

## Iodine concentration in Norwegian milk and dairy products

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The present study was conducted to determine the iodine concentration in Norwegian-produced milk and a selection of dairy products. The iodine concentration of eighty-five samples of milk and dairy products was analysed by inductively coupled plasma–MS. Low-fat milk and organic milk were sampled from nineteen and seven different locations in Norway, respectively, during the summer and winter season of 2000. Other milk and dairy products were chiefly collected during the summer season. Low-fat milk from the summer season had significantly lower median iodine concentration (88 µg/l, range 63–122 µg/l) compared with low-fat milk from the winter season (232 µg/l, range 103–272 µg/l). The median iodine concentration of organic summer milk (60 µg/l) was significantly lower than the iodine concentration of organic winter milk (127 µg/l). There were no significant differences in the low-fat-milk samples with regard to geographical sampling location. Whey cheese (Tine Gudbrandsdalsost) iodine concentration was significantly higher (803 µg/kg) than the median iodine concentration in casein cheeses such as Jarlsberg and Norvegia of 201 and 414 µg/kg, respectively. With a recommended iodine intake of 150 µg/d for adults, a daily intake of 0.4 litres milk meets the requirement with 25 % during the summer and more than 60 % during the winter season. Thus, milk and dairy products are important determinants of iodine intake in Norway.

**Iodine: Milk: Dairy products: Inductively coupled plasma–mass spectrometry analysis: Norway**

The trace element iodine is essential for normal activity of the thyroid hormones thyroxine and triiodothyronine. Clinical and sub-clinical manifestations of iodine deficiency, termed iodine-deficiency disorders, affect all stages of human life and are characterised by a variety of conditions (Hetzl, 1983; Delange, 1994). Iodine deficiency remains the single greatest cause of preventable brain damage and mental retardation worldwide (World Health Organization, 1996). Means to alleviate iodine-deficiency disorders are therefore of both national and international interest. Salt iodisation has been recommended as the preferred strategy to control and eliminate iodine-deficiency disorders by the WHO (World Health Organization, 1994).

Data on the iodine nutrition of the Norwegian population are very scarce. Iodine deficiency has been eradicated in Norway since 1950, when fortifying cattle fodder with iodine was started (Frey, 1986). Milk and dairy products have since then been considered to be the primary source of iodine in the Norwegian diet (Frey *et al.* 1993). The only good natural source of iodine is seawater fish and other marine products (Julshamn *et al.* 2001). Although Norwegians traditionally have had a high average intake of fish, this did not secure the iodine intake of inland population groups without access to the sea. Thus goitre used to be widespread wherever fish intake was low (Frey, 1986).

Food regulations permit the addition of 5 µg I/g NaCl to salt (Ministry of Health, 2002); however, the amount is too low to contribute significant amounts of iodine compared with milk and fish. This is due to the fact that only some brands of table salt are fortified with iodine and industrial salt used in food production is not fortified with iodine (NNR, 1996). With an estimated average intake of 3 g table salt/d, iodine-fortified salt only contributes 15 µg I, if used.

There has never been a systematic monitoring of the iodine concentration of Norwegian foods or the iodine intake from the Norwegian diet. The most recent studies were performed in 1971–2 and in 1985 in small groups of Norwegians and showed satisfactory iodine excretion in urine (Frey *et al.* 1993).

The iodine content of animal feedstuff is controlled by legislation in Norway. The fortification of cow fodder with 2 mg I/kg as calcium iodate, Ca(IO<sub>3</sub>)<sub>2</sub>, to protect animal health (Ministry of Agriculture, 2002) has also provided a source of iodine in human diets (Frey, 1986). In 1996, a new regulation in Norway was passed ensuring that cattle should spend at least 8 weeks each summer out of doors, grazing naturally (Ministry of Agriculture, 1996). The regulation probably contributes to better animal welfare, but also makes the animals' feed more vulnerable to local soil-mineral conditions.

