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Evaluation of EC Interlaboratory Comparison on Radionuclides in Soil

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Summary

In 2010, an interlaboratory comparison (ILC) on radionuclides in soil within the framework of the Euratom Treaty was conducted among 75 laboratories monitoring radioactivity in the environment and foodstuff. The performance of the laboratories in determining activity concentrations of up to 15 natural and anthropogenic radionuclides (^{40}K, ^{90}Sr, ^{137}Cs, ^{212}Pb, ^{212}Bi, ^{214}Pb, ^{214}Bi, ^{226}Ra, ^{230}Th, ^{232}Th, ^{234}U, ^{235}U, ^{238}U, ^{238}Pu, and $^{239+240}$Pu) were compared.

The certified reference material IAEA-375 Soil (originating from the area affected by the Chernobyl accident) was used in this ILC as basis for the testing material. It was reprocessed at IRMM, further homogenized and rebottled, such that its origin was not visible to the participants. Reference values traceable to the International System of Units (SI) and the International Reference System for gamma-ray emitting radionuclides (SIR) were determined at IRMM, and the homogeneity of the material was demonstrated.

The methods of the sample preparation and measurement techniques used by the participating laboratories are described. A robust evaluation of the performance of laboratories is performed using three different approaches: relative deviations, E_n numbers and PomPlots.

The performance of the participating laboratories varied depending on the radionuclide determined and method used. Gamma-ray spectrometry with respect to ^{137}Cs and ^{40}K is well controlled. The determination of ^{90}Sr proved difficult for about two-thirds of the participants, who submitted results outside the acceptable range. Several laboratories need to improve their analytical procedures for the uranium isotopes and ^{226}Ra. Moreover, the results for thorium isotopes are far from satisfactory mainly for the ^{230}Th.

The evaluation based on the E_n criterion revealed that the uncertainty estimation is poor in many laboratories and there is a need to improve their application of uncertainty propagation. A few laboratories were identified as highly unreliable and their measurement routines should be promptly revised.

Contents

Summary ... 3
Contents ... 5
Glossary ... 6
1 Introduction ... 7
  1.1 Reporting of the results ... 8
  1.2 Participating laboratories ... 8
  1.3 Questionnaire ... 9
2 Reference values ... 10
  2.1 Test material ... 10
  2.2 Measurements of ^{40}K and ^{137}Cs by gamma-ray spectrometry ... 12
  2.3 Measurements of ^{90}Sr by liquid scintillation counting ... 15
  2.4 Measurements of ^{226}Ra by alpha-particle spectrometry ... 16
  2.5 Measurements of actinides by alpha-particle spectrometry ... 18
  2.6 Homogeneity measurements ... 24
3 Methods used by the participating laboratories ... 29
  3.1 Water content determination ... 29
  3.2 Gamma-ray spectrometry: ^{137}Cs, ^{40}K, ^{212}Pb, ^{212}Bi, ^{214}Pb, and ^{214}Bi ... 29
  3.3 Chemical separation and source preparation: ^{230}Th and ^{232}Th ... 31
  3.4 Chemical separation and source preparation: ^{234}U, ^{235}U, ^{238}U ... 32
  3.5 Chemical separation and source preparation: ^{238}Pu, $^{239+240}$Pu ... 33
  3.6 Determination of ^{226}Ra ... 33
  3.7 Alpha-particle spectrometry ... 34
  3.8 Determination of ^{90}Sr ... 34
4 Reported results ... 37
  4.1 Data treatment ... 37
  4.2 Uncertainty budgets ... 46
  4.3 Additional radionuclides ... 47
  4.4 Participants' comments ... 49
5 Evaluation and comparison of data ... 51
  5.1 Identification of outliers and normal distribution check ... 51
  5.2 Scores and evaluation criteria ... 58
  5.3 Evaluation of laboratory performances ... 60
  5.4 Comparison of different types of laboratories ... 87
6 Conclusions ... 99
Acknowledgements ... 100
References ... 101
Appendix 1: Invitation letter ... 104
Appendix 2: Information letter ... 105
Appendix 3: Registration (e-mail) ... 106
Appendix 4: Registration guidelines ... 107
Appendix 5: Reporting (e-mail) ... 109
Appendix 6: Reporting guidelines ... 110
Appendix 7: Communication on a preliminary results ... 112
Appendix 8: List of participating laboratories ... 116
Appendix 9: Questionnaire ... 126
Appendix 10: Results, methods and scores of laboratories ... 130
Appendix 11: Additional radionuclides ... 151

Glossary

AC	accession countries
A_{lab}	mean laboratory result of activity concentration
A_{ref}	reference value of activity concentration
BIPM	Bureau International des Poids et Mesures
CCRI(II)	Comité Consultatif des Rayonnements Ionisants, Section 2
CRP	co-ordinated research project
D	difference between the reported and the reference activity concentration
$\mathrm{E_n}$	performance statistic $\mathrm{E_n}$ number
Euratom	European Atomic Energy Community
GM	Geiger-Müller counting
GUM	Guide to the Expression of Uncertainty in Measurement
HPGe	high-purity germanium detector
IRMM	Institute for Reference Materials and Measurements
IAEA	International Atomic Energy Agency
ICP-MS	inductively coupled plasma mass spectrometry
ILC	interlaboratory comparison
ISO	International Organization for Standardization
k	coverage factor according to GUM
LOD	limit of detection
LSC	liquid scintillation counter, liquid scintillation counting
MAD	median absolute deviation
MS	member states
NIM	nuclear instrument module
SI	Système International d'Unités, International System of Units
SIR	Système International de Référence, International Reference System for radionuclides
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 average laboratory result
U_{ref}	expanded uncertainty of reference value

1 Introduction

Within the framework of the European Atomic Energy Community (Euratom) Treaty the European Union member states (MS) are obliged to perform measurements of the radioactivity in their environment and to report the results to the European Commission (EC). To verify the performance of monitoring laboratories and to ensure the comparability of reported results regular interlaboratory comparisons (ILCs) were introduced by the EC. Since 2003, the JRC Institute for Reference Materials and Measurements (IRMM) has had the responsibility for their organization.

The metrological approach of IRMM in conducting comparisons relies on its participation in key comparisons among National Metrology Institutes (Wätjen et al., 2008) as shown in Fig. 1. This allows IRMM to work with intercomparison samples for which it determines the reference values traceable to SI units and the International reference System (SIR) for gamma-ray emitting radionuclides (Ratel, 2007). In terms of physical properties as well as radioactivity concentrations, the IRMM intercomparison samples are generally closer to the real samples measured in monitoring laboratories than calibration standards. Therefore, they offer a realistic estimate of the performance of these laboratories in their monitoring tasks.

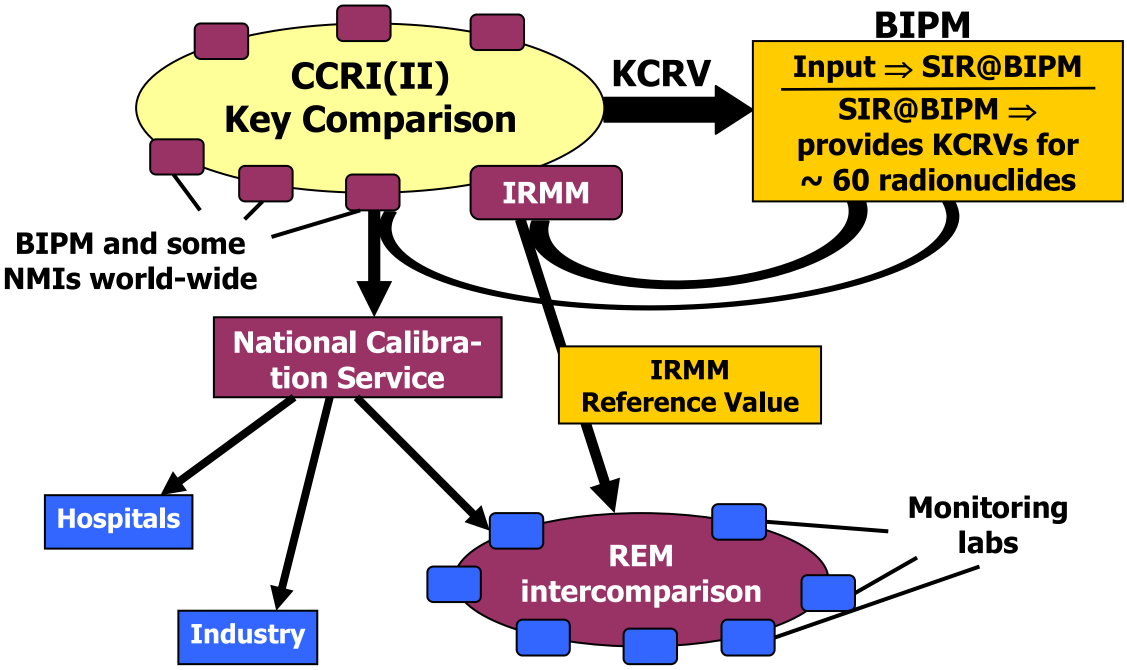

Fig. 1. Key comparisons of CCRI(II) and traceability of the reference values for samples provided by IRMM for the intercomparisons amongst monitoring laboratories (KCRV = key comparison reference value).

The aim of this ILC was to investigate the abilities of monitoring laboratories to measure a wide range of radionuclides in soil. It was organised according to the agreement at the national expert meeting under the Euratom Treaty Art. 35/36 on 12-13 October 2009 at JRC-Ispra. Depending on the participating laboratory's capabilities, the determination of activity concentrations in soil material was requested for 15 radionuclides: ^{40}K, ^{90}Sr, ^{137}Cs, ^{212}Pb, ^{212}Bi, ^{214}Pb, ^{214}Bi, ^{226}Ra, ^{230}Th, ^{232}Th, ^{234}U, ^{235}U, ^{238}U, ^{238}Pu, and $^{239+240}$Pu. The certified reference material IAEA-375 Soil was used as a base material for the comparison samples.

This report describes in detail all phases of the ILC organised in 2010. It starts with the determination of the reference values and the demonstration of the material homogeneity. Afterwards, the analytical methods used at the laboratories are described. Next, the treatment of the data reported by the participants is described and, finally, the participants' results are evaluated and compared. A robust evaluation of the performance of laboratories is performed using three different approaches: relative deviations, E_n numbers (ISO, 2005a) and PomPlots (Spasova et al., 2007).

1.1 Reporting of the results

All results of activity concentrations must be reported normalised to dry mass ($Bq \cdot kg^{-1}$) with the associated expanded uncertainty U ($U = k \cdot u_c$, 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) subsamples. These should be taken from the bottles at about the same time as the samples for radionuclide analysis to be representative for their water content.

For those radionuclides requiring chemical preparation we requested that a total digestion of the samples is performed, except for the plutonium isotopes. For the plutonium analysis an aqua regia leach was considered to be sufficient since these radionuclides are expected to originate from the fallout.

The reference date for all results was 1 January 2010. As source of nuclear decay data, the Monographie BIPM-5 (2004 and 2006) was recommended.

Timetable of ILC:

Dec 2009	invitation letter (Appendix 1) sent to the national representatives
Jan 2010	laboratories are nominated by the national representatives
Mar 2010	soil samples are sent to the participants via express mail (DHL) together with the information on the ILC (Appendix 2)
Jun 2010	on-line reporting system is set up according to the requirements of the exercise (Appendices 3-6)
Aug 2010	laboratories submit their results to IRMM
Sep 2010	laboratories submit their questionnaire to IRMM
Nov 2010	preliminary results sent to participants (Appendix 7)

1.2 Participating laboratories

The participating laboratories were mainly national research institutes, authorities and monitoring laboratories. The national representatives in the expert group according to the Euratom Treaty Art. 35/36 nominated 50 laboratories from EU member states (MS). Traditionally, also other European counties (7 from pre-accession countries (AC) and 2 from Switzerland) were invited by IRMM to participate in the ILC. In addition, 18 laboratories worldwide were nominated by the International Atomic Energy Agency (IAEA). One of the MS nominated laboratories declined to participate due to work overload. This laboratory is not mentioned in the list of participating laboratories.

In total 75 laboratories (49 from MS, 7 from AC, 2 from Switzerland, 17 IAEA) registered for the participation in the exercise and 73 laboratories finally reported results. Table 1 gives an overview of the numbers of laboratories. The list of all participating laboratories is shown in Appendix 8. Since the anonymity is a requirement in this programme of ILC, the identity of the laboratories is not shown in the compilation and evaluation of the results. The order of the listing of participants in Appendix 8 is not the same as the laboratory number used throughout the data evaluation in this report.

Table 1. Overview of the numbers of participating laboratories

	MS	AC + Switzerland	IAEA	Total
Nominated	50	9	18	77
Registered	49	9	17	75
Reported	49	9	15	73

1.3 Questionnaire

Participants were asked to fill in a questionnaire (Appendix 9). It was composed of seven parts concerning the information on the laboratory, its routine measurements, determination of water content, sample treatment, measurement methods, uncertainty budgets and some additional information. Information in the questionnaire is essential in order to evaluate the results of the intercomparison. Although, the laboratories were urged several times to submit the questionnaire, 9 out of 73 participating laboratories (labs: 4, 19, 23, 26, 40, 49, 51, 54, 68) did not do so.

2 Reference values

2.1 Test material

Top soil to a depth of 20 cm was collected on the field of the collective farm “Staryi Vishkov”, Novozybkov district, Bryansk region, Russia, in July 1990 (IAEA, 1996). This region was affected by the Chernobyl nuclear power plant accident in 1986. Therefore, the levels of anthropogenic radioactivity are elevated. It is important to note, that the material was not spiked in the laboratory.

The dried soil material of approximately 500 kg was milled in the Brjansk Centre of Agricultural Radiology and Chemicalization. Then it was sieved (0.3 mm) and filled into 25 polyethylene bags, all from the same batch process. Afterwards, the bulk material was homogenized in IAEA's laboratories in Seibersdorf, Austria, distributed in plastic bottles and sterilized by gamma-ray irradiation to a total dose of 25 kGy using a ^{60}Co source. The homogeneity of the material was tested on randomly selected bottles by measuring uranium in 200 mg aliquots, ^{134}Cs, ^{137}Cs and ^{90}Sr in about 6 g aliquots. It was shown by IAEA that the variance between samples (7 bottles) does not differ significantly from the variance within samples (3 determinations in each bottle) at the level of significance $\alpha = 0.05$. Therefore, the material was considered sufficiently homogeneous. However, there is evidence for the presence of small hot particles which can seriously influence the measured activity concentration of transuranium elements (no reference values for plutonium isotopes were finally used).

Fig. 2. Intercomparison material after reprocessing and rebottling in IRMM.

The soil material is known as reference material IAEA-375 (IAEA, 2000). The recommended values (^{40}K, ^{90}Sr, ^{106}Ru, ^{125}Sb, ^{129}I, ^{134}Cs, ^{137}Cs, ^{226}Ra, ^{232}Th) were established on the basis of results of an ILC organized by IAEA during 1992-93. They represent overall mean values (excluding outliers) calculated on the basis of at least 10 laboratory averages. The information values (^{228}Th, ^{234}U, ^{238}U, ^{238}Pu, $^{239+240}$Pu, ^{241}Am) were calculated on the basis of at least 5 laboratory averages. However, these values are based only on statistical analysis and are not directly traceable to the SI units.

Therefore, IAEA started a co-ordinated research project (CRP) with the aim to upgrade the entitled intercomparison materials to reference materials with assigned property values traceable to the SI. IRMM participated in this CRP. The full traceability was ensured by means of calibrated standard weights, standard calibration sources for gamma-ray and alpha-particle spectrometry, the Extended SIR for liquid scintillation counting and the use of standard time. These new – traceable – activity concentrations were used as reference values for the purposes of this soil ILC.

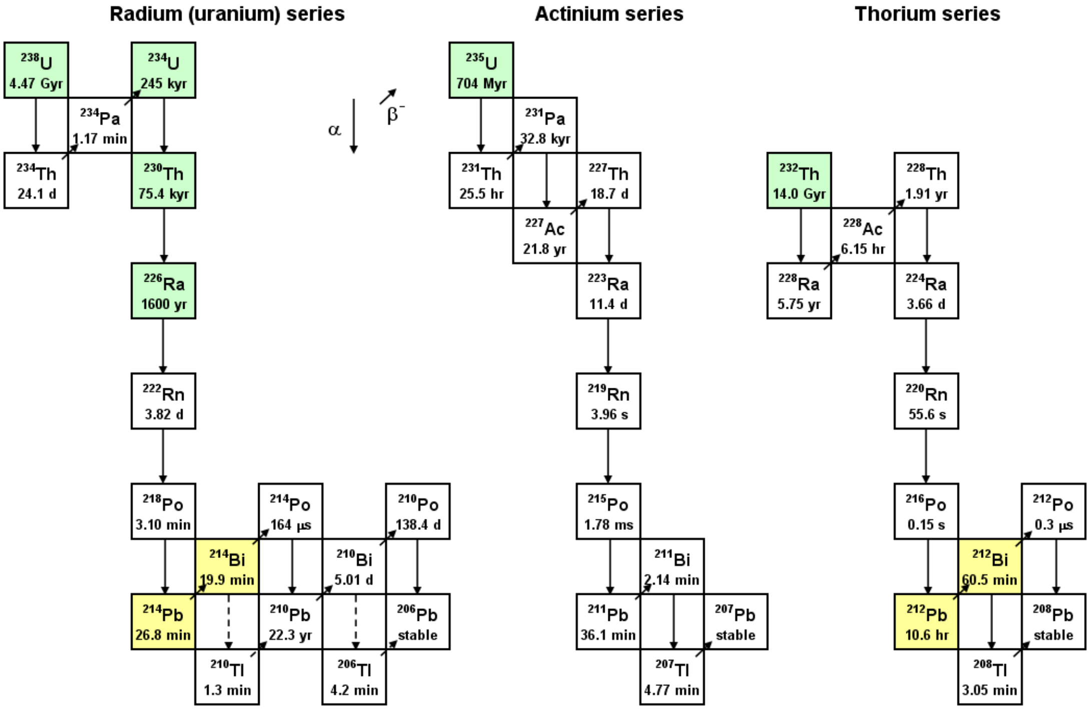

Fig. 3. Radium (uranium), actinium and thorium series. Green colour indicates radionuclides for which reference values were determined, yellow colour indicates radionuclides with indicative values.

At IRMM the material received from IAEA was reprocessed by drying, mixing (Dyna Mix CM200 mixer during 2 hours), and filled in small units of approximately 250 g into 280 mL amber glass bottles (Fig. 2). This way, the material was further homogenised, and – as a side effect – the origin of the material (IAEA-375) could be concealed from the participants. The sample for water content determination was taken and analysed with a Sartorius MA150 moisture analyser (Karl-Fischer titration). The water content was low and varied from 3.33 to 3.91 g/100 g. The mean value from all measurements was 3.49 g/100 g, with a standard deviation of 0.13 g/100 g.

The activity concentrations were determined for primordial ^{40}K; four members of the radium (uranium) series ^{238}U, ^{234}U, ^{226}Ra and ^{230}Th; following ^{232}Th and ^{235}U from the thorium and actinium series (Fig. 3), respectively; and anthropogenic ^{90}Sr and ^{137}Cs. The results were published in Altzitzoglou et al. (2006). Some details from this CRP report are presented in the following chapters (2.2 – 2.5). For ^{212}Pb, ^{212}Bi, ^{214}Pb, and ^{214}Bi, the activity concentrations of their mother radionuclides (^{232}Th and ^{226}Ra) were used as indicative values. For the plutonium isotopes no reference values were established due to an apparent presence of hot particles.

2.2 Measurements of ^{40}K and ^{137}Cs by gamma-ray spectrometry

High-resolution gamma-ray spectrometry was used to determine the activity concentrations of the radionuclides ^{40}K and ^{137}Cs. Since the samples are measured without any prior chemical treatment, the source preparation is very rapid and it does not alter the original sample. Therefore, before any chemical manipulation, the samples were measured by this method. These measurements were realized in 2006 (Altzitzoglou et al., 2006).

Sample preparation

The soil sample was shaken vigorously, using a 3D Turbula mixer (type T2C, Willy A. Bachofen AG Maschinenfabrik, Basel, Switzerland) and was dried in an oven for 48 hours at 105 °C to constant weight. After cooling to room temperature in a desiccator, an amount of the soil was transferred to a cylindrical container (125 mL polypropylene, Nalgene, USA) and weighed, using an analytical balance (type 1712, Sartorius GmbH, Göttingen, Germany). This balance is calibrated with a standard weight set (Weight set Mettler Toledo M7), traceable to the IRMM kilogram, which is directly traceable to the BIPM kilogram by regular direct comparisons. Two samples were prepared from each bottle. The sample mass of the soil ranged from 39.4 to 42.0 gram. The containers were tapped 5000 times, using a tapping machine (Dual Autotap, model DA-1, QuantaChrome, Syosset, N.Y., USA) and then placed directly on top of the detector end-cap and measured for 4 to 12 days each.

Measurement equipment

A high-purity germanium (HPGe) detector system was used for the measurements. The co-axial detector (CANBERRA GC3518-7500SL S/N b93106, Canberra Eurisys Benelux N.V., AREVA Group, Zellik, Belgium) consisted of a HPGe crystal, 58.5 mm in diameter and 53.5 mm in length, with 36 % relative efficiency and an aluminium end-cap window. The detector was housed in a 10 cm thick Pb shield of circular intersection, lined with 1 mm Cd and 1 mm Cu. The inner 2 cm of the Pb shield was made of highly radio-pure Pb.

The pulse processing electronic setup consisted of a multichannel analyser (MCA) and a set of scalers. The signal was digitised after amplification and the pulse height spectrum was acquired by a National Instruments PCIDIO-32HS 32-bit, high-speed parallel digital I/O interface (National Instruments, Austin, Texas, USA). In addition, the dead-time output signals from the amplifier and the analog-to-digital converter (ADC) were processed by a live-time clock gate (CBNM model LGN 7732), which processed the system clock pulses and establishes the live-time of the measurement. A very stable quartz oscillator with a frequency of 100 kHz provided the time base of the live-time clock gates. As a time base the legal time in Germany on the basis of Coordinated World Time (UTC) generated at the Physikalisch-Technische Bundesanstalt (PTB, Braunschweig, Germany) by caesium atomic clocks utilised as primary standard of time and frequency and broadcasted through the LF transmitter DCF77 was used. A computer-resident National Instruments PXI-6602 timing and digital I/O module with eight 32-bit counters was used to accumulate the event pulses and clock pulses generated in the system.

Efficiency calibration

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, actual samples were spiked with known amounts of standard ^{54}Mn, ^{60}Co, ^{65}Zn, ^{137}Cs and ^{241}Am solutions, mixed thoroughly and measured. In this way information on the matrix self-absorption of the soil material was obtained. The final efficiency calibration was based on the calculation of the efficiency using the Monte Carlo computer code GEOLEP (Solé, 1990; Lépy, 2000). The geometry and detector parameters necessary for the program were adjusted, so that the output of the calculation matches the experimental results obtained with the standard point sources, the liquid standards and the spiked soil. The efficiencies calculated using GEOLEP agreed within less than 2.2 % with the experimentally measured efficiencies.

Measurement reproducibility

The measurement reproducibility was tested by placing a sample in front of the detector and performing the measurement and by re-positioning the sample in front of the detector and repeating the measurement. The former tested the statistical reproducibility, while the latter tested in addition the geometrical (repositioning) repeatability. The standard deviation for the statistical reproducibility was 0.15 % for 6 one-day long measurements and that for the geometrical repeatability was 0.23 % for 2 one-day long measurements. The latter includes the statistical reproducibility as well. In the uncertainty budget a contribution of 0.2 % is included for the geometrical repeatability.

Calculations and results

The measured data either for the efficiency or the activity determination were corrected for background, decay and decay during measurement. The activity concentration values for both radionuclides and for each sample are presented in Fig. 4-5. In addition, the information on weighted mean, absolute and relative standard deviations are presented in the figures. Statistical tests were applied to check the results for consistency. The tests utilised 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 these tests. The uncertainty budgets for the typical single measurement of a sample at the 1σ level ($k = 1$) are given in Table 2.

Table 2. Uncertainty budgets for ^{137}Cs and ^{40}K assessed in soil by gamma-ray spectrometry show the typical uncertainties for a single measurement of a sample at the 1σ level. The combined uncertainty is the quadratic sum of all components ($k = 1$).

Component	^{137}Cs (%)	^{40}K (%)
Counting statistics (incl. background)	0.01	0.15
Weighing	0.02	0.02
Geometry repeatability	0.2	0.2
Dead time	0.005	0.005
Detection Efficiency	2.2	2.2
Gamma-ray emission prob.	0.235	1
Timing	0.005	0.005
Half-life	0.14	0.001
Combined uncertainty	2.2	2.4

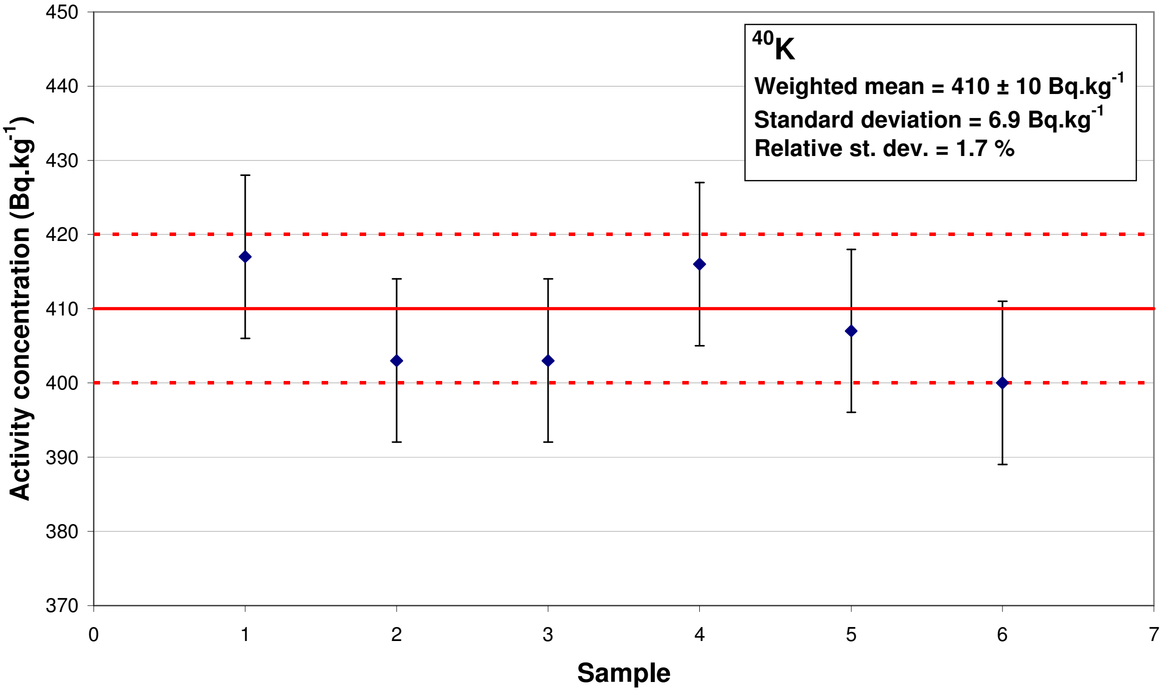

Fig. 4. Activity concentration results of ^{40}K in the soil. The activity values are calculated for the reference date 31 December 1991. All uncertainties are combined uncertainties at the 1σ level ($k = 1$). The solid red line indicates the weighted mean and the dashed red lines the combined uncertainty of the mean ($k = 1$).

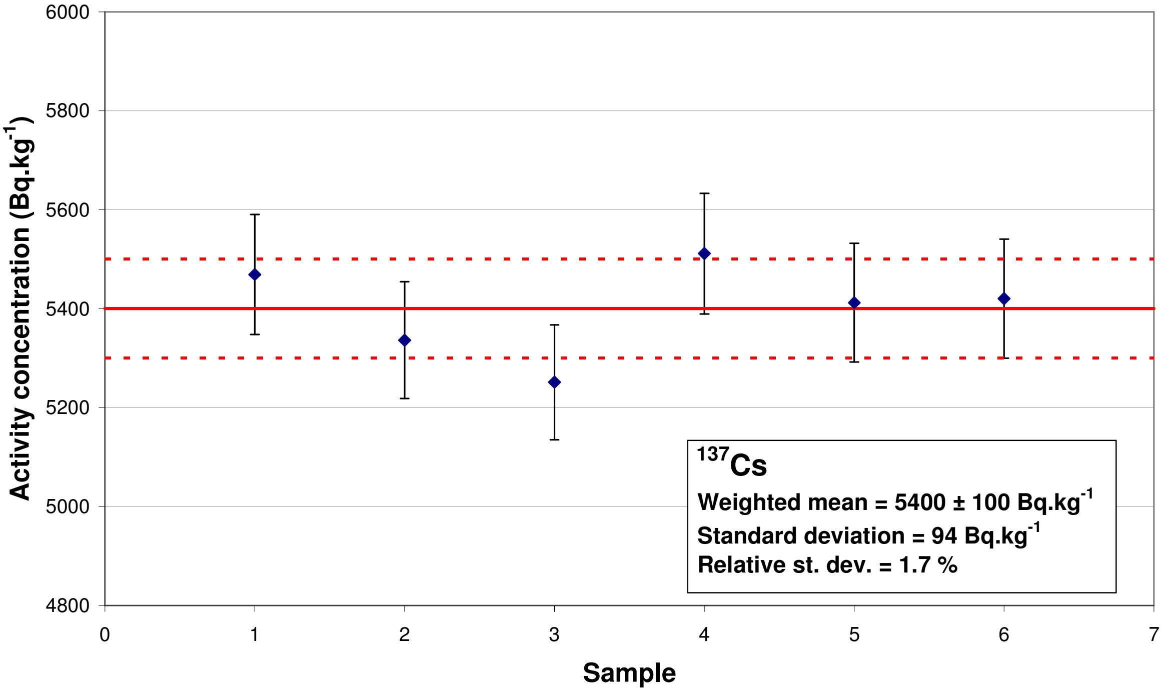

Fig. 5. Activity concentration results of ^{137}Cs in the soil. The activity values are calculated for the reference date 31 December 1991. All uncertainties are combined uncertainties at the 1σ level ($k = 1$). The solid red line indicates the weighted mean and the dashed red lines the combined uncertainty of the mean ($k = 1$).

2.3 Measurements of ^{90}Sr by liquid scintillation counting

The measurement of ^{90}Sr requires Sr to be separated from the matrix and from other interfering radionuclides. The method we applied is 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).

Sample preparation

The material was first dried in an oven at 105 °C to constant weight. The mass of each sample was determined gravimetrically, using an analytical balance (model AT21, Mettler-Toledo, Greifensee, Switzerland), calibrated with standard weights (Weight set Mettler Toledo M7) traceable to the IRMM kilogram. Sample masses of the order of 5 g were used.

The soil was heated to 200 °C for one hour and combusted for at least 4 hours at 550 °C to reduce its mass before the microwave digestion. The sample mass reduction after ashing was about 10 %. After adding the tracer (^{85}Sr) for the chemical recovery determination, wet digestion with concentrated nitric/hydrofluoric acids and hydrogen peroxide was performed with a Mars 5 Digestion System (CEM Corp., Matthews, NC, USA). At the end the solution was visually clear and ready for the chemical separation. The eluent of the group separation (see chapter 2.4) contained Sr and Ra. This solution was passed through an extraction chromatography Eichrom Sr Resin (Eichrom Technologies, Inc., Darien, Il., USA) to obtain a pure Sr fraction. The final Sr eluate was evaporated and the residue taken up by 6 mL 0.05N HNO_3 into a scintillation vial (20 mL High-Performance Packard vial, Perkin Elmer, Boston, MA, USA) containing 14 mL of Insta-Gel Plus LS cocktail (Perkin Elmer, Boston, MA, USA) to be measured by LSC.

Measurement equipment and corrections

The assessment of ^{90}Sr was performed by measuring the samples using a Wallac Quantulus 1220 (Perkin Elmer, Boston, MA, USA) ultra low-level liquid scintillation spectrometer. The samples were measured immediately after the separation of Sr and several times later, with blanks introduced before and after each sample measurement. The blanks were prepared by adding 6 mL 0.05N HNO_3 into 14 mL of Insta-Gel Plus LS cocktail. The data reduction and analysis included the background subtraction, decay correction, decay during measurement correction, correction for the contribution of the tracer (^{85}Sr) and the ingrowth of ^{90}Y. Since the sample went through digestion and chemical separation, in order to isolate the strontium, a tracer for the chemical recovery calculation was used. It was opted for ^{85}Sr, which was then measured by gamma-ray spectrometry and the chemical recovery was calculated as the ratio of the counts under the 514 keV gamma-ray peak of the sample to that of a reference source (in the same geometry).

Efficiency calibration and results

For the instrument efficiency calibration, the CIEMAT/NIST ^{3}H efficiency tracing method (Grau Malonda and Garcia-Toraño, 1982; Grau Malonda et al., 1985) was used, requiring ^{3}H standards only. The ^{90}Sr activity concentration values obtained are given in Fig. 6 together with the weighted mean and standard deviation. The typical uncertainty budget for a single measurement of a sample at the 1σ level ($k = 1$) is presented in Table 3.

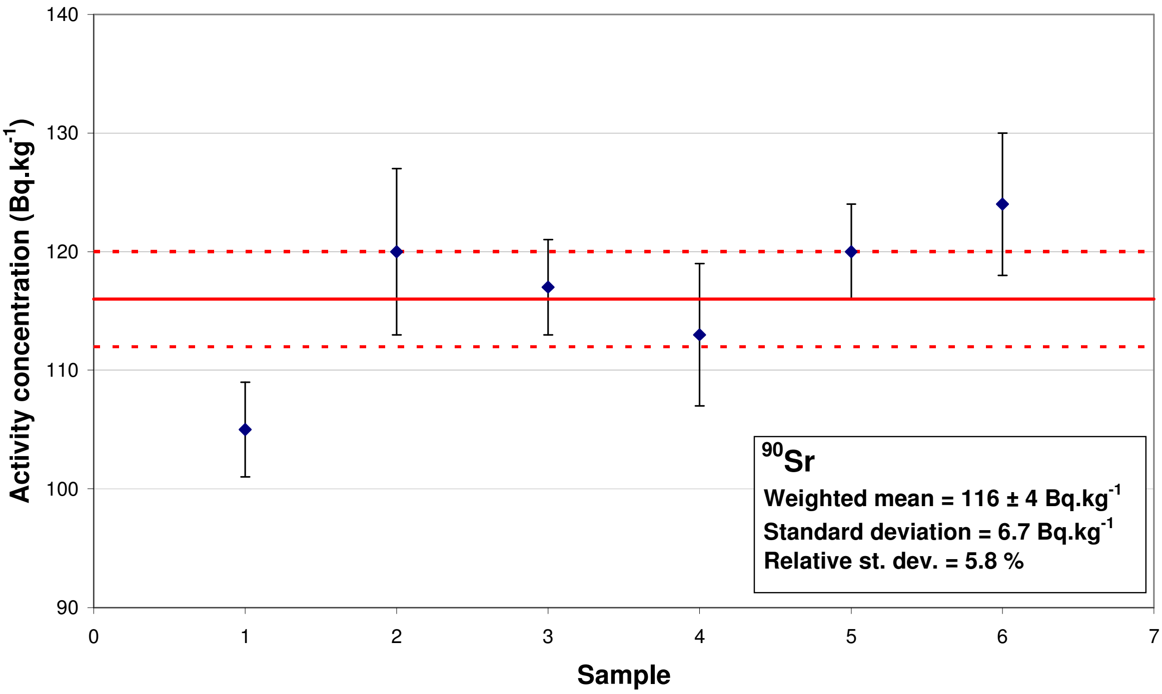

Fig. 6. Activity concentration results of ^{90}Sr in the soil. The activity values are calculated for the reference date 31 December 1991. All uncertainties are combined uncertainties at the 1σ level ($k = 1$). The solid red line indicates the weighted mean and the dashed red lines the combined uncertainty of the mean ($k = 1$).

Table 3. Uncertainty budget for ^{90}Sr assessed in soil by LSC shows the typical uncertainties for a single measurement of a sample at the 1σ level. The combined uncertainty is the quadratic sum of all components ($k = 1$).

Component	^{90}Sr (%)
Counting statistics (incl. background)	1.5
Weighing	0.2
Dead time	0.05
Chemical recovery	3.5
Timing	0.05
Ratio ^{90}Y/^{90}Sr	0.1
Efficiency (incl. quenching and interpolation from curve)	1.0
Half-life	0.11
Sample stability	0.1
Combined uncertainty	3.9

2.4 Measurements of ^{226}Ra by alpha-particle spectrometry

The method, developed and validated (Decaillon et al., 2004) for ^{226}Ra measurement, is based on microwave digestion, separation by extraction and ion exchange chromatography and co-precipitation. The main problem was the large amount of barium present in the soil. That dictated the use of small amounts of sample to avoid the preparation of bulky sources and, therefore, high self-absorption in the alpha sources.

Sample preparation

The material was first dried in an oven at 105 °C to constant weight. The mass of each sample was determined gravimetrically, using an analytical balance (model AX504, Mettler-Toledo, Greifensee, Switzerland) calibrated with standard weights. Sample amounts of the order of 1.3 g were used. Then, the soil was heated to 200 °C for one hour and combusted for at least 4 hours at 550 °C in order to reduce its mass before the microwave digestion or the sample leach. The sample mass reduction after ashing was of the order of 10 %. To determine the chemical recovery of Ra, ^{133}Ba tracer was added at this stage. Barium, as alkaline earth, is assumed to have the same chemical behaviour as radium (Lozano et al., 1997; Baeza et al., 1998). In addition, ^{133}Ba as gamma-ray emitter is easily measured. The digestion was performed on a Mars 5 Digestion System (CEM Corp., Matthews, NC, USA). Afterwards, the solution was visually clear and ready for the chemical separation.

The digested sample was passed through a pre-packed TRU column (Eichrom Technologies, Inc., Darien, Il., USA) in order to separate the Ra and Sr from the actinides. The eluate was evaporated and brought to 0.5N HCl, before loading it onto a Bio-Rad AG 50W-X8 column (Bio-Rad Laboratories, Hercules, CA, USA) to remove the alkaline metal ions. The column was rinsed with 100 mL 1.5N HCl and the barium-radium fraction was eluted with 60 mL 8N HNO_3. The eluate was evaporated to dryness, brought to 0.1N HNO_3 and the co-precipitation of Ra with Ba (as $Ba(Ra)SO_4$) was performed.

Measurement equipment

For the acquisition of the alpha-particle spectra the Canberra Model 7401 VR (Canberra, Meriden, CT, USA) system was used. A stainless steel shelf and sample holder are included with each spectrometer for reproducible detector-to-sample positioning. A Canberra Passivated Implanted Planar Silicon (PIPS) detector was used with an active area of 450 mm^2. Several 7401 alpha spectrometers were supported by a single multichannel analyser through a Canberra multiplexer and the Genie 2000 software (Canberra, Meriden, CT, USA) was controlling the acquisition of the alpha-particle spectra.

Efficiency calibration and measurements

Two sources were used for the efficiency calibration of the alpha-particle detection system: the Pu239-1515 electro-deposited ^{239}Pu source and the Am241-1299 co-precipitated ^{241}Am source. Both sources were measured by alpha-particle counting at defined solid angle, a primary method, to assess their activities with values traceable to the SI units (IRMM certificate 20040810). By calibrating the system this way the measurement and the calibration sample geometry were kept as similar as possible.

Before each measurement, the performance of the instrument was controlled with the previously mentioned ^{239}Pu source. The acquisition time for the ^{226}Ra measurements ranged from 10 to 17 days. In the activity calculations, the ^{226}Ra decay was neglected because of its long half-life (1600 a). The chemical recovery of Ra was assumed to be equal to that of barium which was derived from gamma-ray spectrometric measurements of ^{133}Ba.

Results

The activity concentration of ^{226}Ra was measured in 6 samples from 6 different bottles of the IAEA-375 soil and the results are presented in Fig. 7. The contribution of the uncertainties on the emission probability and the decay of ^{226}Ra were considered negligible and were not included in the uncertainty budget (Table 4). Statistical tests were applied to check the results for consistency. The tests utilised were Dixon's, Grubb's, coefficient of skewness and coefficient of kurtosis. No outliers were found in any of the activity concentration results. The chemical recoveries for radium in the soil were high and ranged from 74 % to 85 %.

Table 4. Uncertainty budget for ^{226}Ra assessed in soil by alpha-particle spectrometry shows the typical uncertainties for a single measurement of a sample at the 1σ level. The combined uncertainty is the quadratic sum of all components (k = 1).

Component	^{226}Ra (%)
Counting statistics (incl. background)	1.5
Weighing	0.3
Dead time	0.05
Chemical recovery	3.9
Timing	0.05
Efficiency	1.1
Combined uncertainty	4.3

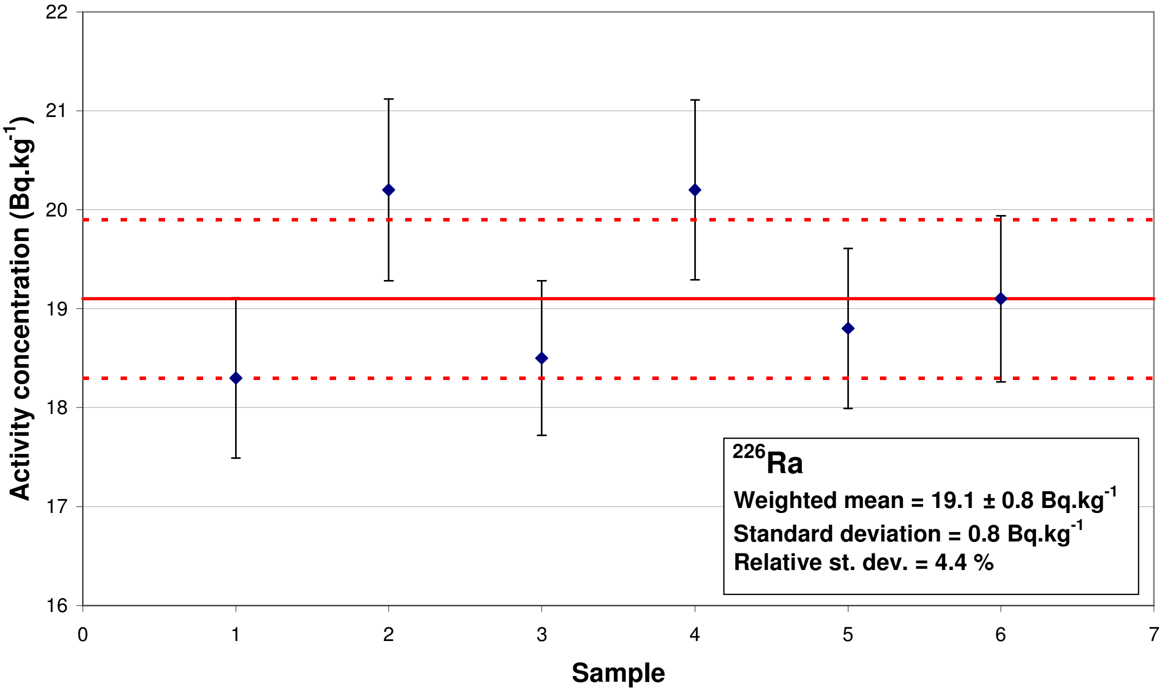

Fig. 7. Activity concentration results of ^{226}Ra in the soil. The activity values are calculated for the reference date 31 December 1991. All uncertainties are combined uncertainties at the 1σ level (k = 1). The solid red line indicates the weighted mean and the dashed red lines the combined uncertainty of the mean (k = 1).

2.5 Measurements of actinides by alpha-particle spectrometry

In the frame of the IAEA-CRP, the traceable determination of the radionuclides ^{230}Th, ^{232}Th, ^{235}U, ^{234}U and ^{238}U in the IAEA-375 soil was performed. The method used in this work, previously developed and validated (Pilviö and Bickel, 1998; Pilviö et al., 1999; Hill et al., 2004), is based on microwave digestion, separation by extraction chromatography and co-precipitation. The quantitative transfer of the analyte from the matrix into solution, the separation of the U and Th from the matrix, and the preparation of sources suitable for alpha-particle spectrometry is described briefly.

Sample preparation and equipment

The material was first dried in an oven at 105 °C to constant weight. The mass of each sample was determined gravimetrically, using an analytical balance (model AX504, Mettler-Toledo, Greifensee, Switzerland), calibrated with standard weights. Sample amounts of the order of 13 to 15 g were used. Then, the soil was heated to 200 °C for one hour and combusted for at least 4 hours at 550 °C in order to reduce its mass before the microwave digestion or the sample leach. The sample mass reduction after ashing was of the order of 10 %.

Prior to the digestion, known amounts of ^{229}Th and ^{232}U were added as tracers (Sibbens et al., 2004) for the thorium and uranium chemical recovery determination, respectively. The digestion was performed on a Mars 5 Digestion System (CEM Corp., Matthews, NC, USA). Afterwards, the solution was visually clear and ready for the chemical separation.

The digested sample was passed through a pre-packed TRU-resin column (Eichrom Technologies, Inc., Darien, Il., USA) in order to separate the actinides from the bulk of the matrix. Then, UTEVA resin column (Eichrom Technologies, Inc.) was used to separate uranium and thorium. Uranium was prepared for the measurement by Ce co-precipitation and thorium by Nd co-precipitation.

For the acquisition of the alpha-particle spectra the same Canberra Model 7401 VR (Canberra, Meriden, CT, USA) system was used as for the ^{226}Ra measurements (Chapter 2.4).

Efficiency calibration and measurements

The efficiency calibration and the alpha-particle spectra measurements were done in the same way as for the determination of ^{226}Ra. The thorium measurements were done with a source-to-detector distance of 13 mm and a pressure of 1.33 kPa in order to decrease the recoil effects. The minimum acquisition time for the thorium and uranium measurements was 7 days and it was extended according to the availability of the detection system and the count rate of the sample.

Results for thorium isotopes

The results from the determination of ^{232}Th and ^{230}Th are presented in Fig. 8-9. One sample (7) was not taken into account in the calculation of the weighted means due to the bad resolution (FWHM > 100 KeV) of the alpha-particle spectrum. The sample was not re-measured due to lack of time. One sample (5) was re-analyzed (6) as suspect for hot particle. However, both results were used for the calculation of the standard deviations. The chemical yield for thorium ranged from 24 % to 56 %. In the activity calculations, the decay of the thorium isotopes was neglected due to their long half-lives. For the same reason, the decay of the chemical recovery tracer during acquisition was neglected. Peak overlapping was also considered as negligible. The typical uncertainty budgets for single measurements at the 1σ level ($k = 1$) are presented in Table 5.

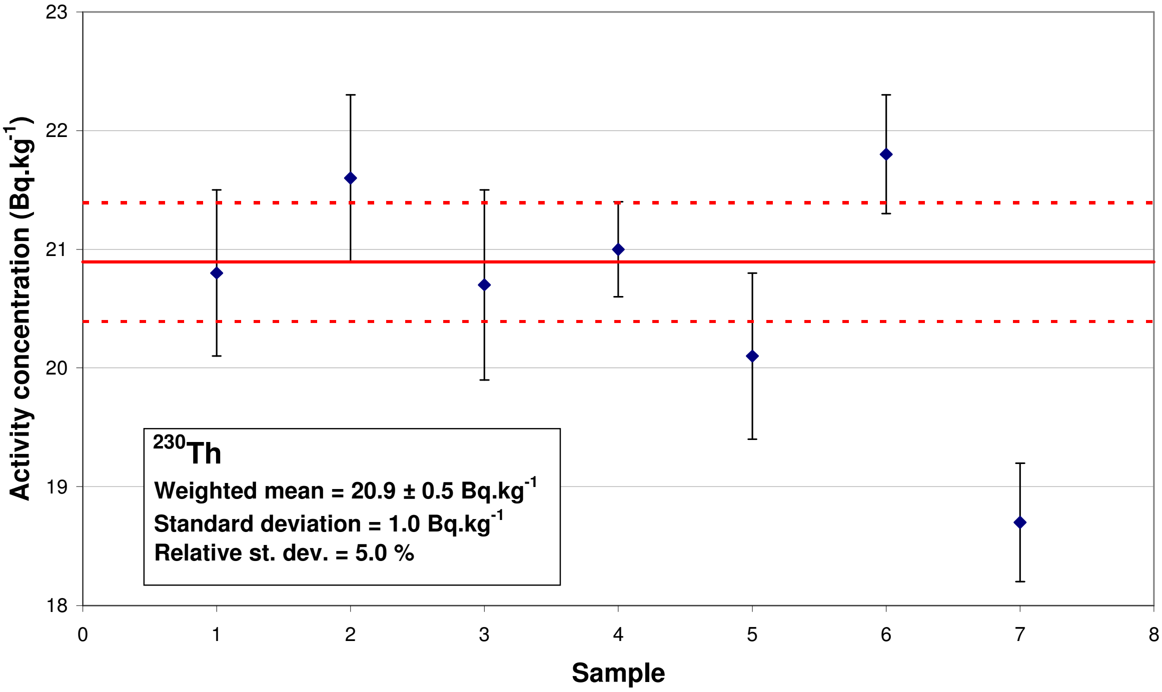

Fig. 8. Activity concentration results of ^{230}Th in the soil. The activity values are calculated for the reference date 31 December 1991. All uncertainties are combined uncertainties at the 1σ level (*k* = 1). The solid red line indicates the weighted mean and the dashed red lines the combined uncertainty of the mean (*k* = 1). The weighted mean is calculated from the first 5 values, while standard deviation is calculated from all values.

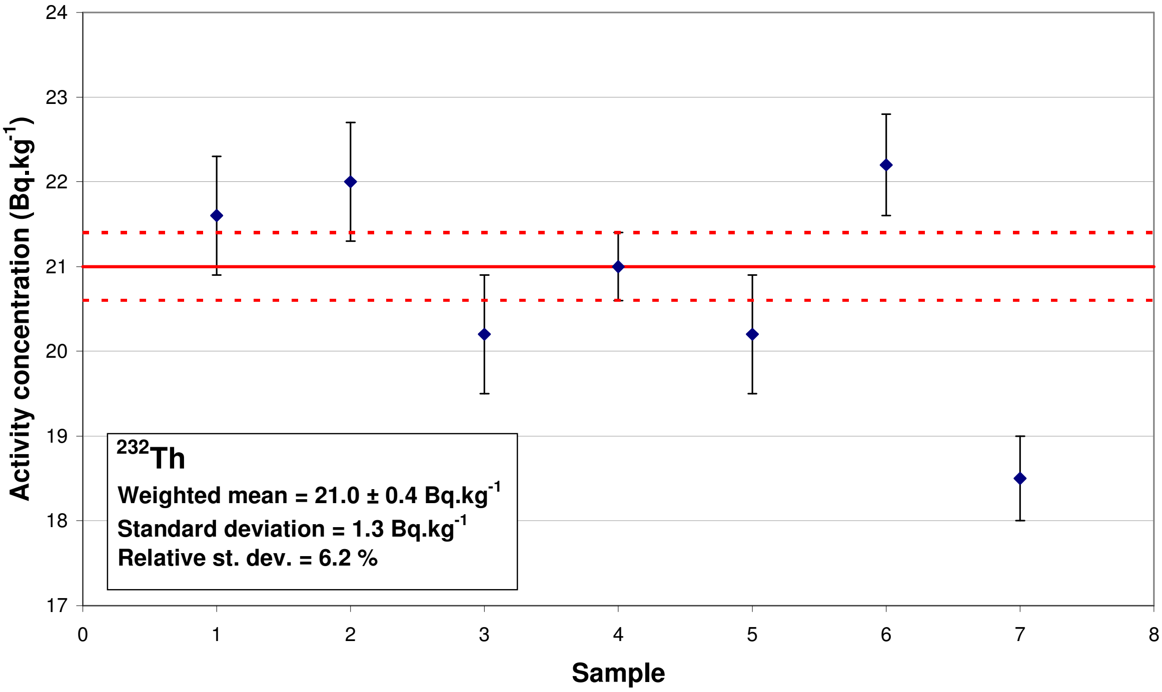

Fig. 9. Activity concentration results of ^{232}Th in the soil. The activity values are calculated for the reference date 31 December 1991. All uncertainties are combined uncertainties at the 1σ level (*k* = 1). The solid red line indicates the weighted mean and the dashed red lines the combined uncertainty of the mean (*k* = 1). The weighted mean is calculated from the first 5 values, while standard deviation is calculated from all values.

Table 5. Uncertainty budgets for ^{230}Th and ^{232}Th assessed in soil by alpha-particle spectrometry show the typical uncertainties for a single measurement of a sample at the 1σ level. The combined uncertainty is the quadratic sum of all components ($k = 1$).

Component	^{230}Th (%)	^{232}Th (%)
Counting statistics (incl. background)	1.3	1.3
Counting statistics for the tracer	2.4	2.4
Tracer activity	0.5	0.5
Weighing	0.1	0.1
Dead time	0.005	0.005
Timing	0.005	0.005
Combined uncertainty	2.8	2.8

Results for uranium isotopes

The chemical yield for uranium ranged from 36 % to 72 %. In the activity calculations, the decay correction for the uranium isotopes was omitted because of the long half-lives. However, the activity of the ^{232}U tracer was corrected for the amount of ^{232}U being generated by the ^{236}Pu tracer (activity concentrations of plutonium isotopes were determined as well). Since the measurements were done immediately after the separation of ^{232}U the ingrowth of ^{228}Th, daughter of ^{232}U, was neglected. The peaks of ^{235}U (4.152 MeV and 4.215 MeV) do overlap with the peak of ^{238}U (4.196 MeV) but the contribution of the interfering ^{235}U to the ^{238}U peak is less than 0.3 % and was considered negligible. The results for uranium isotopes are presented in Fig. 10-12 and the typical uncertainty budgets for single measurements at the 1σ level ($k = 1$) are given in Table 6.

Table 6. Uncertainty budgets for ^{234}U, ^{235}U and ^{238}U assessed in soil by alpha-particle spectrometry show the typical uncertainties for a single measurement of a sample at the 1σ level. The combined uncertainty is the quadratic sum of all components ($k = 1$).

Component	^{234}U (%)	^{235}U (%)	^{238}U (%)
Counting statistics (incl. background)	0.4	1.4	0.4
Counting statistics for the tracer	0.6	0.6	0.6
Tracer activity	0.5	0.5	0.5
Weighing	0.1	0.1	0.1
Dead time	0.005	0.005	0.005
Timing	0.005	0.005	0.005
Combined uncertainty	0.9	1.6	0.9

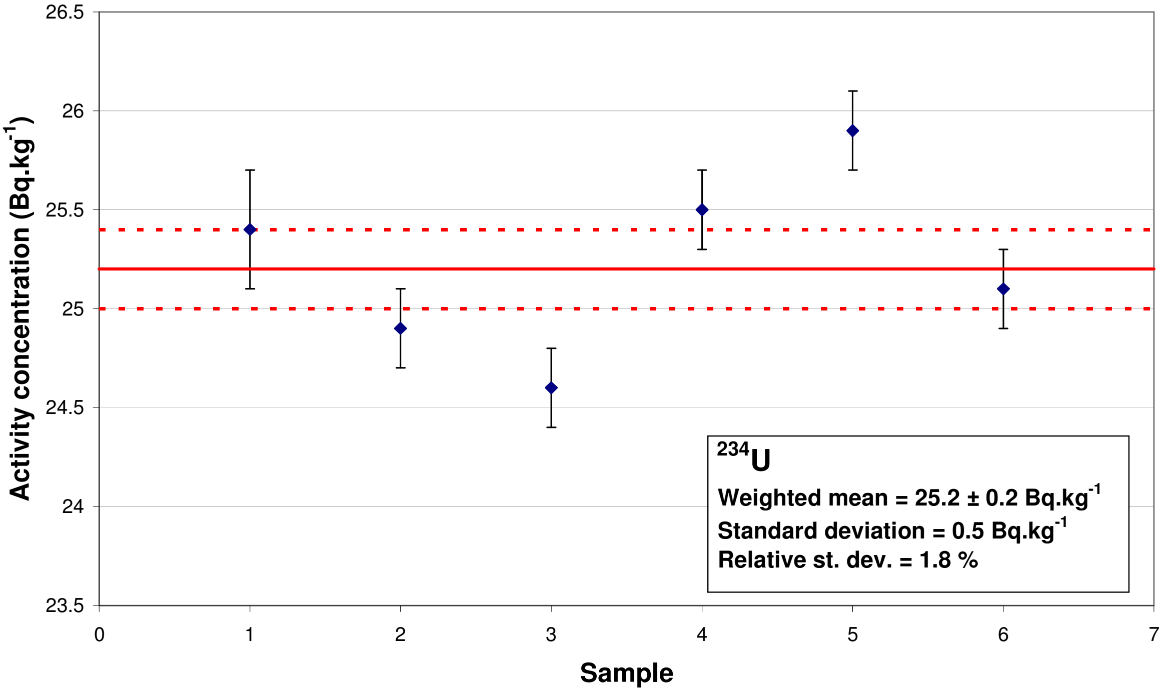

Fig. 10. Activity concentration results of ^{234}U in the soil. The activity values are calculated for the reference date 31 December 1991. All uncertainties are combined uncertainties at the 1σ level (*k* = 1). The solid red line indicates the weighted mean and the dashed red lines the combined uncertainty of the mean (*k* = 1).

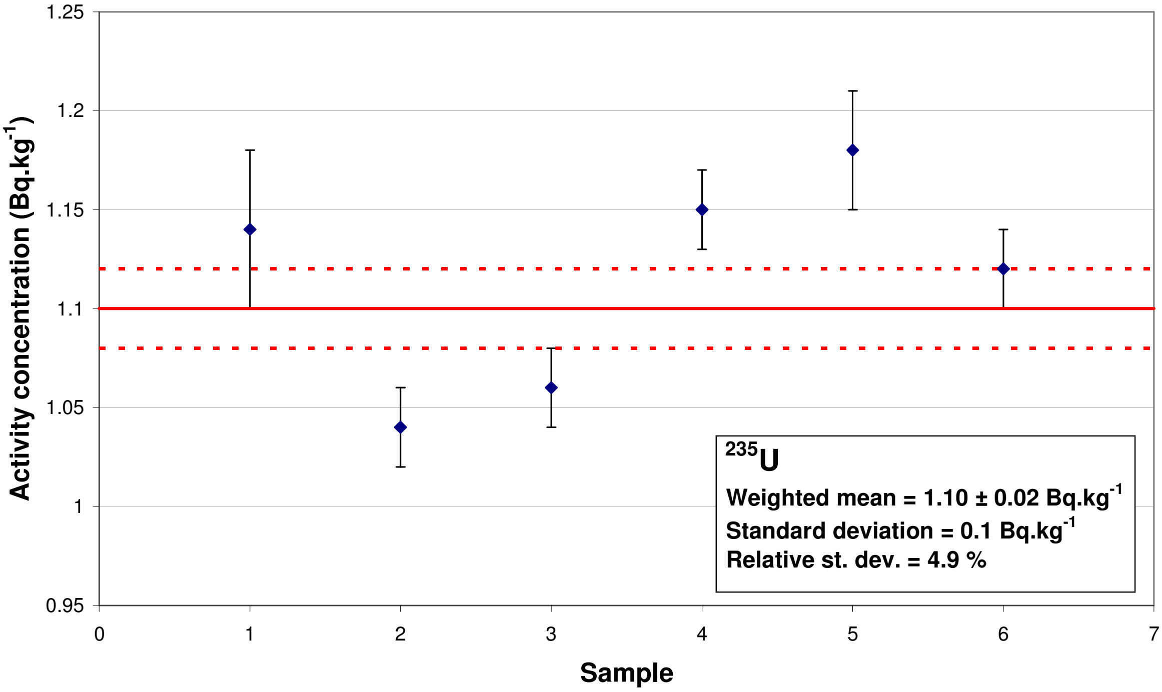

Fig. 11. Activity concentration results of ^{235}U in the soil. The activity values are calculated for the reference date 31 December 1991. All uncertainties are combined uncertainties at the 1σ level (*k* = 1). The solid red line indicates the weighted mean and the dashed red lines the combined uncertainty of the mean (*k* = 1).

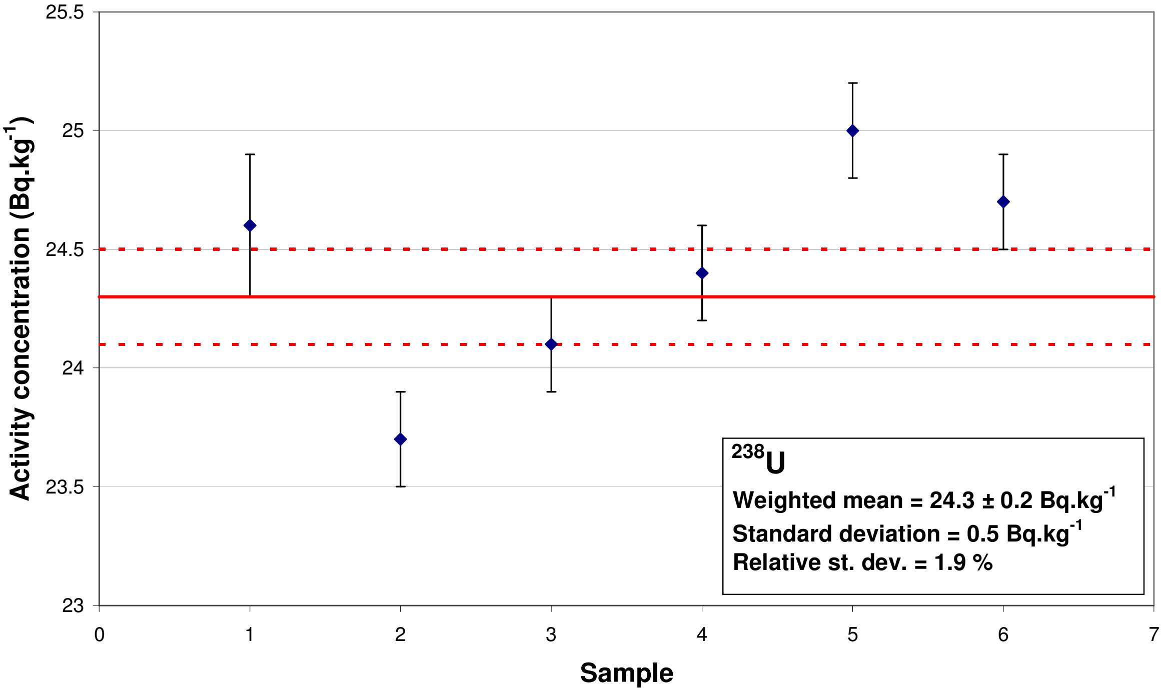

Fig. 12. Activity concentration results of ^{238}U in the soil. The activity values are calculated for the reference date 31 December 1991. All uncertainties are combined uncertainties at the 1σ level ($k = 1$). The solid red line indicates the weighted mean and the dashed red lines the combined uncertainty of the mean ($k = 1$).

All results of the characterisation study are summarized in Table 7. The comparison of the activity concentration values determined within the CRP with those recommended earlier by IAEA is given in Fig. 13 and shows good agreement for most of the radionuclides assessed. More details on the characterisation can be found in Altzitzoglou et al. (2006).

Table 7. Activity concentrations of radionuclides in the IAEA-375 soil reference material determined in the characterisation study (Altzitzoglou et al., 2006). The half-lives from Monographie BIPM-5 (2004, 2006) were used.

	Reference date 31.12.1991		Reference date 1.1.2010	
Radio-nuclide	Activity concentration (Bq·kg^{-1})	Expanded uncertainty (k = 2) (Bq·kg^{-1})	Activity concentration (Bq·kg^{-1})	Expanded uncertainty (k = 2) (Bq·kg^{-1})
^{40}K	410	20	410	20
^{90}Sr	116	8.0	74.5	5.1
^{137}Cs	5400	200	3565	132
^{226}Ra	19.1	1.6	19.0	1.6
^{230}Th	20.9	1.0	20.9	1.0
^{232}Th	21.0	0.8	21.0	0.8
^{234}U	25.2	0.4	25.2	0.4
^{235}U	1.10	0.04	1.10	0.04
^{238}U	24.3	0.4	24.3	0.4

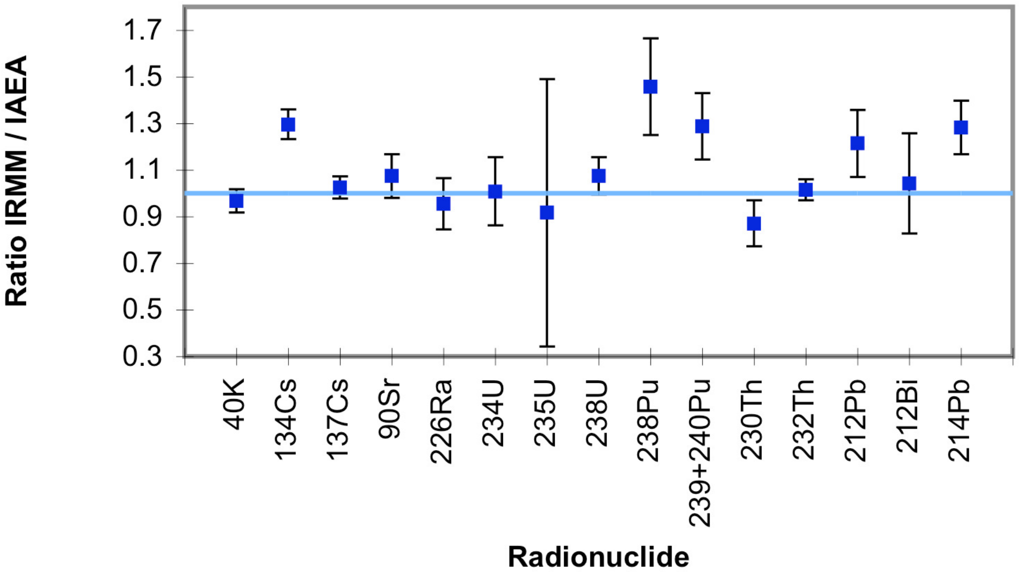

Fig. 13. Comparison of the activity concentration values determined within the IAEA-CRP (Altzitzoglou, 2006) with those recommended earlier by IAEA (IAEA, 1996). The expanded uncertainties include those for the IAEA activity concentration values.

2.6 Homogeneity measurements

The reference value of a comparison material is assumed to be valid for the whole batch at the level of a subsample with a minimum mass. Therefore, an in-homogeneity in the radionuclide concentration increases the uncertainty of the corresponding reference value.

For gamma-ray emiting radionuclides, a dedicated homogeneity study of the soil material was carried out at IRMM (Spasova and Vasile, 2010). For this purpose, 10 bottles of the batch with intercomparison samples, distributed over the entire range of filled bottles, were chosen for performing the homogeneity measurements. The homogeneity of the activity distribution of ^{137}Cs and ^{40}K in these samples was evaluated using gamma-ray spectrometry.

Sample preparation

All samples were prepared gravimetrically. After the water content determination (Karl-Fischer titration), the soil samples were filled into containers on a balance (type 1712, Sartorius GmbH, Göttingen, Germany). For the measurements, cylindrical polypropylene beakers were used with a diameter of about 63 mm, a height of about 73 mm and volume of 125 mL (Nalgene, USA). From each bottle with soil material, two parallel samples of approximately 40 g were taken (in total 20 samples). During the sample filling an electrostatic discharge blower (Ion-care, Sartorius) was used to help avoiding dispersion of the material. To create a denser sample a “tapper” (Dual Autotap, model DA-1, QuantaChrome, Syosset, N.Y., USA) was used (5000 taps/sample). The samples were prepared in the same manner as the ones used for the characterisation of the material and the determination of the reference values (Altzitzoglou et al., 2006).

Measurement equipment

A low-background HPGe detector system was used for the measurements. The semi-planar detector (EURISYS EGMP 80-30-R No. 81086, Canberra Eurisys S.A., AREVA Group, Montigny-le-Bretonneux, France) 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 highly radio-pure Pb. The activity concentration of ^{222}Rn in the laboratory during the period of the measurements was monitored.

The samples were placed directly on top of the detector end-cap and measured for 4 days each. 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. As the task of the performed measurements was to study the homogeneity of the samples, only relative measurements were carried out. Therefore, no efficiency calibration was performed and only net peak intensities were determined.

Results

The results from the gamma-ray spectrometry measurements of ^{137}Cs and ^{40}K in the soil material are presented in Fig. 14-15. The uncertainties are combined standard uncertainties and the major contribution comes from the counting statistics and the geometry repeatability. In the figures, the red solid horizontal line indicates the average and the dashed lines indicate the ±1σ level ($k = 1$).

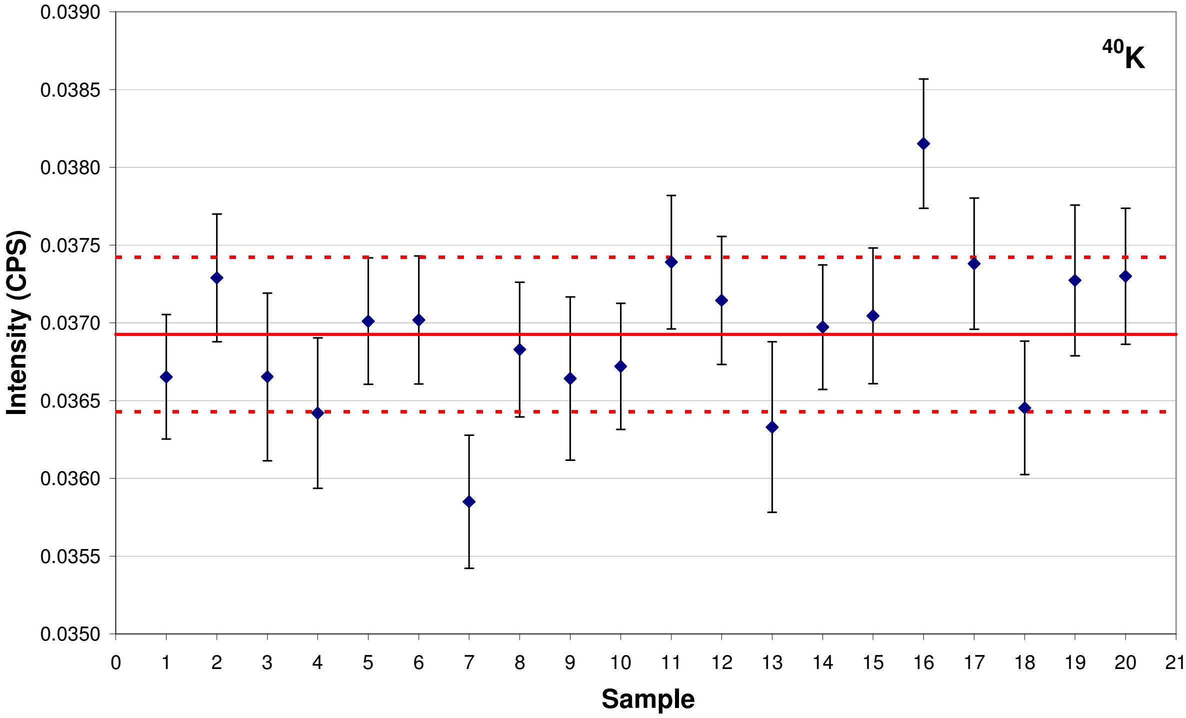

Fig. 14. Count rate of ^{40}K (1460 keV peak) in the soil. All uncertainties are combined standard uncertainties at the 1σ level ($k = 1$). The red solid line indicates the average and the dashed lines indicate the ± 1σ range ($k = 1$).

