

Rapid Simultaneous Determination of ^{89}Sr and ^{90}Sr in Milk: A Procedure Using Cerenkov and Scintillation Counting

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International Atomic Energy Agency

RAPID SIMULTANEOUS DETERMINATION
OF ^{89}Sr AND ^{90}Sr IN MILK:
A PROCEDURE USING CERENKOV
AND SCINTILLATION COUNTING

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FOREWORD

Since 2004, the IAEA programme related to the terrestrial environment has included activities aimed at developing and testing a set of procedures for the determination of radionuclides in environmental samples. Both ^{89}Sr and ^{90}Sr are fission products that can be, and have been, released to the environment during nuclear explosions and nuclear reactor accidents. Since strontium uptake from milk is an important pathway for incorporation of radioactive strontium into the human body, the rapid and accurate analysis of radioactive strontium isotopes in milk is of crucial importance in emergency situations in order to protect the public from radiation hazards.

This report describes a new approach for the rapid determination of ^{89}Sr and ^{90}Sr in milk using Cerenkov and scintillation counting methods that was tested and validated by the analysis of four spiked milk samples in terms of repeatability, reproducibility and trueness (relative bias) in accordance with ISO guidelines. The report also describes the calculation of the uncertainty budget.

This research was proposed in 2007 at the Asia–Pacific regional meeting of the IAEA Analytical Laboratories for the Measurement of Environmental Radioactivity (ALMERA) network. Several ALMERA network laboratories participated in the validation procedure by performing reproducibility tests.

The resulting recommended procedure is designed to be of general use to a wide range of laboratories, including those in the ALMERA network. It is expected that this rapid method for determining ^{89}Sr and ^{90}Sr in milk will be useful in emergency conditions and for routine environmental monitoring of elevated levels of radioactivity.

The IAEA wishes to thank all the participants for their valuable contributions to the development of the procedure and the review of this report, in particular C.-K. Kim. The IAEA officer responsible for this publication was A. Pitois of the IAEA Environment Laboratories.

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1. INTRODUCTION

Radioactive strontium (^{89}Sr and ^{90}Sr) is one of the most biologically hazardous radionuclides produced in nuclear fission processes. If released into the environment due to nuclear power plant accidents such as a reactor meltdown with or without failed containment, fallout from nuclear detonations or radiological attacks, radioactive strontium becomes incorporated into the calcium pool. The radioactive strontium can then enter the human body through the principal pathway: soil to plant to cow's milk to humans. Milk, a major constituent of the human diet, is an important indicator of the transfer of radionuclides from the environment to humans. Therefore, in emergency situations the rapid analysis of ^{89}Sr and ^{90}Sr in milk is essential to protect humans from radiation exposure due to the intake of contaminated milk. The analysis time is an important economic factor, as rapid identification of contamination with radioactive material helps decision makers introduce protective measures according to the intervention levels.

The analysis procedure for radioactive strontium is always complicated because ^{89}Sr , ^{90}Sr and its daughter ^{90}Y are pure beta emitters, and prior to measurement they must be separated from the sample. Some methods using ion exchange resins have been attempted for rapid determination of radioactive strontium in milk in order to avoid the usual process of drying, ashing and acid leaching [1-2]. Melin and Suomela [3] reported a rapid procedure for the determination of ^{89}Sr and ^{90}Sr in food and environmental samples by Cerenkov counting. After measuring ^{89}Sr there is an additional step to separate ^{90}Y . The method has not been applied to samples that have $^{89}\text{Sr}/^{90}\text{Sr}$ activity ratio higher than 5, which would be expected to be present in real samples collected under emergency conditions.

Brun et al. [4] summarized rapid methods of radioactive strontium analysis in milk, compared several methods in terms of analysis time, and reported that the procedure combining cation exchange resin, Sr-resin, oxalate and BaCrO_4 precipitations is appropriate for the analysis of ^{90}Sr in milk within 48 hours [5]. However, it was difficult to apply this method to milk samples containing both ^{89}Sr and ^{90}Sr . In order to simultaneously measure ^{89}Sr and ^{90}Sr , there have been several attempts using a spectrum-unfolding method [6] (Hong et al.), other full spectrum methods [7] and the constrained optimization function in three energy windows with MATLAB software [8]. Eikenberg et al. [9] applied the three energy window approach to measure ^{89}Sr and ^{90}Sr in environmental samples. However, these methods often fail when the ^{89}Sr activity is much higher than the ^{90}Sr activity, because a shift in the spectrum towards the low energy region caused by a small variation in the quenching level leads to a significant counting error in the ^{90}Sr energy region. This problem was demonstrated in the work of Kim et al. [10] where the direct counting method of ^{89}Sr and ^{90}Sr using two energy windows was applied and higher than 20% relative bias was found when the $^{89}\text{Sr}/^{90}\text{Sr}$ activity ratio exceeded 10 due to spectrum shifting. Recently, a rapid method of ^{89}Sr and ^{90}Sr in milk using Cerenkov counting was introduced by Heckel and Vogl [11], but it has not yet been applied to samples that have a high activity ratio of $^{89}\text{Sr}/^{90}\text{Sr}$. To reduce the high relative bias of ^{89}Sr and ^{90}Sr activities observed in the previous study, Kim et al. [12] introduced a combined counting method where the sample was sequentially measured in Cerenkov and liquid scintillation counting modes. The analytical results and measurement uncertainties of ^{89}Sr and ^{90}Sr activity in spiked milk samples using the rapid method were compared with those using the conventional method. According to the rapid method, the ^{89}Sr and ^{90}Sr activities in milk samples of $^{89}\text{Sr}/^{90}\text{Sr}$ activity ratio as high as 35 could be measured within 1 day with less than 20% and 40% relative biases, respectively. The method was applied to four kinds of spiked milk powders with different $^{89}\text{Sr}/^{90}\text{Sr}$ activity ratios for the validation of the method.

A review paper on the analytical methodologies for the determination of radioactive strontium isotopes was recently published by Vajda and Kim [13].

2. SCOPE

This document describes a method for the simultaneous determination of ^{89}Sr and ^{90}Sr in milk samples sequentially using Cerenkov and scintillation counting techniques following the chemical separation of strontium. The method can be used for the relatively fast detection of contamination of milk in emergency situations or for the measurement of elevated activity levels in the environment.

The required sample mass for the analysis is typically 0.25–0.5 L.

3. NORMATIVE REFERENCES

- ISO 18589-5:2009. Measurement of radioactivity in the environment – Soil – Part 5: Measurement of strontium 90.
- ISO 11929:2010. Determination of the characteristic limits (decision threshold, detection limit and limits of the confidence interval) for measurements of ionizing radiation – Fundamentals and application.
- ISO/TS 21748:2004. Guidance for the use of repeatability, reproducibility and trueness estimates in measurement uncertainty estimation.
- ISO Guide 1995. Guide to the expression of uncertainty in measurement.
- ISO 5725-1:1994. Accuracy (trueness and precision) of measurement methods and results – Part 1: General principles and definitions.
- ISO 5725-2:1994. Accuracy (trueness and precision) of measurement methods and results – Part 2: Basic method for the determination of repeatability and reproducibility of a standard measurement method.
- ISO 5725-4:1994. Accuracy (trueness and precision) of measurement methods and results – Part 4: Basic methods for the determination of the trueness of a standard measurement method.
- ISO 7870:1993. Control charts – General guide and introduction.
- ISO 7966:1993. Acceptance control charts.
- QUAM:2012.P1 EURACHEM/CITAC Guide, 2012. Quantifying Uncertainty in Analytical Measurement.
- ISO/IEC Guide 43-1:1997. Proficiency testing by interlaboratory comparisons – Part 1: Development and operation of proficiency testing schemes.

4. RESPONSIBILITIES

It is the responsibility of the analyst to follow this procedure and to establish appropriate safety and health practices. Compliance with national and/or internal chemical and radiological safety rules is obligatory.

5. TERMS AND DEFINITIONS, SYMBOLS AND ABBREVIATIONS

a_t^{Sr89} : massic activity of ^{89}Sr in the spiked milk powder at the target date (or sampling date) (Bq kg^{-1} , as dry mass).

a_t^{Sr90} : massic activity of ^{90}Sr in the spiked milk powder at the target date (or sampling date) (Bq kg^{-1} , as dry mass).

a^* : decision threshold (Bq kg^{-1} , as dry mass).

$a^\#$: detection limit (Bq kg^{-1} , as dry mass).

$A_{t_0}^{Sr89}$: total activity of ^{89}Sr in source at the separation date (Bq).

$A_{t_1}^{Sr89}$: total activity of ^{89}Sr in source at the first measurement time in Cerenkov counting mode (Bq).

$A_{t_2}^{Sr89}$: total activity of ^{89}Sr in source at the second measurement time in scintillation counting mode (Bq).

$A_{t_0}^{Sr90}$: total activity of ^{90}Sr in source at the separation time (Bq).

$A_{t_1}^{Sr90}$: total activity of ^{90}Sr in source at the first measurement time in Cerenkov counting mode (Bq).

$A_{t_2}^{Sr90}$: total activity of ^{90}Sr in source at the second measurement time in scintillation counting mode (Bq).

C : the assigned massic activity of the analyte in the reference material (Bq kg^{-1} , as dry mass).

f_1 : correction factor for ^{90}Y built up for the time interval between the separation time and the first measurement time in Cerenkov counting mode.

f_2 : correction factor for ^{90}Y built up for the time interval between the separation time and the second measurement time in scintillation counting mode.

f_3 : decay correction factor of ^{89}Sr for the time interval between the 1st measurement time in Cerenkov counting mode and the 2nd measurement time in scintillation counting mode.

$N_{t_1}^c$: net count rates of the first measurement at the window (10–360 channels) in Cerenkov counting (cps) as a result of subtraction of background count rate from gross count rate
$$N_{t_1}^c = N_{G1}^c - N_{B1}^c$$

N_{t2}^s : net count rates of the second measurement at the window (120–910 channels) in scintillation counting mode with cocktail (cps) as a result of subtraction of background count rate from gross count rate $N_{t2}^s = N_{G2}^s - N_{B2}^s$.

r : chemical recovery in the rapid method.

r_L : repeatability limit (Bq kg⁻¹, as dry mass).

R_L : reproducibility limit (Bq kg⁻¹, as dry mass).

S_r : standard deviation of repeatability (Bq kg⁻¹, as dry mass).

S_R : standard deviation of reproducibility (Bq kg⁻¹, as dry mass).

$u(f_1), u(f_2), u(f_3)$: uncertainties of correction factors f_1, f_2, f_3 .

$u(a_i^{Sr89})$: combined uncertainty of the massic activity of ⁸⁹Sr at the target date (Bq kg⁻¹).

$u(a_i^{Sr90})$: combined uncertainty of the massic activity of ⁹⁰Sr at the target date (Bq kg⁻¹).

$u(\lambda_{Sr89}), u(\lambda_{Sr90}), u(\lambda_{Y90})$: uncertainty of the decay constants of ⁸⁹Sr, ⁹⁰Sr, ⁹⁰Y (s⁻¹).

$u(N^{Sr89}), u(N^{Sr90})$: uncertainties of the net counts of ⁸⁹Sr and ⁹⁰Sr, respectively.

$u(V)$: uncertainty of mass of sample (kg).

t : reference date and time.

t_0 : separation time.

t_1 : date and time of the first measurement of sample in Cerenkov counting mode.

t_2 : date and time of the second measurement of sample in scintillation counting mode.

V : dry mass of the sample (kg).

\bar{X} : the mean of all measured massic activity values (Bq kg⁻¹, as dry mass).

δ : mean bias of the method (Bq kg⁻¹, as dry mass).

$\delta(\%)$: relative mean bias of the method (%).

ε_c^{Sr89} : counting efficiency of ⁸⁹Sr in the ROI (10–360 channels) in Cerenkov counting mode.

ε_c^{Sr90} : counting efficiency of ⁹⁰Sr in the ROI (10–360 channels) in Cerenkov counting mode.

ε_c^{Y90} : counting efficiency of ⁹⁰Y in the ROI (10–360 channels) in Cerenkov counting mode.

ε_s^{Sr89} : counting efficiency of ⁸⁹Sr in the ROI (120–910 channels) in scintillation counting mode with cocktail.

ε_s^{Sr90} : counting efficiency of ⁹⁰Sr in the ROI (120–910 channels) in scintillation counting mode with cocktail.

ϵ_s^{Y90} : counting efficiency of ^{90}Y in the ROI (120–910 channels) in scintillation counting mode with cocktail.

$\lambda_{\text{Sr}89}$: decay constant of ^{89}Sr (s^{-1}).

$\lambda_{\text{Sr}90}$: decay constant of ^{90}Sr (s^{-1}).

$\lambda_{\text{Y}90}$: decay constant of ^{90}Y (s^{-1}).

σ : standard deviation of all measured values (Bq kg^{-1} , as dry mass).

