples, PFHxS was detected in one sample only and close to the LOQ.

Globally, concentrations (calculated on the mean values of replicates per species) ranged from 11.28 to 100.07 ng g^{-1} dw, 1.20 to 62.49 ng g^{-1} dw, 0.12 to 3.49 ng g^{-1} dw and 3.07 to 55.69 ng g^{-1} dw for ∑ PCBs, ∑ OCPs, ∑ PBDEs and ∑ PFASs, respectively (Table 2). In crustaceans, the intra-species variability of concentrations was low between the 3 *Pasiphaea sivado* replicate pools (6–11% rsd for PFASs, PCBs and OCPs and 45% for PBDEs), but huge variations were found at the inter-species level, with rsd values of 55, 67, 107 and 97% for PFASs, PCBs, OPCs and PBDEs, respectively. The different OC families' contributions showed also some variations between crustacean species. PFASs showed the highest contamination levels of all OC families in *Pasiphaea sivado* (mean of 23.83 ± 2.70 ng g^{-1} dw, representing 65% of the OCs) followed by PCBs (11.28 ± 0.69 ng g^{-1} dw, 31%), OCPs (1.20 ± 0.69 ng g^{-1}

dw, 3%) and PBDEs (0.12 ± 0.05 ng g^{-1} dw, 0.3%). This species showed the lowest contamination levels of PCBs, OCPs and PBDEs, i.e. 6 times, 10–30 times and 5 times lower, respectively, than the concentrations determined in *Sergia robusta* and *Ephyrina figueirai*. These OC relative contributions could be partially explained by TLC values, as *Pasiphae sivado,* with the lowest TLC values, was hence expected to show lower bioaccumulation of lipophilic OCs such as PCBs, OCPs and PBDEs. In the two other crustacean species, chlorinated OCs (and PCBs in particular) were predominant, while PBDEs contributed the least. The contaminant concentrations and TLCs in crustaceans showed significant correlations for ∑ DDTs ($p < 0.0001$), endrine ($p = 0.015$), HCB ($p = 0.003$), ∑ HCHs ($p = 0.024$) and ∑ PBDEs ($p = 0.010$). However, the significance was driven by the high differences in TLCs found between the 3 studied crustacean species (4 ± 1%, 15% and 51% in *Pasiphaea sivado*, *Sergia robusta* and *Ephyrina figueirai*, respectively), while no correlation was evidenced for the three replicates of *Pasiphaea sivado*. When normalised to TLCs, OCP concentrations were still lower in *Pasiphaea sivado* versus the other two species (both being similar), while PCB concentrations were still higher in *Sergia robusta* > *Pasiphaea sivado* > *Ephyrina figueirai*. PBDE concentrations were similar in both *Sergia robusta* and *Pasiphaea sivado* (not determined in *Ephyrina figueirai* because of a matrix effect). PFAS concentrations showed similar levels in *Pasiphaea sivado* and *Ephyrina figueirai* but a two times higher level in *Sergia robusta*.

In fish, PCBs presented the highest concentrations (range of 18.62 ng g^{-1} dw to 115.37 ng g^{-1} dw, mean of 54.42 ± 28.57 ng g^{-1} dw calculated on the replicates), followed by OCPs (5.69–98.57 ng g^{-1} dw, mean of 21.73 ± 21.26 ng g^{-1} dw, of which 83% were DDTs), PFASs (1.54–37.25 ng g^{-1} dw, mean of 11.95 ± 9.58 ng g^{-1} dw) and PBDEs (0.43–5.31 ng g^{-1} dw, mean of 1.57 ± 1.17 ng g^{-1} dw). These results agree with previous studies showing PCBs and DDTs as the major chlorinated contaminants in deep-sea fish worldwide, reflecting their high persistence and hydrophobicity, while PBDE levels are usually reported to be several orders of magnitude lower (Koenig et al., 2013; Webster et al., 2014).

The intra-species variability of concentrations was globally low for all contaminant families (21% rsd on average for PFASs and 29, 38% and 36% for PCB, OCP and PBDE concentrations in dw). For all

contaminant families, the highest variabilities (44–75% in dw, 64–80% in lw) were obtained between *Notoscopelus kroeyeri* replicates. The inter-species variability (calculated on the species means) was between 43% (PCBs) and 87% (PFASs) depending on the OC family. *Xenodermichthys copei* and *Myctophum punctatum* were the least-contaminated species in PCBs, OCPs and PBDEs while *Notoscopelus kroeyeri* showed the lowest PFAS concentrations. The highest levels were determined in *Chauliodus sloani* for PCBs, in *Stomias boa* for OCPs and PBDEs and *Searsia koefoedi* for PFASs.

Only a limited number of lipophilic OCs (i.e. dieldrin, endrin, ∑ HCHs and each isomer, HCB) showed significant ($p < 0.0001$ to 0.022) positive correlations with TLCs, while ∑ DDTs, ∑ PCBs (whether i-PCBs, dl-PCBs or ∑ all congeners were considered) and ∑ PBDEs did not. Very few individual PCB congeners (CBs -18, -28, -31, -44, -49, -52 and -66, i.e. the less-chlorinated ones) and individual PBDEs (BDEs -66 and -77) showed a significant linear relationship between their concentrations and TLCs. These results are somewhat surprising for lipophilic contaminants. However, TLCs determined in the present samples were a proxy of total lipids and it has been proven that lipophilic contaminant accumulation is not governed only by lipid content but also by lipid composition (Xie et al., 2020). Our results suggest complex relationships between the species' lipidic composition and the various studied chlorinated and brominated OC families and would argue in favour of more investigations into the specific tissue biochemical composition of species. Despite this lack of significant correlations, concentrations of lipophilic OCs were normalised to TLCs to compare the concentrations between replicate samples within a species (intra-species variability) and between species (inter-species variability). Normalising the concentrations to TLCs did not decrease the intra-species variability (30, 29 and 39% for PCBs, OCPs and PBDEs respectively) nor the inter-species variability (73–75%). However, these results were highly dependent on the species; the intra-species variability decreased highly for *Argyropelecus olfersii* (ratio between the concentrations in the two samples of 1.0, 1.4 and 1.0 in lw versus 1.4, 1.9 and 1.5 in dw for PCBs, OCPs and PBDEs, respectively), while the concentrations in *Notoscopelus kroeyeri* were highly variable whether they were expressed in dw (60–75%) or lw (64–80%). When both taxa were compared, only PFAS and PBDE concentrations (in ng g^{-1} dw) showed significant differences (KW and

MW) between crustaceans and fish (PFASs higher in crustaceans p = 0.0034, PBDEs higher in fish p = 0.0021). Although PCBs and OCPs showed higher levels in fish than in crustaceans, the lack of significant differences between both taxa could be explained by both the high inter-species variability within each taxon and the smaller difference in concentrations between taxa.