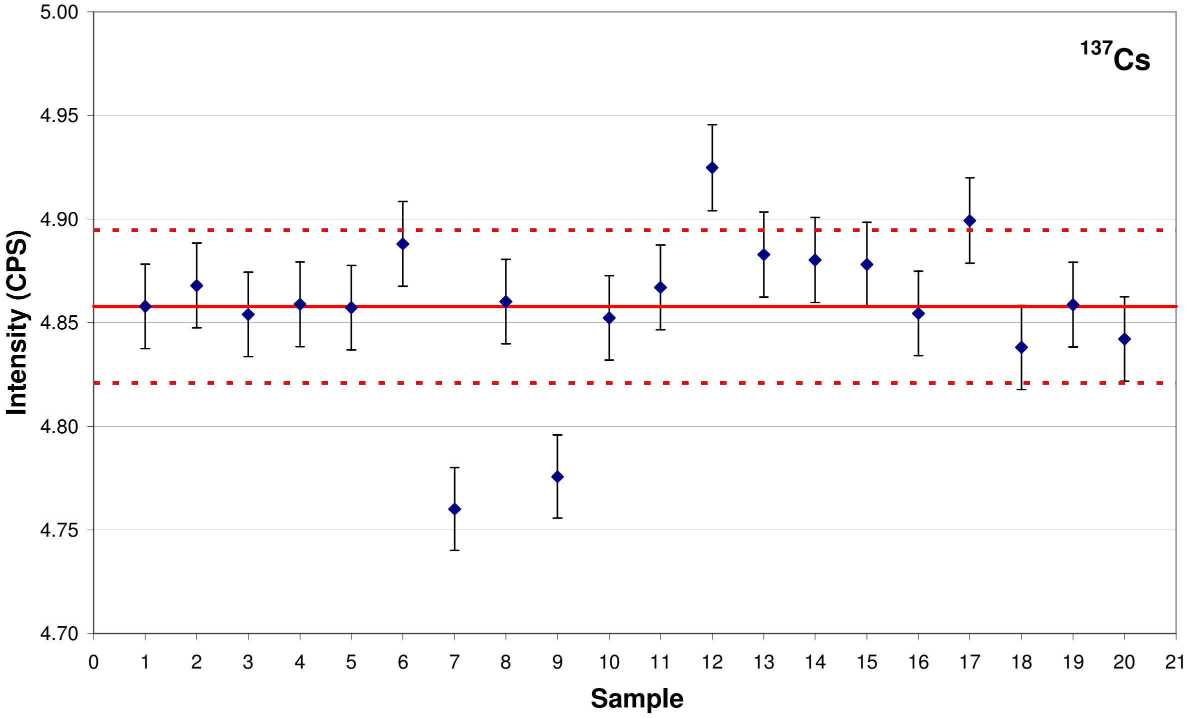

Fig. 15. Count rate of ^{137}Cs (662 keV peak) in the soil. All uncertainties are combined standard uncertainties at the 1σ level ($k = 1$). The red solid line indicates the average and the dashed lines indicate the ± 1σ range ($k = 1$).

Evaluation of data

The in-homogeneity of the radionuclides in the matrix was evaluated using the SoftCRM version 2.0.10 software following the certification principles for reference materials as given in ISO/IEC Guide 35 (ISO, 2006). The data were first tested whether they follow a normal, or at least unimodal distribution. This was done by visual inspection of normal probability plots and histograms. If the data do not follow at least a unimodal distribution, the calculation of standard deviations is doubtful or impossible. All individual results were normally and unimodally distributed.

Grubbs' test was performed to detect potentially outlying individual results. Sample 7 was flagged as an outlier at a level of significance $\alpha = 0.05$ in the case of ^{137}Cs. No outliers were detected for ^{40}K. As no technical reason for the outlier could be found, all the data were retained for the statistical analysis.

The results were then evaluated by a one-way analysis of variance (ANOVA). The between-bottle standard deviation s_{bb} and within bottle standard deviation s_{wb} were calculated with the following formulae (ISO, 2006)

$$s_{bb} = \sqrt{\frac{MS_{between} - MS_{within}}{n}} \quad \text{and} \quad s_{wb} = \sqrt{MS_{within}} \qquad (1)$$

where $MS_{between}$ is the between bottle variance;
MS_{within} is the within bottle variance of the measurements used in the between-bottle homogeneity study;
n is the number of observations per group.

In some cases s_{bb} cannot be estimated because the calculations render unphysical (imaginary) results (i.e. $MS_{\text{between}} < MS_{\text{within}}$). These results could be due to inadequate repeatability of the method used for the homogeneity study. In the case of ^{137}Cs and ^{40}K, the between-bottle standard deviations were calculated as 0.23 % and 0.6 %, respectively. The estimated within bottle standard deviations were 0.73 % and 1.2 % for ^{137}Cs and ^{40}K, respectively. All standard deviations are presented in Table 8.

The in-homogeneity that could be hidden by the method repeatability is calculated by following equation (ISO, 2006)

$$u_{bb}^{*} = \sqrt{\frac{MS_{within}}{n}} \sqrt[4]{\frac{2}{\nu_{MS_{within}}}} \qquad (2)$$

where $\nu_{MSwithin}$ is the degree of freedom of MS_{within}. This expression is based on the consideration that a confidence interval can be established for s_{bb}, and that the half-width of the 95 % confidence interval, converted to a standard uncertainty, can be taken as a measure of the impact of the repeatability of the method on the estimate of s_{bb} (ISO, 2006). The evaluated relative uncertainties between units u_{bb}^{*}, hidden by the method repeatability, are given in Table 8. The uncertainty related to a possible between-bottle variation u_{bb} is then the larger of u_{bb}^{*} and s_{bb}.

The finally adopted uncertainty contributions u_{bb} due to in-homogeneity are presented in the Table 8.

Table 8. ANOVA test results for the gamma-ray emitting radionuclides.

Radionuclide		^{40}K	^{137}Cs
ISO/IEC 13528:2005	s_s	0.6 %	0.23 %
ANOVA	s_{bb}	0.6 %	0.23 %
	s_{wb}	1.2 %	0.73 %
	u_{bb}^{*}	0.6 %	0.34 %
	u_{bb}	0.6 %	0.34 %

An alternative evaluation uses only the standard deviation u_{bb} of all measured sub-samples. This results in an overestimation of real physical in-homogeneity, since the reproducibility of the measurements (in particular counting statistics) is not accounted for. The mean values and the standard deviations of the measured activity concentrations are indicated in Figs. 14-15 by solid and dashed red lines. In relative terms, these standard deviations correspond to $u_{bb}(^{40}\text{K}) = 1.34$ % and $u_{bb}(^{137}\text{Cs}) = 0.76$ % which are consistent with the results given in Table 8 when considering the intrinsic overestimation.

Homogeneity evaluation for other radionuclides

The latter approach was used in the case of radionuclides for which no additional homogeneity study was realized (^{90}Sr, ^{226}Ra, ^{230}Th, ^{232}Th, ^{234}U, ^{235}U and ^{238}U). The standard deviations of six (seven in the case of Th isotopes) bottles analysed during the characterisation study were taken as indicators of homogeneity u_{bb} (chapters 2.3 – 2.5). These values of u_{bb}, possibly overestimating the physical in-homogeneity, are presented in Table 9.

Final reference values

The expanded uncertainty U_{ref} of the reference value is estimated as

$$U_{ref} = k \cdot \sqrt{u_{char}^2 + u_{bb}^2 + u_{lts}^2 + u_{sts}^2} \quad (3)$$

where u_{char} is the combined standard uncertainty from the characterisation study;
u_{bb} is the in-homogeneity contribution;
u_{lts} is the long-term stability;
u_{sts} is the short term stability contribution.

For comparison samples, the long-term stability or the long storage periods of the material are not applicable, hence $u_{lts} = 0$. Uncertainty due to transport conditions was found to be negligible (Wätjen, 2008). Moreover, considering the physical characteristics of the soil material the short-term instability can be neglected and $u_{sts} = 0$. Thus, the final expanded uncertainty U_{ref} can be simplified as

$$U_{ref} = k \cdot \sqrt{u_{char}^2 + u_{bb}^2} \quad (4)$$

The reference values of activity concentrations for the nine radionuclides with their expanded uncertainties are presented in Table 9. It is evident that for some radionuclides (^{90}Sr, Th and U isotopes) the possible in-homogeneity u_{bb} is the dominant contribution to the uncertainty of the reference values. Due to the chosen approach (standard deviation of characterisation data, see above), the resulting uncertainty may be overestimated.

Table 9. Reference values of activity concentrations A_{ref} with expanded uncertainties U_{ref} ($k = 2$) in the soil for the reference date 1 January 2010, together with the relative combined standard uncertainties from the characterisation study u_{char} and the relative homogeneity contributions u_{bb}.

Radionuclide	$A_{ref} \pm U_{ref}$ (Bq·kg^{-1})	u_{char} (%)	u_{bb} (%)
^{40}K	410 ± 21	2.4	0.6
^{90}Sr	74.5 ± 10.1	3.4	5.8*
^{137}Cs	3565 ± 134	1.9	0.34
^{226}Ra	19.0 ± 2.3	4.2	4.4*
^{230}Th	20.9 ± 2.3	2.4	5.0*
^{232}Th	21.0 ± 2.7	1.9	6.2*
^{234}U	25.2 ± 1.0	0.8	1.8*
^{235}U	1.10 ± 0.11	1.8	4.9*
^{238}U	24.3 ± 1.0	0.8	1.9*

*u_{bb} standard deviation of the characterisation dataset was used

3 Methods used by the participating laboratories

Participants were free to use measurement methods of their own choice, preferably the routine procedures used in their laboratories. Depending on their laboratory capabilities they were asked to determine activity concentrations of the following radionuclides: ^{40}K, ^{90}Sr, ^{137}Cs, ^{212}Pb, ^{212}Bi, ^{214}Pb, ^{214}Bi, ^{226}Ra, ^{230}Th, ^{232}Th, ^{234}U, ^{235}U, ^{238}U, ^{238}Pu, and $^{239+240}$Pu. From 73 laboratories, which reported results, only nine (labs: 3, 4, 5, 20, 22, 27, 56, 59, 63) determined activity concentrations of all 15 radionuclides. However, some of these reported values were below the limits of detection (LOD).

Participants were requested to fill in the questionnaire (Appendix 9). Although the laboratories were urged several times to submit the questionnaire, 9 out of 73 participants (labs: 4, 19, 23, 26, 40, 49, 51, 54, 68) did not do so. The information in this chapter is extracted from the submitted questionnaires.

3.1 Water content determination

All results of activity concentrations were instructed to be reported normalized to dry weight. Separate moisture determination of small sample(s) not undergoing further analysis was recommended for this purpose. No special protocol to calculate the moisture content was distributed among participants since this is the common practice in the laboratories and it is not a critical step for the further measurements in this type of sample matrix.

Most of the laboratories determined the water content by oven drying until constant weight at the temperatures 102 °C (49 labs), 105 °C (6 labs), 80 °C (2 labs), 85 °C (1 lab), or 110 °C (1 lab). Only one laboratory (32) used the Karl-Fischer titration and one laboratory (33) used the infrared lamp moisture determination balance in addition to oven drying. One laboratory (47) did not determine the water content at all.

The mass of soil samples used for performing this determination varied from 1 g up to 250 g with the median of 9 g. Some laboratories (1, 18, 33, 67) determined the water content separately for each measurement method and some (5, 6, 11, 17, 25, 46, 56, 57, 61) used more than one (up to 5) aliquot.

The water content expressed as percentage of the weighed sample was reported between 0.15 % and 6.0 %, with the average 2.6 % and median 2.8 %. The water content determined by the Karl-Fischer titration was 3.2 %. Four laboratories (13, 20, 70, 71) used directly the mass of dried sample for the calculations of activity concentrations. Surprisingly, 11 laboratories (9, 12, 14, 17, 30, 31, 42, 44, 52, 58, 73) did not apply any correction although they determined the water content. For four laboratories (7, 63, 67, 69) it is not clear how they applied the correction. The correction factors applied were reported to be between 1.00 and 1.12, with a median of 1.03. One laboratory (37) reported value of the applied correction factor 4.52 (1/0.2212). Three of these values are not consistent with the reported moisture contents (labs: 22, 29, 37).

3.2 Gamma-ray spectrometry: ^{137}Cs, ^{40}K, ^{212}Pb, ^{212}Bi, ^{214}Pb, and ^{214}Bi

Gamma-ray spectrometry was the most often used method in this ILC. All laboratories, except two (31, 64), used this technique. Activity concentrations of ^{137}Cs, ^{40}K, ^{212}Pb, ^{212}Bi, ^{214}Pb, and ^{214}Bi were determined exclusively by direct gamma-ray spectrometry. Only one laboratory (31) measured the ^{137}Cs activity via beta decay. The vast majority of participants used methods routinely applied in their laboratories. One laboratory (47) is accredited for gamma-measurements but only in water samples. Other three laboratories (3, 30, 61) do not measure routinely gamma-ray emitting radionuclides relevant for this ILC.

Sample preparation

In most of the laboratories the sample was not specially treated before the gamma-ray spectrometry measurement. Several laboratories (15, 24, 30, 42, 44, 52, 60, 61, 65, 73) dried the soil before it was placed into the measurement vessel. Few laboratories (15, 17, 39, 53, 61, 66) put some efforts to – additionally - homogenise the material. One laboratory (27) mixed the soil with activated charcoal in order to trap radon daughters. Most of the laboratories prepared for the gamma-ray spectrometry measurement just one sample with the mass varying from 15 g up to 600 g, whereas some participants used parallel, up to 5, samples.

All laboratories used cylindrical containers placed directly on the detector end-cap. The volume of the used beakers varied from 11 mL to 750 mL. In three cases (labs: 20, 30, 45) Marinelli beakers with a volume from 200 to 500 mL were used. One laboratory (20) used in addition a 6 mL container for a well type detector. Apparently, laboratory 5 also used some kind of well type container since they used well detector without, however, giving any details.

Only 23 laboratories indicated in their questionnaires the precautions made in order to achieve the secular radioactive equilibrium between ^{226}Ra, ^{224}Ra and their daughter products. However, we believe that also other laboratories hermetically sealed the containers although they did not specify it in the survey. The storing time before the measurement varied from 20 to 40 days.

Measurement equipment

The measurements were performed mainly with commercially available gamma-ray spectrometry systems (Canberra, Ortec, Eurisys, etc.) consisting mostly of coaxial high purity germanium detectors (HPGe). Four laboratories (11, 12, 17, 33) used extended range coaxial germanium detectors (XtRa), two (15, 67) used broad energy germanium detectors (BEGe), one laboratory (33) used a low energy germanium detector (LEGe) and one laboratory (5) used a germanium well detector. One participating laboratory (13) used two homemade HPGe detectors, true coaxial (10 % efficiency) and planar (~5 % efficiency). The nominal relative efficiency of the detectors varied from 5 % to 150 %.

The acquisition time varied from 1 hour up to 11 days, with the average 49 hours and median 28 hours. The data evaluation was made using commercial software. The most often used was Genie (37 labs), then GammaVision (9 labs), Apex and InterWinner (5 labs each), etc. In three cases (13, 33, 65), in-house developed programmes were used.

Efficiency calibration

Most frequently, the efficiency calibration was made with the use of multiple-nuclide standard solutions or other, unspecified certified reference materials (CRM) in different measurement geometries. Usually, the same or similar geometry as for the sample was used. Five laboratories (1, 14, 16, 20, 30) used soil or soil-like CRM for calibration of their detectors. Three participants (18, 27, 71) performed calibration using point sources and laboratory 27, in addition, used efficiency transfer codes for geometry and matrix correction. Two laboratories (1, 22) determined the efficiency of their detector systems using a CRM in combination with the commercially available software (e.g. LabSOCS). Laboratories 12, 21, 28 and 35 used exclusively LabSOCS (or ISOCS) for performing the efficiency calibration. Laboratories 32 and 36 used Monte Carlo simulations for the efficiency determination.

Efficiency curves were corrected for true coincidence summing and/or self-absorption by laboratories 3, 10, 15, 27 and 46 using software (Gespecor or Genie) or some other calculation methods.

Corrections

Approximately one third of the participating laboratories check the background regularly. However, the periodicity of these measurements varies significantly from weekly checks up to one background measurement per year. Another big group of laboratories measured the background prior to the measurement of the soil sample and typically for approximately the same acquisition time as the sample. Some laboratories used blank samples represented by pure water in a cylindrical beaker of the same geometry as the sample. Other laboratories measured background without any sample placed on the detector.

Only 25 participants responded to the question about the source of the nuclear decay data. In our information letter we recommended to use the Monographie BIPM-5 (2004, 2006), only 12 laboratories claimed to use this source or the on-line library of Laboratoire National Henri Becquerel (LNBH): Nucléide-LARA. Three participants used the IAEA database; other three used the libraries of software used for the spectra analysis. The rest used some other resources.

3.3 Chemical separation and source preparation: ^{230}Th and ^{232}Th

Alpha-particle spectrometry was applied in 19 laboratories (1, 3, 4, 5, 13, 15, 18, 20, 26, 27, 39, 49, 50, 56, 59, 62, 63, 67, 69) in order to measure the activity concentration of ^{230}Th. Gamma-ray spectrometry was used in six laboratories (22, 35, 45, 60, 70, 72), however three of them (22, 35, 60) reported results below LOD. The radionuclide ^{230}Th has no gamma lines; therefore it can be determined (when using gamma-ray spectrometry) only via its daughters assuming secular equilibrium. Most of the laboratories used routine methods in order to determine ^{230}Th activities, except three laboratories (3, 45, 69).

In the case of ^{232}Th, 25 laboratories (1, 2, 7, 9, 14, 22, 29, 30, 33, 34, 35, 37, 38, 40, 41, 42, 45, 46, 48, 51, 57, 61, 70, 72, 73) used gamma-ray spectrometry and 20 participants (1, 3, 4, 5, 13, 15, 18, 20, 26, 27, 39, 43, 49, 50, 56, 59, 62, 63, 67, 69) applied alpha-particle spectrometry. When using gamma-ray spectrometry, ^{232}Th can only be measured via ^{228}Ac assuming equilibrium between the two radionuclides. One laboratory used spectrophotometry without giving further specifications. Except five laboratories (3, 14, 31, 45, 61) all participants used the routine methods.

The procedure followed for gamma-ray spectrometry was the same as described in section 3.2. In this section, sample treatment and pre-concentration techniques used prior to alpha-particle spectrometry are discussed.

Most of the participants used one or two samples with amounts varying from 0.4 g to 5.7 g. Prior to dissolution, 12 laboratories (3, 5, 13, 18, 22, 50, 56, 59, 62, 63, 67, 69) ashed the soil in the muffle furnace at temperatures varying from 450 to 700 °C. The majority of laboratories (1, 3, 13, 15, 18, 20, 22, 43, 50, 59, 63, 69) applied different mixtures of acids in order to dissolve the soil. Two laboratories (9, 62) used microwave digestion and two (5, 56) used fusion digestion.

Chromatography was used in all laboratories in order to separate thorium from the sample. The Eichrom resins (UTEVA, TRU, TEVA) were applied in most laboratories (1, 3, 5, 9, 15, 18, 43, 50, 59, 62, 63). The ion exchange resin Dowex was used in five laboratories (3, 13, 22, 56, 69) and the Bio-Rad AG resin was used in one laboratory (20). Three laboratories (22, 27, 67) used in addition the co-precipitation method.

For source preparation, the majority of participants (1, 3, 5, 15, 20, 27, 43, 50, 56, 62, 69) used electrodeposition onto a stainless steel disc. Six laboratories co-precipitation with fluorides and filtration onto a filter: NdF_3 (13, 18, 22), CeF_3 (59, 63), and LaF_3 (67).

As tracer, the radioisotope ^{229}Th was used in all laboratories except laboratory 62, which used ^{228}Th. Chemical recovery of thorium isotopes varied between 30 % and 97 %.

3.4 Chemical separation and source preparation: ^{234}U, ^{235}U, ^{238}U

The vast majority of participating laboratories determined activity concentrations of ^{234}U by alpha-particle spectrometry. Two laboratories (52, 67) used inductively-coupled plasma mass-spectrometry (ICP-MS) and another two (35, 45) used gamma-ray spectrometry, however the result of laboratory 35 was below LOD. Participants used routine methods except for three laboratories (3, 14, 61).

For the determination of ^{235}U three different methods were used. Alpha-particle spectrometry was applied in 21 laboratories, two of them (13, 56) reported results below LOD. In 20 laboratories gamma-ray spectrometry was used and three (35, 37, 60) reported results below LOD. The third method ICP-MS was used in two laboratories (52, 67). All participants stated that they used the routine analysis procedures.

A similar situation exists for ^{238}U, the most often applied method is alpha-particle spectrometry in 25 laboratories. The second one is gamma-ray spectrometry with 23 laboratories; however two laboratories (7, 35) reported values below LOD. Since the radionuclide ^{238}U does not emit gamma-rays, its activity must be measured (when using gamma-ray spectrometry) via its daughter nuclides, usually ^{234}Th, assuming secular equilibrium. Two laboratories (52, 67) used ICP-MS. One laboratory (31) declared to use the spectrophotometer method, but without giving any further details. Laboratory 58 calculated the activity concentration of ^{238}U from the ^{238}U/^{235}U activity ratio, assuming the presence of natural uranium (the natural activity ratio is 4.6 %). Mostly routine methods were used, except in laboratories 3, 18 (only for gamma-ray spectrometry) and 61.

In the following, the separation and source preparation techniques related to alpha-particle spectrometry and ICP-MS are discussed. The gamma-ray spectrometry procedures are described in section 3.2.

Typically, one or two independent samples were prepared, with the amounts varying from 0.4 g to 5.7 g. In 13 laboratories (3, 5, 6, 13, 18, 2, 33, 50, 52, 56, 59, 62, 63, 67) the soil was ashed before dissolution. The temperatures varied from 450 °C to 700 °C. In order to dissolve the sample, a combination of acids (1, 3, 6, 13, 15, 18, 20, 22, 33, 39, 43, 50, 52, 59, 61, 63) was most often used. Fusion digestion was applied in two laboratories (5, 56) and another two (9, 62) used microwave digestion technique.

Most of the participants (5, 6, 9, 15, 18, 20, 27, 43, 50, 59, 61, 62, 63, 67) used extraction chromatography using Eichrom resins to separate uranium from the sample. Four laboratories (13, 22, 39, 56) used Dowex ion exchange resins, three laboratories used Bio-Rad AG resins (20, 33, 64). Some laboratories (6, 13, 20, 22, 27,) used specific co-precipitation in combination with the separation on resins. Laboratory 3 used liquid/liquid extraction of acid extract with methyl trioctyl ammonium nitrate in xylene followed by back-extraction under reducing conditions and U(VI) chloro-complex purified by anion exchange.

A majority of laboratories (1, 3, 5, 6, 15, 20, 27, 33, 43, 50, 56, 61, 64) used the electrodeposition technique for sample preparation. Six laboratories used co-precipitation with CeF_3 (59, 62, 63, 67) or NdF_3 (13, 22) on membrane filters. Laboratories 52 and 67 determined uranium also via ICP-MS. In this case, the sample was diluted in ultra-pure nitric acid after separation and measured with ICP-MS.

All laboratories used ^{232}U as an internal tracer. In addition, laboratory 50 added iron as a carrier. The chemical recovery of uranium varied from 44 % to 98 %.

3.5 Chemical separation and source preparation: ^{238}Pu, $^{239+240}$Pu

Activity concentrations of ^{238}Pu and $^{239+240}$Pu were determined by 21 and 26 laboratories, respectively. All laboratories which reported results of plutonium isotopes stated alpha-particle spectrometry as technique used. Except for two laboratories (48, 66), all of them used the routine methods.

Most of the laboratories used one or two samples with amounts varying from 1 g to 42 g. Participants applied different methods for the sample pretreatmet and dissolution. A great number of laboratories (3, 5, 6, 13, 22, 37, 48, 52, 56, 59, 62, 63, 67) incinerated the soil material before dissolution at temperatures varying from 450 °C to 700 °C. The majority of participants (1, 6, 37, 39, 43, 56, 59, 64, 66, 13) applied different mixtures of acids in order to dissolve the sample completely. Seven laboratories (15, 20, 22, 48, 61, 62, 63) extracted plutonium with aqua regia. Three laboratories (3, 5, 9) used microwave digestion.

Mainly, the ion exchange technique using resins was applied for the separation of plutonium from the sample. Six laboratories (13, 22, 37, 39, 48, 56) used ion exchange resins of Dowex, three laboratories used the Bio-Rad AG resins (20, 64, 66). One particular Eichrom ion exchange resin (UTEVA, TRU, TEVA, DGA) or their combination was applied in following laboratories: 1, 3, 5, 15, 43, 59, 63. Also other laboratories (9, 27, 61, 62, 67) used extraction chromatography, but without any detailed specifications. Only one laboratory (6) applied the method of anion exchange in HNO_3 medium followed by liquid-liquid extraction in benzene.

Generally, two different methods were used for the source preparation. Electrodeposition onto a stainless steel disc was used by 15 laboratories (1, 5, 6, 9, 15, 20, 27, 37, 43, 48, 56, 61, 64, 66, 67). The second method was co-precipitation and filtration onto a membrane filter. Plutonium was co-precipitated with fluorides: CeF_3 (3, 59, 62, 63), NdF_3 (13, 22) or LaF_3 (39, 67).

Tracer was added to the sample before the separation was carried out. All participating laboratories used ^{242}Pu as a plutonium tracer, only laboratory 39 used ^{236}Pu. Chemical recovery of plutonium isotopes varied between 10 and 100 %.

3.6 Determination of ^{226}Ra

In order to measure the activity concentrations of ^{226}Ra, gamma-ray spectrometry was applied in 43 laboratories. The same procedure as described in section 3.2 was followed. All participants used the routine procedures, except laboratories 45 and 18.

Laboratories 27 and 50 applied the emanation technique using a Lucas cell. The daughter radionuclide ^{222}Rn is allowed to grow in for a specific period of time. The radon gas is then transferred into a Lucas Cell and the alpha activity of the radon and its daughters is measured by scintillation counting. Laboratory 31 described their method as an installation for measuring radon and thoron, but from the description we assume that its basis is also the emanation technique. However, their result was below LOD.

Laboratory 4 used a flow proportional counter, but since no questionnaire was submitted, no further information is available.

In four laboratories (1, 18 - applied both methods, 43, 59) alpha-particle spectrometry was applied. Laboratory 18 ashed the soil sample at a temperature of 650 °C while laboratory 59 did not ash the sample at all. Two laboratories (18, 59) used acid digestion and one laboratory (43) applied microwave digestion. For the separation of radium, all participants used the $BaSO_4$ co-precipitation method. Radionuclide ^{133}Ba was used as a tracer and laboratory 1 reported a chemical recovery of 60 %.

3.7 Alpha-particle spectrometry

Alpha-particle spectrometry was used in order to determine several radionuclides: ^{238}Pu, $^{239+240}$Pu, ^{234}U, ^{235}U, ^{238}U, ^{230}Th, ^{232}Th and ^{226}Ra. Sample treatment and separation techniques were discussed in the previous sections. In this section we discuss the aspects of alpha-particle spectrometry measurements.

Most of the participants used passivated implanted planar silicon (PIPS) detectors with an active area varying from 450 mm^2 to 1700 mm^2. However, the most commonly used PIPS detectors are those of 450 mm^2. Seven laboratories (15, 20, 37, 43, 50, 56, 59) used a silicon surface barrier (SSB) detector. Laboratory 52 used a grid chamber (only for Pu isotopes). Laboratory 63 indicated the use of Si detectors without further specification.

The total acquisition time varied from 20 hours to 21 days. The data evaluation was made mainly using commercial software like Genie (10 labs), Alpha Analyst (3 labs), AlphaVision, MAESTRO-32, Apex-Alpha and InterWinner (2 labs each). Two laboratories (13, 33) used in-house developed software.

Most of the laboratories used internal tracers for determination of the chemical recovery. Mixed alpha sources prepared from the CRMs or commercial solid sources were used for the energy calibration of alpha-detectors.

Some laboratories check the background regularly; the periodicity, however, varies significantly from weekly checks up to one or two background measurements per year. Other laboratories measured the background just before the measurement of the soil sample for approximately the same acquisition time. Laboratory 27 used as background the spectrum based on the average data of multiple background spectra taken over longer periods (> year). Several participants found the background and reagent blank corrections negligible.

3.8 Determination of ^{90}Sr

In total 40 laboratories reported results of ^{90}Sr activity concentrations, all except one (61) used routine procedures. The majority of laboratories used two or three parallel samples for the strontium determination. The amounts of soil varied from 1 g up to 100 g. Prior to the chemical separation of ^{90}Sr from the soil matrix, the samples were ashed in a muffle furnace in most of the laboratories (3, 5, 6, 11, 13, 20, 22, 28, 33, 37, 41, 43, 44, 50, 52, 55, 56, 59, 61, 63, 67, 69). Temperatures varied from 450 °C up to 700 °C.

Sample preparation

All laboratories used acids in order to dissolve the soil material. Only laboratory 5 used pressurised micro-wave digestion of 10 g of ash in 3 aliquots and borate fusion of 1 g of ash in 3 aliquots.

Strontium and yttrium were separated from the sample mainly by two methods: precipitation (labs 6, 10, 11, 18, 24, 34, 37, 39, 53, 55, 61, 66) and the use of Sr Resin (Eichrom) (labs 13, 14, 20, 33, 38, 44, 50, 59) or by combination of both (labs 22, 67, 63, 64, 69). Laboratory 5 used ion-exchange chromatography in combination with oxalate precipitation. Five participants (3, 28, 41, 43, 56) used solvent extraction. Laboratory 56 applied ion-exchange chromatography on Dowex prior to extraction.

The source preparation was dependant on the measurement method. In total, five different counting methods were applied for the strontium determination. Most of the laboratories (5, 6, 11, 18, 20, 24, 27, 38, 39, 44, 50, 52, 53, 55, 63, 66, 67) used gas flow proportional counting. In this case, different precipitation procedures were used: oxalate (5, 6, 24, 38, 39, 50, 53, 67), hydroxide (27, 50, 67), carbonate (11, 18), fluoride (63), or sulphate (66). Afterwards, the precipitate was filtered (5, 50, 63) or deposited on a stainless steel disk by drying (11, 18, 24, 39, 55). The acquisition time varied from 1 hour to 5 days, with the average of 33 hours.

Measurement equipment

In eight laboratories (3, 13, 14, 22, 26, 28, 59, 69) liquid scintillation counting (LSC) was applied. The use of three different liquid scintillators was indicated in questionnaires: Ultima Gold AB (14, 28), HiSafe 3 (22, 69) and ProSafe (13). The precipitate with strontium was either dissolved and mixed with the scintillator or the filter paper with precipitate was put into a vial and dissolved with acid and then mixed with scintillator (69). Packard Tri-Carb (3, 28), Wallac Guardian (13, 69) and Quantulus (22, 59) counting instruments were used. The measuring time ranged from 3 hours to 1 day, with the average of 11 hours.

Four laboratories (10, 34, 37, 56) used the plastic scintillation counting method. In this case, similar procedures of source preparation were applied as in the case of proportional counting. Laboratory 10 deposited the residue on a plate of stainless steel. In laboratories 37 and 56 yttrium was precipitated as oxalate, and in laboratory 34 strontium was precipitated as oxalate. Laboratory 10 used a gross-alpha and -beta counter equipped with a ZnS(Ag) detector and a plastic detector in anti-coincidence. The anti-coincidence technique was applied also in laboratory 56. The counting time varied from 5 to 17 hours, with the average of 10 hours.

In three laboratories (33, 41, 43) ^{90}Sr was determined by measuring its daughter ^{90}Y using Cherenkov counting. Before the measurement, the hydroxide precipitate was dissolved in 1 mL of concentrated nitric acid and transferred into a scintillation vial without addition of any scintillation cocktail. The samples were counted between 2 and 17 hours, with the average of 7 hours. In two cases (41, 43) a Quantulus counter was used and laboratory 33 used the Hydex-Oy Triathler Multilabel Tester.

Two participants (61, 65) applied counting using Geiger-Müller (GM) detectors. Laboratory 61 used the low level GM beta counter system, model RISO, and measured the sample in 4 cycles for 180 minutes. Unfortunately, laboratory 65 did not provide any detailed information on the used procedure.

Calibration and corrections

The chemical recovery was determined either gravimetrically or by using radioactive tracers. In the first case, the inactive strontium (10, 11, 14, 18, 20, 22, 37, 55, 64, 67, 69) or yttrium (5, 24, 28, 39, 41, 43, 67) carrier was used, in the second case the sample was spiked with a reference solution of ^{85}Sr (6, 13, 50, 63, 67) or ^{90}Sr+^{90}Y (28, 33). Chemical recovery of strontium varied from 46 % to 100 %. The yield of yttrium varied from 69 % to 99 %.

Detection systems were calibrated mostly with certified solutions which were treated either under the same conditions as real samples or used for spiking. Laboratories 10 and 66 used calibrated solid sources for this purpose.

For the background control, most of the participants (6, 14, 22, 28, 33, 39, 41, 43, 44, 59, 66) used blank samples undergoing the same chemical treatment as the soil sample. Others (10, 55) used an empty stainless steel disk or filter. And some laboratories (24, 37, 53, 56, 69) applied both approaches. Four participants (6, 11, 53, 56) check background on a routine basis, others (10, 13, 18) control it only before and after each sample measurement.

Several different software (QuantaSmart, Eclipse, etc.) were used for the evaluation of measured data. One laboratory (22) used homemade software and five participants (24, 33, 38, 52, 56) treated the data manually.

4 Reported results

In total, 1000 results were reported, including double results and values beow LOD. Double results are two values reported by one laboratory for one radionuclide determined by the same method (257 results were sent as double values). In case the two reported results were determined via two different techniques, they were treated separately. In the third column of Table 10 the numbers of values LOD are reported and in parenthesis are the numbers without double results. In the fourth column, the numbers of results without double results and values below LOD are presented for each radionuclide.

Table 10. Numbers of reported results per radionuclide

Radionuclide	Number of all reported results	Number of reported results LOD (without double results)	Number of results (without double results and those below LOD)	Number of reported double results
^{40}K	99	0	70	29
^{90}Sr	61	0	40	21
^{137}Cs	102	0	72	30
^{212}Pb	85	0	63	22
^{212}Bi	77	1	57	19
^{214}Pb	85	2(1)	62	21
^{214}Bi	87	2(1)	63	22
^{226}Ra	75	9(5)	50	16
^{230}Th	33	4(3)	22	7
^{232}Th	63	0	46	17
^{234}U	39	3(2)	28	8
^{235}U	56	7(5)	38	11
^{238}U	68	3(2)	50	15
^{238}Pu	32	7(5)	17	8
$^{239+240}$Pu	38	3	24	11
Total	1000	41(23)	690	257

The individual activity concentrations (normalised to dry mass) with expanded uncertainties (coverage factor $k = 2$), as they were reported by the participants, are presented in Appendix 10. If a coverage factor different from 2 was reported, we recalculated the expanded uncertainty for $k = 2$.

4.1 Data treatment

Most results were reported as single results with expanded uncertainties, which were directly taken into the further evaluations.

In case double results were submitted, the arithmetic average A_{lab} was calculated and taken into account for the further evaluations. The values below LOD were excluded from the results analysis. Two different approaches were used for the estimation of the expanded uncertainties U_{lab} of the laboratory mean values. In the first approach the expanded uncertainty U_{lab} ($k = 2$) of the mean activity concentration A_{lab} was calculated based on the information given in the uncertainty budget submitted by the participating laboratory. The following formula was applied

$$U_{lab} = k \cdot \sqrt{\frac{u_{count}^2}{n} + u_{other}^2} \qquad (5)$$

where

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

$$u_{other} = \sqrt{u_{sample}^2 + u_{tracer}^2 + u_{eff}^2 + u_{st}^2 + u_{blank}^2 + u_{rest}^2} \qquad (6)$$

where

u_{sample} is the propagated uncertainty due to uncertainty in sample preparation, ashing, separation, etc.;
u_{tracer} is the propagated uncertainty due to uncertainty in tracer activity;
u_{eff} is the propagated uncertainty due to uncertainty in efficiency of the detection system;
u_{st} is the propagated uncertainty due to uncertainty in the activity efficiency standards;
u_{blank} is the propagated uncertainty due to uncertainty in blank and background measurement;
u_{rest} represents other propagated uncertainty contributions.

Only the counting uncertainty was considered as a random component (type A uncertainty according to the GUM) and, consequently, divided by $\sqrt{n}$ in the combined uncertainty calculation. Equation (6) may lead to an overestimation of u_{other} and U_{lab} in cases where a propagated contribution, e.g. u_{sample}, represents a random component if this is not taken care of by dividing by $\sqrt{n}$. However, the u_{other} was considered as a systematic component for all samples. The counting uncertainty u_{count} was calculated from the submitted combined uncertainty u_{lab} and the uncertainty u_{other}

$$u_{count} = \sqrt{u_{lab}^2 - u_{other}^2} \qquad (7)$$

Nevertheless, only about 16 % of double results were treated using the first approach since most of the uncertainty budgets were inconsistent or missing.

For the majority of laboratories the expanded uncertainty U_{lab} (with coverage factor $k = 2$) of the mean activity concentration A_{lab} was calculated as an arithmetic average of the individual reported expanded uncertainties (second approach).

In Figs. 16 to 30 the mean activity concentrations A_{lab} with corresponding expanded uncertainties U_{lab} ($k = 2$) are plotted in ascending order, in so-called S-plots. Again, the solid red lines indicate the reference activity concentrations and the dashed lines the expanded uncertainties ($k = 2$) of the reference values. Laboratories' codes are indicated with the results. In Appendix 10 tables with all reported and averaged values are presented.

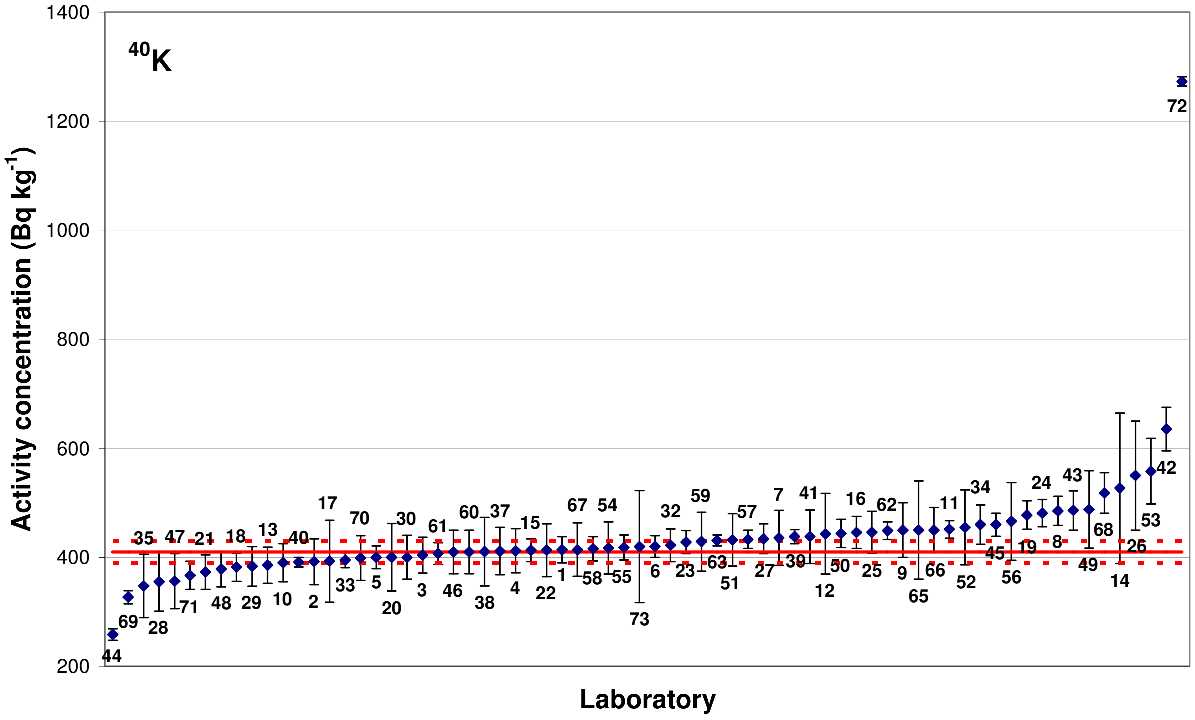

Fig. 16. Laboratory results A_{lab} of ^{40}K activity concentration with expanded uncertainties U_{lab} ($k = 2$) sorted in ascending order. Red lines represent reference value $A_{ref} \pm U_{ref}$ ($k = 2$). Numbers indicate the laboratory code.

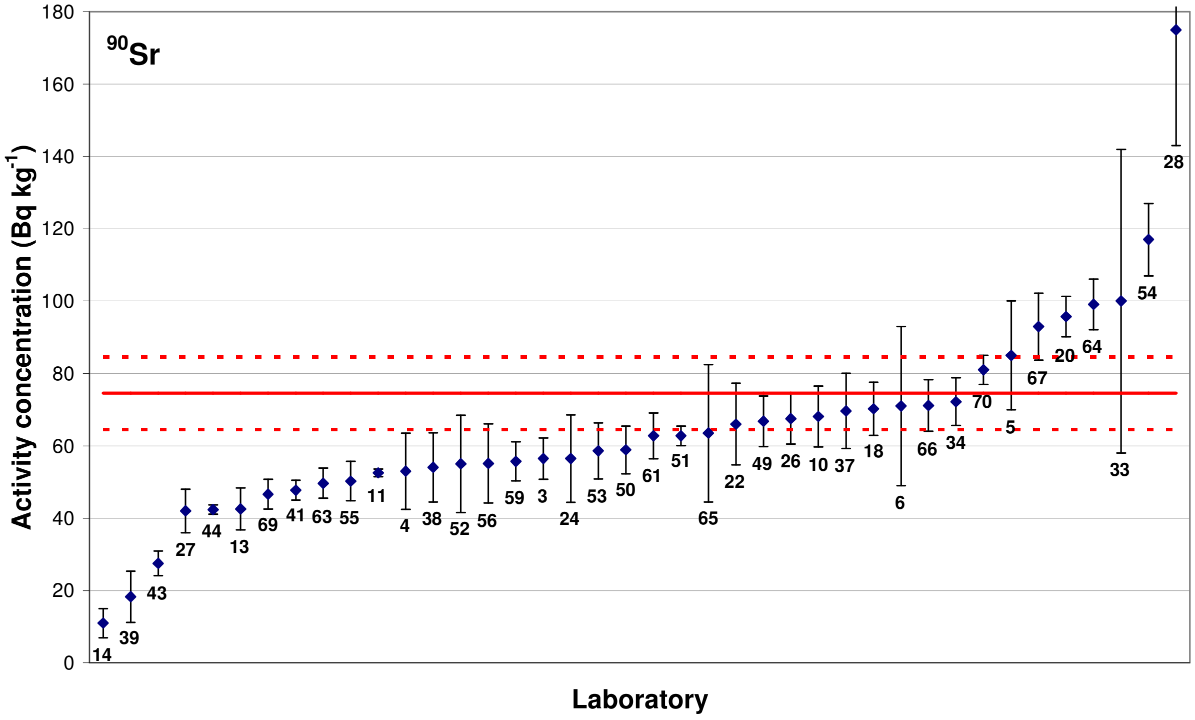

Fig. 17. Laboratory results A_{lab} of ^{90}Sr activity concentration with expanded uncertainties U_{lab} ($k = 2$) sorted in ascending order. Red lines represent reference value $A_{ref} \pm U_{ref}$ ($k = 2$). Numbers indicate the laboratory code.

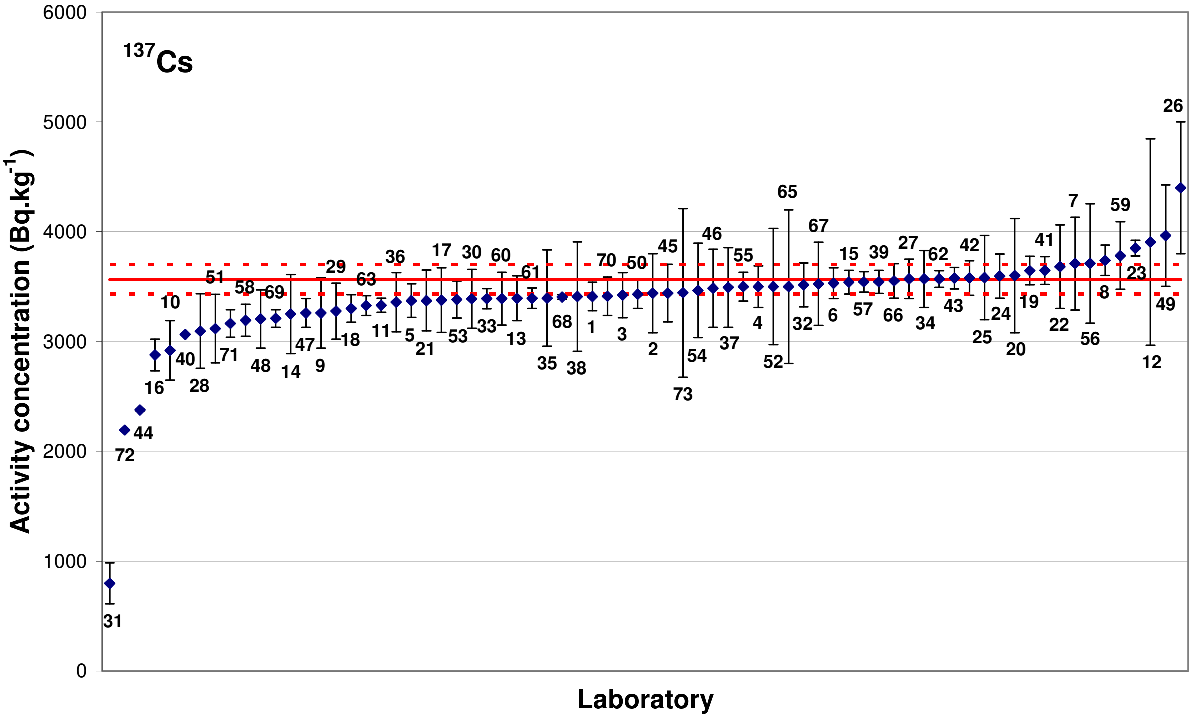

Fig. 18. Laboratory results A_{lab} of ^{137}Cs activity concentration with expanded uncertainties U_{lab} ($k = 2$) sorted in ascending order. Red lines represent reference value $A_{ref} \pm U_{ref}$ ($k = 2$). Numbers indicate the laboratory code.

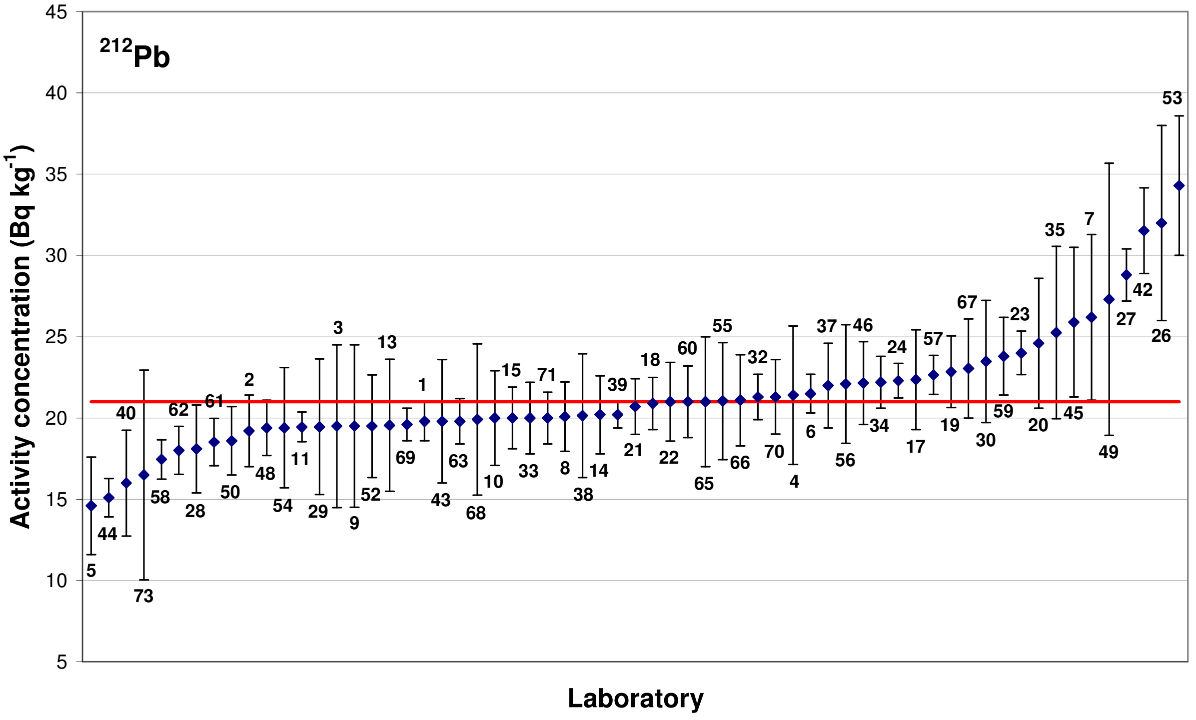

Fig. 19. Laboratory results A_{lab} of ^{212}Pb activity concentration with expanded uncertainties U_{lab} ($k = 2$) sorted in ascending order. Red line represents the indicative value of the mother radionuclide (^{232}Th). Numbers indicate the laboratory code.

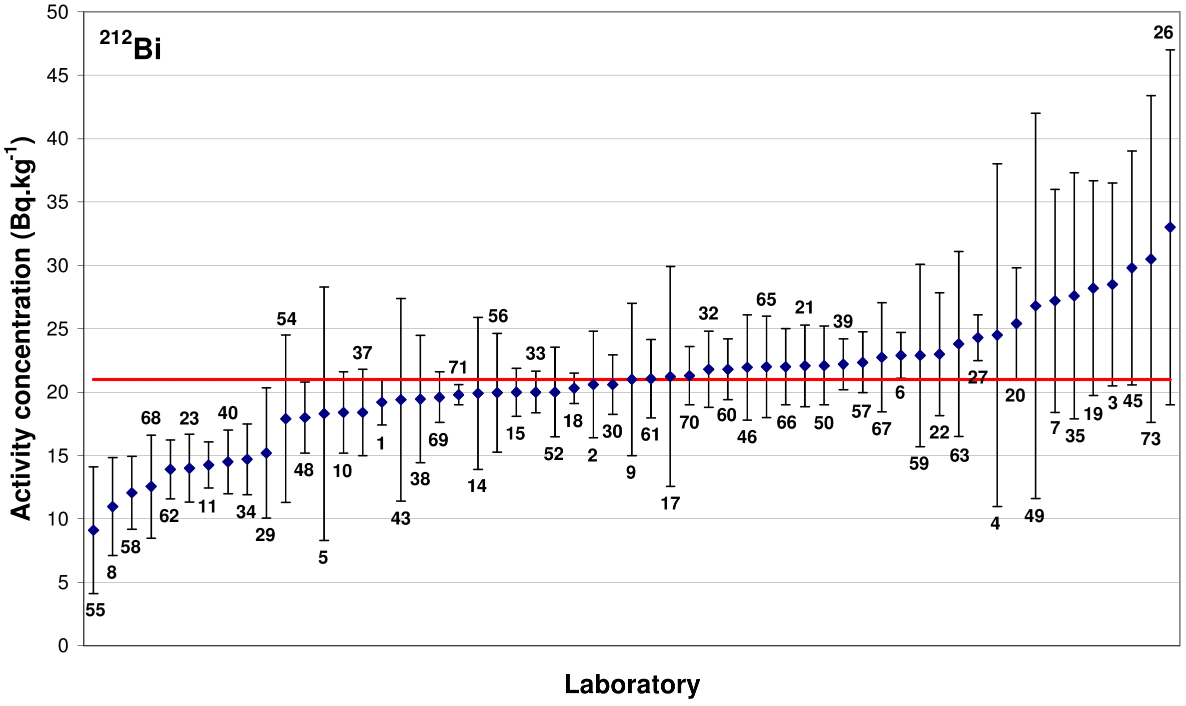

Fig. 20. Laboratory results A_{lab} of ^{212}Bi activity concentration with expanded uncertainties U_{lab} ($k = 2$) sorted in ascending order. Red line represents the indicative value of the mother radionuclide (^{232}Th). Numbers indicate the laboratory code.

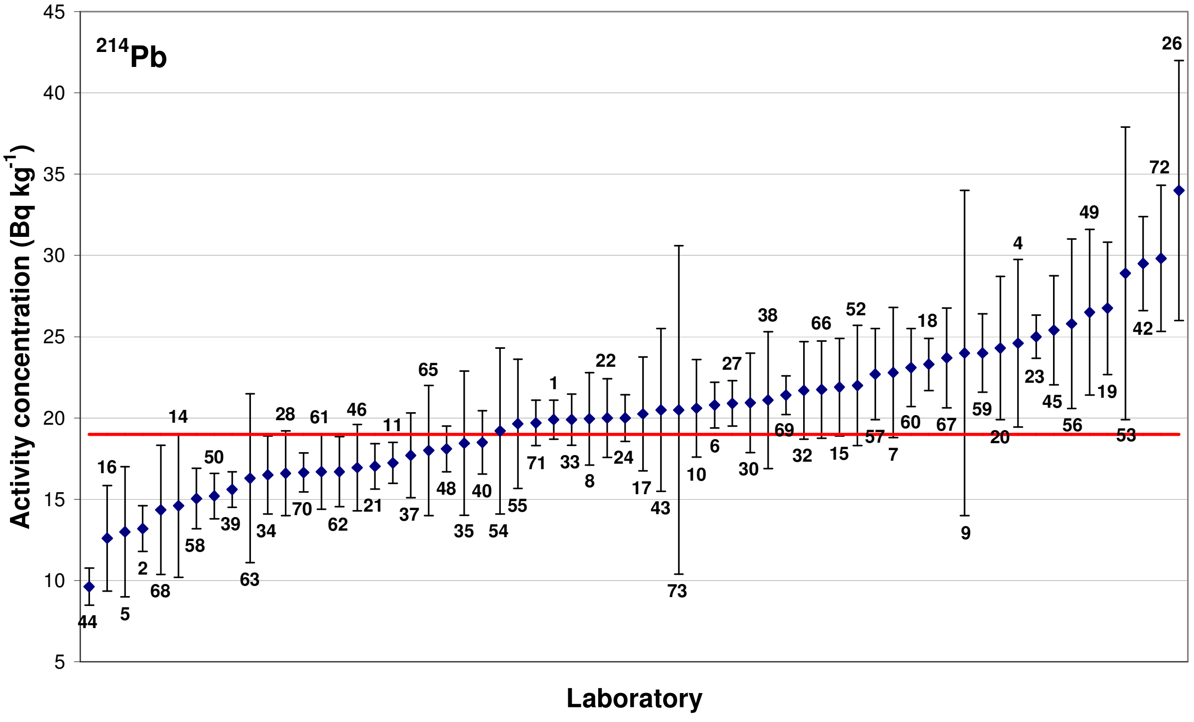

Fig. 21. Laboratory results A_{lab} of ^{214}Pb activity concentration with expanded uncertainties U_{lab} ($k = 2$) sorted in ascending order. Red line represents the indicative value of the mother radionuclide (^{226}Ra). Numbers indicate the laboratory code.

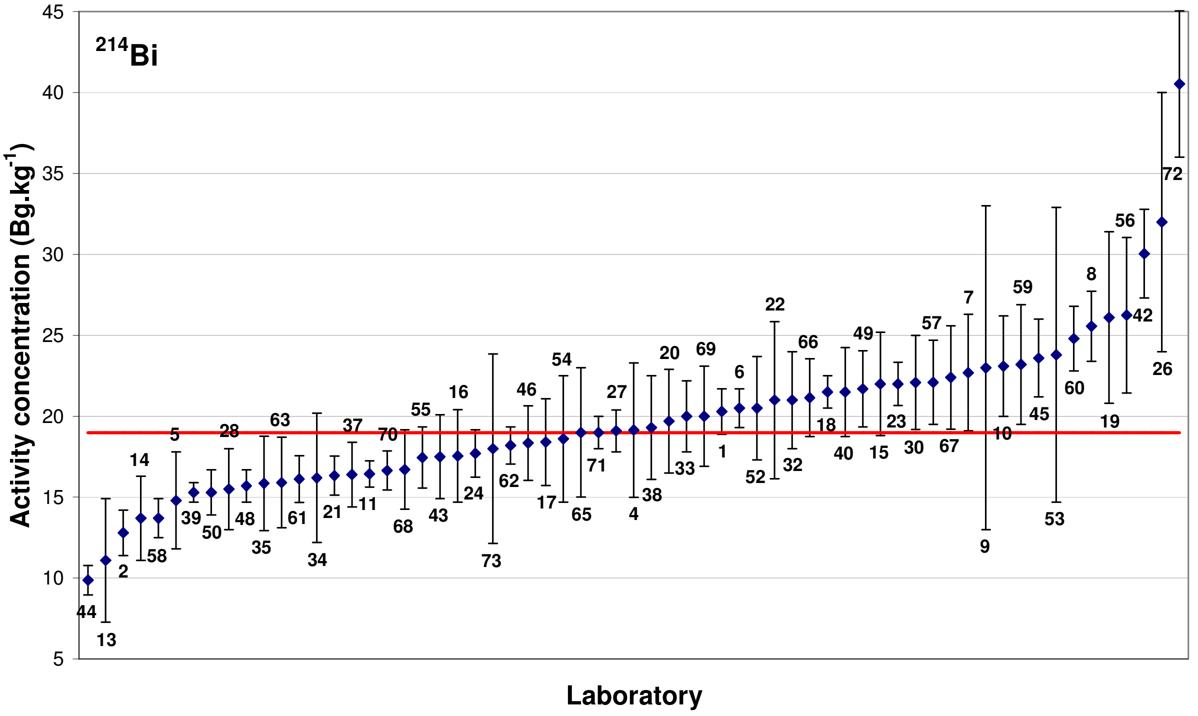

Fig. 22. Laboratory results A_{lab} of ^{214}Bi activity concentration with expanded uncertainties U_{lab} ($k = 2$) sorted in ascending order. Red line represents the indicative value of the mother radionuclide (^{226}Ra). Numbers indicate the laboratory code.

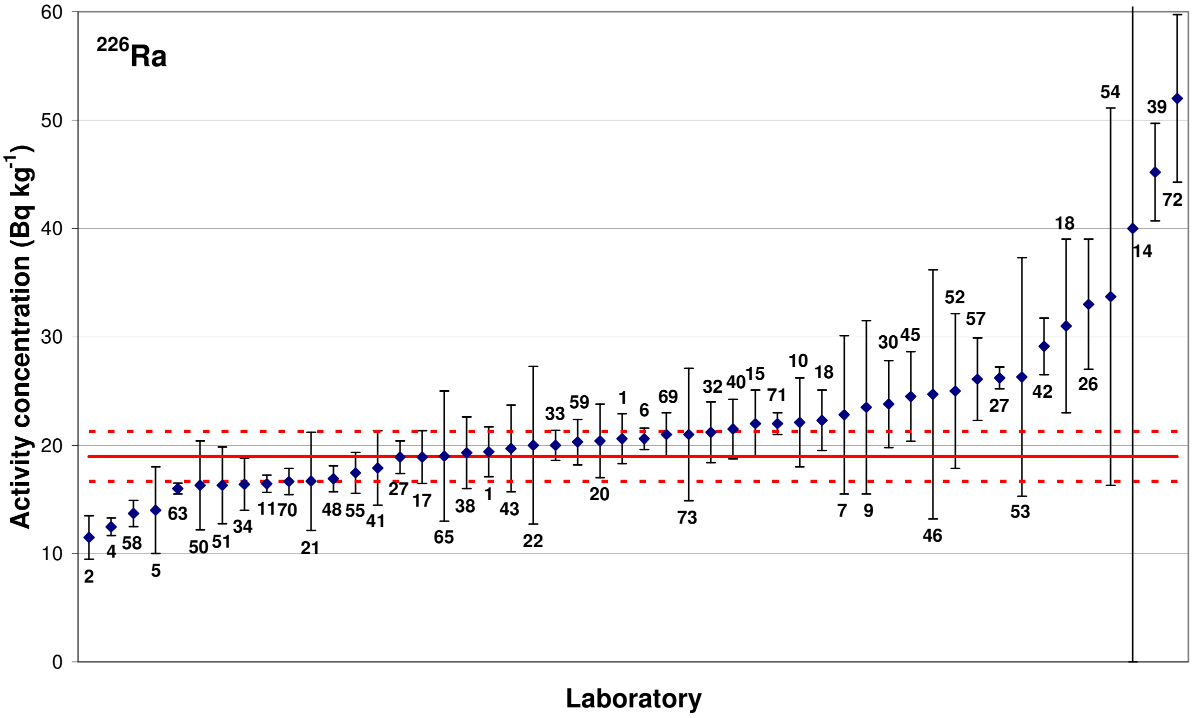

Fig. 23. Laboratory results A_{lab} of ^{226}Ra activity concentration with expanded uncertainties U_{lab} ($k = 2$) sorted in ascending order. Red lines represent reference value A_{ref} ± U_{ref} ($k = 2$). Numbers indicate the laboratory code.

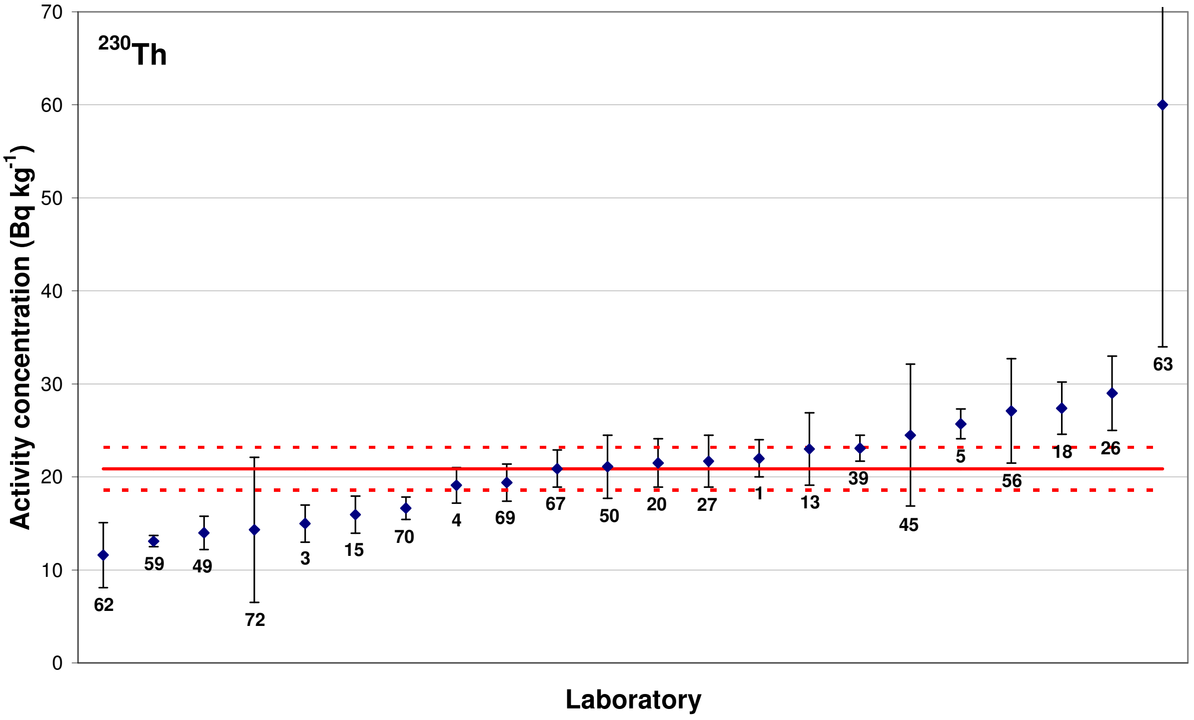

Fig. 24. Laboratory results A_{lab} of ^{230}Th activity concentration with expanded uncertainties U_{lab} ($k = 2$) sorted in ascending order. Red lines represent reference value $A_{ref} \pm U_{ref}$ ($k = 2$). Numbers indicate the laboratory code.

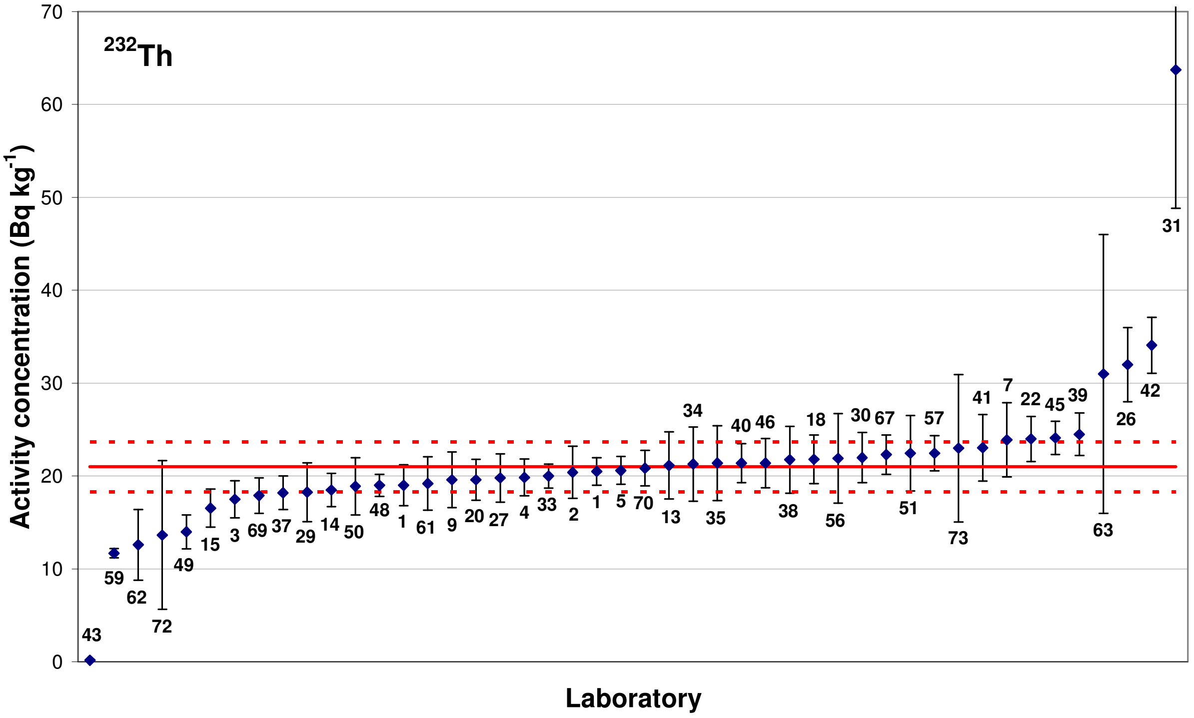

Fig. 25. Laboratory results A_{lab} of ^{232}Th activity concentration with expanded uncertainties U_{lab} ($k = 2$) sorted in ascending order. Red lines represent reference value $A_{ref} \pm U_{ref}$ ($k = 2$). Numbers indicate the laboratory code.

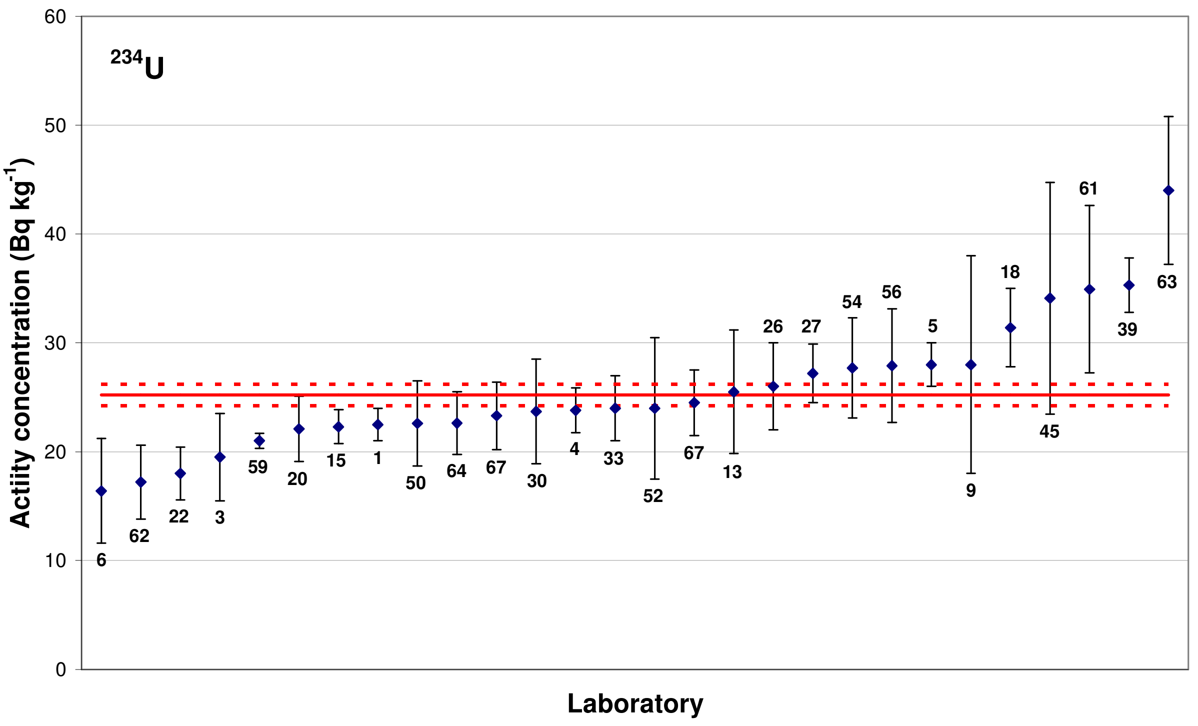

Fig. 26. Laboratory results A_{lab} of ^{234}U activity concentration with expanded uncertainties U_{lab} ($k = 2$) sorted in ascending order. Red lines represent reference value $A_{ref} \pm U_{ref}$ ($k = 2$). Numbers indicate the laboratory code.

