Use of liquid scintillation counting for fast determination of ^{89}Sr and ^{90}Sr in milk.

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Introduction

^{90}Sr ($t_{1/2}$ = 28,5 Jahre) is one of the radiologically most relevant radionuclides occuring in nuclear fallout.[1,2] Fresh fallout, however, also contains ^{89}Sr ($t_{1/2}$ = 50,5 Tage) at activities considerably greater than that of ^{90}Sr.[3] If an area is contaminated with fresh fallout, therefore, the activities of both of these nuclides must be determined in various media including foodstuffs. Milk is a particularly important medium for several reasons: The Sr isotopes migrate efficiently through the food chain "soil-plant-cow-milk" and accumulate especially in the bone cells of humans; milk is usually consumed as a fresh product; the level of contamination of the milk is an indication of the radiostrontium deposition over a wide area.

^{89}Sr and ^{90}Sr are not readily determined in the presence of each other because both are pure b-emitters. The end point b-energies are 1492 KeV and 546 KeV for ^{89}Sr and ^{90}Sr.[4], respectively. While ^{89}Sr decays to an inactive product, ^{90}Sr decays to ^{90}Y, which is also a pure β-emitter with an endpoint energy of 2284 KeV.[4] Since ^{90}Y has a $t_{1/2}$ of 64,1 hours, this mother-daughter pair approaches secular equilibrium in about 14-17 days, i.e., after this time, an initially pure ^{90}Sr sample will contain an equal activity of ^{90}Y. This effect is used to estimate the ^{90}Sr content of mixtures of ^{89}Sr and ^{90}Sr: When secular equilibrium is attained, the ^{90}Y is separated, measured and the activity determined. The ^{89}Sr activity is then calculated by subtracting this ^{90}Y activity (= ^{90}Sr activity) from that of the initial $^{89}Sr/^{90}Sr$ mixture measured before significant amounts of ^{90}Y have formed.

Sr preparations suitable for such measurements must be free of the organic constituents of the milk; the inorganic cations Na^+, K^+, Mg^{2+}, Ca^{2+} (present in g/l concentrations), Fe und Zn (in mg/l concentrations)[5]; the naturally occurring radionuclides such as ^{40}K, ^{89}Rb, ^{14}C and ^{3}H; as well as other fallout nuclides that are transferred to milk, i. e., various isotopes of I, Te, Cs and ^{140}Ba with its daughter ^{140}La.[6] The traditional separation scheme of Harley, still the most frequently used method[4,7], fulfills all of these conditions. It involves drying and ashing the milk, a procedure requiring several days, then carrying out a lengthy series of precipitation reactions on a solution of the ash. There are many variations of this method in which the time consuming drying and ashing procedure is replaced by ion exchange techniques typically with Dowex-type strong cation exchange resins,[8-10] However, rather concentrated solutions of strong acids are required to elute the Sr. Elution is seldom quantitative. Barrata and Knowles[11] have described an interesting approach in which milk is conserved by adding formaldehyde then stored until ^{90}Sr and ^{90}Y have attained secular equilibrium. Sodium citrate is added to bind Y as an anionic citratecomplex which is separated by treating the milk with an anion exchange resin. The Y is subsequently eluted and purified by extraction into tri-n-butyl-phosphate.

The same milk sample is further treated with cation exchange resin to bind ^{89}Sr and ^{90}Sr. These and other metal cations are eluted and the Sr purified by the precipitation reactions described in Harley´s scheme. This includes the difficult separation of Sr^{2+} from Ca^{2+} and Ba^{2+}. Ca^{2+} is removed by precipitating $Sr(NO_3)_2$ from fuming nitric acid, an unpleasant and hazardous procedure. Ba^{2+} is removed by precipitating it as $BaCrO_4$. This reaction, however, provides variable decontamination factors for Ba and frequently precipitates large amounts of Sr with the $BaCrO_4$.

The procedures mentioned above rely on achieving secular equilibrium between ^{90}Sr and ^{90}Y and require two b-measurements. Moreover, because large losses of strontium can occur, overall yields must be determined and the results corrected for these losses.

Clearly, a method is needed for separating strontium reproducibly and in large yield from liquid milk, thus avoiding the lengthy drying and ashing procedures as well as the yield determinations. Furthermore, a measurement technique is required that eliminates the need to carry out two measurements and is independent of the $^{90}Sr/^{90}Y$ equilibrium.

