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Brief Report

Iodine Concentrations in Conventional and Organic Milk in the Northeastern U.S.

Nobumitsu Sakai, Ola Yetunde Esho and Motoko Mukai *

Department of Food Science, College of Agriculture and Life Sciences, Cornell University, Ithaca, NY 14853, USA; ns579@cornell.edu (N.S.); oye2@cornell.edu (O.Y.E.)
* Correspondence: mm2295@cornell.edu

Abstract: Milk is a major source of dietary iodine in the United States. Due to a relatively low margin of safety for iodine intake, there are concerns for both deficiency and over-exposure. Iodine concentrations of raw milk samples from farms and retail milk in the Northeastern U.S. region were compared between seasons (winter vs. summer) and farming practices (conventional vs. organic). Overall, mean iodine concentration was 46.2% higher in raw milk from conventional farms vs. organic farms. An interaction effect between season and farming practices was observed. Organic raw milk had higher iodine content in the winter than in the summer (423 ± 54 µg/L vs. 273 ± 24 µg/L), whereas conventional raw milk had higher iodine content in the summer than in the winter (618 ± 75 µg/L vs. 398 ± 27 µg/L). Milk samples from conventional farms had 2.27-fold higher average iodine concentration compared to milk coming from organic farms in the summer but did not differ in the winter. Out of 68 and 98 raw milk samples originating from conventional and organic farms, 22 (32.4%) and 19 (19.4%) respectively, had iodine concentrations > 500 µg/L, reaching as high as 1928 µg/L. In contrast, the overall mean concentration of iodine in retail milk did not differ between conventional and organic milk (345 ± 23 vs. 320 ± 42 µg/L, respectively). The current study confirms dairy milk remains to be a good source of iodine to U.S. consumers. However, dairy farms should be aware of the potential adverse health effects of excess iodine intake. Careful considerations in dairy management may be necessary to not exceed the recommended level of iodine supplementation in both conventional and organic operations at the farm level—to maintain optimal iodine concentrations in retail fluid milk accessible to the consumers.

Keywords: iodine; marketed milk; organic; season

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1. Introduction

Iodine is an essential trace element required for the synthesis of thyroid hormones, thyroxine (T4) and triiodothyronine (T3), and play critical roles in a variety of organs. Iodine is variably distributed in the earth's environment and certain regions have historically been deficient in iodine. The success of salt iodization programs in the early 1920s resulted in the elimination of the regional epidemic known as 'goiter belt' in the U.S., and many other countries have also benefited from similar programs [1]. However, the National Health and Nutrition Examination Surveys (NHANES) reported that 56.9% of pregnant women in the U.S. may have inadequate iodine intake based on urinary iodine concentrations [2], indicating problem of deficiency still remains in certain populations.

Iodine is also known to have a relatively low margin of safety and excessive iodine intakes from iodized salt have also resulted in adverse health outcomes with increased incidences of hypothyroidism, hyperthyroidism, and autoimmune thyroiditis especially in susceptible individuals with underlying thyroid disease [3–5]. Food and Nutrition Board of National Academies' Institute of Medicine (IOM) has established an average requirements ranging from 65 µg/day in 1–3 years old toddlers (demographic with lowest need) to 209 µg/day in lactating women (demographic with highest need), with tolerable upper intake level ranges from 200 μg/day to 1100 μg/day, respectively [6]. Therefore, it is crucial to monitor iodine concentrations in food products to ensure an adequate iodine intake preventing both deficiency and excess.

Milk and milk products are currently one of the major sources of dietary iodine in the U.S. [7] and many other countries, accounting for, on average, 40–70% of total iodine consumed from one's diet [8–10]. The iodine content in cow milk is influenced by (1) diet—the concentration of iodine in cow feed, including the presence of goitrogens, and drinking water; (2) farm management—such as the use of iodophor sanitizers and processing, and (3) the environment—including the season of milk production and the iodine concentration in the soil where they graze [11]. In recent years, a rise in milk iodine has been observed in some countries to above the maximum recommended level of 500 μg/L. A study of Canadian raw milk indicated iodine concentrations as high as 1902 μg/L [12]. These concentrations may potentially lead to dietary intake of iodine exceeding the tolerable upper intake level (UL), especially in small children, and can induce thyroid dysfunction in milk consumers.