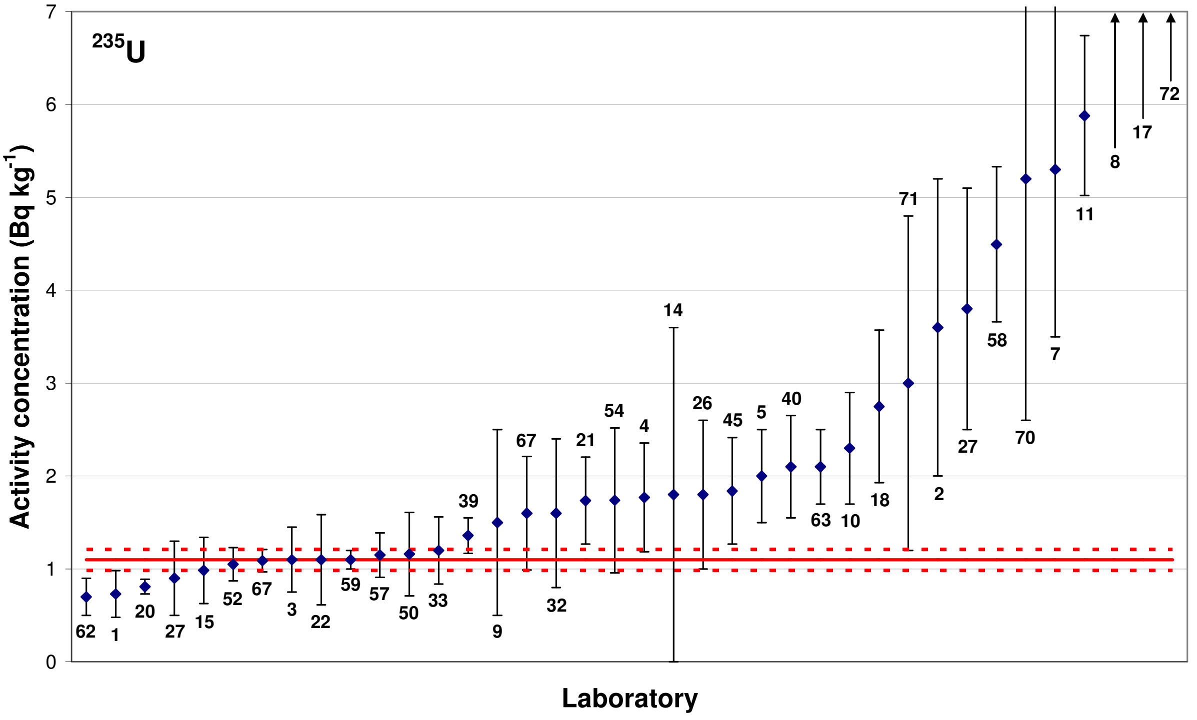

Fig. 27. Laboratory results A_{lab} of ^{235}U activity concentration with expanded uncertainties U_{lab} ($k = 2$) sorted in ascending order. Red lines represent reference value $A_{ref} \pm U_{ref}$ ($k = 2$). Numbers indicate the laboratory code.

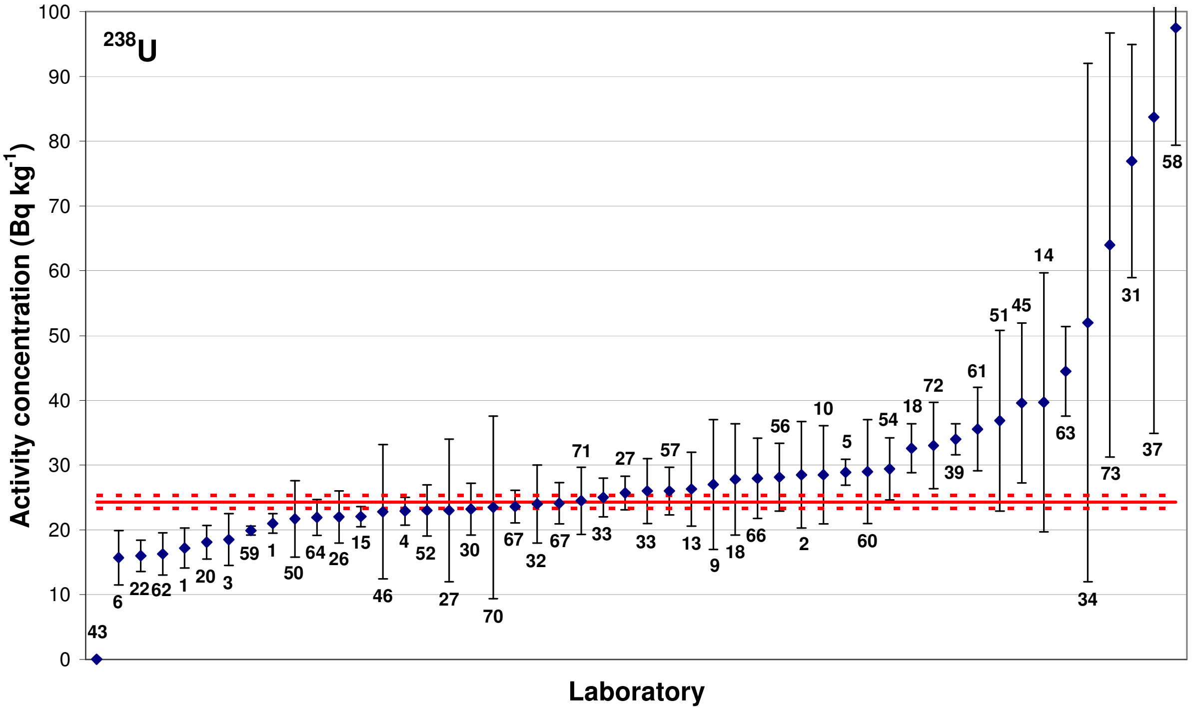

Fig. 28. Laboratory results A_{lab} of ^{238}U activity concentration with expanded uncertainties U_{lab} ($k = 2$) sorted in ascending order. Red lines represent reference value $A_{ref} \pm U_{ref}$ ($k = 2$). Numbers indicate the laboratory code.

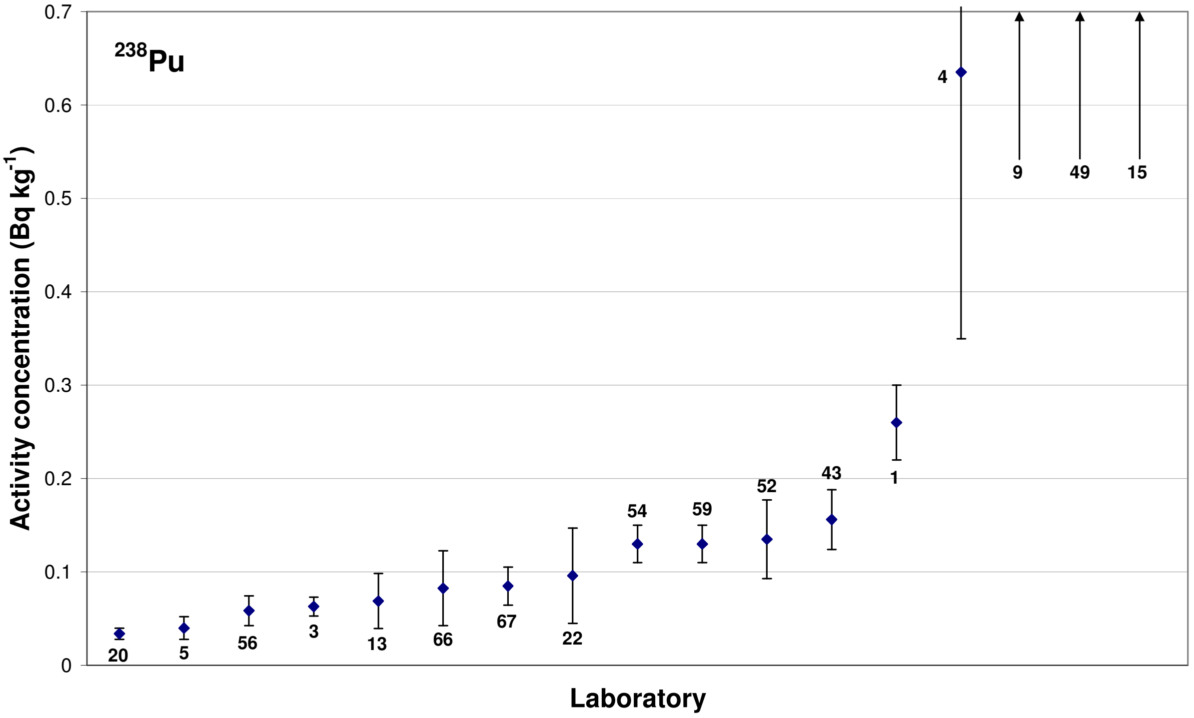

Fig. 29. Laboratory results A_{lab} of ^{238}Pu activity concentration with expanded uncertainties U_{lab} ($k = 2$) sorted in ascending order. Numbers indicate the laboratory code.

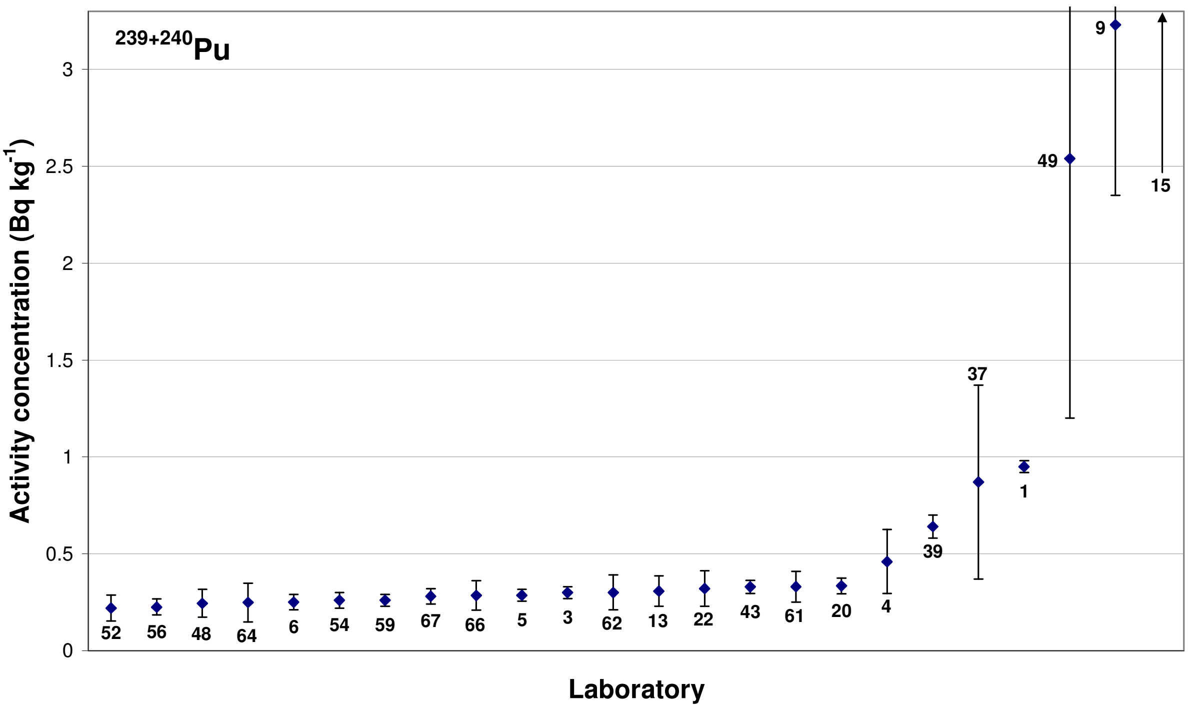

Fig. 30. Laboratory results A_{lab} of $^{239+240}$Pu activity concentration with expanded uncertainties U_{lab} ($k = 2$) sorted in ascending order. Numbers indicate the laboratory code.

4.2 Uncertainty budgets

Participants were requested to provide the uncertainty budget, which was a part of the questionnaire, for each used method. Although 64 out of 73 participating laboratories submitted the questionnaire, 11 of them did not fill in the uncertainty budget (labs: 8, 13, 16, 29, 31, 42, 44, 53, 63, 65, 72). The uncertainty budgets of two laboratories (47 and 60) were not complete or were presented in general manner and, therefore, it was not possible to evaluate them. For these reasons we were not able to analyse and evaluate the abilities to treat the uncertainties of approximately 30 % of the participating laboratories.

The submitted uncertainty budgets were analysed and compared with the numerical values reported with the measurement results. In average 40 % of analysed uncertainty budgets (about 30 % of all results), ranging from 25 % (for ^{212}Bi) up to 60 % (for ^{90}Sr), were inconsistent. Surprisingly, in most of these cases the simple recalculation of the relative combined standard uncertainty u_C according to the submitted uncertainties did not agree with the value of u_C. The cases where the effect of rounding could play a role were considered as consistent. The high number of discrepant uncertainty budgets most probably results from the fact that the determination of the uncertainties is not well treated in these laboratories or not enough attention is paid to these calculations. The final overview of submitted uncertainty budgets, consistent and inconsistent budgets submitted per radionuclide is given in Table 11.

During the analysis of the uncertainty budgets we observed one of the mistakes in reporting of uncertainties. Laboratory 39 submitted uncertainties indicating the coverage factor $k = 2$, but from the uncertainty budget it was clear that these values correspond to the $k = 1$. In spite of this observation the reported uncertainties were used in further analysis. This example shows the importance of careful and attentive reporting.

Table 11. Overview of the uncertainty budgets per radionuclide

	Submitted unc. budget	Consistent	Inconsistent	Inconsistent (%)
^{40}K	25*	12	12	50
^{90}Sr	25	10	15	60
^{137}Cs	42*	25	16	39
^{212}Pb	14	8	6	43
^{212}Bi	12	9	3	25
^{214}Pb	16	10	6	38
^{214}Bi	15*	9	5	36
^{226}Ra	17*	11	5	31
^{230}Th	9	5	4	44
^{232}Th	11	5	6	55
^{234}U	9	6	3	33
^{235}U	11*	7	3	30
^{238}U	16*	9	6	40
^{238}Pu	8	4	4	50
$^{239+240}$Pu	14	8	6	43
total	244	138	100	41#

* one uncertainty budged was not analyzed
\# average

As can be seen in S-plots (Figs. 16-30), several laboratories (11, 43, 44, 59, 63, 71) consistently or for some radionuclides reported lower uncertainties. In this case, it is possible that although their result is close to the reference value, the E_n number is not compatible (see chapter 5.2). On the other hand, many laboratories (4, 7, 9, 14, 26, 45, 49, 53, 54, 63, 65, 73) reported large uncertainties for several radionuclides. Therefore, these results may appear as E_n satisfactory although the value is distant from the reference value. Most of these participants (except labs 9, 45, 73) submitted incompatible uncertainty budgets or did not submit any budget at all.

4.3 Additional radionuclides

Within the questionnaire the participants had the option to report additional radionuclides measured in the soil sample. Activity concentrations of different radionuclides (^{134}Cs, ^{241}Am, ^{234}Th, ^{210}Pb, ^{210}Po, ^{228}Ra, ^{228}Ac, ^{228}Th, and ^{208}Tl) were reported by 21 laboratories (2, 5, 8, 9, 15, 18, 20, 21, 27, 31, 32, 33, 45, 46, 48, 57, 63, 66, 67, 69, and 71). Laboratories 52 and 13 declared that they detected ^{134}Cs and ^{241}Am, respectively, but reported no values.

The most frequently reported radionuclide (10 laboratories) was ^{210}Pb. The presence of this radionuclide is natural since it originates from the uranium series (^{238}U). The average value calculated from 10 reported results is (28 ± 3) Bq·kg $^{-1}$. The data are presented in Fig. 31. From the same decay series originate also ^{234}Th and ^{210}Po, but these radionuclides were only reported by laboratories 21 and 31, respectively. Radionuclides ^{228}Ra, ^{228}Ac, ^{228}Th, and ^{208}Tl originate from the Thorium series (^{232}Th). The reported activity concentrations correspond quite well as can be seen in Fig. 32.

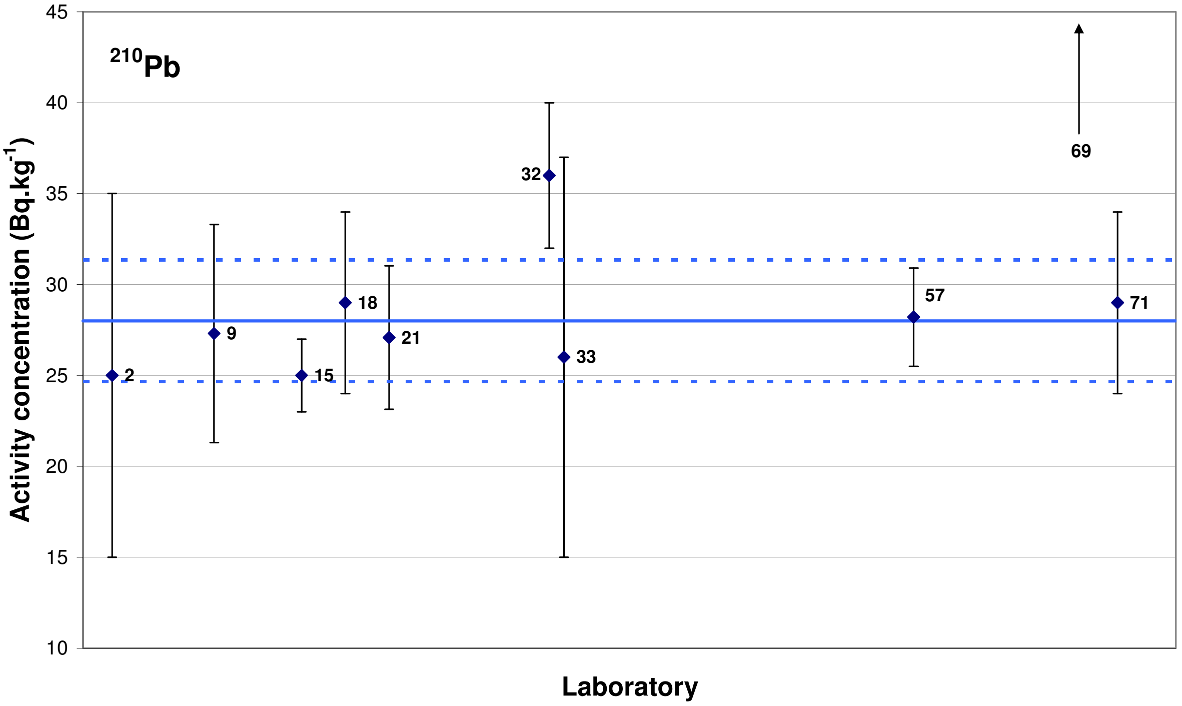

Fig. 31. Reported activity concentrations of ^{210}Pb with expanded uncertainties ($k = 2$). The blue lines indicate the average (without the result of lab 69) activity concentration with the standard deviation of the data. Numbers indicate the laboratory code.

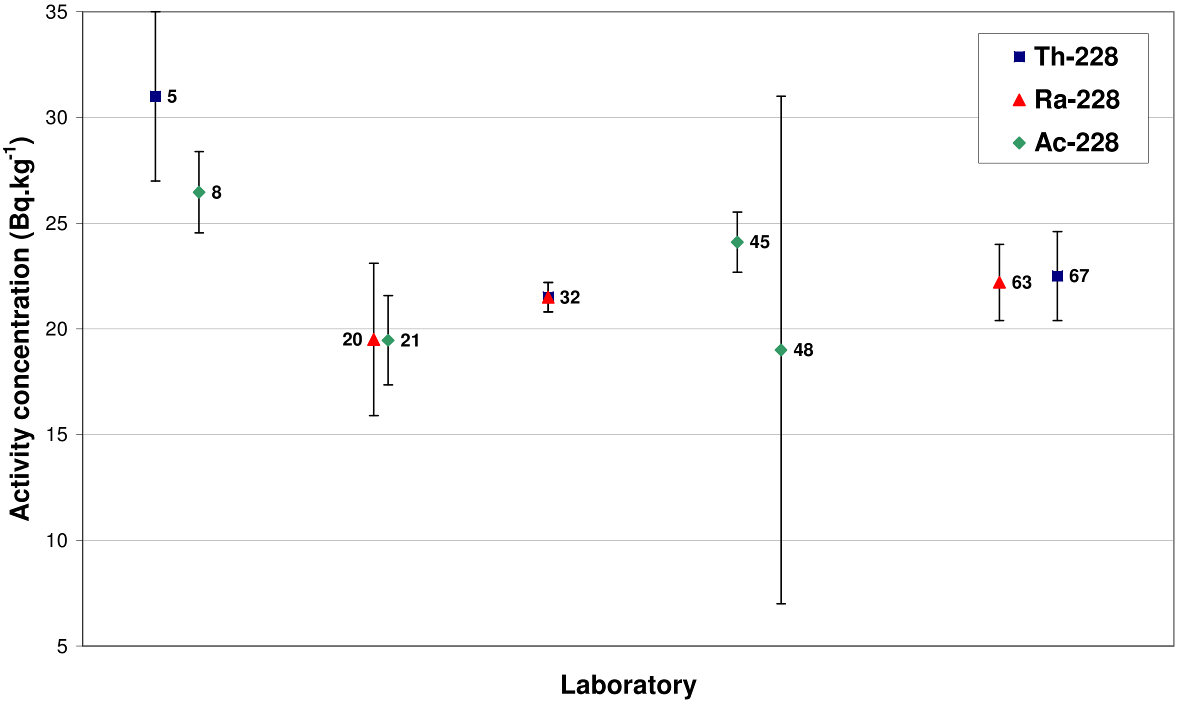

Fig. 32. Reported activity concentrations of ^{228}Th, ^{228}Ra and ^{228}Ac with expanded uncertainties ($k = 2$). Numbers indicate the laboratory code.

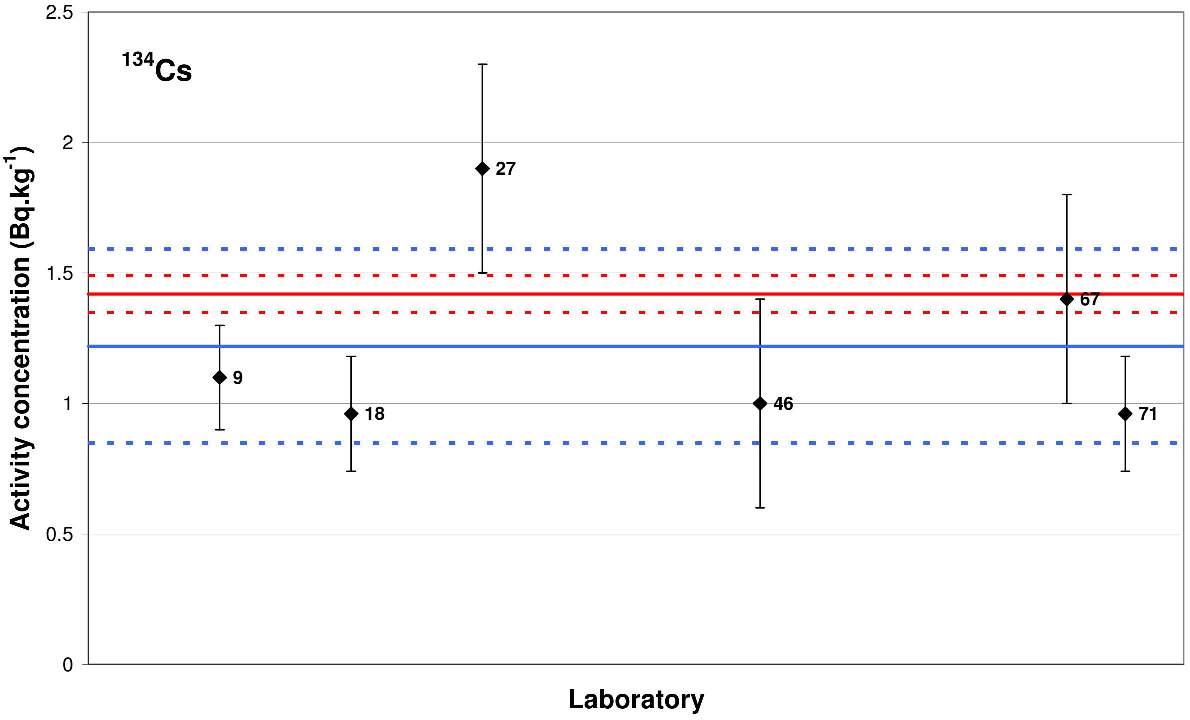

Fig. 33. Reported activity concentrations of ^{134}Cs with expanded uncertainties ($k = 2$). The solid blue line indicates the average activity concentration. The solid red line represents the "reference activity" concentration determined by IRMM (CRP). Corresponding dashed lines represent the expanded uncertainty ($k = 2$) of the "reference value" and the standard deviation of the reported data. Numbers indicate the laboratory code.

Anthropogenic ^{134}Cs was reported six times (Fig. 33) and its presence in the sample results from the soil origin – the area affected by the Chernobyl accident. In the original reference sheet of the reference material IAEA-375 (IAEA, 2000), ^{134}Cs is among the certified radionuclides with the recommended value (463 ± 9) Bq·kg^{-1} for the reference date 31 December 1991. Recalculating this value for the date 1 January 2010, the activity concentration of ^{134}Cs is equal to (1.10 ± 0.02) Bq·kg^{-1}. The activity concentration of ^{134}Cs was also measured at IRMM and its value was determined to be (1.42 ± 0.07) Bq·kg^{-1}. The average value calculated from the six reported results is 1.22 Bq·kg^{-1} with standard deviation 0.37 Bq·kg^{-1} which agrees with the reference value.

Two very different activity concentrations of ^{241}Am were reported: (0.24 ± 0.05) Bq·kg^{-1} and (5.6 ± 0.9) Bq·kg^{-1} by laboratories 5 and 15, respectively. More detailed information on the additional radionuclides can be found in Appendix 11.

4.4 Participants' comments

Within the questionnaire participants had the opportunity to express their difficulties with the measurements and share their comments on ILC. Most often they expressed difficulties with the determination of ^{235}U by gamma-ray spectrometry where the ^{226}Ra peak and Compton backscatter peak interfere. Possible solutions to this problem can be found in chapter 5.3 (subchapter of ^{226}Ra).

Several participants complained about insufficient amounts of the soil sample. The amount of the intercomparison material was limited to maximum two bottles (~ 500 g) per laboratory.

We believe that this amount of sample should be sufficient for the determination of all requested radionuclides.

Some laboratories reported problems with the dissolution of the soil material and one participant reported problems with the determination of the water content. Another participant had difficulties with the interference of the high content of ^{137}Cs in the sample in the measurement of ^{90}Sr. In this case, the method used for the Sr separation (combination of precipitation and Sr resin) was most probably not selective enough for Sr.

Laboratories 67 and 5 expressed their doubts about the homogeneity of the ^{90}Sr and in general, respectively, in the intercomparison material. However, only one laboratory reported to us the detection of a hot particle in one of six sample aliquots used. This sample aliquot contained Pu and Am in large excess (16 $Bq{\cdot}kg^{-1}$ in a 10 g aliquot) and traces of ^{244}Cm (0.05 $Bq{\cdot}kg^{-1}$). This information and other observations led us to withdraw the reference values for Pu isotopes. This laboratory suggested, for future ILCs, to avoid using samples taken from the areas influenced by the Chernobyl accident or a reprocessing plant because of the possible presence of hot particles and hence insufficient uniform distribution of the corresponding radionuclides. However, we believe that samples from these areas are of the highest interest and relevance for most of the monitoring laboratories. The sufficient homogeneity of the intercomparison material is guaranteed by the proper homogenization treatment. And in case of the presence of hot particles, the needed interventions will be made, as in the case of Pu isotopes in this ILC.

Several participants expressed their interest to participate also in future ILCs and would welcome samples with various types of matrix. They appreciate this kind of exercises with the emphasis on the possible future improvement and strengthening of their measurement routines. One laboratory stated also the need of more workshops on ILCs. Another participant found this particular ILC challenging and interesting due to the measurement of several radionuclides which are not determined in their laboratory on the routine basis.

One participant complained about too many questions in the questionnaire without a clear purpose. We believe that this comprehensive report proves the benefit of all the questions asked.

5 Evaluation and comparison of data

Initially, the results were tested for normality and presence of outliers. However, the outlying values were not discarded and were included in further evaluations. 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. Individual laboratory performance is expressed in terms of relative deviations and E_n numbers (ISO, 2005a). The "PomPlot" graphical method is used for producing a summary overview of the participants' results (Spasova et al., 2007).

5.1 Identification of outliers and normal distribution check

The presence of statistical outliers among the reported results was investigated using Grubb's test at a level of significance $\alpha = 1$ %, as suggested in ISO/IEC 5725-2 (1994). Only in the case of two radionuclides, ^{212}Bi and ^{214}Pb, no extreme values were found. The frequency histograms and normal probability plots (Figs. 34-35) showed that the data of these two radionuclides are unimodally and normally distributed.

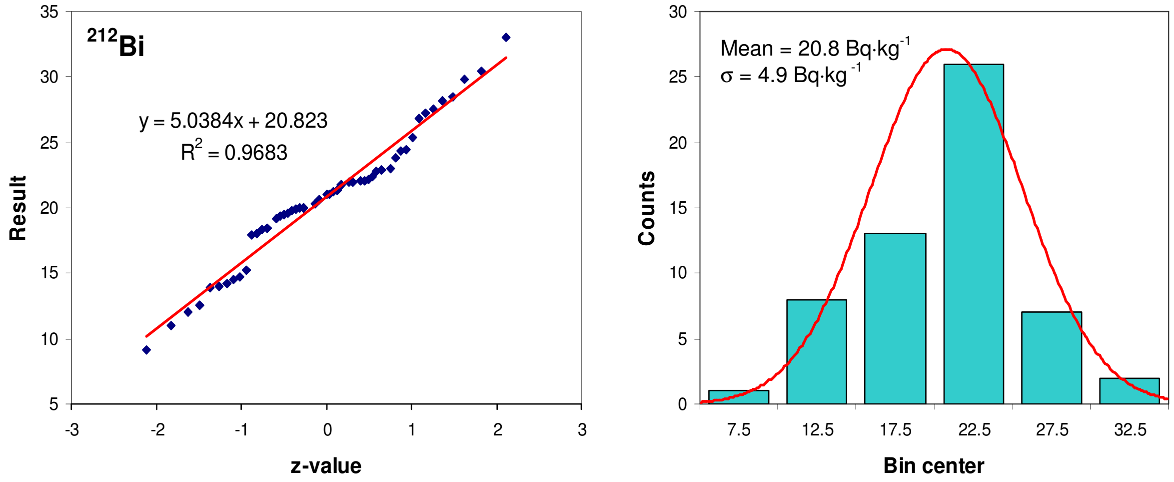

Fig. 34. Normal probability plot and frequency histogram of the ^{212}Bi data.

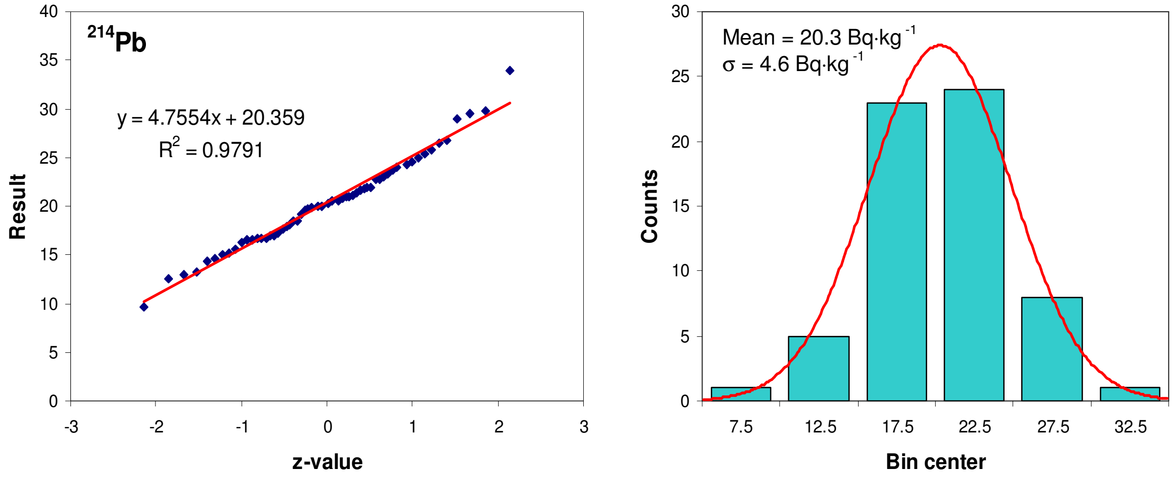

Fig. 35. Normal probability plot and frequency histogram of the ^{214}Pb data.

For ^{214}Bi, the result of laboratory 72 was flagged as an outlier in the first instance. The reported value was about twice the average value reported by other participants. No further outliers were identified on statistical grounds. After exclusion of the outlier the normal probability plot showed (Fig. 36) that the data are normally distributed.

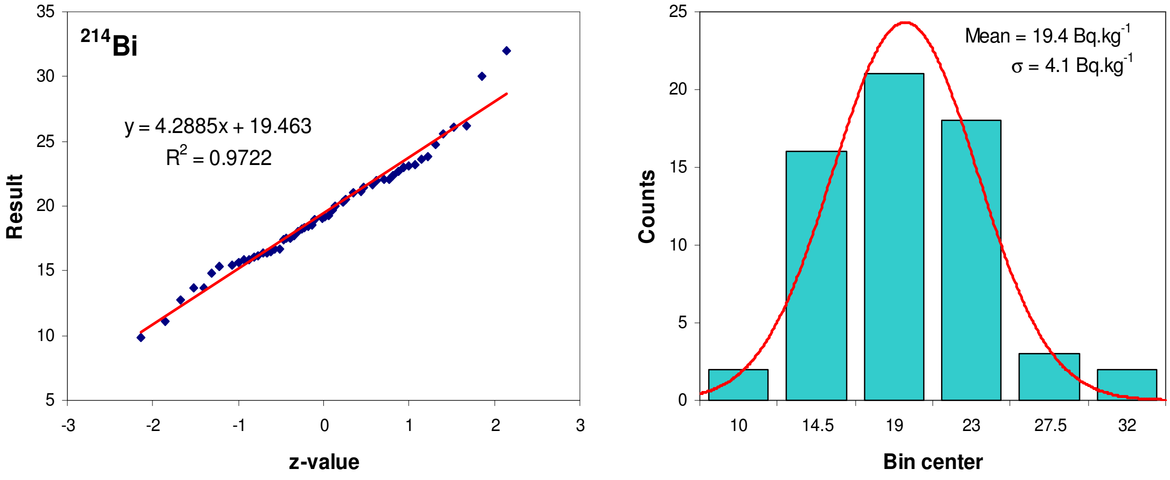

Fig. 36. Normal probability plot and frequency histogram of the ^{214}Bi data after exclusion of the outlier.

In the dataset of ^{212}Pb, the first run of the Grubb's test identified one outlier (lab 53). In the second and third run laboratories 26 and 42, respectively, were tagged as outliers. All three extreme results were overestimated in comparison to the average of reported values. The data distribution can be seen in Fig. 37.

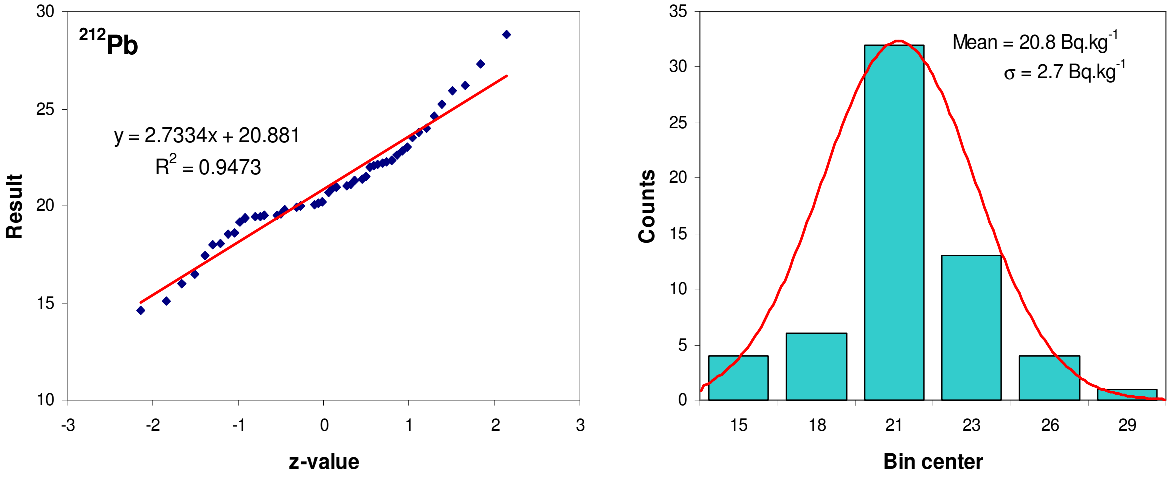

Fig. 37. Normal probability plot and frequency histogram of the ^{212}Pb data after exclusion of the three outliers.

For ^{137}Cs, four results were indicated as outliers by the Grubb's test. In the first round of testing it was the result of laboratory 31 with very low value. In the second round results of laboratories 44 and 72 were indicated as extreme values and in the third round, it was the result of laboratory 26. The data set is unimodal and normally distributed after the exclusion of the outliers as shown in Fig. 38.

In the case of ^{40}K, in the first round of testing the result of laboratory 72 was pointed out as an outlier and in the second round the result of laboratory 42 was indicated. The result of laboratory 44 was pointed out as an outlier in the third round of Grubb's test. Again, the frequency histogram and normal probability plot (Fig. 39) showed that the data are unimodally and normally distributed.

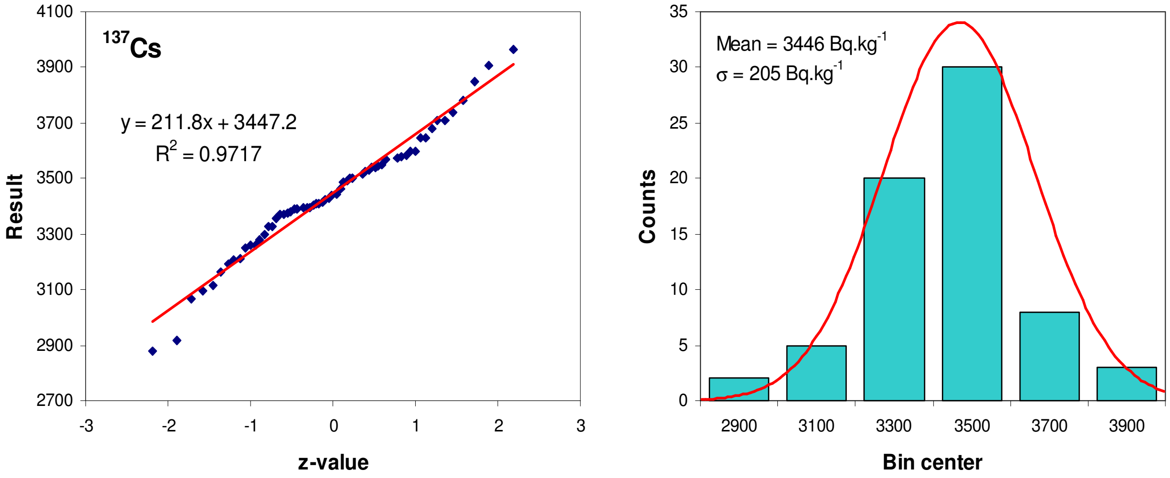

Fig. 38. Normal probability plot and frequency histogram of the ^{137}Cs data after exclusion of the four outliers.

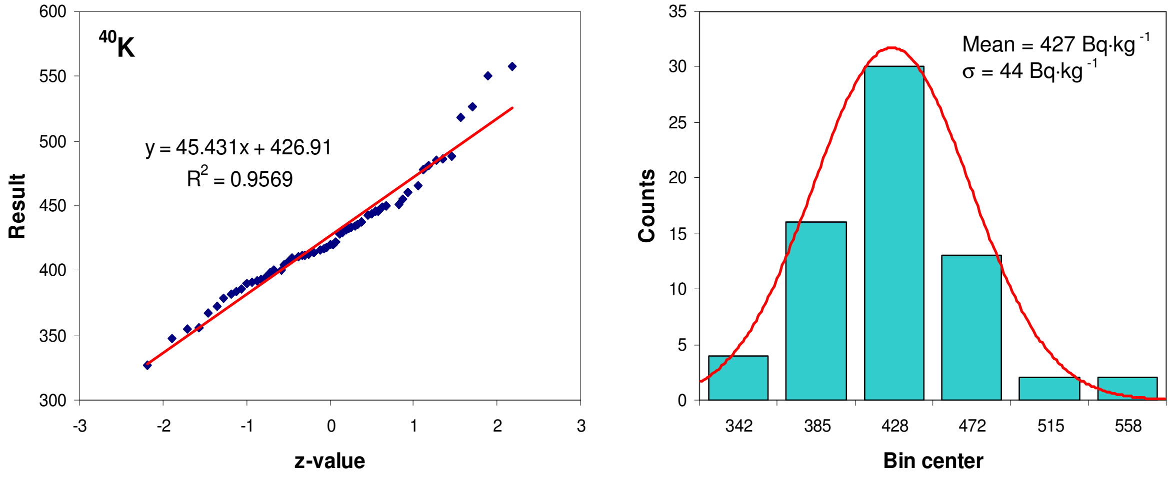

Fig. 39. Normal probability plot and frequency histogram of the ^{40}K data after exclusion of the three outliers.

Due to the possible presence of hot particles no reasonable statistical evaluation could be done for the plutonium isotopes. After the announcement of the preliminary results laboratory 15 realised that for plutonium isotopes they submitted the results concerning a reference sample instead of the intercomparison sample and asked for the correction. However, we could not replace nor withdraw their result in that stage of ILC. This is an example of the importance of attentive and careful reporting.

The results of laboratories 72 and 39 were detected as outliers in the first round of testing for ^{226}Ra. In the second round the result of laboratory 14 was marked as an outlier. The frequency histogram and normal probability plot confirmed that the data are unimodally and normally distributed as can be seen in Fig. 40.

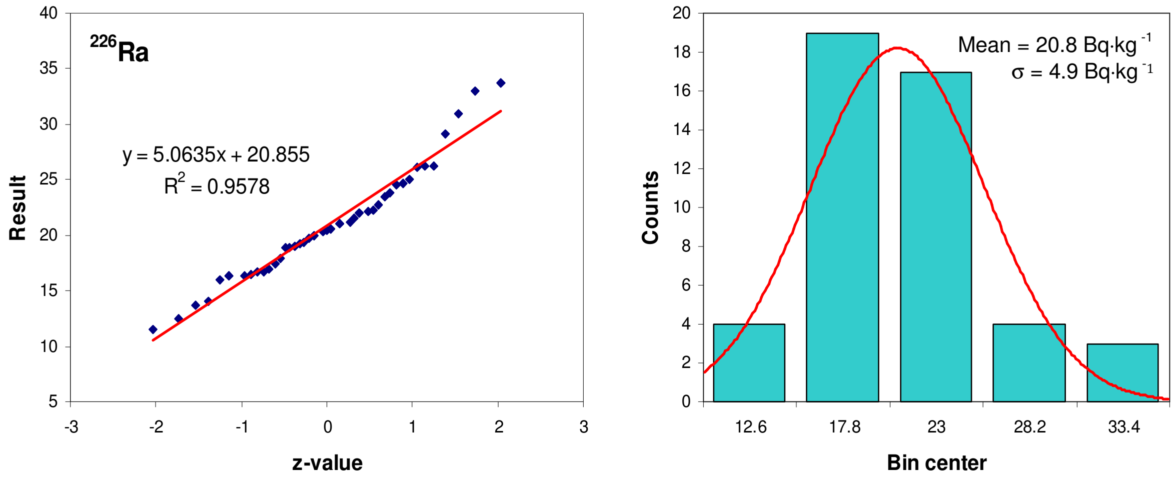

Fig. 40. Normal probability plot and frequency histogram of the ^{226}Ra data after exclusion of the three outliers.

Only one outlier was indicated among the reported data of ^{90}Sr activity concentrations. A too high value was reported by laboratory 28. Both, frequency histogram and normal probability plots (Fig. 41) after exclusion of the outlier showed unimodal and normal distribution of the data.

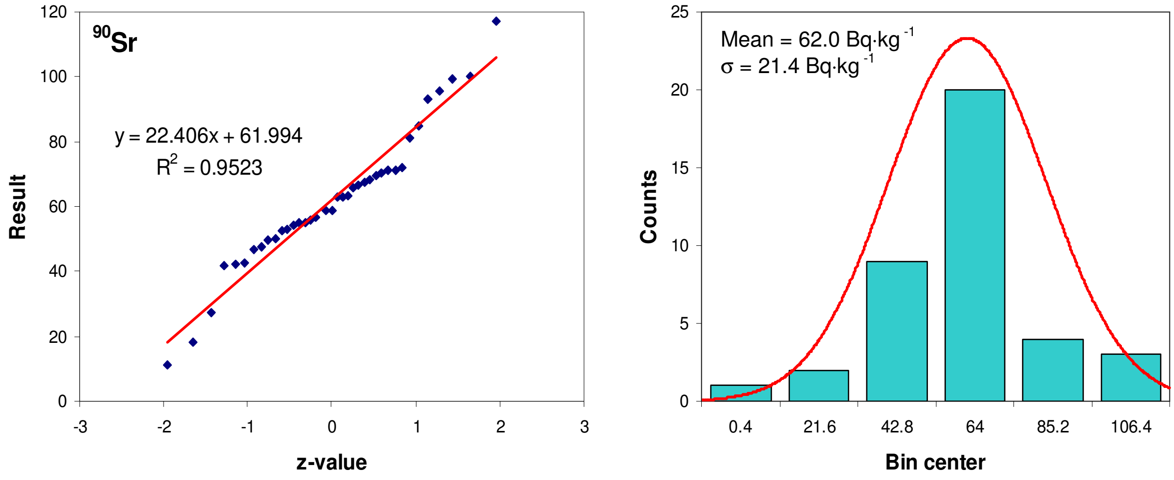

Fig. 41. Normal probability plot and frequency histogram of the ^{90}Sr data after exclusion of the outlier.

One extreme value (lab 63) was indicated in the ^{230}Th data set. The data of ^{230}Th activity concentration show normal distribution, however, the frequency histogram reveals a bimodal distribution (Fig. 42). In the case of ^{232}Th, three values (labs: 31, 43, 42) were identified as outliers in three consecutive runs of the Grubb's test. The normal probability plot is not perfectly straight and the frequency diagram shows the distribution to be left-skewed (Fig. 43). This is due to a few data with lower values in comparison to the average.

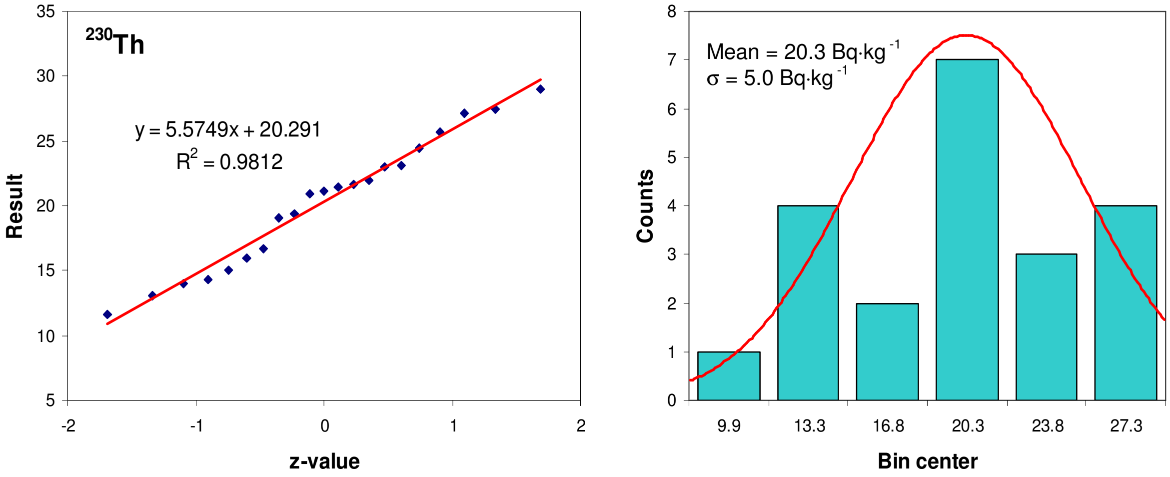

Fig. 42. Normal probability plot and frequency histogram of the ^{230}Th data after exclusion of the outlier.

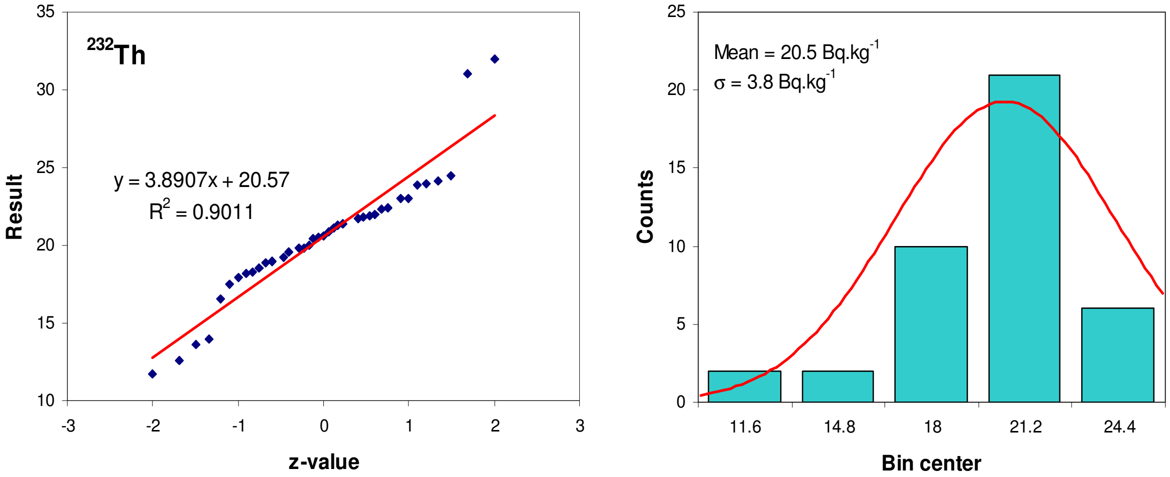

Fig. 43. Normal probability plot and frequency histogram of the ^{232}Th data after exclusion of the three outliers.

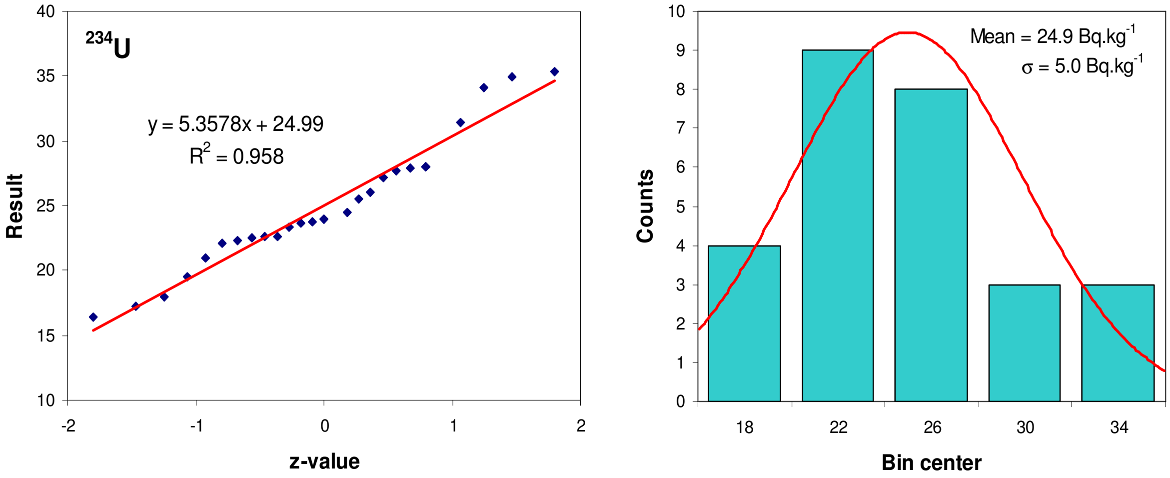

Fig. 44. Normal probability plot and frequency histogram of the ^{234}U data after exclusion of the outlier.

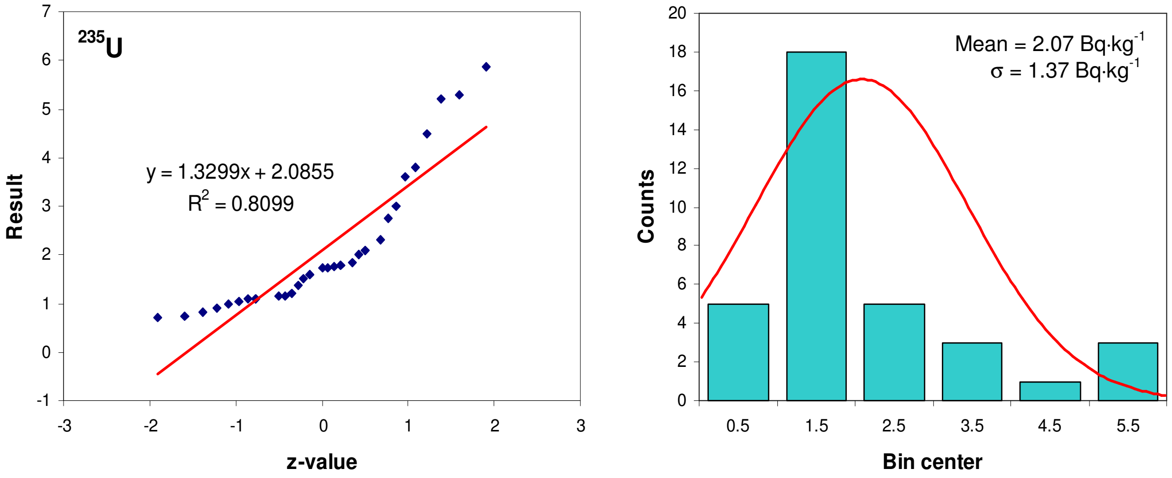

Fig. 45. Normal probability plot and frequency histogram of the ^{235}U data after exclusion of the three outliers.

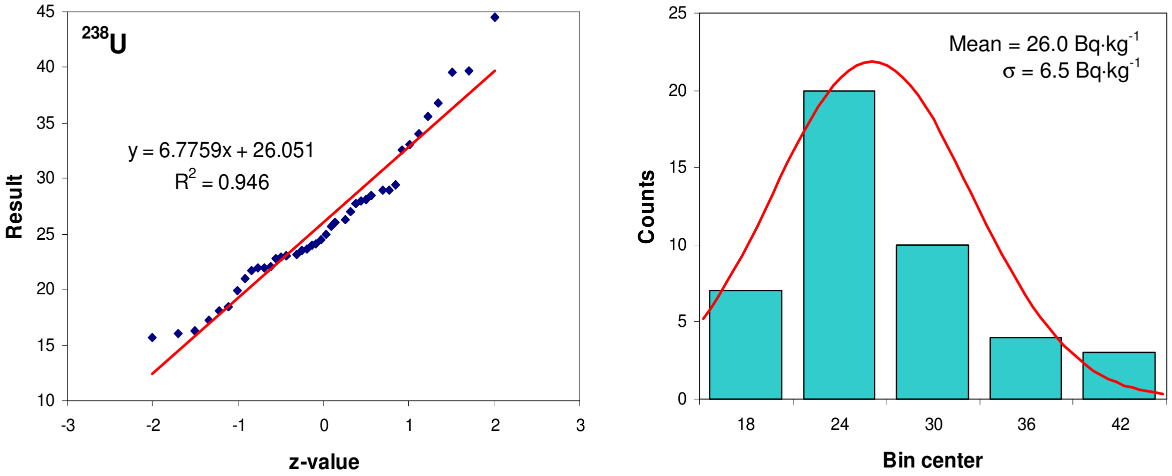

Fig. 46. Normal probability plot and frequency histogram of the ^{238}U data after exclusion of the six outliers.

For ^{234}U, one outlying value was identified (lab 63). The normal probability plot shows Gaussian distribution as shown in Fig. 44. In three consecutive runs of the Grubb's test the results of laboratories 72, 17 and 8 were indicated as outliers for ^{235}U. In the case of ^{238}U, six outliers were identified. Values reported by laboratories 37 and 58 were indicated in the first round, laboratories 31 and 73 were indicated in the second round, laboratories 34 and 43 in the third round of the test. According to the normal probability data plots the distribution of ^{235}U and ^{238}U is more or less normal. However, the frequency plots show slightly right-skewed distribution of the data for both uranium isotopes (Fig. 45-48).

Although some results were indicated as outliers, they are removed only for calculation of the summary statistics. These results are still evaluated and are given the appropriate performance ratings. In Table 12 some basic results of the statistical analysis are presented.

Table 12. Statistical analysis of the laboratory results in soil.

		^{40}K	^{90}Sr	^{137}Cs	^{212}Pb	^{212}Bi	^{214}Pb	^{214}Bi	^{226}Ra	^{230}Th	^{232}Th	^{234}U	^{235}U	^{238}U	^{238}Pu	$^{239+240}$Pu
All data	Num. of results	70	40	72	63	57	62	63	50	22	46	28	38	50	17	24
	Minimum	258	11	796	15	9.1	9.6	9.9	12	12	0.16	16	0.7	0.023	0.034	0.22
	Maximum	1273	175	4400	34	33	34	41	52	60	64	44	57	98	34	100
	Median	420	61	3435	21	21	20	19	21	21	21	24	1.8	26	0.13	0.30
	Mean	439	65	3390	21	21	20	20	22	22	21	26	4.2	31	2.43	4.7
	Standard deviation	115	28	432	3.6	4.9	4.6	4.9	7.7	9.8	8.2	6.1	9.2	17	8.4	20
Without outliers (5%)	Num. of outliers	3	1	4	3	0	0	1	3	1	2	1	3	4	1	7
	Mean	427	62.0	3446	20.8	20.8	20.3	19.4	20.8	20.3	20.8	24.9	2.07	26.0	0.477	0.28
	Standard deviation	44	21.4	205	2.7	4.9	4.6	4.1	4.9	5.0	4.3	5.0	1.37	8.4	0.930	0.04
Without outliers (1%)	Num. of outliers	3	1	4	3	0	0	1	3	1	3	1	3	6	5	7
	Mean	427	62.0	3446	20.8	20.8	20.3	19.4	20.8	20.3	20.5	24.9	2.07	26.0	0.090	0.28
	Standard deviation	44	21.4	205	2.7	4.9	4.6	4.1	4.9	5.0	3.8	5.0	1.37	6.5	0.040	0.04
	Ref. value	410	74.5	3565	21.0*	21.0*	19.0*	19.0*	19.0	20.9	21.0	25.2	1.10	24.3	-	-
	Expanded unc.(*k*=2)	21	10.1	134	-	-	-	-	2.3	2.3	2.7	1.0	0.11	1.0	-	-
	Rel. exp. unc.(*k*=2)	5%	13%	4%	-	-	-	-	12%	11%	13%	4%	10%	4%	-	-

* indicative value

5.2 Scores and evaluation criteria

Results of the participating laboratories were evaluated against the reference values using three different approaches: relative deviations, E_n numbers and PomPlots. Details on these methods are described in this chapter.

Relative deviations

The relative deviations (the percentage differences in ISO 13528:2005 (ISO, 2005a)) are calculated as

$$D_{\%} = 100 \frac{A_{lab} - A_{ref}}{A_{ref}} \tag{8}$$

where A_{lab} is the participant's result, mean activity concentration;
A_{ref} is the reference value.

These values are plotted in ascending order in deviation charts and the laboratories reporting too low or too high values become more visible. For the environmental radioactivity measurements the criterion of ± 20 % from the reference value is usually used.

E_n numbers

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}) (ISO, 1997; ISO, 2005a). Strictly speaking, tests including measurement uncertainty should be used with caution when participants may have poor understanding of the uncertainty estimation. We have already observed (chapter 4.2) that this is applicable for about one third of the participating laboratories. Nevertheless, the selected performance test using E_n numbers proves to be robust enough justifying its use in this evaluation. Moreover, incorporating information on uncertainty into the interpretation of results can play a major role in improving the understanding of this difficult subject (ISO, 2005a).

The performance statistic E_n number is calculated as

$$E_n = \frac{A_{lab} - A_{ref}}{\sqrt{U_{lab}^2 + U_{ref}^2}} \tag{9}$$

where A_{lab} is the participant's result, mean activity concentration;
A_{ref} is the reference value;
U_{lab} is the expanded uncertainty of the participant's result;
U_{ref} is 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, 2008), 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} with about 95 % probability. Therefore, E_n numbers are interpreted in the following way:

$|E_n| \leq 1$, satisfactory, the laboratory values are compatible with the reference value (green colour in E_n numbers' charts);

$\lvert E_n \rvert > 1$,	unsatisfactory, “warning signal”, the laboratory values differ significantly from the reference value, sources of deviation should be investigated and corrected (orange colour in E_n numbers' charts);

In analogy to the interpretation of zeta-scores, a second level of critical value can be defined:

$\lvert E_n \rvert > 1.5$,	“action signal”, there is urgent need to investigate and find the sources of the large deviation (red colour in E_n numbers' charts).

PomPlots

In order to compare the results, a modern type of graph – PomPlot – that underlines the importance of the assigned uncertainties is applied. The PomPlot, an intuitive graphical method, is used for producing a summary overview of the participants' results (Spasova et al., 2007). It displays the relative deviations (D/MAD) of the individual results A_{lab} from the reference value A_{ref} on the horizontal axis and relative uncertainties (u/MAD) on the vertical axis (Fig. 47). For both axes, the variables are expressed as multiples of MAD, which is defined as the median of absolute deviation from the reference value

$$MAD = Median\left|D_i\right|, (i = 1, \ldots, n) \qquad (10)$$

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

$$D_i = A_{lab,i} - A_{ref} \qquad (11)$$

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

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

$$u_i^2 = u_c^2\left(A_{lab,i}\right) + u_c^2\left(A_{ref}\right) \qquad (12)$$

where $u_c\left(A_{lab,i}\right) = U_{lab,i}/k$ and $u_c\left(A_{ref}\right) = U_{ref}/k$ (13)

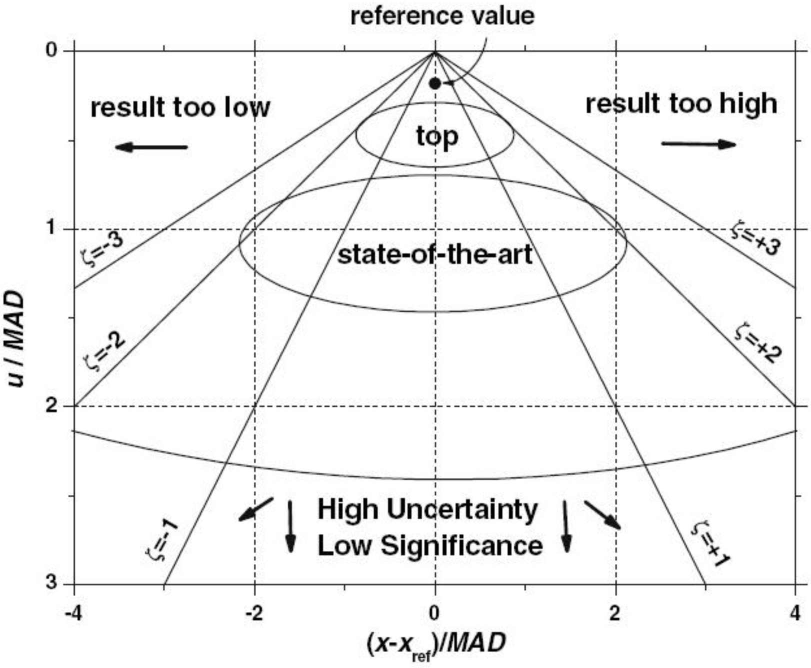

Fig. 47. Interpretation of a PomPlot (Spasova et al., 2007).

The ζ-scores, $\left|\xi\right| = \left|D/u\right| =$ 1, 2 and 3, are represented by diagonal solid lines, creating the aspect of a pyramidal structure. The ζ-score is a measure for the deviation between laboratory result and reference value relative to total uncertainty (ISO, 2005a). The points 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 uncertainty is small, 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 $\zeta = \pm 3$ lines are probably inconsistent with the reference value.

5.3 Evaluation of laboratory performances

Above mentioned statistical tools were used to evaluate the performances of the participating laboratories. This chapter is divided into sub-chapters according to the radionuclides or groups of radionuclides.

^{40}K and ^{137}Cs

^{40}K and ^{137}Cs are the most often determined radionuclides in environmental samples like soil or sediments. Out of 73 participants, 70 and 72 reported results for ^{40}K and ^{137}Cs, respectively. Both are usually determined via gamma-ray spectrometry. Only one laboratory (31) determined the activity concentration of ^{137}Cs via beta decay.

The determination of ^{40}K and ^{137}Cs is the least problematic among all analysed radionuclides with 11 % and 6 % of the results, respectively, deviating more than 20 % from the reference values. However, few laboratories reported too low values for ^{137}Cs (Fig. 51) and, on the contrary, some too high values for ^{40}K (Fig. 48) were submitted. The results of some laboratories (26, 44, 72; lab 31 did not report a result for ^{40}K) deviated more than 20 % from the reference values for both radionuclides although they used routine analytical procedures. In particular, laboratory 72 reported a value of ^{40}K higher by 210 % from the reference value, and for ^{137}Cs a value lower by 38.5 %. The methods used in these laboratories should be thoroughly investigated and the source of error identified. It is quite obvious that background correction for ^{40}K from the laboratory environment and efficiency calibration at the relatively high gamma-ray energy of ^{40}K need special attention.

In terms of E_n numbers, 71 % of the results are compatible with the reference value for ^{40}K, 11 % of results trigger a warning signal and 18 % an action signal. In the case of ^{137}Cs, also 71 % of the laboratory values are satisfactory, while 13 % trigger a warning signal and 16 % trigger an action signal. The evaluation of ^{40}K and ^{137}Cs results based on the E_n criterion (Fig. 49 and 52) and its comparison with the relative deviations (Fig. 48 and 51) shows that several laboratories underestimated their uncertainties. Although, their activity concentrations are acceptable within 20 % from the reference values, their performance in terms of E_n numbers is triggering the action signal (labs: 8, 11, 19, 24, 43, 45 for ^{40}K and labs: 10, 11, 16, 23, 40, 47, 58, 69, 71 for ^{137}Cs).

As presented in the PomPlots (Fig. 50 and Fig. 53), even though the results of ^{40}K and ^{137}Cs are generally very good, there are many points outside the $|\xi| = 1$, 2 and 3 indicating that a large fraction of the laboratories underestimates the uncertainties.

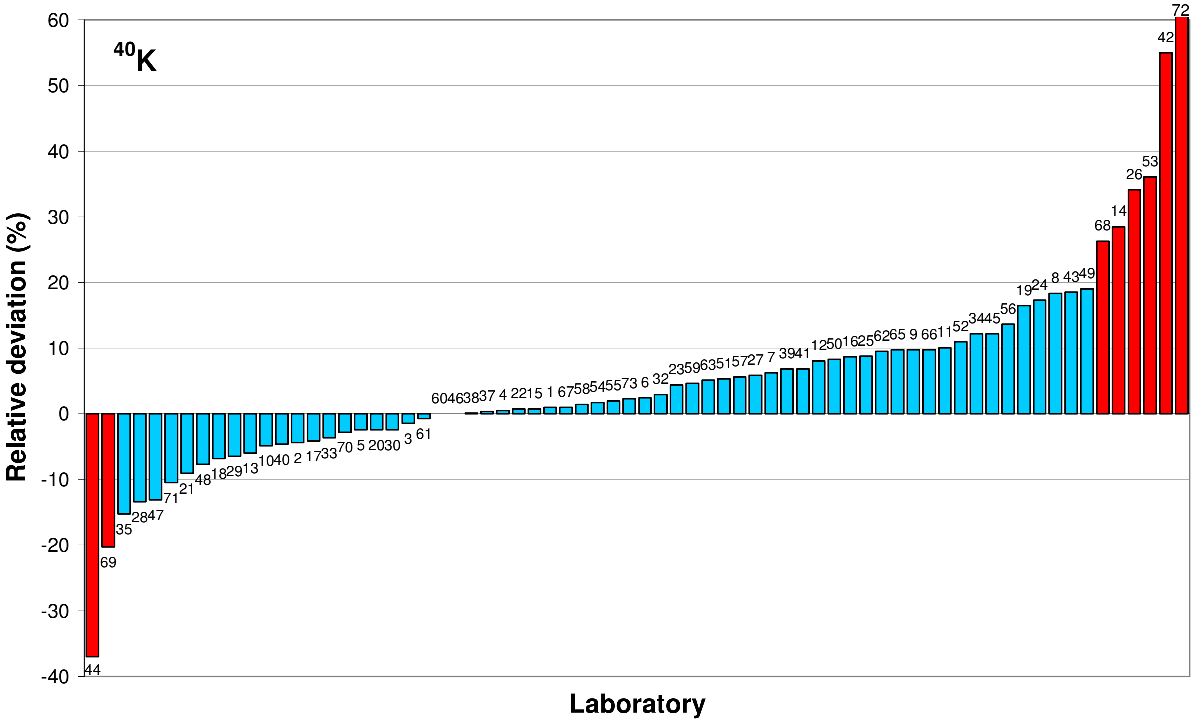

Fig. 48. Deviation chart of the participants' results of ^{40}K plotted in ascending order. Blue colour indicates results within the range ± 20 % from the reference value and red indicates results outside this range. Numbers indicate the laboratory code.

