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Evaluation of EC measurement comparison for ^{137}Cs, ^{40}K and ^{90}Sr in milk powder

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SUMMARY

This report describes in detail the measurement comparison of ^{137}Cs, ^{40}K and ^{90}Sr in milk powder among 60 European laboratories monitoring radioactivity in the environment and foodstuff. Milk as important component of human nutrition is an object of regular monitoring with respect to its radioactivity content. Milk powder reference material (IAEA-152) with elevated levels of radioactivity (not spiked but metabolised from contaminated feed) was bought from the International Atomic Energy Agency (IAEA) in form of hard lumps and re-processed to the form of free-flowing powder. Reference values traceable to SI units were established at IRMM and the homogeneity of the batch of distributed samples was demonstrated. The sample preparation and measurement processes applied in the participating laboratories are described and the results of the intercomparison are presented and discussed in detail.

A robust evaluation of the performance of individual laboratories is performed using three different approaches: relative deviations, E_n numbers and 'POMPlots'. One and four (for ^{137}Cs and ^{40}K, respectively) out of 59 reported measurement results have relative deviations larger than ± 20 % from the IRMM reference value. In addition to the results from this group of laboratories, another 6 and 9 results for these two radionuclides, respectively, do not fulfil the criteria of the compatibility test based on E_n numbers. For ^{90}Sr, ten laboratory results deviate by more than 30 % from the reference value, and about one third out of all 45 reported results were not compatible with the E_n criterion. These are problems which need to be addressed by the concerned laboratories.

Furthermore, many of the reported uncertainty values are not estimated as combined uncertainty of the whole measurement process following the concepts of the "GUM approach" [3]. Some laboratories, however, are able to provide realistic uncertainty estimates consistent with the reported results.

CONTENTS

GLOSSARY

BIPM	Bureau International des Poids et Mesures
CCRI(II)	Comité Consultatif des Rayonnements Ionisants, Section 2
CIEMAT	Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas
DG	Directorate General of the European Commission
GUM	Guide to the Expression of Uncertainty in Measurement [3]
IRMM	Institute for Reference Materials and Measurements
IAEA	International Atomic Energy Agency
ICP-MS	inductively coupled plasma mass spectrometry
ICP-OES	inductively coupled plasma optical emission spectrometry
ICS-REM	Interlaboratory comparison scheme for radioactivity environmental monitoring
IDF	International Dairy Federation
ISO	International Organization for Standardization
LS	liquid scintillation
LSC	liquid scintillation counter, liquid scintillation counting
NIST	National Institute of Standards and Technology
SI	Système International d'Unités, International System of Units
SIR	Système International de Référence, International Reference System for radionuclides
UTC	Coordinated Universal Time
A_{lab}	mean laboratory result of activity concentration
A_{ref}	reference value of activity concentration
D	difference between the reported and the reference activity concentration
E_n	performance statistic E_n number
k	coverage factor according to GUM
MAD	median absolute deviation
s	standard deviation
u	standard uncertainty according to GUM
u_c	combined standard uncertainty according to GUM
U	expanded uncertainty according to GUM
U_{lab}	expanded uncertainty of mean laboratory result
U_{ref}	expanded uncertainty of reference value

1. Introduction

Under the EURATOM Treaty of 1957, covering responsibilities in the peaceful use of nuclear energy in the European Union, Member States are obliged to monitor radioactivity levels in the environment of their countries (Art. 35) and to regularly report the measured values to the European Commission (Art. 36). Networks for routine and emergency measurement and communication of radioactivity values have been established. The Commission Recommendation 2000/473/Euratom of 8 June 2000 specifies in detail what environmental and food matrices should be monitored for which radionuclides "for the purpose of assessing the exposure of the population as a whole". Milk is mentioned as one of the foodstuffs to be monitored.

In order to obtain more information on the measurement methods and on the quality of the values reported by the Member States, measurement comparison exercises have been conducted regularly by the European Commission through its Joint Research Centre (JRC), Institute for Environment and Sustainability (IES). Since 2003, the JRC Institute for Reference Materials and Measurements, IRMM, organises these measurement comparisons as support to the Directorate General for Energy and Transport (DG TREN H.4).

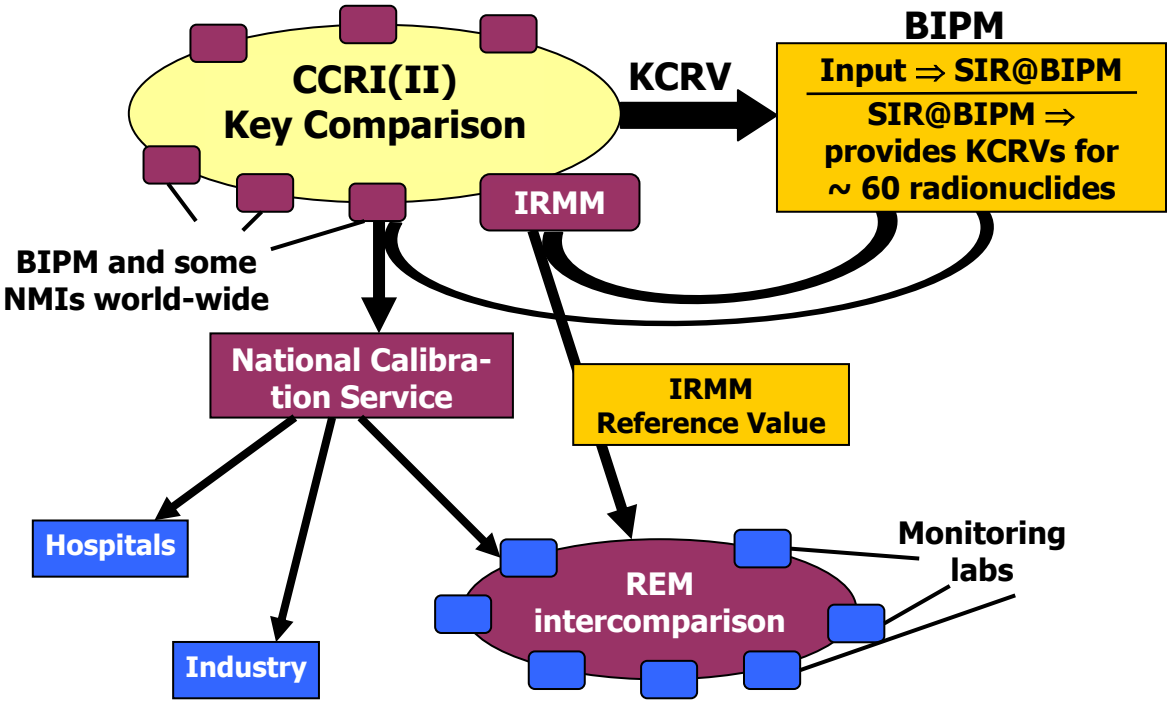

Fig. 1: Key comparisons of CCRI(II) and traceability of reference values for samples provided by IRMM for intercomparisons amongst monitoring laboratories (KCRV = key comparison reference value)

The approach of IRMM in organising the comparisons is sketched in Figure 1. As member of the Consultative Committee for Ionising Radiation (CCRI), IRMM is participating in key comparisons among National Metrology Institutes (NMIs), which serve to "realise" the unit of radioactivity, the becquerel. Results of key comparisons,

which are based on primary measurement techniques, i.e. without resorting to other activity standards, are introduced in the International Reference System for radionuclides (SIR) to determine the SIR calibration factor for that particular nuclide. Since each nuclide has its own decay scheme, all calibration factors are different and need to be experimentally determined for each nuclide. Thus, the SIR system at BIPM, the Bureau International des Poids et Mesures in Sèvres close to Paris, is the world-wide standard for radioactivity, realised as an ionisation chamber.

Due to IRMM's participation in key comparisons and the direct link to the SIR, it can work with standardised solutions which are directly traceable to the SI unit. National metrology institutes, having the same short traceability link to the SIR, usually provide calibration standards through their national calibration services. Such standards can be used for example by radioactivity monitoring laboratories to calibrate their measurement equipment. Parallel to this traceability chain, IRMM offers intercomparisons with samples which have their own traceable reference value. Usually such samples are, in physical properties as well as amount of radioactivity, closer to the routine measurement conditions of a monitoring laboratory than the calibration standards. Thus, this kind of intercomparison can serve as an independent and impartial performance check with samples of high credibility.

This report presents the results of the comparison exercise organized by IRMM on ^{137}Cs, ^{40}K and ^{90}Sr activity concentration in milk powder.

The principal objective of this exercise was to give the opportunity to individual laboratories for checking their procedures of measurement and at the same time to provide direct evidence of the comparability of the results from different laboratories to the benefit of both users (EC DG TREN, Member State authorities) and laboratories. In addition, where possible, the extent of variation should be quantified, and possible causes pointed out.

Description of the sample:

Nature: milk powder reference material (IAEA-152) with elevated levels of radioactivity, not spiked, but metabolised from contaminated feed

Reference date: 1 January 2005, 0:00 UTC

Recommended half-life of:

***^{137}Cs**:* $T_{1/2}$ = (10981 ± 11) days = (30.065 ± 0.030) years [1]

^{40}K: $T_{1/2} = (1.265 \pm 0.013) \cdot 10^9$ years [2]

^{90}Sr: $T_{1/2}$ = (10551 ± 14) days = (28.89 ± 0.038) years [1]

where the numbers following the symbol ± are the numerical values of an expanded uncertainty *U* with a coverage factor *k* = 1, corresponding to a level of confidence of about 68 % [3].

Reference activity concentrations:

***^{137}Cs*:** (1480 ± 110) Bq.kg^{-1}

***^{40}K*:** (540 ± 40) Bq.kg^{-1}

***^{90}Sr*:** (4.9 ± 0.4) Bq.kg^{-1}

where the numbers following the symbol ± are the numerical values of an expanded uncertainty *U* with a coverage factor *k* = 2, corresponding to a level of confidence of about 95 %.

Shipping: in amber glass bottles via express mail (by DHL)

Reporting of the results:

^{137}Cs, ^{40}K and ^{90}Sr activity concentrations normalised to dry mass reported as measured values (Bq·kg^{-1}) with the associated expanded uncertainty *U* (expanded uncertainty $U = k \cdot u_c$, where *U* is determined from the combined standard uncertainty u_c with a coverage factor k=2, corresponding to a level of confidence of about 95 %).

The necessary correction to dry mass had to be determined on separate (small) sub-samples, taken from the bottles at about the same time as the samples for radionuclide analysis to be representative for their water content. Recommended methods for determination of the water content / moisture were [4]:

- Karl-Fischer titration (preferred), or

- drying 1 to 3 g in an oven at 102 °C ± 2 °C during 2 hours at atmospheric pressure, testing for constant mass (< 0,5 mg difference) by additional drying steps of 1 hour, according to the drying method of the International Dairy Federation (IDF method).

Participating laboratories:

The intercomparison exercise was designed to evaluate the performance of the participating laboratories in measuring the ^{137}Cs, ^{40}K and pure beta-decaying ^{90}Sr activity concentrations in milk powder. Participating laboratories were mainly national research institutes and authorities in the EU member states and the accession countries. The laboratories were nominated by the national representatives* in the expert group according to Euratom Treaty Art. 35 and 36.

In total 63 laboratories (49 from the member states**, 11 from the candidate countries** and Western Balkans, 2 from Switzerland, and 1 from Iceland) registered for participation and 60 reported measured values. The list and addresses of all 63 laboratories are presented in chapter 11. Since anonymity is a requirement in this programme of measurement comparisons, the identity of the laboratories is not shown in the compilation of the results. The order of the listing of participants in chapter 11 is not the same as the laboratory number used throughout the data evaluation and comparison.

Timetable and deliverables:

30 Sept. 2004	re-processing of the milk powder material at IRMM
20 Oct. 2004	the participating laboratories are nominated by the national representatives
31 Dec. 2004	milk powder samples are sent to the participants
31 March 2005	the on-line reporting system is set up according to the requirements of the current exercise
30 Apr. 2005	laboratories submit their results and questionnaire to IRMM

* They generally represent their national regulatory bodies for radiological protection.
** Status as of end of 2005.

2. Processing and packaging of the material batch

20 kg (80 plastic bottles of 250 g each) of a milk powder reference material (IAEA-152) with elevated levels of radioactivity (not spiked, but metabolised from contaminated feed) was bought from the International Atomic Energy Agency (IAEA) in form of hard lumps and re-processed to the form of free-flowing powder. The re-processing of the material was done by the Reference Materials Unit of the IRMM.

The bottles were cut open and the material was collected in a big plastic drum where it was crushed as much as possible using a heavy polytetrafluoroethylene (PTFE) pestle. The homogenization was done in a Turbula mixer for 30 minutes. Then the powder was sieved through a 125 μm stainless steel sieve. What remained on the sieve was further crushed with the PTFE pestle and the remaining fraction of about 500 g of hard lumps was crushed in a conventional Moulinex mixer and put together with the rest of the material. Finally the material was homogenized in the Turbula mixer for an additional 30 minutes.

Prior to bottling, the material was again homogenized for another 30 minutes in the Turbula mixer. The milk powder was filled manually in amber glass bottles of 280 mL. In total 183 bottles were filled with approximately 103 g of powder.

Particle size analysis using laser light diffraction demonstrated the homogeneity of the powder in particle size over the full sequence of bottles. The top particle size was found to be approximately 200 μm, which is generally considered small enough to expect a reasonably homogeneous distribution of the measurands in the powder.

The water content of the milk powder was determined by Karl-Fischer titration [4]. Two parallel determinations for ten bottles were done. The water content was found to be low with an average of (3.89 ± 0.13) g/100g, but carrying an expanded uncertainty U = 0.5 g/100g (Annex 1). Furthermore, water activity measurement (measurement of water sorption) [4] showed that the material was not grossly hygroscopic.

3. Establishment of reference values traceable to SI units

The reference values of the activity concentrations of the three radionuclides in the milk powder were established at IRMM [5]. The reference measurements were carried out in the frame of the IAEA co-ordinated research project "Upgrading of Analytical Quality Control Services intercomparison materials to reference materials with assigned property values traceable to the International System of Units". The activity concentrations in the milk powder of ^{137}Cs and ^{40}K were measured by γ-ray spectrometry and that of ^{90}Sr by liquid scintillation counting (LSC) after chemical separation.

Before the measurement, the bottles with milk powder were homogenised, using a 3D Turbula mixer and the powder was dried in an oven for 48 hours at 105°C to constant weight. After cooling to room temperature in a desiccator, a known amount of milk powder (about 50 g dry mass) was placed in cylindrical containers (125 mL polypropylene, Nalgene, USA). The samples were weighed using a Sartorius analytical balance (type 1712), which was calibrated with the weighing set Mettler Toledo M7 traceable to the IRMM kilogram, which in turn is directly traceable to the SI unit through comparison measurements at the BIPM (Bureau International des Poids et Mesures, Sèvres, France). In total 30 samples were prepared.

After tapping (compacting) the samples were placed directly on the detector end-cap and measured by γ-ray spectrometry for 4 to 12 days each. A low-background HPGe detector system was used for the measurements. The semi-planar detector (EURISYS) consisted of a HPGe crystal, 30 mm in length and 80 mm in diameter, with 45 % relative efficiency and a carbon epoxy end-cap window. The detector was housed in a 10 cm thick Pb shield of square intersection, lined with 1 mm Cu; the inner 5 cm of the Pb shield was made of high radiopurity Pb.

The detector system was calibrated for peak efficiency using single-nuclide point sources, as well as multi-nuclide liquid standards prepared in the same geometry as the actual samples. In addition, an actual sample was spiked with known amounts of standard ^{54}Mn, ^{60}Co, ^{65}Zn, ^{137}Cs and ^{241}Am solutions, mixed thoroughly and measured in identical geometry in order to obtain information on the matrix self-absorption of the actual samples. The standard radionuclide solutions used to prepare the calibration standards (both point sources and volume sources) originated from standardisation campaigns - usually key comparisons organised by BIPM/CCRI(II) (Bureau International des Poids et Mesures/Comité Consultatif des Rayonnements Ionisants) [5]. By comparing the experimentally determined efficiency calibrations for the point sources, liquid standards and the spiked milk powder with Monte Carlo simulations for these geometries using the code GEOLEP [6, 7], the uncertainty of the calibrations could be assessed. A relative combined standard uncertainty of 3.6 % was estimated for the efficiency calibration in the chosen geometry of milk powder samples [5].

The reproducibility of the measurements was tested by placing a sample in front of the detector and performing a series of measurements, then replacing the sample in front of the detector and repeating the measurements. The former tested the statistical reproducibility, while the latter included the geometrical (repositioning) repeatability. The standard deviation for the statistical reproducibility was 0.15 % for 10 one-day long measurements and that for the geometrical repeatability 0.32 % for

5 one-day long measurements. Since the latter contains the statistical uncertainty as well, a contribution of 0.2 % was assigned to the geometrical repeatability alone in the uncertainty budget.

The measured data were corrected for background, decay and decay during measurement. Statistical tests were applied to check the results for consistency. The tests applied were Dixon's, Grubb's, coefficient of skewness and coefficient of kurtosis. No outliers were found in any of the activity concentration results, according to the Dixon's and Grubb's tests.

The measurement of ^{90}Sr requires Sr to be separated from the matrix and from other interfering radionuclides first. The method applied was based on the digestion of the sample, the separation of Sr by extraction chromatography and the subsequent measurement of the activity by liquid scintillation counting (LSC).

Because of the low ^{90}Sr activity concentration in the milk powder, it was necessary to use large amounts of sample, i.e. of the order of 50 g. The material was first dried in an oven at 95°C to constant weight. The mass of each aliquot used was determined gravimetrically, using a Mettler-Toledo analytical balance (model AT21), calibrated with standard weights traceable to the IRMM kilogram. After adding the tracer (^{85}Sr), the sample was decomposed by wet digestion with concentrated nitric and hydrochloric acids, in combination with ashing at high temperatures (up to 450°C) in a muffle furnace. Stable Sr was added as carrier and a co-precipitation with calcium oxalate was performed to remove Sr from most of the matrix. Then, Sr was separated from the re-dissolved precipitate by extraction chromatography by means of the Eichrom Sr resin (Eichrom Technologies, Inc., Darien, IL, USA). The pure Sr fraction was evaporated and the residue transferred with 6 mL 0.05 mol/L HNO_3 into a scintillation vial (20-mL High-Performance Packard vial) containing 14 mL of Insta-Gel Plus LS cocktail to be measured by LSC.

For the determination of ^{90}Sr the samples were measured using a Wallac Quantulus 1220 ultra low-level LS spectrometer immediately after separation of Sr and several times later, with blanks introduced before and after each sample measurement. The blanks were prepared by adding 6 mL 0.05 mol/L HNO_3 into 14 mL of Insta-Gel Plus LS cocktail. The data reduction and analysis included the background subtraction, decay correction, correction for decay during measurement, correction for the contribution of the tracer (^{85}Sr) and the ingrowth of ^{90}Y.

Since the samples went through digestion and chemical separation in order to isolate the strontium, a tracer for the chemical recovery calculation was used (^{85}Sr). It was measured by γ-ray spectrometry and the chemical recovery was calculated as the ratio of the counts under the 514-keV γ-ray peak of the sample to that of a reference source in the same geometry.

The activity concentration of ^{90}Sr was determined by LSC using the CIEMAT/NIST ^{3}H efficiency tracing method [8, 9, 10], requiring ^{3}H standards only for the instrument efficiency calibration.

The final results of the activity concentrations for ^{137}Cs, ^{40}K and ^{90}Sr are presented in Table 1. The uncertainties of Table 1 are expanded uncertainties U (k = 2). Special care was taken to ensure traceability to the SI units by means of the calibrated standard weights, the standard efficiency calibration sources for γ-ray spectrometry, the chemical recovery tracer ^{85}Sr and the efficiency tracer ^{3}H, and the use of the Universal Time Coordinated (UTC), generated and distributed by Physikalisch-

Technische Bundesanstalt (PTB), Germany [11]. Whereas the reference values were originally established [5] for the reference date of 31 August 1987, 0:00 UTC, they are re-evaluated in Table 1 for the purposes of this comparison exercise to the reference date 1 January 2005, 0:00 UTC. It should be noted that using either set of half-lives renders the same activity concentrations (within the significant number of digits reproduced in Table 1) for the reference date 1 January 2005.

Table 1: Reference values of activity concentration for ^{137}Cs, ^{40}K and ^{90}Sr calculated for different reference dates. The expanded uncertainty *U* includes a contribution from homogeneity (see Table 2 and chapter 4).

Nuclide	Reference date* 31-08-1987, 0:00 UTC			**Reference date** 01-01-2005, 0:00 UTC**		
	activity conc. / (Bq·kg^{-1})	expanded unc. *U* / (Bq·kg^{-1})	$T_{1/2}$ / d	**activity conc. A_{ref} / (Bq·kg^{-1})**	**expanded unc. U_{ref} / (Bq·kg^{-1})**	**$T_{1/2}$ / d**
^{137}Cs	2210	160	11020	1480	110	10981
^{40}K	540	42	$4.66 \cdot 10^{11}$	540	50	$4.68 \cdot 10^{11}$
^{90}Sr	7.4	0.7	10519	4.9	0.5	10551

* using IAEA-recommended half-lives
** using half-lives recommended to the participants

The uncertainty budgets for the reference values given in Table 1 are presented in detail in Tables 2a to 2c. The values in the second column are stated in the form of propagated uncertainty contribution to the final result. This is not necessarily the uncertainty in the corresponding component itself. The estimation of the material homogeneity contributing to the combined and expanded uncertainties in Tables 1 and 2 is described in chapter 4.

Table 2: Uncertainty budgets for the reference values of the three radionuclides (a) ^{137}Cs, (b) ^{40}K and (c) ^{90}Sr given in Table 1. Here $u_{c\text{-}hom}$ denotes combined uncertainty without taking homogeneity into account.

a) Cs-137

Component	Uncertainty / %
Counting statistics (incl. background)	0.2
Weighing	0.01
Sample positioning	0.2
Dead time	0.005
Efficiency (incl. interpolation and simulation comparisons)	3.6
Gamma-ray emission probability	0.24
Timing	0.005
Half-life	0.2
Combined standard uncertainty $u_{c\text{-}hom}$	3.62
Homogeneity at 40 g sample intake (see chapter 4)	0.45
Combined standard uncertainty u_c	3.65
Expanded uncertainty U (with coverage factor k=2)	7.3

b) K-40

Component	Uncertainty / %
Counting statistics (incl. background)	0.3
Weighing	0.01
Sample positioning	0.2
Dead time	0.005
Efficiency (incl. interpolation and simulation comparisons)	3.6
Gamma-ray emission probability	1.0
Timing	0.005
Half-life	0.0
Combined standard uncertainty $u_{c\text{-}hom}$	3.75
Homogeneity at 40 g sample intake (see chapter 4)	0.7
Combined standard uncertainty u_c	3.8
Expanded uncertainty U (k=2)	7.7

c) Sr-90

Component	Uncertainty / %
Counting statistics (incl. background)	0.9
Weighing	0.2
Dead time	0.05
Chemical recovery	3.5
Timing	0.05
Efficiency (incl. quenching and interpolation from curve)	1.0
Ratio Y-90/Sr-90	0.1
Sample stability	0.1
Half-life	0.11
Combined standard uncertainty $u_{c\text{-}hom}$	3.8
Homogeneity at 50 g sample intake (see chapter 4)	2.0
Combined standard uncertainty u_c	4.3
Expanded uncertainty U (k=2)	8.6

4. Homogeneity measurements

An inhomogeneity in radionuclide concentration increases the uncertainty of the corresponding reference value, which – by definition – is assumed to be valid for the whole batch of comparison material. The procedure given in ISO/FDIS 13528:2005 [12] Annex B was used to check for the homogeneity of the batch of samples with respect to ^{137}Cs and ^{40}K activity concentration. In addition, (in)homogeneity was quantified to estimate its contribution as standard uncertainty to the uncertainty budget of both reference values (Table 2a and 2b). In order to evaluate the within-samples and between-samples standard deviations, ten bottles of the comparison material selected from the entire batch were taken to prepare two parallel samples of approximately 40 g from each bottle. 40 g of sample were considered to be a practical minimum sample mass for γ-ray spectrometry.

