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

Munsch Catherine <sup>1,\*</sup>, Spitz J. <sup>2,3</sup>, Bely Nadege <sup>1</sup>, Héas-Moisan Karine <sup>1</sup>, Olivier Nathalie <sup>1</sup>, Pollono Charles <sup>1</sup>, Chouvelon Tiphaine <sup>1,3</sup>

<sup>1</sup> Ifremer, CCEM Contamination Chimique des Ecosystèmes Marins, F-44000 Nantes, France

<sup>2</sup> Centre d'Etude Biologique de Chizé (CEBC), UMR 7372, Université de La Rochelle / CNRS, 79360 Villiers-en-Bois, France

<sup>3</sup> Observatoire PELAGIS, UAR 3462, Université de La Rochelle / CNRS, 17000 La Rochelle, France

\* Corresponding author: Catherine Munsch, email address : [cmunsch@ifremer.fr](mailto:cmunsch@ifremer.fr)

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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  $\delta^{15}\text{N}$  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 PFCA showed biodilution with increasing  $\delta^{15}\text{N}$  values.

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

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## 55 1. Introduction

56 The deep-sea pelagic environment (< 200 m) is one of the largest ecosystems on Earth and supports a  
57 high diversity and abundance of marine species, especially in the bathyal horizon (< 2000 m) (Rogers,  
58 2015). In particular, meso- (< 1000 m) and bathypelagic (1000–2000 m) communities represent  
59 essential components of oceanic biomass and important prey for higher trophic levels including large  
60 pelagic fish (tunas, sharks), marine mammals and seabirds. They play a key role in the cycling and  
61 vertical transfer of matter and energy in oceans (Bernal et al., 2015; Bianchi et al., 2013; Catul et al.,  
62 2011). As a consequence of their strong diel vertical migrations (DVM) during which some species  
63 (Chouvelon et al., 2022; Eduardo et al., 2021; Takahashi et al., 2000) feed in surface waters (epipelagic  
64 zone, 0–200m) at night and move back to meso- and bathypelagic zones during the day, meso- and  
65 bathypelagic organisms release particulates, organic matter and associated contaminants *via* faecal  
66 pellet egestion and may therefore increase transfers from surface waters to deeper horizons (Belcher  
67 et al., 2019; Bernal et al., 2015). Furthermore, their upward migration, during which they become  
68 available to surface predators, leads to the transfer of organic matter and contaminants accumulated  
69 in their tissues from deep horizons back to epipelagic layers. In addition to their essential role in  
70 biogeochemical cycles, mesopelagic organisms have also raised interest regarding their exploitation as  
71 new resources for human consumption as well as the fish meal and oil industry (Berntssen et al., 2021;  
72 Grimaldo et al., 2020).

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

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

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

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

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

132

133 **2. Materials and methods**

134 **2.1. Sampling**

135 Samples were collected on the French slope of the Bay of Biscay (NE Atlantic) during the annual EVHOE  
136 fishery survey, on board the Ifremer R/V Thalassa in October 2017  
137 (<https://doi.org/10.17600/17002300>). Fish (11 species) and crustacean (3 species) samples were  
138 collected at night using a 25 m vertical opening pelagic trawl in the deep scattering layer (ca 800 m  
139 depth in the water column; 1330 m bottom floor). All samples were collected during the same haul of  
140 60 min at a speed of approximately 4 knots. The selected fish individuals belonged to the most  
141 abundant species, including one species of Sternoptychidae and three species of Myctophidae, two of  
142 the most abundant mesopelagic fish families globally (Catul et al., 2011; Valinassab et al, 2007).

143 Handling of samples was conducted on-board using rigorous protocols to avoid external  
144 contamination. Samples were stored at  $-20^{\circ}\text{C}$  until further processing in the laboratory. To obtain  
145 sufficient material for the quantification of OCs, the whole bodies of individuals belonging to the same  
146 species were pooled by specimens of similar sizes (Table 1). Considering the sizes reported in the  
147 literature, most fish except *Aphanopus carbo* were adult fish. A small piece of white muscle (< 3% of  
148 individual total weight) was also collected for the analysis of stable isotopes of carbon and nitrogen as  
149 trophic tracers. Whenever possible, fish individuals' sexes were determined and noted in the  
150 composition of each pool; all pools were made of both male and female individuals except for  
151 *Serrivomer beanii* (males only) and *Stomias boa* (females only). All fish stomachs were emptied from  
152 their major visible content; however, most stomachs were found to be empty, suggesting either a delay  
153 between the sampling and the last feeding or a fast stomach emptying. When large enough, fish were  
154 analysed individually; this was the case for 3 samples of *Serrivomer beanii*, 2 samples of *Stomias boa*  
155 and 3 samples of *Aphanopus carbo* (Table 1). After pooling, the samples were homogenised using a  
156 blender with stainless steel arms, freeze-dried and finely ground up with a ball mill MM400 (Retsch)  
157 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- $\mu$  (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  $\Sigma$  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<sup>®</sup>,  
183 Waters Corp.) coupled to a triple quadrupole mass spectrometer (Xevo<sup>®</sup> TQ-S micro, Waters Corp.)



