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24 Abstract

25 Although seafood is a nutritious protein source, due to marine environmental pollution, seafood may 26 also be a source of contaminants. The results obtained within the FP7-ECsafeSEAFOOD-project show 27 that among the range of studied environmental contaminants certainly methylmercury (MeHg) requires deeper investigation. This paper presents the results of a probabilistic risk assessment for 28 29 MeHg based on: (1) primary concentration data, as well as secondary data from published papers, 30 and (2) primary species-specific consumption data collected in five European countries (Belgium, 31 Ireland, Italy, Portugal and Spain). The results indicated that in the southern European countries, 32 larger subgroups of the population (up to 11% in Portugal) are potentially at risk for a MeHg 33 exposure above the Tolerable Weekly Intake (TWI) value, while this risk is much lower in Ireland and Belgium. This research confirms the substantial contribution of tuna to MeHg exposure in each of the 34 countries. Also hake, cod, sea bream, sea bass and octopus are identified as important contributors. 35 36 From this study, it is concluded that a country-specific seafood consumption frequency advice is 37 needed. Policy makers may adopt the results of this study in order to develop consumer advices that 38 optimise health benefits versus potential health risks by providing species-specific information.

39

40 Keywords: European consumers, exposure, methylmercury, risk assessment, seafood species

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43 Highlights:

• Highest exposure to methylmercury (MeHg) is assessed in Portugal, followed by Spain, Italy,

45 Ireland and Belgium

- 11% of the Portuguese population is potentially at risk for a MeHg exposure above the
- 47 Tolerable Weekly Intake (TWI) value
- Species contributing most to MeHg exposure are tuna, hake, cod, sea bream, sea bass and
- 49 octopus

51 **1. Introduction**

52 In recent years, a number of studies have focused on the nutritional-toxicological conflict of frequent 53 seafood consumption. While the health benefits of consuming seafood are well established, 54 attention has also been paid to environmental contamination of seafood, as well as the impact of this contamination on consumers' health. Guidelines regarding seafood consumption frequency in 55 different countries follow a consensus, however differing to a small extent. In a review about risk-56 benefit analysis of seafood consumption, it was stated that the benefits outweigh the risks when a 57 58 variety of fish is consumed at least twice per week (Hellberg et al., 2012). In addition, a targeted communication approach for certain populations (e.g. pregnant women and young children) is 59 warranted in order to ensure that these groups consume fish species that are low in contaminants, 60 but high in omega-3 fatty acids (Hellberg et al., 2012). In turn, Hoekstra et al. (2013) concluded that 61 the overall benefits to the Dutch population of eating 200 g of fish per week, instead of the current 62 (lower) consumption amounts in the Netherlands, outweigh the risks. According to the method used 63 64 in this study, eating 500 g of fish per week would even be more beneficial, despite the larger risks of 65 being exposed to higher levels of contaminants. However, Domingo (2016) highlights the importance of specific seafood species consumed, the frequency of consumption, as well as the portion and meal 66 67 size, in order to adequately balance the health benefits and risks of regular seafood consumption. Moreover, the EFSA Scientific Committee compared the benefits of seafood consumption regarding 68 69 omega-3 long-chain polyunsaturated fatty acids (LCPUFA), with the risks to MeHg in seafood and 70 recommended a weekly intake of fish of 1-2 servings (equivalent to 150-300 g) to meet the Dietary 71 Reference Value (DRF) for n-3 LCPUFA. Notwithstanding, when consuming species with a high MeHg 72 content, only a few servings (<1-2) per week can be consumed before reaching the Tolerable Weekly 73 Intake (TWI). Hence, EFSA emphasized the need that seafood species with a high content of MeHg 74 should be limited in a healthy diet (EFSA, 2015).

In this study, we present the results of a risk assessment on MeHg present in seafood. The possible
 health risks from exposure through the seafood consumption pattern were assessed in five European

77 countries: Belgium, Ireland, Italy, Portugal and Spain. The reason to focus on MeHg was twofold. Firstly, within the ECsafeSEAFOOD-project, exposure and risk assessment estimations were 78 performed for different environmental contaminant groups present in seafood, namely for 79 80 methylmercury, polycyclic aromatic hydrocarbons, musks, endocrine disruptors, perfluorinated 81 compounds, brominated flame retardants, UV-filters, inorganic arsenic and pharmaceuticals. The 82 assessment was conducted to screen for which contaminants the seafood consumption pattern 83 could be of concern for public health. Based on the pragmatic approach and the obtained results, it 84 was concluded that refinement of the exposure assessment through seafood intake and risk 85 reduction measurements are certainly needed for MeHg. Secondly, in a recent EFSA report, it was recommended that risk assessment of MeHg should be performed at national level considering the 86 87 national seafood consumption pattern, focussing on the specific species consumed within each 88 country (EFSA, 2015).

89 MeHg exposure assessment has been the main topic of a number of studies (Afonso et al., 2015; 90 Brambilla et al., 2013; Cardoso et al., 2013; Cardoso et al., 2010; Di Leo et al., 2010; Kuballa et al., 91 2011; Maycock and Benford, 2007; Miklavcic et al., 2011; Nunes et al., 2014; Olmedo et al., 2013; 92 Ortega-Garcia et al., 2009; Perello et al., 2014; Ruiz-de-Cenzano et al., 2014; Sioen et al., 2008; 93 Storelli et al., 2003; Storelli et al., 2005; Strom et al., 2011). However, it is stated that the substantial intake of MeHg, which is linked to the high consumption of fish and shellfish, deserves further 94 investigation, while several population subgroups need better guidelines to base their seafood 95 96 choices explicitly on mercury content (Groth, 2010; Perello et al., 2014). On the other hand, until recently, few research considered risk assessment in different countries with distinct seafood 97 98 consumption patterns, and using a detailed collection of consumption data covering the diversity of 99 species with different contamination levels.

