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Total diet study of iodine and the contribution of milk in the exposure of the catalan population



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1 Introduction

The Scientific Food Safety Advisory Committee of the Catalan Food Safety Agency (ACSA) approved the 'Assessment of the risk of exposure to iodine in cow's milk' report. The report stated that milk could be the main contributing food, even more so than fish and fishery products. It also stated that the intake of iodine in the diet through all foods would be unlikely to surpass the tolerable upper intake levels (UL) of iodine in the adult population in Catalonia (ACSA,¹ 2015). The Committee recommended that the ACSA perform a study on exposure to iodine in the diet in order to have current data to reassert the assessment.

Iodine is an essential chemical element for mammals, as it is used to synthesise thyroid hormones. Mammals obtain this element through food and water.

Iodine is a ubiquitous element and is found mainly in seawater. In general, soils contain a very small concentration, and plants barely absorb iodine because they do not need it to grow. Iodine is therefore added to animal feed so that they will produce food with a good performance. Part of this iodine is accumulated in meat, milk and eggs (ACSA, 2015).

In natural terms, the main source of iodine are sea products, fish, shellfish, algae and salt. Nevertheless, in the industrialised countries, milk and dairy products are the main source, followed by eggs and cereals. The content of iodine in cereals is very variable and depends on the type of soil, the use of soil conditioners, fertilisers and pesticides with formulas containing iodine (EFSA, 2006).²

Iodine is a volatile element, so that cooking it reduces its concentration in foods: 20% in fried foods, 23% in roast foods and 58% in boiled food (EFSA, 2006).

The small intestine absorbs more than 90% of iodine, although the presence of large quantities of iron, calcium, magnesium, fluorides, nitrates and glucosinolates (compounds present in cabbage, broccoli, soy and other plants of the Brassica or Crucifer family) reduces the absorption of iodine by the intestine and the thyroid gland or interferes in the action of hormones. A deficiency of vitamin A, iron, copper, zinc, selenium or vanadium may cause hypothyroidism and exacerbate the effects of an existing deficiency (EFSA, 2006).

¹ Avaluació del risc de l'exposició al iode pel consum de llet de vaca. Agència Catalana de Seguretat Alimentària (ACSA); 2015.

² Tolerable upper intake levels for vitamins and minerals. Scientific Committee on Food and Scientific Panel Products, Nutrition and Allergies. European Food Safety Authority (EFSA); 2006.

The synthesis of normal amounts of thyroid hormones requires an adequate dietary intake. Iodine deficiency causes severe clinical effects in children, newborns, the unborn child and pregnant women (growth retardation, miscarriage, congenital abnormalities and cretinism). In adults, it generally causes goitre associated with hypothyroidism. In cases of mild deficiency, symptoms include reduced intellectual capacity, apathy and weight gain. In moderate cases, the symptoms are, besides those already mentioned, intolerance of the cold, hoarse voice, swollen face and body oedemas. In severe cases, goitre can cause obstruction of the trachea and the oesophagus and lead to severe gland disorders (hypo- and hyperthyroidism) and cancer (EFSA, 2006).

An excessive intake can also cause severe thyroid gland operating disorders (goitre, hypothyroidism with or without goitre, hyperthyroidism) and in some cases sensitivity reactions (urticaria, conjunctivitis, fever, headache...), intoxication (diarrhoea, vomiting, abdominal pain...) and autoimmune thyroiditis (EFSA, 2006).

The latest studies on the nutritional status of the Spanish population with regard to iodine, TiroKid (Vila et al., 2012) and Di@bet.es (Soriguer et al., 2012) indicate that the average intake of iodine in Spain is adequate (ACSA, 2015).

The Tirokid study assessed this nutritional condition indirectly, analysing ioduria in a sample of 1,981 children aged between 6 and 7 years. In a normal health status, urinary excretion of iodine (ioduria) accounts for more than 90% of the intake and is therefore a good indicator of recent iodine intake. The results showed that 17.9% presumably had an inadequate intake ($<100 \mu\text{g/L}$), 69.4% (average of $173 \mu\text{g/L}$) an adequate intake and 10.5% ($>300 \mu\text{g/L}$) an excessive intake. This study showed that the group of children that did not take iodised salt, even although they drank two or more glasses of milk a day, had an adequate level of iodine that was equal to the group of children that did take iodised salt but who only drank one glass of milk or less a day (ACSA, 2015).

The Di@bet.es study estimated an average ioduria of $117 \mu\text{g/L}$ in a sample of 5,072 adult individuals from all over Spain. This value is very close to the lower limit of adequate ioduria – $100 \mu\text{g/L}$ – so the existence of other population groups with an inadequate iodine intake cannot be ruled out. The study relates an adequate intake to the consumption of iodised salt (43.9% of the sample individuals) or to the consumption of at least one glass of milk a day (ACSA, 2015).

2 Objectives

The main objective of this study is to estimate the dietary exposure of iodine, with particular emphasis on milk, and to evaluate the possible risk to health due to an excessive or insufficient intake of iodine in the diet by the Catalan population.

The specific objectives of this study are:

- To analyse the concentration of iodine in samples of whole, semi-skimmed and skimmed milk acquired in Catalonia in 2015.
- To estimate exposure to iodine through milk consumption.
- To estimate exposure to iodine in the diet for the population of Catalonia based on food samples taken in 2012.
- To evaluate the risks to health associated with exposure through the consumption of milk and other foods and to determine the contribution of milk to total iodine intake.

3 Materials and methods

3.1 Selection of foods and milks

The analytical samples come from the foods selected in the total diet study in Catalonia 2012, except the milk samples, which were obtained in 2015. The foods' representativeness was estimated based on the population's consumption data obtained in the Enquesta sobre l'estat nutricional de la població catalana i avaluació dels hàbits alimentaris 2002-2003 ['Survey on the nutritional status of the Catalan population and evaluation of food habits 2002-2003'] (ENCAT 2002-2003).

The 67 foods studied are detailed in Table 1. In this case, the milk analysed in the study comes from the samples collected in the most recent study (Table 2).

Table 1. Selection of foods studied

Grup	Aliments	Grup	Aliments	
1. Meat and meat products	Veal: steak, hamburger, pork, loin, Llonganissa (cold sausage) Chicken: breast Lamb: leg/rib Cooked ham Hotdog Chorizo (cold spicy sausage) Parma ham	7. Milk	lwhole milk semi-skimmed milk skimmed milk	
2. Fish	sardine tuna fish anchovy mackerel swordfish salmon hake tinned sardines	mullet sole cuttlefish squid clam mussel prawn tinned tuna fish	8. Dairy products	plain yoghurt type fresh cheese type-I cheese semi-cured type-II cheese cured type III
			9. Soy yoghurt*	
3. Vegetables	lettuce tomato green bean cauliflower onion pepper carrot aubergine	10. Bread and cereals	white bread loaf bread rice food pasta brown bread	
4. Tubers	potato	11. Pulses	lentil bean chickpea peas	
5. Fruit	apple orange pear banana tangerine strawberry peach	12. Fats	olive oil sunflower seed oil margarine butter	
6. Eggs	hen eggs	13. Bakery products	Croissant biscuit breakfast cereals	

* It is regarded as a new group because it is not a dairy product. Its consumption will be evaluated as a fermented soy drink according to the ENCAT 2002-2003 survey.

