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Detection Capability for Determination of ^{89}Sr and ^{90}Sr by Liquid Scintillation Counting

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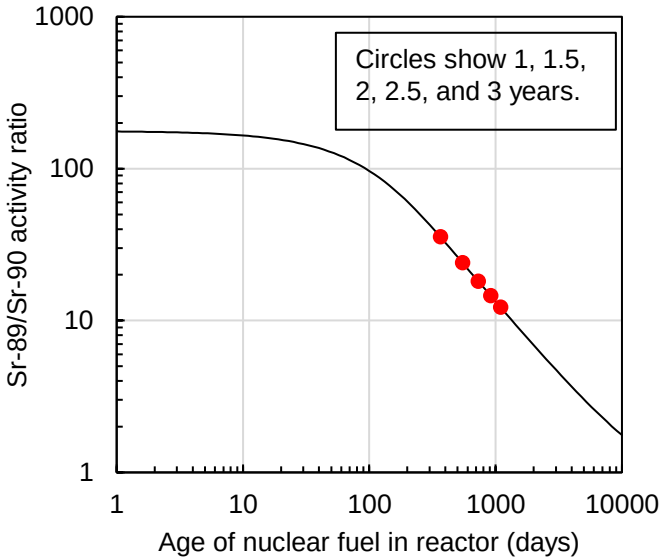
Overview

A method has been developed for detection of ^{89}Sr and ^{90}Sr on liquid scintillation counting that can be used to:

- Analyze up to 50 g of food and 0.1-1 L of water in 2 days, with additional time for evaporating > 1 L of water
- Detect ^{89}Sr activity if at least 1/20 that of ^{90}Sr
- Detect ^{90}Sr activity if at least 1/10 that of ^{89}Sr



Outline

- **Motivation**
- **Experimental Setup**
- **Data Analysis**
- **Results**
- **Conclusions**