184 interfaced with an electrospray ionisation source Z-spray™ (Waters Corp.). The mass spectrometer  
185 was operated in negative ionisation mode using multiple reaction monitoring (MRM) with argon as the  
186 collision gas. PFASs were analysed for five C<sub>4</sub>- to C<sub>10</sub>-perfluoroalkyl sulfonates (PFASs) and nine C<sub>6</sub>- to  
187 C<sub>14</sub> perfluorocarboxylic acids (PFCAs), namely: perfluorobutane sulfonate (PFBS); perfluorohexane  
188 sulfonate (PFHxS); perfluoroheptane sulfonate (PFHpS); perfluorooctane sulfonate (PFOS);  
189 perfluorodecane sulfonate (PFDS); perfluorohexanoic acid (PFHxA); perfluoroheptanoic acid (PFHpA);  
190 perfluorooctanoic acid (PFOA); perfluorononanoic acid (PFNA); perfluorodecanoic acid (PFDA);  
191 perfluoroundecanoic acid (PFUnDA); perfluorododecanoic acid (PFDoDA); perfluorotridecanoic acid  
192 (PFTrDA) and perfluorotetradecanoic acid (PFTeDA).

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

### 200 **2.3. Statistical analyses**

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

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

### 214 **3. Results and discussion**

#### 215 **3.1. Trophic markers**

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

#### 226 **3.2. Total lipid contents (TLCs)**

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

235 *Myctophum punctatum* and *Notoscopelus kroeyeri*, the Stomiidae *Stomias boa* and the Platytroctidae  
236 *Searsia koefoedi*. These results agree with the energy densities reported by Spitz et al. (2010) in various  
237 forage fish species from the Bay of Biscay. Except in *Stomias boa* for which the replicate made from 3  
238 individuals showed extremely different values compared to the two other replicates (made of 1  
239 individual each), TLCs were fairly similar between replicate pool samples of the same species, with an  
240 average rsd of 24% (12-34% range excluding *Stomias boa*). The TLC values in *Stomias boa* replicates (all  
241 female individuals) showed a higher rsd (69%), with high TLC values of 17.6% dw and 19.5% dw for the  
242 two 35 cm and 32 cm individuals respectively, whilst the replicate made from 3 individuals of  $27.7 \pm$   
243 1.5 cm (on average) exhibited a much lower TLC value, i.e. 2.7% dw. The only noticeable difference  
244 noted during dissection was a slight difference between the maturity stages (based on gonad visual  
245 observation) with a less mature stage for the smaller individuals. Concomitantly to the maturity stage,  
246 ontogenic differences may also explain the difference observed in the lipid contents of *Stomias boa*  
247 samples, as energy reserves were shown to be positively correlated with size at the fish species level  
248 (Anthony et al., 2000; Cargnelli & Gross, 1997). However, at the scale of the fish community analysed  
249 here, TLCs showed a significant linear inverse relationship with fish lengths ( $p = 0.014$ ), indicating a  
250 higher energy content for the smaller species. Classically, TLCs showed an inverse significant linear  
251 relationship ( $p < 0.001$ ) with humidity percentages (Spitz et al., 2010).  
252 TLC values were similar to those reported previously in the literature, even though different extraction  
253 methods were used (chloroform:methanol:water (1:2:1) in Sen Özdemir et al., 2019 and ether-ethyl in  
254 Spitz et al., 2010 versus hexane and acetone (80:20 v:v) for our samples).

### 255 **3.3. Bioaccumulation of organohalogen contaminants in the deep pelagic ecosystem**

#### 256 **3.3.1. Organic contaminant concentrations and relative contributions show high inter-species** 257 **variabilities**

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

261 In both crustaceans and fish species, PCBs showed high detection frequencies. Most PCB congeners  
262 showed detection frequencies of 100%, except for CB-77 and CB-189 (97%), CB-126 and CB-169 (91%  
263 and 58% of the samples, respectively) (Table S3), whilst CB-81 was seldom detected above the LOQ  
264 (6% of the samples). CBs -29, -30, -112 and -114 were never detected. Among OCPs, dieldrin, endrin,  
265 and mirex were detected in all samples, similarly to the DDT isomer *p,p'*-DDE, whilst the other DDT  
266 isomers showed detection frequencies in the 88–94% range (Table S4). Among HCHs,  $\beta$ -HCH was  
267 detected in 100% of the samples, above  $\gamma$ -HCH (73%),  $\alpha$ -HCH (58%) and  $\delta$ -HCH (6%). HCB and PeCB  
268 were detected in 97% and 52% of the samples, respectively. The other OCPs, namely, aldrin, isodrin,  
269  $\alpha$ -endosulfan,  $\beta$ -endosulfan and endosulfan sulphate, were never detected. BDEs -28, -47, -49, -66, -  
270 99, -100, -119, -126, -153, -154 and -155 were the most frequently detected (> 70% of the samples),  
271 while BDEs -77, -183, -184, -202, -205, -207 and -209 showed intermediate detection frequencies (27–  
272 55%). The other PBDE congeners, i.e. BDEs 30, -71, -85, -138, -171 and -204 were below LOQs in all  
273 samples. The non-PBDE BFRs HBB, BB-153 and DBDPE were detected in 61, 73 and 76% of the samples,  
274 respectively. BTBPE was only detected in 7 samples (21%). Among PFASs, PFOS, PFNA, PFDA, PFUnDA,  
275 PFDoDA, PFTTrDA and PFTeDA were detected in 100% of the samples. PFOA was detected in 45% of the  
276 samples, PFDS in 27% and PFHpA was seldom detected (12%) (Table S4). PFHpS and PFHxA were below  
277 LOQs in all samples, PFHxS was detected in one sample only and close to the LOQ.

