JRC Scientific and Technical Reports

Evaluation of EC comparison on the determination of ^{226}Ra, ^{228}Ra, ^{234}U and ^{238}U in mineral waters

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EUR 24694 EN - 2011

EUROPEAN COMMISSION

Institute for Reference
Materials and Measurements

The mission of the IRMM is to promote a common and reliable European measurement system in support of EU policies.

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JRC 62731

EUR 24694 EN
ISBN 978-92-79-19069-8
ISSN 1018-5593
DOI 10.2787/36764

Luxembourg: Publications Office of the European Union

Printed in Belgium

SUMMARY

In anticipation of new European requirements for monitoring radioactivity concentration in drinking water, IRMM organised this comparison. The report describes in detail all phases of the comparison for the determination of natural radionuclides in mineral waters among 45 European laboratories monitoring radioactivity in the environment and foodstuff. Commercial mineral waters were purchased as their activity concentration of natural radionuclides is higher (and, therefore, more reliably measurable) than in most drinking waters. Reference values traceable to SI units were established at IRMM and the homogeneity of the batch of distributed samples was demonstrated. The sample preparation and measurement processes applied in the participating laboratories are described and the results of the comparison are presented and discussed in detail. A small selection of these results was also presented at the most recent ICRM conference (Wätjen et al., 2010).

A robust evaluation of the performance of individual laboratories is performed using three different approaches: relative deviations, E_n numbers and 'POMPlots'. The comparison results show that there are many highly discrepant measurement results for the radium isotopes; about one fourth and more than one third of the laboratories have severe problems with the determination of ^{226}Ra and ^{228}Ra, respectively. For radium, 19 results, corresponding to 14 % of all, are even off by a factor of two or more. For uranium, this proportion is slightly better, with 6 % (9 results out of 150) off by a factor of two or more. In terms of the E_n criterion, still 14 % to 23 % of the uranium results are not compliant, and as many as 25 % to 35 % of all radium results are incompatible. These are problems which need to be addressed by the concerned laboratories.

As these samples had rather low activity concentrations – around the detection limits required by the draft EC directive – and not all laboratories are routinely analysing water for these radionuclides yet, unsatisfactory comparison results for ^{226}Ra and ^{228}Ra may not be unexpected. The comparison clearly demonstrates, however, that a number of monitoring laboratories need to improve their analysis procedures for radium in order to correctly identify drinking water sources for which remedial action (with respect to their natural radioactivity concentration) needs to be taken.

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GLOSSARY

BIPM	Bureau International des Poids et Mesures
CCRI(II)	Comité Consultatif des Rayonnements Ionisants, Section 2
DG	Directorate-General of the European Commission
GUM	Guide to the Expression of Uncertainty in Measurement [3]
IRMM	Institute for Reference Materials and Measurements
ICP-MS	inductively coupled plasma mass spectrometry
ICRM	International Committee for Radionuclide Metrology
ISO	International Organization for Standardization
LSC	liquid scintillation counter, liquid scintillation counting
SI	Système International d'Unités, International System of Units
SIR	Système International de Référence, International Reference System for radionuclides
UTC	Coordinated Universal Time
WHO	World Health Organization
A_{lab}	mean laboratory result of activity concentration
A_{ref}	reference value of activity concentration
D	difference between the reported and the reference activity concentration
E_n	performance statistic E_n number
k	coverage factor according to GUM
MAD	median absolute deviation
s	standard deviation
u	standard uncertainty according to GUM
u_c	combined standard uncertainty according to GUM
U_{lab}	expanded uncertainty of mean laboratory result
U_{ref}	expanded uncertainty of reference value

1. Introduction

The activity concentration of natural radionuclides in drinking water is quite variable, depending on the properties of the aquifer rock and the prevailing lithology. Whereas the radionuclide activity concentration in waters usually poses no health concern, there are regions in which the geological situation can render radioactivity levels which need to be monitored in order to reduce the potential health risk of the public. Concerns about the radioactivity concentration of water intended for human consumption have been described (L. Benedik et al., 2008, 2009), and a variety of regulations implemented (WHO, 1993, 2006; European Communities, 1998, 2001) to limit public exposure to radioactivity from drinking water. In anticipation of new requirements (European Communities, 2011) for monitoring radioactivity in drinking water, IRMM has organized this comparison among member state laboratories requesting the determination of low levels of activity concentrations (around the detection limits prescribed in the future legislation) of the naturally occurring radionuclides ^{226}Ra, ^{228}Ra, ^{234}U and ^{238}U in three commercially available mineral waters.

The European Atomic Energy Community (EURATOM) Treaty obliges the member states of the European Union to monitor (Art. 35) and regularly report (Art. 36) the levels of environmental radioactivity on their territory. Some details of sample taking and measurement requirements (sample types, sampling intervals, radionuclides, etc.) are regulated at European level (e.g. Commission Recommendation 2000/473/Euratom of 8 June 2000). In order to verify the quality and in particular comparability of the values reported by the member states, comparison exercises were introduced by the European Commission after the reactor accident of Chernobyl. Since 2003, the JRC Institute for Reference Materials and Measurements, IRMM, organises these comparisons in support to the Directorate-General for Energy, Unit Radiation Protection (DG ENER D.4).

The metrological approach of IRMM in conducting such comparisons using samples carrying reference values traceable to SI units and to the International Reference System for gamma–ray emitting radionuclides (SIR) was presented at a recent ICRM conference (Wätjen et al., 2008), and is sketched in Fig. 1. As member of the Consultative Committee for Ionising Radiation (CCRI), IRMM is participating in key comparisons among National Metrology Institutes (NMIs), which serve to "realise" the unit of radioactivity, the becquerel. Results of key comparisons, which are based on primary measurement techniques, i.e. without resorting to other activity standards, are introduced in the International Reference System for radionuclides (SIR) to determine the SIR calibration factor for that particular nuclide. Thus, the SIR system at BIPM, the Bureau International des Poids et Mesures in Sèvres close to Paris, is the world-wide standard for radioactivity.

