



# Mercury Exposure Assessment from the First Harmonised Total Diet Study in Portugal

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## Abstract

The aim of this study was to estimate the Portuguese population's baseline exposure to methyl and inorganic mercury by a harmonised total diet study (TDS) methodology and the risk of exceeding the Tolerable Weekly Intake (TWI). TDS food samples representative of the whole diet of the population were prepared as consumed, and analysed for total mercury. European Food Safety Authority's (EFSA) conservative approach was used to estimate methylmercury and inorganic mercury and exposure was estimated using Monte Carlo Risk Assessment (MCRA) software. Mean, median and P95 exposure of the overall population (18 to 74 years old) to methylmercury and to inorganic mercury were 1.25, 0.01 and 5.45  $\mu\text{g}/\text{kg}$  bw/week, and 0.37, 0.15 and 1.27  $\mu\text{g}/\text{kg}$  bw/week, respectively. The percentage of individuals exceeding TWI was 27.6 for methylmercury and 3.5 for inorganic mercury. Regarding childbearing age women (18 to 45 years old), methylmercury mean exposure was 1.13  $\mu\text{g}/\text{kg}$  bw/week with 25% of women exceeding the TWI. Cod and hake were the main contributors to mercury intake.

**Keywords** Total Diet Study · Methylmercury exposure · Inorganic mercury exposure · Monte Carlo Risk Assessment · FoodEx2 Classification System · Whole diet

## Introduction

Mercury, a naturally occurring metal found in the Earth's crust, exists in various chemical forms including elemental mercury, inorganic mercury compounds, and organic mercury compounds, the most notable of which is methylmercury (World Health Organization (WHO), 2011). Methylmercury, formed by the microbial conversion of inorganic mercury in aquatic systems, is the primary concern when assessing the risks of mercury exposure, as it accumulates in the food chain and bioaccumulates in the tissues of fish and other aquatic organisms (Clarkson 2002). Exposure to mercury occurs through multiple routes, including inhalation of elemental mercury vapor, ingestion of contaminated food and water, and dermal contact with mercury-containing products, with the consumption of fish and seafood being a major source of exposure for the population (European Food Safety Authority [EFSA], 2012). Apart from dietary intake, other potential sources of mercury exposure include

dental amalgam fillings, occupational settings (e.g., certain industrial processes), and ambient air pollution from natural and anthropogenic sources (Clarkson 2002) (EFSA 2012). Adverse effects of mercury on human health are well-documented. Chronic exposure to methylmercury, primarily through fish consumption, can lead to neurodevelopmental deficits in children, including cognitive impairments, learning disabilities, and delays in motor skills. In adults, exposure to high levels of mercury can result in neurological symptoms, cardiovascular effects, and renal dysfunction (EFSA 2012). To protect human health from exposure to chemical substances, international entities like the Joint FAO/WHO Expert Committee on Food Additives (JECFA) and European Food Safety Authority (EFSA) established health-based guidance values, such as the Tolerable Weekly Intake (TWI) for methyl and inorganic mercury (EFSA 2012; Joint FAO/WHO Expert Committee on Food Additives [JECFA], 2004).

Portugal's unique cultural and culinary traditions place a substantial emphasis on seafood and fish consumption. In 2021, Portugal ranked sixth globally in terms of average fish and seafood consumption per capita, with 59.41 kg consumed annually, based on FAO data cited by Our World in Data (Our World in Data 2021). This high consumption

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makes the Portuguese population particularly exposed to mercury and underscores the need to perform exposure and risk assessment studies that provide scientific evidence to support adequate measures for public health protection.

A Total Diet Study (TDS) is a valuable public health tool for assessing the exposure of a population to various chemical substances such as contaminants and nutrients, reflecting real dietary habits. These studies provide a comprehensive view of dietary exposure by analyzing food items as typically consumed. They are essential for designing effective strategies to manage and reduce exposure (Vasco et al. 2021). Initiatives to harmonise TDS methodologies contribute to a more standardised assessment of dietary exposure and improves comparability across countries (EFSA, Food and Agriculture Organization of the United Nations [FAO], WHO 2011; Pité et al. 2018). In summary, mercury exposure might be a significant concern for the Portuguese population, particularly through seafood consumption, and its impact on human health underscores the importance of assessing and managing exposure. The main objective of the present study was to assess the baseline chronic dietary exposure of the Portuguese population to methylmercury and inorganic mercury using a harmonised TDS methodology and to estimate the population's risk of exceeding the health-based guidance values.

## Material and Methods

The first Portuguese TDS described by Vasco (Vasco et al. 2021) was conducted by the Food and Nutrition Department of the National Institute of Health Doctor Ricardo Jorge, IP (INSA) using the harmonised TDS methodology developed during the TDS-Exposure project (Dofkova et al. 2016; Pité et al. 2018).

### Food Consumption Data and Food Sampling

The consumption survey characteristics, the definition and preparation of TDS samples and the collection of food items was described in detail by Vasco (Vasco et al. 2021). Briefly, based on the food consumption survey results (Poínhos et al. 2009) a core food list containing 1072 food items, representative of the general Portuguese population diet (male and female from 18 to 93 years old,  $n=3529$ ), was classified and mapped by the exposure hierarchy of the FoodEx2 food classification and description system (EFSA 2011) into 20 food groups. For 'adults and elderly' (18 to 74 years old) 'male and female', from now on named 'overall population' ( $n=3272$ ), a TDS food list was compiled. This list included 528 food items, covering 95% of the whole diet. These food items were selected based on two criteria applied at food group level: firstly, they had to be the most consumed foods

contributing to at least 90% of the general food consumption, and secondly, they were foods known or suspected to be the primary sources of exposure or intake to one or more substances of interest (Vasco et al. 2021). The foods items were aggregated and resulted in 164 different TDS samples. Each TDS sample was classified by FoodEx2 and was composed by twelve subsamples. Potential seasonal and regional variations in chemical occurrence were also considered resulting in 227 TDS samples (2724 subsamples). Foods were collected between April 2014 and March 2016 in the selected shops of the Greater Lisbon region (hyper-or supermarket, discount, local markets for fish, fruits and vegetables, butchers, bakeries, pastries, restaurants and takeaway) selected according to market information. The food items were prepared "as consumed" by the population, respecting the most common food preparation and cooking habits (e.g. washing, peeling, removing inedible parts like bones, scales, shells, seeds, and boiling, grilling, roasting, baking, frying). The edible part of the subsamples were homogenised, pooled into the respective TDS samples that were further aliquoted and stored at  $-20\text{ }^{\circ}\text{C}$ , until analysis (Vasco et al. 2021).

### Mercury Determination

The determination of total mercury occurrence in TDS samples was performed by a ISO/IEC 17025 accredited method using a direct mercury analyser DMA-80 (Milestone Inc., Shelton, CT, USA), based on the principle of thermal decomposition, amalgamation and atomic spectrometric detection (254 nm). Slight modifications were made, to the analytical DMA-80 programme concerning the temperatures and times, to optimize for the different food matrices (Martins et al. 2013). Two calibration curves with working ranges between 2.5 and 30 ng and 100 and 500 ng were used. The limits of detection (LOD) and quantification (LOQ) were performed on 10 blanks analysed at different days (LOD:  $3 \times \text{SD}$  and LOQ:  $10 \times \text{SD}$  of the sample boat blanks) (EN 13804). LOD ranged between 0.07 and 4.6  $\mu\text{g}/\text{kg}$  and LOQ between 0.2 and 13.8  $\mu\text{g}/\text{kg}$ . About 0.2–0.3 g (exceptions: popcorn 0.02 g; fruit salad 0.83 g) of each TDS sample was directly weighed into the pre-cleaned sample boat and placed into the auto-sampler. The results were expressed as micrograms per kilogram ( $\mu\text{g}/\text{kg}$ ). The measurement precision expressed by relative standard deviation was below 0.70% for repeatability (RSDr) and 1.16% for intermediate precision (RSDR). The participation in Proficiency Test Schemes launched by Accredited Proficiency Testing Providers (Fapas®, York, UK) and the analysis of the certified reference material (MRC) NIST 1566b Oyster tissue were used to evaluate the laboratory Performance ( $z$ -scores between  $-2.9$  and  $1.2$ ). The relative uncertainty of the results was 5%.

## Occurrence, Exposure Assessment and Risk Characterization

Occurrence results reported below limit of detection and/or limit of quantification (left-censored data) were managed using the upper bound (UB) and lower bound (LB) substitution methods (EFSA 2010; GEMS/Food-EURO 1995). In the UB approach the non-detected results were set to the LOD value and the results between the limit of detection and quantification were set to the LOQ value. In the LB approach the non-detected results were set to zero and results between the limit of detection and quantification were set to the LOD value. For the TDS samples collected in four seasons, the mean occurrence value was used for exposure estimation after applying the left-censored substitution method. For total mercury occurrence data, minimum (min), maximum (max) mean and median values were calculated for each FoodEx2 group. To estimate methylmercury level from total mercury occurrence values, EFSA's conservative approach was used assuming 100% methylmercury in fish and 80% in seafood other than fish. Inorganic mercury was estimated as 20% of total mercury in fish as well as, 50% in seafood other than fish. In other foods, mercury was considered to be 100% inorganic (EFSA 2012). A semiprobabilistic approach was used to perform methylmercury and inorganic mercury dietary exposure assessment by combining the estimated UB and LB occurrence data with the corresponding consumption recorded in the food consumption survey. Occurrence and consumption data were linked by coding both with the FoodEx2 food classification and description system. The exposure calculation was carried out with the Monte Carlo Risk Assessment (MCRA) software using the observed individual mean (OIM) model (Boon et al. 2018). Exposure was estimated for food as measured (TDS sample) and for food as eaten (TDS subsamples) and was expressed as mean, median (P50) and 95th percentile (P95), representing the average and highly exposed population. Exposure estimates, expressed in  $\mu\text{g}/\text{kg}$  bw/week, were conducted for the 'overall population' and segmented by age group ('adults and elderly', 18–74 years old; 'adults', 18–64 years old; 'elderly', 65–74 years old) for both males and females, and separately for males and females. Exposure to methylmercury was also estimated for the sub-population of 18 to 45 years old childbearing age women (905 individuals). A deterministic approach was used to evaluate exposure of the 'overall population' to inorganic mercury through the consumption of tap water. Mean tap water consumption was combined with the average maximum total mercury occurrence value (2018 to 2022) obtained from the water quality legal control of the Greater Lisbon municipalities. For risk characterization, estimated exposure values of methylmercury and inorganic mercury were compared with tolerable weekly intakes (TWI) of 1.3 and 4  $\mu\text{g}/\text{kg}$  of body weight

(bw), respectively, the most recent health based guidance values (HBGV) set by EFSA (EFSA 2012). The percentages of individuals that exceeded the TWI were also estimated. The contribution of each food item to the total exposure was estimated and the main contributors were highlighted.