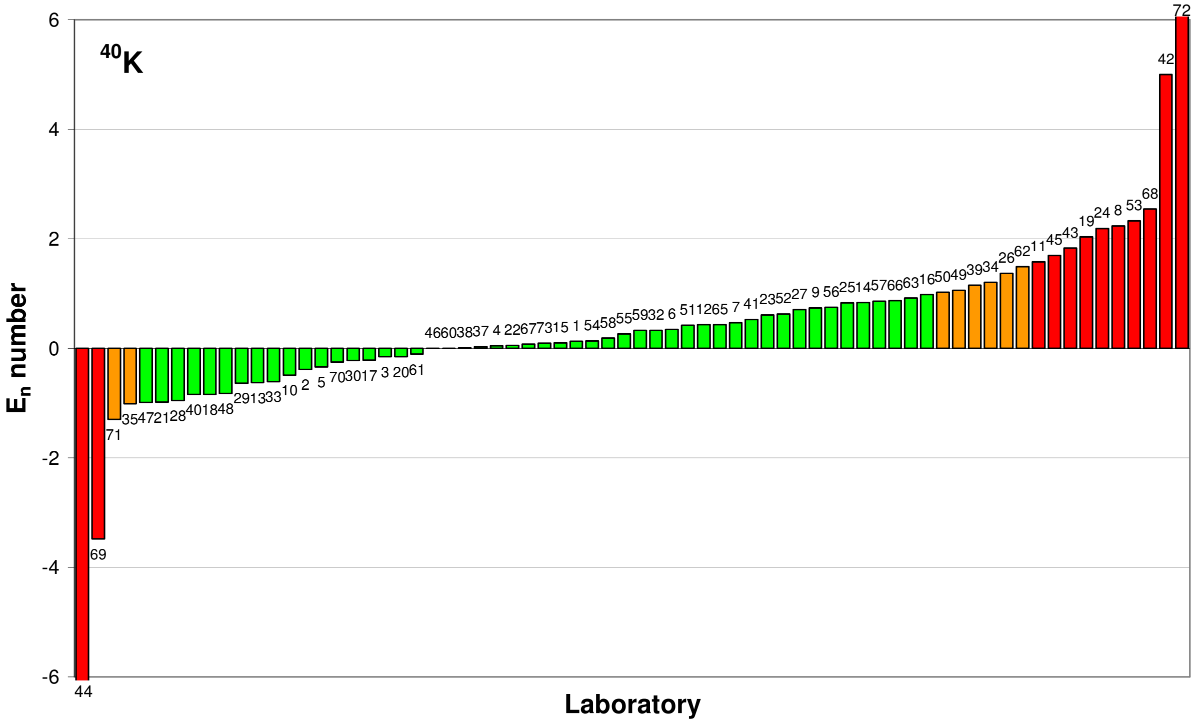

Fig. 49. E_n numbers' chart of ^{40}K activity concentrations plotted in ascending order. Green colour indicates compatible results, orange indicates warning signal and red indicates action signal. Numbers indicate the laboratory code.

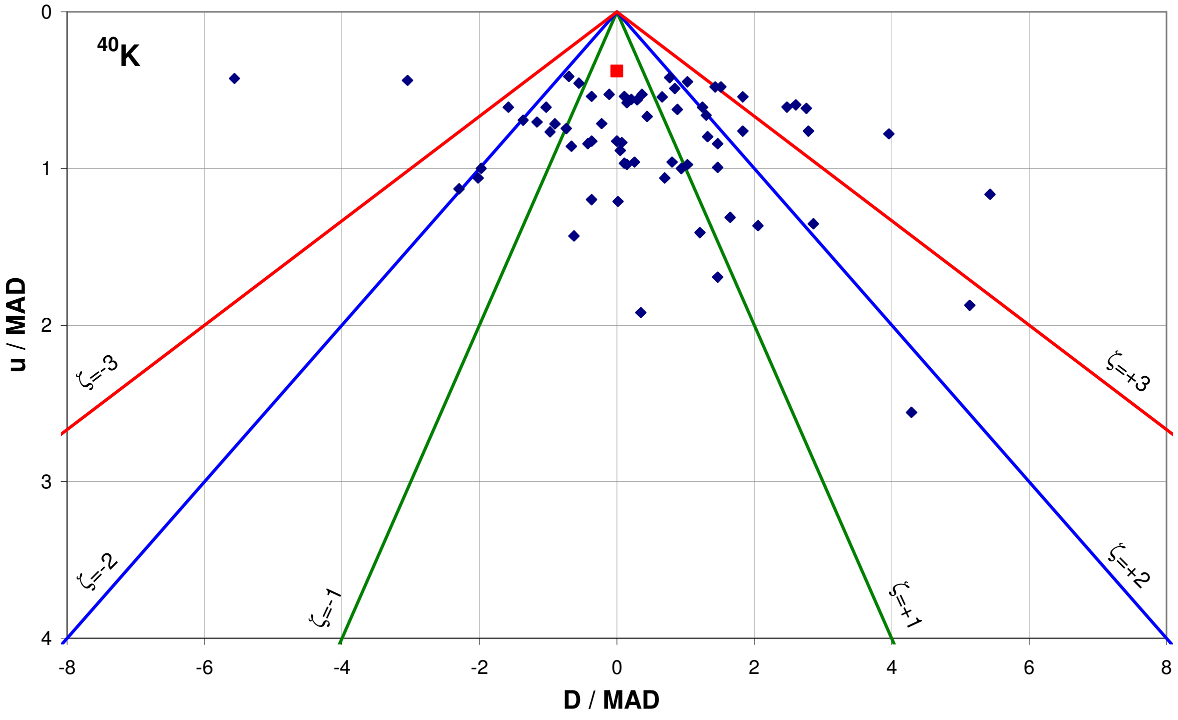

Fig. 50. PomPlot of the ^{40}K data. Red point indicates the reference value. Green, blue and red solid lines indicate ζ-scores = 1, 2 and 3, respectively.

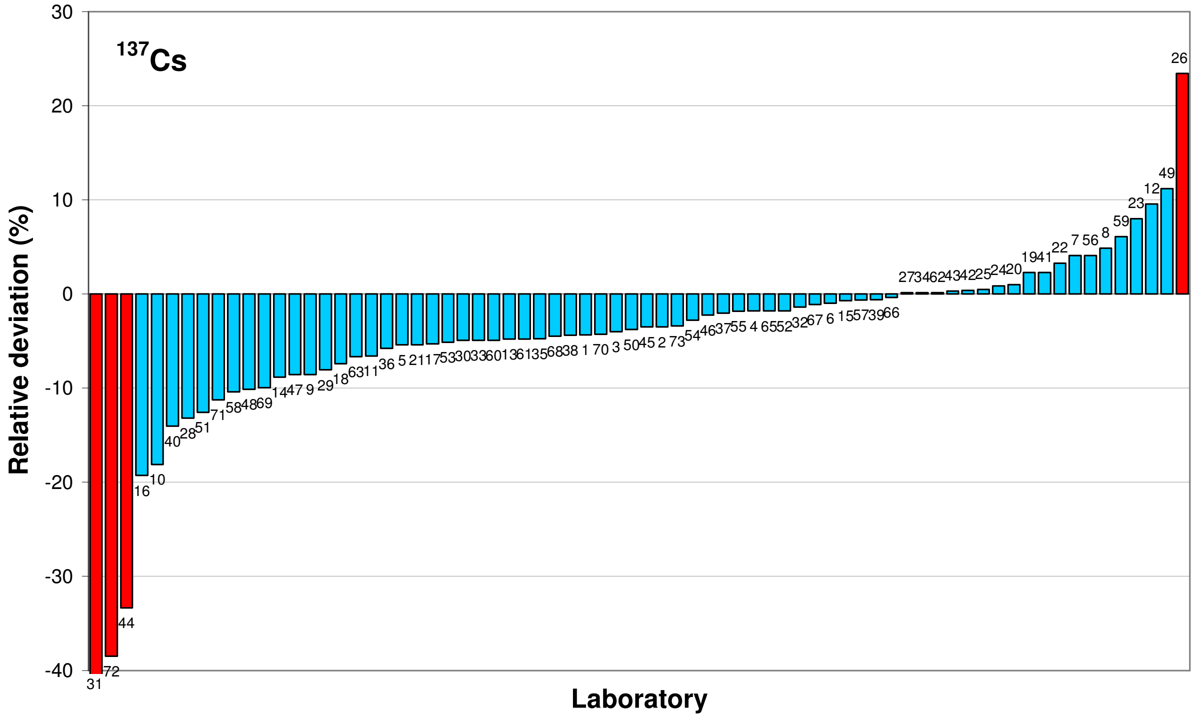

Fig. 51. Deviation chart of the participants' results of ^{137}Cs plotted in ascending order. Blue colour indicates results within the range ± 20 % from the reference value and red indicates results outside this range. Numbers indicate the laboratory code.

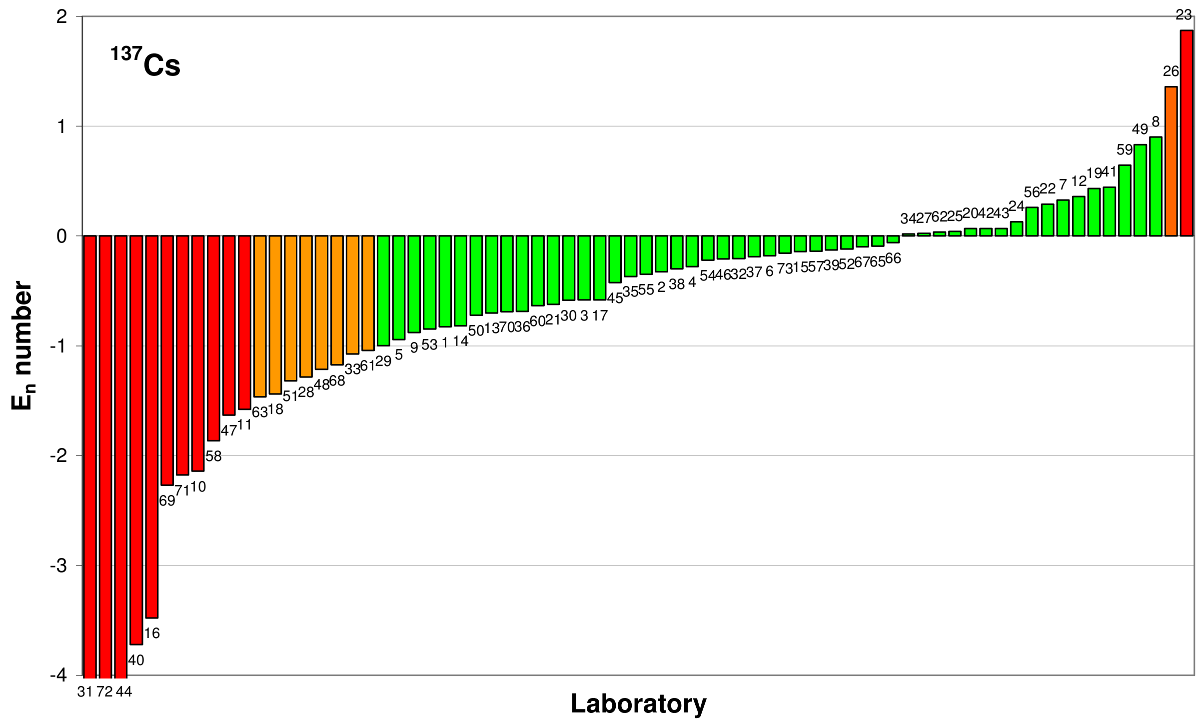

Fig. 52. E_n numbers' chart of ^{137}Cs activity concentrations plotted in ascending order. Green colour indicates compatible results, orange indicates warning signal and red indicates action signal. Numbers indicate the laboratory code.

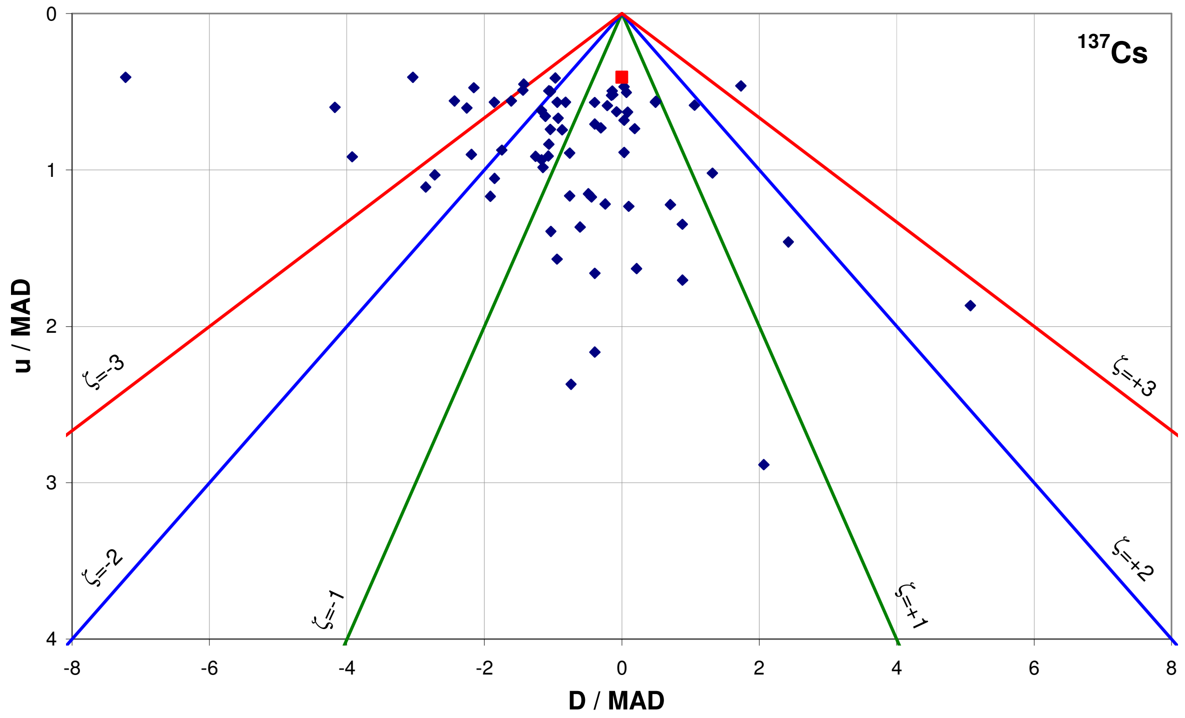

Fig. 53. PomPlot of the ^{137}Cs data. Red point indicates the reference value. Green, blue and red solid lines indicate ζ-scores = 1, 2 and 3, respectively.

^{90}Sr

The determination of ^{90}Sr activity concentrations obviously presents difficulties in the majority of laboratories, since 65 % of the ^{90}Sr results deviate more than 20 % from the reference value as can be seen in Fig. 54. Even if taking into account the high complexity of these measurements, and using a less strict criterion (30 %), only 65 % of the participants' results would fall within 30 % from the reference value. This is even worse performance than what was observed in a previous ILC of milk powder where about 23 % of laboratories deviated more than 30 % from the reference value (Spasova et al., 2008). The difference may be attributed to the easier extraction of strontium from the milk powder compared to the soil material.

Based on the E_n criterion (Fig. 55), two thirds of all results triggered action and warning signals, 42 % and 23 %, respectively. Four laboratories (3, 11, 20, 59) determined the ^{90}Sr concentrations within 30 %; however they underestimated their uncertainties and therefore failed in terms of the E_n numbers. For example, the relative expanded uncertainty reported by laboratory 11 was only 2 % which is a highly unrealistic value.

Among all radionuclides compared in this exercise, ^{90}Sr was determined by the highest number of different measurement techniques. Although difficult due to the variety and complex nature of separation methods, an attempt was made to group the results by separation methods (Fig. 56). In each group there are outlying values but only for some methods there are also results within the uncertainty of the reference value. The precipitation technique appears to be slightly more reliable as can be seen also in the PomPlot (Fig. 57) where actually only results obtained by this separation method are situated close to the top of the pyramid. Nevertheless, some laboratories obtain far too low results also with this method. All other separation techniques rendered unsatisfactory results. As can be seen in Fig. 56, only single results (labs: 22 and 33) are compatible in terms of E_n numbers when the Sr resin or the combination of precipitation with Sr resin were applied. The result of laboratory 5, which used the AG chromatography column followed by oxalate precipitation, is 14 % higher than the reference value and is compatible in terms of E_n number. However, we can not draw any final conclusion since it is the only value obtained via this method. The method of solvent extraction showed the worst results with no value within 20 % from the reference value and no compatible E_n number. Unfortunately, several laboratories (4, 26, 27, 49, 51, 52, 54, 65, 70) did not provide information on the separation methods used. Nevertheless, almost 60 % of the results using the precipitation technique for separation of Sr from the soil matrix deviated less than 20 % from the reference value (Table 13).

The most frequently used counting method was gas flow proportional counting with some outlying results but also with 66 % of results within 30 % from the reference value (Fig. 58). The group of eight laboratories using LSC is the second largest. However, only half of the results lie within ± 30 % from the reference value and only 25 % of values are E_n compatible. All results obtained by LSC (except lab 28) lie below the reference value. Similarly, results obtained with plastic scintillators and Geiger-Müller counting are lower than the reference value but the general performance is more favourable. The Cherenkov counting technique was applied in 3 laboratories and none of these results lies within 30 % from the reference value and only one (lab 33) is E_n compatible. In general, as can be seen in the PomPlot (Fig. 59) and Table 14 no particular counting method can be identified as superior or inferior to others. The reason of the discrepancies must be sought in the individual laboratories concerned.

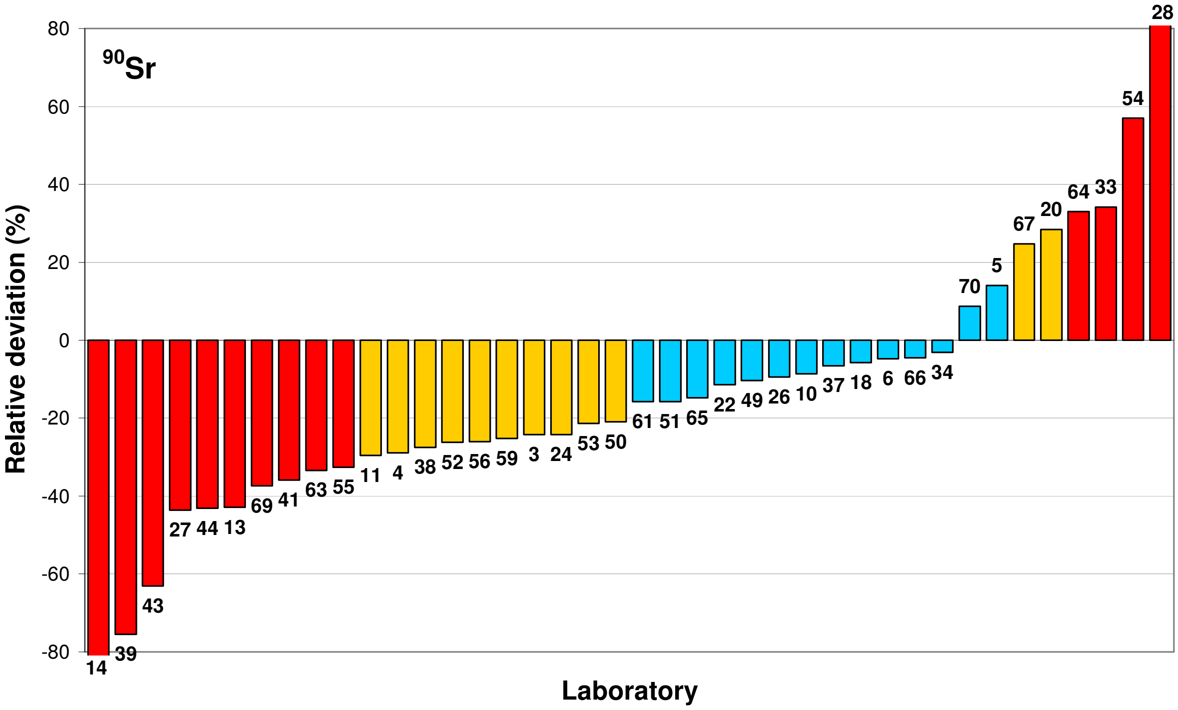

Fig. 54. Deviation chart of the participants' results of ^{90}Sr plotted in ascending order. Blue colour indicates results within the range ± 20 % from the reference value, yellow indicates results between ± 20 % and ± 30 % and red indicates results deviating more than ± 30 %. Numbers indicate the laboratory code.

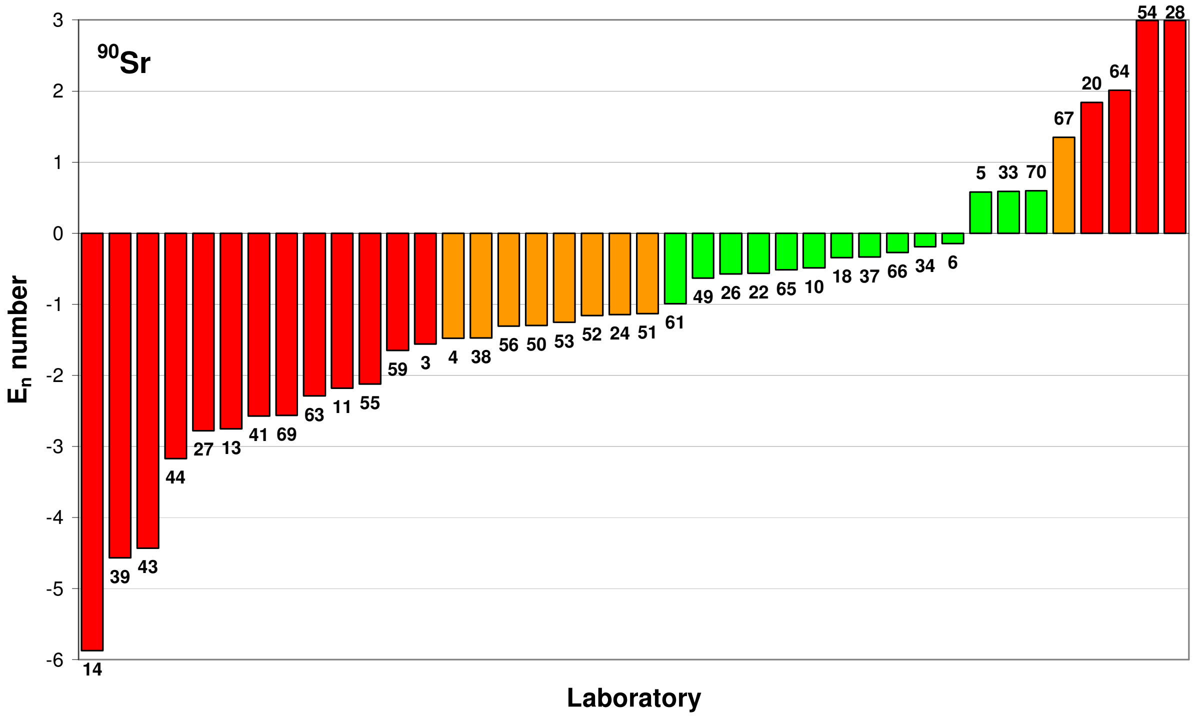

Fig. 55. E_n numbers' chart of ^{90}Sr activity concentrations plotted in ascending order. Green colour indicates compatible results, orange indicates warning signal and red indicates action signal. Numbers indicate the laboratory code.

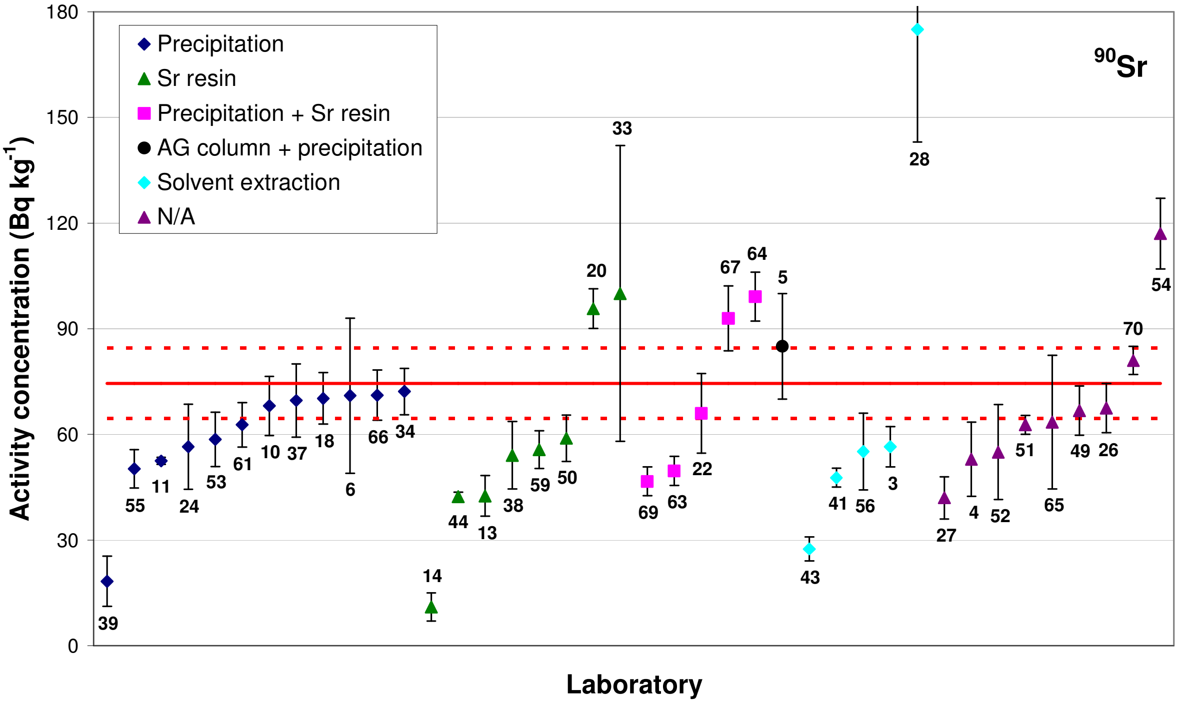

Fig. 56. Laboratory results A_{lab} of ^{90}Sr activity concentration with expanded uncertainties U_{lab} ($k = 2$) sorted according to separation methods. Red lines represent reference value $A_{ref} \pm U_{ref}$ ($k = 2$). Numbers indicate the laboratory code.

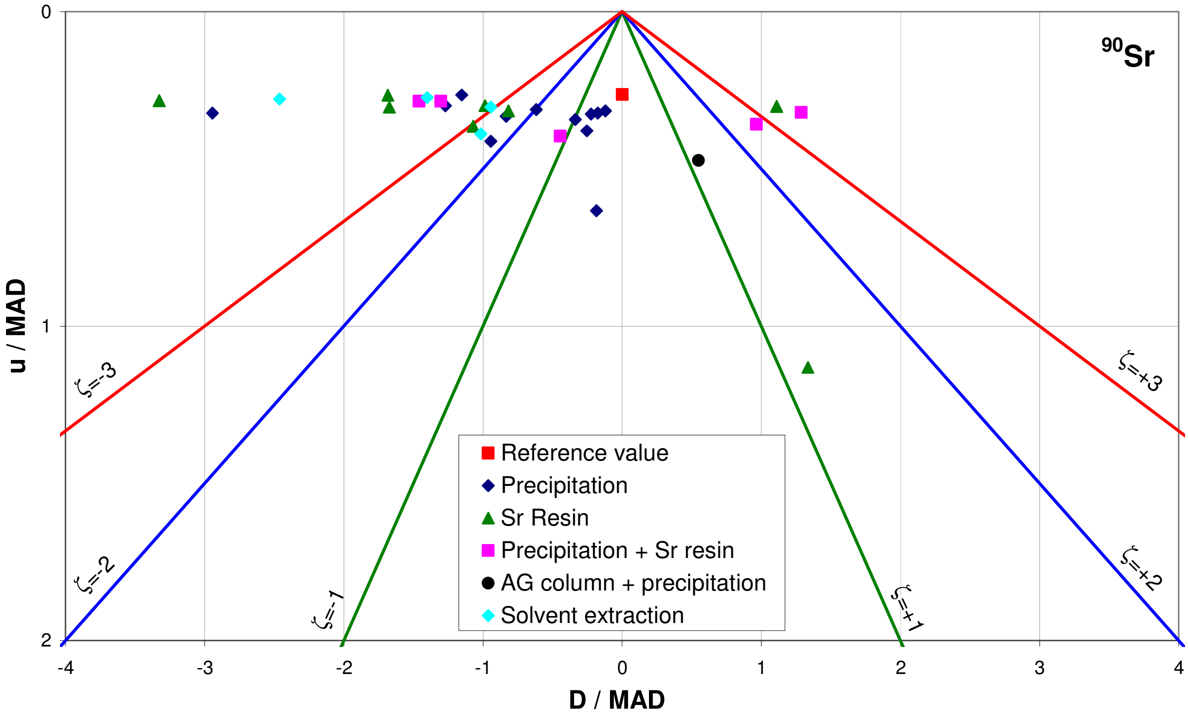

Fig. 57. PomPlot of the ^{90}Sr data sorted according to separation methods. Red point indicates the reference value. Green, blue and red solid lines indicate ζ-scores = 1, 2 and 3, respectively. Values with unidentified separation method are not included in this plot.

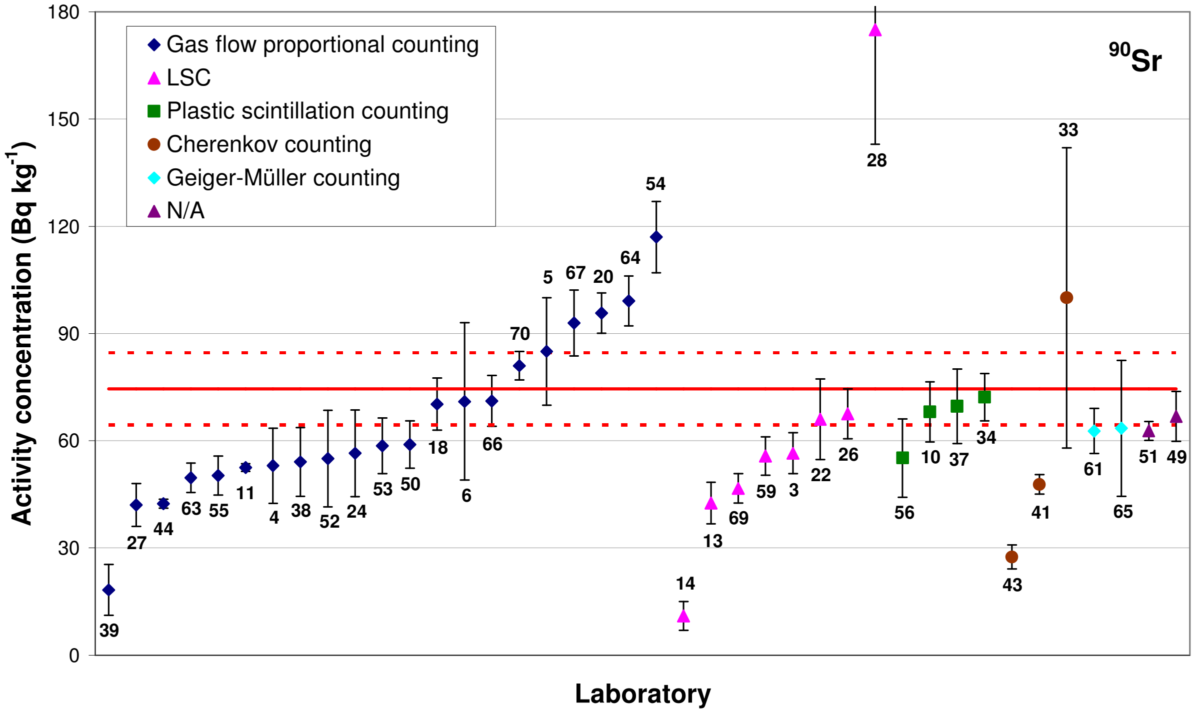

Fig. 58. Laboratory results A_{lab} of ^{90}Sr activity concentration with expanded uncertainties U_{lab} ($k = 2$) sorted according to counting methods. Red lines represent reference value $A_{ref} \pm U_{ref}$ ($k = 2$). Numbers indicate the laboratory code.

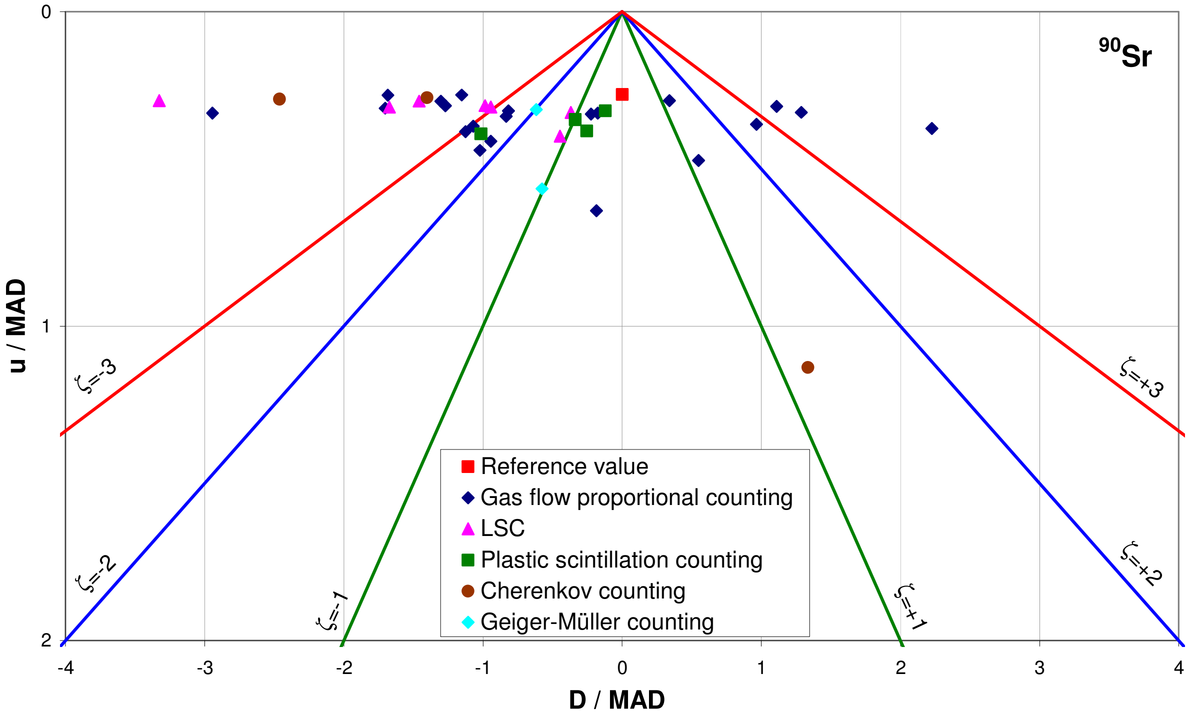

Fig. 59. PomPlot of the ^{90}Sr data sorted according to counting methods. Red point indicates the reference value. Green, blue and red solid lines indicate ζ-scores = 1, 2 and 3, respectively. Values with unidentified counting method are not included in this plot.

Table 13. Overview of the laboratory performances regarding the relative deviation and E_n numbers sorted according to separation methods of ^{90}Sr. The number in parentheses indicates number of laboratories.

	Precipitation (12)	Sr resin (8)	Precipitation + Sr resin (31)	AG + precipitation (1)	Solvent extraction (5)
Within ± 20 %	58 % (7)	0 % (0)	20 % (1)	100 % (1)	0 % (0)
<20 %, 30 %>	25 % (3)	50 % (4)	20 % (1)	0 % (0)	60 % (3)
Outside ± 30 %	17 % (2)	50 % (4)	60 % (3)	0 % (0)	40 % (2)
Compatible $\|E_n\| < 1$	58 % (7)	13 % (1)	20 % (1)	100 % (1)	0 % (0)
Warning sig. $\|E_n\| > 1$	17 % (2)	25 % (2)	20 % (1)	0 % (0)	20 % (1)
Action sig. $\|E_n\| > 1.5$	25 % (3)	63 % (5)	60 % (3)	0 % (0)	80 % (4)

Table 14. Overview of the laboratory performances regarding the relative deviation and E_n numbers sorted according to counting methods of ^{90}Sr. The number in parentheses indicates number of laboratories.

	Gas flow proportional counter (21)	LSC (8)	Plastic scintillation counting (4)	Cherenkov counting (3)	Geiger Müller counting (2)
Within ± 20 %	24 % (5)	25 % (2)	75 % (3)	0 % (0)	100 % (2)
<20 %, 30 %>	43 % (9)	25 % (2)	25 % (1)	0 % (0)	0 % (0)
Outside ± 30 %	33 % (7)	50 % (4)	0 % (0)	100 % (3)	0 % (0)
Compatible $\|E_n\| < 1$	24 % (5)	25 % (2)	75 % (3)	33 % (1)	100 % (2)
Warning sig. $\|E_n\| > 1$	33 % (7)	0 % (0)	25 % (1)	0 % (0)	0 % (0)
Action sig. $\|E_n\| > 1.5$	43 % (9)	75 % (6)	0 % (0)	67 % (2)	0 % (0)

The results of the comparison for ^{90}Sr are not favourable at all, although all participants, except for lab 61, treated the samples according to their routine measurement procedures. The sources of errors must be identified and corrected in these laboratories.

^{212}Pb, ^{212}Bi, ^{214}Pb and ^{214}Bi

No reference values for ^{212}Pb, ^{212}Bi, ^{214}Pb and ^{214}Bi were established. However, for the evaluation of the reported results we used the reference values of their mother radionuclides ^{232}Th and ^{226}Ra as indicative values. No E_n number evaluation and PomPlots were used since there were no reliable uncertainties for the indicative values. Therefore, only relative deviation charts are presented for the evaluation of laboratories' performance (Fig. 60-63).

Success in the determination of activity concentrations of shortlived progenies of radon (^{214}Pb, ^{214}Bi) and thoron (^{212}Pb, ^{212}Bi) varied from 81 % for ^{212}Pb to 61 % for ^{214}Pb. In the case of ^{212}Bi and ^{214}Bi, 67 % and 71 %, respectively, of the reported values deviated less than 20 % from the indicative values. All four radionuclides are beta-decaying and emit several gamma lines. All participants used gamma-ray spectrometry and most of them treated the samples according to their routine procedures.

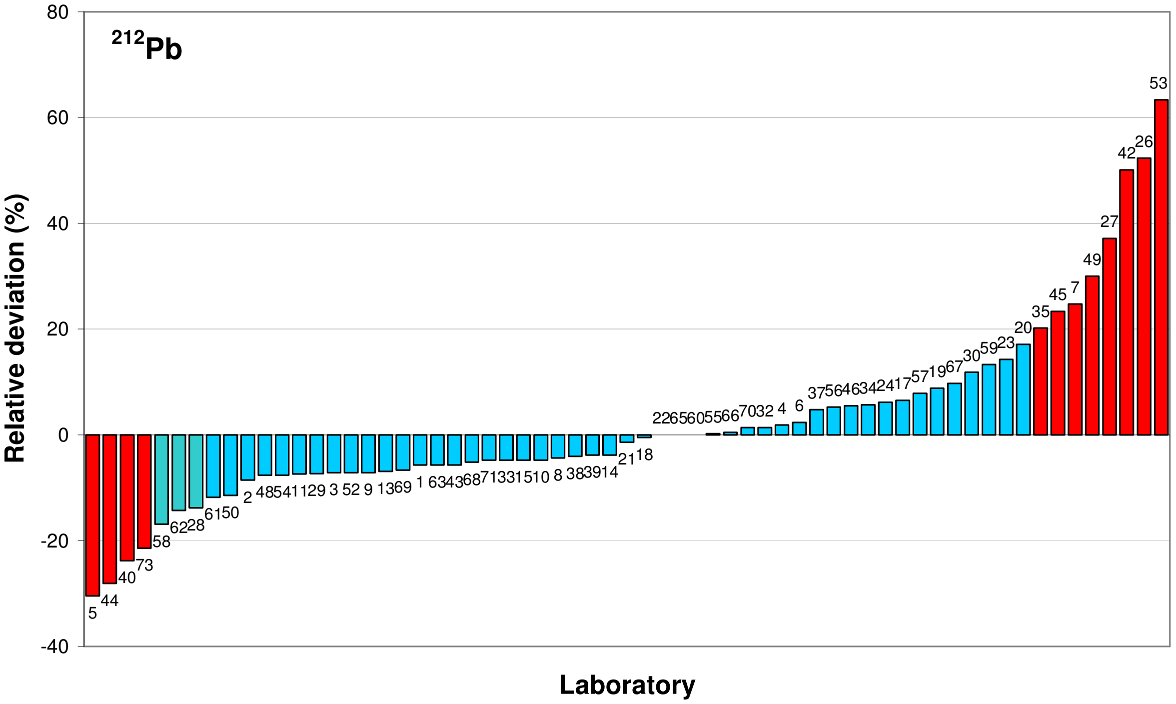

Fig. 60. Deviation chart of the participants' results of ^{212}Pb plotted in ascending order. Blue colour indicates results within the range ± 20 % from the reference value of ^{232}Th and red indicates results outside this range. Numbers indicate the laboratory code.

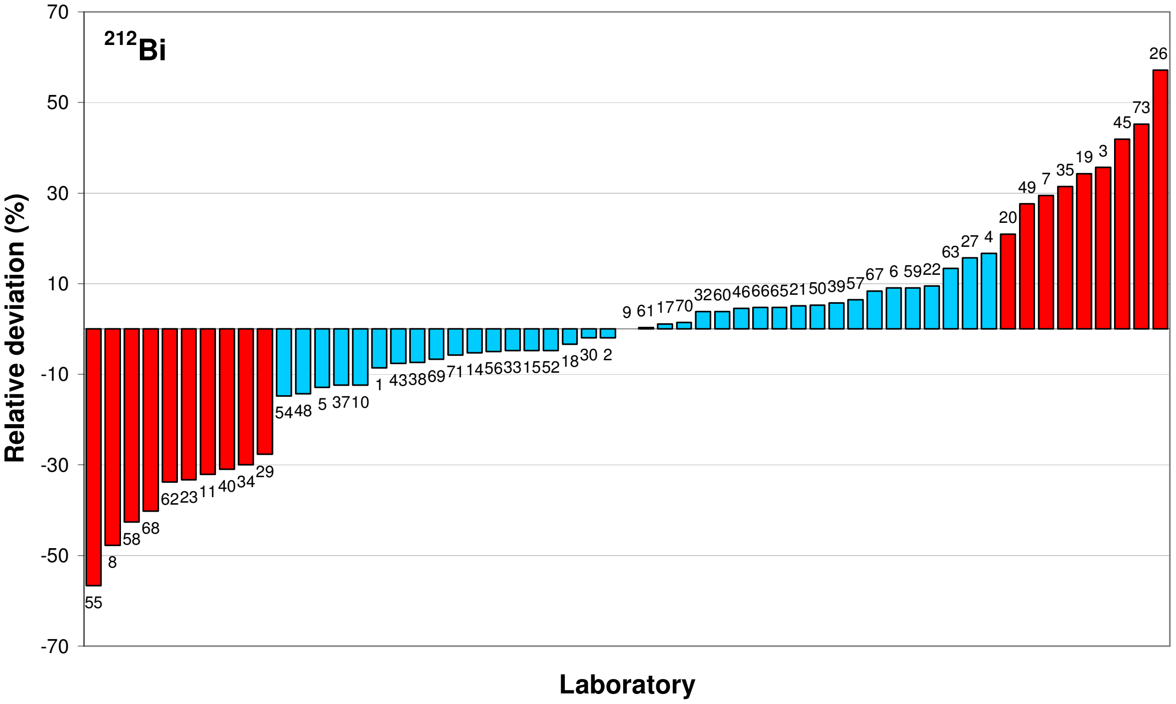

Fig. 61. Deviation chart of the participants' results of ^{212}Bi plotted in ascending order. Blue colour indicates results within the range ± 20 % from the reference value of ^{232}Th and red indicates results outside this range. Numbers indicate the laboratory code.

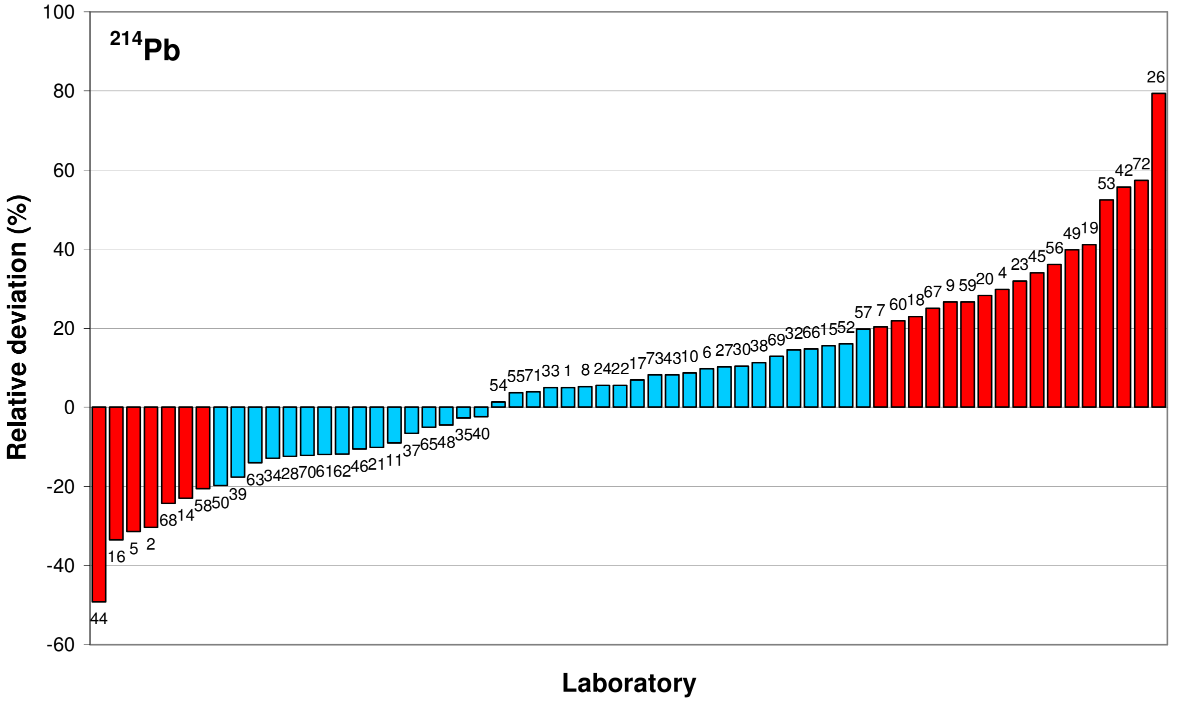

Fig. 62. Deviation chart of the participants' results of ^{214}Pb plotted in ascending order. Blue colour indicates results within the range ± 20 % from the reference value of ^{226}Ra and red indicates results outside this range. Numbers indicate the laboratory code.

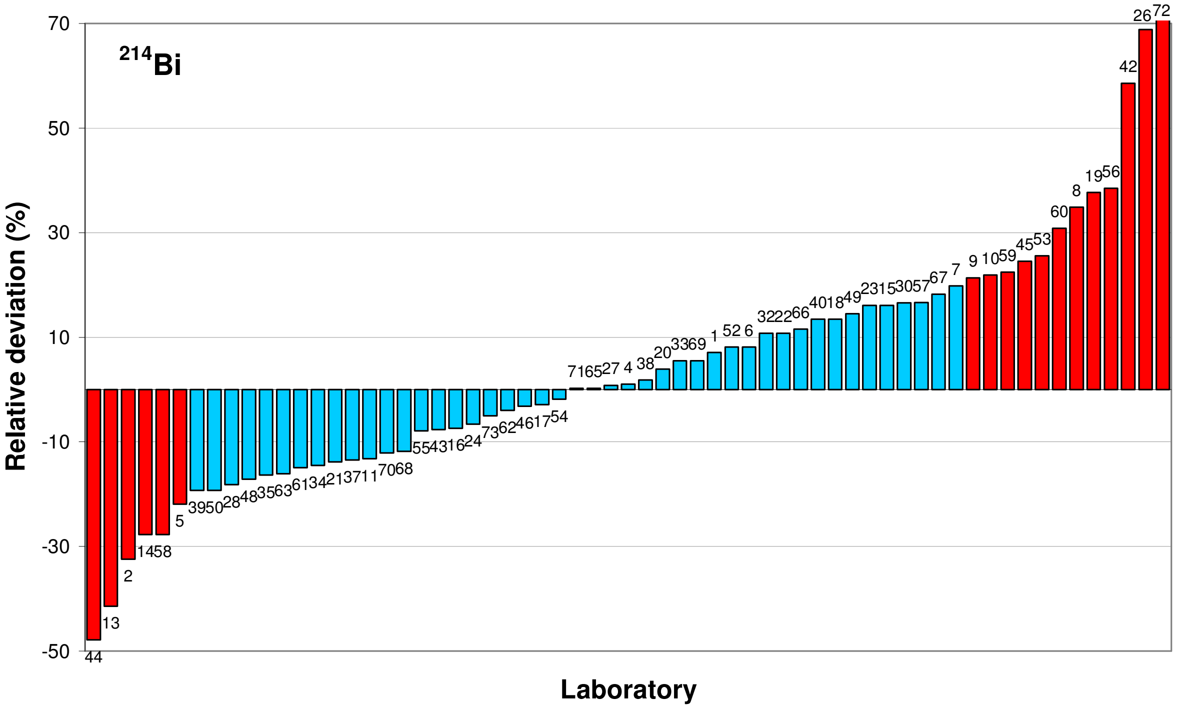

Fig. 63. Deviation chart of the participants' results of ^{214}Bi plotted in ascending order. Blue colour indicates results within the range ± 20 % from the reference value of ^{226}Ra and red indicates results outside this range. Numbers indicate the laboratory code.

^{226}Ra

The results for ^{226}Ra are far from satisfactory. Only 60 % of the participants determined the concentrations within ± 20 % from the reference value and 68 % of the results are compatible in terms of the E_n numbers (Fig. 64-67). These results are even worse than those obtained in the ILC on mineral water, where 70 % (Water-1) and 61 % (Water-2) of the results scattered around the reference value within ± 20 % (Wätjen et al., 2010). In the water comparison, the reference values were even two to three orders of magnitude smaller. Main difference lies in the fact that with the current soil ILC most participants (43 labs or 86 %) applied gamma-ray spectrometry (only 15 % in the water ILC) and only four laboratories used alpha-particle spectrometry (Fig. 66). In most of the participating laboratories, except for labs 45 and 18 (for gamma-ray spectrometry), routine procedures were applied.

The most consistent results (all four) are derived from alpha-particle spectrometry (Table 15, Fig. 66). In the case of gamma-ray spectrometry, participants tended to report results higher than the reference value. The single laboratory (4), which applied gas flow proportional counting, reported a value lower by 34 % than the reference value and the E_n number triggered an action signal. In the PomPlot (Fig. 67), the results of gas flow proportional counting and Lucas cell are found outside of the top of the pyramid indicating incorrect values.

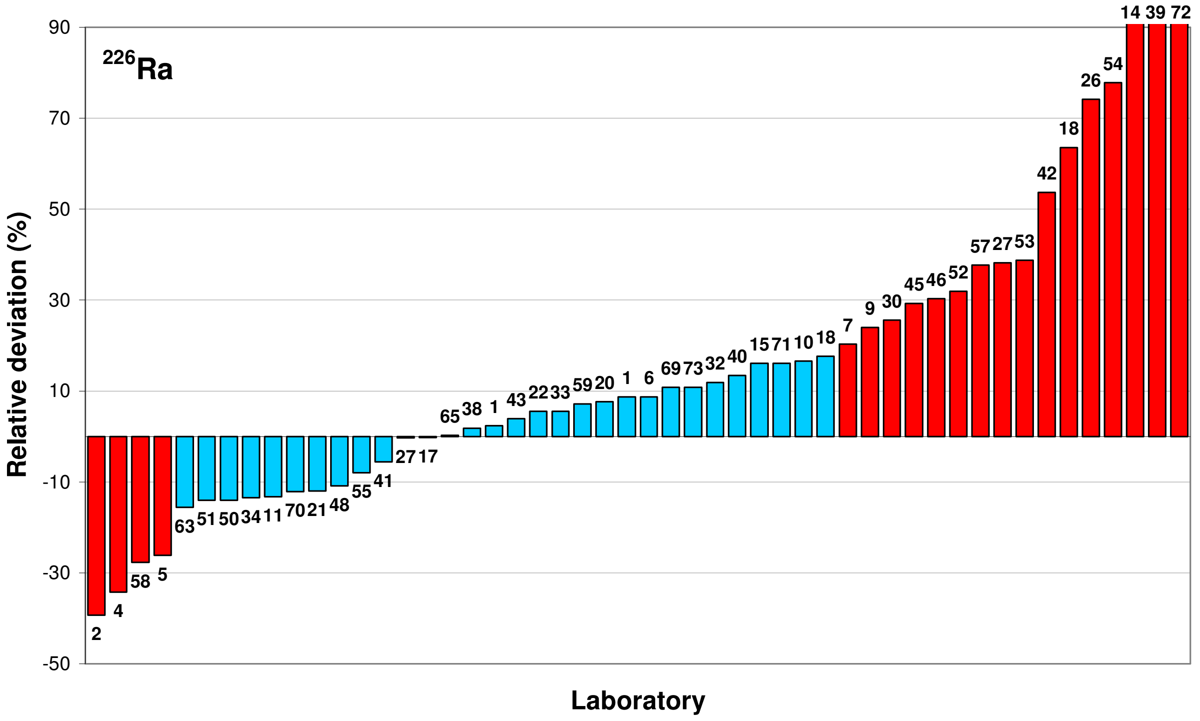

Fig. 64. Deviation chart of the participants' results of ^{226}Ra plotted in ascending order. Blue colour indicates results within the range ± 20 % from the reference value and red indicates results outside this range. Numbers indicate the laboratory code.

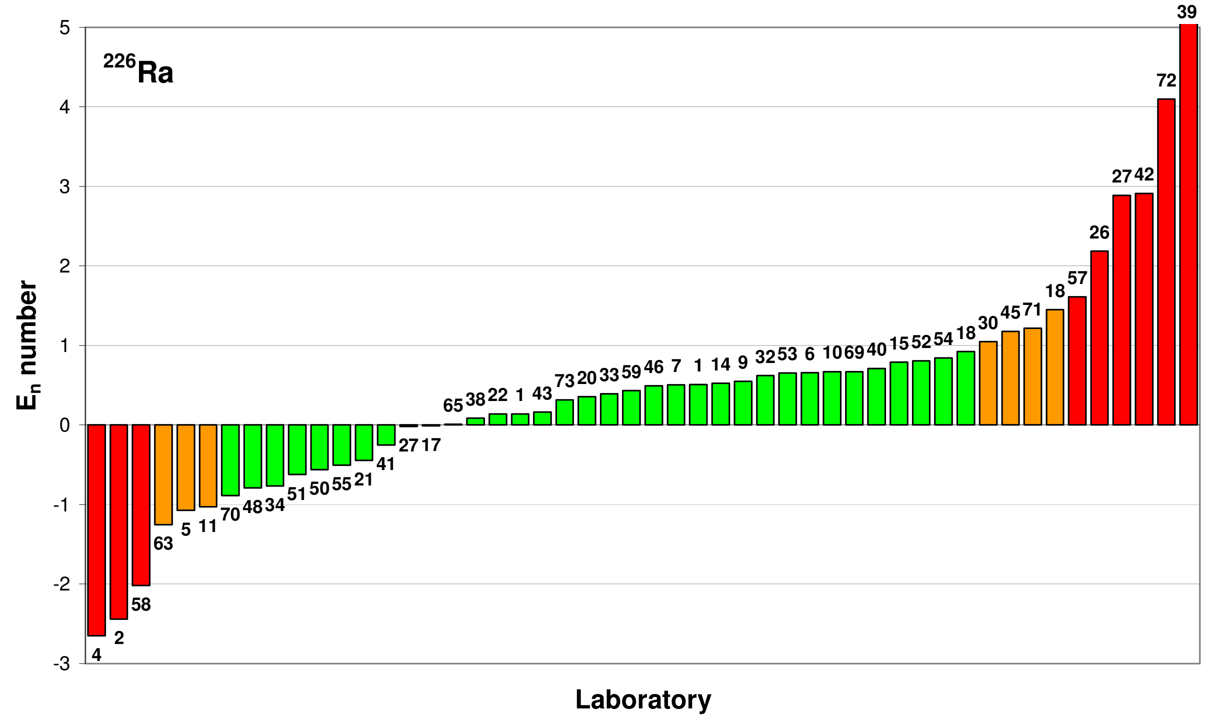

Fig. 65. E_n numbers' chart of ^{226}Ra activity concentrations plotted in ascending order. Green colour indicates compatible results, orange indicates warning signal and red indicates action signal. Numbers indicate the laboratory code.

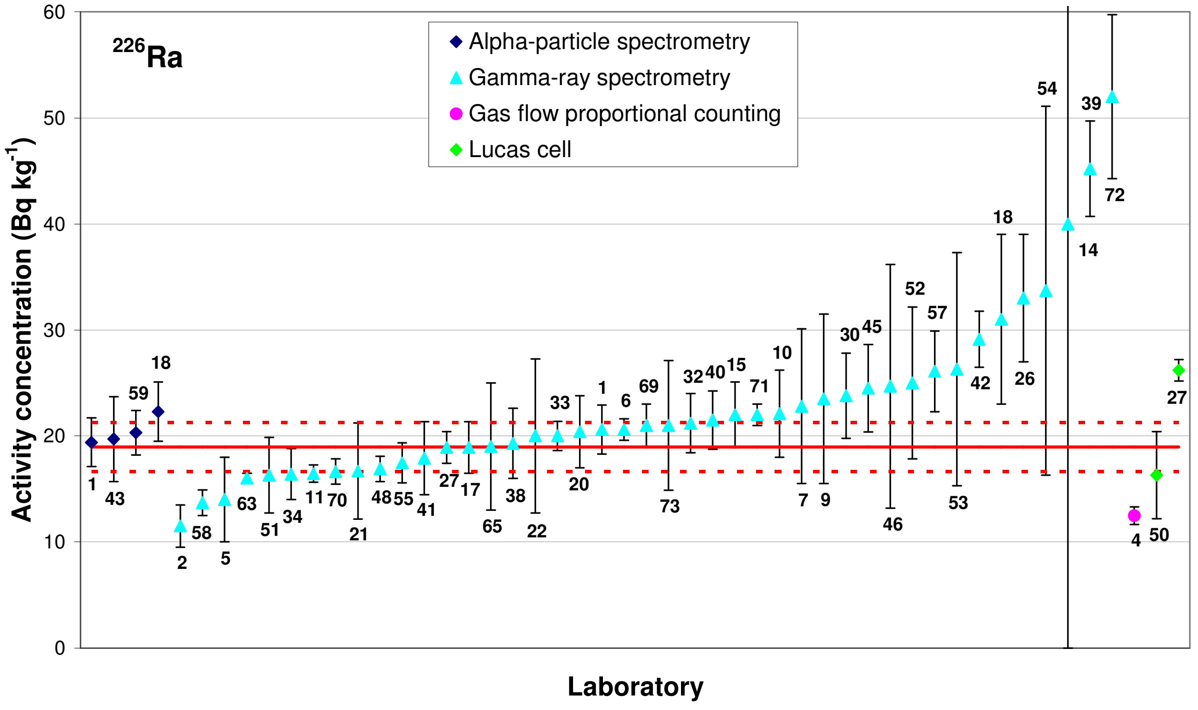

Fig. 66. Laboratory results A_{lab} of ^{226}Ra activity concentration with expanded uncertainties U_{lab} ($k = 2$) sorted according to counting methods. Red lines represent reference value $A_{ref} \pm U_{ref}$ ($k = 2$). Numbers indicate the laboratory code.

Since the ^{226}Ra peak at 186.1 keV is, in environmental samples, interfered by the gamma emission from ^{235}U (185.7 keV) and these two gamma lines are observed as one doublet peak, superimposed on the backscatter peak of the Compton continuum (Fig. 68), an alternative determination of ^{226}Ra via its short-lived daughter products ^{214}Pb and ^{214}Bi is often chosen. This approach, however, offers several pitfalls as well. Not only must the sample container be airtight in order to prevent escape of ^{222}Rn gas, also any air space in the sample container must be avoided to prevent ^{222}Rn and its daughter products from accumulating in that space and, hence, for geometry reasons, being measured with a different efficiency than that of the soil sample (Carconi et al., 2012).

Table 15. Overview of the laboratory performances regarding the relative deviation and E_n numbers sorted according to measurement method of ^{226}Ra. The number in parentheses indicates number of laboratories.

	Alpha-particle spectrometry (4)	Gamma-ray spectrometry (43)	Gas flow proportional counting (1)	Lucas cell (2)
Within ± 20 %	100 % (4)	58 % (25)	0 % (0)	50 % (1)
Outside ± 20 %	0 % (0)	42 % (18)	100 % (1)	50 % (1)
Compatible $\|E_n\| < 1$	100 % (4)	67 % (29)	0 % (0)	50 % (1)
Warning sig. $\|E_n\| > 1$	0 % (0)	16 % (7)	0 % (0)	0 % (0)
Action sig. $\|E_n\| > 1.5$	0 % (0)	16 % (7)	100 % (1)	50 % (1)

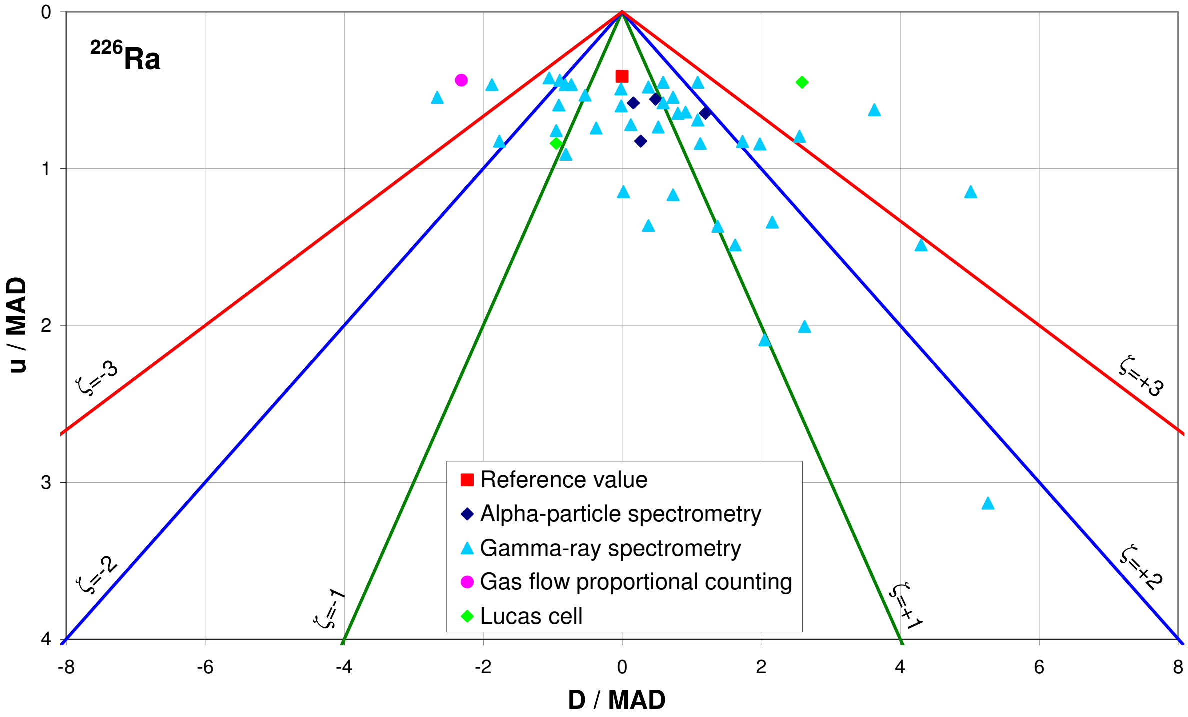

Fig. 67. PomPlot of the ^{226}Ra data sorted according to measurement methods. Red point indicates the reference value. Green, blue and red solid lines indicate ζ-scores = 1, 2 and 3, respectively.

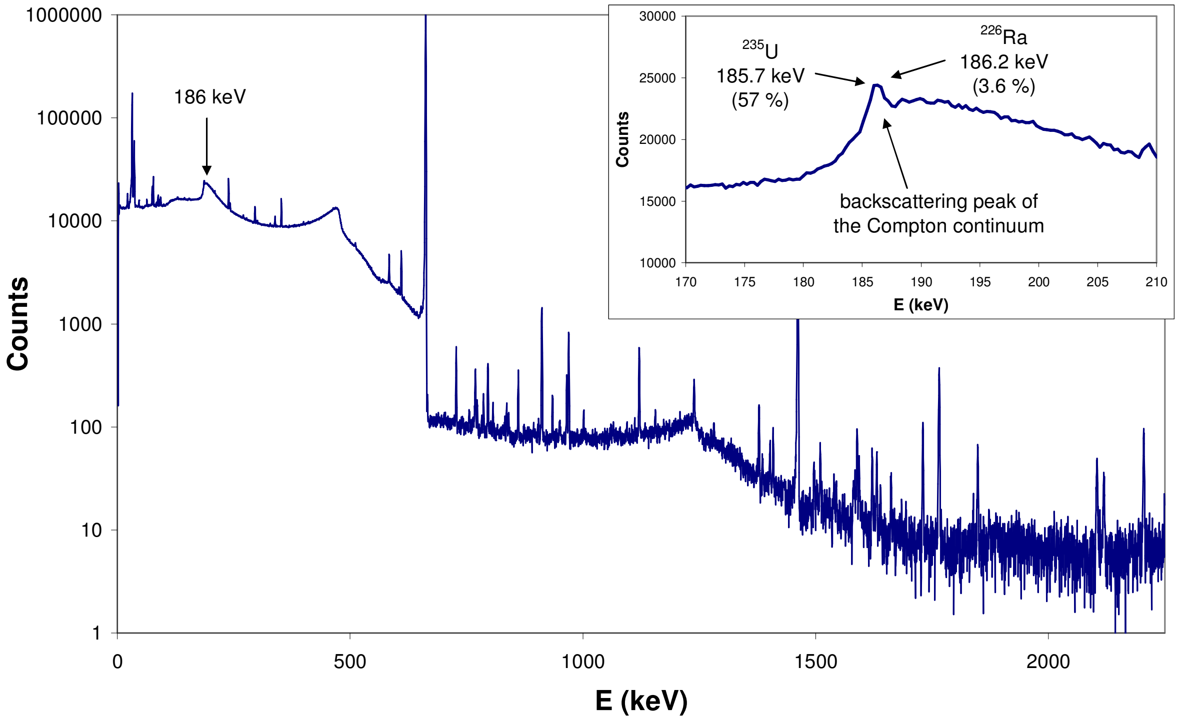

Fig. 68. Gamma-ray spectrum of the soil sample measured with the HPGe detector.

^{230}Th and ^{232}Th

The results of two thorium isotopes are presented in Fig. 69-76. While for ^{232}Th 78 % of the results lie within ± 20 % from the reference value, for ^{230}Th it is only 45 %. This difference may result from the fact that almost half of the participants do not measure activity concentration of ^{230}Th on routine basis. Also, in terms of E_n numbers the performance is much better for ^{232}Th with 80 % compatible values and 16 % triggering action signal than for ^{230}Th with 50 % compatible results and 41 % triggering action signal (Fig. 70 and Fig. 72).

Both isotopes were determined via gamma-ray or alpha-particle spectrometry. As can be seen in Table 16, for ^{230}Th the performance is more or less equal for both measurement methods although only three laboratories used gamma-ray spectrometry (Fig. 73). In the case of ^{232}Th (Fig. 75, Table 16), the results of gamma-ray spectrometry are significantly better in comparison to alpha-particle spectrometry. Comparison of the measurement methods for both thorium isotopes is presented also in the PomPlots (Fig. 74 and Fig. 76).

Table 16. Overview of the laboratory performances regarding the relative deviation and E_n numbers sorted according to measurement method of ^{226}Ra. The number in parentheses indicates number of laboratories.