Homogeneity measurements were performed by γ-ray spectrometry using an extended range (XtRa) p-type HPGe coaxial detector from Canberra. The system is a low-background, high-resolution detector for the energy range 40 to 3000 keV with a high relative efficiency of 90 %. The detector crystal has a diameter of 77 mm and 78 mm length, housed in an end-cap with an Al window 1 mm thick. The detector system is shielded by 5 cm of Pb.

All 20 samples were prepared gravimetrically, corrected to dry mass based on the water content determined for each of the 10 bottles. Cylindrical polypropylene beakers with lid were used with a diameter of about 63 mm, a height of about 73 mm and volume of 125 mL (Nalgene, USA). During the sample filling an electrostatic discharge blower helped to avoid uncontrolled dispersion of the powder. To create a denser sample a “tapper” was used.

Each sample was measured once for 24 hours (86 400 s). Data acquisition was done using the “MCA – Measurement System v1.0” of MK System BVBA (custom made for IRMM). The spectra were evaluated with GammaVision - 32 Software.

In order to study homogeneity of samples only relative measurements of high precision are necessary. Therefore, no efficiency calibration was performed, and the net peak counting rates of ^{137}Cs and ^{40}K were determined under reproducible measurement geometry. Figures 2 and 3 illustrate the achieved homogeneity by the very small variations observed. The measured net peak counting rates of ^{137}Cs varied from 2.26 to 2.31 counts·s^{-1}, those of ^{40}K in the range from 0.166 to 0.172 counts·s^{-1}. The error bars indicate combined standard uncertainty of the individual measurement. The individual relative deviation, i.e. the deviation of the individual counting rate from the mean value, is less than 2 % in all cases.

A homogeneity check strictly limited to the principles of the ISO standard [12] requires that the *between-samples standard deviation* $s_s = \sqrt{s_x^2 - (s_w^2/2)}$ would contribute less than 10 % to the *standard deviation for proficiency assessment* σ (which is in our evaluation approach replaced by the uncertainty u_c of the reference value) without, however, actually using s_s as contribution to σ or u_c. Here s_x is the standard deviation of sample averages and s_w the within-samples standard deviation. Applying this evaluation renders the result *“homogeneous”* for the nuclide ^{40}K, but the nuclide ^{137}Cs cannot be evaluated, because the square root of s_s renders an

unphysical (imaginary) result. If only the (unreduced) expression $\sqrt{s_x^2}$ were taken, then the result for ^{137}Cs would also be *"homogeneous".* This limitation in validity of

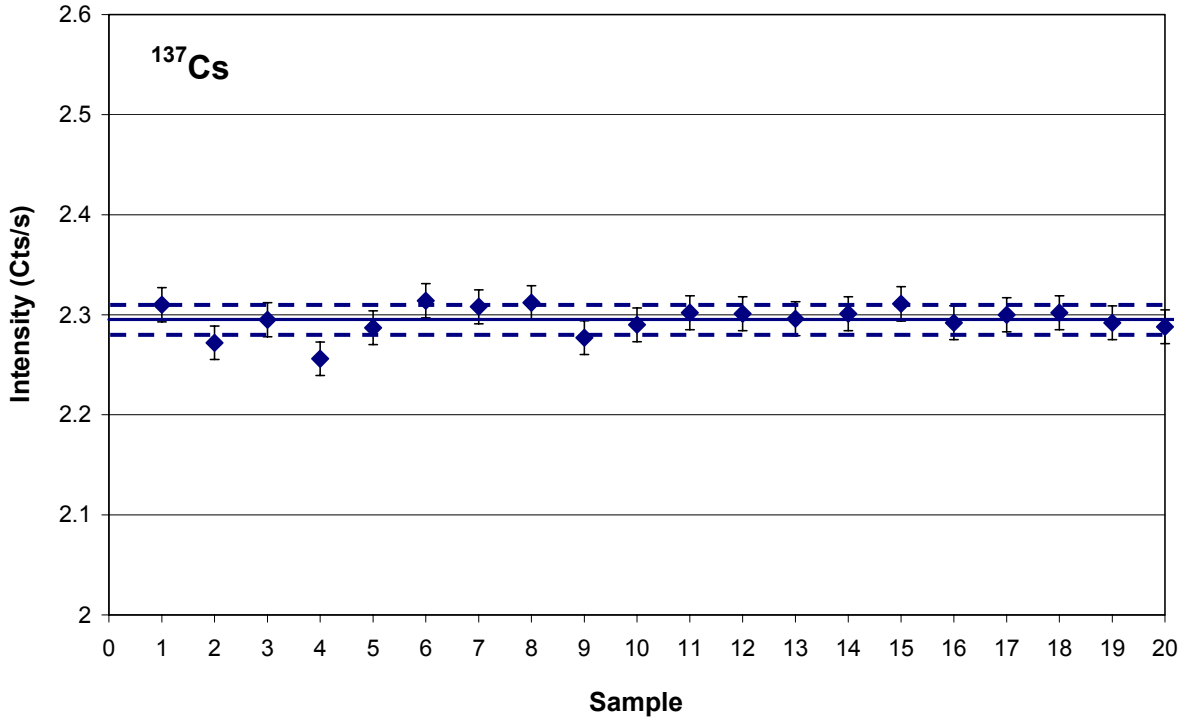

Fig. 2: ^{137}Cs count rate in milk powder samples of 40 g mass

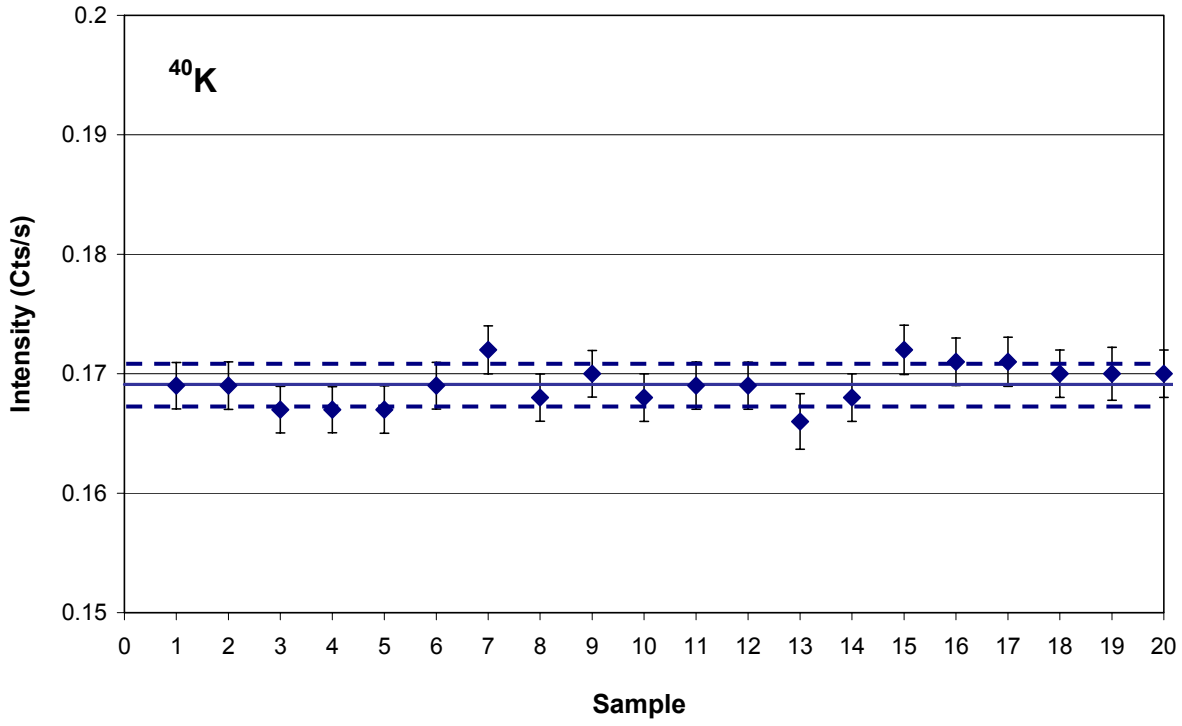

Fig. 3: ^{40}K count rate in milk powder samples of 40 g mass

the ISO standard formalism is not surprising for a well-prepared homogeneous batch of material. With modern homogenisation and filling techniques, the variation between bottles can be of the same magnitude as the variation within a bottle.

Beyond the strict application of the ISO standard, the relative quantitative results for $s_s(^{40}K)$ = 0.7% and $\sqrt{s_x^2}$ (^{137}Cs) = 0.45% can now be used as standard uncertainties, describing the (in)homogeneity of the comparison material at the chosen minimum sample intake of 40 g and contributing to the combined uncertainty u_c of the reference values (Table 2a and 2b).

An alternative evaluation uses only the standard deviation s_{bb} of all 20 sub-samples, knowing that this results in an overestimation of real physical inhomogeneity, since the reproducibility of the measurements (in particular counting statistics) is not accounted for. The mean value and the standard deviation of the measured counting rates (indicated in Figs. 2 and 3 by solid and dashed blue lines) are (2.295 ± 0.015) counts·s^{-1} for ^{137}Cs, and (0.1690 ± 0.0017) counts·s^{-1} for ^{40}K. In relative terms, these standard deviations correspond to s_{bb} (^{137}Cs) = 0.64% and s_{bb} (^{40}K) = 1.0%, well consistent with the results above when considering the intrinsic overestimation.

During the characterisation of the material [5] the homogeneous distribution of ^{137}Cs and ^{40}K activity concentration in the samples were studied as well. Two samples each of about 50 g were prepared from 15 bottles and measured for homogeneity. A one-way ANOVA statistical test was performed on the activity concentration results to test the hypothesis that all sets of data represent the same mean, using the computer program Igor (version 5.04B, Wavemetrics). The one-way ANOVA test performed [5] demonstrates that the material has an inhomogeneity of ≤ 1 % at 50 g sample intake with respect to both radionuclides. These results are fully consistent with the homogeneity results determined above.

The homogeneous distribution of ^{90}Sr in the comparison material was evaluated together with the determination of the reference value [5]. No separate homogeneity measurements (like in the case of ^{137}Cs and ^{40}K) were done. The sample preparation and the measurements of ^{90}Sr in the milk powder are described in chapter 3. The measured activity concentration of ^{90}Sr (in sub-samples of 50 g) varied from 7.0 to 8.1 Bq·kg^{-1} and the mean activity concentration was (7.4 ± 0.3) Bq·kg^{-1}, indicated in Fig. 4 by a solid and dashed blue lines. The error bars indicate combined standard uncertainties of the individual measurements with the major contributions being the chemical recovery and the counting statistics. These results are decay corrected to the reference date 31 August 1987 0:00 UTC.

Since the determination of the ^{90}Sr reference value and with it the evaluation of its homogeneity were performed on bulk material prior to its packaging in final form (bottles of about 100 g), the procedure described in the ISO standard [12] could not strictly be applied. Instead, a one-way ANOVA statistical test was performed to test the hypothesis that all sets of data are represented by the same mean (material is *"homogeneous"*), and in addition to estimate the uncertainty contribution due to inhomogeneity in ^{90}Sr. Using the computer program Igor (version 5.04B, Wavemetrics), the 15 samples were put in random order and divided into 3 "treatments" (groups) of 5 and the test was repeated for each group. This separates the common measurement reproducibility from the physical differences between samples. The results of the one-way ANOVA test demonstrate that the null hypothesis has high credibility (material *"homogeneous"*) and estimate the material inhomogeneity in ^{90}Sr to be 2 % at 50 g sample intake. This

value is contributing as standard uncertainty to the combined standard uncertainty of the reference value in Table 2c.

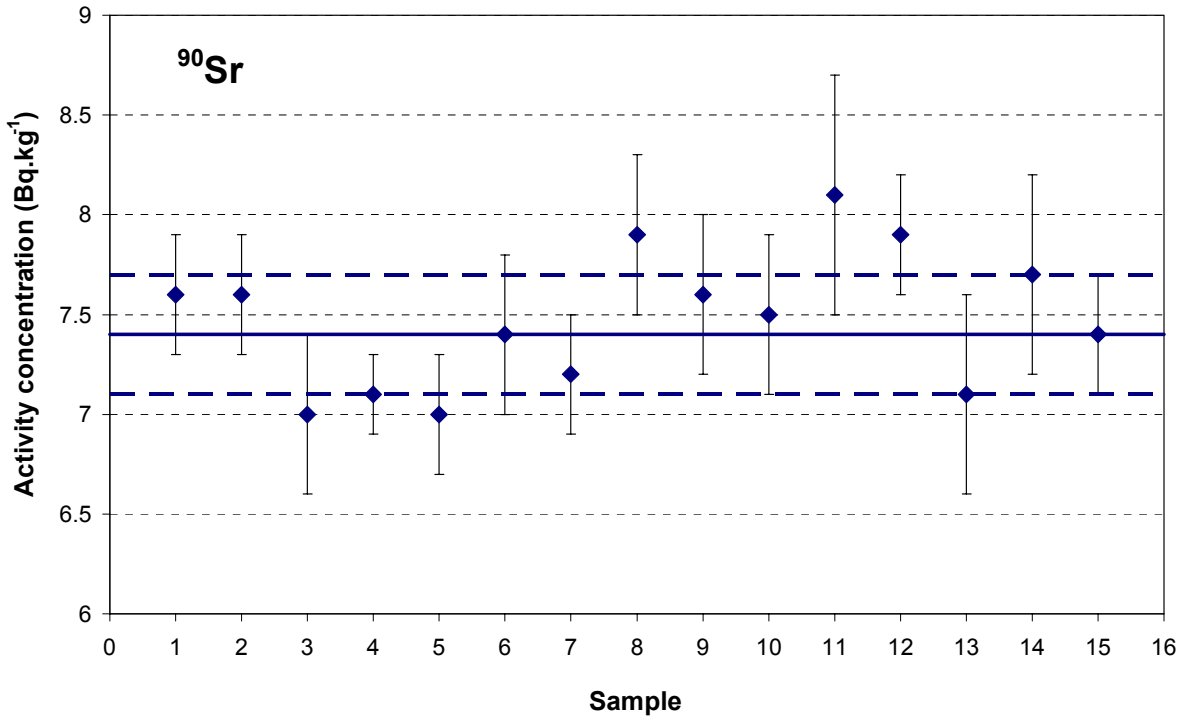

Fig. 4: ^{90}Sr activity concentration in milk powder samples of 50 g mass

5. Preparation, separation and measurement procedures used in the participating laboratories

Together with the samples a letter with short instructions (Annex 2) was sent to the participating laboratories asking them to report the activity concentration related to dry mass. It was recommended to determine the necessary correction factor to dry mass by Karl-Fischer titration or with the oven drying method of the International Dairy Federation (IDF) using separate sub-samples. None of the participants used the Karl-Fischer titration method, 54 laboratories dried the milk powder following the IDF method. Laboratory 20 dried the powder during an extended period of 24 hours. Five laboratories (labs 11, 12, 37, 53, 57) did not determine the moisture content at all. The moisture content expressed as percentage of the weighed sample was reported between 0.16 % and 8.7 %. The vast majority of laboratories (32) determined values between about 3 and 5 %, which is consistent with the findings of IRMM (3.9 % with Karl-Fischer titration). A rather significant number of 15 laboratories, however, under-determined moisture content with values ≤ 2 %. The correction factors applied to determine dry mass were reported to be between 0.99 and 0.92. Not all of these values are consistent with the reported moisture content, moreover, four laboratories said not to apply any correction factor although they reported correct moisture contents. The mass of milk powder used for performing this determination varied from 0.38 to 109 g, and 43 laboratories used sub-samples of 1 to 5 g as recommended.

In order to avoid the consumption of too much sample material for optimising the analysis conditions, additional information was sent to all participants shortly after distribution of samples (Annex 3). In this note the order of magnitude of the ^{90}Sr activity concentration was indicated as 10 Bq·kg^{-1}.

Together with the result reporting form (Annex 4), the participating laboratories were asked to fill in a questionnaire (Annex 5). The questionnaire was composed of 36 questions which were divided in two main parts - on determination of ^{137}Cs and ^{40}K (questions 3 - 20) and on ^{90}Sr separation and measurement (questions 21 - 34). In total 60 out of 63 laboratories provided data and some conclusions are presented here.

The γ-ray emitting nuclides ^{137}Cs and ^{40}K were measured by 59 laboratories. The intercomparison samples were mainly treated with the same analytical procedures as routinely used in the laboratories for this type of samples. Thirteen laboratories do not routinely analyse milk powder but fresh milk in the liquid phase (labs 13, 15, 21, 22, 23, 26, 28, 39, 41, 44, 45, 47, 48) or after calcination (lab 33). The ^{137}Cs activity concentration routinely measured by the laboratories in milk is varying from 0.1 to 30 Bq·kg^{-1}. For ^{40}K, the routinely measured values are on average 500 to 600 Bq·kg^{-1} in powder or 40 to 60 Bq·L^{-1} in fresh milk. Some of the laboratories (labs 14, 48 and 54) do not routinely measure ^{40}K.

In 90 % of the cases the samples were not chemically or physically pre-treated before measurement. In labs 11 and 17 the sample material was incinerated and another 5 laboratories tried to reconstitute milk from the milk powder by addition of distilled water but only one of the samples was actually measured in the liquid phase (laboratory 41). In two of these labs gelatine and sodium benzoate were added to form a gel sample (labs 58 and 59) and in another two the reconstituted suspensions

were found not to be homogeneous enough. Laboratories 11 and 41 performed measurements of both the powder samples as well as the treated ones (ashed and reconstituted, respectively).

Four out of five laboratories have used a cylindrical beaker placed directly on the detector end-cap as measurement geometry for their samples. The volume of the used beakers varied from 40 to 260 mL, and their sizes were between 42 and 95 mm in diameter and 9 and 100 mm in height. In 8 cases Marinelli beakers with a volume from 0.25 to 1 L were used. One laboratory pressed the milk powder to pellets before measurement, another one incinerated 33 g of powder to 2.7 g of ash and pressed this to form a pellet, and two others measured the milk powder in Petri dishes. The mass of the milk powder measured with γ-ray spectrometry varied from 23 g to 1 kg, while one laboratory used a very small mass of only 8 g in a Petri dish. In the latter case, the representativity of this sub-sample of 8 g may be questionable.

The measurements were performed with commercially available γ-ray spectrometry systems (Canberra, Ortec, Eurisys etc.) consisting of HPGe (in 90 % of the cases), BEGe or Ge(Li) detectors, p- or n-type, in coaxial, well or planar configuration using different counting geometries. Three laboratories (labs 2, 37, 56) used two different or different types of detectors for the measurement of the milk powder samples and in another 3 laboratories three types of detectors were used (labs 11, 39, 42). The nominal relative efficiency of the detectors varied from 10 to 100 %.

The detector systems were calibrated for efficiency with standard volume sources or multi-nuclide standard solutions. Six laboratories performed calibration using a radioactive point source (labs 5, 33, 42, 54, 55, 56). Laboratories 13, 38, 57 determined the efficiency of their detector systems for ^{137}Cs and ^{40}K using volume radioactivity standards in combination with commercially available calculation and simulation programmes (e.g. LabSOCS). Laboratory 31 used only LabSOCS for performing the efficiency calibration of their system. Laboratories 10, 42 and 56 used two different types of standards - standard volume and liquid sources (lab 10) or standard volume and point sources (labs 42 and 56).

The data acquisition was done using mainly analog signal processing (43 laboratories versus 20 with digital signal processing). Some of the participants used both (labs 2, 4, 10, 37, 39, 50, 63). The data evaluation was made using commercial software - most often Genie and Gamma Vision, but also InterWinner, EMCA+, Gamma-track and SAMPO. In some cases locally developed programmes were applied. Several laboratories took sample density and geometry corrections into account using Monte-Carlo simulation.

Twenty-nine laboratories also determined ^{134}Cs with activity concentrations varying from 1.4 to 3 Bq·kg^{-1} dry mass. Two participants (labs 35 and 57) claimed to have measured an activity concentration of ^{60}Co of 4 and less than 10 Bq·kg^{-1}, respectively.

One laboratory used a different method than gamma-ray spectrometry to determine ^{40}K: Lab 20 calculated the ^{40}K activity concentration from a determination of the total potassium content with atomic absorption spectrometry.

Only 45 laboratories submitted results for ^{90}Sr in the milk powder. ^{90}Sr in the comparison samples was determined with the methods routinely used by the laboratories. Thirteen laboratories (15, 21, 23, 26, 28, 30, 39, 40, 41, 44, 45, 47 and 48), however, do not normally measure milk powder but fresh milk. Trying to

reconstitute milk from the milk powder posed problems to several labs (explicitly mentioned by labs 26, 28, 30 and 41) because the resulting suspension was not homogeneous. In every third laboratory the activity concentration of ^{90}Sr found in routine monitoring of milk or milk powder is below the detection limits or decision thresholds. The reported detection limits are for milk powder typically around 0.3 to 0.5 $Bq \cdot kg^{-1}$ with a range between 0.03 and 5 $Bq \cdot kg^{-1}$. The measured values reported from routine monitoring of milk powder are typically around 0.3 to 0.5 $Bq \cdot kg^{-1}$, whereas in some Eastern European countries they are slightly higher reaching maxima in routine circumstances of 3 and 4.5 $Bq \cdot kg^{-1}$ (labs 31, 58 and 55, respectively). The routinely measured activity concentrations of ^{90}Sr in liquid milk are reported to be between 10 and 100 $mBq \cdot L^{-1}$.

Prior to the chemical separation of radiostrontium from the milk powder matrix the samples were dry-ashed in the vast majority of the laboratories (about 30), usually followed by dissolving the ash in concentrated nitric or in some cases hydrochloric acid. Three laboratories (no. 36, 47 and 51) applied wet digestion for sample destruction, two of them followed by separation on Eichrom Sr resin, one (lab 47) by solvent extraction (HDEHP). At least two laboratories performed cation exchange chromatography on a Dowex resin prior to entering further separation steps, which were oxalate precipitation and extraction chromatography on Sr resin in the case of lab 20 or solvent extraction with HDEHP (30).

