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Original Article

Iodine content of food groups

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Abstract

The iodine content of several kinds of foods representing different product groups available on the Swiss market was analyzed by isotope dilution inductively coupled plasma mass spectrometry using the enriched long-lived nuclide ^{129}I. Considerable variations in levels of iodine between single foodstuffs within food groups were found, which also applied for levels in different food groups. The contribution of the food groups to the average daily iodine intake for the Swiss population was estimated from recent food consumption data. Bread and milk were identified as significant sources of iodine in the Swiss diet as they contributed 58 and 29 μg/day, respectively. The estimated contribution of all basic food groups to the per capita intake of iodine was approximately 140 μg/day, which was somewhat below the amount recognized for adequate nutrition (150 μg/day). In view of the additional consumption of iodized kitchen salt, an average of 140 μg/day underestimates the actual iodine intake.

Keywords: Iodine; Isotope dilution; ICP-MS; Food groups; Dietary intake

1. Introduction

Iodine is an important micronutrient element and is required for the synthesis of thyroid hormones, thyroxine (T4) and triiodothyronine (T3), which are iodinated molecules of the amino acid tyrosine. The thyroid hormones regulate a variety of important physiological processes including the cellular oxidation. Adolescents and adults need iodine in amounts of 150 μg per day (WHO, 1996). An iodine-deficient diet causes a wide spectrum of illnesses, including goiter and mental retardation. The oral intake also includes iodine from water and beverages; however, food

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provides by far the major contribution to the total iodine exposure (Park et al., 1981). The nutritional status of a population group can be assessed by analyzing the urinary excretion of iodine, which is the most widely used biochemical indicator. However, information on the contribution of iodine by food groups is also necessary for making recommendations to improve the dietary status of individuals. In addition, such information is useful for predicting the effect of nutrition policies on the dietary status. Presently, the main additional iodine source beside the food is iodized kitchen salt (Bürgi, 1998).

Furthermore, the analysis of individual foods gives information on iodine variation of dietary iodine contents. Iodine variability is of practical significance when it occurs in foods that are normally relied on as important sources. The iodine uptake of food plants is in proportion to the iodine present in the environment; which explains why the same food item may have widely different iodine contents depending on the locality where it was produced. Plants grown in iodine-rich soil will contain substantial amounts of iodine. In general, European inland soils are depleted in iodine relative to soils from costal areas.

Data on iodine contents of food items are relatively scarce, except for milk and fish (Julsham et al., 2001; Sieber et al., 1999). A detailed survey of iodine levels in foods of the US food supply was published (Pennington et al., 1995). In this paper the authors acknowledge analytical problems with the method for iodine. The determination of iodine in food has been a challenging analytical problem for a long time (Heckman, 1979). The concentration of iodine in most foods is low, therefore, accurate determination requires a sensitive analytical method and freedom from contamination. Inductively coupled plasma mass spectrometry (ICP-MS) was useful to determine iodine in biological samples (Gélinas et al., 1998). To overcome difficulties normally encountered with iodine, such as poor signal stability and memory effects, a new ICP-MS method was developed (Haldimann et al., 2000). The key improvements were the use of a miniaturized sample introduction system and the application of isotope dilution analysis. Isotope dilution is an explicit method, where only the ratio between the added isotope and a selected reference isotope is measured. Therefore this method is not susceptible to sensitivity changes induced by the matrix. As iodine is a mono isotopic element consisting only of the isotope ^{127}I, the sufficiently stable radioisotope ^{129}I of that element is added deliberately to the samples and the isotope ratio is measured in the mixture. The isotope ^{129}I is now extinct in nature and not detectable in food samples.

The objectives of the study were to obtain information on the actual iodine content of foods and on iodine nutrition by way of identifying the major sources of dietary iodine.

