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Further studies on the iodine concentration of conventional, organic and UHT semi-skimmed milk at retail in the UK

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ABSTRACT

Milk is the largest source of iodine in UK diets and earlier studies showed organic summer and winter milk to be significantly lower in iodine than conventional milk. One study also showed UHT milk to have lower iodine concentration. The study on winter and UHT milk was small and accordingly a new study is reported here involving conventional, organic and UHT semi-skimmed milk from four supermarkets over a six-month period in summer and winter in two regions of the UK. The results showed organic milk to be 44% lower in iodine than conventional milk (427 vs. 241 μg/L, $P < 0.001$) and UHT milk was 27% lower in iodine than conventional milk (427 vs. 314 μg/L, $P < 0.001$) although the differences tended to be less in the summer. The results indicate that replacement of conventional milk by organic or UHT milk will increase the risk of sub-optimal iodine status especially for pregnant/lactating women.

1. Introduction

Iodine is an essential micronutrient in the human diet, required for thyroid hormone synthesis which controls key aspects of metabolism, brain and bone development (Grau et al., 2015). The recommended daily intake of iodine varies; the World Health Organisation (WHO, 2007) advises 150 μg/d for adults and 250 μg/d for pregnant and lactating women, whilst the UK Reference Nutrient Intake (RNI) for iodine is 130 μg/d for children aged 11–14 and 140 μg/d for adults, with no increase during pregnancy or lactation (Department of Health, 1991). The availability of iodised salt has helped to lower the number of countries with iodine deficient populations, however globally, 30% of school children are still believed to consume sub-optimal amounts of the nutrient (Andersson, Karumbunathan, & Zimmermann, 2012). Iodised salt has never been compulsory in the UK (Phillips, 1997) and recent public health advice to reduce salt intake suggests this situation will not change.

A review of data from the recent UK National Diet and Nutrition Survey showed that many sections of the population are still not consuming adequate amounts of iodine (Miller, Spiro, & Stanner, 2016). In particular, 22% of girls aged 11–18 years old and 10% of women aged 19–64 have iodine intakes below the lower reference nutrient intake (70 μg iodine/d) (Bates et al., 2014; Miller et al., 2016). Cohort studies in the UK have shown 51% of schoolgirls to be mildly iodine deficient based on urinary iodine concentrations (Vanderpump et al., 2011) and a large cohort of pregnant women were classified as being mild-to-moderately iodine deficient (Bath, Walter, Taylor, Wright, & Rayman, 2014). In addition, the UK Avon Longitudinal Study of Parents and Children (ALSPAC) showed an association between mild-moderately iodine deficient pregnant women and lower verbal IQ, reading accuracy and comprehension in their children compared to those born from mothers of adequate iodine status (Bath, Rayman, Steer, Golding, & Emmett, 2013). A similar outcome was seen in an Australian study where children from mothers with urinary iodine concentrations during pregnancy <150 μg/L, had significantly lower ability in spelling, grammar and English-literacy at 9 years old than children whose mothers' urinary iodine concentrations were >150 μg/L (Hynes, Otahal, Hay, & Burgess, 2013).

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In the UK, semi-skimmed milk is by far the primary source of dietary iodine for children and adults alike with other dairy products also making important contributions (Bates et al., 2014). Whilst milk is a good source of iodine, the type of milk, time of year, dairy cow diet and the human diet can all affect the amount available for thyroid hormone synthesis. Organic milk has significantly less iodine than conventional milk (Bath, Button, & Rayman, 2012; Payling, Juniper, Drake, Rymer, & Givens, 2015) whilst milk produced in summer months has consistently lower iodine concentrations than winter milk (Food Standards Agency, 2008; Crnkić, Haldimann, Hodžić, & Tahirović, 2015). Furthermore, the presence of thiocyanate in the diet, found in plants belonging to the *Brassica* genus and in tobacco, can prevent the uptake of iodine into the thyroid gland due to competitive inhibition of the sodium-iodine symporter (NIS) (Bivolarska, Gatseva, Nikolova, Argirova, & Atanasova, 2015; Trøan et al., 2015).