Although inadequate iodine intake is a health concern, iodine is not routinely tested in the dairy industry and its concentrations are not labeled on dairy products. There have been only a few reports of iodine concentrations in milk in the U.S. in recent years [7,10,13]. In European countries, there have been reports of organic milk having a lower concentration of iodine than conventional milk [11,14,15], but this has not been investigated in the U.S. Therefore, in this study, we hypothesized that conventional milk in the Northeastern U.S. have elevated iodine levels similarly to Canadian milk, considering similar farming practices, but organic milk would have lower iodine levels compared to conventional milk based on previous reports from Europe. We tested the iodine level in raw milk and retail milk samples from the Northeastern U.S. market and examined differences in milk iodine concentrations between conventional vs. organic milk across two seasons—winter and summer.

2. Materials and Methods

2.1. Collection of Raw Milk Samples

Raw milk samples were collected into 50 mL sampling vials at two Northeast dairy plants at the time of delivery from bulk milk of individual farms. Samples consisted of a total of 49 individual conventional farms and 60 organic farms across two seasons, in the winter (February 2018) and in the summer (July–August 2018). Upon arrival to the lab, raw milk samples were stored in 4 °C fridge and were analyzed within 3 days of collection. In a previous pilot study, we have shown soluble iodide concentrations remain unchanged when raw milk samples were stored in 4 °C for up to 14 days [16].

2.2. Iodine Assay

An ion-selective electrode (ISE) method was used for measuring iodine content in milk samples following the official AOAC method 992.24 (AOAC 2006) with slight modification as reported previously [16]. The electrical potential in each sample was measured by an ion-selective electrode (Orion 9653BNWP, Thermo Fisher), and the soluble iodide concentration was calculated using a linear standard curve ($R^2 > 0.99$), prepared by dilution of the certified iodide standard (41271, Sigma-Aldrich, St. Louis, MO) stock solution. The precision of the results was verified by inter-assay variability test over six days (%RSD = 1.72), and intra-assay variability test of 12 samples (%RSD = 1.19). Extraction efficiency was evaluated by spiking a known amount of certified iodide standard (500 μg/L; mid-range of the generated standard curve) to the reference milk before the procedure with each assay. Data from an assay with percent recovery < 75% (n = 16; 10 conventional and 6 organic samples) and farm samples that were not collected in both seasons (n = 10; 5 conventional and 5 organic samples) were excluded in the final analysis. This resulted in exclusion of total 15 conventional farms and 11 organic farms, with a final data set originating from 34 conventional and 49 organic farms across two different seasons.

2.3. Retail Milk Analysis

Due to concern of elevated iodine in conventional milk in the summer, retail whole milk (n = 21/farm type; total 42 samples) were collected from a total of 15 supermarkets and retail stores in the Northeast in late July to early August in 2018 to evaluate the iodine concentrations in the marketed milk available directly to the consumers. Samples were mixed well and aliquoted into 15 mL tubes and immediately stored at −20 °C. Free-sulfhydryl groups are generated when milk is heated above normal pasteurization temperature and this is known to interfere with ISE activity (Lacroix and Wong, 1980). Therefore, ICP-MS was used instead of ISE for measuring iodine in retail milk. Samples were shipped on dry ice to Eurofins at Food Integrity Innovation-Madison, and total iodine as measured by ICP-MS as previously described [17,18]. Intraassay variability (%RSD) was <11.4% and extraction efficiency was >92.4% (with a matrix spike of a known value). All analysis was performed in a single-day run.