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

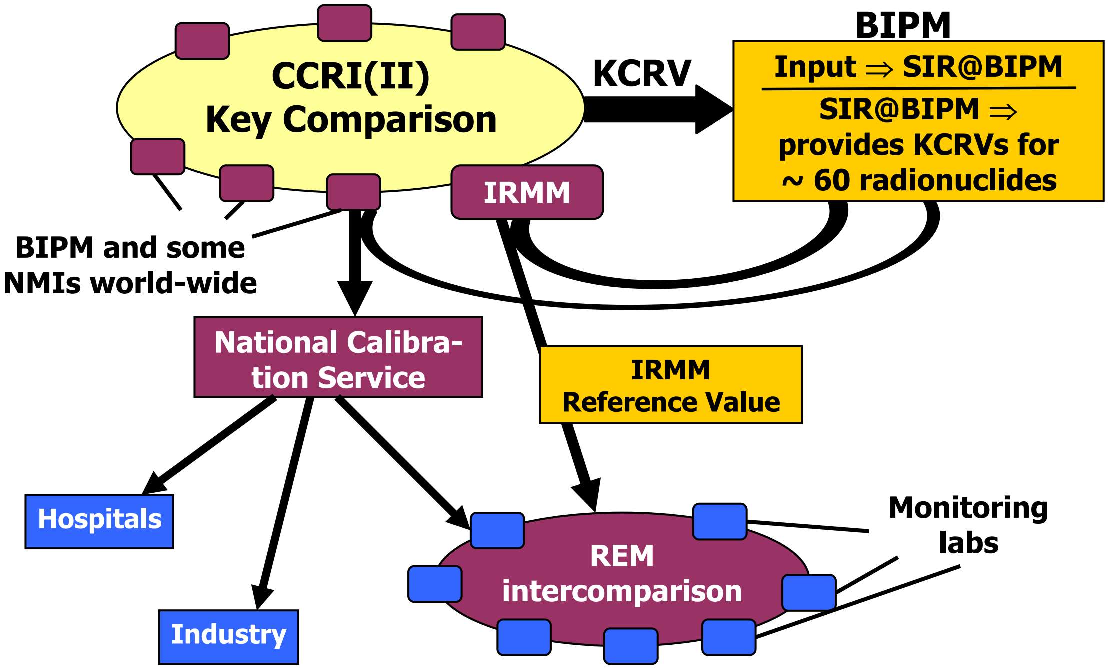

Fig. 1: Key comparisons of CCRI(II) and traceability of reference values for samples provided by IRMM for intercomparisons amongst monitoring laboratories (KCRV = key comparison reference value)

<u>Intercomparison material</u>

In order to cover a range of different radionuclide concentrations (well above the detection limits given in the future regulation on radionuclides in drinking water as well as close to them), mineral water from three different companies in Hungary was bought. All water samples were bottled in 1.5 L PET (polyethylene terephthalate) bottles. Whereas drinking water is falling under the European regulation, it was decided to use mineral waters in this comparison for practical reasons: next to its higher levels of radioactivity, mineral water can be purchased in larger batches bottled at the same time, thus offering an intrinsic homogeneity of the activity concentration. Furthermore, the determination of natural radioactivity was preferred over the analysis of spiked drinking water, since spiked samples were considered to differ too much from realistic water samples measured in the monitoring routine of the laboratories.

The companies were requested to produce a batch with bottled mineral water without any label or other indication on the origin of the water. Prior to bottling the water was treated as routinely done at the companies. No information on the routine treatment of the water was available from the producers. During bottling no precautions were taken to prevent adsorption of radionuclides on the walls of the bottles, e.g. by adding an acid. Chapter 2 of this report summarises the control measurements performed at IRMM in this respect.

Table 1 gives a list of the waters with sample identifications. Waters 2 and 3 were bought from one company, but in two separate deliveries of more than 2 months in between. Therefore, these two waters are divided into sub-batches having different sample numbers.

Table 1: Mineral water samples purchased for the comparison exercise

Sample ID	Sample name	Supplier	Sample number	Quantity
Water 1 - **W1**	Norbi Aqua	Buszesz Rt.	IM-RN-2006-02-001446	912 bottles
Water 2 – **W2**	Balfi	Pet-Pack Kft	IM-RN-2006-05-001448	504 bottles
			IM-RN-2006-07-001449	504 bottles
Water 3 – **W3**	Pannon Aqua	Pet-Pack Kft	IM-RN-2006-05-001450	504 bottles
			IM-RN-2006-07-001451	504 bottles
Water 4 – **W4**	Szentkiralyi	Szentkiralyi Kft	IM-RN-2006-06-001447	1008 bottles

Nominated and participating laboratories

Most of the laboratories (47) in this comparison were nominated to participate by the national representatives* in the expert group according to EURATOM Treaty Art. 35 and 36. These laboratories represent 24 of the 27 EU member states. In addition, four laboratories from three neighboring countries participated on a voluntary basis. All 51 laboratories were requested to fill in a questionnaire (Annex 1). Information was requested about the type and accreditation status of the laboratory, the methods it intended to use in the comparison, and the volume of water it expected to need for duplicate analysis of all radionuclides (see Question 8, Annex 1). Of the 51 laboratories, 48 sent back a filled in questionnaire. Four of the nominated laboratories declared that they were not able to participate, since their analysis methods were not sensitive enough for the low activity concentrations (gamma-ray spectrometry), or not specific for the determination of Ra-226 (precipitation method and proportional counting) or of the uranium isotopes (spectrophotometry). These four laboratories are not mentioned in the list of participating laboratories below.

According to their statements regarding the type of the laboratory, 18 laboratories do monitoring of the radioactivity in the environment (RE), 1 laboratory performs strictly Research and Development (R&D), 12 laboratories combine R&D with RE

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* They generally represent their national regulatory bodies for radiological protection.

measurements, 3 labs combine RE measurements with monitoring of nuclear facilities (MNF), 7 labs perform R&D, RE and MNF, 1 laboratory is an activity calibration lab, 2 laboratories do RE measurements together with other activities, and the last laboratory did not declare anything.

Regarding the status of accreditation, authorization or certification, the laboratories gave the following declarations:

- accredited – 11 laboratories with Quality Management System (QMS) according to ISO 17025;
- accredited, authorised and certified – 2 laboratories: 1 lab with QMS according to ISO 17025 and 1 lab with QMS according to ISO 9000 (!);
- accredited and authorised – 5 laboratories: 4 labs with QMS according to ISO 17025 and 1 lab without QS (!);
- accredited and certified – 3 laboratories with QMS according to ISO 17025 and ISO 9000;
- authorised and certified – 1 laboratory with QMS according to ISO 17025;
- certified – 2 laboratories: 1 lab with QMS according to ISO 17025 (!) and 1 lab without QMS (!)
- authorised – 14 laboratories: 5 without QMS (one with ISO 17025 in progress), 9 with QMS (one according to ISO 9000, one according to ISO 9000 and 17025, one with a “Home quality management system”, one according to “GLP with SOP” and 5 labs according to ISO 17025)
- no accreditation, authorization, or certification – 7 labs: 5 working under a QMS according to ISO 1705 and 2 labs without a QMS.