100 The present study means a novel contribution to the evaluation of the potential risk of MeHg 101 exposure through seafood consumption by considering five European countries with different

102 seafood consumption patterns, and by considering detailed data on consumption levels of 32 different seafood species. For this purpose, MeHg concentration data collected within the 103 104 ECsafeSEAFOOD-project, as well as data from the scientific literature were used. Seafood species 105 vary greatly in their level of MeHg while there is also a wide variation in the seafood species 106 consumed in different countries across the EU, as well as in the overall seafood consumption 107 frequency (Cardoso et al., 2010; EFSA, 2015; Storelli et al., 2003). Consequently, an additional goal of 108 this investigation was to provide insight in the contribution of different seafood species to the MeHg 109 exposure, and hence to the potential health risk, in each country individually. Thus, we addressed the 110 advice of EFSA that each country should consider the specific species consumed in order to make a recommendation for each country, specifically regarding the human health risks and benefits (EFSA, 111 2015). 112

113 **2. Material and methods**

114 2.1. Concentration data

115 A database for MeHg in seafood was compiled based on concentration data (from pooled samples) 116 collected within the ECsafeSEAFOOD-project (52 data points) (methodology; Aznar et al., submitted) combined with additional data from scientific literature (Afonso et al., 2015; Barrento et al., 2008; 117 118 Brambilla et al., 2013; Cardoso et al., 2010; Kwasniak et al., 2012; Miklavcic et al., 2011; Perello et al., 119 2014; Ruiz-de-Cenzano et al., 2014; Storelli et al., 2003; Storelli et al., 2005). In total, 94 data points 120 from scientific literature data were collected. The concentration data includes data originating from 121 measurements performed in raw and canned samples of species commercially relevant in Europe. 122 Data from two species were missing: dry/salted cod and lobster. To bridge this gap, missing 123 concentration data for dry/salted cod were completed by assigning the data of fresh cod. For lobster, 124 it was assumed that the MeHg concentration would be equal to the reported total mercury (Hg) concentration. Contaminant data for the different species considered in the exposure estimations 125 126 are summarized in Table 1 .

Insert Table 1. MeHg contents (mean, standard deviation, μg/kg ww) in the different species
 analysed.

129 2.2. Consumption data

130 In the framework of the ECsafeSEAFOOD-project, a web-based consumer survey was performed in 131 October 2013 in five European countries: Belgium, Ireland, Italy, Portugal and Spain. These countries 132 were selected to cover western, northern and southern Europe. Consequently, the data cover a 133 heterogeneous population in terms of seafood consumption frequency and dietary habits. The 134 samples were nationally representative regarding gender, region and age within the range 18-75 135 years (Table 2) (Jacobs et al., 2015).

136 Insert Table 2. Socio-demographic profile of the sample (n=2824).

Within this survey, the total seafood consumption frequency, as well as the seafood consumption 137 frequency of 32 different seafood species, were inquired using self-reported items. The 32 species 138 139 selected in collaboration with the study partners, were based on the seafood consumption pattern in 140 the five involved countries, and also on the susceptibility of certain species to contain considerable 141 concentrations of certain environmental contaminants. The participants reported their consumption frequency as the number of portions per week or per month, assuming that a portion of seafood per 142 143 meal is about 150-200 g. The information about portion size was explicitly communicated to the 144 study participants. The reported seafood consumption frequencies were transformed into a 145 continuous scale. As a result of this transformation, mean scores (frequency per week) were 146 calculated. For this purpose "Daily" was replaced by a value of 7.0, "5-6 times a week" was replaced by 5.5, "3-4 times a week" by 3.5, "2 times a week" by 2.0, "Once a week" by 1.0, "Less frequently" 147 148 by 0.25, and "Never" by 0. The same was done for the consumption frequency of each of the 32 149 species. Therefore, "2-3 times a month", "Once a month", "1-5 times every 6 months", were also 150 replaced by 0.6, 0.25, 0.15, respectively. Subsequently, consumption frequencies of the 32 species 151 were corrected based on the total consumption frequency reported in the first question because of

overestimation of the consumption when considering separate species. The consumption frequency
was transformed from times per week to grams per week by multiplying by 175 g, assuming this
value as the mean portion size.

155 For each country, at least 85% of the total seafood diet (based on the median) is represented by the 156 15 most consumed species. Consequently, only these 15 most consumed seafood species in each 157 country were considered for exposure assessment. For each country, a distribution was fitted to the 158 consumption frequency data of each species using @RISK version 6 (Palisade Corporation, USA). The distribution fitting was performed in two subsequent steps. Firstly, a distribution was fitted to the 159 160 consumption frequency data of the species, when only the consumers of the species were 161 considered. This distribution was truncated with the lowest consumption frequency as the truncated 162 minimum, and the maximum consumption frequency as the truncated maximum. Secondly, the distribution was combined with the data of the non-consumers (zero intake), taking into account the 163 164 proportion of consumers and non-consumers of the considered species. Table 1 to Table 5 presented in Appendix I show, for each country, the best fitted distributions to the consumption data. 165

166 Furthermore, the body weight (bw) of the participants was also assessed in this survey and a 167 distribution was fitted to these data using @RISK version 6. Table 6 in Appendix I shows the best fitted distributions for body weight for each country. The distribution was truncated with the 168 minimum body weight and the maximum body weight of the data. The ratios of mean body weights 169 170 between countries, and the ranking of the mean values across countries, are fairly compatible with 171 the Eurobarometer data (Special Eurobarometer 246, 64.3 Health and food, 2006). However, mean values reported in the Eurobarometer are lower. A potential explanation is that the Eurobarometer 172 173 sample includes respondents from the age of 15 years onwards, while our study included 174 respondents from the age of 18 years onwards. Given the fact that weight increases with age, this 175 would explain (at least partially) the higher mean values in this study. In addition, the fieldwork of 176 the Eurobarometer was done in 2005, while the fieldwork of the current survey was conducted in

177 2013. Within the Belgian population, a significant linear increase in the mean Body Mass Index (BMI)

is shown based on data from 1997 until 2013 (Scientific Institute of Public Health (WIV-ISP), 2013).

- 179 Hence, it is likely that population weight continued to increase during the time period of 2005-2013.
- 180 The best fitted distributions for the consumption and body weight data were determined using the
- 181 Chi-square statistics, probability/probability (P/P) and quantile/quantile (Q/Q) plots.
- 182 The seafood consumption frequency distributions were divided by the body weight distributions,
- 183 resulting in a consumption dataset (expressed in kg/kg bw/day) for each country.

184 2.3. Exposure assessment

To estimate the exposure to MeHg through seafood consumption in each country, the consumption frequency data of the species are combined with the concentration data of the contaminants in the samples according to the following formula:

$$Y_{i,c} = \sum_{\nu=1}^{\nu=15} C_{c,\nu} \times X_{i,\nu}$$

- 188 $C_{c,v}$ = concentration of contaminant c in seafood species v [µg/kg ww]
- 189 $X_{i,v}$ = consumption of seafood species v by individual i [kg/kg bw/day]
- 190 $Y_{i,c}$ = exposure to contaminant c for individual i [µg/kg bw/day]

191

No adjustments were made for intra-individual correlations in this aggregated exposure model,
meaning that an "upper bound" estimation of the exposure was calculated.

