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Semi-automatic Method for the Determination of Total Iodine in Food

R. E. D. Moxon and E. J. Dixon

Department of Industry, Laboratory of the Government Chemist, Cornwall House, Stamford Street, London, SE1 9NQ

A simple method for the determination of total iodine in food, based on the catalytic destruction of thiocyanate by nitrite in the presence of iodide, has been evaluated and the colorimetric finish technique automated. Optimum conditions for the destruction of organic matter and the liberation of iodine have been investigated and a set of conditions applicable to the automated finish technique developed. The method, when applied to 160 foods, had a precision of about 10%, a detection limit for iodine of 1 μg per 100 g of food, a mean recovery of added iodide of 90% and an output of about 70 samples per week. The effect of storage conditions on the iodine content of fish and eggs has also been examined, and the effect of some possible interfering substances, such as chloride, bromide and mercury, investigated.

Keywords: *Semi-automatic method*; *total iodine determination*; *foods*

In preparation for a proposed survey on the iodine content of food, a reliable and sensitive method for determining total iodine, which could be applied to a wide range of foods, was sought. This survey has been arranged and funded by the Ministry of Agriculture, Fisheries and Food; the results will be published elsewhere.

A literature search showed that colorimetric methods[1,2] based on the catalytic effect of iodide were widely used. Various other methods have also been reported, including atomic-absorption spectrophotometry,[3] gas - liquid chromatography,[4] neutron activation[5] and X-ray fluorescence.[6] In terms of productivity, sensitivity and ease of operation, the colorimetric methods appeared to be most suitable for our requirements.

Modifications of the colorimetric methods of Sandell and Kolthoff,[1] which make use of the catalytic effect of iodide on the reduction of cerium(IV) sulphate by arsenious acid, and of the method of Sveikina,[2] in which iodide catalyses the destruction of thiocyanate by nitrite, with an accompanying decrease in the colour of the iron(III) thiocyanate produced by the addition of iron(III) ions, were both considered sufficiently sensitive and simple for automation. The Sveikina method gave a straight-line calibration graph, whereas the Sandell and Kolthoff method gave a curved calibration graph. Proskuryakova *et al.*[7] have compared these two methods for the analysis of waters and concluded that the Sveikina method has marginally better sensitivity and precision. The Sveikina method was therefore selected for further investigation.

Destruction of Organic Matter

The following three techniques have been considered for the destruction of organic matter prior to iodine determinations. Bomb calorimetry[8,9] is too time consuming for large numbers of samples, and it has been reported that combustion is often incomplete and that the ash residue can interfere with catalytic finish techniques.[10]

Acid combustion[11–14] has been widely used for foods and clinical specimens, but it requires the distillation of iodine into a suitable medium and so is only applicable to small numbers of samples.

Alkaline dry ashing has been widely used for foods, clinical specimens and animal feeding-stuffs. It is simple, adequately sensitive and has a productivity potential that is suited to our requirements. In this technique the recommended temperature of combustion varies from 400–480 °C[2,15] to 600 °C[16–18] and the time of ashing from 30 min[2] to 3 h or more.[16] Fioravanti and Halmi[10] studied these variables and established combustion conditions to give the minimum loss of iodine. They obtained optimum recoveries using the following

procedure. To 1 g of sample in a porcelain crucible add 2 ml of 1 M potassium hydroxide solution and 1 ml of 10% m/V zinc sulphate solution. Dry the mixture completely in a drying cabinet, then place a porcelain lid on the crucible and heat at 450 °C for 1.5 h. Cool the crucible, dampen the residue with a few drops of water, add a further 1 ml of 10% m/V zinc sulphate solution and repeat the ashing procedure for 1.5 h.