## Results and Discussion

### Mercury

Total mercury occurrence levels below the LOD and LOQ represented 77% of the analysed samples, however all TDS samples composed by fish and/or seafood showed occurrence values above LOQ. Data on total mercury occurrence summarised by food group is presented in Table 1. The 'fish, seafood, amphibians, reptiles and invertebrates' food group showed the highest total mercury mean occurrence value (0.136 mg/kg) and included the TDS sample with the highest value (0.574 mg/kg, scabbardfish). Other food groups had mean occurrence values equal or below 0.0195 mg/kg.

### Methylmercury

Negligible differences were found for methylmercury estimates calculated using the UB and LB occurrence values, therefore only the UB estimates are reported.

The estimated dietary exposure of Portuguese 'overall population' to methylmercury with the detailed contribution of each TDS sample is presented in Table 2. The corresponding data for 'male' and 'female' population groups are presented in Tables 3 and 4, respectively. The 'overall population' revealed a mean weekly exposure to methylmercury of 1.25  $\mu\text{g}/\text{kg}$  bw/week, being lower for 'adults' (1.20  $\mu\text{g}/\text{kg}$  bw/week) and higher for 'elderly' (1.48  $\mu\text{g}/\text{kg}$  bw/week) (Table 2). 'Elderly' males showed the highest mean methylmercury exposure (1.77  $\mu\text{g}/\text{kg}$  bw/week) whereas male 'adults' showed the lowest value (1.14  $\mu\text{g}/\text{kg}$  bw/week) of all population groups studied (Table 3). Females did not reveal differences in exposure according to age (1.26  $\mu\text{g}/\text{kg}$  bw/week 'adults' and 1.27  $\mu\text{g}/\text{kg}$  bw/week 'elderly') (Table 4). The median weekly exposure (P50) ranged from 0 to 0.25  $\mu\text{g}/\text{kg}$  bw/week for all sex and age groups, with the highest value observed in male elderly individuals (Table 2, 3, and 4). At the 95th percentile, weekly methylmercury exposure ranged from 4.97 to 5.68  $\mu\text{g}/\text{kg}$  bw/week for all sex and age groups except for male elderly individuals, who showed a weekly exposure of 7.29  $\mu\text{g}/\text{kg}$  bw/week (Table 2, 3, and 4).

The mean methylmercury exposure for the 'overall population' reached 96% of the TWI of 1.3  $\mu\text{g}/\text{kg}$  bw/week. However, for elderly males, this exposure surpassed the TWI at 136%. At the 95th percentile, estimated exposures

**Table 1** Total mercury content (minimum, maximum, mean and median) of TDS samples expressed by FoodEx2 food group

FoodEx2 Food Group Nr	FoodEx2 level 1 food groups	Total Nr of food items	Nr of food items selected for the TDS food list	Nr of TDS Samples	Nr of TDS sam- ples ≤ LOD		Nr of TDS sam- ples > LOD and ≤ LOQ		Nr of TDS sam- ples > LOQ		Total mercury (mg/kg)		Mean		Median					
					LB	UB	LB	UB	LB	UB	LB	UB	LB	UB	LB	UB	LB	UB	LB	UB
2	Alcoholic beverages	28	7	2	2	2	0	0	0	0	0	0	0	0	0	0	0			
3	Animal and vegetable fats and oils	7	3	2	2	0	0	0	0	0	0	0	0	0	0	0	0			
4	Coffee, cocoa and tea and infusions	12	9	4	4	0	0	0	0	0	0	0	0	0	0	0	0			
5	Composite dishes	360	191	34	16	16	7	7	11	11	0	0	0.45	0.0191	0.0195	0.0003	0.0009			
6	Eggs and egg product	2	1	1	1	0	0	0	0	0	0	0.00047	0	0.00047	0	0.00047	0			
7	Fish, seafood, amphibians, reptiles and invertebrates	68	41	25	0	0	0	0	25	25	0.0029	0.0029	0.574	0.136	0.136	0.0984	0.0984			
9	Fruit and fruit products	65	15	14	11	11	3	3	0	0	0	0.00017	0.0006	0.0018	0.0001	0.0006	0	0.00034		
10	Fruit and veg- etable juices and nectars	43	38	2	2	2	0	0	0	0	0	0.0008	0	0.0016	0	0.0012	0	0.0012		
11	Grains and grain-based products	158	67	21	10	10	11	11	0	0	0	0.00033	0.00120	0.0046	0.0003	0.0014	0.0003	0.0015		
12	Legumes, nuts, oilseeds and spices	21	11	8	5	5	3	3	0	0	0	0.00027	0.00047	0.0014	0.00013	0.00063	0	0.0005		
13	Meat and meat products	68	49	11	6	6	5	5	0	0	0	0.00057	0.00067	0.002	0.00025	0.0011	0	0.00067		
14	Milk and dairy products	89	30	6	4	4	2	2	0	0	0	0.0004	0.00053	0.0016	0.00016	0.0008	0	0.00058		

Table 1 (continued)

FoodEx2 Food Group Nr	FoodEx2 level 1 food groups	Total Nr of food items	Nr of food items selected for the TDS food list	Nr of TDS Samples	Nr of TDS sam- ples ≤ LOD		Nr of TDS sam- ples > LOD and ≤ LOQ		Nr of TDS sam- ples > LOQ		Total mercury (mg/kg)													
					LB	UB	LB	UB	LB	UB	Minimum		Maximum		Mean		Median							
					LB	UB	LB	UB	LB	UB	LB	UB	LB	UB	LB	UB	LB	UB	LB	UB				
15	Products for non-standard diets, food imitates and food sup- plements or fortifying agents	9	4	2	0	0	2	2	0	0	0	0	0.00037	0.0011	0.00037	0.0011	0.00037	0.0011	0.00037	0.0011	0.00037	0.0011	0.00037	0.0011
16	Seasoning, sauces and condiments	22	14	5	1	1	4	4	0	0	0	0	0.0004	0.0012	0.000274	0.0008	0.0004	0.0004	0.0012	0.0004	0.0012	0.0004	0.0012	
17	Starchy roots or tubers and products thereof, sugar plants	7	1	1	1	0	0	1	0	0	0	0	0.0015	0	0.0015	0	0.0015	0	0.0015	0	0.0015	0	0.0015	
18	Sugar, confec- tionery and water-based sweet des- serts	23	12	3	0	0	3	3	0	0	0	0.00033	0.001	0.00067	0.002	0.0005	0.0015	0.00047	0.0014	0.00047	0.0014	0.00047	0.0014	
19	Vegetables and vegetable products	46	22	19	14	14	4	4	1	1	0	0.00023	0.0027	0.0027	0.0002	0.0006	0	0.00037	0.00067	0.0002	0.0006	0	0.00037	
20	Water and water-based beverages	40	13	4	4	4	0	0	0	0	0	0	0	0	0	0.0000	0	0.0000	0	0.0000	0	0.0000	0	0.0000
Total	%	1068	528	164	83	82	44	45	37	37	-	-	-	-	-	-	-	-	-	-	-	-	-	-
					51	50	27	27	23	23	-	-	-	-	-	-	-	-	-	-	-	-	-	-

TDS Total Diet Study, LOD Limit of Detection, LOQ Limit of Quantification, LB Lower Bound Approach, UB Upper Bound Approach

**Table 2** Estimated dietary exposure of Portuguese overall population ('male and female') to methylmercury

FoodEx2 Food Group Nr	FoodEx2 Level 1 Food Group	TDS Sample Nr	Sex	Male and Female		Nr Individuals	('overall population')		18-64 ('adults')		65-74 ('elderly')		µg/kg bw/week		Contribution (%)		µg/kg bw/week		
				18-74	18-64		18-64	65-74	Contribution (%)	Mean	Median	P95	Contribution (%)	Mean	Median	P95	Contribution (%)	Mean	Median
5	Composite dishes	12		0.00	0.00	3272	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
5	Beans and meat meal (Beans and gut dish)	13		0.00	0.00		0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
5	Beans, meat, and vegetables meal (Meat and vegetables boiled dish, Portuguese style)	14		0.00	0.00		0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
5	Beans, meat, and vegetables meal (Beans stewed with pork and cow meat)	15		0.00	0.00		0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
5	Beans, meat, and vegetables meal (Pork meat, chickpea, pasta and vegetables stewed)	9		0.38	0.34		0.0047	0.0000	0.0000	0.0040	0.0000	0.0000	0.0000	0.0082	0.0000	0.0000	0.0000	0.0000	0.0000
5	Dishes, incl. Ready to eat meals (excluding soups and salads) (Bread based dish)	30		0.05	0.06		0.0006	0.0000	0.0000	0.0007	0.0000	0.0045	0.02	0.0002	0.0000	0.0000	0.0000	0.0000	0.0000

Table 2 (continued)

FoodEx2 Food Group Nr	FoodEx2 Level 1 Food Group	TDS Sample Nr	Sex	Male and Female		Contribution (%)	µg/kg bw/week	Mean	Median	P95	Contribution (%)	µg/kg bw/week	Mean	Median	P95			
				Age group	Age group													
				18–74	18–64	65–74												
				('overall population')														
			Nr Individuals	3272	2752	520												
			TDS Sample	Contribution (%)	Contribution (%)	Contribution (%)	µg/kg bw/week	Mean	Median	P95	Contribution (%)	µg/kg bw/week	Mean	Median	P95			
5		22	Fish and potatoes meal (Cod fish dishes)	1.57	0.0195	0.0000	0.0000	0.0000	1.70	0.0205	0.0000	0.0000	0.0000	0.0000	0.98	0.0146	0.0000	0.0000
5		23	Fish and potatoes meal (Mixed fish stew)	11.64	0.1453	0.0000	0.0000	0.0000	10.50	0.1264	0.0000	0.0000	0.0000	0.0000	16.57	0.2456	0.0000	0.0000
5		24	Fish and potatoes meal (Potato puree with fish)	0.89	0.0111	0.0000	0.0000	0.0000	1.00	0.0120	0.0000	0.0000	0.0000	0.0000	0.40	0.0059	0.0000	0.0000
5		25	Fish and rice meal	1.85	0.0231	0.0000	0.0000	0.0000	1.93	0.0233	0.0000	0.0000	0.0000	0.0000	1.49	0.0221	0.0000	0.0000
5		19	Fish and seafood based dishes	0.77	0.0097	0.0000	0.0000	0.0000	0.90	0.0109	0.0000	0.0000	0.0000	0.0000	0.22	0.0033	0.0000	0.0000
5		40	Fish soup	0.31	0.0039	0.0000	0.0000	0.0000	0.28	0.0033	0.0000	0.0000	0.0000	0.47	0.0070	0.0000	0.0000	
5		31	Lasagna	0.00	0.0000	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	
5		36	Legume (beans) soup	0.00	0.0000	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	
5		18	Meat balls	0.00	0.0000	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	
5		16	Meat based dishes	0.00	0.0000	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	
5		17	Meat burger (no sand-wich)	0.00	0.0000	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	
5		39	Meat soup, with pieces (Chicken soup)	0.00	0.0000	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	