6. PRINCIPLE

The Sr content of milk samples of about 0.25 L (or milk powder in equivalent amount) is concentrated with 35 mL strong acidic cation exchange resin at a pH of 5.3–5.5. The resin is washed with de-ionized water and Sr is eluted with 200 mL 4M NaCl. To pre-concentrate Sr with the alkaline earth metals a carbonate precipitate is formed that is dissolved in 8M HNO_3 . Sr is selectively separated from the alkaline earth metals and other elements by extraction chromatography (EC) using 3 g of Sr resin that contains bis-4,4'(5')-t-butylcyclohexano-18-crown-6 (crown ether) on an inert support material (product of Eichrom Technologies, LLC). Capacity factors of the resin as determined by Horwitz et al. [14] are shown in Appendix I. The column is washed with 8M HNO_3 and Sr is stripped with de-ionized water. Strontium is precipitated as SrCO_3 and the chemical recovery is determined by gravimetry. Corrections are made according to the Sr content of the milk that is measured from the original sample solution by any adequate elemental analytical technique, e.g. atomic absorption spectrometry (AAS), optical emission spectrometry (OES), or inductively coupled plasma mass spectrometry (ICP-MS).

The massic activities of ^{89}Sr and ^{90}Sr are determined by the consecutive counting of the Sr source by Cerenkov and LSC techniques. Nuclear parameters of various Sr isotopes are listed in Appendix II. Measurements are performed for a relatively short counting interval (e.g. 5 times 20 min), as soon as possible after finishing the chemical procedure and in the given order, to obtain prompt information. From the two measurements the activities of the two unknown parameters are calculated. The presence of ^{90}Y is taken into account by the calculation of the actual in-growth factors.

The basic interferences due to various Sr and Y nuclides, i.e. ^{89}Sr , ^{90}Sr and ^{90}Y are eliminated or taken into account by the present counting protocol. The procedure has no significant interferences from other radionuclides. Decay characteristics of the relevant radionuclides are shown in Appendix II. Yttrium is usually well separated but ^{90}Y grows into the Sr source from its ^{90}Sr parent nuclide after the separation according to the law of radioactive decay. Very “fresh” samples may contain short-lived ^{91}Sr ($T_{1/2}=9.6$ h) and ^{92}Sr ($T_{1/2}=2.7$ h); their presence can be controlled by gamma-ray spectrometry, by repeated measurements or eliminated simply by starting the first measurement after a relatively short cooling period of a couple of days. The selectivity of the chemical procedure (especially that of EC) is responsible for eliminating possible interferences due to the alkaline earth metals, e.g. Ca and Ba. Lead is strongly retained on the Sr resin, but is not eluted by the small volume of de-ionized water.

7. CHEMICAL EQUIPMENT AND REAGENTS

7.1. EQUIPMENT

- Standard laboratory equipment;
- Low background glass scintillation vials;
- Ultra low background liquid scintillation counter;
- Vacuum pump;
- Filtering set suitable for a filter of diameter 23 mm;
- Analytical balance with accuracy of 0.1 mg;
- Hot plate with magnetic stirrer;
- Micropipettes;
- pH papers.

7.2 CHEMICALS

- 10M HCl (32%);
- 14.4M HNO₃ (65%);
- H₂O₂ (25%);
- 4M NaCl;
- NH₃ solution (25%);
- 6M NaOH;
- 0.25M citric acid;
- Na₂CO₃;
- Oxalic acid;
- Ethanol;
- Sr carrier solution (10 mg Sr mL⁻¹ in 1M HNO₃);
- Y carrier solution (10 mg Y mL⁻¹ in 1M HNO₃);
- Dowex 50W-X8 resin, 50–150 mesh, Na-form;
- Sr resin (50–100 μm);
- Scintillation cocktail suitable for alpha/beta separation (for example, UltimaGold AB cocktail, Packard, USA, Cat. No.: 6013309);
- ⁸⁹Sr and ⁹⁰Sr standard solutions.

Equipment and chemicals used for preparation of the spiked milk samples and performance of homogeneity tests are described elsewhere.

All the chemical reagents needed for carrying out this procedure must be of analytical grade.

8. PROCEDURE

8.1. CALIBRATION OF THE INSTRUMENTS

8.1.1. Preparation of ⁸⁹Sr calibration source

- (1) Add a few μL of a known activity of ⁸⁹Sr standard solution (total activity: about 50 Bq) into a 20 mL glass vial.
- (2) Gently evaporate the solution to dryness with an infrared lamp.

- (3) Add a few drops of 65% HNO₃ then evaporate to dryness to change the chemical form to a nitrate.
- (4) Add 8 mL of 0.05M HNO₃, 13 mg of ⁹⁰Sr-free SrCO₃ and an empty filter into the vial and gently shake to dissolve the SrCO₃ precipitate.

8.1.2. Preparation of pure ⁹⁰Sr and ⁹⁰Y calibration sources

- (1) Add a few μL of a known activity of ⁹⁰Sr standard solution (total activity: about 20 Bq), 1 mL of Sr and Y carrier (10 mg mL⁻¹) into a 50 ml beaker.
- (2) Add 10 mL 3M HNO₃.
- (3) Load the solution on the Sr resin column (3 g Sr resin, inner diameter: 10 mm, length: 120 mm) preconditioned in advance with 100 mL of 3M HNO₃.
- (4) Wash the column with 100 mL 3M HNO₃.
- (5) Combine the effluent from the column for determining the ⁹⁰Y counting efficiency.
- (6) Elute Sr from the column with 30 mL of H₂O.
- (7) Adjust the pH of the solution to about 7 with 25% NH₄OH.
- (8) Add 5 mL of saturated (NH₄)₂CO₃ to the solution while stirring to form SrCO₃ precipitation.
- (9) Heat the solution on a hot plate for a few minutes.
- (10) After cooling the solution, filter the precipitate through a pre-weighed filter paper (for example, Whatman 42, pore size 2.5 μm, diameter of filter 23 mm (manually prepared)).
- (11) Wash the precipitate with a few mL of 80% ethanol then dry at 65°C for 30 min.
- (12) Determine the chemical recovery of Sr by gravimetric method.
- (13) Transfer the SrCO₃ precipitate into a 20 mL glass vial together with the filter.
- (14) Add 8 mL of 0.05M HNO₃ into the vial to dissolve the precipitate.
- (15) Evaporate ⁹⁰Y fraction to dryness, then add a few mL of 65% nitric acid and a few drops of 25% H₂O₂ to decompose organic material from Sr resin column.
- (16) Dissolve the residue in 8 mL of 0.05M HNO₃ and transfer the solution into 20 mL glass vial.
- (17) Take 100 μL from the solution to determine the chemical recovery of Y by ICP-MS.

8.1.3. Preparation of mixed source of ⁹⁰Sr and ⁹⁰Y in secular equilibrium

- (1) Add dozens of μL of a known activity of ⁹⁰Sr standard solution (total activity: about 20 Bq) in equilibrium with ⁹⁰Y to a 20 mL glass vial.
- (2) Prepare the mixed source of ⁹⁰Sr and ⁹⁰Y in the same way as described for the preparation of the ⁸⁹Sr source.
- (3) Calculate the Cerenkov counting efficiency of ⁹⁰Sr by subtracting the counting efficiency of ⁹⁰Y from the total counting efficiency for the mixed source of ⁹⁰Sr and ⁹⁰Y.

8.1.4. Scintillation and Cerenkov counting efficiency of ⁸⁹Sr, ⁹⁰Sr and ⁹⁰Y

Sources are measured for 100 minutes (10 min for 10 cycles) in Cerenkov counting mode at the window (10–360 channels). After counting sources in Cerenkov counting mode, sources are mixed with 12 mL of UltimaGold AB cocktail, then measured for 100 minutes (10 min for 10 cycles) at the window (120–910 channels) in high energy mode of scintillation counting.

Counting efficiencies of ⁸⁹Sr, ⁹⁰Sr and ⁹⁰Y in Cerenkov and scintillation counting modes are shown in Table 1.

TABLE 1. COUNTING EFFICIENCIES OF STRONTIUM-89, STRONTIUM-90 AND YTTRIUM-90 IN SCINTILLATION AND CERENKOV COUNTING MODES

Nuclides	Maximum β energy (MeV)	Scintillation*	Cerenkov**
		Window (120–910)	Window (10–360)
^{89}Sr	1.495	$0.978 \pm 0.007^{***}$	$0.347 \pm 0.002^{***}$
^{90}Sr	0.546	$0.937 \pm 0.006^{***}$	$0.017 \pm 0.003^{***}$
^{90}Y	2.280	$0.997 \pm 0.003^{***}$	$0.581 \pm 0.005^{***}$

*: LS Cocktail composition: 8 mL 0.05M HNO_3 + 12 mL UltimaGold AB

** : Cerenkov source composition: 8 mL 0.05M HNO_3

*** : Combined uncertainty at $k=1$

8.2 SEPARATION

8.2.1. Preparation of spiked milk from spiked milk powder

Spiked milk was prepared from spiked milk powder, because of the lack of milk samples that contain ^{89}Sr and ^{90}Sr .

- (1) Heat 250 mL of de-ionized water at 65°C in a 500 mL beaker.
- (2) Add strontium carrier (1 mL of 10 mg mL^{-1} solution in 1M HNO_3) to the solution.
- (3) Precisely adjust the pH of the solution to 5.3–5.5 with some drops of 6M NaOH (if the pH is too high, adjust it using 0.25M citric acid).
- (4) Taking into account that the fat content in milk is on average 3.5%, dissolve 33.5 g of spiked milk powder in the solution while stirring intensively. If the milk powder does not completely dissolve, use an ultrasonic homogeniser. Then cool the milk at $20\text{--}23^\circ\text{C}$.
- (5) Check if the pH of the milk is in the range of 5.3–5.5 (precaution: the milk curdles at a pH less than 5.2). If not, adjust the pH to this range.

8.2.2. Rapid separation of Sr from milk

The flowchart of the procedure is given in Appendix III. The preparation of the chemical reagents is given in Appendix IV.

- (1) Add 30 mL of cation exchange resin (Dowex 50W-X8, 50–150 mesh, Na-form) to 250 mL milk prepared in advance.
- (2) Stir the solution for 30 minutes and then allow the resin to settle down.
- (3) Decant the supernatant (milk fraction) then wash the remaining resin in the beaker with 3 portions of warm de-ionized water of a total volume of 150 mL to remove traces of the milk.
- (4) Transfer the resin from the beaker into a glass column (length: 350 mm, inner diameter: 20 mm) which contains 5 mL of fresh cation exchanger resin with 50 mL warm water.
- (5) Wash the column with 100 mL warm de-ionized water.
- (6) Elute Ca, Sr and Ba from the column with 200 mL 4M NaCl.
- (7) Add 3 g of Na_2CO_3 to the eluate solution, then heat on a hot plate to form a SrCO_3 precipitate.

- (8) Centrifuge for 10 min at 3000 rpm.
- (9) Transfer the precipitate into a 100 mL beaker with about 1 mL of 65% HNO₃ then evaporate to dryness.
- (10) Dissolve the residue in 20 mL of 8M HNO₃.
- (11) Load the solution on a Sr resin column (3 g Sr resin, inner diameter: 10 mm, length: 120 mm) preconditioned in advance with 50 mL of 8M HNO₃.
- (12) Wash the column with 100 mL of 8M HNO₃ to remove interfering elements then record the time (separation time of Sr).
- (13) Elute Sr from the column with 30 mL of de-ionized water.
- (14) After adjusting the pH of the eluate solution to about 7 with 25% NH₄OH, add 5 mL of saturated (NH₄)₂CO₃ to the solution while stirring to form a SrCO₃ precipitate.
- (15) Heat the solution on a hot plate for a few minutes.
- (16) After cooling the solution, filter the precipitate through a pre-weighed filter paper (for example, Whatman 42, pore size: 2.5 μm, diameter of filter: 23 mm).
- (17) Wash the precipitate with a few mL of 80% ethanol then dry it at 65°C for 30 minutes.
- (18) Determine the chemical recovery of Sr by a gravimetric method.
- (19) Transfer the SrCO₃ precipitate into a 20 mL glass vial together with the filter.
- (20) Add 8 mL of 0.05M HNO₃ to the vial to dissolve the precipitate.

8.3. MEASUREMENT

The massic activity concentration is calculated by counting the sample sources for an appropriate time in both the Cerenkov and scintillation modes.

- (1) Immediately measure the sample in Cerenkov counting mode of the liquid scintillation counter. The first measurement data among 6 replicate measurement results is discarded to avoid extra counts due to chemiluminescence.
- (2) After counting in Cerenkov counting mode, add 12 mL of Ultima Gold AB cocktail into the vial, and re-measure it in scintillation counting mode.

Using ⁸⁹Sr and ⁹⁰Sr standard solutions, counting efficiencies are determined as mentioned in the paragraph 8.1.4.

Typical spectra of ⁸⁹Sr, ⁹⁰Sr and ⁹⁰Y obtained in Cerenkov and liquid scintillation counting modes are shown in Appendix V.