2. Materials and methods

2.1. Sampling and preparation

Food products were purchased from retail outlets in Bern (Switzerland) during the years 1999–2001. The samples included commonly consumed foods such as cereals, meat, dairy products, fruit and vegetables. These were chosen mainly because they form a major part of the Swiss diet (Nestlé, 2000), or because previous studies indicated that they might be rich sources of iodine (Pennington et al., 1995). The preparation procedure of the food samples for chemical analysis consisted of the following steps: cutting larger pieces into small portions, homogenizing with a laboratory mixer (B-400, Büchi, Uster, Switzerland), weighing a sub-sample of approximately 100 g, freeze-drying (Lyolab BII, LSL Secfroid, Lausanne, Switzerland) for 48 h at $-50\,^{\circ}C$ and again weighing the dried sub-sample. Any fat, bones and other hard bits were removed from meat, poultry or fish before mixing. Food samples such as vegetables, salads and mushrooms were washed with demineralized water prior to homogenization. The skin or rind of certain fruits and vegetables was peeled. Bread samples were air-dried in a clean room (class 100,000) and then finely ground using an ultra centrifugal mill (Retsch, Haan, Germany). The water content and the composition of food groups are listed in Table 1.

2.2. Reagents

Water was purified and deionized (18 MΩ) using the Barnstead cartridge system Easy Pure LF (Skan, Basel, Switzerland). Suprapure nitric acid was obtained from Merck (Darmstadt, Germany). The radioactivity standard SRM 4949 C ^{129}I (National Institute of Technology, Gaithersburg, MD, USA) was used as a spike for isotope dilution calibration. The certified activity of the solution was 3451 Bq/g, which corresponds to a concentration of 4.09 ± 0.03 mmol/L (528 mg/L) of the isotope ^{129}I. This material does not represent a radiological hazard and no extra shielding was necessary to prevent personnel exposure.

2.3. Iodine analysis

The iodine contents in dried samples were measured by an ICP-MS method previously described in detail (Haldimann et al., 2000). The method involves sample digestion with nitric acid by using a high-pressure autoclave (130 bar) at $230\,^{\circ}C$ (Paar, Graz, Austria) in the presence of definite quantities of the ^{129}I isotope standard. The nitric acid digests were then diluted with water for subsequent $^{127}I/^{129}I$ isotope ratio measurements by ICP-MS (Elan 5000, Perkin-Elmer, Norwalk, CT, USA). The method makes use of a new miniature cyclonic spray chamber and a concentric glass nebulizer that is designed for low sample uptakes (Glass Expansion, Vevey, Switzerland).

Food and agricultural standard reference materials (SRM) were obtained from the major suppliers (NIST and BCR) with either a certified or an indicative value for iodine. They were used to control accuracy and precision. Quality control was maintained by analyzing standard reference materials with every batch of samples. Food and SRM samples were grouped according to similarity of the matrix.

2.4. Statistical analysis

The descriptive statistics and analysis of variance (ANOVA) were calculated using the statistical software Systat (SPSS Inc., Chicago, IL, USA). One-way ANOVA was carried out to uncover a possible effect of the animal from which the meat originates (beef, lamb, pork, veal and horse) on the iodine concentration.

