Iodine concentration of organic and conventional milk: implications for iodine intake
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Running title: Organic and conventional milk-iodine content

Key words: Iodine, Organic, Milk, UK,
Abstract:
Iodine is required for adequate thyroid hormone production which is essential for brain development, particularly in the first trimester of pregnancy. Milk is the principal source of iodine in UK diets, and while small studies in Europe have shown organic milk to have a lower iodine concentration than conventional milk, no such study has been conducted in Britain. In view of the increasing popularity of organic milk in the UK, we aimed to compare the iodine concentration of retail organic and conventional milk and to evaluate regional influences on iodine levels. Ninety-two samples of organic and 80 samples of conventional milk, purchased from retail outlets in 16 areas of the UK (Southern England, Wales and Northern Ireland), were analysed for iodine concentration using inductively-coupled plasma mass spectrometry. The region of origin of the milk was determined from information on the label. Organic milk was 42.1% lower in iodine content than conventional milk (median iodine concentration 144.5 vs. 249.5 ng/g; \( P < 0.001 \)). There was no difference in iodine concentration of either conventional or organic milk by area of purchase. However a difference was seen in iodine concentration of organic milk by region of origin (\( P < 0.001 \)). The lower iodine concentration of organic milk has public-health implications, particularly in view of emerging evidence of iodine deficiency in UK population sub-groups, including pregnant women. Individuals who choose organic milk should be aware that their iodine intake may be compromised and should ensure adequate iodine intake from alternative sources.
Introduction

Iodine is needed for the production of thyroid hormones which are vitally important during pregnancy and infancy owing to their role in brain and neurological development\(^1\). While severe iodine deficiency can cause cretinism and infant mortality\(^1\), there is emerging evidence that mild-to-moderate maternal iodine deficiency is associated with impaired infant development, including lower IQ and increased incidence of Attention Deficit Hyperactivity Disorder\(^2\). For these reasons, pregnant women are vulnerable to iodine deficiency\(^1\).

The iodine content of foods is dependent on the source of the food, whether from sea or land, the soil content (affected by geology, geography, pH, soil leaching) and farming practice\(^1\). In the UK, milk and dairy products are the most important sources of dietary iodine, contributing as much as 42% of adult intake\(^3\). Higher milk-iodine concentration and increased milk consumption have been cited as the reasons for the eradication of endemic goitre in the UK in the 1960s, which was labelled as an “accidental public health triumph” by Phillips\(^4\). Iodine sufficiency has been assumed in the UK for many years\(^5, 6\) to such an extent that iodised salt is scarcely available\(^7\). However, there is emerging concern that the iodine status of the population, particularly of women of childbearing age\(^8, 9\) and pregnant women\(^7\), may be inadequate.

Although conventional milk is the usual choice, organic milk is increasing in popularity because of perceived health and environmental benefits\(^10\), though a systematic review concluded that there are no nutritional differences between organic and conventionally produced foodstuffs\(^11\). Due to the strict organic farming regulations that govern the use of mineral supplements in livestock feed\(^12, 13\), organic milk may contain lower concentrations of trace minerals, thereby reducing or even reversing the potential health benefits of organic milk. Studies in Europe have shown conventional milk to have a higher concentration of iodine than organic milk\(^14, 15\), but no such study has been explicitly conducted in Britain.

This study therefore aimed to compare the iodine content of organic and conventional milk available for purchase in the UK. A secondary aim was to determine regional variations in the iodine content of milk. Our hypotheses were that: (i) the iodine content of organic milk would be lower than that of conventional milk owing to organic farming practices and, (ii) iodine concentration of milk would differ between regions due to likely variations in soil-iodine content and farming practice across the UK.
Materials and methods

