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Original Research Article

Iodine in foods and dietary supplements: A collaborative database developed by NIH, FDA and USDA[☆]

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ABSTRACT

Data on the iodine content of foods and dietary supplements are needed to develop general population intake estimates and identify major contributors to intake. Samples of seafood, dairy products, eggs, baked products, salts, tap water, other foods and beverages, and dietary supplements were collected according to established sampling plans of the U.S. Department of Agriculture (USDA) and the U.S. Food and Drug Administration (FDA). Samples were assayed for iodine content using inductively coupled plasma mass spectrometry with rigorous quality control measures. The food data were released through a collaboration of USDA, FDA, and the Office of Dietary Supplements-National Institutes of Health (ODS-NIH) as the USDA, FDA, and ODS-NIH Database for the Iodine Content of Common Foods at www.ars.usda.gov/mafcl. Iodine data for dietary supplements are available in the ODS-USDA Dietary Supplement Ingredient Database and the ODS Dietary Supplement Label Database. Data from the iodine databases linked to national dietary survey data can provide needed information to monitor iodine status and develop dietary guidance for the general U.S. population and vulnerable subgroups. This iodine information is critical for dietary guidance development, especially for those at risk for iodine deficiency (i.e., women of reproductive age and young children).

1. Introduction

Iodine functions as an essential component of thyroid hormones, regulating biochemical reactions, especially protein synthesis and enzymatic activity, and playing a critical role in fetal growth and in physical and neurological development of infants and children (Rohner et al., 2014; Zimmermann, 2009). The Recommended Dietary Allowance for iodine per day is 90 µg for children ages 1–8 years, 120 µg for children ages 9–13, 150 µg for males and most females ages 14 and older, 220 µg for pregnant women and 290 µg for lactating women (Institute of Medicine (IOM)., 2001). The Tolerable Upper Level per day for people over age 18 not receiving iodine for medical reasons is 1100 µg (Institute of Medicine (IOM)., 2001). The foods generally considered the highest dietary contributors of iodine are fish, seaweeds, dairy foods, and eggs (Lee et al., 2016). Seawater - and in some locations, groundwater - is a predominant source of iodine in the environment, exerting a strong influence on the eventual concentration of iodine in foods via a variety of factors (Fuge and Johnson, 2015). Worldwide, dietary deficiencies are associated with inland continental regions and especially rain shadow areas (blocked precipitation creating desert-like conditions) in mountainous areas and areas of frequent flooding, while coastal areas are generally iodine sufficient. The interaction of geological

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factors and dietary choices confound these generalizations (Fuge and Johnson, 2015; Panth et al., 2019). Iodine status in the U.S. has continued to be generally adequate; however, studies have shown that urinary iodine levels dropped by about half between the early 1970 and the early 1990s. Recently, iodine deficiency has slightly re-emerged in pregnant women which is of concern for developing fetuses (Thyroid.org, 2021; Caldwell et al., 2013.)

Iodine can be an additive in some foods, e.g., iodized salt, breads (in which iodate is used as dough conditioner), and confections and other foods that contain FD&C Red No.3 as a colorant (Lee et al., 1994; Prestwich and Gerrard, 2005; Wenlock et al., 1982). The use of iodine-supplemented feed contributes to the iodine content of meat, milk, and eggs, and iodine-containing sanitizing agents used during milk processing influence iodine in dairy products (Flachowsky, 2007; Flachowsky et al., 2014; Sarlak et al., 2020). Vegetables grown in soils enriched with iodized fertilizers also contain higher amounts of iodine (Fuge and Johnson, 2015). Iodine is also present in some dietary supplements, such as many adult and children's multivitamin/mineral products (MVM), and some prenatal MVM (Patel et al., 2019).

Salt iodization in the US was adopted on a voluntary basis in 1924 to reduce incidence of goiter, an enlargement of the thyroid gland. Globally, over 124 countries have salt iodization programs, but only about 30 international food composition databases include iodine (Ershow et al., 2018). These factors become important in evaluating dietary intake and population iodine status, especially because various diet-related shifts impact overall iodine intakes, and because iodine content can vary widely among different products within a particular food or food group. Dietary trends in recent years include decreased use of salt at the table and in cooking in efforts to reduce sodium intake, lower consumption of dairy foods, shifts toward use of sea salt as a replacement for iodized table salt, and increased consumption of foods prepared with non-iodized salt such as commercial and packaged products (Krela-Kazmierczak et al., 2021; Panth et al., 2019). In fact, most of the salt consumed in the US is not iodized (Dasgupta et al., 2008). In the United States, insufficient iodine intake among pregnant women has been identified (Panth et al., 2019; Pearce et al., 2016). Despite public health recommendations that women of reproductive age take an iodine-containing dietary supplement due to increased needs during pregnancy and lactation, data from NHANES 2011–2014 indicated that less than 20 % of US pregnant and lactating women used a dietary supplement containing iodine (Gupta et al., 2018).

Accurate assessment of total iodine intake or exposure is important in determining the health status of individuals and populations (Lee et al., 2016; Swanson et al., 2012). Development of an analytical US dataset encompassing a broad array of iodine-contributing foods has become critical for estimating dietary intakes, because the existing US food composition database provides very few iodine values (Ershow et al., 2018; Pehrsson et al., 2016; Swanson et al., 2012). It is important to assess the concentrations of iodine in foods with an adequate sample size and to follow sound analytical protocols when analyzing commercially prepared foods because these foods comprise at least half of the US diet. Additionally, iodine content varies considerably among plant and animal commodity foods (agricultural products), and – on a global perspective – deficiency of iodine is not directly correlated with the economic status of a country (Fuge and Johnson, 2015). Dietary guidance derived from intake estimates starts with high quality estimates of iodine in foods and dietary supplements. Iodine data bases exist globally (Ershow et al., 2018), and are vital for providing relevant data for their respective populations, since iodine levels can vary among countries due to differences in food production and processing methods. High-quality databases include information on analytical methods, sampling plans and sample handling, and provide results in data formats that facilitate links to individual and population intake surveys (Ershow et al., 2018).

