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## **Estimating Contaminants tRansfers Over Complex food webs (ESCROC): An innovative Bayesian method for estimating POP's biomagnification in aquatic food webs**

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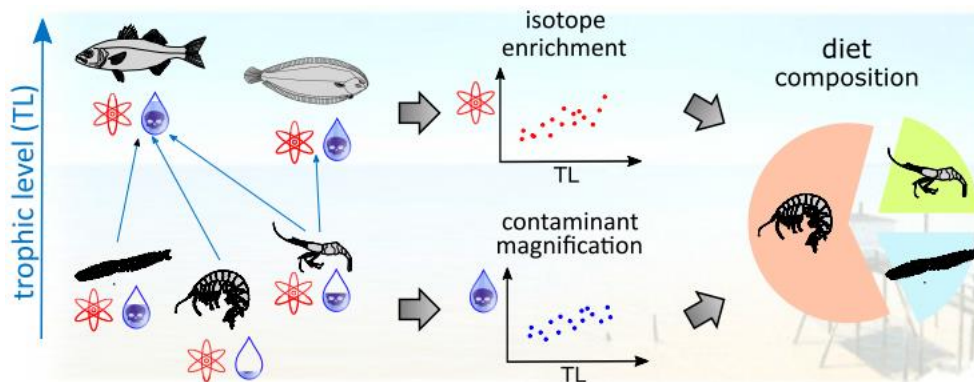
27 proposes a relevant and innovative TMF estimation method accounting for its many variability  
28 sources. The ESCROC model (EStimating Contaminants tRansfers Over Complex food webs), which is  
29 implemented in a Bayesian framework, allows for a more reliable and rigorous assessment of  
30 contaminants trophic magnification, in addition to accurate estimations of isotopes trophic  
31 enrichment factors and their associated uncertainties in food webs. Similar to classical mixing models  
32 used in food web investigations, ECSROC computes diet composition matrices using isotopic  
33 composition data while accounting for contamination data, leading to more robust food web  
34 descriptions.

35 As a demonstration of the practical application of the model, ESCROC was implemented to revisit the  
36 trophic biomagnification of 5 polyfluoroalkyl substances (PFAS) in a complex estuarine food web (the  
37 Gironde, SW France). In addition to the TMF estimate and 95% confidence intervals, the model  
38 provided biomagnification probabilities associated to the investigated contaminants —for instance,  
39 92% in the case of perfluorooctane sulfonate (PFOS) — that can be interpreted in terms of risk  
40 assessment in a precautionary approach, which should prove useful to environmental managers.

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43 **Graphical abstract**



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47 **Highlights**

- 48 • Estimating the trophic magnification potential of chemicals is a key issue for management
- 49 • This biomagnification potential is usually estimated through a Trophic Magnification Factor
- 50 (TMF)
- 51 • ESCROC is an innovative Bayesian mixing model for estimating TMFs
- 52 • It provides rigorous diagnoses on contamination and associated uncertainty
- 53 • The example of PFASs in the Gironde estuarine food web was used as a case study

54

55

56 **Keywords**

57 Trophic magnification – food webs – Bayesian mixing model – organic micropollutants – stable  
58 isotopes - Gironde estuary

## 59 **1 Introduction**

60 Increased nutrients, pollutants, and agrochemicals due to industries, urbanization and agriculture  
61 exert dramatic impacts on ecosystems (Köhler and Triebkorn, 2013; Verhoeven et al., 2006). Aquatic  
62 ecosystems and, among them, coastal and estuarine ecosystems are particularly vulnerable to these  
63 changes: they are increasingly exploited and polluted, and their biodiversity is decreasing (Budzinski  
64 et al., 1997; Matthiessen and Law, 2002). At the individual and population scales, some pollutants can  
65 lead to deleterious effects, such as altered metabolism, immunotoxicity, endocrine disruption or  
66 neurotoxicity (Köhler and Triebkorn, 2013). This issue is exacerbated by the fact that some pollutants  
67 tend to be accumulated by organisms, a process known as bioaccumulation. Bioaccumulation is a  
68 fundamental process in environmental toxicology and risk assessment because it controls the internal  
69 dose of potential toxicants in individual organisms (Arnot and Gobas, 2004). Additionally, some  
70 contaminants also become ecologically harmful because they accumulate through food webs, a  
71 process known as biomagnification. In these instances, pollutants found at low concentrations in  
72 natural environments can achieve harmful concentration for high-order organisms including human  
73 beings (Kelly et al., 2007; Köhler and Triebkorn, 2013; Van Oostdam et al., 2005). Studies aquatic  
74 ecosystems –marine systems (e.g. Romero-Romero et al., 2017; Sun et al., 2017), lakes (e.g. Liu et al.,  
75 2018; Mazzoni et al., 2018), coastal environments (Bodin et al., 2007; Loizeau et al., 2001a; Loizeau et  
76 al., 2001b; Munschy et al., 2011) and rivers (Lopes et al., 2011)- but also in terrestrial environments  
77 (e.g. Daley et al., 2011; Wang and Gao, 2016) demonstrated such biomagnification process occurs for  
78 hydrophobic organohalogenated contaminants. Being potentially persistent, bioaccumulative and  
79 toxic (due for instance to neurotoxic properties and/or endocrine disruption), Persistent Organic  
80 Pollutants (POPs) are of particular concern (see the Stockholm Convention on POPs as amended in  
81 2009 - UNEP, 2009) for human health (Belpaire et al., 2016; Berger et al., 2009) as well as for animal  
82 populations' viability (Gilliers et al., 2006a; Gilliers et al., 2006b; Rochette et al., 2010). In this context,  
83 a comprehensive understanding of the ecodynamics of human-induced chemicals in coastal and

84 estuarine ecosystems is needed to better manage the ecological functions associated with these  
85 areas.

86 Potential for bioaccumulation in organisms and biomagnification in food webs differ depending  
87 on the investigated contaminants, environmental contexts, and physiological characteristics of species  
88 (Bodiguel et al., 2009; Connolly and Glaser, 2002; Gobas, 1993). Therefore, accurate *in situ* assessment  
89 of bioaccumulation and biomagnification potential of pollutants in aquatic food webs is required, in  
90 order to inform management actions.

91 Most empirical approaches used to understand trophic transfers of pollutants rely on the  
92 estimation of a Trophic Magnification Factor (TMF) from field data (Borgå et al., 2012). The TMF is  
93 used to assess the biomagnification of a given pollutant in a food web and to define environmental  
94 quality norms in some instances (see french environmental quality norms (NQE) - Migne-Fouillen et  
95 al., 2010). It usually corresponds to the slope of the statistical regression between the chemical  
96 concentration and the trophic level of organisms within a food web. Although TMF is increasingly used  
97 to describe trophic dynamics of xenobiotics, its estimates present many uncertainties, reviewed by  
98 Borga et al. (2012) and Mackay et al. (2016). Among the many assumptions, potential bias, pitfalls, and  
99 vigilance points reported in these reviews, uncertainty in measurements of contaminant  
100 concentrations, temporal or spatial variability of these concentrations, , inter and intraspecific  
101 variability in the bioaccumulation process, and uncertainty about the food web structure and trophic  
102 levels of individuals were emphasized. Recently, Munoz et al. (2017) evaluated different statistical  
103 methods to address these above-mentioned sources of uncertainty and bias, based on data on 19  
104 polyfluoroalkyl substances (PFASs) in the Gironde estuarine food web. The statistical approaches  
105 compared included linear mixed models from the 'NADA' (Lee, 2017) and 'LMEC' (Vaida and Liu, 2012)  
106 R-packages, accounting for censored responses and a random effect 'species', respectively. Both  
107 methods however assumed that the trophic level of each individual —and consequently, the structure  
108 of the trophic network— are perfectly known, an assumption that rarely verifies in the real world, and  
109 particularly not in the Gironde estuary.

110           Theoretically, estimating the trophic level of a consumer species (or an individual) requires  
111 that its diet be estimated, i.e., the proportion, in biomass, of each prey and its respective trophic level,  
112 in the overall consumer's diet. This is usually done through stomach content analyses, which reflect  
113 the quantitative and qualitative ingestion of species at a specific time, but sometimes raise problems  
114 of prey identification, suffers from some biases such as differential digestibility, and requires many  
115 samples to be analyzed. Stable isotope analyses represent more integrative records of food intake  
116 over longer time scale (Post, 2002) and are now widely used to explore food web structure (Boecklen  
117 et al., 2011; Layman et al., 2012). Stable isotopes of nitrogen are discriminated during digestion and  
118 assimilation, leading to an enrichment of the  $^{15}\text{N}/^{14}\text{N}$  ratio ( $\delta^{15}\text{N}$ ) of the consumer with respect to its  
119 prey. Since the enrichment factor is generally in the range 3 –4 ‰ (DeNiro and Epstein, 1981;  
120 Minagawa and Wada, 1984; Peterson and Fry, 1987), linear regressions can be used to convert stable  
121 isotope composition into trophic level (Post, 2002). This is the usual method chosen to estimate  
122 trophic level when investigating contaminant biomagnification (Borgå et al., 2012). Combined with the  
123 consideration of the isotopic ratio of other elements (e.g.  $^{13}\text{C}/^{12}\text{C}$  or  $^{34}\text{S}/^{33}\text{S}$ ), it can be used to estimate  
124 the diet of species among a set of potential prey. This is the approach used by MixSIR, SIAR and  
125 MixSIAR, the mixing models classically used to estimate a species diet based on its isotopic  
126 composition, which is assumed to be a mixture of the isotopic compositions of the different prey  
127 (Moore and Semmens, 2008; Parnell et al., 2008; Parnell et al., 2010; Parnell et al., 2013; Stock et al.,  
128 2016). This approach allows accounting for the uncertainty of stable isotope enrichment factors, and  
129 for intra- and interspecific variability. However, since the number of isotopic tracers is small (i.e.,  
130 usually two) compared to the number of potential prey, isotope mixing models are generally  
131 underdetermined (Fry, 2013; Phillips and Gregg, 2003; Phillips et al., 2014). Consequently, precise  
132 estimates of enrichment factors must be provided for the mixing model to work, although these  
133 estimates are very sensitive parameters (Bond and Diamond, 2011). Furthermore, it is not possible to  
134 carry out the analysis using those mixing models over a full trophic network but only one predator  
135 after another. Finally, since biomagnification and food web analyses are generally carried out into two

136 independent steps, it is usually impossible (or very difficult) to propagate the uncertainty on estimated  
137 trophic levels to the estimation of biomagnification factors. For both scientific and management  
138 reasons, it is therefore necessary to provide an alternative rigorous method based on sound statistics  
139 to evaluate biomagnification factors (TMF) and associated uncertainty.