The study by Payling et al. (2015) on iodine concentration in organic and conventional winter milk at retail was subject to scrutiny. There was concern that by the time of publication, the results were out of date as it was reported that recently permitted iodine additions to the diet of many organic dairy herds was now leading to organic milk with comparable iodine levels to conventional milk (OMSCO, 2015, 2016). The current study therefore aimed to further the research of Payling et al. (2015) by incorporating into one study the effect on iodine content of conventional, organic and UHT milk of month purchased, supermarket of purchase and geographical area.

2. Materials and methods

2.1. Milk samples

On approximately the same date every month from July to December 2015 milk samples were collected from four leading supermarkets in Reading (South East England) and Stratford-upon-Avon (Midlands, England). From each supermarket a conventionally produced pasteurised, organic produced pasteurised and conventionally produced ultra-high temperature (UHT) processed milk sample (all semi-skimmed) was bought, yielding 24 samples each month and a total of 144 samples collected for analysis. After every collection, a 40 ml aliquot of each milk sample was taken and stored at −20 °C in the same freezer until required.

2.2. Sample analysis

Milk samples were left to thaw overnight, before thorough mixing in a vortex machine prior to analysis. Iodine was measured in all samples by alkali extraction followed by inductively coupled plasma mass spectrometry (ICP-MS) based on the method of Fecher, Goldmann and Nagengast (1998). In brief, 100 μl of each milk sample was diluted to 10 ml using a diluent of 0.22 M tetramethylammonium hydroxide (TMAOH) in ultra-pure water and containing 5 μg/L Rh (as $RhCl_3{\cdot}3H_2O$) as an internal standard. The samples were then filtered using Whatman 0.45 μm PVDF w/pp filters to remove any fat which could block the nebuliser in the ICP-MS instrument. All samples were analysed in the same way, using the same ICP-MS instrument (iCAP Q, Thermoscientific Inc. Waltham, MA, USA). Certified ammonium iodide calibration standards were made up to 0, 2, 4, 6, 8, and 10 μg/L iodine in the same standard 5 μg/L Rh 0.22 M TMAOH diluent.

The accuracy of results obtained was verified using a certified reference material of skim milk powder (LGC Standards, Teddington, TW11 0LY, UK, 2015; certified iodine content 1.73 mg/kg [±0.14 mg/kg]). In triplicate 0.87 g of skim milk powder was dissolved in 10 g of distilled water to yield a normal homogenous solution of cow's milk with an anticipated iodine concentration of 150 μg/L. Dilutions with 5 μg/L Rh in 0.22 M TMAOH were then made to give 10 sample solutions with anticipated iodine concentrations in the range 0–5 μg/L. Iodine concentration was then measured in the same way as for the milk samples. Anticipated and observed values for iodine concentration of the certified reference material showed no significant differences (P = 0.857), indicating a good level of accuracy.

2.3. Statistical analysis

The effect of milk product type (conventional production, organic production, UHT, supermarket of origin (1–4), area of the country (Midlands, South East) and month purchased (July-December) were determined by fixed effects analysis of variance using a general linear model (Mintab Version 17; Minitab Inc., State College, PA, USA). Tukey's pairwise multiple comparison test was then used to identify which treatments were significantly different from each other when the significance was P < 0.05.

3. Results

The iodine concentrations of the milk types by supermarket and overall are shown in Table 1. Within each supermarket, milk iodine concentration was significantly influenced by milk type (P < 0.001) and in three supermarkets by month purchased but there was no effect of area of the country and few interactions. Overall, milk iodine concentration was significantly influenced by milk type, supermarket and month of purchase (all P < 0.001) but not by area. The effect of milk type was pronounced, with the iodine concentration of organic and UHT milk being 43.5 and 26.6% lower than conventional milk respectively. There was a notable month × milk type interaction (P = 0.028) showing that for all milk types iodine concentration progressively increased from summer to winter but with an overriding impact of milk type (Fig. 1). A supermarket × milk type interaction ($P < 0.001$) was also seen, mainly resulting from a progressive rise in conventional and organic milk iodine concentrations in the order of supermarkets 3, 4, 1 and 2 which was not the case for UHT milk (Fig. 2).