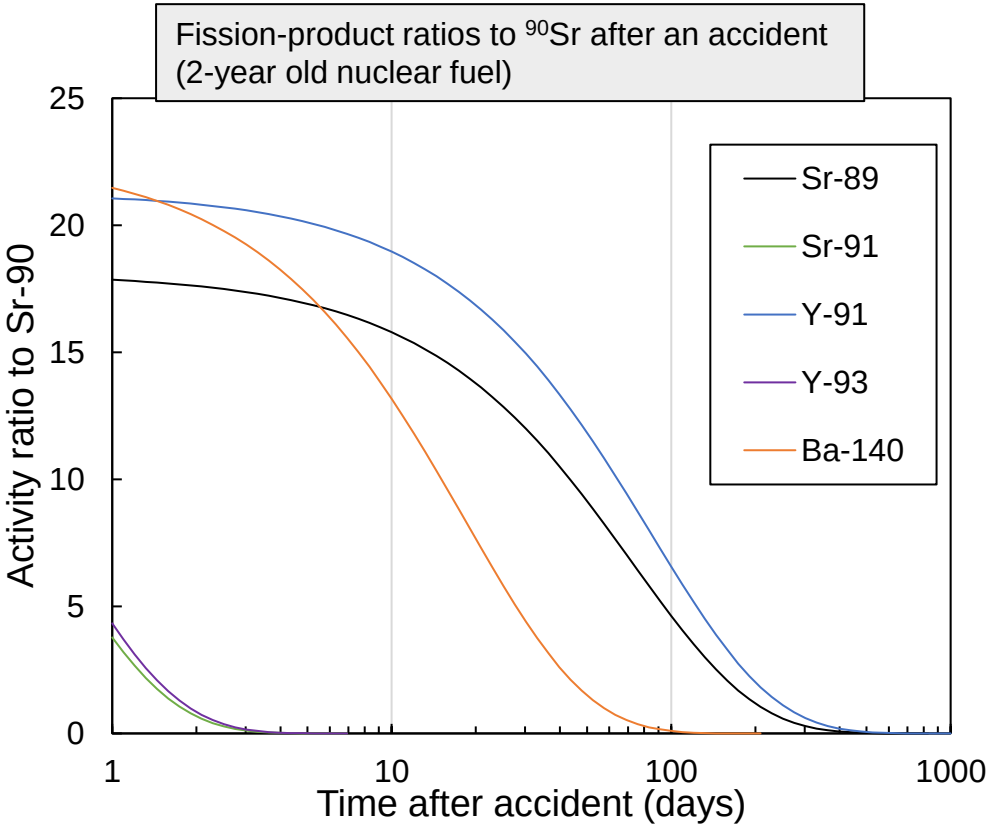
Motivation: Health Hazards of Strontium

- ^{90}Sr isotopes and the decay product ^{90}Y are all beta emitters
- Particles released to environment can be consumed through water, milk and food
- Ingestion leads to internal dose, more difficult to control than external dose
- Strontium is a bone seeker depositing in bone and not being released from body
- ^{90}Sr major health concern due to long half-life
- Animal studies show bone sarcoma from radiostrontium exposure
- Current methods utilize ingrowth of ^{90}Y and can take several weeks

Motivation: Historical Data

Mass number	Isotope	$T_{1/2}$	Decay mode	β_{max} (keV)	<β> (keV)	Chernobyl initial activity[a] (MCi)	Chernobyl released activity[a] (MCi)	U235T chain yield[b] (%)
85	^{85}Sr	64.8 d	EC	—	8.4	—	—	—
89	^{89}Sr	50.6 d	β^-	1492	583	81	2.8	4.99
90	^{90}Sr	28.9 y	β^-	546	196	4.6	0.18	5.89
	↓							
	^{90}Y	64.1 h	β^-	2281	934	4.7	—	
91	^{91}Sr	9.65 h	βγ	2686	648	60	—	5.84
	↓							
	^{91}Y	58.5 d	β^-	1546	603	104	—	
93	^{93}Y	10.2 h	βγ	2884	1171	—	—	6.12
140	^{140}Ba	12.8 d	βγ	1034	276	146	4.7	6.25
	↓							
	^{140}La	1.68 d	βγ	3761	525	155	—	

a: DOE (1987) Health and Environmental Consequences of the Chernobyl Nuclear Power Plant Accident. US Department of Energy Report DOE/ER-0332, Washington, DC, June.
b: Wahl, A.C. (1988) Nuclear-charge distribution and delayed-neutron yields for thermal-neutron-induced fission of 235U, 233U, and 239Pu and for spontaneous fission of 252Cf. At. Data Nucl. Data Tables 39, 1-160.



Tovedal, A., Nygren, U., Ramebäck, H. J. (2009) Radioanal. Nucl. Chem. 282, 455-459.
Tovedal, A., Nygren, U., Lagerkvist, P., Vesterlund, A., Ramebäck, H. (2009) J. Radioanal. Nucl. Chem. 282, 461-466.
Holmgren, S., Tovedal, A., Jonsson S., Nygren, U., Ramebäck, H. (2014). Appl. Radiat. Isot. 90, 94-101.
Zhu, S., Ghods, A., Veselsky, J.C., Mirna, A., Schelenz, R. (1990) Radiochim. Acta 51, 195-198.

