Field Sampling of Iodine 131 in Milk With Ion Exchange Cartridges

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SINCE iodine and many of its compounds are quite volatile, iodine 131 is one of the radioelements most likely to escape to the environment through normal discharges and accidents at nuclear reactor plants, through industrial accidents, and through the detonation of nuclear devices. In unseparated fresh fission products, iodine 131 is the critical nuclide causing the radiation dose which reaches man through the diet (*1*). For these reasons and because milk is the most significant dietary vehicle, the rapid determination of iodine 131 in this foodstuff is important.

One method for the rapid determination of iodine 131 in milk consists of adsorbing the iodine 131 on anion exchange resin followed by gamma counting (*2–4*). On the basis of this method, a field technique has been developed using a small cartridge, containing anion exchange resin, to separate radioiodine from milk (*5*). This technique only measures ionic forms of iodine, but approximately 95 percent of the iodine 131 in milk is present as the iodide. Also this resin has a very high capacity for adsorbing iodine 131. For example, approximately 160–200 resin bed volumes may be passed through the resin bed at a flow rate of up to 1 resin bed volume a minute and yield a total efficiency of 92 percent (personal comunication, Dr. G. K. Murthy, Robert A. Taft Sanitary Engineering Center, Cincinnati, Ohio). This paper describes the recent trial application of this technique in the field.

In addition, since some fluid milk samples were brought back to the laboratory for duplicate processing, the study permitted an investigation of the suitability of thimerosal (*6*) as a preservative for samples awaiting analysis. Previous experience showed that formaldehyde, the preservative used in samples collected under the Public Health Service's Pasteurized Milk Network (*7*), binds radioiodine to the milk protein and reduces its adsorption by anion exchange resin (*8*).

The October 1964 nuclear test on the Chinese mainland introduced fresh fission products, including iodine 131, into the atmosphere. This material, carried by winds and deposited on open pasture, led to contamination of milk supplies in several areas within the United States. The study was conducted in the vicinity of Burlington, Vt. Milk was sampled from the major dairy plants in that city. Samples were collected from bulk tank trucks arriving at milk processing plants and from silo tanks in the plants and passed through the cartridges as a test of the applicability of the cartridge technique to routine environmental monitoring. Dr. Harry L. Wildasin, Forest Norton, and Martin Lamson of the H. P. Hood Milk Company and Benjamin Barrows, dairy division, Vermont State Department of Agriculture, assisted in arranging for the collection of samples.

In the second phase, a more intensive program was conducted by sampling milk from individual cows on a farm with relatively high iodine 131 activity. This phase was designed to test the application to more detailed surveillance or

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research investigations. E. J. Baratta, K. C. Lamson, and P. R. Groulx of the Northeastern Radiological Health Laboratory, assisted in collecting and analyzing the samples.

Sample Collection and Analysis

The ion exchange cartridge was a modification of a model developed at the Southeastern Radiological Health Laboratory (*5*). The unit consists of a polyethylene vial containing approximately 40 milliliters of strongly basic quarternary amine type anion exchange resin—20 to 50 mesh, chloride form, suspended in distilled water. Milk is introduced through a separatory funnel attached to the top of the cartridge, passed through the resin bed and out through ten 2 millimeter diameter holes drilled in the bottom of the vial (fig. 1). Using the modified cartridge and the separatory funnel, a liter of fresh milk, the quantity used in this study, will pass through a cartridge in about 20 to 30 minutes.