PFASs exhibited the highest contribution among the OC families in the crustacean species *Pasiphaea sivado* (65 ± 2% on average), while PCB contribution was the highest in all fish species (between 47% and 72%, with a mean of 62 ± 10%). PFAS contribution decreased significantly with both $\delta^{15}N$ values (p = 0.0001) and TLC (p = 0.004), reflecting a relationship with the trophic magnification factors (TMFs) of the studied contaminants, which are reported to be globally lower for PFASs than for PCBs (Won et al., 2018) and suggesting a potential link with the biochemical compositions of the studied species. Unlike the lipophilic contaminants studied here (i.e. PCBs, OCPs and PBDEs), PFASs have an affinity for specific proteins and phospholipids (Armitage et al., 2013; Ng and Hungerbühler, 2013). PFASs presented the highest relative concentrations in the crustacean species that showed the lower TLC (lipophilic contaminants being consequently less predominant), namely *Pasiphaea sivado*, while the lowest PFAS relative contribution (17%) was found in the crustacean species *Ephyrina figueirai,* which showed the highest TLC (and the highest lipophilic contaminant contribution). However, the two fish species *Serrivomer beanii* and *Xenodermichthys copei,* despite them having been reported to have lower protein contents than the other studied fish species sampled from the Bay of Biscay (Spitz et al., 2010), showed higher PFAS contribution than the other fish (30 ± 8% versus 10 ± 6% in the other species), a result that could rather be explained by their lower trophic level (see section 3.3.6).

3.3.2. OC concentrations are in the range of those reported in other deep pelagic species in the NE Atlantic

Although PCB, OCP and PBDE concentrations in deep-sea organisms from various oceanic regions have been published previously, most of the data refer to bathydemersal and benthopelagic species, i.e. living and feeding close to the bottom seafloor. In addition, the studied periods refer to sampling dates of more than 10 years ago which could bias the comparison for the legacy POPs. Data on contaminants

365 of emerging concern such as the ones we report here are still very scarce, making comparisons
366 impossible. With these precautions in mind, the following are some comparisons with previously
367 published data focusing specifically on deep pelagic species.

368 $\sum_6$ PCB and $\sum_{15}$ PBDE concentrations were determined in the muscles of 4 deep pelagic fish species
369 similar to ours (namely *Benthosema glaciale, Xenodermichthys copei, Argyropelecus hemigymnus,*
370 *Gonostoma bathyphilum*) and collected at 1200–1500 m depths in a submarine canyon in the south
371 Bay of Biscay in 2012–2013 ranged between 134 and 756 ng g^{-1} lw and between <LOD and 23 ng g^{-1} lw,
372 respectively (Romero-Romero et al., 2017). These levels compare with our concentration ranges (i.e.,
373 31.2–1072.9 ng g^{-1} lw and 0.92–43.37 ng g^{-1} lw for PCBs and PBDEs respectively). $\sum_7$ ICES PCB (i.e. the
374 International Council for the Exploration of the Sea PCBs, the major and mostly-used congeners) was
375 found in mean concentrations of between 91.6 ± 116 and 613 ± 739 ng g^{-1} lw in the muscle of adult
376 *Aphanopus carbo* (black scabbardfish) collected from the west of Scotland in 2006–2012 and of
377 between 267 ± 150 and 521 ± 301 ng g^{-1} lw in their livers (Webster et al., 2014), which compares closely
378 to our results in whole fish (524–651 ng g^{-1} lw), although younger individuals were considered in our
379 study. In the same individuals, the $\sum_9$ PBDEs were between 1.1 ± 3.0 and 42.5 ± 26.4 ng g^{-1} lw in the
380 flesh and between 2.4 ± 3.9 and 25.8 ± 8.2 ng g^{-1} lw in the liver (Webster et al., 2014), while our result
381 showed concentrations of a similar order of magnitude with 13.0–17.1 ng g^{-1} lw range in whole fish.
382 Various organic contaminant concentrations (namely, PCBs, HCB, dieldrin and DDTs) were reported in
383 the livers of black scabbardfish collected from the NE Atlantic (from Madeira off the coasts of Marocco
384 to Ireland) in 1999 (Mormede and Davies, 2003). Surprisingly, these contaminant concentrations did
385 not differ drastically from our data, considering the difference in time of sampling, studied areas and
386 targeted tissues ($\sum_7$ ICES PCBs: 91 ng g^{-1} lw to 7660 ng g^{-1} lw versus 524–651 ng g^{-1} lw in our fish samples;
387 HCB: 5.3 to 30.3 ng g^{-1} lw versus 11.4–18.8 ng g^{-1} lw; dieldrin: 22.0-40.9 ng g^{-1} lw versus 20.5–25.0 ng
388 g^{-1} lw) except for the ∑ DDTs, which was lower in our samples (239.5–397.8 ng g^{-1} lw versus 384–4350
389 ng g^{-1} lw in Mormede and Davies' study), potentially reflecting a higher decrease of DDT inputs over
390 the studied periods.

Specific congener analysis reveals untypical BFR profiles

Indicator PCBs (i-PCBs) were predominant compared to dioxin-like congeners (dl-PCBs) in all species, and i-PCB and dl-PCB concentrations were highly correlated ($r = 0.93$, $p < 0.05$). The average ratio (i-PCBs/dl-PCBs) was 7 ± 1 and showed no significant difference between taxa. Detailed PCB mean concentrations per congener and taxon are given in Table S3. The most abundant congener was CB-153 with concentrations ranging from 1.73 to 15.03 ng g^{-1} dw (mean of 6.663 ± 6.776 ng g^{-1} dw) in crustaceans and from 2.40 to 23.27 ng g^{-1} dw (mean of 10.71 ± 6.10 ng g^{-1} dw) in fish. Hexa- and heptachlorinated congeners (i.e. CB-153, CB-138, CB-180 and CB-187, each one contributing to 10-19% of the ∑ PCBs) were the most abundant ones, which is consistent with previous findings in deep-sea ecosystems (Koenig et al., 2013; Romero-Romero et al., 2017; Storelli et al., 2009; Takahashi et al., 2010; Webster et al., 2014). The i-PCBs contributed to 47% of the ∑ PCBs while dl-PCBs counted for 7% of the ∑ PCBs, pointing out the importance of determining more than the classic 18 congeners to better assess total PCB bioaccumulation in marine organisms. Congeners with 5 and more chlorine atoms showed the highest cross-correlations, which underline their similar sources and behaviours.