Table 1
Description of the analyzed food samples

Food group	Description and composition of the subgroups[a]	Water[b] (%)
Meat		
Red meat	Beef (42), lamb(12), pork (11), veal (10), horse (11)	72
Poultry	Chicken (30),	73
Game	Red deer (7), rabbit	73
Processed meat	Air-dried meat (8), Hamburger (10), Ham (5), salami (5), sausage (11)	54
Vegetarian alternatives	Cereal burger (4), quorn, seitan, soy (4), vegetable burger (5)	
Fish		
Freshwater	Dace, grayling, hake, pike-perch, trout (5), whitefish, yellow-perch (4)	74
Marine, shellfish	Cod (5), dab, salmon (7), flounder, mackerel, plaice, redfish (4), scorpion fish, shrimps (4), sole fish, whiting (5)	77
Eggs		
Whole	Farm eggs	80
White	Industrial egg white	87
Yolk	Separated yolks from farm eggs	51
Dairy products		
Milk	Pasteurized milk (5), raw milk (5), dried milk (12)[c]	84
Cheese	Blue cheese, cottage cheese, fondue mixture, Feta, French hard cheese, herb cheese, Mascarpone, Swiss hard cheese (8), Swiss semi-hard cheese (5), Swiss soft cheese	43
Yoghurt and quark	Plain unflavored yoghurt (6), quark (6)	72
Vegetables and fruits		
Fresh fruits	Apple, apricot, avocado, banana, berries (blackberries, blueberries, cherry, citrus fruits (clementine, grapefruit, lime, lemon, nectarine, orange) currants, grape, kiwi, mango, melon, raspberries, strawberries), papaw, peach pear, pineapple, plum	83
Nuts	Cashew, hazelnut, nut mix (cashew, peanut, walnut) peanut	nd
Nightshade vegetables	Tomato, eggplant, bell peppers (green, yellow and red)	92
Fresh vegetables	Asparagus, cauliflower, carrot, corn, cucumber, fennel, green beans, kohlrabi, leek, lentils, onion, peas, radish, red cabbage, sprouting broccoli, pumpkin, zucchini	89
Salads	Endive, lamb's lettuce, iceberg lettuce, lettuce, lollo rosso	95
Herbs	Basil, chives, marjoram, mint, parsley, rosemary, water cress	90
Mushrooms	Boletus, black fungus, champignon, chanterelle, shiitake, truffle	88
Cereals		
Bread	Brown bread (11), corn bread (6), croissant (8), kernel bread (5), milk bread (6), potato bread, roll (12), White bread (22), whole meal bread (4), zwieback	21
Breakfast cereals	Plain, high fiber, sweetened, wholegrain	
Baked confection	Biscuit, cake (8), cookies (4)	

Table 1 (*continued*)

Food group	Description and composition of the subgroups[a]	Water[b] (%)
Pasta	Durum pasta (10), Durum pasta with egg (6), whole meal pasta	nd
Potato[d]	Bintje new potatoes	81
Rice	Long grain, brown rice, white rice (8)	nd

[a] From one to three different samples of the same kind of food were measured; otherwise the number of samples is given in parentheses.

[b] Measured as water loss after freeze drying, nd = not dried.

[c] Milk powder samples obtained from the Swiss Federal Dairy Research Station.

[d] Potato as a starchy food was not classified under nightshade vegetables (solanaceae) but under cereals.

3. Results and discussion

3.1. Accuracy of the measurements

The results obtained using the isotope dilution ICP-MS method for the analysis of certified reference materials and other reference materials in parallel with the food samples are listed in Table 2. The values obtained for the certified reference materials agree with the certified concentrations at the 95% confidence limit or with the available literature findings. The measured iodine concentrations in food were only accepted when the results of the quality assurance material fell within the 95% confidence limit of the certified mean. Even materials with low iodine concentrations such as bovine muscle yielded good results. In addition, drift effects in long series of measurements that might influence accuracy by normal external calibration had no deleterious effect on isotope ratio measurement, and thus on the accuracy of the results.

3.2. Iodine content of Swiss foods

The concentration levels of iodine measured in dry matter of basic food groups are summarized in Table 3, in which the samples are grouped according to similarity of the food matrix. The concentration of iodine in the foods examined in this work varies not only between samples but also within sample groups over a relatively wide range. The disparity in the data makes average values somewhat uncertain in relating the probable iodine content to a particular food group. For most foods, the mean iodine concentration exceeded the corresponding median, indicating that the distribution is skewed towards higher values. If high outlying values are present in a sample, the median will be a better centering constant to describe the iodine content, e.g. frozen vegetables. However, both values indicate in a general way the range of iodine concentrations in food.