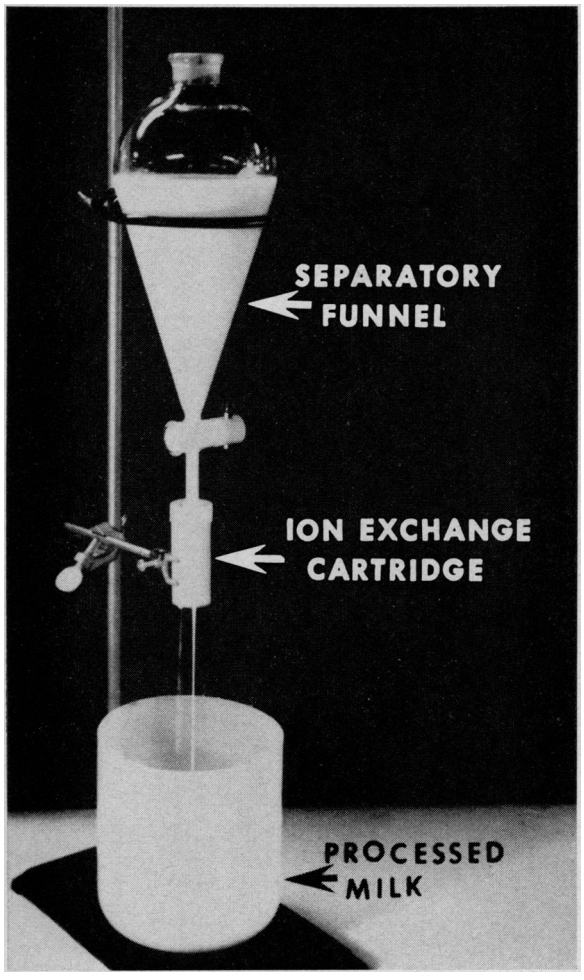


Figure 1. The cartridge as set up for field use

Although "spot" counting of some samples was performed in the field using a portable multichannel gamma spectrometer, all data reported in this paper were based on radioactivity determinations made on the resins in the laboratory. For the laboratory determination, the resin containing the adsorbed iodine 131 was emptied into a cottage cheese container and counted on a solid cylindrical 4-inch by 4-inch sodium iodide (thallium activated) crystal, coupled to a multichannel gamma spectrometer. Subsequent to this study, the northeastern laboratory staff developed a cartridge of smaller diameter (0.99 inches) which can be inserted directly into a well-type crystal for counting (fig. 2). The authors believe that this approach will have two advantages; the well-type crystal will yield a greater counting efficiency and the cartridges can be readily adapted to counting procedures using automatic sample changers.

Results

In the initial phase of this study, the ion exchange cartridge technique was applied to samples of milk from processing plants serving the area around Burlington, Vt. The iodine 131 levels observed in these samples are shown in table 1. The average concentration of this nuclide in the ten 1-liter samples, weighted according to the production of the given plant, was 38 picocuries per liter. The iodine 131 concentration was 31 picocuries per liter in the composite Pasteurized Milk Network sample, collected at the same time and location and analyzed in liquid form (3.5 liters) by gamma spectrometry (*2*). Since the counting error for each measurement was ± 10 picocuries per liter, the two results agree. This indicates that the ion exchange technique is reliable even under field conditions.

To evaluate whether thimerosal preservative would affect anion exchange processing of milk