DDTs were by far the most abundant OCPs in all species (0.53–86.23 ng g^{-1} dw range, mean of 16.35 ± 18.90 ng g^{-1} dw), followed by dieldrin (0.25–6.83 ng g^{-1} dw range, mean of 1.96 ± 1.34 ng g^{-1} dw) > HCB (0.32–3.93 ng g^{-1} dw range, mean of 1.07 ± 0.91 ng g^{-1} dw) > endrin (0.028–0.73 ng g^{-1} dw range, mean of 0.25 ± 0.19 ng g^{-1} dw) = ∑ HCHs (0.01–0.61 ng g^{-1} dw range, mean of 0.20 ± 0.18 ng g^{-1} dw) (Table S4). These results bring evidence that DDT is still a major organochlorine pesticide in marine ecosystems, which is in line with its global past usage and environmental persistence (Li and Macdonald, 2005). ∑ DDTs showed significant positive correlations with all other organochlorine pesticides except ∑ HCHs, possibly related to HCH physico-chemical properties and environmental behaviour (Salvado et al., 2019). The highest correlations were between ∑ DDTs and mirex (0.91) while dieldrin and endrine showed weaker ones (0.70 and 0.42, respectively).

Among DDTs, the *p,p'-DDE* isomer was the most prevalent in all samples (80 ± 11% of ∑ DDTs), in accordance with the common profiles observed in marine biota including deep-sea organisms (Koenig

et al., 2013; Ramu et al., 2006; Storelli et al., 2009). The *o,p'*-DDT/*p,p'*-DDT concentration ratio was 0.36 ± 0.17, on average, in all fish species and showed high inter-species variations (0.03–0.55 range). This ratio is commonly used to distinguish DDT sources, with values in the 0.2–0.3 range in technical DDT (Kalantzi et al., 2001) whereas a ratio above 0.34 is usually attributed to DDT's origin from dicofol acaricide impurities (Suarez et al., 2013). This ratio was particularly low in *Stomias boa* (0.03 ± 0.01) because of peculiar low contributions of *o,p'*-DDT (0.4 ± 0.2% versus 3.3 ± 1.9% in the other fish species). These results show that the use of this ratio should be interpreted with caution and suggest that species-specific parameters may also influence DDT isomeric profiles.

BFR profiles were dominated by BDE-209 (mean contribution of 38% of the 12 summed congeners quantified in more than 50% of the samples) followed by BDE-47 (21%), BDE-155 (19%) and BDE-154 (14%). The other mostly-detected congeners (i.e. BDEs -28, -49, -66, -99, -100, 119, -126, -153) contributed each to less than 10%, on average, to the summed 12 BDEs. As BDE-47 is generally reported as the major PBDE congener in biota, the present PBDE profile is therefore atypical due to the high contribution of BDE-209. The BDE-209 congener is usually reported as poorly bioaccumulable in fish because of its very high log K_{ow} (12.1, Kelly et al., 2008), high molecular size and degradation propensity *via* metabolism (Stapleton et al. 2004a, Roberts et al., 2011). Following our results, this highly brominated congener has been previously identified at higher abundance (17% of ∑ PBDEs) in deep-sea organisms compared to those from the shelf (Romero-Romero et al., 2017). Although all fish stomachs and intestines were emptied of major debris (after visual inspection) before analysis, it cannot be ruled out that high BDE-209 concentrations were also due to non-ingested particles still present in the digestive tracts. However, the presence of a high diversity of several high-brominated congeners, as well as species-specific profiles, rather suggests BDE-209 bioaccumulation and biotransformation, as detailed in section 3.3.4 below.

Another peculiar result was the high contribution of DBDPE, quantified in 76% of the samples at concentrations as high as 16.48 ng g^{-1} dw in *Searsia koefoedi*. Similar to BDE-209, DBDPE is characterised by a very high log K_{ow} (11.1, Covaci et al. 2011) that should limit its bioaccumulation at

high levels in fish and generally leads to its biodilution in trophic webs (Tao et al., 2019). However, unlike for BDE-209, no debromination of DBDPE has been reported in fish and its bioaccumulation factor was reported to be 10 times higher than BDE-209's one (He et al., 2012), which would argue in favour of its occurrence at higher levels than BDE-209. Similar to BDE-209, the DBDPE occurrence at particularly high levels in some of the studied samples (i.e. *Serrivomer beanii, Stomias boa, Aphanopus carbo* and *Searsia koefoedi*) could be the result of undigested particles in their digestive tracts. DBDPE sources in the marine environment include plastics from electronic and electrical equipment wastes (Stubbings et al., 2021). However, whether DBDPE found in some fish originated from its adsorption on particulate matter or plastics, or plastics themselves (eventually extracted following whole fish analyses) cannot be proven. In oceanic waters off the Californian coast (Monterey Bay, US), the highest concentrations of microplastics have been reported between 200 m and 600 m (Choy et al., 2019), and microplastics have been reported in the digestive tracts of mesopelagic fish feeding at these depths, although at very different occurrence frequencies depending on locations (35% in Boerger et al., 2010; 9% in Davison and Asch, 2011; 11% in Lusher et al., 2016; 24% specifically in Myctophidae in Savoca et al., 2020; 73% in Wieczorek et al., 2018). Among the various species studied by Wieczorek et al. (2018), *Serrivomer beanii* and *Lampanyctus macdonaldi* were among those presenting the highest frequency of plastic occurrence, while *Serrivomer beanii* and *Myctophum punctatum* were two species presenting the highest number of plastic debris in their gut. Despite the digestive tracts of fish from our study being emptied from visible material before analysis, the presence of small parts of food residues (including those of plastic origin) that could have still be present in the digestive tracts (i.e. not assimilated/absorbed into the organism's tissues) is not unrealistic. The fact that high variations were sometimes found between replicates of the same species (as in *Aphanopus carbo*, with concentrations of 81, 4762 and 7803 pg g^{-1} dw in each of the three replicates made from individual fish, or in *Xenodermichthys copei* with < LOQ, 148 and 1940 pg g^{-1} dw in pooled samples) argues in favour of undigested particles with a short-time and erratic occurrence rather than a long-term accumulation in tissues. On the opposite hand, species such as *Stomias boa* showed high DBDPE concentrations in all

469 replicates (3075, 5963 and 7840 pg g^{-1} dw, rsd = 43%), while others such as *Myctophum punctatum* or
470 *Lampanyctus crocodilus* exhibited systematically lower concentrations (< LOQ, 42 and 193 pg g^{-1} dw
471 and 436, 682 and 938 pg g^{-1} dw, respectively).

