

## Selenium content of a range of Irish foods

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### Abstract

Over the past two decades selenium (Se) intakes have fallen in the UK and elsewhere in Europe, as a result of reduced imports of Se-rich, high protein wheat for bread-making flour from North America and Canada. However, no analyses of the Se content of Irish flours/breads or other foods have been published, thus making it difficult to estimate the daily dietary Se intakes in Ireland. In the present study, the Se content of selected Irish foods, especially breads and flours, was determined by hydride generation atomic absorption spectrometry after acid digestion. Less refined Irish wheat flours (wheatmeal, course wholemeal, wheatbran) had higher Se levels (7.7–9.9  $\mu\text{g}/100\text{ g}$ ) than the more refined flours (plain, self-raising, baker's, strong; 6.0–6.9  $\mu\text{g}/100\text{ g}$ ). Irish brown breads had higher Se levels (8.6–12.9  $\mu\text{g}/100\text{ g}$ ) than those of white bread (6.6  $\mu\text{g}/100\text{ g}$ ). In conclusion, it appears that Irish flours and breads do not contain as much Se as North American or Canadian flours/breads and contain only slightly more Se compared with those currently used in the UK. © 2001 Elsevier Science Ltd. All rights reserved.

*Keywords:* Selenium content; Bread; Flour; Irish diet

### 1. Introduction

The trace mineral selenium (Se) is an essential nutrient of fundamental importance to human biology. It is a key component of a number of functional selenoproteins required for human health. There is evidence that Se has an antioxidant role through the selenoenzyme glutathione peroxidase (*EC* 1.11.1.9; GSHPx), but also through other more recently discovered selenoenzymes (Arthur & Beckett, 1994). Se is also involved in thyroid metabolism through the enzymes, types I and II iodothyronine deiodinase, although the latter is not itself a selenoenzyme (Arthur, 1991). Selenoproteins have also been associated with maintenance of fertility and possibly some anti-cancer effects (Rayman, 2000). In all, about 35 selenoproteins have been identified, though many have roles that have not yet been fully elucidated (Rayman, 2000). The activity of these selenoproteins depends on an adequate Se supply from the diet.

Some epidemiological studies have revealed an inverse correlation between Se status and cardiovascular disease (Kok et al., 1989; Oster et al., 1986; Salonen & Huttu-

nen, 1986). Epidemiological evidence has also been cited that links certain types of cancer with low Se status (Kok, De Bruijin, Hofman, Vereemen, & Valkenburg, 1987; Willet et al., 1983). More recently, Se deficiency has been suggested to have a role in the aetiology of other pathologies, such as oxidative stress or inflammatory conditions, diabetes mellitus, hepatopathies, HIV infection (for reviews, see Holben & Smith, 1999; Navarro-Alarcón & López-Martínez, 2000; Rayman, 2000). In the context of these health effects, low or diminishing Se status in some parts of the world, notably in some European countries, is giving cause for concern. For example, about 20 years ago, dietary Se intakes in the UK were 60  $\mu\text{g}/\text{day}$  on average, whereas a 1994 survey commissioned by the UK Ministry of Agriculture, Fisheries, and Foods found that Se intake had dropped to 34  $\mu\text{g}/\text{day}$  (Barclay, McPherson, & Dixon, 1995; Rayman, 1997), considerably less than the reference nutrient intakes of 75  $\mu\text{g}/\text{day}$  for men and 60  $\mu\text{g}/\text{day}$  for women. There are similar findings elsewhere in Europe (Rayman, 1997). The falling intakes are reflected in lowered Se concentrations in whole blood and serum. In seven EU countries average serum Se levels fell between 1983 and 1993 to 79  $\mu\text{g}/\text{l}$  (Alfthan & Néve, 1996; Rayman, 1997), which is less than the value

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of 100 µg/l believed to be required for optimal activity of cytosolic GSHPx, an indicator of Se depletion.

