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Fish Discards Management: Pollution Levels and Best Available Removal Techniques

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40 **Abstract**

41 Fish discards and by-catch issues are highly topical subjects nowadays permanently
42 under a social focus. To manage this issue, two main approaches are being considered to
43 address this discard problem: reducing by-catch and increasing by-catch utilization. As
44 these two harvesting strategies may be complementary, an appropriate balance between
45 by-catch reduction and utilization is desirable for any fishery.

46 Increased by-catch valorization interest may come from a greater demand for fish
47 products: the development of new markets for previously discarded species; the use of
48 low-value by-catch specimens for aquaculture and animal feed or the creation of value-
49 added fish products from by-catch or discarded fish for food, pharmaceutical or
50 cosmetic industries. In this valorization framework, and always targeting the aim of
51 promoting the responsible and sustainable management of marine resources, pollutant
52 levels in catches of European fisheries (including target and main discarded species), as
53 well as the best available decontamination techniques of marine valorized discards/by-
54 products are compiled and analyzed in this work. This is due to the fact that a wide
55 different distribution of pollutant concentration between tissues in fish can be found,
56 especially in detoxifying organs, like kidney and liver and in other fractions of high
57 lipid content, like skin. Therefore, contaminants present in fish discards may be
58 transferred to the valorized products obtained from them, leading to possible long-term
59 bioaccumulation and subsequent adverse health effects.

60 The objective of the present work is to provide a general view of the present
61 discards/by-catch valorization-based management options.

62

63 **Keywords** Fish discards; sustainability; pollutants; removal techniques; valorization

64 **1. Introduction**

65 Most of Persistent Organic Pollutants (POP) and heavy metals emitted to air or
66 water as products or by-products of industrial activities, or applied directly on land (i.e.
67 pesticides) can travel long distances from its primary source, and can finally end up in
68 the marine environment (1,2). Most common examples of these substances are
69 polychlorinated dibenzo-p-dioxins (PCDD) and dibenzofurans (PCDF), polychlorinated
70 biphenyls (PCB), polycyclic aromatic hydrocarbons (PAH), organochlorine pesticides
71 (OCP) (like hexachlorobenzene - HCB, hexachlorocyclohexanes - HCHs,
72 dichlorodiphenyltrichloroethane - DDTs, and metabolites like
73 dichlorodiphenyldichloroethylene - DDE), polybrominated diphenyl ethers (PBDE),
74 hexabromocyclododecanes (HBCDs) and metals like As, Cu, Cd, Zn, Pb and Hg. Due
75 to their persistence and toxicity, they can accumulate in biota and biomagnify through
76 trophic webs, being biomagnification especially important for aquatic organisms (3,4).

77 Many studies in the scientific literature, like surveys of fish and fish products in
78 markets of different countries (5-8), monitoring reports of Public Administrations and
79 the EU (9), as well as web tools like the EcosystemData of ICES
80 (<http://ecosystemdata.ices.dk/>), reported significant levels of this kind of pollutants
81 (especially of dioxins, PCBs and heavy metals) in several cases for commercial species
82 of different fisheries. Many of these studies have been developed in heavily polluted
83 areas like the Baltic and North Seas (10-12). Hence, it is logical to assume the presence
84 of contaminants in other non-commercial species, although contamination levels in
85 these non-targeted and/or discarded species are not usually assessed. However, a
86 sustainable management of discards passes through the evaluation of their pollutant
87 content, since the most common uses of discards are oriented to both the production of
88 fish oil and meal (for aquaculture/animal feeds) or as additives in human direct
89 consumption products (food supplement, margarines, gelatine, etc.). In fact, pollutant

90 amounts found in some marine valorized by-products are of concern (13). On the other
91 hand, some studies revealed that concentration of POPs is significantly higher in farmed
92 fish (mainly salmon) than in wild fish (14-16). This is due to the presence of pollutants
93 in feed, which comprise fish oil and meal (17). Concerns on this issue have led EU to
94 set maximum levels for dioxins and dioxin-like (DL-) PCBs for aquaculture feeds (18),
95 fish and fish products (19)

96 For fulfilling these regulations, different options are available to the fish farming
97 industry. One possibility is to use fish oils or meals presenting low levels of these
98 pollutants for fish feed, for example, from the southern hemisphere (20). However, this
99 is not the optimal approach, since fish oil availability is already limited (21). Another
100 solution is to employ vegetable oils in the feed, but these oils do not contain the fatty
101 acids that represent the positive nutritional properties of marine food, and thus, fish
102 breeding with vegetable oils results in specimens with worse performance, health and
103 quality. In fact, a mixture of oils is usually employed as compromise solution
104 (20,22,23). Finally, pollutants could be removed from the oils and meals used in fish
105 feeds, while retaining the nutritive components of the oil (21). Therefore, research and
106 development of technologies for the removal of these contaminants has gained
107 considerable importance (17), since market demand for decontaminated fish feeds in
108 aquaculture has increased during last years (20). Most studies on pollutant removal
109 techniques are focused on the reduction/elimination of POPs (dioxins and dioxin-like
110 polychlorinated biphenyls) in fish oils, especially those produced for salmon breeding.
111 Less attention has been paid to fishmeal, and none to other valorized fish products, like
112 gelatin or hydrolizates. Taking into account the lipophilic character of pollutants, their
113 levels on this type of proteic products should not be of concern.

114 In the aim of promoting the responsible and sustainable management of the
115 European fishing activity, actions were directed to the development of policies to reduce

116 unwanted by-catches and eliminate discards in European fisheries, as well as to make
117 the best possible use of the captured resources avoiding its waste. In this sustainability
118 framework, FAROS project, co-funded under the LIFE+ Environmental Program of the
119 European Union (LIFE08 ENV/E/000119 – www.farosproject.eu), aims as one of its
120 main objectives to analyze the valorization potential of fish discards in order to
121 contribute to their sustainable management by minimizing discards/by-catch through
122 their optimal valorization to recover and to produce valuable chemicals of interest in the
123 food and pharmaceutical industry (24). In order to properly define these adding-value
124 processes, the key issues of pollutant levels in catches of European fisheries (including
125 target and main discarded species), as well as the best available decontamination
126 techniques of marine valorized by-products were compiled and analyzed from several
127 studies, with the objective of providing a general picture of the present management
128 options.

129

130 **2. Pollutant content in species of European fisheries**

131 2.1. Existing pollutant profiles

132 To date, Ecosystemdata web tool of ICES (<http://ecosystemdata.ices.dk/>) can be
133 considered one of the most complete infrastructure of marine data compilation
134 corresponding to the European fishing area. A search within this database was
135 developed for all the discarded species (159) identified in the fisheries considered in
136 FAROS project (25):

137 a) Galician bottom otter trawl fleet vessels authorized to fish in Community waters
138 targeting flat fish, and basically operating in Great Sole Bank.

139 b) Galician coastal bottom otter trawl fleet vessels targeting a variety of demersal
140 species.

141 c) Portuguese coastal bottom trawl vessels for demersal fish that operate along the
142 year, with hauls directed to a variety of species.

143 The objective was to check which “FAROS species” (main discarded species in
144 these fisheries) were found in the database, and if present, which ones were monitored
145 on pollutant profiles. The qualitative results of this query are shown in Table S1 of the
146 Supplementary Material. It can be seen that pollutant analyses are only available for 25
147 of the 159 species reviewed. Among these 25 species, only 7 correspond to the 29 main
148 species discarded in the Spanish and Portuguese *métiers*, marked on grey in Table S1
149 (25). These species are: *Chimaera monstrosa* (rabbit fish), *Lepidorhombus whiffiagonis*
150 (megrim), *Melanogrammus aeglefinus* (haddock), *Merluccius merluccius* (hake),
151 *Micromesistius poutassou* (blue whiting), *Scomber scombrus* (Atlantic mackerel) and
152 *Scyliorhynchus canicula* (small-spotted catshark). Moreover, *Micromesistius poutassou*
153 and *Scyliorhynchus canicula* were only monitored for heavy metals and Hg alone,
154 respectively. The remaining 18 pollutant-monitored species included in Table S1 are
155 considered as discards in these *métiers* only in few occasions, i.e., at a very low discard
156 rate. In fact, most of them have an important commercial value, hence the reason of
157 their exhaustive monitoring on pollutants. Examples of these species are *Gadus morhua*
158 (cod), *Merluccius merluccius* (hake), *Coryphaenoides rupestris* (roundnose grenadier),
159 *Hoplostethus atlanticus* (orange roughy) or *Lophius piscatorus* (monkfish). Besides,
160 flatfish or species that live on or within the sediment layer (bottom dwelling fish) and
161 deep sea fish are among the monitored species, not only because of its commercial
162 value, but also because they are known to highly bioaccumulate pollutants. Examples
163 are *Brosme Brosme* (tusk), *Coryphaenoides rupestris* (roundnose grenadier),
164 *Glyptocephalus cynoglossus* (witch flounder), *Hippoglossoides platessoides* (American
165 plaice), *Microstomus kitt* (lemon sole) or *Pleuronectes platessa* (European plaice). This
166 type of species lives on the sediment layer at the sea bottom (benthic organisms), and

167 directly uptake contaminants from sediment particles apart from diet (26,27). Pollutants
168 tend to be associated with organic matter due to their lipophilic character (3), and at the
169 same time, organic matter is bound to suspended particles in the water column that end
170 up in the sediment layer (marine snow) (28). Moreover, deep water fish have a
171 significant potential for the accumulation of POPs because many of the deep water
172 species feed at higher trophic levels and live longer than pelagic fish (29).

