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Evaluation of different methods for measuring ^{89}Sr and ^{90}Sr: Measurement uncertainty for the different methods as a function of the activity ratio

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HIGHLIGHTS

- Compares three measurement methods with regards to combined measurement uncertainty.
- Methods were tested on spiked water samples with ^{89}Sr/^{90}Sr ratios between 0.3-170.
- Suggests independent measurement of ^{89}Sr and ^{90}Sr to get reliable results for ratios > 3.
- Suggests that a few representative samples should be measured with regards to ^{90}Sr.

ARTICLE INFO

ABSTRACT

In case of a radiological emergency situation involving *e.g.* fission of uranium or plutonium, analysis of radioactive strontium will be of importance. The primary radionuclides of interest are ^{90}Sr, its progeny ^{90}Y and ^{89}Sr. A few days following an event, ^{89}Sr will be the predominant radioisotope of strontium. Most methods found in the literature are valid and applicable when measuring ^{90}Sr, but when samples contain both ^{89}Sr/^{90}Sr interference problematics arise. How these interferences are dealt with will have an effect on the uncertainty of the ^{90}Sr determination.

This work aims at evaluating three measurement approaches, all mentioned in the literature, with respect to the measurement uncertainty when determining ^{90}Sr in an emergency preparedness situation and to propose a suitable measurement strategy.

1. Introduction

Decision makers will, in case of a radiological emergency (*e.g.* a nuclear reactor accident or a nuclear weapons detonation) need reliable measurement results as a base for their choice of action in order to mitigate short- and long term consequences. One of the most important radionuclides to consider in a radiological emergency is the long-lived ^{90}Sr. Its high priority is mainly due to three specific features; its chemical similarity to calcium, its relatively long half-life ($t_{1/2} = 28.8$ y) and the radiotoxicity through its daughter nuclide, ^{90}Y. Works presenting rapid and robust determination methods have been thoroughly covered in the literature (Rondahl et al., 2017; Herranz et al., 2017; Maxwell et al., 2017; Jiang et al., 2017; Sáez-Muños et al., 2018; Habibi et al., 2016; Holmgren et al., 2016, 2014; Maxwell et al., 2013; Grahek et al., 2012, Groska et al., 2012; Herranz et al., 2011; Vajda and Kim, 2010; Maxwell et al., 2010; Maxwell and Culligan, 2009; Tovedal et al., 2009a, 2009b; Tovedal et al., 2008, Chobola et al., 2006; Ramebäck

et al., 1994). However, methods using inductively coupled plasma mass spectrometry (ICP-MS) has not been considered in this work. The literature presents a possible approach to determine ^{90}Sr (Takagai et al., 2014; Feuerstein et al., 2008; Taylor et al., 2007) but due to its short half-life ICP-MS is not a suitable alternative for determining ^{89}Sr.

The most common approach, following the close to mandatory chemical separation, is to perform measurements using a liquid scintillation counter (LSC). An alternative to LSC is proportional counting (Habibi et al., 2016).

There are a number of ways to determine ^{90}Sr and ^{89}Sr using liquid scintillation counting. One method is spectrum deconvolution, *i.e.* to mathematically fit two or more component spectra to an experimentally obtained composite spectrum. This approach has been investigated in the literature as a feasible determination method for emergency response. By using deconvolution it has been shown that it is possible to determine ^{89}Sr and ^{90}Sr from a single activity measurement by LSC (Kabai et al., 2017; Kanisch, 2016; Kabai et al., 2011; Remetti and

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Sessa, 2011; Heckel and Vogl, 2009, Altzitzoglou, 2008).

A similar method is spectrum subtraction. Where one can use set measurement windows to discriminate the ^{90}Sr signal from the ^{89}Sr signal in a LSC spectrum. Or, makes use of the fact that ^{89}Sr has a high $E_{max}(\beta^-)$ and thus determining the ^{89}Sr activity *via* an initial Cherenkov measurement. The latter has been used in this work. Spectrum subtraction is a well-established method for determining ^{89}Sr and ^{90}Sr (Tayeb et al., 2016; IAEA/AQ/27, 2013; Salonen, 1978,; Kim et al., 2009; Heilgeist, 2000).

The third method relies on independent measurements of ^{89}Sr and ^{90}Sr, using Cherenkov counting, which is achieved by a second chemical separation in order to isolate the in-grown ^{90}Y. This can be done by strontium or yttrium chemistry (Horwitz et al., 1992; Tovedal et al., 2009a; Holmgren et al., 2014).

The literature published within this field has one primary objective: to ensure that decision making processes are not limited by sample throughput, regardless of the sample matrix. The range of analytical methods available for strontium analysis is vast. The method of choice is dependent on action limits, sample matrices, and how the method deal with both chemical as well as radioactive interferences, and of course on the experience of the laboratory. However, the aspect of combined measurement uncertainty and its impact on the reliability of the analysis has taken a back seat, not all authors take into consideration how the method of choice effects the combined measurement uncertainty of, in particular, ^{90}Sr.

By theoretical calculations and empirical tests, we aim to show how the combined measurement uncertainty is affected by evaluating the most commonly published methods for determining ^{90}Sr in the presence of ^{89}Sr. The evaluations were done with respect to the combined measurement uncertainty on water samples, spiked with ^{89}Sr and ^{90}Sr at different ratios, thereby bypassing any complications stemming from sample preparation (*e.g.* quench) as well as any other chemical or radiological interferences (Tovedal et al., 2009a; Holmgren et al., 2014). However, this does not mean that these factors can be disregarded when working on real samples. In order to perform correct ^{89}Sr and ^{90}Sr measurements it is paramount that the method of choice can handle interferences as well as complications stemming from sample preparation.

This work aims to contribute to an increased awareness regarding the combined measurement uncertainty, associated with the methods mentioned, in order to assure that an appropriate measurement strategy is used in an emergency situation.

2. Theory

2.1. Initial chemical separation

All methods in this work require an initial chemical separation of strontium from possible interferences as well as ^{90}Y (time of separation equals t_0 for ^{90}Y). How the different methods differ from each other can be seen in Fig. 1.