Experimental

In order to match this technique to an automated finish technique, and make it applicable to a wide variety of foodstuffs, certain aspects of the ashing procedure proposed by Fioravanti and Halmi had to be modified. Comparison of sodium carbonate, potassium hydroxide and potassium carbonate as ashing aids showed that all of them gave quantitative recoveries of iodine when using a proprietary milk powder as the test material. Potassium carbonate was found to be best suited to an automated finish method as it had the lowest blank value. Potassium hydroxide gave a higher background reading and an unsteady base line when the determination was completed automatically, but gave good results with a manual finish technique. It was confirmed that the use of a lid on the crucible during the ashing procedure was essential to prevent losses of iodine and it was shown that for cod, and a proprietary milk powder, an ashing temperature of 550 °C was required to destroy all of the organic matter when using 1 ml of 30% m/V potassium carbonate solution as the alkaline reagent.

Use of Zinc Sulphate as an Ashing Aid

It was found that when the amount of 10% m/V zinc sulphate solution, which is used to assist the combustion of organic matter, was increased from 2 to 3 ml some loss of iodine occurred, and that with 4 ml, major losses were encountered. This loss is thought to be associated with the change in alkalinity that occurs when the zinc carbonate formed is converted into zinc oxide at a temperature of 550 °C.

Effect of Alkali on the Colorimetric Reaction

When sample solutions obtained by using the ashing procedure of the proposed method were titrated with 0.2 N nitric acid, using phenolphthalein as indicator, they were found to have undergone a 50% reduction in alkalinity compared with non-ashed standard solutions made up in 0.6% m/V potassium carbonate solution. Two standard solutions of iodine containing 20 ng ml^{-1} were made up in 0.3 and 0.6% m/V potassium carbonate solution. The chart response (percentage of full-scale deflection) for the solution containing 0.3% potassium carbonate was 10% higher than for that containing 0.6% potassium carbonate when run using the automated finish technique. The alkalinity of ash solutions from seven different foods showed a mean variation of 10%. Thus, even though there was a considerable excess of nitric acid in the ammonium iron(III) sulphate reagent, it was considered necessary to match the alkalinity of the standard and sample solutions, and this was done by making up the standards in 0.3% m/V potassium carbonate solution.

Extraction of Iodide From the Ash Residue

Following the destruction of organic matter, the residue is dissolved in 50 ml of distilled water. Any trace amounts of carbonaceous matter must be removed as they can affect the catalytic reaction. Some batches of filter-paper were found to adsorb iodide, so it was decided that the removal of carbonaceous matter would be best achieved by centrifrugation at 50 Hz for 5 min.

Colorimetric Reaction

The method of Sveikina[2] involves the addition of 0.4 ml of 0.006 M potassium thiocyanate solution, 1 ml of 0.3 M sodium nitrite solution and 1.6 ml of 0.2 M ammonium iron(III) sulphate in 2 M nitric acid to 4 ml of the standard or sample solution containing 0–60 ng of iodide and 0.012 g of potassium carbonate. The mixture is shaken and, after standing for 20 min, the absorbance is read at 430 nm. The reaction was found to be dependent on

hydrogen ion concentration, temperature and time, and in order to control these variables more closely, and also increase the precision and productivity, this manual method was automated by use of standard Technicon equipment (see Fig. 1).

Automation of the Colorimetric Determination

In the course of automating the colorimetric determination several problems arose, which did not occur in the manual method. Erratic and drifting base lines were obtained from day to day, a problem that was solved by changing the absorption wavelength from 430 to 450 nm, thereby greatly reducing the background absorbance of the reagents. A regular interference associated with the sampler was eliminated by removal of the bubble introduced during the sampling action. An interfering peak was suppressed by changing the sampler wash solution from distilled water to a potassium carbonate solution of the same concentration as that used in the standards. By this means, reproducible calibration graphs were obtained for iodide concentrations up to 20 ng ml^{-1}. The equipment and all reagents were kept in a room maintained at 20 $\pm$ 1 °C.