We have recently described a fast method for isolating strontium from raw milk.[12] The technique involves binding the divalent metal cations including Sr^{2+} to a chelating resin containing aminomethyl phosphonate groups. These divalent cations (e. g., Ca^{2+}, Sr^{2+}, Ba^{2+}) are quantitatively recovered by eluting the resin with dilute mineral acid. Ba^{2+} is removed by extracting it from an aqueous picrate solution into a solution of 21- crown-7 in chloroform. Sr^{2+} is isolated from Ca^{2+} by extracting the Sr^{2+} complex with dicyclohexyl-18-crown-6 into chloroform, a method similar to that described by Kimura et al.[13,14] After extracting back into the aqueous phase, Sr^{2+} can be precipitated as carbonate.

The present application note describes some improvements to this method along with its combination with a technique for measuring simultaneously ^{89}Sr, ^{90}Sr and ^{90}Y by liquid scintillation spectrometry. The studies[15,16] have shown the feasibility of such an approach. In this procedure, the $SrCO_3$ preparation containing all three isotopes is reacted with aqueous toluene sulphonic acid and dispersed in a liquid scintillation cocktail. The Revvity LAS TriCarb 2260XL low level scintillation counter is used to record the scintillation spectrum which is then resolved by a simple spectral stripping technique. We report here a test on five 2 liter samples of milk contaminated with known activities of ^{89}Sr and ^{90}Sr. Identical results can be obtained using the Revvity TriCarb model 3100TR with ultra low level option. This instrument belongs to the latest series of LSC Counters with Windows XP user interface.

Materials

All reagents except the following were obtained as analytical grade from Merck (Darmstadt, Germany). Standard solutions of ^{89}Sr and ^{90}Sr were provided by the Physikalisch-Technische-Bundesanstalt (Braunschweig, Germany), while the radioactive tracers ^{45}Ca, ^{85}Sr, ^{85}Y, ^{133}Ba and ^{137}Cs were purchased from Amersham-Buchler (Braunschweig, Germany). The scintillation cocktail Insta-Gel and the high performance glass scintillation vials were from Revvity LAS (Rodgau, Germany). For detailed information about scintillation cocktails please read application note 16.[17] The crown ether 21-crown-7 was from Parish (Orem, Utah, USA), the blue-band filters were from Schleicher and Schüll (Dassel, Germany) and the chelating resin Chelite P was from Serva (Heidelberg, Germany). Raw milk was provided by the Federal Dairy Research Centre´s Experimental Farm.

Methods

Contamination of the milk with ^{89}Sr and ^{90}Sr.

39.55 Bq ^{89}Sr and 8.34 Bq ^{90}Sr along with 1–2 µg Sr^{2+} and Y^{3+} carriers as aqueous solutions of their chlorides were added per liter of milk. The milk was stirred and allowed to equilibrate at room temperature for at least one hour before proceeding with the analysis.

Isolation of strontium from the milk

The overview of the analysis in Figure 1 summarizes the main stages of the separation. The details are as follows:
Stage 1: Each 2 liter sample was stirred with 300 g (450 ml wet volume) of Chelite P in the Na^+ form for one hour at 70 °C. The milk was decanted off and discarded.

Stage 2: The resin was transferred to a 66 * 3 cm inner diameter glass column fitted at the outlet with a glass wool filter and stopcock. The resin was washed free of milk residue with about two liters of deionized water at 70 °C and an additional two liters at 95 °C. These washings were discarded.

Stage 3: Divalent cations were eluted from the column of Chelite P with about 550–600 ml of 2 M CCl at 70 °C at a flow rate of 10–20 ml/minute. This eluate was collected.

Stage 4: The pH of the eluate was adjusted to about 12 with aqueous carbaminate added and the solution was heated (uncovered) in a boiling water bath for 45–60 minutes to coagulate the precipitate. The solution was cooled and the precipitate was collected under mild suction on a filter paper (blue-band) in a Buchner-funnel. The precipitate and filter were washed free of Na^+ and NH_4^+ ions with deionized water. The filtrate and the washings were discarded.

The precipitate was dissolved by reacting it with the minimum quantity of approximately 5–6 M HCl. The acid was added dropwise and the Buchner covered with a watchglass to prevent losses due to effervescence. The resulting solution was then washed with deionized water.

