EVALUATION OF METHODS FOR THE ANALYSIS OF Sr-89 AND Sr-90 IN ENVIRONMENTAL SAMPLES

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I. INTRODUCTION

As a continuing effort to improve its services, Radiation Management Corporation has evaluated its procedure for Sr-89 and -90 on environmental samples. Three areas have been looked into specifically. These areas are:

1. Methods to increase the sensitivity of the analysis for certain sample types.

2. Methods to better differentiate, discrete Sr-89 and Sr-90 contributions to the total radio-strontium activity, and

3. Methods to improve the chemical yields for preserved milk samples.

A new analytical method is proposed to achieve the above objectives.

II. EVALUATION OF EXISTING PROCEDURE

Radiation Management Corporation's methodology for Sr-89/90 determination in environmental samples utilizes classic wet chemistry means to purify strontium from competing elements. A final strontium carbonate precipitate is prepared for beta counting. A second count of the prepared samples is done 7-14 days later. The Sr-89 and -90 activities are calculated from equations using Sr-89 decay and Y-90 ingrowth terms in a solution of 2 simultaneous equations with two unknowns. An evaluation was initiated to review the various aspects of the analytical method for Sr-89/90. The chemistry procedures used for Sr-89/90 analysis for various media were internally reviewed. In addition, an independent evaluation was conducted by our radiochemistry consultant, Dr. K.K.S. Pillay. As a result of these reviews, it was concluded that the radiochemistry methods were scientifically sound in chemical theory. Dr. Pillay was then requested to observe and evaluate the RMC technician in performance of the analyses, and also present a training seminar on the radiochemistry of strontium. From his technological review, Dr. Pillay found no outstanding technique problems, but he was able to provide some suggestions to handle certain specific conditions.

III. RESULTS OF QUALITATIVE AND QUANTITATIVE TESTING

In order to evaluate some of the proposed improvements to the analytical method, testing was performed to accomplish this goal.

TEST 1

The first testing to be performed was to make a more accurate determination of the chemical yield by means of a radioactive tracer Sr-85. A standard solution of Sr-85 was obtained to perform this test. Radioactive Sr-85, which is not a β^- emitter would be added along with stable strontium carrier so that the Sr-85 would act as a tracer for the chemical yield of the prepared samples. By knowing the amount of Sr-85 added, the chemical yield could be determined from a gamma count of each prepared sample to quantitate the Sr-85 remaining in the sample. The ratio of measured and actual Sr-85 amounts would be the chemical yield of the analysis. The chemical yield could thus be determined almost immediately for a prepared

sample. Before performing any actual tests for use of Sr-85 as a radioactive tracer, an aliquot of the Sr-85 standard was counted on the gas-flow proportional counter to determine the counting efficiency of the Sr-85 photons in the proportional counter. Since Sr-85 was going to be added to both samples and blanks, the Sr-85 should not be efficiently counted in the gas-flow proportional counter since the blank is used in the LLD calculation. One additional important concern for this initial test was an evaluation of the potential for the Sr-85 to become a contamination problem and thus have an adverse affect on other samples being prepared and counted. An activity of approximately 50 nCi was used and resulted in a beta counting efficiency of approximately 0.84% (890 cpm/ 105700 dpm). Since a quantity of about 10 nCi was to be added to each sample and blank to provide a suitable quick determination on the finished planchet, the activity of the sample would be masked and a higher LLD result. Approximately 100 cpm additional would result from the use of this amount of activity if a yield of approximately 60% was achieved. The use of Sr-85 was thus not feasible as a determination of yield for low-level determination of Sr-89 and -90. The more convential gravimetric or atomic absorption spectrophotometric methods will be utilized for chemical yield determinations for the strontium procedures.

TEST 2

The next test performed was an evaluation of the Sr-90 results for some previously analyzed samples by means of a separate determination for the Y-90 daughter activities of these samples. Since a condition of secular equilibrium exists in these samples, the Sr-90 activity can be inferred from the Y-90 determination. This test was not intended to make quantitative comparisons to the initial Sr-90 measurement, but was done to confirm the Sr-90 results. The following data in Table 1 show results from previous Sr-90 analyses (with some duplicates) and the Y-90 results.

TABLE 1

Sr-90 CONFIRMATION BY Y-90 COUNTING

SAMPLE #	Sr-90 RESULT(s)	Y-90 RESULT
1	$<2.36 \times 10^{-4}$	$<2.17 \times 10^{-4}$
2	$<2.75 \times 10^{-4}$	$<2.20 \times 10^{-4}$
3	$<3.8 \times 10^{-4}$	3.5×10^{-4}
4	$<4.1 \times 10^{-4}$	$<3.54 \times 10^{-4}$
5	14.9	10.7
6	<.572/<.53	<.449
7	<.62	<.51
8	<.51/<.43	<.46
9	<.50/<.43	<.52
10	<.67	<.58
11	2.06	2.34
12	4.2	<1.32

The testing showed that the initial Sr-90 results were in general agreement with the Y-90 result. The Sr-90 results was not verified in a few of the tests, but the method of analysis of Sr-90 by Y-90 (which RMC has not previously been doing) shows promise. To further test this the Y-90 analysis, a series of Y-90 known spikes were analyzed. Results of this testing appear in Table 2. Results show that the spike recoveries were in decent agreement with the known activities. We expect improved agreement

with future refinements in the technique.