The laboratories were also asked how many measurements of this type they perform per year, and if the comparison samples were treated following the same analytical

Table 2: Number of laboratories performing this type of analysis more or less on a routine basis

Radionuclide	Number of measurements per year			
	Not declared	<25	25-100	>100
^{226}Ra	5	10	16	14
^{228}Ra	13	16	10	6
^{234}U	6	17	14	8
^{238}U	4	15	16	10

Table 3: Comparison samples treated with same procedure as in the routine

Radionuclide	Number of labs not declaring	Number of labs treating the sample in the same way	Number of labs NOT treating the sample in the same way
^{226}Ra	5	37	3
^{228}Ra	13	27	5
^{234}U	7	36	2
^{238}U	4	38	3

procedure as routinely used in the laboratory for the same type of samples. The answers are summarised in Tables 2 and 3.

The names and addresses of all nominated (and voluntary) participants are given in chapter 10 (except for the four laboratories which declared early that their methods did not allow them to participate). As this programme of comparisons requires anonymity, the identity of the participating laboratories is not shown in the compilation of the results. The order of the listing of participants in chapter 10 is not the same as the laboratory code number used throughout the data evaluation and presentation of comparison results.

Distribution of samples

According to the filled in questionnaires, the minimum required amount for the analyses of all four radionuclides was 4.5 L while the maximum volume was 30 L. The majority of the laboratories requested 10-20 L of mineral water. Subsequently, more than 1000 bottles of mineral water were dispatched from IRMM to the participating laboratories. In the comparison exercise only three kinds of water were used. Water 4 was not included in the comparison, because the activity levels of the radionuclides were far below the detection limits given in Annex III to the proposal for a Council Directive on radioactive substances in water intended for human consumption (European Communities, 2011).

Reporting of the results

The reporting was done through a WEB-based reporting system, consisting of two parts, the numerical results and a description of the procedures used (see annexes 2 and 3). The latter part also repeated the questions concerning laboratory type and accreditation status. Of the 51 nominated (and voluntary) laboratories, 45 reported measured values.

Time table and deliverables

November 2007	national representatives nominate the participating laboratories
November 2007	the nominated laboratories are requested to fill in a questionnaire to obtain an overview of used methods and to determine the water volume needed
December 2007	more than 1000 bottles of mineral water of three different kinds (W1, W2 and W3) are dispatched to the participating laboratories
29 February 2008	on-line registration
15 April 2008	deadline for submitting the results and questionnaire to IRMM
June 2008	preliminary results reported to the laboratories in the frame of a workshop held at IRMM
May 2009	provisional reference values sent to all participants by email

2. Homogeneity and stability

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, any inhomogeneity in the radionuclide concentration increases the uncertainty of the corresponding reference value. Likewise, any instability in an activity concentration would contribute additionally to the uncertainty of the corresponding reference value. Accordingly, the expanded uncertainty, U_{ref} (k = 2), of the reference value can be expressed as the quadratic sum of the three independent components

- combined standard uncertainty of the mean of measurement results contributing to the reference value (u_{char}),
- inhomogeneity among bottles of the whole batch (s_{bb}),
- short-term instability of samples for the duration of the comparison (u_{sts}).

More precisely, the expanded uncertainty is given by the formula

$$U_{\mathrm{ref}} = k \cdot \sqrt{u_{\mathrm{char}}^2 + s_{\mathrm{bb}}^2 + u_{\mathrm{sts}}^2} \quad . \qquad (1)$$

In order to determine the inhomogeneity s_{bb} among bottles of the whole batch, a dedicated homogeneity study was performed which is documented in a separate report (Spasova and Vasile, 2011). The results can be summarised as

- s_{bb} (^{226}Ra) are 3.5 % and 10 % in W1 and W2, respectively;
- s_{bb} (^{228}Ra) are 7.2 % and 5.0 % in W1 and W2, respectively;
- s_{bb} (^{234}U) are 7.0 %, 3.0 % and 2.0 % in W1, W3-50 and W3-51, respectively;
- s_{bb} (^{238}U) are 7.0 %, 4.0 % and 3.0 % in W1, W3-50 and W3-51, respectively.

The stability of the intercomparison samples was checked in the following way. Since during bottling no special treatment of the water was carried out, the adsorption of uranium on the walls of the bottles was checked as possible source of instability. This was done by filling empty bottles of mineral water with distilled water and addition of few mL of concentrated nitric acid. The bottles were stored for a period of at least 1 month. Then solid sources were prepared following the same radiochemical procedure as the one used for evaluating the homogeneity. The sources were counted for 4 days. The measured activity concentrations were below the detection limit (< 0.2 $\mathrm{mBq \cdot L^{-1}}$). Therefore, the samples were considered to be stable over time of the complete comparison exercise (u_{sts} = 0), and no uncertainty due to adsorption was taken into account when evaluating the combined uncertainty of the reference value.

The (in)homogeneity values, s_{bb}, mentioned above were taken into account in determining the uncertainty of the reference values (see chapter 3, pp. 48/49).

3. Determination of the reference values

The reference values for these comparison samples (activity concentration) were determined by consensus between two independent expert laboratories according to clause 5.5 of the standard ISO 13528 (ISO, 2005) with the additional constraint that the analyses had to be performed in such a way that the determined values are traceable to SI units and the SIR (Wätjen et al., 2008). IRMM and the Bundesamt für Strahlenschutz (BfS), Department for Radioprotection and the Environment, Berlin, as expert laboratories applied two largely different, independent methods (three in the case of ^{228}Ra).

Preparation, separation and measurement procedures used at IRMM and BfS

Determination of ^{234}U and ^{238}U

At IRMM, a known amount of ^{232}U tracer was added, for monitoring the chemical recoveries and correct results and to improve precision and accuracy. Uranium is coprecipitated with Fe(III) as $Fe(OH)_3$, acid leached (as a Cl-form) and isolated by anion exchange chromatography (Benedik et al., 2008). This fraction is converted to nitric form (with HNO_3) and U is separated and cleaned from other actinides using UTEVA resin. At the end uranium sources are prepared by coprecipitation with Nd in the fluoride form. The sample volume was 1.5 and 3 liters of mineral water. The samples were acidified using concentrated HCl to pH of approximately 1, a magnetic stirrer was used for a good homogenisation. We added Fe (III) carrier for uranium coprecipitation and also U-232 tracer for determination of recovery. All the time the samples were stirred and heated. The pH was adjusted near to 9 using NH_4OH and the precipitate was settled down overnight. The supernatant was decanted and the precipitate centrifugated. Again the supernatant was decanted.

Traceability of the ^{234}U and ^{238}U results to the International Reference System (SIR) for radioactivity measurement was ensured by primary standardization of the tracer nuclide ^{232}U with defined solid angle α-particle counting (Sibbens et al., 2004) and subsequent submission to the SIR following appropriate procedures described elsewhere (Ratel, 2007).