2.2. Method A: Second chemical separation

This method represents independent measurement of both ^{89}Sr and ^{90}Sr. After the first separation ^{89}Sr can be quantified according to:

$$A_{Sr89} = \frac{R_{Sr89,n} - R_{Sr89,bg}}{Y_{Sr} \cdot \Psi_{Cher,Sr89}} \tag{1}$$

Where $R_{Sr89,n}$ is the net count rate (cps) of the ^{89}Sr measurement, $R_{Sr89,bg}$ is the net background count rate (cps) for the ^{89}Sr measurement, Y_{Sr} is the chemical yield of the strontium separation, $\Psi_{Cher,Sr89}$ is the measurement efficiency for ^{89}Sr using Cherenkov counting (cps/Bq). If there is no ^{89}Sr present in the sample the determination of ^{90}Sr can be performed, either during ingrowth of ^{90}Y using Cherenkov counting,

directly using LSC counting or by other means, without need for further precautions.

However, if ^{89}Sr is present further steps are needed in order to determine ^{90}Sr. Apart from spectrum deconvolution or spectrum subtraction (described in the sections below) the most common approach in the literature to date is another chemical separation (SRW01, 2003; Horwitz et al., 1992; Holmgren et al., 2014). The chemical separation can be performed by isolation of ^{90}Y by yttrium separation chemistry, *e.g.* DGA or Ln-resin (Maxwell et al., 2017; Tovedal et al., 2009a; McAlister and Horwitz, 2007), or by rinsing out the ^{90}Y that has grown into the ^{90}Sr solution by strontium separation chemistry, and collecting the rinse fraction. The latter method has been shown to give a 100% yield of yttrium, so there is no need for a second yield determination (EiChrom, 2003; Holmgren et al., 2014). In order to calculate the activity concentration of ^{90}Sr the following equations were used:

$$A_{Y90} = \frac{R_{Y90,n} - R_{Y90,BG}}{\Psi_{Y90}} \tag{2}$$

Where the parameters are the same as in Eq. (1) but for ^{90}Y instead of ^{89}Sr. By calculating the activity of ^{90}Y at time t_i, *i.e.* the time allowed for ingrowth of ^{90}Y from t_0 (time of separation), one can determine the activity of ^{90}Sr according to:

$$A_{Sr90} = \frac{A_{Y90}}{(1-e^{-\lambda_{Y90}t_i}) \cdot Y_{Sr}} \tag{3}$$

Where λ_{90Y} is the decay constant of ^{90}Y.

2.3. Method B: Spectrum subtraction

The calculation approach for spectrum subtraction is, in accordance with IAEA/AQ/27 (2013), expressed for the ^{90}Sr and ^{89}Sr activities at the reference time (t) using the following equations:

$$A_t^{Sr90} = \frac{R_{LSC,t2} \cdot \Psi_{Cher,Sr89} - R_{Cher,\ t1} \cdot \Psi_{LSC,Sr89} \cdot f_3}{y_{subtr} \cdot Y_{Sr}} \cdot e^{-\lambda_{Sr90}(t_1-t)} \tag{4}$$

$$A_t^{Sr89} = \frac{\begin{array}{l} R_{Cher,t1}(y + \Psi_{Cher,Yr90}\Psi_{LSC,Sr89} \cdot f_1 \cdot f_3 + \Psi_{Cher,Sr90} \cdot \Psi_{LSC,Sr89} \cdot f_3) \\ - R_{LSC,t2}(\Psi_{Cher,Sr89} + \Psi_{Cher,Y90} \cdot \Psi_{Cher,Sr89} \cdot f_1) \end{array}}{y_{subtr} \cdot \Psi_{Cher,Sr89} \cdot Y_{Sr}}$$
$$\cdot e^{-\lambda_{Sr90}(t_1-t)} \tag{5}$$

where $R_{Cher,t1}$ is the count rate at t_1 for the Cherenkov measurement, and $R_{LSC,t2}$ the count rate at t_2 for the liquid scintillation counting measurement. As for the other two methods Ψ represents measurement efficiencies. In this case the efficiencies are for ^{89}Sr, ^{90}Sr and ^{90}Y when performing Cherenkov- or liquid scintillation counting, respectively (in cps/Bq). Concerning f_1, f_2, f_3 and y_{subtr} they are defined as:

$$y_{subtr} = \begin{array}{l} \Psi_{Cher,Sr89} \cdot \Psi_{LSC,Sr90} + \Psi_{Cher,Sr89} \cdot \Psi_{LSC,Y90} \cdot f_2 \\ - \Psi_{LSC,Sr89} \cdot \Psi_{Cher,Sr90} \cdot f_3 \cdot \Psi_{LSC,Sr89} \cdot \Psi_{Cher,Y90} \cdot f_1 \cdot f_3 \end{array} \tag{6}$$

$$f_1 = 1-e^{-\lambda_{Y90}(t_1-t_0)} \tag{7}$$

$$f_2 = 1-e^{-\lambda_{Y90}(t_2-t_0)} \tag{8}$$

$$f_3 = e^{-\lambda_{Sr89}(t_2-t_1)} \tag{9}$$

Where y_{subtr} is an auxiliary parameter used to simplify the presentation and calculation of A^{Sr89} and A^{Sr90}.

f_1 and f_2 are correction factors for ingrowth of ^{90}Y between the time of separation (t_0) and the Cherenkov measurement (t_1) or the LSC measurement (t_2), and f_3 is the correction factor for ^{89}Sr decay between the Cherenkov measurement and the LSC measurement. The decay constants (s^{-1}) for ^{90}Y and ^{89}Sr are represented by λ_{Y90} and λ_{Sr89}.
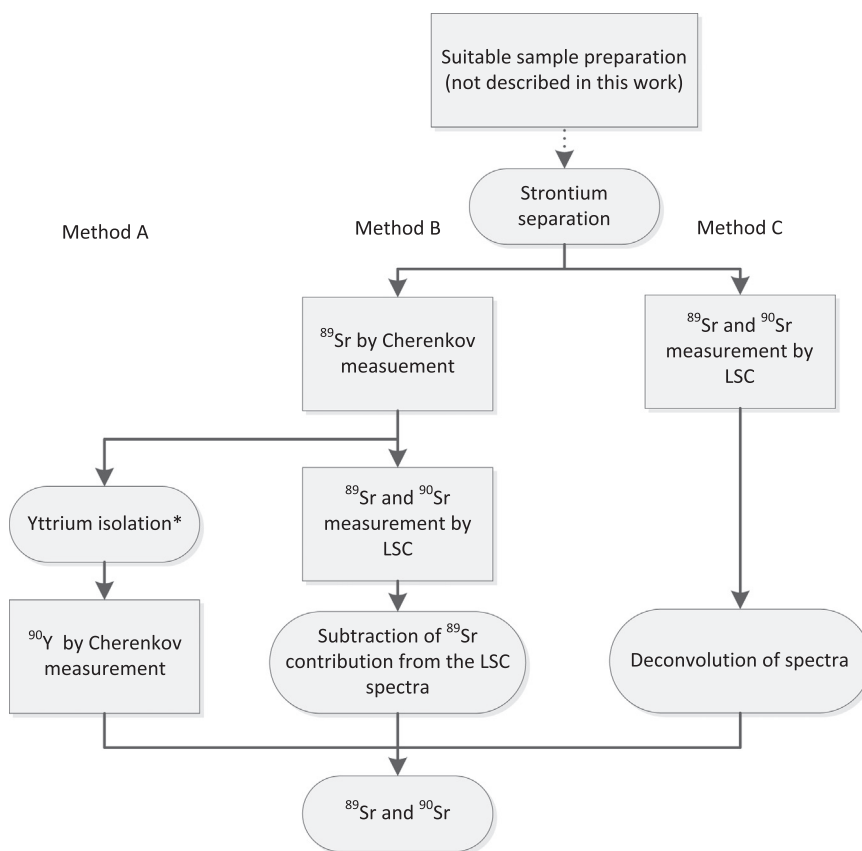
Fig. 1. A schematic representation of the methods used in this work, a second chemical separation (Method A, left), spectrum subtraction (Method B, middle) and spectrum deconvolution (Method C, right).