The substantial fall in Se intake can largely be explained by the drop in imports of Se-rich, high-protein wheat for bread-making flour from North America and Canada (Barclay et al., 1995). Due to EU policies and changes in bread-making technology, there has been an increased use of low-Se, low protein UK and European wheat varieties (Rayman, 1997). While, in 1985, bread was a major source (47%) of dietary Se in the UK, it now only supplies ~20% of the dietary provision (Barclay et al., 1995). Similar trends have been noted in other European countries (Rayman, 1997). In Ireland, bread, which was also formerly made with this high protein, Se-rich imported Canadian/North American flour, is now being made with a higher proportion of home-grown and European flours. However, no analyses of the Se content of Irish flours and breads, or any other foods grown or produced in Ireland, have been published.

Therefore, the purpose of this study was to obtain data on the Se content of selected Irish foods, in particular Irish breads and flours. This information is urgently required so that the daily dietary Se intakes in the Irish population can be estimated from available food consumption data. While Se intake levels are documented for numerous European countries (see reviews by Navarro-Alarcón & López-Martínez, 2000; Rayman, 2000), there is as yet no information available for Ireland.

## 2. Materials and methods

### 2.1. Food selection and preparation

Foods were purchased from retail outlets across Cork City. On purchase, the foods were given a laboratory coding and stored, as appropriate (ambient, chill, or frozen) until required for sampling and analysis. The foods were milled, homogenized, or macerated in a food processor to obtain a uniform material which could be readily sub-sampled for Se analysis. Specifically, in the case of flours, suitably sized samples of flour were achieved by mixing and quartering as described previously by Foster and Sumar (1995).

### 2.2. Sample digestion

Representative food samples were placed in Pyrex glass digestion tubes which had previously been soaked overnight in 20% v/v nitric acid (Aristar, BDH Ltd, Poole, Dorset, UK), rinsed in double distilled, deionized water and left to air-dry. In the case of milks, samples were digested by the method of Foster and Sumar (1996). In brief, 10 ml of a nitric acid (S.G. 1.42 g/ml;

Aristar, BDH) — perchloric acid (S.G. 1.70 g/ml; Aristar, BDH Ltd) mixture (4:1) were added and the digesting tubes stoppered and left in a fume cupboard at ambient temperature for digestion overnight. The following day, the tubes were placed in an ultrasonic bath for 2–3 min. The tube contents were heated to 150°C (without stoppers) in a Kjeldatherm digesting block (Tecator, Sweden) for approximately 30 min until the evolution of brown fumes of NO<sub>2</sub> had ceased. Then the temperature was gradually increased to 220°C for 60 min after the appearance of dense white fumes of perchloric acid. Care was taken to ensure that the solution was not heated to complete dryness because of the explosive nature of metal perchlorates (Analytical Methods Committee, 1979). On cooling, the tubes were made to 5 ml volume with double distilled, deionized water. Five millilitres of hydrochloric acid (S.G. 1.18 g/ml; Aristar, BDH Ltd) were added and heated at 100°C for 30 min to reduce Se(VI) to Se(IV).

For food samples other than milk, a nitric–perchloric–sulphuric acid digestion method was used as described by Hershey and Oostdyk (1988).

Background analytical levels of Se were assessed by running blank acid digestions; the levels detected were insignificant. The analytical method was verified by analysis of BCR Reference materials (Skim milk powder [CRM 063R], Bovine muscle [CRM 184], Wholemeal flour [CRM 189], Brown bread [CRM 191]; Laboratory of the Government Chemist, The Office of Reference Materials, Teddington, Middlesex, UK), samples of which were treated similarly throughout the entire digestion process (Table 1).

### 2.3. Hydride generation atomic absorption spectrometry

After the sample digest had cooled, it was transferred into a 25-ml volumetric flask and made up to volume with double-distilled, deionised water. This was placed in the hydride generation reaction chamber (Varian, VGA-76 model, Victoria, Australia) and 5 ml (2.5% w/v) sodium borohydride (98% crystalline, Sigma Chemical Company Ltd, Poole, Dorset, UK), dissolved in 0.5 N-NaOH (May and Baker Ltd, Essex, UK), was injected

Table 1  
Comparison of the selenium concentration of reference samples as determined in our laboratory with the certified values

Material	Se concentration (µg/kg) <sup>a</sup>	
	This study	Certified value
Skim milk powder [CRM 063R]	133±8	(129) <sup>b</sup>
Bovine muscle [CRM 184]	186±15	183±12
Wholemeal flour [CRM 189]	128±12	132±10
Brown bread [CRM 191]	23±4	(25) <sup>b</sup>

<sup>a</sup> Mean and standard deviation.

