



U.S. ATOMIC ENERGY COMMISSION

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# REGULATORY GUIDE

DIRECTORATE OF REGULATORY STANDARDS

## REGULATORY GUIDE 4.3

### MEASUREMENTS OF RADIONUCLIDES IN THE ENVIRONMENT ANALYSIS OF I-131 IN MILK

#### A. INTRODUCTION

General Design Criterion 64, "Monitoring Radioactivity Releases," of Appendix A to 10 CFR Part 50, "General Design Criteria for Nuclear Power Plants," requires that means be provided for monitoring the plant environs of light-water-cooled nuclear power plants for radioactivity that may be released from normal operations, including anticipated operational occurrences, and from postulated accidents.

Paragraph (a)(2) of § 50.36a of 10 CFR Part 50 provides that technical specifications for each license will include a requirement that the licensee submit a report to the Commission within 60 days after January 1 and July 1 of each year which, in addition to specifying the quantity of each of the principal radionuclides released to unrestricted areas in liquid and airborne effluents during the last six months of operation, provides sufficient information to estimate annual radiation doses to the public resulting from effluent releases.

Paragraph (e) of § 20.106 of 10 CFR Part 20 provides that the Commission may limit the quantities of radioactive materials released in air or water by licensees during a specified period of time if it appears that the daily intake of radioactive materials from air, water, or food by a suitable sample of an exposed population group, averaged over a time period not exceeding one year, would otherwise exceed specified quantities. Section 20.201 of 10 CFR 20 requires that a licensee conduct surveys of levels of radiation or concentrations of radioactive material as necessary for compliance with AEC regulations in Part 20. Paragraph (c) of § 20.1 of 10 CFR Part 20 states that every reasonable effort should be made by AEC licensees to maintain radiation exposures, and releases of radioactive materials in effluents to unrestricted areas, as far below the limits specified in Part 20 as practicable, i.e., as low as is practicably achievable, taking into account the state of technology, and the economics of improvements in

relation to benefits to the public health and safety and in relation to the utilization of atomic energy in the public interest.

The procedure for the determination of I-131 in milk described in this guide constitutes a method acceptable to the Regulatory staff for measuring I-131 as may be required by the above regulations or orders or license conditions imposed pursuant thereto.

#### B. DISCUSSION

In accord with the Commission's policy of keeping exposure of man to radioactive materials released from licensed activities, including the operation of production or utilization facilities, as low as practicable, licensees should make every reasonable effort to limit the release of radioactive materials in effluents to unrestricted areas as far below the limits specified by the Commission as practicable. Iodine-131 requires special attention because it is one of the more significant radionuclides present in releases from nuclear power plants, and it can be concentrated through the air-vegetation-cow-milk pathway and potentially contribute to the thyroid dose. Although most milk data have indicated little or no I-131, milk monitoring at the suggested improved level of sensitivity would provide direct evidence that the potential exposure due to I-131 does not exceed predicted values.

Current guidelines<sup>1</sup> for I-131 dose to the thyroid of infants require that the sensitivity for measuring I-131 in milk be improved over conventional direct gamma counting of the raw milk sample. Low-level beta counting techniques with a detection sensitivity about an order of magnitude better than conventional gamma counting techniques can be used to attain the desired sensitivity. In the analysis of I-131 in milk, it may be

<sup>1</sup>Regulatory Guide 1.42, "Interim Licensing Policy on As Low As Practicable for Gaseous Radioiodine Releases from Light-Water-Cooled Nuclear Power Reactors."

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Published guides will be revised periodically, as appropriate, to accommodate comments and to reflect new information or experience.

Copies of published guides may be obtained by request indicating the divisions desired to the U.S. Atomic Energy Commission, Washington, D.C. 20545, Attention: Director of Regulatory Standards. Comments and suggestions for improvements in these guides are encouraged and should be sent to the Secretary of the Commission, U.S. Atomic Energy Commission, Washington, D.C. 20545, Attention: Chief, Public Proceedings Staff.