2.4. Method C: Spectrum deconvolution

In this work deconvolution was done by least square fitting of pure background subtracted component LSC spectra of ^{89}Sr and ^{90}Sr to the sample composite spectra, *i.e.*:

$$ESS = \sum A_i^2 = \sum (M_{m,i} - M_i)^2 \qquad (10)$$

Where A_i is the residual, M_i is the count rate (cps) in channel i for the sample spectra, and $M_{m,i}$ (the model) is described as:

$$M_{m,i} = \Psi_{LSC,Sr90,i} \cdot A_{Sr90} + \Psi_{LSC,\ Sr89,i} \cdot A_{Sr89} \qquad (11)$$

Where $\Psi_{LSC,Sr90,i}$ and $\Psi_{LSC,Sr89,i}$ are the counting efficiencies (cps/Bq) of ^{90}Sr and ^{89}Sr, respectively, in channel i (for LSC measurements the efficiencies over the entire energy window are equal to 1 in this case), A_{Sr90} and A_{Sr89} are the fitted activities (Bq) for ^{90}Sr and ^{89}Sr. The resulting activities for ^{89}Sr and ^{90}Sr were corrected with regards to chemical yield and decay.

The uncertainty of the fit was evaluated by performing a χ^2-test according to Press et al. (2007), *i.e.* individually adjusting the component variables until the ESS had doubled its value compared to the minimum. For each component variable the value when the ESS is twice its minimum represents the variance, and the standard uncertainty is then simply the square root of the variance.

3. Methods and materials

3.1. Reagents and standards

The reagents used for the chemical separations were all of p.a. grade (Merck, Darmstadt, Germany).

Radioactive standards used for spiking and reference spectra were ^{90}Sr, with ^{90}Y in equilibrium, (SIZ44, Amersham International, UK) and ^{89}Sr (SMYA72, Eckert & Ziegler Analytics, Atlanta, GA, USA). The radiochemical separation was performed using Sr and Ln-resin in 2 ml pre-packed columns, with a particle size of 50–100 µm (TrisKem International, Bruz, France). For LSC measurements Optiphase HiSafe 3 (PerkinElmer, Boston, MA, USA) was used. Stable Sr (1 000 µg/ml), for yield determination using ICP-OES, and Ho (10 000 µg/ml), as internal standard, were supplied by Teknolab Sorbent (Kungsbacka, Sweden).

3.2. Instrumentation

LSC and Cherenkov measurements were performed on a low background system, Wallac Quantalus 1220 (Perkin Elmer, Boston, MA, USA). The yield determination for the separations were performed using ICP-OES (iCap 7400, Thermo Scientific, Bremen, Germany).

3.3. Calibrations

Six (6) separate calibrations were made in order to determine the different measurement efficiencies of ^{90}Sr, ^{90}Y and ^{89}Sr for Cherenkov counting and LSC, see Table 1.

The efficiencies for ^{90}Sr and ^{90}Y were determined after chemical separation.

3.4. Experimental

One round of experiments, for one method, consisted of seven spiked water samples (^{89}Sr/^{90}Sr activity ratio of 170, 100, 30, 10, 3, 1 and 0.3) and a blank sample. Each method was run three times with

Table 1
Counting efficiencies for ^{90}Sr, ^{89}Sr and ^{90}Y of the Cherenkov and LSC measurements.

Radionuclide	Counting efficiency (Ψ)	
	Cherenkov counting	**Liquid Scintillation counting**
^{90}Sr	$(4.34 \pm 0.25) \cdot 10^{-4}$	1.005 ± 0.030
^{89}Sr	0.3890 ± 0.0062	1.005 ± 0.010
^{90}Y	0.586 ± 0.015	1.000 ± 0.028

each round having different added activities, but still in the ratios mentioned above. Experimental round 1 had variable ^{89}Sr activities, and a set ^{90}Sr activity (approximately 9 Bq). Round 2 and 3 had set ^{89}Sr activities (approx. 17 and 60 Bq respectively) and variable ^{90}Sr activities. The added activities are listed with the measured ditto in the *Result*-section. In total 72 samples were prepared and measured. The total volume of the samples were 10 ml in an 8 M HNO$_3$ matrix, stable strontium (100 μg) was added as stable yield tracer. An aliquot of 0.1 ml was taken out of the sample before chemical separation for yield determination.

All samples were chemically separated, initially, in order to remove ^{90}Y. To facilitate the chemical separations vacuum boxes (VacMaster system, Biotage, Sweden) were used. The separation procedure consisted of loading the sample onto a pre-conditioned (5 ml 8 M HNO$_3$) Sr-resin cartridge using a plastic column (25 ml) and then rinsing the sample beaker (2 × 2.5 ml 8 M HNO$_3$). Following the sample loading the resin was rinsed using two 5 ml 8 M HNO$_3$ aliquots. The strontium fraction was then eluted into plastic scintillation vials (18 ml) using 4 ml 0.05 M HNO$_3$. This was done to assure equal treatment of all samples and still allowing an addition of 12 ml scintillation cocktail for LSC measurement. An aliquot of 0.04 ml was taken out of the sample after the chemical separation for yield determination.