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Analysis of iodine concentrations in foods in other countries has shown that the iodine content of dairy products varies according to season of the year and geographical origin of the milk (Baumann, 1990; Pennington, 1990; Lee *et al.* 1994; Karl & Munker, 1999; Larsen *et al.* 1999; Ministry of Agriculture, Fisheries and Food, 2000). The aim of the present study was to determine the iodine concentrations in Norwegian-produced milk and a selection of dairy products, and discuss eventual variations in results in relation to sampling site, seasonal production and production method.

## Materials and methods

### Collection of samples

Low-fat-milk samples (15 g fat/l) were collected from nineteen different locations in Norway (Fig. 1 and Table 1) during the winter and the summer seasons of 2000. Organic milk (not homogenised; 15 g fat/l) was sampled from seven different locations during the same period (Fig. 1 and Table 2). Organic milk is produced by farmers who practise organic farming. The organic products are controlled and certified by Debio, a government-approved authority,

which also provides a guarantee seal for the organic agricultural production in Norway (Debio, 2003). In Norway there are few dairies producing organic milk and the geographical range is therefore less than for non-organic milk.

The choice of low-fat milk was based on the widespread consumption of low-fat milk in Norway i.e. 54% of the total milk sales (AC Nielson, unpublished results). In addition, the iodine concentration of milk is shown not to vary in relation to the fat content in different types of milk (Varo *et al.* 1982; Lee *et al.* 1994). The dairies were divided into five groups dependent on geographical region in Norway: north, central, west, south and east (Fig. 1).

Furthermore, processed dairy products were collected during the summer season. Among the samples analysed were extra-low-fat milk (7 g fat/l), curdled milk (38 g fat/l) and probiotic milk with *Lactobacillus rhamnosus* GG (Biola; TINE, Oslo, Norway; 15 g fat/l), dairy products such as yoghurt (32 g fat/l), soured cream (200 g fat/l) and cheese. Several types of cheese were analysed, i.e. Jarlsberg (270 g fat/kg), Norvegia (270 g fat/kg) and whey cheese (Tine Gudbrandsdalsost; 290 g fat/kg). These three cheeses account for approximately 60% of the cheese sales in Norwegian grocery shops (AC Nielson, unpublished results).



**Fig. 1.** Map of Norway showing positions of the nineteen dairies where low-fat milk and organic milk were sampled. For locations of the dairies 1–19, see Table 1.

**Table 1.** Geographical location and iodine concentration of low-fat milk and organic milk sampled during the winter and summer seasons of 2000 in Norway

Geographical location§	Iodine concentration ( $\mu\text{g/l}$ )			
	Low-fat milk		Organic milk‡	
	Winter	Summer	Winter	Summer
North Mean	252	104		
Tana (1)	237	88		
Tromsø (2)	246	106		
Bodø (3)	272	117		
Central Mean	248	100		
Tunga (4)	270	101		
Høgset (5)	226	98		
West Mean	237	94		
Ålesund (6)	269	105	163	62
Byrkjelo (7)	223	76		
Bergen (8)	247	121		
Haugesund (9)	207	73	115	28
South Mean	225	72		
Sola (10)	232	73		
Setesdal (11)	220	78	127	87
Kristiansand (12)	249	73		
Porsgrunn (13)	199	63	264	60
East Mean	214	99		
Sem (14)	260	81	35	36
Ål (15)	103	nd		
Hamar (16)	232	117		
Odal (17)	225	85	365	68
Oslo (18)	213	88		
Indre Østfold (19)	253	122	102	17
Mean	231*	93	167†	51
SD (all)	34	17	111	25

nd, not determined.

\* Mean value was significantly different to that for low-fat milk from the summer season ( $P < 0.001$ ).

† Mean value was significantly different to that for organic milk from the summer season ( $P = 0.02$ ).

‡ Organic milk was sampled at only seven different locations.

§ The numbers in parentheses relate to the positions on the map (Fig. 1).