TABLE 2

Y-90 SPIKE RECOVERIES

SAMPLE #	SPIKE ACTIVITY pCi/l	RECOVERED ACTIVITY pCi/l	% DIFFERENCE
1	10.2	10.9	+ 6.9
2	10.2	12.0	+ 17.7
3	10.2	12.7	+ 24.5
4	10.2	11.1	+ 8.8
5	20.4	23.2	+ 13.7
6	20.4	22.6	+ 10.8
7	20.4	22.8	+ 11.8
8	20.4	22.8	+ 11.8
		AVERAGE	+ 13.3%

TEST 3

One additional series of tests that was conducted was determination of any upward or downward trends of a repeated second count on prepared samples. A recount of the second count was done on approximately 57 samples. The count was done after Sr-90/Y-90 equilibrium was reached, but before any significant Sr-89 decay. A review of this data showed that the original second count and the repeat second count were not significantly different. This test was thus inconclusive to further improve the analysis or identify any particular problems in the present analysis.

TEST 4

Tests of a method to improve the chemical yield for preserved milk samples is currently being conducted. This test involves denaturation of a fluid milk sample with Trichloroacetic Acid (TCA) prior to performing the remainder of the analysis rather than performing the analysis directly on an ashed milk sample. Results of this test will be presented upon its completion.

IV. PROPOSED NEW METHOD FOR ANALYSIS OF Sr-89 AND Sr-90

Radiation Management Corporation's present procedure for analyzing Sr-89 and Sr-90 has been previously discussed in section II of this report. One major advantage of this method was to eliminate the need for a second chemical separation for Yttrium. One apparent disadvantage is that the method yields higher calculated LLD values than a two separation method since the LLDs are calculated from the two sequential counts rather than individually from single counts performed for each isotope.

Radiation Management Corporation thus investigated alternative calculational methods for the determination of Sr-89 and -90 using a more classic two separation chemical method rather than the simultaneous equation method currently in use. Two principle methods for determining the quantitative Sr-89 and -90 contributions to the total radiostrontium activity in a sample are in popular use. The Sr-89 activity can be determined by counting a freshly prepared strontium sample through an absorber to shield the Sr-90 soft beta and then determing the Sr-90 by Y-90 separation. A second method is to determine the Sr-89 activity by subtraction from the total radiostrontium

activity by first measuring the Sr-90 (by Y-90). The prepared method for the Sr-89/90 analyses will utilize a technique based upon the absorbance of the soft Sr-90 beta to determine Sr-89 activity and a separate Y-90 determination to infer Sr-90 activity.

The Beckmen Wide Beta II gas-flow proportional counters can be equipped with absorbers of different thicknesses for beta counting. An absorber of 90.6 mg/cm^2 thickness was chosen since it reduced the Sr-90 counting efficiency to <1% (44 cpm/4575 dpm) and allowed a Sr-89 counting efficiency of approximately 12.1% (201 cpm/1665 dpm). The use of an absorber should thus be suitable for the measurements to be done.

V. ACHIEVABLE SENSITIVITIES FOR SELECTED SAMPLE TYPES

Typical sensitivities achievable by this method have been calculated for selected sample types. The equation for LLD as described in Regulatory Guide 4.8 has been used with suitable additional terms to correct for Y-90 ingrowth and the second chemical separation required from the Sr-90 analysis. The sample media addressed include water, milk and air particulates. Real data has been used for the variables such as chemical yield and background count rates. Prepared Y-90 samples will be counted within 24 hours to minimize its decay term. Prepared strontium samples will also be counted within 24 hours to minimize Y-90 ingrowth.

Milk

Sr-90 Achievable Sensitivity (LLD): 1 pCi/l

$$LLD_{90} = \frac{4.66\sqrt{B}}{t \cdot V \cdot Ys \cdot Yy \cdot Ey \cdot 2.22 \cdot IF \cdot DF}$$

Where: B = Background counts = 1 cpm x 200 min = 200
t = Counting time = 200 min
V = Sample volume = 1.5 liters
Ys = Strontium chemical yield ≅ .33
Yy = Yttrium chemical yield ≅ .85
Ey = Counter efficiency for Y-90 ≅ .45
2.22 = dpm/pCi (constant)
IF = Y-90 ingrowth factor (time from initial Sr separation to Y separation) = $1 - e^{-\lambda t}$ = .974 for 14 days
DF = Y-90 decay factor (time from Y separation to counting) = $e^{-\lambda t}$ = .878 for 12 hours

$$LLD_{90} = \frac{4.66\sqrt{200}}{(200)\ (1.5)\ (.33)\ (.85)\ (.45)\ (2.22)\ (.974)\ (.878)}$$