A second chemical separation was performed in Method A in order to separate ^{90}Y for the measurement of ^{90}Sr. 4 ml of concentrated HNO$_3$ was added to the eluate after the Cherenkov measurement of ^{89}Sr in order to adjust the molarity of the sample prior the chemical separation using Sr-resin. After allowing for 16 h of ingrowth of ^{90}Y the samples were taken to a second round of chemical separation. The samples were loaded onto a pre-conditioned (5 ml 8 M HNO$_3$) Sr-resin cartridge using a plastic column. The sample container was rinsed with 2 × 2.5 ml 8 M HNO$_3$. The rinse from the loading (total of 13 ml) was collected in a scintillation vial and subsequently measured for ^{90}Y.

Unless stated otherwise the Cherenkov measurement times were 2 × 10 min for ^{89}Sr, 4 h for ^{90}Y, and 6 × 10 min for the LSC measurements.

Yield determination was performed by diluting the sample aliquots to 10 ml with added internal standard (Ho 1000 μg) and 2% HNO$_3$.

3.5. Combined measurement uncertainty

The combined measurement uncertainties for all methods were calculated in accordance with the Guide to the Expression of Uncertainty in Measurement (GUM), ISO, 1995, using the software GUM Workbench (GUM Workbench, 2010). The combined measurement uncertainties presented in this work represent an estimation of all uncertainty components associated with the analysis method. The uncertainties throughout this paper are presented with a coverage factor k = 2, representing an approximate confidence level of about 95%, unless otherwise stated.

The combined measurement uncertainty for Method A was calculated using GUM Workbench and the equations associated with calculating the activity from the Cherenkov measurements. The same approach was applied for Method B, but using Eqs. (4)–(9) as the model equations. For Method C the activity and the associated uncertainty for

each measurand was obtained by the χ^2-method, which was subsequently corrected using the chemical yield. All results were corrected for decay to a set reference date.

In order to determine whether or not a method gives reliable results, with uncertainties that are fit-for purpose, it will need to limit the combined uncertainty to 25% (k = 2). A minimum acceptable relative bias (MARB) of approximately 25% is what the IAEA usually allow for ^{90}Sr determination in their proficiency tests (IAEA/AQ/3, 2009).

Based on the works by Currie (1968) and Lochamy (1976), Tovedal et al. (2008) show that samples at detection limit will have a relative combined uncertainty of about 30% (k = 1). Therefore, all results presented with a relative combined uncertainty of 60% (k = 2) or more will be judged as below detection limit.

Results with uncertainties between 25% and 60% may under certain circumstances be considered to be fit-for-purpose, and that its method can be applicable to some extent. However, the methods that result in relatively high uncertainties in this work will almost certainly give higher uncertainties for real samples, as they present other sources of uncertainty, such as measurements with high statistical uncertainty, matrix effects and interferences.

4. Results and discussion

As expected Method B and C both delivered results more rapidly than Method A, seeing as the latter requires ^{90}Y ingrowth. However, contrary to what has been stated by many authors, one does not need to await radioactive equilibrium, which would take about two weeks, the second separation can be performed much earlier. Of course, the detection limit of ^{90}Sr will be lower for longer ingrowth times for ^{90}Y as it scales with the inverse of the fractional ingrowth of ^{90}Y (the higher fractional ingrowth the lower the detection limit). After a radiological incident higher detection limits compared to environmental monitoring programs might still be fit-for-purpose, and therefore one can significantly reduce the ingrowth time of ^{90}Y resulting in a more rapid method. In this work, the time used for ingrowth of ^{90}Y was limited to 16 h, yielding a total analysis time of 23 h for the analysis of one sample and 44 h for six samples. For Method B and C the total analysis time came in at 5 h and 5.3 h respectively for one sample, 10 h and 12 h respectively for six samples. The increase in total analysis time is due to the time needed for the additional sample measurements, *i.e.* five more ^{90}Y measurements for Method A and five ^{89}Sr/^{90}Sr measurements for Method B and C, respectively. Resulting in a relative increase of the total analysis time, when measuring one versus six samples, which is approximately the same for Method A–C.

These time frames are valid under the assumption that the separation of the six samples occur at the same time, that yield determination is performed during the ingrowth of ^{90}Y and that the Cherenkov/LSC measurements are performed subsequently on one instrument. However, according to the work of Tovedal et al. (2009b) it will not be possible to measure either ^{89}Sr or ^{90}Sr with any certainty within the first few days of an incident involving fission of plutonium or uranium. This is due to the fact that short lived strontium and yttrium isotopes, *e.g.* ^{91}Sr, ^{91}Y and ^{92}Y, will dominate the measurements. The recommended waiting time, for those radionuclides to decay to a manageable level, is 4–5 days in order to measure ^{89}Sr.

The chemical yields (Y_{Sr}) of all three methods were around 70–90%, the differences were between the experimental rounds rather than between methods.

4.1. Determined ^{89}Sr activity

All three methods performed well. The measured ^{89}Sr activities were in agreement with added activities, see Table 2. This was no surprise as two of the methods (A and B) consist of separate ^{89}Sr measurements and for the third one, Method C, ^{89}Sr is not as influenced by the ^{90}Sr content as vice-versa (as ^{89}Sr has a higher β-energy than

Table 2

The added and measured activity for ^{89}Sr, all samples, with their combined measurement uncertainty ($k = 2$). The number following the ^{89}Sr/^{90}Sr ratio in parenthesis gives which of the three experimental rounds the samples are from.