An iodine initiative to develop a tool for estimating dietary intake began in 2015 through collaboration between the US Department of Agriculture (USDA) Methods and Application of Food Composition Laboratory (MAFCL), the US Food and Drug Administration (FDA), and the Office of Dietary Supplements-National Institutes of Health (ODS-NIH). Phase 1, summarized in this report, included identifying, analyzing, and reporting the iodine data content of roughly 420 foods. Phase 2 will link the iodine food composition data of Phase 1 to dietary intake data (24 -h dietary recalls) from the NHANES (National Health and Nutrition Examination Survey) (Ahluwalia et al., 2016), to estimate the iodine intake of the US population.

2. Materials and methods

2.1. Food samples

Foods were collected and analyzed by USDA and FDA and data were combined for this project. Both FDA and USDA used inductively coupled plasma mass spectrometry (ICP-MS) with established AOAC methods 2012.14 & 2012.15 for all the iodine data presented in the database.

2.1.1. USDA sample collection and analyses

Selected food samples collected by USDA as part of its National Food and Nutrient Analysis Program (NFNAP) (Haytowitz and Pehrsson, 2018; Patterson et al., 2020) were analyzed for iodine. Priorities focused on key iodine contributors to the US diet including dairy foods, eggs, fish, breads, and other baked goods (Haytowitz, 2015), which are estimated to comprise over two-thirds of the iodine intake in the US diet. Home-prepared foods, retail mixed dishes, and various table salts were also prioritized. As part of the NFNAP plan, representative sample units for most foods were acquired using statistically robust nationwide designs (Pehrsson et al., 2013) from large supermarkets in up to 24 sampling locations (Fig. 1a). Although NFNAP sample units have been collected for over 20 years, analyses for this study were limited to only archived samples and samples of foods collected in the past few years. Samples were prepared for analysis by Virginia Tech scientists, using established protocols (Trainer et al., 2010). The stability of iodine in the homogenized subsamples of the foods, stored at −60 °C was assessed by ICP-MS for both time points.

The samples were chemically analyzed for iodine at validated laboratories (ISO 17025, analytical certification), using ICP-MS (Sullivan and Zywicki, 2012). Rigorous quality control measures were followed, including analysis of reference control materials along with the collected food samples, to monitor accuracy and precision (Phillips et al., 2006). The control materials were either certified Standard Reference Materials® (SRM) from the National Institute of Standards and Technology (Gaithersburg, MD) or secondary matrix-specific control composite materials (CC) that had been cross validated to SRMs. Data were thoroughly reviewed by quality control teams to meet scientific criteria, which included assessing acceptability of precision for duplicate analyses and of results for SRM and CC relative to certified limits for SRM and to established tolerance limits for CC (Phillips et al., 2006). Re-analysis of subsamples of select food samples stored at −60 °C for five years, along with reference materials, indicated that there had been no change in iodine concentrations during the storage period. Additionally, results from CC representing different food matrices were evaluated by assessing results obtained over time.

2.1.2. FDA sample collection and analyses

FDA obtained samples of foods through its Total Diet Study (TDS), a program that was initiated in 1962 (Abt et al., 2018). TDS data collected from 2016 to 2018, analyzed using ICP-MS (Todorov and Gray, 2016, 2017), were included in the USDA, FDA, and ODS-NIH Database for the Iodine Content of Common Foods. In 2016–2017, TDS samples were collected from three cities in each of four U.S. regions, and foods were sampled from each region each year in the same specific season (e.g., Western U.S. samples were collected in the winter season each year). Beginning in 2018, the TDS used a population-based sample design, with samples collected in each of six regions (Fig. 1b) and each of two

a. Locations of USDA's representative sample units collected through the National Food and Nutrient Analysis Program (Haytowitz and Pehrsson, 2018; Patterson et al., 2020).

b. Sampling regions, with population (as of 2012) for FDA samples collected in two-year periods over six regions through the Total Diet Study (Abt et al., 2018; Murray et al., 2008).

Fig. 1. a Locations of USDA's representative sample units collected through the National Food and Nutrient Analysis Program (Haytowitz and Pehrsson, 2018; Patterson et al., 2020). Sampling regions, with population (as of 2012) for FDA samples collected in two-year periods over six regions through the Total Diet Study (Abt et al., 2018; Murray et al., 2008).

six-month long seasons, for food designated as "regional", i.e., possibly varying in nutrient concentration by region or season. Foods were sampled from three cities per region per collection period. Counties were selected as sample locations based on population proportional to size. Retail outlets were determined based on sales and proximity to households. Samples of "national" foods, i.e., those less likely to vary by region or season, such as processed foods, were obtained in Kansas City or online. FDA TDS samples were analyzed by FDA's Kansas City Laboratory, which is ISO 17025 accredited. Good Laboratory Practices (GLPs) were followed, using protocols like those followed by FDA for enforcement purposes. NIST certified reference materials, selected to match the matrices for TDS foods as closely as possible, were used in the analyses.

2.2. *Database preparation*

USDA and FDA data were combined for specific foods when descriptions of those foods were closely matched. If a given food did not have data from both sources, those data were indicated as originating from either USDA or TDS. Food items were grouped into categories similar to groupings used for the legacy USDA database system, to enable users to find items easily. Microsoft ACCESS database software was used to organize data and to calculate means and standard deviations.

3. Results and discussion

Precision of USDA measurements was supported by results for replicate analysis of select control materials (CC and SRM) that were analyzed with samples throughout the study. For a total of 21 materials assayed throughout the study, 19 had data from replicate analysis within-day. The within-day mean, range, and median for HorRat values for each material (the assayed/expected relative standard deviation based on nutrient concentration (Horwitz and Albert, 2006)) were 0.3, 0.0–1.2, and 0.2, respectively, suggesting acceptable precision (expected HorRat $<|3.0|$, Horwitz and Albert, 2006). Seven of these materials had values obtained over a period of several years and are illustrated in Fig. 2. The low HorRat values, with a mean, range, and median of 0.8, 0.2–2.5, and 0.6, respectively, and the lack of any downward trend over time support the stability of iodine in stored samples, given that the controls were stored under the same conditions as food samples. Similarly, no loss of iodine was found in samples of four foods (American cheese, salami, fish sticks, and fast-food egg sandwich) assayed after storage for 5 years. Since no significant differences in results were found, analytical iodine data for stored NFNAP samples were considered valid. Accuracy of the data is supported by results for SRMs relative to the certified ranges (Table 1). In all cases, assayed means were within the certified ranges.