Table 2 (continued)

FoodEx2 Food Group Nr	FoodEx2 Level 1 Food Group	TDS Sample Nr	Sex	Male and Female		Contribution (%)	µg/kg bw/week	Mean	Median	P95	Contribution (%)	µg/kg bw/week	Mean	Median	P95
				18–74	18–64										
				('overall population')		('adults')		('elderly')							
Nr Individuals				3272	2752	520									
TDS Sample				Contribution (%)	µg/kg bw/week	Mean	Median	P95	Contribution (%)	µg/kg bw/week	Mean	Median	P95		
5	Rice and veg- etables meal	33		0.00	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
5	Rice based dishes cooked	32		0.00	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
5	Sandwich with meat and vegetable top- ping/filling	28		0.00	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
5	Seafood-based meals	20		0.08	0.0010	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
5	Tomato soup	35		0.00	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
5	Total			18.65	0.23	–	–	–	18.13	0.22	–	–	20.90	0.31	–
7	Fish, seafood, amphibians, reptiles and invertebrates	47		0.40	0.0050	0.0000	0.0000	0.0000	0.46	0.0056	0.0000	0.0000	0.12	0.0017	0.0000
7	Bivalve mol- luscs	62		0.08	0.0010	0.0000	0.0000	0.0000	0.07	0.0009	0.0000	0.0000	0.13	0.0019	0.0000
7	Canned fish in oil (Tuna)	67		7.31	0.0913	0.0000	0.0000	0.0000	8.45	0.1017	0.0000	0.0000	2.45	0.0363	0.0000
7	Canned fish in oil (Sardine)	68		0.05	0.0007	0.0000	0.0000	0.0000	0.05	0.0006	0.0000	0.0000	0.06	0.0009	0.0000
7	Catfishes (freshwater)	45		0.01	0.0001	0.0000	0.0000	0.0000	0.01	0.0001	0.0000	0.0000	0.01	0.0002	0.0000
7	Cod, atlantic	52		0.24	0.0029	0.0000	0.0000	0.0000	0.29	0.0035	0.0000	0.0000	0.00	0.0000	0.0000
7	Cod, dried	65		18.30	0.2284	0.0000	1.9444	17.30	0.47	0.2083	0.0000	1.7015	22.59	0.3348	0.0000
7	Conger Euro- pean	50		0.55	0.0068	0.0000	0.0000	0.47	0.47	0.0057	0.0000	0.0000	0.87	0.0129	0.0000
7	European Sardine	57		0.91	0.0114	0.0000	0.0000	0.92	0.92	0.0110	0.0000	0.0000	0.91	0.0134	0.0000
7	Fish fingers, breaded	66		0.10	0.0013	0.0000	0.0000	0.12	0.12	0.0015	0.0000	0.0000	0.01	0.0002	0.0000

Table 2 (continued)

7	53	Hakes	23.65	0.2952	0.0000	2.6675	24.86	0.2993	0.0000	2.7359	18.48	0.2739	0.0000	2.4674
7	55	Horse mackerel	2.38	0.0297	0.0000	0.0000	2.14	0.0258	0.0000	0.0000	3.43	0.0509	0.0000	0.0000
7	54	Ling	0.58	0.0072	0.0000	0.0000	0.40	0.0048	0.0000	0.0000	1.36	0.0201	0.0000	0.0000
7	56	Mackerel, chub	0.40	0.0050	0.0000	0.0000	0.30	0.0036	0.0000	0.0000	0.82	0.0122	0.0000	0.0000
7	61	Marine shrimps, or prawns, cooked	0.18	0.0023	0.0000	0.0000	0.23	0.0027	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
7	46	Nile perch	2.11	0.0264	0.0000	0.0000	2.02	0.0243	0.0000	0.0000	2.52	0.0373	0.0000	0.0000
7	63	Octopus, common	1.64	0.0205	0.0000	0.0000	1.81	0.0218	0.0000	0.0000	0.89	0.0132	0.0000	0.0000
7	49	Other coastal marine fishes (wrasse, pouting, blackspot seabream, red porgy)	1.70	0.0213	0.0000	0.0000	1.65	0.0199	0.0000	0.0000	1.93	0.0286	0.0000	0.0000
7	60	Other demersal marine fishes (Scabbard-fish)	4.54	0.0567	0.0000	0.0000	4.65	0.0560	0.0000	0.0000	4.07	0.0603	0.0000	0.0000
7	59	Other pelagic marine fishes (Forkbeard, red fish)	11.89	0.1484	0.0000	0.0000	11.52	0.1387	0.0000	0.0000	13.50	0.2000	0.0000	0.0000
7	51	Plaice, european	1.33	0.0166	0.0000	0.0000	1.29	0.0156	0.0000	0.0000	1.50	0.0222	0.0000	0.0000
7	48	Sea Bream	1.40	0.0175	0.0000	0.0000	1.23	0.0148	0.0000	0.0000	2.12	0.0314	0.0000	0.0000
7	64	Squid, common	0.34	0.0042	0.0000	0.0000	0.39	0.0047	0.0000	0.0000	0.12	0.0017	0.0000	0.0000
7	69	Terrestrial snails, edible	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
7	58	Tuna	1.23	0.0154	0.0000	0.0000	1.24	0.0149	0.0000	0.0000	1.23	0.0182	0.0000	0.0000
7	-	Total	81.35	1.02	-	-	81.87	0.99	-	-	79.10	1.17	-	-
7	-	Mean Exposure (µg/kg bw)	-	1.25	-	-	-	1.20	-	-	-	1.48	-	-
7	-	% of TWI (Mean Exposure)	-	96.0	-	-	-	92.6	-	-	-	114.0	-	-
7	-	P50 (µg/kg bw)	-	0.01	-	-	-	0.00	-	-	-	0.13	-	-



**Table 3** Estimated dietary exposure of Portuguese male population to methylmercury

FoodEx2 food group Nr	FoodEx2 level 1 food group	TDS Sample Nr	Sex	Male	18–74		18–64 ('adults')		65–74 ('elderly')			
					Nr	Individuals	1363	220	Contribution (%)	Contribution (%)	Contribution (%)	Contribution (%)
				TDS sample		Contribution (%)		Contribution (%)		Contribution (%)		
				Mean	Median	P95	Mean	Median	P95	Mean	Median	P95
				µg/kg bw/week	µg/kg bw/week	µg/kg bw/week	µg/kg bw/week	µg/kg bw/week	µg/kg bw/week	µg/kg bw/week	µg/kg bw/week	µg/kg bw/week
5	Composite dishes	12		0.00	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
				0.00	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
5	Beans, meat, and vegetables meal (Meat and vegetables boiled dish, Portuguese style)	13		0.00	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
5	Beans, meat, and vegetables meal (Beans stewed with pork and cow meat)	14		0.00	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
5	Beans, meat, and vegetables meal (Pork meat, chickpea, pasta and vegetables stewed)	15		0.00	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
5	Dishes, incl. Ready to eat meals (excluding soups and salads) (Bread based dish)	9		0.42	0.0052	0.0000	0.0000	0.41	0.0047	0.0000	0.0000	0.0082
5	Finger food	30		0.06	0.0008	0.0000	0.0065	0.08	0.0009	0.0000	0.0069	0.0003
5	Fish and potatoes meal (Cod fish dishes)	22		1.77	0.0218	0.0000	0.0000	1.91	0.0218	0.0000	0.0000	0.0218
5	Fish and potatoes meal (Mixed fish stew)	23		13.59	0.1672	0.0000	0.0000	12.45	0.1425	0.0000	0.0000	0.3205
5	Fish and potatoes meal (Potato puree with fish)	24		0.90	0.0111	0.0000	0.0000	1.02	0.0117	0.0000	0.0000	0.0072



Table 3 (continued)

FoodEx2 food group Nr	FoodEx2 level 1 food group	TDS Sample Nr	Sex	Age group	Male 18–74	18–64 ('adults')	65–74 ('elderly')	Nr Individuals	TDS sample			Contribution				
									Contribution (%)	µg/kg bw/week	P95	Contribution (%)	µg/kg bw/week	P95		
5	Potatoes and meat meal (Potato puree withmeat)	11	Male	18–74	1583	1363	220	0.00	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	
5	Prepared fish salad	21	Male	18–74	1583	1363	220	1.34	0.0164	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	
5	Quiche	26	Male	18–74	1583	1363	220	0.00	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	
5	Rice and meat meal	34	Male	18–74	1583	1363	220	0.00	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	
5	Rice and vegetables meal	33	Male	18–74	1583	1363	220	0.00	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	
5	Rice based dishes cooked	32	Male	18–74	1583	1363	220	0.00	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	
5	Sandwich with meat and vegetable topping/filling	28	Male	18–74	1583	1363	220	0.00	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	
5	Seafood-based meals	20	Male	18–74	1583	1363	220	0.07	0.0009	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	
5	Tomato soup	35	Male	18–74	1583	1363	220	0.00	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	
5	Total	–	Male	18–74	1583	1363	220	21.18	0.26	–	–	20.94	0.24	–	22.17	0.39
FoodEx2 food group Nr	FoodEx2 level 1 food group	TDS Sample Nr	Sex	Age group	Male 18–74	18–64 ('adults')	65–74 ('elderly')	Nr Individuals	TDS sample			Contribution				
7	Fish, seafood, amphibians, reptiles and invertebrates	47	Male	18–74	1583	1363	220	0.31	0.0038	0.0000	0.0000	0.39	0.0044	0.0000	0.0000	0.0000
7	Bivalve molluscs	62	Male	18–74	1583	1363	220	0.07	0.0008	0.0000	0.0000	0.02	0.0003	0.0000	0.0000	0.0000

Table 3 (continued)