140 The present work aimed at presenting such alternative method combining both biomagnification and  
141 food web analyses into a single model. We assume that using contaminants as additional diet tracers  
142 within trophic networks could mitigate the issue of underdetermination of mixing models. Conversely,  
143 incorporating the inference of trophic levels within biomagnification analysis allows propagating the  
144 uncertainty over trophic levels when estimating biomagnification factors such as TMF. Diet tracers  
145 such as isotopes can then be used to estimate contaminant transfers. Our model aims at (1)  
146 accounting for most of the sources of variability listed above on both biomagnification and isotopic  
147 fractionation, (2) estimating diets and related uncertainty for all predators of the food web at once, (3)  
148 estimating the biomagnification of contaminants and related uncertainty. Our model, named ESCROC  
149 (EStimating Contaminants tRansfers Over Complex food webs), is based on a generic mixing model,  
150 similar to those used for deriving diet composition from isotopic data, but allows incorporating  
151 contamination measures.

152 To illustrate the relevance of this model, a large dataset describing the occurrence of a family  
153 of Persistent Organic Pollutants (POPs) in the Gironde estuary was used. Located on the French  
154 Atlantic coast, in SW France and largest estuary in Western Europe (Lobry et al., 2003), the Gironde  
155 estuary case study is especially relevant since POPs are now an increasing issue in this area (Munoz et  
156 al., 2017; Tapie et al., 2011). Among those substances, the target selected compounds were  
157 polyfluoroalkyl substances (PFASs). Few studies have addressed the contamination of estuarine food  
158 webs by these emerging contaminants (de Vos et al., 2008; Naile et al., 2012; Munoz et al. 2017). As  
159 the dataset was previously described and analyzed by Munoz et al. (2017) with a set of various  
160 modelling approaches, this would also allow a comparison of the outcomes provided by the ESCROC  
161 model with those obtained with more traditional methods.

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## 2 Material and methods / description of the model

### 2.1 Context on TMF estimation

#### 2.1.1 Basis of TMF estimation

Basically, TMF estimation consists in assessing the average factor of change in contaminant concentration per Trophic Level (TL). In that sense, it is similar to the enrichment factor in isotopic analysis. Considering biomass distribution in aquatic food webs as well as contaminants transfer efficiencies, the relationship between contaminant concentrations [C] and TL has an exponential form (Borgå et al., 2012). Therefore, TMF estimations are based on the linear regression between Log-transformed contaminant concentrations  $\text{Log}_{10}[\text{C}]$  and TL. The TMF is subsequently obtained as  $10^b$ , with b being the slope of the linear regression (Eq. 1, Figure 1) usually estimated from simple regression models.

$$\text{Log}_{10}[\text{C}] = a + b \cdot \text{TL} \qquad \text{Eq. 1}$$

Modifications to improve TMF estimation calculation were however recently proposed to better account for different sources of uncertainty and variability. For instance, Munoz et al. (2017) suggested to use linear mixed models with censored data (to take into account non-detected data), with random effects to integrate both inter-specific variabilities in physiological properties, errors in chemical concentration measurements and potentially low sampling effort.. These methods however still assumed that trophic levels were perfectly known.

184 If initially, the trophic positions of organisms in the food webs were directly assessed using stable N  
 185 isotope ratios ( $\delta^{15}\text{N}$ ), most recent studies (see Borga et al. 2012 or for instance Munoz et al. 2017)  
 186 refined the estimation of trophic position by using integer-based TL. The TL of a primary producer  
 187 being fixed to 1 by convention, a primary consumer has a TL of 2. Thus, for secondary consumers, the  
 188 trophic level of a particular individual ( $\text{TL}_i$  see Eq. 2) is estimated using the difference between its own  
 189 stable N isotope ratio ( $\delta^{15}\text{N}_i$ , obtained using tissue measures of  $^{15}\text{N}/^{14}\text{N}$ ) and a source isotope ratio  
 190  $\delta^{15}\text{N}_{\text{base}}$  at the base of the food web weighted by the trophic enrichment between TLs ( $\Delta^{15}\text{N}$ ). Different  
 191 sources for benthic and pelagic food chains ( $\delta^{15}\text{N}_{\text{base1}}$  and  $\delta^{15}\text{N}_{\text{base2}}$ ) are often considered in marine  
 192 coastal and/or estuarine environments, to reflect the complexity of trophic food webs in those  
 193 systems. Each individual has to be allocated to one or the other of these food chains through an  $\alpha$   
 194 coefficient (from totally benthic:  $\alpha=1$  to totally pelagic  $\alpha=0$ ), which has to be fixed *a priori* using expert  
 195 knowledge on species' feeding ecology and food web structure.

196

$$\text{TL}_i = 2 + \frac{[\delta^{15}\text{N}_i - \{\alpha \cdot \delta^{15}\text{N}_{\text{base1}} + (1-\alpha) \cdot \delta^{15}\text{N}_{\text{base2}}\}]}{\Delta^{15}\text{N}} \quad \text{Eq. 2}$$

197 Then, a linear mixed model has to be fitted for each food web (benthic and pelagic) independently, by  
 198 selecting species based on  $\alpha$  values For species feeding on both pelagic and benthic preys and sources,  
 199  $\alpha$  values can be comprised between 0 and 1. When  $\alpha > 0.5$  species are allocated to benthic food chain  
 200 and to the pelagic one for  $\alpha < 0.5$  for pelagic.

201

202 Estimating trophic levels is therefore not straightforward, and subjected to multiple sources of  
 203 uncertainty, including measurements of  $\delta^{15}\text{N}$  of individuals and sources at the basis of the food web,  
 204 and estimations of  $\alpha$  values associated to each species considered in the food web. Moreover, while a  
 205 linear increase in  $\delta^{15}\text{N}$  with TL probably oversimplified the mechanism of isotope discrimination  
 206 (Hussey et al., 2014) and equation 2 is not necessarily relevant in a situation where trophic chains are  
 207 intertwined in complex interaction trophic networks.

208

## 209 2.2 ESCROC modeling framework

210 The proposed model (ESCROC) was developed in a Bayesian framework. The Bayesian theorem allows  
211 (1) combining objectively different core metrics accounting for their sensitivity and variability and (2)  
212 providing rigorous uncertainty quantification.

213 The approach in ESCROC was based on the same conceptual framework as stable isotope mixing  
214 models such as MixSIAR (Parnell et al., 2013; Stock et al., 2016). In such models, consumer species are  
215 assumed to feed on a combination of prey items (or sources) that are all known, and that the isotopic  
216 composition  $y_{i,e,t}$  of an individual  $i$  of species  $e$  for tracer  $t$  results from the combined isotopic  
217 composition of the assimilated prey items.

218

### 219 2.2.1 Model formulation

220 In ESCROC, we combine isotope values and contaminant concentrations (in log scale) as chemical  
221 tracers of food web structure.

222 We can therefore describe the mean concentration  $Y$  of a tracer  $t$  for a species  $e$  as a combination of  
223 its consumed prey ( $p$ ) concentrations. In lieu of raw tracer concentrations, scaled values were used by  
224 subtracting the average value and dividing by the standard deviation:  $y'_{i,e,t} = (y_{i,e,t} - \bar{y}_t) / \sigma_t$  so that  
225 all tracers share a common scale. This facilitates the integration of priors and statistical inference of  
226 the model (Bolker et al., 2013).

227

228 Based on stable isotope mixing model assumptions, the tracer composition of a predator was  
229 calculated using the tracer compositions of its prey items:

230

$$Y_{e,t} = \frac{\sum_{p \in \text{prey}(e)} (\rho_{e,p} \cdot q_{p,t} (Y_{p,t} + \Delta'_t))}{\sum_{p \in \text{prey}(e)} (\rho_{e,p} \cdot q_{p,t})} + E_{e,t} \text{ with } E_{e,t} \sim N(0, s_t^2) \quad \text{Eq. 3}$$

231

232 where

233  $Y_{e,t}$  is the average value of tracer (either contaminant or isotope)  $t$  value for species  $e$

234  $\rho_{e,p}$  is the dietary contribution of prey  $p$  for consumer  $e$

235  $q_{p,t}$  is the concentration of  $t$  in prey  $p$

236  $Y_{p,t}$  is the measured mean tracer  $t$  value for prey  $p$

237  $\Delta'_t$  is the enrichment factor for tracer  $t$ . Note that this corresponds to the enrichment for the

238 scaled values, which can be converted to the enrichment in the original scale:  $\Delta_t = \sigma_t \cdot \Delta'_t$

239  $E_{e,t}$  is the species random effect for species  $e$  and marker  $t$  that accounts for inter-specific

240 physiological variability

241 As ESCROC is implemented in a Bayesian framework, priors can be defined for unknown parameters.

242 Priors, corresponding to possible *a priori* distributions of the parameters, can be constructed using

243 knowledge from various sources (expert knowledge, meta-analyses, other field data...). In the absence

244 of external knowledge, uninformative or weakly informative priors can be built.