194 2.3.1. Probabilistic exposure assessment

195 Calculations were performed using the software package @RISK version 6 (Palisade Corporation, US) 196 for Microsoft Excel. As earlier described, best fit distributions are used for the consumption 197 frequency and for the body weight data. For the concentration data a probabilistic approach is used 198 instead of a deterministic approach (point estimate, mean value) when distribution fitting is possible

199 and a good fit is obtained in order to take into account the variability and uncertainty in both 200 consumption and contaminant concentration. Distribution fitting was feasible when at least five 201 concentration data points were available, among which three data values had to be above the 202 detection limit and above the quantification limit. A probabilistic approach was possible for 11 203 species (see Appendix II for the applied distributions). Best fit distributions for the concentration data 204 were determined using Kolmogorov-Smirnov and Anderson-Darling statistics, the P/P and Q/Q plots. 205 The distributions were truncated with the truncated minimum equal to the lowest concentration at 206 the lower end of the distribution, and with the truncated maximum equal to two times the highest 207 concentration, at the higher end of the distribution. A deterministic approach was used for the species Alaska pollock, canned sardine, clams, cuttlefish, haddock, herring, lobster, mussels, 208 209 pangasius, sea bass, squid and tuna.

First order Monte Carlo simulations were performed considering 100,000 iterations to estimate the MeHg intake through the seafood diet for the two scenarios: lower bound (LB) and upper bound (UB). Non-detects (<LOD) and non-quantified (<LOQ) were considered as zero and LOD or LOQ for LB and UB scenarios, respectively. Only for pangasius, the measured concentration was lower than the LOQ, while for all the other species, the measured concentrations were above the LOQ. The estimated daily intake was expressed in µg/kg bw/day.

216 **2.4.** Risk characterisation

To evaluate the potential health risks of MeHg exposure, a health based guidance value can be applied. The European Food Safety Agency (EFSA) established a Tolerable Weekly Intake (TWI) for oral exposure to MeHg in humans equal to 1.3 μ g/kg bw/day (based on neurodevelopmental outcomes) (EFSA, 2012). The use of this TWI value was chosen for this study as the value is generally accepted in a European context, whereas in the US a Reference Dose (RfD) of 1.0 μ g/kg bw/day is generally applied to evaluate the potential health risk of MeHg exposure (Rice et al., 2000).

223 3. Results and discussion

224 3.1. Occurrence of MeHg in seafood species

225 Data for the different species considered in the exposure estimations are summarized in Table 1. The 226 variation between species is due to biotic (size, sex, longevity, growth rate, feeding habits, trophic position, habitat) and abiotic parameters (e.g. process of sedimentation and persistence of MeHg in 227 228 sea depths, environmental conditions) (Kasper et al., 2009; Ruiz-de-Cenzano et al., 2014; Storelli et 229 al., 2005). The highest mean levels of MeHg were found in tuna (462 μ g/kg ww), monkfish (227 µg/kg ww), sea bass (222 µg/kg ww), sea bream (208 µg/kg ww), canned tuna (167 µg/kg ww), 230 231 octopus (126 μg/kg ww), hake (123 μg/kg ww), lobster (121 μg/kg ww) and cuttlefish (104 μg/kg 232 ww).

233 3.2. Exposure assessment

The most common form of human exposure to MeHg is from seafood consumption (Hellberg et al., 2012). Table 3 shows the results of the exposure assessment for MeHg in the study countries. Mean values, standard deviations and the percentiles (P 50, P 75, P 90, P 95, P 99) of the exposure distributions are described in Table 3.

238

Insert Table 3. Results of the exposure assessment for MeHg in the five countries.

239 The assessed exposure estimates from this study are comparable with the estimates provided by 240 EFSA (EFSA, 2012). EFSA assessed MeHg exposure across different age groups based on dietary 241 surveys from 17 EU countries. The mean medium bound MeHg exposure ranged from 0.06 µg/kg 242 bw/week (for elderly) to 1.57 μ g/kg bw/week (for toddlers). The P 95 ranged from 0.14 μ g/kg bw/week (very elderly) to 5.05 μ g/kg bw/week (adolescents). It must be noted that higher exposure 243 244 levels were assessed for children, while dietary exposure to MeHg in women of childbearing age was 245 reported not to be different from adults in general (EFSA, 2012). Children may be relatively more 246 exposed than adults due to their relatively higher food consumption in relation to their lower body 247 weights. Table 3 indicates that Portuguese adults have the highest exposure to MeHg, followed by

Spanish adults. By contrast, Irish and Belgian adults show the lowest exposure to MeHg throughseafood consumption.

250 3.3. Risk characterisation

251 The estimated mean seafood exposure to MeHg is lower than the provided TWI of 1.3 μ g/kg 252 bw/week in each of the study countries (Table 3), which is in agreement with the results of EFSA 253 (EFSA, 2012). However, the P 95 for Irish, Italian, Portuguese and Spanish adults are close to, or 254 above, the TWI. Specifically, exposure to MeHg through seafood consumption may be of concern for 255 about 11% of the Portuguese population, 5% of the Spanish population, 4% of the Italian population, 256 3% of the Irish population and 1% of the Belgian population (Table 3). These numbers follow the 257 order of the total seafood consumption frequency as Portugal has the highest seafood consumption 258 (2.8 times per week; about 490 g), followed by Spain (2.6 times per week; about 455 g) and Italy (2.1 259 times per week; about 368 g), whereas the Irish and Belgian populations have the lowest total 260 seafood consumption frequency: 1.6 (about 280 g) and 1.1 (193 g) times per week, respectively.

The finding that a larger subgroup of the southern European country's population is potentially at risk due to a MeHg exposure through seafood intake is in agreement with the results of a previous investigation (Cardoso et al., 2010), indicating that the probability of exceeding the MeHg toxicological reference value was higher for Portugal (6.7%) and Spain (4.5%), compared to Germany (0.2%), the Netherlands (0.2%) and the UK (0.04%).

Most of the research on MeHg exposure via seafood consumption has been performed in southern European countries such as Portugal, Spain and Italy (countries with a high seafood consumption frequency) (Afonso et al., 2015; Brambilla et al., 2013; Cardoso et al., 2013; Cardoso et al., 2010; Nunes et al., 2014; Olmedo et al., 2013; Ortega-Garcia et al., 2009; Perello et al., 2014; Ruiz-de-Cenzano et al., 2014; Storelli et al., 2003; Storelli et al., 2005). The results of these studies are comparable with our current findings, namely that a potential health concern exists regarding MeHg exposure via seafood consumption for certain subpopulations in some countries, especially for

vulnerable groups including children and women of childbearing age. It must be noted that children may have a higher exposure to MeHg than that estimated for the adult population in the considered countries. Moreover, it was shown that women aged 18 - 45 years (women of childbearing age) participating in the consumption surveys included in the EFSA report appeared to have similar dietary exposure as the general adult population (EFSA, 2012). Furthermore, pregnant women can be present in the group of high and frequent seafood consumers and unborn children constitute the most vulnerable group.

The results of the present study support and emphasize the need for targeted, species-specific and country-specific recommendations in order to mitigate the risk of MeHg exposure through seafood consumption.