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necessary in some instances to measure I-131 in the presence of I-129<sup>2</sup> as well as other radioisotopes of iodine; since the usual beta counting methods are nonspecific, provisions for the resolution of such a potential mixture of isotopes should be included in the procedure. In most instances, however, radioisotopes of iodine other than I-131 and possibly I-129 are not expected to be of significance in the milk pathway because of their short half-lives. Also, under normal operating conditions of nuclear power plants, the contribution from I-129 will be negligible, the potential significance of I-129 being associated primarily with fuel reprocessing plants. It is probable, therefore, that any radioiodine detected in milk sampled from the environs of nuclear power plants will be primarily I-131. Nevertheless, because of the nonspecificity of beta counting methods, as a minimum, decay checks should be performed.

A simple check for isotopic purity can be performed by recounting the sample for radioactive decay. A half-time of 8 days would be a good indication that the activity is due to I-131. However, since it is not practical to attempt to monitor decay half-times when the sample net counting rate is less than about 0.3 cpm, it would be

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<sup>2</sup> A regulatory guide for the analysis of I-129 in milk is in preparation.

worthwhile to do this only when the net count is more than about 0.3 cpm. If, after the second count, it is determined that the activity is not decaying with a half-time of about 8 days, a third count made 4-5 days after the second count can provide the basis for a rough graphical resolution of I-131 activity. In any case, it would be desirable to hold the sample for at least a year to permit possible identification of the long-lived component should the need arise.

### C. REGULATORY POSITION

The procedure for the analysis of I-131 in milk described in the appendix to this guide is acceptable to the Regulatory staff as a basis for meeting analytical performance standards which are compatible with established guidelines.<sup>1</sup> Other procedures selected for analyzing I-131 in milk should provide levels of analytical sensitivity and specificity similar to those for the method described in the appendix.

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<sup>1</sup> See footnote on page 4.3-1



UNITED STATES  
NUCLEAR REGULATORY COMMISSION  
WASHINGTON, D. C. 20555

December 1, 1976

REGULATORY GUIDE DISTRIBUTION LIST (DIVISION 4)

The Nuclear Regulatory Commission staff has withdrawn Regulatory Guide 4.3, "Measurements of Radionuclides in the Environment, Analysis of I-131 in Milk." This guide was issued in September 1973 to describe one acceptable method for the determination of iodine-131 in milk which would meet the guidelines for iodine-131 dose to the thyroid of infants as given in Regulatory Guide 1.42, "Interim Licensing Policy on as Low as Practicable for Gaseous Radioiodine Releases from Light-Water-Cooled Nuclear Power Reactors." With the adoption of Appendix I to 10 CFR Part 50 as an effective rule and the adoption of a series of implementing guides, there was no longer a need for Regulatory Guide 1.42, and it was withdrawn. Since Regulatory Guide 4.3 was issued, a number of acceptable alternative methods and techniques for the determination of low levels of iodine-131 in milk have been demonstrated, including the analysis of milk containing preservatives and the use of beta-gamma coincidence counting techniques. Also since Regulatory Guide 4.3 was issued, there has been a recognition of the need to consider the effects of relatively high and variable concentrations of stable iodine in milk on the calculated radiochemical recovery and of the resultant need for determinations of stable iodine in milk. Because of the existence of acceptable alternative procedures, and the general recognition of the practicality of determining low levels of iodine-131 in milk, the detailed procedural guidance of Regulatory Guide 4.3 is no longer needed. Current licensing commitments based on Regulatory Guide 4.3 will not need to be reexamined as a result of this withdrawal.

Regulatory guides may be withdrawn when they are superseded by the Commission's regulations, when equivalent recommendations have been incorporated in applicable approved codes and standards, or when changes in methods and techniques have made them obsolete.

Sincerely,

A handwritten signature in cursive script, reading "Robert B. Minogue".