9. EXPRESSION OF RESULTS

9.1. CALCULATION OF ACTIVITY OF SR-89 AND SR-90 PER UNIT OF MASS ON SAMPLING DATE

Activities of ⁸⁹Sr and ⁹⁰Sr in milk can be calculated from sequential measurement data of Cerenkov and scintillation counting. Net count rates for ⁸⁹Sr and ⁹⁰Sr in the ROI 10–360 channels in Cerenkov and in the ROI 120–910 channels in scintillation counting mode can be expressed by equations (1) and (2):

$$N_{t1}^c = \varepsilon_c^{Sr89} \cdot A_{t1}^{Sr89} + \varepsilon_c^{Sr90} \cdot A_{t1}^{Sr90} + \varepsilon_c^{Y90} \cdot A_{t0}^{Sr90} \cdot f_1 \quad (1)$$

$$N_{t2}^s = \varepsilon_s^{Sr89} \cdot A_{t2}^{Sr89} + \varepsilon_s^{Sr90} \cdot A_{t2}^{Sr90} + \varepsilon_s^{Y90} \cdot A_{t0}^{Sr90} \cdot f_2 \quad (2)$$

If the time interval between t_0 , t_1 and t_2 is much less than the half-life of ^{90}Sr , $A_{t_0}^{\text{Sr}90} \cong A_{t_1}^{\text{Sr}90} \cong A_{t_2}^{\text{Sr}90}$, equation (1) and (2) can be expressed by equations (3) and (4):

$$N_{t_1}^c = \varepsilon_c^{\text{Sr}89} \cdot A_{t_1}^{\text{Sr}89} + \varepsilon_c^{\text{Sr}90} \cdot A_{t_1}^{\text{Sr}90} + \varepsilon_c^{\text{Y}90} \cdot A_{t_1}^{\text{Sr}90} \cdot f_1 \quad (3)$$

$$N_{t_2}^s = \varepsilon_s^{\text{Sr}89} \cdot A_{t_1}^{\text{Sr}89} \cdot f_3 + \varepsilon_s^{\text{Sr}90} \cdot A_{t_1}^{\text{Sr}90} + \varepsilon_s^{\text{Y}90} \cdot A_{t_1}^{\text{Sr}90} \cdot f_2 \quad (4)$$

$A_{t_1}^{\text{Sr}89}$ can be expressed as follow:

$$A_{t_1}^{\text{Sr}89} = \frac{N_{t_1}^c - A_{t_1}^{\text{Sr}90} \cdot (\varepsilon_c^{\text{Sr}90} + \varepsilon_c^{\text{Y}90} \cdot f_1)}{\varepsilon_c^{\text{Sr}89}} \quad (5)$$

Inserting equation (5) to equation (4) and solving for $A_{t_1}^{\text{Sr}90}$ we obtain:

$$A_{t_1}^{\text{Sr}90} = \frac{N_{t_2}^s \cdot \varepsilon_c^{\text{Sr}89} - N_{t_1}^c \cdot \varepsilon_s^{\text{Sr}89} \cdot f_3}{\varepsilon_c^{\text{Sr}89} \varepsilon_s^{\text{Sr}90} + \varepsilon_c^{\text{Sr}89} \varepsilon_s^{\text{Y}90} \cdot f_2 - \varepsilon_s^{\text{Sr}89} \cdot \varepsilon_c^{\text{Sr}90} \cdot f_3 - \varepsilon_s^{\text{Sr}89} \cdot \varepsilon_c^{\text{Y}90} \cdot f_1 \cdot f_3} \quad (6)$$

The auxiliary parameter y can be defined and introduced:

$$y = \varepsilon_c^{\text{Sr}89} \cdot \varepsilon_s^{\text{Sr}90} + \varepsilon_c^{\text{Sr}89} \cdot \varepsilon_s^{\text{Y}90} \cdot f_2 - \varepsilon_s^{\text{Sr}89} \cdot \varepsilon_c^{\text{Sr}90} \cdot f_3 - \varepsilon_s^{\text{Sr}89} \cdot \varepsilon_c^{\text{Y}90} \cdot f_1 \cdot f_3 \quad (7)$$

Thus, the following equation is obtained for $A_{t_1}^{\text{Sr}90}$:

$$A_{t_1}^{\text{Sr}90} = \frac{N_{t_2}^s \cdot \varepsilon_c^{\text{Sr}89} - N_{t_1}^c \cdot \varepsilon_s^{\text{Sr}89} \cdot f_3}{y} \quad (8)$$

Substituting equation (8) into equation (5) we get the following expression:

$$A_{t_1}^{\text{Sr}89} = \frac{N_{t_1}^c (y + \varepsilon_c^{\text{Y}90} \cdot \varepsilon_s^{\text{Sr}89} \cdot f_1 \cdot f_3 + \varepsilon_c^{\text{Sr}90} \cdot \varepsilon_s^{\text{Sr}89} \cdot f_3) - N_{t_2}^s \cdot (\varepsilon_c^{\text{Sr}90} \cdot \varepsilon_c^{\text{Sr}89} + \varepsilon_c^{\text{Y}90} \cdot \varepsilon_c^{\text{Sr}89} \cdot f_1)}{\varepsilon_c^{\text{Sr}89} \cdot y} \quad (9)$$

where:

$$f_1 = 1 - \exp(-\lambda_{\text{Y}90} \cdot (t_1 - t_0)) \quad (10)$$

$$f_2 = 1 - \exp(-\lambda_{\text{Y}90} \cdot (t_2 - t_0)) \quad (11)$$

$$f_3 = \exp(-\lambda_{\text{Sr}89} \cdot (t_2 - t_1)) \quad (12)$$

The massic activities of ^{89}Sr and ^{90}Sr on the target date can be calculated as follows:

$$a_t^{\text{Sr}89} = \frac{A_{t_1}^{\text{Sr}89}}{r \cdot V} \cdot e^{\lambda_{\text{Sr}89} \cdot (t_1 - t)} \quad (13)$$

$$a_t^{\text{Sr}90} = \frac{A_{t_1}^{\text{Sr}90}}{r \cdot V} \cdot e^{\lambda_{\text{Sr}90} \cdot (t_1 - t)} \quad (14)$$

9.2. COMBINED UNCERTAINTY

The method for calculating the standard uncertainty of ^{89}Sr and ^{90}Sr measurements on the target/sampling date is given in Appendix VI. The combined uncertainty is calculated by the spreadsheet method according to the ISO Guide to the Expression of Uncertainty in Measurement (1995), and the EURACHEM/CITAC Guide (2012). The results of the calculations are given in Table 5 in Appendix VII.

Typically, results of ^{89}Sr and ^{90}Sr determination are comparable with the assigned values (see in Appendix VIII). Standard uncertainties are smaller for higher activities of ^{89}Sr (1–2%). In the case of ^{90}Sr , lower activities and spectral interferences result in larger standard uncertainties (10–15%). Pulses due to ^{90}Sr in Cerenkov counting mode have little effect on the spectra of ^{89}Sr (the efficiency of ^{90}Sr is much lower than that of ^{89}Sr in the Cerenkov counter) while pulses of ^{90}Sr and ^{89}Sr are mutually interfering in the LS spectra (their efficiencies are comparable).

9.3. DECISION THRESHOLD

In accordance with ISO 11929:2010, the decision threshold, a^* , is obtained from the following equation:

$$a^* = k_{1-\alpha} \cdot u(0) \quad (15)$$

where:

$u(0)$ is the uncertainty of the measurement when the activity of the analyte is equal to zero, and $k_{1-\alpha} = 1.645$ is chosen referring to the probability that error of the first kind (wrongly rejecting the hypothesis that radioactive strontium is present) is not greater than 5%.

In equation (15), $u(0)$ has to be calculated for each analyte using the general expression of the standard uncertainty. In the present case, we do not have an explicit expression for the activity (and the uncertainty) of the analytes. The activities of ^{89}Sr and ^{90}Sr are expressed as a function of two major input parameters, i.e. the count rates in Cerenkov and LS counters. Because of the great number of input parameters, the uncertainty is calculated by the spreadsheet method. The same spreadsheet can be used to determine the $u(0)$ and thus the decision threshold.

For ^{90}Sr , the value of $u(0)$ can be calculated in the following way:

From equation (8) with the assumption that $A_{t1}^{\text{Sr}90} = 0$, we obtain:

$$N_{t2}^s \cdot \varepsilon_c^{\text{Sr}89} = N_{t1}^c \cdot \varepsilon_s^{\text{Sr}89} \cdot f_3 \quad (16)$$

Thus, the count rate ratio is constant and can be determined according to equation (17):

$$N_{t2}^s / N_{t1}^c = \varepsilon_s^{\text{Sr}89} \cdot f_3 / \varepsilon_c^{\text{Sr}89} \quad (17)$$

This expression is introduced into the modified spreadsheet to calculate the value of N_{t2}^s from N_{t1}^c , thus the activity of ^{90}Sr becomes zero. The value of N_{t1}^c can be selected by iteration so that the value of $A_{t1}^{\text{Sr}89}$ equals with the actual activity of ^{89}Sr measured in the sample. The value $u(0)$ belonging to $A_{t1}^{\text{Sr}90} = 0$ is taken from the spreadsheet.

For ^{89}Sr , the value of $u(0)$ can be calculated in a similar way.

From equation (9) with the assumption that $A_{t1}^{Sr89} = 0$, we obtain equation (18):

$$N_{t1}^c \cdot (y + \varepsilon_c^{Y90} \cdot \varepsilon_s^{Sr89} \cdot f_1 \cdot f_3 + \varepsilon_c^{Sr90} \cdot \varepsilon_s^{Sr89} \cdot f_3) = N_{t2}^s \cdot (\varepsilon_c^{Sr90} \cdot \varepsilon_c^{Sr89} + \varepsilon_c^{Y90} \cdot \varepsilon_c^{Sr89} \cdot f_1) \quad (18)$$

Thus, the count rate ratio can be determined according to equation (19):

$$N_{t2}^s / N_{t1}^c = \frac{(y + \varepsilon_c^{Y90} \cdot \varepsilon_s^{Sr89} \cdot f_1 \cdot f_3 + \varepsilon_c^{Sr90} \cdot \varepsilon_s^{Sr89} \cdot f_3)}{(\varepsilon_c^{Sr90} \cdot \varepsilon_c^{Sr89} + \varepsilon_c^{Y90} \cdot \varepsilon_c^{Sr89} \cdot f_1)} \quad (19)$$

This expression is introduced into the modified spreadsheet to calculate N_{t2}^s , and N_{t1}^c can be selected by iteration so that the value of A_{t1}^{Sr90} equals with the actual activity of ^{90}Sr measured in the sample. The value $u(0)$ belonging to $A_{t1}^{Sr89} = 0$ is taken from the spreadsheet.

Using this calculation approach, the decision threshold for each radioactive strontium isotope is obtained as a function of the activity of the other radioactive strontium isotope. The method takes into consideration that the signals due to ^{90}Sr are superposed on the “background” signals that are produced basically from ^{89}Sr , and vice versa.

Results of calculations of the decision threshold are shown for the example of one double spiked milk sample analysed by the rapid method in Table 5 in Appendix VII.

9.4. DETECTION LIMIT

In accordance with ISO 11929:2010, the detection limit $a^\#$ is calculated by the following equation (20).

Assuming $\alpha = \beta$ then $k_{1-\alpha} = k_{1-\beta} = k$,

$$a^\# = 2 \cdot k_{1-\alpha} u(0) + \frac{k_{1-\beta}^2}{a_t^{Sr}} (u^2(a_t^{Sr}) - u^2(0)) \quad (20)$$

where a_t^{Sr} refers to a_t^{Sr90} or a_t^{Sr89} , respectively, and $u(0)$ refers to the uncertainty for ^{90}Sr or ^{89}Sr , respectively.

Results of calculations of uncertainties and determination of detection limit are shown for the example of one double spiked milk sample analysed by the rapid method in Table 5 in Appendix VII.

The decision threshold is higher for ^{90}Sr than for ^{89}Sr when the $^{89}\text{Sr}/^{90}\text{Sr}$ activity ratio at the time of measurement is high (in the present case between 10 and 20). The corresponding values are about 150 and 5 Bq kg⁻¹, respectively, revealing the spectral interferences in both spectra, i.e. in Cerenkov and LS counting modes. ^{90}Sr pulses are detected in the presence of ^{89}Sr pulses of much higher activity in the same spectrum ROIs. Detection limits are about double the relevant decision threshold values. See spectra in Appendix V.

10. VALIDATION OF THE PROCEDURE

Due to the lack of reference materials that contain certified amounts of ^{89}Sr and ^{90}Sr in milk the following working protocol was designed in order to perform the method validation procedure as part of the development of this procedure:

- Characterization of milk powder samples by measurement of gamma-emitting natural radionuclides, e.g., natural ^{40}K , as well as the stable Sr content of the samples;
- Preparation of spiked milk powder samples with four different activity ratios using standard solutions of ^{89}Sr , ^{90}Sr and ^{137}Cs ;
- Homogeneity tests based on gamma-ray spectrometric determination of ^{137}Cs , and the total beta counting of the sample;
- Application of the rapid method for the analysis of the spiked water samples (where homogeneity is inherently assured) to determine repeatability and trueness at the IAEA Terrestrial Environment Laboratory;
- Application of the rapid method for the analysis of spiked milk samples to determine repeatability and trueness at the IAEA Terrestrial Environment Laboratory;
- Application of the rapid method for the analysis of spiked milk samples to determine reproducibility and trueness at selected laboratories of the ALMERA network;
- Comparative evaluation of the results based on the measured and the assigned values taking into account all possible sources of measurement uncertainties as well as uncertainties related to the preparation of the spiked samples.