The USDA, FDA, and ODS-NIH Database for the Iodine Content of Common Foods was released on July 31, 2020, in the Database Resources section at www.ars.usda.gov/mafcl (Patterson et al., 2020). The release includes access to data and documentation for 422 foods with corresponding mean iodine, standard deviation, minimum and maximum values (μg per 100 g), and source(s) of data per item. Food categories having the most items analyzed included dairy and eggs (n = 41 foods), vegetables (n = 42), baked goods (n = 35), seafood (n = 32), and restaurant items (including fast foods) (n = 32) (Table 2), due to their prominence in the US diet or presumed iodine content. None of the fruits (n = 21), vegetables (excluding nori) (n = 41), legumes (n = 11), fats and oils (n = 4), or nuts and seeds (n = 3) had median iodine greater than 7 μg/100 g. Across the remaining 13 food groupings (Table 2), those with the highest median iodine content (in μg/100 g) were iodized salt, dairy/eggs, seafood, fast foods/restaurant foods, and mixed dishes. However, within these groups there was a wide range in iodine content of specific foods (Figs. 3 and 4). Within the groups with low (<10 μg/100 g) median iodine content were various foods having notable iodine content (Figs. 5 and 6).

Overall findings on the iodine content of foods reveal considerable variability among specific foods within certain food groups, e.g., seafood (by species) and commercial breads (by dough conditioner type), and among samples of certain foods (e.g., milk and eggs) due to production variables.

3.1. *Major food sources of iodine*

3.1.1. *Seafood and seaweed*

Iodine naturally occurs at relatively high levels in seaweed and many saltwater fish and other seafood, due to their ability to concentrate iodine from seawater (Panth et al., 2019; Rohner et al., 2014), although other factors such diet of the fish, specific harvest location, and biochemical mechanisms can also affect iodine concentration of marine-sourced fish (Nerhus et al., 2018). We measured iodine concentrations in multiple samples of 32 different types of fish and shellfish. Seafood samples that were measured had a wide range of values and iodine content was below the limit of quantification for some products. Measurable iodine (mean ± SD) in seafood items ranged from 4.2 ± 1.9 μg per 100 g in catfish, a freshwater fish, to 227 ± 88 μg per 100 g in haddock, a saltwater fish species (Fig. 3). Translated to amounts per serving, the iodine content of these seafood (mean ± SD) ranged from 3.6 ± 1.6–249.7 ± 97 μg (85 g cooked or 110 g raw). Nori (dried seaweed) averaged 116 ± 309 (mean ± SD) μg iodine per 5 g serving (n = 3; data not shown). Our findings demonstrate that not all seafood items are rich sources of iodine.

3.1.2. *Eggs*

Eggs are a source of dietary iodine because iodine may be present in the chicken feed and shell cleansing agents (Sarlak et al., 2020; U.S. Food and Drug Administration (US FDA), 2020; Wu, 2014). For the shell eggs sampled in 24 retail locations in USDA's national study conducted in 2019 (Roseland et al., 2022), iodine concentration (mean ± SD; μg/100 g) was 49.2 ± 4.3, with individual sample values ranging from 27.0–115.0 (Fig. 7). Values for individual samples varied within± 1.67 μg of the mean 95 % of the time, suggesting relatively low variability. In addition, processed (frozen whole) egg samples from five major producers (up to 3 lots each; n = 14) were analyzed by USDA in 2018. Iodine (mean ± SE) was 70.6 ± 12.2 μg per 100 g, ranging from 44 to 104 μg per 100 g at the 95% confidence level (Roseland et al., 2020b). Iodine content and variability of shell versus processed eggs are reported by Roseland et al. (2022).

3.1.3. *Salt types*

Salt used in home cooking is often iodized, although some currently marketed salt varieties are not iodized. In fact, only 53 % of table salt in US retail outlets was iodized, according to a recent analysis (Maalouf et al., 2015). Salt in commercial products is not typically iodized. Since roughly half of the US diet is composed of commercially packaged or restaurant foods, iodine consumption may not be sufficient for some Americans (Saksena et al., 2018).

Iodine was < 1 μg per 1.5 g serving in all 27 samples of non-iodized sea salt samples. Among the 33 samples of iodized salt (9 sea salt, 24

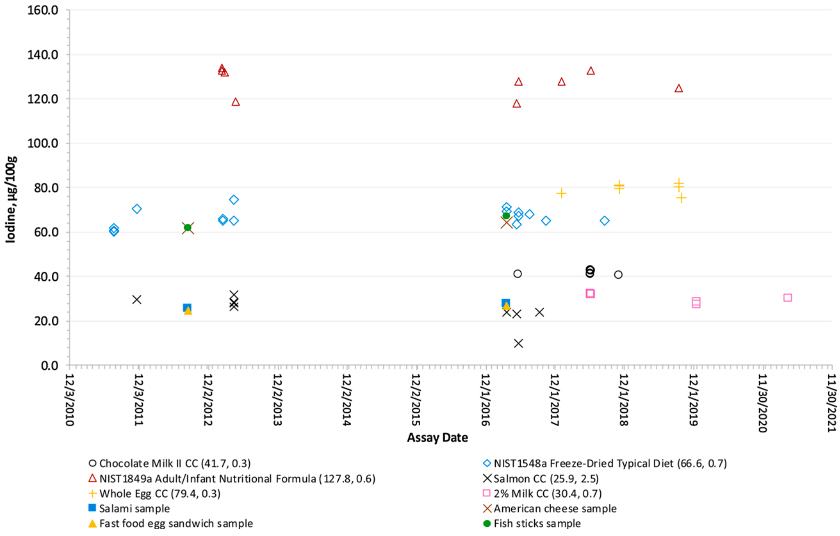

Fig. 2. Results for control materials (CC) and commercial Standard Reference Materials® (SRM) stored at −60 °C analyzed with food samples. Each food sample was analyzed two different times. Certified ranges (μg/100 g) for SRM (National Institute of Standards and Technology, Gaithersburg, MD) NIST1849a: 118-140; NIST 1548a: 65-6-86.2. Mean and HorRat values (Horwitz and Albert, 2006) shown in parentheses in legend for each material.