<sup>b</sup> The selenium concentration in these materials is not certified for Se.

into the chamber. This solution was freshly prepared for every analysis as it is only stable for approximately 2 h. Atomic absorption measurements were performed on a Varian atomic absorption spectrophotometer (Varian, Spectra-AA 600 model, Mulgrave, Victoria, Australia) attached to an autosampler (Varian, SPS-5 sample preparation system, Victoria, Australia). Instrument conditions were the following: *Radiation source*: Se hollow-cathode lamp, 7 mA; *Wavelength*: 196.0 nm; *Spectral band pass*: 1.0 nm; *Flame*: Air-acetylene, 10 psi, flow rate 3 l/min; *Hydride generation* — *Tube temperature*: approx. 900°C; *Carrier gas*: Nitrogen at 10 psi; *Reaction time*: 20 s; *Reaction volume*: 20 ml. An external calibration curve was prepared, using a 1000 ppm selenium [Se(IV)] standard in 0.5 M nitric acid (Spectrosol<sup>®</sup>, BDH Ltd) over the range of 0–0.02 µg/ml from a 1 ppm working stock solution using 6 N hydrochloric acid (Aristar, BDH Ltd) as a diluent. Peak area response was plotted against the Se concentrations.

#### 2.4. Protein determination of flours

Protein content was determined by a modification of the Kjeldahl method.

#### 2.5. Statistical analysis

Data are presented as means with their standard errors and range. Se content data within a distinct food grouping (e.g. flours, breads, meats, etc.) was subjected to one-way ANOVA (Snedecor & Cochran, 1967) or unpaired *t*-tests (in the case of comparisons between only two foods or food groupings, e.g. white versus brown flours). To follow up the ANOVA, all pairs of means were compared by the method of least significant difference (Snedecor & Cochran, 1967).

### 3. Results

The mean Se concentration in 36 separate foods and food ingredients, typically found in the Irish diet, together with the range and standard error is shown in Table 2. The results, blank corrected, are expressed in micrograms of Se/100 g (wet weight) for all foods.

Mean concentrations of Se in retail and bakers' wheat flours commonly used in Ireland, ranged from 1.3 to 9.9 µg/100 g (overall mean, 6.4 µg/100 g). The less refined flours (e.g. wheatmeal, course wholemeal and wheatbran) had significantly higher ( $P < 0.0001$ ) mean Se concentrations (7.0–9.9 µg/100 g) than those of the more refined white flours (e.g. plain, bakers', self-raising, and strong white, 6.0–6.9 µg Se/100 g). The gluten-free flour had dramatically lower Se and protein concentration (1.3 µg and 4.2 g/100 g for Se and protein, respectively) compared with the other flours (6.0–9.9 µg

and 9.3–14.2 g/100 g for Se and protein, respectively). There was a significant ( $P < 0.001$ ) correlation of Se content with protein ( $r = 0.96$ ) in the flours examined in the present study.

Mean concentrations of Se in Irish breads ranged from 1.8 to 12.9 µg/100 g (overall mean, 7.6 µg/100 g). The amount of Se in brown breads (8.6–12.9 µg/100 g) was significantly higher ( $P < 0.0001$ ) than that of the white bread (6.6 µg Se/100 g). Of particular note was that the gluten-free and soda breads were relatively low in Se (1.8 and 3.6 µg/100 g, respectively).

Mean concentrations of Se in meats (which were all raw and lean) ranged from 8.1 to 11.5 µg/100 g. The mean Se content of pork (10.4 µg/100 g) was significantly higher ( $P < 0.01$ ) than that of the lamb and beef (8.8 and 8.1 µg/100 g, respectively), with no significant difference in Se concentration between the latter two meats. Chicken (white breast meat) and pork had similar Se levels, but chicken had a higher Se concentration (11.5 µg/100 g) than that of lamb and beef ( $P < 0.0001$ ). The fish samples, which included representative quantities of white fish (plaice and cod), shellfish (crab) and oily fish (tuna), were richest in Se, ranging from 28 to 70 µg/100 g (overall mean, 41 µg/100 g). Canned fish (tuna and crab) had significantly higher Se concentrations ( $P < 0.0001$ ) than white fish (plaice and cod). Of all the foods analysed in the present study, canned tuna was the richest source of Se.