FoodEx2 food group Nr	FoodEx2 level 1 food group	TDS Sample Nr	Sex	Male		18–64 ('adults')		65–74 ('elderly')								
				Age group	18–74	18–64	65–74									
Nr Individuals				1583	1363	220										
TDS sample				Contribution (%)	Mean	Median	P95	Contribution (%)	Mean	Median	P95					
				µg/kg bw/week	µg/kg bw/week	µg/kg bw/week	µg/kg bw/week	µg/kg bw/week	µg/kg bw/week	µg/kg bw/week	µg/kg bw/week					
7		67		Canned fish in oil (Tuna)	6.67	0.0820	0.0000	0.0000	7.63	0.0873	0.0000	0.0000	2.80	0.0494	0.0000	0.0000
7		68		Canned fish in oil (Sardine)	0.02	0.0002	0.0000	0.0000	0.02	0.0003	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
7		45		Catfishes (freshwater)	0.01	0.0001	0.0000	0.0000	0.01	0.0001	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
7		52		Cod, atlantic	0.36	0.0045	0.0000	0.0000	0.45	0.0052	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
7		65		Cod, dried	19.75	0.2430	0.0000	1.8279	18.18	0.2081	0.0000	1.5841	26.03	0.4596	0.0000	3.3991
7		50		Conger European	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
7		57		European Sardine	0.97	0.0119	0.0000	0.0000	1.04	0.0119	0.0000	0.0000	0.68	0.0120	0.0000	0.0000
7		66		Fish fingers, breaded	0.05	0.0006	0.0000	0.0000	0.07	0.0007	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
7		53		Hakes	19.67	0.2421	0.0000	2.3223	21.50	0.2460	0.0000	2.3223	12.32	0.2175	0.0000	2.2431
7		55		Horse mackerel	1.89	0.0233	0.0000	0.0000	1.68	0.0192	0.0000	0.0000	2.75	0.0485	0.0000	0.0000
7		54		Ling	1.22	0.0150	0.0000	0.0000	0.85	0.0097	0.0000	0.0000	2.69	0.0475	0.0000	0.0000
7		56		Mackerel, chub	0.46	0.0056	0.0000	0.0000	0.33	0.0038	0.0000	0.0000	0.96	0.0170	0.0000	0.0000
7		61		Marine shrimps or prawns, cooked	0.27	0.0033	0.0000	0.0000	0.34	0.0038	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
7		46		Nile perch	2.09	0.0257	0.0000	0.0000	2.14	0.0244	0.0000	0.0000	1.90	0.0336	0.0000	0.0000
7		63		Octopus, common	2.24	0.0275	0.0000	0.0000	2.45	0.0281	0.0000	0.0000	1.37	0.0242	0.0000	0.0000
7		49		Other coastal marine fishes (wrasse, pouting, blackspot seabream, red porgy)	1.72	0.0212	0.0000	0.0000	1.80	0.0206	0.0000	0.0000	1.42	0.0250	0.0000	0.0000

Table 3 (continued)

FoodEx2 food group Nr	FoodEx2 level 1 food group	TDS Sample Nr	Sex	Male	18–74		18–64 (‘adults’)		65–74 (‘elderly’)						
					Nr Individuals	1583	1363	220							
TDS sample					Contribution (%)		µg/kg bw/week		Contribution (%)		µg/kg bw/week				
					Mean	Median	Mean	P95	Mean	P95	Mean	P95			
7		60		5.43	0.0669	0.0000	0.0000	0.0000	0.0627	0.0000	0.0000	0.0000	0.0927	0.0000	0.0000
			Other demersal marine fishes (Scabbard- fish)					5.48							
7		59		11.37	0.1399	0.0000	0.0000	10.19	0.1166	0.0000	0.0000	16.08	0.2839	0.0000	0.0000
			Other pelagic marine fishes (Forkbeard, red fish)												
7		51		1.20	0.0148	0.0000	0.0000	1.11	0.0127	0.0000	0.0000	1.56	0.0275	0.0000	0.0000
			Plaice, euro- pean												
7		48		0.92	0.0114	0.0000	0.0000	1.15	0.0132	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
			Sea Bream												
7		64		0.39	0.0048	0.0000	0.0000	0.45	0.0052	0.0000	0.0000	0.13	0.0023	0.0000	0.0000
			Squid, common												
7		69		0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
			Terrestrial snails, edible												
7		58		1.74	0.0214	0.0000	0.0000	1.77	0.0202	0.0000	0.0000	1.62	0.0285	0.0000	0.0000
			Tuna												
7			Total	78.82	0.97	-	-	79.06	0.90	-	-	77.83	1.37	-	-
			Mean Exposure (µg/kg bw)	-	1.23	-	-	-	1.14	-	-	-	1.77	-	-
			% of TWI (Mean Expo- sure)	-	94.7	-	-	-	88.0	-	-	-	135.8	-	-
			P50 (µg/kg bw)	-	0.01	-	-	-	0.01	-	-	-	0.25	-	-
			% of TWI (P50)	-	0.5	-	-	-	0.4	-	-	-	19.2	-	-
			P95 (µg/kg bw)	-	5.39	-	-	-	5.24	-	-	-	7.29	-	-
			% of TWI (P95)	-	414.7	-	-	-	403.0	-	-	-	561.1	-	-
			% of persons above the TWI	-	27.3	-	-	-	25.8	-	-	-	36.4	-	-

TDS Total Diet Study, TWI Tolerable Weekly Intake, P50 Percentile 50, P95 Percentile 95

**Table 4** Estimated dietary exposure of Portuguese female population to methylmercury

FoodEx2 food group Nr	FoodEx2 level 1 food group	TDS Sample Nr	Sex	Female		Contribution (%)	µg/kg bw/week		Contribution (%)	µg/kg bw/week	
				18–74	18–64 ('adults')		65–74 ('elderly')	Mean		Median	Mean
				18–74	18–64 ('adults')	65–74 ('elderly')					
				1689	1389	300					
				TDS sample							
5	Composite dishes	12	Beans and meat meal (Beans and gut dish)	0.00	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
5		13	Beans, meat, and vegetables meal (Meat and vegetables boiled dish, Portuguese style)	0.00	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
5		14	Beans, meat, and vegetables meal (Beans stewed with pork and cow meat)	0.00	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
5		15	Beans, meat, and vegetables meal (Pork meat, chickpea, pasta and vegetables stewed)	0.00	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
5		9	Dishes, incl. Ready to eat meals (excluding soups and salads) (Bread based dish)	0.34	0.0043	0.0000	0.0000	0.27	0.0034	0.0000	0.64
5		30	Finger food	0.04	0.0005	0.0000	0.0000	0.04	0.0005	0.0000	0.02
5		22	Fish and potatoes meal (Cod fish-dishes)	1.37	0.0174	0.0000	0.0000	1.52	0.0191	0.0000	0.73
5		23	Fish and potatoes meal (Mixed fish stew)	9.87	0.1248	0.0000	0.0000	8.76	0.1106	0.0000	14.96
5		24	Fish and potatoes meal (Potato puree with fish)	0.87	0.0110	0.0000	0.0000	0.98	0.0124	0.0000	0.38
5		25	Fish and rice meal	1.88	0.0237	0.0000	0.0000	1.89	0.0239	0.0000	1.80

Table 4 (continued)

FoodEx2 food group Nr	FoodEx2 level 1 food group	TDS Sample Nr	Sex	Female		18–64 (‘adults’)		65–74 (‘elderly’)							
				18–74		1389		300							
				Contri- bution (%)	µg/kg bw/week	Mean	Median	P95	Contribution (%)	µg/kg bw/week	Mean	Median	P95		
5		19		0.74	0.0094	0.0000	0.0000	0.85	0.0107	0.0000	0.0000	0.26	0.0033	0.0000	0.0000
	Fish and seafood based dishes														
5		40		0.24	0.0031	0.0000	0.0000	0.20	0.0025	0.0000	0.0000	0.46	0.0059	0.0000	0.0000
	Fish soup														
5		31		0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
	Lasagna														
5		36		0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
	Legume (beans) soup														
5		18		0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
	Meat balls														
5		16		0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
	Meat based dishes														
5		17		0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
	Meat burger (no sandwich)														
5		39		0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
	Meat soup, with pieces (Chicken soup)														
5		42		0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
	Mixed vegetable salad (Tomato and lettuce salad)														
5		43		0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
	Mixed vegetable salad (Russian salad)														
5		37		0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
	Mixed vegetables soup (Green cabbage soup)														
5		38		0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
	Mixed vegetables soup, with puree or pieces														
5		27		0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
	Omelette, plain														
5		29		0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
	Pizza and pizza-like dishes														
5		10		0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
	Potato based dishes (mashed potatoes)														

Table 4 (continued)

FoodEx2 food group Nr	FoodEx2 level 1 food group	TDS Sample Nr	Sex	Female		18–64 (‘adults’)		65–74 (‘elderly’)						
				18–74		1389		300						
				Contribution (%)		µg/kg bw/week		Contribution (%)		µg/kg bw/week				
		Mean		Median		P95		Mean		Median		P95		
5		11		0.00	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
	Potatoes and meat meal (Potato puree with meat)			0.91	0.0116	0.0000	0.0000	1.03	0.0131	0.0000	0.0000	0.36	0.0046	0.0000
5		21		0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000
	Prepared fish salad			0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000
5		26		0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000
	Quiche			0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000
5		34		0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000
	Rice and meat meal			0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000
5		33		0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000
	Rice and vegetables meal			0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000
5		32		0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000
	Rice based dishes cooked			0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000
5		28		0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000
	Sandwich with meat and vegetable topping/filling			0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000
5		20		0.09	0.0011	0.0000	0.0000	0.11	0.0013	0.0000	0.0000	0.02	0.0003	0.0000
	Seafood-based meals			0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000
5		35		0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000
	Tomato soup			16.35	0.21	0.0060	0.0000	15.64	0.20	0.0067	0.0000	19.62	0.25	0.0030
5		47		0.48	0.0060	0.0000	0.0000	0.53	0.0067	0.0000	0.0000	0.24	0.0030	0.0000
	Atlantic salmon			0.09	0.0012	0.0000	0.0000	0.11	0.0014	0.0000	0.0000	0.00	0.0000	0.0000
7	Fish, seafood, amphibians, reptiles and invertebrates	62		7.90	0.1000	0.0000	0.0000	9.17	0.1158	0.0000	0.0000	2.09	0.0266	0.0000
		Bivalve molluscs		0.09	0.0012	0.0000	0.0000	0.11	0.0014	0.0000	0.0000	0.00	0.0000	0.0000
7		67		0.09	0.0011	0.0000	0.0000	0.08	0.0010	0.0000	0.0000	0.12	0.0016	0.0000
		Canned fish in oil (Tuna)		0.01	0.0001	0.0000	0.0000	0.01	0.0001	0.0000	0.0000	0.02	0.0003	0.0000
7		45		0.12	0.0015	0.0000	0.0000	0.15	0.0019	0.0000	0.0000	0.00	0.0000	0.0000
		Catfishes (freshwater)		16.97	0.2146	0.0000	2.0170	16.51	0.2084	0.0000	1.8180	19.09	0.2432	0.0000
7		52		1.05	0.0132	0.0000	0.0000	0.89	0.0112	0.0000	0.0000	1.76	0.0224	0.0000
		Cod, atlantic		0.86	0.0109	0.0000	0.0000	0.80	0.0102	0.0000	0.0000	1.14	0.0145	0.0000
7		65		1.05	0.0132	0.0000	0.0000	0.89	0.0112	0.0000	0.0000	1.76	0.0224	0.0000
		Cod, dried		0.86	0.0109	0.0000	0.0000	0.80	0.0102	0.0000	0.0000	1.14	0.0145	0.0000
7	50		1.05	0.0132	0.0000	0.0000	0.89	0.0112	0.0000	0.0000	1.76	0.0224	0.0000	
	Conger European		0.86	0.0109	0.0000	0.0000	0.80	0.0102	0.0000	0.0000	1.14	0.0145	0.0000	
7	57		0.86	0.0109	0.0000	0.0000	0.80	0.0102	0.0000	0.0000	1.14	0.0145	0.0000	
	European Sardine													