10.1. CHARACTERIZATION OF MILK POWDER SAMPLES

Two creamy milk powder samples which contained 26% fat were purchased at a supermarket in Hungary, in 2009 and 2010. Prior to using the milk powder samples, they were characterized to determine the background data, namely the ^{40}K , ^{137}Cs , ^{90}Sr and stable Sr content.

^{137}Cs and ^{40}K in milk powder were measured by gamma-ray spectrometry. For the determination of ^{90}Sr in the blank milk powder, 0.5 kg of milk powder was ashed at 600°C after addition of 10 mg of Sr carrier, and then digested with 65% nitric acid and a few ml of 25% H_2O_2 . ^{90}Sr in the solution was separated using a Sr resin and measured by LSC (Moreno et al.) [15].

The results are given in Table 2. The stable Sr concentrations in the blank milk powder were measured by ICP-MS. The values were used for calculation of the chemical recovery of Sr in spiked milk samples.

TABLE 2. RESULTS OF DETERMINATION OF CAESIUM-137, POTASSIUM-40, STRONTIUM-90 AND STABLE STRONTIUM IN BLANK MILK POWDER

Sample name	Purchase year of sample	Massic activity, Bq kg^{-1} (as dry weight)			Concentration of stable Sr (mg kg^{-1})
		^{137}Cs	^{90}Sr	^{40}K	
M1	2009	$2.04 \pm 0.09^*$	$0.28 \pm 0.02^*$	$374 \pm 3^*$	$2.74 \pm 0.06^*$
M2	2010	$0.88 \pm 0.14^*$	$0.28 \pm 0.03^*$	$331 \pm 18^*$	$3.76 \pm 0.10^*$

* combined uncertainty ($k=1$)

10.2. PREPARATION OF SPIKED MILK POWDER SAMPLES

There are no available milk samples contaminated with ^{89}Sr and ^{90}Sr that could be used to validate the rapid simultaneous procedure. Therefore, six spiked milk powder samples were prepared using the two characterized milk powders (M1 and M2). Four of the milk powder samples were spiked with standard solutions of ^{89}Sr and ^{90}Sr in different $^{89}\text{Sr}/^{90}\text{Sr}$ activity ratios. Milk powders were also spiked with ^{137}Cs as it is one of the interfering radionuclides in an emergency situation.

A list of prepared spiked milk powders is shown below:

- Sample M1-1: spiked with ^{137}Cs and ^{89}Sr ;
- Sample M1-2: spiked with ^{137}Cs and ^{90}Sr ;
- Sample M1-3: spiked with ^{137}Cs , ^{89}Sr and ^{90}Sr ($^{89}\text{Sr}/^{90}\text{Sr}$ activity ratio: 5.8);
- Sample M1-4: spiked with ^{137}Cs , ^{89}Sr and ^{90}Sr ($^{89}\text{Sr}/^{90}\text{Sr}$ activity ratio: 10.6);
- Sample M2-1: spiked with ^{137}Cs , ^{89}Sr and ^{90}Sr ($^{89}\text{Sr}/^{90}\text{Sr}$ activity ratio: 5.0);
- Sample M2-2: spiked with ^{137}Cs , ^{89}Sr and ^{90}Sr ($^{89}\text{Sr}/^{90}\text{Sr}$ activity ratio: 10.2);
- Blank.

Activity ratios refer to the reference time.

2 kg aliquots of milk powder were spiked with ^{89}Sr , ^{90}Sr and ^{137}Cs layer by layer (each layer was about 1 kg) using a 50 μL dispenser with 8 pin tips. After drying at 40°C for 4 hours in a drying oven, the spiked milk powder was homogenised using 1.5 kg of stainless steel balls in a stainless steel vessel of a large volume TURBULA mixer for about 48 hours. As soon as the sample was homogenised, subsamples were bottled in 1 L plastic bottles.

Ten bottles of subsamples were prepared for each spiked milk powder. For the homogeneity testing, 40 g of bulk sample were taken from each bottle and 5 g pellets were prepared from three subsamples from each bulk material. The pellets were prepared under a pressure of five tons (SpectropressTM, Chemplex Industries, Inc.) in order to reduce probable geometry error.

10.3. HOMOGENEITY TESTS BASED ON GAMMA-RAY SPECTROMETRY AND GROSS BETA COUNTING

In order to determine the homogeneity of the samples gross beta and gamma-ray spectrometric measurements were performed. The pellet samples were measured using a low background gas proportional counter (LB 5500, Tennelec, USA) for 30 minutes to get gross beta counts and using a 15% relative efficiency n-type HPGe detector for 10000 seconds to get total counts at 661.7 keV peak of ^{137}Cs . Each sample was measured three times to check the repeatability of the measurement results. The deviation of masses between pellet samples was within $\pm 1\%$.

The homogeneity test results of all spiked milk samples are summarized as an ANOVA table (Table 3). The relative standard deviations (s_r) of three replicate measurements of six spiked samples using the low background gas proportional counter and the gamma-ray spectrometer were less than 3%. The repeatability (S_{bb}) between subsamples was lower than the relative standard deviations (s_r) of three replicates measurements for each subsample. The homogeneity uncertainties of samples were less than 0.3%. These results imply that the homogeneity of the spiked milk powders was acceptable for the validation of the rapid

method of ^{89}Sr and ^{90}Sr in milk.

Therefore, the assigned values of ^{89}Sr and ^{90}Sr in spiked milk powders were determined based on the certificate values of ^{89}Sr and ^{90}Sr standard solutions used for spiking of milk powder. The results of repeated analysis of ^{89}Sr and ^{90}Sr in mixed standard solutions with different ratios are given in Table 6 in Appendix VIII. The uncertainties of massic activity theoretical assigned values of ^{89}Sr and ^{90}Sr were calculated based on the uncertainties of ^{89}Sr and ^{90}Sr standard solutions, the uncertainty of dilution factor of each standard solution and heterogeneity of the spiked milk powder sample. Tables 7–9 in Appendix VIII list the calculated assigned values and uncertainties that varied within the range of 0.3% to 4.3%.

TABLE 3. ANOVA RESULTS OF SPIKED MILK POWDER SAMPLES

Sample	M1-1		M1-2		M1-3		M1-4		M2-1		M2-2	
	Between*	Within**	Between	Within	Between	Within	Between	Within	Between	Within	Between	Within
SS	7.25	9.44	2.26	6.00	9.53	22.4	18.4	51.1	113.9	353.1	992	3669
Df	7	16	9	20	9	20	9	20	9	20	9	20
MS	1.04	0.59	0.25	0.30	1.06	1.12	2.04	2.58	12.6	17.7	100.2	183.4
F	1.76		0.84		0.95		0.79		0.72		0.601	
F crit	2.66		2.39		2.39		2.39		2.39		2.93	
S _{bb} (%)	0.39		0.13		0.14		0.42		0.57		1.11	
S _r (%)	0.77		0.55		1.06		1.6		1.84		3.01	
U _{bb} (%)	0.09		0.06		0.11		0.16		0.19		0.31	

*: Between bottles

**: Within bottle

- SS Sum of Squares
MS Mean Squares (Variance) SS/df
df degree of freedom
F ratio of the variances, $MS_{\text{between}}/MS_{\text{within}}$
F_{crit} theoretical values of F from the F-test table according to the degrees of freedom at P=5%.
S_{bb} repeatability between sub-samples
S_r repeatability of 3 replicates measurement for a sub-sample
u_{bb} uncertainty due to the homogeneity calculated as follows

$$u_{bb} = \sqrt{\frac{MS_{\text{within}}}{n} \cdot 4 \sqrt{\frac{2}{df_{MS_{\text{within}}}}}}$$

where:

 n is the number of observations.

The quality control measurements of the procedure were carried out by the analysis of:

- Double spiked water samples;
- Single spiked milk samples (M1-1 and M1-2 milk spiked with ^{89}Sr and ^{90}Sr , respectively);
- Double spiked milk samples M1-3, M1-4, M2-1, M2-2.

The results were evaluated in terms of relative bias and repeatability.

Results of repeated measurements of double spiked water samples are shown in Table 6 in Appendix VIII. Results of repeated measurements of single spiked milk samples are shown in Table 7 in Appendix VIII. Results of repeated measurements of double spiked milk samples are shown in Tables 8 and 9 in Appendix VIII.

The control charts showing the repeatability of the rapid method are depicted in Fig. 4 and 5 in Appendix VIII, and the major performance parameters (relative bias, repeatability limits) are shown in Tables 10 and 11 in Appendix VIII.

10.4. LINEARITY, RANGE OF MEASUREMENT

The method was tested using samples with a wide range of activities and activity ratios in order to represent a real emergency situation following a nuclear reactor accident. The massic activities of ^{89}Sr and ^{90}Sr varied in the range of 455–6380 Bq kg⁻¹ and 78–628 Bq kg⁻¹, respectively, while $^{89}\text{Sr}/^{90}\text{Sr}$ activity ratios at the time of measurement were between 2 and 35. Linearity of the measurements under the experimental conditions was proven by the good agreement between the measured data and the reference values (assigned values). It is likely that the linearity of the method is valid over a much broader range than that used in this study. It is expected that the range would cover activity starting from the detection limit up to high activities where the dead time becomes high and dead time corrections are necessary (above 1% dead time level).

10.5. RELATIVE BIAS OF THE METHOD (TRUENESS)

For samples M1-1, M1-2, M1-3, M1-4, M2-1, M2-2; 4, 3, 5, 5, 10 and 10 replicates, respectively, were analyzed in order to evaluate the trueness. The bias δ and the relative bias $\delta(\%)$ of the method are calculated by the following formula:

$$\delta = \bar{X} - C \quad (21)$$

$$\delta(\%) = \frac{\bar{X} - C}{C} \times 100 \quad (22)$$

The relative biases of repeatability and reproducibility measurements are summarized in Tables 6–8 in Appendix VIII and Tables 12–13 in Appendix IX.

10.6. REPEATABILITY (WITHIN-RUN PRECISION)

The repeatability limit was estimated by analyzing the replicates of the spiked samples of “known” massic activities of ^{89}Sr and ^{90}Sr in repeatability conditions (instrument, analyst, shortest time, etc.). The repeatability limit was calculated by the following formula:

$$r_L = S_r \times 2.8 \quad (23)$$

Acceptance criteria: The method can be accepted if $|\delta| < 2.8 S_r$.

Results of repeatability tests are summarized in Tables 10 and 11 in Appendix VIII.

10.7. REPRODUCIBILITY (BETWEEN-RUN PRECISION)

The reproducibility limit was estimated by analyzing several replicates of the spiked milk samples of known massic activity using the same method in various laboratories. The reproducibility limit was calculated by the following formula:

$$R_L = S_R \times 2.8 \quad (24)$$

Acceptance criteria: the method can be accepted if $|\delta| < 2.8 S_R$.

Reproducibility tests were performed with the participation of seven laboratories of the ALMERA network. Results of reproducibility tests are summarized in Table 14 in Appendix IX.

10.8. DETECTION LIMIT

Results of calculation of the detection limits for three sample types spiked with different activity ratios of $^{89}\text{Sr}/^{90}\text{Sr}$ are summarized in Table 15 in Appendix X.

11. RESULTS OF METHOD VALIDATION

Before applying the rapid method for the analysis of milk samples, the new counting approach using Cerenkov and scintillation counting was tested with mixed standard solutions of different $^{89}\text{Sr}/^{90}\text{Sr}$ activity ratios (from 2.5 to 46). ^{89}Sr and ^{90}Sr were separated from ^{90}Y with Sr resin. According to the results shown in Table 6 in Appendix VIII, relative biases for ^{89}Sr and ^{90}Sr were acceptably low, within 7% up to the $^{89}\text{Sr}/^{90}\text{Sr}$ activity ratio of 25. Increasing the activity ratio to 40, the relative bias of ^{90}Sr determination became 15% which can be still regarded as acceptable in an emergency situation.

In order to validate the rapid chemical procedure, single spiked milk samples were analyzed using liquid scintillation technique. Results of analysis shown in Table 7 in Appendix VIII reveal good agreement between measured and assigned values both for ^{89}Sr and ^{90}Sr , the relative biases being less than 6% and 15%, respectively. Higher relative biases of ^{90}Sr determination are due to lower activities.