472 Interestingly, in our study, the highest DBDPE concentrations (in both dw and lw) were determined in
473 the longest fishes studied, i.e. *Serrivomer beani* (64.3 ± 9.2 cm), *Stomias boa* (31.6 ± 3.7 cm) and
474 *Aphanopus carbo* (61.7 ± 0.6 cm). These species are all characterised by elongate shapes and therefore
475 have potentially longer digestive tracts, which would lead to a higher retention potential in the gut. A
476 higher occurrence of microplastics in larger fish compared to smaller ones has previously been
477 observed in various freshwater species from a lake in Ontario, Canada (McIllwraith et al., 2021).
478 Despite its relatively smaller length (15 cm), *Searsia koefoedi* exhibited the highest concentration of all
479 species (16.48 ng g^{-1} dw) but these results would need to be confirmed as only one pool of 3 individuals
480 was analysed. However, this result is consistent with this species' non-migratory behaviour (at least
481 towards epipelagic waters at night for feeding, but possible migrations deeper into the bathypelagic
482 zone; Novotny, 2018), as DBDPE has been shown to be high in deeper waters (Zhen et al., 2021). The
483 ratio between DBDPE and BDE-209 concentrations has been suggested as a useful tracer of sources
484 and processes, with a higher DBDPE/BDE-209 ratio being observed in deep waters compared to the
485 surface ones because of the higher affinity to fine particles and stability of DBDPE compared to BDE-
486 209 (Zhen et al., 2021). However, using this ratio in biota has limitations and no correlation was found
487 between BDE-209 and DBDPE in our set of samples. Among the species reputed to undergo no DVM
488 (i.e., *Arctozenus risso* and *Searsia koefoedi*), only the latter one showed a high DBDPE/BDE-209 ratio
489 (16 versus 1.2 in *Arctozenus risso*). Indeed, this ratio was highly variable between species (median value
490 of 3.0 in all fish samples), ranging from 0.11 in *Myctophum punctatum* (the only species in which BDE-
491 209 was higher than DBDPE) to 19 in *Aphanopus carbo*. Globally, our results highlight that mesopelagic
492 fish could, in addition to transferring very hydrophobic contaminants to higher trophic level organisms,
493 contribute to their transfer from surface waters to deeper waters and eventually to the bottom sea.

3.3.3. Chlorinated and brominated OC profiles and diagnostic ratios reveal species-specific metabolic capacities

Because PCB bioaccumulation depends on their chlorine number and position, which both are source- and metabolism-related, PCB profiles can be examined by their number of Cl atoms and their structure-activity group (SAG) classification (Boon et al., 1997; Yunker et al., 2011). While the lower-chlorinated congeners are associated with atmospheric transport and feeding at lower trophic levels, the heavier congeners reflect continental inputs from rivers and prey of higher trophic levels. In our samples, 3-Cl and 4-Cl showed higher contributions to the ∑ PCBs in crustaceans compared to fish, which is consistent with crustaceans' lower trophic levels (i.e. lower $\delta^{15}N$ values), although the difference was significant only for the 3-Cl congeners (10 ± 5% in crustaceans versus 4 ± 5% in fish, MW and KW tests). However, inter-species differences were high when only fish were considered, and similar contributions to those of crustaceans were found in some fish species such as *Arctozenus risso*, *Argyropelecus olfersii* and *Myctophum punctatum* (Fig. 1A). These species are among those showing the lowest $\delta^{15}N$ values among the studied fish species, but other species with low $\delta^{15}N$ values (such as *Serrivomer beanii* or *Xenodermichthys copei*) did not show such a high 3-Cl contribution (Fig. 1A), suggesting that the trophic level does not solely explain these results. Indeed, the lower chlorinated congeners are also the most metabolisable ones (SAG III and IV), while the most refractory ones (6-,7- and 8-Cl) belong to SAG I and II groups. No significant differences were observed between taxa regarding SAG groups. The three crustacean species showed discrepancies in their PCB profiles, with *Sergia robusta* being characterised by higher SAG I and II group contributions (72% in total) than the other two species (52 ± 5%). Among fish species, *Arctozenus risso*, *Argyropelecus olfersii* and *Myctophum punctatum* showed the highest SAG III and IV (metabolisable congeners) and the lowest SAG I and II (refractory congeners) contributions. On the opposite hand, *Serrivomer beanii*, *Xenodermichthys copei*, *Aphanopus carbo*, *Lampanyctus crocodilus* and *Searsia koefoedi* were characterised by higher SAG I and II (6 to 8 Cl) contributions (Fig. 2). More specifically, the CB-149/CB-153 and CB-132/CB-153 concentration ratios, which could be used as metabolism tracers (i.e. a ratio

between hexachlorinated congeners from SAG IV and V and a typical refractory one from SAG I), showed higher values in *Arctozenus risso*, *Argyropelecus olfersii*, *Myctophum punctatum* and *Notoscopelus kroeyeri,* indicating a lower metabolic activity towards CB-149 and CB-132 in these species (Fig. 2).

The (*p,p'*-DDE + *p,p'*-DDD)/Σ DDTs ratio was 0.85 ± 0.06 in all samples, which globally indicates old DDT inputs (Suarez et al., 2013) and is consistent with the ban of DDT usage since the 1970s or 1980s in most countries worldwide (Kalantzi et al., 2001). The *p,p'*-DDE isomer contribution was significantly (MW and KS $p < 0.1$) higher in crustaceans and particularly high in *Pasiphaea sivado* replicates, which was mainly due to *p,p'*-DDD and *p,p'*-DDT < LOQs in these samples. Some variations in *p,p'*-DDE contributions were also observed between fish species (Fig. 1B). Among Myctophidae, both *Myctophum punctatum* and *Notoscopelus kroeyeri* exhibited similar DDT profiles (*p,p'*-DDE and *p,p'*-DDD at 65 ± 5% and 12 ± 1%, respectively) and were different from those of the third Myctophid species *Lampanyctus crocodilus*, in which higher *p,p'*-DDE (84 ± 2%) and lower *p,p'*-DDD (5 ± 1%) contributions were found. These results could suggest a species-specific DDT profile due to different metabolic capacities, which would be in line with the higher metabolic capacity of PCBs observed in *Lampanyctus crocodilus.* In addition, *Lampanyctus crocodilus* was the leanest of the three studied Myctophid species (by a factor of 4 to 5, Table 2), which could influence DDT isomer's relative bioaccumulation. Indeed, in the studied fish species, *p,p'*-DDE contribution showed a significant decrease with increasing TLC ($p < 0.0001$).