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

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

304 In fish, PCBs presented the highest concentrations (range of  $18.62 \text{ ng g}^{-1} \text{ dw}$  to  $115.37 \text{ ng g}^{-1} \text{ dw}$ , mean  
305 of  $54.42 \pm 28.57 \text{ ng g}^{-1} \text{ dw}$  calculated on the replicates), followed by OCPs ( $5.69\text{--}98.57 \text{ ng g}^{-1} \text{ dw}$ , mean  
306 of  $21.73 \pm 21.26 \text{ ng g}^{-1} \text{ dw}$ , of which 83% were DDTs), PFASs ( $1.54\text{--}37.25 \text{ ng g}^{-1} \text{ dw}$ , mean of  $11.95 \pm$   
307  $9.58 \text{ ng g}^{-1} \text{ dw}$ ) and PBDEs ( $0.43\text{--}5.31 \text{ ng g}^{-1} \text{ dw}$ , mean of  $1.57 \pm 1.17 \text{ ng g}^{-1} \text{ dw}$ ). These results agree  
308 with previous studies showing PCBs and DDTs as the major chlorinated contaminants in deep-sea fish  
309 worldwide, reflecting their high persistence and hydrophobicity, while PBDE levels are usually reported  
310 to be several orders of magnitude lower (Koenig et al., 2013; Webster et al., 2014).

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

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

319 Only a limited number of lipophilic OCs (i.e. dieldrin, endrin,  $\Sigma$  HCHs and each isomer, HCB) showed  
320 significant ( $p < 0.0001$  to  $0.022$ ) positive correlations with TLCs, while  $\Sigma$  DDTs,  $\Sigma$  PCBs (whether i-PCBs,  
321 di-PCBs or  $\Sigma$  all congeners were considered) and  $\Sigma$  PBDEs did not. Very few individual PCB congeners  
322 (CBs -18, -28, -31, -44, -49, -52 and -66, i.e. the less-chlorinated ones) and individual PBDEs (BDEs -66  
323 and -77) showed a significant linear relationship between their concentrations and TLCs. These results  
324 are somewhat surprising for lipophilic contaminants. However, TLCs determined in the present  
325 samples were a proxy of total lipids and it has been proven that lipophilic contaminant accumulation  
326 is not governed only by lipid content but also by lipid composition (Xie et al., 2020). Our results suggest  
327 complex relationships between the species' lipidic composition and the various studied chlorinated  
328 and brominated OC families and would argue in favour of more investigations into the specific tissue  
329 biochemical composition of species. Despite this lack of significant correlations, concentrations of  
330 lipophilic OCs were normalised to TLCs to compare the concentrations between replicate samples  
331 within a species (intra-species variability) and between species (inter-species variability). Normalising  
332 the concentrations to TLCs did not decrease the intra-species variability (30, 29 and 39% for PCBs, OCPs  
333 and PBDEs respectively) nor the inter-species variability (73–75%). However, these results were highly  
334 dependent on the species; the intra-species variability decreased highly for *Argyropelecus olfersii* (ratio  
335 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  
336 for PCBs, OCPs and PBDEs, respectively), while the concentrations in *Notoscopelus kroeyeri* were highly  
337 variable whether they were expressed in dw (60–75%) or lw (64–80%). When both taxa were  
338 compared, only PFAS and PBDE concentrations (in  $\text{ng g}^{-1}$  dw) showed significant differences (KW and

339 MW) between crustaceans and fish (PFASs higher in crustaceans  $p = 0.0034$ , PBDEs higher in fish  $p =$   
340  $0.0021$ ). Although PCBs and OCPs showed higher levels in fish than in crustaceans, the lack of  
341 significant differences between both taxa could be explained by both the high inter-species variability  
342 within each taxon and the smaller difference in concentrations between taxa.

343 PFASs exhibited the highest contribution among the OC families in the crustacean species *Pasiphaea*  
344 *sivado* ( $65 \pm 2\%$  on average), while PCB contribution was the highest in all fish species (between 47%  
345 and 72%, with a mean of  $62 \pm 10\%$ ). PFAS contribution decreased significantly with both  $\delta^{15}\text{N}$  values ( $p$   
346  $= 0.0001$ ) and TLC ( $p = 0.004$ ), reflecting a relationship with the trophic magnification factors (TMFs) of  
347 the studied contaminants, which are reported to be globally lower for PFASs than for PCBs (Won et al.,  
348 2018) and suggesting a potential link with the biochemical compositions of the studied species. Unlike  
349 the lipophilic contaminants studied here (i.e. PCBs, OCPs and PBDEs), PFASs have an affinity for specific  
350 proteins and phospholipids (Armitage et al., 2013; Ng and Hungerbühler, 2013). PFASs presented the  
351 highest relative concentrations in the crustacean species that showed the lower TLC (lipophilic  
352 contaminants being consequently less predominant), namely *Pasiphaea sivado*, while the lowest PFAS  
353 relative contribution (17%) was found in the crustacean species *Ephyrina figueirai*, which showed the  
354 highest TLC (and the highest lipophilic contaminant contribution). However, the two fish species  
355 *Serrivomer beanii* and *Xenodermichthys copei*, despite them having been reported to have lower  
356 protein contents than the other studied fish species sampled from the Bay of Biscay (Spitz et al., 2010),  
357 showed higher PFAS contribution than the other fish ( $30 \pm 8\%$  versus  $10 \pm 6\%$  in the other species), a  
358 result that could rather be explained by their lower trophic level (see section 3.3.6).