Stage 5: If necessary, the volume of the above solution was adjusted to less than 70 ml by evaporation, before neutralizing to pH 3–5 with LiOH solution. It is important that the total volume at this point does not exceed 100 ml. Ba (5 mg) and Sr (1 mg) carriers were added along with 1 mmol of picric acid. This solution was extracted in a separating funnel for two minutes with 100 ml $5*10^{-4}$ M 21–crown–7 in dichloromethane. The organic phase containing the barium was discarded.

Stage 6: To the aqueous phase remaining after Stage 5 were added 5 mg Sr carrier and 5 ml aqueous 2 M sodium acetate and acetic acid. This was now extracted for two minutes with 100 ml $2*10^{-3}$ M dicyclohexyl–18–crown–6 in chloroform. The chloroform phase was collected. 5 mg of Sr carrier were added to the aqueous phase and the extraction was repeated with fresh dicyclohexyl–18–crown–6 chloroform solution. A third extraction was similarly carried out on the aqueous phase enriched with 5 mg Sr carrier.

Stage 7: The combined chloroform phases from Stage 6 were extracted for two minutes with 100 ml of aqueous solution 1 M in HCl and NaCl and 0.1 M in NH_4Cl. The aqueous phase was collected. The chloroform phase was extracted with 50 ml of fresh aqueous HCl/NaCl/NH_4Cl containing 5 mg of Sr carrier. The aqueous phase was collected and combined with that from the first extraction. The total amount of Sr carrier present is now 20 mg.

Stage 8: The aqueous phase (i. e., 1 M in HCl and NaCl and 0.1 M in NH_4Cl) was transferred to a 250 ml centrifuge glass and pH adjusted to 12 by adding 10 M NaOH. 1–2 g of ammonium carbaminate were added and the solution heated in a boiling water bath for 30–45 minutes. The solution was briefly centrifuged before collecting the precipitate on a blue-band filter paper under suction. The precipitate and filter were washed with deionized water, then with a few ml of methanol.

Stage 9: The $SrCO_3$ (from Stage 8) and the filter were dried for 2-3 minutes in an oven at 80 °C. If the outer edge of the filter paper was contaminated with traces of picric acid, this was cut away before placing the $SrCO_3$ and filter into a glass scintillation vial. The carbonate was dissolved by reacting with 2.0 ml of an aqueous 25% W/V toluene sulphonic acid solution. 19.0 ml of Insta-Gel were added and the mixture shaken vigorously. The resulting emulsion, which is stable for several months, was equilibrated in darkness for at least 15 minutes at the working temperature of the scintillation counter before carrying out the measurement.

Stage 10: Details of the measurement and calculation of results are discussed below.