Regarding PBDE profiles (Fig. 1C), the nona-BDEs BDE-207 and BDE-208, the octa-BDEs BDE-202, BDE-205 and BDE-197 and the hepta-BDEs BDE 184 and BDE-183 were quantified in 18–48% of our samples, although at low levels (their total contribution was 6% of the sum of all congeners determined above LOQs). In addition, the hexa-BDEs BDE-155 and BDE-154, ranking third and fourth in our samples, have both been reported as debromination products of octa-BDEs (Stapleton et al., 2004b; Zeng et al., 2012). As the nona- to hexa-brominated congeners identified in our samples are not present in technical mixtures (La Guardia et al., 2006; Munschy et al., 2011), their occurrence might be explained

by either i) assimilation and further metabolisation by fish or ii) metabolisation by the prey and bioaccumulation in fish *via* trophic transfer. Indeed, these congeners are among those (ranging from nona- to tetra-BDEs) that have been reported in various fish species following BDE-209 metabolic degradation (Roberts et al., 2011). BDE-154 could result from the debromination in the *meta* position of BDE-183, while BDE-155 could result from successive debrominations in the *meta* positions of BDE-207, BDE-197 and BDE-184, which were all detected in our samples, although at low concentrations compared to their debrominated counterparts. If these congeners originate from BDE-209 metabolism, their detection at concentrations above the LOQs reveals exposure of the studied species to BDE-209. The high variability in PBDE profiles between species could therefore result from species-specific metabolism, as evidenced by various studies in fish (Roberts et al., 2011; Stapleton et al., 2006; Yokota et al., 2021). The results suggest that BDE-183, BDE-197 and BDE-207 could be potential intermediate debromination products of BDE-209 resulting in the formation of BDE-154, and that the removal of Br atoms in the *meta* position was favoured. However, no significant relationship was found between BDE-209 and either BDEs -183, -197 or -207 (although the latter two congeners showed low detection frequencies), nor with BDE-154 or BDE-155. Interestingly, BDE-154 and BDE-155 were highly correlated ($n = 28$, $r = 0.96$) in fish, which could reflect their common origin (i.e. BDE-209 biotransformation).

Specific ratios such as BDE-99/BDE-100 concentration ratios (BDE-99 being metabolised while BDE-100 is not, a low ratio is indicative of a high metabolic capacity towards BDE-99) or congener relative contributions are commonly used to reveal metabolic capacities in fish (Koenig et al., 2013; Voorspoels et al., 2003). In our samples, BDE-99/BDE-100 ratios were highly variable depending on species and were highly consistent between replicate pools within the same species. The lowest ratios were identified in *Lampanyctus crocodilus* (0.11 ± 0.03), while the highest were found in *Argyropelecus olfersii* (1.56 ± 0.56). In *Xenodermichthys copei*, BDE-99 was below the LOQ in the three replicate pools, suggesting high degradation of BDE-99 in this species. This ratio was not trophic level-dependent but rather highly species-dependent (inter- and intra-family). Indeed, within the Myctophydae family, high

variations were found, with both *Myctophum punctatum* and *Notoscopelus kroeyeri* showing high values (1.36 ± 0.23 and 1.29 ± 0.18, respectively), while *Lampanyctus crocodilus* showed a mean ratio of 0.11 ± 0.03 (i.e. a higher metabolism). Concurrently, BDE-154's highest contributions were determined in *Lampanyctus crocodilus* (20 ± 6.2), indicating a higher degradation capacity in this species. Indeed, as BDE-154 could potentially originate from the debromination of higher-brominated congeners, its relative contribution would be indicative of the metabolic capacities of fish, with a high contribution being indicative of a higher degradation capacity into BDE-154.

3.3.4. Perfluorinated substances' molecular profiles are dominated by odd chain length PFCAs

PFOS was the only PFSA detected in 100% of the samples, at concentrations ranging from 0.38 to 10.61 ng g^{-1} dw (mean of 3.72 ± 3.9615 ng g^{-1} dw) in crustaceans and between 0.350 and 10.04 ng g^{-1} dw (mean of 3.00 ± 2.22 ng g^{-1} dw) in fish. PFDS was detected in 27% of the samples (crustaceans and fish) at levels ranging from 0.04 to 0.09 ng g^{-1} dw (mean of 0.06 ± 0.02 ng g^{-1} dw). Among PFCAs, the long-chain PFCAs perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUnDA), perfluorododecanoic acid (PFDoDA), perfluorotridecanoic acid (PFTrDA) and perfluorotetradecanoic acid (PFTeDA) were detected in 100% of the samples, while perfluorooctanoic acid (PFOA) was detected in 45% of the samples. Perfluoroheptanoic acid (PFHpA) was detected in only four samples (*Sergia robusta*, *Xenodermichthys copei* and *Searsia koefoedi*) at a mean concentration of 0.05 ± 0.03 ng g^{-1} dw. The mean concentrations of the most-detected PFCA ranged from 0.18 ± 0.17 ng g^{-1} dw (PFOA) to 4.40 ± 4.51 ng g^{-1} dw (PFTrDA). PFCA concentrations ranked in the order PFTrDA > PFUnDA > PFNA = PFDA > PFTeDA > PFDoDA > PFOA, showing higher bioaccumulation with increasing carbon chain length, which is consistent with their higher BAF (Houde et al., 2006; Martin et al., 2003; Ng and Hungerbühler, 2014). In fish, all PFCA concentrations except PFOA's were: i) significantly inter-correlated and ii) correlated with PFOS concentrations but iii) not correlated to the other contaminant families. This could reflect different sources and/or bioaccumulation processes. In most samples, PFCAs were predominant compared to PFOS (by far the predominant PFSA) with a mean contribution

598 of 75 ± 12% (mean PFCAs/PFOS ratio = 5.4 considering all species), although some variations were
599 observed within taxon and species (Fig. 1D). PFCA contributions were significantly ($p = 0.002$) higher in
600 crustaceans (89 ± 6%) than in fish (72 ± 11%). The crustacean species *Ephyrina figueirai* showed a
601 peculiar high PFCA contribution (Σ PFCAs/PFOS ratio of 59) due to a low PFOS concentration (0.379 ng
602 g^{-1} dw), while its ∑ PFCA concentration was similar to the ones determined in the other crustacean
603 species *Pasiphae sivado*. On the opposite hand, *Chauliodus sloani* (although only one sample was
604 analysed) was the only species with lower ∑ PFCA concentrations than PFOS ones, with a ∑ PFCAs/PFOS
605 ratio of 0.75, due to a particularly high PFOS contribution (57% of ∑ PFASs). The ∑ PFCA contribution
606 was also higher (87 ± 2%) in *Serrivomer beanii*, the fish species with the lowest $\delta^{15}N$ values and
607 comparable with the ratio determined in crustaceans.