Table 4 (continued)

FoodEx2 food group Nr	FoodEx2 level 1 food group	TDS Sample Nr	Sex	Female		18–64 ('adults')		65–74 ('elderly')							
				18–74		1389		300							
				Contribution (%)	µg/kg bw/week	Mean	Median	Mean	Median	Mean	Median				
7		66		0.14	0.0018	0.0000	0.0000	0.17	0.0022	0.0000	0.0000	0.02	0.0003	0.0000	0.0000
7		53		27.28	0.3450	0.0000	2.9908	27.84	0.3515	0.0000	3.1951	24.73	0.3152	0.0000	2.5673
7		55		2.83	0.0358	0.0000	0.0000	2.55	0.0322	0.0000	0.0000	4.13	0.0526	0.0000	0.0000
7		54		0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
7		56		0.35	0.0044	0.0000	0.0000	0.28	0.0035	0.0000	0.0000	0.68	0.0086	0.0000	0.0000
7		61		0.11	0.0014	0.0000	0.0000	0.13	0.0017	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
7		46		2.14	0.0270	0.0000	0.0000	1.92	0.0242	0.0000	0.0000	3.14	0.0401	0.0000	0.0000
7		63		1.09	0.0138	0.0000	0.0000	1.24	0.0157	0.0000	0.0000	0.41	0.0052	0.0000	0.0000
7		49		1.69	0.0214	0.0000	0.0000	1.52	0.0192	0.0000	0.0000	2.45	0.0312	0.0000	0.0000
7		60		3.73	0.0472	0.0000	0.0000	3.92	0.0495	0.0000	0.0000	2.87	0.0365	0.0000	0.0000
7		59		12.37	0.1565	0.0000	0.0000	12.70	0.1604	0.0000	0.0000	10.87	0.1385	0.0000	0.0000
7		51		1.45	0.0183	0.0000	0.0000	1.45	0.0183	0.0000	0.0000	1.43	0.0183	0.0000	0.0000
7		48		1.83	0.0232	0.0000	0.0000	1.30	0.0164	0.0000	0.0000	4.27	0.0544	0.0000	0.0000
7		64		0.29	0.0036	0.0000	0.0000	0.33	0.0041	0.0000	0.0000	0.10	0.0013	0.0000	0.0000
7		69		0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000	0.00	0.0000	0.0000	0.0000
7		58		0.77	0.0098	0.0000	0.0000	0.76	0.0096	0.0000	0.0000	0.83	0.0106	0.0000	0.0000
7				83.65	1.06	-	-	84.36	1.07	-	-	80.38	1.02	-	-
				-	1.26	-	-	-	1.26	-	-	-	1.27	-	-
				-	97.3	-	-	-	97.1	-	-	-	98.0	-	-



(European Food Safety Authority [EFSA], 2012). Nasreddine (Nasreddine et al. 2006) estimated a mean methylmercury exposure of 0.28  $\mu\text{g}/\text{kg}$  bw/week for the urban adult Lebanese population (25 to 54 years old) corresponding to 21.5% of the TWI of 1.3  $\mu\text{g}/\text{kg}$  bw/week (EFSA 2012). Methylmercury mean exposure by the Czechia's adult population was reported as 4.3% of 1.3  $\mu\text{g}/\text{kg}$  bw/week TWI corresponding to 0.056  $\mu\text{g}/\text{kg}$  bw/week (Sokolov et al. 2022). Exposure of Germany's adult population (14 to 79 years old) to methylmercury was estimated by Sarvan as 0.072 (mean) and 0.49 (P95)  $\mu\text{g}/\text{kg}$  bw/week (Sarvan et al. 2021), corresponding respectively 5.5 and 38% of the 1.3  $\mu\text{g}/\text{kg}$  bw/week TWI (EFSA 2012). When consumers only were considered, methylmercury exposure was estimated by Kolbaum as 0.28 (mean) and 0.85 (P95)  $\mu\text{g}/\text{kg}$  bw/week (Kolbaum et al. 2019) and by Sarvan as 0.339 (mean) the later corresponding to 26% of the 1.3  $\mu\text{g}/\text{kg}$  bw/week TWI (Sarvan et al. 2021). Marín (Marin et al. 2017) reported a methylmercury average exposure of 0.60  $\mu\text{g}/\text{kg}$  bw/week (UB) estimated based on consumption of fish and seafood by the adult population (16 to 95 years old) of Valencia region, Spain. Methylmercury mean exposure of 1.25  $\mu\text{g}/\text{kg}$  bw/week estimated in this study was well above the values reported by the previously referred studies that ranged from 0.048 to 0.60  $\mu\text{g}/\text{kg}$  bw/week. The same was observed when consumers only mean exposure (2.64  $\mu\text{g}/\text{kg}$  bw/week) was compared with German population values.

The considerably higher methylmercury exposure estimates found in the present study as compared to those reported in other countries might be explained by the significantly higher fish and seafood consumption by the Portuguese population and not by the methylmercury occurrence levels of the fish consumed.

Adverse neurodevelopmental outcomes in infants and young children during pregnancy are associated with methylmercury, therefore, exposure to methylmercury was estimated for 905 Portuguese women of childbearing age (18 to 45 years old). Table 6 shows the estimated dietary exposure to methylmercury of Portuguese childbearing age women, detailing the contribution of each TDS sample to exposure. The estimated mean weekly exposure to methylmercury was 1.13  $\mu\text{g}/\text{kg}$  bw/week corresponding to 86.9% of the TWI (1.3  $\mu\text{g}/\text{kg}$  bw/week). Whereas, the estimated median weekly exposure (P50) was 0  $\mu\text{g}/\text{kg}$  bw/week. At the 95th percentile, methylmercury weekly exposure was 5.56  $\mu\text{g}/\text{kg}$  bw/week, approximately four fold the TWI. The percentage of childbearing age women exceeding the TWI was 25.0%. When only women of childbearing age who consumed fish and seafood either classified as 'fish, seafood, amphibians, reptiles and invertebrates' or 'composite dishes' (409 individuals) were considered (consumers only) the estimated mean weekly exposure was 2.50  $\mu\text{g}/\text{kg}$  bw/week (192% of TWI), two times higher than that of all women of childbearing age

(Table 6). Hake (20.2%), dried cod (14.5%) and canned tuna (12.8%) were the three main contributors to methylmercury exposure.

Carvalho (Carvalho et al. 2022) reported a methylmercury mean exposure of 0.58  $\mu\text{g}/\text{kg}$  bw/week in Portuguese women of childbearing age (15 to 49 years old), corresponding to 44.6% of the TWI (1.3  $\mu\text{g}/\text{kg}$  bw/week). The percentage of childbearing age women exceeding the TWI was 12.7%. The values reported by Carvalho (Carvalho et al. 2022), using a different methodology, were approximately half of those found in the present study. A methylmercury mean exposure of 0.98  $\mu\text{g}/\text{kg}$  bw/week was reported by Ortega-García (Ortega-García et al. 2009) in childbearing age women of Murcia, Spain corresponding to 75.4% of the TWI (1.3  $\mu\text{g}/\text{kg}$  bw/week). P95 exposure was estimated at 2.44  $\mu\text{g}/\text{kg}$  bw/week (188% of the TWI) (Ortega-García et al. 2009). The mean exposure level estimated in the present study was slightly higher than the one reported by Ortega-García (Ortega-García et al. 2009) whereas P95 exposure level was approximately twice. Arnich (Arnich et al. 2012) reported a methylmercury mean exposure of 0.13  $\mu\text{g}/\text{kg}$  bw/week in women of childbearing age (18 to 45 years old) in France, corresponding to 10% of the TWI (1.3  $\mu\text{g}/\text{kg}$  bw/week). P95 exposure was estimated at 0.47  $\mu\text{g}/\text{kg}$  bw/week (36% of the TWI) (Arnich et al. 2012). The mean and P95 exposure levels reported by Arnich (Arnich et al. 2012) were one order of magnitude lower than those found in the present study.

Consumers-only childbearing age women (15 to 49 years old) of Germany showed a mean exposure to methylmercury of 0.21  $\mu\text{g}/\text{kg}$  bw/week corresponding to 16% of the TWI (1.3  $\mu\text{g}/\text{kg}$  bw/week) and a P95 exposure of 0.72  $\mu\text{g}/\text{kg}$  bw/week, 55% of the TWI (Kolbaum et al. 2019). The mean exposure level estimated in the present study was approximately one order of magnitude higher than the one reported by Kolbaum (Kolbaum et al. 2019).

A biomonitoring study in childbearing women (25 to 44 years old) in Portugal reported a mean blood mercury of 5.9  $\mu\text{g}/\text{L}$  (Santiago et al., n.d.) corresponding to a weekly exposure to methylmercury of 0.81  $\mu\text{g}/\text{kg}$  bw/week calculated according to the average ratios between intake and blood levels indicated by WHO (WHO 2008). Other biomonitoring studies focusing on specific Portuguese regions and pregnant women (Alves et al. 2017; Caetano et al. 2019; Namorado et al. 2023; Nunes et al. 2014) or childbearing age women (Castaño et al. 2015) presented results of the same order of magnitude.