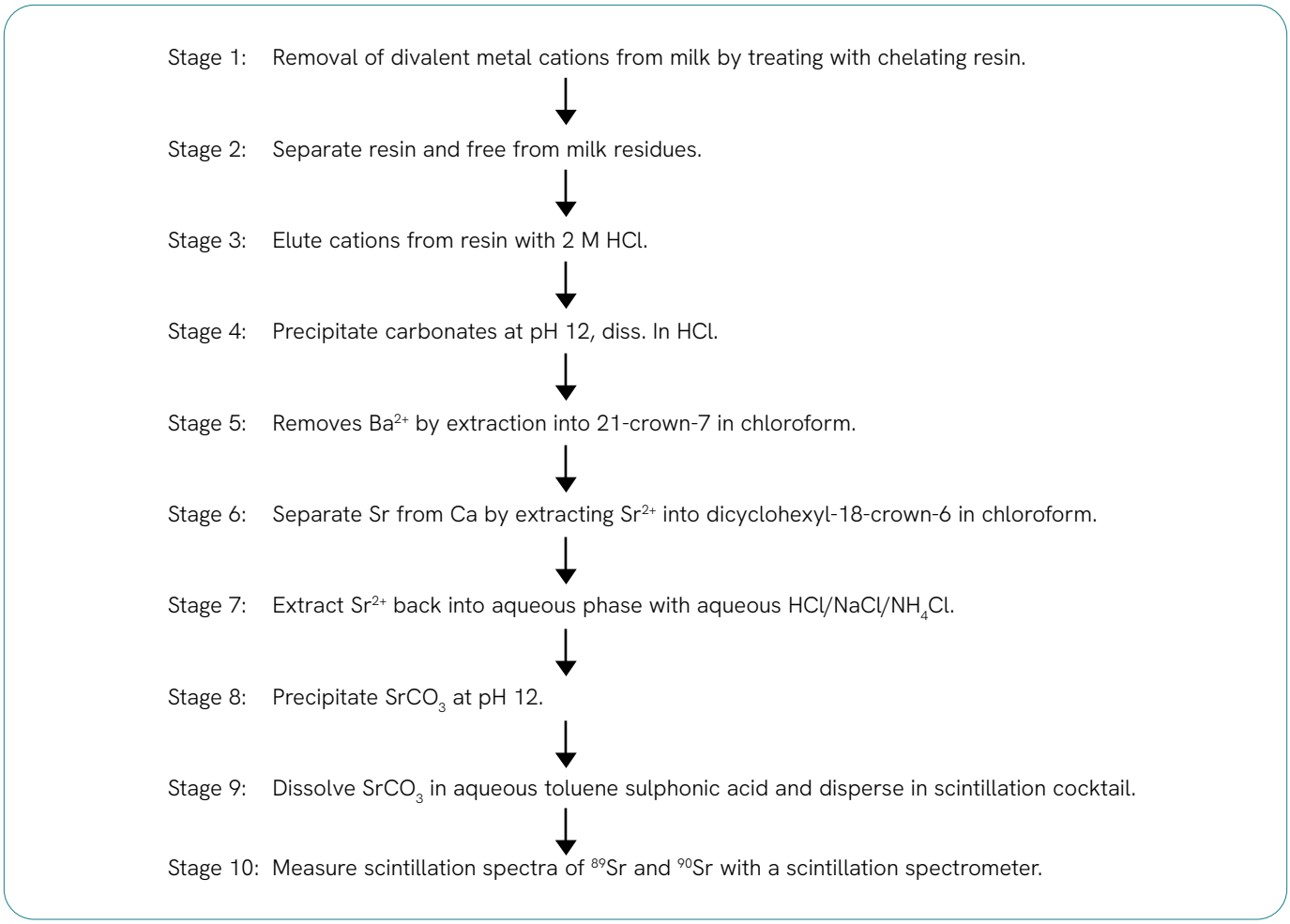

Figure 1: Outline of method for analysis of ^{89}Sr and ^{90}Sr.

Präparation of standards

^{89}Sr: 100-500 µl aliquots of an ^{89}Sr standard solution (200 Bq/ml) were added to 1.0 ml of 50% W/V aqueous toluene sulphonic acid containing 20 mg Sr carrier in a glass scintillation vial. The volume was made up to 2.0 ml with deionized water before adding 19.0 ml Insta-Gel.

^{90}Sr: 100-500 µl aliquots of ^{90}Sr standard solution (150 Bq/ml) were transferred by pipette to a separating funnel and diluted with 100 ml of a solution 0.1 M in both sodium acetate and acetic acid. 5 mg Sr carrier, 6 µg Y carrier and 1 mmol of picric acid were also added. This solution was extracted twice with dicyclohexyl-18- crown-6 chloroform solution as described in stage 6.

The aqueous phase was collected for extraction of ^{90}Y (see below), while the organic phase was treated as described in Stages 7, 8 and 9. An additional 5 mg Sr carrier were added prior to the carbonate precipitation (Stage 8) to bring the total Sr carrier present to 20 mg.

^{90}Y: 20 mg of Sr carrier were added to the aqueous phase remaining after extraction of the ^{90}Sr standard with dicyclohexyl-18-crown-6 as described above. $SrCO_3$ was precipitated as described under Stage 8. Under these conditions Y precipitates with $SrCO_3$. If more than 6 µg Y carriers are present, however, the $SrCO_3$ may have a slightly yellowish colour and have increased quench in the scintillation cocktail mixture (prepared as in Stage 9).

Background: 2 ml of 25% W/V aqueous toluene sulphonic acid containing 20 mg Sr carrier were dispersed in 19.0 mL Insta-Gel in a glass scintillation vial.

Measurement of scintillation spectra and calculation of results.

The scintillation spectra of standard and background cocktails and sample cocktails were measured with a TriCarb 2260XL Low-Level Liquid Scintillation Counter from Revvity LAS (former Packard Instrument Company). Figure 2 shows spectra of the cocktails containing ^{89}Sr, ^{90}Sr and ^{90}Y standards.