For milk, 20 commercial brands of milk were selected; three varieties – whole, semi-skimmed and skimmed – were analysed. Of these 20 brands, 16 were UHT, 3 fresh milk and 1 organic. This enables us to study the possible existence of differences caused by heat treatment or the way the milk is obtained, thus guaranteeing representativeness of the consumption of these types of milk.

3.2 Sample collection

Foods

Samples were taken in 12 towns/cities of Catalonia. The overall sample is representative of 72% of the Catalan population that lives in towns or cities of more than 20,000 inhabitants with a clearly urban profile.

The populations are as follows, grouped by the following territorial areas:

Metropolitan Area: Barcelona, l'Hospitalet de Llobregat, Vilanova i la Geltrú, Mataró, Sabadell and Terrassa; Counties of Girona Area: Girona; Camp de Tarragona Area: Tarragona and Reus; Terres de l'Ebre: Tortosa; Western Area: Lleida; Central Counties Area: Manresa.

Individual samples of each food were taken in July 2012. In each town or city where the samples were obtained, the sampling was distributed over a minimum of four different-sized establishments (market, shop, small supermarket, large supermarket, hypermarket) in order to maximise the diversity and the origin of the food acquired and to make the sample as representative as possible with regard to all types of shoppers. Samples that required refrigerated conditions were transported accordingly.

The composite samples were prepared following the same methodology as the previous study (WHO guidelines):

- 24 individual samples acquired independently formed a composite sample.
- Cleaning and separation of the edible parts, raw, from the individual samples, which were used to make a composite sample.
- Weighing of equal parts of each individual sample. When the sample presented very different parts in terms of texture, amount of fat, etc., such as different parts of lamb ribs, all the parts of each individual piece were used in a balanced way in the composite sample.
- Grinding and homogenisation of the samples using food mixers, with analytical-grade care taken in cleaning between samples to prevent cross contamination. Parts of equal weight of the edible parts of meat and the meat products, greens, tubers, fruit, eggs (whisked), cheese, cereals, pulses, fats (margarine and butter) and bakery products were mixed and ground until a homogeneous paste or meal was obtained.
- Formation of aliquots in laboratory test tubes and frozen storage until the time of the analysis.

The samples of yoghurt, cheese, milk, soy yoghurt, butter and margarine were stored in their original form and in refrigerated conditions. The corresponding composite sample was formed just before it was sent to the analysis laboratory.

The oils were stored at ambient temperature, away from the light, until the analysis, in the course of which equal volumes of each unit sample were mixed and distributed into the corresponding aliquots, after which the analysis was performed as described above.

A total of 1,584 individual food samples were processed.

Drinking milks

In July 2015, 180 drinking milk samples were purchased at different establishments in the cities of Tarragona and Reus. "Composite" samples were made up with three individual samples, making a total of 60 samples corresponding to 20 different commercial brands. A sample of whole, semi-skimmed and skimmed milk was obtained for each one of the brands.

Once all the samples had been taken and the composites had been formed, two duly identified aliquots were stored and frozen at $-20\text{ }^{\circ}\text{C}$ until they were analysed.

3.3 Analytical procedure

The iodine content analysis in both the milk samples and the rest of the foods was performed at the Laboratory of the Public Health Agency of Barcelona. The iodine analytical method is based on a pressurised extraction in a microwave and at a maximum temperature of $180\text{ }^{\circ}\text{C}$, using a basic medium with tetramethylammonium hydroxide. The extract is measured instrumentally by inductively-coupled plasma-mass spectrometry (ICP- MS).

ICP-MS methods use an internal standard, in this case, tellurium, to minimise matrix effects, and a collision cell is used in all cases to eliminate polyatomic interferences, and isotopes free of isobaric interferences are always used also. In all the analysis sequences, a process blank was analysed in parallel to guarantee the absence of contaminations, external solutions of a known concentration in order to confirm the goodness of the calibration straight line, standard solutions at the end of each sequence in order to guarantee the absence of instrumental drift, as well as added samples to continuously control the method's correct recovery percentage.

3.4. Population groups studied

According to the conditions established in previous studies, and in accordance with the WHO guidelines (1985), the same age groups that had been evaluated in previous total diet studies were studied. These groups reflected, out of the overall population, the groups that are regarded as standard individuals.

This study breaks down both sexes into all the age groups (except for children) to adapt them to the data structure of the Survey on the nutritional status of the Catalan population and evaluation of food habits 2002-2003, which showed certain gender-specific food consumption differences, whereas the EnKid study was chosen to evaluate children aged between 4 and 9 years.

Table 3 presents the population groups study and the body weight assumed for each one of them.

Table 3. Population groups, age intervals and weight

Group	Age (years)	Body weight (kg)
Children	4 to 9	24
Adolescent boys	10 to 19	56
Adolescent girls	10 to 19	53
Men	20 to 65	70
Women	20 to 65	55
Men above the age of 65 years	65 to 80	65
Women above the age of 65 years		60

3.5. Daily food consumption data

This study also used the data obtained in the Enquesta sobre l'estat nutricional de la població catalana i avaluació dels hàbits alimentaris 2002-2003 ['Survey on the nutritional status of the Catalan population and evaluation of food habits 2002-2003'] (ENCAT 2002-2003). Moreover, this study sought to compare the results of the intake of an adult man and woman to the consumption data of the ENIDE study; a study on food habits performed in Spain by the Spanish Food Safety Authority (AESAN, 2011). The data from the EnKid (1998-2000) study were used for boys and girls aged 4 to 9 years, a group which was not envisaged in the ENCAT study.

Tables 4, 5 and 6 detail the data pertaining to the consumption (g/day) of the different foods and for the different age groups considered, according to surveys performed in Catalonia and Spain.

Table 4. Food consumption in the child population between 6 and 9 years (EnKid)

Food group	g/day	Food group	g/day
Total meat	134,7	Total milk	364,8
Total fish	34,33	Total dairy products	108,6
Total greens	60,20	Total cereals	155,9
Total tubers	70,84	Total pulses	22,29
Total fruit	196,6	Total fats	31,14
Total eggs	22,98	Total bakery products	48.25

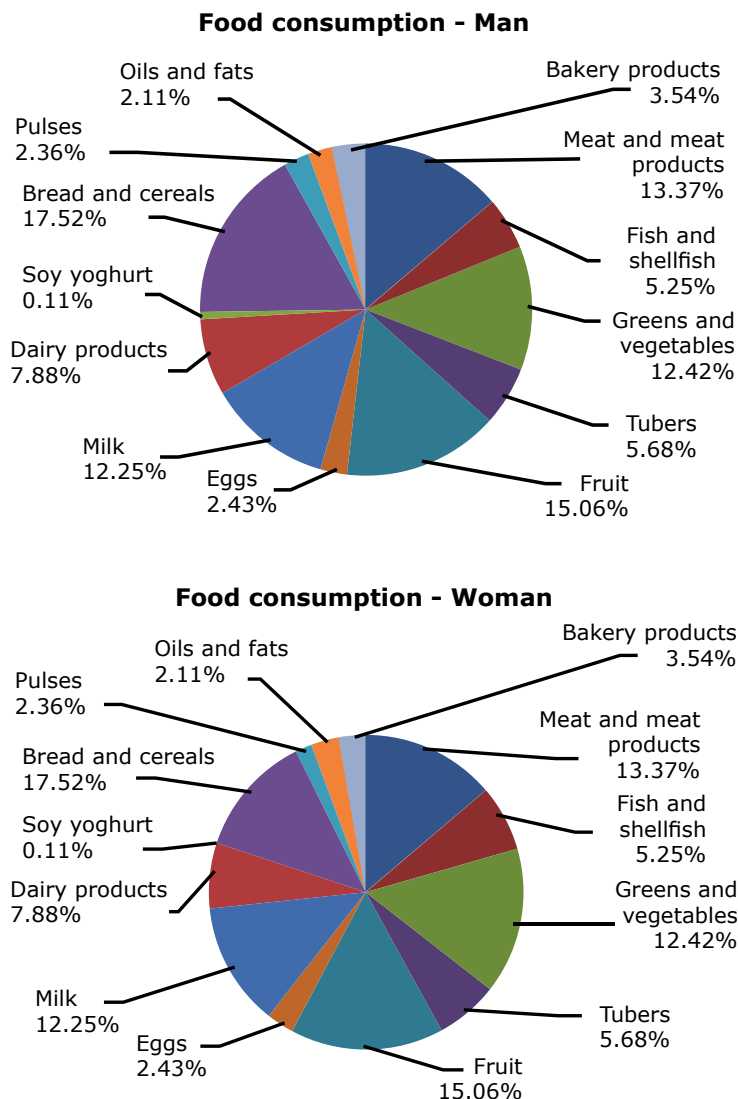
Table 5. Food consumption in different population groups (ENCAT)