173 2.2. Pollutant levels in discarded FAROS species

174 A review of studies presenting the quantitative pollutant contents in fish was
175 developed. Values of either heavy metals and/or POPs (PCDD/Fs, PCBs and OCPs) in
176 European commercial species are reported in Tables 1 and 2. These commercial species
177 are considered as discards in Great Sole Bank and Atlantic Spanish and Portuguese
178 coast métiers (although most of them usually at a very low rate) for different reasons.
179 The main ones are: i) legal reasons related to the quota system; ii) strategic or
180 commercial reasons; iii) lack of quality in the case of damaged specimens or in poor
181 condition; etc (25).

182 Data were collected from market surveys in different countries and from other
183 relevant studies available on commercial fish species (flesh or viscera). Origin was also
184 included when possible. Although many studies on pollutant monitoring are available
185 for the area of the Baltic Sea, they were not included in Tables 1 and 2 since the species
186 monitored (herring, cod, sprat, etc.) are different from those considered in the selected
187 fishing area (Great Sole Bank and coastal waters of the Atlantic side of the Iberian
188 Peninsula). Pollutant concentration values for a total of 43 species were recorded, 14 of
189 them corresponding to the most discarded species in the métiers considered in this
190 analysis (marked on grey in Tables 1 and 2).

191 In Table 1, metal concentrations present in 30 fish species identified as target and
192 main discarded on the Atlantic fisheries considered in FAROS framework are

193 summarized. In general, a wide range of concentration values were found for all the
194 metals considered. Zn, As and Cu reached the highest concentrations in the liver of
195 *Aphanophus carbo*. This deep sea fish presented in most of occasions the highest
196 concentrations for all metals.

197 Heavy metal bioaccumulation is related to biotic and abiotic factors such as water
198 temperature, fish biological habitat, chemical form of metal in the water, fish species,
199 gender and length or age (50). In general, it was observed that concentrations of metals
200 are significantly higher in liver tissues than in muscle for the monitored species. This is
201 particularly of concern when thinking in produce fish oil from livers. Concentration
202 values ranges can vary widely among species and even for the same species in the same
203 study, which implies a clear influence of location (as expected). Among the most-
204 monitored species, specimens of *Aphanophus carbo* (black scabbard), *Coryphaenoides*
205 *rupestris* (roundnose grenadier) and in less proportion *Merluccius merluccius* (hake),
206 *Sardina pilchardus* (sardine) and *Scomber scombrus* (Atlantic mackerel) presented
207 levels of pollution that can be of concern when thinking in further valorization
208 technologies, since concentration steps are always present in these processes.

209 Concentrations of PCDD/Fs, PCBs, HCB, DDTs, chlordanes, PBDEs, DDE and
210 HBCD are shown in Table 2. PCBs and PCDD/Fs were the most frequently analyzed
211 pollutant, due to their higher toxic effects on human health.

212 In many of the studies shown in Tables 1 and 2, the exposure to a variety of
213 pollutants by fish ingestion was assessed (6,7,41,42). The conclusions were similar in
214 most of them: moderate fish consumption not only does not pose a risk to human health
215 but also has numerous nutritional benefits. However, production of fish oil and meal
216 involves concentration processes that could increase pollutant concentration in valorized
217 products, becoming an important problem.

218

219 **3. Pollutant removal techniques**

220 As previously mentioned, the most common uses of discards are the production of
221 fish oil and meal. As shown in Table 3, pollutant concentration values available in the
222 literature for fish oil and meal produced from species of different origin and location
223 reveals that, in some cases, these levels can be of concern when compared to the limits
224 established on the European Commission Directive 2006/13/EC on undesirable
225 substances in animal feed as regards dioxins and dioxin-like PCBs (18). Therefore,
226 purification step/s would be needed before consumption. In the next section, a revision
227 of available decontamination techniques for fish valorized products (fish oil and meal)
228 and other marine solid by-products is presented.

229 3.1. Fish oil

230 A key factor during fish oil refining is to remove contaminants without altering the
231 levels of present nutritionally valuable compounds and the oxidative status of the oil
232 (54,55). A reduction of some type of pollutants associated to fish oil during refining has
233 been assessed. This is due to the fact that crude fish oils are usually refined to reduce the
234 content of free fatty acids, metal traces, pigments, etc. In particular, the deodorization
235 step (steam distillation at high temperature and vacuum) causes a decrease not only in
236 residual pigments and other volatile compounds, but also the almost total removal of
237 most volatile pollutants like organochlorine pesticides (α -HCH, lindane, etc) and the
238 reduction to a half of the initial concentration of PCBs and less volatile organochlorine
239 pesticides (56). However, standard conditions (180 °C and 2 hours of contact) of
240 deodorization are found to be inefficient for the removal of dioxins and furans.

241 Most up-to-date efficient removal methodologies for fish oil involve the use of a
242 solid apolar adsorbent (like activated carbon), distillation processes, extraction
243 processes or a combination of these techniques.

244 3.1.1. Extraction with solid adsorbents

245 Generally, purification of fish oil by solid adsorption is performed by mixing the oil
246 with the adsorbent in a rotavapor under different experimental conditions, usually at
247 mild pressure and temperature. The oil is subsequently separated from the adsorbent by
248 filtration over a paper filter.

249 Eppe et al. (57) investigated oil purification by solid adsorbents for decreasing
250 PCDD/Fs and dioxin-like (DL-) PCBs levels in order to be in compliance with
251 European Legislation. The adsorbents tested were bleaching earths (polar adsorbents),
252 acid activated silica powder, Diatomaceous earth, and several types of activated carbon
253 (apolar adsorbents). Among the tested adsorbents, only activated carbon could
254 significantly remove the PCDD/Fs (up to 99%) and DL-PCBs (up to 50%)
255 concentration in cod liver oil. Optimum pressure and temperature of 0.05 bar and 74 °C
256 were established, respectively. Besides, it was concluded that reaction time within the
257 range of 10 to 50 minutes had virtually no influence on the adsorption of pollutants,
258 while the dose of activated carbon was the most influential variable, since the higher the
259 dose, the higher the adsorption of contaminants.

260 A similar study was developed by Maes et al. (55), who evaluated the efficiency of
261 different grades and doses (0.1 to 0.5% ww) of activated carbon to remove PCDD/Fs
262 and DL-PCBs from cod liver oil. The process was performed at reduced pressure (0.05
263 bar) and moderate temperature (70 °C) with a 30-minute reaction time. An almost
264 complete elimination of PCDD/Fs and 80% removal of DL-PCBs were achieved with
265 0.5% of high-grade activated carbon. The lower PCBs removal was due to the minor
266 adsorption of the mono-ortho fraction (30% or less), since their noncoplanar
267 geometrical structure present low affinity to activated carbon.

268 Optimization of activated carbon adsorption for the reduction of POPs in
269 commercial fish oil was performed by response surface methodology (58). PCDD/Fs
270 were eliminated in a 99%, while non-ortho PCBs and mono-ortho PCBs were reduced

271 to a maximum of 87 and 21%, respectively, operating at 80 °C and 15 minutes of
272 contact time. However, PBDEs reduction was not observed with this treatment. The
273 authors stated that an increase in the adsorption temperature will probably enhance DL-
274 PCBs reduction. However, they advise that it is necessary to have a compromise
275 between optimal processing conditions, capacity utilisation, target Toxic Equivalent
276 Quantity (TEQ) and oil quality specifications in large-scale industrial operations.

