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Scientific update on the iodine content of Portuguese foods

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Abstract

Iodine is an essential trace element in human and animal diets. However, mild to moderate iodine deficiency has been reported in several countries. Food is the natural source of iodine. Detectable analytical values, expressed in SI units (µg/kg), are required to guarantee reliable measurement results used to estimate iodine intake over time at national and international level. The aim of this work, conducted as an activity of the WHO Collaborating Centre for Nutrition and Childhood Obesity, was to develop a database of the iodine content of foods in order to predict nutritional adequacy of dietary intake. This database may be used as a tool to promote iodine intake through consumption of foods rich in iodine.

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Abbreviations

EuroFIR	European Food Information Resource
ICP-MS	inductively coupled plasma mass spectrometry
LC	left-censored
LoD	limit of detection
LoQ	limit of quantification
RNI	recommended nutrient intake

Executive summary

Iodine is an essential trace element in human and animal diets. However, mild to moderate iodine deficiency has been reported in several countries. Food is the natural source of iodine. Detectable analytical values, expressed in SI units (μg/kg), are required to guarantee reliable measurement results used to estimate iodine intake over time at national and international level. The aim of this work, conducted as an activity of the WHO Collaborating Centre for Nutrition and Childhood Obesity, was to develop a database of the iodine content of foods in order to predict nutritional adequacy of dietary intake. This database may be used as a tool to promote iodine intake through consumption of foods rich in iodine.

The specific objective of this report is to provide updated data on the iodine content of Portuguese foods as consumed within, and as representative of, the Portuguese diet. The methodology selected for quantification of this nutrient was inductively coupled plasma mass spectrometry (ICP-MS) assisted by microwave after alkaline digestion.

Eight groups of food were analysed: (1) meat; (2) fish and seafood; (3) milk and milk products, eggs; (4) fruit; (5) vegetables and pulses; (6) sweets and beverages; (7) cereals and tubers; and (8) meals. These foods were collected on the basis of consumption patterns drawn from national food consumption surveys (Fabrice Elegbede et al., 2017). The proportion of samples beyond the limit of detection (LoD) ranged between 0% in fish, seafood and dairy products to 81.3% in fruit. The food samples with detectable iodine content showed a wide variation: 2.4–7.8 μg/100 g for meat; 2.8–289.3 μg/100 g for fish and seafood; 15.8–39.4 μg/100 g for milk, milk products and eggs; 3.3–26.6 μg/100 g for fruit; 0.3–6.5 μg/100 g for vegetables and pulses; 0.3–22.7 μg/100 g for sweets and beverages; 0.9–4.7 μg/100 g for cereals and tubers; and 0.7–56.8 μg/100 g for meals. The results showed that in Portugal a diet rich in fish, seafood and dairy products supplies the recommended daily intake of iodine for a healthy adult.

1. Background

Iodine is an essential trace element and its nutritional importance is well established. It is required for the synthesis of the thyroid hormones thyroxine and its active form T3, as well as the precursor iodotyrosines. Iodine deficiency is the most common cause of preventable mental impairment. Chronic iodine deficiency can lead to disorders including mental impairment and retardation, and formation of goitre (thyroid dysfunction), an enlargement of the thyroid gland which implies inadequate production of thyroid hormones (Rohner et al., 2014). In Europe, several initiatives have been implemented to decrease incidence of iodine deficiency. The iodized salt programme has been successful in reducing iodine deficiency. However, many countries are following salt reduction programmes, and as a consequence work has to be undertaken to identify other dietary iodine sources (Gärtner, 2016; WHO, 2012).

Food is the natural source of iodine. Detectable analytical values, expressed in SI units (µg/kg), are required to guarantee reliable measurement results which are used to estimate iodine intake over time at national and international level (Leufroy et al., 2015).

High intakes of iodine can cause some of the same symptoms as iodine deficiency, including goitre, elevated thyroid-stimulating hormone (TSH) levels, and hypothyroidism, because excess iodine in susceptible individuals inhibits thyroid hormone synthesis and thereby increases TSH stimulation, which can produce goitre. Iodine-induced hyperthyroidism can also result from high iodine intakes, usually when iodine is administered to treat iodine deficiency. Studies have also shown that excessive iodine intakes cause thyroiditis and thyroid papillary cancer. Cases of acute iodine poisoning are rare and are usually caused by doses of many grams. Acute poisoning symptoms include burning of the mouth, throat and stomach; fever; abdominal pain; nausea; vomiting; diarrhoea; weak pulse; and coma (Zimmermann & Boelaert, 2015).