4. Discussion

Previous studies (Bath et al., 2012; Payling et al., 2015; Rey-Crespo, Miranda, & López-Alonso, 2013) and meta-analysis (Średnicka-Tober et al., 2016) have reported that organic milk has a lower iodine concentration than conventionally produced milk. At the time of publication, the data of Payling et al. (2015) were the most up to date available for UK retail milk produced in the winter. It used milk purchased in January 2014, was a relatively small study and there was some concern that by the time of publication the results were out of date as a result of recently permitted iodine additions to the diet of many organic dairy herds (OMSCO, 2015). The results of the present study involving both summer and winter produced milk confirm the same outcome, with retail organic milk having a 43.5% lower iodine concentration than conventionally produced milk.

The overall mean iodine concentration in organic milk in the present study (241.4 μg/L) is markedly lower than in the study of Payling et al. (2015) (343 μg/L) although this is likely since the current study involves milk produced in winter and summer whereas that of Payling et al. (2015) was restricted to winter milk. It has been recognised for some time that summer milk contains less iodine than winter milk (Crnkić et al., 2015; Food Standards Agency, 2008) and this effect was seen in the present study with the progressive increase in iodine content from summer to winter

Table 1
Least square mean (±SE) iodine concentrations of retail milk as influenced by type (T), supermarket (S), area of purchase (A) and month of purchase (M).

Supermarket	Mean iodine concentration (μg/L)				*P*-value for*						
	Conventional n = 48	Organic n = 48	UHT n = 48	SED	T	S	M	SxT	AxM	AxT	MxT
1	494 ± 31[a]	272 ± 32[b]	337 ± 20[b]	28.4	<0.001	–	0.024	–	NS	NS	NS
2	522 ± 19[a]	269 ± 26[b]	280 ± 11[b]	17.3	<0.001	–	0.003	–	NS	NS	NS
3	339 ± 15[a]	198 ± 15[b]	343 ± 16[a]	20.7	<0.001	–	NS	–	NS	NS	NS
4	353 ± 16[a]	227 ± 30[c]	294 ± 18[b]	17.5	<0.001	–	<0.001	–	NS	0.024	NS
Overall	427 ± 16[a]	241 ± 14[c]	314 ± 9[b]	11.9	<0.001	<0.001	<0.001	<0.001	0.036	0.026	0.028

[a,b,c]Means within a row with a common superscript are not significantly different (P > 0.05); NS, not significant (P > 0.05); SED, standard error of the difference.
* Excluding effects of area, area × supermarket and month × supermarket, all NS.