The further separation and extraction was performed by a variety of methods. In many cases (at least 12 times), an oxalate (co-)precipitation was carried out followed by extraction chromatography (on Eichrom Sr resin and other crown ethers, or on a different resin) or by other separation schemes. In two labs (4 and 5) phosphate co-precipitation was combined with separation on Eichrom Sr resin. All in all, at least 7 laboratories used the Eichrom Sr resin. Other extraction schemes were based on solvent extraction such as di-(2-ethylhexyl) phosphate (HDEHP) in toluene (labs 15, 26, 28, 29, 30, 31, 40, 47, 55 and 63), tributylphosphate (TBP, no. 25, 46 and 56) and EDTA (labs 16 and 24). At least three laboratories (no. 7, 44 and 50) applied the oxalate method, with ammonium oxalate precipitation from the original leachate or dissolved ash in the first place and the final conversion (after ^{90}Sr-^{90}Y equilibrium) of the $Y(OH)_3$ precipitate into oxalate.

At least ten laboratories (10, 11, 21, 23, 30, 39, 42, 43, 45 and 48) applied the classic separation method with fuming nitric acid. (Lab 30 used indeed two methods: it analysed one sample with HDEHP solvent extraction and four samples with fuming nitric acid.) After the ^{90}Sr-^{90}Y equilibrium had been established, the ^{90}Y was precipitated as hydroxide and converted to oxalate. In most of these cases laboratories worked with a preceding ashing step for matrix destruction, but two of them (no. 23 and 39) worked without. Lab 39 had separated and discarded the milk protein by hydrochloric and silicic acid.

The chemical recovery of the whole process of sample preparation and radiochemical separation was evaluated mainly with gravimetric methods (16 times). The use of ^{85}Sr tracer, titration and atomic absorption spectrometry of natural Sr or Y or added carrier were each named about 6 to 8 times. Furthermore, about one lab each carried out ICP-MS and ICP-OES measurement of Sr or relied on a total method validation with spiked milk or on the validity of a standard method as such. The values reported for the radiochemical yield of strontium or yttrium, respectively, spanned a wide range between 22 and 105 %. The median value was 79 %.

An attempt is made in Table 3 to sort the reported radiochemical yields according to the separation methods used. A clear distinction of yield between methods, however, can not be deduced. The radiochemical yield appears to be primarily dependent on how exactly to every small detail the separations are performed in a particular laboratory.

The number of independent samples treated for ^{90}Sr determination varied from 1 to 10 while 25 laboratories (more than 50 %) performed two independent analyses. The mass of milk powder consumed for each analysis cannot be unambiguously deduced from the answers given in the questionnaire, because some pertain to the milk powder, others apparently to the ashed material after incineration.

In the vast majority of laboratories, namely in 24, the measurements were performed with gas flow proportional counters from at least eight different suppliers. At least three laboratories (no. 11, 38 and 56) measured with low-level Geiger-Müller counters made by Risø National Laboratory. Seven laboratories used liquid scintillation counting on equipment by Wallac or Packard, applying liquid scintillation cocktails such as UltimaGold (labs 4, 18, 36, 51, 57) (produced by Perkin Elmer) or OptiPhase Hi-Safe (supplier Wallac) (labs 1 and 32). Cherenkov counting was carried out by another eight laboratories (no. 15, 26, 31, 34, 40, 41, 49 and 55) making use of liquid scintillation counters from Wallac or Packard. Interesting to note is the use of plastic scintillators for gross beta counting in three laboratories (no. 28, 30 and 50).

The measurement systems were normally calibrated using standard solutions of ^{90}Sr in equilibrium with ^{90}Y or after separation of ^{90}Y. The accumulated measurement acquisition time per unknown sample ranged from 500 to 300000 s distributed over 1 to 24 acquisition cycles. In many cases between 3 and 5 cycles were applied. The data analysis was done with the software of the instrument supplier (20 laboratories), custom made software or manually (25 laboratories).

A literature review of determination methods for radiostrontium in milk was recently published by Brun et al. [13].

Table 3: Radiochemical yields reported for separation procedures of strontium or yttrium grouped according to separation methods

Separation method		Yield η	Lab codes
fuming nitric acid		(0.84 for Sr; 0.83 and 1.0 for Y); 0.80; (0.62 to 0.91 Sr; 0.59 to 0.74 Y); 0.95; (0.68 Sr; 0.97 Y); 0.64; 0.60; (0.55 to 0.86 Sr; 1.05 Y); (0.56 Sr; 0.92 Y); (0.75 to 0.85)	11; 45; 23; 21; 39; 42; 48; 30 (4 samples); 43; 10
oxalate method		0.94; 0.76; (0.85 Sr; 0.91 Y)	7; 44; 50
extraction chromatography on resins		0.92; (0.80 to 0.86); 0.60; 0.87; (0.55 to 0.60); 0.93; (0.62 and 0.86); 0.65; 0.90; 0.78; 0.85; 0.59	58; 1; 4; 5; 51; 57; 18; 20; 36; 41; 61; 32
solvent extraction	HDEHP	0.90; 0.62; (0.92 Sr; 0.88 Y); 0.57; 0.90; 0.97; 0.76; 0.99; 0.64; (0.63 Sr; 0.89 Y)	15; 26; 28; 31; 40; 55; 47; 63; 29; 30 (1 sample)
	TBP	0.31; 0.51; 0.80	56; 25; 46
	EDTA	0.96; 0.80	24; 16
other method		0.62	49
not identified		0.85; 0.96; 0.87; 0.87; 0.22	3; 9; 34; 38; 17

6. Reported results

In total, 59 sets of results were returned from the participating laboratories for ^{137}Cs and ^{40}K, but only 45 laboratories submitted results for ^{90}Sr. In case several measurements were performed with the same or different analysis or measurement scheme, it was possible to report up to 6 individual results per radionuclide in the reporting form. If more than one result was submitted the unweighted laboratory mean A_{lab} of the reported values was calculated by us and taken into account in the evaluations below (starting with Fig. 8).

Figures 5 to 7 show the individual activity concentrations (normalised to dry mass and decay-corrected to the reference date 1 January 2005) and expanded uncertainties (with a coverage factor k=2) as they were reported by the participants. If a coverage factor different from 2 was reported, we recalculated the expanded uncertainty for k=2. The solid red lines indicate the reference activity concentration and the dashed lines the expanded uncertainty ± U_{ref} (k=2) of the reference value. As seen from Figs. 5 and 6 the results obtained for ^{137}Cs and ^{40}K are distributed (with one exception for ^{137}Cs and several for ^{40}K) in a narrow band around the reference value. One laboratory (no. 35) reported far too high values for ^{137}Cs as well as ^{40}K.

In the case of ^{90}Sr the distribution of the results was not as uniform as for the other two radionuclides measured (Fig. 7). A large number of laboratories reported results deviating from the reference value by more than the sum of its expanded uncertainty U_{ref} and the expanded uncertainty of the individual measurement results. Laboratory 26 with far too low results is one of the laboratories that encountered problems with

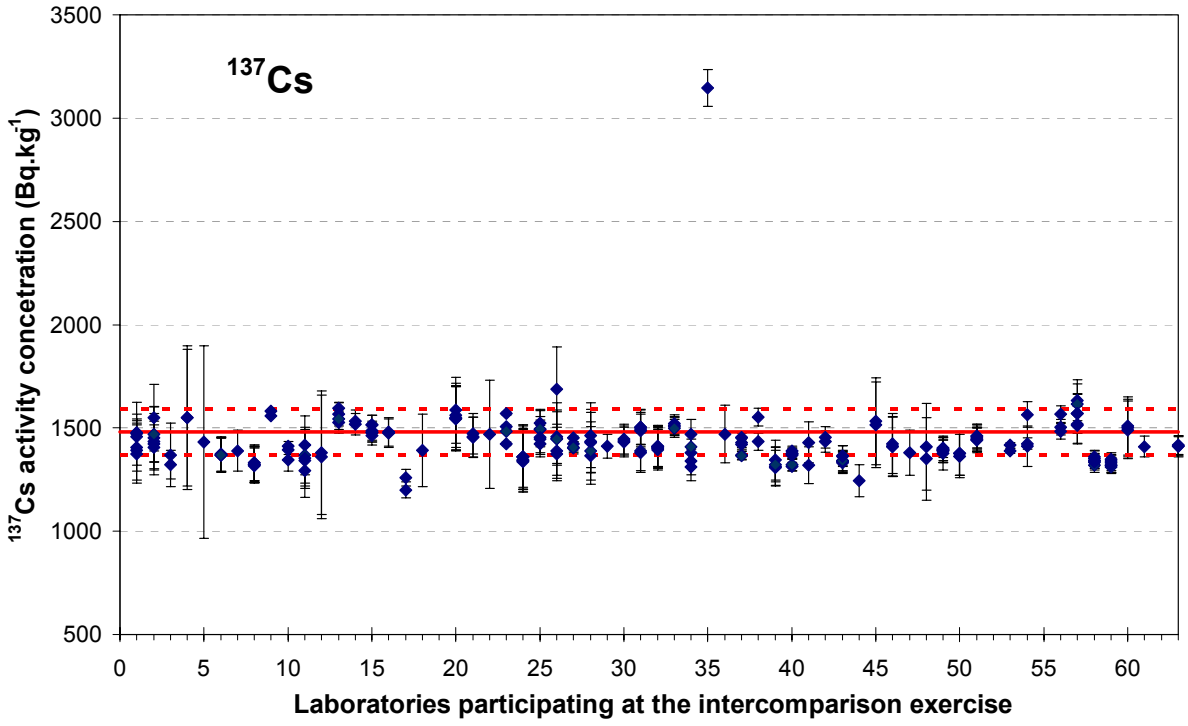

Fig. 5: Measured ^{137}Cs activity concentration (normalised to dry mass). Error bars represent expanded uncertainty (with k=2) of individual measurements

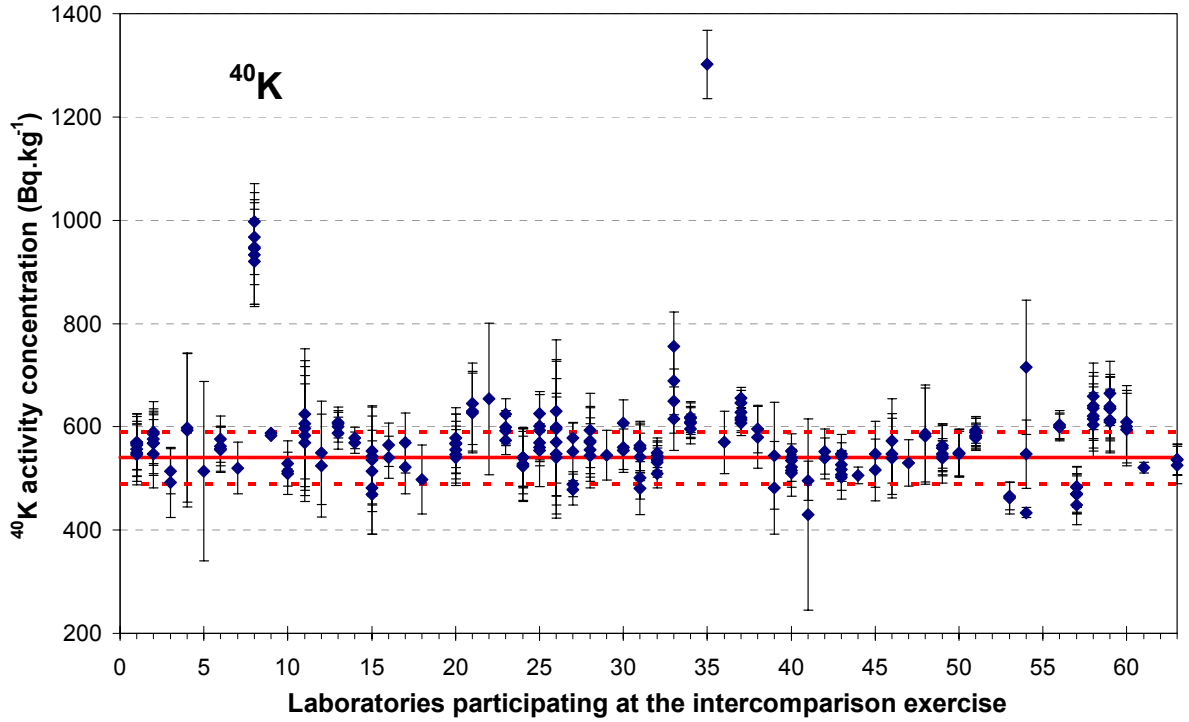

Fig. 6: Measured ^{40}K activity concentration (normalised to dry mass). Error bars represent expanded uncertainty (with *k*=2) of individual measurements

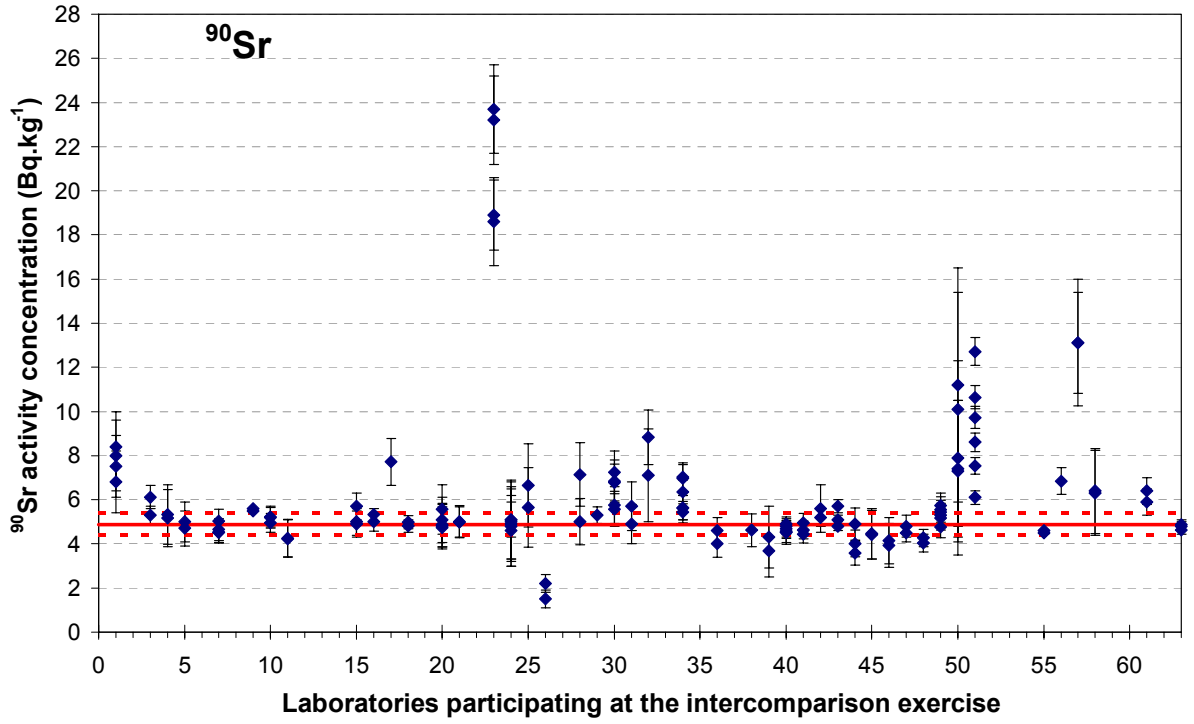

Fig. 7: Measured ^{90}Sr activity concentration (normalised to dry mass). Error bars represent expanded uncertainty (with *k*=2) of individual measurements

the reconstitution of milk from the powder, and for that reason asked to withdraw the results, albeit after having been informed about a large deviation of its results.

The estimation of the expanded uncertainty U_{lab} of the mean laboratory result was based on the information given by the participants in the result reporting form (Annex 4) and in the questionnaire (Annex 5). Laboratories were requested to give the individual expanded uncertainty (and the coverage factor applied) together with the measurement results in the reporting form, and to provide the full uncertainty budget for one measurement of each nuclide in the questionnaire. A non-exhaustive list of uncertainty contributions served as budget template for information. Moreover, participants were invited to send their complete uncertainty budget in a free format of their own choice.

The submitted uncertainty budgets were analysed and compared with the numerical values reported with the measurement results. For gamma-ray spectrometry, every two of three laboratories reported uncertainty values which were consistent with the given budget. For 30 % of the gamma-spectrometric results, discrepancies between the given uncertainty budget and the reported uncertainty values were observed, or no budget was given at all (10 laboratories). In the case of ^{90}Sr determination about half of the concerned laboratories provided consistent uncertainty budgets and uncertainty values with the measurement results. Admittedly, the budget template provided by IRMM in the questionnaire was not of great help to the laboratories for estimating correctly the uncertainty for ^{90}Sr determination.

When using the submitted uncertainty budgets to estimate the expanded uncertainties U_{lab} of the laboratory mean values, two cases had to be distinguished. If the uncertainties reported with the individual results were found to be consistent with the budget, then the expanded uncertainty U_{lab} (with k=2) of the mean activity concentration A_{lab} was calculated based on the information given in the uncertainty budget (approach 1). The following formula was applied:

$$U_{lab} = k \cdot \sqrt{\frac{u_{count}^2}{n} + u_{other}^2} \qquad (1)$$

where

u_{count} the counting uncertainty as given in the uncertainty budget,
n number of measurements,
u_{other} calculated from the reported uncertainty budget.

It is obvious from Eq. (1) that only the counting uncertainty was considered as a random component (type A uncertainty according to the GUM [3]) and, consequently, is divided by $\sqrt{n}$ in the combined uncertainty.

In the case of ^{137}Cs and ^{40}K u_{other} is determined from:

$$u_{other} = \sqrt{u_{moisture}^2 + u_{sample.prep}^2 + u_{act.stardards}^2 + u_{geometry}^2 + u_{other.contrib}^2} \qquad (2)$$

where u_{index} denotes the *propagated* uncertainty contribution to the combined standard uncertainty of A_{lab}, in detail:

$u_{moisture}$ propagated uncertainty due to uncertainty in moisture content, mass etc.;
$u_{sample\ prep}$ propagated uncertainty due to uncertainty in sample preparation;
$u_{act.\ standards}$ propagated uncertainty due to uncertainty in the activity of standards;

$u_{geometry}$ propagated efficiency uncertainty due to uncertainty in geometry, sample density etc.;

$u_{other\ contrib}$ other propagated uncertainty contributions.

Equation (2) may lead to an overestimation of u_{other} and U_{lab} in cases where a propagated contribution, e.g. $u_{sample\ prep}$, represents a random component if this is not taken care of by dividing by $\sqrt{n}$.

In the case of ^{90}Sr the propagated contribution of the uncertainty due to blank and background measurement (u_{blank}) and the uncertainty in efficiency of the counter ($u_{efficiency}$) were added, whereas the geometry effect is not applicable. While $u_{sample\ prep}$ is now also including components due to separation and chemical recovery, the applied formula for u_{other} reads:

$$u_{other} = \sqrt{u^2_{moisture} + u^2_{sample.prep} + u^2_{act.stardards} + u^2_{blank} + u^2_{efficiency} + u^2_{other.contrib}} \qquad (3)$$

When discrepancies between the individual uncertainty values and the uncertainty budget were observed (or no budget at all was presented), a second approach was used. Then U_{lab} was calculated by us as an arithmetic mean (if n>1) of the reported individual expanded uncertainties (setting the coverage factor, if not given like that, to k=2).

Tables 4 to 6 give in detail for each participating laboratory the number of measurements n, the unweighted laboratory mean A_{lab} of the reported values and its standard deviation s. Furthermore, the expanded uncertainty U_{lab} and its relative value U_{lab}/A_{lab} are given. The uncertainty values U_{lab} printed in bold were calculated by us based on the reported uncertainty budget (approach 1 above). Normally printed values of U_{lab} were generated using approach 2.

For laboratory 2, a slight variation in equation (2) was applied: Since two different gamma-ray detectors were used, it is reasonable to reduce the uncertainty component due to geometry effects by $\sqrt{2}$. This could also apply to other laboratories, but the effect would be negligible since the geometry effect is estimated by these labs as very small from the outset. Laboratory 20 did not individually estimate the uncertainty of its measurement results, it based its uncertainty statement on a global assessment: $U = 2 \cdot S_R$ where S_R is the accuracy of the method, apparently obtained from an external validation of the method.

In Figures 8 to 10 the mean activity concentration A_{lab} with its corresponding expanded uncertainty U_{lab} (k=2) obtained by one of the approaches described above are plotted in ascending order. Again, the solid line indicates the reference activity concentration A_{ref}, and its expanded uncertainty ± U_{ref} (k=2) is plotted with dashed lines. Laboratory numbers are indicated with the results.

Apart from clearly wrong results (lab 35 for ^{137}Cs, labs 35 and 8 for ^{40}K and several for ^{90}Sr), it is obvious from Figs. 8 to 10 that several laboratories underestimate their uncertainties for one or more radionuclides (labs 14, 17, 23, 27, 29, 37, 48, 53, 55, 58, 59 and 61). Interesting to note, however, there are also several laboratories apparently overestimating the measurement uncertainty considerably (labs 4, 5, 22 and 48 for both gamma-ray emitters, labs 11, 26 and 41 for ^{40}K and labs 12, 38 and 45 for ^{137}Cs).