245  $Y_{e,t}$  as described in Eq3 therefore corresponds to a weighted average of the prey tracer compositions

246 (with weights corresponding to the importance of the prey in the predator's diet and to the

247 concentration of the tracer in the prey), to which we added an enrichment and a species effect. If  $q_{p,t}$

248 are equal among prey items, equation 3 simplifies to:

249

$$Y_{e,t} = \sum_{p \in \text{prey}(e)} (\rho_{e,p} \cdot Y_{p,t}) + \Delta'_t + E_{e,t} \text{ with } E_{e,t} \sim N(0, s_t^2) \quad \text{Eq. 4}$$

250

251

252 Note that the model does not work if two species are both prey and predator of each other because  
 253 equations 3 or 4 become circular.

254

255 Similarly, the variance of the values of the tracer  $t$  for a species  $e$ , denoted  $V_{e,t}$ , can be calculated from  
 256 the variances of the different prey:

$$V_{e,t} = \frac{\sum_{p \in \text{prey}(e)} (\rho_{e,p}^2 \cdot q_{p,t}^2 \cdot V_{p,t})}{[\sum_{p \in \text{prey}(e)} (\rho_{e,p} \cdot q_{p,t})]^2} + B_t \quad \text{Eq. 5}$$

257

258 With  $B_t$  a variable to add potential noise at each trophic level.

259 If the concentration parameters  $q_{p,t}$  are all equals, then the equation simplifies to:

$$V_{e,t} = \sum_{p \in \text{prey}(e)} (\rho_{e,p}^2 \cdot V_{p,t}) + B_t$$

260 In equation 3, if  $Y_{e,t}$  is the concentration of a contaminant (in log10 scale) and assuming that all  
 261 concentration parameters are equal (as in equation 4), then  $\Delta_t$  corresponds to the enrichment in  
 262 contaminant between a prey and its predator, i.e., the enrichment along the food web when trophic  
 263 level increases by 1. Consequently, it corresponds to the slope of the line in Figure 1 and we obtain  
 264  $\Delta_t = \log_{10}(TMF)$ .

265

266 The measured values of a given tracer on a given dataset are supposed to follow a Normal law whose  
 267 parameters are calculated from previous equations. We thus assume that the distribution of the tracer  
 268  $t$  value, for an individual  $i$  of the species  $e$  can be written as:

269

$$y_{i,e,t} \sim \text{Normal}(Y_{e,t}, V_{e,t}) \quad \text{Eq. 6}$$

270

271 In case of the concentration measurement of a given tracer is null or not reliable for analytical reasons  
 272 (e.g. below the detection threshold of a given measurement method), it can be considered as a left-

273 censored data. The cumulative distribution of the normal distribution is thus used instead of the  
274 density of probability.

275

## 276 *2.2.2 Model calibration and priors construction*

277 We propose here a selection of possible priors that can be implemented for most of the model  
278 applications, but adaptation can be made in relation with available data or expert knowledge.

279

### 280 *Selection of priors for main parameters*

281 Priors can be supplied for the enrichment factors either on the transformed scale or on the original  
282 scale. Informative priors for Nitrogen and Carbon TEF ( $\Delta_N$  and  $\Delta_C$  see Eq. 3) can be inferred from the  
283 literature (e.g. Post 2002). For instance, these priors can be implemented as follows:

284

$$\Delta_N \sim \text{Normal}(3,1) \quad \text{Eq. 7}$$

$$\Delta_C \sim \text{Normal}(0,1) \quad \text{Eq. 8}$$

285

286 corresponding to a TEF value for N around 3 and around 0 for C.

287

288 A non-informative prior can be used for the enrichment factors of all tracers corresponding to PFAS  
289 concentrations:

290

$$\Delta'_t \sim \text{Normal}(0,10) \quad \text{Eq. 9}$$

291

292 Finally, non-informative priors can be used for other parameters (residual variation and random  
293 effect) of model formulation (Eq. 3 and Eq. ):)

294

$$B_t \sim \text{Inverse Gamma}(0.01, 0.01) \quad \text{Eq. 10}$$

$$\sigma_t \sim \text{Uniform}(0.01, 10) \quad \text{Eq. 11}$$

295

296 *Diet matrix*

297 Information on trophic interactions should be implemented in the model. At first, we can only specify  
298 if a prey  $p$  can be predated by a consumer  $e$  based on the evidence of predator-prey relationships  
299 using field data.

300

301 An uninformative prior for the dietary contribution of prey  $p$  for consumer  $e$  ( $\rho_{e,p}$  see Eq. 3) can then  
302 be constructed by assuming that it follows a Dirichlet distribution:

303

$$\{\rho_{e,1}, \dots, \rho_{e,n}\} \sim \text{Dirichlet}(\{\lambda_{e,1}, \dots, \lambda_{e,n}\}) \quad \text{Eq. 12}$$

304

305 where  $\lambda_{e,p} = 1$  if  $e$  feeds on  $p$  and  $\lambda_{e,p} = 0$  otherwise;  $n$  being the number of species in the considered  
306 food web.

307

308 Informative priors can be implemented if external data, such as stomach contents, are available.  
309 However, considering the implementation framework, trophic loops cannot be included in the food  
310 web description.

311 *Constructing priors for tracer values using data*

312 Average tracer compositions of predators are calculated from prey tracer compositions. However, if a  
313 species does not have prey (e.g. primary producers), or if some prey of a species are not present in the  
314 dataset (i.e., no contamination data or isotopic measurements), it is necessary to provide priors for

315 this species. We will see in the case study how an informative prior can be constructed. However, in  
316 the absence of external data, a weakly informative prior can be implemented as follows:

317

$$Y_{e,t} \sim \text{Normal}(0,10) \quad \text{Eq. 13}$$

$$V_{e,t} \sim \text{Gamma}(0.01,0.0) \quad \text{Eq. 14}$$

318

### 319 2.2.3 *Outputs and Implementation*

#### 320 *Outputs*

321 Main outputs of ESCROC consist in posterior distributions of the estimated parameters. Three main  
322 types of outputs can be obtained from ESCROC: diet compositions of each consumer of the  
323 investigated food web, enrichment factors for each tracer, and TMF estimates (with associated  
324 credibility interval).

325

326 As a mixing model, ESCROC allows estimating a distribution of the proportion of each prey in the diet  
327 of the predators in the investigated food web.

328

329 Furthermore, by considering posterior distribution of enrichment factors (see Eq. 3, Figure 4) for N  
330 and C isotopic ratios, ESCROC allowed re-estimating TEF values, which are usually empirically fixed in  
331 the literature on isotope-based trophic studies, with aforementioned uncertainty. TEF can indeed be  
332 estimated using the median of the distribution and 2.5% and 97.5% quantiles, providing associated  
333 95%-credibility intervals. In the same way, estimated posterior distributions of enrichment factors for  
334 contaminants can be used to estimate TMFs and associated credibility intervals. In this case, TMFs  
335 corresponds to  $10^{\Delta t}$ . Similarly, an estimation of TMFs can be provided using the median of the  
336 distribution and bounds of the 95%-credibility interval, computed from 2.5% and 97.5%-quartiles.  
337 Associated with TMF estimates, the probability of a contaminant to be biomagnified in the

338 investigated food web can then be computed by estimating the probability of  $\Delta_t$  to be positive (*i.e.*  
339 corresponding to a TMF value greater than 1).

340

### 341 *Implementation*

342 ESCROC was implemented using the R software (R Development Core Team, 2006) and the integrated  
343 development environment (IDE) R-studio (the model being run using *coda* and *runjags* packages). For  
344 the Bayesian part of the model, the JAGS language was used (Plummer et al., 2016). The model  
345 convergence can be checked using Gelman and Rubin tests (Brooks and Gelman, 1997; Gelman and  
346 Rubin, 1992). A first beta version of the R-package (escrocR) implementing the method is available on  
347 GitHub (<https://github.com/lrstea/escroc>). It can be cited as follows:

348 Hilaire Drouineau, Marine Ballutaud and Jeremy Lobry (2018). EscrocR: a R package implementing the  
349 model ESCROC. R package version 0.0.0.9000.

350

## 351 **2.3 Illustrative example: PFAS in the Gironde estuarine food web**

352 In this illustrative example, the method was applied to a dataset on PFAS contamination in the  
353 Gironde estuarine food web. The main aims were to estimate (1) TMF values for a set of PFAS  
354 previously described and analyzed (Munoz et al. 2017), (2) enrichment factors of two isotopes of  
355 nitrogen and carbon and (3) the diets of all species within the trophic network.

356

357

### 358 **2.3.1 Dataset**

359 Data used in this study are taken from Munoz (2015) and Munoz et al (2017). Samples were collected  
360 between May and November 2012 in the mesohaline zone of the Gironde estuary. Amongst the initial  
361 dataset of 147 biological samples from 18 species or group of species, a subset of data was used

362 comprising 138 samples from 16 species, for which both isotopic data ( $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$ ) and PFAS  
363 concentrations were available. We selected 5 PFAS (L-PFOS, FOSA, PFOA, PFNA and PFUnDA, see Table  
364 S1) based on their occurrence in biota samples, the existence of censored data (considering PFAS with  
365 different proportions of censored data), the TMF values previously estimated, and the variety of  
366 chemical structures (ie., fluoroalkyl chain length or functional group - Table 1).