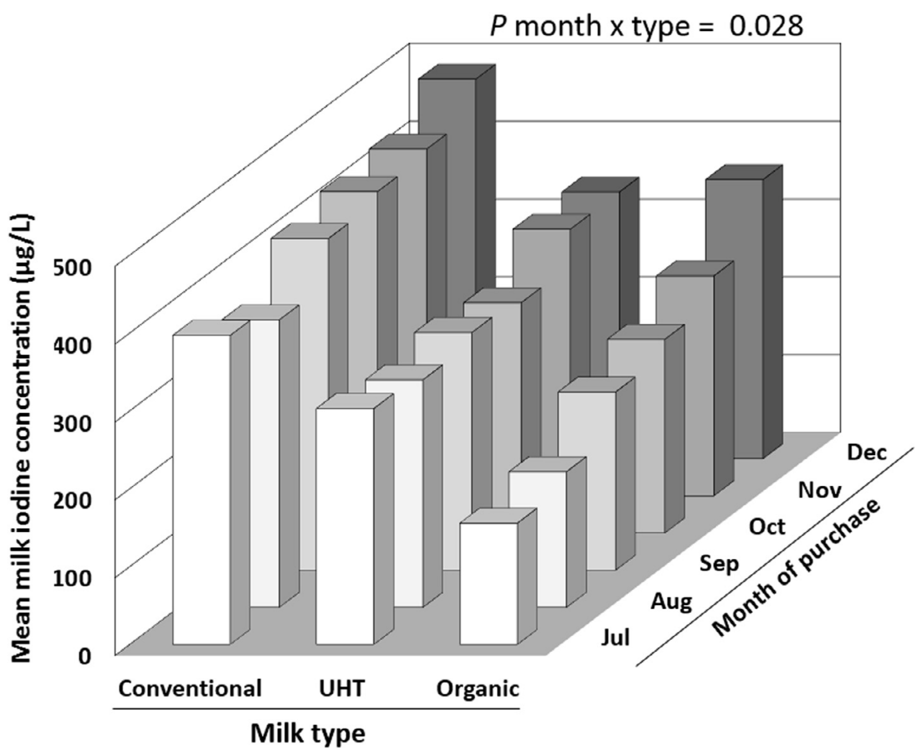

Fig. 1. Interaction effect of milk type and month of purchase on the iodine content of conventional, organic and UHT milk.

moderated by milk type shown in Fig. 1. The effect of season of milk production is mainly a result of the dairy cows receiving less iodine containing supplementary feed in the summer (Flachowsky, Franke, Meyer, Leiterer, & Schöne, 2014). Indeed it is likely that this is one reason why organic milk has lower iodine content than conventional. In organic dairy systems in the EU, the diet of the cows must generally contain a minimum of 600 g/kg forage dry matter (European Union, 2008) and with greater reliance on forage legumes such as clover and limited options for adding iodine to the diet. Whilst there is general agreement that a key factor influencing milk iodine concentration is iodine intake by the dairy cow (Flachowsky et al., 2014), it can be substantially reduced by in-feed iodine antagonists such as glucosinolates in rapeseed meal (Trøan et al., 2015) and potentially, cyanogenic glycosides in clover. This effect is mediated by these compounds being converted to thiocyanates which competitively inhibit the Na+/I- symporter (De La Vieja, Dohan, Levy, & Carrasco, 2000) thus reducing iodine uptake by both the mammary and thyroid glands. Rapeseed products and clover can be used in diets for organic milk production and may contribute to the lower iodine content of organic milk and although the impact of glucosinolates and rapeseed cake/meal have been modelled (Trøan et al., 2015) there is little experimental evidence for effects of cyanogenic glycosides or clover. The influence of in-feed iodine antagonists on milk iodine concentration is clearly in need of detailed research not least because if thiocyanates are also transferred to milk they may reduce iodine uptake by the human thyroid. It is of note that Bivolarska et al. (2015) reported that pregnant women exposed to thiocyanates from tobacco smoke and who had a urinary thiocyanate/creatinine ratio greater than a median value of 3.57 mg/g, had substantially greater risk of being of sub-optimal iodine status (urinary iodine <150 μg/L) than the pregnant women with lower urinary thiocyanate/creatinine ratios (odds ratio = 3.882, 95%CI 1.402–10.751, P = 0.009).

Payling et al. (2015) also reported that UHT processed milk had a significantly lower iodine concentration than conventional pasteurised but not significantly different to organic milk. The present study also showed that UHT milk had a significantly lower iodine concentration than conventional (Table 1) but was significantly higher than organic milk. The reasons for the lower iodine concentrations in UHT than conventional pasteurised milk remain largely unknown although the review of Flachowsky et al. (2014) suggests that higher temperature processing may lead to a loss of iodine. Whilst consumption of UHT milk in the UK is low, in many other parts of the EU it represents up 90% of milk consumed (Arrizabalaga, Jalón, Espada, Cañas, & Latorre, 2015). Interestingly,