Mean concentrations of Se in dairy produce ranged from 1.4 to 10.1 µg/100 g (overall mean, 5.0 µg/100 g). The three milk types (i.e. skim, low-fat, and full-fat cow's milk) had similar Se concentrations (1.4–1.8 µg/100 g), but all three were considerably lower in Se content than other dairy produce. For example, cheese contained between 6.1 and 10.1 µg/100 g, on average. Egg white contained 6.4 µg Se/100 g, whereas egg yolk had a much higher ( $P < 0.0001$ ) Se content (24.1 µg/100 g).

Breakfast cereals (Rice Krispies, Alpen's muesli, and porridge) were relatively poor sources of dietary Se (1.4–3.1 µg/100 g). Similarly, the two vegetable foodstuffs examined in the present study, i.e. red kidney beans and mushrooms, were also relatively poor sources of Se (1.8 and 3.3 µg/100 g, respectively).

### 4. Discussion

Plant species do not require Se for growth and can be very low in Se, in contrast to animal species, for which Se is an essential nutrient, and which will not survive if tissue levels are too low (Barclay et al., 1995). It is not surprising, therefore that, of the foods analysed in the present study, meat and fish were the richest sources of Se, whereas vegetables, such as red kidney beans and mushrooms, were among the poorest.

Table 2  
Selenium content of selected foods included in the Irish diet<sup>a</sup>

Food	n	Se content ( $\mu\text{g}/100\text{ g}$ )*		
		Mean	S.E.M.	Range
<i>Wheat flour</i>				
Gluten-free	10	1.3a	0.3	1.0–2.0
Plain white	21	6.2b	1.3	4.6–9.5
Self-raising	15	6.0b	0.9	4.4–7.5
Bakers	12	6.0b	0.4	5.5–6.7
Strong white	18	6.9c	1.4	4.8–9.0
Wheatmeal	14	7.0c,d	1.0	5.2–8.7
Course wholemeal	14	7.8d	1.6	6.2–10.1
Wheatbran	12	9.9e	1.0	8.8–12.1
<i>Bread</i>				
Gluten-free	20	1.8a	0.3	1.5–2.6
White bread	18	6.6b	0.9	5.3–9.5
Soda bread	22	3.9c	0.8	2.0–5.0
Brown bread	20	11.8d	1.4	9.4–14.6
Wholegrain	26	12.9e	2.6	8.4–15.8
Mixed grain	26	8.6f	2.2	5.6–14.6
<i>Meat</i>				
Beef (raw)	14	8.1a	1.5	6.1–10.5
Pork (raw)	14	10.4b	1.8	8.2–12.9
Lamb (raw)	16	8.8a	1.2	7.1–10.9
Chicken (raw, white)	13	11.5b	2.1	8.6–14.7
<i>Fish</i>				
Plaice (raw)	10	28.2a	1.2	26.8–29.8
Cod (raw)	10	26.5a	2.5	29.9–23.3
Tuna (canned in brine)	10	70.1b	5.0	63.7–78.9
Crab (canned in brine)	22	39.0c	2.4	34.7–43.7
<i>Milk and dairy</i>				
Milk (full-fat)	10	1.8a	0.3	1.4–2.2
Milk (low-fat)	10	1.8a	0.4	1.3–2.3
Milk (skimmed)	12	1.4a	0.7	1.0–2.8
Cheese (red cheddar)	10	10.1b	0.9	8.6–11.1
Cheese (white cheddar)	12	9.1c	1.4	7.5–11.5
Processed cheese	10	6.1d	1.1	4.5–7.5
Egg white (raw)	19	6.4d	0.6	5.6–8.1
Egg yolk (raw)	11	24.1e	2.1	22.2–28.2
<i>Breakfast cereals</i>				
Rice Krispies	10	1.4a	0.2	1.0–1.7
Oatmeal	28	3.1b	1.8	2.8–3.4
Muesli	10	2.3c	0.3	1.8–2.6
<i>Pasta</i>				
Spaghetti (raw, wholewheat)	20	6.6	0.8	4.9–7.8
<i>Vegetables</i>				
Red kidney beans (raw)	18	1.8a	0.5	1.0–2.8
Mushroom (raw)	31	3.3b	1.5	2.5–3.8

<sup>a</sup> n = number of samples analyzed; S.E.M. = standard error of the mean. \*Concentration expressed on a wet weight basis. a,b,c,d,e,f Mean values within a column for any one food grouping which have different letters are significantly different using least significant difference ( $P < 0.05$ ) as follow-up to ANOVA, or by unpaired *t*-test in the case of comparisons between only two foods or food groupings.