367

### 368 2.3.2 *Specific priors*

369 The priors for diet compositions were directly built from the trophic network illustrated in Figure 2.

370

371 For two groups (gammarids and copepods), instead of using uninformative prior as proposed in  
372 equations 13 and 14, we built an informative prior to take advantage of pre-existing data on nitrogen  
373 and carbon isotopic ratios (David, unpublished data). We specified a normal prior parameterized with  
374 the mean and the standard errors in pre-existing available tracer composition data:

375

$$\frac{Y_{e,t} - \bar{y}_t}{\sigma_t} \sim \text{Normal}(\mu_{e,t}, se_t^2) \quad \text{Eq. 15}$$

376

377 with

378 e species or group of organism (gammarid or copepods)

379 t type of chemical tracer (carbon or nitrogen)

380  $\mu_{e,t}$  arithmetic mean of tracer values in an independent dataset

381  $se_{e,t}$  associated standard error

382

383 Regarding the variances, the estimator of the variances in the samples follows the distribution:

384

$$u_{e,t} \sim \frac{V_{e,t}}{\sigma_t^2 \cdot n_{e,t} - 1} \chi^2_{(n_{e,t}-1)} \quad \text{Eq. 16}$$

385

386 with

387  $v_{e,t}$  the estimator of the variance in the samples

388  $n_{e,t}$  number of samples

389  $e$  species or group of organism (gammarid or copepods)

390  $t$  type of chemical tracer (carbon or nitrogen)

391

392 Specific gammarid and copepod priors were thus built as follows:

393

$$\frac{1}{V_{e,t}} \sim \frac{u_{e,t}}{\sigma_t^2 \cdot n_{e,t} - 1} \chi^2_{(n_{e,t}-1)} \quad \text{Eq. 17}$$

394

### 395 2.3.3 *Outputs and model implementation*

396 We set  $q_{p,t}=1$  for all species in the model. We also computed an index  $\alpha_e$  for each predator to compare

397 to the  $\alpha$  values (see section 2.1.1) arbitrarily assigned in the Munoz et al. dataset (2017).

$$\alpha_e = \sum_{p \in \text{prey}(e)} (\rho_{e,p} \cdot \alpha_e)$$

398 For gammarids and copepods, we set  $\alpha_e=0$  (pelagic), and we set  $\alpha_e=0$  (benthic) for nereids and crabs.

399 The model was fitted using a MCMC (Monte Carlo Markov Chain) method. Three chains were used in

400 parallel with 1 million preliminary iterations – burnin – followed by 150,000 iterations to assess

401 posterior distributions. We run the model with JAGS software version 4.1.0 and check convergence

402 using the Gelman-Rubin diagnostic ( $\text{gelman.test} < 1.05$ ).

403

## 404 **3 Results**

### 405 **3.1 Model calibration and convergence**

406 Values of Gelman indices confirmed the model convergence for 68 parameters out of 70. The only two  
407 parameters for which the model did not converge corresponded to tracer composition of prey items  
408 with 100% of censored data.

409

410 The model fitted observations as suggested by the plot of predicted *posterior* distributions of mean  
411 species tracer composition against observed values (Figure 3).

412

### 413 **3.2 Outputs**

#### 414 **3.2.1 Diet compositions**

415 Diet compositions were estimated for each predator of the Gironde estuarine food web for which  
416 both isotope and PFAS data were available (see Figure 4 as an example). A global diet matrix can then  
417 be obtained by compiling all the diet values estimated by ESCROC using both N and C isotopic ratios  
418 and the 5 PFAS concentrations (Table 2).

419

420 For most species, the coefficients relating the species to the pelagic and benthic food webs were  
421 rather consistent with the expert knowledge used in Munoz et al. (2017) though the model tends to  
422 consider less species as benthic (Table 3). Those results confirm that it is very difficult to separate  
423 species into a set of two independent food-webs, as required by usual TMF estimation methods.

424

#### 425 **3.2.2 Enrichments and TMF**

426 TEF values and the associated uncertainty were estimated, as well as TMFs estimates and  
427 biomagnification probabilities (Figure 4 and Table 4).

428

429 Non-informative priors were used for the PFAs enrichment factors, as highlighted by the flat curve  
430 lines in Figure 4. In these cases, ESCROC provided informative posterior estimates for contaminants  
431 TMFs. Conversely, informative priors based on literature were used for isotopes TEFs. Although the  
432 estimated posterior distribution appeared consistent for  $\delta^{15}\text{N}$ , it was significantly different for  $\delta^{13}\text{C}$   
433 even if the classical value used in the literature and in our prior definition (TEF = 0) is comprise in the  
434 posterior credibility interval.

435

436

## 437 **4 Discussion**

### 438 **4.1 Limits and benefits of the ESCROC model formulation**

#### 439 *4.1.1 Bayesian framework, a priori information and uncertainty propagation*

440 ESCROC was implemented in an innovative and flexible Bayesian framework to estimate TMFs and  
441 associated uncertainty. This modeling approach presents several advantages.

442

443 Unlike frequentist methods previously mentioned (e.g. linear models and mixed models), the Bayesian  
444 framework allows to account rigorously for uncertainty propagation in measurements (e.g., TL  
445 estimates) to TMF estimates. Starrfelt et al. (2013), for instance, recommended the use of Bayesian  
446 inference to account for measure uncertainty in contaminants concentrations and isotope ratios  
447 ( $\delta^{15}\text{N}$ ), as well as for variability and uncertainty related to TL estimates, in order to improve the  
448 precision of TMF estimations. ESCROC goes even further, by providing credibility intervals for all  
449 parameters, in particular for contaminants TMFs and isotopes TEFs. By doing so, ESCROC represents a  
450 noteworthy methodological advance as compared to traditional methods for TMF estimation.

451

452 Based on both a contaminant propagation model and an isotope mixing model, ESCROC combines and  
453 enriches both modeling approaches. The contaminant propagation model therefore benefits from  
454 integrated diet estimations. As such, TMF estimates are of more generic nature, as they no longer  
455 depend on a pre-specified trophic chain structure that the model user provides (and usually simplify).  
456 This model rather accounts for the whole food web complexity at once, and provides generic TEF  
457 estimates. Similar to widely-used mixing models (Parnell et al., 2008; Parnell et al., 2010; Parnell et al.,  
458 2013; Stock et al., 2016), ESCROC allows estimating diet compositions in investigated food webs (See  
459 Supplementary Materials S2 for a preliminary comparison of both approaches). It allows going even  
460 further than classical mixing models. Indeed, using contaminants in addition to isotopes data increases  
461 the number of chemical tracers and allow the estimation of isotope enrichment factors, a significant  
462 improvement as compared to the use of fixed *a priori* -values from the literature (usually from Post  
463 (2002)), values which are known to not perform as well in various environmental contexts or for  
464 contrasted food webs). Furthermore, our modelling approach partly addresses some of the  
465 recommendations listed by Hussey et al. (2014) for estimating isotope discrimination. The full  
466 Bayesian estimation framework indeed provides a pragmatic and very flexible estimation of  
467 enrichment factors. The framework is thus generic and can be applied in all ecosystem contexts.  
468 Furthermore, a random effect 'Species' is added in the model. Thus, although the TEF and TMF  
469 estimation are still considered globally constant through the food web, they are modulated species by  
470 species. By doing this, ESCROC did not specifically consider that isotope discrimination varies with  
471 trophic position but it allowed the estimations to vary for every species.

472 In the Gironde estuarine case study, the TEF for  $\delta^{13}\text{C}$  estimated from ESCROC considerably differs from  
473 the one in literature. This is mostly explained by the fact we used data from studies on marine  
474 environments to compute the prior distribution while estuarine ecosystems are usually enriched on  
475 continental organic carbon which signature is different. However the model allowed not only  
476 estimating more accurately the TEF distribution for  $\delta^{13}\text{C}$  in the estuarine context but the estimates  
477 remains consistent with the reference literature (e.g. Post, 2002). This result further highlights the

478 need for accurate TEF estimates based on the best available knowledge, as already advocated in  
479 multiple reviews of isotope-based ecological studies (Martinez del Rio et al., 2009; Layman et al.,  
480 2012; Bastos et al., 2017).

481

482 Additionally, prior knowledge incorporation is a really significant advantage of both ESCROC and  
483 MixSIAR approaches (Parnell et al., 2010; Parnell et al., 2013). Indeed, the Bayesian framework is  
484 especially well-adapted to integrate *a priori* information such as expert knowledge or external  
485 datasets. For instance, in the present case study, external information about copepods isotopic  
486 composition were combined in an informative prior as available data were uncertain. Similarly, expert  
487 knowledge and food web data (such as stomach contents) could have been used to compute more  
488 informative prior on trophic interactions, instead of using an uninformative prior for the contribution  
489 of each prey in each consumer diet.

490

#### 491 **4.1.2 Computing TMF and associated bioaccumulation probabilities**

492 Although the initial purpose of the model was to compute TMF estimates and associated uncertainty,  
493 the ECROC modeling approach provides a comprehensive framework for the understanding of  
494 contaminants transfers in a complex food web. In relation with the Bayesian framework used, ECSROC  
495 indeed provides a biomagnification probability, which corresponds to the probability of a particular  
496 contaminant to be biomagnified in the investigated food web. This innovative feature is especially  
497 relevant for risk assessment. In fact, this probability directly expresses the risk a manager would take  
498 by classifying a given contaminant as “biomagnifiable” in a given food web. As such, ESCROC model  
499 represents an important tool to support decision making. For instance, estimated thresholds of risks  
500 could be used to define contamination levels for which additional monitoring is required, as well as  
501 levels for which specific management measures appear mandatory.

502

### 503 4.1.3 *Limitations*

504 Despite the aforementioned advances and advantages, ESCROC modeling approach also presents two  
505 major limitations.

506 First, considering the implementation framework, trophic loops cannot be considered in the food web  
507 description, although such phenomena may exist in nature. We can cite cannibalism as an illustrative  
508 example, a process relatively common in aquatic food webs, in which adult consumers sometimes  
509 prey upon juveniles from the same species (Livingston, 2002). In our case example, we assumed such  
510 flows to be negligible or, at least much weaker than direct prey-predator trophic flows. This  
511 assumption seems reasonable with regards to available knowledge on food webs in our study system  
512 (see for instance Lobry et al., 2008; Selleslagh et al., 2012; Tecchio et al., 2015 for French estuaries).