Table 1
Certified values for Standard Reference Materials® (SRM®) from the National Institutes of Standards and Technology (NIST) (Gaithersburg, MD, USA) with assay results. Iodine concentrations for the materials shown are certified except for 2383a, which are reference values. Certificates of analysis (COA)[1] accessed 7/1/2021, except for SRM® 1549, which is no longer available, for which archived values were used. For materials marked with *, the certified range is on a dry mass basis, and those corresponding assayed values were corrected for residual moisture for comparison.

	Certified or Reference Iodine Values (µg/100 g)				**Assayed Iodine Values (µg/100 g)**			
Material	**Mean**	**Uncertainty**	**Low**	**High**	**Mean**	***n***	**Low**	**High**
1548a Freeze-Dried Typical Diet*	75.9	10.3	65.6	86.2	73.6	*17*	66.8	82.9
1849a Adult/Infant Nutritional Formula	129	11	118	140	127.8	*9*	118.0	134.0
3530 Iodized Table Salt*	5220	420	4800	5640	5736	*3*	5696	5796
1549 Nonfat Milk Powder	338	20	318	358	352.5	*6*	329.0	380.0
2383a Baby Food	7.37	0.83	6.54	8.20	< 10	*1*		

[1] COA dates are as follows: NIST 1548a: 10/6/2020; NIST 1849a: 7/12/2018; NIST 3530: 9/9/2020; NIST 2383a: 8/28/2019.

Table 2
Summary of number of foods analyzed (*n*) and overall iodine content (µg per 100 g) by food category.

Food Group	*n*	Median	Minimum	Maximum
Salt, iodized	*3*	4865	4660	5070
Dairy/Eggs	*41*	42.3	0.7	349
Seafood	*32*	22.0	4.2	227
Fast Foods, Restaurant Foods	*32*	12.2	1.8	278
Mixed Dishes	*21*	10.7	1.8	31.0
Meats (including processed meats)	*22*	3.8	0.4	29.8
Baked goods	*35*	3.2	0.5	1196
Candy, Sweets	*23*	1.5	0.1	46.8
Condiments, Sauces, Soups	*29*	1.5	0.3	20.7
Grains	*9*	1.1	0.1	28.9
Fats, Oils (excluding dairy)	*4*	1.0	0	5.9
Legumes	*11*	0.8	0	9.7
Nuts, Seeds	*3*	0.6	0.3	1.0
Cereals	*14*	0.6	0	14.2
Baby Foods	*43*	0.6	0	27.9
Vegetables	*42*	0.5	0.1	2320[a]
Beverages (excluding milk)	*37*	0.4	0	75.8
Fruits	*21*	0.3	0.1	6.2

[a] Maximum for the vegetable category was 6.2 excluding 2320 µg/100 g for nori (dried seaweed).

table salt), there was no statistically significant difference ($p = 0.360$) in mean iodine content of iodized table salt and iodized sea salt. The mean iodine per 1.5 g serving in iodized salt was 74 µg (median 75 µg) with a 95 % confidence interval (CI) of 69–80 µg per 1.5 g serving. The variability in iodine among individual samples was large (range 34–111 µg) (Fig. 8). Only nine samples fell within the 95 % CI. Dasgupta et al. (2008) reporting on the iodine content of salt in the US (88 samples) found a mean and standard deviation, respectively, of 44.1 and 18.5 mg/kg (66 and 28 µg per 1.5 g serving), and that iodine could vary widely within a container and was affected by humidity.

3.1.4. *Milk*

Dairy foods are an important source of iodine in the US diet. In a nationwide USDA study of whole, skim, 1%, and 2% fat milk from 24 different retail locations ($n = 96$) based upon USDA's statistically based nationwide sampling plan (Roseland et al., 2020a), no significant differences were found among mean iodine concentration or variances of the different milk fat levels ($\alpha = 0.05$). The mean iodine content, plus or minus standard error was 35.4 ± 2.3 µg per 100 g serving, but wide variability was observed among individual samples (13−105 µg per 100 g) (Fig. 9). The results suggest that while the mean iodine content might be a reliable estimate of the average iodine in milk for the US population, such as for predicting population-based intakes, it could be a poor predictor of iodine for individual or local population milk consumption. Possible causes of variability include various aspects of dairy cows' dietary intakes and the use of iodophors for sanitation during milking, with a combination of these factors likely to be the main sources of variability among samples (Roseland et al., 2020a). Research is underway by USDA scientists to investigate potential systematic differences in the retail locations sampled. Iodine content of plant-based beverages marketed as milks will also be investigated by the scientists in this collaboration.

3.2. *Other food sources*

Ancillary studies were conducted by USDA and Virginia Tech scientists to examine the absorption of iodized salt from boiling pasta along with the iodine content of water used in cooking, tap water samples from a nationwide study that had not previously been analyzed for iodine, and presence of iodate-containing dough conditioners in commercial breads. These data will be useful in future evaluations of diet where household salt iodization or the presence of dough conditioners in bread are not known. Results are summarized below.

3.2.1. *Pasta*

Samples of pasta cooked with varying amounts of salt (iodized) in the cooking water, from a study on sodium in cooked pasta (Bianchi et al., 2019), were analyzed for iodine. Iodine concentration in the cooking water was calculated using the analyzed iodine content of salt used. There was no iodine in the pasta cooked without salt, and the content increased linearly with increasing salt concentration in the cooking water. Iodine ranged from 11.3–64.7 ug/100 g for salt concentration ranging from 3.2–12.7 g/L (iodine 0–535 u g/L) for one pound of pasta cooked in 5.68 L water (Fig. 10). Like sodium, the amount of iodine in the pasta varied as a function of iodized salt concentration in the water. However, unlike sodium, the iodine content of iodized salt varied widely (section 3.1.3), and the iodine content of the salt used in this study (2397 ug/100 g) was about one-third of the maximum (7430 ug/100 g) found in the iodized salt samples analyzed for the iodine database ($n = 25$). Therefore, further work prospectively designed to test a wider range of iodine in the cooking water would be needed to fully understand iodine uptake by cooked pasta across a wider range of iodine in the water.