Method Summary

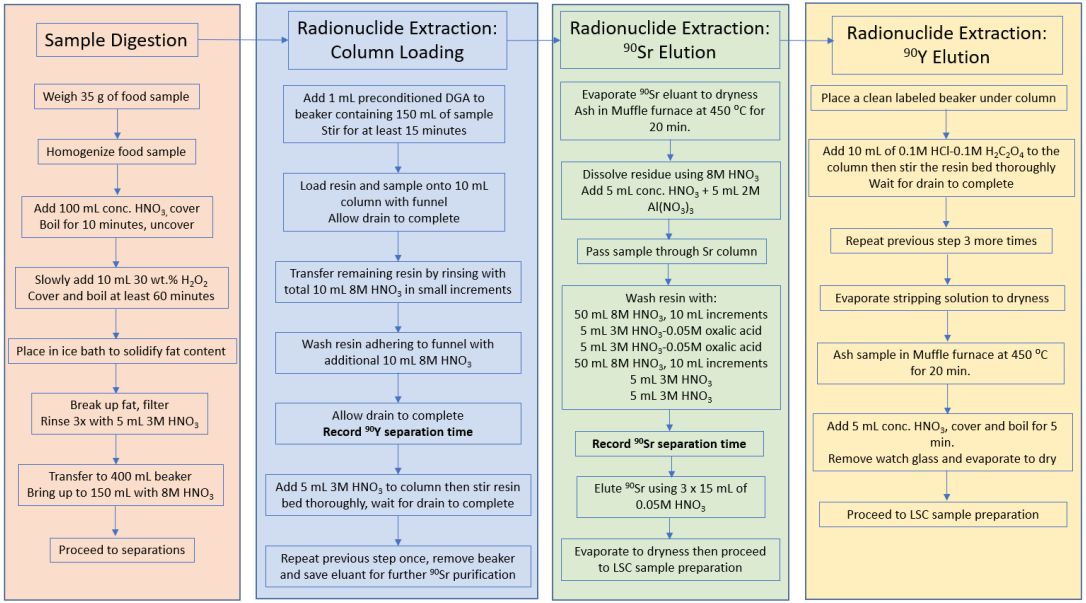
Chemistry: Acid digestion and column chromatography using Eichrom Sr resin, with Sr recovery using gamma counting of ^{85}Sr tracer.

Calibration: Standard calibrations were performed for ^{85}Sr and ^{89}Sr. ^{90}Sr and ^{90}Y calibration was performed by following an ingrowth of ^{90}Y in a freshly prepared ^{90}Sr fraction.

Counting: Single count on a 3-window LSC.

Analysis: Activity equations are solved numerically, with covariances used for uncertainty and detection-limit calculations.

Experimental Setup: Chemical Separations



Experimental Setup: Sample Digestion

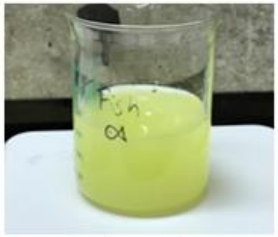
Acid Digestion

35 g samples of food were acid digested with concentrated HNO_3 and 30% H_2O_2

Filtration

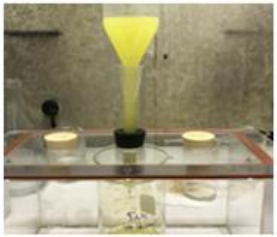
Remaining fat content was skimmed and filtered through a Whatman 1 filter

Resin Contact



1 g of preconditioned DGA resin was added to sample and stirred for at least 15 minutes

Separation



Sample was added to column, eluent collected for further analysis

Experimental Setup: Hidex Liquid Scintillator

- Hidex SL600 Detector Used
- Detector utilizes triple coincidence to reduce low level noise
- Additional muon guard installed to further reduce cosmic background
- Internal 152Eu source used to quantify quench parameter QPE
- External chiller ensures operation at 20 degrees C

https://lablogic.com/life-sciences/instruments/hidex-600-sl?region=us#overview

Experimental Setup: Determination of Recovery

- Chemical recovery is important for accurate determination of activity
- ^{85}Sr used as a recovery tracer
- ^{85}Sr decays through electron capture and emits 514 keV gamma ray
- Gamma spectroscopy on 514 keV gamma used to determine chemical recovery
- Measurement samples are compared to pure tracer samples and ratio used to find chemical recovery
- Recoveries ranged from approximately 80-95%

Experimental Setup: 3 Window Analysis

For data analysis, 3 windows were chosen to highlight the counts from the three separate isotopes