283 3.4. Contribution of seafood species to MeHg exposure

284 To develop and optimise country-specific risk mitigation communication strategies and dietary 285 recommendations, the contribution of the seafood species to the total MeHg exposure was 286 determined for each country (Table 4). In each of the five countries, tuna and canned tuna are the 287 biggest contributors to MeHg exposure through seafood consumption. Tuna has the highest mean 288 concentration level, while canned tuna has a relatively high mean MeHg concentration (Table 1). Canned tuna is highly consumed, especially in Italy, Portugal and Spain, while fresh tuna is consumed 289 290 to a lesser, but still substantial extent in those countries. Discouraging the consumption of top predator fish species, such as tuna, is a frequently drawn conclusion, especially for susceptible 291 292 groups (Brambilla et al., 2013; Cardoso et al., 2010; EFSA, 2012; Nunes et al., 2014; Olmedo et al., 293 2013; Ortega-Garcia et al., 2009; Ruiz-de-Cenzano et al., 2014).

Insert Table 4. Contribution of the considered species to MeHg exposure through the
 seafood pattern in the five countries (based on the mean exposure). The shading indicates
 the five species contributing most to MeHg exposure.

297 The highest potential health risk for the Portuguese population may be attributed to the higher total seafood consumption frequency in Portugal compared to the other four countries. However, the 298 299 Portuguese population has not a substantial higher seafood consumption than that of Spain, but the 300 amount of consumers potentially at risk is about two times higher for Portugal than for Spain. 301 Comparing the seafood consumption pattern in these two southern European countries, the 302 consumption of cod (and to some extent that of monkfish) in Portugal is an important explanatory 303 factor for the higher amount of consumers potentially at risk. Cardoso et al. (2010) recently 304 concluded that cod consumption in Portugal should be reduced in order to reduce the potential risks 305 related with MeHg exposure. In Belgium and Ireland, cod is also identified as an important 306 contributor to MeHg exposure since in both countries, cod is one of the most consumed species. 307 Furthermore, EFSA also reports cod as one of the most important contributors to MeHg exposure 308 through seafood consumption (EFSA, 2012).

309 Table 4 shows that hake is also an important contributor to MeHg exposure in Spain, Portugal and 310 Ireland, which is in agreement with the results of previous surveys (Cardoso et al., 2013; Cardoso et 311 al., 2010; EFSA, 2012; Nunes et al., 2014; Storelli et al., 2005). In Italy, octopus is an important 312 contributor to MeHg exposure through seafood consumption. Research performed regarding MeHg 313 exposure through cephalopods consumption in Portugal revealed that squid does not present a 314 serious health concern, but cuttlefish and octopus consumption should not exceed two 150 g meals per week (Cardoso et al., 2012). Noteworthy, due to the high MeHg levels in sea bass and sea bream 315 316 (Table 1), sea bass is identified as an important contributor to MeHg exposure for Italy and Ireland, 317 and both species being important contributors in Italy, Portugal and Spain. To the best of our 318 knowledge, the latter finding has not been observed in previous studies, and hence, to some extent it 319 is surprising. This result likely reflects the dynamics of seafood consumption trends in the last years 320 in the southern European countries compared to previous consumer surveys. Nonetheless, in a 321 consumer guide focusing on mercury levels in seafood developed by Groth (2010), sea bream (porgy) and sea bass have been listed as "higher-mercury fish". 322

323 **3.5.** Uncertainties and limitations

Although this study includes the consumption frequency of a rather large set of seafood species indifferent regions of Europe, some limitations should be acknowledged.

326 When consuming seafood, people are exposed to both contaminants and nutrients influencing 327 specific health outcomes (i.e. neurological, cardiovascular, immunological systems). Therefore, 328 balanced assessments of contaminants and nutrients are recommended (Domingo, 2016; Gribble et 329 al., 2016). This study aimed at assessing the potential health risks due to MeHg exposure. However, 330 results from previous studies focussing on both MeHg and nutrients (omega-3 LCPUFA and/or 331 selenium), highlighted that seafood species high in MeHg levels should be reduced -or avoided- to a 332 certain extent (especially in vulnerable groups) in order to limit MeHg exposure. A frequent consumption of these seafood species may imply exceeding the toxicological threshold value for 333 334 MeHg before reaching the recommended level of omega-3 LCPUFA and/or selenium (Cardoso et al., 335 2013; Cardoso et al., 2010; Hellberg et al., 2012; Nunes et al., 2014; Strom et al., 2011). Therefore, 336 this study investigated whether a potential health risk exists in different European countries. The 337 contribution of about 85% most consumed seafood species to MeHg exposure was assessed in five 338 countries representing different seafood consumption patterns. Such approach is highly relevant in 339 order to optimise species- and country- specific recommendations to assure that consumers benefit 340 from the nutritional assets of seafood while lowering the potential health risks.

An important limitation of the current study is that the exposure and risk assessments were performed based on data of MeHg concentrations measured in raw (and canned) samples. Of course, it is of interest to know the concentrations of MeHg after processing, as in most cases and in line with most EU-consumers' seafood consumption habits, seafood is not consumed raw. Within another part of the ECsafeSEAFOOD-project, 18 steamed seafood samples were analysed regarding MeHg levels. In about 55% of the samples, the concentrations of MeHg increased at least 10%, compared to the same raw samples (Alves et al., submitted). This emphasises the need for more data

348 on processed seafood samples in order to be able to draw more robust conclusions based on a larger data set of processed samples. On the other hand, Afonso et al. (2015) showed that bioaccessibility 349 350 also influences the risk-benefit evaluation of tuna regarding selenium and MeHg. Recently, Cano-351 Sancho et al. (2015) reported a rather low MeHg bioaccessibility in marine species, concluding that 352 potential health risks for the adult population might be overestimated if bioaccessibility is not 353 considered. Therefore, further monitoring and exposure studies taking into account processing and 354 bioaccessibility are advised, especially for those seafood species identified as high contributors to 355 MeHg exposure.

Inherently associated to exposure assessments are uncertainties that should be considered for the 356 357 interpretation of the current results. Intrinsic factors related with food consumption such as 358 misreporting of consumed foods, and erroneous estimation of consumed quantities (based on 359 portion size) contribute to uncertainty. In addition, the MeHg concentration data file is composed of 360 primary data obtained from the ECsafeSEAFOOD-project, as well as data from available scientific 361 literature. The primary data collected within the project is expected to be of good quality, as quality 362 assurance procedures and validated techniques were employed, and therefore, the uncertainty in 363 the values is considered to be low as a specific sampling plan/framework was followed to collect the 364 samples in the different countries. This primary data was combined with data from literature 365 (different sources), in which for example other analytical methods might have been used (Kroes et 366 al., 2002). Nonetheless, combining the data was essential to have a higher degree of 367 representativeness of contaminant levels in the seafood species addressed. Despite not explicitly considered in the current study, the data included in the exposure model corresponded to levels of 368 369 MeHg from seafood species collected in different seasons and in geographical locations, since both 370 aspects are of relevance for European consumers.