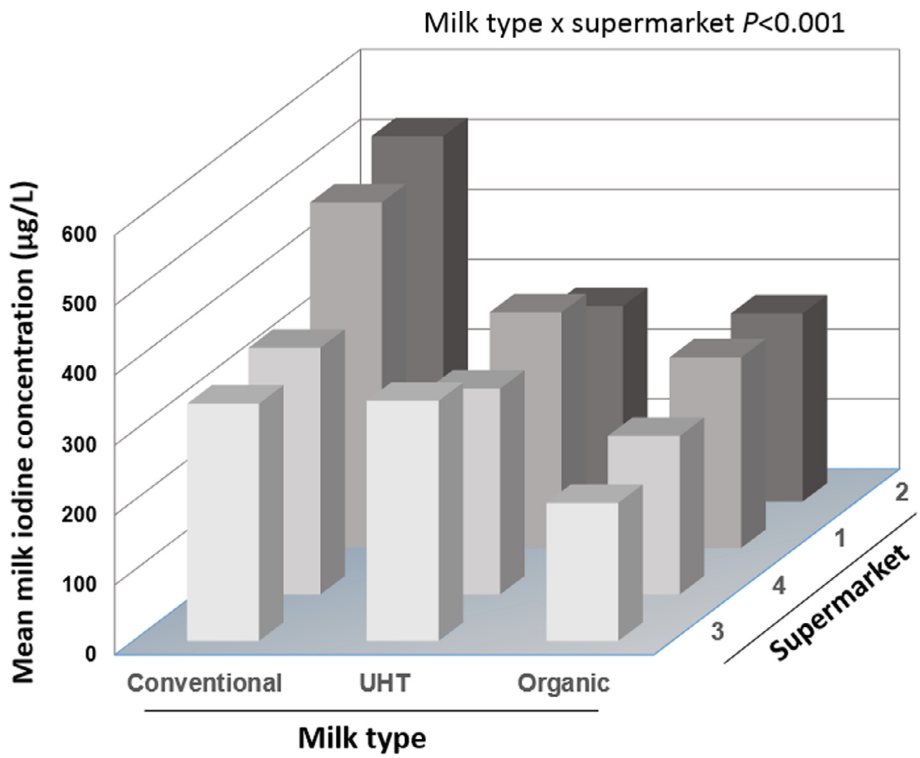

Fig. 2. Interaction effect of milk type and supermarket on the iodine content of conventional, organic and UHT milk.

a study in 2008 of 489 samples of conventional UHT retail milk from the Basque Region of Spain reported a mean (SD) iodide concentration of 197.6 (58.1) µg/L, equivalent, according to the authors, of 227.1 µg/L of total iodine (Arrizabalaga et al., 2015), lower than in the current study. There was no conventional pasteurised milk for comparison but the mean (SD) iodide concentration in a small set of 12 organic UHT milks was 56.4 (8.6) µg/L (equivalent to 64.8 µg/L iodine) significantly ($P < 0.001$) lower than conventional UHT milk. The effect of UHT processing clearly needs further investigation.

Because of the nature of a retail study the reasons for the effects of supermarket and the supermarket × milk type interaction cannot be identified. It is however important to note that conventional milk from two supermarkets (1 and 2) were significantly ($P < 0.05$) higher in iodine than milk from supermarkets 3 and 4. It may be that supermarkets 1 and 2 have greater influence on the primary production of the milk including the dietary ingredients used and iodine supplementation for the dairy cows involved. Despite the effect of supermarket on conventional milk iodine content there was no effect of area of purchase both within supermarkets and overall. This agrees with the study of Bath et al. (2012) who suggested that this was probably because retail milk is a relatively homogenous product as result of mixing collections from wide geographical locations and after processing, distribution to supermarkets over a wide area. Whilst this is certainly the case, the present findings suggest that despite this, some supermarkets are able to maintain a higher conventional milk iodine content through the production chain than others.

The results from this study confirm earlier findings that pregnant women in particular should be aware of the lower concentrations of iodine in UHT and especially organically produced milk. This knowledge will allow pregnant women who wish to consume organic milk to obtain the recommended daily intake of iodine by consuming more of it. Based on the current results the UK RNI for iodine of 140 µg/d could be achieved by consumption of 328 ml and 580 ml of conventional and organic milk respectively, although if the WHO (2007) recommended iodine intake during pregnancy of 250 µg/d was aimed for, daily milk consumption would need to rise to 585 ml and 1035 ml respectively. It is questionable whether a milk intake of about 1 L/d of organic milk is likely to be consistently achieved but it highlights the need for careful design of the whole diet, including supplements and lifestyle, including possible thiocyanate load from diet and smoking.