608 The occurrence of the longer chain PFCAs (> C_8) in biota is commonly explained by their release in the
609 environment as impurities from fluorotelomer and C_9-based products (Wang et al., 2014) and their
610 subsequent higher bioaccumulation compared to the shorter chain ones (Houde et al., 2006; Conder
611 et al., 2008; Ng and Hungerbühler, 2014). Besides, PFCAs might also originate from the *in vivo*
612 biotransformation of precursors, which would increase their biomagnification in food webs (Gebbing
613 et al., 2016). Various experimental studies showed that when selected fluorotelomer alcohol (FTOH)
614 precursors were administrated to fish, PFOA, PFNA, PFDA and PFHpA were formed, although with low
615 yields (Brandsma et al., 2011; Butt et al., 2014). However, recent results showed that PFCAs could also
616 be formed by yet-unidentified precursor degradation (Simmonet-Laprade et al., 2019) and suggest that
617 direct exposure to PFCAs is not a predominant source to explain PFCA profiles in fish. The
618 predominance of the odd-numbered PFCAs PFTrDA (C_{13}) and PFUnDA (C_{11}) has been previously
619 reported in marine fish worldwide (Fujii et al., 2015, 2019; Munschy et al., 2020b; Schultes et al., 2020
620 and references therein) and explained by direct inputs from industrial releases (Gewurtz et al., 2013;
621 Simonnet-Laprade et al., 2019) and/or by the degradation of precursors (FTOHs), either in the
622 atmosphere, soil or organisms, followed by preferential accumulation of the longer chain length PFCAs
623 (i.e. $C_{11} > C_{10}$ and $C_{13} > C_{12}$). However, FTOH degradation leads to 10-times more abundant even chain

length PFCAs than odd chain length PFCAs (Franklin et al., 2016; Gebbink et al., 2016). In our samples, the mean $(C_{11} + C_{13})/(C_{10} + C_{12})$ ratio was 4.2 ± 1.4, showing a higher occurrence of odd chain length PFCAs in all species (no significant difference was observed between taxa). Washington et al. (2020) used the $(C_{11} + C_{13}) - (C_{10} + C_{12})$ values to reveal PFAS sources in soils, allowing them to distinguish between direct release (higher values) and fluorotelomer degradation. In biota, this parameter could be influenced by *in vivo* degradation of precursors into PFCAs, which are not expected to further degrade in organisms. This ratio could therefore give an integrated view of direct exposure and subsequent bioaccumulation, combined with the effect of precursor biotransformation. Interestingly, this ratio showed strong differences between i) the crustaceans and the fish species *Serrivomer beanii* and *Searsia koefoedi* that show high values (14420 ± 3968) and ii) the other fish species that presented lower values (3243 ± 2360). The higher $(C_{11} + C_{13}) – (C_{10} + C_{12})$ values suggest that these PFCAs were less derived from precursors in crustaceans and the fish species *Serrivomer beanii* and *Searsia koefoedi* than in the other fish species. This assumption would agree with species-specific precursor biotransformation as already reported for PFOS in freshwater invertebrates and fish (Babut et al., 2017) and would suggest a lower precursor metabolism in these species.

3.3.5. Most of the chlorinated and brominated OCs show biomagnification across the studied food chain, while PFASs exhibit a contrasted behaviour

Despite legacy POP concentrations in deep-sea organisms (mostly demersal or benthic) having been reported in the literature, very few studies have examined their biomagnification in deep-sea food webs (but see Cresson et al., 2016) and especially in the deep pelagic. In the present study, significant linear relationships were found between PCB log-transformed concentrations in lw and $\delta^{15}N$ values (Table S6). However, CBs -18, -28 and -31 showed significant negative linear relationships with $\delta^{15}N$ values, indicating their biodilution in the food web, while the higher chlorinated congeners (5 chlorine atoms and above) showed significant positive linear relationships (Fig. 3A, Table S7). Among DDT isomers, *p,p'*-DDE, *p,p'*-DDD and *p,p'*-DDT, but neither *o,p'*-DDT or *o,p'*-DDD, concentrations showed significant positive linear relationships with $\delta^{15}N$ values, as well as dieldrin, endrin, HCB and mirex (Fig.

650 3A, Table S8). On the opposite hand, neither ∑ HCHs nor individual isomers showed any significant
651 relationship. ∑ PBDEs and individual congeners, except BDEs -28, -66, -77, and -183, showed significant
652 positive linear relationships with $\delta^{15}N$ values (Fig. 3B, Table S9). Among the alternative brominated
653 flame retardants, BB-153 showed significant positive linear relationships ($p = 0.005$), while HBB did
654 not. These results agree with the biomagnification of lipophilic compounds in marine pelagic food webs
655 reported previously, including in deep-sea ecosystems (Romero-Romero et al., 2017; Takahashi et al.,
656 2010). Indeed, the congeners showing no significant relationship with $\delta^{15}N$ values are those with the
657 lowest reported trophic magnification factors (TMFs) (Walters et al., 2016). The lack of
658 biomagnification of the lower-chlorinated PCB congeners such as CBs -18, -28, -31, HCHs and HBB
659 might be related to their moderate hydrophobicity and potential elimination *via* respiration (Kelly et
660 al., 2007; Takahashi et al., 2010). Besides, low TMFs might also result from the compound
661 hydrophobicity and size, which limit their bioaccumulation (e.g. BDE-209). The limited extent of the
662 trophic level covered by the food web studied here (i.e. one theoretical trophic level if referring to the
663 average difference of 3.4‰ reported between two trophic levels, Post, 2002) might also have
664 restrained the possibility of detecting high contaminant biomagnification for lipophilic compounds
665 (Brandsma et al., 2015; Won et al., 2018).

666 Peculiar results were obtained for BDE-209 and DBDPE: both compounds are superhydophobic ones
667 and their concentrations (in lw) showed contrasting relationships with $\delta^{15}N$ values; while BDE-209
668 concentrations were not significantly related to $\delta^{15}N$ values, DBDPE exhibited biomagnification (Fig.
669 3B, Table S9), which could be due to its presence at high levels in *Stomias boa*, *Aphanopus carbo* and
670 *Searsia koefoedi* (see section above).

671 Among PFASs, individual PFCAs showed a significant decrease with increasing $\delta^{15}N$ values when both
672 crustaceans and fish were considered, while PFOS showed no significant trend. However, no significant
673 trend was detected in fish only, whether individual PFCAs or PFOS were considered (Fig. 3C, Table S10),
674 which shows that the significant relationship was due to the influence of the high concentrations
675 observed in crustaceans. This lack of observed biomagnification for both PFOS and PFCAs in fish

676 contrasts with some previously reported results. Indeed, various PFASs, including PFOS and long-chain
677 PFCAs have been reported to biomagnify in marine trophic webs (Loi et al., 2011; Munoz et al., 2017;
678 Pan et al., 2021; Tomy et al., 2004), but some opposing results lacking evidence of long-chain PFCA
679 biomagnification have also been reported when only piscivore food webs were considered (Du et al.,
680 2021; Kelly et al., 2009; Mazzoni et al., 2020; Miranda et al., 2021; Pan et al., 2021). This has partially
681 been explained by PFCA's high aqueous solubility (due to the carboxylate functional group) and
682 preferential distribution in blood, leading to their efficient respiratory elimination *via* blood-water
683 exchange in the gills (Kelly et al., 2009). Our results show the complexity of interpretation of PFAS
684 bioaccumulation along marine trophic webs in a given ecosystem, as the observed results could be
685 reflecting different accumulation and/or depuration processes depending on species and potential
686 metabolic capacities towards precursors.