The referred biomonitoring studies were in line with the methylmercury exposure estimates obtained by the present study and by Carvalho (Carvalho et al. 2022) and support the need of special consideration regarding fish consumption by the Portuguese childbearing age women.

The European Commission (EC) has advised the establishment of specific national guidelines regarding fish and

seafood consumption to maximize their health benefits while minimizing the risk of mercury toxicity, specifying which species and how often they should be consumed (European Commission [EC], 2022). Subsequently, recent recommendations for fish and seafood consumption in Portugal have been issued, highlighting pregnant and breastfeeding women, as well as children up to 10 years old, as vulnerable groups (General Directorate for Food and Veterinary [DGAV] et al., 2023). To adhere to these recommendations, the vulnerable population groups are advised to avoid species with high mercury levels such as blue shark, fresh tuna, ling, scabbardfish, small-spotted catshark, smooth-hound, and swordfish, and to consume species with low to medium mercury content three to 4 times a week for optimal health benefits. For the general population a consumption up to 7 times a week regardless of the species is advocated. The recommendations were developed by a working group promoted by the General Directorate for Food and Veterinary which included the National Institute of Health Doctor Ricardo Jorge, the Economic and Food Safety Authority, the Faculty of Nutrition and Food Sciences of the University of Porto, the Portuguese Institute for Sea and Atmosphere and the Public Health Institute of the University of Porto, and were based on a risk–benefit assessment associated with fish consumption by the Portuguese population (Carvalho et al. 2022).

### Inorganic Mercury

The estimated dietary exposure of Portuguese overall population ('male and female') to inorganic mercury is presented in Table 7. Corresponding data for 'male' and 'female' population groups are presented in Tables 8 and 9, respectively.

Mean exposure to inorganic mercury for the 'overall population' was 0.30 µg/kg bw/week (0.30 for males and 0.33 for females) using the LB approach, and 0.37 µg/kg bw/week (0.36 for males and 0.40 for females) using the UB approach. Among other sex and age group combinations, mean exposure ranged from 0.29 µg/kg bw/week (for male adults and female elderly) to 0.40 µg/kg bw/week for elderly males at LB, and from 0.35 µg/kg bw/week (for male adults and female elderly) to 0.46 µg/kg bw/week for elderly males at UB. At the 95th percentile, inorganic mercury weekly exposure of the 'overall population' was 1.26 (LB) and 1.27 (UB) µg/kg bw/week. Across all population groups, the mean estimated exposure to inorganic mercury ranged from 7.1 to 10.0% (LB) and from 8.7 to 11.5% (UB) of the TWI of 4 µg/kg bw/week. At the 95th percentile, this exposure ranged from 25.8 to 37.2% (LB) and from 27.3 to 38.7% (UB). The TWI was exceeded by 3.4% (LB) and 3.5% (UB) individuals of the 'overall population'. The food groups 'fish, seafood, amphibians, reptiles and invertebrates' and 'composite dishes' contributed 78 to 88% for inorganic

mercury exposure of all sex and age groups. Detailed data on the contribution of each TDS sample to inorganic mercury exposure are presented as supplementary information (Supplementary Table S1, S2, S3). The main foods as eaten contributing to inorganic mercury exposure are presented in Table 10. Cod (dried) and hake were the two main contributors (food as eaten) ranging from 11.2 to 20.0% and from 6.7 to 12.3%, respectively.

Arnich (Arnich et al. 2012) reported for the French adults a dietary (except seafood) inorganic mercury mean exposure of 0.042 µg/kg bw/week (LB) and 1.26 µg/kg bw/week (UB) and 95th percentiles of 0.182 (LB) and 2.03 (UB) µg/kg bw/week. The TWI was not exceeded by any individual. The mean exposure level estimated in the present study was approximately ten times higher at LB and three times lower at UB than the one reported by Arnich (Arnich et al. 2012), whereas at P95 it was seven times higher at LB and half at UB. The disparity in results may be due to methodological differences between the two studies, such as: (i) the inclusion of seafood TDS samples in the present study using EFSA's conservative approach (EFSA 2012); (ii) percentage of left censored data; and (iii) analytical limits.

The population of Germany (14 to 80 years old) showed a mean exposure to inorganic mercury of 0.02 (LB) and 0.07 (UB) µg/kg bw/week corresponding to 1 and 2% of the TWI (4 µg/kg bw/week), respectively, and a P95 exposure of 0.07 (LB) and 0.14 (UB) µg/kg bw/week, corresponding to 2 and 4% of the TWI (4 µg/kg bw/week), respectively (Kolbaum et al. 2019). The mean exposure level estimated in the present study was approximately five times higher than the one reported by Kolbaum (Kolbaum et al. 2019) and seven times higher at P95. However, Kolbaum study did not include fish and seafood samples in the estimation of inorganic mercury exposure and considered a different age group.

As previously mentioned, tap water was excluded from the TDS food list and, therefore, a deterministic approach was used to evaluate its contribution to inorganic mercury exposure of the Portuguese population. The exposure of the 'overall population' to inorganic mercury through the consumption of tap water was estimated at 0.006 µg/kg bw/week (0.15% of TWI), corresponding roughly to 1.6% of the total inorganic mercury exposure.

### Strengths and Limitations

TDSs are very useful to assess chronic exposure to chemicals because they allow covering the whole diet with limited costs by using food sample pooling. Furthermore, foods are analysed after being prepared according to common consumption habits, thus, the chemicals in the foods analysed are closely representative of what is actually ingested by the consumer. The use of a TDS harmonised methodology provides data that facilitates assessing time trends and country

**Table 5** Main foods contributing to methylmercury exposure

Sex	Male and Female			Male			Female		
	18–74 (%)	18–64 (%)	65–74 (%)	18–74 (%)	18–64 (%)	65–74 (%)	18–74 (%)	18–64 (%)	65–74 (%)
Age group									
Contribution									
Cod	18.30	17.30	22.59	19.75	18.18	26.03	16.97	16.51	19.09
Hake	14.33	14.75	12.51	10.54	11.01	8.67	17.79	18.09	16.42
Mixed fish stew	8.69	8.66	8.83	9.93	9.25	12.64	7.56	8.12	4.96
Scabbardfish	8.66	8.24	10.44	8.67	7.60	12.95	8.64	8.81	7.88
Canned fish in oil (Tuna)	7.17	8.27	2.45	6.67	7.63	2.80	7.63	8.84	2.09
Fish	5.32	5.45	4.75	4.42	4.96	2.23	6.14	5.88	7.30
Mixed cod stew	1.23	0.43	4.65	1.46	0.92	3.60	1.02	0.00	5.72

comparison of the dietary exposure of populations to chemical substances. Nevertheless, uncertainties are present at the different steps of exposure assessment and may lead to over or underestimation. The consumption data used in this study was gathered in 2009, and it is possible that food habits have evolved since then. However, the consumption of fish and seafood per year per capita, which is the primary contributor to mercury exposure, remained relatively stable from 2009 (57.17 kg) to the years when the food collection took place, from 2014 (53.22 kg) to 2016 (57.33 kg), according to Our World in Data (Our World in Data 2021). Another uncertainty arises from the fact that only one day of consumption was recorded, potentially resulting in an overestimation of the higher percentiles of chronic exposure. Regarding the choice of foods to analyse, using the food group approach when applying the selection criteria to include foods in the TDS food list avoided the loss of some less consumed foods that could contribute considerably to exposure. If foods were ranked according to the amount consumed across the whole diet, some less consumed foods would never be selected even if they were important contributors to exposure. Occurrence data used to perform the exposure assessment estimation was based on total mercury analytical determination and a high left-censored data rate was observed for food groups other than those containing fish and seafood. To further refine the exposure assessment to mercury, analytical improvements would be desirable, such as lowering LODs for total mercury determination and/or preferably conducting speciation analysis.

## Conclusions

The mean estimated baseline exposure of the Portuguese overall population (adults and elderly) to methylmercury and inorganic mercury did not exceed the respective HBGV. However, the percentage of individuals exceeding the HBGV was 27.6% for methylmercury and 3.5% for inorganic

mercury (UB approach). According to these results the possibility of adverse health effects cannot be excluded. The methylmercury mean and P50 exposure results of the overall population evidence a distribution markedly skewed to the right with only half of the individuals being exposed. Consequently, when focusing on consumers only, the mean and P50 values were observed at 189% and 121% of the Tolerable Weekly Intake (TWI), respectively. The findings are particularly of concern for women of childbearing age, as their exposure to methylmercury poses risks of adverse neurodevelopmental outcomes to the child during pregnancy. The previous findings pose a double concern on the one hand the very high exposure to methylmercury of fish and seafood consumers, as discussed before, and on the other hand the potential low intake of omega 3 fatty acids by the non-consumers, with particular relevance to women of childbearing age. Omega 3 fatty acids has been associated with a reduction of coronary heart disease mortality in adults and with neurodevelopmental improvement in infants and young children derived from mother's fish consumption during pregnancy (FAO 2011) (EFSA 2015).

Future assessment of methylmercury exposure by the Portuguese population and particularly of the vulnerable groups (pregnant and lactating women and children up to ten years old) identified in the fish and seafood consumption recommendations issued recently in Portugal will be needed to evaluate the impact of this risk management measure.