^{89}Sr/^{90}Sr ratio	Method A Chemical separation		Method B Subtraction		Method C Deconvolution	
	Added [Bq]	Measured [Bq]	Added [Bq]	Measured [Bq]	Added [Bq]	Measured [Bq]
170 (1)	1526 ± 28	1478 ± 160	1523 ± 28	1466 ± 160	1524 ± 28	1520 ± 180
100 (1)	903 ± 19	889 ± 91	894 ± 19	848 ± 93	904 ± 20	920 ± 110
30 (1)	282 ± 10	290 ± 33	264.1 ± 9.4	270 ± 30	281 ± 10	284 ± 35
10 (1)	90.6 ± 1.8	85.2 ± 9.6	90.2 ± 1.8	89.4 ± 9.8	89.9 ± 1.8	84 ± 12
3 (1)	27.15 ± 0.58	25.7 ± 3.1	26.92 ± 0.58	29.1 ± 3.2	27.27 ± 0.58	25.1 ± 4.6
1 (1)	9.28 ± 0.30	9.3 ± 1.3	8.81 ± 0.28	9.1 ± 1.2	9.27 ± 0.30	9.5 ± 3.6
0.3 (1)	2.47 ± 0.22	2.32 ± 0.40	2.53 ± 0.22	3.16 ± 0.76	2.63 ± 0.22	2.7 ± 2.9
170 (2)	20.13 ± 0.61	20.8 ± 2.7	20.12 ± 0.61	24.3 ± 2.3	14.14 ± 0.42	14.0 ± 3.5
100 (2)	20.11 ± 0.61	19.6 ± 2.5	20.08 ± 0.61	20.3 ± 2.2	14.38 ± 0.44	14.2 ± 3.7
30 (2)	20.16 ± 0.61	22.5 ± 2.9	20.05 ± 0.61	22.2 ± 2.2	14.94 ± 0.44	15.8 ± 4.2
10 (2)	20.13 ± 0.61	21.3 ± 2.8	20.12 ± 0.61	20.1 ± 2.2	15.20 ± 0.46	15.1 ± 3.7
3 (2)	20.11 ± 0.61	20.4 ± 2.6	20.14 ± 0.61	20.1 ± 2.2	15.20 ± 0.46	15.7 ± 5.1
1 (2)	20.16 ± 0.61	21.7 ± 2.8	20.11 ± 0.61	19.8 ± 2.1	15.29 ± 0.46	18.6 ± 4.9
0.3 (2)	20.14 ± 0.61	23.1 ± 3.5	20.10 ± 0.61	22.3 ± 2.0	15.71 ± 0.48	26.0 ± 7.8
170 (3)	61.5 ± 1.2	55.0 ± 6.9	58.6 ± 1.2	46.8 ± 5.2	60.1 ± 1.2	53.6 ± 11
100 (3)	61.2 ± 1.2	57.2 ± 7.2	58.3 ± 1.2	51.7 ± 5.1	59.6 ± 1.2	59.7 ± 12
30 (3)	61.1 ± 1.2	53.5 ± 6.7	58.6 ± 1.2	49.7 ± 5.4	59.4 ± 1.2	57.2 ± 11
10 (3)	60.9 ± 1.2	57.8 ± 7.3	58.3 ± 1.2	48.4 ± 5.2	59.3 ± 1.2	57.8 ± 12
3 (3)	60.1 ± 1.2	66.5 ± 8.5	58.7 ± 1.2	54.3 ± 7.1	60.2 ± 1.2	55 ± 12
1 (3)	59.6 ± 1.2	58.0 ± 6.7	58.9 ± 1.2	55.9 ± 7.3	58.7 ± 1.2	55 ± 12
0.3 (3)	59.4 ± 1.2	67.0 ± 7.8	58.9 ± 1.2	67.3 ± 9.4	59.3 ± 1.2	59 ± 15

^{90}Sr).

However, the relative uncertainty for the ^{89}Sr activity determined by Method C increases with decreasing activity. This is due to the fact that the ^{89}Sr determination is not done independently from ^{90}Sr, so the uncertainty will increase with higher ^{90}Sr activities *i.e.* lower ^{89}Sr/^{90}Sr ratios.

The results presented in Table 2 and Fig. 2 imply that, given that the ^{89}Sr activity of the sample fraction measured on the instrument is higher than 70 Bq all three methods will perform equally well for ^{89}Sr determinations at all activity ratios.

Methods A and B provide results that are fit-for-purpose over the entire range of ^{89}Sr/^{90}Sr activity ratios, *i.e.* the relative combined uncertainty is < 25%. Method C can be used for ratios 3–170, depending on the counting statistics.

4.2. Determined ^{90}Sr activity using Method A: A second chemical separation

The results for the ^{90}Sr determination using Method A are presented in Table 3. The results are in agreement with the added ^{90}Sr activity for all samples. The performance of this method was proven earlier by Tovedal et al. (2009a) and by Holmgren et al. (2014).

The resulting relative uncertainties for ^{90}Sr determination by the chemical separation method are between 11–20% depending on the activity level of the sample. The main contributor to the combined measurement uncertainty, excluding counting statistics for low activity
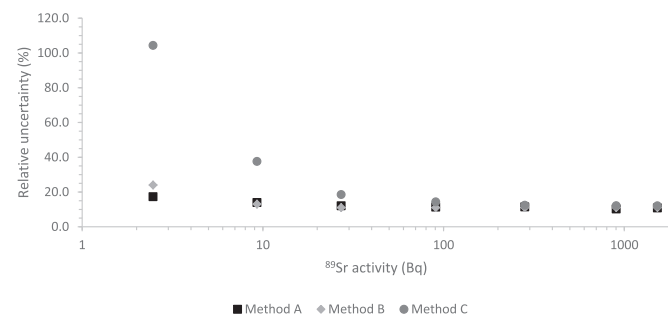
samples, is the yield determination which contributes to 80–90% of the total variance. For measurements in need of lower uncertainties or lower detection limits it is recommended to extend the time for ingrowth of ^{90}Y to approximately 64 h instead of the 16 h used in this work. During the first 24 h ^{90}Y grows into the sample at a rate of approximately 1% per hour. By extending the allowed time for ingrowth to 64 h the detection limit, calculated according to Currie (1967), will be 30% lower than for 16 h.

Method A provides results that are fit-for-purpose and above detection limit, according to the previous definition, for ^{90}Sr determination at the entire range of ^{89}Sr/^{90}Sr-ratios. This is due to the fact that ^{90}Sr is measured independently, *i.e.* without the presence of ^{89}Sr since a second chemical separation is done in order to isolate and measure ^{90}Y.

4.3. Determined ^{90}Sr activity using Method B: Spectrum subtraction

In this work, this method gave generally good results for activity ratios between 0.3–3.

The relative combined uncertainties for the method as a function of the activity ratio can be seen in Fig. 3.

Method B is not fit-fit for purpose when determining ^{90}Sr for ^{89}Sr/^{90}Sr-ratios above about 3, as they present relative combined uncertainties larger than 25% throughout all experimental rounds. Moreover, the method gives results below detection limit (rel. combined uncertainties > 60%, $k = 2$) for ratios 30–170. However, even though the precision in some cases is poor, the accuracy for round 1 is at least fair, see Table 4.