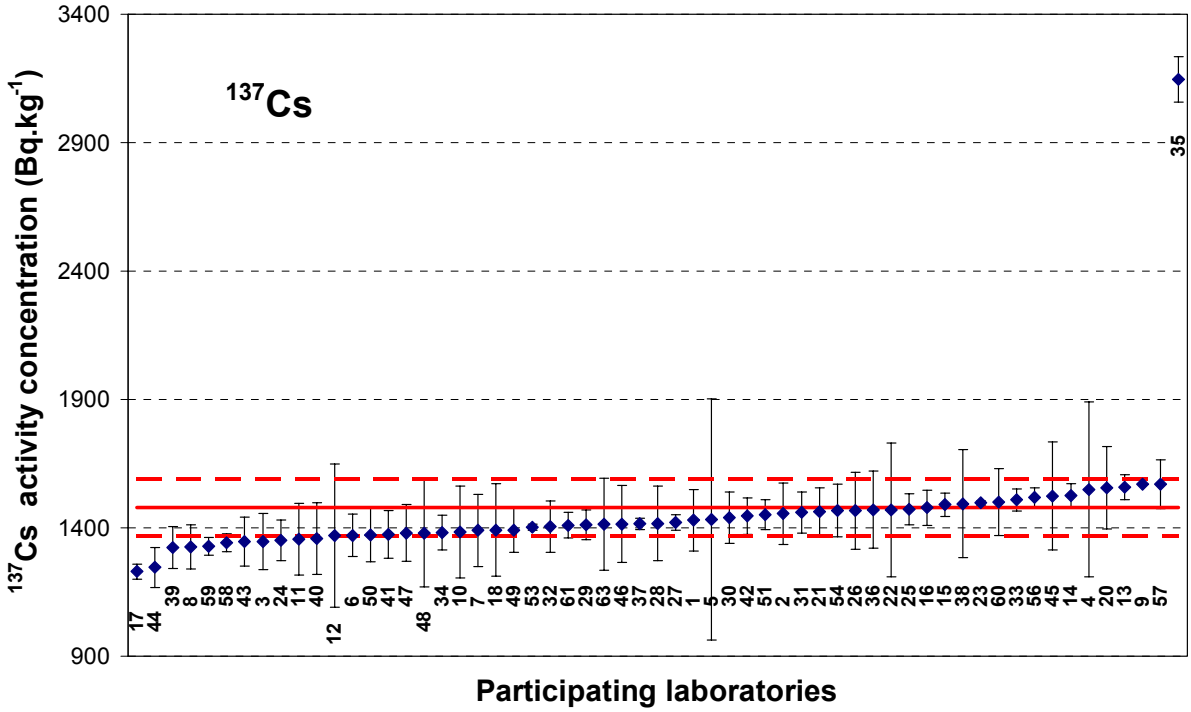

Fig. 8: Mean laboratory results A_{lab} for ^{137}Cs activity concentration (normalised to dry mass). Error bars indicate expanded uncertainty U_{lab} (k=2) of laboratory mean, red lines are reference value A_{ref} ± expanded uncertainty U_{ref} (k=2). Laboratory numbers are indicated

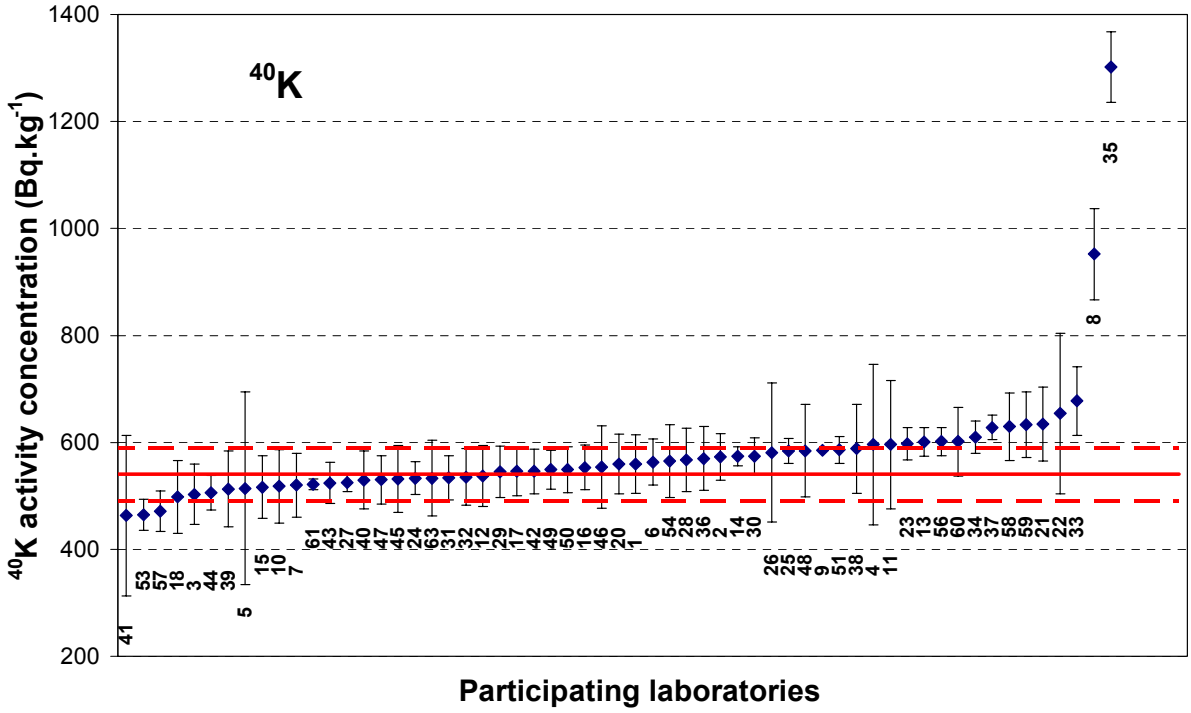

Fig. 9: Mean laboratory results A_{lab} for ^{40}K activity concentration (normalised to dry mass). Error bars indicate U_{lab} (k=2) of laboratory mean, red lines are reference value A_{ref} ± U_{ref} (k=2). Laboratory numbers are indicated

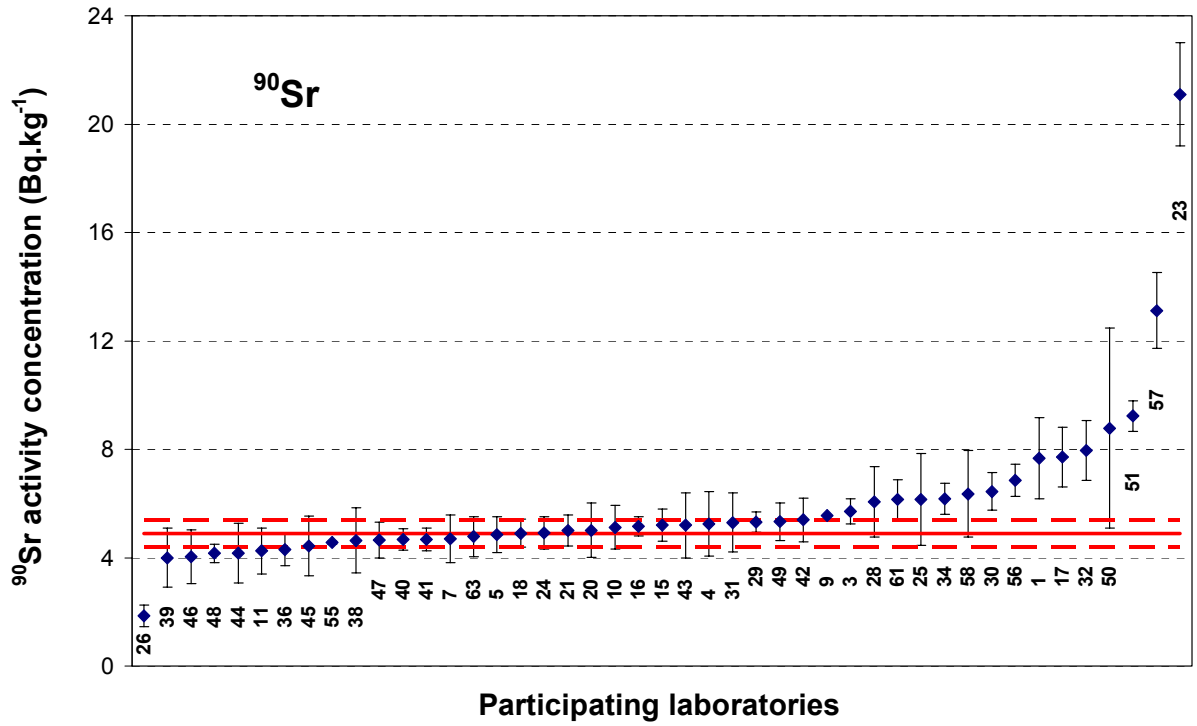

Fig. 10: Mean laboratory results A_{lab} for ^{90}Sr activity concentration (normalised to dry mass). Error bars indicate U_{lab} (k=2) of laboratory mean, red lines are reference value $A_{ref} \pm U_{ref}$ (k=2). Laboratory numbers are indicated

For ^{90}Sr an attempt is made to distinguish results obtained with the different methods of separation and measurement. Fig. 11 shows the results plotted in groups of counting methods. It is obvious that in each group there are outlying values but almost always also laboratories which manage to obtain results within the uncertainty limits of the reference value. The group of laboratories using liquid scintillation counting (LSC) renders the relatively largest proportion of deviating results. This result may be taken as a warning that the correct application of LSC is far from trivial, although – it must be said – three laboratories obtained results with only small or no deviation from the reference value, which itself was established using LSC. The results of this comparison, however, do not justify earmarking a particular counting method as being superior or inferior to others. Maybe with one exception: Although the three laboratories applying plastic scintillators for gross beta counting (no. 28, 30 and 50) are too small a group to allow reliable conclusions to be drawn, a critical review of the implementation of this counting method should be done by them.

Although difficult due to the variety and complex nature of separation methods, an attempt is made to group the results by separation procedures (Fig. 12). The most important conclusion is similar to that for counting methods: For each separation procedure there is a reasonably large number of laboratories which succeed to obtain results close to the reference value, even though some labs in most groups produce largely deviating results. Again, no particular separation procedure can be identified as the source for deviating results, but the reason must be sought in the individual laboratories concerned. Laboratory no. 30 occurs twice, since it analysed four samples with the fuming nitric acid method and one with HDEHP. Only a small number of results could not be attributed to a particular separation method (“not identified”).

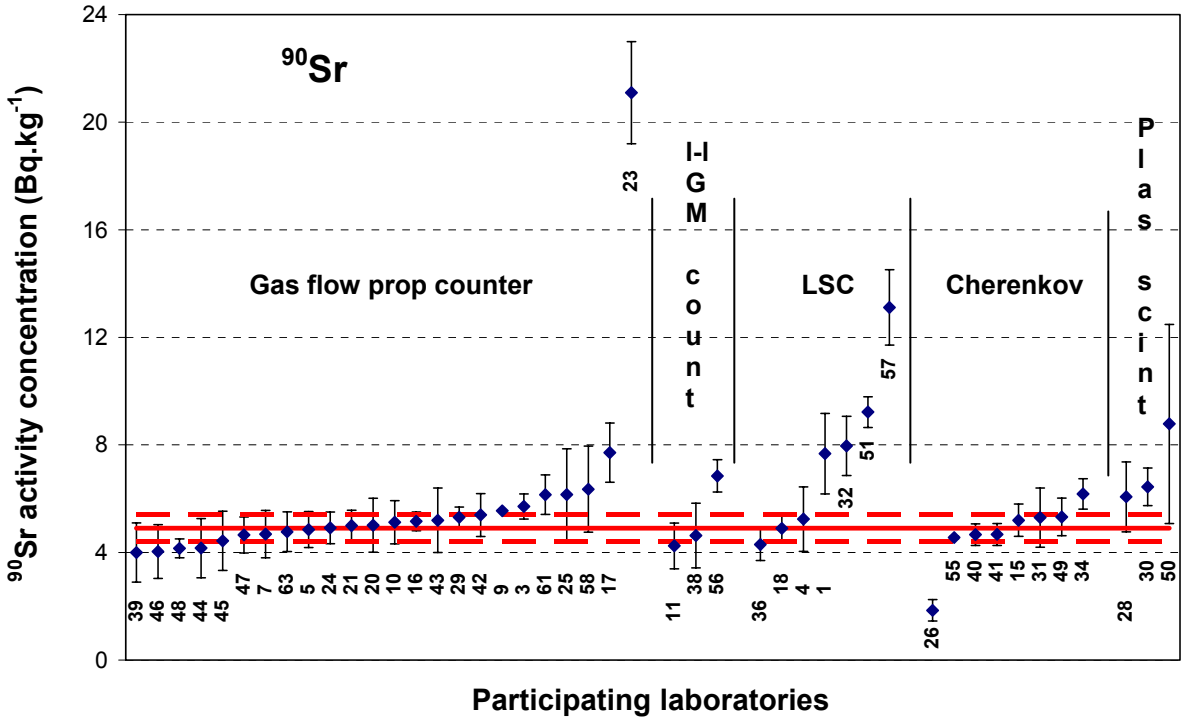

Fig. 11: Same as Fig. 10 but mean laboratory results A_{lab} for ^{90}Sr activity concentration sorted according to counting method. Error bars indicate U_{lab} (k=2) of laboratory mean, red lines are reference value $A_{ref} \pm U_{ref}$ (k=2). Laboratory numbers are indicated

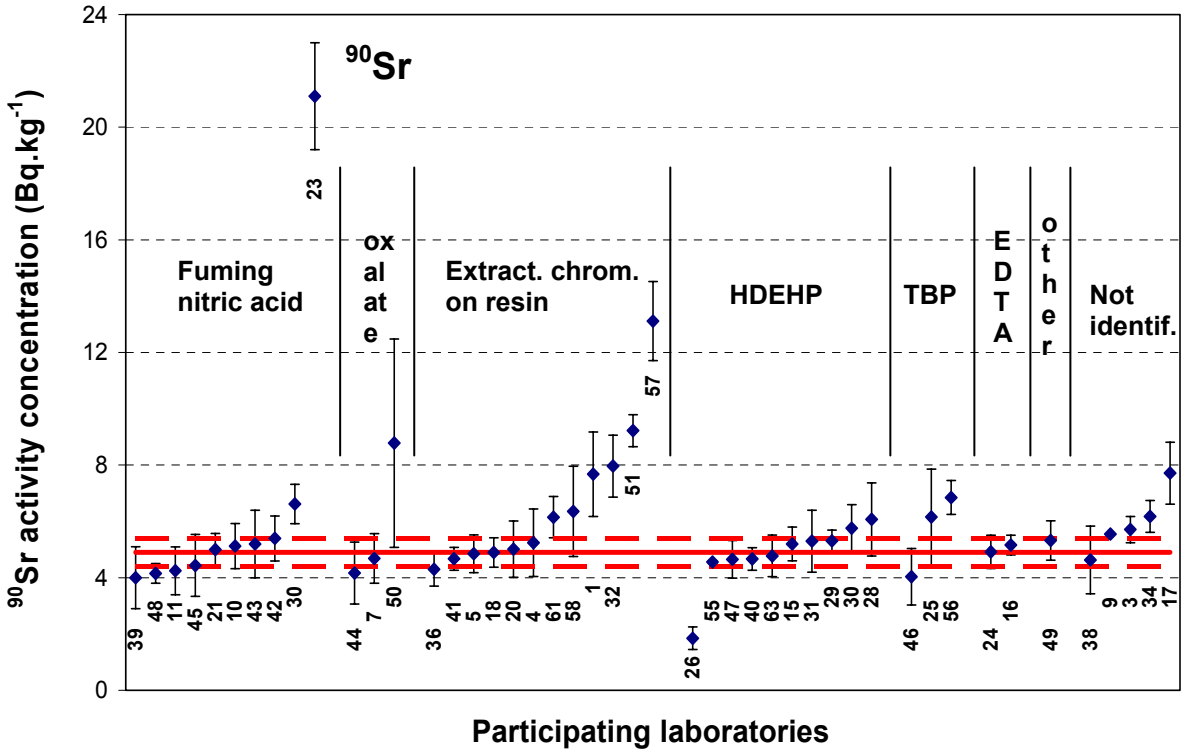

Fig. 12: Same as Fig. 10 but mean laboratory results A_{lab} for ^{90}Sr activity concentration sorted according to separation method

Table 4a: Mean ^{137}Cs activity concentrations (normalised to dry mass), their standard deviations and estimated expanded uncertainties

lab no.	*n*	mean activity concentration A_{lab} / (Bq·kg^{-1})	standard deviation *s* / (Bq·kg^{-1})	expanded uncertainty U_{lab} / (Bq·kg^{-1})	rel. expanded uncertainty U_{lab}/A_{lab} / (%)
1	6	1430	44	120	9
2	6	1456	51	**120**	8
3	2	1347	33	110	8
4	2	1550	-	340	22
5	1	1432	-	**470**	33
6	4	1370	3	83	6
7	1	1390	-	**140**	10
8	6	1325	5	86	7
9	2	1569	17	-	-
10	3	1384	35	**180**	13
11	5	1355	45	140	10
12	2	1370	14	**280**	20
13	4	1559	30	**49**	3
14	2	1526	11	**47**	3
15	6	1490	21	46	3
16	2	1478	4	69	5
17	2	1230	42	**29**	2
18	1	1391	-	**180**	13
19	-	-	-	-	-
20	6	1556	16	160	10
21	3	1462	10	**93**	6
22	1	1470	-	260	18
23	4	1497	60	9	1
24	6	1351	10	**79**	6
25	6	1473	38	**61**	4
26	6	1467	113	150	10
27	4	1422	23	30	2
28	5	1417	39	**145**	10
29	1	1411	-	58	4
30	3	1439	7	**100**	7
31	6	1460	58	**81**	6
32	6	1405	6	**100**	7
33	4	1509	10	43	3
34	5	1382	62	68	5
35	1	3146	-	88	3
36	1	1470	-	**150**	10
37	6	1415	40	22	2
38	2	1494	84	**210**	14
39	4	1323	16	**81**	6
40	6	1358	32	**140**	10

Table 4b: Mean ^{137}Cs activity concentrations (normalised to dry mass), their standard deviations and estimated expanded uncertainties (continued)

lab no.	*n*	mean activity concentration A_{lab} / (Bq·kg^{-1})	standard deviation *s* / (Bq·kg^{-1})	expanded uncertainty U_{lab} / (Bq·kg^{-1})	rel. expanded uncertainty U_{lab}/A_{lab} / (%)
41	2	1375	78	**93**	7
42	2	1446	15	**70**	5
43	6	1346	15	**95**	7
44	1	1246	-	**78**	6
45	2	1524	13	**210**	14
46	3	1415	8	**150**	10
47	1	1380	-	110	8
48	2	1380	42	**210**	15
49	6	1392	9	**87**	6
50	3	1373	8	**105**	8
51	6	1450	9	58	4
52	-	-	-	-	-
53	2	1403	20	11	1
54	3	1467	85	**103**	7
55	-	-	-	-	-
56	3	1518	44	38	3
57	6	1570,4	49	95	6
58	6	1341	14	35	3
59	6	1328	14	34	3
60	3	1500	10	**130**	9
61	1	1410	-	50	4
62	-	-	-	-	-
63	3	1414	4	**180**	13

Table 5a: Mean ^{40}K activity concentrations (normalised to dry mass), their standard deviations and estimated expanded uncertainties

lab no.	*n*	mean activity concentration A_{lab} / (Bq·kg^{-1})	standard deviation *s* / (Bq·kg^{-1})	expanded uncertainty U_{lab} / (Bq·kg^{-1})	rel. expanded uncertainty U_{lab}/A_{lab} / (%)
1	6	560	10	55	10
2	6	573	15	**44**	8
3	2	503	16	56	11
4	2	596	3	150	25
5	1	514	-	**180**	34
6	4	563	9	43	8
7	1	520	-	**60**	12
8	6	952	27	85	9
9	2	585	3	-	-
10	3	518	10	**69**	13
11	5	596	21	120	20
12	2	538	18	**57**	11
13	4	601	9	**27**	4
14	2	574	6	**18**	3
15	6	516	34	**59**	11
16	2	553	17	42	8
17	2	546	33	**45**	8
18	1	498	-	**68**	14
19	-	-	-	-	-
20	6	559	15	56	10
21	3	634	10	**69**	11
22	1	654	-	150	22
23	4	598	21	30	5
24	6	533	8	**31**	6
25	6	584	28	**23**	4
26	6	581	34	130	22
27	4	525	49	**16**	3
28	5	567	19	**59**	10
29	1	545	-	48	9
30	3	574	29	**35**	6
31	6	534	35	**41**	8
32	6	535	14	**53**	10
33	4	678	60	64	9
34	5	610	9	30	5
35	1	1302	-	66	5
36	1	570	-	**60**	11
37	6	628	19	23	4
38	2	588	11	**83**	14
39	2	513	44	**71**	14
40	6	530	16	**54**	10

Table 5b: Mean ^{40}K activity concentrations (normalised to dry mass), their standard deviations and estimated expanded uncertainties (continued)

lab no.	*n*	mean activity concentration A_{lab} / (Bq·kg^{-1})	standard deviation *s* / (Bq·kg^{-1})	expanded uncertainty U_{lab} / (Bq·kg^{-1})	rel. expanded uncertainty U_{lab}/A_{lab} / (%)
41	2	463	47	**150**	31
42	2	546	9	**42**	8
43	6	524	18	**39**	7
44	1	506	-	**32**	6
45	2	532	22	**62**	12
46	3	553	18	**77**	14
47	1	530	-	45	8
48	2	585	4	**87**	15
49	6	549	10	**36**	7
50	3	549	1	**43**	8
51	6	586	6	25	4
52	-	-	-	-	-
53	2	464	2	29	6
54	3	565	141	**68**	12
55	-	-	-	-	-
56	3	601	3	26	4
57	6	471	13	38	8
58	6	629	20	63	10
59	6	633	20	61	10
60	3	602	8	**64**	11
61	1	521	-	10	2
62	-	-	-	-	-
63	3	533	6	**71**	13

Table 6a: Mean ^{90}Sr activity concentrations (normalised to dry mass), their standard deviations and estimated expanded uncertainties

lab no.	*n*	mean activity concentration A_{lab} / (Bq·kg^{-1})	standard deviation *s* / (Bq·kg^{-1})	expanded uncertainty U_{lab} / (Bq·kg^{-1})	rel. expanded uncertainty U_{lab}/A_{lab} / (%)
1	4	7.68	0.69	1.5	20
2	-	-	-	-	-
3	2	5.71	0.58	0.47	8
4	2	5.25	0.12	**1.2**	22
5	2	4.85	0.21	**0.67**	14
6	-	-	-	-	-
7	4	4.69	0.24	**0.88**	19
8	-	-	-	-	-
9	2	5.55	0.07	-	-
10	3	5.12	0.15	**0.80**	16
11	2	4.25	0.02	0.85	20
12	-	-	-	-	-
13	-	-	-	-	-
14	-	-	-	-	-
15	3	5.20	0.44	0.60	12
16	2	5.16	0.23	0.35	7
17	1	7.71	-	**1.1**	14
18	2	4.90	0.12	**0.52**	11
19	-	-	-	-	-
20	5	5.01	0.33	1.0	20
21	2	5.00	0.03	**0.57**	11
22	-	-	-	-	-
23	4	21.10	2.72	1.9	9
24	6	4.92	0.19	**0.59**	12
25	2	6.16	0.70	**1.7**	28
26	2	1.85	0.49	0.40	22
27	-	-	-	-	-
28	2	6.07	1.51	**1.3**	21
29	1	5.31	-	0.38	7
30	5	6.44	0.73	**0.70**	11
31	2	5.30	0.57	**1.1**	21
32	2	7.97	1.22	**1.1**	14
33	-	-	-	-	-
34	6	6.18	0.71	0.57	9
35	-	-	-	-	-
36	2	4.30	0.42	0.60	14
37	-	-	-	-	-
38	1	4.63	-	**1.2**	26
39	2	4.00	0.42	**1.1**	27
40	6	4.67	0.15	0.40	9

Table 6b: Mean ^{90}Sr activity concentrations (normalised to dry mass), their standard deviations and estimated expanded uncertainties (continued)

lab no.	*n*	mean activity concentration A_{lab} / (Bq·kg^{-1})	standard deviation *s* / (Bq·kg^{-1})	expanded uncertainty U_{lab} / (Bq·kg^{-1})	rel. expanded uncertainty U_{lab}/A_{lab} / (%)
41	3	4.67	0.27	**0.41**	9
42	2	5.40	0.28	0.8	14
43	3	5.20	0.46	**1.2**	22
44	3	4.16	0.67	**1.1**	26
45	2	4.44	0.04	**1.1**	24
46	2	4.04	0.15	**1.0**	25
47	2	4.65	0.21	**0.67**	14
48	2	4.16	0.16	**0.35**	8
49	6	5.32	0.34	**0.70**	13
50	5	8.78	1.77	**3.7**	43
51	6	9.22	2.34	**0.57**	6
52	-	-	-	-	-
53	-	-	-	-	-
54	-	-	-	-	-
55	2	4.56	0.71	0.05	1
56	1	6.85	-	0.60	9
57	2	13.12	0.01	**1.4**	11
58	2	6.36	0.06	**1.6**	24
59	-	-	-	-	-
60	-	-	-	-	-
61	2	6.15	0.35	**0.73**	12
62	-	-	-	-	-
63	3	4.77	0.12	**0.74**	16

Twenty-nine laboratories also reported results for the ^{134}Cs activity concentration, a nuclide for which no reference value was determined in the frame of this intercomparison. The reported results incl. their expanded uncertainties are plotted in Fig. 13. For laboratory no. 13 and 25 the mean values of their four and six individual measurements, respectively, are shown. In view of only incomplete information requested in the questionnaire, the plotted uncertainty for these two values is the mean of the expanded uncertainties stated for the individual measurements.