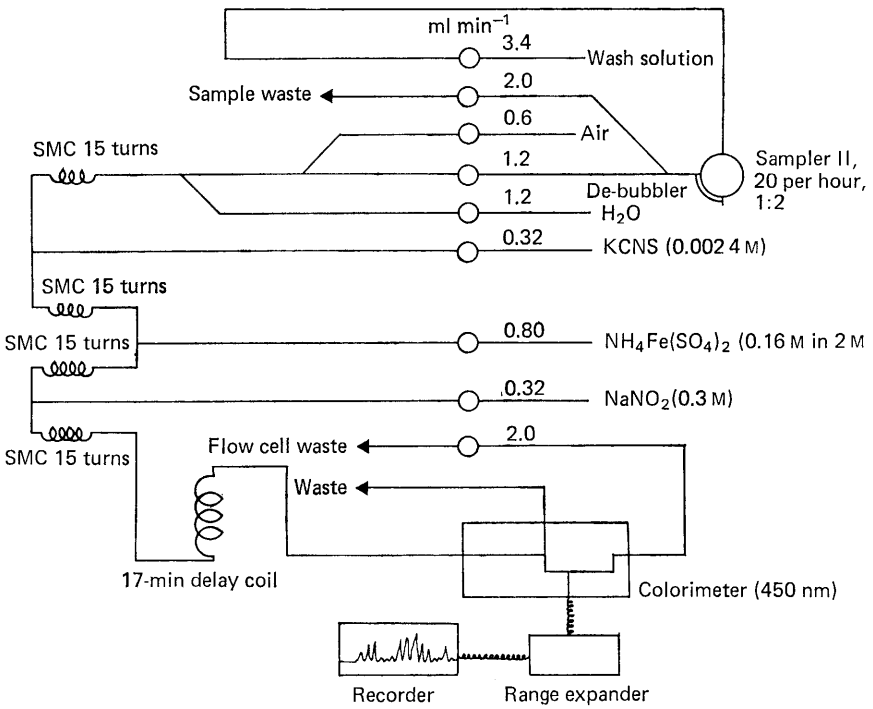

Fig. 1. Flow diagram for the determination of iodine in foods.

Method for the Determination of Iodine in Food

Preparation and Storage of Samples

When a determination is carried out using only 1 g of food, care must be taken to ensure that the sample is properly homogenised by thorough mincing, grinding or blending. Samples should be examined when fresh, but if this is not possible they should be deep frozen and stored in sterile plastic containers. In order to avoid contamination, all bench surfaces should be clean, and glass, porcelain and polythene apparatus should be washed with concentrated nitric acid and rinsed thoroughly with distilled water before use.

Preparation of Standard Iodide Solutions

Standard iodide solution (4 g l^{-1}). Dissolve 0.523 2 g of potassium iodide, previously dried in a desiccator, in distilled water and dilute the solution to 100 ml in a calibrated flask (stable for at least 1 month).

Standard iodide solution (40 mg l^{-1}). Dilute 10 ml of the standard iodide solution (4 g l^{-1}) to 1 000 ml with distilled water in a calibrated flask (stable for at least 1 month).

Standard iodide solution (200 ng ml^{-1}). Dilute 5 ml of the standard iodide solution (40 mg l^{-1}) to 1 000 ml with distilled water in a calibrated flask. Store the solution in a glass bottle away from light (stable for 1 month only).

Into a series of 100-ml calibrated flasks, pipette 10, 8, 6, 4, 2 and 0 ml of standard iodide solution (200 ng ml^{-1}). To each solution add an amount of 30% *m/V* potassium carbonate solution so that when diluted to 100 ml it is of the same alkalinity as that of the sample solutions (usually 1 ml of 30% *m/V* potassium carbonate solution). These are the working standards; store them in glass bottles away from light and prepare them freshly at weekly intervals.

Reagents

All of the reagents should be of analytical-reagent grade and distilled water should be used in preference to de-ionised water.

Potassium carbonate solution (30% *m/V*). Dissolve 30 g of potassium carbonate in water and dilute to 100 ml.