2.4. Statistical Analysis

Statistical analysis (linear mixed effect model with repeated measure analysis for raw milk, Student's *t*-test for retail milk) was performed using JMP 15 (SAS, Cary, NC, USA). Data were tested for the assumption of normality with quantile-quantile plots and tested for the assumption of variance using Levene's test. When these assumptions were not met, $\log_{10}$ transformed data were used. After a significant effect in the main model was detected, the significance of each group comparisons for raw milk were determined with Tukey's post hoc tests at a *p*-value of 0.05. All data are shown in mean ± SEM μg/L.

3. Results and Discussion

The overall average raw milk iodine concentration in the winter was 412.62 ± 33.30 μg/L and was not significantly different from raw milk collected in the summer (414.38 ± 38.55 μg/L; Figure 1A and Table 1). Overall average raw milk iodine concentration from organic farms was significantly lower at 347.74 ± 30.16 μg/L than milk from conventional farms at 508.28 ± 41.85 μg/L (31.6% less, $p < 0.001$; Figure 1B and Table 1). There was a significant interaction between season and farming practices ($p < 0.0001$). Although there were no differences between organic and conventional farms in the winter, organic milk (272.76 ± 24.05 μg/L) had significantly lower (55.90%) mean iodine level in the summer compared to conventional milk (618.48 ± 75.27 μg/L; Figure 1C and Table 1). The overall mean of the organic milk was below what is generally regarded as a safe level (<500 μg/L) in the dairy industry. However, substantial numbers of raw milk from both individual conventional and organic farms had iodine concentrations > 500 μg/L. A total of 21 out of the 83 winter samples tested (25.3%) had iodine concentrations above 500 μg/L (originating from 8 conventional and 13 organic farms ranging as high as 1768 μg/L). A total of 20 out of the 83 summer samples tested (24.1%) had iodine concentrations above 500 μg/L (originating from 14 conventional and 6 organic farms ranging as high as 1929 μg/L).

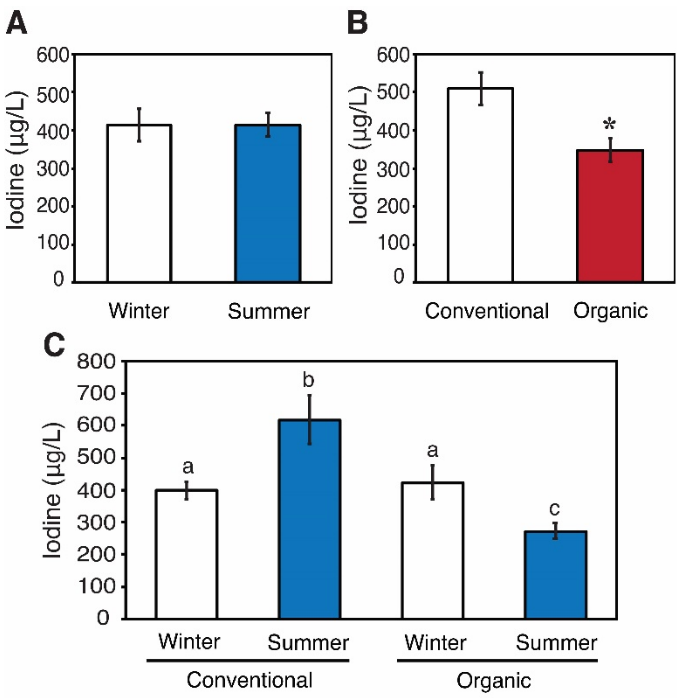

Figure 1. Iodine concentrations in raw milk originating from conventional and organic farms across different seasons using an ion-selective electrode method. (**A**) Average iodine concentrations in the winter vs. summer; (**B**) Average iodine concentrations in conventional vs. organic; (**C**) Average iodine concentrations in each of the four groups (across types and season). Data are shown as mean ± SEM. Asterisks (*) represents statistical significance on the main effect of farming practice ($p < 0.0001$) with a linear mixed-effect model with repeated measures analysis. Interactive effect of farming practice x season was also observed ($p < 0.0001$). Different letters (a–c) stand for statistical differences ($p < 0.05$) between each group shown by post hoc Tukey's test.