277 Usydus et al. (59) employed activated carbon at industrial scale to purify fish oil
278 from different origins (sprat, herring and salmon). Optimum purification parameters
279 were obtained in previous studies at laboratory scale (60), and were: dose of 1.2%
280 weight, temperature of 85 °C and 90 minutes of contact time. Although the oil loss
281 produced during purification was approximately 20%, the removal of PCDD/Fs and dl-
282 PCBs was 77.0-99.6% and 42.7-50.0%, respectively, depending on the raw material.
283 These authors also evaluated the reduction of OCPs, PBDEs and Cd, Pb and Hg, which
284 was negligible.

285 11 silicon-based and 9 carbon-based adsorbents were tested for the elimination of
286 PCDD/Fs, PCBs, PBDEs, DDTs and HCBs from refined salmon oil by Ortiz et al. (61).
287 Silicon-based adsorption was developed at 1.5% w/w, 50 °C of temperature and
288 atmospheric pressure, according to previous studies (54). However, silicon-based
289 adsorbents and graphitized carbon were not suitable for the removal of POPs. On the
290 contrary, activated carbon adsorbents showed a very high removal capacity for these
291 pollutants. DL-PCBs were also better removed by activated than by graphitized carbon,
292 although efficiency was lower than in the case of PCDD/Fs (18-24%). For the
293 remaining POP evaluated, low effectiveness of both types of carbon-based adsorbent
294 was observed, due to their non-planar structure. However, optimization of the
295 adsorption process with coconut-shell activated carbon by response surface
296 methodology resulted in an increase of the POP removal efficiencies. Operating at a

297 dose of 2.5% w/w during 37.5 minutes with a temperature of 80 °C and 1 bar resulted in
298 eliminations of 99% PCDD/Fs, 70% HCBs, 36% dioxin-like PCBs, 27% DDTs, 11%
299 marker PCBs and 9% PBDEs.

300 In general, activated carbon adsorption is an appropriate method for removing
301 dioxins and furans, but low elimination efficiencies are obtained for DL-PCBs. Efficient
302 activated carbon adsorption depends on a planar molecular conformation, and this will
303 strongly limit the number of possible POP to remove based on this technology (58).
304 Therefore, complete decontamination of fish oil could only be achieved by a
305 combination of activated carbon with other extraction (stripping) process (55,62).

306 3.1.2. Supercritical CO₂ extraction

307 Supercritical CO₂ extraction (SCE) has been applied to several different processes,
308 like selective extraction of valuable natural products, separation of contaminants and
309 other processes because of its extraction selectivity, low critical point and lack of
310 flammability, toxicity and corrosiveness (17). SCE for removal of pollutants from fish
311 oil has been performed in semi-batch and counter current installations. One of the first
312 attempts of investigating the feasibility of supercritical counter current fluid extraction
313 for pollutant removal in fish oil was the study conducted by Krukonis (63). The author
314 found that PCBs can be extracted from cod oil with supercritical carbon dioxide at quite
315 modest temperature and pressure, with little yield loss of the fish oil. During the
316 experiment, temperature was held constant at 70 °C and pressure was gradually
317 increased from 172.4 to 448.2 bar. However, it was stated that subsequent increases
318 from 241.3 bar did not increase the extractability of the pollutants. Although the results
319 obtained in this lab-scale study were promising, it was advised to subject the process to
320 a detailed economic viability evaluation, as well as to analyze factors such as stability to
321 autoxidation during storage and product performance.

322 Some years later, Jakobsson et al. (64) investigated the elimination of dioxins and
323 dibenzofurans from cod liver oil. The main objective of their study was to test the
324 counter current method and to establish the influence of the feed oil/CO₂ ratio on the
325 extractability of these compounds from the fish oil. The results obtained showed that the
326 higher the carbon dioxide/oil ratio, the smaller the recovery of oil. The most effective
327 extraction (80% of dioxins together with 17% of the oil) was achieved at ratios of 100.
328 However, improvements in this technique have led to a consequent increase in pollutant
329 removal efficiencies and to a decrease in oil loss.

330 Kawashima et al. (62) investigated the removal of PCDD/Fs and coplanar PCBs
331 from menhaden oil by SCE and by activated carbon adsorption. Experimental
332 conditions of SCE were 60 °C of temperature, 280 bar of pressure and a CO₂ flow
333 volume of 50 L/g oil. This method proved effective to remove PCBs, with elimination
334 percentages ranging from 70% to 90%. However, removal efficiency decreases as the
335 molecular weight of PCDD and PCDF congener increases, being in the range of 15-
336 90% depending on the molecule. For the effective removal of high-chlorinated
337 PCDD/Fs, the authors considered an adsorption process with activated carbon. Removal
338 ratios of this process were higher than 90% for all of the isomers of PCDD/Fs, while
339 removal percentages for PCBs were within 1% (mono-ortho) and 30% (non-ortho).
340 Consequently, a combined removal process (SCE with CO₂ followed by activated
341 carbon adsorption) was more effective, since almost 100% of the total TEQ value was
342 reduced.

343 The same authors advanced in this field, assessing the use of continuous counter
344 current supercritical CO₂ extraction and activated carbon adsorption for removing
345 pollutants from fish oil (17). In their previous work (62), SCE was found to have high
346 efficiency in DL-PCBs removal. However, semi-batch processes require long operation
347 times, producing a purified oil yield not enough for practical use. Thus, extraction

348 conditions and contaminant removal efficiency of counter current SCE was
349 investigated. As in Kawashima et al. (62), removal efficiencies decrease with an
350 increase in the molecular weights of pollutants. Process efficiency also increased with
351 extraction pressure, being the optimal operating conditions 300 bar of pressure, 70 °C
352 and a CO₂/oil ratio of 72. These conditions proved effective for the remove of DL-PCBs
353 (93%) and PCDD/Fs congeners that have molecular weights less than 400. Fish oil
354 refined by counter current SCE was subsequently treated with activated carbon for the
355 elimination of PCDD/Fs, reaching values higher than 80% for each congener. Thus, the
356 combined process reduces the pollutant concentration by 94%, while presenting a
357 minimal influence on the fatty acid content of the oil. When compared with the semi-
358 batch type process, counter current SCE uses 40% less CO₂ and yields 30% more
359 refined oil. However, it is necessary to consider that fish oils extracted with CO₂ can
360 lose much of its unsaturation during storage and can polymerize (63).

361 3.1.3. Short-path distillation

362 Short-path distillation (SPD) technology is characterized by operation with short
363 residence times and high vacuum level. Breivik & Thorstad (21) presented an improved
364 method based on this technology to eliminate POPs from marine oils. They found that
365 with the addition of 3-6% of an ester mixture (working fluid) prior to the distillation,
366 pollutants are removed in a much more efficient manner. In this case, the working fluid
367 was a light ethyl ester fraction of transesterified fish oil produced as a by-product from
368 commercial production of omega-3 concentrates. With this technique, concentration of
369 dioxins and PCBs were reduced by more than 90%, including DL-PCBs. Moreover,
370 DDT, toxaphene and PBRDs were removed to a level below the analytical detection
371 limit.

372 Decontamination of sprat oil by SPD technology was evaluated by Oterhals et al.
373 (65). The objective was to quantify the effect of evaporator temperature, feed rate and

374 addition of a working fluid (21) on the reduction of PCDD/Fs, DL-PCBs and PBDEs.
375 Furthermore, a model of SPD based on process parameters and quantitative structure
376 properties relationship was proposed by the authors to relate removal efficiency with
377 congener volatility. The results obtained in this study indicated that is not possible to
378 define optimum operation conditions for POP reduction in fish oil by SPD due to the
379 large variance in vapor pressures for the multicomponent mixture of organic
380 compounds. However, as TEQ reduction is mainly influenced by the removal of
381 PCDD/Fs and DL-PCBs, the best decontamination effect was obtained with a
382 combination of low feed rate and high evaporator temperature and working fluid
383 conditions.

384 The feedstock used in the study by Oterhals et al. (65) was the same as earlier
385 reported on activated carbon-based decontamination of fish oil (58). Therefore, these
386 authors were able to compare both removal methods. Activated carbon is an appropriate
387 method for removing compounds like PCDD/Fs, PAHs and some congeners of PCBs,
388 which present a coplanar structure, since effective adsorption is dependent on dispersive
389 electronic interactions affected by sorbate planarity and steric effects (66). However,
390 compounds like most of organochlorine pesticides and PBDEs are not adsorbed and
391 removed by this method. On the contrary, the efficiency of a SPD based
392 decontamination process is mainly dependent on the volatility of the respective
393 compounds and the selection of favorable process conditions. SPD is less influenced by
394 the conformation and chemical nature of POPs to be removed when compared to
395 activated carbon adsorption (65).