### 359 **3.3.2. OC concentrations are in the range of those reported in other deep pelagic species in** 360 **the NE Atlantic**

361 Although PCB, OCP and PBDE concentrations in deep-sea organisms from various oceanic regions have  
362 been published previously, most of the data refer to bathydemersal and benthopelagic species, i.e.  
363 living and feeding close to the bottom seafloor. In addition, the studied periods refer to sampling dates  
364 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  $\Sigma_6$  PCB and  $\Sigma_{15}$  PBDE concentrations were determined in the muscles of 4 deep pelagic fish species  
369 similar to ours (namely *Benthoosema 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<sup>-1</sup> lw and between <LOD and 23 ng g<sup>-1</sup> lw,  
372 respectively (Romero-Romero et al., 2017). These levels compare with our concentration ranges (i.e.,  
373 31.2–1072.9 ng g<sup>-1</sup> lw and 0.92–43.37 ng g<sup>-1</sup> lw for PCBs and PBDEs respectively).  $\Sigma_7$  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<sup>-1</sup> 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<sup>-1</sup> lw in their livers (Webster et al., 2014), which compares closely  
378 to our results in whole fish (524–651 ng g<sup>-1</sup> lw), although younger individuals were considered in our  
379 study. In the same individuals, the  $\Sigma_9$  PBDEs were between 1.1 ± 3.0 and 42.5 ± 26.4 ng g<sup>-1</sup> lw in the  
380 flesh and between 2.4 ± 3.9 and 25.8 ± 8.2 ng g<sup>-1</sup> 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<sup>-1</sup> 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 ( $\Sigma_7$  ICES PCBs: 91 ng g<sup>-1</sup> lw to 7660 ng g<sup>-1</sup> lw versus 524–651 ng g<sup>-1</sup> lw in our fish samples;  
387 HCB: 5.3 to 30.3 ng g<sup>-1</sup> lw versus 11.4–18.8 ng g<sup>-1</sup> lw; dieldrin: 22.0–40.9 ng g<sup>-1</sup> lw versus 20.5–25.0 ng  
388 g<sup>-1</sup> lw) except for the  $\Sigma$  DDTs, which was lower in our samples (239.5–397.8 ng g<sup>-1</sup> lw versus 384–4350  
389 ng g<sup>-1</sup> lw in Mormede and Davies' study), potentially reflecting a higher decrease of DDT inputs over  
390 the studied periods.



391 ***Specific congener analysis reveals untypical BFR profiles***

392 Indicator PCBs (i-PCBs) were predominant compared to dioxin-like congeners (dl-PCBs) in all species,  
393 and i-PCB and dl-PCB concentrations were highly correlated ( $r = 0.93$ ,  $p < 0.05$ ). The average ratio (i-  
394 PCBs/dl-PCBs) was  $7 \pm 1$  and showed no significant difference between taxa. Detailed PCB mean  
395 concentrations per congener and taxon are given in Table S3. The most abundant congener was CB-  
396 153 with concentrations ranging from 1.73 to 15.03  $\text{ng g}^{-1}$  dw (mean of  $6.663 \pm 6.776$   $\text{ng g}^{-1}$  dw) in  
397 crustaceans and from 2.40 to 23.27  $\text{ng g}^{-1}$  dw (mean of  $10.71 \pm 6.10$   $\text{ng g}^{-1}$  dw) in fish. Hexa- and  
398 heptachlorinated congeners (i.e. CB-153, CB-138, CB-180 and CB-187, each one contributing to 10-19%  
399 of the  $\sum$  PCBs) were the most abundant ones, which is consistent with previous findings in deep-sea  
400 ecosystems (Koenig et al., 2013; Romero-Romero et al., 2017; Storelli et al., 2009; Takahashi et al.,  
401 2010; Webster et al., 2014). The i-PCBs contributed to 47% of the  $\sum$  PCBs while dl-PCBs counted for 7%  
402 of the  $\sum$  PCBs, pointing out the importance of determining more than the classic 18 congeners to better  
403 assess total PCB bioaccumulation in marine organisms. Congeners with 5 and more chlorine atoms  
404 showed the highest cross-correlations, which underline their similar sources and behaviours.

405 DDTs were by far the most abundant OCPs in all species (0.53–86.23  $\text{ng g}^{-1}$  dw range, mean of  $16.35 \pm$   
406  $18.90$   $\text{ng g}^{-1}$  dw), followed by dieldrin (0.25–6.83  $\text{ng g}^{-1}$  dw range, mean of  $1.96 \pm 1.34$   $\text{ng g}^{-1}$  dw) > HCB  
407 (0.32–3.93  $\text{ng g}^{-1}$  dw range, mean of  $1.07 \pm 0.91$   $\text{ng g}^{-1}$  dw) > endrin (0.028–0.73  $\text{ng g}^{-1}$  dw range, mean  
408 of  $0.25 \pm 0.19$   $\text{ng g}^{-1}$  dw) =  $\sum$  HCHs (0.01–0.61  $\text{ng g}^{-1}$  dw range, mean of  $0.20 \pm 0.18$   $\text{ng g}^{-1}$  dw) (Table  
409 S4). These results bring evidence that DDT is still a major organochlorine pesticide in marine  
410 ecosystems, which is in line with its global past usage and environmental persistence (Li and  
411 Macdonald, 2005).  $\sum$  DDTs showed significant positive correlations with all other organochlorine  
412 pesticides except  $\sum$  HCHs, possibly related to HCH physico-chemical properties and environmental  
413 behaviour (Salvado et al., 2019). The highest correlations were between  $\sum$  DDTs and mirex (0.91) while  
414 dieldrin and endrine showed weaker ones (0.70 and 0.42, respectively).

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

417 et al., 2013; Ramu et al., 2006; Storelli et al., 2009). The *o,p'*-DDT/*p,p'*-DDT concentration ratio was  
418  $0.36 \pm 0.17$ , on average, in all fish species and showed high inter-species variations (0.03–0.55 range).  
419 This ratio is commonly used to distinguish DDT sources, with values in the 0.2–0.3 range in technical  
420 DDT (Kalantzi et al., 2001) whereas a ratio above 0.34 is usually attributed to DDT's origin from dicofol  
421 acaricide impurities (Suarez et al., 2013). This ratio was particularly low in *Stomias boa* ( $0.03 \pm 0.01$ )  
422 because of peculiar low contributions of *o,p'*-DDT ( $0.4 \pm 0.2\%$  versus  $3.3 \pm 1.9\%$  in the other fish  
423 species). These results show that the use of this ratio should be interpreted with caution and suggest  
424 that species-specific parameters may also influence DDT isomeric profiles.