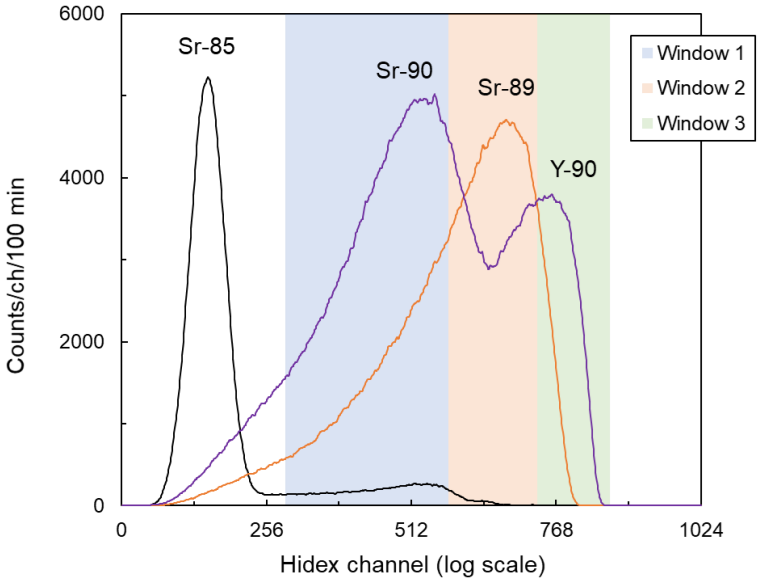
The given count windows are:

- Window 1 (^{90}Sr): Channel 289-576
- Window 2 (^{89}Sr): Channel 577-734
- Window 3 (^{90}Y): Channel 735-864

The recovery tracer ^{85}Sr is mostly outside of these windows

Corrections for the additional ^{85}Sr background in windows 1 and 2 are applied in the full analysis

Since the locations of these windows are constant, accurate determination of detector efficiency is vitally important for this method



Experimental Setup: Detector Calibration

To accurately determine the activity of unknown samples the detector efficiency must be measured

The efficiency for detection will be different for each isotope and for each window

To determine the efficiency for ^{85}Sr and ^{89}Sr is relatively straightforward

- A sample of known activity is counted in the LSC for a given period of time
- The measured count rate is compared to the known activity of the sample and the conversion between the two will yield the efficiency
- Three samples are measured, and the average of the samples is taken as the efficiency with the uncertainty being the measured standard deviation

Experimental Setup: Detector Calibration

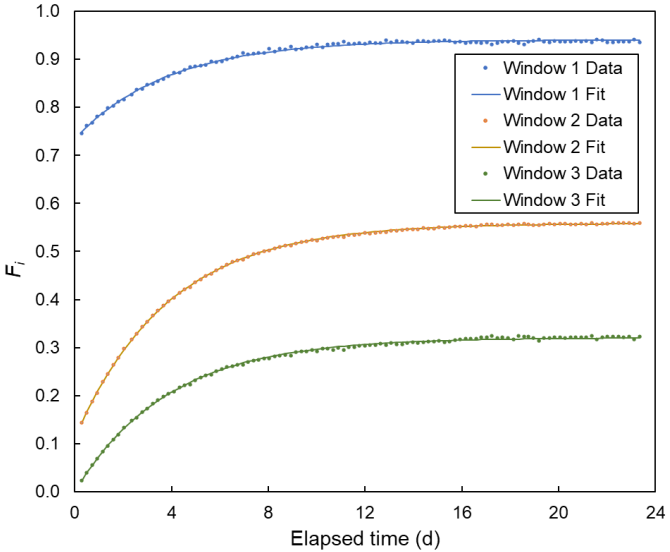
Determining the efficiency of the ^{90}Sr and ^{90}Y is more complicated due to the decay process

To determine these efficiencies the counts as a function of time were plotted following the separation of ^{90}Sr from a given sample

The ingrowth of ^{90}Y can be seen in the plot for all three counting windows

A χ^2 fitting of the data yielded the efficiency values for Sr and Y in the three windows

The agreement between the fitted curve and the data points shows very good agreement for the fitted efficiencies



Data Analysis: Analysis Equations

To determine the activity of the sample we must take into account the efficiency of the detector and the radioactive buildup of the sample

To solve for all isotopes simultaneously we will utilize a matrix formalism

We define our efficiency matrix $\mathbb{E}$ as follows where ε_{ij} is the efficiency for isotope *i* in window *j*:

$$\mathbb{E} = \begin{pmatrix} \varepsilon_{11} & \varepsilon_{12} & \varepsilon_{13} \\ \varepsilon_{21} & \varepsilon_{22} & \varepsilon_{23} \\ \varepsilon_{31} & \varepsilon_{32} & \varepsilon_{33} \end{pmatrix}$$