The overall mean and standard deviation *s* of the 29 measurement results for ^{134}Cs is (2.2 ± 0.4) Bq·kg^{-1} indicated in Fig. 13 as solid and dashed blue lines.

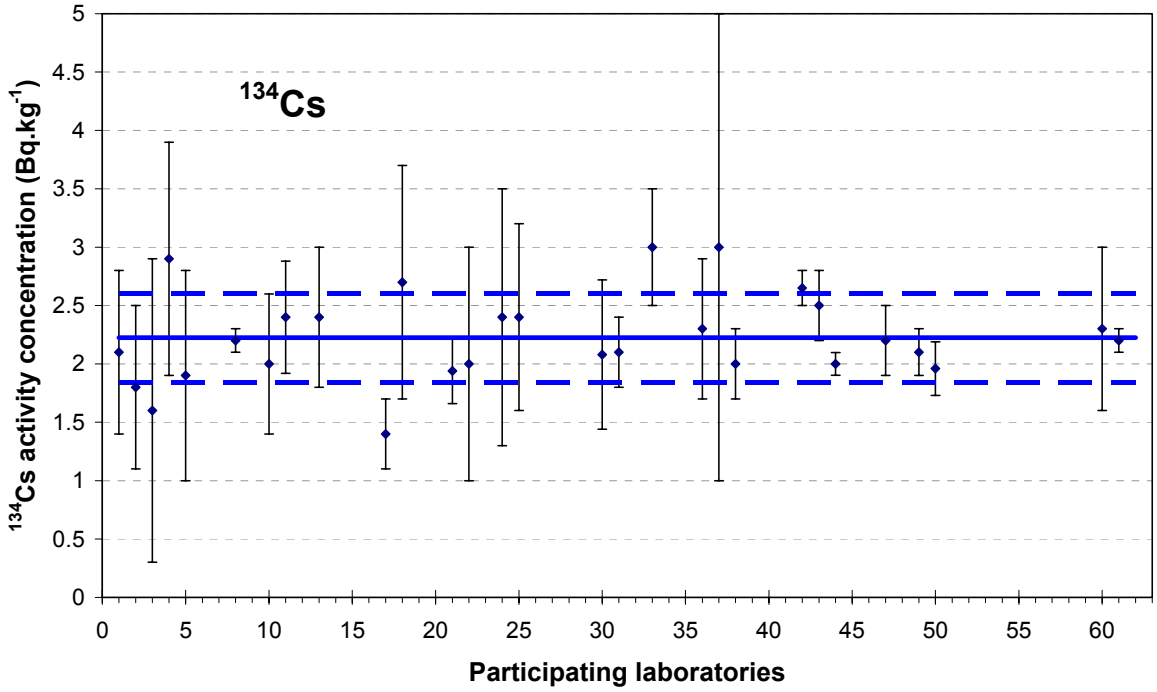

Fig. 13: ^{134}Cs activity concentration determined additionally by some laboratories. Error bars represent expanded uncertainty (*k*=2) of individual measurements, mean value and standard deviation *s* are indicated in blue

7. Data evaluation and comparison of data

In order to compare the results, a modern type of plot, PomPlot, that underlines the importance of the assigned uncertainties, is applied. The 'PomPlot', an intuitive graphical method, is used here for producing a summary overview of the participants' results [14]. It displays relative deviations, *D/MAD*, of the mean results A_{lab} from the reference value on the horizontal axis and relative uncertainties, *u/MAD*, on the vertical axis. For both axes, the variables are expressed as multiples of *MAD*, which is defined as the median absolute deviation from the reference value:

$$MAD = \text{Median } |D_i|, \ (i=1,\ldots,n), \tag{4}$$

where D_i is the difference between the reported and the reference activity concentration:

$$D_i = A_{lab,i} - A_{ref} \tag{5}$$

The median absolute deviation *MAD* is used because of its robustness.

For every data point the uncertainty was calculated as independent sum of the reported combined uncertainties on $A_{lab,i}$ and A_{ref}:

$$u_i^2 = u_c^2\left(A_{lab,i}\right) + u_c^2\left(A_{ref}\right) \tag{6}$$

where $u_c(A_{lab,i}) = U_{lab,i} / k$ and $u_c(A_{ref}) = U_{ref} / k$.

The ζ-scores, $|\zeta| = |D/u| = 1$, 2 and 3, are represented by diagonal solid lines, creating the aspect of a pyramidal structure. Dots on the right-hand side of the graph correspond to results that are higher than the reference value whereas lower values are situated on the left. When the claimed uncertainty is low, the corresponding point is situated high in the graph. The most accurate results should be situated close to the top of the pyramid. Points outside of the $\zeta=\pm3$ lines are probably inconsistent with the reference value.

Figure 14 shows the PomPlot for the ^{137}Cs results. For laboratories not providing an uncertainty value, u_i was artificially set equal to zero. The reference value is indicated by a horizontal red dash. Outliers are indicated with an arrow.

Two features of the reference value are immediately apparent from the plot:
1) the reference value of ^{137}Cs is higher than the mean of the participants' results;
2) its relative uncertainty is not insignificant in the total uncertainty *u* (hence, the dots cannot reach the top).
There is no proportionality between the stated uncertainty $u_c(A_{lab})$ and the experimental deviation *D*. Moreover there seems to be a slight anti-correlation.

This is studied in more detail in Figure 15. Here the data (excluding lab 35) are grouped according to the *u/MAD*-value and for each group the mean values of the normalised deviation $|D|/MAD$ is calculated, as well as the corresponding ratio of $|D|/u$. In the group of laboratories claiming the smallest uncertainty ($0.7<u/MAD<0.8$), the mean deviation is among the highest of all results, hence the uncertainty clearly underestimated ($\langle|D|/u\rangle$ is higher than 1).

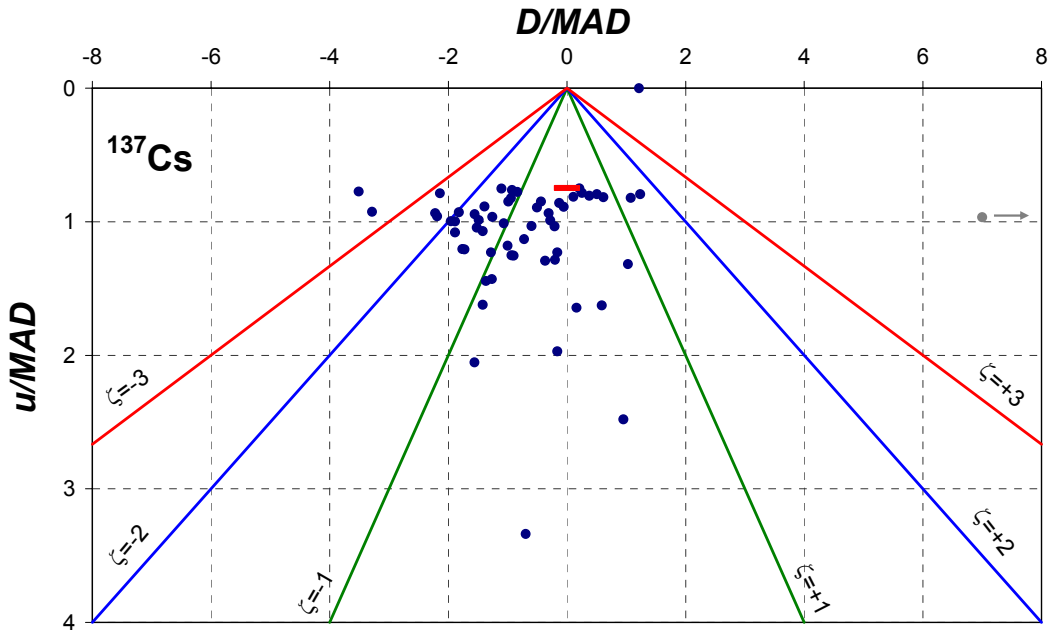

Fig. 14: PomPlot of the ^{137}Cs data

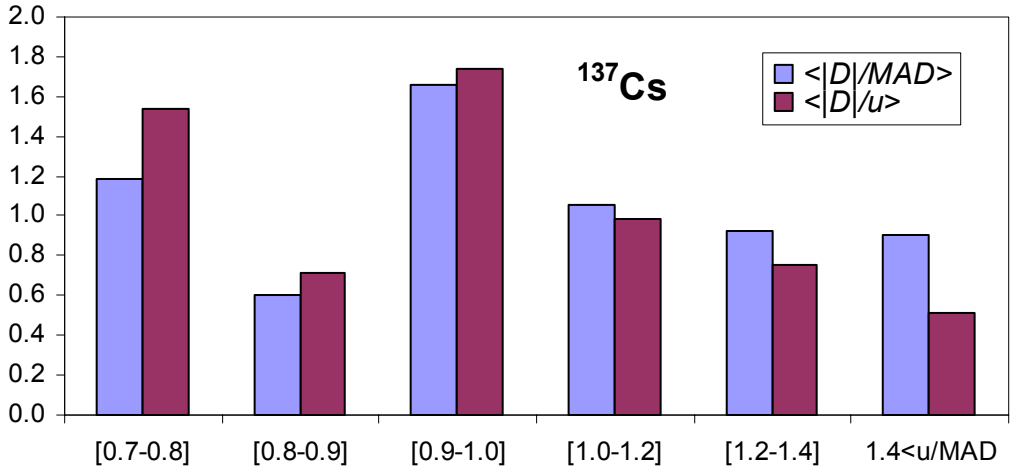

Fig. 15: Measures of average deviation <|*D*|/*MAD*>, <|*D*|/*u*> for different intervals of *u/MAD* in the case of ^{137}Cs

The best results concerning the activity measurement as well as uncertainty assessment can be found in the group with 0.8 ≤ *u/MAD* < 0.9. Some laboratories seem to assign conservative uncertainty values, even though their results are not worse than those of others. Indeed, nearly all results can be found between -2 < *D/MAD* < +2, irrespective of the claimed uncertainty.

Similar plots have been made for ^{40}K and ^{90}Sr (Figs. 16 - 19). Here the uncertainty on the reference value is relatively smaller, and corresponds well with the median of lab results. Yet, from a statistical point of view, there are too many points outside the |ζ|=1, 2 and 3 levels, which indicates that a significant fraction of the laboratories underestimates the uncertainties involved.

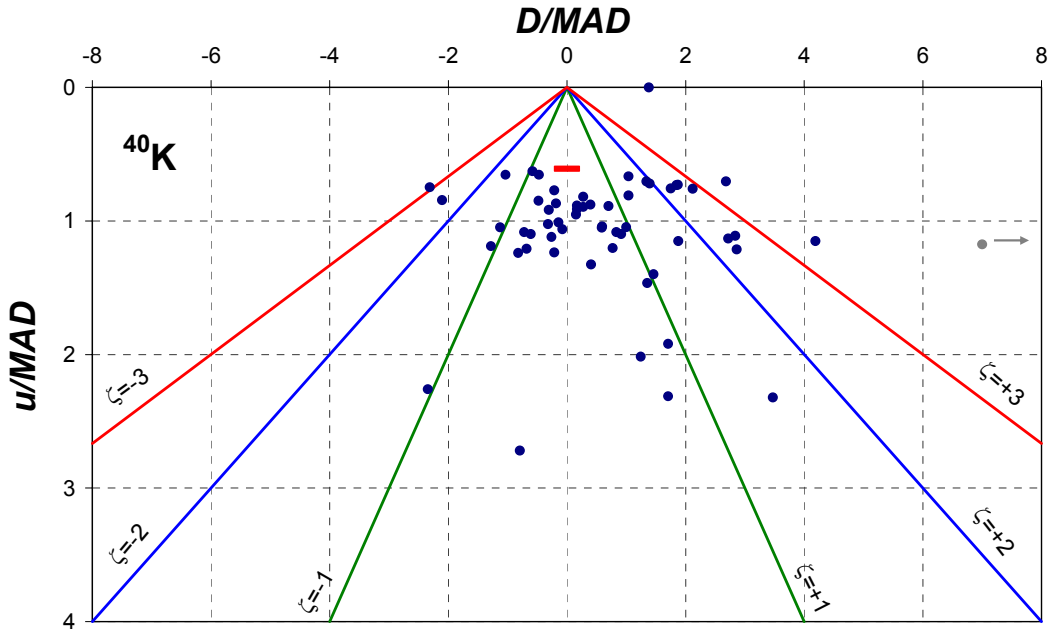

Fig. 16: PomPlot of the ^{40}K data

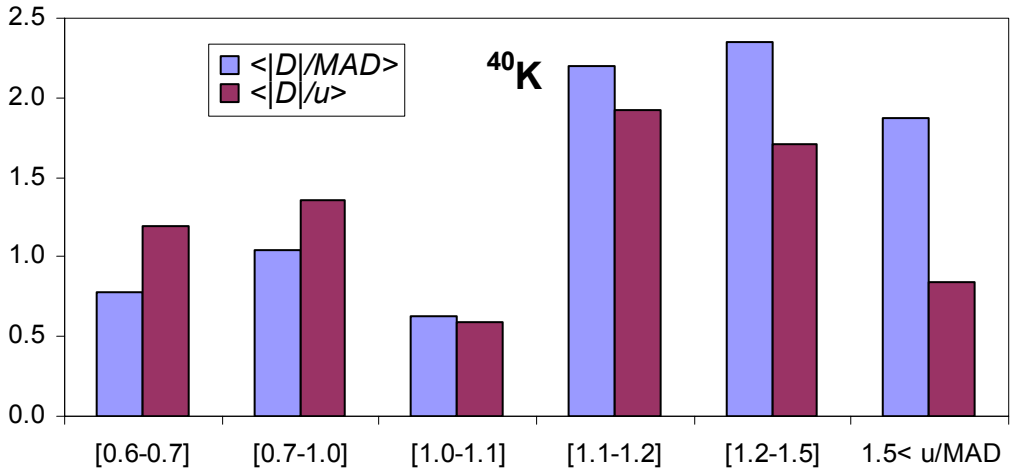

Fig. 17: Measures of average deviation <|*D*|/*MAD*>, <|*D*|/*u*> for different intervals of *u/MAD* in the case of ^{40}K

In particular, the ^{90}Sr results look problematic, as they show more outliers. Assuming that uncertainties would be assessed correctly by the laboratories, one should have an equal distribution of points above and below the *u/MAD* = 1 line. This is not the case for ^{90}Sr, showing that for many laboratories the determination process is not under statistical control. Again, when analyzing this in more detail in Figures 17 and 19, one finds the best and the most realistic results around 0.7 ≤ *u/MAD* < 1.1. It seems that there is a group of laboratories that manages to provide accurate results together with a comprehensive uncertainty assessment.

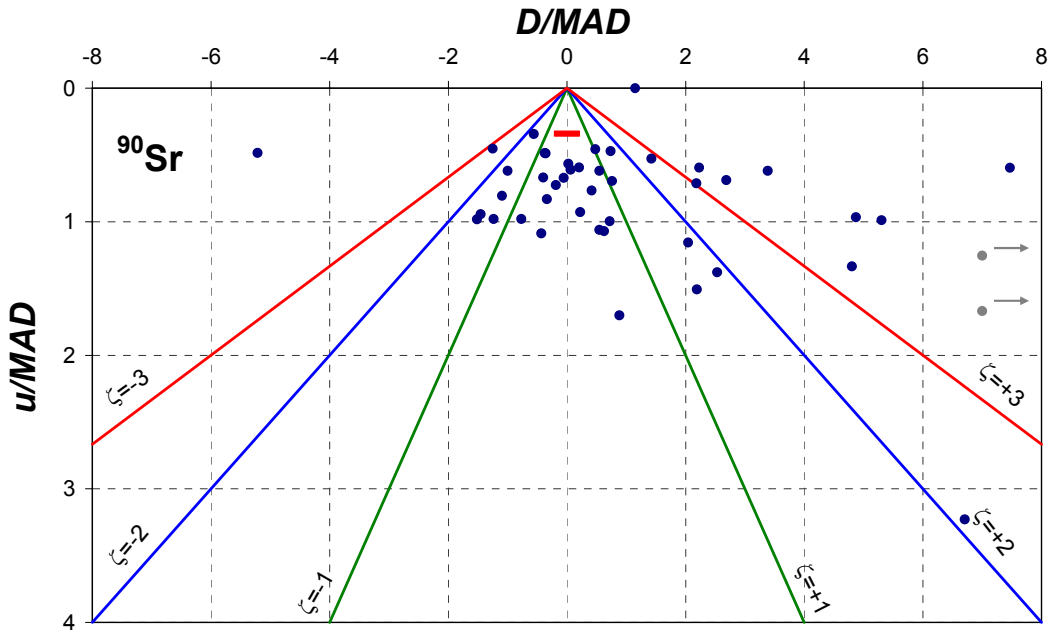

Fig. 18: PomPlot of the ^{90}Sr data

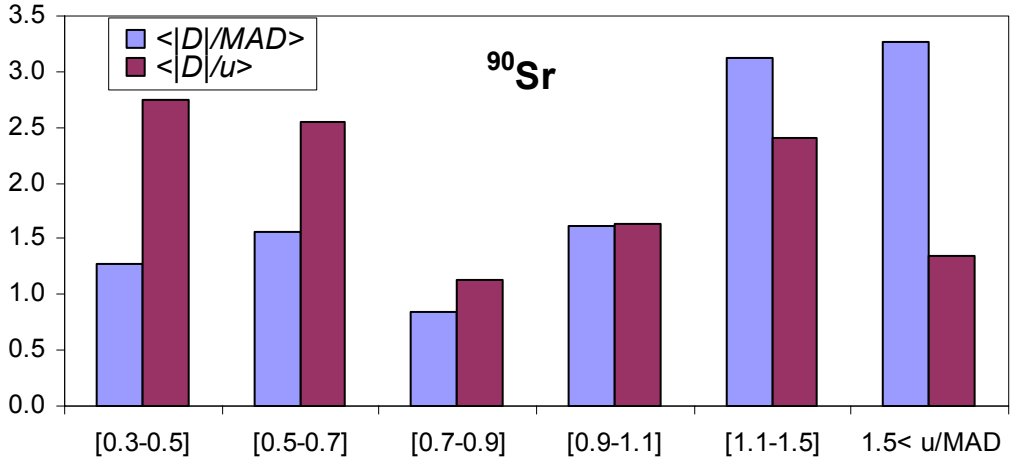

Fig. 19: Measures of average deviation <|*D*|/*MAD*>, <|*D*|/*u*> for different intervals of *u/MAD* in the case of ^{90}Sr

The presented plots suggest that there is no clear positive correlation between stated uncertainty and deviation of the result from the reference value. Yet, there appears to be a group of laboratories that succeeds at combining a good accuracy with a realistic uncertainty assessment. For some laboratories, there is still some work to be done in identifying and correcting errors in the activity measurement process and, beyond that, correctly quantifying the uncertainty components.

Table 7a: Relative deviation of mean laboratory results A_{lab} from reference value A_{ref} and compatibility test (E_n numbers) for ^{137}Cs

lab no.	mean activity concentration A_{lab} / (Bq·kg^{-1})	expanded uncertainty U_{lab} / (Bq·kg^{-1})	relative deviation D/A_{ref} / %	E_n	compatibility
1	1430	120	-3.4	-0.31	YES
2	1456	120	-1.7	-0.15	YES
3	1347	110	-9.0	-0.86	YES
4	1550	340	4.7	0.20	YES
5	1432	470	-3.2	-0.10	YES
6	1370	83	-7.4	-0.80	YES
7	1390	140	-6.1	-0.51	YES
8	1325	86	-10.5	-1.11	NO
9	1569	-	6.0	-	-
10	1384	180	-6.5	-0.46	YES
11	1355	140	-8.4	-0.70	YES
12	1370	280	-7.4	-0.37	YES
13	1559	49	5.3	0.66	YES
14	1526	47	3.1	0.38	YES
15	1490	46	0.7	0.08	YES
16	1478	69	-0.1	-0.02	YES
17	1230	29	-16.9	-2.20	NO
18	1391	180	-6.0	-0.42	YES
19	-	-	-		
20	1556	160	5.1	0.39	YES
21	1462	93	-1.2	-0.13	YES
22	1470	260	-0.7	-0.03	YES
23	1497	9	1.1	0.15	YES
24	1351	79	-8.7	-0.95	YES
25	1473	61	-0.5	-0.06	YES
26	1467	150	-0.9	-0.07	YES
27	1422	30	-3.9	-0.51	YES
28	1417	145	-4.2	-0.35	YES
29	1411	58	-4.7	-0.55	YES
30	1439	100	-2.8	-0.28	YES
31	1460	81	-1.4	-0.15	YES
32	1405	100	-5.0	-0.50	YES
33	1509	43	2.0	0.25	YES
34	1382	68	-6.6	-0.76	YES
35	3146	88	112.6	11.83	NO
36	1470	150	-0.7	-0.05	YES
37	1415	22	-4.4	-0.58	YES
38	1494	210	0.9	0.06	YES
39	1323	81	-10.6	-1.15	NO
40	1358	140	-8.3	-0.69	YES

Table 7b: Relative deviation of mean laboratory results A_{lab} from reference value A_{ref} and compatibility test (E_n numbers) for ^{137}Cs (continued)

lab no.	mean activity concentration A_{lab} / (Bq·kg^{-1})	expanded uncertainty U_{lab} / (Bq·kg^{-1})	relative deviation D/A_{ref} / %	E_n	compatibility
41	1375	93	-7.1	-0.73	YES
42	1446	70	-2.3	-0.26	YES
43	1346	95	-9.0	-0.92	YES
44	1246	78	-15.8	-1.74	NO
45	1524	210	3.0	0.19	YES
46	1415	150	-4.4	-0.35	YES
47	1380	110	-6.8	-0.64	YES
48	1380	210	-6.8	-0.42	YES
49	1392	87	-6.0	-0.63	YES
50	1373	105	-7.3	-0.71	YES
51	1450	58	-2.0	-0.24	YES
52	-	-	-		
53	1403	11	-5.2	-0.70	YES
54	1467	103	-0.9	-0.09	YES
55	-	-	-	-	-
56	1518	38	2.6	0.33	YES
57	1570.4	95	6.1	0.62	YES
58	1341	35	-9.4	-1.20	NO
59	1328	34	-10.3	-1.32	NO
60	1500	130	1.4	0.12	YES
61	1410	50	-4.7	-0.58	YES
62	-	-	-		
63	1414	180	-4.5	-0.31	YES

An alternative way of presentation, albeit yielding no new information versus Figures 8-10, is the deviation chart. Tables 7-9 contain the relative deviation of the lab mean values A_{lab} from the reference activity concentration A_{ref}:

$$Rel.dev. = \frac{A_{lab} - A_{ref}}{A_{ref}} \cdot 100 = \frac{D}{A_{ref}} \cdot 100 \qquad (7)$$

These values are plotted in ascending order in a deviation chart, and the laboratories reporting too low or too high values become more visible (Figs.20 - 22). Dashed red lines indicate a deliberately chosen range of ±20 % in the case of ^{137}Cs and ^{40}K and ±30 % in the case of ^{90}Sr. Laboratories which have reported values outside of the specified range were considered to deviate considerably from the reference value. These laboratories were contacted after the deadline for reporting results and were asked to critically review their analysis and measurement procedures and calculations, and to report back if the review would lead to changed values. Some of the contacted laboratories corrected their results and the newly submitted results were used for the final evaluation of the laboratory performance as presented here.