371 **4. Conclusion**

372 Regarding MeHg exposure through seafood consumption, the results of the present study indicate 373 that a country-specific approach is highly relevant in risk management and communication 374 interventions. The largest subgroup of population exposed to potential health risks due to MeHg exposure was identified in Portugal. Taking into account that vulnerable groups may be present 375 376 among this potential risk subgroup, species-specific advice is recommended in order to reduce the 377 health risk. Our results confirm that an excessive consumption of large predatory fish species, such as 378 tuna, should be discouraged, while consumption of hake and cod in Portugal should be moderated. 379 In Spain, although a similar advice should be followed, the consumption of cod is of less concern. 380 Among the southern European countries, the potential risk of MeHg exposure through seafood is the 381 lowest for Italy. However, to decrease the potential risk to MeHg exposure for a certain subpopulation, it should be advised to reduce the consumption of octopus, in addition to that of tuna 382 383 and hake. The finding that sea bream and sea bass, due to the high MeHg levels, are substantial 384 contributors to MeHg exposure in these three southern countries is surprising. Due to the important 385 contribution of these species, further monitoring of the MeHg levels in sea bream and sea bass 386 would be relevant, however, this also applies for other species. For the countries with a lower 387 general seafood consumption frequency, such as Ireland and Belgium, the potential health risks due to MeHg exposure are lower, being for Belgium almost negligible. However, in both countries, 388 389 seafood consumption advice to increase the seafood consumption frequency in order to profit from 390 the health benefits should focus on species low in MeHg content and high in omega-3 LCPUFA levels. 391 The latter applies to all countries and it generally implies the recommendation to consume lower 392 trophic, small, fatty seafood species, such as sardine, Atlantic mackerel, herring and salmon, while 393 minimising the risk of exposure to methylmercury without reducing the benefits of the intake of 394 nutrients (Cardoso et al., 2013; Cardoso et al., 2010; Gribble et al., 2016; Nunes et al., 2014; Strom et 395 al., 2011). Earlier studies in particular European countries indicated that consuming fatty fish more 396 than twice per week can result in an intake of dioxin-like compounds approximating the TWI

(Hellberg et al., 2012; Sioen et al., 2008). Consequently, to balance the potential health benefits and
risks of seafood consumption, small fatty fish are recommended to be consumed instead of large
predatory fish and large white fish, but not to an unlimited extent. The findings and conclusions of
the present study, and their integration in food and health policy should be especially relevant for
vulnerable groups of the population, such as pregnant women, children and women of childbearing
age.

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Tables

Species	Ν		MeH	g (µg/kg ww)
	Analysed	≥ LOD	Mean	Std. Deviation
Alaska pollock	1	1	95.00	0.00
Canned sardine	4	4	45.25	18.46
Canned tuna	13	13	166.65	135.75
Clams	2	2	14.00	2.83
Cod (dry/salted) ^a	0		85.46	71.79
Cod	9	9 🔺	85.46	71.79
Cuttlefish	2	2	104.00	100.41
Haddock	1	1	66.00	0.00
Hake	17	17	123.45	42.02
Herring	3	3	28.67	13.32
Lobster ^b	2	2	121.28	26.23
Mackerel	19	19	80.64	71.32
Monkfish	7	7	227.04	78.62
Mussels	3	3	11.33	6.03
Octopus	6	6	126.47	98.94
Pangasius ^c	1	0	1.33	0.00
Salmon	6	6	20.74	8.40
Sardine	5	5	59.40	25.33
Sea bass	2	2	221.50	183.14
Sea bream	5	5	208.12	167.85
Shrimps and prawns	5	5	53.12	57.67
Sole (and plaice)	17	17	50.21	21.00
Squid	4	4	46.25	27.15
Tuna	12	12	461.98	338.89

Table 1. MeHg contents (mean, standard deviation, µg/kg ww) in the different species analysed.

^a The concentration value of fresh cod is assigned to dry/salted cod due to missing data.

^c Only for pangasius a concentration level < LOQ was measured. For pangasius the concentration <LOQ is replaced by the LOQ value, hence the upper bound (UB) scenario is presented for pangasius in this table.

^b data from total Hg are used

		<u>Belgium</u> (n=540)	<u>Ireland</u> (n=575)	<u>Italy</u> (n=560)	<u>Spain</u> (n=561)	<u>Portugal</u> (n=588)	<u>Total</u> (n=2824)
Gender (%)	Female	49.8	50.4	51.1	49.2	49.5	50.0
	Male	50.2	49.6	48.9	50.8	50.5	50.0
Age (%)	18-24 years	11.5	15.7	10.0	10.3	12.1	11.9
	25-39 years	28.9	36.0	27.3	35.1	35.0	32.5
	40-50 years	22.4	21.4	23.9	21.6	22.6	22.4
	51-60 years	17.4	15.3	18.9	16.9	17.2	17.1
	61-75 years	19.8	11.7	19.8	16.0	13.1	16.0

Table 2. Socio-demographic profile of the sample (n=2824).

Table 3. Result of the exposure assessment for MeHg in the five countries.

MeHg intake (µg/kg bw/week)								
	Mean	Std Dev	P50	P75	P90	P95	P99	% > 1.3 (TWI)
Belgium								
Seafood diet (UB) ^a	0.198	0.217	0.140	0.231	0.374	0.524	1.098	0.7
Ireland				$\langle \rangle$				
Seafood diet	0.360	0.393	0.249	0.424	0.714	1.003	2.019	2.8
<u>Italy</u>			0	7				
Seafood diet	0.546	0.355	0.459	0.685	0.979	1.207	1.785	3.8
Portugal								
Seafood diet	0.796	0.426	0.707	0.997	1.347	1.604	2.220	11.3
<u>Spain</u>								
Seafood diet	0.641	0.365	0.560	0.793	1.086	1.316	1.918	5.2

^a For Belgium, the UB scenario is presented as pangasius is only part of the top 15 most consumed species in Belgium, and only for pangasius a concentration level < LOQ was measured. For pangasius the concentration <LOQ is replaced by the LOQ value.