513 Another limitation of the ESCROC modeling approach lies in technical aspects. First, computing time  
514 can reveal quite long depending on food web complexity and computer devices used. Second, data  
515 about multiple tracers need to be included to avoid any underdetermination issues, as TMFs and TEFs  
516 are estimated together and at the whole trophic network scale, since the number of tracers (either  
517 isotopes or contaminants) should be large enough to avoid any underdetermination issue. This implies  
518 considerable efforts in sample collection, preparation, and chemical analysis, an even higher than for a  
519 classical mixing model such as MixSIAR.

520

## 521 4.2 **Diagnosis about PFAS in the Gironde estuary**

522 The computed TMF values of the five investigated PFAS are not significantly greater than 1 with a  $\alpha$ -  
523 risk at 95%. This implies that, considering the results of the present study, none of the five  
524 investigated contaminants can be considered as 'biomagnifiable' in the Gironde estuarine food web.  
525 However, when considering the biomagnification probabilities associated with TMF estimates, results  
526 are more contrasted. The diagnosis actually depends on the risk-level a manager is ready to accept.  
527 For instance, if a risk-level was fixed to 30% in a precautionary approach, PFUnDA, FOSA, and more

528 particularly L-PFOS would be considered to be biomagnified in the food web with 72%, 86% and 92%  
529 of certainty, respectively (Table 4). These considerations give sense to the computation of  
530 biomagnification probabilities within the ESCROC modeling tool.

531

532 Our ESCROC-based diagnosis about PFAS contamination in the Gironde estuarine food web are slightly  
533 different from previous assessment from Munoz et al. (2017). In the latter study, the 5 investigated  
534 contaminants were considered to have been magnified at both the benthic chain and the whole food  
535 web levels, whereas only one TMF estimates (FOSA) was greater than one when considering the  
536 pelagic chain. Differences between Munoz et al. (2017) and our interpretation probably arises from  
537 differences in the methodological approaches. First, pelagic and benthic data were combined in  
538 ESCROC's estimates as well as in the Munoz's pooled estimates (Table S17 in Munoz et al., 2017). As  
539 we saw in section 3.2.1, such combination is probably more realistic than separating benthic and  
540 pelagic food chains, and this difference can lead to significant contrasts in TMF estimates. Second,  
541 the Bayesian model formulation provides a better integration of uncertainty propagation than  
542 traditional linear models. This leads to greater associated uncertainties and larger credibility intervals  
543 than the confidence intervals estimated with the LMEC method used in Munoz et al. (2017) for pooled  
544 TMFs. Third, Munoz et al. (2017) study was based on TMF estimates only whereas ESCROC provides  
545 both TMFs estimates and biomagnification probabilities. Considering only TMFs estimates however,  
546 both studies also led to contradictory results. When considering biomagnification probabilities as well,  
547 our and Munoz et al. (2017)'s diagnoses remain different, but partly converge for PUnDA, FOSA and  
548 L-PFOS (see above). Finally, previous diagnoses from Munoz et al. (2017) based on classical TMF  
549 estimations through linear regression appear questionable, with regards to aforementioned statistical  
550 consideration, although such a method can still be seen as a simple and useful approach to perform  
551 comparative studies for comparing biomagnification of selected chemicals in a given ecosystem.  
552 Nevertheless, the results obtained in the present study also plead for a precautionary approach when

553 interpreting contaminant transfer data, and for using biomagnification probabilities rather than TMF  
554 values alone.

555

#### 556 4.3 Perspectives for the ESCROC modeling approach

557 More than an innovative estimation framework for TMFs in complex food webs, ESCROC can also be  
558 viewed as an improved mixing model for food web analyses. Considering more chemical tracers than  
559 the classical N and C isotopes indeed clearly improves diet matrix estimations. Results obtained for the  
560 Gironde estuarine food web are in line with those previously obtained by Pasquaud et al. (2008) and  
561 Pasquaud et al. (2010) using both stomach contents and isotope data, and by Lobry et al. (2008) using  
562 literature compilation and mass-balance modeling. Moreover, in the ESCROC modeling approach  
563 presented herein, tracers are contaminants, but other kind of isotopes (e.g.  $\delta^{34}\text{S}$ ) can be used as well.  
564 As highlighted by Mackay et al. (2016), several processes related for instance to hydrophobicity or  
565 rates of biotransformation and growth can influence contaminant biomagnification. However, as far as  
566 they biomagnify, any type of tracer can be used in the ESCROC modeling framework. In their *Best*  
567 *practice in Ecopath with Ecosim food-web models for ecosystem-based management*, Heymans et al.  
568 (2016) underlined that: "Diet estimates for functional groups can also be obtained from stable isotopic  
569 analyses using Bayesian isotopic mixing models." By providing rigorous estimates of diet matrices (and  
570 associated uncertainty) based on chemical tracers (eventually combined with expert knowledge  
571 and/or external information), diet composition estimates from ESCROC modeling could so be used to  
572 calibrate diet matrices in mass-balanced food web models.

573

574 Other perspectives could also relate to mass-balance equations (similar than the ones used in  
575 Ecopath) which could also be implemented in the ESCROC model formulation, in order to provide an  
576 innovative modeling framework of 'biomass propagation'. This would allow a very integrated view of

577 aquatic ecosystem food webs, with simultaneous estimations of biomass, contaminants, and isotopes  
578 transfers..

579

580

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586

587

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1 **Table 1.** Data summary with Average [Min; Max] values for each tracer (5 PFAS, d13C and d15C). n: number of samples on which contaminants and isotopes were measured for each species. –  
 2 corresponds to censored values (*i.e.* values below the limit of quantification for each specific contaminant). Contaminant concentrations are given in ng g<sup>-1</sup> wet weight of the whole-body (from  
 3 Munoz et al., 2017).

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Species	L-PFOS (ng g <sup>-1</sup> )		FOSA (ng g <sup>-1</sup> )		PFOA (ng g <sup>-1</sup> )		PFNA (ng g <sup>-1</sup> )		PFUnDA (ng g <sup>-1</sup> )		δ <sup>13</sup> C (‰)		δ <sup>15</sup> N (‰)		n
Anchovy	3.58	[1.6;6.3]	1.52	[0.9;2.2]	0.12	[-;0.1]	0.25	[0.0;0.1]	0.31	[-;0.2]	-18.71	[-19.4;-18.2]	7.99	[8.8;12.0]	6
Brown shrimp	6.57	[3.8;8.0]	3.75	[1.5;5.4]	0.37	[0.3;0.5]	1.44	[0.8;2.1]	0.51	[0.5;0.6]	-18.07	[-18.7;-17.1]	12.60	[11.5;13.3]	3
Common seabass	6.68	[3.0;14.3]	1.39	[0.4;2.2]	0.30	[-;0.2]	0.39	[0.1;0.5]	0.63	[0.3;1.4]	-17.58	[-19.8;-15.7]	12.65	[10.7;15.0]	9
Copepods	1.24	[0.8;1.6]	0.33	[0.3;0.4]	0.31	[0.1;0.5]	0.56	[-;0.1]	0.32	[0.1;0.2]	-23.43	[-26.4;-21.6]	9.04	[10.6;13.1]	3
Crabs	2.42	[1.8;3.0]	0.22	[0.2;0.2]	2.42	[1.7;3.0]	1.40	[0.8;2.3]	0.31	[-;0.4]	-14.84	[-15.8;-13.3]	6.03	[7.9;10.1]	3
Flounder	5.71	[0.7;21.7]	0.90	[0.1;3.8]	0.40	[-;1.6]	1.18	[0.2;7.9]	0.73	[0.1;1.9]	-18.00	[-23.8;-14.1]	10.53	[10.7;15.2]	13
Gammarids	2.36	[1.5;2.8]	0.52	[0.4;0.7]	1.00	[0.3;2.1]	0.48	[0.3;0.6]	0.44	[0.4;0.5]	-22.51	[-24.2;-19.4]	9.10	[8.4;9.1]	3
Goby	2.35	[2.0;2.4]	0.18	[0.2;0.2]	-	[-;-]	0.14	[0.1;0.2]	0.41	[-;0.4]	-19.21	[-19.2;-19.0]	11.53	[11.2;11.8]	3
Meagre	4.39	[2.5;10.7]	3.30	[2.3;5.4]	-	[-;-]	0.34	[-;0.3]	0.39	[0.2;0.8]	-16.89	[-18.4;-16.3]	14.19	[13.1;14.8]	12
Mullet	2.53	[0.8;4.0]	0.36	[0.1;0.8]	-	[-;-]	0.36	[-;0.3]	0.94	[-;4.0]	-21.55	[-28.2;-16.5]	10.11	[8.7;13.6]	12
Mysids	3.14	[2.4;3.8]	1.20	[0.9;1.5]	0.86	[0.1;0.1]	0.15	[0.1;0.2]	0.15	[0.1;0.2]	-21.35	[-22.8;-19.9]	10.75	[8.2;13.3]	2
Nereis	2.90	[2.0;21.0]	0.59	[0.4;0.8]	5.21	[3.6;8.2]	6.01	[3.7;8.3]	0.49	[0.1;0.3]	-16.40	[-18.2;-15.2]	4.85	[9.9;10.7]	5
Oyster	0.52	[0.1;0.1]	0.74	[0.4;0.8]	0.16	[-;0.0]	0.11	[-;0.0]	0.16	[-;0.0]	-19.66	[-21.2;-18.7]	7.86	[6.1;8.5]	4
<i>Scrobicularia</i>	0.31	[0.2;0.5]	0.27	[0.2;0.3]	0.26	[-;0.0]	0.31	[0.0;0.0]	0.49	[0.0;0.1]	-16.25	[-17.5;-15.5]	7.65	[7.6;7.7]	3
Sole	9.12	[0.7;19.2]	1.22	[0.1;2.4]	0.83	[0.0;2.5]	3.73	[0.2;11.8]	0.47	[0.1;1.3]	-14.19	[-20.8;-14.3]	13.11	[11.1;14.9]	31
Spotted seabass	4.87	[2.2;10.5]	2.22	[1.0;4.2]	0.41	[-;0.7]	0.27	[-;0.7]	0.39	[0.1;1.0]	-16.45	[-20.1;-14.8]	13.78	[11.8;15.4]	28
Sprat	1.64	[0.3;3.8]	3.03	[1.7;4.7]	-	[-;-]	0.15	[-;0.1]	-	[-;-]	-17.19	[-17.4;-16.8]	11.60	[11.3;11.9]	3
White shrimp	3.02	[2.7;3.0]	3.24	[2.7;3.6]	0.39	[0.3;0.4]	0.39	[0.3;0.4]	0.42	[0.3;0.5]	-14.00	[-20.9;-19.4]	8.00	[10.5;11.2]	3