We define our buildup matrix as:

$$\mathbb{P} = \begin{pmatrix} 1 & 0 & 0 \\ \frac{\lambda_2}{\lambda_2 - \lambda_1} & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad \mathbb{P}^{-1} = \begin{pmatrix} 1 & 0 & 0 \\ -\frac{\lambda_2}{\lambda_2 - \lambda_1} & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}$$

We can then define a new matrix $\mathbb{G}$ where:

$$\mathbb{G} = \mathbb{E}\mathbb{P}\mathrm{diag}\left(e^{-\lambda_i t}\right)\mathbb{P}^{-1}$$

Here t is the elapsed time from separation

We can solve for the activity based on measured counts using the following equation. This will yield the activity at separation time

$$\boldsymbol{C} = \mathbb{G}\boldsymbol{A}$$

Data Analysis: Analysis Equations

Once the activity at separation time is determined the activity at sampling time can be found

This will take into account the chemical recovery of the sample as well as the quantity and decay from sampling to separation time

The separation matrix is as follows:

$$\mathbb{S} = \begin{pmatrix} S_1 & 0 & 0 \\ 0 & S_2 & 0 \\ 0 & 0 & S_3 \end{pmatrix} = S_S \mathbb{W}$$

Where:

$$\mathbb{W} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & f_s & 0 \\ 0 & 0 & 1 \end{pmatrix}, \qquad \mathbb{W}^{-1} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1/f_s & 0 \\ 0 & 0 & 1 \end{pmatrix}$$

The value f_s is the recovery ratio of ^{90}Sr to ^{90}Y and S_S is the recovery of ^{90}Sr

The activity at sampling time can be found using the following where Q is the sample quantity and f_c is the conversion from dpm to activity reporting units

$$\boldsymbol{A_r} = \frac{1}{Q S_S f_c} \mathbb{P}\mathrm{diag}\left(e^{\lambda_i t_r}\right) \mathbb{P}^{-1} \mathbb{W}^{-1} \boldsymbol{A}$$

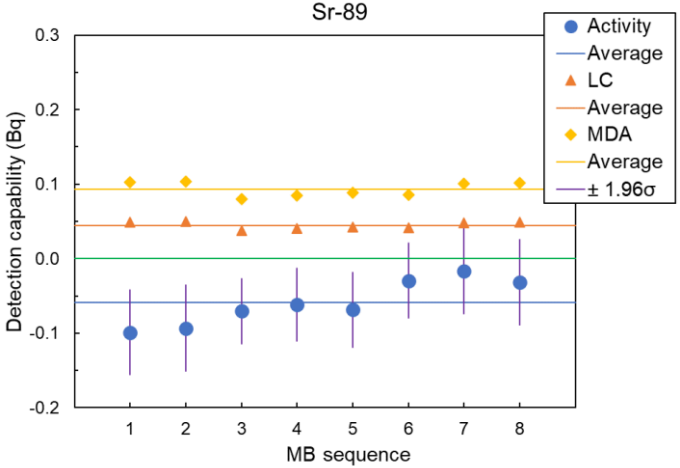
Results: Method Blank Results

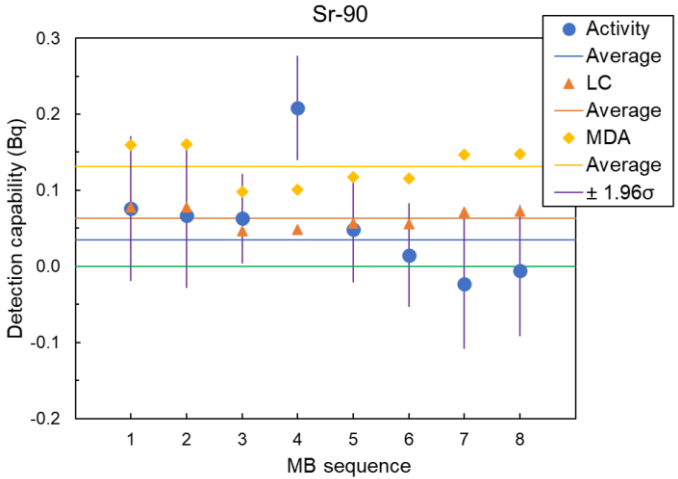
8 Method blanks (MB) were measured over the 7 month span of the project