Responses to excess iodine and the doses required to cause adverse effects vary. Some people, such as those with autoimmune thyroid disease and iodine deficiency, may suffer adverse effects with iodine intakes considered safe for the general population (Zimmermann & Boelaert, 2015).

Nutritional needs should be met primarily from foods. A healthy eating pattern is one that includes a variety of vegetables, fruits, whole grains, fat-free or low-fat milk and milk products, and oils (WHO, 2015). Milk is an excellent source of iodine. Fruits, vegetables and bread also provide small quantities of iodine. Some fish contain high amounts of iodine, while eggs are a good source too (Leufroy et al., 2015).

Different analytical methods and principles for gathering qualitative and quantitative information on iodine food samples are reported. Inductively coupled plasma mass spectrometry (ICP-MS) is the gold standard for determination of iodine in foods; this is due to the method performance parameters, such as a high level of accuracy, reproducibility and repeatability, associated with a low limit of quantification (Moreda-Piñeiro, Romarís-Hortas, & Bermejo-Barrera, 2011). A data quality evaluation system (DQES) was developed to ensure comparability and data management and to allow interchange with available systems already in place, such as the European Food Information Resource (EuroFIR) (Castanheira et al., 2016; Westenbrink et al., 2009).

In Portugal, recent studies have shown iodine deficiencies in pregnant women and a low iodine intake in more than 50% of Portuguese children, particularly in the Azores and Madeira regions (Costa Leite et al., 2017; Limbert et al., 2012). In Madeira only 8.2% of pregnant women presented adequate iodine intake values, and in the Azores this value was even lower, at only 1.4% (Limbert et al., 2012).

Thus, in 2013 the Health General Directorate (Direção Geral de Saúde (DGS)) recommended that the diet of pregnant women be supplemented with potassium iodate (Direção-Geral da Saúde, 2013); and in the same year the Education General Directorate (Direção Geral da Educação (DGE)) introduced iodized salt in school meals (Direção-Geral da Educação, 2013).

2. Materials and methods

2.1 Sampling plan

The collection of foods for analysis followed a sampling plan representative of the Portuguese diet, as outlined in the Total Diet Study Exposure project (Dofkova et al., 2016; Fabrice Elegbede et al., 2017). This plan included eight food groups: (1) meat; (2) fish and seafood; (3) milk and milk products, eggs; (4) fruit; (5) vegetables and pulses; (6) sweets and beverages; (7) cereals and tubers; and (8) meals. Before collection, the sampling plan was designed on the basis of a food consumption survey, representing 85% of the foods most consumed in Portugal. The shopping list was created in order to collect the most consumed brands across all regions. After reception in the laboratory, samples were divided into two groups: those consumed raw and those consumed cooked (Supplementary table 1). The 1284 foods collected were analysed, as consumed, in 107 pooled samples, each consisting of 12 identical foods.

2.2 Sample acquisition and categorization

Foods were categorized in eight groups in accordance with the food classification system adopted by EFSA (Food Ex2) (EFSA, 2015). The classification (as detailed in the previous section) covered most significant food items. This was created on the basis of consumption patterns shown in national food consumption surveys that represent the average diet of the Portuguese population (70–95% of the total diet) (Fabrice Elegbede et al., 2017). Food samples, either raw or as consumed, were purchased from various popular supermarkets in the Lisbon metropolitan area.

2.3 Sample preparation

Samples were analysed in the form that they are consumed. For this purpose, raw samples were cooked following traditional recipes without iodized salt and according to known consumption patterns. After preparation, the edible parts of the samples, raw or cooked, were grouped into pools of 12 samples composed of identical foods. However, a single pool could contain foods prepared in a number of different ways. For example, the pool containing chicken comprised four different chicken recipes according to the customary Portuguese food consumption pattern. Pooled samples combined groups of different brands or cultivars (e.g. 12 apples or 12 yoghurts). Others, such as the carrot pool, contained both raw and cooked forms, because they are consumed in both ways by the Portuguese population.