Several important findings can be drawn from these descriptive statistics. Clearly, the richest natural food sources of iodine are marine fish. Concentrations were found to be in the range 0.39–6.9 μg/g. Also, high iodine concentration ranging from 0.15–2.1 μg/g were found in milk and

Table 2
Iodine in standard reference materials compared with the certified and reported values

Standard Reference Material	*n*	Obtained value (μg/g)	Certified value (μg/g)
Whole Milk Powder, NIST RM 8435	44	2.42 ± 0.07	2.30 ± 0.4
Whole Egg Powder, NIST RM 8415	26	1.99 ± 0.07	1.97 ± 0.46
Peach Leaves, NIST SRM 1547	21	0.31 ± 0.009	0.32 ± 0.02 a
Hay Powder, BCR CRM 129	9	0.18 ± 0.027	0.167 ± 0.024
Bovine Muscle NIST RM 8414	71	0.041 ± 0.003	0.035 ± 0.012
Durum Wheat Flour, NIST RM 8436	9	0.005 ± 0.004	0.006 ± 0.004

[a]Obtained value = Mean and 95% confidence interval for n independent determinations, the iodine concentration of 0.32 μg/g in SRM 1547 was taken from literature (Gélinas, 1998).

dairy products and eggs, whereas the values obtained for plant foods were generally low and fell in the range 0.002–0.7 μg/g.

The iodine content in fishes varies considerably at high levels. Classification of fish populations according to iodine concentration is however difficult as there is large variation between fishes of the same species (Karl et al., 2001). Iodine in fish reflects the content in the water they inhabit. Examining the fish data in Table 3, the influence of seawater and its mineral composition on iodine concentration is evident. Marine fish had about a six-fold higher iodine level than freshwater fish; however, the data have overlapping ranges of values.

Milk contained relatively high amounts of iodine and its iodine was partly derived from iodinated cattle feed supplements used in the dairy industry. The amount of iodine that is secreted in milk is not limited as it is with many other trace elements. The response of iodine concentration in milk to changes in iodine intake through feed is rapid (Swanson et al., 1990; Herzig et al., 2003). Seasonal variations reflect the amount of indoor feeding during the colder months versus outdoor pasture feeding during the warmer months. The samples measured in this study were winter milk with a higher iodine level than comparable summer milk (Sieber et al., 1999).

The content of cheese does not reflect the iodine level of the milk from which it was produced (Sieber, 1998). At some point in the cheese manufacturing the curd is separated from the whey, and most of the iodine present in the milk partitions into whey. Added salt used for cheese manufacturing is generally iodized and the salting will affect the concentration and the distribution of iodine within the cheese, e.g. soaking the pressed cheese in brine (Hoffmann et al., 1997).

Iodine is readily enriched in milk, whereas meat of dairy cows does not usually have high concentrations. Swanson et al. (1990) demonstrated that the concentration of iodine in muscle tissue of cows was not significantly affected by an increase of supplemental iodine of up to 2 μg/g in the feed. Consequently, the type of meat (beef, veal, pork, lamb and horse) had no significant effect on the iodine concentration ($P = 0.74$). For this reason all red meats were pooled, as presented in Table 3.

Kaufmann et al. (1998) conducted a similar study on laying hens and found that iodine supplementation of feed enriched iodine in eggs, but not in the breast muscle tissue. It is interesting to note in this respect that the medians of red meat and poultry compare quite well with game meat although the iodine intake of deer is based on natural feed. The results of the