Zinc sulphate solution (10% *m/V*). Dissolve 10 g of zinc sulphate ($ZnSO_4.7H_2O$) in water and dilute to 100 ml.

Potassium thiocyanate solution (0.023% *m/V*). Dissolve 0.23 g of potassium thiocyanate in water and dilute to 1 l.

Sodium nitrite solution (2.07% *m/V*). Dissolve 2.07 g of sodium nitrite in water and dilute to 100 ml (stable for 1 d only).

Ammonium iron(III) sulphate reagent. Dissolve 77 g of ammonium iron(III) sulphate [$NH_4Fe(SO_4)_2.12H_2O$] in approximately 400 ml of water. Add 167 $\pm$ 1 ml of concentrated nitric acid (sp. gr. 1.42) and dilute to 1 l, warming until all traces of the solid dissolve.

Potassium carbonate wash solution (0.3% *m/V*). Dissolve 3 g of potassium carbonate in water and dilute to 1 l.

Apparatus

Muffle furnace with thermostat (0–1 000 °C).
AutoAnalyzer system for colorimetric analysis (*Technicon*).
Porcelain crucibles, 45 mm *diameter, with lids.*
Centrifuge, operated at 50 Hz.
Centrifuge tubes (*plastic or glass*) *of* 150-ml *capacity.*
Vortex mixer (*for manual method only*).

Procedure

Into a clean, dry crucible, accurately weigh approximately 1 g of food with a content of iodine not exceeding 1 μg. (For some samples, such as fish, a lower mass can be used; alternatively, the final ash solution can be diluted with potassium carbonate solution to match the alkalinity of the standards. For samples with a high water content and a low iodine level, up to 10 g can be taken, provided that a white ash is obtained.) Add 1 ml of 30% *m/V* potassium carbonate solution and then 1 ml of zinc sulphate solution. Slurry the mixture with a glass rod and wash any residue left on the rod back into the crucible with a jet of distilled water. Next, place the crucible in a drying cabinet at 95 °C until dry, preferably overnight. Cover the crucible with a lid and place it in a muffle furnace at 100 °C. Then raise the temperature evenly to 550 °C in approximately 90 min and maintain this temperature for 1 h. Remove the crucible immediately and allow it to cool to room temperature.

Add 1 ml of zinc sulphate solution and slurry the charred residue. Repeat the drying and ashing procedure as before. Next, transfer the cooled ash, normally white or grey in colour, to a centrifuge tube with 50 $\pm$ 0.5 ml of distilled water, and spin the tube at 50 Hz for 5 min. Decant about half of the solution and store it in a clean polyethylene container before analysing it by use of the AutoAnalyzer. Also take three reagent blanks through the whole procedure.

Place the standards, samples and blanks into their respective sample cups and run them at a rate of 20 per hour as shown on the flow diagram, in a room maintained at a constant temperature of 20 ± 1 °C. Install the 17-min delay coil in a constant temperature water-bath if no suitable room is available.

A Technicon Mark I AutoAnalyzer System was used for this work; it was operated and maintained in accordance with the procedure described in the Technicon Mark I Assembly and Operating Instruction Manual, published in 1962 by the Technicon Chromatography Corporation, Research Park, Chauncey, New York, USA.

A manual version of this procedure would operate as follows. Pipette 4 ml of each standard, sample and reagent blank solution into its respective 15 × 1.5 cm test-tube. To each tube add 1 ml of distilled water, 1 ml of potassium thiocyanate solution and 2 ml of ammonium iron(III) sulphate reagent. Mix the orange solutions in the tubes well on a vortex mixer. At exactly 90-s intervals add 1 ml of sodium nitrite solution and again mix on a vortex mixer. Measure the colour at a wavelength of 450 nm on a spectrophotometer after 20 min, still at exactly 90-s intervals. It is essential that all solutions should be maintained at the same temperature:

Calculation

By using a range of standards, prepare a calibration graph. From the absorbance values of the sample and blank solutions, determine the iodine content of the solutions, in ng ml^{-1}, from the calibration graph. Calculate the iodine content of the sample in μg per 100 g using the following equation:

$$\text{Iodine content } (\mu\text{g per } 100\text{ g}) = [(A - B) \times 5]/W$$

where A is the iodine content of the sample solution (ng ml^{-1}), B is the mean iodine content of the blank solution (ng ml^{-1}) and W is the amount of sample in grams.