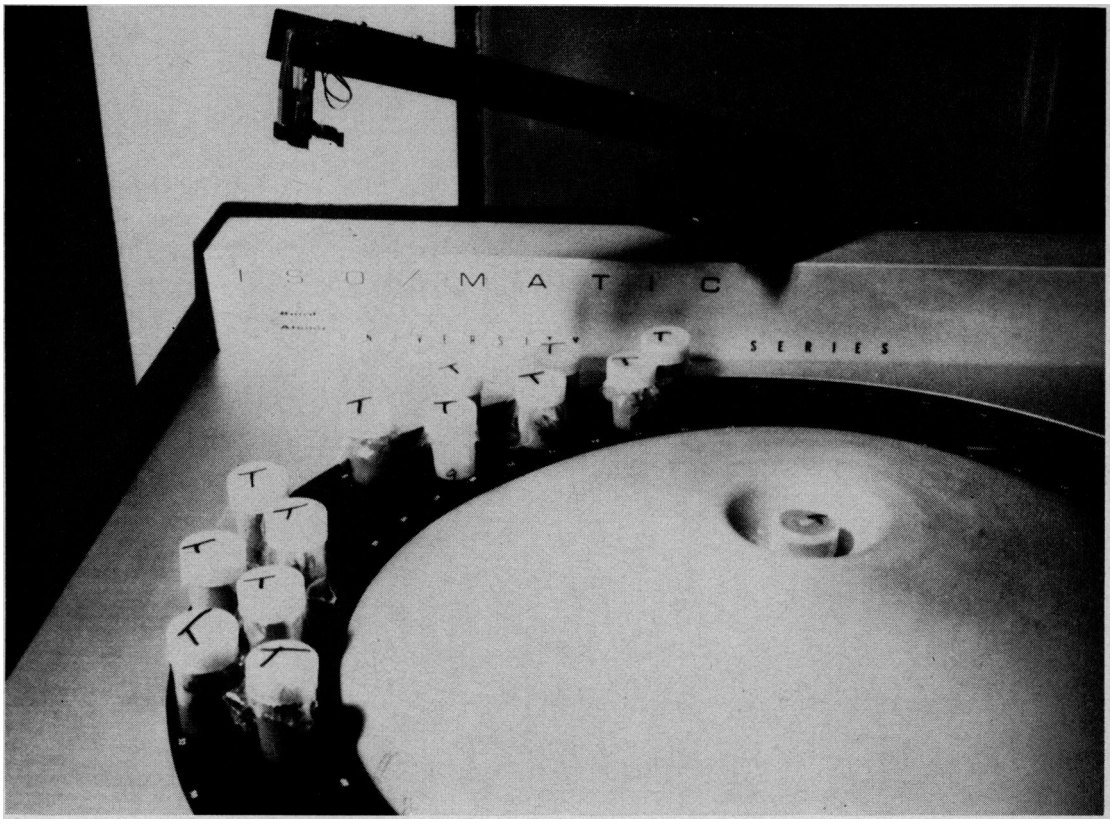


Figure 2. Well-type counting crystal with automatic sample changer

after storage, an additional series of samples were collected from three different tank trucks arriving at one of the larger plants. The preservative, 3 milliliters of a 10 percent aqueous solution of thimerosal per liter of milk, was added to 3 samples in the field. They were returned to the laboratory where 3.5-liter aliquots were analyzed for iodine 131 by gamma spectrometry; this method is not affected by type or quantity of preservative. Later, 1-liter volumes were passed through resin cartridges for analysis by the ion exchange technique. The average iodine 131 levels determined by these two methods are compared in table 2. Considering the counting error for each value, the two methods agree. This indicates that thimerosal is an effective preservative which does not impair the accuracy of iodine 131 measurements by the ion exchange technique.

For the second phase, milk samples were obtained from 36 cows at a dairy farm in Williston, Vt. One-liter portions of 23 samples were passed through ion exchange cartridges in the field. Additional 1-liter portions, some preserved with thimerosal and some not, were passed through similar cartridges 1 to 4 days later in the laboratory. All resins were counted under identical conditions. A summary of the data for the three sets of samples is shown in table 3.

Of interest in table 3 is the range of values of iodine 131 in the milk from the individual cows. All the cows were Holsteins and had been on open pasture until 3 days before sampling. The data show that the iodine 131 levels in the milk samples varied by a factor of 10. Two conditions possibly contributed to this wide variation: (*a*) the proportion of supplemental (stored) feed consumed by each cow while still on open pasture and (*b*) the fact that the biological half-life of iodine 131 is shorter than the 3 days the cows had been off pasture. For this reason, the precise length of time individual cows had been indoors and con-

suming stored feed before milk samples were collected could be important. No correlation could be found between the individual radionuclide levels and the age, time of lactation, or milk production of each cow.