396 However, the use of SPD to eliminate POPs from fish oils will also remove other
397 volatile compounds and decrease the nutritional value and oxidative stability of the oil.
398 This fact is due to the high temperature levels (>200 °C) applied to the fish oil during
399 the process. Oterhals and Berntssen (67) quantified the effects of alternative SPD

400 process conditions on the oil nutritional and oxidative properties and identified the
401 optimal process conditions by combining decontamination effects in compliance with
402 legislation levels and maximum retention of nutritional quality. Some reduction in the
403 oxidation level was observed, but with preservation of PUFA level and quality. Only
404 76% of reduction of the TEQ level was achieved in the fish oil to be in accordance with
405 international quality standards, with a final loss of vitamins lower than 20%. If a higher
406 decontamination level is required (90%), vitamin retentions can vary between 60-90%.

407 3.1.4. Other procedures

408 Other volatilization procedures (steam deodorization) were tested to eliminate POPs
409 from fish oils. Carbonelle et al. (68) used a combination of an activated carbon
410 adsorption treatment with either packed column stripping or cross-flow stripping. The
411 last two methods were introduced with the aim of improving the removal of mono-ortho
412 PCBs, since activated carbon is not adequate for this task. Packed column and cross-
413 flow stripping are procedures that involve high temperature, low pressure and injection
414 of a stripping agent (steam). Differences between them are residence time (very short in
415 packed column), pressure (pressure drop in packed column and constant in cross-flow)
416 and contact between oil and steam (counter-current in packed column). The authors
417 decided to combine cross-flow stripping (instead of packed column) with activated
418 carbon because this technique is preferable in an industrial process. With an optimum
419 combination of these methods, removal of 100% of dioxins and furans, more than 95%
420 of non-ortho PCBs and between 48 and 74% of mono-ortho PCBs was achieved.
421 However, the final nutritional quality (PUFA content, etc) of the product was not
422 evaluated.

423 3.2. Fishmeal and other marine solid wastes

424 Although several alternatives have been tested for the elimination of pollutants from
425 fish oil, less emphasis has been given to the development of purification alternatives for

426 fishmeal and marine solid by-products/wastes without decreasing its nutritional/reusable
427 value. Regarding fishmeal, decontamination techniques like ultraviolet (UV) light,
428 extraction with either organic solvents or oil, and enzymatic treatments have been
429 evaluated.

430 Baron et al. (69) applied UV on contaminated fishmeal to photodegrade dioxins.
431 After 5 days of exposure to UVB light, degradation of 70% of PCDD/Fs content was
432 obtained. However, the photodegradation mechanism triggered lipid oxidation and
433 increase the content of non- and non-ortho PCBs as reaction products in the treated
434 fishmeal. This fact, linked to the high exposure time required, make the application of
435 this methodology at industrial scale unfeasible. Although an increase in the light
436 intensity should decrease exposure time, oxidation of long-chain fatty acids would also
437 be enhanced. The authors proposed the addition of antioxidants to avoid this undesirable
438 process.

439 As previously mentioned, persistent organic pollutants are lipophilic compounds,
440 and the reduction of the fat content in fishmeal will result in the concurrent reduction of
441 these undesirable compounds. Baron et al. (70) studied several techniques to remove
442 dioxin and DL-PCBs from fishmeal. The studied methods were fat extraction with
443 organic solvents, oil, protease and direct breakdown of dioxin and PCBs using
444 oxidoreductase. Low reduction of pollutant content was observed with fat separation
445 after protease treatment (30%) and with PCDD/Fs and DL-PCBs degradation by
446 oxidoreductase (10-15%). The organic solvents (ethanol, isopropanol and isohexane)
447 reduce the fat content to 80%, with a proportional reduction of pollutants, but the
448 fishmeal had a low nutritional quality and might content traces of solvents. Extraction
449 of dioxin and PCBs using olive oil or fish oil resulted in 60%-75% of decontamination
450 effect. Enrichment of the oily phase in pollutants was observed for all congeners and
451 both oil types. Increasing the time of extraction (24 hours) only resulted in a minor

452 increase in the levels of POPs in oils, indicating that equilibrium and partitioning of the
453 contaminant into the oil is a fast process (70). Oterhals and Nygard (71) also
454 investigated the reduction of persistent organic pollutants in fishmeal at pilot scale by
455 organic solvent and soybean oil extraction. Both extraction agents provided fishmeal
456 products with a TEQ value below the maximum permitted levels. However, lowest
457 levels were observed with soybean oil (reduction of 97% of TEQ). Moreover, the
458 estimated fat content of the extracted product presented a value close to the respective
459 one in ordinary processed fishmeal. This was considered as a critical success factor if
460 the process should be used in industrial application. The obtained decontamination rates
461 in this work were higher than those reported by Baron et al. (70) on the basis of olive oil
462 extraction. According to Oterhals and Nygard (71), the difference might be explained by
463 the combined use of a higher extraction temperature (88 °C vs room temperature) and
464 oil/matrix ratio (1:3 vs 1:1). From the industrial point of view, the oil extraction process
465 has several advantages when compared to organic solvent extraction, like for example,
466 easy integration in an existing fishmeal processing line and the use of a safe and non-
467 flammable extraction medium.

468 Other type of pollutants found in marine solid by-products are metals. Removal of
469 these compounds from the solid matrix is critical for its subsequent reuse and
470 valorization as fodder. Tavakoli and Yoshida (72) investigated the use of sub- and
471 supercritical water treatment as a method for recovering heavy metals from squid
472 wastes. Reaction temperature ranged between 443 and 653 K and the pressure range
473 between 7.92-300 bar, and produced four phases (unreacted solid, aqueous, fat and oil).
474 Distribution coefficients of the metals considered (Cd, Cu and Zn) followed the
475 decreasing order fat>solid>oil>aqueous phase. The proposed decontamination waste
476 process is energy efficient according to the authors, and produces valuable products (oil,
477 soluble proteins, organic acids and aminoacids) from waste. In addition, metal ions can

478 be recovered from waste streams and recycled back to the related industries. Other
479 techniques for the removal of metals from marine by-products involve a coagulation
480 process. Ghimire et al. (73) proposed an environmental friendly removal process of
481 heavy metals from Cd-contaminated scallop waste by using apple waste and astringent
482 persimmon extract (kaki-shibu), which has the advantage of a coagulating effect
483 independent of pH. The process consists of three steps: 1) leaching of all metals
484 contained in scallop waste by dilute sulphuric acid; 2) removal by kaki-shibu
485 coagulation of turbid organic materials from the leach liquor and; 3) adsorption of
486 heavy metals onto a gel prepared with apple waste. To adjust pH (lower in the leaching
487 process and higher during the adsorption), the authors proposed a counter current
488 process, recycling the leach liquor to the feed waste. The obtained cadmium free scallop
489 waste can be used as cattle and fish fodder, while the cadmium free apple waste can be
490 reused as fertilizer.

491 Discards are one of the most important topics in fisheries, both from an economic
492 and environmental point of view. The contribution to a sustainable management of this
493 biomass through their optimal valorization highly depends on the quality of the products
494 to be obtained from them. The products of discard valorization are mainly concentrates,
495 being the most common ones fish oil and meal. Pollutants contained in the raw material
496 are usually present at higher concentrations in the valorized product, especially if the
497 product has a high fat content (oil). Therefore, reduction and/or elimination of
498 undesirable and toxic compounds from fish oil and fishmeal is a key factor for its safe
499 reuse, either as feed in aquaculture or as additive in food products.

500 Three techniques are currently available to reduce POPs from fish oil: 1) solid
501 adsorbers (activated carbon), 2) SCE and, 3) SPD. Although SPD remove a wider
502 variety of pollutants (PCDD/Fs, DL-PCBs, PBDEs, OCPs) than SCE or activated
503 carbon adsorption, this technique is performed under experimental conditions

504 (especially of temperature) that can affect the positive properties of the oil if a high
505 degree of decontamination is needed. SCE has a minimal environmental impact and
506 preserve the PUFA content of the treated oil, although some authors found unstable
507 behavior of the oil during storage. However, it is not efficient for the removal of high-
508 chlorinated PCDD/Fs, and for that reason, can be combined with activated carbon
509 adsorption, that effectively removes dioxins and furans. The selection of the most
510 appropriate technique for oil decontamination mainly depends on the pollutant type and
511 congener found in the oil, and the percentage of TEQ value reduction needed to comply
512 with legislation, which is different according to the final use (feed or food). Regarding
513 fishmeal, the most effective method to remove pollutants is the reduction of the fat
514 content. Extraction or separation of fat content can be achieved by extraction with
515 organic solvent and oil, or with enzymatic treatment. The most promising method so far
516 is the extraction with oil (olive, soybean, etc.), which does not alter the nutritional
517 properties of the meal and does not involve the use of toxic solvent. For metal
518 elimination, critical extraction or coagulation methods can be used.