<u>Belgium</u>	Ireland	Italy	Portugal	<u>Spain</u>
Tuna (40.76)	Tuna (40.04)	Tuna (32.51)	Tuna (26.48)	Tuna (29.53)
Canned tuna (18.34)	Canned tuna (16.16)	Canned tuna (19.59)	Canned tuna (14.83)	Canned tuna (21.17)
Cod fresh (11.02)	Cod fresh (8.29)	Sea bream (11.76)	Sea bream (11.48)	Hake (10.46)
hrimps and prawns (5.86)	Sea bass (6.83)	Sea bass (8.53)	Hake (9.66)	Sea bream (9.28)
Alaska pollock (5.48)	Hake (4.42)	Octopus (5.49)	Sea bass (7.70)	Sea bass (7.49)
Mackerel (3.12)	Haddock (4.28)	Hake (3.74)	Cod dry/salted (7.37)	Octopus (4.88)
Salmon (2.95)	Cod dry/salted (3.49)	Cuttlefish (3.54)	Octopus (5.78)	Cuttlefish (3.58)
Sole (2.58)	Shrimps and prawns (3.45)	Cod fresh (3.39)	Monkfish (4.77)	Shrimps and prawns (2.75
Lobster (2.37)	Mackerel (3.16)	Cod dry/salted (2.80)	Sardine (2.92)	Sardine (2.57)
Sardine (2.16)	Salmon (2.60)	Shrimps and prawns (2.58)	Cuttlefish (2.65)	Squid (2.48)
Canned sardine (1.93)	Sardine (2.46)	Squid (2.00)	Squid (1.92)	Canned sardine (1.87)
Herring (1.36)	Canned sardine (1.66)	Sole (1.83)	Shrimps and prawns (1.76)	Sole (1.77)
Squid (1.28)	Sole (1.50)	Salmon (1.08)	Salmon (1.34)	Salmon (1.09)
Mussels (0.71)	Lobster (1.42)	Clams (0.69)	Canned sardine (1.02)	Mussels (0.57)
Pangasius (0.08)	Mussels (0.24)	Mussels (0.48)	Clams (0.33)	Clams (0.49)

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Table 4. Contribution of the considered species to MeHg exposure through the seafood pattern in the five countries (based on the mean exposure). The shading indicates the five species contributing to the highest extent.

Appendix I

Table 1. Best fit distributions, minimum, maximum and median determined for the consumption (g/week) of the 15 most consumed seafood species in Belgium (fit on the consumption

data > 0).

Species	Variable (fraction consumers)	Function	Min	Max	Median
Salmon	Intake consumers (0.84)	RiskInvgauss(22.724;11.73;RiskTruncate(0.95583;240.04583);RiskShift(-0.51583))	0.44	239.53	11.32
Shrimps and prawns	Intake consumers (0.86)	RiskInvgauss(19.303;12.731;RiskTruncate(1.2798;227.4198);RiskShift(-0.5698))	0.71	226.85	10.68
Cod	Intake consumers (0.80)	RiskLognorm(22.721;46.871;RiskTruncate(0.16341;947.22341);RiskShift(0.28659))	0.45	947.51	10.21
Canned tuna	Intake consumers (0.77)	RiskLognorm(19.974;29.083;RiskTruncate(0.29591;408.56591);RiskShift(-0.23591))	0.06	408.33	11.07
Mussels	Intake consumers (0.80)	RiskInvgauss(12.232;12.784;RiskTruncate(1.23568;91.80567);RiskShift(-0.89568))	0.34	90.91	7.50
Tuna	Intake consumers (0.71)	RiskPearson5(1.6907;15.157;RiskTruncate(2.5748;478.6048);RiskShift(-2.2148))	0.36	476.39	8.95
Sole	Intake consumers (0.69)	RiskPearson5(1.8126;11.213;RiskTruncate(1.7125;93.3225);RiskShift(-1.4225))	0.29	91.90	6.06
Alaska pollock	Intake consumers (0.55)	RiskLognorm(15.716;24.963;RiskTruncate(0.25242;250.19242);RiskShift(-0.19242))	0.06	250.00	8.18
Pangasius	Intake consumers (0.55)	RiskInvgauss(16.75;9.0888;RiskTruncate(1.13685;167.46685);RiskShift(-0.79685))	0.34	166.67	8.22
Herring	Intake consumers (0.55)	RiskLognorm(12.608;23.121;RiskTruncate(0.15351;239.39351);RiskShift(0.13649))	0.29	239.53	6.17
Mackerel	Intake consumers (0.51)	RiskInvgauss(11.917;5.9171;RiskTruncate(0.45494;127.14494);RiskShift(-0.29494))	0.16	126.85	5.80
Squid	Intake consumers (0.51)	RiskLoglogistic(0.24353;4.5708;1.529;RiskTruncate(0.29;68.85))	0.29	68.85	4.73
Lobster	Intake consumers (0.51)	RiskInvgauss(5,601;3,6832;RiskTruncate(0,254089;98,384089);RiskShift(-0,094089))	0.16	98.29	3.16
Canned sardine	Intake consumers (0.50)	RiskInvgauss(12.431;6.29;RiskTruncate(0.55558;215.67558);RiskShift(-0.22558))	0.33	215.45	6.21
Sardine	Intake consumers (0.50)	RiskInvgauss(10.636;5.9735;RiskTruncate(0.60303;91.53303);RiskShift(-0.24303))	0.36	91.29	5.52

Table 2. Best fit distributions, minimum, maximum and median determined for the consumption (g/week) of the 15 most consumed seafood species in Ireland (fit on the consumption data > 0).

Species	Variable (fraction consumers)	Function		Min	Max	Median
Salmon	Intake consumers (0.79)	RiskInvgauss(39.722;19.859;Ris	kTruncate(2.211;453.161);RiskShift(-1.841))	0.37	451.32	18.66
Cod	Intake consumers (0.82)	RiskInvgauss(33.678;19.065;Ris	kTruncate(2.1142;351.8542);RiskShift(-1.8542))	0.26	350.00	16.55
Canned tuna	Intake consumers (0.70)	RiskInvgauss(37.408;16.614;Ris	kTruncate(2.0238;512.0738);RiskShift(-1.6538))	0.37	510.42	16.64
Haddock	Intake consumers (0.74)	RiskGamma(0.91177;25.344;Ri	skTruncate(;175);RiskShift(0.15))	0.15	175.00	15.54
Shrimps and prawns	Intake consumers (0.69)	RiskLognorm(28.728;54.734;Ris	kTruncate(0.34758;257.02758);RiskShift(-0.28758))	0.06	256.74	12.92
Tuna	Intake consumers (0.68)	RiskLognorm(36.562;86.595;Ris	kTruncate(0.21396;674.37396);RiskShift(-0.01396))	0.20	674.36	14.17
Mackerel	Intake consumers (0.64)	RiskInvgauss(18.167;7.5802;Ris	kTruncate(0.68891;173.46891);RiskShift(-0.62891))	0.06	172.84	7.86
Sardine	Intake consumers (0.58)	RiskInvgauss(19.596;7.689;Risk	Truncate(0.77411;209.98411);RiskShift(-0.58411))	0.19	209.40	8.31
Sea bass	Intake consumers (0.60)	RiskLognorm(14.961;29.051;Ris	kTruncate(0.122;154.922);RiskShift(0.13805))	0.26	155.06	6.93
Hake	Intake consumers (0.57)	RiskInvgauss(17.382;7.0422;Ris	kTruncate(0.666;255.816);RiskShift(-0.60613))	0.06	255.21	7.45
Sole	Intake consumers (0.58)	RiskInvgauss(14.056;6.2994;Ris	kTruncate (0.68949; 138.35949); Risk Shift (-0.35949))	0.33	138.00	6.49
Canned sardine	Intake consumers (0.55)	RiskInvgauss(18.513;6.4035;Ris	kTruncate(0.76942;279.25942);RiskShift(-0.39942))	0.37	278.86	7.51
Cod dry/salted	Intake consumers (0.53)	RiskInvgauss(23.878;5.8664;Ris	kTruncate(0.641;233.711);RiskShift(-0.38106))	0.26	233.33	7.76
Mussels	Intake consumers (0.51)	RiskInvgauss(12,025;6,3474;Ris	kTruncate(0,50353;97,66353);RiskShift(-0,44353))	0.06	97.22	5.85
Lobster	Intake consumers (0.51)	RiskLoglogistic(0,22282;3,3542	:1,5968;RiskTruncate(0,26;84,7))	0.26	84.7	3.56