FOODS	Men			Women		
	10-19	20-65	65-80	10-19	20-65	65-80
Total meat and meat products	188,9	171,9	109,1	143,2	122,4	102,8
Total fish and shellfish	45,0	67,5	73,3	45,4	65,0	55,6
Total greens and vegetables	91,0	159,7	176,2	99,6	182,4	162,5
Total tubers	81,1	73,1	63,8	71,8	52,8	57,7
Total fruit	110,8	193,6	327,5	119,5	204,3	269,8
Total eggs	25,8	31,3	23,5	22,7	23,2	20,2
Total milk	246,6	157,5	192,7	209,9	198,1	210,1
Total dairy products	110,0	101,3	71,8	99,8	97,8	77,2
Total soy yoghurt	0,0	1,4	0,0	0,0	0,5	0,0
Total bread and cereals	269,5	225,3	192,7	207,7	156,8	132,6
Total pulses	31,4	30,4	34,0	20,1	25,7	28,9
Total oils and fats	24,0	27,2	26,6	21,3	24,6	23,7
Total bakery products	81,7	45,5	22,6	61,0	41,2	26,0
Total	1.306	1.285	1.314	1.122	1.195	1.167

Table 6. Food consumption for an adult man and woman (ENIDE)

FOODS	Man	Woman
	20-65	20-65
Total meat and meat products	176.3	140.5
Total fish and shellfish	92.4	80.0
Total greens and vegetables	197.5	208.1
Total tubers	85.0	62.7
Total fruit	199.8	194.4
Total eggs	34.9	28.0
Total milk	176.5	193.4
Total dairy products	87.6	96.4
Total soy yoghurt	0.7	1.1
Total bread and cereals	165.9	119.4
Total pulses	23.0	18.4
Total oils and fats	36.3	31.9
Total bakery products	35.5	26.6
Total	1,311.4	1,200.9

Figure 1. Percentage distribution of daily food intake in an adult man. aENCAT and bENIDE



In all age groups, and in a non-homogeneous mode, it should be recalled that there is one food group, in both this ENCAT and in the ENIDE, that falls outside the study, which we refer to as "Other". For example, in the meat group it would be: raw bacon and pork ribs, grilled belly of pork, grilled pig's trotters, whole roast duck, roast skinned young pigeon, turkey breast, boiled hen, whole quail, raw or stewed rabbit. In order to evaluate this type of datum, the percentage of representativeness of our selection was calculated taking the total data for each group of foods as 100%. These data are shown in Tables 6 and 7. In any case, the data provided in this study refer exclusively to the "total analysed".

Table 6. Representativeness of the selection of foods analysed, expressed as a percentage out of the total of the group in ENCAT.

FOODS	Men			Women		
	10-19	20-65	65-80	10-19	20-65	65-80
Meat and meat products	93	87	87	91	88	83
Fish and shellfish	85	78	76	88	78	70
Vegetables	90	81	79	86	80	70
Tubers	100	100	100	100	100	100
Fruits	87	83	89	80	77	81
Eggs	98	99	97	99	99	100
Milk	100	100	100	100	100	100
Dairy products	62	60	73	74	67	78
Soy Yoghurt	—	100	—	—	100	—
Cereals	98	98	98	97	96	97
Pulses	100	94	89	100	95	91
Oils and fats	100	100	100	100	100	100
Bakery products	49	31	23	41	33	28

Table 7. Representativeness of the selection of foods analysed, expressed as a percentage out of the total of the group of adult men and/or women in ENIDE.

FOODS	Men	Woman
	20-65	20-65
Meat & meat products	61	51
Fish and shellfish	67	69
Vegetables	67	66
Tubers	91	91
Fruit	71	70
Eggs	97	99
Milk	100	100
Dairy products	52	48
Soy Yoghurt	100	100
Cereals	88	86
Pulses	94	95
Oils and fats	78	77
Bakery products	40	44

Some soy yoghurt data are left blank. This is due basically to the fact that in the ENCAT 2002-2003 study there were some population groups that consumed a very small proportion of this type of products.

The data pertaining to the consumption (g/day) of the different types of milk and for the different age groups considered are presented in Tables 8 and 9, except for the children (4-9 years), where total milk consumed was considered (364.80 g/day).

Table 8. Milk consumption in different population groups (ENCAT)

MILKS	Men			Women		
	10-19	20-65	65-80	10-19	20-65	65-80
Whole milk	200.20	86.93	59.38	132.50	83.68	51.71
Semi-skimmed milk	41.81	40.18	62.47	52.04	63.43	72.44
Skimmed milk	4.61	30.38	70.82	25.31	51.00	85.92
Total	246.61	157.50	192.67	209.85	198.10	210.08

In g/day

Table 9. Milk consumption for an adult man and woman (ENIDE)

MILK	Man	Woman
	20-65	20-65
Whole milk	98.52	74.40
Semi-skimmed milk	45.33	57.14
Skimmed milk	32.65	61.89
Total	176.5	193.4

In g/day

3.6. Estimate of daily iodine intake

The intake of iodine through the consumption of foods and milks can be calculated by multiplying the concentration of the iodine in each sample of food or individual milk by the daily intake and adding up all the products obtained.

Daily intake = Σ (concentration of the iodine \times amount of food taken)

Or else, expressed by unit of body weight:

Daily intake = Σ (concentration of the iodine \times amount of food taken)/body weight

In this study, intake was calculated by applying two different databases related to food consumption by the population: ENCAT and ENIDE. The main objective is to estimate if both of them are equivalent and similar results are obtained.

3.7. Estimate of results below the limit of detection

For the processing of results, in cases in which a food type presented an iodine concentration below the limit of detection (LOD), the concentration was assumed to be half the LOD ($ND = \frac{1}{2}LOD$), following the WHO recommendations (medium-bound).

3.8 Statistical analysis

The XLStat statistical application was used to evaluate whether there were statistically significant differences between the concentrations of the different food groups and the milks studied. A probability of 0.05 or less ($P < 0.05$) was regarded as significant.