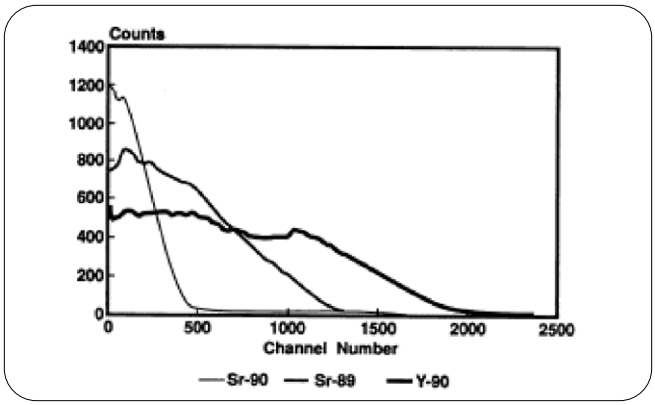

Abb. 2: Counting regions for ^{89}Sr, ^{90}Sr and ^{90}Y

The total spectral region of 2500 channels shown in Figure 2 can be divided into three sub-regions A, B and C according to the sources of the counts that can be registered there, as follows:

Region	Possible source of counts
A	^{90}Y, ^{89}Sr, ^{90}Sr
B	^{90}Y, ^{89}Sr
C	^{90}Y

The spectrum of a sample containing all three nuclides will therefore contain counts distributed in regions A, B and C as shown above. Background counts will also be registered in all three regions.

To evaluate ^{89}Sr and ^{90}Sr in such a complex spectrum, information is needed from spectra of background and each of the nuclides measured alone. To allow ready comparison of these spectra, the counts in each region are divided by the measurement times of given count rates (R). The net count rate of ^{89}Sr in region B of the sample spectrum (^{89}Sr RB) is calculated from **Equation 1:**

$$^{89}\text{Sr RB} = \text{Total RB} - \text{BG RB} - {}^{90}\text{Y RB} \quad (1)$$

where Total RB, BG RB and ^{90}Y RB are the total observed count rate, the background count rate, and the count rate attributable to ^{90}Y in region B of the sample spectrum.

Similarly, the net count rate of ^{90}Sr in region A of the sample spectrum (^{90}Sr RA) is given by **Equation 2:**

$$^{90}\text{Sr RA} = \text{Total RA} - \text{BG RA} - {}^{90}\text{Y RA} - {}^{89}\text{Sr RA} \quad (2)$$

Values of Background RA and RB are readily obtained from measurements of the background cocktail. To calculate ^{90}Y RA and ^{90}Y RB a cocktail containing standard ^{90}Y only must be measured. From the spectrum of such a preparation, proportionality constants (k) relating the count rate in region C with those in regions A and B can be obtained, as shown in **Equations 3 and 4:**

$$k_1 = \frac{^{90}\text{Y Standard RA}}{^{90}\text{Y Standard RC}} \quad (3)$$

$$k_2 = \frac{^{90}\text{Y Standard RB}}{^{90}\text{Y Standard RC}} \quad (4)$$

These constants can be assumed to be valid also for sample spectra (i. e., those containing all three nuclides). Therefore, ^{90}Y RA and ^{90}Y RB for a sample can now be calculated by combining these constants with the ^{90}Y RC observed for the sample as shown in **Equations 5 and 6:**

$$^{90}\text{Y RA} = k_1 * {}^{90}\text{Y RC} \quad (5)$$

$$^{90}\text{Y RA} = k_2 * {}^{90}\text{Y RC} \quad (6)$$

All the components required to solve Equations 1 can thus be determined.

The net count rate of ^{89}Sr in region A of the sample can be derived by a procedure similar to that described in Equations 3-6: from a spectrum of a cocktail containing ^{89}Sr standard only, a proportionality constant is obtained **(Equation 7):**

$$k_3 = \frac{^{89}\text{Sr Standard RA}}{^{89}\text{Sr Standard RB}} \quad (7)$$

Combining k_3 with the value of the sample ^{89}Sr RB given by Equation 1 (shown in **Equation 8**):

$$^{89}\text{Sr RA} = k_3 * {}^{89}\text{Sr RB} \quad (8)$$

Equation 2 can now be readily solved for ^{90}Sr RA.

To convert the count rates of ^{89}Sr and ^{90}Sr given by Equations 1 and 2 into activities, the counting efficiencies of these nuclides in regions B and A, respectively, are required.