Table 3. Best fit distributions, minimum, maximum and median determined for the consumption (g/week) of the 15 most consumed seafood species in Italy (fit on the consumption data > 0).

Species	Variable (fraction consumers)	Function		Min	Max	Median
Canned tuna	Intake consumers (0.95)	RiskInvgauss(48.412;41.508;	RiskTruncate(4.4281;616.6081);RiskShift(-4.1081))	0.32	612.50	27.15
Tuna	Intake consumers (0.85)	RiskGamma(0.80043;38.127	RiskTruncate(;448.71);RiskShift(0.24))	0.24	448.71	19.37
Shrimps and prawns	Intake consumers (0.90)	RiskExpon(20.184;RiskTrunc	ate(0.039968;236.179968);RiskShift(0.050032))	0.09	236.23	14.08
Clams	Intake consumers (0.89)	RiskExpon(20.459;RiskTrunc	ate(0.04125;161.37125);RiskShift(0.12875))	0.17	161.50	14.34
Salmon	Intake consumers (0.88)	RiskGamma(0.99039;19.599	;RiskTruncate(;170.93);RiskShift(0.08))	0.08	170.93	13.48
Squid	Intake consumers (0.89)	RiskExpon(17.898;RiskTrunc	ate(0.035725;154.615725);RiskShift(0.094275))	0.13	154.71	12.53
Mussels	Intake consumers (0.90)	RiskGamma(0.96087;18.243	RiskTruncate(;161.49);RiskShift(0.17))	0.17	161.49	12.12
Sea bream	Intake consumers (0.86)	RiskGamma(0.85325;22.937	;RiskTruncate(;271.12);RiskShift(0.24))	0.24	271.36	12.90
Octopus	Intake consumers (0.85)	RiskExpon(15.157;RiskTrunc	ate(0.03171;128.03171);RiskShift(0.23829))	0.27	128.27	10.77
Sea bass	Intake consumers (0.82)	RiskGamma(0.8343;20.715;F	RiskTruncate(;139.66);RiskShift(0.34))	0.34	140.00	11.39
Sole	Intake consumers (0.82)	RiskInvgauss(17.201;10.182;	RiskTruncate(1.14429;140.84429);RiskShift(-0.84429))	0.30	140.00	8.71
Cuttlefish	Intake consumers (0.80)	RiskInvgauss(17.367;10.979;	RiskTruncate(1.2633;161.7533);RiskShift(-1.1733))	0.09	160.58	8.79
Cod	Intake consumers (0.77)	RiskGamma(0.75897;24.622	RiskTruncate(;140);RiskShift(0.33))	0.33	140.00	11.69
Cod dry/salted	Intake consumers (0.78)	RiskInvgauss(16,211;8,6989;	RiskTruncate(0,99471;265,46471);RiskShift(-0,80471))	0.19	264.66	7.87
Hake	Intake consumers (0.71)	RiskInvgauss(16,715;9,2571;	RiskTruncate(1,06801;250,14801);RiskShift(-0,87801))	0.19	249.27	8.20

Table 4. Best fit distributions, minimum, maximum and median determined for the consumption (g/week) of the 15 most consumed seafood species in Portugal (fit on the consumption data > 0).

Species	Variable (fraction consumers)	Function	Min	Max	Median
Canned tuna	Intake consumers (0.92)	RiskInvgauss(60.15;79.898;RiskTruncate(9.04;399.35);RiskShift(-8.7685))	0.27	390.58	35.74
Cod dry/salted	Intake consumers (0.95)	RiskExpon(48.888;RiskTruncate(0.08777;487.79777);RiskShift(0.42223))	0.51	488.22	34.39
Hake	Intake consumers (0.90)	RiskExpon(47.264;RiskTruncate(0.08935;426.80935);RiskShift(0.11065))	0.20	426.92	32.96
Salmon	Intake consumers (0.84)	RiskExpon(37.125;RiskTruncate(0.074849;342.644849);RiskShift(0.015151))	0.09	342.66	25.82
Tuna	Intake consumers (0.79)	RiskExpon(39.696;RiskTruncate(0.085;287.735);RiskShift(0.195))	0.28	287.93	27.77
Sardine	Intake consumers (0.85)	RiskGamma(0.90182;33.632;RiskTruncate(;331.81);RiskShift(0.09))	0.09	331.90	20.22
Squid	Intake consumers (0.85)	RiskInvgauss(29.729;28.094;RiskTruncate(3.2158;316.1058);RiskShift(-3.1458))	0.07	312.96	16.75
Sea bream	Intake consumers (0.85)	RiskExpon(28.965;RiskTruncate(0.05793;289.90793);RiskShift(0.03207))	0.09	289.94	20.17
Octopus	Intake consumers (0.86)	RiskGamma(1.0169;23.38;RiskTruncate(0.000835;180.890835);RiskShift(0.019165))	0.02	180.91	16.60
Shrimps and prawns	Intake consumers (0.89)	RiskExpon(20.838;RiskTruncate(0.039995;201.709995);RiskShift(0.050005))	0.09	201.76	14.53
Sea bass	Intake consumers (0.77)	RiskInvgauss(26.907;14.677;RiskTruncate(1.8803;322.6203);RiskShift(-1.7903))	0.09	320.83	12.77
Clams	Intake consumers (0.80)	RiskGamma(1.0252;15.656;RiskTruncate(0.00094;103.78094);RiskShift(0.11906))	0.12	103.90	11.33
Cuttlefish	Intake consumers (0.73)	RiskExpon(19.11;RiskTruncate(0.045;139.365);RiskShift(0.12545))	0.17	139.49	13.40
Monkfish	Intake consumers (0.71)	RiskInvgauss(16,271;9,9532;RiskTruncate(1,01388;234,86388);RiskShift(-0,92388))	0.09	233.94	8.27
Canned sardine	Intake consumers (0.64)	RiskInvgauss(21.75;10.476;RiskTruncate(1.12711;172.28711);RiskShift(-0.95711))	0.17	171.33	9.97

RiskInvgauss(21.75;10.470

Table 5. Best fit distributions, minimum, maximum and median determined for the consumption (g/week) of the 15 most consumed seafood species in Spain (fit on the consumption data > 0).