The Irish foods selected for Se determination in the present study were chosen to include those frequently-consumed foods that contribute a major proportion of Se to the diets of the Irish population. Meat, poultry, and fish, bread and cereals, and milk and dairy products, are the food groups that provide most of the total Se in the diets of northern Europeans (see review by van Dokkum, 1995).

The Se content of Irish beef (8.1  $\mu\text{g}/100\text{ g}$ ) was similar to that recently reported for UK beef (7.6  $\mu\text{g}/100\text{ g}$ ; Barclay et al., 1995), but about half that of US beef (19.0  $\mu\text{g}/100\text{ g}$ ; Pennington, Scheon, Salmon, Young, Johnson, & Marst, 1995; Zhang, Shi, & Spallholz, 1993). Irish pork (10.4  $\mu\text{g}/100\text{ g}$ ) had a slightly lower Se concentration than that of UK pork (14.0  $\mu\text{g}/100\text{ g}$ ; Barclay et al., 1995) and much lower than USA pork (26.5  $\mu\text{g}/100\text{ g}$ ; Pennington et al., 1995; Zhang et al., 1993). Irish lamb (8.8  $\mu\text{g Se}/100\text{ g}$ ), on the other hand, had about double the Se concentration of UK lamb (3.8  $\mu\text{g}/100\text{ g}$ ; Barclay et al., 1995), but had a much lower Se content than lamb in the USA (21.0  $\mu\text{g}/100\text{ g}$ ; Schubert, Holden, & Wolf, 1987). Irish chicken (11.5  $\mu\text{g}/100\text{ g}$ ) had a slightly lower Se level than US chicken (13.0  $\mu\text{g}/100\text{ g}$ ; Pennington et al., 1995; Zhang et al., 1993), but had a higher Se content than UK chicken (7  $\mu\text{g}/100\text{ g}$ ; Holland, Welch, Unwin, Buss, Paul, & Southgate, 1991).

Differences in Se concentrations of carcass meats from the different countries presumably reflect the soil Se availability in their areas of origin, which in turn affects the intake of Se by grazing animals. In general, the Se concentrations of meats of US origin was much higher than that of Irish and UK meats. On the other hand, the Se concentrations of meats of Irish and UK origin were relatively similar, with the exception of lamb. It is not clear why Irish lamb contained nearly twice as much Se as lamb of UK origin (Barclay et al., 1995). It is possible that differences exist between the two countries in the Se content of the soil on which crops are grown and livestock and poultry are raised; however, this seems unlikely, as the levels of Se in beef and pork from both countries were similar. Another possibility is that differences exist between the two countries in terms of the level of Se supplementation of animal feeds, particularly for lambs.

The levels of Se in fish found in the present study varied widely with the type of fish. For example, tinned fish had a much higher Se content than fresh fish. In general, the levels of Se in meat and fish are relatively high and meat and fish could be considered a good source of dietary Se for humans.

Cow's milk can make a significant contribution to Se intake where it is consumed regularly but, like cereals, its Se content shows marked geographical variations (Hojo, 1982). The Se content of Irish dairy produce (milks, cheeses, egg yolk and egg white) was similar to

that reported in UK dairy products (Barclay et al., 1995; Holland et al., 1991; Thorn, Robertson, Buss, & Bunton, 1978).