4 Results and discussion

4.1. Concentrations of iodine in drinking milk

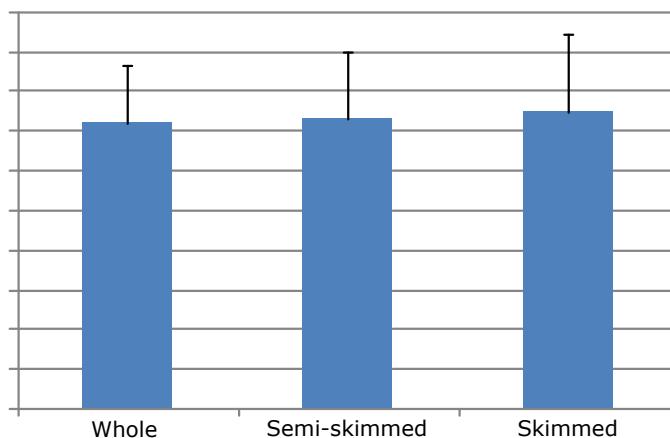
Table 10 details the individual concentrations of iodine in each one of the 60 milk samples analysed. The iodine levels were in an interval of 45 to 204 µg/kg. By type, organic milk contains much less Iodine (47 µg/kg) than the non-organic milk (average iodine concentration of 146 ± 34 µg/kg).

Table 10. Concentration of iodine in cow's milks for human consumption (µg/kg).

Sample brand	Whole	Semi-skimmed	Skimmed	Average according to milk brand
1	170	170	179	173
2	167	170	204	180
3	169	167	179	172
4	151	117	172	147
5 Fresh milk	139	120	126	128
6	128	140	131	133
7	141	139	103	128
8	149	101	142	131
9	151	165	194	170
10	140	189	148	159
11	163	182	160	168
12	180	165	170	172
13 Fresh milk	132	163	166	154
14 Organic milk	45	49	48	47
15 Fresh milk	148	154	168	157
16	119	118	124	120
17	139	149	107	132
18	173	177	184	178
19	116	118	96	110
20	155	166	180	167
Average according to milk type	144	146	149	

Figure 2 graphically represents the average iodine concentration according to milk type. Although skimmed milk presented slightly higher levels, no significant differences are observed ($P>0.05$).

Figure 2. Concentration (average and standard deviation) of iodine in milk according to type.



The EFSA evaluated the use of iodine in animal feed in 2005³. The finding indicated that the intake in food from animals, particularly milk and eggs, produced from animals fed with feed containing the maximum authorised amount of iodine, 10 mg/kg, could constitute an intake above the tolerable upper intake levels (UL) for adults and adolescents. Consequently, in 2005, the European Commission⁴ established a new maximum iodine content in animal feed of 5 mg/kg. Subsequently, in 2013, the EFSA⁵ reviewed the safety and efficacy of four iodine additives. This evaluation concluded that the maximum content should be further reduced from 5 mg/kg to 2 mg/kg, in the case of feeds intended for dairy cows, since the major milk consumers (>1.5 l/day) could surpass the UL. The evaluation was made on the supposition that all manufacturers added the maximum authorised amount of iodine to animal feed.

Nevertheless, the most recent ioduria data from European populations do not seem to indicate a risk situation, as the EFSA finding indicates, hence the European Commission has not reduced the maximum iodine content in animal feed any further.

³ Opinion of the Scientific Panel on additives and products or substances used in animal feed (FEEDAP) on the use of iodine in feedingstuffs. EFSA, 2005.

⁴ Commission Regulation (EC) No 1459/2005 of 8 September 2005 amending the conditions for authorisation of a number of feed additives belonging to the group of trace elements.

⁵ Scientific Opinion on the safety and efficacy of iodine compounds (E2) as feed additives for all animal Species: calcium iodate anhydrous and potassium iodide, based on a dossier submitted by Ajay Europe SARL. Scientific Opinion on the safety and efficacy of iodine compounds (E2) as feed additives for all animal species: calcium iodate anhydrous and potassium iodide, based on a dossier submitted by Helm AG.

If we compare the concentration of iodine in milk of this study to that of other European countries (ACSA⁶, 2015), the data obtained are within the average European values (Table 11).

Table 11. Average concentrations of iodine in milk in different European countries

Country	Concentration of iodine in milk ($\mu\text{g}/\text{kg}$ or $\mu\text{g}/\text{L}$)	Author	Observations
Catalonia	146 $\mu\text{g}/\text{kg}$	This study	Range 45-204 $\mu\text{g}/\text{kg}$
Austria	74 $\mu\text{g}/\text{kg}$	Rysava et al., 2007 (2)	Range 45-92 $\mu\text{g}/\text{kg}$
Poland	90 $\mu\text{g}/\text{kg}$	Rysava et al., 2007 (2)	Range 86-93 $\mu\text{g}/\text{kg}$
Switzerland	90 $\mu\text{g}/\text{kg}$	Rysava et al., 2007 (2)	Range 79-106 $\mu\text{g}/\text{kg}$
Germany	130 $\mu\text{g}/\text{kg}$	Rysava et al., 2007 (2)	Range 93-159 $\mu\text{g}/\text{kg}$
Belgium	158 $\mu\text{g}/\text{kg}$	Rysava et al., 2007 (2)	
Slovakia	240 $\mu\text{g}/\text{kg}$	Rysava et al., 2007 (2)	Range 180-310 $\mu\text{g}/\text{kg}$
	325 $\mu\text{g}/\text{kg}$	Rysava et al., 2007 (2)	Range 305-345 $\mu\text{g}/\text{kg}$
England	311 $\mu\text{g}/\text{kg}$ (1998-1999)	COT 20004	1991/1992 Summer: 90 $\mu\text{g}/\text{kg}$ Winter: 210 $\mu\text{g}/\text{kg}$ 1998-1999 Summer: 200 $\mu\text{g}/\text{kg}$ Winter: 430 $\mu\text{g}/\text{kg}$
Czech Republic	471 $\mu\text{g}/\text{kg}$ 324 $\mu\text{g}/\text{L}$ 489 $\mu\text{g}/\text{L}$	Rysava et al., 2007 (2) Kursa et al., 2004 (1) Travnicek et al., 2006 (1)	Range 387-601 $\mu\text{g}/\text{kg}$
Norway	190 $\mu\text{g}/\text{L}$ 88 $\mu\text{g}/\text{kg}$ - summer 2000 232 $\mu\text{g}/\text{kg}$ - winter	Haug et al., 2012 (1) Dahl et al., 2003 (2)	
Denmark	243 $\mu\text{g}/\text{L}$	Haug et al., 2012 (1)	
Sweden	140 $\mu\text{g}/\text{L}$	Haug et al., 2012 (1)	
Finland	170 $\mu\text{g}/\text{L}$	Haug et al., 2012 (1)	
Iceland	112 $\mu\text{g}/\text{L}$ 259 $\mu\text{g}/\text{L}$	Haug et al., 2012 (1) Soriguer et al., 2011	Summer: 247 $\mu\text{g}/\text{L}$ Winter: 270 $\mu\text{g}/\text{L}$
Spain	197.6 $\mu\text{g}/\text{L}$ (UHT milk) 56.4 $\mu\text{g}/\text{L}$ (organic milk)	Arrizabalaga et al., 20145	2008: 84 - 428 $\mu\text{g}/\text{L}$ 50.5 - 61.5 $\mu\text{g}/\text{L}$
	113 $\mu\text{g}/\text{kg}$	(ANSES 2005) (3) Aquaron (1986-1988)	Range 10-272 $\mu\text{g}/\text{kg}$ Summer: 62 $\mu\text{g}/\text{kg}$ Winter: 203 $\mu\text{g}/\text{kg}$
France	135 $\mu\text{g}/\text{kg}$	Valeix (1999-2001) (3)	Range 59-258 $\mu\text{g}/\text{kg}$ Summer: 107 $\mu\text{g}/\text{kg}$ Winter: 167 $\mu\text{g}/\text{kg}$
	207 $\mu\text{g}/\text{kg}$	Rysava et al., 2007 (2)	Range 192-221 $\mu\text{g}/\text{kg}$

Sources: (1) EFSA FEEDAP 2013; (2) Lohmann 2007; (3) ANSES 2005; (4) COT 2000. (5) Medicina clínica, 2015:145(2):55-61

Note: the concentrations expressed in litres can be converted into kilograms by multiplying by the coefficient of 1.03.