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**Table 2.** Diet matrix of the Gironde estuarine food web. Each value corresponds to the proportion of the prey in line in the diet of the predator in column. It was computed as the median value of the proportion of each of the listed preys in the diet composition of the predators estimated by ESCROC using both N and C isotopic ratios and 5 PFAS concentrations.

Predators																
Prey	Anchovy	Common seabass	Spotted seabass	Flounder	Goby	Meagre	Mullet	Sole	Sprat	White shrimp	Brown shrimp	Mysids	Gammarids	Copepods	Nereis	Crab
Anchovy	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Common seabass	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Spotted seabass	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Flounder	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Goby	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Meagre	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Mullet	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Sole	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Sprat	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
White shrimp	0.00	0.08	0.23	0.00	0.00	0.17	0.00	0.10	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Brown shrimp	0.00	0.09	0.00	0.00	0.00	0.16	0.00	0.13	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Mysids	0.41	0.38	0.15	0.00	0.83	0.11	0.06	0.00	0.00	0.49	0.54	0.00	0.00	0.00	0.00	0.00
Gammarids	0.12	0.29	0.19	0.71	0.17	0.09	0.00	0.47	0.59	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Copepods	0.46	0.00	0.13	0.00	0.00	0.10	0.94	0.00	0.41	0.51	0.46	1.00	0.00	0.00	0.00	0.00
Nereis	0.00	0.06	0.28	0.05	0.00	0.35	0.00	0.12	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Crab	0.00	0.00	0.00	0.22	0.00	0.00	0.00	0.07	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

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**Table 3.**  $\alpha_e$  coefficients (1=benthic, 0=pelagic) estimated by the model (quantiles of the posterior distributions) for the different species and comparison with the values estimated a priori by Muñoz et al. (2017).

Species	ESCROC (quantiles of posterior distributions)			Muñoz et al. (2017)
	2.5%	50%	97.5%	
Anchovy	0.00	0.00	0.00	0.00
Common seabass	0.00	0.06	0.26	0.13
Spotted seabass	0.06	0.28	0.52	0.23
Flounder	0.11	0.29	0.64	0.82
Goby	0.00	0.00	0.00	0.05
Meagre	0.10	0.35	0.60	0.44
Mullet	0.00	0.00	0.00	0.96
Sole	0.04	0.22	0.51	0.67
Sprat	0.00	0.00	0.00	0.00
White shrimp	0.00	0.00	0.00	0.00
Brown shrimp	0.00	0.00	0.00	0.00
Mysids	0.00	0.00	0.00	0.00
Gammarids	0.00	0.00	0.00	0.00
Copepods	0.00	0.00	0.00	0.00
Nereis	1.00	1.00	1.00	1.00
Crab	1.00	1.00	1.00	1.00

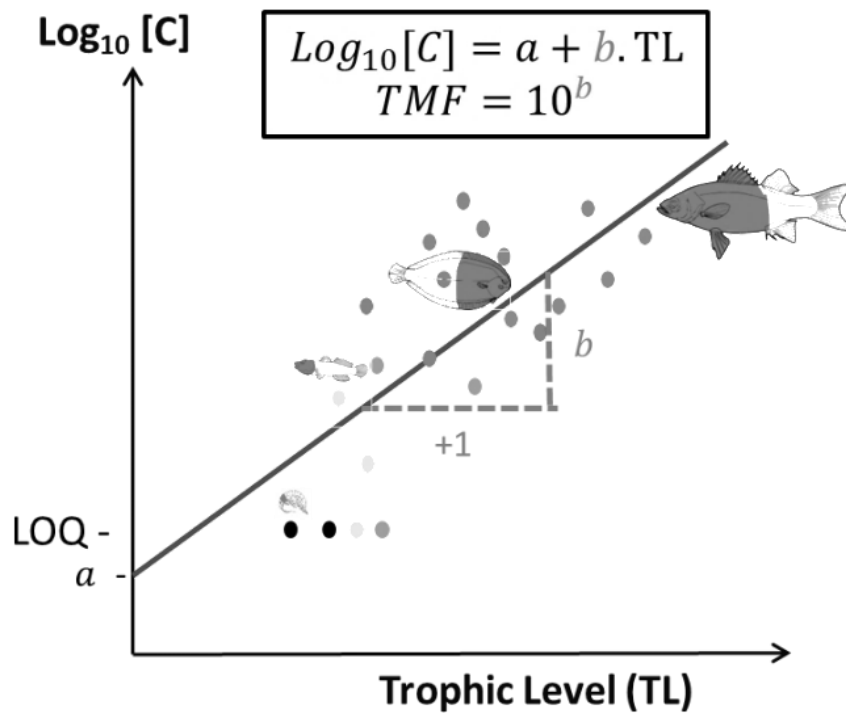
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**Table 4.** Estimates and associated 95% -credibility interval of isotopes TEF and contaminants TMF in the investigated Gironde estuary food web with associated biomagnification probabilities. Munoz et al. (2017) are TMF values via LMEC methods when pooling all samples. See text for details.

Tracers	Median	Bounds of the 95% credibility interval		Biomagnification probability	Munoz et al. (2017)
Isotopes TEF					
$\delta^{15}\text{N}$	2.76	1.55	3.80	NA	
$\delta^{15}\text{C}$	1.60	-0.23	3.09	NA	
Contaminants TMF					
L-PFOS	1.65	0.77	3.28	0.92	1.5 [1.5;1.6]
FOSA	2.29	0.48	6.50	0.86	1.9 [1.9;2.0]
PFOA	0.28	0.04	1.50	0.06	2.0 [1.9;2.1]
PFNA	0.69	0.13	4.22	0.32	1.5 [1.4;1.6]
PFUnDA	1.30	0.47	3.02	0.72	1.1 [1.0;1.2]

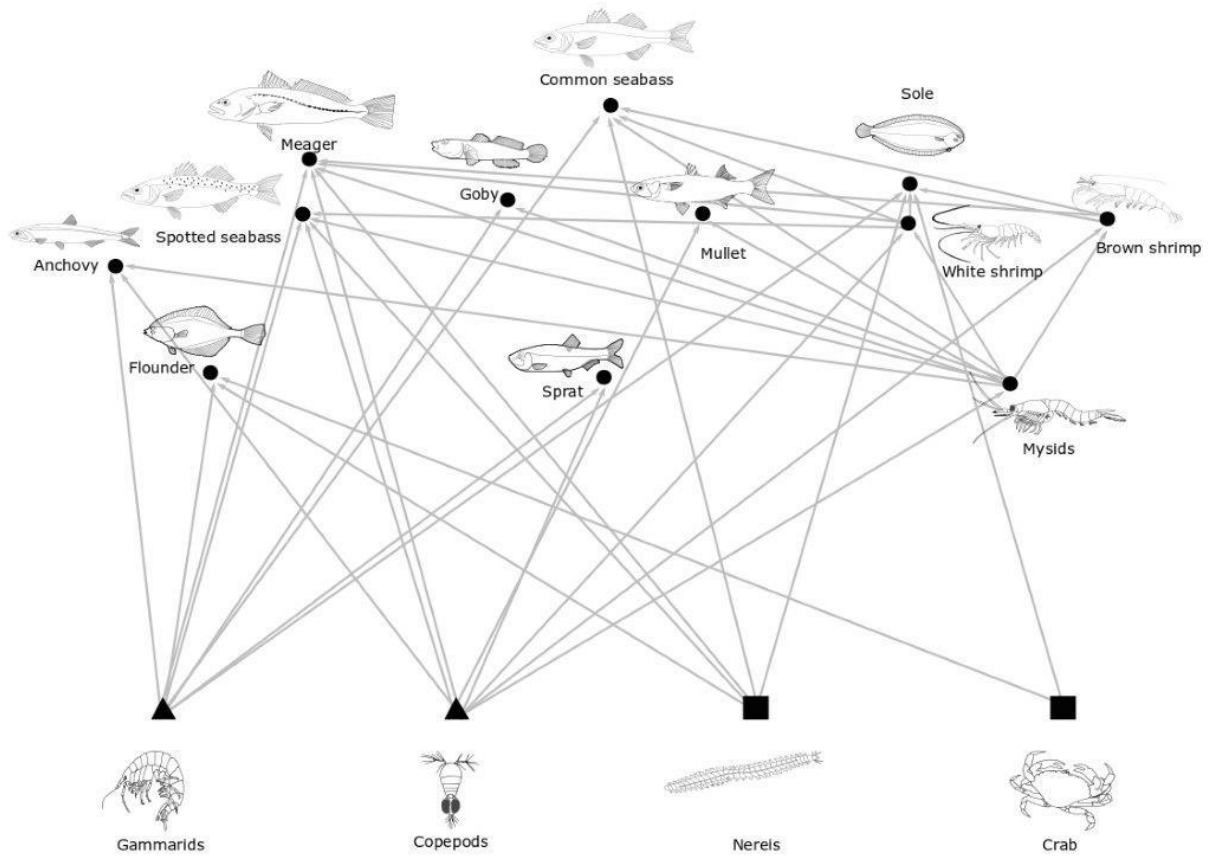
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**Figure 1.** Basis of Trophic Magnification Factor (TMF) estimation.  $\text{Log}_{10}[C]$ : log-transformation of the contaminant concentrations [C]; a: intercept of the regression ; b: slope of the regression. LOQ: limit of quantification. See text for details.