Results and Discussion

Application of the Method to a Standard Reference Material

A sample of human serum was obtained from the International Atomic Energy Authority in Vienna, which had been analysed by 11 laboratories for total iodine by various methods. Table I shows the value obtained by this laboratory compared with those obtained by other laboratories.

TABLE I

COMPARISON OF TOTAL IODINE VALUES OF HUMAN SERUM OBTAINED BY 12 LABORATORIES USING CHEMICAL AND RADIOCHEMICAL METHODS

Laboratory code	Method of analysis	Number of determinations	Mean value/μg per 100 ml ± standard deviation
Laboratory of Government Chemist (present work)	Chemical	3	7.00 ± 1.40
A	Chemical	3	5.7 ± 0.0
B	Chemical	7	5.40 ± 0.46
C	Activation analysis	1	82 ± 20
D	Chemical	2	7.0
E	Activation analysis	6	121 ± 2
F	Activation analysis	20	5.74 ± 0.38
G	Activation analysis	3	9.50 ± 1.49
H	?	3	6.87 ± 0.06
I	Activation analysis	6	6.17 ± 0.20
K	Activation analysis	9	8.68 ± 1.97
L	Chemical	3	6.53 ± 1.62

The value tentatively estimated from selected results by the author of the IAEA report[19] for the sample of human serum is 5.92 ± 0.20 μg per 100 ml. If values from laboratories C and E only are disregarded, the mean of the mean values for serum becomes 6.8 ± 1.4 μg per 100 ml, which is not significantly different from the value found by the proposed method.

Effect of Interfering Ions

Possible interfering substances were thought to include metals, such as mercury, which form insoluble iodides, trace metals, which are common in foods, and other halides, which might act similarly to iodide in the finish reaction. Standard iodide solutions, containing 16 ng ml^{-1}, were prepared containing 0.01, 0.05 and 0.1 mg l^{-1} of mercury, added as mercury(II), 250 mg l^{-1} of chloride and 0.1 mg l^{-1} of bromide. The iodine contents of these solutions were determined by the proposed method, omitting the alkaline ashing stage, and were then compared with a standard iodine solution containing 16 ng ml^{-1} of iodine treated similarly. The results, which are shown in Table II, indicate that chloride and bromide at 250 and 0.1 mg l^{-1}, respectively, did not interfere, but that mercury at a concentration of 0.01 mg l^{-1} gave a value 18% lower than the standard. In order to determine whether the ashing stage of the proposed method would remove or suppress mercury and so obviate interference at the colorimetric stage, and also to investigate the possible interference of other trace metals commonly found in foods, 10 μg of mercury(II) and 0.2 mg of manganese(II), iron(III), copper(II), zinc(II) and lead(II) were added to 0.5 g of a proprietary milk powder. When these samples were analysed in duplicate by the proposed method no significant changes were found compared with the mean of five untreated samples.

TABLE II

EFFECT OF MERCURY, CHLORIDE AND BROMIDE ON THE DETERMINATION OF 16 ng ml^{-1} OF IODINE IN STANDARD SOLUTIONS BY THE PROPOSED METHOD, OMITTING THE ALKALINE ASHING STAGE

Added ions	None	Hg^{2+}	Hg^{2+}	Hg^{2+}	Cl^-	Br^-
Amount added/mg l^{-1}		0.01	0.05	0.1	250	0.1
Iodine found/ng ml^{-1}	16.0	13.1	6.8	2.1	16.0	16.0
Reduction, %	0.0	18.1	57.5	87.1	0.0	0.0

The effect of iodate on the catalytic finish method was investigated by using a solution containing 12 ng ml^{-1} of iodine made up from potassium iodate. This solution was found to have no effect on the catalytic finish method, but when the same solution was taken through the ashing procedure of the proposed method the iodate was reduced to iodide and was quantitatively recovered.