Table 8a: Relative deviation of mean laboratory results A_{lab} from reference value A_{ref} and compatibility test (E_n numbers) for ^{40}K

lab no.	mean activity concentration A_{lab} / (Bq·kg^{-1})	expanded uncertainty U_{lab} / (Bq·kg^{-1})	relative deviation D/A_{ref} / %	E_n	compatibility
1	560	55	3.6	0.26	YES
2	573	44	6.1	0.49	YES
3	503	56	-6.9	-0.49	YES
4	596	150	10.4	0.35	YES
5	514	180	-4.8	-0.14	YES
6	563	43	4.3	0.35	YES
7	520	60	-3.7	-0.26	YES
8	952	85	76.3	4.18	NO
9	585	-	8.4	-	-
10	518	69	-4.1	-0.26	YES
11	596	120	10.4	0.43	YES
12	538	57	-0.5	-0.03	YES
13	601	27	11.2	1.07	NO
14	574	18	6.3	0.64	YES
15	516	59	-4.4	-0.31	YES
16	553	42	2.4	0.20	YES
17	546	45	1.0	0.08	YES
18	498	68	-7.8	-0.50	YES
19	-	-	-		
20	559	56	3.6	0.26	YES
21	634	69	17.4	1.10	NO
22	654	150	21.1	0.72	YES
23	598	30	10.6	0.99	YES
24	533	31	-1.3	-0.12	YES
25	584	23	8.1	0.80	YES
26	581	130	7.6	0.29	YES
27	525	16	-2.9	-0.30	YES
28	567	59	5.1	0.35	YES
29	545	48	0.9	0.07	YES
30	574	35	6.3	0.56	YES
31	534	41	-1.1	-0.09	YES
32	535	53	-0.9	-0.06	YES
33	678	64	25.5	1.69	NO
34	610	30	12.9	1.19	NO
35	1302	66	141.1	9.20	NO
36	570	60	5.6	0.38	YES
37	628	23	16.3	1.60	NO
38	588	83	8.9	0.50	YES
39	513	71	-5.0	-0.31	YES
40	530	54	-1.9	-0.14	YES

Table 8b: Relative deviation of mean laboratory results A_{lab} from reference value A_{ref} and compatibility test (E_n numbers) for ^{40}K (continued)

lab no.	mean activity concentration A_{lab} / (Bq·kg^{-1})	expanded uncertainty U_{lab} / (Bq·kg^{-1})	relative deviation D/A_{ref} / %	E_n	compatibility
41	463	150	-14.3	-0.49	YES
42	546	42	1.0	0.08	YES
43	524	39	-2.9	-0.25	YES
44	506	32	-6.3	-0.57	YES
45	532	62	-1.6	-0.11	YES
46	553	77	2.5	0.15	YES
47	530	45	-1.9	-0.15	YES
48	585	87	8.2	0.44	YES
49	549	36	1.7	0.15	YES
50	549	43	1.7	0.14	YES
51	586	25	8.5	0.82	YES
52	-	-	-		
53	464	29	-14.1	-1.31	NO
54	565	68	4.7	0.30	YES
55	-	-	-	-	-
56	601	26	11.4	1.09	NO
57	471	38	-12.8	-1.10	NO
58	629	63	16.5	1.11	NO
59	633	61	17.3	1.18	NO
60	602	64	11.4	0.76	YES
61	521	10	-3.5	-0.37	YES
62	-	-	-		
63	533	71	-1.3	-0.08	YES

Table 9a: Relative deviation of mean laboratory results A_{lab} from reference value A_{ref} and compatibility test (E_n numbers) for ^{90}Sr

lab no.	mean activity concentration A_{lab} / (Bq·kg^{-1})	expanded uncertainty U_{lab} / (Bq·kg^{-1})	relative deviation D/A_{ref} / %	E_n	compatibility
1	7.68	1.5	57.2	1.77	NO
2	-	-	-	-	-
3	5.71	0.47	17.0	1.21	NO
4	5.25	1.2	7.4	0.28	YES
5	4.85	0.67	-0.6	-0.04	YES
6	-	-	-	-	-
7	4.69	0.88	-4.0	-0.19	YES
8	-	-	-	-	-
9	5.55	-	13.7	-	-
10	5.12	0.80	5.0	0.26	YES
11	4.25	0.85	-13.0	-0.65	YES
12	-	-	-	-	-
13	-	-	-	-	-
14	-	-	-	-	-
15	5.20	0.60	6.5	0.41	YES
16	5.16	0.35	5.7	0.46	YES
17	7.71	1.1	58.0	2.34	NO
18	4.90	0.52	0.3	0.02	YES
19		-	-		
20	5.01	1.0	2.7	0.12	YES
21	5.00	0.57	2.4	0.16	YES
22	-	-	-	-	-
23	21.10	1.9	332.3	8.26	NO
24	4.92	0.59	0.7	0.05	YES
25	6.16	1.7	26.1	0.72	YES
26	1.85	0.40	-62.1	-4.73	NO
27	-	-	-	-	-
28	6.07	1.3	24.3	0.85	YES
29	5.31	0.38	8.8	0.68	YES
30	6.44	0.70	32.0	1.82	NO
31	5.30	1.1	8.6	0.35	YES
32	7.97	1.1	63.2	2.55	NO
33	-	-	-	-	-
34	6.18	0.57	26.5	1.71	NO
35	-	-		-	-
36	4.30	0.60	-11.9	-0.74	YES
37	-	-	-	-	-
38	4.63	1.2	-5.2	-0.19	YES
39	4.00	1.1	-18.1	-0.73	YES
40	4.67	0.40	-4.4	-0.34	YES

Table 9b: Relative deviation of mean laboratory results A_{lab} from reference value A_{ref} and compatibility test (E_n numbers) for ^{90}Sr (continued)

lab no.	mean activity concentration A_{lab} / (Bq·kg^{-1})	expanded uncertainty U_{lab} / (Bq·kg^{-1})	relative deviation D/A_{ref} / %	E_n	compatibility
41	4.67	0.41	-4.3	-0.32	YES
42	5.40	0.8	10.5	0.54	YES
43	5.20	1.2	6.5	0.25	YES
44	4.16	1.1	-14.7	-0.59	YES
45	4.44	1.1	-9.1	-0.37	YES
46	4.04	1.0	-17.3	-0.76	YES
47	4.65	0.67	-4.7	-0.28	YES
48	4.16	0.35	-14.9	-1.19	NO
49	5.32	0.70	9.1	0.51	YES
50	8.78	3.7	79.9	1.04	NO
51	9.22	0.57	88.9	5.73	NO
52	-	-	-		
53	-	-	-	-	-
54	-	-	-	-	-
55	4.56	0.05	-6.7	-0.65	YES
56	6.85	0.60	40.3	2.52	NO
57	13.12	1.4	168.7	5.54	NO
58	6.36	1.6	30.2	0.88	YES
59	-	-	-	-	-
60	-	-	-	-	-
61	6.15	0.73	26.0	1.43	NO
62	-	-	-		
63	4.77	0.74	-2.2	-0.12	YES

For completeness, the expanded uncertainty U_{ref} of the reference value is plotted as dotted red lines in Figs. 20-22. In the figures as well as in Tables 7-9, large relative deviations are highlighted by colours: orange/yellow for D/A_{ref} > 15 % (> 25 % in case of ^{90}Sr), red/violet for D/A_{ref} > 20 % (> 30 % in case of ^{90}Sr).

As seen from the distribution of ^{137}Cs results, laboratory no. 35 reported a value which deviates more than 110 % from the reference value (Fig. 20). The ^{40}K result of the same participant deviates more than 140 % from the assigned value as well (Fig. 21). Laboratory 35 followed its routine analytical procedure and no sample preparation was applied. The ^{137}Cs activity concentration in the comparison samples, however, was many times higher than the activities measured routinely. Another laboratory (no. 8) reported a far too high activity concentration (> 70 % too high) for ^{40}K. Also in this laboratory the sample was treated according to the routine measurement procedure. Another two laboratories deviate more than 20 % from the ^{40}K reference value (cf. Fig. 21).

Considering the fact that the vast majority of laboratories (95 %) are able to determine ^{137}Cs at this high activity concentration with a relative deviation of < 11 %,

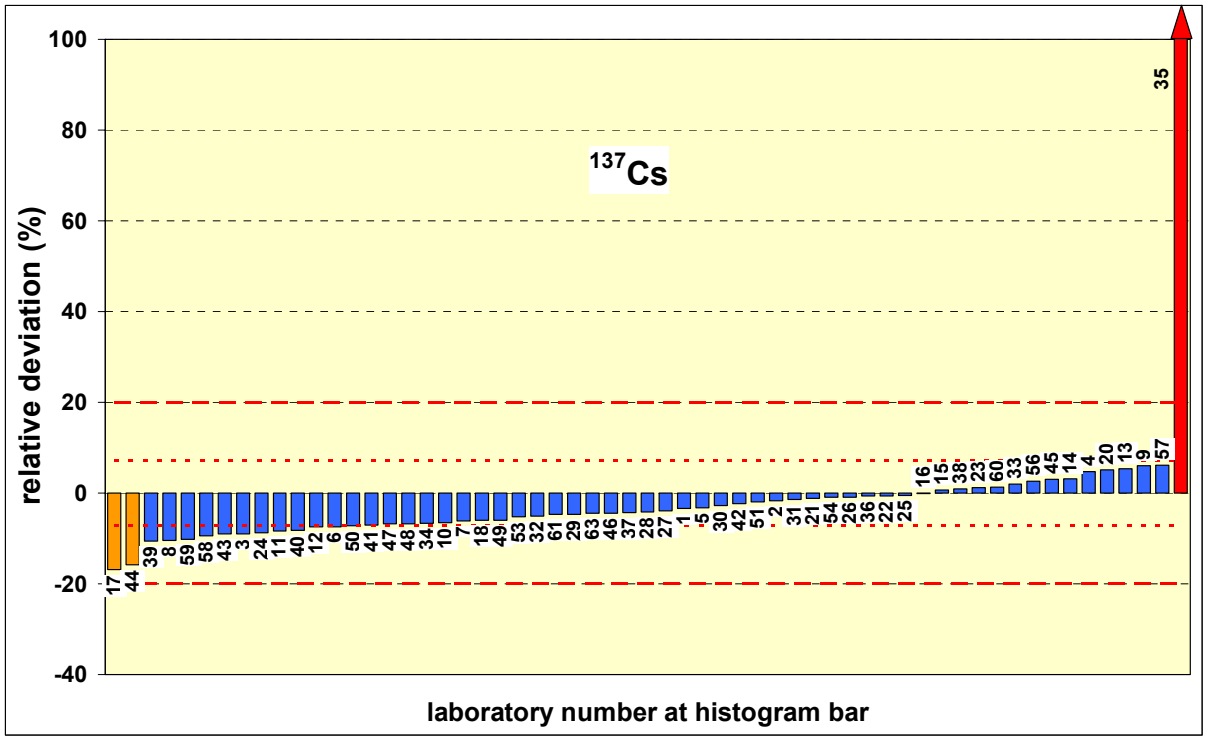

Fig. 20: Deviation chart of the 59 ^{137}Cs measurement results plotted as D/A_{ref} in ascending order. Range of ±20 % (dashed) and expanded uncertainty U_{ref} (dotted) are indicated

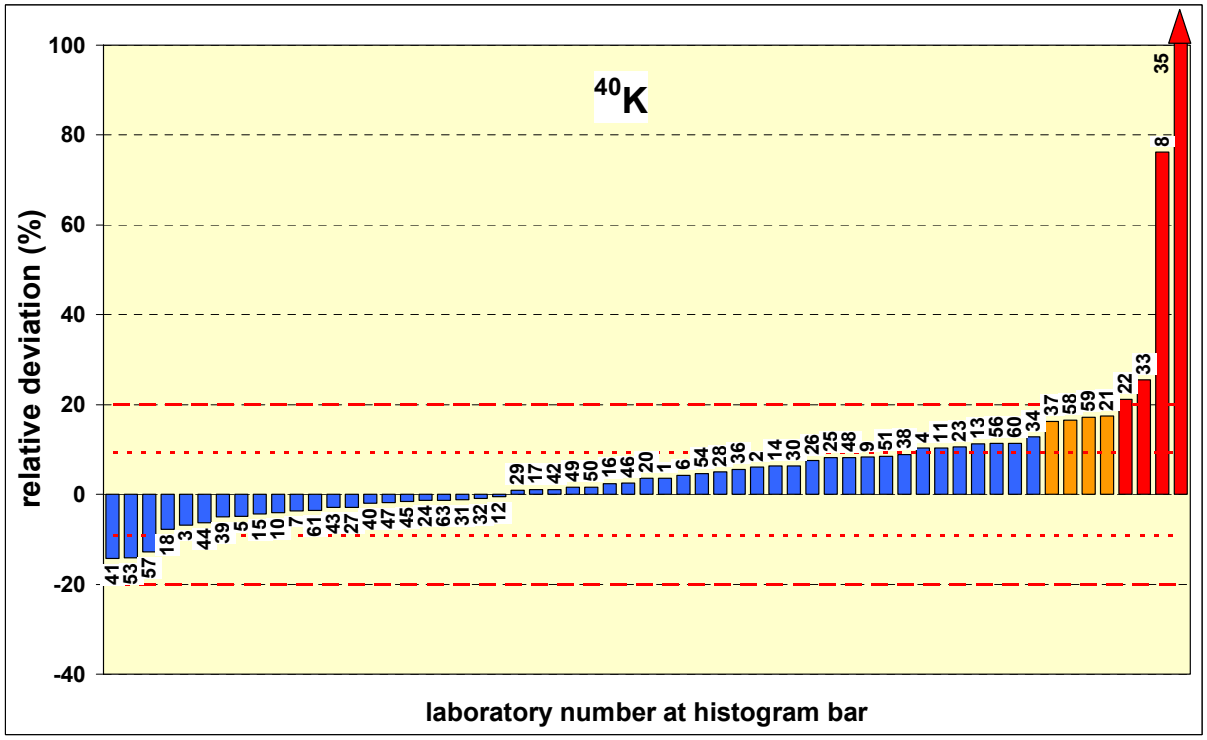

Fig. 21: Deviation chart of the 59 ^{40}K measurement results plotted as D/A_{ref} in ascending order. Range of ±20 % (dashed) and expanded uncertainty U_{ref} (dotted) are indicated

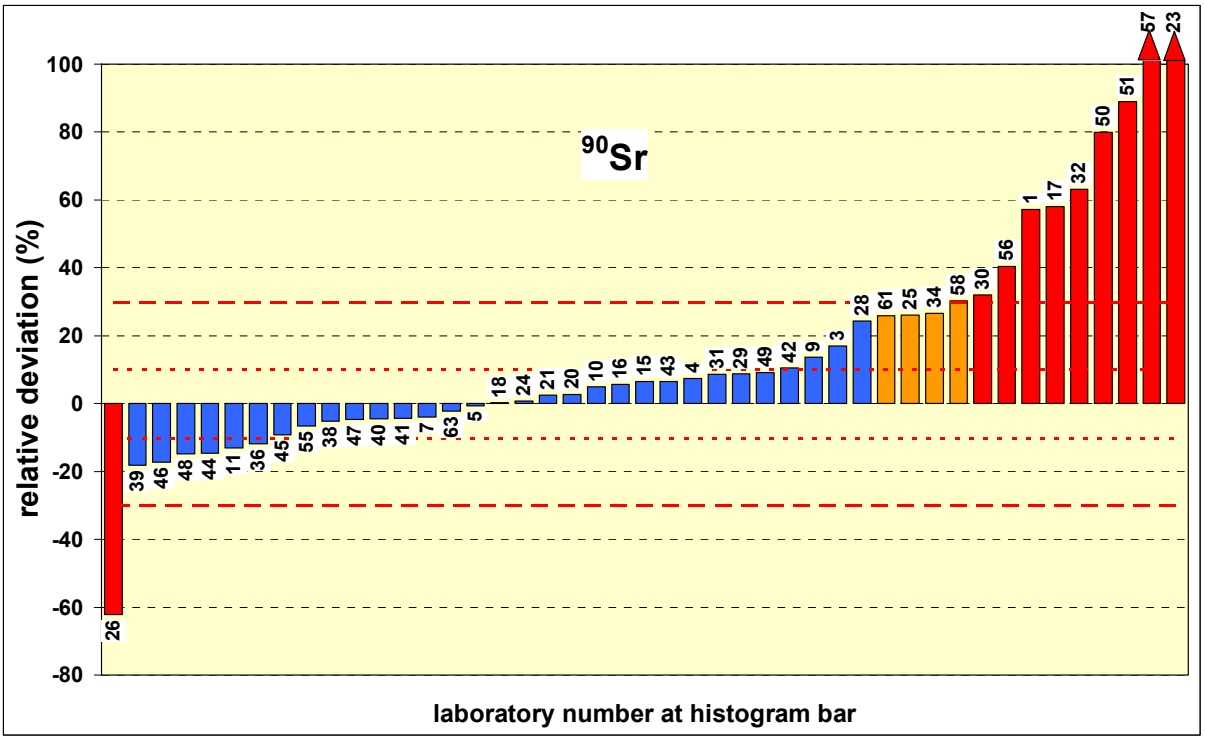

Fig. 22: Deviation chart of the 45 ^{90}Sr measurement results plotted as D/A_{ref} in ascending order. Range of ±30 % (dashed) and expanded uncertainty U_{ref} (dotted) are indicated

one can only conclude that the arbitrarily chosen criterion of 20 % deviation was too unselective in this case. A similar conclusion can be drawn for ^{40}K. Worrying, however, is the observation that one laboratory (no. 35) reported results for both gamma-ray emitters twice as large as the reference values.

For ^{90}Sr the distribution of the results (Fig. 22) is not as uniform as for the other two radionuclides. Two laboratories with results deviating more than ± 30 % from the reference value (labs 26 and 32) reported problems during the ^{90}Sr determination. A probable factor influencing the measurement of laboratory 26 may be the inability to reconstitute milk from the milk powder which led to an inhomogeneous liquid sample, whereas in the second laboratory (no. 32) impurities of ^{137}Cs were found in the sample prepared for LSC. Another eight participants reported activity concentrations deviating more than 30 % from the reference value as well (labs 1, 17, 23, 30, 50, 51, 56, 57). In no. 1, 32, 51 and 57 the ^{90}Sr activity concentration was determined after radiochemical separation by liquid scintillation counting, while in laboratories 17 and 23 gas flow proportional counters were used. Laboratory 56 used a low-level Geiger-Müller counter, lab 26 carried out Cherenkov counting and laboratories 30 and 50 applied plastic scintillators for gross beta counting (cf. also Fig. 11). In all these cases the ^{90}Sr activity was determined with the routine analytical procedures, and no difficulties during determination were reported by the participants. It must be pointed out, however, that the ^{90}Sr results of laboratories 57 and 23 deviate by as much as 170 % and 330 %, respectively.

Just as all counting methods are represented among these ten largely deviating results, there is not a single sample preparation or radiochemical separation procedure which could be pin-pointed as being particularly frequent among or even responsible for largely deviating results (cf. also Fig. 12).

In order to allow a more detailed analysis, several statistical tests – taking the measurement uncertainty and that of the reference values into account – have been applied. Strictly speaking, tests including measurement uncertainty must be used with caution when the uncertainty estimation is poorly understood. We have already observed, in chapter 6, that this holds for one third up to half of the participating laboratories. Nevertheless, the selected performance test using E_n numbers [12, 15] of the activity concentrations proves to be robust enough justifying its use in this evaluation.* The E_n number takes into account the absolute deviation of the activity concentration value reported by each laboratory (A_{lab}) from the reference value (A_{ref}) and the combination of expanded uncertainties associated to them (U_{lab} and U_{ref}) [12, 15]. It will become clear below that the conclusions from the previous discussion (which ignored uncertainty) will in general be confirmed and some additional insight will be gained.

The performance statistic "E_n number" is calculated as [12, 15]:

$$E_n = \frac{A_{lab} - A_{ref}}{\sqrt{U_{lab}^2 + U_{ref}^2}} \qquad (8)$$

where

A_{lab} the participant's result, mean activity concentration;
A_{ref} the reference value;
U_{lab} the expanded uncertainty of the participant's result;
U_{ref} the expanded uncertainty of the reference value.

When the estimation of uncertainties is consistent with the Guide to the Expression of Uncertainty in Measurement (GUM) [3], a measurement result with its uncertainty interval giving a level of confidence of 95 % (corresponding to $A_{lab} \pm U_{lab}$ with an expanded uncertainty $U_{lab} = k \cdot u_c$ with a coverage factor of $k \approx 2$) will overlap with the reference value A_{ref} (and its expanded uncertainty U_{ref}).

Therefore, E_n numbers are interpreted in the following way:

If $|E_n| \leq 1$, the laboratory values are compatible with the reference value;

If $|E_n| > 1$, "warning signal", the laboratory values differ significantly from the reference value, sources of deviation should be investigated and corrected (yellow colour in Tables 7-9);

In analogy to the interpretation of *z*-scores [12], a second level of critical value can be defined:

If $|E_n| > 1.5$, "action signal", there is urgent need to investigate and find the sources of the large deviation (orange colour in Tables 7-9).

The E_n numbers and test results are given for each laboratory in the last two columns of Tables 7-9. Sorted in ascending order, the E_n numbers are graphically presented in Figures 23-25. Laboratory 9, which did not provide any uncertainty estimate, could not be considered for the E_n criterion.