⁶ Opinion of the Scientific Panel on additives and products or substances used in animal feed (FEEDAP) on the use of iodine in feedingstuffs. EFSA, 2005.

According to the available data about milk marketed in Spain, the average values of our study are below the values obtained in the study by Arrizabalaga et al (2014) and Soriguer et al. (2011), who studied milk samples collected in 2008 in the Basque Country and all over Spain, respectively. Both our and Arrizabalaga's study indicate that organic milk contains substantially less iodine than non-organic milk.

4.2 Intake of iodine through the consumption of milk

The average intake of iodine through milk by the Catalan population has been estimated as 29.5 µg/day, according to the ENCAT. The group of boys and girls presented the maximum iodine intake through the consumption of milk with an average value of 54.4 µg/day (60% of the adequate intake established by the EFSA). The group of adolescent boys presented a maximum daily intake of 35.6 µg/day (29-27% of the adequate intake), whereas the intake of iodine through milk in pregnant women is lower, around 14% the adequate intake.

Table 12. Daily intake of iodine through the consumption of milk in the Catalan population and percentage between the daily iodine intake through milk and the adequate intake recommended by the EFSA.

Population group	Daily iodine intake (µg/day)	Adequate intake (µg/day)	Contribution of milk consumption to the adequate intake (%)	Consumption study
Children (4-9 years)	54.40	90	60.5%	EnKid
Adolescent boys (10-19 years)	35.06	120-130b	29 - 27%	
Adult men (20-65 years)	22.90	150	15.2%	
Men above 65 years	28.20	150	18.8%	
Adolescent girls (10-19 years)	30.40	120-130b	25.3 - 23.3%	ENCAT
Adult women (20-65 years)	28.90	150-200a	29.2 -14.5%	
Women above 65 years	30.80	150	20.5%	
Adult men (20-65 years)	25.60	150	17%	
Adult women (20-65 years)	28.30	150-200a	18.9 -14%	ENIDE

a. Pregnant woman

b. Adequate intake of adolescents between 15-17 years

4.3 Iodine concentrations in food (total diet study)

Table 13 details the concentrations of iodine in each one of the 67 food samples analysed.

Table 13. Individual concentration of iodine in consumption food for the Catalan population

Group	Food	Concentration ($\mu\text{g}/\text{kg}$)
Meat and meat products	Steak veal	124
	Hamburger veal	81
	Loin pork	8.4
	Fresh sausage	42
	Chicken breast	<7
	Lamb leg/rib	26
	Cooked ham	34
	Hotdog	218
	Chorizo	9.6
	Parma ham	46
	Sardine	292
	Tinned sardine	230
	Tuna fish	167
	Tinned tuna fish	153
	Anchovy	315
Fish and shellfish	Mackerel	282
	Swordfish	223
	Salmon	70
	Hake	112
	Mullet	624
	Sole	416
	Cuttlefish	96
	Squid	89
	Clam	667
	Mussel	1510
Vegetables	Prawn	737
	Lettuce	21
	Tomato	5.7
	Cauliflower	<4
	Green bean	6.5
	Onion	4.9
	Pepper	21
Tubers	Carrot	15
	Aubergine	<4
	Potato	7.8

Group	Food	Concentration ($\mu\text{g}/\text{kg}$)
Fruit	Apple	<4
	Orange	<4
	Pear	<4
	Banana	<4
	Strawberry	<4
	Tangerine	<4
	Peach	<4
Eggs	Hen eggs	461
Milk*	Whole milk	144
	Semi-skimmed milk	146
	Skimmed milk	149
	Yoghurt	137
Dairy products	Cheese 1, fresh	284
	Cheese 2, semi-cured	469
	Cheese 3, cured	980
Soy yoghurt	Soy yoghurt	74
Bread and cereals	White bread	98
	Loaf bread	17
	Brown bread	113
	Rice	<7
	Food pasta	19
	Lentils	5.7
	Green bean	4.2
Pulses	Chickpea	8.3
	Pea	8.5
	Olive oil	<1
Fats	Sunflower seed oil	<1
	Margarine	2.8
	Butter	8.8
Bakery products	Croissant	38.5
	Biscuit	67
	Breakfast cereals	19.1

*Concentration obtained in the samples acquired in 2015.

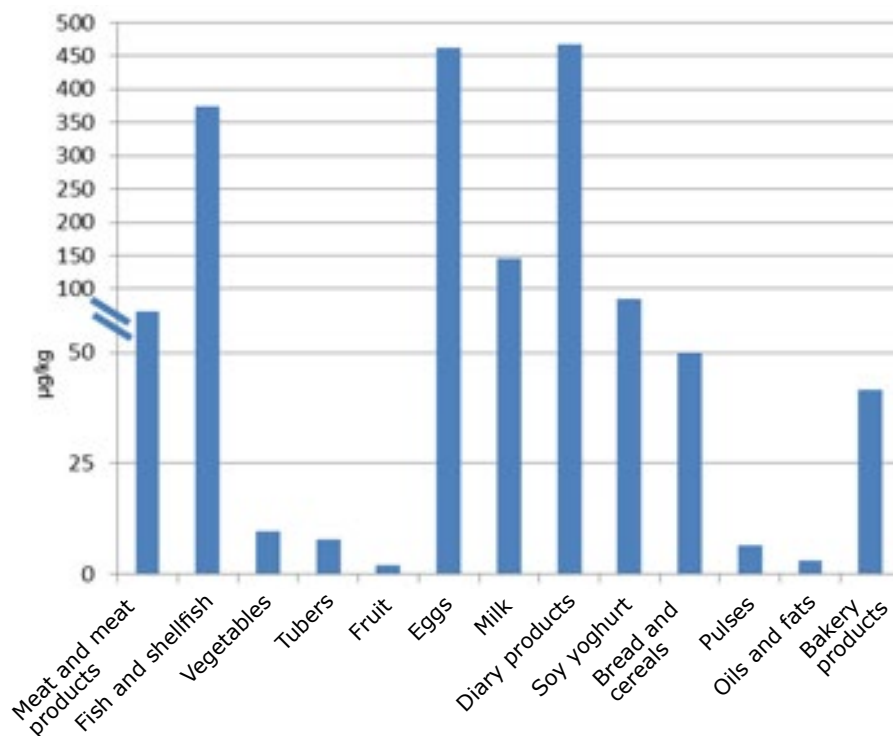
Figure 3 shows the concentrations of iodine ($\mu\text{g}/\text{kg}$) according to the group of foods studied, also considering, for milks, the values obtained in the samples collected in 2015.