687 ***3.3.6. PCBs and PBDEs exceed OSPAR BACs in the majority of samples***

688 To identify marine regions of potential environmental concern, OSPAR (Oslo/Paris Convention for the
689 Protection of the Marine Environment of the North-East Atlantic) defined Background Assessment
690 Concentrations (BACs) for PCBs (OSPAR, 2017). BACs are used as thresholds to assess whether
691 concentrations are close to background levels. Although these values were initially defined in the
692 muscle of flatfish in coastal areas, a comparison was made with the present set of samples based on
693 the determination of concentrations in whole fish. All samples showed higher concentrations (by a
694 factor of 11-24 on average) than the corresponding BACs for CBs -138 (0.09 ng g^{-1} ww), -153 (0.10 ng
695 g^{-1} ww) and -180 (0.11 ng g^{-1} ww), while CBs -52, -105 and -118 levels were above the BACs in 86% of
696 the samples (by a factor of 3, 2 and 5, respectively). CBs -28 and -156 were above BACs in 57% of the
697 samples (3 and 1.5 times respectively). Similarly, a BAC of 0.065 ng g^{-1} lw was recommended for each
698 PBDE congener BDEs -28, -47, -66, -85, -99, -100, -126, -153, -154, -183 and -209 (OSPAR, 2021). The
699 BACs were exceeded for the predominant congeners BDEs -47, -9, -100 and -154 in more than 80% of
700 the samples (by 33, 7, 17 and 27 times, respectively), while BDEs -28, -66, -126, -153 and -209 BACs
701 were exceeded in 34% (by 1.4 times, on average), 44% (1.4 times), 44% (1.8 times), 63% (3 times) and

53% (100 times) of the samples, respectively. BDE-183 was barely above the BAC (3% of the sample by 1.6 times) and BDE-85 was never detected above the LOQ. In addition, Environmental Assessment Criteria (EACs, in lw) defined as concentrations below which biological effects are unlikely to occur have also been established in fish. EACs were exceeded only for CB-118 (25 ng g^{-1} lw) in 30% of the samples (*Lampanyctus crocodilus*, *Chauliodus sloani*, *Stomias boa* and *Aphanopus carbo*), in agreement with the OSPAR intermediate assessment conducted within the period 1995–2015 (OSPAR, 2017).

4. Conclusions and implications for higher trophic level consumers

The results obtained in the present study bring evidence of the contamination of deep-sea pelagic organisms from the Bay of Biscay by both legacy POPs and substances of emerging concern, showing that major organohalogen contaminant families (whether chlorinated, brominated or fluorinated) reach meso- and bathypelagic ecosystems. Despite being regulated for decades, PCBs were the major organic contaminant family in fish followed by OCPs, making chlorinated organic contaminants the major contributors to the targeted halogenated ones. On the opposite hand, PBDEs contributed the least to the contaminant load in both taxa. PFASs ranked third in fish while in crustaceans, PFAS and chlorinated contaminant contributions were similar. The significant contribution of PFASs (and among them the long-chain ones) to the load of organic contaminants in the studied deep-sea species, in addition to the lack of data on a large number of emerging contaminants, emphasises the importance of considering this family of compounds together with the legacy POPs in future studies.

Altogether, contaminant profiles and specific ratios suggest that the studied species exhibited metabolic capacities, especially towards PBDEs, and that the metabolic activity was highly species-dependent. Selective bioaccumulation of the investigated OC families was evidenced and shown to be related to taxa and species, trophic parameters and potential metabolic capacities. While most chlorinated and brominated contaminants showed biomagnification along the studied trophic assemblage, most PFASs showed biodilution.

This high inter-species variability observed in terms of OC concentrations induce important consequences in terms of matter fluxes in oceanic ecosystems. Variability in species abundance will

have a direct impact on the total amount of the different OCs that will be spatially transferred in the environment during vertical migration, as well as transfer to higher trophic level through food webs. For example, in oceanic waters of the Bay of Biscay, the lipid-rich Myctophid species *N. kroeyeri* was shown to largely dominate the diet of oceanic common dolphins (55% by mass of fresh reconstructed diet, Pusineri et al., 2007) while the lean Alepocephalid *X. copei* is not consumed at all. At a similar fresh mass of prey ingested, the consumption of *N. kroeyeri* thus represents a much higher intake of PCBs, OCPs and PBDEs for common dolphins than if they consumed *X. copei* (1.8 to 2.6 higher on average depending on OCs). On the other hand, *N. kroeyeri* provides 6 times less PFASs than *X. copei* at equivalent fresh mass ingested. Finally, in the perspective of the developpement of fishing activities targeting these mesopelagic ressources, our study pinpoints a potential issue regarding matter transfers, considering an exploitation of a selection of species and given the high inter-species variability in OC concentrations.

In conclusion, our results provide essential data for understanding and predicting some impacts of anthropogenic activities on deep pelagic ecosystems, filling a gap regarding the need to increase knowledge on the fate of human-induced organic contamination in the deep ocean. These original results may also allow a better assessment of contaminant vertical transport and transfer to higher trophic levels. In the actual context of climate change and global increase of human pressures, which might affect contaminant cycle dynamics and increase chemical pressures, it appears crucial to better monitor, characterise and understand chemicals' behaviour in offshore marine environments including in the deep sea. To these ends, more efforts are still needed to further assess the impact of the anthropogenic chemical contamination on deep-sea species and, ultimately, on ecosystem functioning.

Credit author statement

Catherine Munschy: Conceptualisation, Investigation, Formal analysis, Writing - original draft, review & editing. Jérôme Spitz: Conceptualisation, Funding acquisition, Sample acquisition, Writing - review & editing. Nadège Bely: Data acquisition. Karine Héas-Moisan: Data acquisition. Nathalie Olivier: Data acquisition. Charles Pollono: Data acquisition, Validation. Tiphaine Chouvelon: Conceptualisation, Funding acquisition, Sample acquisition, Writing - review & editing.