Precision of the Proposed Method

In order to obtain a measure of the precision of the method, five samples of a proprietary baby-milk powder and five samples from a bottle of milk were ashed on each of two separate days. The iodine content of each sample was then determined by using the proposed automated method three times on one day and then once on each of four consecutive days. The iodine contents were calculated, and a summary of the results is shown in Table III. The variation in the values for individual samples between determinations, expressed by the standard deviation from the mean, gives an indication of the general precision of the method.

The mean values of 35 determinations of iodine, expressed in μg per 100 g, for milk powder and liquid milk are 142 $\pm$ 10.5 and 17.8 $\pm$ 3.5, respectively. However, if the value for liquid milk 6 ashed on day 2 is excluded, then the mean value for liquid milk (34 determinations) becomes 16.8 $\pm$ 2.2.

In Table IV a measure of the precision of the catalytic finish method can be obtained from the figures for the calibration graphs run in conjunction with the samples of milk powder and bottled milk shown in Table III.

Limit of Detection

The limit of detection was calculated by using twice the standard deviation of the iodine value, in nanograms per millilitre, of the three reagent blanks that were included with each set of samples. The mean blank value was generally 1 ng ml^{-1} and the 2 $\times$ s.d. value ranged from 0.2 to 0.5 ng ml^{-1}. The limit of detection for a 1-g sample varied from 1 to 2 μg of iodine per 100 g of food. This value represents 1/100 of the Recommended Daily Allowance[20]

and so is acceptable for nutritional purposes. However, should it be necessary to operate at a greater sensitivity, it would be possible to lower the detection limit by drying the sample and increasing the sample size. In addition, the sensitivity of the finish method could be increased by using the scale expansion facility, or by raising the temperature at which the reaction occurred.

TABLE III

IODINE CONTENT (μg per 100 g) OF MILK POWDER AND LIQUID MILK

Sample		Mean value of seven determinations	Standard deviation	Coefficient of variation, %
Ashed on day 1—				
Milk powder	1	149	9.0	6.0
	2	137	4.6	3.3
	3	145	7.2	5.0
	4	146	4.5	3.1
	5	146	7.3	5.0
Liquid milk	1	16.0	1.9	12.0
	2	14.9	2.1	14.2
	3	15.3	2.1	14.0
	4	17.1	1.6	9.2
	5	17.3	2.7	15.6
Ashed on day 2—				
Milk powder	6	147	5.6	3.8
	7	122	8.1	6.6
	8	134	7.3	5.4
	9	139	2.5	1.8
	10	153	4.5	3.0
Liquid milk	6	26.0	1.7	6.7
	7	15.9	1.6	9.9
	8	17.0	1.5	9.0
	9	18.9	1.5	7.7
	10	18.6	1.1	6.1

Recovery of Iodide Added to Food

During a survey of the iodine content of foods 400 ng of iodide were added to each sample and the recovery of iodide was measured. A representative summary of the recoveries obtained is given in Table V.

TABLE IV

VALUE FOR IODINE CALIBRATION GRAPHS RUN ON FIVE CONSECUTIVE DAYS

Iodine/ng ml^{-1}	20.0	16.0	12.0	8.0	4.0
Mean value	74.0	56.5	37.0	25.0	12.0
Standard deviation of 14 readings/ng ml^{-1}	3.4	2.6	2.4	1.7	1.1
Coefficient of variation, %	4.6	4.6	6.5	6.8	9.2

Some 163 samples of food were analysed with and without the addition of 400 ng of iodide, and the mean recovery was found to be 360 $\pm$ 70 ng of iodine (90 $\pm$ 19%). The high standard deviation is partly caused by the fact that the recoveries were calculated using the difference between two analyses.