At BfS, the determination of uranium isotopes was performed with proven methods of radiochemical separation and activity measurement (Bundesminister, 2006). Whereas the radiochemical separation by extraction chromatography and the use of ^{232}U as a tracer is very similar to the method used at IRMM, the preconcentration consisted of evaporating water samples of 1 L to dryness and wet ashing of the residues. The thin sources for α-particle spectrometric measurement were prepared by electrodeposition on stainless steel discs (Spasova et al., 2009).

Determination of ^{226}Ra

At IRMM an analytical scheme adapted from Lozano et al.(1997) was applied.

The method is specified for the determination of ^{226}Ra in water with activity (drinking water, rain water, ground water and surface water) in the following range: 0.01 Bq/L to 10 Bq/L. In certain cases, the range of application may be changed by variations in the working conditions (e.g. sample volume, pre-concentration techniques, sensitivity ranges of detectors, etc.).

In the case of ^{226}Ra measurement by alpha spectrometry, the sample requires extensive chemical separation prior to counting in order to remove peak interferences from other alpha emitters.

The procedure is based on coprecipitation of radium as $Pb(Ra)(Ba)SO_4$ from acidified water samples between 0.7 L and 3 L. Adding, together with barium carrier, a known amount of ^{133}Ba as tracer for the chemical yield of radium and, furthermore, Pb^{2+} allowed to coprecipitate radium and barium with lead sulfate. Dissolution of the precipitate with EDTA/NAOH and micro-coprecipitation of $Ba(Ra)SO_4$ at pH = 4.5 led to a suspension which was filtered (polypropylene), whereas Pb^{2+} ions remained in solution. The filter, mounted on a stainless steel disc, was measured by α-particle spectrometry to determine ^{226}Ra and by γ-ray spectrometry for the ^{133}Ba yield. A chemical recovery between 74 % and 98 % was obtained.

At BfS, the standardized radon emanation method H-Ra-226-TWASS-01-1 (Bundesminister, 2006) was applied. Radium was preconcentrated by coprecipitation with $BaSO_4$ from water samples of 1 L, using the natural Ba carrier addition as a tracer by determination with atomic absorption spectroscopy. The precipitate was dissolved in $EDTA/NH_4OH$ and transferred to a radon bubbler. The sample was de-emanated and stored for a minimum of 14 days to allow ^{222}Rn to grow in. The grown-in ^{222}Rn was then transferred to a Lucas-type scintillation chamber, where the α-emission of ^{222}Rn and its short-lived decay products were counted. The system was calibrated with a ^{226}Ra standard solution.

Determination of ^{228}Ra

At IRMM, two independent methods were applied.

The first method is based on the chemical separation procedure described by Nour et al. (2004).

Sample preparation

Two types of bottled mineral water were analysed, W1 and W2. Because of the low activity concentration of ^{228}Ra in the waters under investigation, volumes of 1.5 L and 3 L were used for each single determination. Pre-concentration of ^{228}Ra was done using MnO_2 co-precipitation. The pH was adjusted to 2 with concentrated hydrochloric acid and then a known amount of ^{133}Ba tracer was added to be used for determination of the chemical yield of the radiochemical separation procedure. Depending on volume of the water taken for the analysis, precipitation of radium with MnO_2 was done by adding 0.375 or 0.75 mL of saturated solution of $KMnO_4$ and 0.75 or 1.5 mL of 0.5M $MnCl_2$, and adjusting the pH to 8 - 9 with 2M NaOH. The sample was stirred for two hours and then left overnight to allow the precipitate to settle. The supernatant was decanted, the MnO_2 precipitate was centrifuged for 5 minutes at 3500 rpm and the precipitate was dissolved in the mixture of concentrated HCl and H_2O_2.

After the pre-concentration of radium isotopes from the mineral water samples with MnO_2, ^{228}Ra was separated applying the following procedure.