425 BFR profiles were dominated by BDE-209 (mean contribution of 38% of the 12 summed congeners  
426 quantified in more than 50% of the samples) followed by BDE-47 (21%), BDE-155 (19%) and BDE-154  
427 (14%). The other mostly-detected congeners (i.e. BDEs -28, -49, -66, -99, -100, 119, -126, -153)  
428 contributed each to less than 10%, on average, to the summed 12 BDEs. As BDE-47 is generally  
429 reported as the major PBDE congener in biota, the present PBDE profile is therefore atypical due to  
430 the high contribution of BDE-209. The BDE-209 congener is usually reported as poorly bioaccumulable  
431 in fish because of its very high  $\log K_{ow}$  (12.1, Kelly et al., 2008), high molecular size and degradation  
432 propensity *via* metabolism (Stapleton et al. 2004a, Roberts et al., 2011). Following our results, this  
433 highly brominated congener has been previously identified at higher abundance (17% of  $\Sigma$  PBDEs) in  
434 deep-sea organisms compared to those from the shelf (Romero-Romero et al., 2017). Although all fish  
435 stomachs and intestines were emptied of major debris (after visual inspection) before analysis, it  
436 cannot be ruled out that high BDE-209 concentrations were also due to non-ingested particles still  
437 present in the digestive tracts. However, the presence of a high diversity of several high-brominated  
438 congeners, as well as species-specific profiles, rather suggests BDE-209 bioaccumulation and  
439 biotransformation, as detailed in section 3.3.4 below.

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

443 high levels in fish and generally leads to its biodilution in trophic webs (Tao et al., 2019). However,  
444 unlike for BDE-209, no debromination of DBDPE has been reported in fish and its bioaccumulation  
445 factor was reported to be 10 times higher than BDE-209's one (He et al., 2012), which would argue in  
446 favour of its occurrence at higher levels than BDE-209. Similar to BDE-209, the DBDPE occurrence at  
447 particularly high levels in some of the studied samples (i.e. *Serrivomer beanii*, *Stomias boa*, *Aphanopus*  
448 *carbo* and *Searsia koefoedi*) could be the result of undigested particles in their digestive tracts. DBDPE  
449 sources in the marine environment include plastics from electronic and electrical equipment wastes  
450 (Stubbings et al., 2021). However, whether DBDPE found in some fish originated from its adsorption  
451 on particulate matter or plastics, or plastics themselves (eventually extracted following whole fish  
452 analyses) cannot be proven. In oceanic waters off the Californian coast (Monterey Bay, US), the highest  
453 concentrations of microplastics have been reported between 200 m and 600 m (Choy et al., 2019), and  
454 microplastics have been reported in the digestive tracts of mesopelagic fish feeding at these depths,  
455 although at very different occurrence frequencies depending on locations (35% in Boerger et al., 2010;  
456 9% in Davison and Asch, 2011; 11% in Lusher et al., 2016; 24% specifically in Myctophidae in Savoca et  
457 al., 2020; 73% in Wieczorek et al., 2018). Among the various species studied by Wieczorek et al. (2018),  
458 *Serrivomer beanii* and *Lampanyctus macdonaldi* were among those presenting the highest frequency  
459 of plastic occurrence, while *Serrivomer beanii* and *Myctophum punctatum* were two species presenting  
460 the highest number of plastic debris in their gut. Despite the digestive tracts of fish from our study  
461 being emptied from visible material before analysis, the presence of small parts of food residues  
462 (including those of plastic origin) that could have still be present in the digestive tracts (i.e. not  
463 assimilated/absorbed into the organism's tissues) is not unrealistic. The fact that high variations were  
464 sometimes found between replicates of the same species (as in *Aphanopus carbo*, with concentrations  
465 of 81, 4762 and 7803 pg g<sup>-1</sup> dw in each of the three replicates made from individual fish, or in  
466 *Xenodermichthys copei* with < LOQ, 148 and 1940 pg g<sup>-1</sup> dw in pooled samples) argues in favour of  
467 undigested particles with a short-time and erratic occurrence rather than a long-term accumulation in  
468 tissues. On the opposite hand, species such as *Stomias boa* showed high DBDPE concentrations in all

469 replicates (3075, 5963 and 7840  $\text{pg g}^{-1}$  dw,  $\text{rsd} = 43\%$ ), while others such as *Myctophum punctatum* or  
470 *Lampanyctus crocodilus* exhibited systematically lower concentrations ( $< \text{LOQ}$ , 42 and 193  $\text{pg g}^{-1}$  dw  
471 and 436, 682 and 938  $\text{pg g}^{-1}$  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* ( $64.3 \pm 9.2$  cm), *Stomias boa* ( $31.6 \pm 3.7$  cm) and  
474 *Aphanopus carbo* ( $61.7 \pm 0.6$  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 (McIlwraith et al., 2021).  
478 Despite its relatively smaller length (15 cm), *Searsia koefoedi* exhibited the highest concentration of all  
479 species (16.48  $\text{ng g}^{-1}$  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.