In order to validate the rapid method, double spiked milk samples of various $^{89}\text{Sr}/^{90}\text{Sr}$ activity ratios in the range of 2–35 were analyzed. Results of the analysis of a total of 30 samples are summarized in Tables 7–9 in Appendix VIII and show that the method is fit for purpose, i.e.:

- The relative biases of the results of ^{89}Sr and ^{90}Sr in M1-3 and M1-4 subsamples were less than about $\pm 20\%$;
- The relative biases of the results of ^{89}Sr and ^{90}Sr in M2-1 and M2-2 subsamples ranged from $\pm 0.2\%$ to $\pm 20\%$ and from $\pm 3\%$ to $\pm 41\%$, respectively, increasing with increasing activity ratio of $^{89}\text{Sr}/^{90}\text{Sr}$.

Typically, biases were higher in case of higher $^{89}\text{Sr}/^{90}\text{Sr}$ activity ratios due to the smaller activity of ^{90}Sr . In order to receive acceptable high measurement accuracies, e.g., to keep relative biases of ^{90}Sr determination below 30%, the $^{89}\text{Sr}/^{90}\text{Sr}$ activity ratio (at the time of measurement) should not exceed the value of 30.

The relative biases measured in milk samples were higher than those of standard solutions showing that the chemical processing of real milk samples contributes to the uncertainty of the method.

Chemical recoveries obtained by processing milk samples (M2) were relatively high with average values of 71 and 75% in case of M2-1 and M2-2 samples, respectively.

The results of samples M1 and M2 indicate that the rapid method does not show matrix dependence.

According to Tables 9 and 10 in Appendix VIII and Table 14 in Appendix IX, the test results met the requirements of repeatability and reproducibility in each studied case, i.e. the repeatability and reproducibility bias $|\delta|$ was smaller than the repeatability limit (r_L) and the reproducibility limit (R_L), respectively.

According to the criteria of ISO/TS 21748:2004, the rapid method performs well in repeatability tests for both nuclides with one exception in case of ^{90}Sr measurement in M2-2 sample (PI% = 31%). This exception can be interpreted by the high $^{89}\text{Sr}/^{90}\text{Sr}$ activity ratio (46) that is the highest tested value and shows the limitation of the rapid method in extreme cases when the $^{89}\text{Sr}/^{90}\text{Sr}$ activity ratio exceeds the value of 30.

According to Table 15 in Appendix X, the detection limit of ^{89}Sr and ^{90}Sr determination in fresh milk varied in the range of 2 to 5 Bq L⁻¹ and 2 to 70 Bq L⁻¹, respectively when the activity ratio of $^{89}\text{Sr}/^{90}\text{Sr}$ varied in the range of 3–20.

12. TEST REPORT

The test report must conform to ISO 17025 requirements and should contain the following information:

- Sample code;
- Sampling date (if available);
- Date of separation of the analyte;
- Reference date of the tracer solution;
- Analysis request code;
- Responsible person;
- Reference to the measurement and evaluation procedure used;
- Identification of the sample;
- Units in which the results are expressed.

Unless otherwise required, the test result should be stated together with the expanded uncertainty u [as defined in the International Vocabulary of Basic and General terms in Metrology, 2nd ed., ISO 1993] calculated using a coverage factor k . The following form is recommended:

$a \pm u(a)$ with the associated k value (coverage factor). Results in the present document were calculated at the uncertainty level $k=1$.

13. CONCLUSION

A rapid procedure was developed and tested for the simultaneous determination of ^{89}Sr and ^{90}Sr in milk samples using Cerenkov and liquid scintillation counting techniques. Strontium is radiochemically separated using sequential ion exchange, extraction chromatography and precipitation operations.

Chemical recoveries are acceptably high (in average 70-75%). Results are obtained within one day.

According to the repeatability and reproducibility tests, the measurement process is considered to perform adequately.

The method is recommended for application in the analysis of milk samples contaminated by elevated levels of the radionuclides, especially in emergency situations.

APPENDIX I

CAPACITY FACTORS (k') OF VARIOUS ALKALI AND ALKALINE EARTH METAL IONS ON SR RESIN IN NITRIC ACID

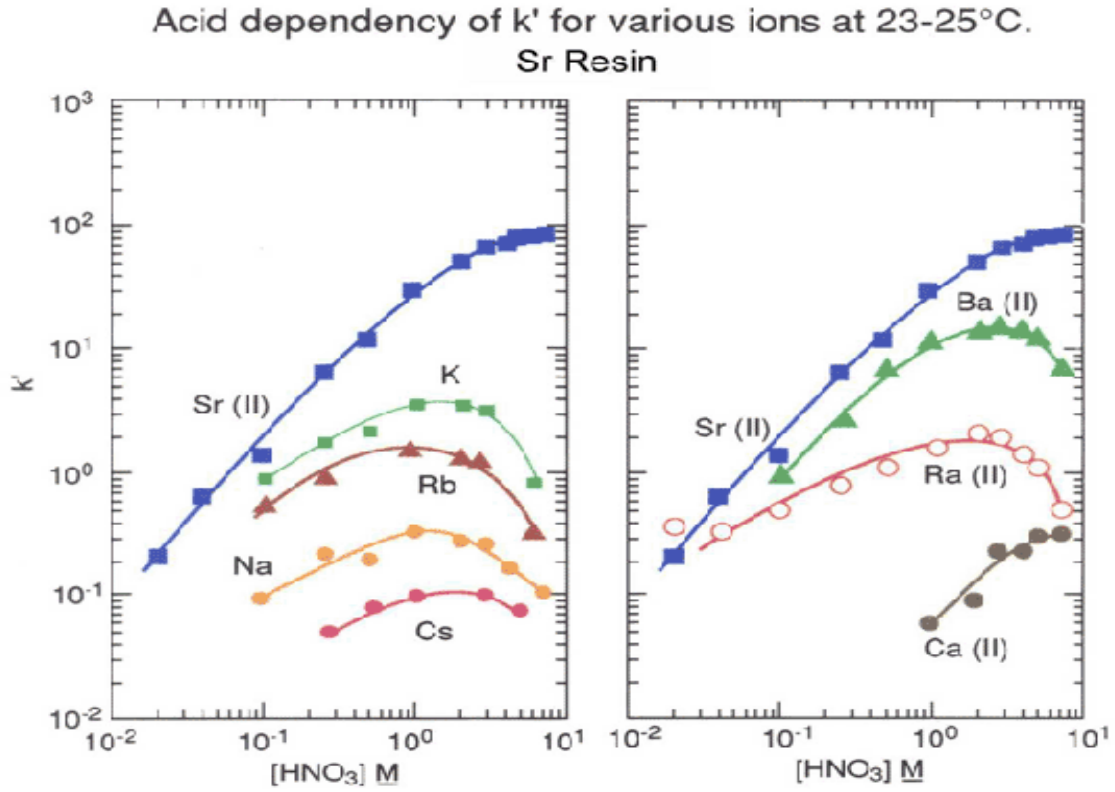


FIG. 1. Acid dependency of the capacity factor (k') for various ions at 23–25°C on Sr resin according to the Eichrom Technologies homepage (www.eichrom.com).

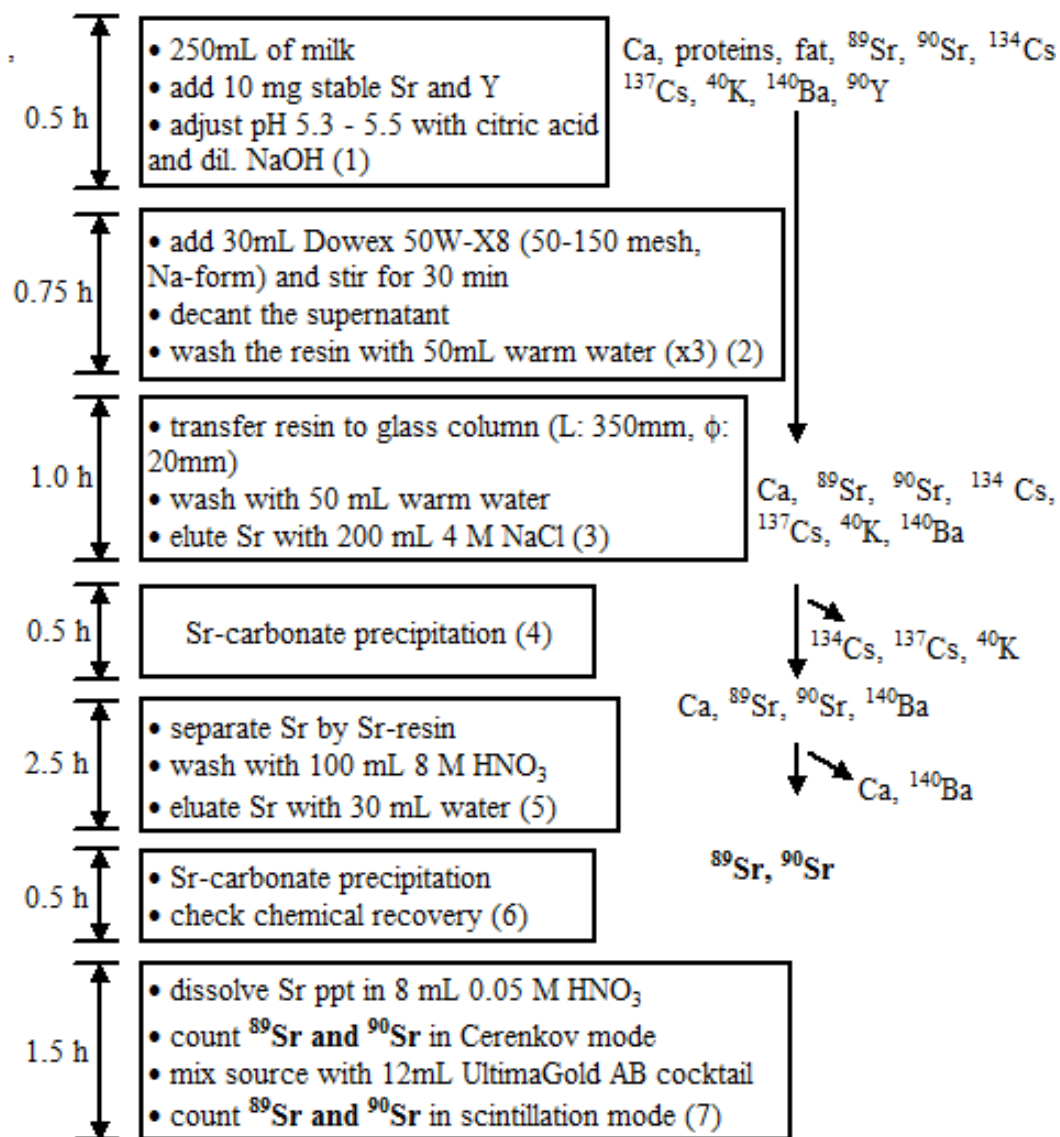
APPENDIX II

TABLE 4. DECAY DATA OF STRONTIUM ISOTOPES AND YTTRIUM-90
(SOURCES: [16] AND [17])

Radionuclide	Half-life	Disintegration modes	$E_{\beta \text{ Max}}$ (keV)	Intensity (%)
^{90}Sr	$28.80 \pm 0.07 \text{ y}$	$\beta^- : 100\%$	545.9	100
^{90}Y	$2.664 \pm 0.0013 \text{ d}$	$\beta^- : 100\%$	2279.8	99.98
^{89}Sr	$50.57 \pm 0.03 \text{ d}$	$\beta^- : 100\%$	1495.1	99.99
$^{91}\text{Sr}^*$	9.7 h	$\beta^- : 100\%, \gamma$	2686	29
$^{92}\text{Sr}^*$	2.7 h	$\beta^- : 100\%, \gamma$	1383	90

APPENDIX III

FLOWCHART FOR RAPID SEPARATION OF STRONTIUM FROM MILK USING CATION EXCHANGE AND EXTRACTION CHROMATOGRAPHY



APPENDIX IV

PREPARATION OF CHEMICAL REAGENTS

- The following nitric solutions are prepared by dilution of 65% nitric acid: 8M HNO₃, 3M HNO₃, 1M HNO₃, 0.05M HNO₃.
- 1M HCl is prepared by dilution of 32% HCl.
- 6M NaOH: dissolve 240 g NaOH pellets in de-ionized water and filled up to 1 L.
- 0.25M citric acid solution: dissolve 52.5 g citric acid monohydrate in 1 L de-ionized water.
- Sr carrier (10 mg Sr mL⁻¹ in 1M HNO₃): dissolve 2.415 g Sr(NO₃)₂ in 1M HNO₃ and filled up to 100 mL.
- Y carrier (10 mg Y mL⁻¹ in 1M HNO₃): dissolve 4.307 g Y(NO₃)₃ 6H₂O in 1M HNO₃ and filled up to 100 mL.