2.4 Iodine analysis

The iodine content was determined by ICP-MS, with reference to EN 15111:2007 (European Committee for Standardization, 2007). Briefly, 0.5–1 g samples were weighed into a vessel or a 50 ml tube. The extraction of iodine was performed by a graphite block system over three hours at 90 °C with TMAH (tetramethylammonium hydroxide) solution. After extraction, all samples were centrifuged and filtered through 0.45 μm filters. Starchy samples were pretreated overnight with an α-amylase solution at 37 °C before TMAH extraction.

2.5 Quality control

The results were obtained in triplicate analytical samples under conditions of quality assurance supported by the requirements described in NP EN ISO/IEC 17025:2005 (European Committee for Standardization, 2005). The iodine content was expressed in μg of iodine per 100 g of food, following EuroFIR recommendations for modes of expression for nutrients entered into food composition databases (Westenbrink et al., 2016).

The quantification of iodine content was carried out using a calibration curve between 0.5 μg/L and 50 μg/L prepared with iodine standards. The correlation coefficient was ≥ 0.9995. The limit of detection (LoD) was 0.13 μg/L. To demonstrate laboratory competence, proficiency test schemes (FAPAS) were performed. For FAPAS 1881 (milk powder), 07238 and 07260 (infant formula), the Z-scores obtained were –0.2, –0.1 and 0.0, respectively. Samples were analysed in batches including several internal quality controls to guarantee the reliability of each assay and to monitor the drift of equipment.

2.6 Recommended nutrient intake

The recommended nutrient intake (RNI) of iodine for a healthy adult was assumed to be 150 μg per day (WHO, 2007); the portion size 100 g or 100 ml. On this basis, the contribution of each food was calculated.

3. Results and discussion

The concentration of iodine in Portuguese food samples showed a wide variation according to food group, from < 0.05 µg/100 g in beverages to 289 µg/100 g in fish and seafood. The left-censored (LC) data of our samples were 35% (37/106). The main sources of iodine were shellfish and fish, and milk and milk products; the groups with the lowest iodine content were fruit, and vegetables and pulses. These results are aligned with the literature concerning iodine content by food group (Haldimann et al., 2005; Leufroy et al., 2015; Rose et al., 2001).

3.1 Meat

In this group of foods, seven different types of meat were analysed (Table 1). Four samples presented values below the limit of quantification (LoQ), with 57.1% LC data. The mean iodine content found in meat samples with detectable iodine content is low, ranging from 2.4 µg/100 g in chicken to 7.8 µg/100 g in rabbit, thus contributing, respectively, 1.6% and 5.2% (on average) of recommended daily intake. These values are more or less consistent with the literature. Haldimann et al. (2005) and Rose et al. (2001) found average values for meat higher than ours, at 12 and 10 µg/100 g, respectively.

3.2 Fish and seafood

A total of 27 samples of fish and seafood presented the highest values of iodine content (Table 2). In fish samples, the variation in iodine content is probably due to factors such as the different habitat types of fish – marine, freshwater and aquaculture (Fuge & Johnson, 2015; Lee et al., 1994; Risher & Keith, 2009). In this group there are no LC data. Table 2 shows that in fish and seafood there is a large variation in iodine content, ranging from 2.8 to 289 µg/100 g. In terms of contribution to RNI, the group ranges between 1.9% and 192.9%; mussels and clams, with the highest values for iodine, contribute more than 100% of RNI. The different habitat types of fish are presented in Supplementary table 2.

3.3 Milk and milk products, eggs

In this group, six samples were analysed and, as observed in fish, all samples contained iodine in values above LoQ (Table 3). Iodine content ranged between 15.8 µg/100 g (milk) and 39.4 µg/100 g (cheese). When calculated as a proportion of RNI, dairy foods ranged from 10.5% to 26.3%. These results are in accordance with the literature (Haldimann et al., 2005; Tinggi et al., 2012).