Table 1. Iodine concentrations (µg/L) detected in both raw milk and retail milk samples from conventional and organic farming across different seasons (winter and summer).

	Season and Farming Practice	n	Mean ± SEM (µg/L)	Median	Min	Max	n > 500 µg/L	% >500 µg/L
Raw Milk	Winter (all)	83	412.62 ± 33.30	318.45	113.02	1768.05	21	25.3
	Summer (all)	83	414.38 ± 38.55	312.93	71.37	1928.91	20	24.1
	Conventional (all)	68	508.28 ± 41.85	423.81	182.10	1928.91	22	32.4
	Organic (all)	98	347.74 ± 30.16 *	252.75	71.37	1768.10	19	19.4
	Conventional (Winter)	34	398.08 ± 26.67 [a]	369.01	176.58	780.93	8	23.5
	Conventional (Summer)	34	618.48 ± 75.27 [b]	468.31	182.10	1928.91	14	41.2
	Organic (Winter)	49	422.71 ± 53.51 [a]	299.77	113.02	1768.05	13	26.5
	Organic (Summer)	49	272.76 ± 24.05 [c]	225.54	71.37	741.79	6	12.2
Retail Milk	Conventional (Summer)	21	344.52 ± 22.55	360.00	120.00	554.00	1	4.8
	Organic (Summer)	21	319.70 ± 42.01	252.00	78.60	827.00	4	19.0

* Significant main effect was observed at $p < 0.0001$ with farming practice but not with season. Interactive effect between season and farming practice was observed at $p < 0.0001$. [a–c]: Different alphabet denotes significance at $p < 0.05$ following Tukey's post hoc test.

Elevated iodine level has been reported previously in cow milk in Canada [12], which was attributed to being due to excess feed supplementation [19] and teat-dipping practices [20]. There is a dearth of information on milk iodine concentration in the U.S. in recent years. The mean iodine concentrations we found in our study is similar to those reported previously with milk purchased locally in Boston (n = 18) [7] as well those reported by the FDA Total diet study (n = 20) [10], but much higher than mean concentration of 89.2 µg/L (n = 40) reported by another [13]. Iodine in ~25% raw milk samples we tested had a concentration above 500 µg/L. The current study suggests an elevated raw milk iodine level in the Northeastern U.S. similar to elevated concentrations reported in Canada [12] at the farm level.

Iodine level in raw milk was higher in winter than in summer in organic milk, and the effect was reversed in conventional milk. Iodine increases in the winter have been reported in the U.S. [7,21] as well as in European countries [22–24], which is commonly attributed to the increase in mineral supplementation and decreased feeding of goitrogenic plants in the winter compared to the summer [11]. This is also likely to be the case as reasons for the seasonal differences in organic milk in this study. The reasons for the higher iodine in summer than in winter in conventional milk is unknown and no previous studies have shown an increase of milk iodine in the summer compared to winter. However, it is possible that differences in management practices between summer and winter within conventional farms are causing this increase and may warrant further investigation to unveil the reasons.

This is the first study that compared iodine concentrations in conventional vs. organic milk in North America. We found a lower mean iodine concentration in organic milk compared to conventional milk. This result is consistent with several previous reports from Europe where organic milk was observed to have a lower concentration of iodine when compared to conventional milk [15,22,25,26], although iodine concentrations of organic milk reported in these studies were much lower (<150 μg/L) compared to the current study (272.74 and 422.71 μg/L, summer and winter, respectively). In the current study, we did not investigate feeding and management practices of individual conventional or organic farms, thus the reason for the differences cannot be clearly identified. However, the relative higher concentrations of iodine in organic milk, in both seasons, potentially could be due to different feeding practices used in organic farms in the U.S. (or particularly in the Northeast) compared to European countries.