	^{230}Th		^{232}Th	
	Alpha-particle spectrometry (19)	Gamma-ray spectrometry (3)	Alpha-particle spectrometry (20)	Gamma-ray spectrometry (25)
Within ± 20 %	47 % (9)	33 % (1)	65 % (13)	92 % (23)
Outside ± 20 %	53 % (10)	67 % (2)	35 % (7)	8 % (2)
Compatible $\lvert E_n\rvert < 1$	47 % (9)	67 % (2)	60 % (12)	96 % (24)
Warning sig. $\lvert E_n\rvert > 1$	11 % (2)	0 % (0)	10 % (2)	0 % (0)
Action sig. $\lvert E_n\rvert > 1.5$	42 % (8)	33 % (1)	30 % (6)	4 % (1)

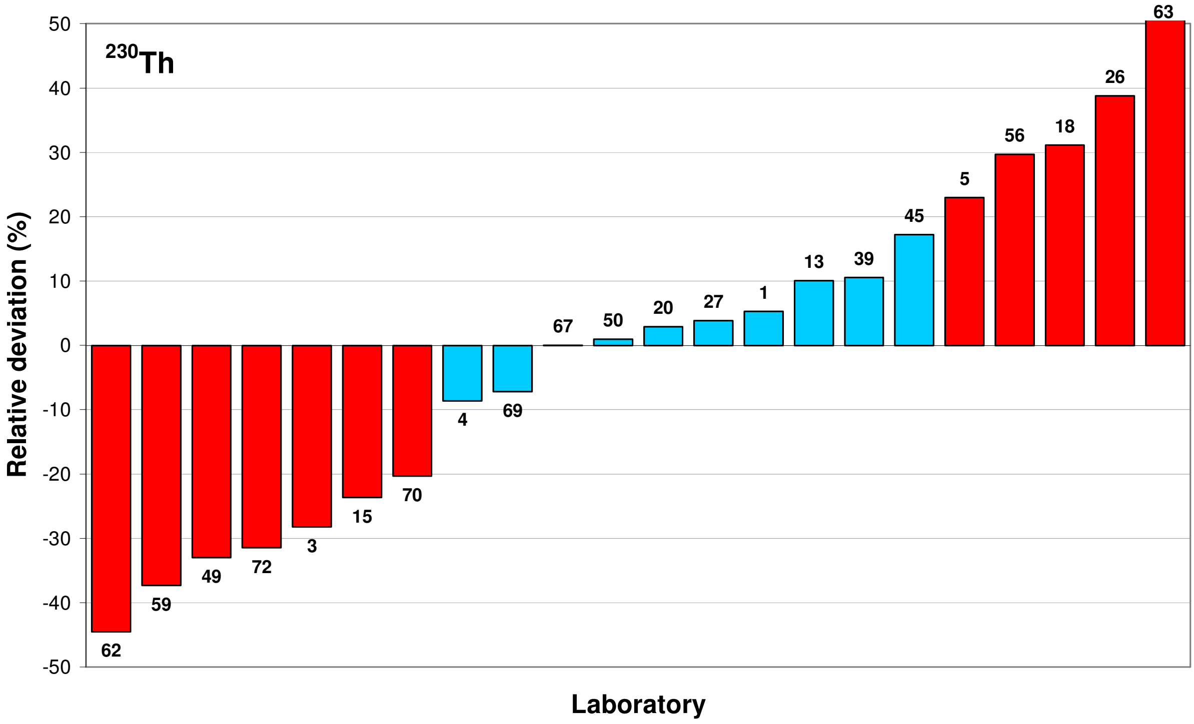

Fig. 69. Deviation chart of the participants' results of ^{230}Th plotted in ascending order. Blue colour indicates results within the range ± 20 % from the reference value and red indicates results outside this range. Numbers indicate the laboratory code.

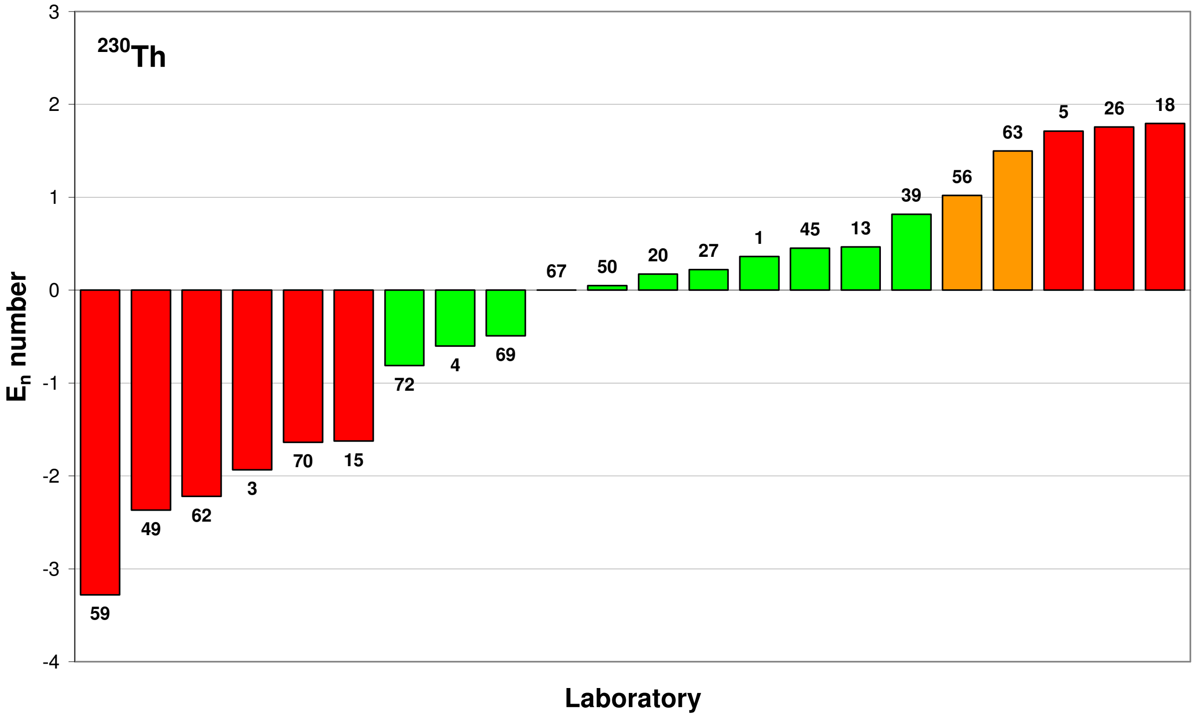

Fig. 70. E_n numbers' chart of ^{230}Th activity concentrations plotted in ascending order. Green colour indicates compatible results, orange indicates warning signal and red indicates action signal. Numbers indicate the laboratory code.

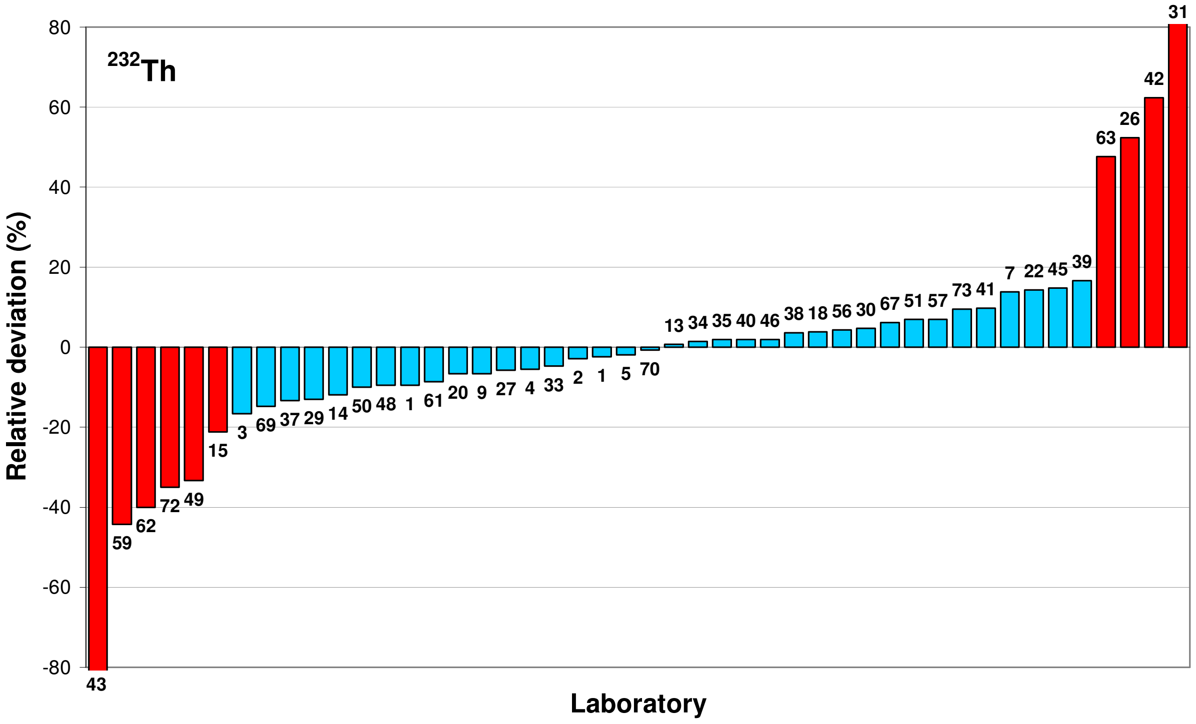

Fig. 71. Deviation chart of the participants' results of ^{232}Th plotted in ascending order. Blue colour indicates results within the range ± 20 % from the reference value and red indicates results outside this range. Numbers indicate the laboratory code.

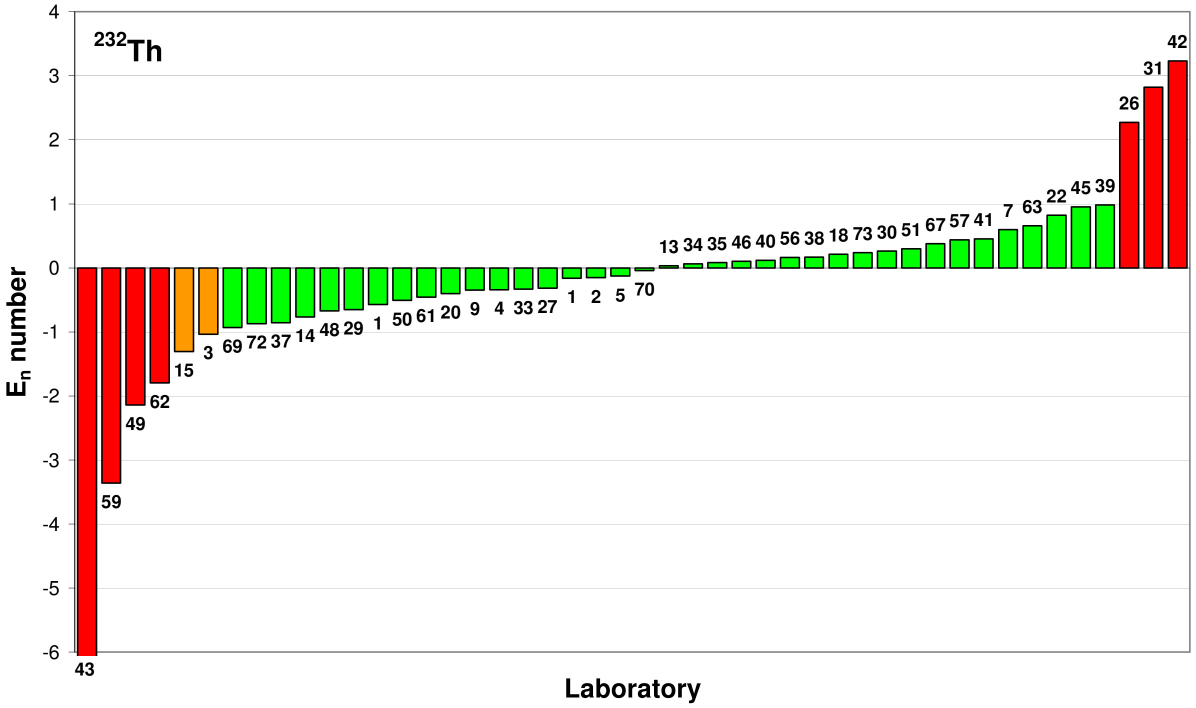

Fig. 72. E_n numbers' chart of ^{232}Th activity concentrations plotted in ascending order. Green colour indicates compatible results, orange indicates warning signal and red indicates action signal. Numbers indicate the laboratory code.

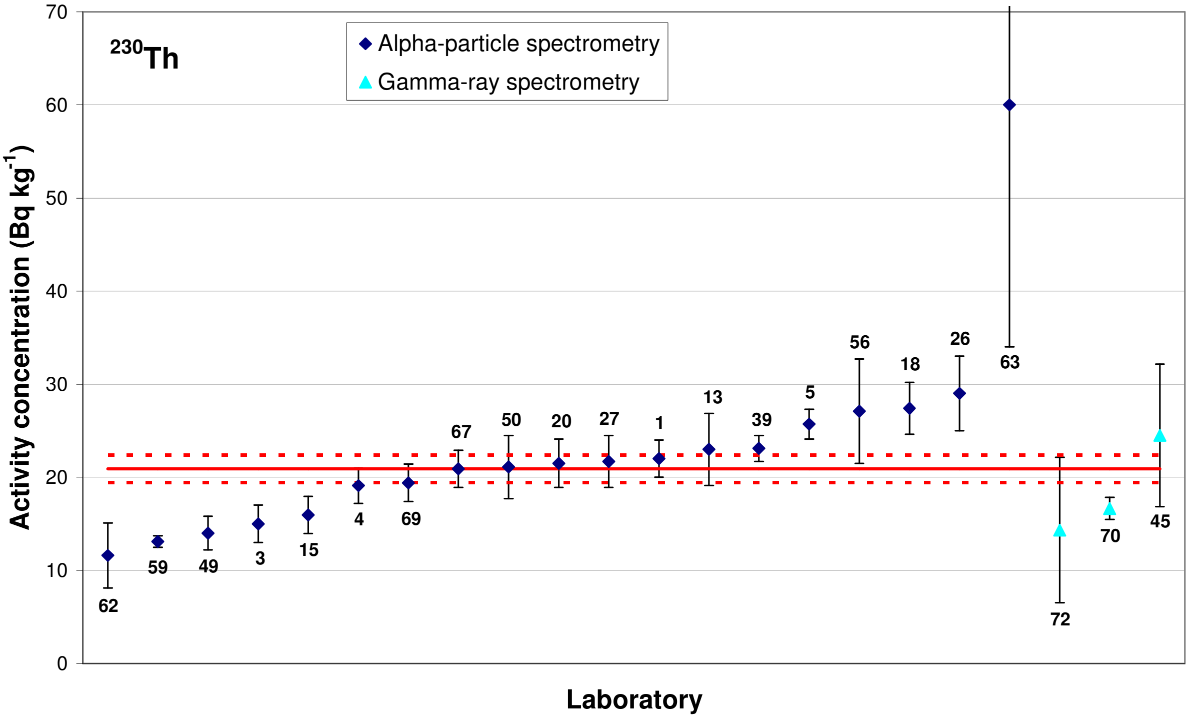

Fig. 73. Laboratory results A_{lab} of ^{230}Th activity concentration with expanded uncertainties U_{lab} ($k = 2$) sorted according to counting methods. Red lines represent reference value $A_{ref} \pm U_{ref}$ ($k = 2$). Numbers indicate the laboratory code.

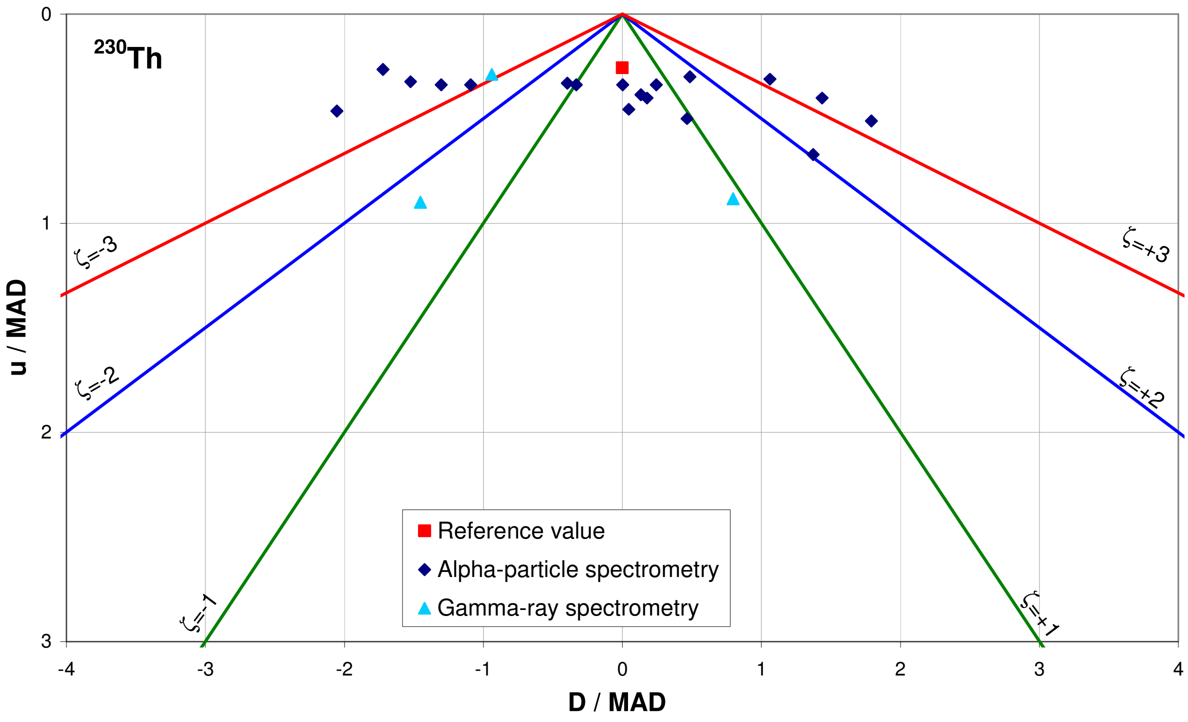

Fig. 74. PomPlot of the ^{230}Th data sorted according to measurement methods. Red point indicates the reference value. Green, blue and red solid lines indicate ζ-scores = 1, 2 and 3, respectively.

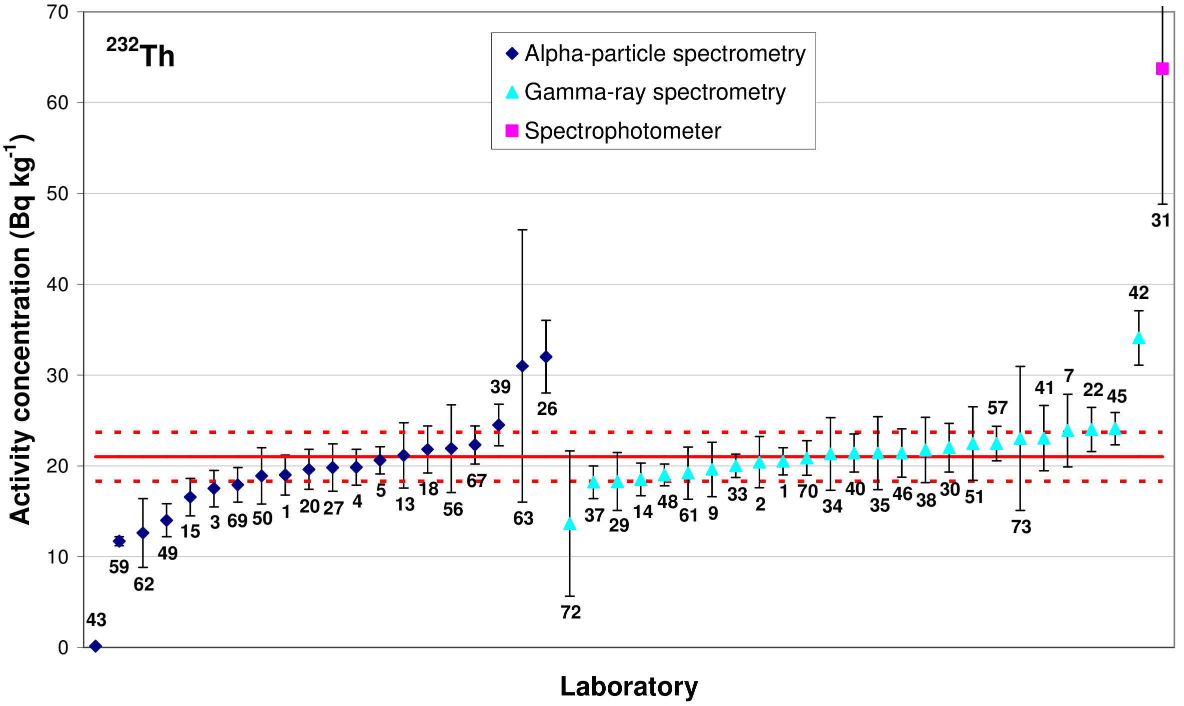

Fig. 75. Laboratory results A_{lab} of ^{232}Th activity concentration with expanded uncertainties U_{lab} ($k = 2$) sorted according to counting methods. Red lines represent reference value $A_{ref} \pm U_{ref}$ ($k = 2$). Numbers indicate the laboratory code.

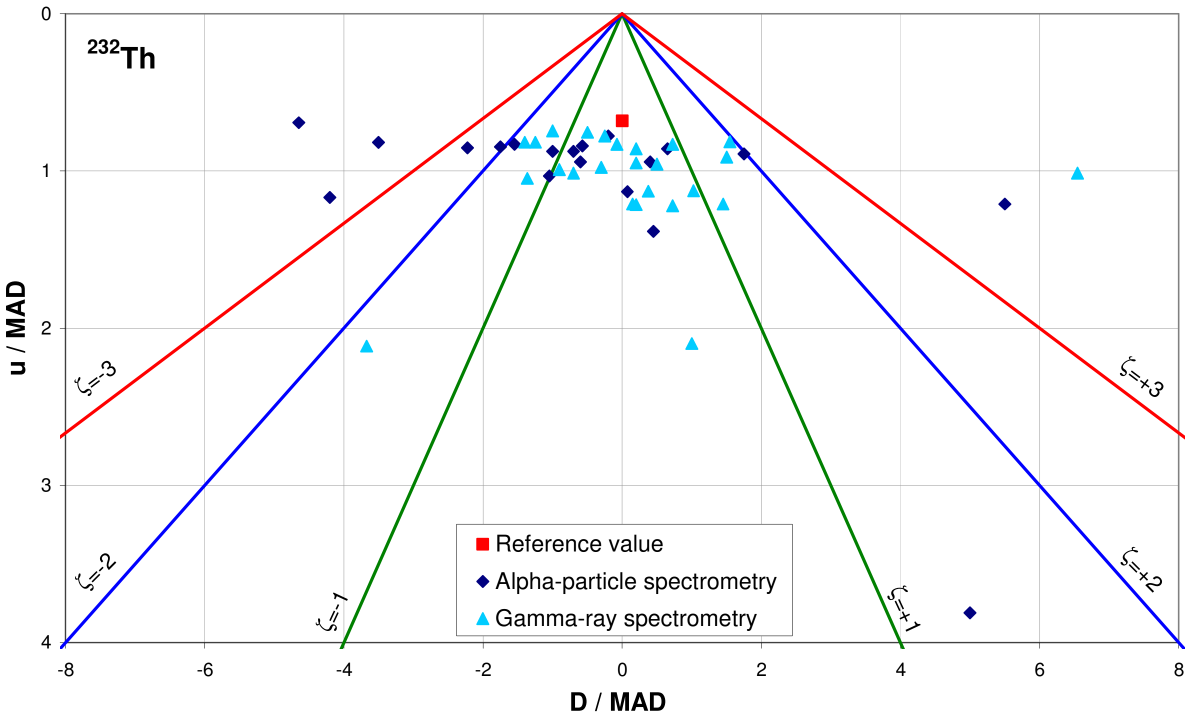

Fig. 76. PomPlot of the ^{232}Th data sorted according to measurement methods. Red point indicates the reference value. Green, blue and red solid lines indicate ζ-scores = 1, 2 and 3, respectively.

^{234}U, ^{235}U and ^{238}U

A mixed performance is observed for the uranium isotopes. Most of the participants treated the samples according to the routine procedures established in their laboratories, except for laboratories 3, 18 (routine procedure for alpha-particle spectrometry but not for gamma-ray spectrometry), 45, 61, and 71.

The worst performance is found for ^{235}U, with 74 % of the values deviating more than 20 % from the reference value (Fig. 77) and only 42 % E_n compatible (Fig. 78). The results are strongly method dependent as can be seen in Fig. 79 and Table 17. Since the equivalent argumentation given above for ^{226}Ra holds also for ^{235}U (interference with ^{226}Ra and superposition on backscatter peak), it is not surprising that the worst performance is observed for gamma-ray spectrometry where a large number of results are far too high. For instance, 11 laboratories (65 %) submitted results more than twice the reference value, and only one submitted a result within 20 %. Obviously, gamma-ray spectrometry is not the optimal method for ^{226}Ra and ^{235}U, although, some laboratories were successful with good determinations, especially for ^{226}Ra.

Results for ^{235}U obtained with alpha-particle spectrometry are comparably better but still not good. Only 7 labs of 19 give results within ± 20 % from the reference value and one result is off by more than a factor of two. Interesting to note, 58 % of alpha-spectrometric results are accompanied by a realistic uncertainty estimate, and, therefore, are compatible with E_n numbers. The best results are those determined by ICP-MS (labs 52, 67) which can be seen in Fig. 79 and 80 where these two values almost overlap with the reference value.

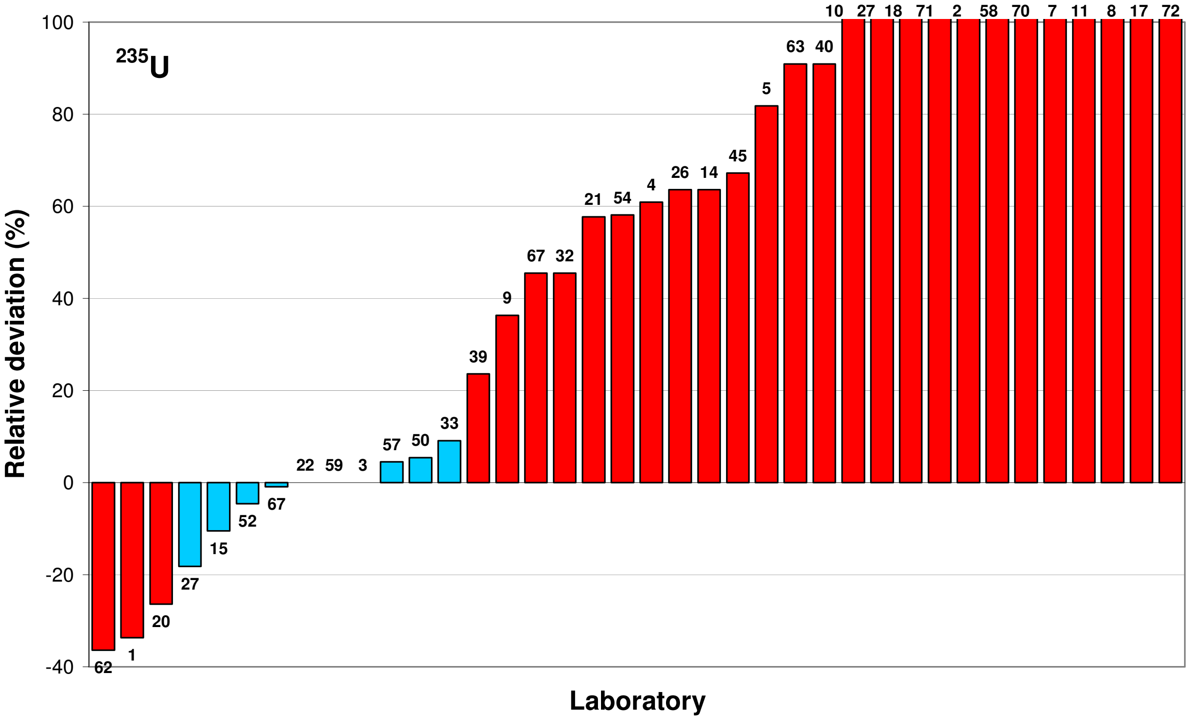

Fig. 77. Deviation chart of the participants' results of ^{235}U plotted in ascending order. Blue colour indicates results within the range ± 20 % from the reference value and red indicates results outside this range. Numbers indicate the laboratory code.

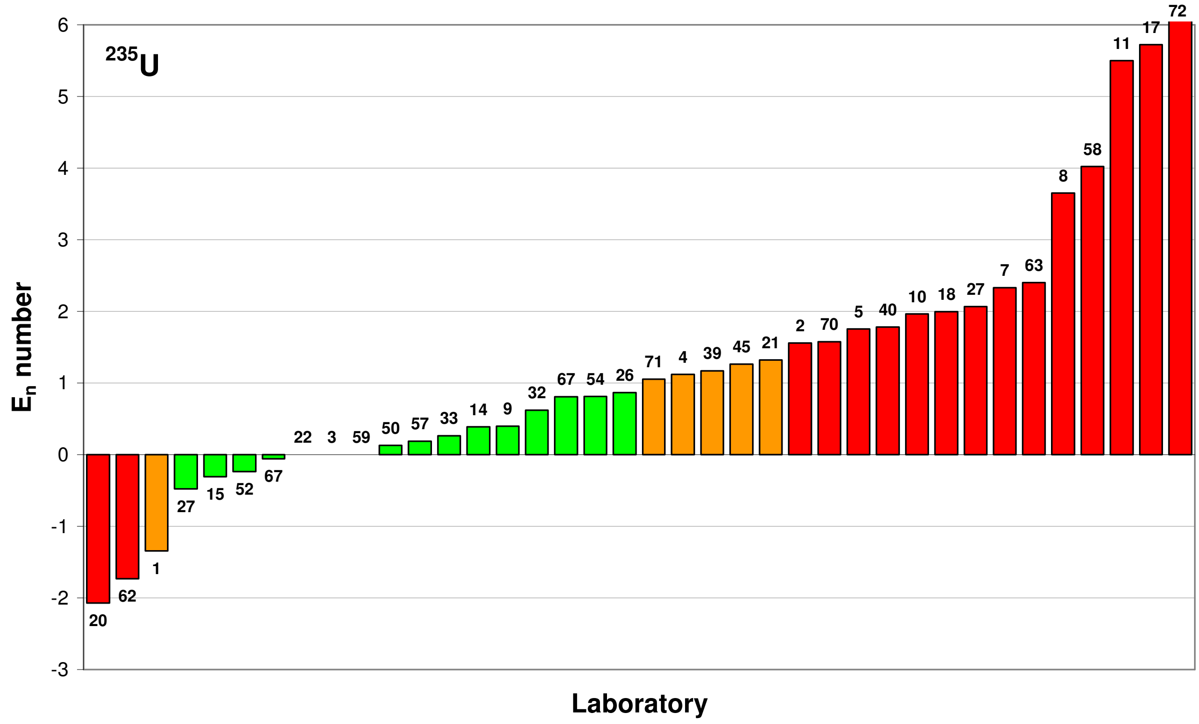

Fig. 78. E_n numbers' chart of ^{235}U activity concentrations plotted in ascending order. Green colour indicates compatible results, orange indicates warning signal and red indicates action signal. Numbers indicate the laboratory code.

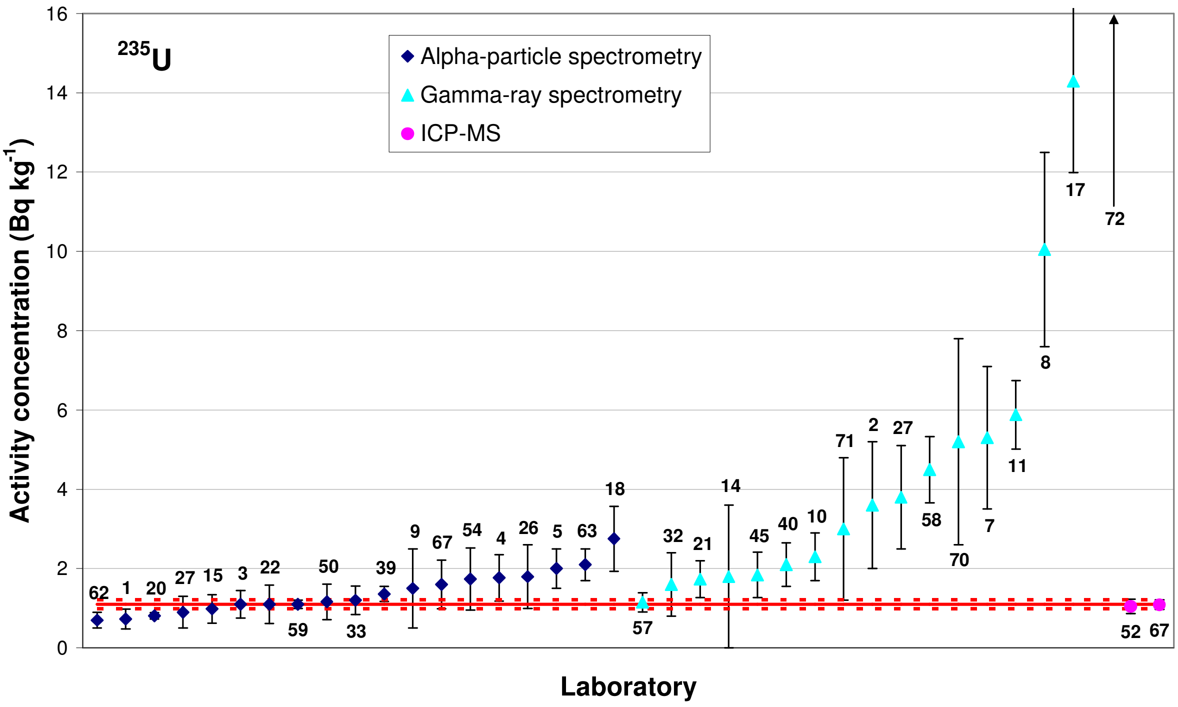

Fig. 79. Laboratory results A_{lab} of ^{235}U activity concentration with expanded uncertainties U_{lab} ($k = 2$) sorted according to counting methods. Red lines represent reference value $A_{ref} \pm U_{ref}$ ($k = 2$). Numbers indicate the laboratory code.

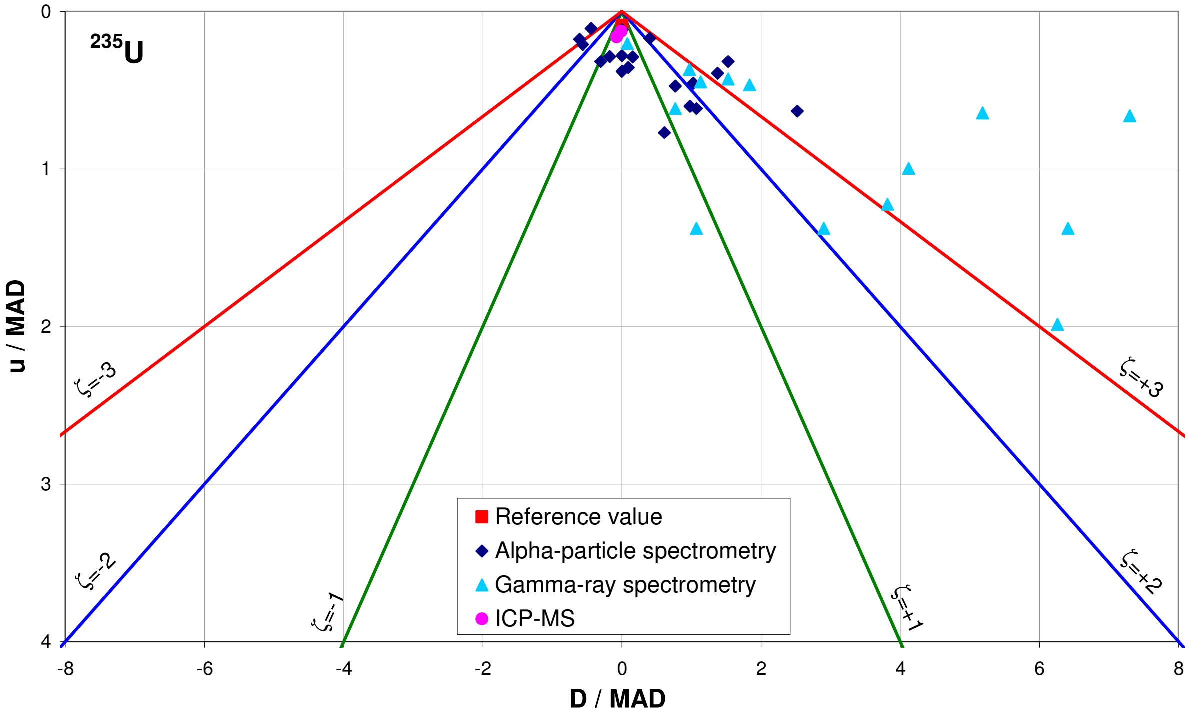

Fig. 80. PomPlot of the ^{235}U data sorted according to measurement method. Red point indicates the reference value. Green, blue and red solid lines indicate ζ-scores = 1, 2 and 3, respectively.

Table 17. Overview of the laboratory performances regarding the relative deviation and E_n numbers sorted according to measurement methods of ^{235}U. The number in parentheses indicates number of laboratories.

	Alpha-particle spectrometry (19)	Gamma-ray spectrometry (17)	ICP-MS (2)
Within ± 20 %	37 % (7)	6 % (1)	100 % (2)
Outside ± 20 %	63 % (12)	94 % (16)	0 % (0)
Compatible $\|E_n\| < 1$	58 % (11)	18 % (3)	100 % (2)
Warning sig. $\|E_n\| > 1$	16 % (3)	18 % (3)	0 % (0)
Action sig. $\|E_n\| > 1.5$	26 % (5)	65 % (11)	0 % (0)

Better, but still not satisfactory, performance is observed for the other two uranium isotopes. For ^{234}U, 68 % of the results, mostly obtained with alpha-particle spectrometry, are within ± 20 % from the reference value and 57 % are compatible in terms of E_n numbers (Fig. 81, Fig. 82 and Table 18). The ICP-MS method was used in two laboratories (52, 67) with very good results. Laboratory 45 calculated the ^{234}U activity via gamma spectrometric measurement of ^{238}U decay products assuming radioactive equilibrium (Fig. 83). Although this result deviates more than 20% from the reference value, due to its high uncertainty it is E_n compatible. Four laboratories (1, 5, 15, 59) underestimated their uncertainties and therefore their results are E_n incompatible. On the other hand laboratories 54 and 61 reported larger uncertainties and hence their results are E_n compatible or triggering warning signal though the reported values deviate more than 30 % from the reference value.

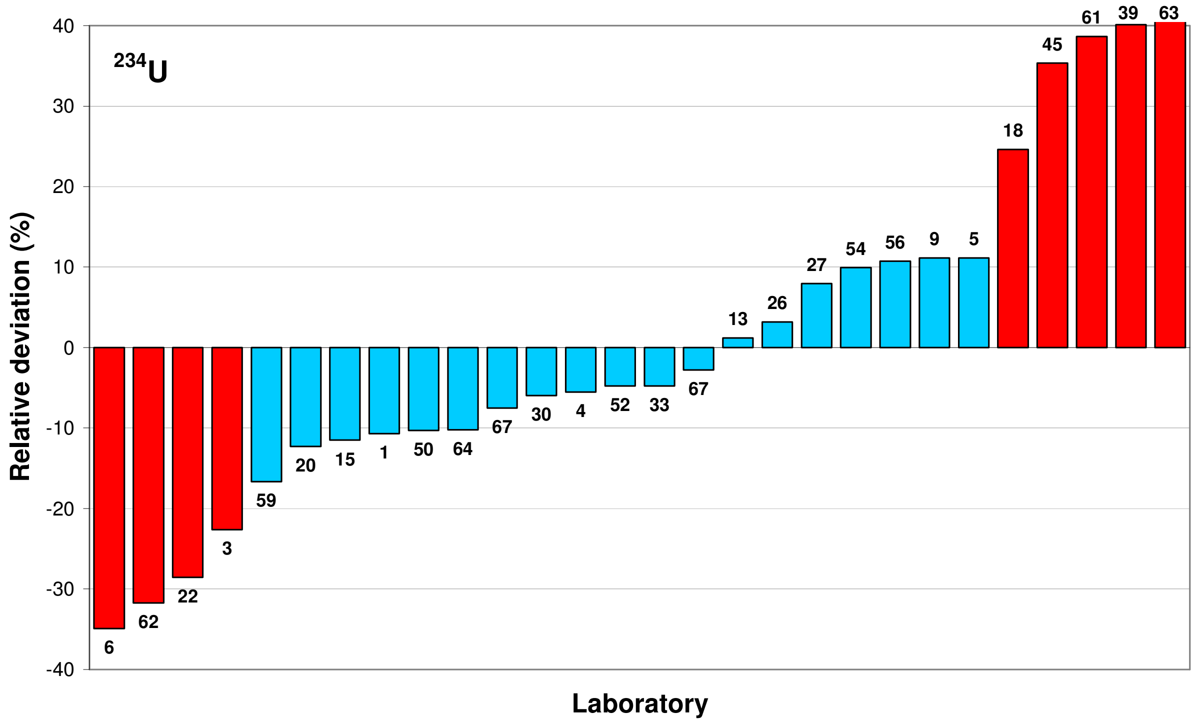

Fig. 81. Deviation chart of the participants' results of ^{234}U plotted in ascending order. Blue colour indicates results within the range ± 20 % from the reference value and red indicates results outside this range. Numbers indicate the laboratory code.

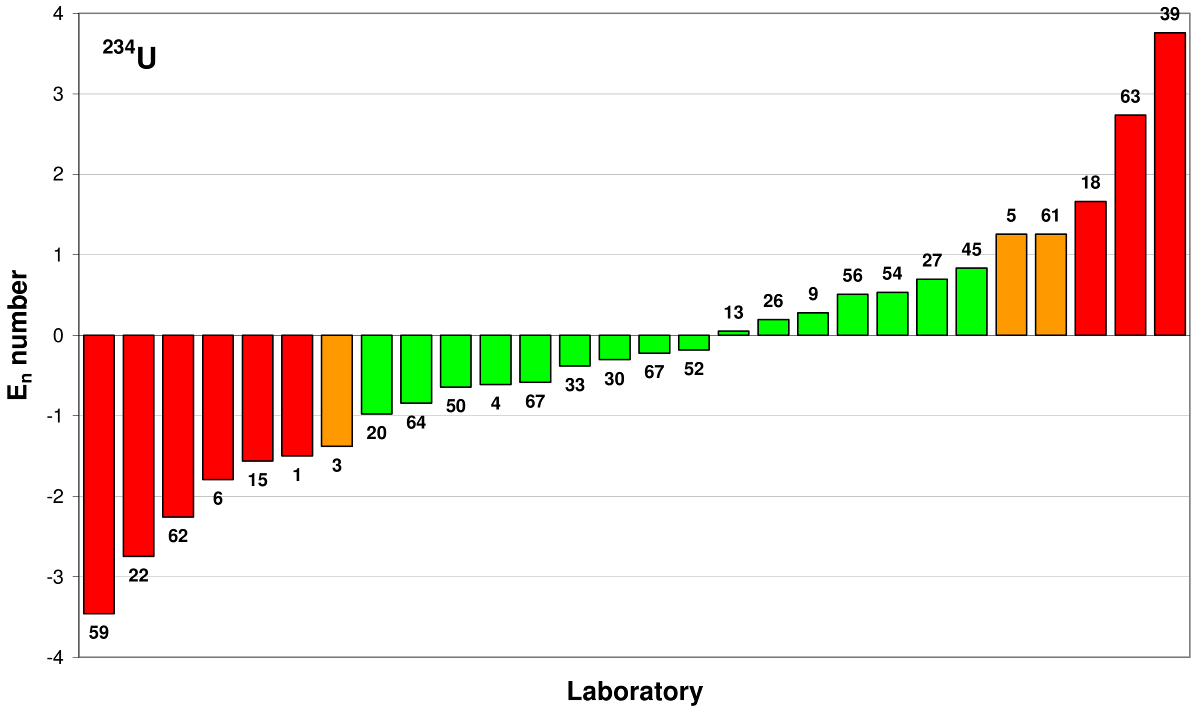

Fig. 82. E_n numbers' chart of ^{234}U activity concentrations plotted in ascending order. Green colour indicates compatible results, orange indicates warning signal and red indicates action signal. Numbers indicate the laboratory code.

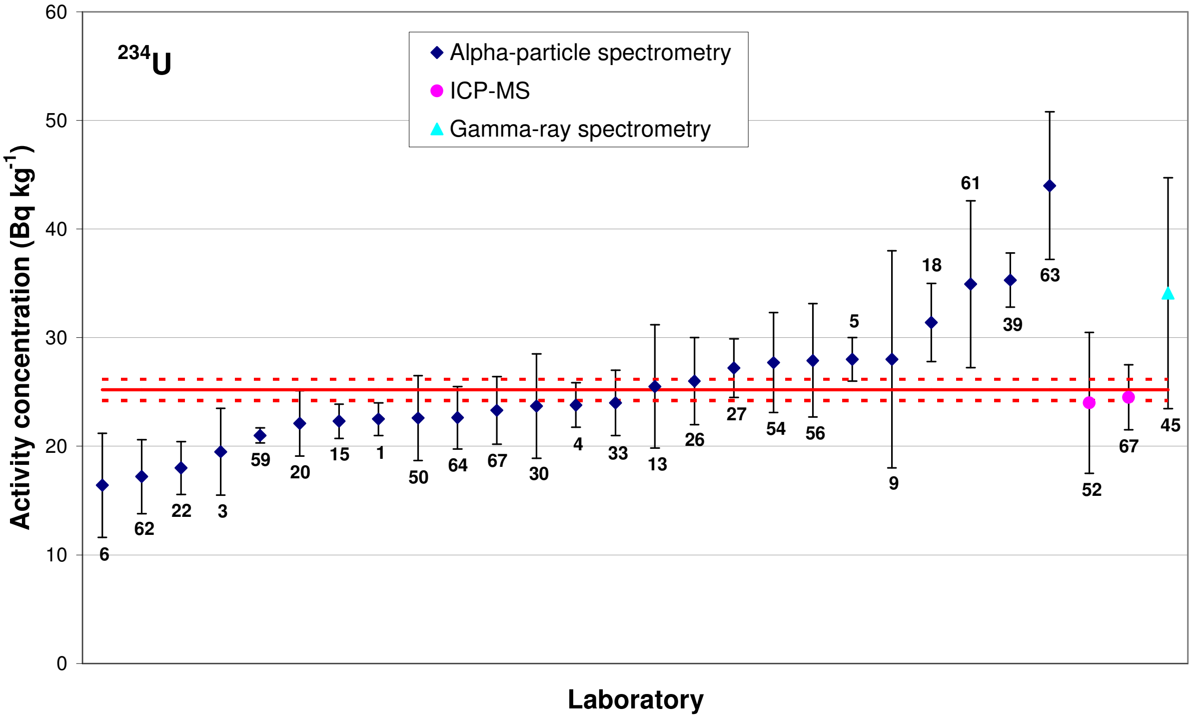

Fig. 83. Laboratory results A_{lab} of ^{234}U activity concentration with expanded uncertainties U_{lab} ($k = 2$) sorted according to counting methods. Red lines represent reference value $A_{ref} \pm U_{ref}$ ($k = 2$). Numbers indicate the laboratory code.

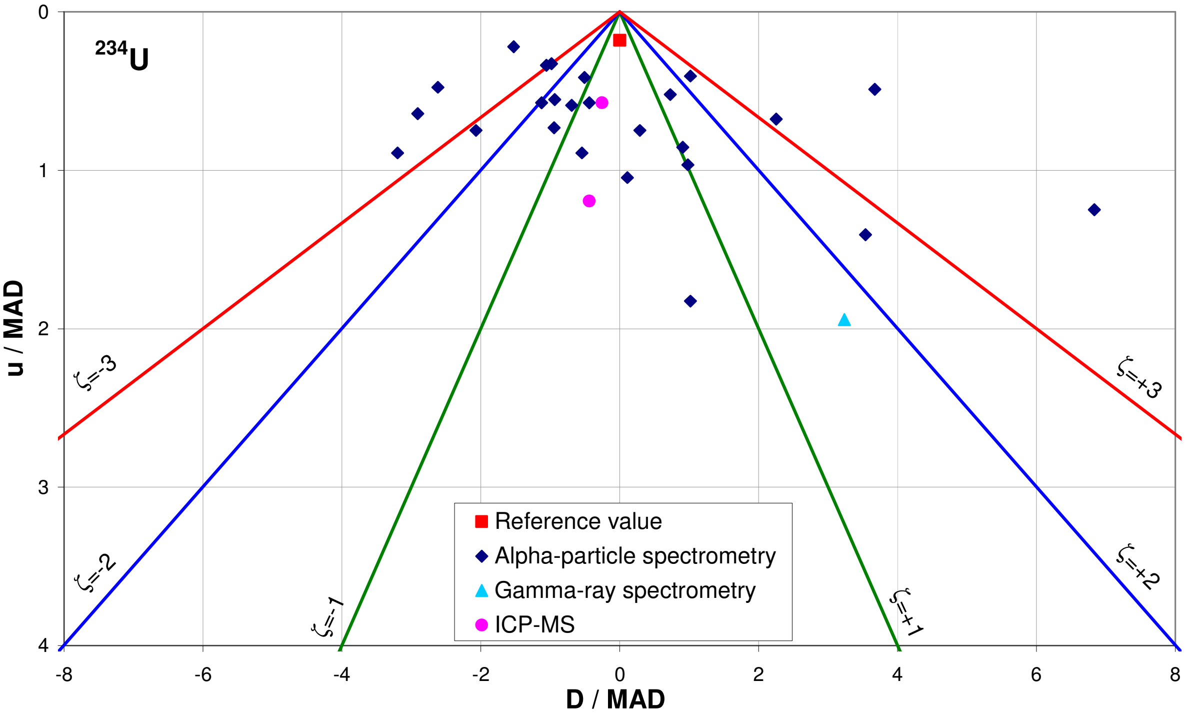

Fig. 84. PomPlot of the ^{234}U data sorted according to measurement method. Red point indicates the reference value. Green, blue and red solid lines indicate ζ-scores = 1, 2 and 3, respectively.

Table 18. Overview of the laboratory performances regarding the relative deviation and E_n numbers sorted according to measurement methods of ^{234}U. The number in parentheses indicates number of laboratories.

	Alpha-particle spectrometry (25)	ICP-MS (2)	Gamma-ray spectrometry (1)
Within ± 20 %	68 % (17)	100 % (2)	0 % (0)
Outside ± 20 %	32 % (8)	0 % (0)	100 % (1)
Compatible $\lvert E_n \rvert < 1$	52 % (13)	100 % (2)	100 % (1)
Warning sig. $\lvert E_n \rvert > 1$	12 % (3)	0 % (0)	0 % (0)
Action sig. $\lvert E_n \rvert > 1.5$	36 % (9)	0 % (0)	0 % (0)

In the case of ^{238}U, 58 % of the results lie within ± 20 % and 56 % are compatible with the E_n criterion as presented in Fig. 85-88. About half of the participating laboratories used gamma-ray spectrometry in order to determine the activity concentration of ^{238}U (Fig. 87). Although the results are comparable (Table 19), the distribution of the results of gamma-ray spectrometry is not as uniform as for alpha-particle spectrometry (Fig. 87 and 88). For the determination of ^{238}U, gamma-ray emissions following the decay of the first daughter ^{234}Th are often used. However, the low-energy gamma lines at 63 keV and 92.5 keV (both doublets) are very difficult to measure due to high attenuation and interferences with X-rays and scattered gamma-rays. Of course, the secular equilibrium between mother and daughter radionuclides must be secured. The gamma-lines, emitted by the second daughter radionuclide ^{234m}Pa, at 767 keV and 1001 keV can not be used due to their very low emission probabilities (0.317 % and 0.842 %, respectively). Detailed information on necessary precautions and measurement conditions are presented in the paper of Hult et al. (2012). It addresses the use of correct decay data, suitable detectors, optimised sample size, enhanced spectral amplification, correction for peak interferences and control of background. Again, the best results are produced by ICP-MS. Nevertheless, since only two participants (52, 67) used this technique, no general conclusion can be made.

Table 19. Overview of the laboratory performances regarding the relative deviation and E_n numbers sorted according to measurement methods of ^{238}U. The number in parentheses indicates number of laboratories.

	Alpha-particle spectrometry (25)	Gamma-ray spectrometry (21)	ICP-MS (2)
Within ± 20 %	56 % (14)	62 % (13)	100 % (2)
Outside ± 20 %	44 % (11)	38 % (8)	0 % (0)
Compatible $\lvert E_n \rvert < 1$	40 % (10)	76 % (16)	100 % (2)
Warning sig. $\lvert E_n \rvert > 1$	12 % (3)	19 % (4)	0 % (0)
Action sig. $\lvert E_n \rvert > 1.5$	48 % (12)	5 % (1)	0 % (0)

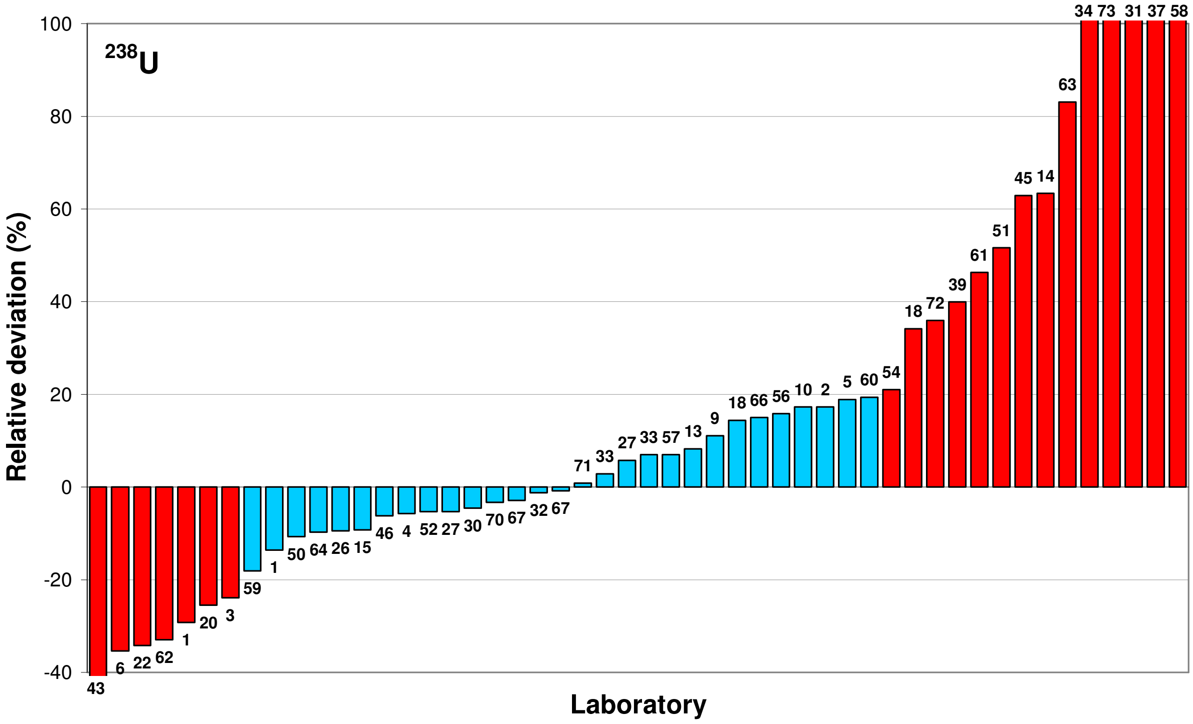

Fig. 85. Deviation chart of the participants' results of ^{238}U plotted in ascending order. Blue colour indicates results within the range ± 20 % from the reference value and red indicates results outside this range. Numbers indicate the laboratory code.

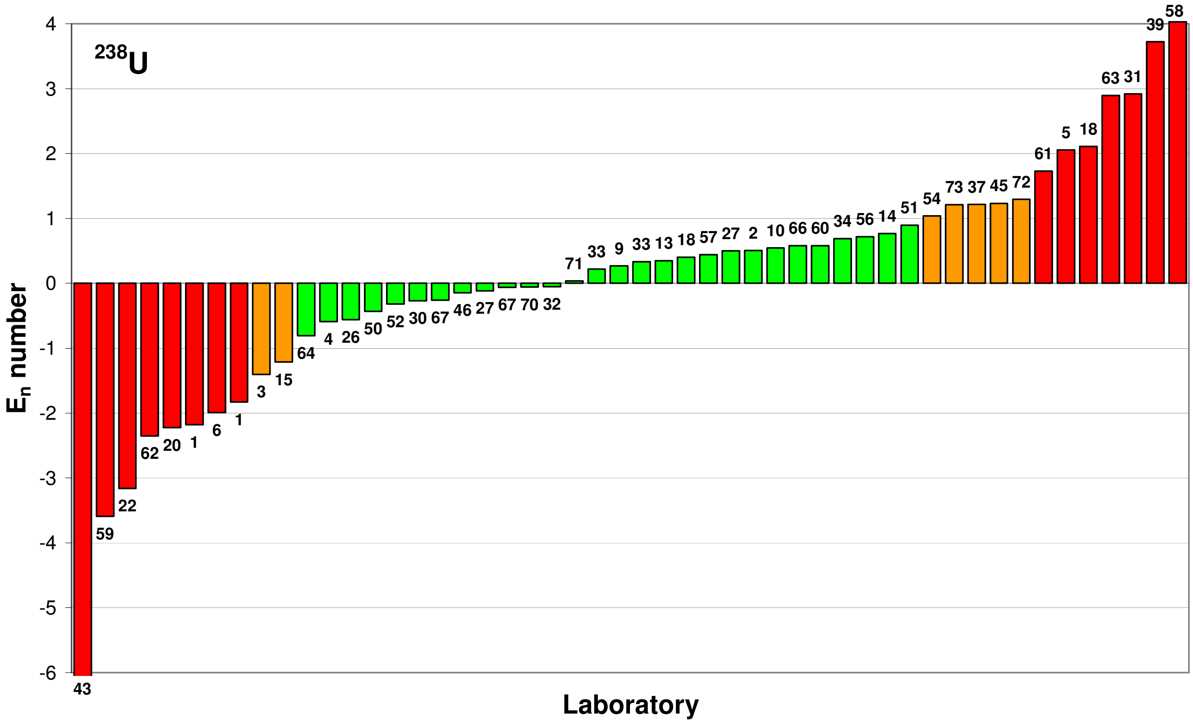

Fig. 86. E_n numbers' chart of ^{238}U activity concentrations plotted in ascending order. Green colour indicates compatible results, orange indicates warning signal and red indicates action signal. Numbers indicate the laboratory code.

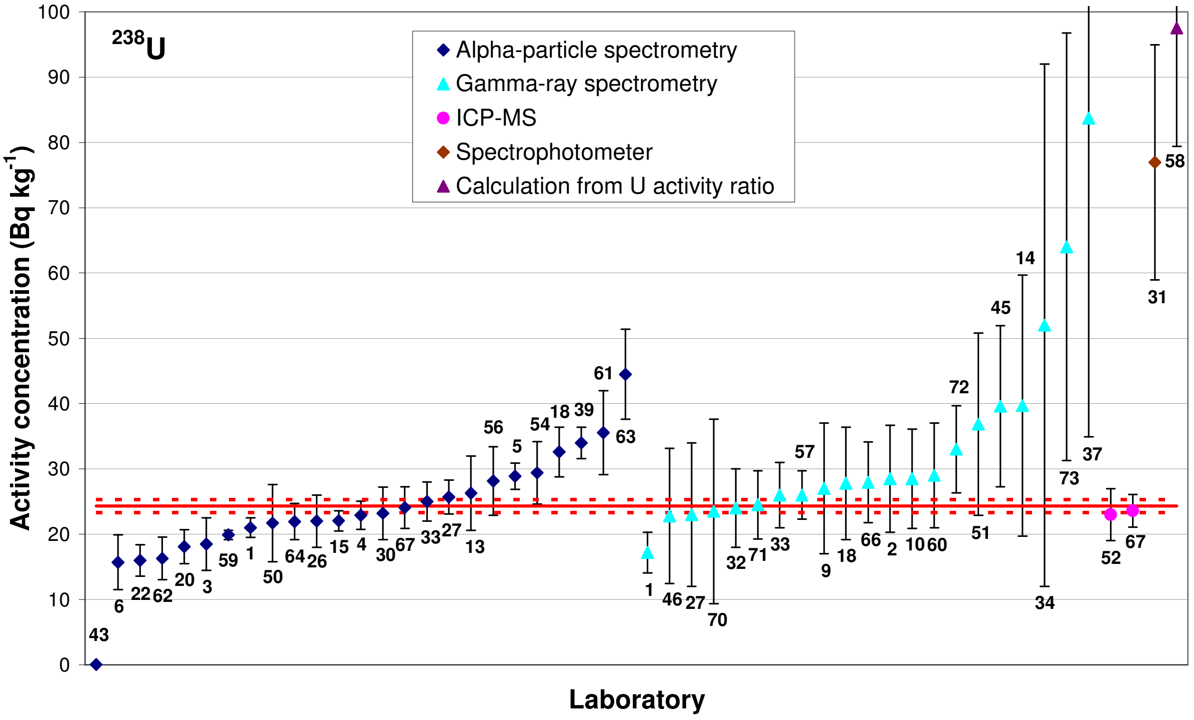

Fig. 87. Laboratory results A_{lab} of ^{238}U activity concentration with expanded uncertainties U_{lab} ($k = 2$) sorted according to counting methods. Red lines represent reference value $A_{ref} \pm U_{ref}$ ($k = 2$). Numbers indicate the laboratory code.

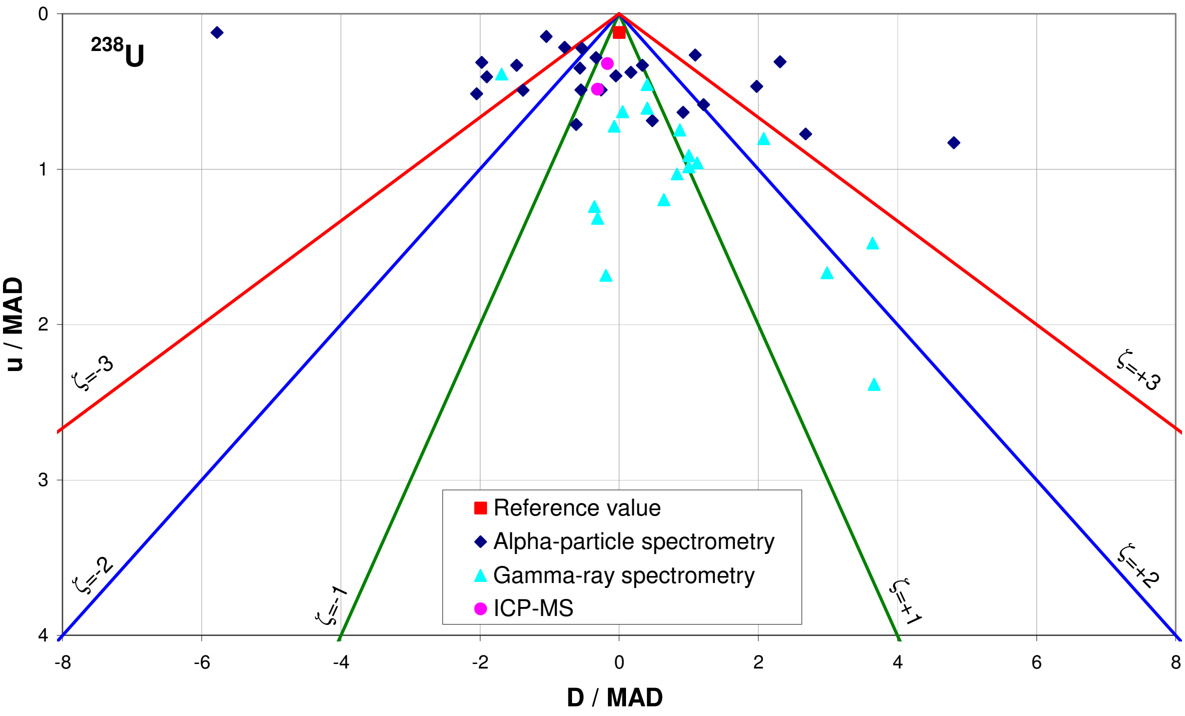

Fig. 88. PomPlot of the ^{238}U data sorted according to measurement method. Red point indicates the reference value. Green, blue and red solid lines indicate ζ-scores = 1, 2 and 3, respectively.

A summary of the performance of participating laboratories for all radionuclides is given in Table 20.

Table 20. Performance of laboratories expressed as percentages of results within the range of ± 20 % from the reference values and in terms of the E_n criterion.

Radionuclide	Number of results	Results within range ± 20% (%)	Compatible $\lvert E_n \rvert \leq 1$ (%)	Warning signal $1.5 \geq \lvert E_n \rvert > 1$ (%)	Action signal $\lvert E_n \rvert > 1.5$ (%)
^{40}K	70	89	71	11	18
^{90}Sr	40	35 (65)*	35	23	43
^{137}Cs	72	94	71	12	17
^{212}Pb	63	81	-	-	-
^{212}Bi	57	67	-	-	-
^{214}Pb	62	61	-	-	-
^{214}Bi	63	71	-	-	-
^{226}Ra	50	60	68	14	18
^{230}Th	22	45	50	9	41
^{232}Th	46	78	81	4	15
^{234}U	28	68	57	11	32
^{235}U	38	26	42	16	42
^{238}U	50	58	56	14	30

* 30 % threshold

Some results of this comparison were published, together with the concept of determining the reference values for the comparison samples, in the open literature (Merešová et al., 2012).

5.4 Comparison of different types of laboratories

An attempt to compare the performance of different types of laboratories, in terms of the accreditation status and source of nomination, was made. Only the scores for main radionuclides (^{40}K, ^{90}Sr, ^{137}Cs, ^{226}Ra and uranium isotopes) were investigated.

Accreditation

One of the questions in the questionnaire (Appendix 9) was whether the laboratory is accredited according to ISO 17025 (ISO, 2005b) and if yes for which radionuclides and methods. This chapter compares participants' results with the reference values, taking into account this aspect.

In the case of gamma-ray spectrometry, 31 laboratories out of 73 (42 %) were accredited to measure activities of gamma-emitting radionuclides in soil matrix, 32 participants declared that their laboratory is not accredited for this method and 10 (14 %) laboratories did not submit a questionnaire or did not answer this particular question.

For ^{40}K and ^{137}Cs, all accredited laboratories reported values within ± 20 % from the reference values. However, up to 20 % of these laboratories do not treat the uncertainties properly, as indicated by the warning and action signals triggered in the E_n evaluation. More details are presented in Table 21. The PomPlot of the ^{40}K data sorted according to the accreditation status is presented in Fig. 89. It can be seen that none of the accredited laboratories reported values outside of the $\zeta = \pm 3$ lines whereas several non-accredited laboratories did. A similar situation can be seen in Fig. 90 for ^{137}Cs.

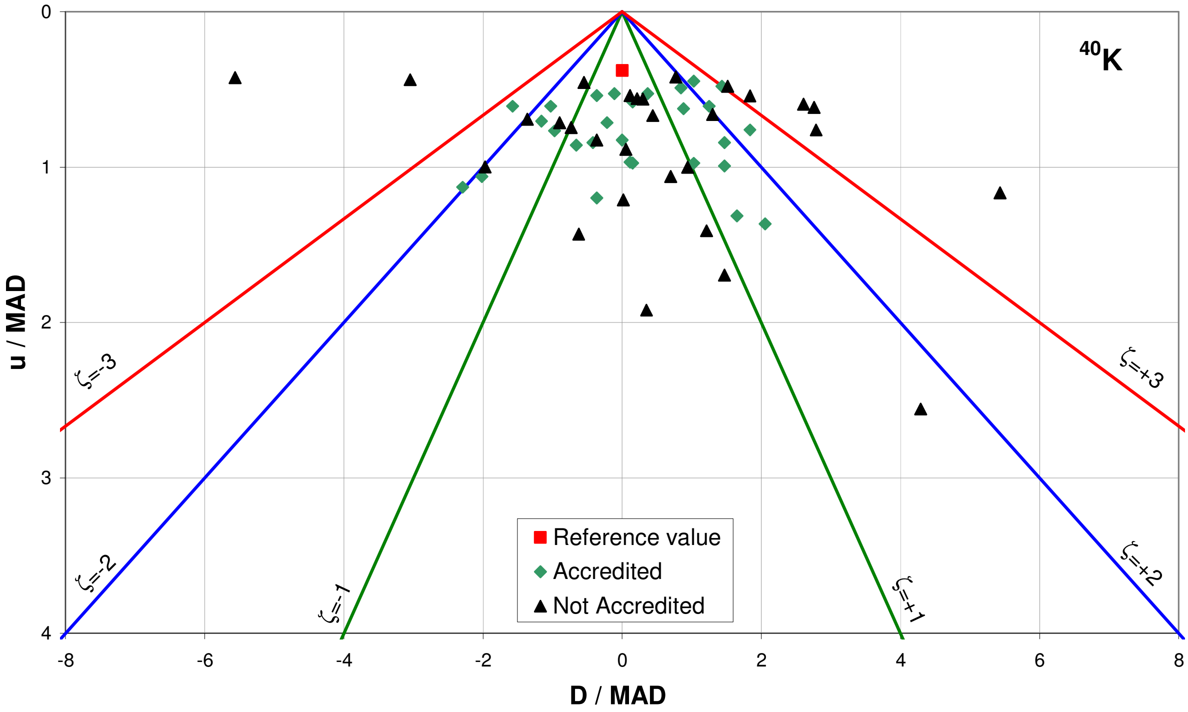

Fig. 89. PomPlot of the ^{40}K data sorted according to accreditation status. Red point indicates the reference value. Green, blue and red solid lines indicate ζ-scores = 1, 2 and 3, respectively.

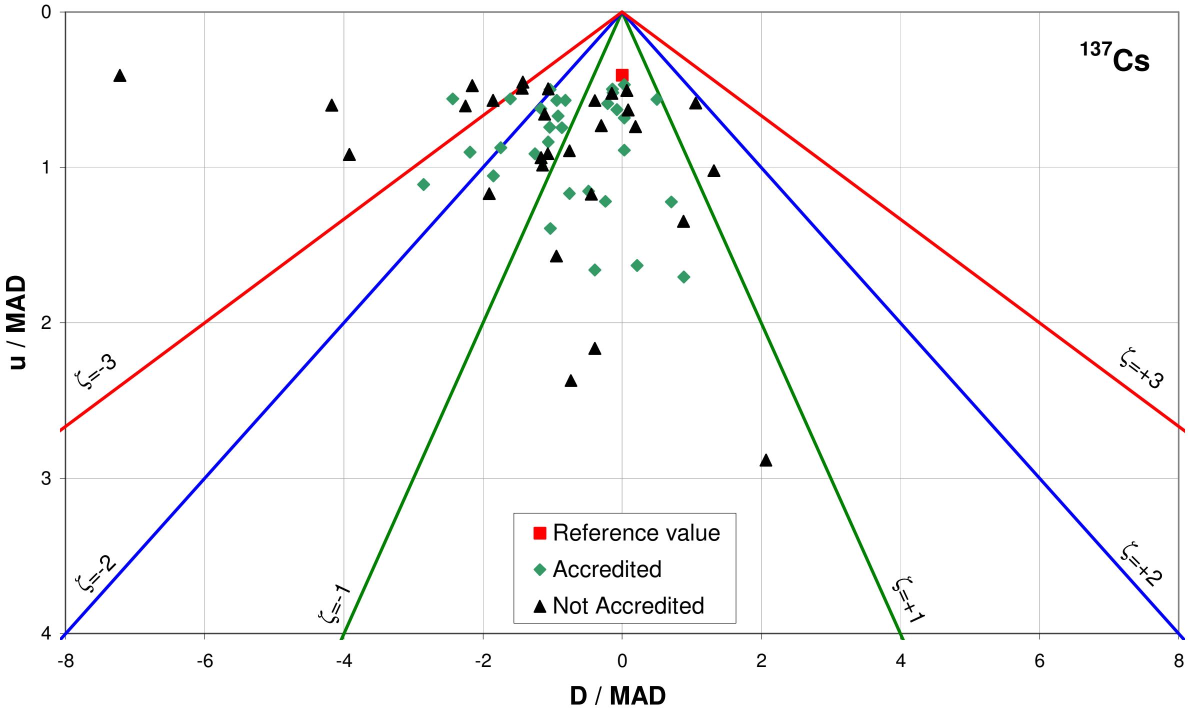

Fig. 90. PomPlot of the ^{137}Cs data sorted according to accreditation status. Red point indicates the reference value. Green, blue and red solid lines indicate ζ-scores = 1, 2 and 3, respectively.