Table 1. Iodine content of meat expressed as mean and standard deviation (SD) of three replicates

Sample	Mean ± SD (µg/100g)	LoD (µg/100 g)	LoQ (µg/100 g)	Contribution to RNI (%)
Beef	<LoQ	0.64	2.01	–
Chicken	2.4 ± 0.2	0.63	1.96	1.6
Lamb	7.5 ± 0.4	0.63	1.97	5.0
Pork	<LoQ	0.65	2.03	–
Rabbit	7.8 ± 0.6	0.64	2.00	5.2
Turkey	<LoQ	0.65	2.03	–
Veal	<LoQ	0.64	2.01	–

Table 2. Iodine content of fish and seafood expressed as mean and SD of three replicates

Sample	Mean ± SD (µg/100 g)	LoD (µg/100 g)	LoQ (µg/100 g)	Contribution to RNI (%)
Canned sardine	24.3 ± 0.1	0.54	2.06	16.2
Canned tuna	19.7 ± 0.6	0.54	2.07	13.1
Catfish	2.8 ± 0.3	0.60	1.89	1.9
Clams	157 ± 6	0.53	2.05	104.6
Codfish	56.5 ± 1.0	0.53	2.05	37.7
European conger	22.5 ± 0.7	0.65	2.04	15.0

Table 2. cont.

Sample	Mean ± SD (µg/100 g)	LoD (µg/100 g)	LoQ (µg/100 g)	Contribution to RNI (%)
Fresh cod	138 ± 1	0.53	2.05	91.9
Fresh tuna	16.7 ± 0.3	0.54	2.06	11.1
Gadiformes Mix 1	131 ± 1	0.54	2.06	87.3
Gadiformes Mix 2	21.5 ± 0.4	0.66	2.06	14.3
Gilt-head bream	17.3 ± 0.4	0.63	1.95	11.5
Hake	9.7 ± 0.3	0.54	2.06	6.5
Horse mackerel	44.4 ± 1.0	0.53	2.05	29.6
Ling	27.2 ± 1.3	0.62	1.95	18.1
Mackerel	40.5 ± 0.3	0.53	2.06	27.0
Mussel	289 ± 18	3.26	10.17	192.9
Nile perch	13.1 ± 0.3	0.63	1.96	8.7
Octopus	13.1 ± 0.5	0.54	2.06	8.7
Red porgy	88.2 ± 0.2	0.64	2.01	58.8
Salmon	10.5 ± 0.6	0.54	2.06	7.0
Sardine	26.0 ± 0.2	0.54	2.06	17.3
Sargo	82.1 ± 4.9	0.65	2.04	54.7
Sea bass	17.9 ± 0.4	0.64	2.01	11.9
Shrimp	71.4 ± 2.2	0.53	2.05	47.6
Sole	19.3 ± 0.2	0.53	2.02	12.9
Squid	22.5 ± 0.5	1.45	4.52	15.0
Swordfish	24.9 ± 1.9	0.63	1.96	16.6

Table 3. Iodine content of dairy products and eggs expressed as mean and SD of three replicates

Sample	Mean ± SD (µg/100 g)	LoD (µg/100 g)	LoQ (µg/100 g)	Contribution to RNI (%)
Cheese	39.4 ± 2.0	0.19	7.44	26.3
Eggs	24.3 ± 2.2	0.21	0.81	16.2
Milk	15.8 ± 1.2	0.08	3.23	10.5
Milk with cereals, milk with chocolate	19.9 ± 0.3	0.14	0.44	13.3
Yoghurt	17.9 ± 0.2	0.08	3.00	11.9
Yoghurt with fruit, cereals	17.2 ± 0.2	0.08	3.03	11.5

3.4 Fruit

The 16 samples analysed in the fruit group present some of the lowest values for iodine content, with 81.3% LC data (Table 4). Fruit salad had the highest content of iodine, at 26.6 μg/100 g. Where detectable, the contribution of fruit to RNI was therefore very low, ranging between 2.2% to 17.7%. Fruits consumed in Portugal are not as good a source of iodine as fruits consumed in other European countries (Leufroy et al., 2015; Rose et al., 2001).

Table 4. Iodine content of fruit expressed as mean and SD of three replicates

Sample	Mean ± SD (μg/100 g)	LoD (μg/100 g)	LoQ (μg/100 g)	Contribution to RNI (%)
Apple	<LoQ	0.54	1.69	–
Banana	<LoQ	0.61	3.84	–
Canned peach and pineapple	<LoQ	0.58	3.34	–
Cantaloupe	<LoQ	0.06	0.18	–
Dried fig	3.3 ± 0.1	0.61	1.90	2.2
Fruit salad	26.6 ± 0.5	0.60	1.86	17.7
Grape	<LoQ	0.47	1.45	–
Kiwi	<LoQ	0.59	3.66	–
Melon	<LoQ	0.09	0.28	–
Orange	<LoQ	0.10	0.61	–
Peach	<LoQ	0.60	3.73	–
Pear	<LoQ	0.11	0.68	–
Pineapple	<LoQ	0.59	3.68	–
Raisins	5.5 ± 0.6	0.47	1.47	3.7
Strawberry	<LoQ	0.60	3.76	–
Watermelon	<LoQ	0.09	0.29	–