The Se content of Irish white flours (plain, self-raising, bakers, and strong), examined in the present study, was at least double that recently reported for UK flours (2.3–2.5 µg/100 g; Barclay et al., 1995). It was, however, much lower than that reported for white flours in the USA (18.7–39.0 µg/100 g; Morris & Levander, 1970; Olson & Palmer, 1984) and Canada (26.0–30.0 µg/100 g; Arthur, 1972). Less-refined Irish wheat flours (wheatmeal, wholemeal, and wheatbran), examined in the present study, had only slightly higher Se concentrations (7.0, 7.8, and 9.9 µg/100 g, respectively) than UK flour (5.9 µg Se/100 g wholemeal flour; Barclay et al., 1995). Again, the Se levels of these Irish wheat flours were much lower than those reported for the USA (62.7–87.0 µg/100 g; Morris & Levander, 1970; Olson & Palmer, 1984) and Canada (56.0–65.0 µg/100 g; Arthur, 1972).

Unfortunately, to our knowledge, there has been no previously published data on the Se content of Irish flours with which to compare the levels observed in the present study. Therefore, it is unclear whether the Se levels of Irish flour blends have dropped over the last two decades, as they have in the UK (Barclay et al., 1995). For example, in comparison with the Se levels reported by Thorn et al. (1978) for plain and self-raising (4 µg/100 g), bread-making (42 µg/100 g), and wholemeal (53 µg/100 g) UK flours, Barclay et al. (1995) recently reported a range of 1.7–3.6 µg Se/100 g for white, of 1.8–3.7 µg Se/100 g for strong white, and 2.3–11.0 µg Se/100 g for wholemeal flour. This reduction in UK flour Se levels, especially in bread-making and wholemeal flours, can largely be explained by the drop in imports of Se rich, high protein wheat for bread-making flour from North America and Canada (Barclay et al., 1995). Levies imposed on foreign imports when the UK joined the European Union, coupled with changes in bread-making technology, have resulted in increased use of low-Se, lower-protein European and UK varieties (Rayman, 1997). A similar situation exists in Ireland, whereby higher proportions of home-produced and European wheat are being used in place of those of Canadian and North American origin. Therefore, while difficult to confirm, owing to the lack of previous data, it is likely that a parallel reduction in Se levels in flour used for bread-making has occurred in Ireland.

The Se content of Irish white bread (6.6 µg/100 g) was only slightly higher than that recently reported for UK bread (4.3–4.4 µg/100 g; Barclay et al., 1995), but was much lower than that for white bread in the USA (27.4–32.0 µg/100 g; Morris & Levander, 1970; Olson & Palmer, 1984; Schubert et al., 1987) and Canada (42–67 µg/100 g; Arthur, 1972). Irish brown breads had more than double the Se concentration (8.6–12.9 µg/100 g) of UK

bread (3.9–4.8 µg/100 g; Barclay et al., 1995), but were much lower in Se content than brown breads in the USA (41.0–67.6 µg/100 g; Morris & Levander, 1970; Olson & Palmer, 1984; Schubert et al., 1987) and Canada (65.0–71.0 µg/100 g; Arthur, 1972).

Bread is one of the important foods in the West as regards sources of Se, owing to its frequency and quantity of consumption. In the UK, while there has been little change in the Se content of white bread, the concentration in wholemeal bread has declined by 50% since 1986 (Barclay et al., 1995), reflecting this increased use of European wheat flour rather than Canadian/North American wheat flours. Since 1978, Barclay et al. (1995) estimated that the daily Se intake coming from bread and cereals dropped from 40.3 µg/day to 9.9 µg/day. Furthermore, the increases in meat (3.7 to 4.7 µg/day), poultry and fish (4.8 to 5.7 µg/day), and milk and milk products (5.1 to 5.5 µg/day) are small in comparison. Again, while difficult to confirm, owing to the lack of previous data, it is likely that this trend also exists in Ireland.

Of particular note, was the fact that the gluten-free flour and bread had extremely low Se levels (1.3 and 1.8 µg/100 g, respectively). This may be of significance in terms of the Se intake of individuals with celiac disease, especially as celiacs have been reported as having lower blood Se levels than healthy individuals (Hinks, Inwards, Lloyd, & Clayton, 1984).

In conclusion, it appears that Irish flours and breads do not contain as much Se as North American or Canadian flours and breads and have only slightly higher concentrations of Se than those currently used in the UK. While there is no data available to investigate whether a decrease in Se content of Irish breads mirrors those in the UK since 1973, based on the similarities in Se levels in Irish and UK breads, it would seem prudent that the Se intakes and Se status of the Irish population be investigated at this time.

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