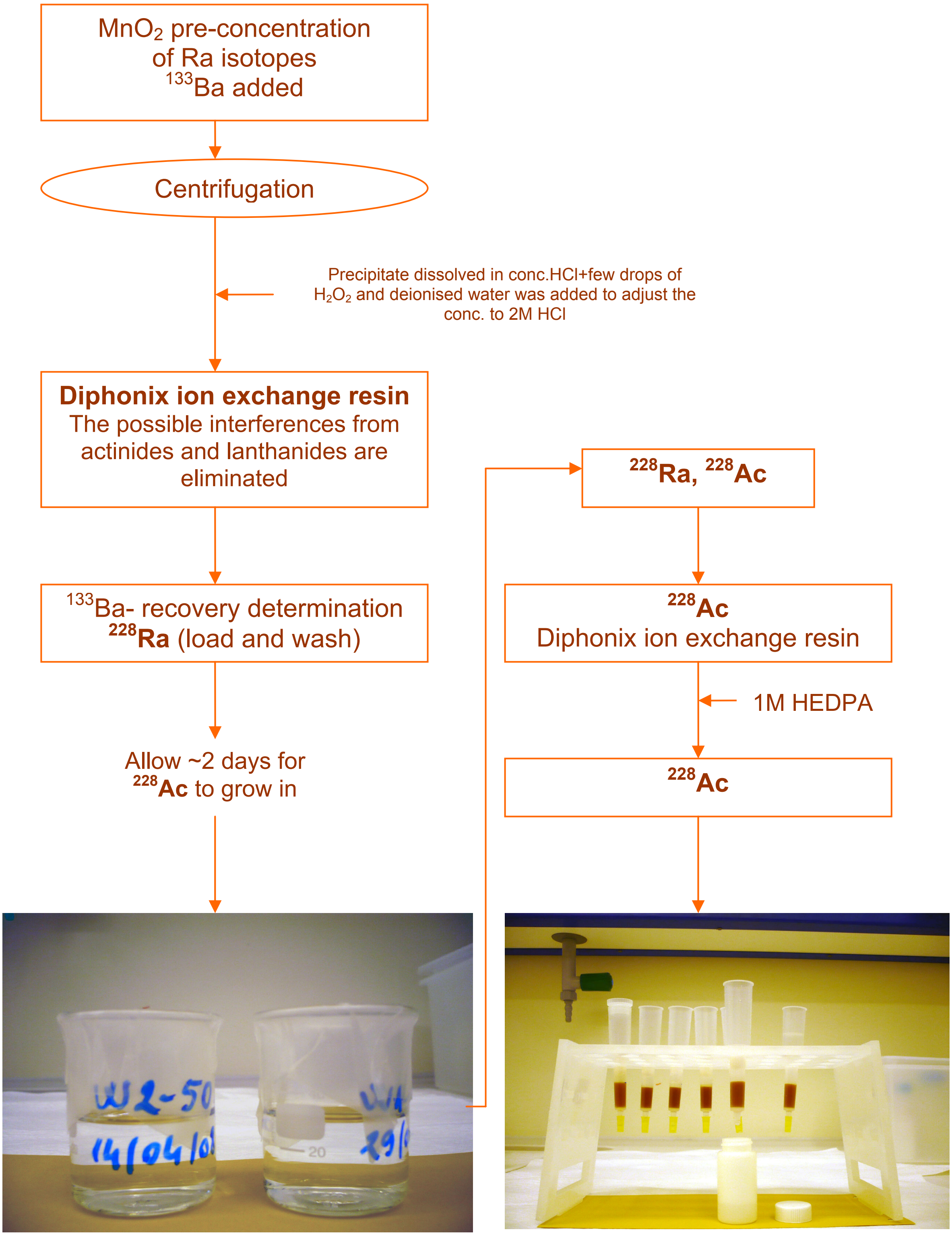

Fig. 2: Radiochemical separation of radium and ^{228}Ac from mineral water

After dissolving the precipitate with concentrated HCl and H_2O_2, de-ionised water was added to adjust to 2M HCl and the sample was ready to be loaded onto the first Diphonix resin column. The resin column (0.5 cm i.d., 15 cm long) was conditioned with 10 mL of 2M HCl, then the sample was loaded onto the column and the non-retained ions (Ra, Ba) were eluted with 15 mL of 2M HCl. The eluted fractions were collected and stored for approximately two days to allow ^{228}Ac to reach equilibrium with its parent ^{228}Ra. A second Diphonix resin column (0.8 cm i.d., 2.3 cm long), conditioned with 10 mL 2M HCl, was used for the separation of ^{228}Ac, which was eluted with 5 mL of 1M of 1-hydroxyethane-1,1-diphosphonic acid (HEDPA) in a polyethylene liquid scintillation vial. A blank was prepared in the same way as the samples using the same reagents and deionised water.

For the second method of IRMM, use was made of sources prepared a year earlier for the determination of ^{226}Ra by α-particle spectrometry as described above. The basic idea is to separate and determine by α-particle spectrometry the ^{228}Th grown in as granddaughter nuclide of ^{228}Ra after more than a year. The Ba(Ra)SO_4 precipitates of the existing sources were dissolved with EDTA/NaOH and ^{229}Th added as tracer for determination of the chemical recovery. Coprecipitation with $Fe(OH)_3$, extraction chromatography on TEVA columns and micro-coprecipitation with CeF_3 were applied as preconcentration, separation and source preparation steps. The procedure is described in detail in Vasile et al. (2009).

The BfS laboratory followed the standard procedure H-Ra-228-TWASS-01-1 (Bundesminister, 2006). Adapted from Burnett et al. (1995), radium and actinium were preconcentrated by coprecipitation with barium sulfate, the sulfate was converted to a more soluble carbonate, and – after a waiting time of at least 30 hours – the grown-in ^{228}Ac was separated by extraction chromatography on RE Spec resin. Sources were prepared using micro-coprecipitation of actinium on cerium fluoride, immediately followed by β-particle measurement in a low-level proportional counter. The chemical yield of the radium separation was determined via atomic absorption spectrometry of the barium carrier. The proportional counter was calibrated with a standardised ^{89}Sr source.

^{226}Ra, ^{234}U and ^{238}U measurements at IRMM

Alpha-particle spectrometry equipment and calibration

The experimental set-up used for ^{226}Ra, ^{234}U and ^{238}U measurements is an alpha spectrometry system for environmental samples which consists of Canberra alpha spectrometer chambers (Model 7401 VR) and PIPS detectors with 450 mm^2 sensitive area.

The measured source was deposited on a flat substrate that was placed in a parallel plane, centred at the symmetry axis of the detector at a distance (varying a bit amongst chambers) of about (5.0 ± 0.5) mm.

Each source was measured 4 days (345600 s); two measurements per source were performed. The data acquisition and analysis was done using the Genie-2000 Alpha

Analysis Software. For the analyses of ^{226}Ra also Alpha Analyst Control Application V.2.1 was used.

The energy calibration of the alpha chambers was performed by measuring single nuclide sources and a mixed source. Each source was measured for 30 minutes and the sum spectrum was used for performing the calibration. The list of the sources and the energy lines used for the energy calibration of the alpha-spectrometry system is given in Table 4.

Table 4: Sources used for the energy calibration of alpha-particle spectrometers

Source	Nuclide	Energy, keV*
IRMM 7.4.89-5	^{243}Am	5275.3
IRMM 310395-1	^{233}U	4824.2
IRMM 070292-1	^{148}Gd	3183$^{\#}$
	^{238}Pu	5499.03
	^{244}Cm	5804.96

* Nucléide (2004)

$^{\#}$ Predefined nuclear library in the software of Canberra

Activity concentration determination and uncertainty estimation (IRMM)

^{226}Ra

The activity concentration of ^{226}Ra ($A_{Ra\text{-}226}$) was calculated by:

$$A_{Ra-226} = \frac{N_{Ra-226}}{t_{Ra-226} \cdot \varepsilon \cdot V_{sample} \cdot R_{chem}} \cdot f_{decay} \qquad (2)$$

where $N_{Ra\text{-}226}$ is the net peak area of ^{226}Ra; $t_{Ra\text{-}226}$ is the counting time of the sample; ε is the detection efficiency; V_{sample} is the sample volume; R_{chem} is the radiochemical yield of the sample; f_{decay} is a correction factor for decay of ^{226}Ra (including decay correction to reference date and decay during measurement).

The half-life of ^{226}Ra used for the calculations is (1600 ± 7) years (Nucléide, 2004).

Uncertainty of ^{226}Ra determination

Uncertainty due to sample preparation and chemical recovery

For the determination of the chemical yield of the radiochemical procedure ^{133}Ba was used as a tracer (see section "Sample preparation"). Its activity concentration in the sample was determined by γ-ray spectrometry with a high-purity germanium detector (HPGe) with an endcap having a 0.15 mm thick Be window. The γ-ray peak analysis was done with the GammaVision-32 software.

The chemical yield was determined by relative measurements based on the comparison of the measured peak areas of the ^{133}Ba tracer in the water samples ($N_{Ba\text{-}133sample}$) and a ^{133}Ba standard source ($N_{Ba\text{-}133Std}$) that was prepared in the same manner. The recovery factor of the standard ($R_{Ba\text{-}133Std}$) was determined by measuring the activity left in the filtrate and the washing solution after the filtration of $BaSO_4$.