Dairy products and eggs are the foods that present the highest concentration of iodine (468 and 461 $\mu\text{g}/\text{kg}$, respectively), followed by fish and shellfish (374 $\mu\text{g}/\text{kg}$). On the other hand, all the fruit samples present values below the analytical limit of detection (<4 $\mu\text{g}/\text{kg}$), whereas neither was it possible to quantify iodine in oils (<1 $\mu\text{g}/\text{kg}$) and other specific foods (chicken breast, cauliflower, aubergine and rice).

Mention should be made of the high levels of iodine obtained in certain sea species, such as the mussel (1510 $\mu\text{g}/\text{kg}$) and the prawn (737 $\mu\text{g}/\text{kg}$), as well as in cured cheese (980 $\mu\text{g}/\text{kg}$).

It should also be mentioned that the average concentration detected in the whole milk samples (144 $\mu\text{g}/\text{kg}$) is much higher than the one contained in the Spanish Food Composition Database (9 $\mu\text{g}/\text{kg}$).

Figure 3. Iodine concentrations in the total diet study foods ($\mu\text{g}/\text{kg}$).



4.4. Iodine intake in food (total diet study)

The intake of iodine through food consumption by the child population was calculated from the EnKid study data (Figure 4). Milk first, followed by dairy products, are the main contributors to the total, with percentages of 38.9% and 30.2%, respectively.

Figure 4 presents iodine intake according to food group (total diet study) and population group based on the Catalan Consumption Survey (ENCAT).

The intake of iodine through milk is the most outstanding finding in all the population groups studied, particularly in adolescent boys (35.06 $\mu\text{g}/\text{day}$) and girls (30.40 $\mu\text{g}/\text{day}$). With regard to the intake of other food groups, soy yoghurt, oils and fats and pulses were the groups with the lowest contribution of iodine to the diet for all the population groups (Figure 5).

Figure 4. Intake of iodine through foods in the child population ($\mu\text{g}/\text{day}$) (EnKid).

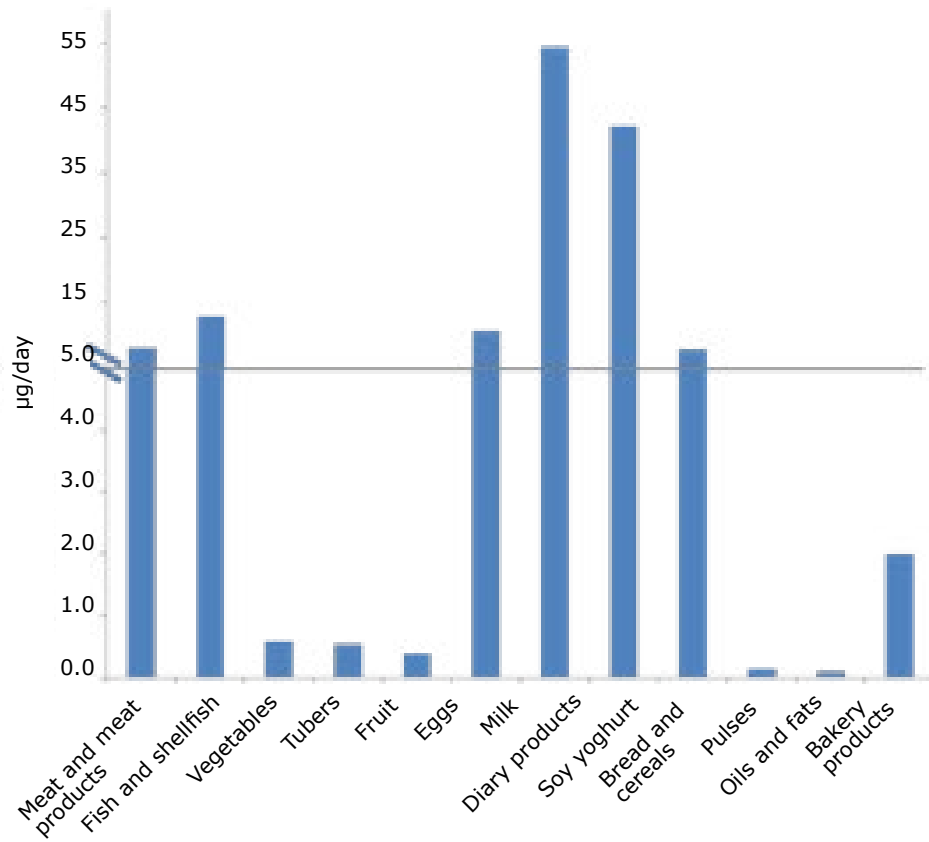


Figure 5. Intake of iodine through foods according to the population group ($\mu\text{g}/\text{day}$) (ENCAT).

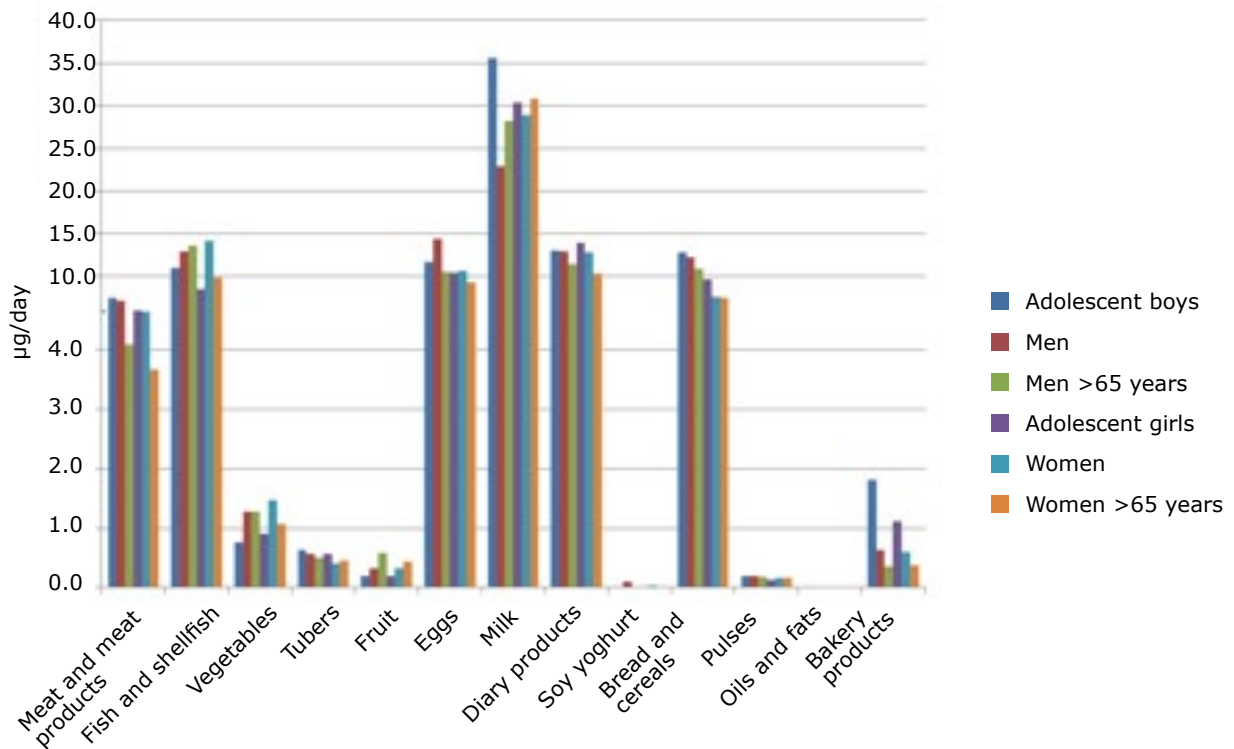
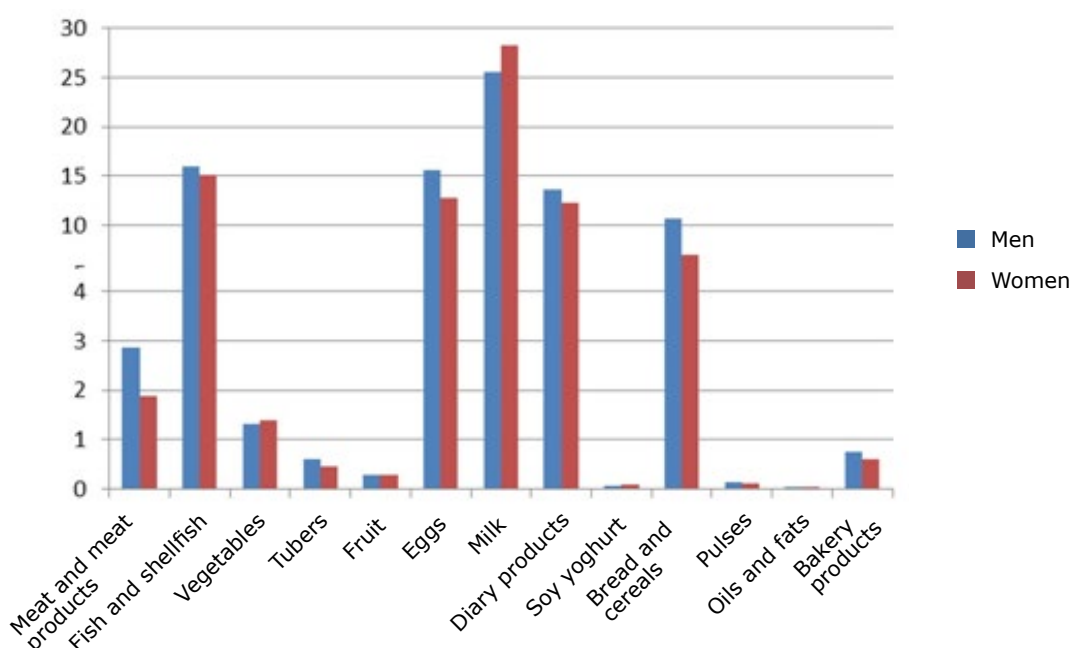