Similar results were presented for the IAEA rapid method, which was validated on spiked water and milk samples (IAEA/AQ/27 2013). For spiked water samples, the relative combined uncertainty was about 25% ($k = 2$) already at an activity ratio of about 2.5 and reached 100% ($k = 2$) for an activity ratio of about 40. Moreover, for spiked milk samples two standard deviations larger than 25% were obtained for replicate samples ($n = 5$) for ^{90}Sr already at an activity ratio of about 2. For higher activity ratios (about 35) that method yielded relative biases of up to about 40%.

As seen in Fig. 3 the relative combined uncertainties are in general higher for round 1 than round 2 and 3. In order to explain it is necessary to look at the uncertainty budgets, see Fig. 4.

At activity ratios higher than about 3 the yield will no longer be the

Fig. 2. The relative uncertainties as a function of activity for experimental round 1.

Table 3
The added and the resulting ^{90}Sr activities, with their associated uncertainties, for all three rounds of experiments using Method A: A second chemical separation.

^{89}Sr/^{90}Sr ratio	Experimental round 1		Experimental round 2		Experimental round 3	
	Added [Bq]	Measured [Bq]	Added [Bq]	Measured [Bq]	Added [Bq]	Measured [Bq]
170	9.08 ± 0.27	7.50 ± 0.83	0.120 ± 0.012	0.154 ± 0.031	0.340 ± 0.034	0.441 ± 0.074
100	9.08 ± 0.27	8.31 ± 0.87	0.210 ± 0.013	0.248 ± 0.041	0.580 ± 0.035	0.66 ± 0.10
30	8.93 ± 0.27	7.85 ± 0.91	0.650 ± 0.020	0.75 ± 0.10	1.960 ± 0.059	2.10 ± 0.28
10	9.12 ± 0.27	7.59 ± 0.87	2.01 ± 0.05	2.20 ± 0.29	5.94 ± 0.15	6.27 ± 0.82
3	9.09 ± 0.27	10.16 ± 1.2	6.68 ± 0.16	6.77 ± 0.89	17.45 ± 0.52	14.9 ± 2.3
1	9.10 ± 0.27	10.36 ± 1.3	19.06 ± 0.19	22.0 ± 2.8	58.5 ± 1.7	68 ± 11
0.3	9.07 ± 0.27	10.48 ± 1.3	62.46 ± 0.26	68.4 ± 9.6	200.8 ± 6.0	192 ± 26
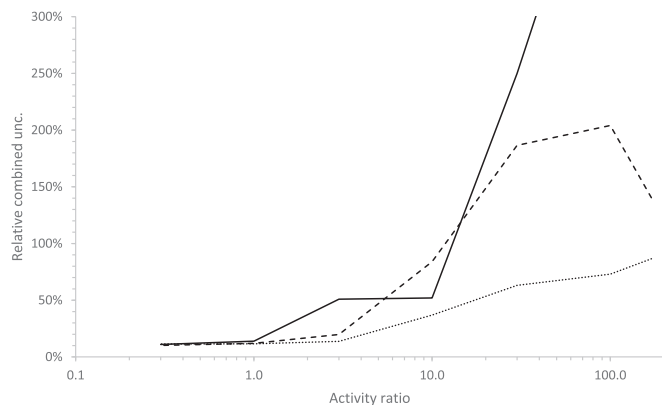
Fig. 3. The relative combined uncertainties in measured ^{90}Sr activities for the subtraction method as a function of activity ratio. The solid line represent samples with a fixed ^{90}Sr activity (round 1), the dashed line is for samples from round 2 and the dotted line represents samples from round 3.

major contributor to the combined uncertainty for ^{90}Sr, regardless of the activity level of the sample. The yield is substituted as the main contributing factor by the measurement efficiencies of the LSC and Cherenkov measurement of ^{89}Sr ($\Psi_{LSC,Sr89}$ and $\Psi_{Cher,Sr89}$) and the counting statistics for the Cherenkov measurement, $R_{Cher,t1}$. The fact that the counting statistics is a more prominent contributor to the uncertainty for round 2 than round 3 is due to the relatively low ^{89}Sr activities, *i.e.* when the ^{89}Sr activity decreases the uncertainty contribution from the Cherenkov counting statistics will increase.

As the activity content of the samples increases the counting statistics becomes insignificant as an uncertainty contributor, as can be seen for round 1, resulting in the ^{89}Sr-efficiencies being the major contributor.

4.4. Determined ^{90}Sr activity using Method C: Spectrum deconvolution

The results from the experiments using deconvolution can be as seen in Table 5. As with the subtraction method the determined ^{90}Sr activities are associated with large measurement uncertainties.

Method C could probably be deemed as fit-for-purpose when determining ^{90}Sr for ^{89}Sr/^{90}Sr-activity ratios 0.3–1. However, for experimental round 1 all ratios show a relative combined uncertainty larger than 25% and for round 2 the only sample which an uncertainty below 25% was for the 0.3 ratio. Furthermore, the method provides results below detection limit for activity ratios 10–170.

The relative combined uncertainties for round 1 are in general lower than for round 2 and 3 for activity ratios > 1. However, at the activity ratio 0.3 the relative combined uncertainty for round 1 is larger than for round 2 and 3, this is due to the relative low ^{90}Sr activity in that sample (Fig. 5). For activity ratios 0.3 to 3 the chemical yield is the main contributor to the combined uncertainty).

The uncertainty in counting statistics is included in the model fit. This is apparent when comparing different counting times, see Fig. 6. The result is an uncertainty of 40–77% ($k = 1$) at an activity ratio of 3. When looking at lower activity ratios, *e.g.* 0.3, the uncertainty (7–9%) is not significantly reduced even for 200 min measurements, *i.e.* the major contributor to the uncertainty in those results is not from counting statistics. For higher activity ratios, *i.e.* ^{89}Sr/^{90}Sr = 10–170, the uncertainty of the model fit is greater than 100%. This is because at those ratios a large value (the combined spectra) is subtracted by a similarly large value (the ^{89}Sr contribution). Kabai et al. (2017), although not presenting any measurement results, stated that deconvolution of LSC spectra would be fit-for-purpose up to an activity ratio of 10, after which the method would become uncertain. This is similar to what was observed in this work.