Robert B. Minogue, Director  
Office of Standards Development

## APPENDIX

### IODINE-131 ANALYSIS OF MILK AT CONCENTRATIONS DOWN TO 0.25 PICOCURIE PER LITER

The direct ion exchange method for separating iodine from raw milk requires that the iodine be in a readily exchangeable anionic form. Radioiodine tracer experiments on cows<sup>1, 2, 3</sup> have shown that 0-10 percent of the iodine in milk may be protein bound, rendering this fraction essentially unavailable for exchange with the ion exchange resin. However, this uncertainty of up to 10 percent in the I-131 determination will not significantly affect data interpretation and therefore is considered to be acceptable. Nevertheless, for the sake of accuracy, it is recommended that a fixed correction factor of 1.05 ( $1/0.95 = 1.05$ ) be applied to the counting data to compensate for this effect.

Also, there may be instances where milk samples may curdle to varying degrees in the interim between collection and analysis. Milk in this condition is unsuitable for analysis by the direct ion exchange method. A fresh sample should be obtained should this occur. If clabbering continues to be a problem, the ion exchange separation should be performed at the sample collection point and the ion exchange resin column shipped to the laboratory for processing. If this cannot be done, the milk sample should be frozen prior to shipment to the analytical laboratory.

The procedure described in this appendix was adapted from a procedure entitled "Determination of I-129 in Milk and Water Samples" developed by the Radiological Sciences Laboratory, Division of Laboratories and Research, New York State Department of Health. It was tested at the Battelle Memorial Institute Pacific Northwest Laboratories and shown to be adequate to measure I-131 in milk at the 0.25 picocurie per liter concentration level. This sensitivity is based on using a 4-liter milk sample and beta counting for 1000 minutes or longer in a low-background counter with a nominal background count rate of 0.5-1 count per minute (cpm). The analytical sensitivity can be further improved by using more than 4 liters of milk and counting for longer than 1000 minutes.

A recount for decay check should be performed 7-10 days after the first count. However, because it is not practical to do this for net counting rates of less than about 0.3 cpm, a recount should be made only when the initial net counting rate is greater than 0.3 cpm. If, after the second count, it is determined that the activity is not decaying with a half-life of 8 days, a third count should be made 4-5 days after the second count.

<sup>1</sup> Murthy, G. K., et al "Method for Removing Iodine-131 from Milk," *Journal of Dairy Science*, 45:1066 (1962)

<sup>2</sup> Salki, M., Omomo, Y., "Determination of Iodine-131 in Milk, Quick Methods for Radiochemical Analysis," Technical Reports Series No. 95, IAEA, 1969.

<sup>3</sup> Bretthauer, E. W., Mullen, A. L., Moghissi, A. A., "Milk Transfer Comparisons of Different Chemical Forms of Radioiodine," *Health Physics*, 22:257 (1972)

A graphical resolution of I-131 activity based on these three points may be performed. The sample should be held for at least a year to permit possible identification of the long-lived component should the need arise.

#### Abstract of the Procedure

Iodine-131 is removed from milk by concentrating the iodine on an anion exchange resin column and subsequently removing it from the resin by batch extraction using NaOCl. After reduction to I<sup>-2</sup> by hydroxylamine hydrochloride, the iodine is extracted into CCl<sub>4</sub>, reduced with bisulfite, and back extracted into water. The iodine is precipitated as palladous iodide. Chemical yield based on the added carrier iodine is determined gravimetrically. The I-131 concentration is determined by counting the palladous iodide precipitate in a low-background beta counter. The yield for the procedure is 70 to 85%.