Table 21. Overview of the laboratory performances for different accreditation status for ^{40}K and ^{137}Cs results. The number in parentheses indicates number of laboratories.

	^{40}K		^{137}Cs	
	Accredited (29)	Non-accredited (31)	Accredited (31)	Non-accredited (31)
Within ± 20 %	100 % (29)	81 % (25)	100 % (31)	90 % (28)
Outside ± 20 %	0 % (0)	19 % (6)	0 % (0)	10 % (3)
Compatible $\|E_n\| < 1$	79 % (23)	68 % (21)	84 % (26)	65 % (20)
Warning sig. $\|E_n\| > 1$	21 % (6)	0 % (0)	13 % (4)	6 % (2)
Action sig. $\|E_n\| > 1.5$	0 % (0)	32 % (10)	3 % (1)	29 % (9)

The activity concentration of ^{226}Ra was measured by gamma-ray spectrometry in 43 out of 50 laboratories (86 %). About half of them are accredited for this method but the quality of results does not differ significantly from the quality of results reported by the non-accredited laboratories (Table 22). On the contrary, four laboratories which applied alpha-particle spectrometry rendered the best results although they are not accredited for this method. The results are presented in Fig. 91.

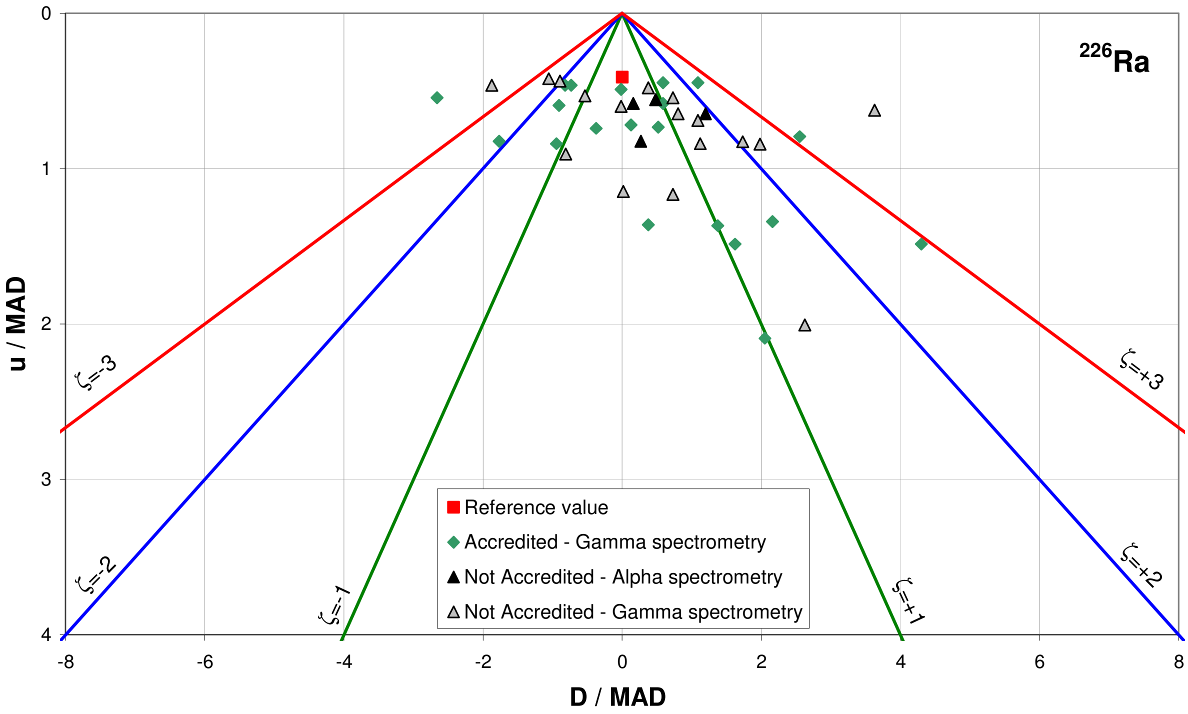

Fig. 91. PomPlot of the ^{226}Ra data sorted according to accreditation status. Red point indicates the reference value. Green, blue and red solid lines indicate ζ-scores = 1, 2 and 3, respectively.

Table 22. Overview of the laboratory performances for different methods and accreditation status for ^{226}Ra results. The number in parentheses indicates number of laboratories.

^{226}Ra	Gamma-ray spectrometry		Alpha-particle spectrometry	
	Accredited (20)	Non-accredited (19)	Accredited (0)	Non-accredited (4)
Within ± 20 %	55 % (11)	63 % (12)	-	100 % (4)
Outside ± 20 %	45 % (9)	37 % (7)	-	0 % (0)
Compatible $\lvert E_n \rvert < 1$	70 % (14)	63 % (12)	-	100 % (4)
Warning sig. $\lvert E_n \rvert > 1$	15 % (3)	21 % (4)	-	0 % (0)
Action sig. $\lvert E_n \rvert > 1.5$	15 % (3)	16 % (3)	-	0 % (0)

All results of ^{234}U, except for three, were determined via the alpha-particle spectrometry method, 9 of which were accredited and 13 not. As can be seen in Table 23 and Fig. 92, the results of accredited methods show poorer performance with only 55.5 % of results deviating less than ± 20 % from the reference value in comparison to the results of non-accredited methods with 69 % of results within the range.

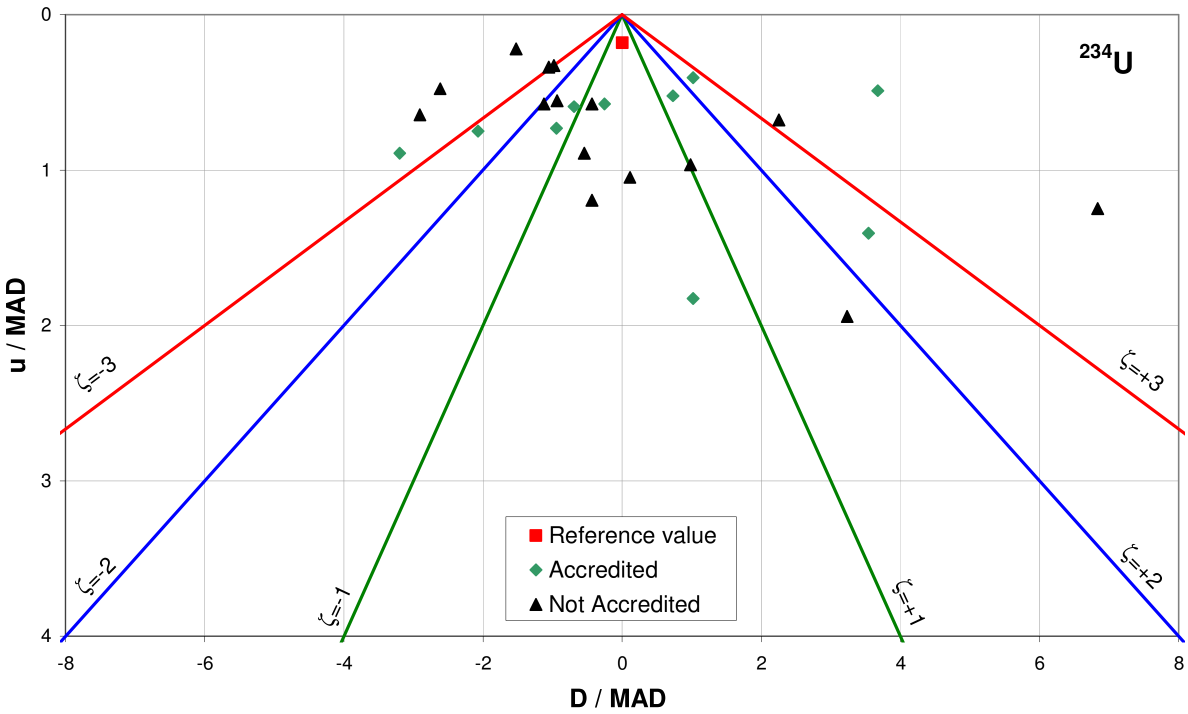

Fig. 92. PomPlot of the ^{234}U data sorted according to accreditation status. Red point indicates the reference value. Green, blue and red solid lines indicate ζ-scores = 1, 2 and 3, respectively.

In the case of ^{235}U (Table 24), 17 laboratories applied gamma-ray spectrometry. Only one out of five (20 %) accredited laboratories determined values within ± 20 % from the reference value and is E_n compatible. None of the 11 non-accredited laboratories reported values deviating less than 20 % from the reference value and only two results (18 %) were E_n compatible. Alpha-particle spectrometry was applied in 19 laboratories out of which seven are accredited, nine are non-accredited and we have no information about the three left. There is no difference between accredited and non-accredited laboratories, with 43 % and 44.5 % of results, respectively, deviating less than 20 % from the reference value. For E_n classification, the difference is significant with 72 % and 44.5 % compatible results for accredited and non-accredited laboratories, respectively. From Table 25 and Fig. 93 it is obvious that many laboratories using gamma-ray spectrometry significantly overestimated activity concentration of ^{235}U, whether accredited or not.

Table 23. Overview of the laboratory performances for different accreditation status for ^{234}U results determined via alpha-particle spectrometry. The number in parentheses indicates number of laboratories.

^{234}U	Accredited (9)	Non-accredited (13)
Within ± 20 %	55.5 % (5)	69 % (9)
Outside ± 20 %	44.5 % (4)	31 % (4)
Compatible $\lvert E_n \rvert < 1$	45 % (4)	46 % (6)
Warning sig. $\lvert E_n \rvert > 1$	33 % (3)	0 % (0)
Action sig. $\lvert E_n \rvert > 1.5$	22 % (2)	54 % (7)

Table 24. Overview of the laboratory performances for different methods and accreditation status for ^{235}U results. The number in parentheses indicates number of laboratories.

^{235}U	Gamma-ray spectrometry		Alpha-particle spectrometry	
	Accredited (5)	Non-accredited (11)	Accredited (7)	Non-accredited (9)
Within ± 20 %	20 % (1)	0 % (0)	43 % (3)	44.5 % (4)
Outside ± 20 %	80 % (4)	100 % (11)	57 % (4)	55.5 % (5)
Compatible $\lvert E_n \rvert < 1$	20 % (1)	18 % (2)	72 % (5)	44.5 % (4)
Warning sig. $\lvert E_n \rvert > 1$	20 % (1)	18 % (2)	14 % (1)	11 % (1)
Action sig. $\lvert E_n \rvert > 1.5$	60 % (3)	64 % (7)	14% (1)	44.5 % (4)

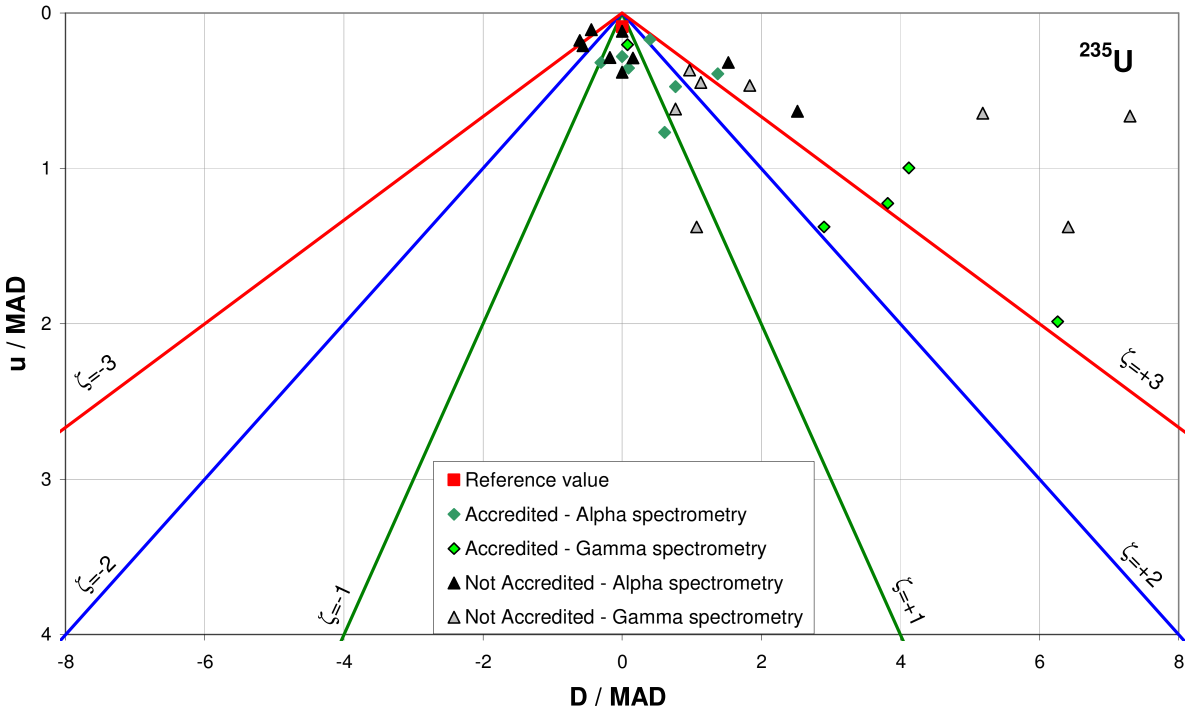

Fig. 93. PomPlot of the ^{235}U data sorted according to accreditation status. Red point indicates the reference value. Green, blue and red solid lines indicate ζ-scores = 1, 2 and 3, respectively.

Almost one half (20 out of 50) of the ^{238}U results were determined via gamma-ray spectrometry, where 11 methods are accredited. The performance of these laboratories, with 91 % of results deviating less than 20 % from the reference value and 100 % of E_n compatible results, is much better than the performance achieved with the non-accredited methods, with only 33 % within ± 20 % from the reference value and 44.5 % E_n satisfactory. Performance is independent from the accreditation status for alpha-particle spectrometry as can be seen in Table 25 and Fig. 94.

Table 25. Overview of the laboratory performances for different methods and accreditation status for ^{238}U results. The number in parentheses indicates number of laboratories.

^{238}U	Gamma-ray spectrometry		Alpha-particle spectrometry	
	Accredited (11)	Non-accredited (9)	Accredited (8)	Non-accredited (14)
Within ± 20 %	91 % (10)	33 % (3)	50 % (4)	57 % (8)
Outside ± 20 %	9 % (1)	67 % (6)	50 % (4)	43 % (6)
Compatible $\|E_n\| < 1$	100 % (11)	44.5 % (4)	37.5 % (3)	36 % (5)
Warning sig. $\|E_n\| > 1$	0 % (0)	44.5 % (4)	12.5 % (1)	7 % (1)
Action sig. $\|E_n\| > 1.5$	0 % (0)	11 % (1)	50 % (4)	57 % (8)

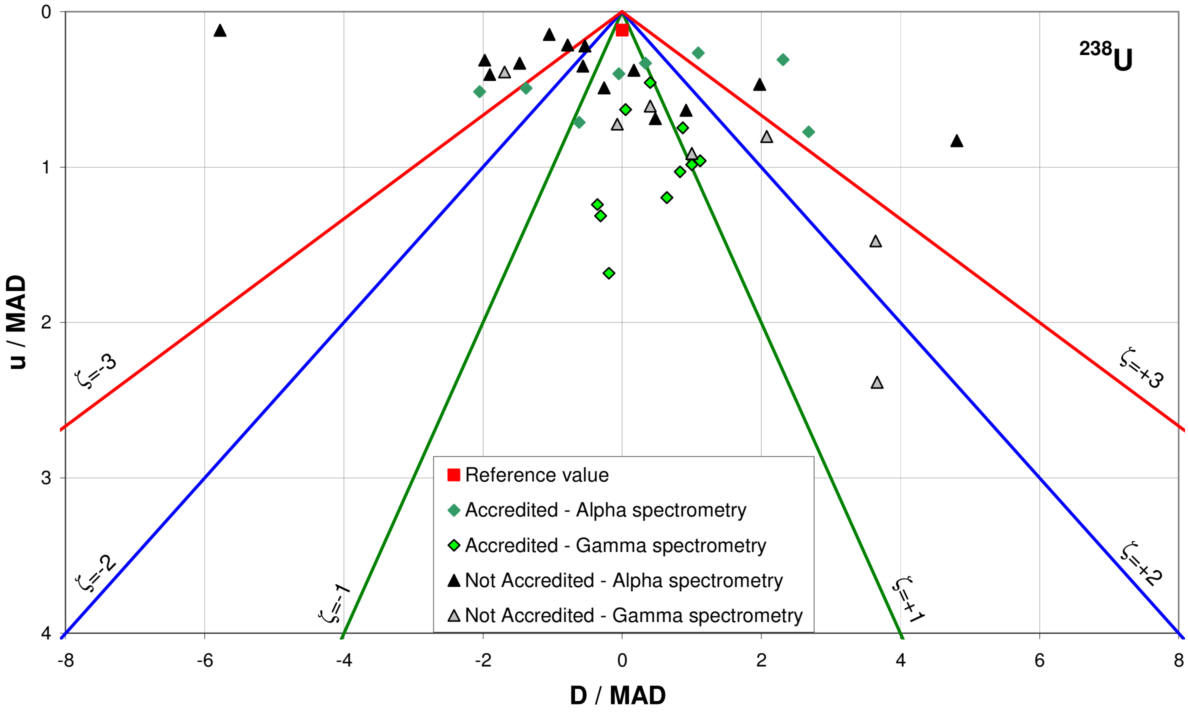

Fig. 94. PomPlot of the ^{238}U data sorted according to accreditation status. Red point indicates the reference value. Green, blue and red solid lines indicate ζ-scores = 1, 2 and 3, respectively.

In the case of ^{90}Sr, a large variety of separation and counting techniques was used. Only 14 out of 40 (35 %) laboratories used accredited methods and for five laboratories we have no information. Although the performance of accredited laboratories is unsatisfactory with only half of the results within ± 20 % from the reference value and compatible with the E_n criterion, in comparison to non-accredited laboratories this performance is still much better (Table 26 and Fig. 95).

Table 26. Overview of the laboratory performances for different accreditation status for ^{90}Sr results. The number in parentheses indicates number of laboratories.

^{90}Sr	Accredited (14)	Non-accredited (21)
Within ± 20 %	50 % (7)	19 % (4)
Outside ± 20 %	50 % (7)	81 % (17)
Compatible $\|E_n\| < 1$	50 % (7)	24 % (5)
Warning sig. $\|E_n\| > 1$	21 % (3)	19 % (4)
Action sig. $\|E_n\| > 1.5$	29 % (4)	57 % (12)

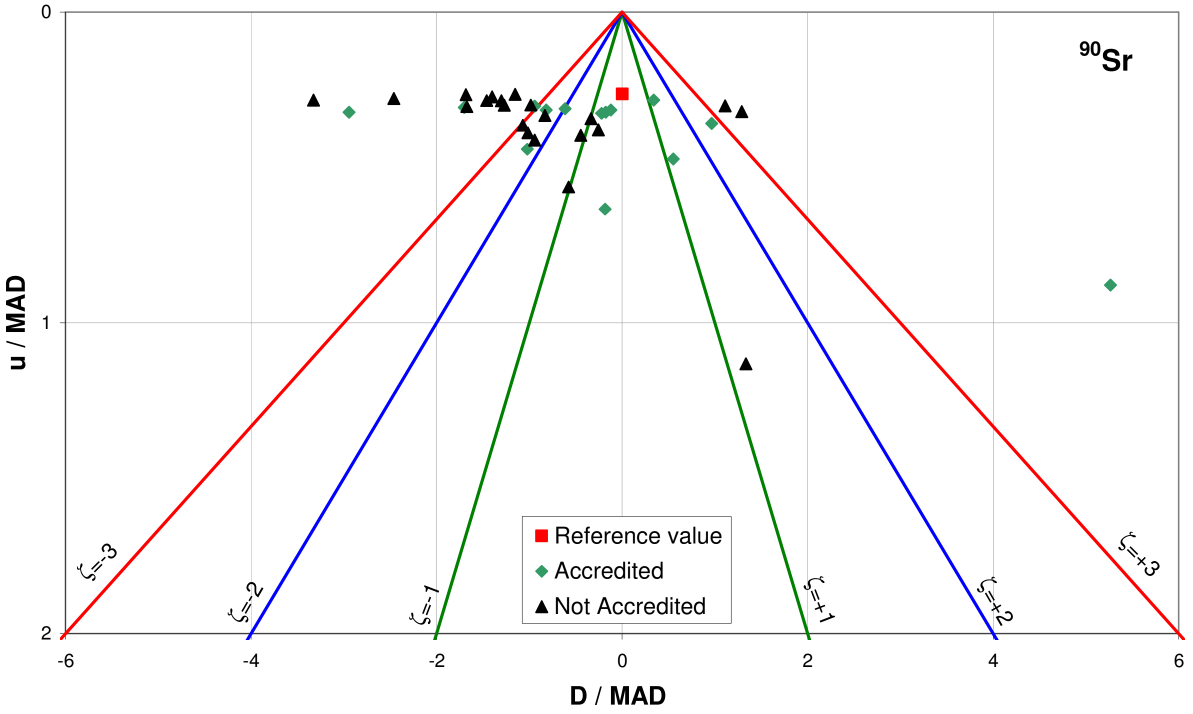

Fig. 95. PomPlot of the ^{90}Sr data sorted according to accreditation status. Red point indicates the reference value. Green, blue and red solid lines indicate ζ-scores = 1, 2 and 3, respectively.

In general, we can conclude that in most cases the performance of laboratories using accredited methods is better; their uncertainty estimation appears to be more realistic as well (E_n compatibility). Surprisingly, in some cases, like gamma-ray spectrometry of ^{226}Ra or alpha-particle spectrometry of ^{234}U and ^{238}U, it is vice versa and laboratories applying non-accredited methods reported better results in terms of the deviation criterion, but not with respect to the E_n criterion due to the weakness in uncertainty estimation.

Group of nomination source

Laboratories were divided into three groups according to the source of their nomination. Group of nomination source EU member state (MS) laboratories were nominated by their national representatives in the expert group according to the Euratom Treaty Art. 35/36. In addition, several laboratories from pre-accession countries (AC) were invited by JRC IRMM to participate in this ILC. Finally, IAEA nominated the last group of laboratories worldwide.

For ^{40}K, the IAEA laboratories show the worst performance with 29 % of results outside the ± 20 % range (Table 27 and Fig. 96). Performance for ^{137}Cs is similar in all three types of laboratories (Table 27 and Fig. 97).

Similarly for ^{90}Sr, the IAEA participants show the worst performance with less than 20 % of the results E_n compatible and within ± 20 % from the reference value. This is also obvious from Fig. 98. On the contrary, for ^{226}Ra the IAEA laboratories are the best in terms of relative differences as well as E_n numbers (Table 28 and Fig. 99).

Table 27. Overview of the laboratory performances for ^{40}K and ^{137}Cs according to nomination source. The number in parentheses indicates number of laboratories.

	^{40}K			^{137}Cs		
	MS (48)	AC (8)	IAEA (14)	MS (49)	AC (8)	IAEA (15)
Within ± 20 %	92 % (44)	100 % (8)	71 % (10)	92 % (45)	100 % (8)	100 % (15)
Outside ± 20 %	8 % (4)	0 % (0)	29 % (4)	8 % (4)	0 % (0)	0 % (0)
Compatible $\|E_n\| < 1$	71 % (34)	88 % (7)	64 % (9)	71 % (35)	75 % (6)	67 % (10)
Warning sig. $\|E_n\| > 1$	13 % (6)	0 % (0)	14 % (2)	12 % (6)	13 % (1)	13 % (2)
Action sig. $\|E_n\| > 1.5$	17 % (8)	13 % (1)	21 % (3)	16 % (8)	13 % (1)	20 % (3)

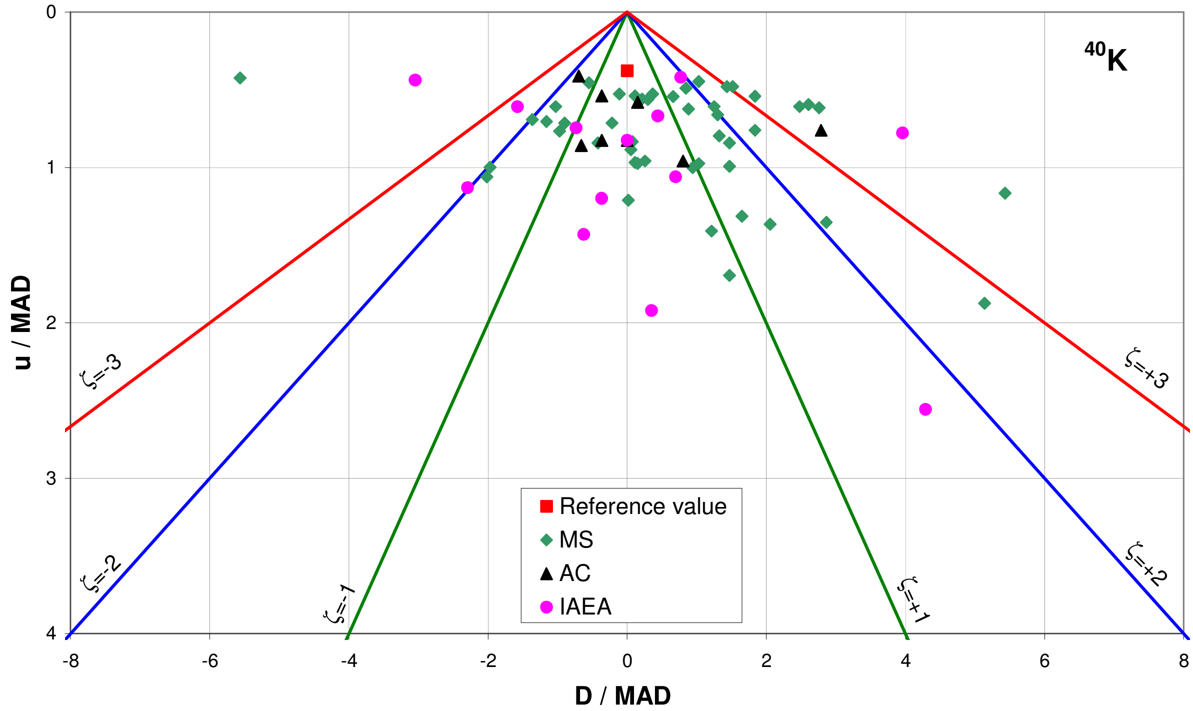

Fig. 96. PomPlot of the ^{40}K data sorted according to source of nomination. Red point indicates the reference value. Green, blue and red solid lines indicate ζ-scores = 1, 2 and 3, respectively.

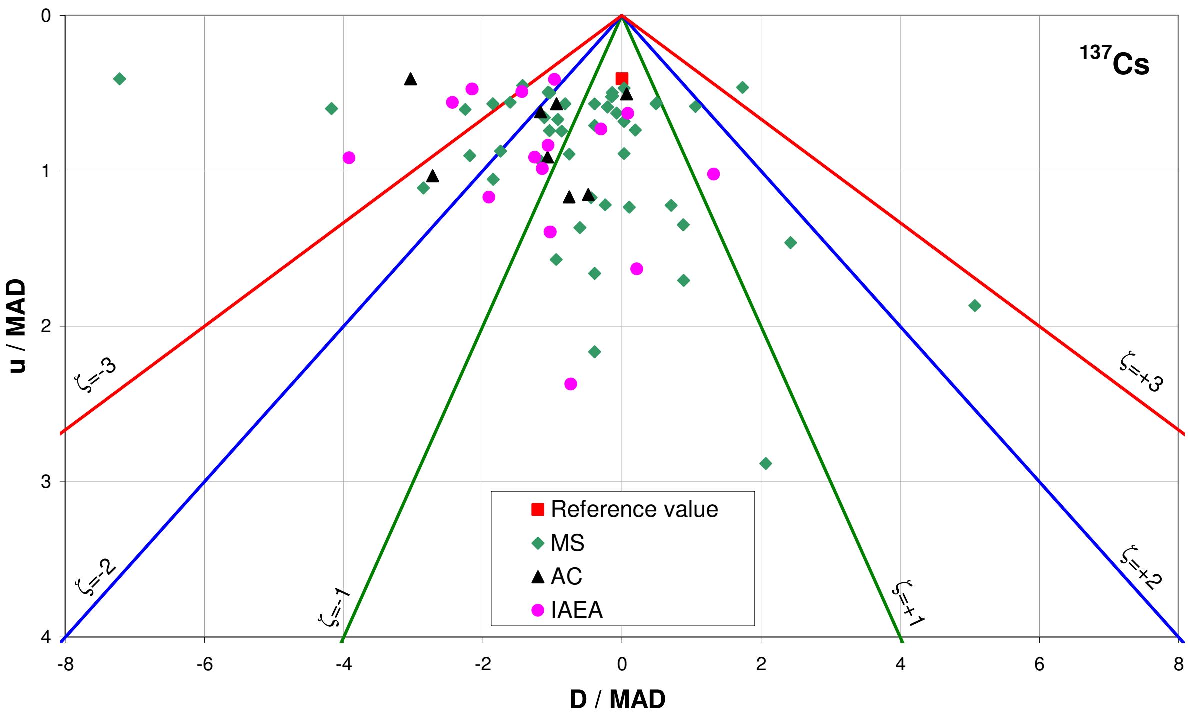

Fig. 97. PomPlot of the ^{137}Cs data sorted according to source of nomination. Red point indicates the reference value. Green, blue and red solid lines indicate ζ-scores = 1, 2 and 3, respectively.

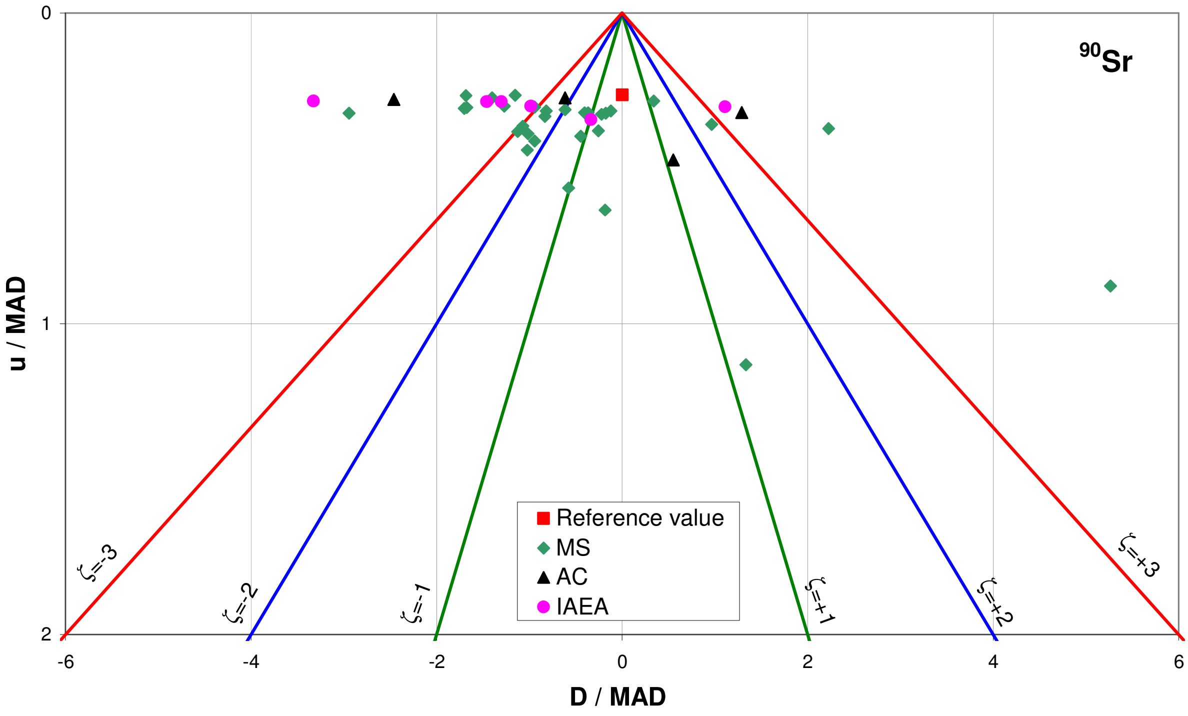

Fig. 98. PomPlot of the ^{90}Sr data sorted according to source of nomination. Red point indicates the reference value. Green, blue and red solid lines indicate ζ-scores = 1, 2 and 3, respectively.

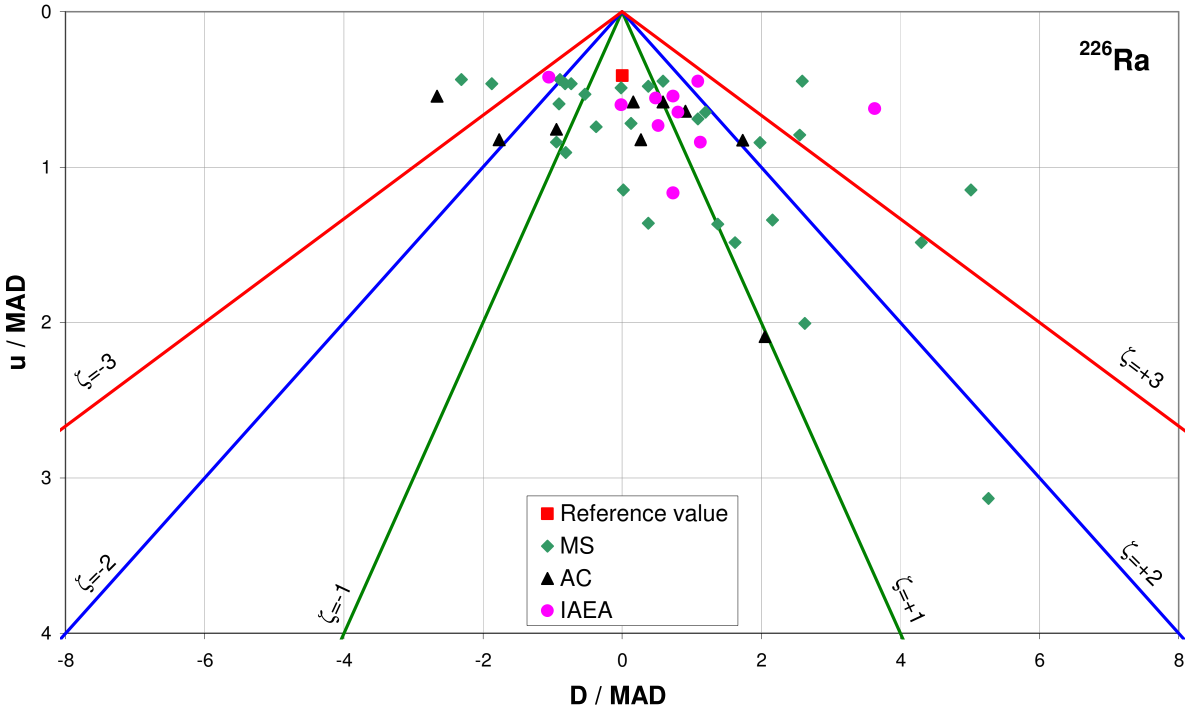

Fig. 99. PomPlot of the ^{226}Ra data sorted according to source of nomination. Red point indicates the reference value. Green, blue and red solid lines indicate ζ-scores = 1, 2 and 3, respectively.

Table 28. Overview of the laboratory performances for ^{90}Sr and ^{226}Ra according to source of nomination. The number in parentheses indicates number of laboratories.

	^{90}Sr			^{226}Ra		
	MS (30)	AC (4)	IAEA (6)	MS (30)	AC (9)	IAEA (11)
Within ± 20 %	37 % (11)	50 % (4)	17 % (1)	53 % (16)	56 % (5)	82 % (9)
Outside ± 20 %	63 % (19)	50 % (4)	83 % (5)	47 % (14)	44 % (4)	18 % (2)
Compatible $\|E_n\| < 1$	40 % (12)	25 % (2)	17 % (1)	67 % (20)	67 % (6)	73 % (8)
Warning sig. $\|E_n\| > 1$	27 % (8)	25 % (2)	0 % (0)	10 % (3)	22 % (2)	18 % (2)
Action sig. $\|E_n\| > 1.5$	33 % (10)	50 % (4)	83 % (5)	23 % (7)	11 % (1)	9 % (1)

All AC laboratories reported results deviating less than 20 % from the reference value of ^{234}U but in E_n statistics MS laboratories are better as can be seen in Table 29. In the case of ^{235}U, all AC participants failed to determine the correct value and performance of the MS laboratories is better than the one of the IAEA participants which reported results (Table 29). For ^{238}U, the performance of all types of laboratories is comparable as shown in Table 30. and Fig. 102.

Table 29. Overview of the laboratory performances for ^{234}U and ^{235}U according to source of nomination. The number in parentheses indicates number of laboratories.

	^{234}U			^{235}U		
	MS (21)	AC (4)	IAEA (3)	MS (26)	AC (4)	IAEA (8)
Within ± 20 %	62 % (13)	100 % (4)	67 % (2)	35 % (9)	0 % (0)	12.5 % (1)
Outside ± 20 %	38 % (8)	0 % (0)	33 % (1)	65 % (17)	100 % (4)	87.5 % (7)
Compatible $\|E_n\| < 1$	62 % (13)	50 % (2)	33 % (1)	50 % (13)	0 % (0)	37.5 % (3)
Warning sig. $\|E_n\| > 1$	10 % (2)	25 % (1)	0 % (0)	15 % (4)	25 % (1)	12.5 % (1)
Action sig. $\|E_n\| > 1.5$	28 % (6)	25 % (1)	67 % (2)	35 % (9)	75 % (3)	50 % (4)

Table 30. Overview of the laboratory performances for ^{238}U according to source of nomination. The number in parentheses indicates number of laboratories.

	^{238}U		
	MS (32)	AC (9)	IAEA (9)
Within ± 20 %	56 % (18)	67 % (6)	56 % (5)
Outside ± 20 %	44 % (14)	33 % (3)	44 % (4)
Compatible $\|E_n\| < 1$	56 % (18)	56 % (5)	56 % (5)
Warning sig. $\|E_n\| > 1$	19 % (6)	0 % (0)	11 % (1)
Action sig. $\|E_n\| > 1.5$	25 % (8)	44 % (4)	33 % (3)

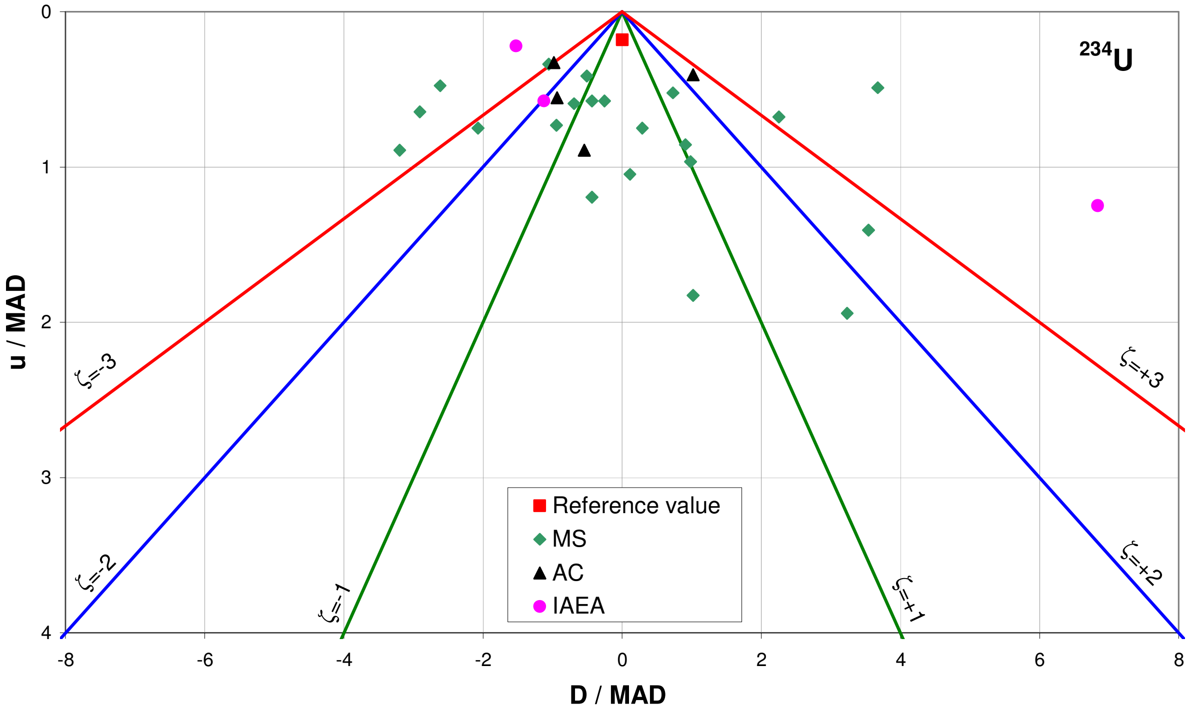

Fig. 100. PomPlot of the ^{234}U data sorted according to nomination source. Red point indicates the reference value. Green, blue and red solid lines indicate ζ-scores = 1, 2 and 3, respectively.

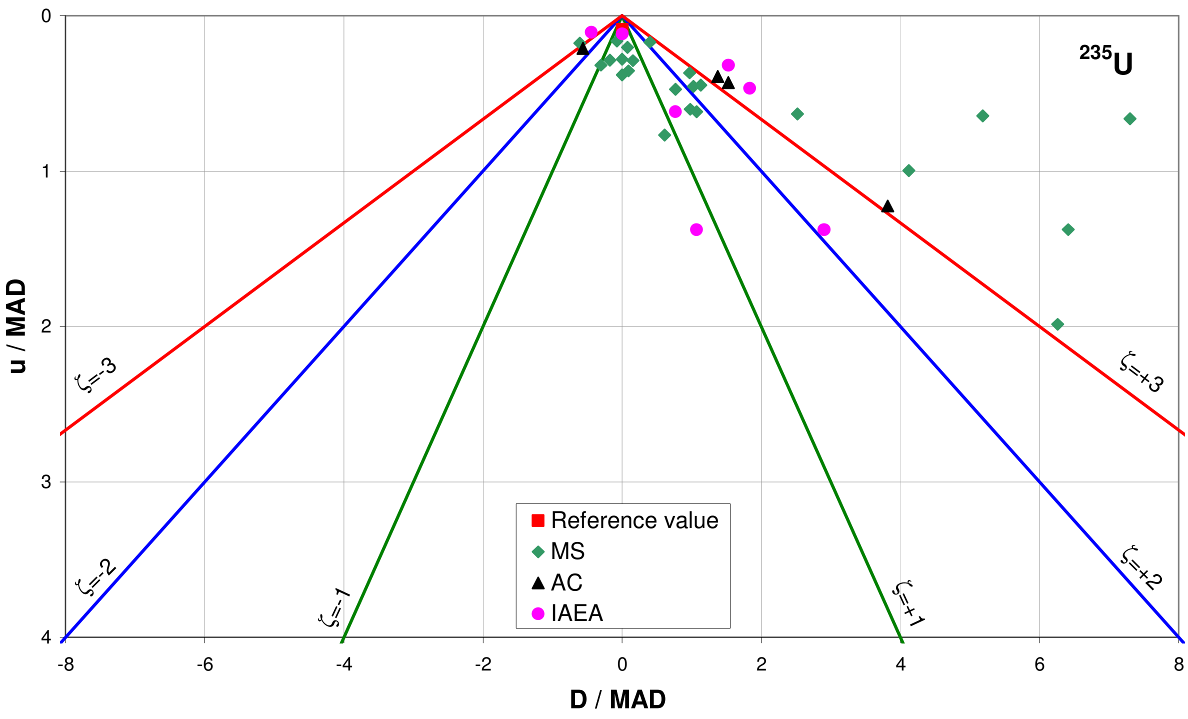

Fig. 101. PomPlot of the ^{235}U data sorted according to nomination source. Red point indicates the reference value. Green, blue and red solid lines indicate ζ-scores = 1, 2 and 3, respectively.

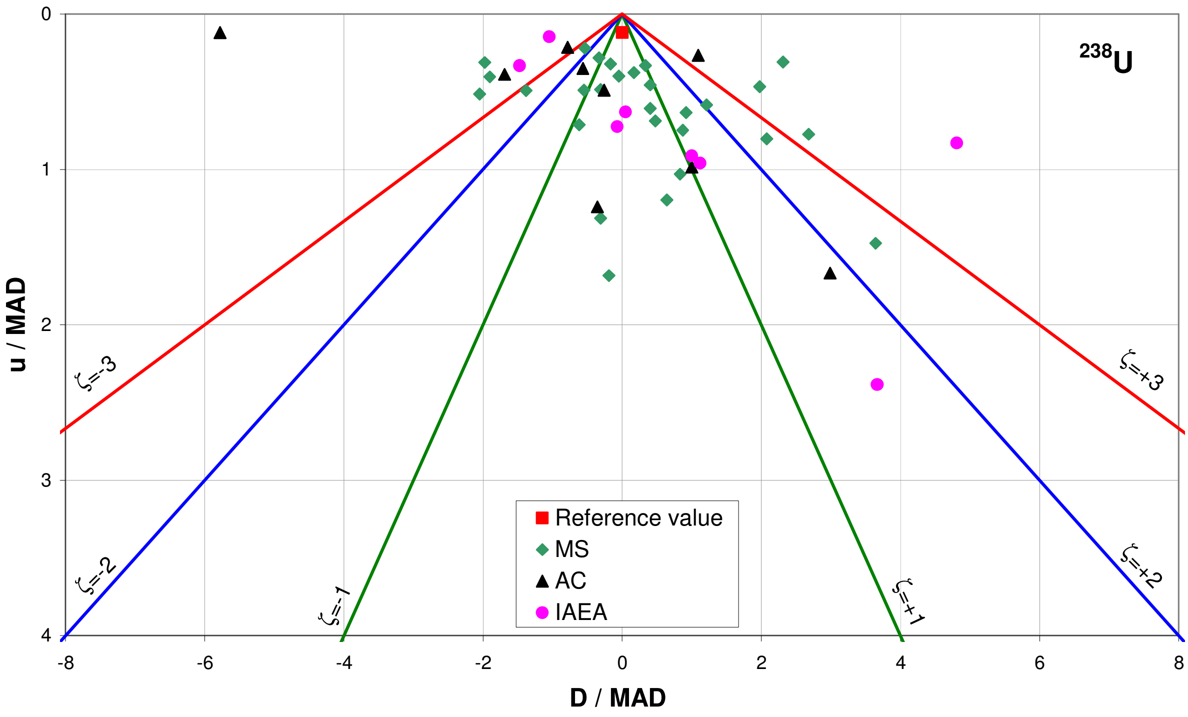

Fig. 102. PomPlot of the ^{238}U data sorted according to nomination source. Red point indicates the reference value. Green, blue and red solid lines indicate ζ-scores = 1, 2 and 3, respectively.

6 Conclusions

Soil from an area affected by the Chernobyl accident was used as comparison material in this interlaboratory comparison. The reference values traceable to SI units were determined at IRMM. The performance of 73 participating laboratories varied depending on the radionuclide determined and method used (Table 20). With a few exceptions, the comparison samples were treated and measured with routine procedures.

Gamma-ray spectrometry with respect to ^{137}Cs and ^{40}K is relatively well controlled in the laboratories. The determination of ^{90}Sr proved difficult for 65 % of the participants which submitted results outside the acceptable range (± 20 %). No improvement could be seen compared to ^{90}Sr determination in one of the previous ILC exercises (Wätjen et al., 2008). The laboratories concerned, i.e. the vast majority of laboratories reporting ^{90}Sr results, are urged to review their analysis procedures. The results clearly demonstrate that several laboratories need to improve their analytical procedures for determination of uranium isotopes. Mainly results of ^{235}U proved to be highly method dependent when gamma-ray spectrometry rendered very poor results. This is most probably due to the lack of application of appropriate corrections in these measurements. A similar situation was observed for ^{226}Ra with unsatisfactory scores for gamma-ray spectrometry results and most probably caused by similar reasons as in the case of ^{235}U. Surprisingly, in the case of ^{232}Th the results of gamma-ray spectrometry are significantly better compared to alpha-particle spectrometry results. However, the performance is more or less equal for both measurement methods for ^{230}Th.

One of the laboratories (lab 72) reported a result of ^{235}U 50-times larger than the reference value. Also the results of ^{40}K and ^{226}Ra were overestimated by a factor of 2.1 and 1.7, respectively, by the same participant (lab 72), while the result of ^{137}Cs was underestimated by almost 40 %. Similar mediocre performance is observed also for other participants (labs: 39, 14, 44). These observations are distressing and indicate that critical and prompt revision of the measurement methods is necessary in these laboratories.

Performance evaluation based on the E_n criterion revealed that uncertainty estimation is unsatisfactory in many laboratories and there is a need to improve their application of uncertainty propagation and implement the concept of the GUM (2008).

The results of this ILC have shown again that proficiency testing with comparison samples bearing reference values traceable to SI units (and SIR, where applicable) remains an important tool to evaluate laboratory performance. More efforts on the side of the participating laboratories are expected to turn bad ILC performance into improvement actions, leading to the revision of existing, obviously insufficient laboratory routines. It must be stressed that – as in any other application – only reliable methods should be used in environmental monitoring, and gamma-ray spectrometry must be applied with the greatest care when it comes to naturally occurring radionuclide material (NORM).

Acknowledgements

This work was possible only with the active participation of 73 laboratories in 39 countries (listed in Appendix 8), and the support of IAEA letting us purchase such a large amount of reference material for the purposes of this ILC. The authors would also like to thank G. Sibbens, M. Bickel, A. Bohnstedt, J.-G. Decaillon, C. Hill, and L. Holmes for their contributions to the establishment of the traceable reference values. Our acknowledgements also go to our colleagues from the Reference Materials Unit of IRMM for the re-processing of the original soil material and for determining the water content of the packaged samples. We are grateful to B. De La Calle for discussions of the manuscript.

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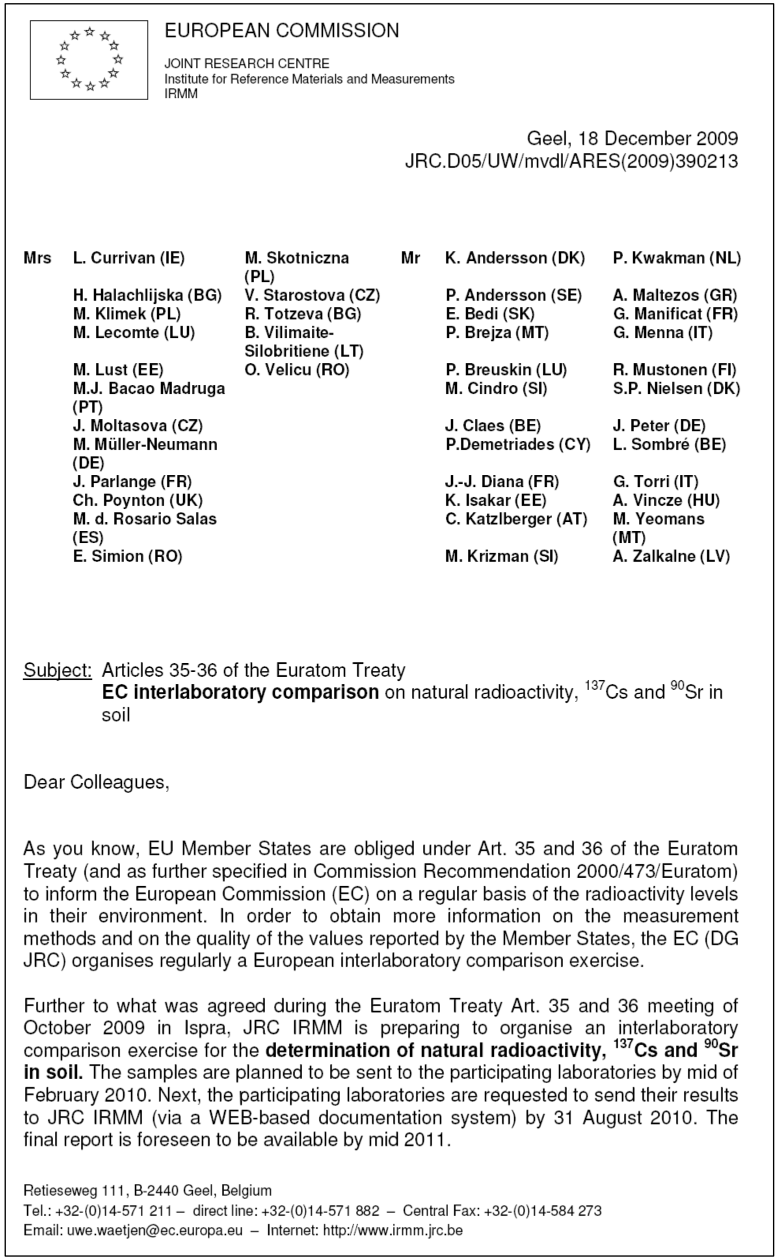
Appendix 1: Invitation letter



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

Geel, 18 December 2009
JRC.D05/UW/mvdl/ARES(2009)390213

Mrs

L. Currivan (IE)
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M. Klimek (PL)
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M. Lust (EE)
M.J. Bacao Madruga (PT)
J. Moltasova (CZ)
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E. Simion (RO)

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O. Velicu (RO)

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G. Torri (IT)
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M. Yeomans (MT)
A. Zalkalne (LV)

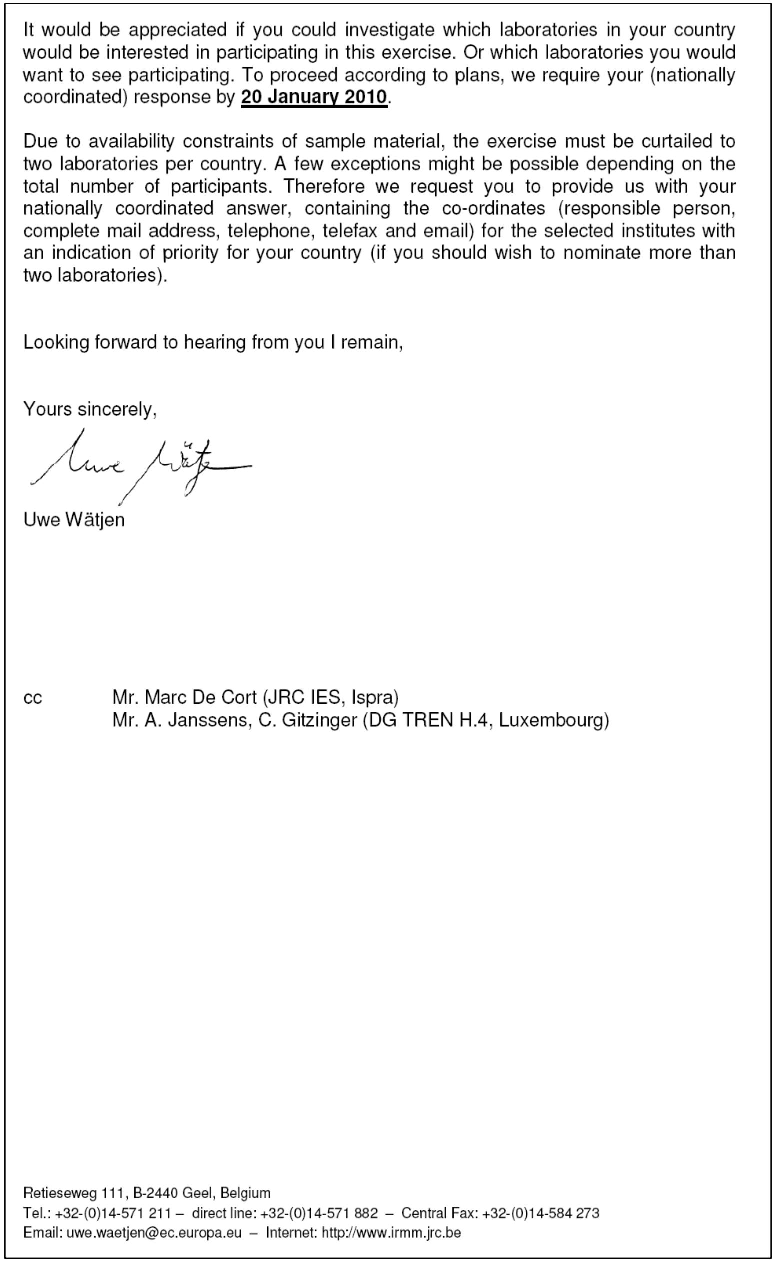
Subject: Articles 35-36 of the Euratom Treaty
EC interlaboratory comparison on natural radioactivity, ^{137}Cs and ^{90}Sr in soil

Dear Colleagues,

As you know, EU Member States are obliged under Art. 35 and 36 of the Euratom Treaty (and as further specified in Commission Recommendation 2000/473/Euratom) to inform the European Commission (EC) on a regular basis of the radioactivity levels in their environment. In order to obtain more information on the measurement methods and on the quality of the values reported by the Member States, the EC (DG JRC) organises regularly a European interlaboratory comparison exercise.

Further to what was agreed during the Euratom Treaty Art. 35 and 36 meeting of October 2009 in Ispra, JRC IRMM is preparing to organise an interlaboratory comparison exercise for the **determination of natural radioactivity, ^{137}Cs and ^{90}Sr in soil.** The samples are planned to be sent to the participating laboratories by mid of February 2010. Next, the participating laboratories are requested to send their results to JRC IRMM (via a WEB-based documentation system) by 31 August 2010. The final report is foreseen to be available by mid 2011.

Retieseweg 111, B-2440 Geel, Belgium
Tel.: +32-(0)14-571 211 – direct line: +32-(0)14-571 882 – Central Fax: +32-(0)14-584 273
Email: uwe.waetjen@ec.europa.eu – Internet: http://www.irmm.jrc.be



It would be appreciated if you could investigate which laboratories in your country would be interested in participating in this exercise. Or which laboratories you would want to see participating. To proceed according to plans, we require your (nationally coordinated) response by **20 January 2010**.

Due to availability constraints of sample material, the exercise must be curtailed to two laboratories per country. A few exceptions might be possible depending on the total number of participants. Therefore we request you to provide us with your nationally coordinated answer, containing the co-ordinates (responsible person, complete mail address, telephone, telefax and email) for the selected institutes with an indication of priority for your country (if you should wish to nominate more than two laboratories).

Looking forward to hearing from you I remain,

Yours sincerely,

Uwe Wätjen

cc Mr. Marc De Cort (JRC IES, Ispra)
Mr. A. Janssens, C. Gitzinger (DG TREN H.4, Luxembourg)

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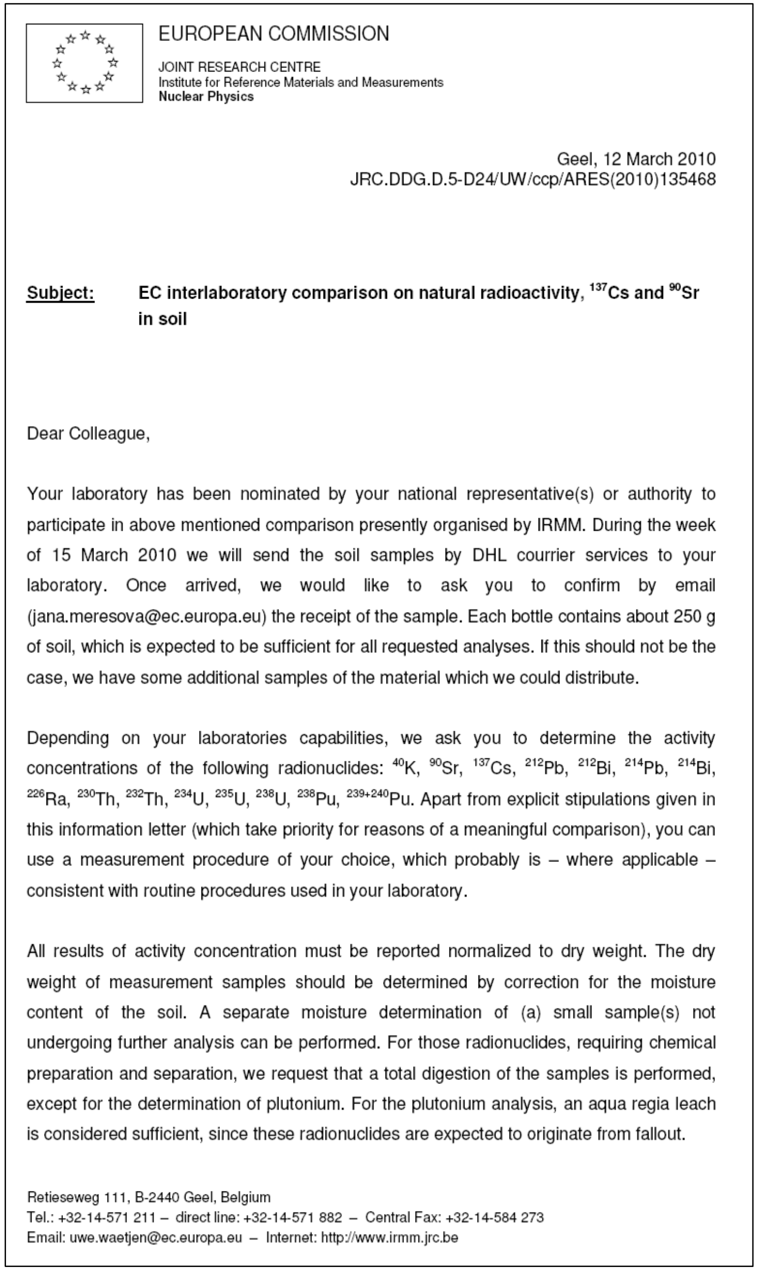
Appendix 2: Information letter



EUROPEAN COMMISSION

JOINT RESEARCH CENTRE
Institute for Reference Materials and Measurements
Nuclear Physics

Geel, 12 March 2010
JRC.DDG.D.5-D24/UW/ccp/ARES(2010)135468

Subject: **EC interlaboratory comparison on natural radioactivity, ^{137}Cs and ^{90}Sr in soil**

Dear Colleague,

Your laboratory has been nominated by your national representative(s) or authority to participate in above mentioned comparison presently organised by IRMM. During the week of 15 March 2010 we will send the soil samples by DHL courrier services to your laboratory. Once arrived, we would like to ask you to confirm by email (jana.meresova@ec.europa.eu) the receipt of the sample. Each bottle contains about 250 g of soil, which is expected to be sufficient for all requested analyses. If this should not be the case, we have some additional samples of the material which we could distribute.

Depending on your laboratories capabilities, we ask you to determine the activity concentrations of the following radionuclides: ^{40}K, ^{90}Sr, ^{137}Cs, ^{212}Pb, ^{212}Bi, ^{214}Pb, ^{214}Bi, ^{226}Ra, ^{230}Th, ^{232}Th, ^{234}U, ^{235}U, ^{238}U, ^{238}Pu, $^{239+240}$Pu. Apart from explicit stipulations given in this information letter (which take priority for reasons of a meaningful comparison), you can use a measurement procedure of your choice, which probably is – where applicable – consistent with routine procedures used in your laboratory.

All results of activity concentration must be reported normalized to dry weight. The dry weight of measurement samples should be determined by correction for the moisture content of the soil. A separate moisture determination of (a) small sample(s) not undergoing further analysis can be performed. For those radionuclides, requiring chemical preparation and separation, we request that a total digestion of the samples is performed, except for the determination of plutonium. For the plutonium analysis, an aqua regia leach is considered sufficient, since these radionuclides are expected to originate from fallout.

Retieseweg 111, B-2440 Geel, Belgium
Tel.: +32-14-571 211 – direct line: +32-14-571 882 – Central Fax: +32-14-584 273
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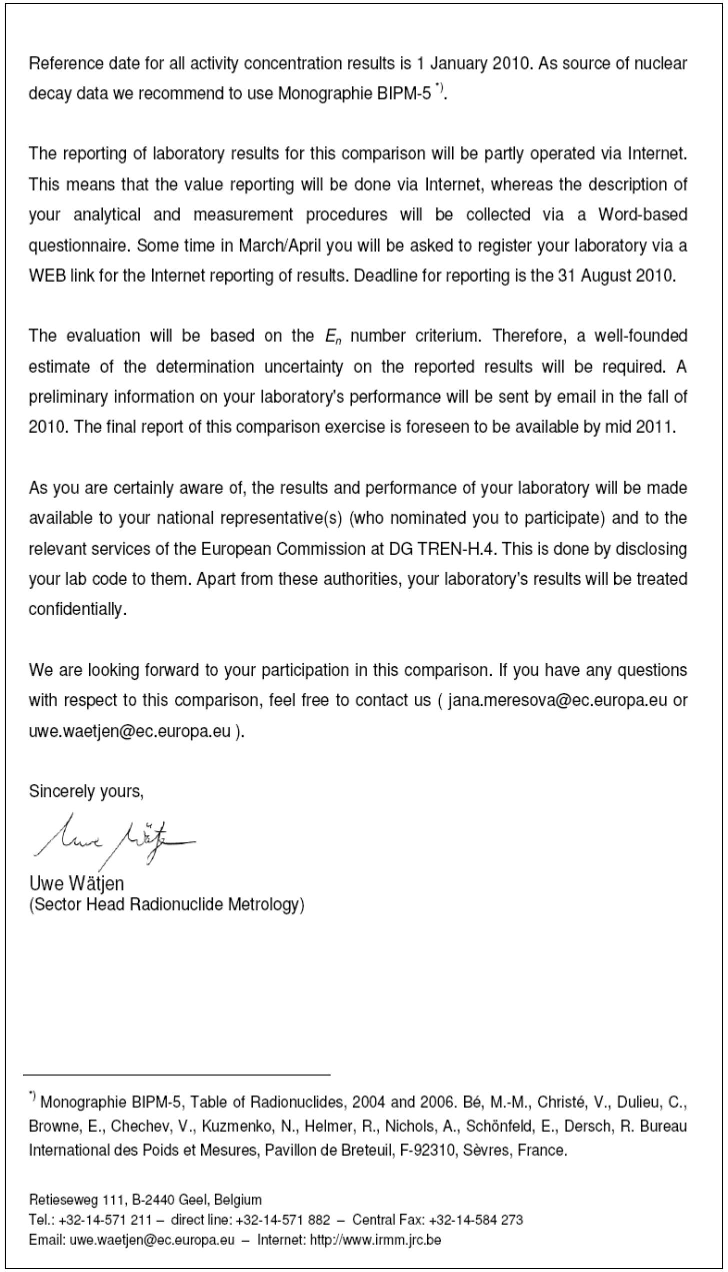

Reference date for all activity concentration results is 1 January 2010. As source of nuclear decay data we recommend to use Monographie BIPM-5 *).

The reporting of laboratory results for this comparison will be partly operated via Internet. This means that the value reporting will be done via Internet, whereas the description of your analytical and measurement procedures will be collected via a Word-based questionnaire. Some time in March/April you will be asked to register your laboratory via a WEB link for the Internet reporting of results. Deadline for reporting is the 31 August 2010.

The evaluation will be based on the E_n number criterium. Therefore, a well-founded estimate of the determination uncertainty on the reported results will be required. A preliminary information on your laboratory's performance will be sent by email in the fall of 2010. The final report of this comparison exercise is foreseen to be available by mid 2011.

As you are certainly aware of, the results and performance of your laboratory will be made available to your national representative(s) (who nominated you to participate) and to the relevant services of the European Commission at DG TREN-H.4. This is done by disclosing your lab code to them. Apart from these authorities, your laboratory's results will be treated confidentially.

We are looking forward to your participation in this comparison. If you have any questions with respect to this comparison, feel free to contact us (jana.meresova@ec.europa.eu or uwe.waetjen@ec.europa.eu).

Sincerely yours,

Uwe Wätjen
(Sector Head Radionuclide Metrology)

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*) Monographie BIPM-5, Table of Radionuclides, 2004 and 2006. Bé, M.-M., Christé, V., Dulieu, C., Browne, E., Chechev, V., Kuzmenko, N., Helmer, R., Nichols, A., Schönfeld, E., Dersch, R. Bureau International des Poids et Mesures, Pavillon de Breteuil, F-92310, Sèvres, France.

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Email: uwe.waetjen@ec.europa.eu – Internet: http://www.irmm.jrc.be

Appendix 3: Registration (e-mail)

Subject: Online registration for EC intercomparison on radioactivity in soil

Dear Colleague,

As you were informed beforehand, the reporting of values will be done via Internet. Therefore, we kindly ask you to register your laboratory via the following web site:

https://irmm.jrc.ec.europa.eu/ilc/ilcRegistration.do?selComparison=460

We have prepared the guidelines for online registration to help you with the procedure.

After the registration and confirmation of all your data you will obtain the **password key** needed for the online reporting of your results. Please be aware that the deadline for registration is **15 June 2010** and the deadline for the reporting of results is **31 August 2010.**

Thank you very much for your participation in the intercomparison and your co-operation in using this online reporting tool. If you have further questions, please contact us. We will be happy to help you.

Kind regards,

Jana Meresova

Appendix 4: Registration guidelines

EUROPEAN COMMISSION JRC

Institute for Reference Materials and Measurements

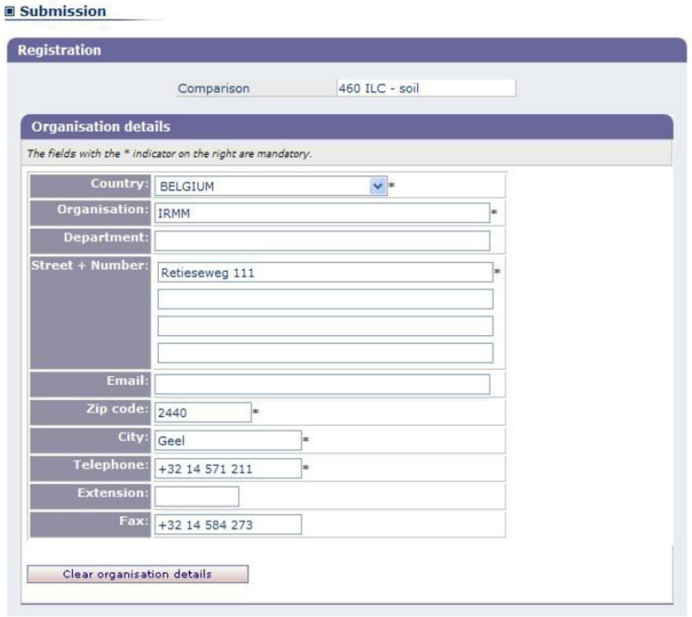
GUIDELINES for Registration for Participation in EC Interlaboratory Comparison on Natural Radioactivity, ^{137}Cs and ^{90}Sr in Soil

We are pleased to inform you that the online registration system is now operational. These guidelines will explain how you can input your organisational and personal details.