* One should keep in mind that other performance tests usually also have constraints, e.g. a normal distribution of results, which are not always met.

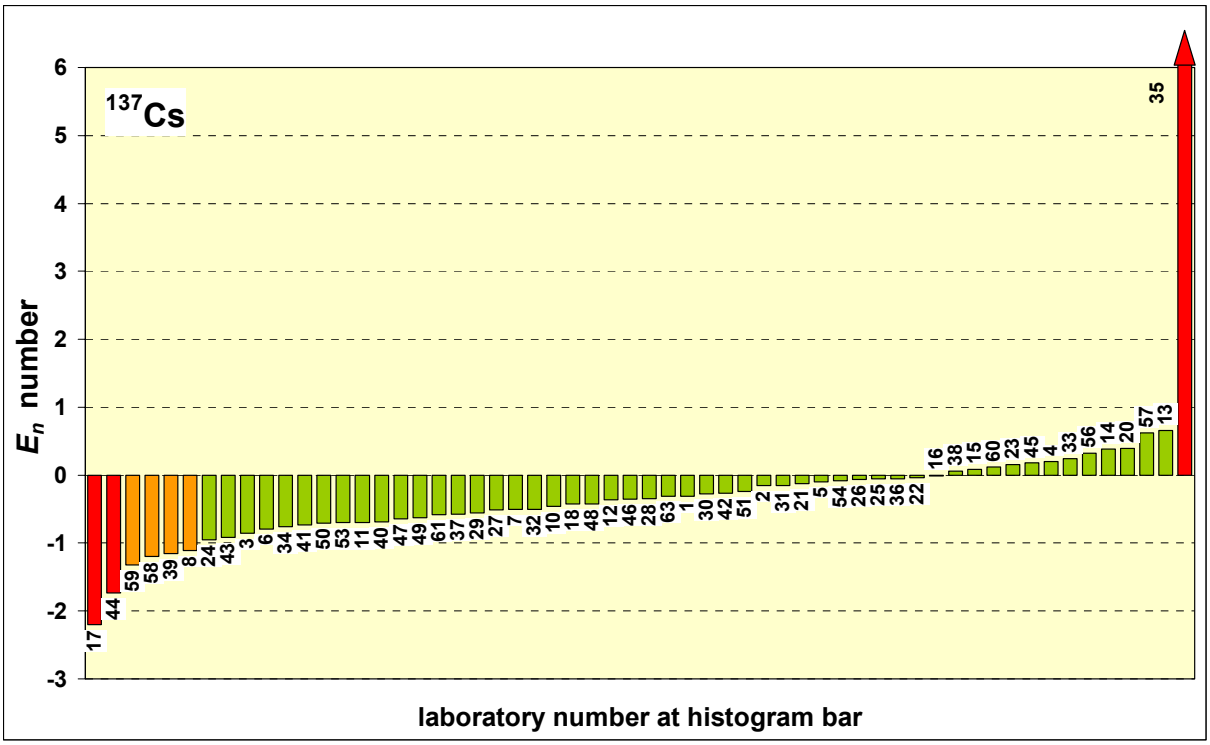

Fig. 23: E_n number of ^{137}Cs activity concentrations obtained by 58 laboratories, sorted in ascending order

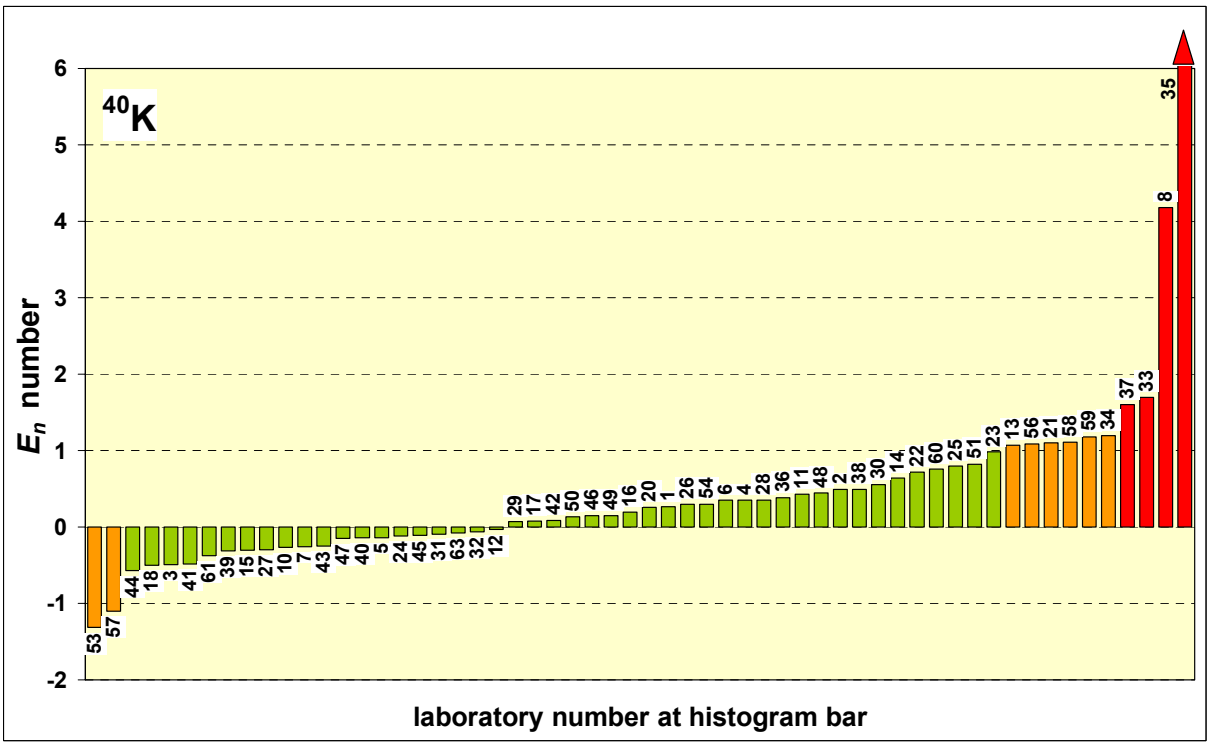

Fig. 24: E_n number of ^{40}K activity concentrations obtained by 58 laboratories, sorted in ascending order

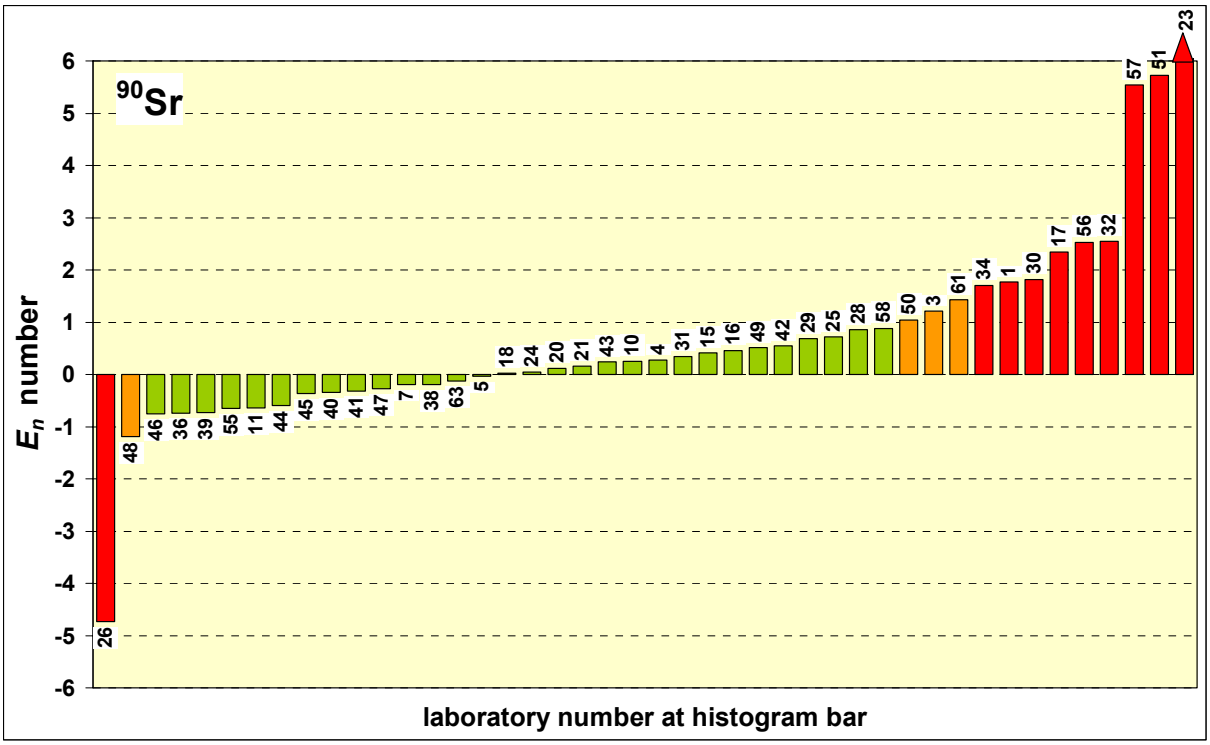

Fig. 25: E_n number of ^{90}Sr activity concentrations obtained by 44 laboratories, sorted in ascending order

Based on the E_n number criterion, 51 results out of 58 (88 %) are compatible with the reference activity concentrations for ^{137}Cs under the conditions of this test, 7 are not. Among those 7, three laboratories (No. 17, 35 and 44) report largely incompatible results ($|E_n| > 1.5$). When comparing Figure 23 with 20, it is clear that results having large deviations from the reference value are scoring bad with E_n numbers as well. That comparison, however, also shows the effect of estimating uncertainty *too small*: Some laboratories with a still acceptable deviation of < 15 % are assigned a critical E_n number, because *not all contributions to uncertainty are correctly estimated* (laboratories No. 8, 39, 58 and 59). If a laboratory controls the measurement process well with only very small deviations *and* obtains a realistic estimate of uncertainty as well*, the E_n number is close to 0. Table 7 and Figure 23 depict several laboratories of this arbitrarily chosen category of $|E_n| < 0.1$: No. 5, 15, 16, 22, 25, 26, 36, 38 and 54.

Similarly for ^{40}K, 46 results (79 %) are compatible with the reference value and 12 are not. Four laboratories (labs 8, 33, 35 and 37) of the incompatible ones have an $|E_n| > 1.5$ and three of these (No. 8, 33 and 35) deviate more than 20 % from the reference value (Fig. 21). In the case of ^{90}Sr, 30 values (68 %) are satisfactory and 14 results are not, 10 of which (labs 1, 17, 23, 26, 30, 32, 50, 51, 56, 57) show deviations larger than 30 % from A_{ref} (cf. Fig. 22).

The preceding discussion is summarised in Table 10.

* Unfortunately, “guessing” a too large uncertainty would reduce the $|E_n|$ number without justification.

Table 10: Brief summary of the evaluation of laboratory results of ^{137}Cs, ^{40}K and ^{90}Sr in milk powder

Measurand	Number of compatible labs $\lvert E_n \rvert \leq 1$	Number of incompatible labs $\lvert E_n \rvert > 1$	Number of largely incompatible labs $\lvert E_n \rvert > 1.5$	Number of labs with "large deviation"
^{137}Cs	51	7	3	1 **(>20%)**
^{40}K	46	12	4	4 **(>20%)**
^{90}Sr	30	14	10	10 **(>30%)**

Some further statistical evaluations serve to characterise this intercomparison in a general way, which do not replace, however, the performance evaluation of individual laboratories. Table 11 summarises the basic characteristics of the distribution of measurement results.

Table 11: Statistical analysis of the laboratory results of ^{137}Cs, ^{40}K and ^{90}Sr in milk powder

Measurand	^{137}Cs	^{40}K	^{90}Sr
Number of results	59	59	45
Minimum value ($Bq \cdot kg^{-1}$)	1230	463	1.9
Maximum value ($Bq \cdot kg^{-1}$)	3146	1302	21
Median ($Bq \cdot kg^{-1}$)	1417	559	5.2
Mean value ($Bq \cdot kg^{-1}$)	1455	578	5.9
Reference value ($Bq \cdot kg^{-1}$)	**1480**	**540**	**4.9**
Expanded uncertainty ($k=2$) ($Bq \cdot kg^{-1}$)	**110**	**50**	**0.5**

The Nalimov test was used to identify purely statistical outliers among the reported results. In the case of ^{137}Cs, the result of laboratory No. 35 was flagged in first instance at a level of significance $\alpha = 0.01$, and the result of No. 17 in the second round of testing and No. 44 in the third round at the same level of significance after excluding the first outlier. No further outliers were identified on statistical grounds (at $\alpha = 0.01$) with neither the Nalimov nor other tests.

After exclusion of the three outlying results identified by the test, the skewness and kurtosis tests indicate the normality of the remaining data at a level of significance of $\alpha = 0.01$.

Performing the Nalimov test on the ^{40}K data, the results of laboratory No. 8 and 35 were marked as outliers at a level of significance $\alpha = 0.01$. The result of lab 33 was indicated in the second round of the test at the same level of significance. The laboratory results of ^{90}Sr were, as expected, statistically more problematic as only after six rounds of the Nalimov test there were no more outliers detected in the remaining data set. Laboratory 23 was indicated as an outlier at $\alpha = 0.01$ in the first round, No. 57 in the second and No.26 and 51 in the third round. Labs 50, 32, 1 and 17 were indicated as outliers in the next rounds of the test. After exclusion of all values indicated by the test as outliers, the skewness and kurtosis tests indicate the normality of the remaining data at a level of significance of $\alpha = 0.01$.

Table 12 reflects the distribution of results after exclusion of the outliers identified by the Nalimov test.

Table 12: Statistical analysis of the laboratory results of ^{137}Cs, ^{40}K and ^{90}Sr in milk powder excluding the identified outliers (see text)

Measurand	**^{137}Cs**	**^{40}K**	**^{90}Sr**
Number of results	56	56	37
Minimum value (Bq·kg^{-1})	1323	463	4.0
Maximum value (Bq·kg^{-1})	1570	654	6.9
Median (Bq·kg^{-1})	1419	553	5.0
Mean value (Bq·kg^{-1})	1433	556	5.1
Reference value (Bq·kg^{-1})	**1480**	**540**	**4.9**
Expanded uncertainty (*k*=2) (Bq·kg^{-1})	**110**	**50**	**0.5**

8. Conclusions

The measurement comparison "^{137}Cs, ^{40}K and ^{90}Sr in milk powder" allowed to obtain a generally realistic estimate of the trueness of radioactivity measured in milk powder during routine monitoring in EU Member States and neighbouring countries.

With few exceptions, the intercomparison samples were radiochemically treated and measured by the participating laboratories as under routine conditions. Of course, in order to be able to establish reference values with credible and acceptably small uncertainty, intercomparison samples with higher levels of the anthropogenic radionuclides ^{137}Cs and ^{90}Sr than what is usually seen in routine monitoring had to be distributed to the laboratories. Only a few laboratories in Eastern European countries are measuring – according to the returned questionnaire – ^{90}Sr at levels in milk powder approaching that of the intercomparison samples. The ^{40}K activity concentration was of the same level as reported under monitoring conditions due to its natural occurrence. A material was chosen which contained the elevated nuclides in a naturally metabolised state.

With a view to the elevated levels, the pure measurement task was thus in principle a bit easier than under routine conditions, in particular referring to counting uncertainty and background subtraction. Whereas the sample preparation and separation procedures were not influenced by elevated – yet still low – radioactivity levels.

The few cases where the distributed samples required an approach different from the routine procedures are mainly due to the fact that many laboratories are monitoring milk in its liquid form, and some also perform the measurements on liquid samples. In these cases, the inability to reconstitute a homogeneous liquid from the milk powder may have been a possible source of error during analysis. Consequently, the results obtained by the corresponding laboratories may not reflect completely their measurement capability under routine conditions.

In general, the measurement results for the ^{137}Cs and ^{40}K activity concentration showed good agreement with the reference values. However, the measurement uncertainty was often underestimated. About 88 % and 79 % (for ^{137}Cs and ^{40}K, respectively) of the laboratories were compatible with the reference values based on the E_n number criterion, and of the, respectively, 7 and 12 incompatible laboratories one and four deviated by more than 20 %. For ^{90}Sr, about one third of the reported results were not compatible following the E_n number, and 10 laboratories deviated by more than 30 % from the reference value. It is hoped that these results will stimulate the necessary actions for improvement – on the side of the analysis methods as well as on the correct propagation of uncertainties following the GUM [3]. Together with uncertainty assignments, which are clearly underestimated in many laboratories and apparently also overestimated in some, the implementation of analysis methods in particular for ^{90}Sr should be critically reviewed and improved in the corresponding laboratories. It must be stressed that the results of this comparison do *not* give evidence that a particular separation method *as such* might perform significantly worse (or better) for ^{90}Sr determination than others.

9. Acknowledgements

This work was possible only with the active participation of 60 laboratories in 32 countries (listed in chapter 11 below), and the support of A. Janssens from the Directorate General for Energy and Transport of the European Commission. The authors would also like to thank A. Bohnstedt for her contribution to the establishment of the traceable reference value of ^{90}Sr and A. Oostra, J. Charoud-Got, P. Conneely and A. Bernreuther from the Reference Materials Unit of IRMM for the re-processing of the milk powder and for determining the moisture content of the packaged samples.

10. References

[1] MacMahon D., Pearce A., Harris P., 2004. Convergence of techniques for the evaluation of discrepant data, *Appl. Radiat. Isot.* **60**, pp. 275–281.

[2] Nucléide 2000, Version 2-2004, Nuclear and Atomic Decay Data software, BNM – LNHB/CEA, http://www.nucleide.org/NucData.htm.

[3] GUM:1995, Guide to the expression of uncertainty in measurement. BIPM, IEC, IFCC, ISO, IUPAC, IUPAP, OIML, corrected and reprinted version. ISBN 92-67-10188-9.

[4] Rückold, S., Grobecker K.H., Isengard H.D., 2000. Determination of the contents of water and moisture in milk powder, *J. Anal. Chem.* **368**, pp. 522-527.

[5] Altzitzoglou T., Bohnstedt A., 2006. Characterisation of the IAEA-152 milk powder intercomparison material for radioactivity with assigned values traceable to the SI units, IRMM Internal Report no. GE/R/IM/15/06.

[6] Lepy, M.C., 2000. EUROMET Action 428; Transfer of Ge detectors efficiency calibration from point source geometry to other geometries. Rapport CEA-R-5894.

[7] Sole, V.A., 1990. A Monte Carlo program to calculate solid angle and transmission corrections in counting systems. IRMM Internal Report no. GE/R/RN/12/90.

[8] Coursey, B.M., Gibson, J.A.B., Heitzmann, M.W., Leak, J. C., 1984. Standardization of technetium-99 by liquid-scintillation counting, *Int. J. Appl. Radiat. Isot.* **35**, pp. 1103-1112.

[9] Grau Malonda, A., Garcia-Toraño, E., 1982. Evaluation of counting efficiency in liquid scintillation counting of pure β-ray emitters, *Int. J. Appl. Radiat. Isot*. **33**, pp. 249- 253.

[10] Grau Malonda, A., Garcia-Toraño, E., Los Arcos, J.M., 1985. Liquid-scintillation counting efficiency as a function of the figure of merit for pure beta-particle emitters, *Int. J. Appl. Radiat. Isot.* **36**, pp. 157-158.

[11] Altzitzoglou, T., Sibbens, G., Bickel, M., Bohnstedt, A., Decaillon, J.-G., Hill, C, Holmes, L., 2004. Characterisation of reference materials for radioactivity with assigned values traceable to the SI units, *Appl. Radiat. Isot.* **61**, pp. 395-399.

[12] ISO/FDIS 13528:2005 (E), Statistical methods for use in proficiency testing by interlaboratory comparisons. ISO – International Organization for Standardization, Geneva.

[13] Brun, S., Kergadallan, Y., Boursier, B., Fremy, J.-M., Janin, F., 2003. Methodology for determination of radiostrontium in milk: a review, *Lait* **83**, pp. 1-15.

[14] Pommé S., 2006. An intuitive visualisation of intercomparison results applied to the KCDB, *Appl. Radiat. and Isot.* **64**, pp. 1158-1162.

[15] ISO Guide 43-1:1997, Proficiency testing by interlaboratory comparisons – Part 1: Development and operation of proficiency testing schemes.