The higher count rate for MB 4 in the ^{90}Sr was attributed to contamination in the MB sample

While there is a slight negative bias to the background results for ^{89}Sr all values are relatively low with only one value yielding a false positive result

The Decision Level (LC) and Minimum Detectable Activity (MDA) are calculated for the MB samples





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Results: Spiked Sample Results

Samples containing a range of $^{89}Sr/^{90}Sr$ activity ratios were measured to determine the effectiveness of the method

Ratios included: pure ^{90}Sr, pure ^{89}Sr and a 10:1, 5:1, 2:1, 1:1, 0.5:1, 0.2:1 and 0.1:1 ratio of $^{89}Sr/^{90}Sr$

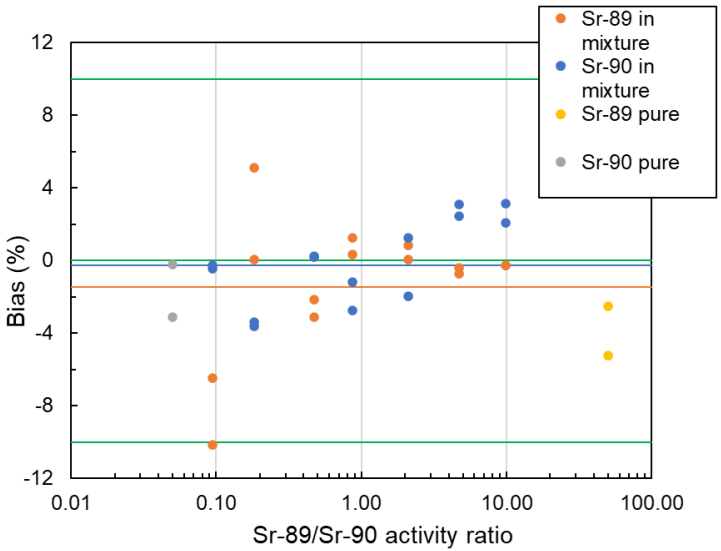
The results on the right show the measurement bias at different activity ratios

Most of the datapoints are within 5% bias showing a very good agreement for the method

Each measurement has two corresponding datapoints for two replicate samples

The orange and blue lines correspond to the average bias of the ^{89}Sr and ^{90}Sr respectively

The green lines are used to highlight the +/- 10% bias region



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Detection Limits

Average Decision Level (L_C), MDA, and SDWA DL (no interference)

Radio-nuclide	L_C (Bq)		MDA (Bq)		SDWA DL (Bq)	
	MB	Spiked	MB	Spiked	MB	Spiked
^{89}Sr	4.477E-02	4.928E-02	9.342E-02	1.025E-01	4.065E-02	4.369E-02
^{90}Sr	6.350E-02	6.931E-02	1.308E-01	1.418E-01	4.792E-02	5.137E-02

SDWA DL and regulatory limits for drinking water

Drinking water		Measured	SDWA	
Sample quantity (L)	Radionuclide	DL (pCi/L)	DL (pCi/L)	MCL[a] (pCi/L)
1	^{89}Sr	1.2	10	—
	^{90}Sr	1.4	2	8

Footnotes: [a]Maximum Contaminant Level

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Conclusions

A method has been developed to measure radiostrontium using a 3 window counting method on LSC

This method has a quick measurement time compared to other radiostrontium counting methods particularly those relying on yttrium counting

Detection limits have been calculated and show adequate detection of radiostrontium based on food requirements by the FDA

The method has been shown to work well on a variety of strontium ratios indicating its usefulness in measuring several different types of samples

This method can be very useful for measurement of radiostrontium following a nuclear plant accident or radioactive contamination event

Acknowledgements

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Questions?