4.5. Suggested measurement approach for a nuclear emergency situation

The results show that Method B and C are associated with large uncertainties for ^{90}Sr at activity ratios greater than 3, which is hardly fit-for purpose in an emergency situation. In case of a radiological emergency resulting in an activity ratio of 170 (instant fission) it will take approximately 320 days until the ratio has decreased to below 3, the respective time for an initial ratio of 10 (nuclear power plant accident) is 95 days. Given the fact that one cannot in advance know the ^{89}Sr/^{90}Sr-activity ratio it is recommended to use the method which consistently delivers fit-for purpose results.

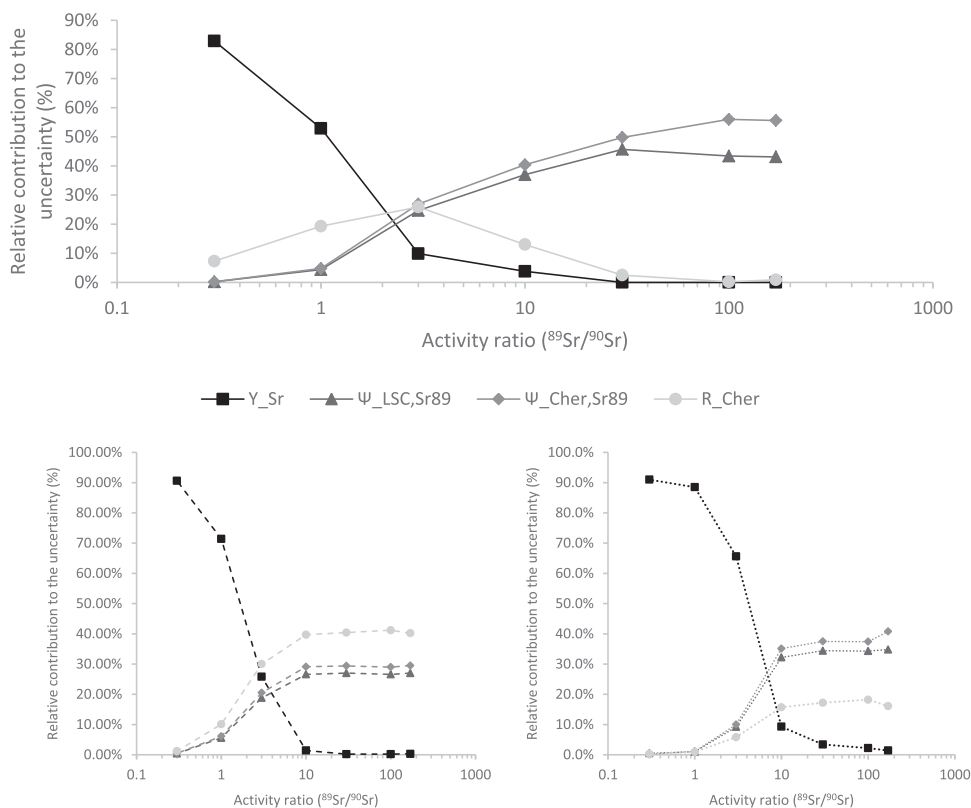The suggested approach is therefore that, in order to optimize the

Table 4
The added and measured ^{90}Sr activities, with their associated uncertainties, for all three rounds of experiments using Method B: Spectrum subtraction.

^{89}Sr/^{90}Sr ratio	Experimental round 1		Experimental round 2		Experimental round 3	
	Added [Bq]	Measured [Bq]	Added [Bq]	Measured [Bq]	Added [Bq]	Measured [Bq]
170	9.14 ± 0.25	9 ± 90	0.100 ± 0.008	− 0.8 ± 1.1	0.339 ± 0.020	2.7 ± 2.3
100	9.18 ± 0.24	9 ± 47	0.170 ± 0.006	− 0.5 ± 1.1	0.577 ± 0.020	3.2 ± 2.3
30	8.15 ± 0.24	4 ± 10	0.543 ± 0.022	− 0.6 ± 1.1	1.955 ± 0.026	3.8 ± 2.4
10	9.14 ± 0.26	8.2 ± 4.3	1.678 ± 0.034	1.3 ± 1.1	5.94 ± 0.28	6.5 ± 2.4
3	9.18 ± 0.24	5.3 ± 2.7	5.59 ± 0.11	6.6 ± 1.3	19.01 ± 0.38	16.4 ± 3.3
1	9.15 ± 0.24	7.9 ± 1.1	19.43 ± 0.39	18.4 ± 2.2	56.7 ± 1.2	50.1 ± 6.5
0.3	9.12 ± 0.24	8.51 ± 0.94	66.5 ± 1.3	65.0 ± 6.6	199.0 ± 4.3	168 ± 22

Fig. 4. Uncertainty budgets for the ⁹⁰Sr determination using Method B (subtraction). The top figure is for round 1 (solid lines), the bottom left figure is for round 2 (dashed lines) and the bottom right is for round 3 (dotted lines).

use of resources and time, the bulk of samples in an emergency are measured only with regards to ⁸⁹Sr using Cherenkov counting, about 4 days after an incident (Tovedal et al. 2009a). In order to determine the long time effect it is recommended that a few representative samples are analyzed with regards to ⁹⁰Sr using the chemical separation method, in which ⁸⁹Sr and ⁹⁰Sr are measured independently and therefore will give reliable results.

5. Conclusions

This work shows the importance of performing independent measurement of ⁹⁰Sr in order to obtain reliable results for both ⁸⁹Sr and ⁹⁰Sr, especially for activity ratios > 3, in case of a radiological incident involving fission of plutonium and uranium. By testing three different methods a conclusion was drawn as to the suitability of simultaneous determination, in this work represented by spectrum subtraction (Method B) and spectrum deconvolution (Method C). It was shown that simultaneous determination is associated with large uncertainties in particular for ⁹⁰Sr activities, and especially for high activity ratios. Therefore, it is arguably more time and cost efficient to deliver reliable
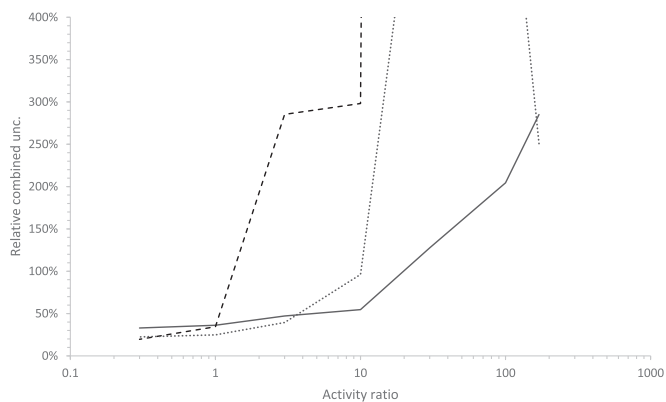

Fig. 5. The relative combined uncertainties obtained by the deconvolution method for the measured ⁹⁰Sr activities as a function of activity ratio. The solid lines represent samples with a fixed ⁹⁰Sr activity (round 1), the dashed line is for samples from round 2 and the dotted line represents samples from round 3.