#### Special Apparatus

1. Ion exchange column, 2 cm dia. by about 10 cm long with standard taper fittings
2. Four-liter reservoir with standard taper fitting
3. Assorted sizes of glass beakers
4. Hot plate
5. Magnetic stirrer
6. Suction filtering apparatus
7. 250-ml and 125-ml separatory funnels
8. Clinical centrifuge
9. Low-background beta counter (nominal background  $\leq 1$  count per min.)
10. Nylon rings and discs, Mylar, glass fiber paper (2.8 cm) (see HASL 300)

#### Reagents and Chemicals

1. Iodide carrier, 10 mg/ml - dissolve 1.181 grams NaI in 100 ml water
2. Dowex 1 x 8, 20-50 mesh, Cl<sup>-</sup> form
3. 2M NaCl - dissolve 116.9 grams NaCl in 1 liter water
4. NaOCl, 5-6% (commercial strength)
5. HNO<sub>3</sub>, concentrated
6. Hydroxylamine hydrochloride, crystals
7. CCl<sub>4</sub>
8. 1M NaHSO<sub>3</sub> - dissolve 1.041 grams of NaHSO<sub>3</sub> in 10 ml water (prepare daily)
9. Palladous chloride, 10 mg/ml - dissolve 1.66 grams of PdCl<sub>2</sub> in 100 ml of water

## Procedure

1. Prepare an ion exchange column 2 cm in diameter by 10 cm long and fill to a height of 5 cm with Dowex 1 x 8, 20-50 mesh,  $\text{Cl}^-$  form. Add the resin from a water slurry.
2. Add 1-2 ml of iodine carrier (10 mg I<sup>-</sup>/ml) to 4-liter milk sample and stir thoroughly. Accurately measure and record exact volume of carrier added. Pass through the column at a flow rate of about 30 ml per minute and discard effluent.
3. Wash the column with 500 ml hot (50°C) distilled water followed by 100 ml of 2M NaCl. Discard washes.
4. Transfer the resin to a 250-ml beaker using 50 ml of 5-6% NaOCl. (Note 1)
5. Place a small (~2 cm long) magnetic stirring bar in the beaker and stir vigorously for 5 minutes on a magnetic stirrer.
6. Filter the resin slurry through a suction filter and retain the NaOCl solution. (Note 2)
7. Reextract the resin by repeating steps 4, 5, and 6.
8. Discard the resin, combine the two 50-ml solutions, and carefully add 20 ml of conc.  $\text{HNO}_3$ . (Note 3)
9. Pour the acidified NaOCl solution into a 250-ml separatory funnel and add 50 ml of  $\text{CCl}_4$ .
10. Add 1.5 gm of hydroxylamine hydrochloride and shake (Note 4). Extract the iodine into the organic phase (about 2 min. equilibration). (Note 5)
11. Drain lower organic phase into a clean 250-ml separatory funnel and save.
12. Add 50 ml  $\text{CCl}_4$  and 1 gm hydroxylamine hydrochloride to the aqueous phase in the first separatory funnel and reextract. Combine organic phases and discard aqueous phase.
13. Add 25 ml  $\text{H}_2\text{O}$  and 10 drops of freshly prepared 1M  $\text{NaHSO}_3$  to the separatory funnel containing the combined  $\text{CCl}_4$  and shake. Equilibrate for 2 minutes. Discard organic (lower) phase. (Note 6)
14. Transfer the aqueous (upper) phase into a clean 50-ml centrifuge tube and add 1 ml of conc.  $\text{HNO}_3$  and 10 ml of  $\text{PdCl}_2$  solution. Stir and let stand for 5 minutes.
15. Centrifuge  $\text{PdI}_2$  precipitate discarding supernate. Wash precipitate by stirring with 10 ml of  $\text{H}_2\text{O}$ .
16. Using a filter funnel setup similar to that (Teflon or polyethylene - nylon) described in HASL 300, "Procedure Manual," USAEC, filter with suction through a tared glass fiber paper (2.8 cm diameter), using a water wash bottle to effect the transfer. (Note 7)
17. Dry precipitate for 20 minutes in an oven set at 110°C and weigh to the nearest 0.1 milligram. (Alternatively, the sample may be dried on a hot plate at low heat for about 30 minutes.)