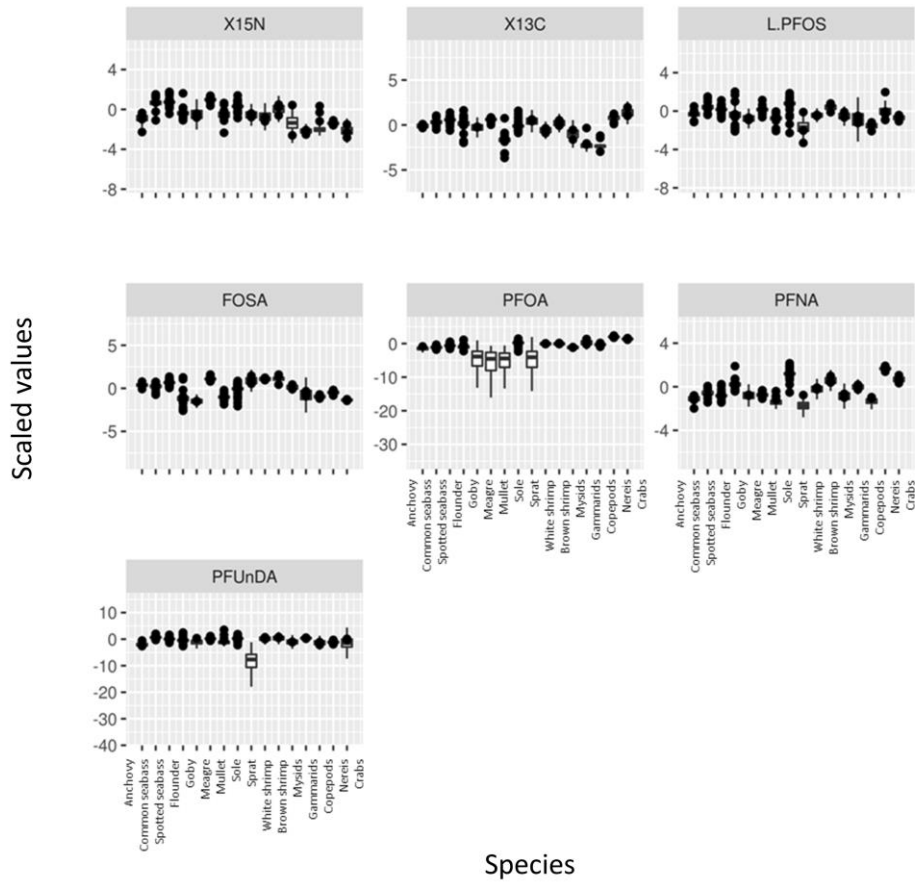
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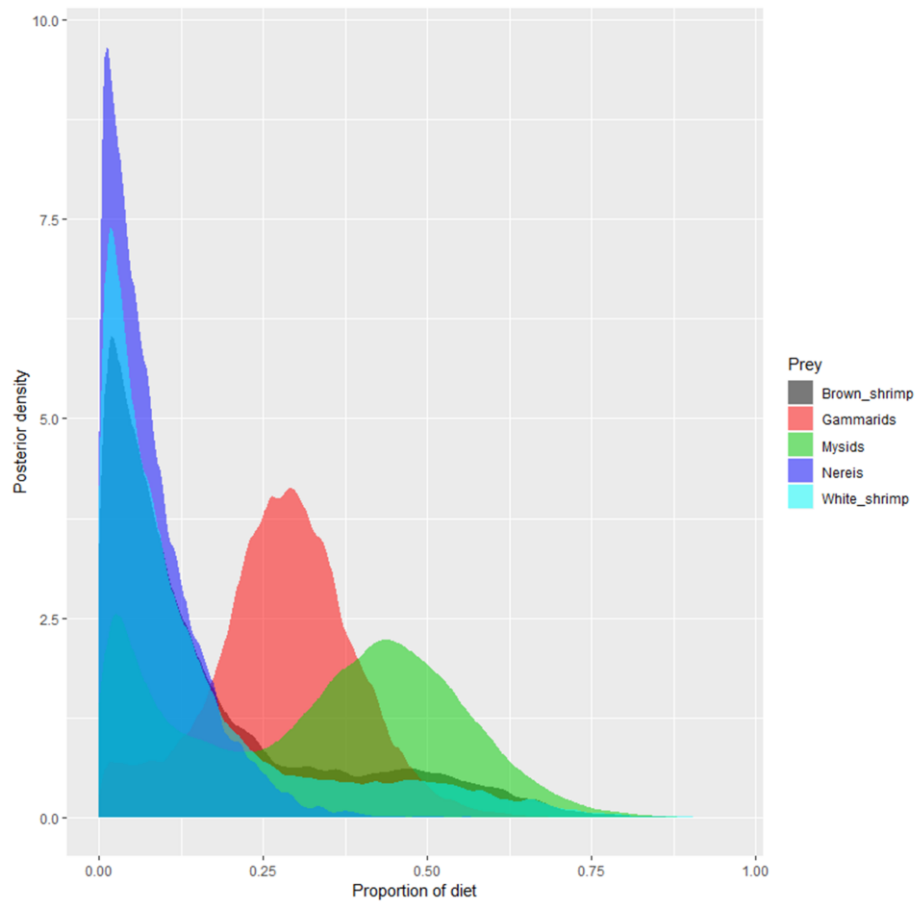
9 **Figure 2.** Synoptic diagram of the Gironde estuarine food web (from Pasquaud et al., 2010)

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11 **Figure 3.** Predicted *posterior* distributions (boxplot) and observations (points) of tracer values by species. Values are scaled  
 12 (see text for details)  
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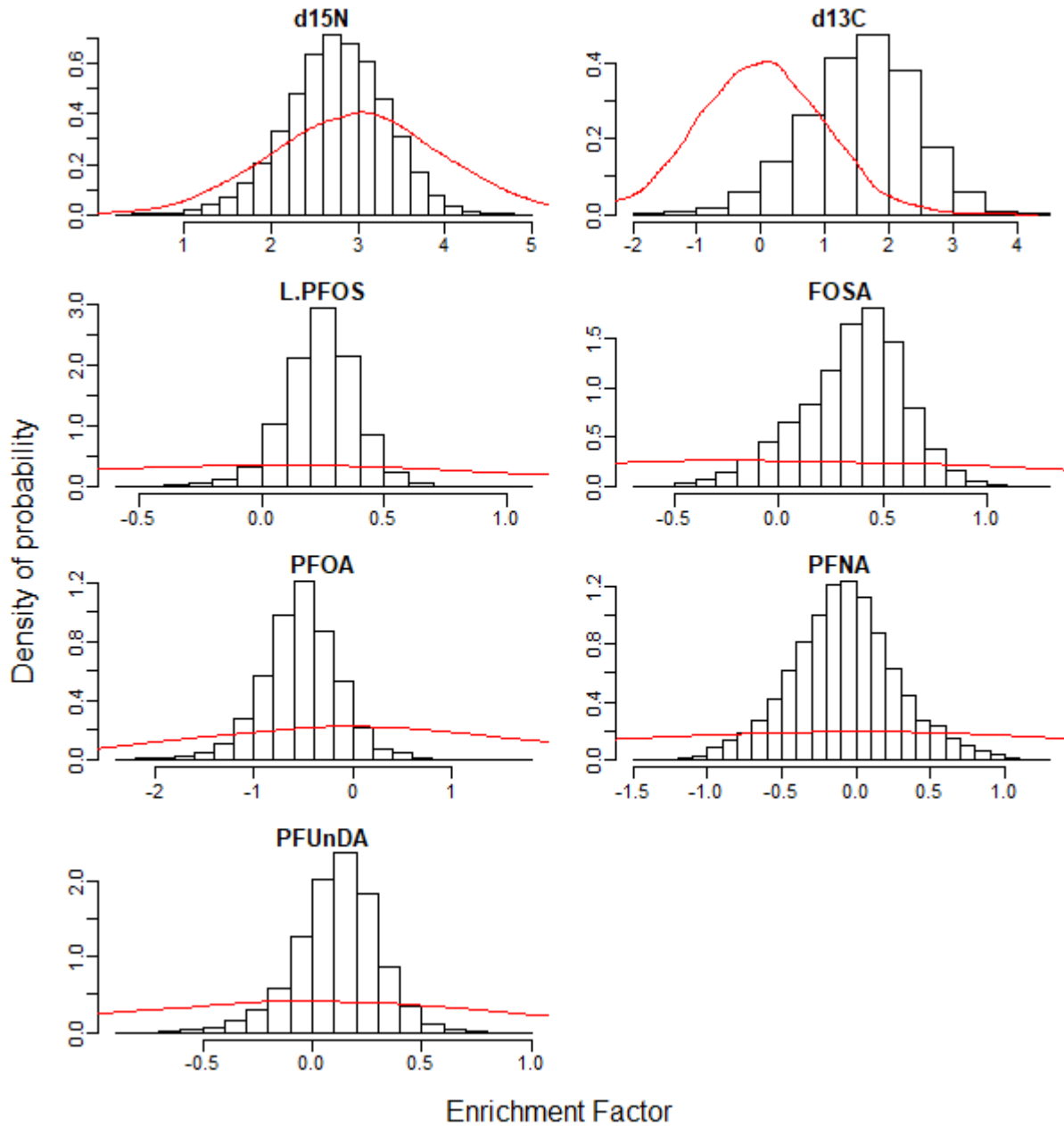
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**Figure 4.** Probability density (in y-axis) of the proportion (in x-axis) of each of the listed preys in the diet composition of the common seabass in the Gironde estuary estimated by ESCROC using both N- and C- isotopic ratios and 5 PFAS concentrations.