Table 5
The added and the measured ⁹⁰Sr activities, with their combined uncertainties, for all three rounds of experiments using Method C: Spectrum deconvolution.

| ⁸⁹Sr/⁹⁰Sr ratio	Experimental round 1		Experimental round 2		Experimental round 3	
	Added [Bq]	Measured [Bq]	Added [Bq]	Measured [Bq]	Added [Bq]	Measured [Bq]
170	9.11 ± 0.25	6 ± 18	0.079 ± 0.002	− 0.1 ± 2.9	0.333 ± 0.010	− 2.2 ± 5.6
100	9.09 ± 0.24	7 ± 14	0.162 ± 0.003	− 0.3 ± 3.1	0.579 ± 0.013	− 0.7 ± 4.5
30	9.12 ± 0.24	7 ± 8	0.494 ± 0.010	− 0.0 ± 3.6	1.988 ± 0.044	0.6 ± 4.8
10	9.12 ± 0.26	11.6 ± 6.4	1.528 ± 0.030	1.0 ± 3.1	6.01 ± 0.13	4.7 ± 4.5
3	9.12 ± 0.24	7.4 ± 3.5	5.015 ± 0.10	1.8 ± 5.1	19.09 ± 0.42	15.0 ± 5.9
1	9.10 ± 0.24	9.5 ± 3.4	14.68 ± 0.29	13.2 ± 4.5	56.8 ± 1.3	51 ± 12
0.3	9.03 ± 0.24	8.8 ± 2.9	50.1 ± 1.0	44.3 ± 8.7	199.5 ± 4.4	174 ± 39
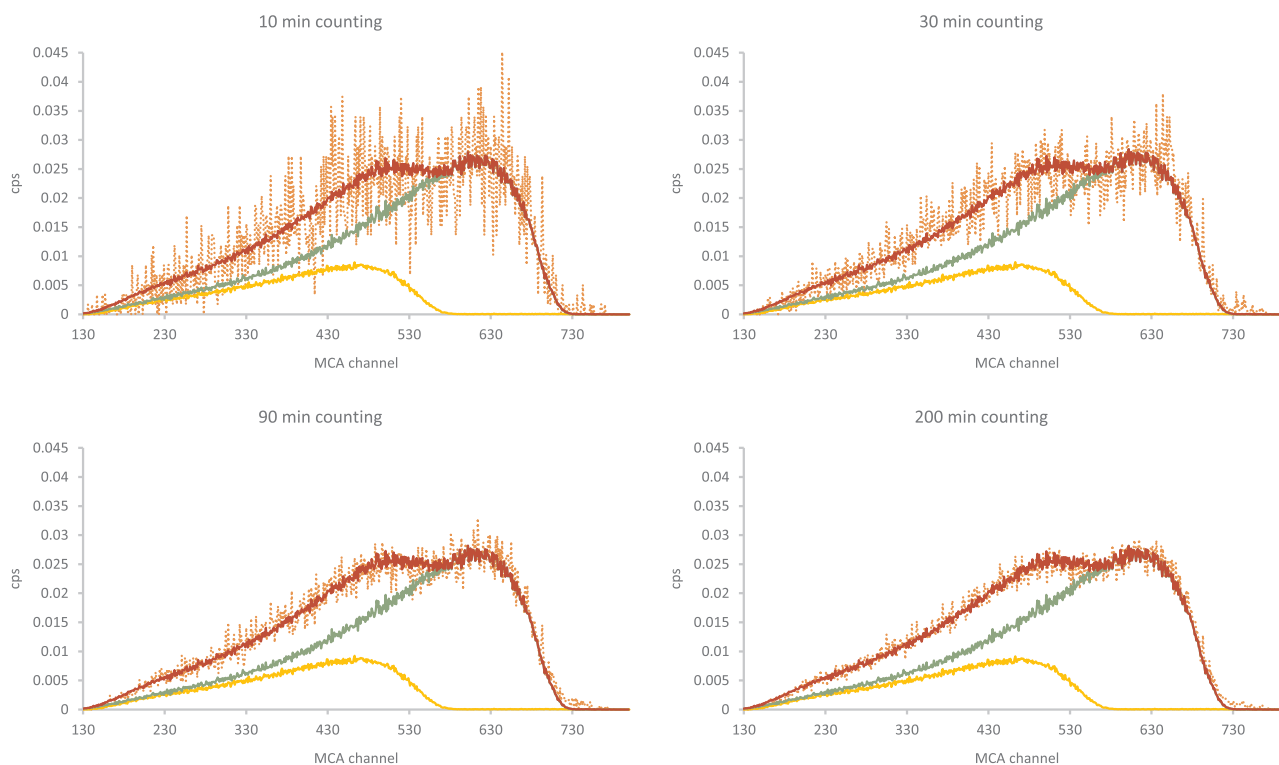
Fig. 6. The difference in fit for measurements of 10, 30, 90 and 200 minutes of the same sample. The measured spectra (dotted, orange), the two component spectra (^{89}Sr, green; ^{90}Sr, yellow) and the fitted model spectra (red). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

results, *i.e.* results with combined uncertainties no greater than 25% ($k = 2$), than saving a day or two on a method that delivers results that are less suitable for decision making. In conclusion, a comparison of the results shows that it is preferable to perform independent determination of ^{90}Sr in a nuclear emergency situation, *i.e.* measure ^{89}Sr after a first chemical separation and thereafter ^{90}Sr via its daughter radionuclide ^{90}Y after a second chemical separation. However, to optimize resources, the majority of the samples could be measured only with respect to ^{89}Sr and only a few representative samples also with respect to ^{90}Sr using the most reliable method which comprises of a second chemical separation (Method A), since this method facilitates independent measurements of ^{89}Sr and [90]Sr. Moreover, this method gives reliable results up to activity ratios representing instant fission, *i.e.* about 170.

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