A comparison between field and laboratory application of ion exchange cartridges is shown in figure 3. The correlation coefficient for these 12 samples was 0.76; however, all the data agreed within the counting error of about ±10 picocuries per liter. The least squares line indicates that the samples processed in the laboratory gave slightly higher iodine 131 concentrations. This may have been due to the temperature of the milk at the time of processing. The warm milk direct from the cow passed through the resin cartidge faster than the refrigerated milk in the laboratory.

Eleven samples were processed by ion exchange both with and without thimerosal preservative. All the samples were refrigerated during the 1- to 4-day processing period. The

Table 1. Concentration of iodine 131 determined by ion exchange technique and fluid gamma spectrometry in milk from processing plants, Burlington, Vt., November 3, 1964

Plant and technique	Concentration of iodine 131 (picocuries per liter)
Ion exchange (1 liter)	
A–1 [1]	57
A–2 [1]	39
A–3 [1]	30
B–1 [1]	33
B–2 [1]	29
C	18
D	21
E	48
F	14
G	19
Weighted average	38±10
Fluid gamma spectrometry (3.5 liters)	
Pasteurized Milk Network sample	31±10

[1] Samples from 3 silo tanks at plant A and 2 tanks at plant B.

Table 2. Comparison of analytical methods for iodine 131 in milk samples from tank trucks serving a milk processing plant, Burlington, Vt.

Time and date of collection	Concentration of iodine 131 (picocuries per liter)[1]	
	Ion exchange technique (1 liter)	Fluid gamma spectrometry (3.5 liters)
Evening, Nov. 3, 1964	43±10	32±10
Morning, Nov. 4, 1964	36±10	35±10
Evening, Nov. 4, 1964	50±10	37±10

[1] Analysis was performed at the laboratory from November 6–10, 1964.

Table 3. Concentration of iodine 131 in milk samples from 36 Holstein cows, Williston, Vt., November 5, 1964

Sample number	Concentration of iodine in picocuries per liter		
	Field determination (unpreserved)	Laboratory determination	
		Unpreserved	Preserved
1	42	73	
2	14		
3			14
4	25	49	
5	43	39	
6	44	47	
7			42
8			21
9	44	78	
10			24
11	47		
12			48
13	36		29
14		58	57
15	132		
16	61		
17	36		27
18	19		34
19		83	80
20			61
21	49		63
22	40		44
23			56
24	15		
25	117		
26	35		
27			10
28	74		
29	27	30	
30	35		
31		69	
32		26	24
33			36
34	65	70	
35	19		29
36	42		48
Composite			44

data for these samples are graphed in figure 4. A correlation coefficient of 0.90 indicates the close agreement in the values and confirms the acceptability of thimerosal as a milk preservative. Apparently, thimerosal does not produce any chemical changes sufficient to affect the measurement of iodine 131 by the ion exchange technique.

Discussion and Conclusions

The major advantages of the use of ion exchange cartridges are their simplicity, low cost, and the fact that all components are reusable. The iodine 131 can be stripped from the resin with 2*M* hydrochloric acid and the resin reconditioned with 2*M* sodium chloride (personal communication, Dr. G. K. Murthy, Robert A. Taft Sanitary Engineering Center, Cincinnati, Ohio). The resin can also be used 5 to 10 times without exceeding the exchange capacity after it is washed clean with water and the adsorbed iodine has decayed.

In contrast to the procedure followed in this study, the cartridges could be shipped in advance to a field location for use and them returned to a laboratory for counting. Since iodine is the only radioelement separated from

Figure 3. Relationship between observed iodine 131 concentrations as determined in the field and the laboratory