In the U.S., feed supplements and additives are allowed in organic farming to meet the adequate nutritional requirements although excessive amounts are not allowed. However, it is possible that some farms in the Northeast, regardless of farming practices, are over-supplementing feed with mineral mixes containing iodine. This was the case in Canada [19], where more than 85% of the farms tested were providing far excess iodine above the NRC's maximum dietary recommendation of 0.5 mg/kg of diet DM for lactating cows [27]. On the other hand, in Europe, although feed supplementations are allowed in organic farming, the average iodine level in organic dairy milk suggests that they may not be supplementing at all, if any, in some countries.

Another possible reason for the increase of iodine, especially in organic milk in the Northeast, is the use of supplementation of natural sources that are high in iodine. For example, according to the Organic Dairy Handbook published by the Northeast Organic Farming Association of New York [28], kelp meals are provided as free choice in many organic dairy farms as supplements of vitamins and minerals. It has been reported that 49~83% of organic dairy producers feed kelp meal in Midwestern and Northeastern U.S. [29–31]. According to a survey [29], organic dairy farmers use kelp meals due to various health benefits claims (some of which are not scientifically proven) such as improving body condition; reducing SCC, reproductive problems, the incidence of pinkeye; and controlling a fly infestation. It has been reported that the dietary supplementation with kelp meal at 57–170 g/day significantly increased milk iodine concentrations (up to 1370 μg/L) with differential effect between seasons but does not result in animal performance [29,32]. Excess consumption of kelp meal or when provided in combination with iodine supplementation through the mineral mix could result in increased iodine concentrations due to naturally high concentrations of iodine in kelp/seaweed.

Increased feeding of products containing glucosinolates, such as rapeseeds, crambe meal, is suspected as one of the potential causes of lower iodine content in European organic milk compared to conventional milk [11,24,33]. Hydrolysis of glucosinolates generates a variety of products, some of which possess goitrogenic effects by inhibiting sodium-iodine-symporter and limiting the availability of iodine to the thyroid and most likely in the mammary gland as well [34]. Organic dairy farms in the Northeast may use limited amounts of glucosinolate-containing products compared to European counterparts. The Organic Dairy Handbook [28] mentions that only limited farms use glucosinolate-containing plants in the Northeast U.S. as feed. Cyanogenic glycoside-containing plants, such as *Trifolium* spp., *Sorghum* spp., and *Lotus* spp., are often found in grazing pasture. White clover (*Trifolium repens* L.) is commonly used not only as nitrogen fixing cover crop but particularily important in organic agriculture due to its use in green-manure, as replacement for synthetic fertilizer [35]. However cyanogenic glycoside-containing plants can release hydrogen cyanide when damaged (including grazing and sudden frost), be metabolized into thiocyanates, and pose goitrogenic effects. Exposure to cyanogenic glycoside-containing plants may be higher in the summer, reducing the milk iodine concentration during summer for organic farms.

Another factor known to affect the milk iodine level is the use of iodine-containing disinfectants [16,20,36]. The use of iodine as disinfectants, sanitizer, medical treatment as well as a topical treatment (such as teat-dips) and external parasiticides, are allowed in organic dairy farming in the U.S. according to the final rule of the National Organic Program (NOP) under 7 CFR 205.6039(a)(14) and (b)(3). Use of iodophore is also allowed in organic farming in Europe under Commission Regulation (EC) No 889/2008 (EU, 2008) but it may be used in moderation in organic dairy farms in European countries, also leading to differences found between this current study in Northeast milk and previous reports from Europe [15,22,25,26].

Despite concerns for the overall elevated level of raw milk iodine from conventional milk in the summer, the average total iodine of conventional retail milk was <500 μg/L and similar to organic retail milk (344.52 ± 22.55 vs. 319.70 ± 42.01 μg/L, respectively; Figure 2 and Table 1). There were 5 samples out of 42 retail samples (11.9%; 4 organic and 1 conventional), that had iodine concentration above 500 μg/L and variability was greater with organic milk. This could be due to a difference in how bulk-tank raw milk is transported from each farm to the processing plant, combined, and be packaged together as one branded retail milk. Milk from conventional farms may be mixed more often, minimizing the effect of any farm-to-farm variability on iodine concentrations in the final retail product. Organic milk, on the other hand, may not be mixed between different farms as much to maintain its authenticity (such as 'Grass-fed') and branding and leading to the larger variability of iodine concentrations reflective of management practices (feed supplementation and sanitation, etc.) in the final retail product. In 1984, average iodine concentration in retail milk in NYS was reported at 394.1 μg/L (with a minimum of 129.2 μg/L and maximum of 1362.2 μg/L) [37], which indicates that farming practices have not largely changed in Northeast to affect the overall iodine content in milk.