APPENDIX V

TYPICAL CERENKOV AND LIQUID SCINTILLATION SPECTRA

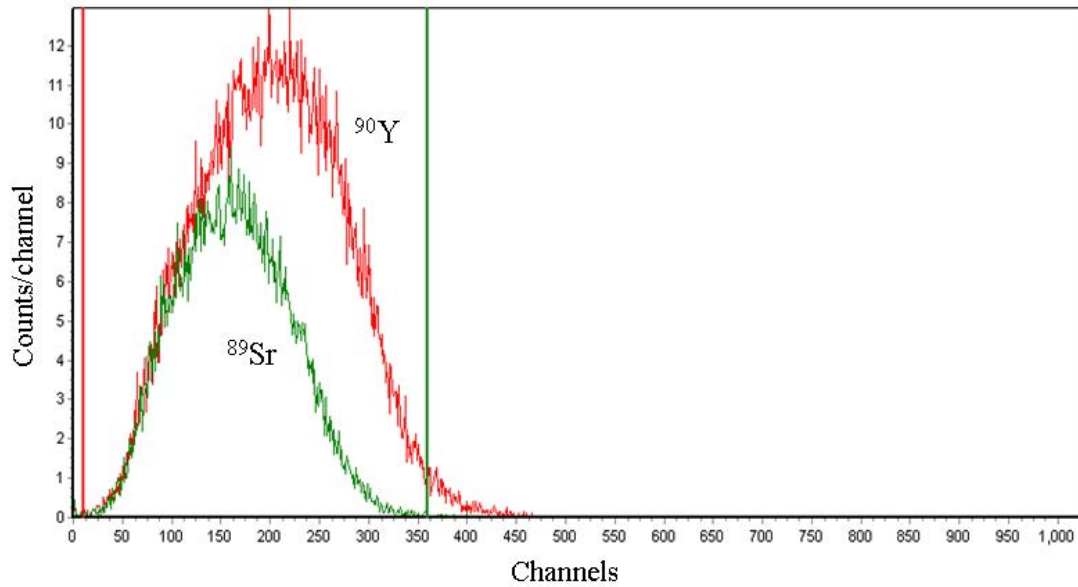


FIG. 2. Typical Cerenkov spectra of ^{89}Sr and ^{90}Y obtained in the ROI 10–360 channels using a Quantulus LSC.

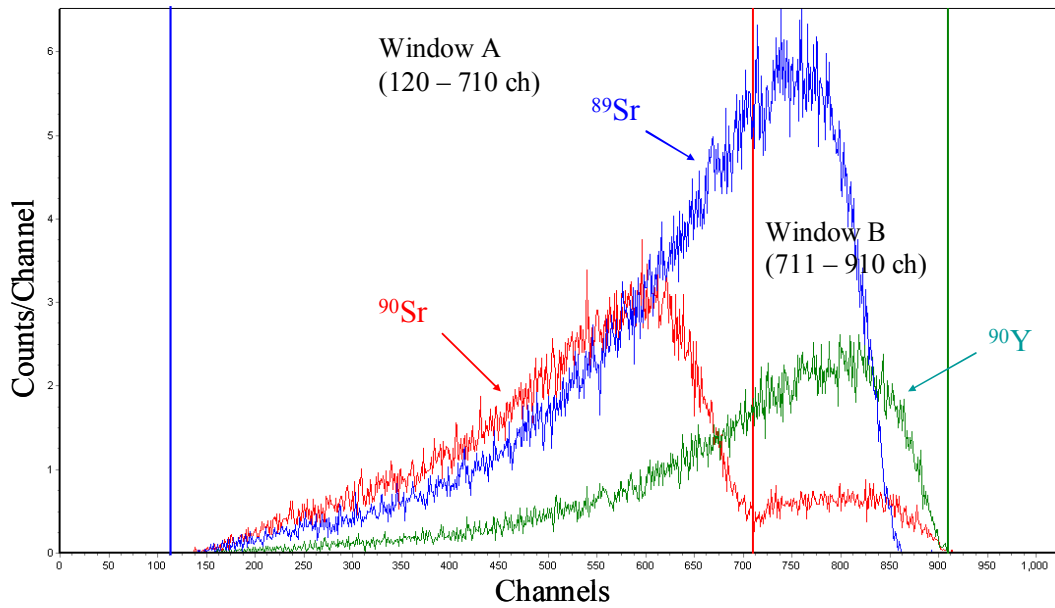


FIG. 3. Typical liquid scintillation spectra of ^{89}Sr , ^{90}Sr and ^{90}Y obtained in the ROI 120–910 channels using a Quantulus LSC.

APPENDIX VI

CALCULATION OF STANDARD UNCERTAINTY OF STRONTIUM-89 AND STRONTIUM-90 ON TARGET/SAMPLING DATE

VI.1. Calculation of individual uncertainties

VI.1.1. Uncertainty in weighing of sample mass

The uncertainty associated with the mass of sample is estimated using the data from the calibration certificate of the analytical balance and the manufacturer's recommendations on the uncertainty estimation of the balance. If it is assumed that a balance with 0.1 mg uncertainty is used for weighing the sample and the distribution is assumed to show a rectangular distribution, the standard uncertainty in the weighing of the sample mass can be calculated by the following formula:

$$\frac{0.1}{\sqrt{3}} = 0.058 \text{ mg} \quad (\text{VI-1})$$

The contribution for the linearity has to be accounted for twice, once for the tare and once for the gross mass (tare + sample), leading to an uncertainty $u(V)$ of

$$u(V) = \sqrt{2 \times (0.058)^2} = 0.082 \text{ mg} \quad (\text{VI-2})$$

VI.1.2. Uncertainty of net count rates of analytes

The uncertainty of net count rates in Cerenkov and analogously for LS counting can be calculated from the instrumental values of count rates and uncertainties as follows:

$$N_{t1}^c = N_{G1}^c - N_{B1}^c \quad (\text{VI-3})$$

$$u(N_{t1}^c)^2 = u^2(N_{G1}^c) + u^2(N_{B1}^c) \quad (\text{VI-4})$$

VI.1.3. The uncertainties for decay correction factors $u(f)$ can be calculated as follows:

$$u(f_1) = (t_1 - t_0) \cdot e^{-\lambda_{Y90}(t_1 - t_0)} \cdot u(\lambda_{Y90}) \quad (\text{VI-5})$$

$$u(f_2) = (t_2 - t_0) \cdot e^{-\lambda_{Y90}(t_2 - t_0)} \cdot u(\lambda_{Y90}) \quad (\text{VI-6})$$

$$u(f_3) = (t_2 - t_1) \cdot e^{-\lambda_{Sr89}(t_2 - t_1)} \cdot u(\lambda_{Sr89}) \quad (\text{VI-7})$$

Uncertainties of counting times and date and time values are neglected.

Uncertainties of other decay correction factors can be calculated in a similar way.

VI.1.4. Other uncertainty components

Uncertainties of counting efficiencies are calculated in the course of the instrumental calibration procedures according to the law of uncertainty propagation and very often as the standard deviation of repeated measurements.

Uncertainty calculation of the chemical recovery (r) is described in detail in the paper of Moreno et al. (2004).

Uncertainty of the auxiliary parameter (y) is calculated according to the law of uncertainty propagation.

VI.2. Combined uncertainty in the determination of ^{89}Sr and ^{90}Sr

The combined uncertainties of the ^{89}Sr and ^{90}Sr massic activities are calculated using the spreadsheet method according to ISO Guide, 1995, EURACHEM/CITAC Guide, 2012. The spreadsheet procedure takes advantage of an approximate numerical method of differentiation, and requires knowledge of the calculation of the final result (equations 3, 9, 13 and 14) and of the numerical values of the parameters and their individual uncertainties.

The method also provides data about the individual sensitivity parameters of the uncertainty budget.

APPENDIX VII

RESULTS OF CALCULATION OF COMBINED STANDARD UNCERTAINTY, DECISION THRESHOLD AND DETECTION LIMIT

TABLE 5. SUMMARY OF RESULTS OF ANALYSES OF SPIKED MILK SAMPLE M2-1-4(A)

Parameters	unit	Parameter value	Standard unc.	Relative standard unc.	Percent contribution to combined standard unc.	
					⁸⁹ Sr	⁹⁰ Sr
Sample mass						
V _w	g	33,31	0,00008	2,45E-06		
f _{d/w}	kg/kg	0,9990	0,000001	1,00E-06	negligible	negligible
V	g	33,28	8,81E-05	2,65E-06	negligible	negligible
Sample counting						
N _{G1}	cpm	4705,9	15,434	3,28E-03		
NB _{t1}	cpm	7,257	0,925	1,27E-01		
N _{t1} ^c	cps	78,311	0,258	3,29E-03	15.2	10.1
N _{G2}	cpm	14061,0	37,757	2,69E-03		
NB _{t2}	cpm	8,358	0,940	1,12E-01		
N _{t2} ^s	cps	234,2	0,629	2,69E-03	0.59	7.8
Decay corrections						
t		2010.07.01 12:00				
t ₀		2010.04.12 9:00				
t ₁		2010.04.12 22:16				
t ₂		2010.04.13 12:28				
λ _{Sr89}	1/d	0,0137	1,90E-05	1,39E-03		
λ _{Sr90}	1/d	0,0000666	6,82E-08	1,04E-03		
λ _{Y90}	1/d	0,260	1,27E-04	4,87E-04		
f ₁		0,134	6,06E-05	4,53E-04	negligible	negligible
f ₂		0,257	1,08E-04	4,18E-04	negligible	negligible
f ₃		0,992	1,12E-05	1,12E-05	negligible	negligible
f _{Sr89}		0,336	5,08E-04	1,51E-03	1.9	negligible
f _{Sr90}		0,995	5,40E-06	5,43E-06	negligible	negligible
Counting efficiencies						
ε _c ^{Sr89}		0,347	0,002	5,76E-03	46	36
ε _c ^{Sr90}	-	0,017	0,003	1,76E-01	0.4	negligible
ε _c ^{Y90}	-	0,581	0,005	8,61E-03	0.02	negligible
ε _s ^{Sr89}	-	0,972	0,0067	6,89E-03	3.3	44.5
ε _s ^{Sr90}	-	0,921	0,009	9,77E-03	negligible	negligible
ε _s ^{Y90}	-	0,997	0,003	3,01E-03	negligible	negligible
Chemical recovery						
m _{car}	g	0,0100	1,11E-05	1,11E-03		
C _{Sr0}	mg/kg	3,760	9,50E-02	2,53E-02		
m _{Sr0}	g	0,0101	1,15E-05	1,14E-03		
m _{Sr} /m _{SrCO3}	-	0,594	0,00E+00	0,00E+00		
m _{SrB}	g	0,049	1,00E-04	2,04E-03		
m _t	g	0,036	1,00E-04	2,77E-03		
m _{Srt}	g	0,0077	4,85E-05	6,28E-03		
r	-	0,764	4,87E-03	6,38E-03	32.5	0.2
y	-	0,317	4,82E-03	1,52E-02	0.1	1.2
Results						
A _{t1} ^{Sr89}	Bq	220,7				
A _{t1} ^{Sr90}	Bq	220,7	1,99	9,01E-03		
a _t ^{Sr89}	Bq kg-1, as dry mass	2914	32,41	1,11E-02		
a _t ^{Sr90}	Bq kg-1, as dry mass	711,4	96,21	1,35E-01		
Decision threshold						
⁸⁹ Sr	Bq kg-1, as dry mass	5.3				
⁹⁰ Sr	Bq kg-1, as dry mass	146				
Detection limit						
⁸⁹ Sr	Bq kg-1, as dry mass	11.2				
⁹⁰ Sr	Bq kg-1, as dry mass	298				

Remark: Results of the individual measurement M2-1-4A are slightly different from the results of the repeated measurements M2-1-4 (given as average ± standard deviation) in Table 9 in Appendix VIII. Input parameters are indicated by yellow or pink background colors, results of calculation are depicted on green background.