Table 3
Iodine in basic food groups

Food group	n	Iodine contents in dry weight samples (ng/g)				
		Mean	±SD	Median	Min.	Max.
Cereals and grains						
Bread	76	393	±213	392	25	1032
Wheat	11	35	±11	37	11	47
Baked confectionery	13	245	±260	148	32	893
Pasta	17	79	±117	45	6	322
Rice	11	333	±341	250	11	934
Potatoes	3	16	±11	18	4	26
Breakfast cereals	10	42	±51	22	9	174
Meat and proteins						
Red meat	86	59	±71	37	7	555
Poultry	30	66	±112	34	10	627
Game	7	34	±14	33	15	48
Processed meat	39	335	±395	106	20	1254
Egg	10	1625	±294	1620	1236	2140
Egg, yolk	4	1413	±886	1170	711	2600
Egg, white	14	219	±74	193	132	347
Fish, marine	34	2112	±1713	1440	387	6926
Fish, freshwater	17	375	±436	205	11	1571
Vegetarian alternatives	17	109	±120	70	14	396
Dairy products						
Milk	22	690	± 170	675	330	1107
Yoghurt	12	670	± 313	556	347	1239
Cheese	27	473	± 289	396	146	1323
Fruit and vegetables						
Fresh fruit	62	18	±15	15	2	75
Fresh vegetables	36	47	±43	33	9	203
Leafy vegetables (Salad)	19	236	±181	153	46	703
Nightshade vegetables	6	130	±95	95	80	322
Frozen or canned vegetables	16	1203	±1571	498	46	1571
Mushrooms	10	211	±108	222	44	426
Nuts	13	218	±107	216	20	374

present study corroborate results from these previous studies and provide further evidence that iodine is not accumulated in muscle tissue.

The iodine content of plant foods varied from species to species (Table 3). Most nutrient components of plants under study such as fruits and vegetables had rather low iodine contents with the exception of herbs, leafy and nightshade vegetables. Frozen and canned vegetables tend to be packaged with iodized salt, which however did not consistently affect the iodine content as the values ranged from vegetable background concentrations of 0.05–5 μg/g. Mushrooms had relatively high iodine contents. Fair amounts of iodine were also detected in nuts. Like most other plant products, wheat grains did not derive much iodine from the soil. However, baked products had high iodine levels. The high iodine content of bread comes principally from the addition of iodized salt to the dough.

In conclusion, it can be pointed out that the bulk of iodine that is found in foods with high contents is incorporated either by the use of iodized salt in the manufacturing process or by fortifying animal feeds. About 70% of salt for the Swiss industrial food production is iodized (20 μg/g).

3.3. Comparison of iodine contents with previously published data

Table 4 compares the results of the present study with published data. The most comprehensive data tables of iodine contents in food are those of the Chilean Iodine Educational Bureau published in 1952. All publications note the variability of the iodine content in foods. In general, the data compare within order of magnitude and some results of different studies are quite close to each other. Similar base levels have been reported for vegetables, fruits and wholemeal. The distribution of iodine throughout the measured milk and egg samples is non-uniform and reflects the use of iodine-containing feed supplements. Some of the American and British samples were cooked prior to measurement. The different methods such as boiling, frying or baking seem to have a limited effect on iodine loss (Chavasit et al., 2002). Interestingly, the iodine content of meat and poultry in the American samples exceeds the levels found in the other studies. Also, the high iodine content in American bread is attributed to the use of iodate as dough conditioner.

Table 4
Comparison of iodine contents from the present study with previous results

Food	Mean iodine content in fresh weight samples with range or ± standard deviation (ng/g)				
	International[a] 1923–1949	Great Britain[b] 1977–1979	Finland[c] 1980	USA[d] 1982–1991	Switzerland 2000–2001
Fish, marine	832 (163–3180)	750 (320–1440)	460	1160 (±880)	486 (89–1593)
Fish, freshwater	30 (17–40)	—	165	—	98 (3–408)
Meat	32 (27–97)	50 (20–90)	<50	180 (±180)	17 (2–155)
Poultry	—	75	90	170 (±190)	18 (10–169)
Milk	47 (35–56)	230 (50–550)	169	200 (±80)	124 (59–199)
Eggs	93 (1–324)	525	170	480 (±390)	324 (247–428)
Wholemeal	59 (2–320)	<50	<100	—	33 (11–47)
Bread	56 (2–132)	(<50–100)	100	910±840	310 (20–815)
Fruits	18 (10–29)	(<20–80)	—	<30	3 (0.3–13)
Vegetables	29 (12–201)	(<20–280)	<10	<10	5 (1–22)

[a] Chilean Iodine Educational Bureau (1952).
[b] Wenlock et al. (1982).
[c] Varo et al. (1982).
[d] Pennington et al. (1995).