Despite the TDS methodology being recommended as the most cost-effective means to evaluate chronic exposure to food contaminants and the international efforts to harmonize procedures, discrepancies persist in published data, making comparisons challenging. Variations arise in food list preparation, food classification, food sampling, and reporting exposure results. Harmonisation initiatives such as those by FAO/WHO/EFSA (EFSA et al. 2011) and TDS-Exposure (Pité et al. 2018) focused mainly on TDS food list preparation and sampling, and the FoodEx2 food classification system was proved adequate by the TDS Exposure project

**Table 6** Estimated dietary exposure of Portuguese of childbearing age women (18 to 45 years old) to methylmercury

FoodEx2 food group Nr	FoodEx2 level 1 food group Nr	TDS sample Nr	TDS sample Description	Female 18–45					
				Contri- bution (%)	All individuals		Consumers only		
					µg/kg bw/day	Mean	Median	Mean	Median
5	Composite dishes	12	Beans and meat meal (Beans and gut dish)	0.00	0.0000	0.0000	0.0000	0.0000	–
5		13	Beans, meat, and vegetables meal (Meat and vegetables boiled dish, Portuguese style)	0.00	0.0000	0.0000	0.0000	0.0000	–
5		14	Beans, meat, and vegetables meal (Beans stewed with pork and cow meat)	0.00	0.0000	0.0000	0.0000	0.0000	–
5		15	Beans, meat, and vegetables meal (Pork mestr, chickpea, pasta and vegetables stewed)	0.00	0.0000	0.0000	0.0000	0.0000	–
5		9	Dishes, incl. Ready to eat meals (excluding soups and salads) (Bread based dish)	0.02	0.0002	0.0000	0.0000	0.0005	–
5		30	Finger food	0.00	0.0000	0.0000	0.0000	0.0000	–
5		22	Fish and potatoes meal (Cod fish dishes)	0.38	0.0043	0.0000	0.0000	0.0094	–
5		23	Fish and potatoes meal (Mixed fish stew)	3.83	0.0433	0.0000	0.0000	0.0958	–
5		24	Fish and potatoes meal (Potato puree with fish)	0.35	0.0039	0.0000	0.0000	0.0087	–
5		25	Fish and rice meal	0.92	0.0104	0.0000	0.0000	0.0230	–
5		19	Fish and seafood based dishes	0.34	0.0038	0.0000	0.0000	0.0085	–
5		40	Fish soup	0.02	0.0002	0.0000	0.0000	0.0004	–
5		31	Lasagna	0.00	0.0000	0.0000	0.0000	0.0000	–
5		36	Legume (beans) soup	0.00	0.0000	0.0000	0.0000	0.0000	–
5		18	Meat balls	0.00	0.0000	0.0000	0.0000	0.0000	–
5		16	Meat based dishes	0.00	0.0000	0.0000	0.0000	0.0000	–
5		17	Meat burger (no sandwich)	0.00	0.0000	0.0000	0.0000	0.0000	–
5		39	Meat soup, with pieces (Chicken soup)	0.00	0.0000	0.0000	0.0000	0.0000	–
5		42	Mixed vegetable salad (Tomato and lettuce salad)	0.00	0.0000	0.0000	0.0000	0.0000	–
5		43	Mixed vegetable salad (Russian salad)	0.00	0.0000	0.0000	0.0000	0.0000	–
5		37	Mixed vegetables soup (Green cabbage soup)	0.00	0.0000	0.0000	0.0000	0.0000	–
5		38	Mixed vegetables soup, with puree or pieces	0.00	0.0000	0.0000	0.0000	0.0000	–
5		27	Omelette, plain	0.00	0.0000	0.0000	0.0000	0.0000	–

Table 6 (continued)

FoodEx2 food group Nr	FoodEx2 level 1 food group Nr	TDS sample Nr	TDS sample	Female 18–45							
				Contri- bution (%)	All individuals		Consumers only				
					µg/kg bw/day	Mean	Median	P95	Mean	Median	P95
5	29		Pizza and pizza-like dishes	0.00	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	–
5	10		Potato based dishes (mashed potatoes)	0.00	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	–
5	11		Potatoes and meat meal (Potato puree with meat)	0.00	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	–
5	21		Prepared fish salad	0.28	0.0032	0.0000	0.0000	0.0000	0.0071	–	–
5	26		Quiche	0.00	0.0000	0.0000	0.0000	0.0000	0.0000	–	–
5	34		Rice and meat meal	0.00	0.0000	0.0000	0.0000	0.0000	0.0000	–	–
5	33		Rice and vegetables meal	0.00	0.0000	0.0000	0.0000	0.0000	0.0000	–	–
5	32		Rice based dishes cooked	0.00	0.0000	0.0000	0.0000	0.0000	0.0000	–	–
5	28		Sandwich with meat and vegetable topping/filling	0.00	0.0000	0.0000	0.0000	0.0000	0.0000	–	–
5	20		Seafood-based meals	0.06	0.0006	0.0000	0.0000	0.0000	0.0014	–	–
5	35		Tomato soup	0.00	0.0000	0.0000	0.0000	0.0000	0.0000	–	–
			Total	6.19	0.07	–	–	–	0.15	–	–

Table 6 (continued)

FoodEx2 food group Nr	FoodEx2 level 1 food group Nr	TDS sample Nr	TDS sample	Female 18–45					
				Contri- bution (%)	All individuals		Consumers only		
					µg/kg bw/day	Mean	Median	Mean	Median
7	Fish, seafood, amphibians, reptiles and invertebrates	47	Atlantic salmon	0.65	0.0073	0.0000	0.0000	0.0162	–
7		62	Bivalve molluscs	0.19	0.0021	0.0000	0.0000	0.0047	–
7		67	Canned fish in oil (Tuna)	13.37	0.1509	0.0000	1.1775	0.3340	–
7		68	Canned fish in oil (Sardine)	0.09	0.0011	0.0000	0.0000	0.0024	–
7		45	Catfishes (freshwater)	0.00	0.0000	0.0000	0.0000	0.0000	–
7		52	Cod, atlantic	0.11	0.0012	0.0000	0.0000	0.0027	–
7		65	Cod, dried	14.49	0.1636	0.0000	0.0000	0.3620	–
7		50	Conger European	1.53	0.0173	0.0000	0.0000	0.0382	–
7		57	European Sardine	1.14	0.0128	0.0000	0.0000	0.0284	–
7		66	Fish fingers, breaded	0.08	0.0009	0.0000	0.0000	0.0019	–
7		53	Hakes	31.55	0.3562	0.0000	3.3329	0.7881	–
7		55	Horse mackerel	2.27	0.0257	0.0000	0.0000	0.0568	–
7		54	Ling	0.00	0.0000	0.0000	0.0000	0.0000	–
7		56	Mackerel, chub	0.35	0.0040	0.0000	0.0000	0.0088	–
7		61	Marine shrimps or prawns, cooked	0.20	0.0023	0.0000	0.0000	0.0051	–
7		46	Nile perch	2.57	0.0291	0.0000	0.0000	0.0643	–
7		63	Octopus, common	1.20	0.0135	0.0000	0.0000	0.0300	–
7		49	Other coastal marine fishes (wrasse, pouting, blackspot seabream, red porgy)	0.76	0.0086	0.0000	0.0000	0.0190	–
7		60	Other demersal marine fishes (Scabbardfish)	4.81	0.0543	0.0000	0.0000	0.1202	–
7		59	Other pelagic marine fishes (Forkbeard, red fish)	14.51	0.1639	0.0000	0.0000	0.3626	–
7		51	Plaice, european	1.86	0.0210	0.0000	0.0000	0.0465	–
7		48	Sea Bream	1.45	0.0164	0.0000	0.0000	0.0362	–
7		64	Squid, common	0.47	0.0053	0.0000	0.0000	0.0118	–
7		69	Terrestrial snails, edible	0.00	0.0000	0.0000	0.0000	0.0000	–
7		58	Tuna	0.15	0.0017	0.0000	0.0000	0.0038	–
			Total	93.81	1.06	–	–	2.34	–
			Mean Exposure (µg/kg bw)	–	1.13	–	–	2.50	–
			% of TWI (Mean Exposure)	–	86.9	–	–	192.3	–
			P50 (µg/kg bw)	–	0.00	–	–	–	–
			% of TWI (P50)	–	0.0	–	–	–	–

**Table 6** (continued)

FoodEx2 food group	FoodEx2 level 1 food group	TDS sample Nr	TDS sample	Nr	Female 18–45
					Contri- bution (%)
					All individuals
					µg/kg bw/day
					Mean
					Median
					P95
					Mean
					Median
					P95
					µg/kg bw/day
					Consumers only
					µg/kg bw/day
					Mean
					Median
					P95
					–
			P95 (µg/kg bw)		5.56
			% of TWI (P95)		427.7
			% of persons above the TWI		25.0

*TDS* Total Diet Study, *TWI* Tolerable Weekly Intake, *P50* Percentile 50, *P95* Percentile 95

**Table 7** Estimated dietary exposure of Portuguese overall population ('male and female') to inorganic mercury

FoodEx2 food group Nr	Sex	Male and Female											
		18-74 ('overall population')				18-64 ('adults')				65-74 ('elderly')			
		LB	UB	LB	UB	LB	UB	LB	UB	LB	UB	LB	UB
	Age group	3272				2752				520			
	Nr Individuals	3272				2752				520			
	FoodEx2 level 1 food group	Mean exposure (µg/kg bw/week)		Contribution (%)		Mean exposure (µg/kg bw/week)		Contribution (%)		Mean exposure (µg/kg bw/week)		Contribution (%)	
		LB	UB	LB	UB	LB	UB	LB	UB	LB	UB	LB	UB
2	Alcoholic beverages	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
3	Animal and vegetable fats and oils	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
4	Coffee, cocoa and tea and infusions	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
5	Composite dishes	0.08	0.08	26.72	21.25	0.08	0.07	26.88	20.61	0.09	0.10	26.00	24.32
6	Eggs and egg product	0.00	0.00	0.00	0.09	0.00	0.00	0.00	0.09	0.00	0.00	0.00	0.06
7	Fish, seafood, amphibians, reptiles and invertebrates	0.22	0.22	70.68	58.73	0.21	0.21	70.34	58.30	0.24	0.24	72.30	60.79
9	Fruit and fruit products	0.00	0.01	0.23	2.40	0.00	0.01	0.22	2.34	0.00	0.01	0.27	2.66
10	Fruit and vegetable juices and nectars	0.00	0.01	0.00	3.00	0.00	0.01	0.00	3.30	0.00	0.01	0.00	1.54
11	Grains and grain-based products	0.00	0.01	0.86	4.08	0.00	0.02	0.94	4.36	0.00	0.01	0.51	2.75
12	Legumes, nuts, oilseeds and spices	0.00	0.00	0.04	0.17	0.00	0.00	0.04	0.17	0.00	0.00	0.03	0.15
13	Meat and meat products	0.00	0.01	0.72	2.71	0.00	0.01	0.78	2.92	0.00	0.01	0.43	1.67
14	Milk and dairy products	0.00	0.02	0.27	5.34	0.00	0.02	0.29	5.52	0.00	0.02	0.20	4.48
15	Products for non-standard diets, food imitates and food supplements or fortifying agents	0.00	0.00	0.03	0.08	0.00	0.00	0.04	0.09	0.00	0.00	0.00	0.01
16	Seasoning, sauces and condiments	0.00	0.00	0.02	0.06	0.00	0.00	0.02	0.06	0.00	0.00	0.02	0.04
17	Starchy roots or tubers and products thereof, sugar plants	0.00	0.00	0.00	0.77	0.00	0.00	0.00	0.80	0.00	0.00	0.00	0.65
18	Sugar, confectionery and water-based sweet desserts	0.00	0.00	0.25	0.63	0.00	0.00	0.28	0.70	0.00	0.00	0.11	0.29
19	Vegetables and vegetable products	0.00	0.00	0.16	0.69	0.00	0.00	0.17	0.72	0.00	0.00	0.11	0.56
20	Water and water-based beverages	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	Mean Exposure (mg/kg bw/week)	0.30	0.37	-	-	0.30	0.36	-	-	0.33	0.40	-	-
	% of TWI (Mean Exposure)	7.6	9.2	-	-	7.5	9.0	-	-	8.4	9.9	-	-
	P50 (mg/kg bw/week)	0.03	0.15	-	-	0.03	0.15	-	-	0.05	0.15	-	-
	% of TWI (P50)	0.8	3.6	-	-	0.8	3.6	-	-	1.3	3.7	-	-
	P95 (mg/kg bw/week)	1.26	1.27	-	-	1.25	1.27	-	-	1.37	1.40	-	-
	% of TWI (P95)	31.5	31.7	-	-	31.2	31.6	-	-	34.4	34.9	-	-
	% of persons above the TWI	3.43	3.50	-	-	2.87	2.87	-	-	2.61	6.16	-	-