519 As stated, the assessment of pollution levels in fish and the application of removal
520 techniques when necessary is a key factor for an effective discards management.
521 Nonetheless, more alternatives apart from fish oil and meal must be provided to the
522 processing sector in order to optimize the reuse of the different species by the
523 production of high-added value products. Hence, valorizing potential of the most
524 discarded species in FAROS project will be evaluated in the second part of this work.
525 The potential presence of contaminants will be discussed in terms of valorization
526 process of the different species.

527

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533

534 **Supporting Information Available**

535 Availability of pollutant monitoring data for discarded species in Spanish and
536 Portuguese fisheries is shown in Table S1.

537

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- 763

Table S1. Presence of Different Pollutants in Discarded Species of Spanish and Portuguese *Métiers*

Specie	Pollutant profile
<i>Actinauge richardi</i>	No data available
<i>Alosa alosa</i>	-
<i>Alosa fallax</i>	-
<i>Alepocephalus rostratus</i>	No data available
<i>Aphanopus carbo</i>	Heavy metals (Al, Cd, Cr, Co, Cu, Fe, Pb, Li, Mn, Hg, Ni, Se, V and Zn)
<i>Argentina silus</i>	-
<i>Argentina sphyraena</i>	-
<i>Argobuccinum olearium</i>	No data available
<i>Arnoglossus imperialis</i>	-
<i>Arnoglossus laterna</i>	-
<i>Aspitrigla cuculus</i>	-
<i>Asteroidea</i>	No data available
<i>Beryx decadactylus</i>	-
<i>Boops boops</i>	-
<i>Brama brama</i>	-
<i>Brosme brosme</i>	PCBs, α -HCH, DDE, DDT, γ -HCH, HCB, octachlorostyrene, pentachlorobenzene, DDD and heavy metals (Cd, Cu, Pb, Hg and Zn)
<i>Buccinum</i> spp.	No data available
<i>Caelorinchus caelorhincus</i>	-
<i>Callionymus lyra</i>	-
<i>Callionymus reticulatus</i>	-
<i>Cancer bellianus</i>	No data available
<i>Cancer pagurus</i>	-
<i>Capros aper</i>	-
<i>Cassidaria tyrrhena</i>	No data available
<i>Centrolophus niger</i>	No data available
<i>Centrophorus granulosus</i>	No data available
<i>Centrophorus squamosus</i>	No data available
<i>Centrocymnus coelolepis</i>	No data available
<i>Centrostephanus longispinus</i>	No data available
<i>Cepola macrophthalma</i>	No data available
<i>Charonia lampas</i>	No data available
<i>Chelidonichthys cuculus</i>	-
<i>Chelidonichthys gurnardus</i>	No data available
<i>Chelidonichthys lucerna</i>	No data available
<i>Chimaera monstrosa</i>	PCBs, α -HCH, DDE, DDT, γ -HCH, HCB, octachlorostyrene, pentachlorobenzene, DDD and heavy metals (Cd, Cu, Pb, Hg and Zn)
<i>Caelorinchus caelorhincus</i>	-
<i>Conger conger</i>	-
<i>Coryphaenoides rupestris</i>	Heavy metals (Al, Cd, Cr, Co, Cu, Fe, Pb, Li, Mn, Hg, Ni, Se, V and Zn)
<i>Crinoidea</i>	No data available
<i>Crustacea</i>	-
<i>Dalatias licha</i>	No data available
<i>Dardanus arrosor</i>	No data available
<i>Deania calcea</i>	No data available
<i>Dipturus batis</i>	-
<i>Echinoidea</i>	No data available
<i>Echinodermata</i>	-
<i>Echinus acutus</i>	No data available
<i>Eledone cirrhosa</i>	-
<i>Etmopterus spinax</i>	-
<i>Eutrigla gurnardus</i>	-
<i>Gadiculus argenteus</i>	-
<i>Gadus morhua</i>	Dioxins, furans, benzenes, bromocyclododecane, naphthalenes, PCBs, PAHs, alpha-endosulfan, α -HCH, α -HBCD, β -endosulfan, β -HCH, β -HBCD, dibenzothiophenes, cesium-134, cesium-137, cis-chlordane, cis-nonachlor, DDE, DDT, PBDEs, dibutyltin, dieldrin, endrin, γ -HCH, γ -HBCD, heptachlor, heptachlor epoxide, HCB, hexachlorobutadiene, methoxychlor, mirex, monobutyltin, monophenyltin, PBTs, oxychlordane, toxaphene, pentachlorothioanisole, perfluorodecanoic acid, perfluoroheptanoic acid, perfluorohexanesulfonic acid, perfluorohexanoic acid, perfluorononanoic acid, perfluorooctanoic acid, perfluorooctanyl sulphonic acid, perfluorooctylsulfonate acid amide, perylene, radium-226, radium-228, N, P, DDD, tetrabromobiphenol, trans-chlordane, trans-nonachlor, tributyltin, triphenyltin and heavy metals (As, Cd, Cr, Co, Cu, Fe, Pb, Mg, Mn, Mo, Hg, Na, Ni, K, Se, Sn and Zn)
<i>Gaidropsarus guttatus</i>	No data available
<i>Galeorhinus galeus</i>	-
<i>Galeus melastomus</i>	-
<i>Gastropoda</i>	-
<i>Geryon longipes</i>	No data available
<i>Glyptocephalus cynoglossus</i>	PCBs, α -HCH, DDE, γ -HCH, HCB, octachlorostyrene, pentachlorobenzene, DDD and heavy metals (Cd, Cu, Pb, Hg and Zn)
<i>Gobiidae</i>	-
<i>Halargyreus johnsonii</i>	No data available
<i>Helicolenus dactylopterus</i>	-