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Fig. 1. Relative contributions (% of the summed compounds) of (A) individual PCBs (presented per number of chlorine atoms), (B) DDTs (5 isomers, DDDs in green, DDTs in red/orange), (C) PBDEs (12 congeners, tri-, tetra, penta-, hexa- and deca-Br congeners are presented in blue, green, orange, purple and brown, respectively) and (D) PFASs (odd-chain PFCAs in green, even-chain PFCAs in orange) in deep pelagic crustacean and fish species collected in the Bay of Biscay in October 2017. Species names according to Table 1. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

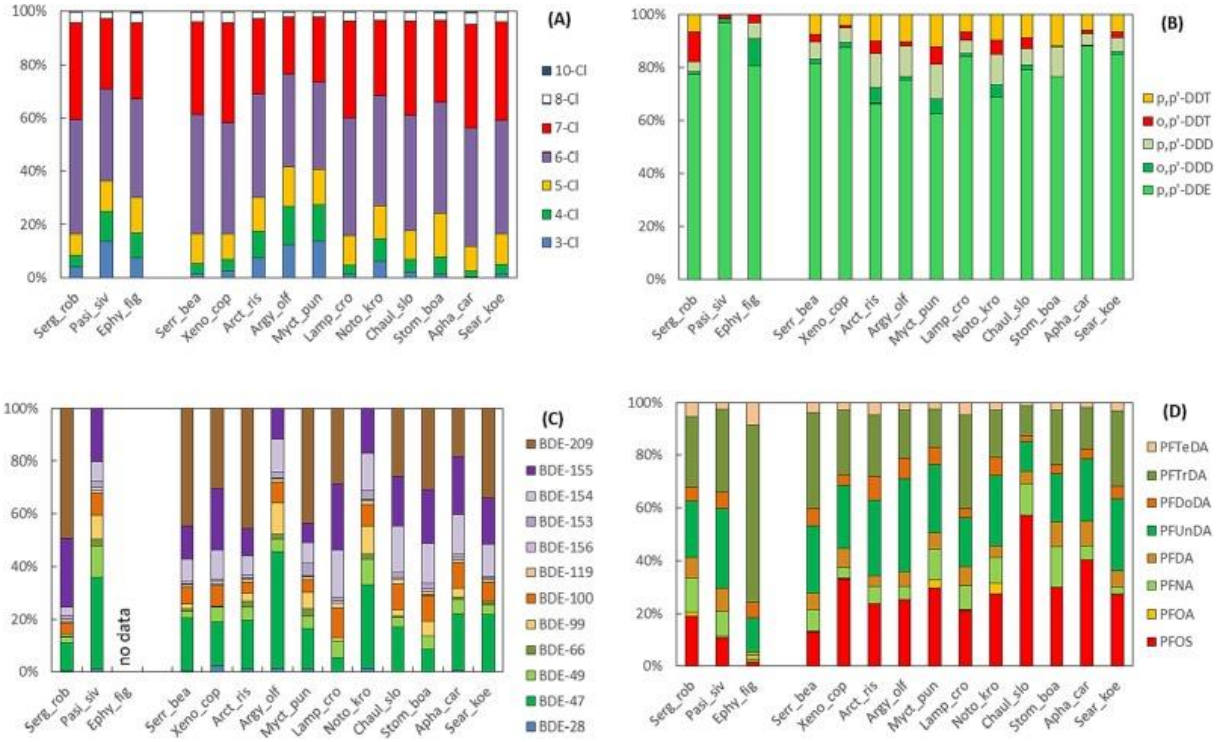

Fig. 2. Projection of the principal component analysis conducted on normalised profiles of PCBs categorized according to their SAG group in deep pelagic fish species collected in the Bay of Biscay in October 2017. Projection of variables (A) and individuals (B) on the first two principal components are presented. CB-149/CB-153 and CB-132/CB-153 were used as supplementary variables. Species abbreviations Ac = *Aphanopus carbo*, Ao = *Argyropelecus olfersii*, Ar = *Arctozenus risso*, Cs = *Chauliodus sloani*, Lc = *Lampanyctus crocodilus*, Mp = *Myctophum punctatum*, Nk = *Notoscopelus kroeyeri*, Sb = *Serrivomer beanii*, Sbo = *Stomias boa*, *Sk* = *Searsia koefoedi*, Xc = *Xenodermichthys copei*.

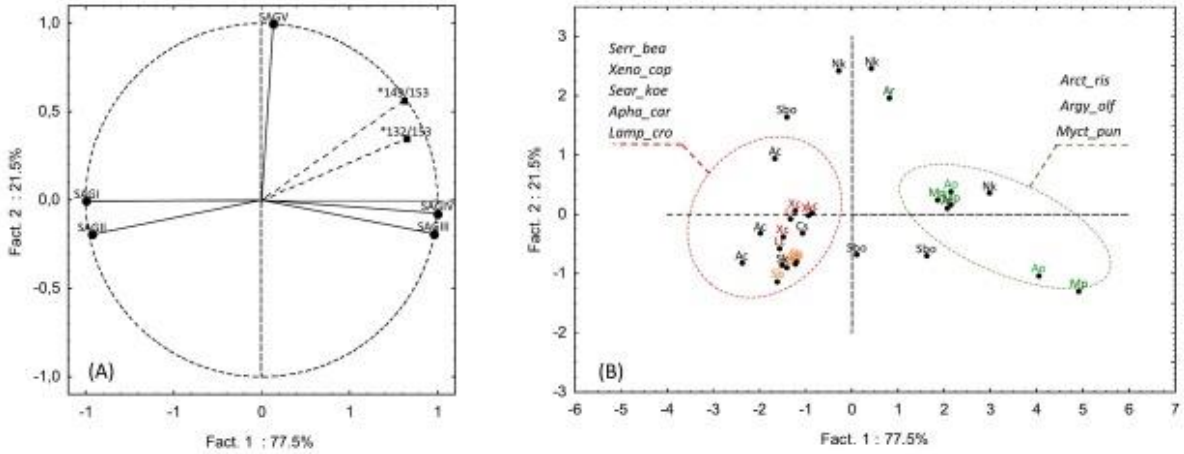

Fig. 3. Relationships between organic contaminant concentrations and $\delta^{15}N$ values (‰) in crustaceans (black dots) and fish (white dots) collected in deep pelagic waters of the Bay of Biscay in October 2017. Concentrations of organochlorinated compounds (A) and organobrominated compounds (B) were log-transformed and expressed in ng g^{-1} lw and pg g^{-1} lw respectively; PFAS concentrations (C) were log-transformed and expressed in ng g^{-1} dw. Significant relationships are indicated by dashed lines when significant for crustaceans and fish together (and for fish alone as well), by continuous lines when significant for fish only, by dotted lines when significant for crustaceans and fish only (i.e. not for fish alone). General linear model statistical parameters are given in Tables S7–S10.