**Table 2.** Iodine concentration in milk ( $\mu\text{g/l}$ ) and dairy products ( $\mu\text{g/kg}$ ) produced in Norway during the winter and summer seasons of 2000

(Median values and ranges)

Product	<i>n</i>	Median	Range
Low-fat milk, winter ( $\mu\text{g/l}$ )	19	232*	103–272
Low-fat milk, summer ( $\mu\text{g/l}$ )	18	88‡	63–122
Organic milk, winter ( $\mu\text{g/l}$ )	7	127†	35–365
Organic milk, summer ( $\mu\text{g/l}$ )	7	60	17–87
Extra-low-fat milk, summer ( $\mu\text{g/l}$ )	5	103	66–128
Extra-low-fat milk, winter ( $\mu\text{g/l}$ )	1	234	
Curdled milk ( $\mu\text{g/l}$ )	6	114	100–221
Probiotic milk with LGG   ( $\mu\text{g/l}$ )	1	87	
Yoghurt ( $\mu\text{g/kg}$ )	5	147	115–179
Soured cream ( $\mu\text{g/kg}$ )	5	87	44–176
Jarlsberg cheese ( $\mu\text{g/kg}$ )	3	201	132–353
Norvegia cheese ( $\mu\text{g/kg}$ )	3	414	169–468
Whey cheese§ ( $\mu\text{g/kg}$ )	5	803	103–1360

LGG, *Lactobacillus rhamnosus* GG.

\* Median value was significantly different to that for low-fat milk from the summer season ( $P < 0.001$ ).

† Median value was significantly different to that for organic milk from the summer season (Mann–Whitney U test for non-parametric samples,  $P = 0.02$ ).

‡ Median value was significantly different to that for organic milk from the summer season ( $P = 0.006$ ).

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Representative samples of all types of milk (1 litre), yoghurt (175 ml), soured cream (300 ml) and cheese, casein and whey (1 kg) were shipped from each dairy to the National Institute of Nutrition and Seafood Research in Bergen in a cooled condition (approximately  $+4^\circ\text{C}$ ). An adequate amount of each sample was collected and frozen at  $-20^\circ\text{C}$  in 50 ml screw-capped polyethylene centrifuge tubes until analysis. Before analysis, milk samples were left at room temperature to thaw. Cheese samples were crushed in a frozen condition to produce small particles. Both cheese and soured-cream samples were weighed in frozen condition.

#### Sample preparation and analysis

The determination of iodine in milk and dairy products was carried out using inductively coupled plasma–MS after digestion in tetra methyl ammonium hydroxide (Fecher *et al.* 1998). A test sample of 1 ml milk or 0.1–0.2 g solid material was added to 5 ml deionised water and 1 ml tetra methyl ammonium hydroxide. Extraction was carried out in a dry oven at  $90^\circ\text{C} \pm 3^\circ\text{C}$  for 3 h. After cooling, 1  $\mu\text{g}$  Te was added as internal standard to the sample solution, which was then diluted to a final volume of 25 ml with water. This gives a Te concentration in the solution analysed of 40  $\mu\text{g/l}$ . In order to avoid blocking of the nebuliser, all samples were filtered through a 0.45  $\mu\text{m}$  Sartorius Minisart RC 25 filter (Göttingen, Germany). A high degree of fat in the sample solution may cause problems; this was avoided by use of a syringe to extract solution before filtration. The iodine concentrations in the samples were determined by standard addition calibration (Julshamn *et al.* 2001). The use of an internal standard compensates for both random and systematic errors (Skoog, 1998). Te as an internal standard is preferred because of its low natural occurrence in biological material and good stability when analysed with inductively coupled plasma–MS (Eckhoff & Maage, 1997).

All samples were analysed in duplicate using a Perkin-Elmer SCIEX Elan 5000A inductively coupled plasma–mass spectrometer (Toronto, Canada). Optimisation and operating conditions of the instrument are described elsewhere (Julshamn *et al.* 2001).

Certified reference material is widely used to expose any systematic errors and assess the trueness of the analytical method in use, i.e. the degree of agreement between the certified value of an analyte and the mean value obtained from analysis (NMKL, 1996). Certified reference material was selected with regard to similarity in concentration and matrix to sample material analysed. For analyses of iodine in milk and dairy products, milk powder (BCR 150 spiked skimmed milk powder, Community Bureau of Reference, Brussels, Belgium) was chosen.