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25 **Figure 5.** Graphical representation of priors (curves in red) and posteriors (histograms in black) distributions of enrichment  
 26 factors for each investigated tracer (d15N:  $\delta^{15}\text{N}$ , d13C:  $\delta^{13}\text{C}$ , and contaminants concentrations: L-PFOS, FOSA, PFOA, PFNA  
 27 and PFUnDA).

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2 **Table S 1.** List of PFAS compounds targeted in the present study

Acronym	Compound name	Molecular formula
L-PFOS	n-perfluoro-1-octanesulfonic acid	$C_8F_{17}SO_3H$
FOSA	perfluorooctane sulfonamide	$C_8F_{17}SO_2NH_2$
PFOA	perfluoro-n-octanoic acid	$C_7F_{15}COOH$
PFNA	perfluoro-n-nonanoic acid	$C_8F_{17}COOH$
PFUnDA	perfluoro-n-undecanoic acid	$C_{10}F_{21}COOH$

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## 6 S2. Comparisons between ESCROC and MixSIAR

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8 As an illustrative example, we calibrated MixSIAR to estimate the sole diet, using the same data that  
9 were used to fit ESCROC.

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11 To be consistent with our ESCROC approach (see text for details),

12 - We used exactly the same isotope data

13 - We used exactly the same a prior for diet composition than in ESCROC

14 - We used two alternative priors for N and C TEFs ( $\Delta_N$  and  $\Delta_C$  in Eq 7 and 8 in the text)

15 o First, we used exactly the same priors as in ESCROC (see Eq. 7 and 8 in the text):

16  $\Delta_N \sim \text{Normal}(3,1)$  and  $\Delta_C \sim \text{Normal}(0,1)$

17 o Second, we used priors corresponding to the TEF posterior distributions from ESCROC

18 (see Figure 5 in the text)

19 Then, we also fitted ESCROC, but, contrary to the article, using only C and N (i.e., ignoring  
20 contaminants concentrations) data to directly compare with MixSIAR outputs. Note that with such a  
21 limited dataset, ESCROC is likely to be underdetermined.

22

23 The results presented in the figure S2 below are based on the direct plotting of the posterior  
24 distributions for both ESCROC and MixSIAR. They first highlight that:

25 (1) Adding contaminants in ESCROC allows to better discriminate the proportions of the main  
26 prey in the sole diet (Figures S2 A and C).

27 (2) Using the posterior distribution of TEF from ESCROC, MixSIAR provides very similar results to  
28 ESCROC (Figures S2 A and D).

29 (3) Using naive priors for TEF and using posterior distributions from ESCROC in MixSIAR provide  
30 quite different diet estimates (Figures S2 B and D). This is due to the fact that the naive prior is  
31 rather different from the posterior distribution (especially for C). This underlines that MixSIAR  
32 is especially sensitive to TEF prior specifications.

33

34 Outputs from ESCROC and MixSIAR (Figures S2 C and D) are different but not contradictory. Actually,  
35 they mainly differ in the proportion of white shrimps in the sole's diet. However, the posterior density  
36 is very flat and rather uninformative for this particular species in the MixSIAR outputs and does not  
37 allow to really conclude on the proportion of this shrimp in the sole's diet. Indeed, most of the  
38 ESCROC simulated results could correspond to the MixSIAR posterior distribution.

39

40 Although based on the same transfer equations, these differences between ESCROC and MixSIAR  
41 could be explained by:

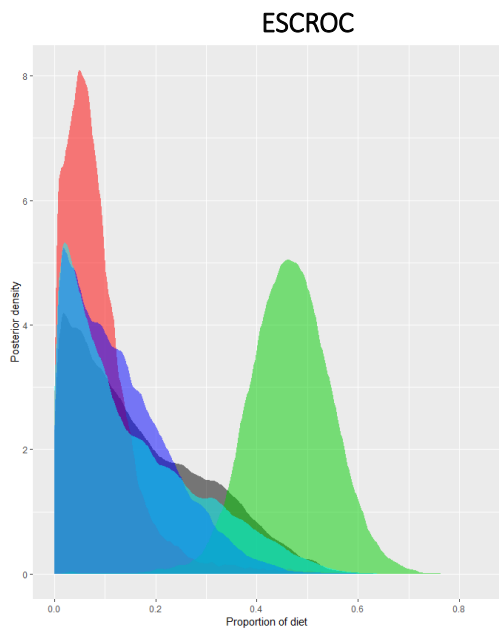
42 (1) the fact that ESCROC estimates are computed for the whole food web at one time. This allows  
43 using mutual information to compute more accurate estimates.

44 (2) the use of a random effect 'Species' Thus, although the TEF and TMF estimation are still  
45 considered globally constant through the food web, they are modulated species by species. By  
46 doing this, we did not specifically consider that isotope discrimination varies with Trophic  
47 Position but we allowed the estimation to vary for every species.

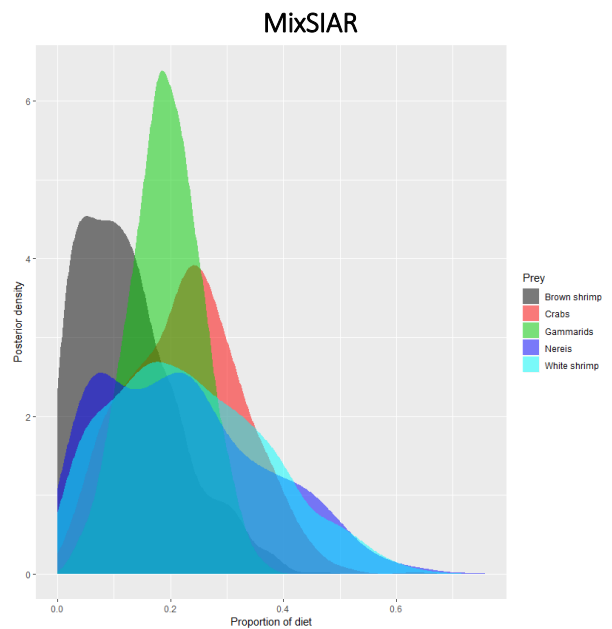
48 (3) a different way to account for uncertainty in both models. In particular, posterior sources and  
49 consumers' isotope compositions are computed in ESCROC using both observation data and  
50 parameters estimates. This leads to an a posteriori quite different set of data between both  
51 approaches.

52

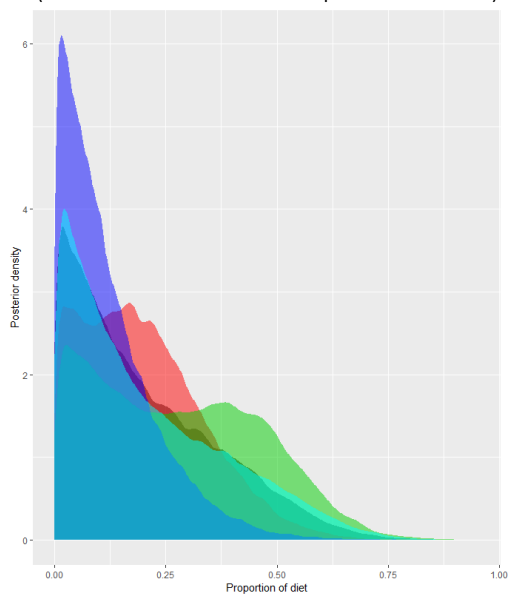
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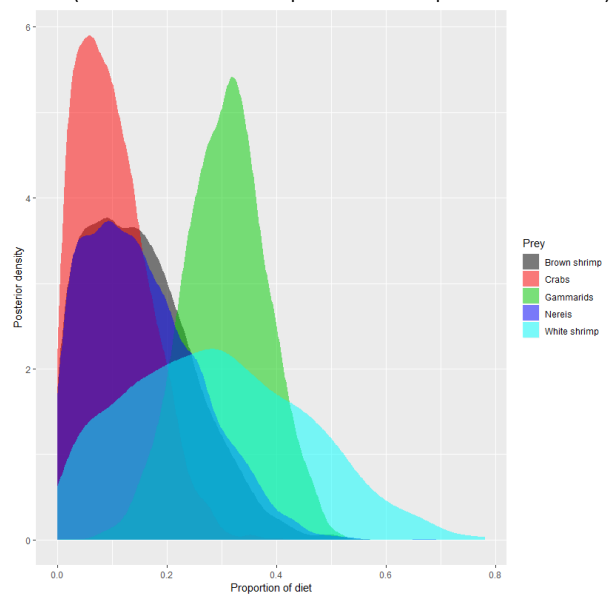
A. (ESCROC with both isotopes and PFASs)



B. (MixSIAR with TEF priors = TEF priors ECSROC)



C. (ESCROC using only C and N isotopes data)



D. MixSIAR (TEF priors = posteriors ESCROC)

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**Figure S2.** Probability density (in y-axis) of the proportion (in x-axis) of each of the listed preys in the diet composition of the sole in the Gironde estuary. Comparisons of outputs from ESCROC (A. based on both isotopes and contaminants data; C. based only on isotopes data) and MixSIAR (B. using initial TEF priors implemented in ESCROC; C. using estimated posterior TEF estimations from ESCROC).