Sampling

Samples of supermarket semi-skimmed (less than 2% fat) own-brand conventional and own-brand organic milk were purchased from five leading supermarkets [total market share 79.4%\(^{(16)}\)] in sixteen areas (largely identifiable as counties) of the UK [fourteen of which were in the South of England, one in Cardiff (Wales) (Figure 1) and one in County Antrim (Northern Ireland)] in June, July and August 2009. The areas were combined into four regions for analysis: South East \(n=8\), South West \(n=6\), Wales \(n=1\) (Figure 1) and Northern Ireland \(n=1\). In addition, three samples of popular milk brands were purchased, the majority of which were organic. Fewer samples of branded milk were purchased reflecting their lower UK market share\(^{(17)}\). As milk-iodine content is known to vary between seasons\(^{(6, 18, 19)}\), sampling was restricted to one season so that milk purchased at the start and end of the collection period would be comparable. Semi-skimmed milk was selected as it is the most popular choice in the UK\(^{(20)}\) and its iodine concentration is not thought to differ from that of skimmed and full-fat milk\(^{(19)}\). Milk was deemed to be organic based on the label claim and if it had an organic certification symbol, such as that of The Soil Association.

The areas sampled were chosen mainly for logistical reasons but also because the inclusion of areas in the South East allowed the assessment of milk from densely populated regions, while the South West is both a major dairy farming region\(^{(17, 21)}\) and a region with historically high goitre rates\(^{(4)}\). Northern Ireland and Wales were sampled to investigate potential regional variations, although sampling in Northern Ireland was restricted, as only two of the five chosen supermarket chains operate there.

The milk origin was categorised as being from Wales, the West Country, Scotland, Northern Ireland, or of unknown origin. The origin was determined from the label; where the geographical source of the milk was stated (e.g. Milk from the West Country), this was assumed to be the case but otherwise, the EU Identification Mark was used to trace the milk to the processing dairy\(^{(22)}\) and in conjunction with internet sources, the origin was determined where possible.

Sample analysis
Sample preparation and analysis was performed at LGC Ltd., Teddington, UK. Sample aliquots (20 ml) were stored at -80°C, before being transported to LGC Ltd. An aliquot of 0.5 g was mixed with 5 ml of 5% tetramethylammonium hydroxide (TMAH) solution, prepared by dissolving solid TMAH (≥97% Sigma Aldrich, UK) in ultrapure water. The vial was then placed in an oven at 90°C for 3 hours. To each sample 0.5 ml of the internal standard was added (1300 ng/ml tellurium, Romil, UK, in 1% TMAH) and samples were made up to 50 g with 1% TMAH solution. The digested samples were analysed for iodine concentration by inductively-coupled plasma mass spectrometry (ICP-MS) (Element2, ThermoFisher Scientific, Bremen, Germany) by external calibration using a stock standard, prepared in-house gravimetrically from high purity potassium iodide (≥99.99%, Alfa Aesar, UK) in 5% TMAH solution. Subsequent dilutions were performed in 1% TMAH. The uncertainty of the method was calculated as ±10% according to in-house United Kingdom Accreditation Service (UKAS) accredited methods which are in accordance with ISO 17025 and Eurachem/CITAC guidelines. Accuracy of the results was verified using the certified reference material (CRM) BCR 063R Skimmed Milk Powder (LGC Standards, UK); [certified iodine content 810 ± 50 ng/g (dry weight basis)]; the mean value for the CRM was 838.9 ng/g (± 23.6), a percentage recovery of 103.6%. The Coefficient of Variation for the 15 measurements of the CRM was 2.8%. The means of the spiked recoveries for the CRM and the milk samples were 101.5 % (±6.3) and 94.5 % (±5.0) respectively.

Statistical analysis
Statistical analysis was with the Statistical Package for Social Sciences (SPSS Version 17.0; Chicago, IL). Iodine concentration was not normally distributed in either organic or conventional milk samples, as determined by the Shapiro-Wilk test; data were transformed using the natural logarithm to allow parametric testing. Geometric means with their 95% confidence intervals (CI) (computed by back transformation of log values) are reported, along with the median. Independent t-tests were used to test the difference between organic and conventional milk-iodine concentration and one-way ANOVA (with Bonferroni correction for post-hoc analysis) was used for comparison of iodine concentration between area of purchase and region of origin of the milk. Significance was set at \( P < 0.05 \).

Results

Differences in iodine concentration
Ninety-two samples of organic and 80 samples of conventional milk were collected. Table 1 shows the iodine concentrations of all organic and conventional milks, of supermarket own-brand milk and other branded milk and by known region of origin.