LB Lower bound approach, UB Upper bound approach, TWI Tolerable Weekly Intake, P50 Percentile 50, P95 Percentile 95

**Table 8** Estimated dietary exposure of Portuguese male population to inorganic mercury

FoodEx2 food group Nr	Sex	Male		18–74		18–64 ('adults')		65–74 ('elderly')		Mean exposure (µg/kg bw/week)	Contribution (%)	Mean exposure (µg/kg bw/week)	Contribution (%)
		Age group	Nr Individuals	LB	UB	LB	UB	LB	UB				
		18–74	1583			1363		220					
				Mean exposure (µg/kg bw/week)	Contribution (%)	Mean exposure (µg/kg bw/week)	Contribution (%)	Mean exposure (µg/kg bw/week)	Contribution (%)	Mean exposure (µg/kg bw/week)	Contribution (%)	Mean exposure (µg/kg bw/week)	Contribution (%)
		LB	UB	LB	UB	LB	UB	LB	UB	LB	UB	LB	UB
2		0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
3		0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
4		0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
5		0.09	0.08	29.08	23.31	0.09	0.08	29.63	22.91	0.11	0.12	26.59	25.18
6		0.00	0.00	0.00	0.09	0.00	0.00	0.00	0.10	0.00	0.00	0.00	0.06
7		0.21	0.21	68.31	57.68	0.20	0.20	67.51	56.61	0.29	0.29	71.94	62.71
9		0.00	0.01	0.20	2.04	0.00	0.01	0.19	1.97	0.00	0.01	0.23	2.36
10		0.00	0.01	0.00	2.76	0.00	0.01	0.00	3.13	0.00	0.00	0.00	1.00
11		0.00	0.01	0.84	4.05	0.00	0.02	0.94	4.43	0.00	0.01	0.39	2.29
12		0.00	0.00	0.05	0.21	0.00	0.00	0.05	0.22	0.00	0.00	0.03	0.15
13		0.00	0.01	0.77	2.99	0.00	0.01	0.85	3.28	0.00	0.01	0.43	1.64
14		0.00	0.02	0.30	4.65	0.00	0.02	0.32	4.93	0.00	0.02	0.20	3.34
15		0.00	0.00	0.02	0.04	0.00	0.00	0.02	0.05	0.00	0.00	0.00	0.00
16		0.00	0.00	0.03	0.07	0.00	0.00	0.03	0.08	0.00	0.00	0.01	0.01
17		0.00	0.00	0.00	0.83	0.00	0.00	0.00	0.90	0.00	0.00	0.00	0.52
18		0.00	0.00	0.26	0.66	0.00	0.00	0.30	0.75	0.00	0.00	0.10	0.25
19		0.00	0.00	0.13	0.59	0.00	0.00	0.15	0.63	0.00	0.00	0.07	0.42
20		0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
		0.31	0.36	-	-	0.29	0.35	-	-	0.40	0.46	-	-
		7.7	9.1	-	-	7.3	8.7	-	-	10.0	11.5	-	-
		0.03	0.14	-	-	0.03	0.13	-	-	0.08	0.15	-	-
		0.8	3.4	-	-	0.7	3.4	-	-	2.1	3.8	-	-
		1.26	1.29	-	-	1.23	1.27	-	-	1.49	1.55	-	-
		31.5	32.2	-	-	30.8	31.7	-	-	37.2	38.7	-	-
		3.64	3.71	-	-	2.66	2.80	-	-	8.47	8.61	-	-

LB Lower bound approach, UB Upper bound approach, TWI Tolerable Weekly Intake, P50 Percentile 50, P95 Percentile 95

**Table 9** Estimated dietary exposure of Portuguese female population to inorganic mercury

Food group Nr	Sex	Female					
		18–74		18–64 ('adults')		65–74 ('elderly')	
		1689		1389		300	
Nr Individuals							
FoodEx2 level 1 food group		Mean exposure (µg/kg bw/week)		Mean exposure (µg/kg bw/week)		Mean exposure (µg/kg bw/week)	
		LB	UB	LB	UB	LB	UB
		Contribution (%)		Contribution (%)		Contribution (%)	
		LB	UB	LB	UB	LB	UB
2	Alcoholic beverages	0.00	0.00	0.00	0.00	0.00	0.00
3	Animal and vegetable fats and oils	0.00	0.00	0.00	0.00	0.00	0.00
4	Coffee, cocoa and tea and infusions	0.00	0.00	0.00	0.00	0.00	0.00
5	Composite dishes	0.07	0.07	24.48	19.35	0.07	0.07
6	Eggs and egg product	0.00	0.00	0.00	0.09	0.00	0.00
7	Fish, seafood, amphibians, reptiles and invertebrates	0.22	0.22	72.94	59.69	0.22	0.22
9	Fruit and fruit products	0.00	0.01	0.25	2.73	0.00	0.01
10	Fruit and vegetable juices and nectars	0.00	0.01	0.00	3.22	0.00	0.01
11	Grains and grain-based products	0.00	0.02	0.89	4.12	0.00	0.02
12	Legumes, nuts, oilseeds and spices	0.00	0.00	0.03	0.13	0.00	0.00
13	Meat and meat products	0.00	0.01	0.66	2.44	0.00	0.01
14	Milk and dairy products	0.00	0.02	0.24	5.97	0.00	0.02
15	Products for non-standard diets, food imitates and food supplements or fortifying agents	0.00	0.00	0.05	0.12	0.00	0.00
16	Seasoning, sauces and condiments	0.00	0.00	0.02	0.05	0.00	0.00
17	Starchy roots or tubers and products thereof, sugar plants	0.00	0.00	0.00	0.72	0.00	0.00
18	Sugar, confectionery and water-based sweet desserts	0.00	0.00	0.24	0.59	0.00	0.00
19	Vegetables and vegetable products	0.00	0.00	0.19	0.78	0.00	0.00
20	Water and water-based beverages	0.00	0.00	0.00	0.00	0.00	0.00
	Mean Exposure (mg/kg bw/week)	0.30	0.37	0.31	0.37	0.29	0.35
	% of TWI (Mean Exposure)	7.5	9.2	7.6	9.3	7.1	8.8
	P50 (mg/kg bw/week)	0.04	0.16	0.04	0.16	0.02	0.15
	% of TWI (P50)	0.9	3.9	0.9	4.0	0.6	3.7
	P95 (mg/kg bw/week)	1.24	1.23	1.29	1.26	1.03	1.09
	% of TWI (P95)	31.1	30.9	32.2	31.5	25.8	27.3
	% of persons above the TWI	3.15	3.15	2.87	2.87	3.01	3.15

LB Lower bound approach, UB Upper bound approach, TWI Tolerable Weekly Intake, P50 Percentile 50, P95 Percentile 95

**Table 10** Main foods contributing to inorganic mercury exposure

Sex	Male and Female						Male						Female					
	18-74		18-64		65-74		18-74		18-64		65-74		18-74		18-64		65-74	
	LB	UB	LB	UB	LB	UB	LB	UB	LB	UB	LB	UB	LB	UB	LB	UB	LB	UB
Cod	15.01	12.47	13.95	11.56	20.03	16.85	15.83	13.37	14.25	11.95	22.96	20.96	14.22	11.64	13.66	11.20	17.03	13.81
Hake	11.76	9.77	11.90	9.86	11.10	9.33	8.45	7.13	8.63	7.23	7.64	6.66	14.91	12.20	14.96	12.27	14.64	11.88
Mixed fish stew	8.97	7.45	8.78	7.28	9.86	8.29	10.02	8.46	9.13	7.65	14.03	12.23	7.97	6.53	8.46	6.94	5.57	4.52
Scabbardfish	7.10	5.90	6.64	5.51	9.26	7.78	10.02	8.46	9.13	7.65	14.03	12.23	7.97	6.53	8.46	6.94	5.57	4.52
Canned fish in oil (Tuna)	5.88	4.89	6.67	5.53	2.17	1.83	5.34	4.51	5.98	5.01	2.47	2.15	6.40	5.23	7.31	6.00	1.86	1.51
Mixed cod stew	1.27	1.06	0.44	0.37	5.19	4.37	1.47	1.24	0.91	0.76	3.99	3.48	1.08	0.88	0.00	0.00	6.42	5.21
Octopus	4.20	3.49	4.57	3.78	2.48	2.08	5.61	4.73	6.01	5.04	3.78	3.29	2.86	2.34	3.21	2.63	1.14	0.93
Cod with cream	5.17	0.43	6.09	0.50	0.82	0.07	5.29	0.45	6.10	0.51	1.63	0.14	5.05	0.41	6.07	0.5	0.00	0.00
Fish	4.35	3.62	4.39	3.64	4.21	3.54	3.54	2.99	3.89	3.26	1.97	1.72	5.14	4.21	4.87	3.99	6.51	5.28

UB - Upper bound approach and LB - Lower bound approach

(Dofkova et al. 2016) to harmonise food and sample lists across countries. However, future agreement on minimum requirements to report exposure assessment results would be valuable to allow for improved international comparison, namely food grouping (food classification), assumptions on occurrence (elements speciation), population age groups, sex, descriptive statistics and overall population vs consumers-only.

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**Data Availability** The authors declare that the data supporting the findings of this study are available within the paper and it Supplementary Information file.

## Declarations

**Conflict of interest** The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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