Measures of the trueness and repeatability (short-term precision) of the method were carried out by analysing ten parallels of the milk powder. The average obtained was 1.23 (SD 0.05) mg/kg compared with the certified value of 1.29 (SD 0.09) mg/kg. The repeatability calculated as relative standard deviation (RSD %) was 4%. The internal reproducibility of the milk powder given as RSD (%) was calculated as 11% and with an average iodine

concentration of 1.19 mg/kg ( $n = 30$ ). The results show clearly that the analytical method in use is under control with regard to systematic and random error.

### Statistics

Statistical analyses (one-way ANOVA and Student's  $t$  test) were carried out using Statistica™ Software, release 4.5 (1999; StatSoft, Inc., Tulsa, OK, USA). A confidence level of 95% was used throughout all statistical tests, i.e.  $P$  values below 0.05 were considered significant. Unless commented on, the data were tested for homogeneity of variance with Levene's test in a non-transformed way. In cases of organic milk, the data showed non-homogeneity. Mann-Whitney U tests for non-parametric samples were therefore used. In cases where ANOVA showed significant differences, *post hoc* tests were used to find which groups showed significant difference to each other. In cases where several groups were tested and the amount of samples was different, Tukey's honest significant difference test for different  $n$  (Spjøtvoll-Stoline test) was used. In the case of two groups, Student's  $t$  test for independent samples was used. Pearson's correlation coefficient was calculated using the correlation function.

## Results

### Seasonal differences in milk

To observe seasonal differences, low-fat milks were, with one exception, collected from the same dairies during the summer and the winter seasons of 2000. The iodine concentration of low-fat milk produced during the summer period was found to be significantly lower than the iodine concentrations in low-fat milk produced during the

winter period ( $P < 0.001$ ; Table 1). Low-fat milk from the winter season showed less variation in the iodine concentration (CV 14.7%) compared with low-fat milk sampled during the summer season (CV 18.3%).

### Geographical location

The iodine concentration of low-fat milk showed no significant differences between the five different regions (Fig. 1) within season (Table 1). Nevertheless, it became apparent that the iodine concentration in low-fat milk, to some degree, was dependent on the location of the dairy. Fig. 2 shows a significant correlation ( $r = 0.60$ ) between the iodine concentration in low-fat milk produced during the summer and the winter seasons ( $P < 0.01$ ). This shows that a dairy with high iodine concentration in low-fat milk sampled during the winter season can be expected to lie in the highest range of iodine concentrations during the summer season.

### Milk from organic farms

Organic milk was sampled from seven dairies during both seasons. The iodine concentration in organic milk from the winter period was significantly higher than organic milk from the summer period ( $P = 0.02$ ; Table 1). Two dairies had high iodine concentrations in the organic milk sampled during the winter period; 365 and 264  $\mu\text{g/l}$ , respectively. The range of iodine concentration in organic milk from the summer season was much smaller than organic milk from the winter season. The median iodine concentration of organic milk sampled during both seasons was lower than non-organic milk (low-fat milk), the summer-season differences being significant ( $P = 0.006$ ; Table 2).