The registration is done on the Internet, using the following URL: https://irmm.jrc.ec.europa.eu/ilc/ilcRegistration.do?selComparison=460

The following page will appear:

Figure 1

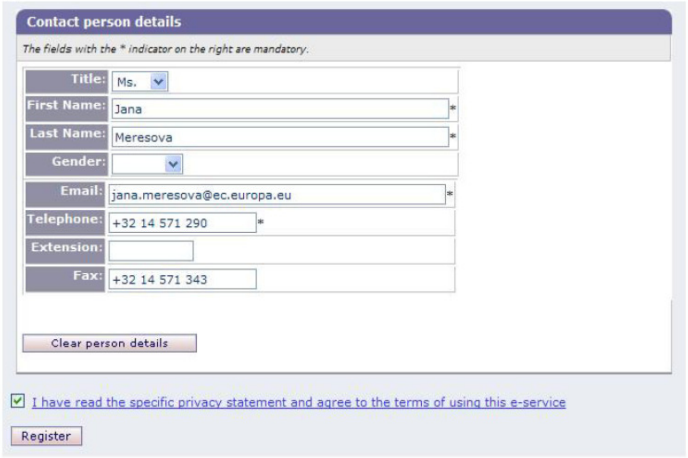


Comparison	460 ILC - soil

*The fields with the * indicator on the right are mandatory.*

Country:	BELGIUM *
Organisation:	IRMM *
Department:	
Street + Number:	Retieseweg 111 *
Email:	
Zip code:	2440 *
City:	Geel *
Telephone:	+32 14 571 211 *
Extension:	
Fax:	+32 14 584 273

Figure 2



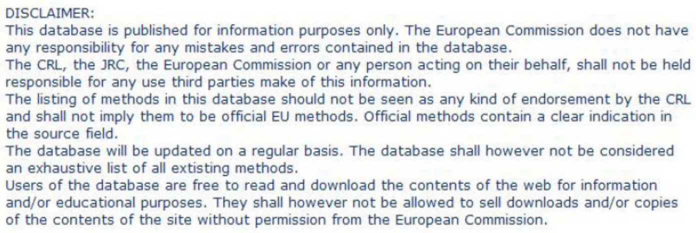
*The fields with the * indicator on the right are mandatory.*

Title:	Ms.
First Name:	Jana *
Last Name:	Meresova *
Gender:	
Email:	jana.meresova@ec.europa.eu *
Telephone:	+32 14 571 290 *
Extension:	
Fax:	+32 14 571 343

☑ I have read the specific privacy statement and agree to the terms of using this e-service

Please fill out all your organisation (Figure 1) and contact details (Figure 2). The fields with the asterisk indicator (*) on the right side are mandatory. Data must be entered in these fields; otherwise, the final registration information will not be submitted. Do not forget to click the checkbox to confirm that you have read the privacy statement and agree with the terms. The full text of the privacy statement is in Figure 3 (DISCLAIMER).

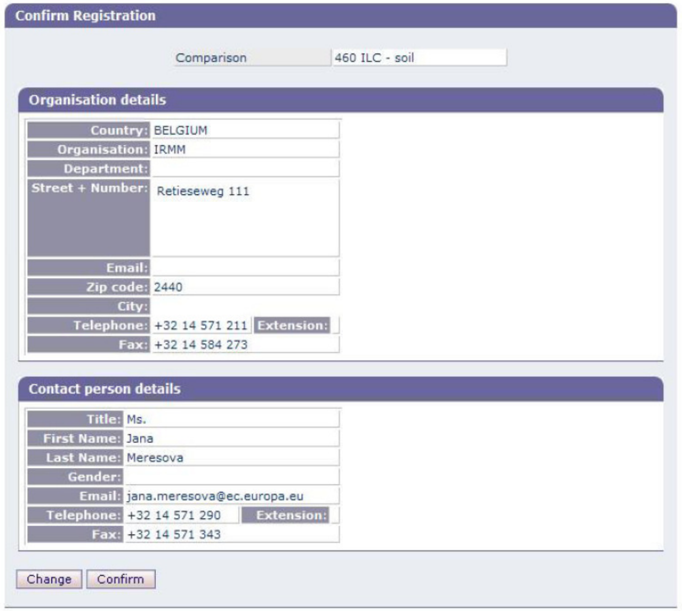
Figure 3



DISCLAIMER:
This database is published for information purposes only. The European Commission does not have any responsibility for any mistakes and errors contained in the database.
The CRL, the JRC, the European Commission or any person acting on their behalf, shall not be held responsible for any use third parties make of this information.
The listing of methods in this database should not be seen as any kind of endorsement by the CRL and shall not imply them to be official EU methods. Official methods contain a clear indication in the source field.
The database will be updated on a regular basis. The database shall however not be considered an exhaustive list of all existing methods.
Users of the database are free to read and download the contents of the web for information and/or educational purposes. They shall however not be allowed to sell downloads and/or copies of the contents of the site without permission from the European Commission.

When all data are entered, press the **REGISTER** button. The screen on the next page (Figure 4) summarises the information. The following options are given at the bottom of the screen: **CHANGE** or **CONFIRM**.

Figure 4


Confirm Registration

Comparison: 460 ILC - soil

Organisation details

Country: BELGIUM
Organisation: IRMM
Department:
Street + Number: Retieseweg 111
Email:
Zip code: 2440
City:
Telephone: +32 14 571 211 Extension:
Fax: +32 14 584 273

Contact person details

Title: Ms.
First Name: Jana
Last Name: Meresova
Gender:
Email: jana.meresova@ec.europa.eu
Telephone: +32 14 571 290 Extension:
Fax: +32 14 571 343

In case any changes need to be made, press the **CHANGE** button. This will take you back to the previous registration page. Apply the amendments and then press the **REGISTER** button again.

If the data are correct, then press the **CONFIRM** button. The final screen concludes that the data have been input into the system (Figure 5). A new window will appear with the printout form.

Figure 5


Submission

Registration confirmation

THE REGISTRATION HAS BEEN SUCCESSFULLY INPUT INTO THE SYSTEM

Please sign the printed registration form and fax it to:

Jana MERESOVA +32 14 584 273

If you didn't see this registration form or you would like to make another print, then click here

This screen is common to all campaigns at IRMM. You may print out your registration form for your own use as many times as you wish. **However, we do not require a copy of a signed printout of the registration form.**

If you notice after the final confirmation that your entries are wrong, you may correct them by sending to us a signed fax or e-mail with the corrected entries.

After your registration, we will send you the **password key** in a separate e-mail. You will need this password key for the online reporting of your results.

REMEMBER:
The DEADLINE for online registration is **15 June 2010**

Appendix 5: Reporting (e-mail)

Subject: Reporting results for EC intercomparison on radioactivity in soil

Dear XXXX,

Thank you very much for your registration. We are pleased to inform you that the online reporting system is now operational. The results reporting is done via the login page using the following URL:

https://irmm.jrc.ec.europa.eu/ilc/ilcReporting.do

To report your results you need a password key which is unique to this intercomparison and your laboratory.

Your password key is: **XXXXX**

Please note that only **SUBMITTED** results will be taken into account, so do not only **SAVE** your results but also select the **SUBMIT** button. Once you have submitted your results, please remember to send us a PDF-file by e-mail or to print the results report form and to fax us a signed copy (Fax: +32 14 584 273). Reference date for all activity concentration results is **1 January 2010**.

The guidelines for online reporting to help you to submit your results are attached. These guidelines explain how you can input them.

We would like to inform you that the **deadline** for reporting your results is **31 August 2010**.

Thank you very much for your participation in the intercomparison and your co-operation in using this online reporting tool. If you have further questions, please contact us. We will be happy to help you.

Kind regards,

Jana Meresova

Appendix 6: Reporting guidelines

JRC
EUROPEAN COMMISSION

irmm
Institute for Reference Materials and Measurements

GUIDELINES for Reporting of Results in EC Interlaboratory Comparison on Natural Radioactivity, ^{137}Cs and ^{90}Sr in Soil

We are pleased to inform you that the online reporting system is now operational. These guidelines will explain how you can input your measurement results.

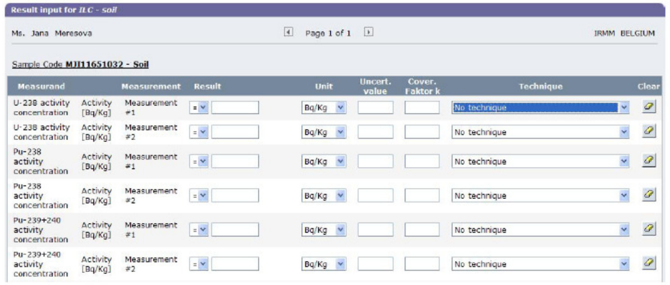
The results reporting is done on the Internet, using the following URL: https://irmm.jrc.ec.europa.eu/ilc/ilcReporting.do

The following page will appear:

Figure 1

Insert the password key, which was sent to you by e-mail after your registration and press the **SUBMIT** button.

Figure 2



Ms. Jana Meresova

IRMM BELGIUM

Sample Code **MJI11651032 - Soil**

Measurand		Measurement	Result	Unit	Uncert. value	Cover. Faktor k	Technique	Clear
U-238 activity concentration	Activity [Bq/Kg]	Measurement #1	=	Bq/Kg			No technique	
U-238 activity concentration	Activity [Bq/Kg]	Measurement #2	=	Bq/Kg			No technique	
Pu-238 activity concentration	Activity [Bq/Kg]	Measurement #1	=	Bq/Kg			No technique	
Pu-238 activity concentration	Activity [Bq/Kg]	Measurement #2	=	Bq/Kg			No technique	
Pu-239+240 activity concentration	Activity [Bq/Kg]	Measurement #1	=	Bq/Kg			No technique	
Pu-239+240 activity concentration	Activity [Bq/Kg]	Measurement #2	=	Bq/Kg			No technique	

When you log in you will access the page for results reporting (Figure 2). There is a possibility to report two measurement results for the duplicate analysis of the specified radionuclide. You may leave blank fields if you do not have results for all radionuclides or if you have performed less than two measurements.

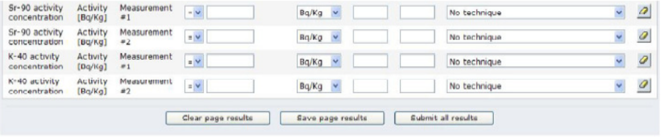
In the column marked "Result", enter the measurement results using the second box from the left. If you need to report an upper limit as a result, you will have to select the "<" from the drop down menu (first box from the left). Please be aware that in this case the uncertainty fields will be disabled.

In the field marked "Uncertainty value", enter the expanded uncertainty following GUM (Guide to the Expression of Uncertainty in Measurement). Do not forget to input the corresponding coverage factor.

In the field "Technique" choose one of the options. The detailed information on your procedures will be requested in a separate Word-based questionnaire.

At this stage you can **CLEAR**, **SAVE** or **SUBMIT PAGE RESULTS** (Figure 3).

Figure 3



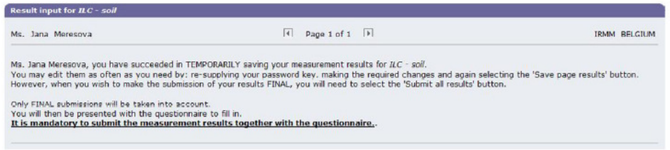
Sr-90 activity concentration	Activity [Bq/Kg]	Measurement #1	=	Bq/Kg			No technique	
Sr-90 activity concentration	Activity [Bq/Kg]	Measurement #2	=	Bq/Kg			No technique	
K-40 activity concentration	Activity [Bq/Kg]	Measurement #1	=	Bq/Kg			No technique	
K-40 activity concentration	Activity [Bq/Kg]	Measurement #2	=	Bq/Kg			No technique	

If any amendments need to be made, press the **CLEAR PAGE RESULTS** button. This will delete all the results on the screen. Make the required changes and SAVE or SUBMIT the data.

To save the results press the **SAVE PAGE RESULTS** button. This will SAVE the data entered with the possibility to edit them before you issue the final SUBMIT.

There is no EXIT button. If you want to leave the Result Reporting Page choose x at the right top corner of the screen. **Be aware that you have to SAVE or SUBMIT entered data otherwise they will be lost.** The following message (Figure 4) will appear informing you that your results have been saved.

Figure 4



Ms. Jana Meresova

IRMM BELGIUM

Ms. Jana Meresova, you have succeeded in TEMPORARILY saving your measurement results for *ILC - soil*.
You may edit them as often as you need by: re-supplying your password key, making the required changes and again selecting the 'Save page results' button.
However, when you wish to make the submission of your results FINAL, you will need to select the 'Submit all results' button.

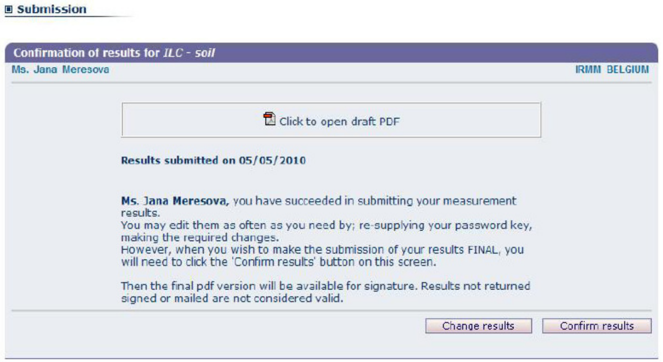
Only FINAL submissions will be taken into account.
You will then be presented with the questionnaire to fill in.
It is mandatory to submit the measurement results together with the questionnaire..

To reconnect to our system use the same URL link https://irmm.jrc.ec.europa.eu/ilc/ilcReporting.do and re-supply the password key. The results form will appear with the data that has already been entered. Make the required changes and select either the **SAVE PAGE RESULTS** button or the **SUBMIT ALL RESULTS** button.

Remember to submit the results before the deadline date, as <u>ONLY submitted results</u> will be accepted.

After the submission of all results, the following message (Figure 5) will appear informing you that your results have been submitted. You can see the draft PDF file with your submitted results. As long as you do not **CONFIRM** the report submission you can always come back to the entered data and change or correct them.

Figure 5

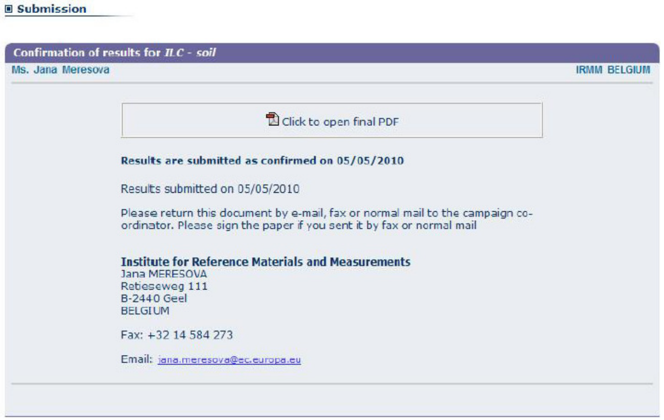


Results submitted on 05/05/2010

Ms. Jana Meresova, you have succeeded in submitting your measurement results.
You may edit them as often as you need by; re-supplying your password key, making the required changes.
However, when you wish to make the submission of your results FINAL, you will need to click the 'Confirm results' button on this screen.

Then the final pdf version will be available for signature. Results not returned signed or mailed are not considered valid.

Figure 6



Results are submitted as confirmed on 05/05/2010

Results submitted on 05/05/2010

Please return this document by e-mail, fax or normal mail to the campaign co-ordinator. Please sign the paper if you sent it by fax or normal mail

Institute for Reference Materials and Measurements
Jana MERESOVA
Retieseweg 111
B-2440 Geel
BELGIUM

Fax: +32 14 584 273

Email: jana.meresova@ec.europa.eu

After the final confirmation of your results you are not allowed to change the results. The following message (Figure 6) will appear with your organizational and personal data. Also the final PDF file with your submitted results can be downloaded. Please send us this PDF file by e-mail or print the Result Report Form and fax to us a signed copy (Fax: +32-14-571 343).

The description of your analytical and measurement procedures will be collected via a Word-based questionnaire. This will be sent to you in a separate e-mail.

REMEMBER:
The DEADLINE for final submission of the results is **<u>31 August 2010</u>**

Appendix 7: Communication on a preliminary results

EUROPEAN COMMISSION
JOINT RESEARCH CENTRE
Institute for Reference Materials and Measurements
Nuclear Physics

Geel, 9 November 2010
JRC.DG.D.5/UW/jm/ARES(2010)xxxxxx

Subject: **Preliminary results of the EC interlaboratory comparison on natural radioactivity, ^{137}Cs and ^{90}Sr in soil**

Dear Dr Xxxxxx,

First of all, thank you for your participation in the EC interlaboratory comparison on radioactivity in soil. You were asked to determine the activity concentrations of the following radionuclides: ^{40}K, ^{90}Sr, ^{137}Cs, ^{212}Pb, ^{212}Bi, ^{214}Pb, ^{214}Bi, ^{226}Ra, ^{230}Th, ^{232}Th, ^{234}U, ^{235}U, ^{238}U, ^{238}Pu, $^{239+240}$Pu depending on your laboratory's capabilities.

At the moment we are working on the evaluation of your results. However, for your early information we are sending you a preliminary evaluation of the results in this comparison. Since the anonymity is a requirement in this programme of measurement comparisons, the identity of the laboratories is not shown in the compilation of the results. Each laboratory was assigned a code number. The code number for your laboratory is **XX**.

In Figures 1 to 15 the reported activity concentrations (normalised to dry weight) with their corresponding expanded uncertainty (k = 2) are plotted in ascending order. The solid red line (where available) indicates the preliminary reference activity concentration and its expanded uncertainty (k = 2) is plotted in dashed lines. Laboratory codes are indicated with the results.

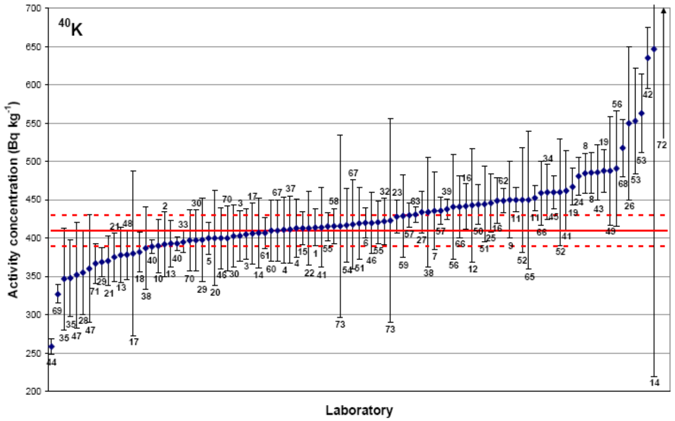


Fig. 1: Laboratory results for ^{40}K activity concentration.

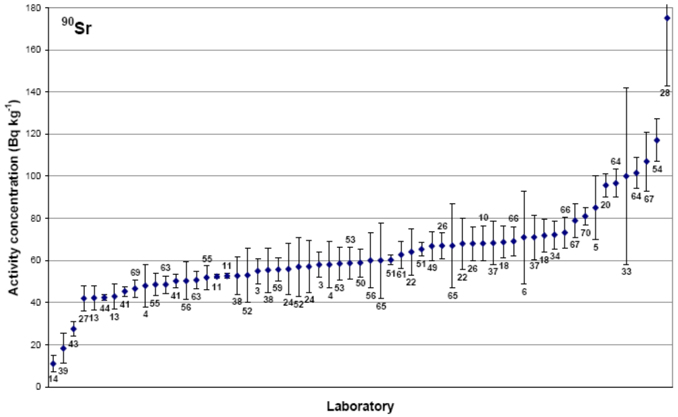


Fig. 2: Laboratory results for ^{90}Sr activity concentration.

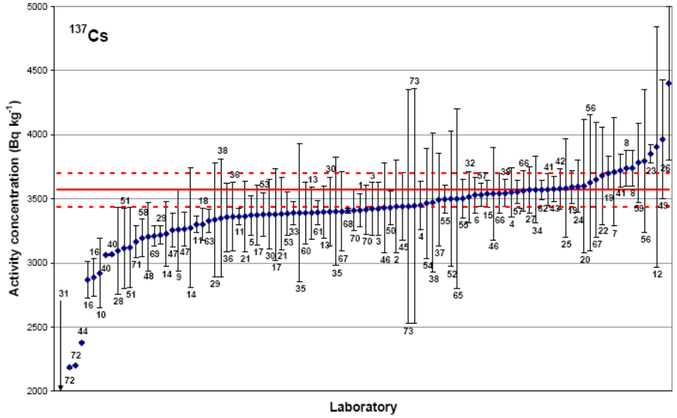


Fig. 3: Laboratory results for ^{137}Cs activity concentration.

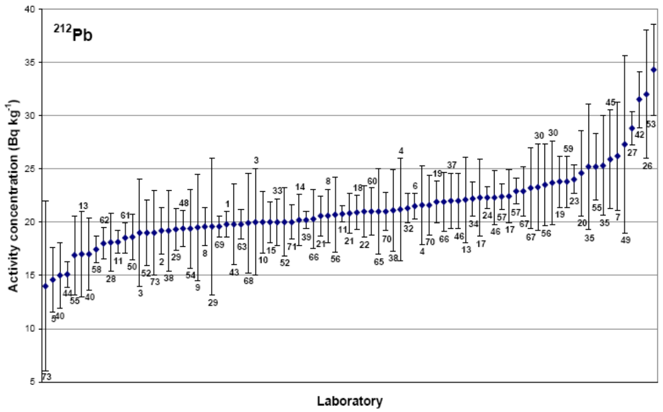


Fig. 4: Laboratory results for ^{212}Pb activity concentration.

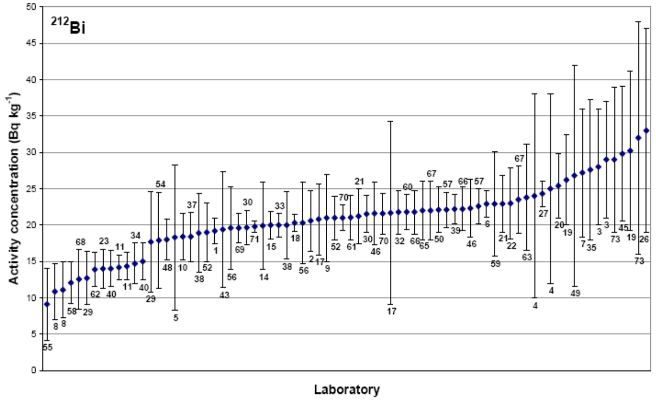


Fig. 5: Laboratory results for ^{212}Bi activity concentration.

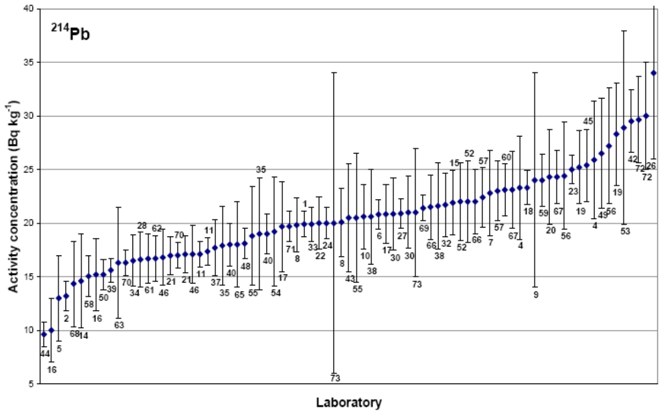


Fig. 6: Laboratory results for ^{214}Pb activity concentration.

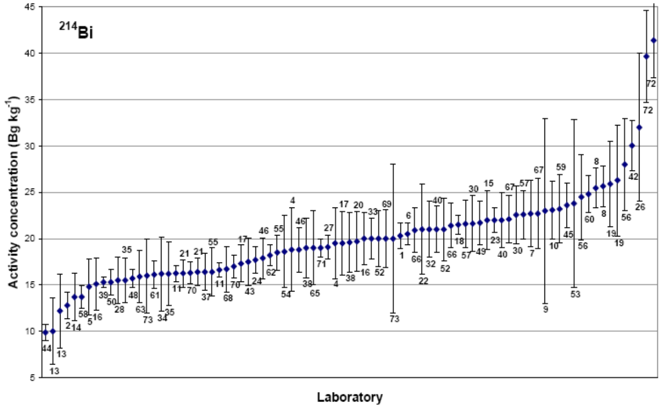


Fig. 7: Laboratory results for ^{214}Bi activity concentration.

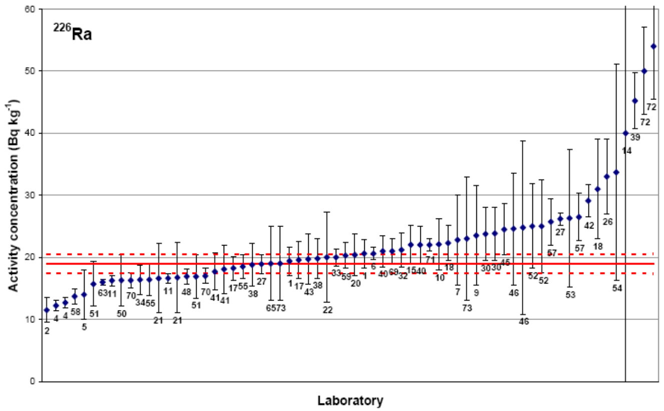


Fig. 8: Laboratory results for ^{226}Ra activity concentration.

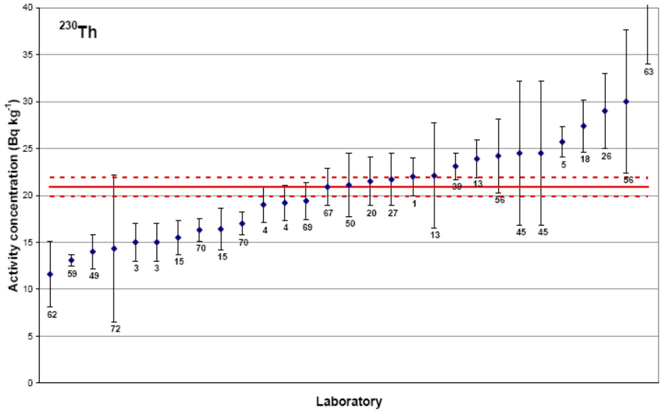


Fig. 9: Laboratory results for ^{230}Th activity concentration.

Retieseweg 111, B-2440 Geel, Belgium
Tel.: +32-14-571 211 – direct line: +32-14-571 882 – Central Fax: +32-14-584 273
Email: uwe.waetjen@ec.europa.eu, jana.meresova@ec.europa.eu – Internet: http://irmm.jrc.ec.europa.eu

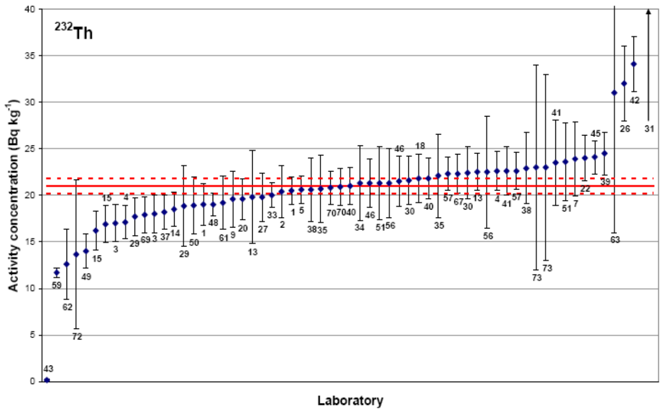


Fig. 10: Laboratory results for ^{232}Th activity concentration.

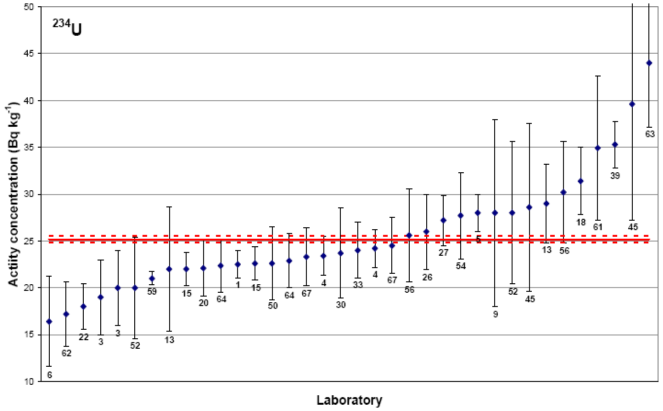


Fig. 11: Laboratory results for ^{234}U activity concentration.

Retieseweg 111, B-2440 Geel, Belgium
Tel.: +32-14-571 211 – direct line: +32-14-571 882 – Central Fax: +32-14-584 273
Email: uwe.waetjen@ec.europa.eu, jana.meresova@ec.europa.eu – Internet: http://irmm.jrc.ec.europa.eu

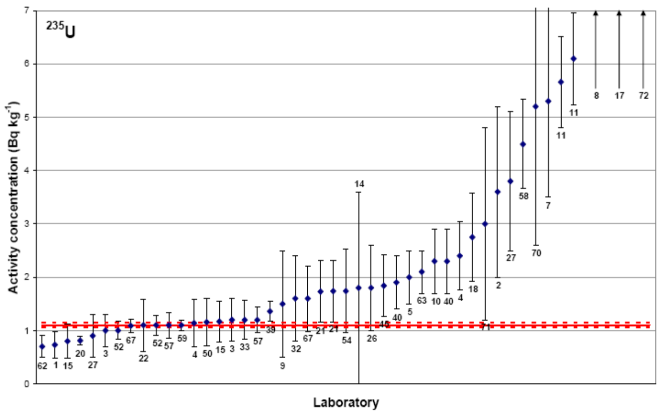


Fig. 12: Laboratory results for ^{235}U activity concentration.

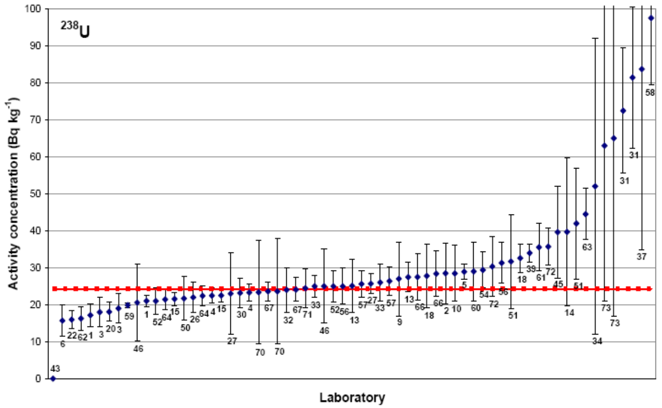


Fig. 13: Laboratory results for ^{238}U activity concentration.

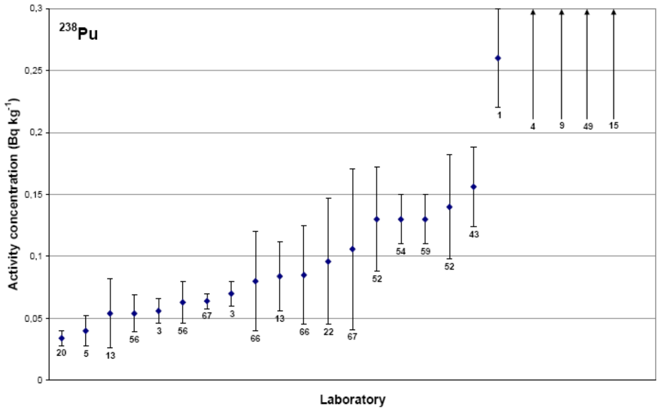


Fig. 14: Laboratory results for ^{238}Pu activity concentration.

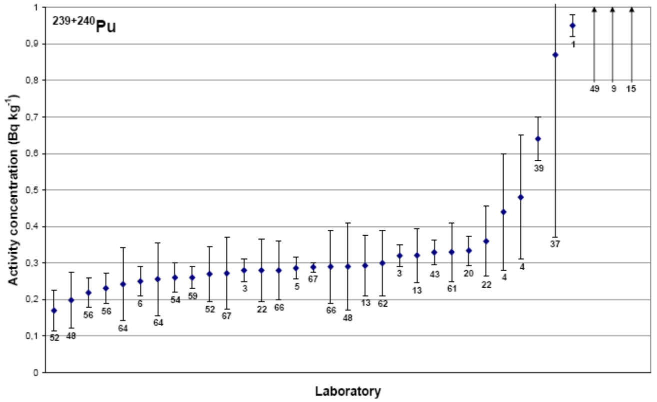


Fig. 15: Laboratory results for $^{239+240}$Pu activity concentration.

The final report of this comparison exercise is foreseen to be available by mid 2011. If you have any further questions with respect to this comparison, please feel free to contact us.

Sincerely yours,
Jana Meresova and Uwe Wätjen

Appendix 8: List of participating laboratories

ALBANIA

Dr Elida Bylyku
Centre of Applied Nuclear Physics
Qesarake street
1000 Tirana

AUSTRALIA

Ms Atun Zawadzki (IAEA)
Australian Nuclear Science and Technology Organisation
New Illawarra Rd Lucas Heights NSW 2232
Mailing address: Locked Bag 2001 Kirrawee DC NSW 2232

AUSTRIA

Mag Claudia Landstetter
Austrian Agency for Health and Food Safety
CC Radiation Protection and Radiochemistry
Spargelfeldstr. 191
1220 Vienna

Mr. Johannes Klimstein
AGES GmbH
CC RARA
Derfflingerstrasse 2
4020 Linz

BANGLADESH

Dr Satyajit Ghose (IAEA)
Bangladesh Atomic Energy Commission
Nuclear Safety & Radiation Con
4 No Kazi Nazrul Islam Avenue
1000 Ramna, Dhaka

BELGIUM

Dr Michel Bruggeman
SCK-CEN
LRM/RNM
Boeretang 200
2400 Mol

Mr Benoit Deconninck
IRE
SEM
Rue de l'Espérance 1
6220 Fleurus

Dr Mikael Hult (IAEA)
IRMM
Nuclear Physics Unit
Retieseweg 111
2440 Geel

BOSNIA - HERZEGOVINA

Ms Delveta Deljkic
Institute for Public Health of Federation of Bosnia and Herzegovina
Radiation Protection Centre
Marsala Tita 9
71000 Sarajevo

BRAZIL

Prof Brigitte Pecequilo (IAEA)
IPEN-Instituto de Pesquisas Energéticas e Nucleares
Radiations Metrology
Av. Prof. Lineu Prestes, 2242
05508-000 São Paulo

BULGARIA

Mr Victor Badulin
National Center of Radiobiology and Radiation Protection
Public Exposure Monitoring Lab
3 Sv. Georgi Sofiyski St.
1606 Sofia

Mr Mihail Shishenkov
Executive Environment Agency
Radioactivity Measurements Laboratory
136 Tzar Boris III Blvd.
1618 Sofia

CHILE

Mr Osvaldo Piñones (IAEA)
Chilean Commission of Nuclear Energy
Environmental Radioactivity
Amunategui 95
Casilla 188D Santiago

CROATIA

Dr Zdenko Franic
Institute for Medical Research and Occupational Health
Radiation Protection Unit
Ksaverska cesta 2
10000 Zagreb

Dr Zeljko Grahek
Rudjer Boskovic Institute
Center for marine and environment
Bijenicka 54
10000 Zagreb

CUBA

Mr Jorge Carrazana González (IAEA)
Centro de Proteccion e Higiene de las Radiaciones (CPHR)
Lab. de Radiologica Ambiental
Calle 20 No. 4113 e/ 41 y 47 Playa
10600 La Habana

CYPRUS

Mrs Anastasia Caballero
State General Laboratory of Cyprus
Radioactivity Laboratory
Kimonos 44
1451 Nicosia

CZECH REPUBLIC

Dr Petr Rulík
National Radiation Protection Institute
Monitoring
Bartoskova 28
14000 Prague

DENMARK

Ms Katrine Berg
National Institute of Radiation Protection
Knapholm 7
2730 Herlev

Dr Sven Nielsen
Riso National Laboratory for Sustainable Energy, Technical University of Denmark
Radiation Research
Frederiksborgvej 399
4000 Roskilde

ESTONIA

Ms Kadri Isakar
University of Tartu, Institute of Physics
Laboratory of Environmental Physics
Riia 142
51014 Tartu

Ms Eia Jakobson
Environmental Board
Radiation Safety Department
Kopli 76
10416 Tallinn

FRANCE

Dr Cédric Aubert
IRSN
DEI/STEME
31 rue de l'ecluse
78116 Le Vesinet

Dr Stéphane Scapolan
French Atomic Energy and Alternative Energies Commission (CEA)
DSM/SAC/UPSE/SPR
Centre de Saclay
Batiment 388
91191 Gif-Sur-Yvette Cedex

GERMANY

Dr Matthias Köhler (IAEA)
VKTA
Analytical Department
Bautzener Landstrasse 400
1314 Dresden

Dr David Tait
Max Rubner-Institut
Bundesforschungsinstitut für Ernährung und Lebensmittel
Institut für Sicherheit und Qualität bei Milch und Fisch (Standort Kiel)
LeitstelleUmweltradioaktivität
Hermann-Weigmann-Str. 1
24103 Kiel

Mr Christoph Wilhelm (IAEA)
Karlsruhe Institute of Technology
Central Safety Management
Hermann-von-Helmholtz-Platz 1
76344 Eggenstein-Leo

GREECE

Dr Marios Anagnostakis
National Technical University of Athens
Nuclear Engineering Department
Heroon Polytechniou 9
Zografou Campus
15780 Athens

Dr Nikolaos Evagnelliou (IAEA)
National Centre for Scientific Research
INT-R / ERL
Patriarchou Grigoriou & Neapoleos
15310 Aghia Paraskevi

Dr Stavros Seferlis
Greek Atomic Energy Commission
Environmental Radioactivity
Patriarchou Grigoriou & Neapoleos
15310 Aghia Paraskevi

HUNGARY

Mr Sandor Tarjan
Central Agricultural Office
Food and Feed Safety Dir.
Fogoly str 13-15
1182 Budapest

Ms Agota Ugron
National Institute for Radiobiology and Radiohygiene
Departement of Radiohygiene
Anna u. 5
1182 Budapest

INDIA

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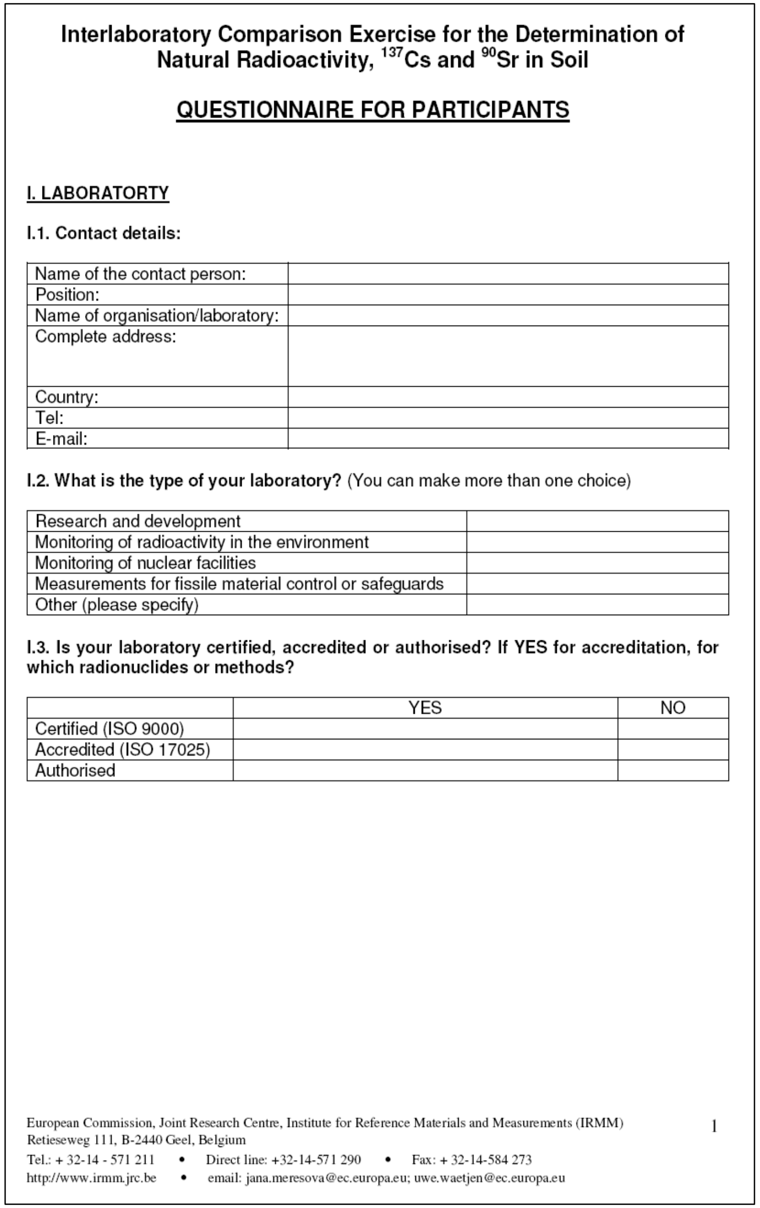
Appendix 9: Questionnaire



Interlaboratory Comparison Exercise for the Determination of Natural Radioactivity, ^{137}Cs and ^{90}Sr in Soil

QUESTIONNAIRE FOR PARTICIPANTS

I. LABORATORTY

I.1. Contact details:

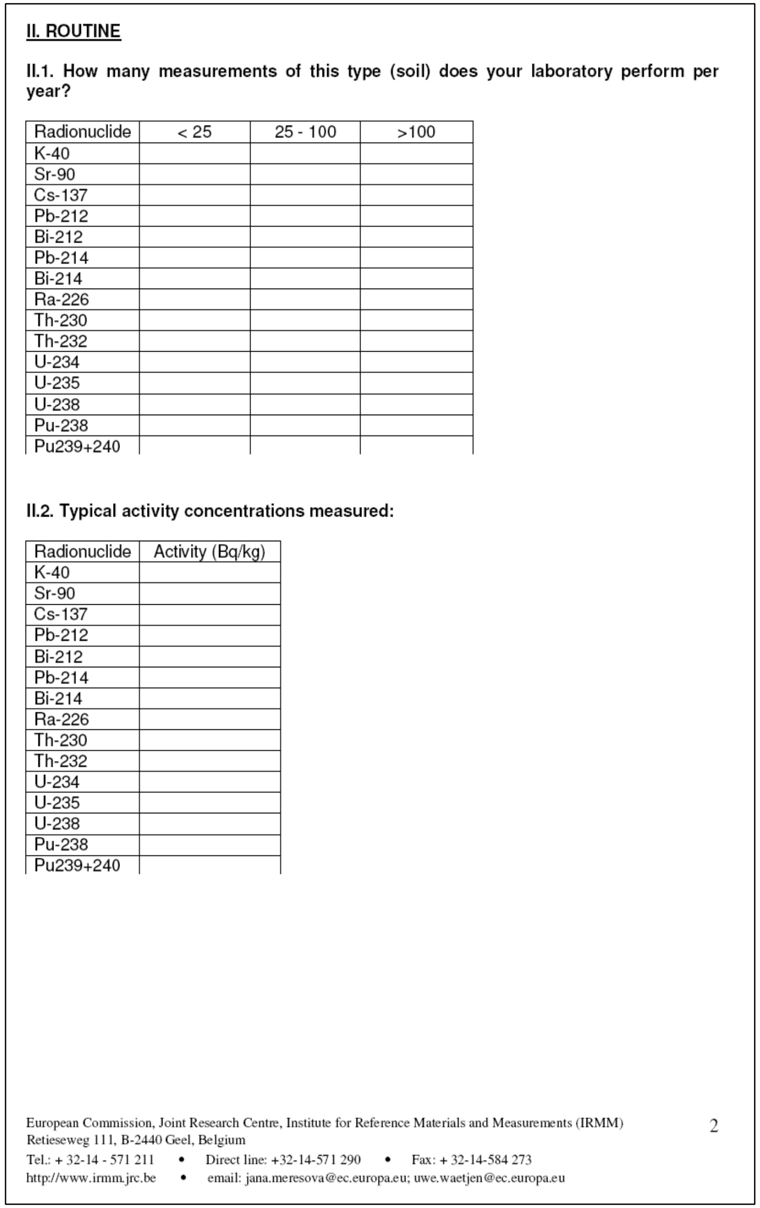
Name of the contact person:	
Position:	
Name of organisation/laboratory:	
Complete address:	
Country:	
Tel:	
E-mail:	

I.2. What is the type of your laboratory? (You can make more than one choice)

Research and development	
Monitoring of radioactivity in the environment	
Monitoring of nuclear facilities	
Measurements for fissile material control or safeguards	
Other (please specify)	

I.3. Is your laboratory certified, accredited or authorised? If YES for accreditation, for which radionuclides or methods?

	YES	NO
Certified (ISO 9000)		
Accredited (ISO 17025)		
Authorised		



II. ROUTINE

II.1. How many measurements of this type (soil) does your laboratory perform per year?

Radionuclide	< 25	25 - 100	>100
K-40			
Sr-90			
Cs-137			
Pb-212			
Bi-212			
Pb-214			
Bi-214			
Ra-226			
Th-230			
Th-232			
U-234			
U-235			
U-238			
Pu-238			
Pu239+240			

II.2. Typical activity concentrations measured:

Radionuclide	Activity (Bq/kg)
K-40	
Sr-90	
Cs-137	
Pb-212	
Bi-212	
Pb-214	
Bi-214	
Ra-226	
Th-230	
Th-232	
U-234	
U-235	
U-238	
Pu-238	
Pu239+240	

III. MOISTURE DETERMINATION

III.1. Which method have you used for the moisture content determination?

	YES	NO
Karl-Fischer titration		
Drying at 102 °C		
Other		

If other, please specify:

III.2. What was the moisture content (%) of the sample?

III.3. What was the uncertainty of the moisture content of the sample?

III.4. What was the applied correction factor for dry-mass?

III.5. Mass of the sample used for determination of moisture content?

IV. SAMPLE TREATMENT

Please group radionuclides according to the used methods for all questions related to sample treatment.

IV.1. Describe methods of sample preparation (and specify mass of sample, number of independent samples, etc.).

IV.2. Describe methods of preconcentration and chemical separation (chemicals and procedures used, chemical recovery obtained, etc.).

IV.3. Describe methods of source preparation for radionuclide determination (geometry, type of LSC cocktail, precipitation method, deposition, etc.).

V. MEASUREMENT

Please group radionuclides according to the used methods for all questions related to measurement.

V.1. Describe the measurement methods.

V.2. What type of detector was used? (For gamma-spectrometers, e.g., specify also its nominal relative efficiency (%).)

V.3. What type of electronics and data acquisition was used (analog/digital signal processing, TR-LSC, etc.)?

V.4. Describe the efficiency calibration of the detector system(s).

V.5. What was the acquisition time per sample and in how many measurement cycles was it counted?

V.6. Provide information on blank and background measurement.

V.7. Specify software for data evaluation, peak area determination, source of nuclear data and equations used.

VI. UNCERTAINTY BUDGET

Here are possible contributions, which should be accounted for in the estimation of the combined uncertainty, all as the standard uncertainty (corresponding to one standard deviation σ) in the form of propagated uncertainty contribution to the final result. Please provide for all used methods.

VI.1. Please state for one measurement result of _____ (radionuclide) and indicate:

Measurement number:	
Number of measurements performed:	
Relative uncertainty due to moisture content (%):	
Relative uncertainty due to sample preparation, separation etc. (%):	
Relative uncertainty on the tracer activity (%):	
Relative counting uncertainty (%):	
Relative uncertainty in efficiency (%):	
Relative uncertainty due to the activity of efficiency standards (%):	
Relative uncertainty due to blank and background measurement (%):	
Possibly other uncertainty contributions (%):	
Relative combined standard uncertainty u^*_C (%):	
Combined standard uncertainty u_C (Bq/kg):	
Expanded uncertainty U (mBq/kg, k=2):	

VI.2. Please state for one measurement result of _____ (radionuclide) and indicate:

Measurement number:	
Number of measurements performed:	
Relative uncertainty due to moisture content (%):	
Relative uncertainty due to sample preparation, separation etc. (%):	
Relative uncertainty on the tracer activity (%):	
Relative counting uncertainty (%):	
Relative uncertainty in efficiency (%):	
Relative uncertainty due to the activity of efficiency standards (%):	
Relative uncertainty due to blank and background measurement (%):	
Possibly other uncertainty contributions (%):	
Relative combined standard uncertainty u^*_C (%):	
Combined standard uncertainty u_C (Bq/kg):	
Expanded uncertainty U (mBq/kg, k=2):	

Etc.

VII. ADDITIONAL INFORMATION

VII.1. Were the inter-comparison samples treated according to the same analytical procedures as routinely used in your laboratory for the same type of samples?

Radionuclide	YES	NO
K-40		
Sr-90		
Cs-137		
Pb-212		
Bi-212		
Pb-214		
Bi-214		
Ra-226		
Th-230		
Th-232		
U-234		
U-235		
U-238		
Pu-238		
Pu239+240		

VII.2. Did you determine any additional radionuclide(s)?

YES NO

If YES, give the name of the nuclide(s) measured and its measured value(s) (Bq/kg) together with the expanded uncertainty(ies) U (Bq/kg, k=2):

Radionuclide	Activity (Bq/kg)

VII.3. Difficulties encountered:

VII.4. Further comments on this interlaboratory comparison exercise:

VII.5. Questionnaire completed by:

Name:
Position:
Date:

VII.6. Further comments:

Appendix 10: Results, methods and scores of laboratories

In Tables 1 to 15 are presented the values reported by participants and in the case of double result the calculated arithmetic means A_{lab}. For the results marked with an asterisk (*) was the expanded uncertainty U_{lab} calculated with the first approach - based on the information given in the uncertainty budget. The information on used method, as reported by participants, is included. Moreover, the information on whether the result was indicated by the Grubbs' test (□ = 1 %) as an outlier is presented. Also, the values of relative deviation and E_n number are present in the last two columns. The red colour indicates results deviating more than 20 % from the reference values or E_n in-compatible results (the action signal). E_n numbers in black colour indicate the warning signal.

Table 1. Activity concentrations of ^{40}K with expanded uncertainties (k = 2), used methods and performance scores of individual laboratories.

Lab. code	Reported values		Arithmetic means		Used method	Outlier	$D_{\%}$ (%)	E_n
	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)				
1	414 ± 24	6	-	-	Direct gamma-spec.	-	1.0	0.1
2	392 ± 42	11	-	-	Direct gamma-spec.	-	-4.4	-0.4
3	403 ± 33	8	404 ± 33	8	Direct gamma-spec.	-	-1.5	-0.2
	405 ± 33	8						
4	411 ± 43	10	412 ± 41	10	Direct gamma-spec.	-	0.5	0.0
	413 ± 38	9						
5	400 ± 21	5	-	-	Direct gamma-spec.	-	-2.4	-0.3
6	420 ± 20	5	-	-	Direct gamma-spec.	-	2.4	0.3
7	435.6 ± 50.5	12	-	-	Direct gamma-spec.	-	6.2	0.5
8	484.72 ± 26.45	5	485 ± 27	5	Direct gamma-spec.	-	18.3	2.2
	485.62 ± 26.59	5						
9	450 ± 50	11	-	-	Direct gamma-spec.	-	9.8	0.7
10	390 ± 35	9	-	-	Direct gamma-spec.	-	-4.9	-0.5
11	450 ± 16	4	451 ± 16	4	Direct gamma-spec.	-	10.1	1.6
	452.5 ± 16.2	4						
12	443 ± 74	17	-	-	Direct gamma-spec.	-	8.0	0.4
13	378 ± 36	10	386 ± 33	9	Direct gamma-spec.	-	-0.6	-0.6
	393 ± 30	8						
14	406.9 ± 46	11	527 ± 13*	26	Direct gamma-spec.	-	28.5	0.8
	646.8 ± 428	66						
15	413 ± 21	5	-	-	Direct gamma-spec.	-	0.7	0.1
16	442 ± 29.7	7	446 ± 30	7	Direct gamma-spec.	-	8.7	1.0
	449 ± 29.4	7						
17	380 ± 108	28	393 ± 75	19	Direct gamma-spec.	-	-4.1	-0.2
	406 ± 40	10						
18	382 ± 26	7	-	-	Direct gamma-spec.	-	-6.8	-0.8
19	467 ± 24	5	478 ± 26	5	Direct gamma-spec.	-	16.5	2.0
	488 ± 28	6						
20	400 ± 62	16	-	-	Direct gamma-spec.	-	-2.4	-0.2
21	370.3 ± 32.36	9	373 ± 32*	8	Direct gamma-spec.	-	-9.1	-1.0
	375.4 ± 31.9	8						
22	413 ± 48.48	12	-	-	Direct gamma-spec.	-	0.7	0.1
23	428 ± 21.33	5	-	-	Direct gamma-spec.	-	4.4	0.6
24	481 ± 25	5	-	-	Direct gamma-spec.	-	17.3	2.2
25	445.99 ± 38.24	9	-	-	Direct gamma-spec.	-	8.8	0.8
26	550 ± 100	18	-	-	Direct gamma-spec.	-	34.1	1.4

27	434 ± 27	6	-	-	Direct gamma-spec.	-	**5.9**	**0.7**
28	355 ± 54	15	-	-	Direct gamma-spec.	-	**-13.4**	**-1.0**
29	369 ± 19.39	5	384 ± 36	9	Direct gamma-spec.	-	**-6.5**	**-0.6**
	398 ± 54.55	14						
30	397.38 ± 39.72	10	400 ± 40	10	Direct gamma-spec.	-	**-2.4**	**-0.2**
	402.76 ± 40.28	10						
32	422 ± 30	7	-	-	Direct gamma-spec.	-	**2.9**	**0.3**
33	395 ± 13.8	3	-	-	Direct gamma-spec.	-	**-3.7**	**-0.6**
34	460 ± 36	8	-	-	Direct gamma-spec.	-	**12.2**	1.2
35	347 ± 66.4	19	348 ± 58	17	Direct gamma-spec.	-	**-15.2**	-1.0
	348 ± 49.6	14						
37	411.4 ± 43.6	11	-	-	Direct gamma-spec.	-	**0.3**	**0.0**
38	387 ± 54	14	411 ± 63	15	Direct gamma-spec.	-	**0.1**	**0.0**
	434 ± 72	17						
39	438 ± 13	3	-	-	Direct gamma-spec.	-	**6.8**	1.1
40	389 ± 9	2	391 ± 9	2	Direct gamma-spec.	-	**-4.6**	**-0.8**
	393 ± 9	2						
41	414 ± 52	13	438 ± 49*	11	Direct gamma-spec.	-	**6.8**	**0.5**
	462 ± 52	11						
42	635.41 ± 40.1	6	-	-	Direct gamma-spec.	yes	**55.0**	**5.0**
43	486 ± 36	7	-	-	Direct gamma-spec.	-	**18.5**	**1.8**
44	258.4 ± 10.59	4	-	-	Direct gamma-spec.	yes	**-37.0**	**-6.5**
45	460 ± 21.16	5	-	-	Direct gamma-spec.	-	**12.2**	**1.7**
46	400 ± 40	10	410 ± 40	10	Direct gamma-spec.	-	**0.0**	**0.0**
	420 ± 40	10						
47	352.21 ± 69.04	20	356 ± 50*	14	Direct gamma-spec.	-	**-13.1**	**-1.0**
	360.33 ± 70.63	20						
48	378.4 ± 32.3	9	-	-	Direct gamma-spec.	-	**-7.7**	**-0.8**
49	488 ± 70.8	15	-	-	Direct gamma-spec.	-	**19.0**	1.1
50	444 ± 26	6	-	-	Direct gamma-spec.	-	**8.3**	1.0
51	419.4 ± 46.8	11	432 ± 48	11	Direct gamma-spec.	-	**5.3**	**0.4**
	444.4 ± 49.2	11						
52	450 ± 68	15	455 ± 69	15	Direct gamma-spec.	-	**11.0**	**0.6**
	460 ± 69	15						
53	553 ± 69	12	558 ± 60	11	Direct gamma-spec.	-	**36.1**	**2.3**
	563 ± 51	9						
54	417 ± 48	12	-	-	Direct gamma-spec.	-	**1.7**	**0.1**
55	415.4 ± 18.2	4	418 ± 23	5	Direct gamma-spec.	-	**2.0**	**0.3**
	420.8 ± 27.3	6						
56	441 ± 68	15	466 ± 72	15	Direct gamma-spec.	-	**13.7**	**0.8**
	491 ± 75	15						
57	430 ± 16	4	433 ± 17	4	Direct gamma-spec.	-	**5.6**	**0.9**
	436 ± 18	4						
58	415.773 ± 22.452	5	-	-	Direct gamma-spec.	-	**1.4**	**0.2**
59	429 ± 54	13	-	-	Direct gamma-spec.	-	**4.6**	**0.3**
60	410 ± 40	10	-	-	Direct gamma-spec.	-	**0.0**	**0.0**
61	407 ± 20	5	-	-	Direct gamma-spec.	-	**-0.7**	**-0.1**
62	449 ± 16.1	4	-	-	Direct gamma-spec.	-	**9.5**	1.5
63	431 ± 10	2	-	-	Direct gamma-spec.	-	**5.1**	**0.9**
65	450 ± 90	20	-	-	Direct gamma-spec.	-	**9.8**	**0.4**
66	441 ± 40	9	450 ± 41	9	Direct gamma-spec.	-	**9.8**	**0.9**
	459 ± 42	9						

67	410 ± 40	10	414 ± 49	12	Direct gamma-spec.	-	1.0	0.1
	418 ± 58	14						
68	517.91 ± 37.08	7	-	-	Direct gamma-spec.	-	26.3	2.5
69	327 ± 12	4	-	-	Direct gamma-spec.	-	-20.2	-3.5
70	397 ± 40	10	399 ± 41	10	Direct gamma-spec.	-	-2.8	-0.3
	400 ± 42	11						
71	367 ± 26	7	-	-	Direct gamma-spec.	-	-10.5	-1.3
72	1235 ± 9.3	1	1273 ± 9	1	Direct gamma-spec.	yes	210.5	38.6
	1311 ± 8	1						
73	416 ± 119	29	420 ± 103*	24	Direct gamma-spec.	-	2.3	0.1
	423 ± 133	31						

Table 2. Activity concentrations of ^{90}Sr with expanded uncertainties (k = 2), used methods and performance scores of individual laboratories.

Lab. code	Reported values		Arithmetic means		Used method	Outlier	$D_{\%}$ (%)	E_n
	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)				
3	55 ± 6	11	56.5 ± 5.7*	10	Liquid-scint. counting	-	-24.2	-1.6
	58 ± 6	10						
4	48 ± 10	21	53.0 ± 10.5	20	Gas flow prop. counting	-	-28.9	-1.5
	58 ± 11	19						
5	85 ± 15	18	-	-	Prop. counting after chemical sep. of ^{90}Y	-	14.1	0.6
6	71 ± 22	31	-	-	Proportional counter	-	-4.7	-0.1
10	68.1 ± 8.4	12	-	-	Total beta counting (^{90}Y) with chemical separation	-	-8.6	-0.5
11	52.4 ± 1	2	52.5 ± 1.0	2	Radiochemical sep. and prop. counter	-	-29.5	-2.2
	52.6 ± 1.1	2						
13	42.2 ± 5.8	14	42.6 ± 5.8	14	Liquid-scint. counting	-	-42.9	-2.8
	42.9 ± 5.8	14						
14	11 ± 4	36	-	-	Liquid-scint. counting	-	-85.2	-5.9
18	68.7 ± 7.6	11	70.3 ± 7.3*	10	Proportional counter	-	-5.7	-0.3
	71.8 ± 7.8	11						
20	95.7 ± 5.6	6	-	-	Low background gross alpha/beta counter	-	28.4	1.8
22	64 ± 10.909	17	66.0 ± 11.3*	17	Liquid-scint. counting	-	-11.4	-0.6
	68 ± 12.121	18						
24	55.9 ± 12	21	56.5 ± 12.1	21	^{90}Sr chemical separation, ^{90}Y beta measurement	-	-24.2	-1.1
	57.1 ± 12.2	21						
26	67 ± 6	9	67.5 ± 7.0	10	Liquid-scint. counting	-	-9.4	-0.6
	68 ± 8	12						
27	42 ± 6	14	-	-	Y-90 ingrowth	-	-43.6	-2.8
28	175 ± 32	18	-	-	Liquid-scint. counting	yes	134.8	3.0
33	100 ± 42	42	-	-	Liquid-scint. counting	-	34.2	0.6
34	72.2 ± 6.6	9	-	-	Chemical sep. of strontium; ^{90}Y counting	-	-3.1	-0.2
37	68.3 ± 10.2	15	69.7 ± 10.4	15	Beta plastic scintillation counter	-	-6.5	-0.3
	71 ± 10.6	15						
38	52.7 ± 8.8	17	54.1 ± 9.6	18	Gas flow	-	-27.5	-1.5

	55.4 ± 10.4	19			proportional counter			
39	18.3 ± 7.1	39	-	-	Liquid-scint. counting	-	**-75.4**	**-4.6**
41	45.3 ± 2.3	5	47.8 ± 2.7	6	Liquid-scint. counting	-	**-35.9**	**-2.6**
	50.2 ± 3.2	6						
43	27.5 ± 3.4	12	-	-	Liquid-scint. counting	-	**-63.1**	**-4.4**
44	42.38 ± 1.246	3	-	-	Proportional counting	-	**-43.1**	**-3.2**
49	66.8 ± 6.99	10	-	-	?	-	**-10.4**	**-0.6**
50	58.9 ± 6.6	11	-	-	Ion exchange chromatography followed by Yttrium separation and beta counting on low background detector	-	**-21.0**	-1.3
51	60.2 ± 2.3	4	62.8 ± 2.7	4	Separation of Sr on anion exchange column, counting on beta counter in ^{90}Sr-^{90}Y equilibrium	-	**-15.8**	-1.1
	65.3 ± 3.1	5						
52	53 ± 13	25	55.0 ± 13.5	25	Radiochemical separation, proportional counting	-	**-26.2**	-1.2
	57 ± 14	25						
53	58.5 ± 7.8	13	58.6 ± 7.8	13	Proportional alfa/beta counter	-	**-21.4**	-1.3
	58.7 ± 7.7	13						
54	117 ± 10	9	-	-	Low beta gas flow proportional counter	-	**57.0**	**3.0**
55	48.6 ± 5.3	11	50.3 ± 5.5	11	Separation - Proportional counter	-	**-32.6**	**-2.1**
	51.9 ± 5.6	11						
56	50.3 ± 9	18	55.2 ± 10.9	20	Gross beta counting with chemical separation	-	**-26.0**	-1.3
	60 ± 13	22						
59	55.7 ± 5.4	10	-	-	Liquid-scint. counting	-	**-25.3**	**-1.6**
61	62.74 ± 6.32	10	-	-	Radiochemistry, beta measurement	-	**-15.8**	**-1.0**
63	48.6 ± 4.1	8	49.7 ± 4.2	8	Proportional counter	-	**-33.4**	**-2.3**
	50.7 ± 4.2	8						
64	101.54 ± 7.2	7	99.1 ± 7.0	7	Crown ether, gas proportional counter	-	**33.0**	**2.0**
	96.68 ± 6.72	7						
65	60 ± 18	30	63.5 ± 19.0	30	Chemical treatment, GM counting of ^{90}Y	-	**-14.8**	**-0.5**
	67 ± 20	30						
66	69.1 ± 7	10	71.2 ± 7.1*	10	Chemical separation and low level beta counting	-	**-4.5**	**-0.3**
	73.2 ± 7.4	10						
67	107 ± 14	13	93.0 ± 9.3*	10	Fuming nitric acid then gas flow proportional counter resin separation then gas flow proportional counter	-	**24.7**	1.3
	78.9 ± 8	10						
69	46.66 ± 4.1	9	-	-	Direct gamma-spec.	-	**-37.4**	**-2.6**
70	81 ± 4	5	-	-	Prop. counter after radiochemical separation	-	**8.7**	**0.6**

Table 3. Activity concentrations of ^{137}Cs with expanded uncertainties (k = 2), used methods and performance scores of individual laboratories.

Lab. code	Reported values		Arithmetic means		Used method	Outlier	$D_{\%}$ (%)	E_n
	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)				
1	3410 ± 130	4	-	-	Direct gamma-spec.	-	**-4.3**	**-0.8**
2	3440 ± 360	10	-	-	Direct gamma-spec.	-	**-3.5**	**-0.3**
3	3422 ± 205	6	3422 ± 205	6	Direct gamma-spec.	-	**-4.0**	**-0.6**
	3422 ± 205	6						
4	3450 ± 190	6	3500 ± 190	5	Direct gamma-spec.	-	**-1.8**	**-0.3**
	3550 ± 190	5						
5	3372 ± 154	5	-	-	Direct gamma-spec.	-	**-5.4**	**-0.9**
6	3530 ± 140	4	-	-	Direct gamma-spec.	-	**-1.0**	**-0.2**
7	3709.8 ± 423	11	-	-	Direct gamma-spec.	-	**4.1**	**0.3**
8	3737.88 ± 138.60	4	3739 ± 139	4	Direct gamma-spec.	-	**4.9**	**0.9**
	3739.36 ± 138.65	4						
9	3260 ± 320	10	-	-	Direct gamma-spec.	-	**-8.5**	**-0.9**
10	2919 ± 270	9	-	-	Direct gamma-spec.	-	**-18.1**	**-2.1**
11	3299 ± 64	2	3330 ± 65	2	Direct gamma-spec.	-	**-6.6**	**-1.6**
	3361 ± 65	2						
12	3905 ± 940	24	-	-	Direct gamma-spec.	-	**9.5**	**0.4**
13	3391 ± 204	6	3394 ± 204	6	Direct gamma-spec.	-	**-4.8**	**-0.7**
	3396 ± 204	6						
14	3226.8 ± 255.6	8	3250 ± 361	11	Direct gamma-spec.	-	**-8.8**	**-0.8**
	3273 ± 467.2	14						
15	3540 ± 108	3	-	-	Direct gamma-spec.	-	**-0.7**	**-0.1**
16	2868 ± 145	5	2878 ± 145	5	Direct gamma-spec.	-	**-19.3**	**-3.5**
	2887 ± 145	5						
17	3373 ± 232	7	3376 ± 295	9	Direct gamma-spec.	-	**-5.3**	**-0.6**
	3379 ± 358	11						
18	3300 ± 126	4	-	-	Direct gamma-spec.	-	**-7.4**	-1.4
19	3592 ± 128	4	3646 ± 130	4	Direct gamma-spec.	-	**2.3**	**0.4**
	3699 ± 132	4						
20	3600 ± 520	14	-	-	Direct gamma-spec.	-	**1.0**	**0.1**
21	3363.3 ± 276.46	8	3373 ± 278*	8	Direct gamma-spec.	-	**-5.4**	**-0.6**
	3382.26 ± 281.4	8						
22	3681 ± 379.39	10	-	-	Direct gamma-spec.	-	**3.3**	**0.3**
23	3850 ± 72	2	-	-	Direct gamma-spec.	-	**8.0**	**1.9**
24	3596 ± 202	6	-	-	Direct gamma-spec.	-	**0.9**	**0.1**
25	3581.5 ± 383.4	11	-	-	Direct gamma-spec.	-	**0.5**	**0.0**
26	4400 ± 600	14	-	-	Direct gamma-spec.	yes	**23.4**	1.4
27	3570 ± 180	5	-	-	Direct gamma-spec.	-	**0.1**	**0.0**
28	3095 ± 340	11	-	-	Direct gamma-spec.	-	**-13.2**	-1.3
29	3217 ± 70.30	2	3278 ± 254	8	Direct gamma-spec.	-	**-8.1**	**-1.0**
	3338 ± 444.85	13						
30	3378.46 ± 270.28	8	3389 ± 268	8	Direct gamma-spec.	-	**-4.9**	**-0.6**
	3399.21 ± 266.64	8						
31	761.4 ± 178.17	23	796 ± 186	23	Measuring beta	yes	**-77.7**	**-12.1**
	831.064 ±	23						

	194.48							
32	3515 ± 200	6	-	-	Direct gamma-spec.	-	-1.4	-0.2
33	3390 ± 92	3	-	-	Direct gamma-spec.	-	-4.9	-1.1
34	3570 ± 260	7	-	-	Direct gamma-spec.	-	0.1	0.0
35	3390 ± 538	16	3395 ± 439	13	Direct gamma-spec.	-	-4.8	-0.4
	3400 ± 422	12						
36	3357 ± 268	8	3359 ± 269	8	Direct gamma-spec.	-	-5.8	-0.7
	3360 ± 270	8						
37	3492 ± 362	10	-	-	Direct gamma-spec.	-	-2.0	-0.2
38	3348 ± 460	14	3410 ± 499	15	Direct gamma-spec.	-	-4.4	-0.3
	3471 ± 540	16						
39	3543 ± 106	3	-	-	Direct gamma-spec.	-	-0.6	-0.1
40	3062 ± 6.8	0,2	3065 ± 7	0.2	Direct gamma-spec.	-	-14.0	-3.7
	3067 ± 6.4	0,2						
41	3572 ± 125	3	3647 ± 127*	3	Direct gamma-spec.	-	2.3	0.4
	3721 ± 130	3						
42	3578.78 ± 158.05	4	-	-	Direct gamma-spec.	-	0.4	0.1
43	3576 ± 98	3	-	-	Direct gamma-spec.	-	0.3	0.1
44	2376 ± 7.428	0,3	-	-	Direct gamma-spec.	yes	-33.3	-8.8
45	3440 ± 261.44	8	-	-	Direct gamma-spec.	-	-3.5	-0.4
46	3430 ± 350	10	3485 ± 355	10	Direct gamma-spec.	-	-2.2	-0.2
	3540 ± 360	10						
47	3254.72 ± 129.96	4	3259 ± 130	4	Direct gamma-spec.	-	-8.6	-1.6
	3264.16 ± 130.56	4						
48	3204.5 ± 264.5	8	-	-	Direct gamma-spec.	-	-10.1	-1.2
49	3964 ± 462	12	-	-	Direct gamma-spec.	-	11.2	0.8
50	3430 ± 130	4	-	-	Direct gamma-spec.	-	-3.8	-0.7
51	3114 ± 312	10	3117 ± 312	10	Direct gamma-spec.	-	-12.6	-1.3
	3120 ± 312	10						
52	3500 ± 530	15	-	-	Direct gamma-spec.	-	-1.8	-0.1
53	3378 ± 170	5	3382 ± 170	5	Direct gamma-spec.	-	-5.1	-0.8
	3385 ± 169	5						
54	3465 ± 429	12	-	-	Direct gamma-spec.	-	-2.8	-0.2
55	3497.1 ± 110.5	3	3500 ± 130	4	Direct gamma-spec.	-	-1.8	-0.3
	3502 ± 150.5	4						
56	3626 ± 530	15	3711 ± 545	15	Direct gamma-spec.	-	4.1	0.3
	3795 ± 560	15						
57	3530 ± 92	3	3542 ± 93	3	Direct gamma-spec.	-	-0.6	-0.1
	3554 ± 94	3						
58	3193.671 ± 146.908	5	-	-	Direct gamma-spec.	-	-10.4	-1.9
59	3782 ± 308	8	-	-	Direct gamma-spec.	-	6.1	0.6
60	3390 ± 240	7	-	-	Direct gamma-spec.	-	-4.9	-0.6
61	3394 ± 94	3	-	-	Direct gamma-spec.	-	-4.8	-1.0
62	3570 ± 75.6	2	-	-	Direct gamma-spec.	-	0.1	0.0
63	3328 ± 90	3	-	-	Direct gamma-spec.	-	-6.6	-1.5
65	3500 ± 700	20	-	-	Direct gamma-spec.	-	-1.8	-0.1
66	3542 ± 156	4	3552 ± 157	4	Direct gamma-spec.	-	-0.4	-0.1
	3562 ± 158	4						
67	3400 ± 310	9	3525 ± 378	11	Direct gamma-spec.	-	-1.1	-0.1
	3650 ± 450	12						

68	3405.27 ± 20.6	1	-	-	Direct gamma-spec.	-	**-4.5**	-1.2
69	3210 ± 80	2	-	-	Direct gamma-spec.	-	**-10.0**	**-2.3**
70	3409 ± 160	5	3413 ± 175	5	Direct gamma-spec.	-	**-4.3**	**-0.7**
	3416 ± 190	6						
71	3164 ± 126	4	-	-	Direct gamma-spec.	-	**-11.2**	**-2.2**
72	2186 ± 7	0,3	2193 ± 7	0.3	Direct gamma-spec.	yes	**-38.5**	**-10.2**
	2200 ± 6	0,3						
73	3441 ± 909	26	3444 ± 769*	22	Direct gamma-spec.	-	**-3.4**	**-0.2**
	3446 ± 915	27						

Table 4. Activity concentrations of ^{212}Pb with expanded uncertainties ($k = 2$), used methods and performance scores of individual laboratories.