494 **3.3.3. Chlorinated and brominated OC profiles and diagnostic ratios reveal species-specific**  
495 **metabolic capacities**

496 Because PCB bioaccumulation depends on their chlorine number and position, which both are source-  
497 and metabolism-related, PCB profiles can be examined by their number of Cl atoms and their structure-  
498 activity group (SAG) classification (Boon et al., 1997; Yunker et al., 2011). While the lower-chlorinated  
499 congeners are associated with atmospheric transport and feeding at lower trophic levels, the heavier  
500 congeners reflect continental inputs from rivers and prey of higher trophic levels. In our samples, 3-Cl  
501 and 4-Cl showed higher contributions to the  $\Sigma$  PCBs in crustaceans compared to fish, which is  
502 consistent with crustaceans' lower trophic levels (i.e. lower  $\delta^{15}\text{N}$  values), although the difference was  
503 significant only for the 3-Cl congeners ( $10 \pm 5\%$  in crustaceans versus  $4 \pm 5\%$  in fish, MW and KW tests).  
504 However, inter-species differences were high when only fish were considered, and similar  
505 contributions to those of crustaceans were found in some fish species such as *Arctozenus risso*,  
506 *Argyrolepecus olfersii* and *Myctophum punctatum* (Fig. 1A). These species are among those showing  
507 the lowest  $\delta^{15}\text{N}$  values among the studied fish species, but other species with low  $\delta^{15}\text{N}$  values (such as  
508 *Serrivomer beanii* or *Xenodermichthys copei*) did not show such a high 3-Cl contribution (Fig. 1A),  
509 suggesting that the trophic level does not solely explain these results. Indeed, the lower chlorinated  
510 congeners are also the most metabolisable ones (SAG III and IV), while the most refractory ones (6-,7-  
511 and 8-Cl) belong to SAG I and II groups. No significant differences were observed between taxa  
512 regarding SAG groups. The three crustacean species showed discrepancies in their PCB profiles, with  
513 *Sergia robusta* being characterised by higher SAG I and II group contributions (72% in total) than the  
514 other two species ( $52 \pm 5\%$ ). Among fish species, *Arctozenus risso*, *Argyrolepecus olfersii* and  
515 *Myctophum punctatum* showed the highest SAG III and IV (metabolisable congeners) and the lowest  
516 SAG I and II (refractory congeners) contributions. On the opposite hand, *Serrivomer beanii*,  
517 *Xenodermichthys copei*, *Aphanopus carbo*, *Lampanyctus crocodilus* and *Searsia koefoedi* were  
518 characterised by higher SAG I and II (6 to 8 Cl) contributions (Fig. 2). More specifically, the CB-149/CB-  
519 153 and CB-132/CB-153 concentration ratios, which could be used as metabolism tracers (i.e. a ratio

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

524 The  $(p,p'$ -DDE +  $p,p'$ -DDD)/ $\Sigma$  DDTs ratio was  $0.85 \pm 0.06$  in all samples, which globally indicates old DDT  
525 inputs (Suarez et al., 2013) and is consistent with the ban of DDT usage since the 1970s or 1980s in  
526 most countries worldwide (Kalantzi et al., 2001). The  $p,p'$ -DDE isomer contribution was significantly  
527 (MW and KS  $p < 0.1$ ) higher in crustaceans and particularly high in *Pasiphaea sivado* replicates, which  
528 was mainly due to  $p,p'$ -DDD and  $p,p'$ -DDT  $< LOQs$  in these samples. Some variations in  $p,p'$ -DDE  
529 contributions were also observed between fish species (Fig. 1B). Among Myctophidae, both  
530 *Myctophum punctatum* and *Notoscopelus kroeyeri* exhibited similar DDT profiles ( $p,p'$ -DDE and  $p,p'$ -  
531 DDD at  $65 \pm 5\%$  and  $12 \pm 1\%$ , respectively) and were different from those of the third Myctophid species  
532 *Lampanyctus crocodilus*, in which higher  $p,p'$ -DDE ( $84 \pm 2\%$ ) and lower  $p,p'$ -DDD ( $5 \pm 1\%$ ) contributions  
533 were found. These results could suggest a species-specific DDT profile due to different metabolic  
534 capacities, which would be in line with the higher metabolic capacity of PCBs observed in *Lampanyctus*  
535 *crocodilus*. In addition, *Lampanyctus crocodilus* was the leanest of the three studied Myctophid species  
536 (by a factor of 4 to 5, Table 2), which could influence DDT isomer's relative bioaccumulation. Indeed,  
537 in the studied fish species,  $p,p'$ -DDE contribution showed a significant decrease with increasing TLC ( $p$   
538  $< 0.0001$ ).

539 Regarding PBDE profiles (Fig. 1C), the nona-BDEs BDE-207 and BDE-208, the octa-BDEs BDE-202, BDE-  
540 205 and BDE-197 and the hepta-BDEs BDE 184 and BDE-183 were quantified in 18–48% of our samples,  
541 although at low levels (their total contribution was 6% of the sum of all congeners determined above  
542 LOQs). In addition, the hexa-BDEs BDE-155 and BDE-154, ranking third and fourth in our samples, have  
543 both been reported as debromination products of octa-BDEs (Stapleton et al., 2004b; Zeng et al.,  
544 2012). As the nona- to hexa-brominated congeners identified in our samples are not present in  
545 technical mixtures (La Guardia et al., 2006; Munschy et al., 2011), their occurrence might be explained