These efficiencies (E) are determined by measuring the net count rates in the regions of interest of cocktails containing known activities of ^{89}Sr and ^{90}Sr standards, than applying **Equations 9 and 10:**

$$^{89}\text{Sr EB} = \frac{^{89}\text{Sr Standard RB}}{^{89}\text{Sr Standard Aktivität}} \quad (9)$$

$$^{90}\text{Sr EA} = \frac{^{90}\text{Sr Standard RA}}{^{90}\text{Sr Standard Aktivität}} \quad (10)$$

where ^{89}Sr EB and ^{90}Sr EA are the efficiencies of counting ^{89}Sr and ^{90}Sr in den regions B and A, respectively. It is important here that the units of count rate and activity are the same, e. g., both in counts per second or in counts per minute.

The activities of ^{89}Sr and ^{90}Sr in the sample are therefore given by Equations 11 and 12:

$$^{89}\text{Sr sample activity} = \frac{^{89}\text{Sr RB}}{^{89}\text{Sr EB}} \quad (11)$$

$$^{90}\text{Sr sample activity} = \frac{^{90}\text{Sr RA}}{^{90}\text{Sr EA}} \quad (12)$$

If the count rates have been calculated in counts per second, then the units of the activities equal Becquerel (Bq).

Region settings:
The results depend on the settings used for the regions A, B and C. Initial settings were chosen by inspection of spectra of cocktails containing the single nuclides.

To optimize these settings, a cocktail was prepared containing known activities of standard ^{89}Sr and ^{90}Sr in equilibrium with ^{90}Y (i. e., no ^{90}Sr/^{90}Y separation was carried out). This cocktail was measured and evaluated with various region settings according to the procedure described above. The single nuclide standards used for comparison were of the same activity as the mixed nuclide standard cocktail. The estimated activities of ^{89}Sr and ^{90}Sr in the mixed standard cocktail were 99.4% and 99.5% of those added, when the following settings were used:

Region	first channel	last channel_
A	20	310
B	310	1112
C	1112	1956

Count statistical error:
To calculate the relative uncertainty in the activities, the statistical errors (σ) associated with the counts measured in each region of the standards (^{89}Sr, ^{90}Sr and ^{90}Y), background and sample were considered. This error was converted to a relative error σ_{rel} for each measurement, e. g., for the counts in region of a given spectrum (shown in **Equations 13 and 14**):

$$\sigma = \sqrt{\text{Counts in region}} \quad (13)$$

$$\sigma \text{ rel} = \frac{\sigma}{\text{Counts in region}} \quad (14)$$

The relative uncertainties of the ^{89}Sr and ^{90}Sr activities are then derived by geometric addition of the srel values for all measurements involved in the calculation. For further details about counting statistics please refer to the literature.[18, 19)]

Measurement of decontamination factors.

These factors were determined for Ca, Y, Ba and Cs using the radiotracers ^{45}Ca, ^{88}Y, ^{133}Ba and ^{137}Cs. The measurement technique is described in reference 12.

Results

In all scintillation measurements, the transformed spectral index of the external standard (tSIE) developed by Packard (now Revvity LAS) was used as the quench indicating parameter. For more details about this and other quench parameters please read also application note 33.[20)]
The value of this parameter (on a scale from 0-1000) varied in the range 343-360. This small variation can be minimized by ensuring that all samples are equilibrated long enough at the operating temperature of the instrument.

Figure 3 shows the effect of quench on the counting efficiencies of the three nuclides in the region A for A for ^{90}Sr, B for ^{89}Sr and C for ^{90}Y. To obtain these plots, the quench of scintillation cocktail containing known activities (about 200 Bq) of the single nuclides was varied either by passing N_2 (to displace O_2 and increase the tSIE quench factor), or by adding 10-50 µl of chloroform (to decrease the tSIE quench factor). The plots indicate that the small variation in quench observed in our sample measurements carried out under normal conditions do not influence the count rates significantly.