≅ 0.92 pCi/liter

Sr-89 Achievable Sensitivity (LLD): 5 pCi/l

$$LLD_{89} = \frac{4.66\sqrt{B}}{t \cdot Es \cdot Ys \cdot V \cdot 2.22 \cdot DF}$$

Where: B = Background counts = 1 cpm x 200 min = 200
t = Counting time = 200 minutes
Ys = Strontium chemical yield = .33
2.22 = Constant
V = Volume = 1.5 liters
Es = Counter efficiency for Sr-89 = .121
DF = Sr-89 decay factor (assumes approximately 30 days between collection and counting) = $e^{-\lambda t}$ = .674

$$LLD_{89} = \frac{4.66\sqrt{200}}{(200)\ (.121)\ (.33)\ (1.5)\ (2.22)\ (.674)}$$

$\cong$ 3.7 pCi/liter

Water

Sr-90 Achievable Sensitivity (LLD): 1 pCi/l

$$LLD_{90} = \frac{4.66\sqrt{B}}{t \cdot V \cdot Ys \cdot Yy \cdot Ey \cdot 2.22 \cdot IF \cdot DF}$$

Where: B = Background counts = 1 cpm x 100 min = 100 counts
t = Counting time = 100 minutes
V = Sample volume = 2 liters
Ys = Strontium chemical yield $\cong$ .80
and all other terms are same as in the Sr-90 milk determination.

$$LLD_{90} = \frac{4.66\sqrt{100}}{(100)\ (2.0)\ (.80)\ (.85)\ (.45)\ (2.22)\ (.974)\ (.878)}$$

$\cong$ 0.40 pCi/liter

Sr-89 Achievable Sensitivity (LLD): 1 pCi/liter

$$LLD_{89} = \frac{4.66\sqrt{B}}{t \cdot Es \cdot Ys \cdot V \cdot 2.22 \cdot DF}$$

Where: Ys = Strontium chemical yield $\cong$ .80
V = Sample volume = 2.0 liters
DF = Sr-89 decay factor for 14 days = .832
and all other terms are the same as in the Sr-89 in milk determination

$$LLD_{89} = \frac{4.66\sqrt{200}}{(200)\ (.121)\ (.80)\ (2.0)\ (2.22)\ (.832)}$$

$\cong$ 0.92 pCi/l

Air Particulates

Sr-90 Achievable Sensitivity (LLD): 1 x 10^{-4} pCi/m^3

$$LLD_{90} = \frac{4.66\sqrt{B}}{t \cdot V \cdot Ys \cdot Yy \cdot Ey \cdot 2.22 \cdot IF \cdot DF}$$

Where: B^* = (.7 cpm) (750 min) = 525 counts
t = 750 minutes
$V \cong 2500\ m^3$
and all other terms are the same as Sr-90 analysis in water
*Low Background Counter Used

$$LLD_{90} = \frac{4.66\sqrt{525}}{(750)\ (2500)\ (.8)\ (.85)\ (.45)\ (2.22)\ (.974)\ (.878)}$$

$$\cong 0.98 \times 10^{-4}\ pCi/m^3$$

Activity calculations for Sr-89 and -90 would be as follows:

$$\text{Sr-90 (pCi/unit volume) Activity} = \frac{\dot{G} - \dot{B}}{V \cdot Ys \cdot Yy \cdot Ey \cdot 2.22 \cdot IF \cdot DF}$$

Where: the terms are identical to those in the Sr-90 LLD equations and
$\dot{G}$ = Gross count rate
$\dot{B}$ = Background count rate

$$\text{Sr-89 (pCi/unit volume)} = \frac{\dot{G} - \dot{B} - Bs - By}{V \cdot Ys \cdot Ey \cdot 2.22 \cdot DF}$$

Where: the terms are identical to those in the Sr-89 LLD equations and
$\dot{G}$ = Gross count rate
$\dot{B}$ = Background count rate (instrument)
Bs = Additional background count rate from fractional Sr-90 counting
By = Additional background count rate from Y-90 ingrowth

VI. CONCLUSIONS

1. RMC has evaluated its current chemistry procedures and have determined them to be scientifically sound.

2. RMC proposes to change the analytical method in the following manner:
   a. A two separation chemical procedure will be performed in lieu of the current single separation method.
   b. An absorber will be utilized in the determination of Sr-89
   c. A Y-90 separation will be performed for the determination of Sr-90

3. Achievable sensitivities will meet client requirements.

	Sr-90	Sr-89
Milk	0.92 pCi/l	3.7 pCi/l
Water	0.57 pCi/l	0.92 pCi/l
Air Particulates	0.98×10^{-4} pCi/m^3	

4. Although the proposed methods will achieve the above sensitivities, more testing (e.g. TCA usage in the milk procedure to improve the chemical yield) is currently being conducted and will be reported upon completion.

Radiation Management Corporation will conduct some additional testing as necessary to make further refinements in its analytical procedures.