546 by either i) assimilation and further metabolisation by fish or ii) metabolisation by the prey and  
547 bioaccumulation in fish *via* trophic transfer. Indeed, these congeners are among those (ranging from  
548 nona- to tetra-BDEs) that have been reported in various fish species following BDE-209 metabolic  
549 degradation (Roberts et al., 2011). BDE-154 could result from the debromination in the *meta* position  
550 of BDE-183, while BDE-155 could result from successive debrominations in the *meta* positions of BDE-  
551 207, BDE-197 and BDE-184, which were all detected in our samples, although at low concentrations  
552 compared to their debrominated counterparts. If these congeners originate from BDE-209  
553 metabolism, their detection at concentrations above the LOQs reveals exposure of the studied species  
554 to BDE-209. The high variability in PBDE profiles between species could therefore result from species-  
555 specific metabolism, as evidenced by various studies in fish (Roberts et al., 2011; Stapleton et al., 2006;  
556 Yokota et al., 2021). The results suggest that BDE-183, BDE-197 and BDE-207 could be potential  
557 intermediate debromination products of BDE-209 resulting in the formation of BDE-154, and that the  
558 removal of Br atoms in the *meta* position was favoured. However, no significant relationship was found  
559 between BDE-209 and either BDEs -183, -197 or -207 (although the latter two congeners showed low  
560 detection frequencies), nor with BDE-154 or BDE-155. Interestingly, BDE-154 and BDE-155 were highly  
561 correlated ( $n = 28$ ,  $r = 0.96$ ) in fish, which could reflect their common origin (i.e. BDE-209  
562 biotransformation).

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

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

### 579 **3.3.4. Perfluorinated substances' molecular profiles are dominated by odd chain length**

#### 580 **PFCAs**

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



598 of  $75 \pm 12\%$  (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 ( $89 \pm 6\%$ ) than in fish ( $72 \pm 11\%$ ). The crustacean species *Ephyrina figueirai* showed a  
601 peculiar high PFCa contribution ( $\Sigma$  PFCAs/PFOS ratio of 59) due to a low PFOS concentration ( $0.379 \text{ ng}$   
602  $\text{g}^{-1} \text{ dw}$ ), while its  $\Sigma$  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  $\Sigma$  PFCa concentrations than PFOS ones, with a  $\Sigma$  PFCAs/PFOS  
605 ratio of 0.75, due to a particularly high PFOS contribution (57% of  $\Sigma$  PFASs). The  $\Sigma$  PFCa contribution  
606 was also higher ( $87 \pm 2\%$ ) in *Serrivomer beanii*, the fish species with the lowest  $\delta^{15}\text{N}$  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 PFTTrDA ( $C_{13}$ ) 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 Simmonet-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

624 length PFCAs than odd chain length PFCAs (Franklin et al., 2016; Gebbink et al., 2016). In our samples,  
625 the mean  $(C_{11} + C_{13})/(C_{10} + C_{12})$  ratio was  $4.2 \pm 1.4$ , showing a higher occurrence of odd chain length  
626 PFCAs in all species (no significant difference was observed between taxa). Washington et al. (2020)  
627 used the  $(C_{11} + C_{13}) - (C_{10} + C_{12})$  values to reveal PFAS sources in soils, allowing them to distinguish  
628 between direct release (higher values) and fluorotelomer degradation. In biota, this parameter could  
629 be influenced by *in vivo* degradation of precursors into PFCAs, which are not expected to further  
630 degrade in organisms. This ratio could therefore give an integrated view of direct exposure and  
631 subsequent bioaccumulation, combined with the effect of precursor biotransformation. Interestingly,  
632 this ratio showed strong differences between i) the crustaceans and the fish species *Serrivomer beanii*  
633 and *Searsia koefoedi* that show high values ( $14420 \pm 3968$ ) and ii) the other fish species that presented  
634 lower values ( $3243 \pm 2360$ ). The higher  $(C_{11} + C_{13}) - (C_{10} + C_{12})$  values suggest that these PFCAs were  
635 less derived from precursors in crustaceans and the fish species *Serrivomer beanii* and *Searsia koefoedi*  
636 than in the other fish species. This assumption would agree with species-specific precursor  
637 biotransformation as already reported for PFOS in freshwater invertebrates and fish (Babut et al.,  
638 2017) and would suggest a lower precursor metabolism in these species.

639 **3.3.5. Most of the chlorinated and brominated OCs show biomagnification across the studied**  
640 **food chain, while PFASs exhibit a contrasted behaviour**

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

650 3A, Table S8). On the opposite hand, neither  $\Sigma$  HCHs nor individual isomers showed any significant  
651 relationship.  $\Sigma$  PBDEs and individual congeners, except BDEs -28, -66, -77, and -183, showed significant  
652 positive linear relationships with  $\delta^{15}\text{N}$  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  $\delta^{15}\text{N}$  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 superhydrophobic ones  
667 and their concentrations (in lw) showed contrasting relationships with  $\delta^{15}\text{N}$  values; while BDE-209  
668 concentrations were not significantly related to  $\delta^{15}\text{N}$  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  $\delta^{15}\text{N}$  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<sup>-1</sup> ww), -153 (0.10 ng  
695 g<sup>-1</sup> ww) and -180 (0.11 ng g<sup>-1</sup> 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<sup>-1</sup> 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

702 53% (100 times) of the samples, respectively. BDE-183 was barely above the BAC (3% of the sample by  
703 1.6 times) and BDE-85 was never detected above the LOQ. In addition, Environmental Assessment  
704 Criteria (EACs, in lw) defined as concentrations below which biological effects are unlikely to occur  
705 have also been established in fish. EACs were exceeded only for CB-118 (25 ng g<sup>-1</sup> lw) in 30% of the  
706 samples (*Lampanyctus crocodilus*, *Chauliodus sloani*, *Stomias boa* and *Aphanopus carbo*), in agreement  
707 with the OSPAR intermediate assessment conducted within the period 1995–2015 (OSPAR, 2017).

#### 708 **4. Conclusions and implications for higher trophic level consumers**

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

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

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

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

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

750 **Credit author statement**

751 Catherine Munsch: Conceptualisation, Investigation, Formal analysis, Writing - original draft, review  
752 & editing. Jérôme Spitz: Conceptualisation, Funding acquisition, Sample acquisition, Writing - review &  
753 editing. Nadège Bely: Data acquisition. Karine Héas-Moisan: Data acquisition. Nathalie Olivier: Data  
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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.)