The present study has weaknesses. Only two areas in England were used although in agreement with the retail study of Bath et al. (2012) it found no significant effect of area of milk purchase. This may not translate to the whole UK however. In addition milk purchases were restricted to one six-month period although this did cover a summer and winter production period and is to our knowledge the only recent UK retail study to examine the progression from summer to winter produced conventional, organic and UHT milk. There were supermarket × milk production system and supermarket × month of purchase interactions which were related to inconsistencies between supermarkets although a feature of retail studies is the inability to fully understand the reasons for them. Retail studies do however have the benefit of examining foods actually purchased by the population and despite some limitations, this study adds to the understanding of the effects of diet on iodine status.

5. Conclusions

The results from this study reinforce the findings of earlier studies (Bath et al., 2012; Payling et al., 2015) showing significantly higher concentrations of iodine in conventional milk compared to organic and UHT milk. The results from the present study showed that retail organic and UHT milk to be 44% and 27% lower in iodine than conventional milk respectively although these values are greater in milk produced in the summer than that produced in winter. This relates to the finding that summer produced milk has a lower iodine content than winter milk. Overall the findings reinforce the message of earlier studies that simple replacement of conventional milk by organic milk will increase the risk of sub-optimal iodine status, especially in periods of increased iodine demand such as pregnancy and lactation. The finding of lower iodine concentrations in UHT milk than in conventional milk is also in agreement with the findings of Payling et al. (2015) but the current study confirms that this effect is consistent across summer and winter produced milk. This is of considerable public health nutrition importance in countries where UHT is the primary form of liquid milk. The reasons for this effect are not fully understood and need to be investigated.

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References

Andersson, M., Karumbunathan, V., & Zimmermann, M. B. (2012). Global iodine status in 2011 and trends over the past decade. *Journal of Nutrition, 142*, 744–750.

Arrizabalaga, J. J., Jalón, M., Espada, M., Cañas, M., & Latorre, P. M. (2015). Iodine concentration in ultra-high temperature pasteurized cow's milk. Applications in clinical practice and in community nutrition. *Medicina Clinica (Barcelona), 145*, 55–61.

Bates, B., Lennox, A., Prentice, A., Bates, C., Page, P., Nicholson, S., & Swan, G. (2014). National Diet and Nutrition Survey, Results from Years 1–4 (combined) of the Rolling Programme (2008/2009-2011/12). A survey carried out on behalf of Public Health England and the Food Standards Agency.

Bath, S. C., Button, S., & Rayman, M. (2012). Iodine concentration of organic and conventional milk: Implications for iodine intake. *British Journal of Nutrition, 107*, 935–940.

Bath, S. C., Rayman, M. P., Steer, C. D., Golding, J., & Emmett, P. (2013). Effect of inadequate iodine status in UK pregnant women on cognitive outcomes in their children: Results from the Avon Longitudinal Study of Parents and Children (ALSPAC). *The Lancet, 382*(9889), 331–337.

Bath, S. C., Walter, A., Taylor, A., Wright, J., & Rayman, M. P. (2014). Iodine deficiency in pregnant women living in the South East of the UK: The influence of diet and nutritional supplements on iodine status. *British Journal of Nutrition, 111*, 1622–1631.

Bivolarska, A., Gatseva, P., Nikolova, J., Argirova, M., & Atanasova, V. (2015). Effect of thiocyanate on iodine status of pregnant women. *Biological Trace Element Research, 172*, 101–107.

Crnkić, Ć., Haldimann, M., Hodžić, A., & Tahirović, H. (2015). Seasonal and regional variations of the iodine content in milk from federation of Bosnia and Herzegovina. *Mljekarstvo, 65*, 32–38.

De La Vieja, A., Dohan, O., Levy, O., & Carrasco, N. (2000). Molecular analysis of the sodium/iodide symporter: Impact on thyroid and extrathyroid pathophysiology. *Physiological Reviews, 80*, 1083–1105.

Department of Health (1991) Dietary Reference Values for Food, Energy and Nutrients in the United Kingdom No. 41. London: HMSO.