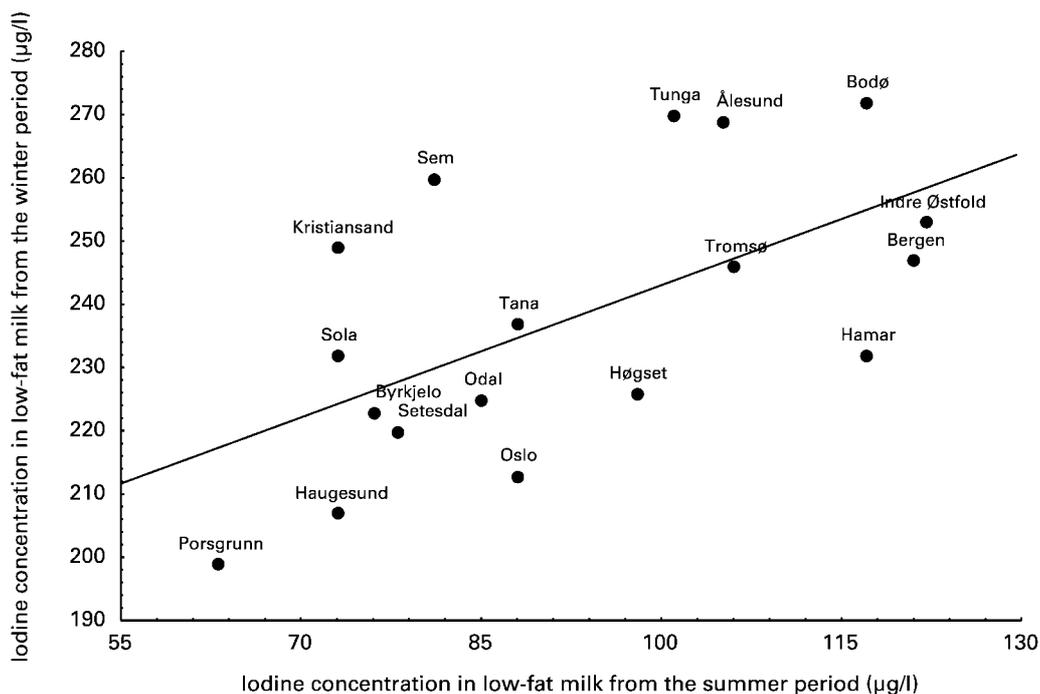


Fig. 2. Correlation between iodine concentrations in milk sampled during the summer and winter seasons of 2000 in Norway ( $r = 0.60$ ;  $P < 0.01$ ).

### *Iodine concentrations in dairy products*

The production of dairy products is centralised and thus limited to certain dairies. The geographical range is therefore less for processed dairy products than for milk samples. The median and the range of iodine concentrations for processed dairy products are given in Table 2. The results indicate that extra-low-fat milk and curdled milk products have similar concentration ranges of iodine as found in low-fat milk. Whey cheese (Tine Gudbrandsdalsost) showed the largest variation in iodine concentration of the dairy products. Jarlsberg cheese generally had a lower iodine concentration than Norvegia cheese, although the differences were not significant. The correlation between the iodine concentration in low-fat milk and the two dairy products (yoghurt and soured cream) produced at the same dairy was significant with correlation coefficients of 0.82 and 0.42, respectively ( $P < 0.01$ ).

### **Discussion**

The seasonal variation in the iodine content of milk was the main finding in the present study. The iodine concentration of low-fat milk found in the present study was higher than values reported in milk sampled from twelve places in Norway in 1971 (T Renaa and K Staveland, unpublished results); however, the seasonal variation showed the same pattern as has been described before (T Renaa and K Staveland, unpublished results). The iodine concentration in milk sampled in 1971 had the lowest iodine concentration in the summer season (65 µg/l) in the inland areas and the highest iodine concentration in milk during the winter season (120 µg/l; T Renaa and K Staveland, unpublished results). Thus, it seems that the iodine concentration of milk has increased in the last 30 years, especially during the winter months. However, the samples were not taken from the exact same dairies in the two studies and results are therefore not directly comparable.

The seasonal variations in the iodine content of milk are usually attributed to the proportion of pasture feeding *v.* the use of cow fodder fortified with iodine (Pennington, 1990). The higher iodine content of winter milk in the present study can be explained by the iodine fortification of cow feed and is in accordance with a number of other studies (Wenlock *et al.* 1982; Pennington, 1990; Lee *et al.* 1994; Ministry of Agriculture, Fisheries and Food, 1997, 1999; Phillips, 1997; Larsen *et al.* 1999). The iodine content of milk reported recently in the UK (Ministry of Agriculture, Fisheries and Food, 2000) was on average 430 µg/l in winter, while summer milk contained 200 µg/l. Two studies from Denmark showed both geographical and seasonal variations in the iodine content of milk (Larsen *et al.* 1999; Rasmussen *et al.* 2000). A study from the USA (Pennington, 1990) showed that the western parts of the continent had a higher iodine concentration in milk (340 µg/l) than milk from the eastern parts (160 µg/l). These differences were found in milk from the summer season (Pennington, 1990).