The chemical yield R_{chem} was calculated from:

$$R_{chem} = \left(\frac{N_{Ba-133sample}}{t_{Ba-133sample} \cdot m_{Ba-133sample}} \right) \cdot \left(\frac{t_{Ba-133Std} \cdot m_{Ba-133Std}}{N_{Ba-133Std}} \right) \cdot R_{Ba-133Std} \qquad (3)$$

in which N_{Ba-133} is the net area of ^{133}Ba in the sample and the standard source, respectively; t_{Ba-133} is the counting time of the sample and the standard source; m_{Ba-133} is the mass of added ^{133}Ba in the sample and in the barium standard source, respectively; and $R_{Ba-133Std}$ is the recovery factor of the standard source.

In evaluating the uncertainty due to the sample preparation and the chemical yield, the contributions of the weighing of the tracer, and peak area determination have to be taken into account. Also the use of ^{133}Ba for yield correction must be taken with caution, as one may suspect a different microscopic chemical behaviour between radium and barium. It has been shown in literature, Lozano et al.(1997), that the yield ratio Y_{Ra-226}/Y_{Ba-133} is slightly higher than 1 (≈1.04), with a typical uncertainty of 8 %. As this value is taken from the literature, it leads to a 4.6 % standard uncertainty contribution (with a rectangular distribution as type B uncertainty (GUM, 1995)) to the combined uncertainty. This is the major uncertainty component in the chemical procedure.

A squared sum was taken of the independent components due to the sample preparation and the chemical recovery, including uncertainty on the ^{133}Ba standard (2 %), weighing (1 %), counting uncertainty (1 %) and yield ratio uncertainty (4.6 %), leading to a 5.2 % combined standard uncertainty for the separation and chemical yield.

Uncertainty on the solid angle

The detection efficiency is obtained directly from the relative solid angle, i.e. the ratio of the solid angle to 4π steradian. The solid angle Ω of the measured sources depends on dimensional parameters such as the source-detector distance d, the detector radius R_D, the source radius R_s and eccentricity of the source. Also, the activity distribution within the sources is very important in estimating the uncertainty on the solid angle (Spasova et al., 2007).

Realistic sources are somewhat inhomogeneous and out-of-centre. Autoradiographs were taken of sixteen ^{226}Ra sources, in order to assess their activity distribution (Sibbens et al., 2003). The sources were not as homogeneous and of reproducible geometry as would be desired. The activity was sometimes concentrated in the centre or in a 'moon shaped' structure. Variation of 3 % in the calculated Ω ($s(\Omega)$ = 3 %) was estimated, which is completely due to a different activity distribution within the sources.

The eventual value of the source-to-detector distance d was evaluated from the measurement of a ^{226}Ra reference source with known activity (source ID: STD-11) and similar geometry, and taking into account possible variation amongst sources. The source was made by mounting a known amount of calibrated ^{226}Ra solution and ^{133}Ba solution, as a sulphate in the same manner as the samples were prepared. It was standardised at IRMM by defined solid angle counting and its activity was 12.0 Bq ± 0.2 % (k = 1). The uncertainty of the distance d (≈ 5 mm) was estimated as

2 %. The uncertainty caused by a possible tilt of the source or detector is considered to be negligible compared to the other uncertainty components.

A squared sum was taken of the independent components due to the radial activity distribution (including source inhomogeneity, internal eccentricity and source radius) (3 %), distance *d* (1.2 %), detector radius and tilt (0.5 %), external eccentricity of 1 mm (1 %), leading to a combined standard uncertainty of 3.4 % for the solid angle.

Uncertainty due to spectral deconvolution

The data acquisition and analysis was done using the Genie-2000 Alpha Analysis Software and Alpha Analyst Control Application V.2.1. The fit was restricted to the energy region 3.7 MeV to 4.8 MeV around the ^{226}Ra peak, hereby not fully subtracting the tailing of the daughter peaks situated at higher energy. The residuals of the fit showed systematic structure, whereas for the uncertainty calculations to be valid it is required that the residuals are randomly distributed (S. Pommé, 2006a).

As an alternative to the fit, the net peak area ($N_{Ra\text{-}226}$) was calculated by taking the numerical integral of the number of counts in the peak region (N_g) instead and correcting for background (B) and subtracting the low-energy tail of the interfering peak at 5.49 MeV (T):

$$N_{Ra-226} = N_g - T - B \tag{4}$$

A tail-to-peak ratio of $T/N_{Ra\text{-}226} \sim 0.05$ was derived from the well isolated peak at the end of the spectrum (7.68 MeV) and this ratio was applied to subtract the tailing from the 4.78 MeV peak. The uncertainty was calculated from the sum of the counting uncertainties (Poisson) and an additional, estimated 10 % uncertainty on the tailing:

$$u(N_{Ra-226}) = \sqrt{N_g + B + T + (T/10)^2} \tag{5}$$

which is only slightly higher than the uncertainty delivered by the software. In the case of Water 1 (W1), a typical relative uncertainty is 1 % to 1.5 %.

Uncertainty budget for ^{226}Ra activity concentration

In the uncertainty budget for the determination of ^{226}Ra in mineral water (Table 5), the typical standard uncertainties (1 *s*) for a single measurement are given. The individual uncertainty contributions are combined by taking the square root of the sum of the squared uncertainty components. The major contributions come from the sample preparation and the chemical recovery (5.2 %) and the solid angle determination (3.4 %). The uncertainties due to system dead time, counting time and decay correction (i.e. half-life) are considered to be negligible compared to the other uncertainty components (in total < 0.1% of the combined uncertainty). In the example presented (Water 1 – W1), the uncertainty on the counting statistics is a minor uncertainty contribution (1.3 %).

Table 5: Uncertainty budget for ^{226}Ra in mineral water sample W1. The uncertainty budget shows the standard uncertainties (1 *s*) for a single measurement. The combined standard uncertainty is the quadratic sum of all components.

Uncertainty component	**Uncertainty ^{226}Ra (%)**
Chemical yield	5.2
Solid angle	3.4
Counting statistics (incl. background)	1.3
Volume of sample	0.1
Counting time	0.005
Dead time	0.005
Half-life ($T_{1/2}$ = 1600 a)	9.3E-05
Combined standard uncertainty u_c	**6.4**
Expanded uncertainty U (k = 2)	**12.7**

^{234}U and ^{238}U

The activity concentrations A_U of ^{234}U and ^{238}U, respectively, were determined by relative measurements based on the comparison of the measured peak areas of these uranium isotopes (N_U) and the ^{232}U tracer (N_{tracer}):

$$A_U = \frac{N_U \cdot m_{tracer} \cdot A_{tracer}}{N_{tracer} \cdot V_{sample}} \cdot f_{decay} \qquad (6)$$

where N_U and N_{tracer} are the net peak areas of the corresponding uranium isotopes; A'_{tracer} is the activity per mass of the tracer solution; m_{tracer} is the mass of the added ^{232}U solution in the sample; V_{sample} is the sample volume; f_{decay} is a correction factor for decay of the uranium isotopes (including decay correction to reference date and decay during measurement).