<i>Hexanchus griseus</i>	-
<i>Hippoglossoides platessoides</i>	PCBs, α -HCH, β -HCH, cis-chlordane, DDE, DDT, γ -HCH, HCB, oxychlordane, DDD, trans-chlordane, trans-nonachlor and heavy metals (As, Cd, Cr, Cu, Pb, Hg, Ni, Se and Zn)
<i>Holothuria</i> spp.	No data available
<i>Holothurioidae</i>	No data available
<i>Hoplostethus atlanticus</i>	Heavy metals (Al, Cd, Cr, Co, Cu, Fe, Pb, Li, Mn, Hg, Ni, Se, V and Zn)
<i>Hoplostethus mediterraneus</i>	No data available
<i>Illex coindetii</i>	-
<i>Lepidion eques</i>	-
<i>Lepidopus caudatus</i>	-
<i>Lepidorhombus boscii</i>	-
<i>Lepidorhombus</i> spp.	No data available
<i>Lepidorhombus whiffiagonis</i>	PCBs, α -HCH, cis-chlordane, DDE, DDT, dieldrin, γ -HCH, HCB, octachlorostyrene, pentachlorobenzene, DDD, trans-chlordane, trans-nonachlor and heavy metals (As, Cd, Cr, Co, Cu, Pb, Hg, Ni, Ag, Sn and Zn)
<i>Leucoraja circularis</i>	-
<i>Leucoraja naevus</i>	-
<i>Liocarcinus depurator</i>	No data available
<i>Loligo vulgaris</i>	-
<i>Lophius budegassa</i>	-
<i>Lophius piscatorius</i>	PCBs, α -HCH, cis-chlordane, DDE, DDT, dieldrin, γ -HCH, HCB, DDD, trans-chlordane, trans-nonachlor and heavy metals (Al, Cd, Cr, Co, Cu, Fe, Pb, Li, Mn, Hg, Ni, Se, V and Zn)
<i>Lophius</i> spp.	-
<i>Macropipus tuberculatus</i>	-
<i>Macropodia tenuirostris longipes</i>	No data available
<i>Macroramphosus scolopax</i>	-
<i>Malacocephalus laevis</i>	-
<i>Melanogrammus aeglefinus</i>	PCBs, PAHs, α -HCH, dibenzothiophenes, naphthalenes, cis-chlordane, DDE, DDT, dieldrin, γ -HCH, HCB, oxychlordane, perylene, DDD, trans-chlordane, trans-nonachlor and heavy metals (Cd, Cu, Pb, Hg and Zn)
<i>Merlangius merlangus</i>	PCBs, PAHs, α -HCH, dibenzothiophenes, naphthalenes, cis-chlordane, DDE, DDT, γ -HCH, HCB, perylene, DDD, trans-chlordane, trans-nonachlor and heavy metals (As, Cd, Cu, Pb, Hg and Zn)
<i>Merluccius merluccius</i>	PCBs, α -HCH, β -HCH, cis-chlordane, DDE, DDT, dieldrin, γ -HCH, HCB, DDD, trans-chlordane, trans-nonachlor and heavy metals (Cd, Cr, Cu, Pb, Hg and Zn)
<i>Microchirus variegatus</i>	-
<i>Micromesistius poutassou</i>	Heavy metals (Cd, Co, Cu, Pb, Li, Hg, Ni, Se, V and Zn)
<i>Microstomus kitt</i>	α -HCH, PCBs, dibenzothiophenes, naphthalenes, chrysene, cis-chlordane, DDE, DDT, dieldrin, γ -HCH, chlorobenzenes, octachlorostyrene, perylene, DDD, trans-chlordane, trans-nonachlor and heavy metals (Cd, Cu, Pb, Hg and Zn)
<i>Mola mola</i>	-
<i>Mollusca</i>	No data available
<i>Molpadiidae</i>	No data available
<i>Molva dypterygia</i>	Heavy metals (Al, Cd, Cr, Co, Cu, Fe, Pb, Mn, Hg, Ni, Se and Zn)
<i>Molva molva</i>	PCBs, α -HCH, γ -HCH, DDE, DDT, benzenes, octachlorostyrene, DDD and heavy metals (Cd, Cu, Pb, Hg and Zn)
<i>Mora moro</i>	-
<i>Mumida</i> spp.	No data available
<i>Mustelus asterias</i>	-
<i>Nephrops norvegicus</i>	Naphthalenes, PCBs, PAHs, dibenzothiophenes, naphthalene, perylene and heavy metals (Cd, Cu, Pb, Hg and Zn)
<i>Nettastoma melanurum</i>	No data available
<i>Nezumia aequalis</i>	No data available
<i>Nezumia sclerorhynchus</i>	No data available
<i>Octopodidae</i>	-
<i>Ommastrephidae</i>	-
<i>Opisthoteuthis agassici</i>	No data available
<i>Ophiothrix fragilis</i>	-
<i>Ophiura</i> spp.	-
<i>Pagellus acarne</i>	-
<i>Pagellus bogaraveo</i>	-
<i>Pagurus alatus</i>	No data available
<i>Pagurus</i> spp.	No data available
<i>Parapenaeus longirostris</i>	No data available
<i>Paromola cuvieri</i>	No data available
<i>Phycis blennoides</i>	-
<i>Phycis</i> spp.	No data available
<i>Pisces</i>	No data available
<i>Plesionika</i> spp.	No data available
<i>Pleuronectes platessa</i>	PAHs, naphthalenes, PCBs, α -HCH, cis-chlordane, DDE, DDT, δ -HCH, γ -HCH, dieldrin, benzenes, perylenes, DDD, trans-chlordane, trans-nonachlor and heavy metals (As, Ba, Cd, Cr, Co, Cu, Pb, Mn, Hg, Ni, Se, Ag, Sr, Sn, V and Zn)
<i>Pollachius virens</i>	DDE, DDT, γ -HCH, HCB, PCBs and heavy metals (Cd, Cu, Pb, Hg and Zn)
<i>Polybius henslowi</i>	No data available
<i>Polychaeta</i>	-
<i>Raja asterias</i>	No data available
<i>Raja brachyuran</i>	No data available
<i>Raja clavata</i>	Hg

<i>Raja montagui</i>	Hg
<i>Rajidae</i>	No data available
<i>Rhizopoda</i>	No data available
<i>Rossia macrosoma</i>	-
<i>Sacoglossa</i>	No data available
<i>Sardina pilchardus</i>	PCBs and heavy metals (Cd, Cu, Pb, Hg and Zn)
<i>Scaphander lignarius</i>	No data available
<i>Scomber colias</i>	No data available
<i>Scomber scombrus</i>	PCBs, α -HCH, γ -HCH, HCB, DDD, trans-chlordane, trans-nonachlor and heavy metals (Cd, Cr, Cu, Pb, Hg and Zn)
<i>Scyliorhinus canicula</i>	Hg
<i>Scymnodon ringens</i>	No data available
<i>Scyphozoa</i>	-
<i>Sepia officinalis</i>	-
<i>Sepia orbignyana</i>	-
<i>Sepia</i> spp.	-
<i>Sepiola</i> spp.	-
<i>Squalus acanthias</i>	-
<i>Spherooides pachygaster</i>	No data available
<i>Stichopus</i> spp.	No data available
<i>Stichopus tremulus</i>	No data available
<i>Tealia</i> spp.	No data available
<i>Todarodes sagittatus</i>	No data available
<i>Todaropsis eblanae</i>	-
<i>Torpedo marmorata</i>	-
<i>Trachurus mediterraneus</i>	No data available
<i>Trachurus picturatus</i>	No data available
<i>Trachurus</i> spp.	No data available
<i>Trachurus trachurus</i>	-
<i>Trachyrincus scabrus</i>	No data available
<i>Trigla lyra</i>	-
<i>Trigla</i> spp.	No data available
<i>Triglidae</i>	No data available
<i>Trisopterus luscus</i>	-
<i>Trisopterus minutus</i>	-
<i>Zeus faber</i>	-

Table 1. Metal Levels in Atlantic Fish Species

Specie, origin and tissue (muscle when not specified)		Pollutant	Concentration (mg/kg ww)	Reference
<i>Aphanopus carbo</i> (black scabbardfish)				
West Scotland	muscle	As	<0.002-26.49	(30)
		Cd	<0.002-0.017	
		Cu	0.07-0.27	
		Pb	0.002-0.052	
		Zn	2.12-3.90	
	liver	As	<0.05-35.79	
		Cd	2.06-18.24	
		Cu	<1.00-39.05	
		Pb	<0.05-0.471	
		Zn	29.42-108.70	
Madeira and Azores	muscle	Hg	0.19-1.43	(31)
		Cd	0.01-0.09	
		Pb	nd-0.10	
	liver	Hg	0.28-1.19	
		skin	Hg	
	Cd		0.02-0.11	
	Pb		nd-0.10	
	<i>Brosme brosme</i> (tusk or cusk)			
Northeast Atlantic		Zn	3.0-3.5	(32)
		Cu	0.13-0.18	
<i>Chelidonichthys gurnardus</i> (grey gurnard)				
Northeast Atlantic		Zn	3.0-4.2	(32)
		Cu	0.23-0.39	
<i>Conger conger</i> (European conger)				
Croatia		Hg	0.864	(7)
<i>Coryphaneoides rupestris</i> (roundnose grenadier)				
West Scotland		Cd	ND-0.01	(33)
		Cu	0.03-0.54	
		Pb	ND-0.06	
		Hg	0.02-0.28	
		Zn	1.7-2.9	
<i>Helicolenus dactylopterus</i> (blackbelly rosefish)				
Portuguese coast		Cr	0.23-0.28	¹ (34)
		Ni	0.038-0.065	

Specie, origin and tissue (muscle when not specified)	Pollutant	Concentration (mg/kg ww)	Reference	
	Hg	0.44-1.35		
	Pb	ND		
	Cd	0.025-0.013		
<i>Hippoglossoides platessoides</i>				
(American plaice)				
Barents Sea	liver	Cd	0.4	¹ (35)
		Hg	0.018	
		Cu	8.0	
		Zn	29.75	
	muscle	Hg	0.093	
		Cu	<0.43	
		Zn	4.75	
<i>Hoplostethus atlanticus</i>				
(orange roughy)				
West Scotland		Cd	ND-0.01	(33)
		Cu	0.04-0.19	
		Pb	ND-0.66	
		Hg	0.11-0.86	
		Zn	2.0-3.4	
<i>Lepidorhombus boscii</i>				
(four-spot megrim)				
Northern Iberian shelf	liver	Cu	2.45-6.93	¹ (36)
		Zn	19.5-40.75	
		Cr	0.25-0.72	
		Fe	25.25-55.5	
		Cd	0.025-0.34	
		Pb	0.0005-0.0028	
		Hg	0.0028-0.11	
<i>Lepidorhombus whiffiagonis</i>				
(megrim)				
Northeast Atlantic		Zn	2.1-2.9	(32)
		Cu	0.13-0.47	
<i>Loligo vulgaris</i>				
(European squid)				
Catalonian markets		As	1.41-4.74	(6)
		Cd	0.05-0.15	
		Hg	0.02-0.03	
		Pb	0.01-0.01	
France		Hg	0.047	(7)
<i>Lophius piscatorus</i>				
(monkfish)				
West Scotland	muscle	As	2.70-21.47	(30)
		Cd	<0.002-0.041	