APPENDIX VIII

RESULTS OF REPEATABILITY MEASUREMENTS

TABLE 6. RESULTS OF REPEATED ANALYSIS OF STRONTIUM-89 AND STRONTIUM-90 IN MIXED STANDARD SOLUTIONS WITH DIFFERENT ACTIVITY RATIOS OF STRONTIUM-89/STRONTIUM-90 USING SEQUENTIAL CERENKOV AND SCINTILLATION COUNTING TECHNIQUES

Assigned values (Bq)		Experimental results (Bq)		Relative bias (%)		⁸⁹ Sr / ⁹⁰ Sr activity ratio in source
⁸⁹ Sr	⁹⁰ Sr	⁸⁹ Sr	⁹⁰ Sr	⁸⁹ Sr	⁹⁰ Sr	
87.1 ± 1.0	35.0 ± 0.1	86.2 ± 4.5	33.9 ± 4.1	-1.0	-3.1	2.5
131 ± 2	34.9 ± 0.1	131 ± 5	34.7 ± 4.6	0	-0.6	3.8
195 ± 2	30.4 ± 0.1	194 ± 5	30.0 ± 4.6	-0.5	-1.3	6.5
143 ± 2	10.21 ± 0.03	145 ± 3	10.5 ± 2.3	1.4	2.9	13.8
264 ± 3	10.21 ± 0.03	269 ± 5	10.9 ± 3.7	1.9	6.7	24.7
219 ± 2	4.76 ± 0.02	221 ± 4	5.47 ± 2.74	0.9	14.9	40.4

TABLE 7. RESULTS OF REPEATED ANALYSIS OF STRONTIUM-89 AND STRONTIUM-90 IN SINGLE SPIKED M1-1, M1-2 MILK SAMPLES DETERMINED BY RAPID METHOD

Subsample code	Chemical yield (%)	Measured massic activity (Bq kg ⁻¹)		Assigned value on reference date [*] (Bq kg ⁻¹)		Relative bias (%)	
		⁸⁹ Sr	⁹⁰ Sr	⁸⁹ Sr	⁹⁰ Sr	⁸⁹ Sr	⁹⁰ Sr
M1-1-1	80.5	399 ± 27	-			-5.5	
M1-1-2	30.6	403 ± 28	-	422 ± 18		-4.5	
M1-1-3	60.3	413 ± 29	-			-2.1	
M1-1-4	57.8	428 ± 30	-			1.4	
M1-2-1	36.0	-	109 ± 4				-8.4
M1-2-2	54.5	-	113 ± 4		119 ± 4		-5.0
M1-2-3	49.6	-	102 ± 4				-14.3
Average		411	108			-2.6	-9.2
SD		13	6				
RSD (%)		3.0	5.0				

*: reference date of assigned values: 01.07.2009

TABLE 8. RESULTS OF ANALYSIS OF STRONTIUM-89 AND STRONTIUM-90 IN DOUBLE SPIKED M1-3, M1-4 MILK SAMPLES DETERMINED BY RAPID METHOD

Subsample code	Chemical yield (%)	Measured massic activity (Bq kg ⁻¹)		Assigned value on reference date [*] (Bq kg ⁻¹)		Relative bias (%)		Activity ratio of ⁸⁹ Sr / ⁹⁰ Sr on measurement time
		⁸⁹ Sr ^{**}	⁹⁰ Sr ^{**}	⁸⁹ Sr	⁹⁰ Sr	⁸⁹ Sr	⁹⁰ Sr	
M1-3-1	89.4	447 ± 38	90.6 ± 16.7			-1.8	16.3	2.0
M1-3-2	93.2	435 ± 40	90.0 ± 17.7			-4.3	15.5	2.0
M1-3-3	74.4	438 ± 42	91.1 ± 18.8	455 ±	77.9 ±	-3.7	16.9	2.0
M1-3-4	90.1	443 ± 25	72.3 ± 9.0	13 ^{***}	2.3 ^{***}	-2.6	-7.1	2.3
M1-3-5	90.5	439 ± 25	69.3 ± 9.2			-3.5	-11.0	2.4
Average		440	82.7			-3.3	6.2	
SD		4.7	10.9					
RSD (%)		1.1	13.2					
M1-4-1	80.6	808 ± 35	88.0 ± 14.7			-2.2	13.3	3.4
M1-4-2	91.7	823 ± 48	95.8 ± 21.9			-0.4	23.3	3.5
M1-4-3	91.2	825 ± 48	90.3 ± 21.8	826 ±	77.7 ±	-0.1	16.2	3.8
M1-4-4	77.3	829 ± 29	75.0 ± 10.7	32 ^{***}	3.0 ^{***}	0.4	-3.5	4.2
M1-4-5	91.3	811 ± 30	74.4 ± 10.9			-1.8	-4.2	4.2
Average		819	84.7			-0.8	9.0	
SD		9.2	9.6					
RSD (%)		1.1	11.3					

*: Reference date of assigned values: 01.07.2009

** : Average and standard deviation of 5 replicate measurements

*** : Combined standard uncertainty (k=1)

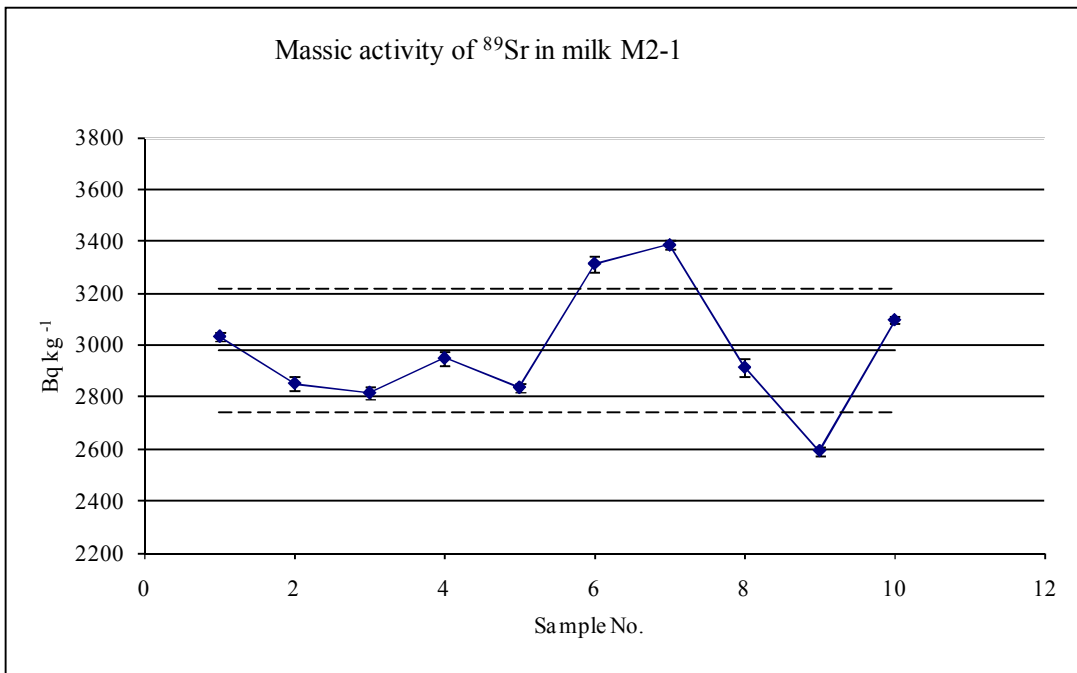
TABLE 9. RESULTS OF ANALYSIS OF STRONTIUM-89 AND STRONTIUM-90 IN DOUBLE SPIKED M2-1, M2-2 MILK SAMPLES DETERMINED BY RAPID METHOD

Subsample code	Chemical yield (%)	Measured massic activity (Bq kg ⁻¹)		Assigned values on reference date*		Relative bias (%)		Activity ratio of ⁸⁹ Sr / ⁹⁰ Sr on measurement time
		⁸⁹ Sr**	⁹⁰ Sr**	⁸⁹ Sr	⁹⁰ Sr	⁸⁹ Sr	⁹⁰ Sr	
M2-1-1	75.6	3034 ± 20	535 ± 66			-4	-15	18.1
M2-1-2	77.8	2853 ± 25	484 ± 74			-10	-23	17.7
M2-1-3	79.9	2816 ± 24	482 ± 75			-11	-23	16.6
M2-1-4	76.4	2950 ± 26	580 ± 48			-7	-8	14.9
M2-1-5	76.4	2837 ± 20	547 ± 52			-10	-13	16.7
M2-1-6	66.1	3314 ± 33	525 ± 57	3160 ± 10***	628 ± 5***	5	-16	11.6
M2-1-7	65.5	3386 ± 17	580 ± 29			7	-8	11.8
M2-1-8	80.8	2915 ± 34	485 ± 61			-8	-23	11.3
M2-1-9	82.0	2593 ± 18	442 ± 30			-18	-30	11.5
M2-1-10	71.4	3098 ± 14	541 ± 30			-2	-14	11.2
Average	75.2	2980	520			-6	-17	
SD	5.8	238	46					
RSD (%)	7.7	8.0	8.8					
M2-2-1	75.5	5812 ± 34	816 ± 72			-9	30	18.5
M2-2-2	80.0	5089 ± 24	643 ± 61			-20	3	20.6
M2-2-3	81.9	5375 ± 45	700 ± 81			-16	12	20.0
M2-2-4	66.7	6369 ± 42	815 ± 62			-0.2	30	20.2
M2-2-5	57.7	6392 ± 23	437 ± 30			0.2	-30	29.3
M2-2-6	68.9	6396 ± 34	506 ± 69	6380 ± 20***	626 ± 5***	0.3	-19	25.5
M2-2-7	73.7	6477 ± 28	370 ± 35			2	-41	35.1
M2-2-8	67.6	6134 ± 21	516 ± 25			-4	-18	23.7
M2-2-9	67.3	5892 ± 18	371 ± 28			-8	-41	31.7
M2-2-10	69.9	5942 ± 34	382 ± 39			-7	-39	31.3
Average	70.9	5988	556			-6	-11	
SD	7.1	467	176					
RSD (%)	10.0	7.8	31.8					

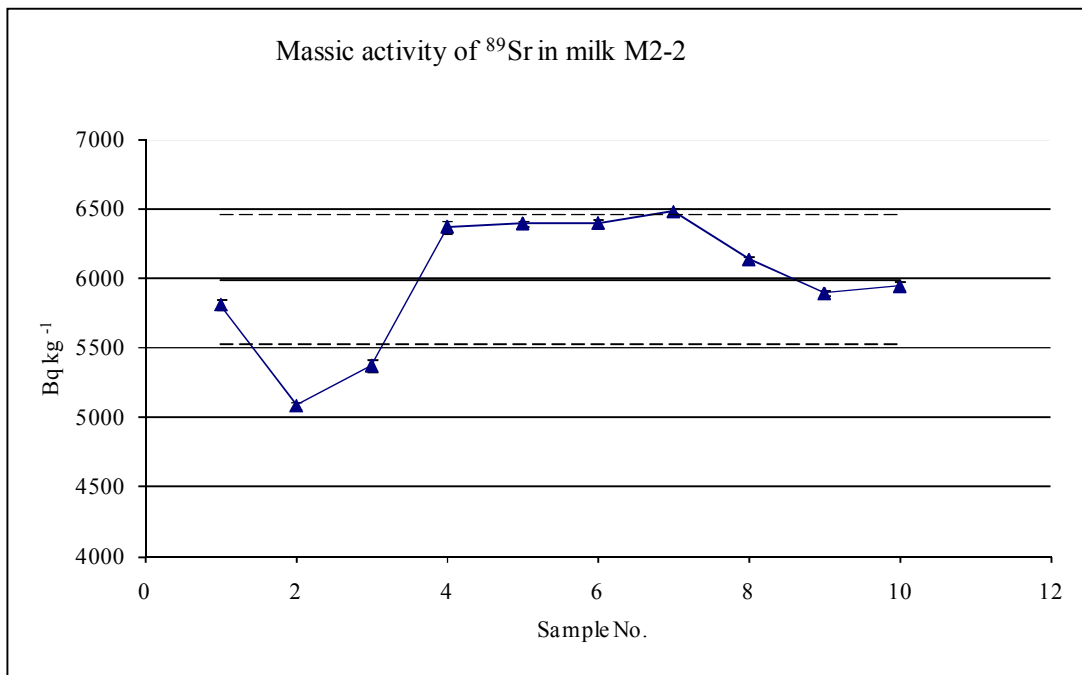
*: Reference date of assigned value: 01.07.2010

**: Average and standard deviation of 5 replicate measurements

***: Combined standard uncertainty (k=1)

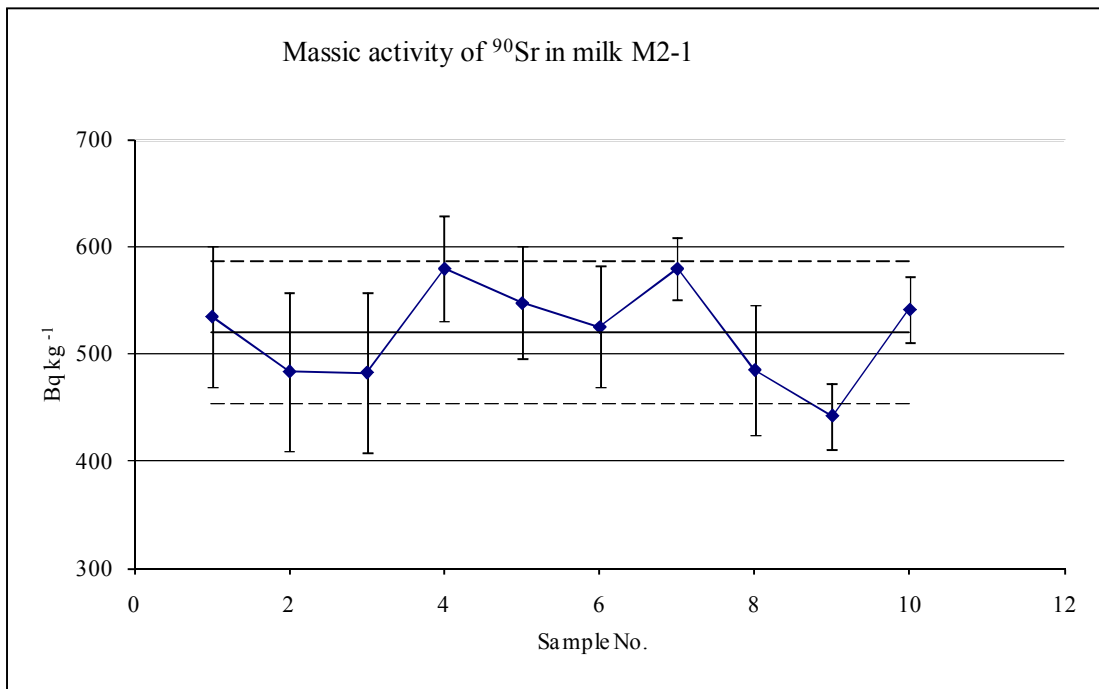


a.)