The net peak area (N) was calculated by taking the numerical integral of the number of counts in the peak region (N_g) and correcting for background (B):

$$N = N_g - B \qquad (7)$$

The half-lives used for the calculations are (245.5 ± 0.6)·10^3 years for ^{234}U, (4.47 ± 0.02)·10^9 years for ^{238}U and (70 ± 1) years for ^{232}U (Nucléide, 2004).

Uncertainty of uranium determination

Table 6 shows a typical uncertainty budget of the ^{234}U and ^{238}U activity concentrations for a single measurement at the 1 *s* level (Spasova et al., 2009). The first uncertainty contributions concern counting statistics in the ^{234,238}U peaks and the tracer peak calculated from the number of gross counts N_g and the background counts B:

$$u(N) = \sqrt{N_g + B} \qquad (8)$$

Also the choice of the regions of interest (ROI) around the analysed peaks plays a role. The corresponding uncertainty was estimated to be 2 %, and taken as a systematic uncertainty, which does not reduce with repeated measurements.

With respect to sample preparation, a ^{232}U tracer solution standardised at IRMM within 0.6 % by defined solid angle counting was used (Sibbens et al., 2004). Furthermore, the estimated uncertainty on the volume of the sample and the uncertainty on the weight of the tracer solution is 0.1 % and 0.5 %, respectively. The propagated uncertainty due to the decay correction factor of the different isotopes (i.e. half-life) is negligible.

The uncertainty on the chemical yield cannot be easily determined. The uranium isotopes in the sample and as tracer should have the same chemical behaviour and, therefore, the same yield. Nevertheless, one cannot completely exclude that the uranium in the mineral water could behave differently from the tracer because of matrix effects; for example, if they were encapsulated in silica grains which are not easily dissolved. At this stage no uncertainty component is added for such possible effects.

Table 6: Uncertainty budget for ^{234}U and ^{238}U in mineral water sample W3-50, giving the standard uncertainties (1 *s*) for a single measurement. The combined standard uncertainty is the quadratic sum of all components.

Uncertainty component	**^{234}U (%)**	**^{238}U (%)**
Counting statistics (incl. background)	2.0	2.5
Counting statistics tracer (incl. background)	1.0	1.0
Choice of ROI	2.0	2.0
Activity of tracer	0.6	0.6
Weighing of tracer	0.5	0.5
Volume of sample	0.1	0.1
Half-life	8.0E-07	8.0E-07
Combined standard uncertainty u_c	**3.2**	**3.5**
Expanded uncertainty *U* (*k* = 2)	**6.4**	**6.9**

The individual uncertainty contributions are combined as quadratic sum. The major contributions come from the counting statistics and the choice of region of interest (ROI, 2 %). The uncertainty due to sample preparation (sample volume, weight and activity of tracer solution) contributes 0.8 % to the combined standard uncertainty.

Results for ^{226}Ra, ^{234}U and ^{238}U (IRMM)

The results from the activity concentration measurements of ^{226}Ra, ^{234}U and ^{238}U in mineral water are presented in the tables and figures below. The uncertainties were estimated according to the GUM (1995) and their components are approximations of the corresponding relative standard deviations for typical measurement parameters (Tables 5 and 6).

Data were corrected for:

- Decay – reference date 1 May 2006, 0:00 h UTC;
- Decay during measurement;
- Dead time (automatically made by the counter) in case of ^{226}Ra.

The activity concentration values obtained for the three studied radionuclides according to the sample volume used are given in Tables 7 to 21. In the tables, the results for each sample as well as the arithmetic mean and the weighted mean are given for all measurements. In addition, the corresponding uncertainties, the absolute and relative standard deviations are presented as well.

During the ^{234}U and ^{238}U analyses, a difference between the two sub-batches of Water 3 (W3) indicated as W3-50 and W3-51 (IM-RN-2006-05-001450 and IM-RN-2006-07-001451) was observed. The difference was almost 10 %. Therefore, these two sub-batches were analysed separately and results are given for each of them. Furthermore, it has to be noted that in all the batches both uranium isotopes are not in equilibrium and the activities measured for ^{238}U are lower than those measured for ^{234}U.

The results from all prepared sources for each radionuclide measured with their corresponding combined standard uncertainties u_c (as given in Tables 5 and 6) are plotted in Figures 3-13. In the figures, the red solid horizontal line indicates the mean and the dashed lines indicate the ± 1 *s* range.

The mean activity concentrations of ^{226}Ra, ^{234}U and ^{238}U in mineral water were calculated as weighted means of the results from all prepared sources (0.7/0.8 L, 1.5 L and 3 L sample volume). Tables 13, 22 and 23 summarise the basic characteristics of the distribution of all measurement results.

Table 7: Measured activity concentrations of ^{226}Ra (mBq·L^{-1}) in sample batch 1 (W1 - IM-RN-2006-02-001446). Sources prepared from a volume of 0.7 L to 0.8 L.

Sample ID	**Measurement 1 (mBq·L^{-1})**		**Measurement 2 (mBq·L^{-1})**		**Weighted mean value (mBq·L^{-1})**	
	Act. conc.	u_c	**Act. conc.**	u_c	**Act. conc.**	u_c
W1-12	98.10	6.25	96.32	6.13	97.17	6.11
W1-13	90.71	5.80	84.53	5.41	87.52	5.52
W1-15	98.40	6.27	97.93	6.25	98.17	6.18
W1-31	91.89	5.84	90.88	5.78	91.39	5.74
W1-32	94.69	6.03	95.49	6.08	95.08	5.98
W1-33	94.64	6.01	91.77	5.81	93.06	5.84
W1-34	101.35	6.45	99.48	6.33	100.38	6.31
W1-35	89.91	5.71	89.66	5.70	89.79	5.64
W1-36	94.74	6.03	95.02	6.03	94.89	5.96
W1-37	94.92	6.04	96.04	6.12	95.46	6.00
W1-38	92.10	5.86	94.54	6.02	93.28	5.86
W1-39	101.91	6.47	106.82	6.78	104.29	6.55
W1-40	98.56	6.28	99.52	6.35	99.03	6.23
W1-41	103.75	6.58	96.29	6.09	99.59	6.25
W1-42	98.00	6.24	94.82	6.05	96.42	6.07
W1-43	97.33	6.20	100.12	6.37	98.73	6.21
W1-44	104.78	6.67	100.45	6.39	102.52	6.44
Mean	96.81	6.02	95.86	5.95	96.28	5.99
Weighted mean	96.52	6.00	95.51	5.94	95.90	5.96
Minimum	89.91		84.53		87.52	
Maximum	104.78		106.82		104.29	
St. dev. (abs)	4.44		5.02		4.44	
Rel. st. dev. (%)	4.59		5.23		4.61	

Table 8: Measured activity concentrations of ^{226}Ra (mBq·L^{-1}) in sample batch 1 (W1 - IM-RN-2006-02-001446). Sources prepared from a volume of 1.5 L.