An independent t-test showed that the iodine concentration of organic milk was significantly lower than that of conventional milk ($P<0.001$), the median value of organic milk being 42.1% lower than that of conventional milk. Branded organic milk samples ($n = 15$) were significantly lower in iodine concentration than the supermarket own-brand conventional milk samples ($n = 77$) ($P<0.001$; Table 1).

**Analysis by area of purchase**
One-way ANOVA showed that there was no difference in iodine concentration between the 16 areas of purchase of either supermarket own-brand organic or conventional milk samples ($P = 0.75$ and $P = 0.49$ respectively) or between the four regions ($P = 0.36$ and $P = 0.66$ respectively), i.e. the three regions shown in Figure 1 and Northern Ireland.

**Analysis by region of origin of the milk**
Organic milk from the West Country, Wales and of unknown origin was significantly lower in iodine than conventional milk of the same origin ($P<0.001$) (Table 1). Due to the small number of samples originating from Wales and Northern Ireland, these regions were excluded from the one-way ANOVA for analysis by milk origin. Though no sampling was carried out in Scotland, eight of the organic milk samples were of Scottish origin. The iodine concentration of organic samples differed by region of origin ($P < 0.001$; Table 1); post-hoc testing revealed that Scottish organic milk was significantly higher in iodine than both organic milk from the West-Country ($P < 0.001$) and organic milk of unknown origin ($P < 0.001$). Conventional milk from the West Country exhibited a lower iodine concentration than milk of unknown origin, but the difference did not reach significance ($P = 0.051$). To prevent Scottish milk samples from skewing the results, statistical testing was repeated with Scottish milk excluded; the difference in iodine concentration between organic and conventional milk remained significant ($P<0.001$) but the difference between the median values increased to 43.1% (data not shown).

**Discussion**
This is the first sizeable study to evaluate differences in iodine concentration between organic and conventional milk. The main finding of our study, that organic milk has a significantly
lower iodine concentration than conventional milk, supports both our original hypothesis and the findings of other, smaller studies (fewer than ten organic samples), in Denmark\textsuperscript{(14)} and Norway\textsuperscript{(15)}. The iodine concentrations of the Norwegian and Danish summer organic milk samples were 31.8\% and 40.8\% lower than those of conventional milk respectively, the latter being close to the 42.1\% found in this study.

Milk iodine concentration in the UK has increased since the 1920s\textsuperscript{(4)} through the use of iodine-supplemented feeds in dairy herds to protect livestock against deficiency\textsuperscript{(18)} and through the use of iodophor disinfectants used in sanitisation and teat dipping\textsuperscript{(24)}, which can contaminate the milk with iodine. The UK has never introduced an iodization programme to ensure optimal iodine status, despite goitre being historically endemic up until the 1960s\textsuperscript{(25)}. Instead, the country has experienced “iodization by default”\textsuperscript{(4)}, through increased milk-iodine concentration and increased milk consumption. Our finding that the iodine content of summer organic milk is over 40\% lower in than that of conventional milk is a concern from a public-health perspective.

The UK adult reference nutrient intake is 140 µg/d\textsuperscript{(26)} with no increment for pregnancy, a recommendation that is clearly outdated in the light of current WHO advice for pregnancy of 250 µg/day\textsuperscript{(27)}. Given that milk and dairy products are the primary source of iodine in the UK, those who switch to organic milk, with its lower iodine concentration, are likely to have a reduced iodine intake. This is of particular concern during pregnancy when a woman needs additional iodine and consumption of organic milk may reduce the chance of her meeting the higher iodine requirement of pregnancy. Based on our median milk-iodine values, a portion (200 g) of conventional milk would provide approximately 50 µg compared to only 29 µg in a portion of organic milk; iodine intakes would be further compromised if other organic dairy products were consumed.