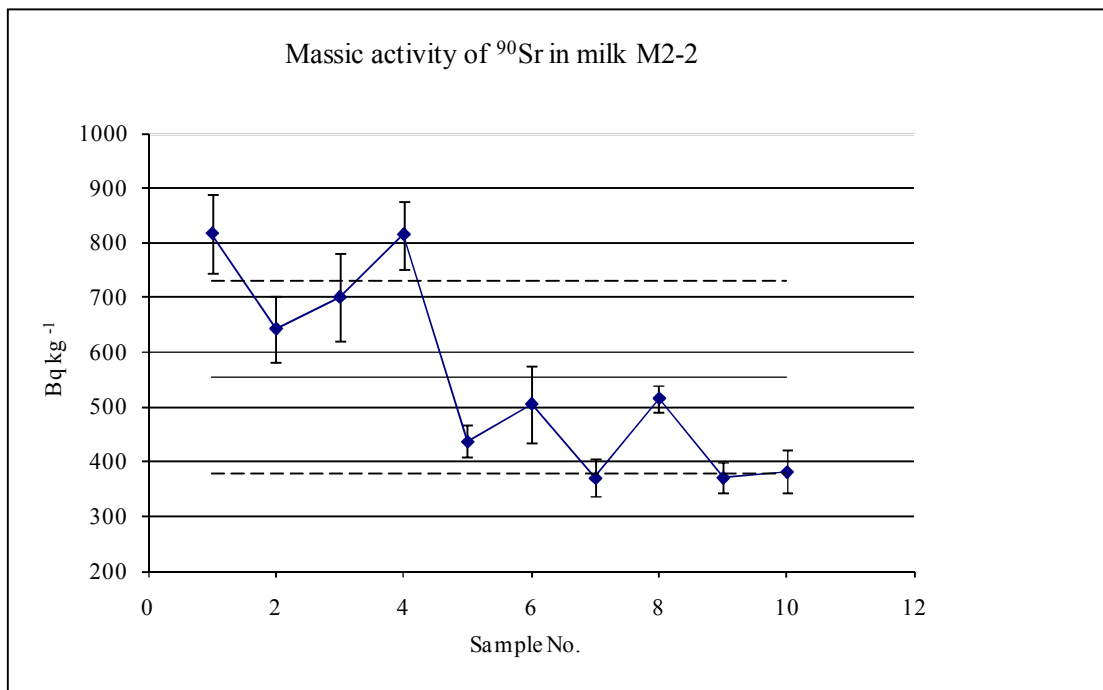


b.)

FIG. 4. Control chart for repeatability of ^{89}Sr determination in double spiked milk samples. a.) M2-1 and b.) M2-2: Measured massic activity of ^{89}Sr (Bq kg^{-1}) is plotted against the number of replicates. The straight line shows the average value, the dotted lines represent the standard deviation from the average ($k=1$), error bars belong to standard deviation of the repeated measurements of individual samples.



a.)



b.)

FIG. 5. Control chart for repeatability of ^{90}Sr determination in double spiked milk sample. a.) M2-1 and b.) M2-2: Measured massic activity of ^{90}Sr (Bq kg^{-1}) is plotted against the number of replicates. The straight line shows the average value, the dotted lines represent the standard deviation from the average ($k=1$), error bars belong to standard deviations of the repeated measurements of the individual samples.

TABLE 10. THE RESULTS OF REPEATABILITY TESTS FOR THE DETERMINATION OF STRONTIUM-89 AND STRONTIUM-90 IN SINGLE SPIKED MILK SAMPLES M1-3 AND M1-4

Repeatability of ^{89}Sr measurement in M1-3 milk sample	
Mean (\bar{X}) in repeatability test, Bq kg^{-1}	440
Repeatability standard deviation (S_r), Bq kg^{-1}	4.7
Repeatability limit (r_L), Bq kg^{-1}	13.2
Relative mean bias (%)	-3.3
Mean bias Bq kg^{-1}	-15
Repeatability of ^{89}Sr measurement in M1-4 milk sample	
Mean (\bar{X}) in repeatability test, Bq kg^{-1}	819
Repeatability standard deviation (S_r), Bq kg^{-1}	9.2
Repeatability limit (r_L), Bq kg^{-1}	25.8
Relative mean bias (%)	-0.8
Mean bias Bq kg^{-1}	-6.6
Repeatability of ^{90}Sr measurement in M1-3 milk sample	
Mean (\bar{X}) in repeatability test, Bq kg^{-1}	82.7
Repeatability standard deviation (S_r), Bq kg^{-1}	10.9
Repeatability limit (r_L), Bq kg^{-1}	30.5
Relative mean bias (%)	6.2
Mean bias Bq kg^{-1}	4.8
Repeatability of ^{90}Sr measurement in M1-4 milk sample	
Mean (\bar{X}) in repeatability test, Bq kg^{-1}	84.7
Repeatability standard deviation (S_r), Bq kg^{-1}	9.6
Repeatability limit (r_L), Bq kg^{-1}	27
Relative mean bias (%)	9.0
Mean bias Bq kg^{-1}	7.0

TABLE 11. THE RESULTS OF REPEATABILITY TESTS FOR THE DETERMINATION OF STRONTIUM-89 AND STRONTIUM-90 IN DOUBLE SPIKED MILK SAMPLES M2-1 AND M2-2

Repeatability of ⁸⁹ Sr measurement in M2-1 milk sample	
Mean (\bar{X}) in repeatability test, Bq kg ⁻¹	2980
Repeatability standard deviation (S _r), Bq kg ⁻¹	238
Repeatability limit (r _L), Bq kg ⁻¹	668
Relative mean bias (%)	-5.7
Mean bias Bq kg ⁻¹	-181
Repeatability of ⁸⁹ Sr measurement in M2-2 milk sample	
Mean (\bar{X}) in repeatability test, Bq kg ⁻¹	5988
Repeatability standard deviation (S _r), Bq kg ⁻¹	467
Repeatability limit (r _L), Bq kg ⁻¹	1308
Relative mean bias (%)	-6.3
Mean bias Bq kg ⁻¹	-400
Repeatability of ⁹⁰ Sr measurement in M2-1 milk sample	
Mean (\bar{X}) in repeatability test, Bq kg ⁻¹	520
Repeatability standard deviation (S _r), Bq kg ⁻¹	66
Repeatability limit (r _L), Bq kg ⁻¹	185
Relative mean bias (%)	-17.2
Mean bias Bq kg ⁻¹	-108
Repeatability of ⁹⁰ Sr measurement in M2-2 milk sample	
Mean (\bar{X}) in repeatability test, Bq kg ⁻¹	556
Repeatability standard deviation (S _r), Bq kg ⁻¹	176
Repeatability limit (r _L), Bq kg ⁻¹	494
Relative mean bias (%)	-11.3
Mean bias Bq kg ⁻¹	-70

APPENDIX IX

RESULTS OF REPRODUCIBILITY MEASUREMENTS

TABLE 12. VALIDATION RESULTS OF RAPID METHOD FOR DETERMINATION OF STRONTIUM-89 AND STRONTIUM-90 IN DOUBLE SPIKED M2-1 BY INTER-LABORATORY TESTING

Sample code	⁸⁹ Sr			⁹⁰ Sr			⁸⁹ Sr/ ⁹⁰ Sr ratio in source at measurement time
	Mean massic activity (Bq kg ⁻¹)	Relative standard deviation (%)	Relative mean bias (%)	Mean massic activity (Bq kg ⁻¹)	Relative standard deviation (%)	Relative mean bias (%)	
Lab-1	3140	4.5	-0.6	528	9.7	-17	2.1
Lab-2	3253	2.8	2.9	580	3.4	-7.6	3.1
Lab-3	3153	2.7	-0.2	623	2.9	-0.8	2.2
Lab-4	2772	6.4	-12.3	543	6.8	-13.6	2.8
Lab-5	3175	2.3	-0.5	613	3.3	-2.4	3.3
Lab-6	2996	3.4	-5.2	554	4.2	-11.8	3.0
Lab-7	2980	8.0	-5.7	520	8.8	-17	14
Assigned value*	3160 ± 10			628 ± 10			

*: Reference date of assigned value: 01. 07. 2010

TABLE 13. VALIDATION RESULTS OF RAPID METHOD FOR DETERMINATION OF STRONTIUM-89 AND STRONTIUM-90 IN DOUBLE SPIKED M2-2 MILK SAMPLES BY INTER-LABORATORY TESTING

Sample code	⁸⁹ Sr			⁹⁰ Sr			⁸⁹ Sr/ ⁹⁰ Sr ratio in source at measurement time
	Mean massic activity (Bq kg ⁻¹)	Relative standard deviation (%)	Relative mean bias (%)	Mean massic activity (Bq kg ⁻¹)	Relative standard deviation (%)	Relative mean bias (%)	
Lab-1	6065	4.0	-4.9	521	8.2	-17	4.3
Lab-2	6463	2.6	0.8	566	2.4	-9.7	5.6
Lab-3	6417	2.3	0.6	599	3.3	-4.3	4.3
Lab-4	5390	13.5	-15.5	478	19.1	-23.6	5.4
Lab-5	6401	3.3	0.3	640	4.4	2.2	6.6
Lab-6	5923	2.2	-7.2	569	7.8	-9.2	4.6
Lab-7	5988	7.8	-6.2	556	31.8	-11.2	25.6
Assigned value*	6380 ± 20			626 ± 5			

*: Reference date of assigned value: 01. 07. 2010

TABLE 14. RESULTS OF REPRODUCIBILITY TESTS OF RAPID METHOD FOR DETERMINATION OF STRONTIUM-89 AND STRONTIUM-90 IN DOUBLE SPIKED MILK SAMPLES M2-1 AND M2-2 BY INTER-LABORATORY TESTING

Reproducibility of ⁸⁹ Sr measurement in M2-1 milk sample	
Mean (\bar{X}) in reproducibility test, Bq kg ⁻¹	3067
Reproducibility standard deviation (S _R), Bq kg ⁻¹	163
Reproducibility limit (R _L), Bq kg ⁻¹	455
Relative mean bias, δ(%)	-2.9
Mean bias (δ), Bq kg ⁻¹	-93
Reproducibility of ⁸⁹ Sr measurement in M2-2 milk sample	
Mean (\bar{X}) in reproducibility test, Bq kg ⁻¹	6092
Reproducibility standard deviation (S _R), Bq kg ⁻¹	381
Reproducibility limit (R _L), Bq kg ⁻¹	1067
Relative mean bias, δ (%)	-4.5
Mean bias (δ), Bq kg ⁻¹	-288
Reproducibility of ⁹⁰ Sr measurement in M2-1 milk sample	
Mean (\bar{X}) in reproducibility test, Bq kg ⁻¹	566
Reproducibility standard deviation (S _R), Bq kg ⁻¹	40.6
Reproducibility limit (R _L), Bq kg ⁻¹	114
Relative mean bias, δ (%)	-9.9
Mean bias (δ), Bq kg ⁻¹	-62
Reproducibility of ⁹⁰ Sr measurement in M2-2 milk sample	
Mean (\bar{X}) in reproducibility test, Bq kg ⁻¹	561
Reproducibility standard deviation (S _R), Bq kg ⁻¹	52.1
Reproducibility limit (R _L), Bq kg ⁻¹	146
Relative mean bias, δ (%)	-10.3
Mean bias (δ), Bq kg ⁻¹	-64.7

APPENDIX X

RESULTS OF DETECTION LIMIT CALCULATIONS

TABLE 15. DETECTION LIMITS EXPRESSED AS MASSIC OR VOLUMIC ACTIVITIES IN DOUBLE SPIKED MILK SAMPLES WITH DIFFERENT STRONTIUM-89/STRONTIUM-90 ACTIVITY RATIO

Sample code	Nuclides	$^{89}\text{Sr}/^{90}\text{Sr}$ activity ratio in source on measurement date	Massic activity on target date (Bq kg^{-1} , dry spiked milk powder)	Volumic activity on target date (Bq L^{-1} , milk)	Detection limit (Bq kg^{-1} , dry spiked milk powder)	Detection limit (Bq L^{-1} , milk)
M1-3	^{89}Sr	3	511 ± 16.4	68.9 ± 2.2	26.4	3.6
	^{90}Sr		71.6 ± 5.4	9.7 ± 0.7	16.4	2.2
M2-1	^{89}Sr	12	2914 ± 32	393 ± 4	11.5	1.6
	^{90}Sr		711 ± 96	96 ± 13	298	40
M2-2	^{89}Sr	20	5826 ± 92	787 ± 12	38	5
	^{90}Sr		749 ± 156	101 ± 21	487	66

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