Species	Variable (fraction consumers)	Function	2	Min	Max	Median
Canned tuna	Intake consumers (0.95)	RiskInvgauss(65.533;82.034;RiskTruncat	e(8.288;503.528);RiskShift(-8.128))	0.16	495.40	39.41
Hake	Intake consumers (0.94)	RiskExpon(39.219;RiskTruncate(0.07414	;336.59414);RiskShift(0.28586))	0.36	336.88	27.54
Tuna	Intake consumers (0.92)	Risk Expon (30.572; Risk Truncate (0.05902	;299.67902);RiskShift(0.15098))	0.21	299.83	21.40
Squid	Intake consumers (0.95)	RiskGamma(1.2051;20.669;RiskTruncate	(0.02236;192.26236);RiskShift(0.23764))	0.26	192.50	18.70
Mussels	Intake consumers (0.91)	RiskGamma(1.0641;22.988;RiskTruncate	e(0.0041;172.0341);RiskShift(0.3559))	0.36	172.39	17.71
Shrimps and prawns	Intake consumers (0.95)	RiskExpon(24.298;RiskTruncate(0.04559	;180.76559);RiskShift(0.21441))	0.26	180.98	17.09
Salmon	Intake consumers (0.91)	RiskExpon(22.386;RiskTruncate(0.04407	;259.91407);RiskShift(0.22593))	0.27	260.14	15.79
Sardine	Intake consumers (0.90)	RiskGamma(1.0584;19.028;RiskTruncate	e(0.0057;142.0857);RiskShift(0.3543))	0.36	142.44	14.61
Canned sardine	Intake consumers (0.83)	RiskExpon(21.79;RiskTruncate(0.04666;	207.82666);RiskShift(0.28334))	0.33	208.11	15.43
Clams	Intake consumers (0.88)	RiskInvgauss(19.609;18.459;RiskTruncat	e(1.8438;336.7938);RiskShift(-1.7938))	0.05	335.00	11.26
Sole	Intake consumers (0.89)	RiskGamma(1.0417;15.823;RiskTruncate	e(0.00161;123.79161);RiskShift(0.20839))	0.21	124.00	11.81
Cuttlefish	Intake consumers (0.87)	RiskExpon(17.636;RiskTruncate(0.03628	8;127.346288);RiskShift(0.013712))	0.05	127.36	12.26
Octopus	Intake consumers (0.89)	RiskInvgauss(16.836;12.798;RiskTruncat	e(1.2567;150.8567);RiskShift(-1.0467))	0.21	149.81	9.32
Sea bream	Intake consumers (0.86)	RiskInvgauss(19.533;10.34;RiskTruncate	(1.388;484.488);RiskShift(-1.038))	0.35	483.45	9.43
Sea bass	Intake consumers (0.83)	RiskExpon(17.78;RiskTruncate(0.03824;	288.95824);RiskShift(0.32176))	0.36	289.28	12.68

Country	Variable	Function	Min	Max	Mean	Median
Belgium	Weight (kg)	RiskWeibull(2.5004;42.274;RiskTruncate(1.522;105.522);RiskShift(38.478))	40	144	75.99	74.99
Ireland	Weight (kg)	RiskExtvalue(69.203;17.219;RiskTruncate(40;150))	40	150	78.51	75.39
Italy	Weight (kg)	RiskWeibull(2.4156;38.056;RiskTruncate(2.607;92.607);RiskShift(37.393))	40	130	71.17	70.12
Portugal	Weight (kg)	RiskLoglogistic(-35.559;105.93;13.437;RiskTruncate(40;150))	40	150	71.68	70.53
Spain	Weight (kg)	RiskGamma(7.6996;5.5585;RiskTruncate(10.5061;102.5061);RiskShift(29.4939))	40	132	72.22	70.45
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Table 6. Best fit distributions, minimum, maximum, mean and median of the best fitted distribution for body weight (kg) for each country.

Appendix II

Table 1. Best fit distributions, minimum, maximum and mean determined for MeHg contents in different species (µg/kg ww).

Species	Variable	Function	Min	Max	Mean
Canned tuna	MeHg LB and UB	RiskPearson5(3.6945;594.75;RiskTruncate(79.89;1129.727);RiskShift(-49.727))	30.16	1080.00	172.45
Cod (and dry/salted cod)	MeHg LB and UB	RiskInvgauss(76.734;108.55;RiskTruncate(15.2215;525.2715);RiskShift(8.7285))	23.95	534.00	86.82
Hake	MeHg LB and UB	RiskGamma(5.41;17.615;RiskTruncate(27.847;404.067);RiskShift(28.153))	56.00	432.22	124.42
Mackerel	MeHg LB and UB	RiskInvgauss(61.073;29.377;RiskTruncate(4.821;614.431);RiskShift(19.569))	24.39	634.00	79.36
Monkfish	MeHg LB and UB	RiskNormal(227.043;78.624;RiskTruncate(127.53;719.8))	127.53	719.80	242.74
Octopus	MeHg LB and UB	RiskExtvalueMin(172.217;84.1765;RiskTruncate(11;532.14))	11.00	532.14	155.36
Salmon	MeHg LB and UB	RiskExtvalueMin(24.6019;6.9599;RiskTruncate(12.17;64))	12.17	64.00	23.44
Sardine	MeHg LB and UB	RiskInvgauss(110.32;2470.76;RiskTruncate(78.916;230.916);RiskShift(-50.916))	28.00	180.00	62.12
Sea bream	MeHg LB and UB	RiskNormal(208.12;167.85;RiskTruncate(41.58;880))	41.58	880.00	256.86
Shrimps and prawns	MeHg LB and UB	RiskExpon(44.515;RiskTruncate(8.90743;274.29743);RiskShift(-0.29743))	8.61	274.00	52.44
Sole	MeHg LB and UB	RiskNormal (50.211; 20.996; Risk Truncate (17.09; 170))	17.09	170.00	52.77

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