European Union (2008). Commission Regulation (EC) no. 889/2008 of 5 September 2008 laying down detailed rules for the implementation of Council Regulation (EC) no. 834/2007 on organic production and labelling of organic products with regard to organic production, labelling and control. Official Journal of the European Union, 51, L250.

Fecher, P. A., Goldmann, I., & Nagengast, A. (1998). Determination of iodine in food samples by inductively coupled plasma mass spectrometry after alkaline extraction. *Journal of Analytical Atomic Spectrometry, 13*, 977–982.

Flachowsky, G., Franke, K., Meyer, U., Leiterer, M., & Schöne, F. (2014). Influencing factors on iodine content of cow milk. *European Journal of Nutrition, 53*, 351–365.

Food Standards Agency (2008). *Retail survey of iodine in UK produced dairy Foods, FSIS 02/08*. London: Food Standards Agency.

Grau, G., Aguayo, A., Vela, A., Aniel-Quiroga, A., Espada, M., Miranda, G., ... Rica, I. (2015). Normal intellectual development in children born from women with hypothyroxinaemia during their pregnancy. *Journal of Trace Elements in Medicine and Biology, 31*, 18–24.

Hynes, K. L., Otahal, P., Hay, I., & Burgess, J. R. (2013). Mild iodine deficiency during pregnancy is associated with reduced educational outcomes in the offspring: 9-year follow-up of the Gestational Iodine Cohort. *The Journal of Clinical Endocrinology and Metabolism, 98*, 1954–1962.

Miller, R., Spiro, A., & Stanner, S. (2016). Micronutrient status and intake in the UK – where might we be in 10 years' time? *Nutrition Bulletin, 41*, 14–41.

OMSCO (2015). OMSCo statement in response to Reading University study into iodine levels in organic milk. Somerset: OMSCo Admin. URL: http://www.omsco.co.uk/_clientfiles/pdfs/OMSCo-statement-280415.pdf Accessed 26.11.15.

OMSCo (2016). Organic dairy makes advancements in Iodine levels. Somerset: OMSCo Admin. URL: http://www.omsco.co.uk/_clientfiles/pdfs/omsco-iodine-levels.pdf Accessed 10.02.16.

Payling, L. M., Juniper, D. T., Drake, C., Rymer, C., & Givens, D. I. (2015). Effect of milk type and processing on iodine concentration of organic and conventional winter milk at retail: Implications for nutrition. *Food Chemistry, 178*, 327–330.

Phillips, D. I. W. (1997). Iodine, milk, and the elimination of endemic goitre in Britain: The story of an accidental public health triumph. *Journal of Epidemiology and Community Health., 31*, 391–393.

Rey-Crespo, F., Miranda, M., & López-Alonso, M. (2013). Essential trace and toxic element concentrations in organic and conventional milk in NW Spain. *Food and Chemical Toxicology, 55*, 513–518.

Średnicka-Tober, D., Barański, M., Seal, C., Sanderson, R., Benbrook, C., Steinshamn, H., ... Leifert, C. (2016). Composition differences between organic and conventional meat: A systematic literature review and meta-analysis. *British Journal of Nutrition, 115*, 994–1011.

Trøan, G., Dahl, L., Meltzer, H. M., Abel, M. H., Indahl, U. G., Haug, A., & Prestløkken, E. (2015). A model to secure a stable iodine concentration in milk. *Food & Nutrition Research, 59*, 29829.

Vanderpump, M. P. J., Lazarus, J. H., Smyth, P. P., Laurberg, P., Holder, R. L., Boelaert, K., & Franklyn, J. A. (2011). Iodine status of UK schoolgirls: A cross-sectional survey. *The Lancet, 377*(9782), 2007–2012.

WHO (2007). *World Health Organization, United Nations Children's Fund & International Council for the Control of Iodine Deficiency Disorders. Assessment of iodine deficiency disorders and monitoring their elimination. A guide for programme managers* (3rd ed.). Geneva: World Health Organization. Available online: http://whqlibdoc.who.int/publications/2007/9789241595827_eng.pdf.