3.4. Contribution of the food groups to dietary exposure to iodine in switzerland

Per capita exposures from food were estimated using statistics on food consumption (Zimmerli et al., 1998; Schweizerischer Bauernverband, 2003) and the corresponding mean iodine concentrations (Table 5). The daily average consumption (dry weight) of bread and milk ranked among the highest of all foods. The results in Table 5 implicate milk and bread baked with iodized salt as significant contributors to iodine intake. Fish is the richest source of iodine; nevertheless the contribution of iodine was low because fish was consumed to a lesser extent. More important dietary sources were eggs and cheese, and the remaining foods were nutritionally less important because they did not contain substantial amounts of iodine or the levels of consumption were low. The total dietary iodine exposure for average consumers amounted to approximately 140 μg/day, which was somewhat below the recommended iodine intake of 150 μg/day. The complementary use of iodized kitchen salt certainly changes the balance from sub-optimal to adequate iodine nutrition. In a recent study Hess et al. (2001) showed in a school-age children survey that the iodine status of the Swiss population was adequate.

In addition to the dietary sources that were estimated, processed or industrially produced foods may further increase the iodine intake. For example, processed meat is also part of the Swiss diet; however, its contribution to the iodine intake could not be quantified (Eichholzer et al., 2000).

The estimate method provides information on per capita levels of iodine intake but fails to address population groups whose food consumption pattern diverge widely from the average. Potential iodine deficiencies of vulnerable population groups such as pregnant women or infants,

Table 5
Per capita iodine exposures from Swiss foods

Food	Daily average food consumption[a] (g/day)	Mean iodine concentration (μg/g)	Daily iodine intake (μg)
Pasta	34.0	0.08	2.7
Bread	148	0.39	57.8
Rice	14.9	0.33	5.0
Potato	150	0.016	2.4
Nuts	24.7	0.22	5.4
Fresh vegetables[b]	19.4	0.11	0.9
Canned vegetables	3.2	1.2	3.9
Fresh fruits	36.8	0.018	0.7
Red meat	30.9	0.059	1.8
Poultry	6.7	0.066	0.4
Game meat	0.5	0.034	<0.1
Eggs	5.8	1.6	9.5
Fish, including frozen fish[c]	5.3	1.2	6.4
Milk[d]	47.8	0.60	28.7
Cheese	26.8	0.47	12.6
All foods			139.5

[a] Mean values from 1999 trough 2001 (dry weight).
[b] All vegetable groups from Table 3 were combined and a weighed mean was calculated.
[c] Marine and freshwater fish were averaged.
[d] The year-round exposure from milk was estimated using previously published data as well (Sieber et al., 1999).

who may be exposed to low iodine levels, are not revealed by intake estimates based on average consumption data. Nevertheless, the estimated intake data are the basis for comparing different groups of foods as nutritional sources of iodine and reveal the fact that bread baked with iodized salt is the most important dietary iodine source in Swiss diet, which had not been generally recognized so far.

4. Conclusions

For the study of the iodine intake in a large population group, calculation from the iodine contents of different foods and the average quantities consumed is a method that is dependant on the quality and completeness of the data. The results confirm previous data on large variability of iodine contents even in foods belonging to similar groups. Consequently, the estimated daily iodine intake cannot be extrapolated safely to the Swiss population and accordingly there is a need to carry out human diet studies, which involve measurement of urinary iodine.

The study has highlighted bread as the major dietary source of iodine, which is a function of the addition of iodized salt. The practice of using iodized salt in bread production has always been voluntary, thus it may change over time.

The information gained from this study is not directly applicable to other countries because of the variety of iodine deficiency control programs. However, the data pattern serves as an example for future studies exploring dietary exposure of iodine aimed at evaluating the contributions of food groups.

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