3.2.2. *Tap water*

Fig. 11 illustrates iodine results in a random subset of 40 tap water samples drawn from a larger sample set collected from 144 US locations in a national study of fluoride and minerals in US drinking water (Pehrsson et al., 2006). Samples were obtained from wells and municipal sources, but homes using water treatment systems were not included. Samples were analyzed by ICP-MS at FDA's laboratory. Overall, iodine in tap water was low. In 36 samples it was ≤ 2 µg per µ100 g (per 240 mL serving), but 4 samples had higher iodine contents of (2.5–11 µg/100 g (6–26 µg per 240 mL serving). These data suggest that iodine in a particular water supply, if consistently higher than the national mean, could significantly impact iodine intake from drinking water (for example, 6–26 µg per 240 mL (8 oz) would contribute 36–156 µg for daily intake of 6 servings of water.

Iodine (as iodide) is widely but unevenly distributed in the earth's environment. Although most iodine is present in the oceans, in many

Fig. 3. Summary of results (mean and range) for items in the seafood category with median iodine content greater than 10 µg/100 g. (Mean values below the limit of quantitation of 10 µg/100 g appear as 0.). Dotted vertical line shows Dietary Reference Intake (IOM, 2001).

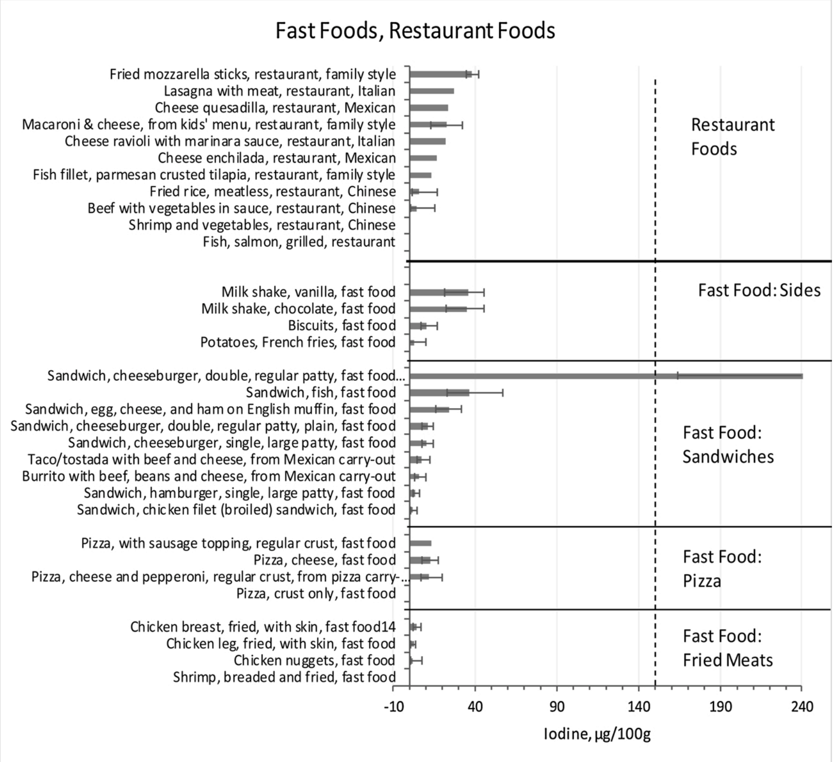

Fig. 4. Summary of results (mean and range) for items in the fast foods and restaurant food categories with median iodine content greater than 10 µg/100 g. (Mean values below the limit of quantitation of 10 µg/100 g appear as 0.) Dotted vertical line shows Dietary Reference Intake (IOM, 2001).

land regions, flooding, leaching from glaciations, flooding, and erosion have used up surface soils of iodide. Seawater iodide ions are oxidized to elemental iodine, which in turn volatilizes into the atmosphere; it is subsequently returned to the soil through rainfall, completing the iodine cycle. Iodine cycling may be incomplete in many areas of the world, impacting repletion of iodine in soils, the crops they bear, and drinking water. Weathering of iodine-containing rocks, and volcanic activity (including under-water volcanoes) contribute to groundwater iodine

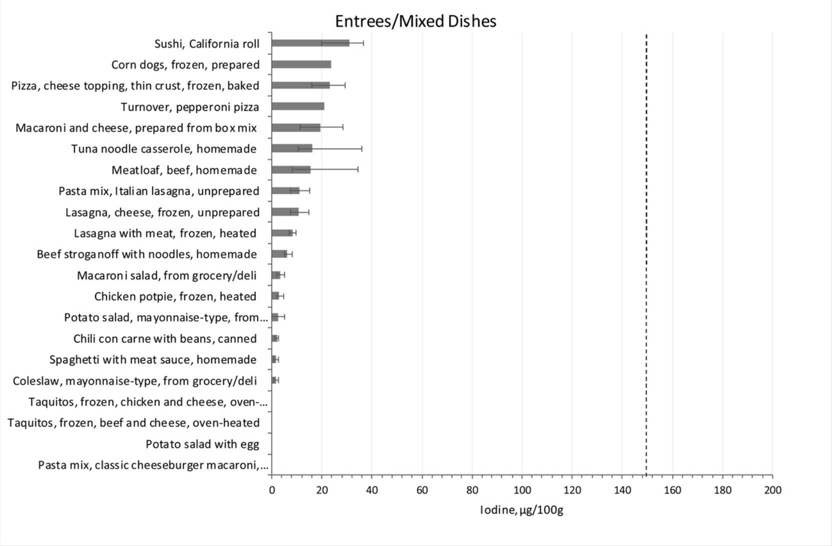

Fig. 5. Summary of results (mean and range) for items in the entrees and mixed dishes categories with median iodine content greater than 10 µg/100 g. (Mean values below the limit of quantitation of 10 µg/100 g appear as 0.) Dotted vertical line shows Dietary Reference Intake (IOM, 2001).