3.5 Vegetables and pulses

As Table 5 shows, vegetables and pulses are a very poor source of iodine. A total of 23 samples were analysed in this group; 14 of these were below LoQ, with 60.9% LC data. The iodine content of vegetables and pulses ranged from 0.3 to 6.5 μg/100 g. In terms of RNI, they range between 0.2% to 4.3% and make a lower contribution than fruit. These results have already been demonstrated in the literature (Haldimann et al., 2005; Leufroy et al., 2015; Rose et al., 2001).

Table 5. Iodine content of vegetables and pulses expressed as mean and SD of three replicates

Sample	Mean ± SD (μg/100 g)	LoD (μg/100 g)	LoQ (μg/100 g)	Contribution to RNI (%)
Asparagus	6.5 ± 0.4	0.53	1.67	4.3
Broadbean	<LoQ	0.27	0.90	–
Broccoli	0.4 ± 0.01	0.08	0.23	0.3
Brussels sprout	<LoQ	0.54	1.70	–
Carrot	0.7 ± 0.01	0.08	0.23	0.5
Cauliflower	<LoQ	0.36	1.13	–
Chickpea	<LoQ	0.07	0.23	–
Corn	<LoQ	0.52	1.62	–

Table 5. cont.

Sample	Mean ± SD (µg/100 g)	LoD (µg/100 g)	LoQ (µg/100 g)	Contribution to RNI (%)
Cowpea	0.3 ± 0.1	0.07	0.22	0.2
Green bean	0.4 ± 0.2	0.08	0.23	0.3
Lettuce	1.5 ± 0.01	0.10	0.32	1.0
Lupines	0.3 ± 0.01	0.07	0.23	0.2
Mushroom	<LoQ	0.52	1.63	–
Olive	4.1 ± 0.1	0.08	0.23	2.7
Onion	<LoQ	0.53	1.67	–
Pea	<LoQ	0.07	0.23	–
Pepper	<LoQ	0.12	0.39	–
Portuguese cabbage	<LoQ	0.56	1.73	–
Red bean, white, butter, black	<LoQ	0.07	0.23	–
Sprout	<LoQ	0.58	1.83	–
Tomato	<LoQ	0.10	0.32	–
Turnip greens	1.9 ± 0.2	0.60	1.87	1.3
White cabbage	<LoQ	0.56	1.74	–

3.6 Sweets and beverages

The samples of beverages analysed showed very low levels of iodine (Table 6); this result had already been reported in the literature (Leufroy et al., 2015). In the case of sweets, one sample presented a value below LoQ. The iodine content of crème brûlée and flan is high, at 22.7 µg/100 g; in terms of RNI, this is a contribution of 15.1%. This high value could be explained by the fact that this type of dessert has eggs and milk (or cream) as ingredients.

Table 6. Iodine content of sweets and beverages expressed as mean and SD of three replicates

Sample	Mean ± SD (µg/100 g)	LoD (µg/100 g)	LoQ (µg/100 g)	Contribution to RNI (%)
Beverage, soy-based	0.6 ± 0.03	0.01	0.31	0.4
Crème brûlée and flan	22.7 ± 0.03	0.30	0.93	15.1
Milk bread	4.2 ± 0.2	0.25	0.78	2.8
Mineral water	0.3 ± 0.03	0.02	0.05	0.0
Quince jam	<LoQ	0.56	1.74	–
Yeast-leavened cakes	4.8 ± 0.1	0.25	0.82	3.2

3.7 Cereals and tubers

A total of seven samples were analysed in this group, with 57.1% LC data (Table 7). Croissants presented the highest iodine content, at 4.7 µg/100 g. Four samples presented values below LoQ; of the rest, the lowest value, 0.9 µg/100 g, was presented by white bread and by breakfast cereals and chocolate breakfast cereals. The RNI contribution of the group ranged between 0.6% and 3.1%.