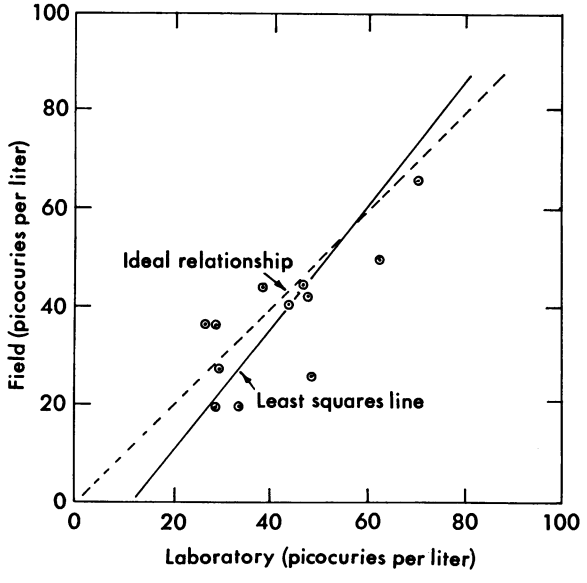
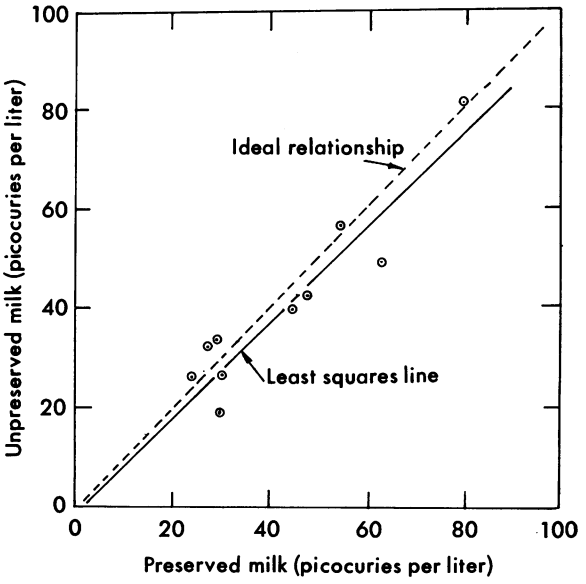


Figure 4. Relationship between observed iodine 131 concentrations in preserved and unpreserved milk



milk by anion resin, it would also be feasible to conduct entire studies of this type in the field. Under such conditions, counting could be done using a well-crystal scintillation detector and a suitable scaler. Or, at higher levels such as those contemplated under emergency conditions (*1*), the iodine 131 activity on the cartridges could be measured in the field with a simple Gieger-Müeller survey meter.

On the basis of this study, it can be concluded that the ion exchange cartridge is an effective means for field sampling of iodine 131 in fluid milk. Because of the simplicity of the method, it appears especially advantageous when it is necessary to collect large numbers of samples. This approach should be seriously considered by all public health personnel responsible for programs of environmental radiation surveillance.

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Surgeon General Stewart

DR. WILLIAM H. STEWART, director of the National Heart Institute since August 1, 1965, has been designated as the new Surgeon General of the Public Health Service. He succeeds Dr. Luther L. Terry, Surgeon General since 1961.

Until his present appointment, Dr. Stewart has held the rank of Assistant Surgeon General. He has been a commissioned officer in the Service since 1951. He has served as assistant to the special assistant to the Secretary (Health and Medical Affairs) of the Department of Health, Education, and Welfare.

His other appointments in the Public Health Service have been: head, Epidemiology Unit, Thomasville (Ga.) Field Station, Communicable Disease Center; Grants and Training Branch, National Heart Institute; chief, Heart Disease Control Program, Division of Special Health Services; assistant director, National Heart Institute; assistant to the Surgeon General; chief, Division of Public Health Methods; and chief, Division of Community Health Services.

Dr. Stewart was born in Minneapolis, Minn., in 1921. He attended the University of Minnesota and received his M.D. degree from Louisiana State University in 1945.

Following his internship at Philadelphia General Hospital, Dr. Stewart served in the U.S. Army Medical Corps and the Veterans Administration Outpatient Clinic in St. Paul, Minn. From 1948 to 1950 he was resident in pediatrics at Charity Hospital, New Orleans, La. He was in private practice in Alexandria, La., from 1950 until he joined the Public Health Service in 1951.

Dr. Stewart is a member of the American Heart Association, the American Medical Association, the American Public Health Association, and the American Academy of Pediatrics.