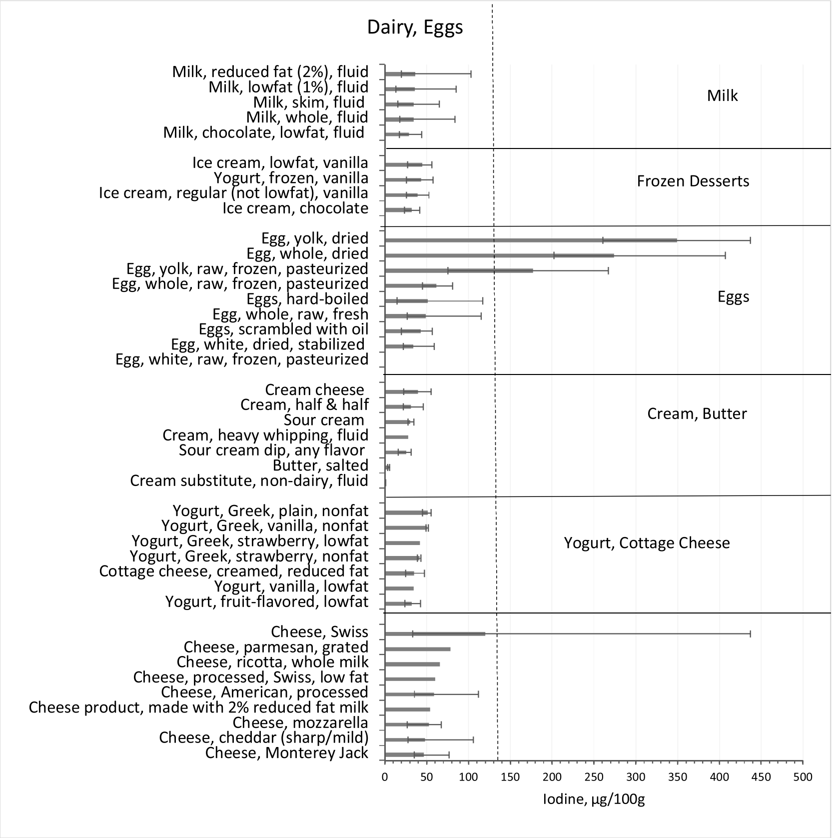

Fig. 6. Summary of results (mean and range) for items in dairy and egg categories with median iodine content greater than 10 µg/100 g. (Mean values below the limit of quantitation of 10 µg/100 g appear as 0.) Dotted vertical line shows Dietary Reference Intake (IOM, 2001).

concentrations. Our water data reflect the influence of these various factors, to be viewed as representing iodine content at a single snapshot in time, according to individual location. The impact of these data is that some individual sources could vary quite significantly from a national mean or median. Populations served by a particular water supply could receive quite a bit of iodine or very little. More work is necessary to determine water sources of iodine and consistency among outlets supplied.

3.2.3. *Breads*

The use of dough conditioners in commercial baked goods serves many purposes: to strengthen texture, accelerate the speed of dough rise, or make the dough more workable. Some dough conditioners are iodate-based, e.g., potassium and calcium iodate, whereas others are not. In our

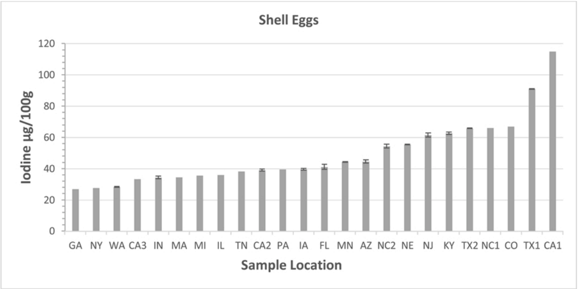

Fig. 7. Iodine concentration (µg per 100 g) of shell eggs sampled at 24 US retail locations (Roseland et al., in press). Error bars show standard deviation for analytical replicates.

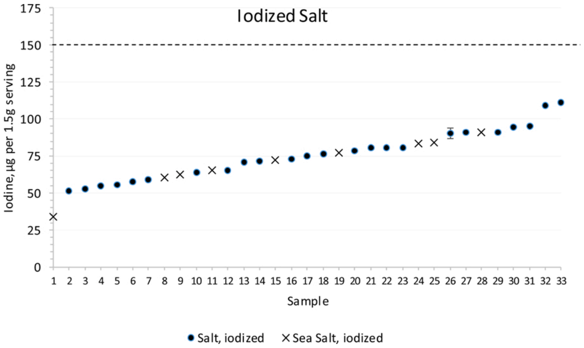

Fig. 8. Iodine content of iodized table and sea salt, µg per serving (1.5 g, 1/4 teaspoon). Error bars show standard deviation of replicate measurement. Dotted line shows the Dietary Reference Intake (IOM, 2001).

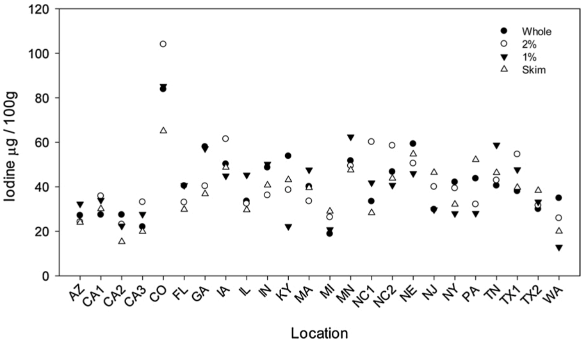

Fig. 9. Iodine concentration in individual samples of milk (µg per 100 g) sampled at 24 US retail locations in different states (from Roseland et al., 2020).

findings, considerable levels of iodine were measured in some bread samples, despite the presumed discontinuation of iodate-containing conditioners in commercial bakeries (Panth et al., 2019; Rappaport et al., 2017). Bread labels are not always clear since iodates can be shown in the ingredient list among dough conditioners that "may be used", but the label does not indicate which one was used. Results are shown in Fig. 12. The mean iodine concentration for bread products with iodate dough conditioners ranged from 618 to 1196 µg per 100 g (309–598 µg per 50 g serving), while counterparts without iodates had <2 µg per serving. Substantial variability of iodine content among commercial breads has also been noted by others (Murray et al., 2008; Pearce et al., 2004; U.S. Food and Drug Administration (US FDA), 2017).