Binnerts (1979) has suggested that higher concentrations of iodine in the winter months might reflect decreased milk

production. Other studies have claimed that iodine may occur in milk as a result of the use of iodophors as cow teat sterilants and equipment sanitisers in dairies (Franke *et al.* 1983; Galton *et al.* 1986; Pennington, 1990). According to Hemken (1979) iodine test dips and sanitisers contribute an insignificant amount to increase the iodine content in milk, unless they are improperly used. The dairies in Norway do not use sanitisation or disinfectant containing iodophores.

The smaller variation of the iodine content in low-fat milk from the winter season compared with the summer milk indicates that cow fodder containing iodine is widely used by farmers in Norway. Even though cows are fed with different brands of fodder, the fortification of iodine (2 mg/kg) is stable and supplies the cows with a standardised amount of iodine.

Although both precipitation and iodine concentrations in the soil vary in Norway (Låg & Steinnes, 1976), these variations were not reflected in the iodine concentrations of milk within season in the present study. The relationship between the iodine concentration in low-fat milk sampled during the summer and winter seasons (Fig. 2) indicates that the farming practices vary within small ranges in the five different geographical locations. The larger variation of the iodine content in milk from the summer season compared with the winter milk is probably accounted for by a longer period of pasture feeding in the South and East regions of Norway combined with differences in the access to iodine-fortified cow fodder during the summer season. In Denmark the regional differences in milk within season were due to differences in the iodine content in the drinking-water resources (Larsen *et al.* 1999). The iodine concentration in drinking water in Norway is on average less than 2 µg/l (L Dahl, HM Meltzer, L Johansson and K Julshamn, unpublished results) and contributes in all probability very little to the iodine content of the milk.

The iodine level in organic milk was expected to be more or less dependent on the iodine concentration of pastureland. Results from the present study showed a wide range of iodine concentrations in organic milk sampled during the winter season (34–367 µg/l). The iodine concentrations in organic milk from both summer and winter seasons were in general lower than the iodine concentration in non-organic milk from the same season. The observed differences between organic milk and non-organic milk (low-fat milk) may display the effect of the variable use of feed to the cows. Farms producing organic milk are allowed to use mineral mixtures and give cows iodine-enriched fodder up to 20% feed energy, according to Norwegian national regulations in the field (Debio, 2003). Organic milk producers also frequently use fodder containing seaweed meal. However, the exact influence cattle fodder has on the iodine level in non-organic milk is difficult to estimate since type and amount of fodder used has not been monitored. According to a study from Denmark (Rasmussen *et al.* 2000), which shows similar seasonal differences between organic and non-organic milk, the differences can mainly be explained by the variation in feeding practices.

Iodine levels in dairy products are expected to be in the same concentration range as the raw material. Studies have shown minimal losses during different processing methods of milk, such as pasteurisation and skimming (Pedriali *et al.* 1997). The great variation in iodine content found in whey cheese (Tine Gudbrandsdalsost) reflects in all probability the use of milk from different seasons, as the storage of the cheese before sale was not registered. Whey cheese also contains added goats' milk. The influence goats' milk has on the iodine concentration in whey cheese is unknown since it was not analysed for iodine. It should be commented that during the production of whey cheese the whey is boiled in order to remove water (resulting in 560–580 g DM/kg). This will obviously increase the concentration of iodine in the remaining product and explains the higher iodine concentration in whey cheese. Iodine compounds are soluble in water; it is therefore probable that whey cheese contains more iodine than cheese made from casein (Jarlsberg and Norvegia).

Knowledge about the variation in iodine concentration of milk and dairy products is important as the consumption of milk and dairy products is very common in Norway (Johansson & Solvoll, 1999). According to a nationwide dietary survey among 2672 representative adult Norwegians in 1997 the mean intake of milk was 438 (SD 319) g/d per individual with a median value of 375 g/d (Johansson & Solvoll, 1999). Assuming a relatively stable intake of milk during a year, the iodine level in Norwegian milk alone will meet the daily recommendations of iodine set at 150 µg with 25 % during the summer and with more than 60 % during the winter season. In addition, dairy products will contribute substantially.

In conclusion, a seasonal variation in iodine content of milk was found in Norway. The highest content of iodine was found in samples from the winter season and reflects the use of cow fodder fortified with iodine. Milk and dairy products are important determinants of iodine intake in Norway.

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