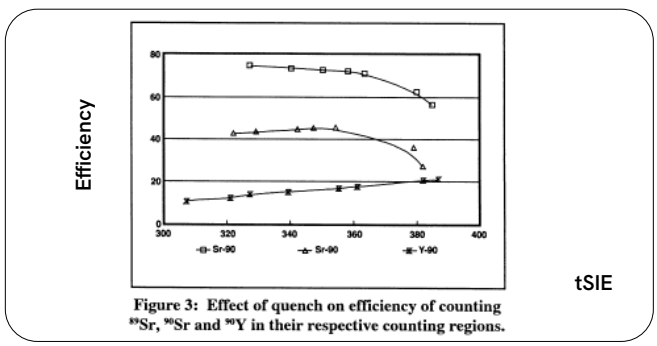

Figure 3: Effect of quench on efficiency of counting ^{89}Sr, ^{90}Sr and ^{90}Y in their respective counting regions.

The decontamination factors for Ca, Y, Ba and Cs are listed in Table 1. These values indicate that Ca and radioisotopes of Ba and Cs are not likely to interfere with the analysis. The large decontamination factor for Y indicates that the correction for ^{90}Y contributions in regions A and B is very small for freshly prepared Sr samples.

Table 2 shows the results of all five analyses of the two liter milk samples contaminated with 39.55 +/- 0.99 Bq/l ^{89}Sr and 8.34 +/- 0.17 Bq/l ^{90}Sr. The averages of the values in the table are 34.7, standard deviation 1.4 Bq/l and 7.54, standard deviation 0.25 Bq/l for ^{89}Sr and ^{90}Sr respectively.

Table 1: Decontamination factors

Decontamination factors*	
Element	**Factor**
Ca	> 6,2 * 102
Y	1,6 * 104
Ba	7,9 * 101
Cs	> 1,8 * 102

* Activity in initial milk/activity in final $SrCO_3$ isolate at Stage 8

These values correspond to 87.7 +/- 3.5% of the ^{89}Sr and 90.4 +/- 3.0% of the ^{90}Sr added to the milk. Studies on the recovery of strontium from two liter milk samples containing ^{89}Sr as tracer indicated that 90.4% of the Sr is recovered. This is similar to the recoveries of 93.5 +/- 0.7%, found for 100 ml milk samples.[12)]

Table. 2: Results of analyses of milk samples of two liter volume contaminated with 39.5 and 8.35 Bq of ^{89}Sr and ^{90}Sr

	Bq/l found*		% Recovery§	
Sample	^{89}Sr	^{90}Sr	^{89}Sr	^{90}Sr
1	34,0	7,32	86,0	87,7
2	40,0	7,89	90,9	94,5
3	34,2	7,71	86,5	92,3
4	36,3	7,34	91,9	88,0
5	33,1	7,42	83,7	88,9

* total relative statistical counting error (3σ) for ^{89}Sr are < 1.6% and for ^{90}Sr < 5.5% in all cases.

§ (Bq nuclide found / Bq nuclide added) multiplied with 100

Conclusion

The results of the analyses agree well with the activities added to the milk samples; the strontium losses which occur in the isolation procedure correspond with the observed discrepancies between the added and measured activities. Small losses of strontium are observed at all stages of this procedure, particularly at the Ba/Sr separation (Stage 4). However, the good reproducibility would permit the use of a factor correcting for these losses.

Decontamination factors for I and Te were not determined because these species are presumably very strongly removed at Stages 1-3. The naturally occurring radionuclides ^{40}K and ^{87}Rb are assumed to behave similarly to Cs because all three are related alkali metals. These elements should be strongly removed at Stages 1-3 and 8. The bulk of the ^{14}C and ^{3}H also present in milk certainly remains in the milk during treatment with the chelating resin at Stage 1. They should not interfere with the results. Milk from the same area as the milk samples used here contains typically 0.10 Bq/l and in any event <0.20 Bq/l ^{90}Sr. Interference from this contamination is therefore negligible, too.

The effect on the detection limit for ^{90}Sr of $^{89}Sr/^{90}Sr$ ratio, ^{90}Y activity and measurement are under investigation. Preliminary studies indicate a detection limit of around 0.3-0.5 Bq at a $^{89}Sr/^{90}Sr$ ratio of 10:1 and with a measurement time of 12 hours. Further work is needed to ascertain whether the method is suitable for monitoring ^{90}Sr at current levels of environmental contamination.

The isolation technique requires 5-8 hours depending on the volume of the milk sample, so that results can be obtained within 24 hours. The findings described here imply that the method is suitable for monitoring milk for radiostrontium in areas contaminated with fresh fallout or with increased levels of ^{90}Sr.

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