TABLE V

REPRESENTATIVE VALUES OF THE RECOVERY OF 400 ng OF IODIDE ADDED TO VARIOUS FOODS

Type of sample	Number of samples examined	Range of iodine content/μg per 100 g	Mean recovery of 400 ng of iodide/ng	Standard deviation/ng
Butter, soup, margarine	8	16–49	370	50
Oxo cube, cheese, salad cream, fruit juice	7	11–58	380	60
Bacon, ham, gammon	12	6–17	420	60
Sausage, beefburger, cooking fat	7	3–7	360	40
Instant coffee, lamb, pork	4	2–9	350	20
Canned meat, cooking oil, fruit, flour	7	2–85	350	60
Fish fingers, fish	14	10–210	300	60

Effect of Storage on the Iodine Content of Foods

During survey work, it is not always possible to examine all of the samples immediately on receipt and so the effect of storage conditions, such as freeze drying and deep freezing, on the iodine contents of two foods was studied. Samples of egg and haddock were examined for iodine content when fresh, after freeze drying and after deep freezing at −15 °C for 1 week. The results, which are shown in Table VI, indicate that freeze drying does not greatly affect the iodine content of either eggs or haddock. However, it was subsequently found that freeze-dried liquid milk lost 20% of its iodine content, while freeze-dried standard iodide solution (200 ng ml^{-1}) lost 75% of its iodine content. When the standard solution was freeze dried after adding 1 ml of 30% potassium carbonate solution and 2 ml of 10% zinc sulphate solution there was no loss of iodide. Thus, it would seem that the degree of ionisation and the presence of a matrix affect the loss of iodine on freeze drying. The low values for deep-frozen haddock shown in Table VI were suspected to be due to the fact that when the fish thawed after deep freezing, some iodine would be present in the liquid produced. A piece of haddock was therefore deep frozen, allowed to thaw, and the liquid collected. The iodine content of this liquid was determined in addition to that of the fresh and thawed fish. The results, which are shown in Table VII, would appear largely to account for the value of deep frozen haddock shown in Table VI and also explain the losses of iodine on deep freezing fish reported by Gurevic.[21]

TABLE VI

EFFECT OF FREEZE DRYING AND DEEP FREEZING ON THE IODINE CONTENT OF EGG AND HADDOCK

Sample	Iodine content of fresh food/ μg per 100 g	Mean value/μg per 100 g	Standard deviation/μg per 100 g
Egg—			
Raw egg	50, 62, 59, 52	55	5
Freeze-dried egg	50, 30, 66, 61, 56, 62	54	13
Deep-frozen egg	56, 58, 56, 46	54	5
Haddock—			
Raw haddock	122, 122, 119, 132	124	6
Freeze-dried haddock	101, 120, 142, 106	117	18
Deep-frozen haddock	109, 115, 110, 116, 111	112	3

The higher standard deviations of the iodine levels for freeze-dried foods shown in Table VI probably reflect the fact that low amounts of sample were used to take account of the concentrating effect of freeze drying on nutrient levels.

TABLE VII

LOSS OF IODINE ON THAWING FROZEN FISH

Iodine content of piece of fresh haddock (35 g)	44 μg
Iodine content of same piece after deep freezing and then thawing	34 μg
Iodine content of the liquid collected on thawing	5 μg

Conclusion

The semi-automatic method, which is simple to operate and has an output of 70 samples per week, has been shown to be applicable to a large range of foodstuffs. It has a precision of 10% and an average recovery of added iodide of 90%, which are considered to be acceptable values for food analysis. A manual method on which the automated method is based requires very simple apparatus and is applicable when only a few samples have to be examined.

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