Specie, origin and tissue (muscle when not specified)	Pollutant	Concentration (mg/kg ww)	Reference	
	Cu	0.06-0.22		
	Pb	<0.002-0.041		
liver	As	1.44-14.33		
	Cd	<0.05-1.4		
	Cu	1.45-36.44		
	Pb	<0.05-0.074		
Northeast Atlantic	Zn	2.6-3.3	(32)	
	Cu	0.10-0.29		
Portuguese coast	Cr	0.0075-0.53	(34)	
	Ni	0.02-0.053		
	Hg	0.118-0.63		
	Pb	<0.0075		
	Cd	0.0025-0.0075		
Croatia	Hg	0.071-0.678	(7)	
<i>Melanogrammus aeglefinus</i>				
(haddock)				
Barents Sea	liver	Cd	0.35	¹ (35)
		Hg	0.013	
		Cu	6.75	
		Zn	14	
	muscle	Hg	0.083	
		Cu	<0.43	
		Zn	3.75	
Northeast Atlantic		Zn	2.2-4.0	(32)
		Cu	0.13-0.34	
<i>Merluccius merluccius</i>				
(hake)				
West Scotland	muscle	As	0.08-3.30	(30)
		Cd	<0.002-0.062	
		Cu	0.16-0.54	
		Pb	<0.002-0.047	
	liver	As	<0.05-7.59	
		Cd	<0.05-1.43	
		Cu	<1.00-17.89	
		Pb	<0.05-0.159	
Northeast Atlantic		Zn	3.2-3.3	(32)
		Cu	0.20-0.61	
Catalonian markets		As	3.22-4.55	(6)
		Cd	0.005-0.01	
		Hg	0.12-0.29	
		Pb	0.01-0.13	
Croatia		Hg	0.052	(7)
<i>Micromesistius pouassou</i>				

Specie, origin and tissue (muscle when not specified)	Pollutant	Concentration (mg/kg ww)	Reference	
<i>(blue whiting)</i>				
West Scotland	muscle	As	0.37–6.10	(30)
		Cd	<0.002–1.178	
		Cu	0.19–0.45	
		Pb	0.005–0.030	
	liver	As	1.52-13.74	
		Cd	0.06-1.29	
		Cu	2.56-10.18	
		Pb	<0.05-0.061	
<i>Microstomus kitt</i>				
<i>(lemon sole)</i>				
Northeast Atlantic	Zn	2.6-2.9	(32)	
	Cu	0.19-0.38		
<i>Molva dypterygia</i>				
<i>(blue ling)</i>				
West Scotland	muscle	As	1.84-13.09	(30)
		Cd	<0.002–0.004	
		Cu	0.10–0.41	
		Pb	<0.002–0.008	
	liver	As	<0.02-32.44	
		Cd	<0.55-1.59	
		Cu	<1.00-7.10	
		Pb	<0.05-0.336	
<i>Molva molva</i>				
<i>(ling)</i>				
Northeast Atlantic	Zn	2.7-3.6	(32)	
	Cu	0.13-0.22		
<i>Pagellus acarne</i>				
<i>(axillary seabream)</i>				
Portuguese coast	Cr	0.15-0.425	¹ (34)	
	Ni	0.023-0.063		
	Hg	0.22-0.96		
	Pb	0.0025-0.018		
	Cd	0.0025-0.01		
<i>Pagellus bogaraveo</i>				
<i>(black spot or red seabream)</i>				
Portuguese coast	Cr	0.15-0.43	¹ (34)	
	Ni	0.023-0.063		
	Hg	0.22-0.96		
	Pb	0.0025-0.018		
	Cd	0.0025-0.01		
<i>Phycis phycis</i>				
<i>(forkbeard)</i>				

Specie, origin and tissue (muscle when not specified)		Pollutant	Concentration (mg/kg ww)	Reference
Portuguese coast		Cr	0.18-0.325	¹ (34)
		Ni	0.02-0.035	
		Hg	0.14-0.59	
		Pb	<0.008	
		Cd	0.0025-0.013	
<i>Pleuronectes platessa</i>				
(European plaice)				
Barents Sea	liver	Cd	0.53	¹ (35)
		Hg	0.045	
Cu		3		
Zn		26.3		
	muscle	Hg	0.06	
		Cu	0.68	
		Zn	5.75	
<i>Pollachius virens</i>				
(saithe)				
Barents Sea	liver	Cd	0.058	¹ (35)
		Hg	0.005	
Cu		1.75		
Zn		9.75		
	muscle	Hg	0.08	
		Zn	7.0	
Northeast Atlantic		Zn	3.7-4.5	(32)
		Cu	0.46-0.65	
<i>Sardina pilchardus</i>				
(sardine)				
Catalonian markets		As	3.53-3.94	(6)
		Cd	0.002-0.01	
		Hg	0.07-0.09	
		Pb	0.01-0.08	
Spanish market		Hg (total)	0.06	(37)
Portuguese waters and markets		Hg	0.0116–0.0280	(8)
		Cd	0.0017–0.0151	
		Pb	0.0029–0.0569	
		As	0.8116–1.336	
<i>Scomber scombrus</i>				
(Atlantic mackerel)				
Northeast Atlantic		Zn	3.3-5.2	(32)
		Cu	0.70-0.97	
Catalonian markets		As	1.73-7.47	(6)
		Cd	0.003-0.01	
		Hg	0.06-0.15	
		Pb	0.01-0.02	

Specie, origin and tissue (muscle when not specified)	Pollutant	Concentration (mg/kg ww)	Reference
Slovenia <i>Scyliorhinus caniculus</i> (small-spotted catshark)	Hg	0.035-0.056	(7)
Northeast Atlantic	Zn	8.5-8.7	(32)
	Cu	0.50-0.53	
<i>Sepia spp</i> (cuttlefish)			
Catalonian markets	As	2.45-5.33	(6)
	Cd	0.01-0.09	
	Hg	0.04-0.08	
	Pb	0.01-0.10	
<i>Trachurus trachurus</i> (Atlantic horse mackerel)			
Mauritania	Cd	0.01	¹ (38)
	Cu	0.4	
	Zn	10.5	
	Hg	0.075	
Portuguese waters and markets	Hg	0.0380–0.3371	(8)
	Cd	0.0030–0.0141	
	Pb	0.0031–0.0215	
	As	0.655–1.941	
<i>Trisopterus luscus</i> (pouting)			
Northern Iberian shelf-liver	Cu	0.58-2.0	¹ (36)
	Zn	2.0-6.25	
	Cr	0.11-0.89	
	Fe	17.0-29.5	
	Cd	0.005-0.085	
	Pb	0.0005-0.013	
	Hg	0.00025-0.0085	
<i>Zeus faber</i> (John dory)			
Northeast Atlantic	Zn	3.1	(32)
	Cu	0.12-0.14	
Morocco	Hg	0.066	(7)

¹assuming wet/dry ratio of 0.25

Table 2. POPs Levels in Atlantic Fish Species

Specie, origin and tissue (muscle when not specified)		Pollutant	Concentration (mg/kg ww for metals and µg/kg ww for POPs)	Reference
<i>Aphanopus carbo</i> (black scabbardfish)				
West Scotland	muscle	PBDE	0.194-0.98	(29)
	liver	PBDE	0.57-11.98	
Ireland-liver		HCB	3.89	(39)
		PCBs	17.03	
		DDTs	75.64	
		chlordanane	22.8	
Madeira-liver		HCB	0.43	(39)
		PCBs	112.34	
		DDTs	107.42	
		chlordanane	15.83	
Meriadzec-liver		HCB	2.13	(39)
		PCBs	93.8	
		DDTs	185.6	
		chlordanane	49.1	
Rockall-liver		HCB	2.62	(39)
		PCBs	15.9	
		DDTs	43.8	
		chlordanane	12.6	
Sesimbra-liver		HCB	3.2	(39)
		PCBs	257.2	
		DDTs	165.4	
		chlordanane	30.7	
<i>Arnoglossus laterna</i> (Mediterranean scaldfish)				
Adriatic Sea		PCBs	2.07	(40)
Southern Mediterranean		dl-PCBs	0.23	(41)
Adriatic Sea		PCBs	2.13	(42)
		PCDD/Fs	ND	
<i>Aspitrigla cuculus</i> (red gurnard)				
Adriatic Sea		PCBs	1.08	(40)
Adriatic Sea		PCBs	1.06	(42)
		PCDD/Fs	0.00022	
<i>Boops boops</i> (bogue)				
Adriatic Sea		PCBs	3.13	(42)
		PCDD/Fs	0.00024	
<i>Brosme brosme</i> (tusk or cusk)				