USDA plans to analyze additional commercially produced breads containing iodate dough conditioners or FD&C Red No.3. USDA is also comparing ingredient labels from breads purchased in stores to the ingredient lists for the same products in the USDA Global Branded Food Products Database (U.S. Department of Agriculture (USDA), Agricultural Research Service, 2021a) to verify whether iodate containing ingredients are currently in the foods and can be matched to foods reported by NHANES participants.

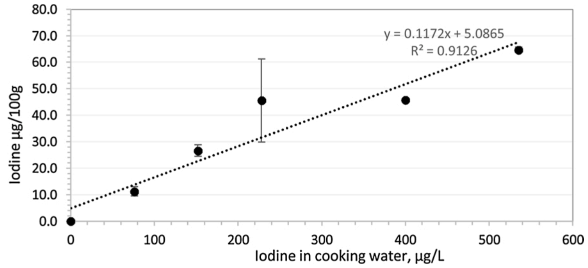

Fig. 10. Iodine in pasta (semolina spaghetti) cooked in water with varying amounts of iodized salt, as a function of iodine concentration in the cooking water. Pasta samples were from a previous study on sodium in cooked pasta (Bianchi et al., 2019), with 454 g pasta cooked in 5.68 L water. Iodine concentration in the water was calculated using the analyzed iodine concentration in the salt (2397 ug/100 g). Data points are the mean $\pm$ standard error of samples from three experiments for each salt level.

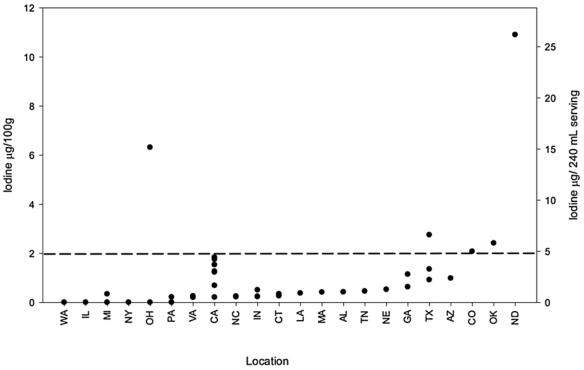

Fig. 11. Iodine concentration in tap water (µg per 100 g and per serving) for individual samples at 40 locations in US states. Dotted line indicates limit of quantitation.

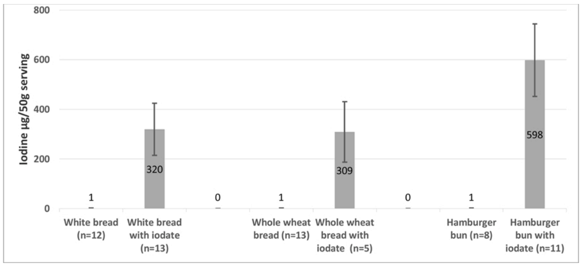

Fig. 12. Iodine concentration (mean and SD; µg per 50 g serving) among bread samples with and without iodate dough conditioners. (50 g serving = 2 slices bread or 1 bun).

4. Estimating iodine intake with USDA, FDA, and ODS-NIH data and survey data

Foods reported in dietary intakes from about 900 survey participants for whom 24 -h urinary iodine measurements are available (NHANES 2014) are being linked to iodine data from the USDA, FDA, and ODS-NIH database. To highlight the importance of developing good analytical data on iodine, these data will be used to assess dietary iodine intake using the recommended indicator of intake adequacy for a population, multiple measures of 24 -h urinary iodine excretion, and to evaluate measurement error from self-report in 24 -h dietary recalls. Foods such as salt, breads, and pasta, can have very different amounts of iodine. Eventually the USDA, FDA, and ODS-NIH database iodine composition data will be linked to all participants and subsequent NHANES cycles and will provide the most comprehensive estimates of dietary iodine intake in the US population and population sub-groups.

A factor affecting iodine status is that some foods contain goitrogens, which are dietary substances that impede thyroid metabolism and can aggravate iodine deficiency (Panth et al., 2019). Cruciferous vegetables such as broccoli, cabbage, kale, and cauliflower contain glucosinolates, the metabolites of which compete with iodine for uptake by the thyroid gland. Similarly, cassava, lima beans, sorghum, and sweet potato contain cyanogenic glucosides which can be metabolized to thiocyanates that compete with iodine for uptake (Zimmermann, 2009). Isoflavones in soy foods also have goitrogenic effects (Doerge and Sheehan, 2002), and higher consumption of soy products has been associated with lower iodine status (Herrick et al., 2018). Additionally, exposure to perchlorate, a chemical which has been detected in US food and water samples, can inhibit iodine uptake (Dasgupta et al., 2008; Murray et al., 2008). In countries where these goitrogens are highly consumed and iodine adequacy is at risk, data on goitrogen content and other interfering compounds in foods would be an important relevant resource, and thus is potentially a future research need.

5. Dietary supplements

The Dietary Supplement Label Database (DSLD; https://dsld.od.nih.gov/dsld/) provides label information from dietary supplements such as ingredient amounts, instructions for use, targeted consumers' age and gender, quality claims, manufacturer, and other information. When we used "advanced search" function and searched for "MVM "products that had the term "prenatal" in their name, were manufactured for consumption by "pregnant and lactating women and that were currently on the market, 73 products were returned, and, of these, only 47 products listed iodine in "Supplement Fact" panels. If we included products that are already off the market and present in NHANES, 98 products were found, and of these, 62 products listed iodine in the "Supplement Fact" panels.