3.8 Meals

A total of 15 samples were analysed in this group, with 6.7% LC data (Table 8). Soups of vegetables and pulses were the meals with the lowest values, 0.7 µg/100 g. Meals with fish and seafood presented the highest iodine content, ranging from 8.4 to 56.8 µg/100 g. In terms of RNI contribution, this group supplied 0.5% to 37.9%.

Table 7. Iodine content of cereals and tubers expressed as mean and SD of three replicates

Sample	Mean ± SD (µg/100 g)	LoD (µg/100 g)	LoQ (µg/100 g)	Contribution to RNI (%)
Breakfast cereals and chocolate breakfast cereals	0.9 ± 0.1	0.25	0.82	0.6
Corn bread	<LQ	0.25	0.78	–
Croissant	4.7 ± 0.2	0.25	0.82	3.1
Pasta	<LQ	0.25	0.77	–
Potato	<LQ	0.11	0.34	–
Rice	<LQ	0.25	0.77	–
Wheat bread	0.9 ± 0.1	0.25	0.82	0.6

Table 8. Iodine content of meals expressed as mean and SD of three replicates

Sample	Mean ± SD (µg/100 g)	LoD (µg/100 g)	LoQ (µg/100 g)	Contribution to RNI (%)
Cod oven-based food	15.9 ± 1.3	0.16	0.60	10.6
Cod-based food	23.6 ± 0.6	0.20	0.61	15.7
Fish and potato meal	9.2 ± 0.02	1.01	3.15	6.1
Fish fingers, breaded	28.1 ± 0.2	0.67	2.08	18.7
Fish rice	8.4 ± 0.8	0.84	2.63	5.6
Fish salad	8.4 ± 0.1	0.26	1.01	5.6
Hamburger	3.5 ± 0.04	0.62	1.94	2.3
Meatballs	8.3 ± 0.6	0.62	1.93	5.5
Pulses soup	0.7 ± 0.04	0.10	0.32	0.5
Seafood rice	10.3 ± 0.3	0.28	0.86	6.9
Seafood soup	15.0 ± 1.4	0.55	1.73	10.0
Soy-based food	<LoQ	0.63	1.98	–
Sushi	56.8 ± 3.2	0.64	2.01	37.9
Vegetables rice	1.3 ± 0.1	0.25	0.77	0.9
Vegetables soup	0.7 ± 0.01	0.10	0.31	0.5

4. Conclusions

The analysis shows that fish and seafood provide a good source of iodine; dairy products also make a significant contribution to the RNI of iodine. In sum, the data allow us to conclude that the RNI of iodine for healthy, non-pregnant adults – 150 µg/day – is best provided by a diet rich in fish, seafood and dairy products.

The analytical method reported in this work shows the importance of determining iodine content in micrograms per portion (µg/100 g). Given its LoQ, the performance of ICP-MS is adequate to assess iodine content across a wide range of foods.

Quantifying iodine in food as it is consumed allows daily intake and relative contribution from different foods and food groups to be estimated; this kind of data is very useful as there is a lack of data on retention of iodine in food when cooked. Furthermore, these results concerning iodine content in representative food groups analysed as they are consumed can be combined with food consumption data to estimate actual dietary intake of iodine for all groups of the Portuguese population.

The data quality procedures applied in this work are very useful. The accuracy of nutrient values produced allows interchange of data across countries. Food composition data on iodine present a particular challenge, as several industrialized countries have introduced iodized salt programmes to reduce iodine deficiency while at the same time following salt-reduction programmes. Accurate and reliable iodine values are crucial if we are to make progress with monitoring the risks and benefits of iodine intake.