Figure 6 shows the intake of iodine according to the Spanish Consumption Study (ENIDE). Milk continues to be the food group with the greatest contribution, both by men (25.6 µg/day) and women (28.3 µg/day). On the other hand, soy yoghurt, oils and fats and pulses are the groups that account for the lowest intake of iodine in both sexes.

In general, women have a lower intake of iodine, since they consume a small amount of the foods studied, except for the greens and vegetables group, which contain little iodine. In this group, the contribution of lettuce (0.46 µg/day, ENIDE) deserves special mention.

Figure 6. Intake of iodine through food according to the population group (µg/day) (ENIDE).



4.5. Contribution of milk to total daily iodine intake

Figures 7-8 present the contribution of iodine intake through milk to total iodine intake through foods.

With regard to the results obtained with the ENCAT consumption study (Figure 6), women above the age of 65 years present the greatest contribution (36.8%) and adult men the lowest (22.7%). The average contribution of milk in the consumption of iodine in the diet was 31.2%, well above that of other foods, such as fish and shellfish, eggs and dairy products (12.3%, 11.7% and 13.1%, respectively).

With regard to the values obtained using the ENIDE study (Figure 8), the contribution of milk accounted for 22.6% in the case of men (values almost identical to that of the ENCAT study), and 26.9% in women. The average contribution of milk as a dietary source of iodine, on the basis of the ENIDE data, would be 24.2%, substantially higher than that of other foods (13.9%, 12.7% and 7.8% for fish and shellfish, eggs and dairy products).

Figure 7. Daily iodine intake through milk and other foods according to the ENCAT consumption study.

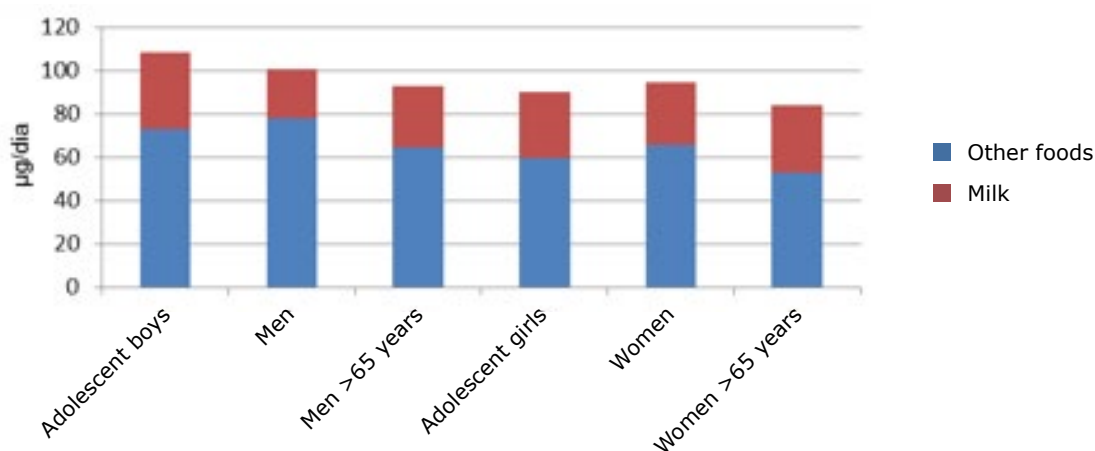
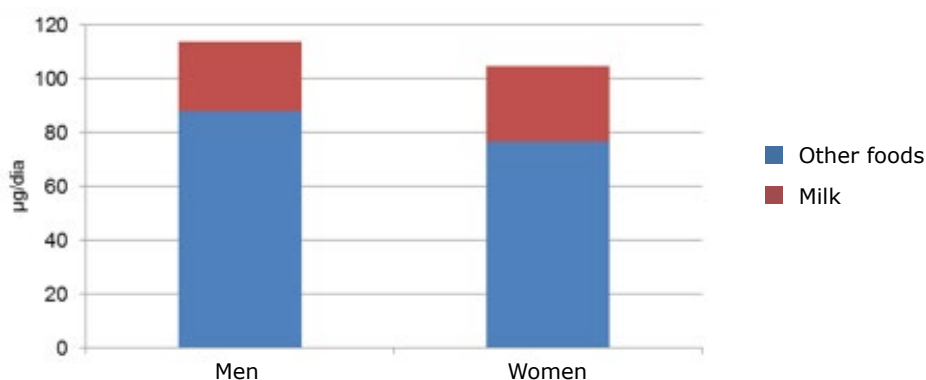


Figure 8. Daily iodine intake through milk and other foods according to the ENIDE consumption study.



4.6. Risk assessment

Table 14 presents the reference dietary values for iodine according to the WHO-UNICEF-ICCIDD (2007) and EFSA (2014).