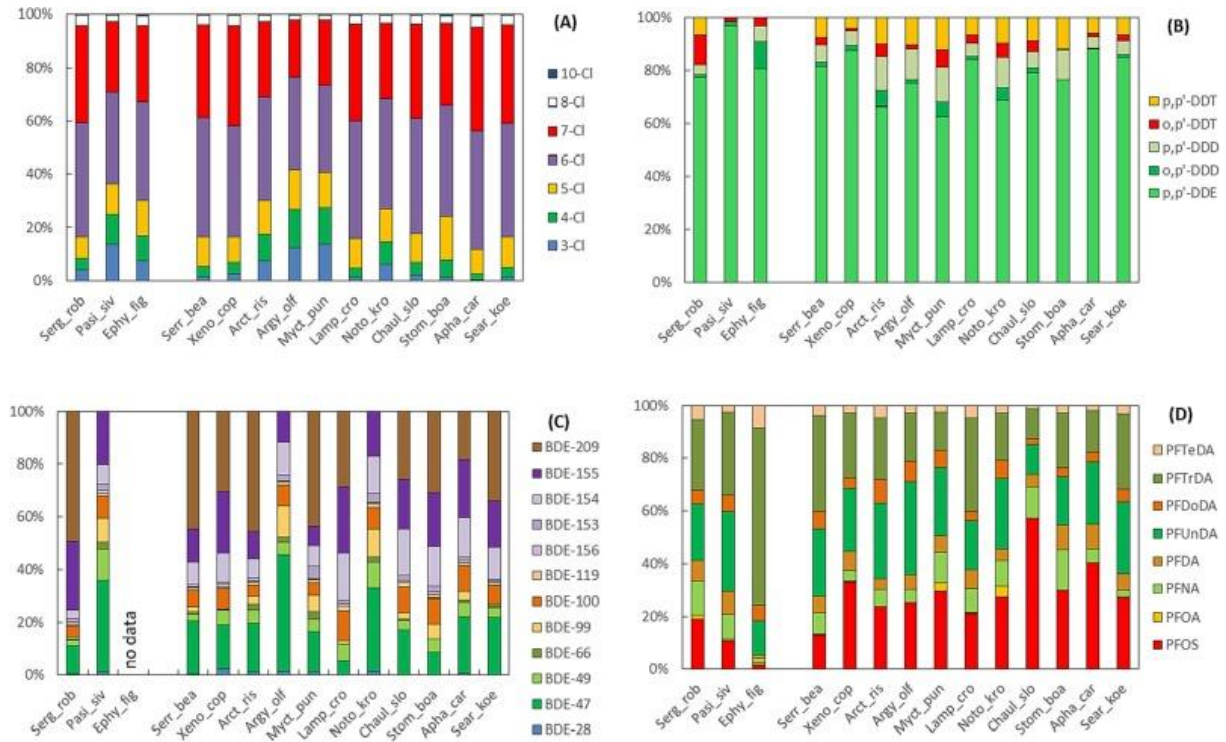


Fig. 2. Projection of the principal component analysis conducted on normalised profiles of PCBs categorized according to their SAG group in deep pelagic fish species collected in the Bay of Biscay in October 2017. Projection of variables (A) and individuals (B) on the first two principal components are 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*.

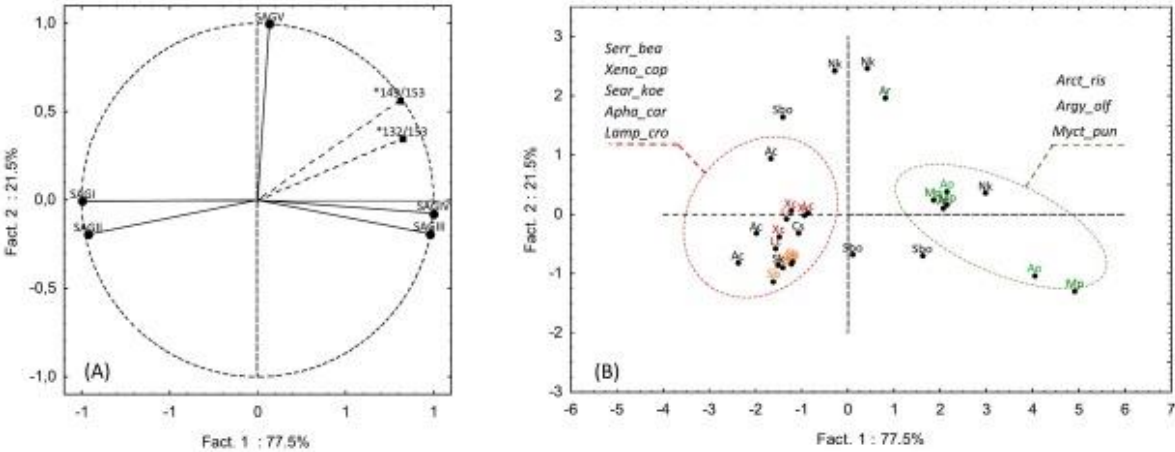




Fig. 3. Relationships between [organic contaminant](#) concentrations and  $\delta^{15}\text{N}$  values (‰) in crustaceans (black dots) and fish (white dots) collected in deep pelagic waters of the Bay of Biscay in October 2017. Concentrations of organochlorinated compounds (A) and organobrominated compounds (B) were log-transformed and expressed in  $\text{ng g}^{-1} \text{lw}$  and  $\text{pg g}^{-1} \text{lw}$  respectively; PFAS concentrations (C) were log-transformed and expressed in  $\text{ng g}^{-1} \text{dw}$ . 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.