Milk processing techniques are also considered as potential factors in affecting iodine concentration in retail milk. High-temperature short-time (HTST) pasteurization has been shown to decrease milk iodine in a few studies by as much as 52.7% [38,39], although such an effect was not observed in other studies [40–42]. Ultra-high temperature sterilization does not seem to affect milk iodine concentrations [38,41]. Out of 42 retail milk collected, 25 were HTST pasteurized (6 organic, 19 conventional), 16 were UHT milk (15 organic, 1 conventional), and one (conventional) was vat-pasteurized. There were no significant differences overall between HTST-pasteurized and UHT milk (370.04 ± 28.66 vs. 286.1 ± 38.39 μg/L, respectively). Although we do not have ways to evaluate the effect of HTST pasteurization on milk iodine in our studies, it is possible HTST used in most conventional milk contributed to the decrease of iodine concentrations, minimizing the difference in iodine concentrations between two farming practices in the final retail milk. Since retail milk sampling was limited, interpretation of the results comparing organic and conventional milk should be taken with caution. Nevertheless, routine iodine testing is recommended either at the farm or plant level to maintain ideal iodine concentrations of retail milk for consumer health benefits, both for organic and conventional farms.

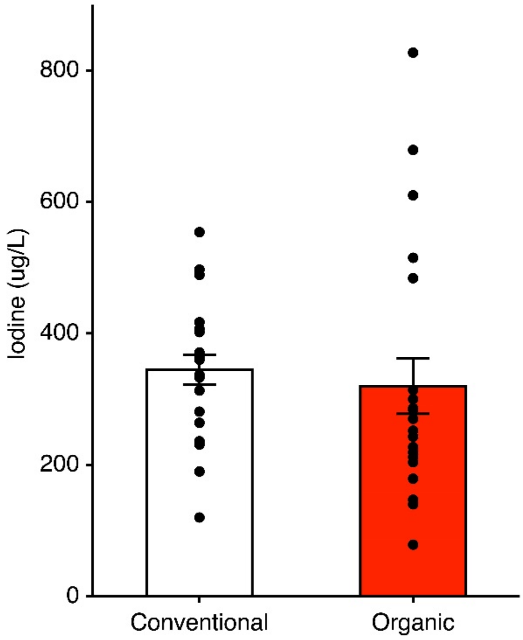

Figure 2. Total iodine concentrations in whole milk (conventional and organic) collected from Northeastern U.S. supermarkets and retail stores using ICP-MS. Bars are shown as mean ± SEM with a scatter plot of individual samples.

4. Conclusions

In summary, this is the first study to investigate iodine concentrations in conventional vs. organic milk in the U.S. and North America. This study offers new information that the situation in the Northeastern U.S. is different from European countries. Although organic milk has overall lower iodine concentrations compared to conventional milk at the farm level, there is an indication of over-supplementation of iodine in both conventional and organic farms, with 25% of samples reported concentrations above 500 µg/L. It would be important to study the difference in organic farming practices to understand the cause of this differential effect on iodine concentrations. Based on milk iodine concentrations found in retail milk, both organic and conventional milk originating from the Northeast remains to be a good source of iodine although dairy farms should be aware of potential adverse health effects of excess iodine. Since the margin of safety of iodine is relatively narrow, this study highlights that it would be important to consider dairy management to ensure that iodine concentrations are in the appropriate range for human consumption.

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Conflicts of Interest: The authors declare no conflict of interest.

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