Although only a small proportion of liquid milk sold in the UK is organic\textsuperscript{(28)}, sales of organic milk increased more than 50-fold over the ten-year period between 1997 and 2007\textsuperscript{(29)}, and remained strong during the recent economic recession\textsuperscript{(10, 28)}. The UK organic dairy sector is planning increased advertising in order to encourage consumers to switch to organic\textsuperscript{(28)}. With increasing use of organic milk, there is potential for exacerbation of the mild iodine deficiency described in the UK, notably in pregnant women\textsuperscript{(30-32)} and women of childbearing age\textsuperscript{(8, 9)}. 
The lower iodine concentration in organic milk can be explained by differing practices on organic and conventional farms. Organic farming regulations do not allow the routine use of vitamin and mineral preparations\(^\text{(12, 13)}\). Regulations also stipulate that at least 60% of feed on organic farms must be fresh or conserved forage\(^\text{(13)}\), thus limiting the use of concentrates and relying on soil minerals, which can be low in some areas. Due to these restrictions, deficiencies in some minerals, including iodine, can occur in organically farmed livestock\(^\text{(33)}\). Nitrogen-fixing crops, such as clover, are important in organic farming and are used in place of artificial fertilisers\(^\text{(21)}\). White clover contains cyanogenic glucosides that are thought to exhibit goitrogenic properties\(^\text{(34)}\) and, as suggested by Rasmussen \textit{et al.}\(^\text{(14)}\), greater use of goitrogenic feed could lower milk-iodine concentration through inhibition of the sodium-iodide symporter in the mammary gland of the cow. As iodophor disinfectants and teat dips are permitted in both organic\(^\text{(13)}\) and conventional farming\(^\text{(35)}\), it is unlikely that the difference in milk-iodine concentration can be explained by their use.

Our hypothesis concerning regional differences in milk iodine concentration was only partially supported. Although regional differences were found when milk was broadly grouped by known milk origin, no difference was found by area of purchase. The latter finding may be explained by the supply-chain logistics in the milk industry, where milk is bulked in the processing dairy and delivered to stores within the same supermarket chain in neighbouring areas\(^\text{(21)}\). The inclusion of Scottish milk was unintentional and was revealed through interpretation of the EU identification mark\(^\text{(22)}\). The finding that Scottish milk is higher in iodine than that from the West Country is interesting in that historically, goitre and cretinism, were common in the West Country but their incidence was lower in Scotland\(^\text{(25)}\). As this study did not collect details on specific farming differences between regions (e.g. soil and feed-type), we are unable to provide answers for the observed differences in iodine between the regions.

Our study has a number of limitations: samples were only collected in the summer months, so findings may not be representative of the levels in winter milk. However, as both conventional and organic cattle are less reliant on mineral-supplemented feed during the summer, any difference observed between the groups in the summer is likely to be matched, or exceeded during the winter. Milk was largely purchased from the South of England, and whilst this could be considered a limitation, the results indicate that the region of origin was a greater influence on iodine concentration than area of purchase. However, sample sizes for regional analysis were small and unequal and regional differences should therefore be interpreted cautiously.
Furthermore, our study used retail milk which is pooled from farms thus substantially masking regional differences.

In conclusion, the fact that organic milk has a lower concentration of iodine than conventional milk is a public-health concern. When individuals make the decision to switch to organic produce they often start with milk\(^{(28)}\), believing this to be the best choice. Individuals who make such a choice should be aware that their iodine intake may be compromised and that, during pregnancy and infancy, this may have implications for infant brain development. The authors are not suggesting that all individuals should switch to conventional milk, as there may be other benefits to organic produce in terms of lower levels of pesticide residues\(^{(36)}\). Rather, individuals consuming organic milk (particularly pregnant women) should be aware of the need for alternative food sources of iodine in order to meet requirements and where these alternatives are not consumed, a nutritional supplement containing iodine should be considered (kelp supplements are not recommended due high iodine concentrations in some preparations\(^{(37)}\)).

It would be prudent for the organic dairy industry to take seriously the deficit in iodine content of organic milk that our study has revealed. The restrictions imposed by organic farming methods have nutritional implications both for organically raised animals\(^{(33)}\) and for populations eating their produce. Though these restrictions may affect other trace mineral concentrations, the issue is likely to be most serious for iodine owing to the fact that milk and dairy products are the principal source of iodine in the UK diet.