Specie, origin and tissue (muscle when not specified)	Pollutant	Concentration (mg/kg ww for metals and µg/kg ww for POPs)	Reference	
<i>Cancer pagurus</i>				
(edible crab)				
South Norway	hepatopancreas	PCDD/Fs	0.639-15.98	(11)
	claw meat	PCDD/Fs	0.125	
<i>Conger conger</i>				
(European conger)				
Adriatic Sea		PCBs	27.72	(42)
		PCDD/Fs	0.00026	
Croatia		PCBs	3.82	(7)
<i>Coryphaneoides rupestris</i>				
(roundnose grenadier)				
Ireland-liver		HCB	17.9	(39)
		PCBs	379.8	
		DDTs	577.7	
		chlordanes	36.55	
West Scotland	muscle	PBDE	<dl-2.11	(29)
	liver	PBDE	2.08-91.9	
<i>Eutrigla gurnardus</i>				
(grey gurnard)				
Adriatic Sea		PCBs	2.08	(40)
<i>Helicolenus dactylopterus</i>				
(blackbelly rosefish)				
Adriatic Sea		PCBs	2.64	(42)
		PCDD/Fs	0.0002	
<i>Hippoglossoides platessoides</i>				
(American plaice)				
Northwest Atlantic		PBDE	0.62	(43)
<i>Hoplostethus atlanticus</i>				
(orange roughy)				
Meriadzec-liver		HCB	5.49	(39)
		PCBs	198.6	
		DDTs	151.4	
		chlordanes	18.7	
Ireland-liver		HCB	9.95	(39)
		PCBs	260.76	
		DDTs	259.3	
		chlordanes	38.64	
<i>Lepidopus caudatus</i>				
(silver scabbardfish)				
Adriatic Sea		dl-PCBs	1.10	(41)
<i>Lepidorhombus boscii</i>				
(four-spot megrim)				
Adriatic Sea		PCBs	0.657	(42)

Specie, origin and tissue (muscle when not specified)	Pollutant	Concentration (mg/kg ww for metals and µg/kg ww for POPs)	Reference
	PCDD/Fs	ND	
<i>Lepidorhombus whiffiagonis</i> (megrim)			
Adriatic Sea	PCBs	2.08	(40)
<i>Loligo vulgaris</i> (European squid)			
<i>Lophius budegassa</i> (blackbellied angler)			
Adriatic Sea	PCBs	0.245	(42)
	PCDD/Fs	0.0000056	
<i>Lophius piscatorus</i> (monkfish)			
Spanish Atlantic Southwest coast	PCBs	2.512-3.112	(44)
	PCDD/Fs	0.00033-0.00086	
North Sea	dl-PCBs	0.09	(41)
Croatia	PCBs	1.8–3.3	(7)
<i>Merluccius merluccius</i> (hake)			
Bay of Biscay	PCDD/F	0.000086	(5)
Southern Italy	HCB	<dl-0.48	(45)
	DDTs	0.98-9.2	
	PCBs	6.72-101.3	
Adriatic Sea	PCBs	3.41	(42)
	PCDD/Fs	0.00008	
Croatia	PCBs	2.7–4.6	(7)
Atlantic Ocean	dl-PCBs	1.33	(41)
<i>Molva molva</i> (ling)			
Ireland-liver	HCB	31.19	(39)
	PCBs	610.8	
	DDTs	505.24	
	chlordanes	160.11	
<i>Mora moro</i> (common mora)			
Mediterranean Sea-liver	DDTs	745–1630	(46)
	PCBs	736–5490	
<i>Mustelus asterias</i> (starry smooth-hound)			
North Sea	dl-PCBs	7.60	(41)
<i>Pagellus acarne</i> (axillary seabream)			
Adriatic Sea	PCBs	1.52	(40)
<i>Phycis blennoides</i>			

Specie, origin and tissue (muscle when not specified)	Pollutant	Concentration (mg/kg ww for metals and µg/kg ww for POPs)	Reference
(greater forkbeard)			
Mediterranean Sea-liver	DDTs	214	(47)
	PCBs	350	
Adriatic Sea	PCBs	0.0011	(42)
	PCDD/Fs	0.0000096	
<i>Pleuronectes platessa</i>			
(European plaice)			
North Sea	dl-PCBs	0.34	(41)
<i>Pollachius virens</i>			
(saithe)			
North Sea	PCDD/F	0.098	(5)
Norway	PCDD/F	0.025	(5)
Mediterraneum Sea	dl-PCBs	0.47	(41)
<i>Raja clavata</i>			
(thornback ray)			
North Sea	dl-PCBs	0.15	(41)
<i>Sardina pilchardus</i>			
(sardine)			
Bay of Biscay	PCDD/F	0.603	(5)
Spanish market	PCDD/Fs	0.00145-0.00239	(48)
	PCBs	0.049-0.0652	
Spanish Atlantic Southwest coast	PCBs	20.9-23.8	(44)
	PCDD/Fs	0.00084-0.00119	
Adriatic Sea	dl-PCBs	0.88	(41)
<i>Scomber scombrus</i>			
(Atlantic mackerel)			
Bay of Biscay	PCDD/F	0.317	(5)
North Sea	PCDD/F	0.330	(5)
North Ionan Sea-liver	HCB	ND-6.07	(49)
	DDE	0.25-104	
	PCBs	0.1-158	
Southern Italy	HCB	<dl-2.83	(45)
	DDTs	<dl-23.8	
	PCBs	2.54-237.8	
Northwest Atlantic	PBDE	7.11	(43)
	HBCD	1.44	
Slovenia	PCBs	8.4-17.4	(7)
<i>Trachurus mediterraneus</i>			
(Mediterranean horse mackerel)			
Adriatic Sea	PCBs	5.2	(42)
	PCDD/Fs	0.000062	
<i>Trachurus trachurus</i>			
(Atlantic horse mackerel)			

Specie, origin and tissue (muscle when not specified)	Pollutant	Concentration (mg/kg ww for metals and µg/kg ww for POPs)	Reference
Adriatic Sea	PCBs	6.15	(42)
	PCDD/Fs	ND	
<i>Trigla lyra</i> (piper gurnard)			
Adriatic Sea	PCBs	0.70	(40)

Table 3. Pollutant content in Fish By-Products

Type and Origin	Pollutant	Concentration (ng/g)	Reference
Fish oil -Mixed (no salmon)	PCDD/Fs	0.00055	(13)
	PBDEs	0.887	
Fish oil-Mixed (including salmon)	PCDD/Fs	0.00157	(13)
	PBDEs	0.898	
Fish oil-Salmon	PCDD/Fs	0.0072	(13)
	PBDEs	3.260	
Fish oil-Shark	PCDD/Fs	0.139	(13)
	PBDEs	57.7	
Fish oil-Menhaden	PCDD/Fs	0.0818	(13)
	PBDEs	50.9	
Fish oil- Fish processing industry blend-Baltic Sea	HCB	12	(51)
	DDTs	337	
	PCBs	197	
Fish oil-unknown	HCH	11.9	(52)
	DDTs	30.0	
	PCB	74.0	
	PBDEs	12.7	
Fish feed-Scottish source	HCH	30.4	(52)
	DDTs	47.9	
	PCBs	157.3	
	PBDEs	16.2	
Fish meal-Peru	PCDDs	0.132	(22)
	PCDFs	0.058	
	DL-PCBs	0.17	
	non DL-PCBs	2.0	
	PBDEs	0.068	
	HCB	0.18	
	DDTs	1.9	
	As	3 (mg/kg)	
	Hg	0.045 (mg/kg)	
	Cd	1.2 (mg/kg)	
	Pb	0.087 (mg/kg)	
Fish oil-Norway	PCDDs	2.48	(22)
	PCDFs	5.2	
	DL-PCBs	21	
	non DL-PCBs	133	
	PBDEs	26	
	HCB	40	
	DDTs	254	
Krill meal-Norway	PCDDs	0.048	(22)
	PCDFs	0.060	
	DL-PCBs	0.072	

Type and Origin	Pollutant	Concentration (ng/g)	Reference
	non DL-PCBs	1.3	
	PBDEs	0.047	
	HCB	1.1	
	DDTs	-	
Fish oil for feed- different sources	HBCD	3.34-26.8	(53)
Fish oil for food-different sources	HBCD	0.19-4.19	(53)