Ingredient content measured in dietary supplements may significantly differ from labeled content; it can be below or above the label claims. To account for such differences in intake calculations in population studies, the Dietary Supplement Ingredient Database (DSID; https://dsid.usda.nih.gov/) provides sampling plan descriptions and nutrient content estimates for popular categories of dietary supplements based on chemical analysis of nationally representative products (U.S. Department of Agriculture (USDA) Agricultural Research Service et al., 2021b). The DSID study results for the iodine content of adult multivitamin/mineral products (MVM), non-prescription prenatal MVM, and children's MVM were previously published (Andrews et al., 2017, 2018; Pehrsson et al., 2016). Mean and variability results for iodine in these categories of dietary supplements are reported in the DSID calculators (for example, for non-prescription prenatal MVM: https://dsid.od.nih.gov/ingredient_calculator/calc_nonrx_prenatal.php).

At the most common label level for iodine of 150 µg per serving, the predicted mean

content of non-prescription MVM was 25.9 % above the labeled value leading to a predicted analytical mean (±SEM) of 189 ± 7.0 µg/per serving. Individual product results, including the listed sources of iodine are shown in Fig. 13.

The iodine content of prenatal MVM sold by prescription was also analyzed. Prescription prenatal MVMs are not categorized as drugs by the FDA, but as dietary supplements.

Two to three lots of twenty-four products were purchased from local pharmacies and comprised 61.2 % of the market. ICP-MS was used to measure iodine. The percentage difference from the label claim was calculated for iodine in each sample: [(analytic value − label value)/label value] ×100 %. Regression analysis, weighted by market share, was conducted to determine the relationship between the percentage differences from labels and label values for iodine.

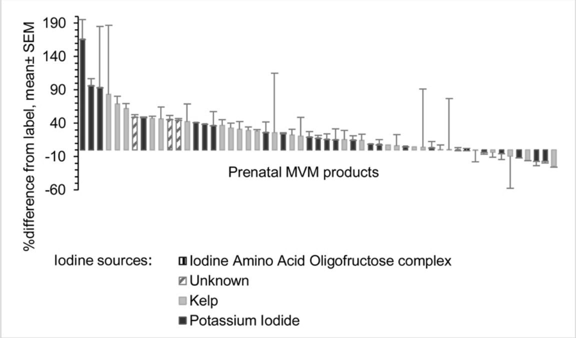

Fig. 13. Iodine sources, mean percentage difference from label claims for iodine content and lot-to-lot variability in 55 non-prescription prenatal multivitamin/mineral (MVM) products (n = 1-3 lots). SEM = standard error of the mean. (Andrews et al., 2014).

In the prescription prenatal MVM purchased for analyses in 2017–2018, only 13 (54.1 %) of 24 of products contained iodine, and their summed market share was quite low, 17.3 %. In comparison, in the non-prescription MVM purchased for analysis in 2009–2010, 77.5 % of the products (n = 55) listed iodine on the labels. Among prescription prenatal MVMs, all products except one were labeled at 100−150 µg per day (which is below the RDA for iodine for pregnant women = 220 µg per day). The World Health Organization (WHO), United Nations Children's Fund (UNICEF), and the International Council for the Control of Iodine Deficiency Disorders (ICCIDD) recommend a slightly higher iodine intake for pregnant women of 250 µg per day (World Health Organization (WHO)., 2007a; World Health Organization (WHO) Secretariat et al., 2007b).

Mean percentage differences from labeled levels for iodine in the 13 prescription prenatal MVM products were quite variable. Nine products contained iodine between 10 and 32 % above label and one was 49 % above label. Two products had iodine at 20 and 39 % below the label, and the most expensive product averaged 95 % below label for iodine (based on 3 lots). Regression analysis (after influence testing and weighting by market share) predicted a mean iodine content in prescription prenatal MVMs of 18.5 ± 6.2 % (±SEM) above the labeled value.

Different categories of MVMs may provide significant amounts of iodine for MVM consumers in the US. The analytical studies performed on different categories of MVM suggest that, on average, the products have similar percentage differences from the iodine content claimed by their labels (approximately 20 % above label), with some products having iodine content significantly above or below the label claims. Because of the widespread deviation in iodine content from the labels, application of analytically derived estimates of iodine content provided by DSID can improve precision of iodine intake calculations in epidemiological studies.

6. Conclusions

Iodine content data for 422 foods are now publicly available at www.ars.usda.gov/mafcl. The USDA, FDA and ODS-NIH Database for the Iodine Content of Common Foods provides a valuable tool for research programs and clinical applications in iodine nutrition. The US population intake estimates derived by linking these iodine data to dietary recalls from individual NHANES participants will address important research needs on assessment of iodine status, help identify iodine-rich foods (means and sources) for at-risk groups and be useful for developing dietary guidance. In addition, labeled and analytically estimated iodine amounts in dietary supplements are provided by the Dietary Supplement Label Database (https://dsld.nlm.nih.gov/dsld/) and the Dietary Supplement Ingredient Database (https://dietarysupplementdatabase.usda.nih.gov/) and will be useful to estimate total iodine intakes. The database of iodine in foods will be updated as new data are accumulated. Related projects include linking of iodine data to subsequent NHANES cycles, DSID and DSLD.

CRediT authorship contribution statement

Pamela R. Pehrsson: Conceptualization, Methodology, Validation, Investigation, Resources, Visualization, Supervision, Project administration, Funding acquisition, Writing - review & editing. **Janet M. Roseland:** Validation, Investigation, Writing - original draft, Visualization, Writing - review & editing. **Kristine Y. Patterson:** Validation, Investigation, Visualization, Writing - review & editing. **Katherine M. Phillips:** Validation, Formal analysis, Data curation, Writing - review & editing. **Judith H. Spungen:** Data curation, Writing - review & editing. **Karen W. Andrews:** Supervision, Project administration, Writing - review & editing. **Pavel A. Gusev:** Conceptualization, Investigation, Writing - review & editing. **Jaime J. Gahche:** Writing - review & editing. **Carol J. Haggans:** Writing - review & editing. **Joyce M. Merkel:** Writing - review & editing. **Abby G. Ershow:** Conceptualization, Resources.

Declaration of Competing Interest

The authors, shown above, have no conflicts of interest.

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Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.jfca.2021.104369.

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