**Acknowledgments**

We thank Sarah Hill, Malcolm Burn and other analysts at LGC Ltd, and Peter Williams in the Department of Mathematics at the University of Surrey for statistical advice. Studentship funding for S. Bath by The Waterloo Foundation and Wassen International supported this work. The funding bodies did not have influence over any part of the study. SCB and MPR designed the study, SCB and SB conducted milk sample collection, SCB performed statistical analysis. MPR had primary responsibility for final content. All authors prepared, reviewed and approved the final manuscript. None of the authors had a personal or financial conflict of interest.
References


Table 1 Iodine concentration of organic and conventional milk samples and by known milk origin.

<table>
<thead>
<tr>
<th>Milk Type</th>
<th>Number of samples</th>
<th>Median</th>
<th>Geometric Mean</th>
<th>95% CI of Geometric Mean</th>
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</thead>
<tbody>
<tr>
<td>All samples</td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>Organic</td>
<td>92</td>
<td>144.5</td>
<td>152.2*</td>
<td>141.7, 163.5</td>
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<tr>
<td>Conventional</td>
<td>80</td>
<td>249.5</td>
<td>256.4</td>
<td>245.0, 268.3</td>
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<td>Supermarket own-brand</td>
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<td></td>
<td></td>
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<tr>
<td>Organic</td>
<td>77</td>
<td>148.0</td>
<td>159.8*</td>
<td>148.1, 172.5</td>
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<tr>
<td>Conventional</td>
<td>77</td>
<td>250.0</td>
<td>258.4</td>
<td>246.9, 270.6</td>
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<td>Branded milk</td>
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<tr>
<td>Organic</td>
<td>15</td>
<td>131.0</td>
<td>118.3†</td>
<td>100.7, 139.0</td>
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<td>Conventional</td>
<td>3</td>
<td>196.0</td>
<td>208.5</td>
<td>135.0, 322.0</td>
</tr>
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<td></td>
<td></td>
<td></td>
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<tr>
<td>Organic</td>
<td>24</td>
<td>140.5</td>
<td>130.3*</td>
<td>117.4, 144.6</td>
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<tr>
<td>Conventional</td>
<td>18</td>
<td>236.0</td>
<td>239.2</td>
<td>215.5, 265.4</td>
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<td>Wales</td>
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<td></td>
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<tr>
<td>Organic</td>
<td>3</td>
<td>83.0</td>
<td>84.0*</td>
<td>48.9, 144.2</td>
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<td>Conventional</td>
<td>4</td>
<td>212.5</td>
<td>217.6</td>
<td>184.9, 256.1</td>
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<tr>
<td>Organic</td>
<td>8</td>
<td>287.5</td>
<td>276.5‡</td>
<td>238.4, 320.7</td>
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<tr>
<td>Conventional</td>
<td>0</td>
<td>NA</td>
<td>NA</td>
<td>NA</td>
</tr>
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<td>Northern Ireland</td>
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<tr>
<td>Organic</td>
<td>2</td>
<td>220.5</td>
<td>220.4</td>
<td>160.5, 302.7</td>
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<tr>
<td>Conventional</td>
<td>2</td>
<td>222.0</td>
<td>221.9</td>
<td>148.6, 331.3</td>
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<td>Unknown</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Organic</td>
<td>55</td>
<td>145.0</td>
<td>152.2*</td>
<td>140.8, 164.4</td>
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<tr>
<td>Conventional</td>
<td>56</td>
<td>275.5</td>
<td>266.6</td>
<td>252.7, 281.3</td>
</tr>
</tbody>
</table>

*Iodine concentration significantly lower than conventional milk of same category ($P<0.001$);
†Iodine concentration significantly lower than conventional supermarket own-brand milk ($P<0.001$);
‡Iodine concentration significantly higher than organic milk from the West Country ($P<0.001$) and organic milk of unknown origin ($P<0.001$)
Figure 1. Map showing the areas (shaded) from which milk was sampled and how the areas were combined into regions. Milk was purchased from supermarkets in towns shown by black dots. Milk was also purchased in County Antrim, Northern Ireland (not shown).