18. Mount the precipitate on a nylon disc, cover with Mylar (1/4 mil thick), and fasten with ring. (This procedure is described in HASL 300.)
19. Count in a low-background counter for 1000 minutes.
20. If net counting rate of sample is greater than 0.3 cpm, recount after 7-8 days.
21. Calculate as picocuries I-131 per liter of milk at time of sampling.

## Notes

1. This is most easily accomplished by using a polyethylene squirt bottle to dispense the NaOCl.
2. Resin should be very light straw color after NaOCl extraction; if not light colored, the NaOCl is below strength, and a fresh solution should be obtained. Commercial grade Clorox has been found to be adequate.
3. Add the acid slowly with stirring until the vigorous reaction subsides. Perform in well ventilated hood.
4. Proceed with caution in this step. Excessive gas formation during the extraction can cause the stopcock or cap on the separatory funnel to "pop" with consequent loss of sample. Start by gently swirling the solution to effect mixing. Invert the separatory funnel with the stopcock pointing up and release the pressure by opening the stopcock. Close the stopcock, shake, and repeat the pressure release sequence.
5. Organic phase should be deep red; if not, add additional gram of  $\text{NH}_2\text{OH}\cdot\text{HCl}$ .
6. After back extraction into water,  $\text{CCl}_4$  should be colorless; if not add additional  $\text{NaHSO}_3$  and reextract.
7. HASL 300 "Procedure Manual" Health and Safety Laboratory, U.S. Atomic Energy Commission, 376 Hudson St., New York, New York 10014.

## Calibration and Standardization of Beta Counter

Counter standardization should be performed with I-131 standard obtained from the National Bureau of Standards (NBS) or with one which is relatable to NBS. An example of a relatable standard is a secondary or tertiary standard which has been calibrated against a primary NBS standard.

A self-absorption calibration curve should be constructed using at least 4 points. A useful curve can be constructed by plotting counter efficiency against mass. The counter efficiency can then be read off the curve as a function of the gravimetric yield.

### Example of Calculation of Results

The activity in picocuries per liter at the time the milk was collected is determined by calculations based on the observed net counts per minute of I-131. The calculation requires correction for chemical yield, protein-bound iodine, counter efficiency, decay since sampling, and the sample volume.

An example of a calculation of the I-131 concentration in a sample and the error (standard deviation) associated with the measurement follows:

$$\text{Chemical yield} = \frac{\text{Wt of ppt, mg}}{14.83^*}$$

\*The ppt is not stoichiometrically  $\text{PbI}_2$ , and this value is a determined empirical yield for 100% recovery; 14.83 mg is the determined weight of  $\text{PbI}_2$  for 10 mg of  $\text{I}^-$ .

1. Chemical yield	= 0.78
2. Counter efficiency	= 0.35
3. Counter background	= 0.6 cpm
4. Counting time	= 1000 minutes
5. Radioactive decay	= 8.05 days
6. Sample net counts	= 0.6 cpm (for a 1-half-life decay period)
Gross counts for 1000-min interval	= (background cpm + net cpm) (counting time) = (0.6 + 0.6)1000 = 1200
The net counts	= 1200 - 600 = 600 counts per 1000 minutes or 0.6 cpm
Standard deviation	= $\sqrt{\text{gross counts} + \text{background counts}}$ = $\sqrt{1200 + 600} = 42$
Net count with error limit	= $600 \pm 42$
Net pCi/liter	= $\frac{(\text{net counts}) (\text{factor for protein-bound fraction})}{(\text{count time})(\text{chem yield})(\text{counter eff})(\text{sample vol})(\text{decay})(\text{dpm/pCi})}$ = $\frac{(600 \pm 42) (1.05)}{(1000)(0.78)(0.35)(4)(0.50)(2.22)}$ = 0.5 pCi/liter with standard deviation of $\pm 7\%$