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Strahlenschutzmessstelle Frankfurt (Oder)
Ringstr. 1030
15236 Frankfurt (Oder)
Responsible: Jörg Dietrich

Landeslabor Brandenburg
Strahlenschutzmessstelle Oranienburg
Sachsenhausener Str. 7
16515 Oranienburg
Responsible: Joachim Beetz

GREECE

Greek Atomic Energy Commission (GAEC)
Department of Environmental Radioactivity
P.O. Box 60092
153 10 Agia Paraskevi, Attikis
Responsible: Konstantinos Potiriadis, Virginia Koukouliou

NCSR "Demokritos"
Institute of Nuclear Technology (INT)
RP/ERL
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153 10 Agia Paraskevi, Attikis
Responsible: Heleny Florou

HUNGARY

National Food Investigation Institute
Department of Radiochemistry
P.O. Box 1740
1465 Budapest
Responsible: Sandor Tarjan

National Research Institute for Radiobiology and Radiohygiene
P.O. Box 101
1775 Budapest
Responsible: Ágota Ugron, Gyula Szábo, Júlia Kónyi

ICELAND

Icelandic Radiation Protection Institute
Raudararstig 10
150 Reykjavik
Responsible: Sigurður Emil Pálsson

IRELAND

Radiological Protection Institute of Ireland (RPII)
Environmnetal Laboratory
3, Clonskeagh Square
Dublin 14
Responsible: Mary Fegan, Savio Sequeira, Alison Dowdall

University College Dublin (UCD)
Department of Experimental Physics
Belfield
Dublin 4
Responsible: Luis Leon Vintro

ITALY

Agenzia Regionale Prevenzione e Ambiente dell'Emilia-Romagna
(ARPA Emilia-Romagna)
Via XXI Aprile, 48
29100 Piacenza
Responsible: Roberto Sogni, Laura Gaidolfi

Agenzia Regionale per la Protezione Ambientale
(ARPA – Piemonte)
Centro regionale per le radiazioni ionizzanti e non ionizzanti
Via Jervis 30
10015 Ivrea
Responsible: Mauro Magnoni

Agenzia Regionale per la Protezione dell'Ambiente della Lombardia
(ARPA Lombardia)
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20129 Milano
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LATVIA

State Veterinary Medicine Diagnostic Centre
Radiology Department
3, Lejupes Str.
1076 Riga
Responsible: Janis Rudzitis

LITHUANIA

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Kalvariju 153
08221 Vilnius
Responsible: Gendrutis Morkunas

Institute of Physics
Nuclear Research and Environmental Radioactivity Research Laboratory
Savanoriu Ave 231
2053 Vilnius
Responsible: Arunas Gudelis

LUXEMBOURG

Direction de la Santé
Division de la Radioprotection
Villa Louvigny, Allée Marconi
2120 Luxembourg
Responsible: Marielle Lecomte

MALTA

Ministry of Health, the Elderly and Community Care (MHEC)
Public Health Laboratory
Evans Bldg., Lower Merchants Street
CMR 02 Valletta
Responsible: Rose Schembri, Raymond Camilleri

THE NETHERLANDS

National Institute for Public Health and the Environment (RIVM)
Laboratory for Radiation Research
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3720 BA Bilthoven
Responsible: P.J.M. Kwakman

RIKILT - Institute of Food Safety
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Postbus 230
6700 AE Wageningen
Responsible: Jasper Hattink

POLAND

Central Laboratory for Radiological Protection (CLOR)
Dosimetry Department
ul. Konwaliowa 7
03-194 Warszawa
Responsible: Pawel Lipiński

PORTUGAL

Instituto Tecnológico e Nuclear (ITN)
Departamento de Protecção Radiológica e Segurança Nuclear (DPRSN)
E.N. 10
2686-953 Sacavém
Responsible: Maria José Madruga

ROMANIA

National Commission for Nuclear Activities Control (CNCAN)
Radioprotection and Radiological Emergencies Section
Bulevardul Libertății no. 14, Sector 5
P.O. Box 42-4
Bucharest
Responsible: Florian Baciu

National Research and Development Institute for Environmental Protection (ICIM)
Environmental Radioactivity Laboratory
Splaiul Independenței no. 294, Sector 6
60031 Bucharest
Responsible: Gyongyi Ruzsa

SLOVAKIA

Public Health Authority of the Slovak Republic
P.O. Box 45
826 45 Bratislava
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Regional Public Health Authority
Department of Radiation Protection
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975 56 Banska Bystrica
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State Veterinary and Food Institute Nitra
Laboratory of Radiometry
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Institute of Occupational Safety (ZVD)
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Responsible: Peter Jovanovič

Jožef Stefan Institute
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1001 Ljubljana
Responsible: Matjaž Korun

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Universidad de A Coruña
Departamento de Química Analítica
Escuela Universitaria Politécnica de Ferrol
Laboratory of Environmental Radioactivity
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15405 A Coruña
Responsible: Alfonso Calleja García

Universidad de León
Departamento de Física
Campus de Vegazana
24071 León
Responsible: Benito de Celis

SWEDEN

Swedish Defence Research Agency (FOI)
NBC – Skydd
Cementvägen 20
901 82 Umeå
Responsible: Annika Tovedal

Swedish Radiation Protection Authority
Dept. of Emergency, Preparedness and Environmental Assessment
171 16 Stockholm
Responsible: Inger Östergren, Lena Wallberg

SWITZERLAND

Institut de Radiophysique Appliquée
Grand Pré 1
1007 Lausanne
Responsible: Pascal Froidevaux

Bundesamt für Gesundheit
Sektion Ueberwachung der Radioaktivität (SUeR)
Chemin du Musée 3
1700 Fribourg
Responsible: Sybille Estier

TURKEY

Çekmece Nuclear Research and Training Center (CNAEM)
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34149 Istanbul
Responsible: Nurdan Güngör

Çekmece Nuclear Research and Training Center (CNAEM)
Nuclear Physics and Chemistry Laboratory
P.K. 1, Atatürk Havalimani
34149 Istanbul
Responsible: Neşet Öztürk, Bektaş Karakelle

UNITED KINGDOM

National Radiological Protection Board
Environmental Assessments Department
Chilton, Didcot
Oxfordshire OX11 0RQ
Responsible: George J. Ham

Veterinary Laboratories Agency
Radiochemistry Unit
New Haw
Surrey KT15 3NB
Responsible: Tony Dell, Peter Hodson

12. Annexes

Annex 1: Moisture content determined by Karl-Fischer titration

EUROPEAN COMMISSION
DIRECTORATE GENERAL JRC
JOINT RESEARCH CENTRE
Institute for Reference Materials and Measurements
Reference Materials Unit
Retiesoweg 2440 Geel Belgium

RM Unit Report of Analysis # 419

Requested by: y.spasova

Applicant sample identification: See attachment

RM Unit sample ID: See Below

Date of receipt of samples: 01/07/2005

Condition of the samples ok

RM sample ID	Result (– expanded uncertainty)		unit	method
2437	3.82 , 4.17	Mean 3.99 U = 0.54	g/100 g	VKFT (WI/0137)
2438	3.59 , 3.77	Mean 3.68 U = 0.50	g/100 g	VKFT (WI/0137)
2439	3.82 , 3.75	Mean 3.79 U = 0.51	g/100 g	VKFT (WI/0137)
2440	3.88 , 3.94	Mean 3.91 U = 0.53	g/100 g	VKFT (WI/0137)
2441	3.84 , 3.89	Mean 3.87 U = 0.52	g/100 g	VKFT (WI/0137)
2442	4.05 , 4.07	Mean 4.06 U = 0.55	g/100 g	VKFT (WI/0137)
2443	3.94 , 3.79	Mean 3.87 U = 0.52	g/100 g	VKFT (WI/0137)
2444	3.90 , 3.82	Mean 3.86 U = 0.52	g/100 g	VKFT (WI/0137)
2445	3.82 , 3.77	Mean 3.80 U = 0.51	g/100 g	VKFT (WI/0137)
2446	4.20 , 3.99	Mean 4.10 U = 0.55	g/100 g	VKFT (WI/0137)

Filename(s) of raw data: J : / BackupTiNet

05/09/05.
Date:

signature responsible Analyst
Patrick Conneely

signature Laboratory responsible
Alex Bernreuther

Note: No feedback within 2 weeks is seen as acceptance of the report. Potential rests of samples will be destroyed after that period.

Annex 2: Letter distributed with samples

EUROPEAN COMMISSION
DIRECTORATE GENERAL JRC
JOINT RESEARCH CENTRE
Institute for Reference Materials and Measurements
IRMM

Geel, 17 December 2004
Ref. UW-EC-04

Subj: EC intercomparison ^{40}K, ^{90}Sr and ^{137}Cs in milk powder
Articles 35-36 of the Euratom Treaty

Dear colleague,

Enclosed please find two bottles of milk powder of about 103 g mass each. The material is homogenized and packed in different bottles for convenience only. We ask you to determine the specific activity (Bq/kg) of the three radionuclides ^{40}K, ^{90}Sr and ^{137}Cs in this milk powder. We hope that the supplied amount of material is sufficient in order to allow you gamma-measurements and the determination of the ^{90}Sr activity after suitable preparation of samples.

We ask you to report the specific activity related to dry mass. The necessary correction to dry mass should be determined on separate (small) sub-samples, separate from those taken for radionuclide analysis, but taken from the bottles at about the same time as the analyzed samples. Recommended methods for determination of the water content/moisture are:
- Karl-Fischer titration (preferred), or
- drying 1 to 3 g in an oven at 102 °C ± 2 °C during 2 hours at atmospheric pressure, testing for constant mass (< 0.5 mg difference) by additional drying steps of 1 hour, according to the drying method of the IDF (International Dairy Federation).

For reporting the results to us, we will set up a WEB-based system, which has proved successful in other intercomparison exercises of our institute. Next to reporting of the measurement results, we will also require some information on the preparation and measurement methods used. We will inform you once the system is available for access.

Due to the enormous delay in sending these samples to you, for which we deeply apologize, we have to post-pone the **deadline for reporting results until 30 April 2005**.

For any further questions, I'm at your disposal.

Good luck and success with your measurements!

Kind regards,
and the Season's Greetings,

Uwe Wätjen

Retieseweg 111, B-2440 Geel, Belgium
Tel.: +32-14-571 211 - Direct line: ...-571 882.... •Fax: +32-14-584 273
Email: uwe.waetjen@cec.eu.int
http://www.irmm.jrc.be

Annex 3: Email message with clarifications

From: WAETJEN Uwe (JRC)
Sent: Monday, January 10, 2005 3:30 PM
Subject: Some clarifications EC milk powder intercomparison

Dear colleague,

I have received several questions asking for some clarifications, which I want to share with every participant:

1. The two bottles you received are taken from the same well homogenized batch of milk powder, in other words the material should be identical in composition.

The numbers on the bottles are only serial numbers from the filling process. The bottles are considered to be the **SAME sample**.

2. The reference date for all measurement results is **1 January 2005.**

3. In order to enable you to select a reasonable amount of sample from the very beginning, I want to inform you that the **specific activity of Sr-90** is of the order of 10 Bq/kg.

Finally, I would like to ask you to **confirm the receipt of the samples** to me. This serves also as an implicit verification of my list of email addresses.

With my best regards,
Uwe Waetjen

Dr. Uwe Wätjen
CEC-JRC
Institute for Reference Materials and Measurements (IRMM)
Radionuclide Metrology Sector
Retieseweg 111
B-2440 Geel, Belgium

Annex 4: Result reporting form

IMPORTANT : Disclaimer, Confidentiality Notice and rules on Privacy Protection

European Commission
Joint Research Centre
Institute for Reference Materials and Measurements

IRMM Interlaboratory Comparison

> Login > Results

Functions

Results

Result input for *REM-04*

Dr. Yana Spasova **IRMM BELGIUM** **Page 1 of 1**

Sample Code SYIZ6484728

■ **Specific activity** **Cs-137** **Optional**

■ Measurement #1

Select measurement unit from the allowable units:

◉ Bq/Kg

Result value value [ ] ± Uncertainty [ ] Coverage factor [ ] *k*

Technique used ○ [ ]

○ OTHER. Please specify, [ ]

■ Measurement #2

Select measurement unit from the allowable units:

◉ Bq/Kg

Result value value [ ] ± Uncertainty [ ] Coverage factor [ ] *k*

Technique used ○ [ ]

○ OTHER. Please specify, [ ]

■ Measurement #3

Select measurement unit from the allowable units:

◉ Bq/Kg

Result value value [ ] ± Uncertainty [ ] Coverage factor [ ] *k*

Technique used ○ [ ]

○ OTHER. Please specify, [ ]

■ Measurement #4

Select measurement unit from the allowable units:

◉ Bq/Kg

Result value value [ ] ± Uncertainty [ ] Coverage factor [ ] *k*

Technique used ○ [ ]

○ OTHER. Please specify, [ ]

■ Measurement #5

Select measurement unit from the allowable units:

◉ Bq/Kg

Result value: value [ ] ± Uncertainty [ ] Coverage factor [ ] k

Technique used: ○ [ ]

○ OTHER. Please specify, [ ]

■ Measurement #6

Select measurement unit from the allowable units:

◉ Bq/Kg

Result value: value [ ] ± Uncertainty [ ] Coverage factor [ ] k

Technique used: ○ [ ]

○ OTHER. Please specify, [ ]

■ **Specific activity** **K-40** **Optional**

■ Measurement #1

Select measurement unit from the allowable units:

◉ Bq/Kg

Result value: value [ ] ± Uncertainty [ ] Coverage factor [ ] k

Technique used: ○ [ ]

○ OTHER. Please specify, [ ]

■ Measurement #2

Select measurement unit from the allowable units:

◉ Bq/Kg

Result value: value [ ] ± Uncertainty [ ] Coverage factor [ ] k

Technique used: ○ [ ]

○ OTHER. Please specify, [ ]

■ Measurement #3

Select measurement unit from the allowable units:

◉ Bq/Kg

Result value: value [ ] ± Uncertainty [ ] Coverage factor [ ] k

Technique used: ○ [ ]

○ OTHER. Please specify, [ ]

■ Measurement #4

Select measurement unit from the allowable units:

◉ Bq/Kg

Result value: value ± Uncertainty; Coverage factor k

Technique used: ○ ○ OTHER. Please specify,

■ Measurement #5

Select measurement unit from the allowable units:

◉ Bq/Kg

Result value: value ± Uncertainty; Coverage factor k

Technique used: ○ ○ OTHER. Please specify,

■ Measurement #6

Select measurement unit from the allowable units:

◉ Bq/Kg

Result value: value ± Uncertainty; Coverage factor k

Technique used: ○ ○ OTHER. Please specify,

■ **Specific activity** **Sr-90** **Optional**

■ Measurement #1

Select measurement unit from the allowable units:

◉ Bq/Kg

Result value: value ± Uncertainty; Coverage factor k

Technique used: ○ ○ OTHER. Please specify,

■ Measurement #2

Select measurement unit from the allowable units:

◉ Bq/Kg

Result value: value ± Uncertainty; Coverage factor k

Technique used: ○ ○ OTHER. Please specify,

■ Measurement #3

Select measurement unit from the allowable units:

◉ Bq/Kg

Result value value ± Uncertainty Coverage factor k

Technique used ○ ○ OTHER. Please specify,

■ Measurement #4

Select measurement unit from the allowable units:

◉ Bq/Kg

Result value value ± Uncertainty Coverage factor k

Technique used ○ ○ OTHER. Please specify,

■ Measurement #5

Select measurement unit from the allowable units:

◉ Bq/Kg

Result value value ± Uncertainty Coverage factor k

Technique used ○ ○ OTHER. Please specify,

■ Measurement #6

Select measurement unit from the allowable units:

◉ Bq/Kg

Result value value ± Uncertainty Coverage factor k

Technique used ○ ○ OTHER. Please specify,

▲

● Save ● Clear

Submit results

This site is managed by the Informatics Unit, IRMM. Last update : 11/02/2005

Annex 5: Questionnaire

IMPORTANT : Disclaimer, Confidentiality Notice and rules on Privacy Protection

European Commission
Joint Research Centre
Institute for Reference Materials and Measurements

IRMM Interlaboratory Comparison

> Login > Results > Questionnaire

Functions

Questionnaire for *REM-04*

Results

1. Method used for moisture determination

○ Drying method: drying at 102 deg C ± 2 deg C under atmospheric pressure for 2 hr ○ Volumetric Karl Fischer titration (KFT) ○ Other

If Other, please specify

2. Moisture determination

What was the moisture content by % of the weighed sample?

What was the uncertainty of the moisture content by % of the weighed sample?

What was the applied correction factor for dry-mass?

Mass of sample used for determination of moisture content (kg)?

3. Gamma-spectrometric measurements. Was the sample chemically pre-separated before the measurements of Cs-137 and K-40?

	Reply
Yes	☐

If Yes, please give a brief description of sample preparation (you may want to send a detailed description as an e-mail attachement to Yana.SPASOVA@cec.eu.int)

No	☐

4. Was the sample physically pre-treated before the measurement of Cs-137 and K-40?

○ Yes ○ No

If Yes, please give a brief description of sample preparation (you may want to send a detailed description as an e-mail attachement to Yana.SPASOVA@cec.eu.int)

5. Geometry of measurement (you may want to send a drawing as an e-mail attachment to Yana.SPASOVA@cec.eu.int)

	Reply
Marinelli beaker	☐

If Marinelli beaker, please specify volume (litre)

Beaker	☐

If Beaker, please specify (diameter cm, height cm etc. of sample)

Pressed pellet	☐

If Pressed pellet, please specify (diameter cm, height cm, distance from detector cm etc.)

Reconstituted liquid sample	☐

If Reconstituted liquid sample, please specify

Other	☐

If Other, please specify

6. Mass of sample measured with gamma-spectrometry (kg)

7. What type of detector have you used for performing gamma-spectrometry?

	Reply
GeLi - detector	☐
HPGe - detector	☐
BEGe - detector	☐
Other	☐

If Other, please specify

8. Nominal relative efficiency (%)

9. Supplier of the detector

	Reply
Ortec	☐
Canberra	☐
Eurisys	☐
Other	☐

If Other, please specify

10. Electronics and data acquisition

	Reply
Analog signal processing (spectroscopy, amplifier etc.)	☐
Digital signal processing	☐
Other	☐

If Other, please specify

11. Data evaluation (software)

	Reply
Genie 2000 family	☐
GammaVision	☐
InterWinner	☐
Gamma-W	☐
EMCA+	☐
Gamma-track	☐
SAMPO	☐
HYPERMET	☐
Other	☐

If Other, please specify (software, peak area determination, nuclear data and equations used or you may send a detailed description as an e-mail attachment to Yana.SPASOVA@cec.eu.int)

12. Efficiency calibration of detector system for Cs-137 and K-40

	Reply
Radioactive point standards	☐
Radioactive volume standards / reference materials	☐
Radioactive liquid standards / reference materials	☐
LabSOCS	☐
Other	☐

If Other, please specify

13. Uncertainty budget: Cs-137. Here are SOME possible contributions to be entered as propagated standard uncertainty contribution to the final result. Feel free to send the complete uncertainty budget as an e-mail attachment to Yana.SPASOVA@cec.eu.int. If any considered contribution is not applicable enter 0 (zero).

In case of multiple measurement results, only state for one and indicate measurement number: #

Relative uncertainty due to moisture content, mass etc. (%):

Relative uncertainty due to sample preparation (%):

Relative counting uncertainty (%):

Relative uncertainty due to the activity of efficiency standards (%):

Relative efficiency uncertainty due to geometry etc. (%):

Possibly other uncertainty contributions (%):

Relative combined standard uncertainty u*sub c (%):

Combined standard uncertainty u sub c (Bq/kg):

Expanded uncertainty U (Bq/kg, k=2):

14. Uncertainty budget: K-40. If any considered contribution is not applicable enter 0 (zero).

In case of multiple measurement results, only state for one and indicate measurement number: #

Relative uncertainty due to moisture content, mass etc. (%):

Relative uncertainty due to sample preparation (%):

Relative counting uncertainty (%):

Relative uncertainty due to the activity of efficiency standards (%):

Relative efficiency uncertainty due to geometry etc. (%):

Possibly other uncertainty contributions (%):

Relative combined standard uncertainty u*sub c (%):

Combined standard uncertainty u sub c (Bq/kg):

Expanded uncertainty U (Bq/kg, k=2):

15. Were the intercomparison samples treated according to the same analytical procedures as routinely used in your laboratory for the same type of samples?

○ Yes ○ No

16. Routine measurements. Is the typical activity of Cs-137 (Bq/kg) in milk powder below the detection limits/decision thresholds?

○ Yes ○ No

If Yes, please give the upper limit (Bq/kg):<

If No, please specify the typical value in Bq/kg:

17. **Routine measurements. Is the typical activity of K-40 (Bq/kg) in milk powder below the detection limits/decision thresholds?**

○ Yes ○ No

If Yes, please give the upper limit (Bq/kg):<

If No, please specify the typical value in Bq/kg:

18. **Did you determine any additional radionuclide?**

○ Yes ○ No

If Yes, please supply the following information.

The name of nuclide(s) measured:

Measured value(s) (Bq/kg):

Expanded uncertainty(s) U (Bq/kg, k=2):

19. **Difficulties encountered. (if not applicable enter NONE)**

20. **Further comments on gamma-spectrometry etc. (if not applicable enter NONE)**

21. **Sr-90 determination. Mass of sample used for analytical procedure (kg).**

22. **Brief description of sample preparation (please send a detailed description as an e-mail attachment to Yana.SPASOVA@cec.eu.int).**

23. **Brief description of chemical separation (please send a detailed description as an e-mail attachment to Yana.SPASOVA@cec.eu.int).**

24. **Number of independent samples treated for Sr-90 determination.**

25. **Chemical recovery method used.**

26. **Chemical recovery (%).**

27. **Type of detection system for Sr-90 determination.**

	Reply
Wallac LSC	☐
Packard LSC	☐
Beckman LSC	☐
Other	☐

If Other, please specify

28. Type of cocktail used for Sr-90 determination in LS Counter.

	Reply
InstaGel (supplier Perkin Elmer)	☐
UltimaGold (supplier Perkin Elmer)	☐
Pico-Fluor (supplier Perkin Elmer)	☐
ReadySolv (supplier Beckman)	☐
ReadySafe (supplier Beckman)	☐
OptiPhase Hi-Safe (supplier Wallac)	☐
Other	☐

If Other, please specify

29. Determination of Sr-90

	Software of instrument supplier	Other
Data evaluation	○	○

If Other, please specify (software, half-life and equations used)

Briefly describe the efficiency calibration of detection system:

Blank and background measurement (CPM):

Acquisition time per sample (sec):

Measurement cycles:

30. Uncertainty budget: Sr-90. Here are SOME possible contributions to be entered as propagated standard uncertainty contribution to the final result. Feel free to send the complete uncertainty budget as an e-mail attachment to Yana.SPASOVA@cec.eu.int. If any considered contribution is not applicable enter 0 (zero).

In case of multiple measurement results, only state for one and indicate measurement number: #

Relative uncertainty due to moisture content, mass etc. (%):

Relative uncertainty due to sample preparation, separation etc. (%):

Relative counting uncertainty (%):

Relative uncertainty due to the activity of efficiency standards (%):

Relative uncertainty in efficiency (%):

Relative uncertainty due to blank and background measurement (%):

Possibly other uncertainty contributions (%):

Relative combined standard uncertainty u*sub c (%):

Combined standard uncertainty u sub c (Bq/kg):

Expanded uncertainty U (Bq/kg, k=2):

31. Were the intercomparison samples treated according to the same analytical procedures as routinely used in your laboratory for the same type of samples?

○ Yes ○ No

32. Routine measurements. Is the typical activity of Sr-90 (Bq/kg) in milk powder below the detection limits/decision thresholds?

○ Yes ○ No

If Yes, please give the upper limit (Bq/kg):<

If No, please specify the typical value in Bq/kg:

33. Difficulties encountered. (if not applicable enter NONE)

34. Further comments on Sr-90 determination. (if not applicable enter NONE)

35. Other comments on this intercomparison exercise. (if not applicable enter NONE)

36. Questionnaire completed by:

Name

Position

European Commission

EUR 23270 EN – Joint Research Centre – Institute for Reference Materials and Measurements
Title: Evaluation of EC measurement comparison for ^{137}Cs, ^{40}K and ^{90}Sr in milk powder
Authors: U. Wätjen, Y. Spasova, T. Altzitzoglou, H. Emteborg and S. Pommé
Luxembourg: Office for Official Publications of the European Communities
2008 – 77 pp. – 21.0 x 29.7 cm
EUR – Scientific and Technical Research series – ISSN 1018-5593
ISBN 978-92-79-08500-0
DOI 10.2787/32777

Abstract
This report describes all details of the measurement comparison for ^{137}Cs, ^{40}K and ^{90}Sr in milk powder among 60 European laboratories monitoring radioactivity in food and the environment. An available IAEA reference material was re-processed at IRMM into suitable intercomparison samples and the homogeneity of the distributed samples together with other quality parameters was determined. Reference values of the three radionuclides under study in this intercomparison were determined at IRMM using tracer techniques and standardised radionuclide solutions and are thus traceable to the SI units. The sample preparation and measurement processes applied in the participating laboratories are described and the results of the intercomparison are presented and discussed in detail. Whereas, in general, the measurement results for ^{137}Cs and ^{40}K show good agreement with the reference value, the results of this comparison point at problems of ^{90}Sr determination in about one third of the laboratories. The corresponding participants should investigate and revise their analytical methods, next to many laboratories needing to improve their estimation of measurement uncertainty.

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