Lab. code	Reported values		Arithmetic means		Used method	Outlier	$D_{\%}$ (%)	E_n
	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)				
1	19.8 ± 1.2	6	-	-	Direct gamma-spec.	-	**-5.7**	-
2	19.2 ± 2.2	11	-	-	Direct gamma-spec.	-	**-8.6**	-
3	19 ± 5	26	19.5 ± 5.0	26	Direct gamma-spec.	-	**-7.1**	-
	20 ± 5	25						
4	21.2 ± 4.8	23	21.4 ± 4.3	20	Direct gamma-spec.	-	**1.9**	-
	21.6 ± 3.7	17						
5	14.6 ± 3	21	-	-	Direct gamma-spec.	-	**-30.5**	-
6	21.5 ± 1.2	6	-	-	Direct gamma-spec.	-	**2.4**	-
7	26.2 ± 5.1	19	-	-	Direct gamma-spec.	-	**24.8**	-
8	19.57 ± 1.79	9	20.1 ± 2.1	11	Direct gamma-spec.	-	**-4.4**	-
	20.6 ± 2.50	12						
9	19.5 ± 5	26	-	-	Direct gamma-spec.	-	**-7.1**	-
10	20 ± 2.9	15	-	-	Direct gamma-spec.	-	**-4.8**	-
11	18.13 ± 1.06	6	19.4 ± 0.9	5	Direct gamma-spec.	-	**-7.4**	-
	20.76 ± 0.74	4						
13	17 ± 4	24	19.6 ± 4.1	21	Direct gamma-spec.	-	**-6.9**	-
	22.1 ± 4	18						
14	20.2 ± 2.4	12	-	-	Direct gamma-spec.	-	**-3.8**	-
15	20 ± 1.9	10	-	-	Direct gamma-spec.	-	**-4.8**	-
17	22.3 ± 3.6	16	22.4 ± 3.1	14	Direct gamma-spec.	-	**6.5**	-
	22.43 ± 2.52	11						
18	20.9 ± 1.6	8	-	-	Direct gamma-spec.	-	**-0.5**	-
19	21.9 ± 2	9	22.9 ± 2.2	10	Direct gamma-spec.	-	**8.8**	-
	23.8 ± 2.4	10						
20	24.6 ± 4	16	-	-	Direct gamma-spec.	-	**17.1**	-
21	20.59 ± 1.86	9	20.7 ± 1.7*	8	Direct gamma-spec.	-	**-1.4**	-
	20.83 ± 1.86	9						
22	21 ± 2.42	12	-	-	Direct gamma-spec.	-	**0.0**	-
23	24 ± 1.33	6	-	-	Direct gamma-spec.	-	**14.3**	-
24	22.3 ± 1.06	5	-	-	Direct gamma-spec.	-	**6.2**	-
26	32 ± 6	19	-	-	Direct gamma-spec.	yes	**52.4**	-
27	28.8 ± 1.6	6	-	-	Direct gamma-spec.	-	**37.1**	-
28	18.1 ± 2.7	15	-	-	Direct gamma-spec.	-	**-13.8**	-
29	19.33 ± 1.94	10	19.5 ± 4.2	21	Direct gamma-spec.	-	**-7.3**	-
	19.59 ± 6.42	33						
30	23.28 ± 4.06	17	23.5 ± 3.8*	16	Direct gamma-spec.	-	**11.8**	-
	23.69 ± 3.96	17						
32	21.3 ± 1.4	7	-	-	Direct gamma-spec.	-	**1.4**	-
33	20 ± 2.2	11	-	-	Direct gamma-spec.	-	**-4.8**	-
34	22.2 ± 1.6	7	-	-	Direct gamma-spec.	-	**5.7**	-
35	25.2 ± 5.92	23	25.3 ± 5.3	21	Direct gamma-spec.	-	**20.2**	-
	25.3 ± 4.68	18						

37	22 ± 2.6	12	-	-	Direct gamma-spec.	-	**4.8**	-
38	19.2 ± 3.8	20	20.2 ± 3.8	19	Direct gamma-spec.	-	**-4.0**	-
	21.1 ± 3.8	18						
39	20.2 ± 0.8	4	-	-	Direct gamma-spec.	-	**-3.8**	-
40	15 ± 3.1	21	16.0 ± 3.3	20	Direct gamma-spec.	-	**-23.8**	-
	17 ± 3.4	20						
42	31.52 ± 2.63	8	-	-	Direct gamma-spec.	yes	**50.1**	-
43	19.8 ± 3.8	19	-	-	Direct gamma-spec.	-	**-5.7**	-
44	15.1 ± 1.18	8	-	-	Direct gamma-spec.	-	**-28.1**	-
45	25.9 ± 4.6102	18	-	-	Direct gamma-spec.	-	**23.3**	-
46	22.3 ± 2.5	11	22.2 ± 2.6	12	Direct gamma-spec.	-	**5.5**	-
	22 ± 2.6	12						
48	19.4 ± 1.7	9	-	-	Direct gamma-spec.	-	**-7.6**	-
49	27.3 ± 8.36	31	-	-	Direct gamma-spec.	-	**30.0**	-
50	18.6 ± 2.1	11	-	-	Direct gamma-spec.	-	**-11.4**	-
52	19 ± 3.1	16	19.5 ± 3.2	16	Direct gamma-spec.	-	**-7.1**	-
	20 ± 3.2	16						
53	34.3 ± 4.3	13	-	-	Direct gamma-spec.	yes	**63.3**	-
54	19.4 ± 3.7	19	-	-	Direct gamma-spec.	-	**-7.6**	-
55	16.9 ± 3.7	22	21.1 ± 3.6	17	Direct gamma-spec.	-	**0.2**	-
	25.2 ± 3.1	12						
56	20.7 ± 3.5	17	22.1 ± 3.7	17	Direct gamma-spec.	-	**5.2**	-
	23.5 ± 3.8	16						
57	22.4 ± 1.2	5	22.7 ± 1.2	5	Direct gamma-spec.	-	**7.9**	-
	22.9 ± 1.2	5						
58	17.452 ± 1.222	7	-	-	Direct gamma-spec.	-	**-16.9**	-
59	23.8 ± 2.4	10	-	-	Direct gamma-spec.	-	**13.3**	-
60	21 ± 2.2	10	-	-	Direct gamma-spec.	-	**0.0**	-
61	18.52 ± 1.46	8	-	-	Direct gamma-spec.	-	**-11.8**	-
62	18 ± 1.48	8	-	-	Direct gamma-spec.	-	**-14.3**	-
63	19.8 ± 1.4	7	-	-	Direct gamma-spec.	-	**-5.7**	-
65	21 ± 4	19	-	-	Direct gamma-spec.	-	**0.0**	-
66	20.3 ± 2.8	14	21.1 ± 2.8	13	Direct gamma-spec.	-	**0.5**	-
	21.9 ± 2.8	13						
67	22.9 ± 2.3	10	23.1 ± 3.0	13	Direct gamma-spec.	-	**9.8**	-
	23.2 ± 3.8	16						
68	19.91 ± 4.66	23	-	-	Direct gamma-spec.	-	**-5.2**	-
69	19.6 ± 1	5	-	-	Direct gamma-spec.	-	**-6.7**	-
70	21 ± 1.8	9	21.3 ± 2.3	11	Direct gamma-spec.	-	**1.4**	-
	21.6 ± 2.8	13						
71	20 ± 1.6	8	-	-	Direct gamma-spec.	-	**-4.8**	-
73	14 ± 8	57	16.5 ± 6.5	39	Direct gamma-spec.	-	**-21.4**	-
	19 ± 4	21						

Table 5. Activity concentrations of ^{212}Bi with expanded uncertainties ($k = 2$), used methods and performance scores of individual laboratories.

Lab. code	Reported values		Arithmetic means		Used method	Outlier	$D_{\%}$ (%)	E_n
	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)				
1	19.2 ± 1.8	9	-	-	Direct gamma-spec.	-	**-8.6**	-
2	20.6 ± 4.2	20	-	-	Direct gamma-spec.	-	**-1.9**	-
3	28 ± 8	29	28.5 ± 8.0	20	Direct gamma-spec.	-	**35.7**	-
	29 ± 8	28						
4	24 ± 14	58	24.5 ± 13.5	55	Direct gamma-spec.	-	**16.7**	-
	25 ± 13	52						
5	18.3 ± 10	55	-	-	Direct gamma-spec.	-	**-12.9**	-
6	22.9 ± 1.8	8	-	-	Direct gamma-spec.	-	**9.0**	-
7	27.2 ± 8.8	32	-	-	Direct gamma-spec.	-	**29.5**	-

8	10.86 ± 3.9	36	11.0 ± 3.9	35	Direct gamma-spec.	-	**-47.7**	-
	11.09 ± 3.83	35						
9	21 ± 6	29	-	-	Direct gamma-spec.	-	**0.0**	-
10	18.4 ± 3.2	17	-	-	Direct gamma-spec.	-	**-12.4**	-
11	14.18 ± 1.73	12	14.3 ± 1.8	13	Direct gamma-spec.	-	**-32.1**	-
	14.33 ± 1.91	13						
14	19.9 ± 6	30	-	-	Direct gamma-spec.	-	**-5.2**	-
15	20 ± 1.9	10	-	-	Direct gamma-spec.	-	**-4.8**	-
17	20.81 ± 4.9	24	21.2 ± 8.7	41	Direct gamma-spec.	-	**1.1**	-
	21.65 ± 12.6	58						
18	20.3 ± 1.2	6	-	-	Direct gamma-spec.	-	**-3.3**	-
19	26.2 ± 6.2	24	28.2 ± 8.5	30	Direct gamma-spec.	-	**34.3**	-
	30.2 ± 11	36						
20	25.4 ± 4.4	17	-	-	Direct gamma-spec.	-	**21.0**	-
21	21.23 ± 3.82	18	22.1 ± 3.2*	15	Direct gamma-spec.	-	**5.1**	-
	22.92 ± 3.96	17						
22	23 ± 4.85	21	-	-	Direct gamma-spec.	-	**9.5**	-
23	14 ± 2.67	19	-	-	Direct gamma-spec.	-	**-33.3**	-
26	33 ± 14	42	-	-	Direct gamma-spec.	-	**57.1**	-
27	24.3 ± 1.8	7	-	-	Direct gamma-spec.	-	**15.7**	-
29	12.7 ± 3.64	29	15.2 ± 5.1	34	Direct gamma-spec.	-	**-27.7**	-
	17.68 ± 6.91	39						
30	19.66 ± 2.36	12	20.6 ± 2.3*	11	Direct gamma-spec.	-	**-1.9**	-
	21.54 ± 2.58	12						
32	21.8 ± 3	14	-	-	Direct gamma-spec.	-	**3.8**	-
33	20 ± 1.64	8	-	-	Direct gamma-spec.	-	**-4.8**	-
34	14.7 ± 2.8	19	-	-	Direct gamma-spec.	-	**-30.0**	-
35	< 23.1		-	-	Direct gamma-spec.	-	**31.4**	-
	27.6 ± 9.7	35						
37	18.4 ± 3.4	18	-	-	Direct gamma-spec.	-	**-12.4**	-
38	18.9 ± 5.4	29	19.5 ± 5.0	26	Direct gamma-spec.	-	**-7.4**	-
	20 ± 4.6	23						
39	22.2 ± 2	9	-	-	Direct gamma-spec.	-	**5.7**	-
40	14 ± 2.5	18	14.5 ± 2.5	17	Direct gamma-spec.	-	**-31.0**	-
	15 ± 2.5	17						
43	19.4 ± 8	41	-	-	Direct gamma-spec.	-	**-7.6**	-
45	29.8 ± 9.238	31	-	-	Direct gamma-spec.	-	**41.9**	-
46	21.6 ± 4.3	20	22.0 ± 4.2	19	Direct gamma-spec.	-	**4.5**	-
	22.3 ± 4	18						
48	18 ± 2.8	16	-	-	Direct gamma-spec.	-	**-14.3**	-
49	26.8 ± 15.2	57	-	-	Direct gamma-spec.	-	**27.6**	-
50	22.1 ± 3.1	14	-	-	Direct gamma-spec.	-	**5.2**	-
52	19 ± 4	21	20.0 ± 3.5	18	Direct gamma-spec.	-	**-4.8**	-
	21 ± 3	14						
54	17.9 ± 6.6	37	-	-	Direct gamma-spec.	-	**-14.8**	-
55	9.1 ± 5	55	-	-	Direct gamma-spec.	-	**-56.7**	-
56	19.6 ± 5.7	29	20.0 ± 4.7*	24	Direct gamma-spec.	-	**-5.0**	-
	20.3 ± 5.6	28						
57	22.1 ± 2.4	11	22.4 ± 2.4	11	Direct gamma-spec.	-	**6.4**	-
	22.6 ± 2.4	11						
58	12.06 ± 2.87	24	-	-	Direct gamma-spec.	-	**-42.6**	-
59	22.9 ± 7.2	31	-	-	Direct gamma-spec.	-	**9.0**	-
60	21.8 ± 2.4	11	-	-	Direct gamma-spec.	-	**3.8**	-
61	21.06 ± 3.1	15	-	-	Direct gamma-spec.	-	**0.3**	-
62	13.9 ± 2.33	17	-	-	Direct gamma-spec.	-	**-33.8**	-
63	23.8 ± 7.3	31	-	-	Direct gamma-spec.	-	**13.3**	-
65	22 ± 4	18	-	-	Direct gamma-spec.	-	**4.8**	-
66	21.8 ± 3	14	22.0 ± 3.0	14	Direct gamma-spec.	-	**4.8**	-

	22.2 ± 3	14						
67	22 ± 4	18	22.8 ± 4.3	19	Direct gamma-spec.	-	**8.3**	-
	23.5 ± 4.6	20						
68	12.55 ± 4.06	32	-	-	Direct gamma-spec.	-	**-40.2**	-
69	19.6 ± 2	10	-	-	Direct gamma-spec.	-	**-6.7**	-
70	21 ± 1.8	9	21.3 ± 2.3	11	Direct gamma-spec.	-	**1.4**	-
	21.6 ± 2.8	13						
71	19.8 ± 0.8	4	-	-	Direct gamma-spec.	-	**-5.7**	-
73	29 ± 10	34	30.5 ± 12.9	42	Direct gamma-spec.	-	**45.2**	-
	32 ± 16	50						

Table 6. Activity concentrations of ^{214}Pb with expanded uncertainties ($k = 2$), used methods and performance scores of individual laboratories.

Lab. code	Reported values		Arithmetic means		Used method	Outlier	$D_{\%}$ (%)	E_n
	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)				
1	19.9 ± 1.2	6	-	-	Direct gamma-spec.	-	**5.0**	-
2	13.2 ± 1.4	11	-	-	Direct gamma-spec.	-	**-30.3**	-
3	< 30	-	-	-	Direct gamma-spec.	-	-	-
	< 30	-						
4	23.3 ± 4.8	21	24.6 ± 5.1	21	Direct gamma-spec.	-	**29.8**	-
	25.9 ± 5.5	21						
5	13 ± 4	31	-	-	Direct gamma-spec.	-	**-31.4**	-
6	20.8 ± 1.4	7	-	-	Direct gamma-spec.	-	**9.8**	-
7	22.8 ± 4	18	-	-	Direct gamma-spec.	-	**20.3**	-
8	19.81 ± 2.51	13	20.0 ± 2.8	14	Direct gamma-spec.	-	**5.3**	-
	20.09 ± 3.19	16						
9	24 ± 10	42	-	-	Direct gamma-spec.	-	**26.6**	-
10	20.6 ± 3	15	-	-	Direct gamma-spec.	-	**8.7**	-
11	17.11 ± 1.2	7	17.2 ± 1.3	7	Direct gamma-spec.	-	**-9.1**	-
	17.36 ± 1.32	8						
14	14.6 ± 4.4	30	-	-	Direct gamma-spec.	-	**-23.0**	-
15	21.9 ± 3	14	-	-	Direct gamma-spec.	-	**15.6**	-
16	10 ± 2.95	30	12.6 ± 3.3	26	Direct gamma-spec.	-	**-33.5**	-
	15.2 ± 3.36	22						
17	19.68 ± 4.2	21	20.3 ± 3.5	17	Direct gamma-spec.	-	**6.9**	-
	20.83 ± 2.74	13						
18	23.3 ± 1.6	7	-	-	Direct gamma-spec.	-	**22.9**	-
19	25.2 ± 3.4	13	26.8 ± 4.1	15	Direct gamma-spec.	-	**41.1**	-
	28.3 ± 4.8	17						
20	24.3 ± 4.4	18	-	-	Direct gamma-spec.	-	**28.2**	-
21	16.97 ± 1.8	11	17.0 ± 1.4*	8	Direct gamma-spec.	-	**-10.1**	-
	17.09 ± 1.7	10						
22	20 ± 2.42	12	-	-	Direct gamma-spec.	-	**5.5**	-
23	25 ± 1.33	5	-	-	Direct gamma-spec.	-	**31.9**	-
24	20 ± 1.44	7	-	-	Direct gamma-spec.	-	**5.5**	-
26	34 ± 8	24	-	-	Direct gamma-spec.	-	**79.4**	-
27	20.9 ± 1.4	7	-	-	Direct gamma-spec.	-	**10.3**	-
28	16.6 ± 2.6	16	-	-	Direct gamma-spec.	-	**-12.4**	-
30	20.86 ± 3.34	16	20.9 ± 3.1*	15	Direct gamma-spec.	-	**10.4**	-
	21 ± 3.36	16						
32	21.7 ± 3	14	-	-	Direct gamma-spec.	-	**14.5**	-
33	19.9 ± 1.58	8	-	-	Direct gamma-spec.	-	**5.0**	-
34	16.5 ± 2.4	15	-	-	Direct gamma-spec.	-	**-12.9**	-
35	17.9 ± 3.66	20	18.5 ± 4.4	24	Direct gamma-spec.	-	**-2.6**	-
	19 ± 5.24	28						
37	17.7 ± 2.6	15	-	-	Direct gamma-spec.	-	**-6.6**	-
38	20.6 ± 4.4	21	21.1 ± 4.2	20	Direct gamma-spec.	-	**11.3**	-

	21.6 ± 4	19						
39	15.6 ± 1.1	7	-	-	Direct gamma-spec.	-	-17.7	-
40	18 ± 2	11	18.5 ± 2.0	11	Direct gamma-spec.	-	-2.4	-
	19 ± 1.9	10						
42	29.5 ± 2.9	10	-	-	Direct gamma-spec.	-	55.7	-
43	20.5 ± 5	24	-	-	Direct gamma-spec.	-	8.2	-
44	9.63 ± 1.128	12	-	-	Direct gamma-spec.	-	-49.2	-
45	25.4 ± 3.3528	13	-	-	Direct gamma-spec.	-	34.0	-
46	16.8 ± 2.6	15	17.0 ± 2.6	16	Direct gamma-spec.	-	-10.6	-
	17.1 ± 2.7	16						
48	18.1 ± 1.4	8	-	-	Direct gamma-spec.	-	-4.5	-
49	26.5 ± 5.1	19	-	-	Direct gamma-spec.	-	39.8	-
50	15.2 ± 1.4	9	-	-	Direct gamma-spec.	-	-19.8	-
52	22 ± 3.6	16	22.0 ± 3.7	17	Direct gamma-spec.	-	16.1	-
	22 ± 3.8	17						
53	28.9 ± 9	31	-	-	Direct gamma-spec.	-	52.5	-
54	19.2 ± 5.1	27	-	-	Direct gamma-spec.	-	1.3	-
55	18.8 ± 4.6	24	19.7 ± 4.0*	20	Direct gamma-spec.	-	3.7	-
	20.5 ± 6	29						
56	24.4 ± 5	20	25.8 ± 5.2	20	Direct gamma-spec.	-	36.1	-
	27.2 ± 5.4	20						
57	22.4 ± 2.8	13	22.7 ± 2.8	12	Direct gamma-spec.	-	19.8	-
	23 ± 2.8	12						
58	15.048 ± 1.866	12	-	-	Direct gamma-spec.	-	-20.6	-
59	24 ± 2.4	10	-	-	Direct gamma-spec.	-	26.6	-
60	23.1 ± 2.4	10	-	-	Direct gamma-spec.	-	21.9	-
61	16.69 ± 2.3	14	-	-	Direct gamma-spec.	-	-11.9	-
62	16.7 ± 2.15	13	-	-	Direct gamma-spec.	-	-11.9	-
63	16.3 ± 5.2	32	-	-	Direct gamma-spec.	-	-14.0	-
65	18 ± 4	22	-	-	Direct gamma-spec.	-	-5.0	-
66	21.5 ± 3	14	21.8 ± 3.0	14	Direct gamma-spec.	-	14.8	-
	22 ± 3	14						
67	23.1 ± 3.6	16	23.7 ± 3.1	13	Direct gamma-spec.	-	25.1	-
	24.3 ± 2.5	10						
68	14.35 ± 3.98	28	-	-	Direct gamma-spec.	-	-24.3	-
69	21.4 ± 1.2	6	-	-	Direct gamma-spec.	-	12.9	-
70	16.3 ± 1.2	7	16.7 ± 1.2	7	Direct gamma-spec.	-	-12.1	-
	17 ± 1.2	7						
71	19.7 ± 1.4	7	-	-	Direct gamma-spec.	-	3.9	-
72	29.64 ± 4	13	29.8 ± 4.5	15	Direct gamma-spec.	-	57.3	-
	30 ± 5	17						
73	20 ± 14	70	20.5 ± 10.1	49	Direct gamma-spec.	-	8.2	-
	21 ± 6	29						

Table 7. Activity concentrations of ^{214}Bi with expanded uncertainties ($k = 2$), used methods and performance scores of individual laboratories.

Lab. code	Reported values		Arithmetic means		Used method	Outlier	$D_{\%}$ (%)	E_n
	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)				
1	20.3 ± 1.4	7	-	-	Direct gamma-spec.	-	7.1	-
2	12.8 ± 1.4	11	-	-	Direct gamma-spec.	-	-32.5	-
3	< 30	-	-	-	Direct gamma-spec.	-	-	-
	< 30	-						
4	18.8 ± 4.5	24	19.2 ± 4.2	22	Direct gamma-spec.	-	1.0	-
	19.5 ± 3.8	19						
5	14.8 ± 3	20	-	-	Direct gamma-spec.	-	-21.9	-
6	20.5 ± 1.2	6	-	-	Direct gamma-spec.	-	8.2	-
7	22.7 ± 3.6	16	-	-	Direct gamma-spec.	-	19.8	-

8	25.46 ± 2.13	8	25.6 ± 2.2	9	Direct gamma-spec.	-	**34.9**	-
	25.66 ± 2.22	9						
9	23 ± 10	43	-	-	Direct gamma-spec.	-	**21.4**	-
10	23.1 ± 3.1	13	-	-	Direct gamma-spec.	-	**21.9**	-
11	16.25 ± 0.87	5	16.4 ± 0.8	5	Direct gamma-spec.	-	**-13.3**	-
	16.63 ± 0.75	5						
13	10 ± 3.6	36	11.1 ± 3.8	34	Direct gamma-spec.	-	**-41.4**	-
	12.2 ± 4	33						
14	13.7 ± 2.6	19	-	-	Direct gamma-spec.	-	**-27.7**	-
15	22 ± 3.2	15	-	-	Direct gamma-spec.	-	**16.1**	-
16	15.1 ± 2.77	18	17.6 ± 2.9	16	Direct gamma-spec.	-	**-7.4**	-
	20 ± 2.85	14						
17	17.31 ± 1.98	11	18.4 ± 2.7	15	Direct gamma-spec.	-	**-2.9**	-
	19.5 ± 3.44	18						
18	21.5 ± 1	5	-	-	Direct gamma-spec.	-	**13.4**	-
19	25.9 ± 4.6	18	26.1 ± 5.3	20	Direct gamma-spec.	-	**37.7**	-
	26.3 ± 6	23						
20	19.7 ± 3.2	16	-	-	Direct gamma-spec.	-	**3.9**	-
21	16.26 ± 1.48	9	16.3 ± 1.2*	7	Direct gamma-spec.	-	**-13.8**	-
	16.4 ± 1.5	9						
22	21 ± 4.85	23	-	-	Direct gamma-spec.	-	**10.8**	-
23	22 ± 1.33	6	-	-	Direct gamma-spec.		**16.1**	
24	17.7 ± 1.46	8	-	-	Direct gamma-spec.	-	**-6.6**	-
26	32 ± 8	25	-	-	Direct gamma-spec.	-	**68.9**	-
27	19.1 ± 1.3	7	-	-	Direct gamma-spec.	-	**0.8**	-
28	15.5 ± 2.5	16	-	-	Direct gamma-spec.	-	**-18.2**	-
30	21.62 ± 3.04	14	22.1 ± 2.9*	13	Direct gamma-spec.	-	**16.6**	-
	22.56 ± 3.16	14						
32	21 ± 3	14	-	-	Direct gamma-spec.	-	**10.8**	-
33	20 ± 2.2	11	-	-	Direct gamma-spec.	-	**5.5**	-
34	16.2 ± 4	25	-	-	Direct gamma-spec.	-	**-14.5**	-
35	15.5 ± 2.44	16	15.9 ± 2.9	18	Direct gamma-spec.	-	**-16.4**	-
	16.2 ± 3.42	21						
37	16.4 ± 2	12	-	-	Direct gamma-spec.	-	**-13.5**	-
38	19 ± 3.2	17	19.3 ± 3.2	17	Direct gamma-spec.	-	**1.8**	-
	19.6 ± 3.2	16						
39	15.3 ± 0.6	4	-	-	Direct gamma-spec.	-	**-19.3**	-
40	21 ± 2.5	12	21.5 ± 2.7	13	Direct gamma-spec.	-	**13.4**	-
	22 ± 3	14						
42	30.05 ± 2.75	9	-	-	Direct gamma-spec.	-	**58.6**	-
43	17.5 ± 2.6	15	-	-	Direct gamma-spec.	-	**-7.7**	-
44	9.875 ± 0.905	9	-	-	Direct gamma-spec.	-	**-47.9**	-
45	23.6 ± 2.4072	10	-	-	Direct gamma-spec.	-	**24.5**	-
46	17.9 ± 2.2	12	18.4 ± 2.3	13	Direct gamma-spec.	-	**-3.2**	-
	18.8 ± 2.4	13						
48	15.7 ± 1	6	-	-	Direct gamma-spec.	-	**-17.2**	-
49	21.7 ± 2.36	11	-	-	Direct gamma-spec.	-	**14.5**	-
50	15.3 ± 1.4	9	-	-	Direct gamma-spec.	-	**-19.3**	-
52	20 ± 3	15	20.5 ± 3.2	16	Direct gamma-spec.	-	**8.2**	-
	21 ± 3.4	16						
53	23.8 ± 9.1	38	-	-	Direct gamma-spec.	-	**25.6**	-
54	18.6 ± 3.9	21	-	-	Direct gamma-spec.	-	**-1.9**	-
55	16.4 ± 2.6	16	17.5 ± 1.9*	11	Direct gamma-spec.	-	**-7.9**	-
	18.5 ± 1.9	10						
56	24.5 ± 4.6	19	26.3 ± 4.8	18	Direct gamma-spec.	-	**38.5**	-
	28 ± 5	18						
57	21.6 ± 2.6	12	22.1 ± 2.6	12	Direct gamma-spec.	-	**16.6**	-
	22.6 ± 2.6	12						

58	13.702 ± 1.206	9	-	-	Direct gamma-spec.	-	**-27.7**	-
59	23.2 ± 3.7	16	-	-	Direct gamma-spec.	-	**22.4**	-
60	24.8 ± 2	8	-	-	Direct gamma-spec.		**30.9**	
61	16.12 ± 1.44	9	-	-	Direct gamma-spec.	-	**-14.9**	-
62	18.2 ± 1.15	6	-	-	Direct gamma-spec.	-	**-4.0**	-
63	15.9 ± 2.8	18	-	-	Direct gamma-spec.	-	**-16.1**	-
65	19 ± 4	21	-	-	Direct gamma-spec.	-	**0.3**	-
66	20.9 ± 2.4	11	21.2 ± 2.4	11	Direct gamma-spec.	-	**11.6**	-
	21.4 ± 2.4	11						
67	22.1 ± 2.6	12	22.4 ± 3.2	14	Direct gamma-spec.	-	**18.2**	-
	22.7 ± 3.8	17						
68	16.71 ± 2.45	15	-	-	Direct gamma-spec.	-	**-11.8**	-
69	20 ± 3.1	16	-	-	Direct gamma-spec.	-	**5.5**	-
70	16.3 ± 1.2	7	16.7 ± 1.2	7	Direct gamma-spec.	-	**-12.1**	-
	17 ± 1.2	7						
71	19 ± 1	5	-	-	Direct gamma-spec.	-	**0.3**	
72	39.67 ± 5	13	40.5 ± 4.5	11	Direct gamma-spec.	yes	**113.9**	-
	41.4 ± 4	10						
73	16 ± 4	25	18.0 ± 5.9	33	Direct gamma-spec.	-	**-5.0**	-
	20 ± 8	40						

Table 8. Activity concentrations of ^{226}Ra with expanded uncertainties (k = 2), used methods and performance scores of individual laboratories.

Lab. code	Reported values		Arithmetic means		Used method	Outlier	$D_{\%}$ (%)	E_n
	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)				
1	19.4 ± 2.3	12	-	-	Alpha spectrometry	-	**2.4**	**0.1**
	20.6 ± 2.3	11			Direct gamma-spec.	-	**8.7**	**0.5**
2	11.5 ± 2	17	-	-	Direct gamma-spec.	-	**-39.3**	**-2.4**
3	< 30	-	-	-	Direct gamma-spec.	-	-	-
	< 30	-	-	-				
4	12.22 ± 0.82	7	12.5 ± 0.8	7	Gas flow prop. counting	-	**-34.2**	**-2.7**
	12.72 ± 0.81	6						
5	14 ± 4	29	-	-	Direct gamma-spec.	-	**-26.1**	-1.1
6	20.6 ± 1	5	-	-	Direct gamma-spec.	-	**8.7**	**0.7**
7	22.8 ± 7.3	32	-	-	Direct gamma-spec.	-	**20.3**	**0.5**
9	23.5 ± 8	34	-	-	Direct gamma-spec.	-	**24.0**	**0.5**
10	22.1 ± 4.1	19	-	-	Direct gamma-spec.	-	**16.6**	**0.7**
11	16.25 ± 0.87	5	16.4 ± 0.8	5	Direct gamma-spec.	-	**-13.3**	-1.0
	16.63 ± 0.75	5						
14	40 ± 40	100	-	-	Direct gamma-spec.	yes	**111.1**	**0.5**
15	22 ± 3.1	14	-	-	Direct gamma-spec.	-	**16.1**	**0.8**
17	18.25 ± 1.88	10	18.9 ± 2.4	13	Direct gamma-spec.	-	**-0.2**	**0.0**
	19.57 ± 3.04	16						
18	22.3 ± 2.8	13	-	-	Alpha spectrometry	-	**17.7**	**0.9**
	31 ± 8	26			Direct gamma-spec.	-	**63.6**	1.4
20	20.4 ± 3.4	17	-	-	Direct gamma-spec.	-	**7.6**	**0.4**
21	16.62 ± 5.6	34	16.7 ± 4.5*	27	Direct gamma-spec.	-	**-12.0**	**-0.4**
	16.74 ± 5.66	34						
22	20 ± 7.273	36	-	-	Gamma-spec. (daughter products)	-	**5.5**	**0.1**
26	33 ± 6	18	-	-	Direct gamma-spec.	-	**74.1**	**2.2**
27	18.9 ± 1.5	8	-	-	Direct gamma-spec.	-	**-0.3**	**0.0**
	26.2 ± 1	4			Lucas cell	-	**38.2**	**2.9**
30	23.75 ± 4.26	18	23.8 ± 4.0*	17	Direct gamma-spec.	-	**25.6**	1.0
	23.85 ± 4.28	18						

31	< 42701.74	-	-	-	The installation of measuring radon and thoron	-	-	-
	< 43384.88	-						
32	21.2 ± 2.8	13	-	-	Direct gamma-spec.	-	**11.9**	**0.6**
33	20 ± 1.38	7	-	-	Direct gamma-spec.	-	**5.5**	**0.4**
34	16.4 ± 2.4	15	-	-	Direct gamma-spec.	-	**-13.5**	**-0.8**
35	< 40.1	-	-	-	Direct gamma-spec.	-	-	-
	< 40.2	-						
38	18.8 ± 3.4	18	19.3 ± 3.3	17	Direct gamma-spec.	-	**1.8**	**0.1**
	19.8 ± 3.2	16						
39	45.2 ± 4.5	10	-		Direct gamma-spec.	yes	**138.5**	**5.2**
40	21 ± 2.5	12	21.5 ± 2.7	13	Direct gamma-spec.	-	**13.4**	**0.7**
	22 ± 3	14						
41	17.7 ± 3	17	17.9 ± 3.4	19	Direct gamma-spec.	-	**-5.5**	**-0.3**
	18.1 ± 3.9	22						
42	29.12 ± 2.63	9	-	-	Direct gamma-spec.	-	**53.7**	**2.9**
43	19.7 ± 4	20	-	-	Alpha spectrometry	-	**3.9**	**0.2**
45	24.5 ± 4.116	17	-	-	Direct gamma-spec.	-	**29.3**	1.2
46	24.6 ± 9	37	24.7 ± 11.5	47	Direct gamma-spec.	-	**30.3**	**0.5**
	24.8 ± 14	56						
48	16.9 ± 1.2	7	-	-	Direct gamma-spec.	-	**-10.8**	**-0.8**
50	16.3 ± 4.1	25	-	-	^{226}Ra by emanation	-	**-14.0**	**-0.6**
51	15.7 ± 3.6	23	16.3 ± 3.6	22	Direct gamma-spec.	-	**-14.0**	**-0.6**
	16.9 ± 3.5	21						
52	25 ± 6.8	27	25.0 ± 7.2	29	Direct gamma-spec.	-	**31.9**	**0.8**
	25 ± 7.5	30						
53	26.3 ± 11	42	-	-	Direct gamma-spec.	-	**38.8**	**0.7**
54	33.7 ± 17.4	52	-	-	Direct gamma-spec.	-	**77.8**	**0.8**
55	16.4 ± 2.6	16	17.5 ± 1.9*	11	Direct gamma-spec.	-	**-7.9**	**-0.5**
	18.5 ± 1.9	10						
56	< 73	-	-		Direct gamma-spec.	-	-	-
	< 92	-						
57	25.7 ± 3.8	15	26.1 ± 3.8	15	Direct gamma-spec.	-	**37.7**	**1.6**
	26.5 ± 3.8	14						
58	13.702 ± 1.206	9	-	-	Direct gamma-spec.	-	**-27.7**	**-2.0**
59	20.3 ± 2.1	10	-	-	Alpha spectrometry	-	**7.1**	**0.4**
60	< 33	-	-	-	Direct gamma-spec.	-	**-**	-
63	16 ± 0.5	3	-	-	Direct gamma-spec.	-	**-15.6**	-1.3
65	19 ± 6	32	-	-	Direct gamma-spec.	-	**0.3**	**0.0**
69	21 ± 2	10	-	-	Direct gamma-spec.	-	**10.8**	**0.7**
70	16.3 ± 1.2	7	16.7 ± 1.2	7	Direct gamma-spec.	-	**-12.1**	**-0.9**
	17 ± 1.2	7						
71	22 ± 1	5	-		Direct gamma-spec.	-	**16.1**	1.2
72	50 ± 7	14	52.0 ± 7.7	15	Direct gamma-spec.	yes	**174.4**	**4.1**
	54 ± 8.5	16						
73	19 ± 6	32	21.0 ± 6.1*	29	Direct gamma-spec.	-	**10.8**	**0.3**
	23 ± 10	43						

Table 9. Activity concentrations of ^{230}Th with expanded uncertainties ($k = 2$), used methods and performance scores of individual laboratories.

Lab. code	Reported values		Arithmetic means		Used method	Outlier	$D_\%$ (%)	E_n
	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_\%$ (%)	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_\%$ (%)				
1	22 ± 2	9	-	-	Alpha spectrometry	-	**5.3**	**0.4**
3	15 ± 2	13	15.0 ± 2.0	13	Alpha spectrometry	-	**-28.2**	**-1.9**
	15 ± 2	13						
4	19 ± 1.9	10	19.1 ± 1.9	10	Alpha spectrometry	-	**-8.6**	**-0.6**

	19.2 ± 1.9	10						
5	25.7 ± 1.6	6	-	-	Alpha spectrometry	-	**23.0**	**1.7**
13	22.1 ± 5.6	25	23.0 ± 3.9	17	Alpha spectrometry	-	**10.1**	**0.5**
	23.9 ± 2	8						
15	15.5 ± 1.8	12	16.0 ± 2.0	13	Alpha spectrometry	-	**-23.7**	**-1.6**
	16.4 ± 2.2	13						
18	27.4 ± 2.8	10	-	-	Alpha spectrometry	-	**31.1**	**1.8**
20	21.5 ± 2.6	12	-	-	Alpha spectrometry	-	**2.9**	**0.2**
22	< 116	-	-	-	Direct gamma-spec.	-	**-**	-
26	29 ± 4	14	-	-	Alpha spectrometry	-	**38.8**	**1.8**
27	21.7 ± 2.8	13	-	-	Alpha spectrometry	-	**3.8**	**0.2**
35	< 157	-	-	-	Direct gamma-spec.	-	-	-
	< 211	-						
39	23.1 ± 1.4	6	-	-	Alpha spectrometry	-	**10.5**	**0.8**
45	24.5 ± 7.644	31	24.5 ± 7.6	31	Direct gamma-spec.	-	**17.2**	**0.5**
	24.5 ± 7.644	31						
49	14 ± 1.79	13	-	-	Alpha spectrometry	-	**-33.0**	**-2.4**
50	21.1 ± 3.4	16	-	-	Alpha spectrometry	-	**1.0**	**0.0**
56	24.2 ± 3.9	16	27.1 ± 5.6	21	Alpha spectrometry	-	**29.7**	1.0
	30 ± 7.6	25						
59	13.1 ± 0.6	5	-	-	Alpha spectrometry	-	**-37.3**	**-3.3**
60	< 175	-	-	-	Direct gamma-spec.	-	-	-
62	11.6 ± 3.5	30	-	-	Alpha spectrometry	-	**-44.5**	**-2.2**
63	60 ± 26	43	-	-	Alpha spectrometry	yes	**187.1**	1.5
67	20.9 ± 2	10	-	-	Alpha spectrometry	-	**0.0**	**0.0**
69	19.4 ± 2	10	-	-	Direct gamma-spec.	-	**-7.2**	**-0.5**
70	16.3 ± 1.2	7	16.7 ± 1.2	7	Direct gamma-spec.	-	**-20.3**	**-1.6**
	17 ± 1.2	7						
72	14.32 ± 7.8	54	-	-	Direct gamma-spec.	-	**-31.5**	**-0.8**

Table 10. Activity concentrations of ^{232}Th with expanded uncertainties ($k = 2$), used methods and performance scores of individual laboratories.

Lab. code	Reported values		Arithmetic means		Used method	Outlier	$D_{\%}$ (%)	E_n
	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)				
1	19 ± 2.2	12	-	-	Alpha spectrometry	-	**-9.5**	**-0.6**
	20.5 ± 1.5	7			Direct gamma-spec.	-	**-2.4**	**-0.2**
2	20.4 ± 2.8	14	-	-	Direct gamma-spec.	-	**-2.9**	**-0.2**
3	17 ± 2	12	17.5 ± 2.0	11	Alpha spectrometry	-	**-16.7**	-1.0
	18 ± 2	11						
4	17.1 ± 1.8	11	19.9 ± 2.0	10	Alpha spectrometry	-	**-5.5**	**-0.3**
	22.6 ± 2.1	9						
5	20.6 ± 1.5	7	-	-	Alpha spectrometry	-	**-1.9**	**-0.1**
7	23.9 ± 4	17	-	-	Direct gamma-spec.	-	**13.8**	**0.6**
9	19.6 ± 3	15	-	-	Direct gamma-spec.	-	**-6.7**	**-0.3**
13	19.8 ± 5	25	21.2 ± 3.6	17	Alpha spectrometry	-	**0.7**	**0.0**
	22.5 ± 2	9						
14	18.5 ± 1.8	10	-	-	From ^{228}Ac activity by gamma spec.	-	**-11.9**	**-0.8**
15	16.2 ± 2.1	13	16.6 ± 2.1	23	Alpha spectrometry	-	**-21.2**	-1.3
	16.9 ± 2	12						
18	21.8 ± 2.6	12	-	-	Alpha spectrometry	-	**3.8**	**0.2**
20	19.6 ± 2.2	11	-	-	Alpha spectrometry	-	**-6.7**	**-0.4**
22	24 ± 2.42	10	-	-	Gamma-spectrometry (^{228}Ac)	-	**14.3**	**0.8**
26	32 ± 4	13	-	-	Alpha spectrometry	-	**52.4**	**2.3**
27	19.8 ± 2.6	13	-	-	Alpha spectrometry	-	**-5.7**	**-0.3**

29	17.7 ± 2.06	12	18.3 ± 3.2	17	Direct-Gamma, assumed ^{232}Th in equilibrium with ^{228}Ac	-	-13.0	-0.7
	18.84 ± 4.36	23						
30	21.59 ± 2.58	12	22.0 ± 2.7	12	Direct gamma-spec.	-	4.8	0.3
	22.41 ± 2.8	12						
31	59.4213 ± 13.91	23	63.7 ± 14.9	23	Spectrophotometers measures	yes	203.5	2.8
	68.034 ± 15.91	23						
33	20 ± 1.3	7	-	-	Direct gamma-spec.	-	-4.8	-0.3
34	21.3 ± 4	19	-	-	Direct gamma-spec.	-	1.4	0.1
35	20.7 ± 3.58	17	21.4 ± 4.0	19	Direct gamma-spec.	-	1.9	0.1
	22.1 ± 4.48	20						
37	18.2 ± 1.8	10	-	-	Direct gamma-spec.	-	-13.3	-0.9
38	20.6 ± 3.4	17	21.8 ± 3.6	17	Direct gamma-spec.	-	3.6	0.2
	22.9 ± 3.8	17						
39	24.5 ± 2.3	9	-	-	Alpha spectrometry	-	16.7	1.0
40	21 ± 2	10	21.4 ± 2.1	10	^{288}Ac - direct gamma-spec.	-	1.9	0.1
	21.8 ± 2.2	10						
41	22.6 ± 2.6	12	23.1 ± 3.6	16	Direct gamma-spec.	-	9.8	0.5
	23.5 ± 4.6	20						
42	34.09 ± 3	9	-	-	Direct gamma-spec.	yes	62.3	3.2
43	0.1603 ± 0.084	52	-	-	Alpha spectrometry	yes	-99.2	-7.6
45	24.1 ± 1.7834	7	-	-	Direct gamma-spec.	-	14.8	1.0
46	21.3 ± 2.6	12	21.4 ± 2.6	12	Direct gamma-spec.	-	1.9	0.1
	21.5 ± 2.7	13						
48	19 ± 1.2	6	-	-	Direct gamma-spec.	-	-9.5	-0.7
49	14 ± 1.81	13	-	-	Alpha spectrometry	-	-33.3	-2.1
50	18.9 ± 3.1	16	-	-	Alpha spectrometry	-	-10.0	-0.5
51	21.3 ± 3.9	18	22.5 ± 4.1	18	Direct gamma-spec.	-	6.9	0.3
	23.6 ± 4.2	18						
56	21.3 ± 3.7	17	21.9 ± 4.8	22	Alpha spectrometry	-	4.3	0.2
	22.5 ± 6	27						
57	22.3 ± 1.8	8	22.5 ± 1.9	8	Direct gamma-spec.	-	6.9	0.4
	22.6 ± 2	9						
59	11.7 ± 0.5	4	-	-	Alpha spectrometry	-	-44.3	-3.4
61	19.19 ± 2.88	15	-	-	Direct gamma-spec.	-	-8.6	-0.5
62	12.6 ± 3.8	30	-	-	Alpha spectrometry	-	-40.0	-1.8
63	31 ± 15	48	-	-	Alpha spectrometry	-	47.6	0.7
67	22.3 ± 2.1	9	-	-	Alpha spectrometry	-	6.2	0.4
69	17.9 ± 1.9	11	-	-	Direct gamma-spec.	-	-14.8	-0.9
70	20.8 ± 1.8	9	20.9 ± 1.9	9	Direct gamma-spec.	-	-0.7	0.0
	20.9 ± 2	10						
72	13.65 ± 8	59	-	-	Direct gamma-spec.	-	-35.0	-0.9
73	23 ± 11	48	23.0 ± 7.9*	34	Direct gamma-spec.	-	9.5	0.2
	23 ± 10	43						

Table 11. Activity concentrations of ^{234}U with expanded uncertainties ($k = 2$), used methods and performance scores of individual laboratories.

Lab. code	Reported values		Arithmetic means		Used method	Outlier	$D_{\%}$ (%)	E_n
	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)				
1	22.5 ± 1.5	7	-	-	Alpha spectrometry	-	-10.7	-1.5
2	< 56	-	-	-	Direct gamma-spec.	-	-	-
3	19 ± 4	21	19.5 ± 4.0	21	Alpha spectrometry	-	-22.6	-1.4
	20 ± 4	20						
4	23.4 ± 2.1	9	23.8 ± 2.1	9	Alpha spectrometry	-	-5.6	-0.6
	24.2 ± 2	8						

5	28 ± 2	7	-	-	Alpha spectrometry	-	11.1	1.3
6	16.4 ± 4.8	29	-	-	Alpha spectrometry	-	-34.9	-1.8
9	28 ± 10	36	-	-	Alpha spectrometry	-	11.1	0.3
13	22 ± 6.6	30	25.5 ± 5.7	22	Alpha spectrometry	-	1.2	0.1
	29 ± 4.2	14						
15	22 ± 1.8	8	22.3 ± 1.6*	7	Alpha spectrometry	-	-11.5	-1.6
	22.6 ± 1.8	8						
18	31.4 ± 3.6	11	-	-	Alpha spectrometry	-	24.6	1.7
20	22.1 ± 3	14	-	-	Alpha spectrometry	-	-12.3	-1.0
22	18 ± 2.42	13	-	-	Alpha spectrometry	-	-28.6	-2.7
26	26 ± 4	15	-	-	Alpha spectrometry	-	3.2	0.2
27	27.2 ± 2.7	10	-	-	Alpha spectrometry	-	7.9	0.7
30	23.7 ± 4.8	20	-	-	Alpha spectrometry	-	-5.9	-0.3
33	24 ± 3	13	-	-	Alpha spectrometry	-	-4.8	-0.4
35	< 443	-	-	-	Direct gamma-spec.	-	-	-
	< 604	-						
39	35.3 ± 2.5	7	-	-	Alpha spectrometry	-	40.1	3.8
45	28.6 ± 8.9232	31	34.1 ± 10.6	31	Direct gamma-spec.	-	35.3	0.8
	39.6 ± 12.3552	31						
50	22.6 ± 3.9	17	-	-	Alpha spectrometry	-	-10.3	-0.6
52	20 ± 5.4	27	24.0 ± 6.5	27	ICP-MS	-	-4.8	-0.2
	28 ± 7.6	27						
54	27.7 ± 4.6	17	-	-	Alpha spectrometry	-	9.9	0.5
56	25.6 ± 5	20	27.9 ± 5.2	19	Alpha spectrometry	-	10.7	0.5
	30.2 ± 5.4	18						
59	21 ± 0.7	3	-	-	Alpha spectrometry	-	-16.7	-3.5
61	34.93 ± 7.68	22	-	-	Alpha spectrometry	-	38.6	1.3
62	17.2 ± 3.4	20	-	-	Alpha spectrometry	-	-31.7	-2.3
63	44 ± 6.8	15	-	-	Alpha spectrometry	yes	74.6	2.7
64	22.36 ± 2.88	13	22.6 ± 2.9	13	Alpha spectrometry	-	-10.2	-0.8
	22.89 ± 2.88	13						
67	23.3 ± 3.1	13	-	-	Alpha spectrometry	-	-7.5	-0.6
	24.5 ± 3	12			ICP-MS	-	-2.8	-0.2

Table 12. Activity concentrations of ^{235}U with expanded uncertainties ($k = 2$), used methods and performance scores of individual laboratories.

Lab. code	Reported values		Arithmetic means		Used method	Outlier	$D_{\%}$ (%)	E_n
	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)				
1	0.73 ± 0.25	34	-	-	Alpha spectrometry	-	-33.6	-1.3
2	3.6 ± 1.6	44	-	-	Direct gamma-spec.	-	227.3	1.6
3	1 ± 0.3	30	1.10 ± 0.35	32	Alpha spectrometry	-	0.0	0.0
	1.2 ± 0.4	33						
4	1.14 ± 0.45	39	1.77 ± 0.59	33	Alpha spectrometry	-	60.9	1.1
	2.4 ± 0.64	27						
5	2 ± 0.5	25	-	-	Alpha spectrometry	-	81.8	1.8
7	5.3 ± 1.8	34	-	-	Direct gamma-spec.	-	381.8	2.3
8	10.01 ± 2.43	24	10.1 ± 2.4	24	Direct gamma-spec.	yes	813.6	3.6
	10.09 ± 2.47	24						
9	1.5 ± 1	67	-	-	Alpha spectrometry	-	36.4	0.4
10	2.3 ± 0.6	26	-	-	Direct gamma-spec.	-	109.1	2.0
11	5.66 ± 0.86	15	5.88 ± 0.86	15	Direct gamma-spec.	-	434.5	5.5
	6.1 ± 0.86	14						
13	< 2	0	-	-	Alpha spectrometry	-	-	-
14	1.8 ± 1.8	100	-	-	Direct gamma-spec.	-	63.6	0.4
15	0.8 ± 0.32	40	0.99 ± 0.36	36	Alpha spectrometry	-	-10.5	-0.3

	1.17 ± 0.38	32						
17	12.43 ± 2.52	20	14.3 ± 2.3*	16	Direct gamma-spec.	yes	**1199**	**5.7**
	16.15 ± 2.66	16						
18	2.75 ± 0.82	30	-	-	Alpha spectrometry	-	**150.0**	**2.0**
20	0.81 ± 0.08	10	-	-	Alpha spectrometry	-	**-26.4**	**-2.1**
21	1.73 ± 0.58	34	1.74 ± 0.47*	27	Direct gamma-spec.	-	**57.7**	1.3
	1.74 ± 0.58	33						
22	1.1 ± 0.48	44	-	-	Alpha spectrometry	-	**0.0**	**0.0**
26	1.8 ± 0.8	44	-	-	Alpha spectrometry	-	**63.6**	**0.9**
27	0.9 ± 0.4	44	-	-	Alpha spectrometry	-	**-18.2**	**-0.5**
	3.8 ± 1.3	34			Direct gamma-spec.	-	**245.5**	**2.1**
32	1.6 ± 0.8	50	-	-	Direct gamma-spec.	-	**45.5**	**0.6**
33	1.2 ± 0.36	30	-	-	Alpha spectrometry	-	**9.1**	**0.3**
35	< 2.51	-	-	-	Direct gamma-spec.	-	-	-
	< 2.52	-						
37	< 4.95	-	-	-	Direct gamma-spec.	-	-	-
39	1.36 ± 0.19	14	-	-	Alpha spectrometry	-	**23.6**	1.2
40	1.9 ± 0.5	26	2.10 ± 0.55	26	Direct gamma-spec.	-	**90.9**	**1.8**
	2.3 ± 0.6	26						
45	1.84 ± 0.57408	31	-		Direct gamma-spec.	-	**67.3**	1.3
50	1.16 ± 0.45	39	-		Alpha spectrometry	-	**5.5**	**0.1**
52	1 ± 0.17	17	1.05 ± 0.18	17	ICP-MS	-	**-4.5**	**-0.2**
	1.1 ± 0.19	17						
54	1.74 ± 0.78	45	-	-	Alpha spectrometry	-	**58.2**	**0.8**
56	< 0.8	-	-	-	Alpha spectrometry	-	-	-
	< 2.6	-						
57	1.1 ± 0.24	22	1.15 ± 0.24	21	Direct gamma-spec.	-	**4.5**	**0.2**
	1.2 ± 0.24	20						
58	4.494 ± 0.836	19	-	-	Direct gamma-spec.	-	**308.5**	**4.0**
59	1.1 ± 0.1	9	-	-	Alpha spectrometry	-	**0.0**	**0.0**
60	< 18	-	-	-	Direct gamma-spec.	-	-	-
62	0.7 ± 0.2	29	-	-	Alpha spectrometry	-	**-36.4**	**-1.7**
63	2.1 ± 0.4	19	-	-	Alpha spectrometry	-	**90.9**	**2.4**
67	1.6 ± 0.61	38	-	-	Alpha spectrometry	-	**45.5**	**0.8**
	1.09 ± 0.12	11			ICP-MS	-	**-0.9**	**-0.1**
70	5.2 ± 2.6	50	-	-	Direct gamma-spec.	-	**372.7**	**1.6**
71	3 ± 1.8	60	-	-	Direct gamma-spec.	-	**172.7**	1.1
72	55.4 ± 9	16	57.0 ± 8.8	15	Direct gamma-spec.	yes	**5077**	**6.4**
	58.5 ± 8.5	15						

Table 13. Activity concentrations of ^{238}U with expanded uncertainties ($k = 2$), used methods and performance scores of individual laboratories.

Lab. code	Reported values		Arithmetic means		Used method	Outlier	$D_{\%}$ (%)	E_n
	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)				
1	17.2 ± 3.1	18	-	-	Direct gamma-spec.	-	**-29.2**	**-2.2**
	21 ± 1.5	7			Alpha spectrometry	-	**-13.6**	**-1.8**
2	28.5 ± 8.2	29	-	-	Direct gamma-spec.	-	**17.3**	**0.5**
3	18 ± 4	22	18.5 ± 4.0	22	Alpha spectrometry	-	**-23.9**	-1.4
	19 ± 4	21						
4	22.5 ± 2.1	9	22.9 ± 2.1	9	Alpha spectrometry	-	**-5.8**	**-0.6**
	23.3 ± 2.2	9						
5	28.9 ± 2	7	-	-	Alpha spectrometry	-	**18.9**	**2.1**
6	15.7 ± 4.2	27	-	-	Alpha spectrometry	-	**-35.4**	**-2.0**
7	< 100	-	-	-	Direct gamma-spec.	-	-	-
9	27 ± 10	37	-	-	Direct gamma-spec.	-	**11.1**	**0.3**

10	28.5 ± 7.6	27	-	-	Direct gamma-spec.	-	17.3	0.5
13	25.1 ± 7.2	29	26.3 ± 5.7	22	Alpha spectrometry	-	8.2	0.3
	27.5 ± 4	15						
14	39.7 ± 20	50	-	-	From ^{234}Th activity by gamma spec.	-	63.4	0.8
15	21.6 ± 1.8	8	22.1 ± 1.6*	7	Alpha spectrometry	-	-9.3	-1.2
	22.5 ± 1.8	8						
18	27.8 ± 8.6	31	-	-	Direct gamma-spec.	-	14.4	0.4
	32.6 ± 3.8	12			Alpha spectrometry	-	34.2	2.1
20	18.1 ± 2.6	14	-	-	Alpha spectrometry	-	-25.5	-2.2
22	16 ± 2.42	15	-	-	Alpha spectrometry	-	-34.2	-3.2
26	22 ± 4	18	-	-	Alpha spectrometry	-	-9.5	-0.6
27	23 ± 11	48	-	-	Direct gamma-spec.	-	-5.3	-0.1
	25.7 ± 2.6	10			Alpha spectrometry	-	5.8	0.5
30	23.2 ± 4	17	-	-	Alpha spectrometry	-	-4.5	-0.3
31	72.4521 ± 16.95	23	76.9 ± 18.0	23	Spectrophotometers measures	yes	216.6	2.9
	81.4342 ± 19.05	23						
32	24 ± 6	25	-	-	Direct gamma-spec.	-	-1.2	0.0
33	26 ± 5	19	-	-	Direct gamma-spec.	-	7.0	0.3
	25 ± 3	12			Alpha spectrometry	-	2.9	0.2
34	52 ± 40	77	-	-	Direct gamma-spec.	yes	114.0	0.7
35	< 153	-	-	-	Direct gamma-spec.	-	-	-
	< 80.3	-						
37	83.7 ± 48.8	58	-	-	Direct gamma-spec.	yes	244.4	1.2
39	34 ± 2.4	7	-	-	Alpha spectrometry	-	39.9	3.7
43	0.023 ± 0.012	52	-	-	Alpha spectrometry	yes	-99.9	-24.1
45	39.6 ± 12.3552	31	-	-	Direct gamma-spec.	-	63.0	1.2
46	20.6 ± 10.5	51	22.8 ± 10.4	45	Direct gamma-spec.	-	-6.2	-0.1
	25 ± 10	40						
50	21.7 ± 5.9	27	-	-	Alpha spectrometry	-	-10.7	-0.4
51	31.7 ± 12.7	40	36.9 ± 14.0	38	Direct gamma-spec.	-	51.6	0.9
	42 ± 15	36						
52	21 ± 3.6	17	23.0 ± 3.9	17	ICP-MS		-5.3	-0.3
	25 ± 4.3	17						
54	29.4 ± 4.8	16	-		Alpha spectrometry	-	21.0	1.0
56	25 ± 4.9	20	28.2 ± 5.2	19	Alpha spectrometry	-	15.8	0.7
	31.3 ± 5.5	18						
57	25.6 ± 3.6	14	26.0 ± 3.7	14	Direct gamma-spec.	-	7.0	0.4
	26.4 ± 3.8	14						
58	97.52 ± 18.138	19	-	-	Calculation from ^{238}U/^{235}U activity ratio	yes	301.3	4.0
59	19.9 ± 0.7	4	-	-	Alpha spectrometry	-	-18.1	-3.6
60	29 ± 8	28	-	-	Direct gamma-spec.	-	19.3	0.6
61	35.56 ± 6.42	18	-	-	Alpha spectrometry	-	46.3	1.7
62	16.3 ± 3.25	20	-	-	Alpha spectrometry	-	-32.9	-2.4
63	44.5 ± 6.9	16	-	-	Alpha spectrometry	-	83.1	2.9
64	22.41 ± 2.74	12	21.9 ± 2.8	13	Alpha spectrometry	-	-9.8	-0.8
	21.44 ± 2.78	13						
66	27.5 ± 6.2	23	28.0 ± 6.2	22	Gamma spec. via ^{234}Th	-	15.0	0.6
	28.4 ± 6.2	22						
67	24.1 ± 3.2	13	-	-	Alpha spectrometry	-	-0.8	-0.1
	23.6 ± 2.5	11			ICP-MS	-	-2.9	-0.3
70	23.4 ± 14	60	23.5 ± 14.1	60	Direct gamma-spec.	-	-3.3	-0.1

	23.6 ± 14.2	60						
71	24.5 ± 5.2	21	-	-	Direct gamma-spec.	-	**0.8**	**0.0**
72	30.35 ± 8	26	33.0 ± 6.7	20	Direct gamma-spec.	-	**35.9**	1.3
	35.71 ± 5	14						
73	63 ± 42	67	64.0 ± 32.7*	51	Direct gamma-spec.	yes	**163.4**	1.2
	65 ± 48	74						

Table 14. Activity concentrations of ^{238}Pu with expanded uncertainties ($k = 2$), used methods and performance scores of individual laboratories.

Lab. code	Reported values		Arithmetic means		Used method	Outlier	D$_{\%}$ (%)	E$_n$
	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)				
1	0.26 ± 0.04	15	-	-	Alpha spectrometry	yes	-	-
3	0.056 ± 0.01	18	0.063 ± 0.010	16	Alpha spectrometry	-	-	-
	0.07 ± 0.01	14						
4	0.57 ± 0.26	46	0.635 ± 0.285	45	Alpha spectrometry	yes	-	-
	0.7 ± 0.31	44						
5	0.04 ± 0.012	30	-	-	Alpha spectrometry	-	-	-
9	2.83 ± 0.9	32	-	-	Alpha spectrometry	yes	-	-
13	0.054 ± 0.028	52	0.069 ± 0.029	43	Alpha spectrometry	-	-	-
	0.084 ± 0.028	33						
15	33.5 ± 3.7	11	33.7 ± 4.2	12	Alpha spectrometry	yes	-	-
	33.8 ± 4.7	14						
20	0.034 ± 0.006	18	-	-	Alpha spectrometry	-	-	-
22	< 0.1	-	-	-	Alpha spectrometry	-	-	-
	0.096 ± 0.051	53						
27	< 0.5	-	-	-	Alpha spectrometry	-	-	-
37	< 0.28	-	-	-	Alpha spectrometry	-	-	-
	< 0.42	-						
43	0.1562 ± 0.032	20	-	-	Alpha spectrometry	-	-	-
48	< 0.025	-	-	-	Alpha spectrometry	-	-	-
	< 0.0387	-						
49	2.83 ± 0.53	19	-	-	?	yes	-	-
52	0.13 ± 0.042	32	0.135 ± 0.042	31	Alpha spectrometry	-	-	-
	0.14 ± 0.042	30						
54	0.13 ± 0.02	15	-	-	Alpha spectrometry	-	-	-
56	0.054 ± 0.015	28	0.059 ± 0.016	27	Alpha spectrometry	-	-	-
	0.063 ± 0.017	27						
59	0.13 ± 0.02	15	-	-	Alpha spectrometry	-	-	-
63	< 0.4	0	-	-	Alpha spectrometry	-	-	-
66	0.08 ± 0.04	50	0.083 ± 0.040	49	Alpha spectrometry	-	-	-
	0.085 ± 0.04	47						
67	0.064 ± 0.006	9	0.085 ± 0.020*	24	Alpha spectrometry	-	-	-
	0.106 ± 0.065	61						

Table 15. Activity concentrations of $^{239+240}$Pu with expanded uncertainties ($k = 2$).

Lab. code	Reported values		Arithmetic means		Used method	Outlier	D$_{\%}$ (%)	E$_n$
	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)	$A_{lab} \pm U_{lab}$ (Bq·kg^{-1})	$U_{\%}$ (%)				
1	0.95 ± 0.03	3	-	-	Alpha spectrometry	yes	-	-
3	0.28 ± 0.03	11	0.300 ± 0.030	10	Alpha spectrometry	-	-	-
	0.32 ± 0.03	9						
4	0.44 ± 0.16	36	0.460 ± 0.165	36	Alpha spectrometry	yes	-	-
	0.48 ± 0.17	35						
5	0.286 ± 0.03	10	-	-	Alpha spectrometry	-	-	-
6	0.25 ± 0.04	16	-	-	Alpha spectrometry	-	-	-

9	3.23 ± 0.88	27	-	-	Alpha spectrometry	yes	-	-
13	0.293 ± 0.082	28	0.307 ± 0.078	26	Alpha spectrometry	-	-	-
	0.321 ± 0.074	23						
15	100 ± 12	12	99.5 ± 10.2*	10	Alpha spectrometry	yes	-	-
	99 ± 10	10						
20	0.334 ± 0.04	12	-	-	Alpha spectrometry	-	-	-
22	0.28 ± 0.085	30	0.320 ± 0.092	29	Alpha spectrometry	-	-	-
	0.36 ± 0.097	27						
27	< 0.5	-	-	-	Alpha spectrometry	-	-	-
37	< 0.89	-	-		Alpha spectrometry	yes	-	-
	0.87 ± 0.5	57						
39	0.64 ± 0.06	9	-	-	Alpha spectrometry	yes	-	-
43	0.3294 ± 0.034	10	-	-	Alpha spectrometry	-	-	-
48	0.1983 ± 0.0762	38	0.244 ± 0.071*	29	Alpha spectrometry	-	-	-
	0.2905 ± 0.1188	41						
49	2.54 ± 1.34	53	-	-	?	yes	-	-
52	0.17 ± 0.056	33	0.220 ± 0.067	31	Alpha spectrometry	-	-	-
	0.27 ± 0.076	28						
54	0.26 ± 0.04	15	-	-	Alpha spectrometry	-	-	-
56	0.219 ± 0.041	19	0.225 ± 0.041	18	Alpha spectrometry	-	-	-
	0.231 ± 0.041	18						
59	0.26 ± 0.03	12	-	-	Alpha spectrometry	-	-	-
61	0.33 ± 0.08	24	-	-	Alpha spectrometry	-	-	-
62	0.3 ± 0.09	30	-	-	Alpha spectrometry	-	-	-
63	< 0.6	-	-	-	Alpha spectrometry	-	-	-
64	0.242 ± 0.1	41	0.249 ± 0.100	40	Alpha spectrometry	-	-	-
	0.256 ± 0.1	39						
66	0.28 ± 0.08	29	0.285 ± 0.076*	27	Alpha spectrometry	-	-	-
	0.29 ± 0.1	34						
67	0.272 ± 0.098	36	0.281 ± 0.040*	14	Alpha spectrometry	-	-	-
	0.289 ± 0.013	4						

Appendix 11: Additional radionuclides

Table 1: The activity concentrations of additional radionuclides reported by some participants. The uncertainties are expanded uncertainties ($k = 2$).

Radionuclide	Lab. code	Activity concentration ($Bq \cdot kg^{-1}$)
^{134}Cs	9	1.10 ± 0.20
	18	0.96 ± 0.22
	27	1.90 ± 0.40
	46	1.00 ± 0.40
	67	1.40 ± 0.40
	71	0.96 ± 0.22
^{241}Am	5	0.24 ± 0.05
	15	5.6 ± 0.9
^{234}Th	21	20.17 ± 6.49
^{210}Pb	2	25± 10
	9	27.3± 6
	15	25± 2
	18	29± 5
	21	27.08± 3.95
	32	36± 4
	33	26± 11
	57	28.2± 2.7
	69	424± 40
	71	29 ± 5
^{210}Po	31	530.25
^{228}Ra	20	19.5 ± 3.6
	32	21.5 ± 0.7
	63	22.2 ± 1.8
^{228}Ac	8	26.46 ± 1.924
	21	19.46 ± 2.11
	45	24.1 ± 1.422
	48	19 ± 12
^{228}Th	5	31 ± 4
	32	21.5 ± 0.7
	67	22.5 ± 2.1
^{208}Tl	2	7.1 ± 0.8
	48	5.6 ± 7
	66	7.1

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Abstract

A comparison was organised by the Institute for Reference Materials and Measurements among 73 environmental radioactivity monitoring laboratories for the determination of 15 radionuclides in soil. The reference material IAEA-375 Soil was reprocessed by IRMM to provide the comparison material and reference values traceable to SI units and SIR were established at IRMM. The analytical procedures used by participating laboratories are described. A robust evaluation of the performance of laboratories is performed using three different approaches: relative deviations, E_n numbers and PomPlots.
The performance of the participating laboratories varied depending on the radionuclide determined and method used. Gamma-ray spectrometry with respect to ^{137}Cs and ^{40}K is well controlled. The determination of ^{90}Sr proved difficult for about two-thirds of the participants, which submitted results outside the acceptable range. These laboratories need to review their analysis procedures. Several laboratories need to improve their analytical procedures for the uranium isotopes and ^{226}Ra. Moreover, the results for thorium isotopes are far from satisfactory mainly for the ^{230}Th. The use of the E_n criterion revealed that the uncertainty estimation of many participating laboratories is poor.

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