Table 14. Reference dietary values for the intake of iodine of the WHO-UNICEF-ICCIDD (2007) and EFSA (2014)

Minimum iodine recommendations according to the WHO-UNICEF-ICCIDD (2007)		Reference values for iodine according to the EFSA (2014)	
Age group	Population Reference Intake ⁷ (PRI) ($\mu\text{g}/\text{day}$)	Age group	Adequate intake ⁸ (SI) ($\mu\text{g}/\text{day}$)
Pre-terms 0-5 months	>30	—	—
Newborns	15	—	—
6-12 months	90	7-11 months	70
1-3 years	90	1-8 years	90
4-6 years	90	9-14 years	120
7-10 years	120	15-17 years	130
Adults	150	≥ 18 years	150
Pregnant and breast-feeding women	250-350	Pregnant and breast-feeding women	200
		Woman of childbearing age	200

Table 15 shows the tolerable upper intake levels (UL) of iodine in healthy populations by the WHO (1989), the IOM-USA (2001) and the SCF (2002).

Table 15. Tolerable upper level intake (UL) of iodine in healthy populations ($\mu\text{g}/\text{day}$) according to different committees.

WHO (1989)	IOM-USA (2001) ($\mu\text{g}/\text{day}$)	SCF (2002) ($\mu\text{g}/\text{day}$)
	0-12 months	—
1 mg	1-3 years	200
PMTDI 0.017 mg/kg	4-8 years	300
of body weight	9-13 years	600
(=1 mg/day/60 kg)	14-18 years	900
	>19 years	1100
		Adults
		600
	Pregnant women	Pregnant and breast-feeding women
	600	600

⁷ The EFSA defines the population reference intake (PRI) as the amount of an individual nutrient that the majority of people in a population need for good health.

⁸ The adequate intake (AI) is the estimated value that is used when a population reference intake cannot be established, representing the average daily intake of a nutrient observed in a healthy population and is assumed to be adequate. It is different from the reference intake in the method of calculation and the solidness of the result, which is less in the case of the AI.

⁹ The Tolerable Upper Intake Level represents the highest level of nutrient intake that is likely to pose no risk of adverse health effect throughout life.

Table 16 shows the reference minimum iodine intakes¹⁰ in healthy populations, without signs of goitre and thyroid disorders or suboptimal gland functioning, established by the UK Committee on Medical Aspects of Food and Nutrition Policy (COMA, 1991), Scientific Committee on Food (SCF, 1993) and Nordic Council of Ministers (NCM, 2014).

Table 16. Reference minimum intake ($\mu\text{g}/\text{day}$)

COMA (1991)	SCF (1993)	NCM (2014)
70	70 (40-100)	70

Table 17 shows the results for the daily iodine intake by population groups according to this study.

Table 17. Daily iodine intakes according to this study per population groups

Population group	Daily iodine intake	Consumption study
Children (4-9 years)	139.6	EnKid
Adolescent boys (10-19 years)	108.3	
Adult men (20-65 years)	100.7	
Men above 65 years	92.7	
Adolescent girls (10-19 years)	90.1	ENCAT
Adult women (20-65 years)	94.7	
Women above 65 years	83.7	
Adult men (20-65 years)	113.6	
Adult women (20-65 years)	104.7	ENIDE

The group of children is the only one that surpasses the recommended daily intake ($90 \mu\text{g}/\text{day}$). The remaining groups are between the minimum reference intake ($70 \mu\text{g}/\text{day}$) and the recommended daily intake ($150 \mu\text{g}/\text{day}$). Consequently, the average adult Catalan population has a sufficient intake of iodine in the diet to maintain proper thyroid function. Nevertheless, it would be suitable the intake levels to reach the recommended value of $150 \mu\text{g}/\text{day}$, guaranteeing a broad safety margin against the intake of strumogenic substances both of natural origin in vegetable foods (glucosinolates, nitrates, etc.), and contaminating substances (perchlorate, nitrates, thiocyanates, etc.) and deficiencies¹¹ of Vitamin A, selenium, zinc, copper, iron and vanadium, which exacerbate the effects of an existing iodine deficiency.

¹⁰ Scientific Opinion on Dietary Reference Values for Iodine. EFSA Panel on Dietetic Products, Nutrition and Allergies (NDA), 2015.

¹¹ Tolerable upper intake levels for vitamins and minerals. Scientific Committee on Food and Scientific Panel on Dietetic Products, Nutrition and Allergies. EFSA, 2006.

It must be mentioned that the estimated intake for women of childbearing age does not cover iodine requirements during pregnancy (200 µg/day).

4.7. Other studies

Table 17 contains data pertaining to the diet intake of iodine found in different studies. Asian countries, such as China and Japan, present the highest intake values of this element. As for the European countries, the iodine intakes found in different studies are similar to those of Catalonia.

Table 17. Iodine intake (µg/day) through food in different studies

Country	Daily intake (µg/day)	Reference
Japan	229 (women) and 273 (men)	Katagiri et al., 2015
China	379	Mao et al., 2015
Italy	62-168* (women) and 60-164* (men)	Pastorelli et al., 2015
United Kingdom	123-167 breastfeeding women	Bath et al., 2014
Belgium	104-164 (preschool children between 2.5 and 6.5 years)	Vandevijvere et al., 2012
Spain	94-102 (adult man) 74-87 (adult woman)	AECOSAN, 2011
Children	139.6	
Adolescent boys	108.3	
Men	100.7	
Men > 65 years	92.7	Present study (ENCAT)
Adolescent girls	90.1	
Women	94.7	
Women > 65 years	83.7	
Men	113.6	
Women	104.7	Present study (ENIDE)

*Intake with iodised salt

5 Conclusions

On the basis of the analysis of the iodine content of 60 "composite" cow's milk samples, an average concentration of iodine in milk was established, namely $146 \pm 34 \mu\text{g}/\text{kg}$, with levels comprised within a range between 45 and 204 $\mu\text{g}/\text{kg}$. The results indicate that there are no significant differences in iodine levels according to the type of milk (whole, semi-skimmed and skimmed).

According to the results of this study, dairy products and eggs are the foods that present the highest concentration of iodine (468 and 461 $\mu\text{g}/\text{kg}$, respectively), followed by fish and shellfish (374 $\mu\text{g}/\text{kg}$).

The average intake of iodine through milk by the Catalan population has been estimated as 29.5 $\mu\text{g}/\text{day}$, according to consumption data of the ENCAT study, and as 26.9 $\mu\text{g}/\text{day}$, according to ENIDE data. The group of adolescent boys presented the maximum daily intake (35.6 μg), whereas men presented the minimum value (22.9 $\mu\text{g}/\text{day}$). In any event, the group of boys and girls presented the maximum iodine intake through the consumption of milk, with an average value of 54.4 $\mu\text{g}/\text{day}$ (according to EnKid).

Milk and dairy products are the foods that contribute the greatest amount of iodine in the diet of any population group. In the child population, the consumption of these foods alone already delivers the adequate intake of iodine established by the EFSA. In pregnant women, consumption accounts for approximately 20% of the adequate intake.

The Catalan population presents a dietary intake of iodine below the general recommendation of 150 $\mu\text{g}/\text{day}$ in adults, established by both the WHO-UNICEF-ICCIDD (2007) and by the EFSA (2014) and above the minimum reference intake of 70 $\mu\text{g}/\text{day}$ (EFSA, 2014). Therefore, the estimated intake entails no health risk whatsoever for the population of Catalonia.

The iodine values obtained in milk obtained in this study are within the intervals found in other countries.

The findings of this study reassert the conclusion of the report by the Scientific Food Safety Advisory Committee of the ACSA, Assessment of the risk of exposure to iodine in cow's milk, which stated that milk could be the main contributor, even ahead of fish.

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