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6. Supplementary tables

Supplementary table 1. Type of processing method used for each sample

Sample	Processing method
Hamburger	Cooked, grilled, roasted and fried
Meatballs	Cooked, fried and stewed
Cod oven-based food	Cooked, roasted
Seafood rice	Cooked, stewed
Fish salad	Raw and cooked, boiled
Cod-based food	Cooked, roasted
Fish and potato meal	Cooked, roasted
Fish rice	Cooked, stewed and boiled
Vegetables rice	Cooked, stewed and boiled
Pulses soup	Cooked, boiled
Vegetables soup	Cooked, boiled
Seafood soup	Cooked, boiled
Eggs	Cooked, scrambled, fried and poached
Catfish	Cooked, fried and roasted
Nile perch	Cooked, boiled, roasted and grilled
Salmon	Cooked, roasted and grilled
Gilt-head bream	Cooked, roasted and grilled
Gadiformes Mix 1	Cooked, fried, roasted and grilled
European conger	Cooked, boiled and roasted
Sole	Cooked, fried, roasted and grilled
Fresh cod	Cooked, boiled, grilled and roasted
Hake	Cooked, boiled, fried and roasted
Ling	Cooked, boiled and stewed
Horse mackerel	Cooked, fried, roasted and grilled
Mackerel	Cooked, boiled, grilled and roasted
Sardine	Cooked, fried and grilled
Fresh tuna	Cooked, grilled
Gadiformes Mix 2	Cooked, boiled and roasted
Swordfish	Cooked, fried and grilled
Shrimp	Cooked, boiled and roasted
Clams	Cooked, boiled
Octopus	Cooked, boiled and roasted
Squid	Cooked, fried, grilled and curried
Codfish	Cooked, boiled and roasted
Fish fingers, breaded	Cooked, fried and roasted

Supplementary table 1 cont.

Sample	Processing method
Canned tuna	Canned in oil
Canned sardine	Canned in oil
Orange	Raw
Apple	Raw
Pear	Raw
Strawberry	Raw
Peach	Raw
Kiwi	Raw
Banana	Raw
Pineapple	Raw
Raisins	Uncooked
Dried fig	Uncooked
Quince jam	Cooked, boiled
Fruit salad	Raw
Canned peach and pineapple	Canned
Rice	Cooked, boiled
Corn bread	Cooked
Wheat bread	Cooked
Pasta	Cooked, boiled
Yeast-leavened cakes	Cooked
Milk bread	Cooked
Croissant	Cooked
Breakfast cereals and chocolate breakfast cereals	Uncooked
Broadbean	Cooked, boiled and stewed
Pea	Cooked, boiled and stewed
Red bean, white, butter, black	Cooked, boiled
Chickpea	Cooked, boiled
Cowpea	Cooked, boiled
Lupines	Processed
Olive	Processed
Beef	Cooked, fried and grilled
Veal	Cooked, fried and grilled
Pork	Cooked, fried, roasted and grilled
Lamb	Cooked, roasted and stewed
Rabbit	Cooked, roasted and stewed
Chicken	Cooked, boiled, roasted and grilled
Turkey	Cooked, grilled and roasted
Milk	Fluid

Supplementary table 1 cont.

Sample	Processing method
Milk with cereals, milk with chocolate	Fluid
Yoghurt	Plain
Yoghurt with fruit, cereals	Plain
Cheese	Soft
Crème brûlée and flan	Cooked
Soy-based food	Cooked, grilled and roasted
Beverage, soy-based	Fluid
Potato	Cooked, boiled
Broccoli	Cooked, boiled
Cauliflower	Cooked, boiled
Brussels sprout	Cooked, boiled
White cabbage	Cooked, boiled
Portuguese cabbage	Cooked, boiled
Onion	Raw
Asparagus	Cooked, boiled
Tomato	Raw
Pepper	Raw
Cantaloupe	Raw
Melon	Raw
Watermelon	Raw
Lettuce	Raw
Sprout	Cooked, boiled
Turnip greens	Cooked, boiled
Green bean	Cooked, boiled
Carrot	Raw and cooked, boiled
Mushroom	Cooked, grilled and roasted
Corn	Cooked, boiled
Mineral water	Thermal water
Sushi	Cooked
Red porgy	Cooked, grilled and roasted
Mussel	Cooked
Sea bass	Cooked, grilled and roasted
Sargo	Cooked, grilled and roasted
Grape	Raw

Supplementary table 2. Habitat types of fish and seafood group

Sample	Open-sea	Aquaculture (open-sea)	Coastal	Aquaculture (coastal)
Catfish				×
Nile perch			×	
Salmon		×		
Gilt-head bream	×	×		
Gadiformes Mix 1	×			
European conger	×			
Sole	×			
Fresh cod	×			
Hake	×			
Ling	×			
Horse mackerel	×			
Mackerel	×			
Sardine	×			
Fresh tuna	×			
Gadiformes Mix 2	×			
Swordfish	×			
Shrimp	×	×		
Clams	×	×		
Octopus	×			
Squid	×			
Codfish	×			
Canned tuna	×			
Canned sardine	×			
Red porgy	×			
Mussel	×	×		
Sea bass	×	×		
Sargo	×			

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