Sample ID	**Measurement 1 (mBq·L^{-1})**		**Measurement 2 (mBq·L^{-1})**		**Weighted mean value (mBq·L^{-1})**	
	Act. conc.	u_c	**Act. conc.**	u_c	**Act. conc.**	u_c
W1-20	104.19	6.57	111.32	7.01	107.69	6.74
W1-21	101.56	6.39	106.82	6.72	104.11	6.51
W1-22	101.23	6.37	99.08	6.23	100.12	6.26
W1-23	96.30	6.07	97.27	6.12	96.82	6.05
W1-24	103.76	6.52	105.02	6.60	104.37	6.52
W1-25	100.88	6.34	99.09	6.23	99.97	6.25

W1-26	104.68	6.60	107.18	6.76	105.90	6.63
W1-27	93.71	5.90	100.33	6.31	97.10	6.07
W1-29	95.05	5.99	97.72	6.16	96.34	6.03
W1-45	98.21	6.17	105.48	6.63	101.47	6.34
Mean	99.96	6.22	102.93	6.40	101.39	6.30
Weighted mean	99.74	6.20	102.51	6.38	101.11	6.29
Minimum	93.71		97.27		96.34	
Maximum	104.68		111.32		107.69	
St. dev. (abs)	3.93		4.83		4.01	
Rel. st. dev. (%)	3.93		4.69		3.99	

Table 9: Measured activity concentrations of ^{226}Ra (mBq·L^{-1}) in sample batch 2 (W2 - IM-RN-2006-05-001448 / IM-RN-2006-07-001449). Sources prepared from a volume of 0.7 L to 0.8 L.

Sample ID	**Measurement 1 (mBq·L^{-1})**		**Measurement 2 (mBq·L^{-1})**		**Weighted mean value (mBq·L^{-1})**	
	Act. conc.	u_c	**Act. conc.**	u_c	**Act. conc.**	u_c
W2-24	43.07	2.80	41.98	2.72	42.48	2.70
W2-25	38.52	2.54	40.35	2.66	39.40	2.52
W2-26	40.39	2.64	38.24	2.50	39.25	2.50
W2-27	42.59	2.82	40.77	2.70	41.63	2.67
W2-30	43.16	2.80	41.08	2.68	42.12	2.68
W2-31	37.17	2.45	39.09	2.58	38.06	2.44
W2-32	41.32	2.69	42.37	2.76	41.83	2.66
W2-33	41.46	2.73	40.13	2.64	40.77	2.61
W2-34	40.89	2.67	39.27	2.57	40.07	2.55
W2-35	42.98	2.80	40.47	2.66	41.76	2.66
W2-36	35.79	2.35	37.77	2.48	36.72	2.35
W2-37	38.95	2.58	39.18	2.58	39.07	2.51
W2-38	40.13	2.62	42.10	2.74	41.11	2.62
W2-39	41.99	2.77	42.86	2.81	42.43	2.72
W2-40	39.86	2.61	40.12	2.63	39.99	2.55
W2-41	36.55	2.42	40.14	2.64	38.24	2.45
W2-42	42.40	2.75	38.61	2.53	40.55	2.58
W2-43	40.70	2.68	39.24	2.57	39.89	2.55
W2-44	39.46	2.58	42.37	2.77	40.84	2.60
W2-45	38.86	2.57	39.18	2.58	39.02	2.50
Mean	40.31	2.50	40.27	2.50	40.26	2.50
Weighted mean	40.24	2.50	40.23	2.50	40.15	2.50
Minimum	35.79		37.77		36.72	
Maximum	43.16		42.86		42.48	
St. dev.	2.18		1.48		1.58	

(abs)						
Rel. st. dev. (%)	5.42		3.69		3.93	

Table 10: Measured activity concentrations of ^{226}Ra (mBq·L^{-1}) in sample batch 2 (W2 - IM-RN-2006-05-001448 / IM-RN-2006-07-001449). Sources prepared from a volume of 1.5 L.

Sample ID	**Measurement 1 (mBq·L^{-1})**		**Measurement 2 (mBq·L^{-1})**		**Weighted mean value (mBq·L^{-1})**	
	Act. conc.	u_c	**Act. conc.**	u_c	**Act. conc.**	u_c
W2-16	53.12	3.38	51.04	3.25	52.03	3.27
W2-17	46.71	2.98	46.69	2.98	46.70	2.94
W2-18	47.38	3.04	44.82	2.88	46.08	2.91
W2-19	44.83	2.87	44.87	2.87	44.85	2.83
W2-20	47.51	3.04	48.04	3.07	47.78	3.01
W2-21	56.20	3.57	53.30	3.41	54.82	3.45
W2-22	50.43	3.22	46.17	2.96	48.18	3.04
W2-23	39.43	2.52	40.79	2.61	40.09	2.53
W2-28	41.61	2.66	41.06	2.62	41.33	2.61
W2-29	38.63	2.47	38.59	2.46	38.61	2.43
W2-48	45.04	2.89	47.00	3.01	46.00	2.90
W2-49	50.54	3.23	50.60	3.24	50.57	3.19
W2-50	42.49	2.73	42.98	2.75	42.75	2.70
W2-51	45.06	2.87	44.49	2.84	44.77	2.82
Mean	46.36	2.88	45.75	2.85	46.04	2.86
Weighted mean	45.58	2.84	45.08	2.80	45.23	2.81
Minimum	38.63		38.59		38.61	
Maximum	56.20		53.30		54.82	
St. dev. (abs)	5.04		4.16		4.54	
Rel. st. dev. (%)	10.87		9.10		9.87	

Table 11: Measured activity concentrations of ^{226}Ra (mBq·L^{-1}) in sample batch 3 (W3 - IM-RN-2006-05-001450 / IM-RN-2006-07-001451). Sources prepared from a volume of 1.5 L.

Sample ID	**Measurement 1 (mBq·L^{-1})**		**Measurement 2 (mBq·L^{-1})**		**Weighted mean value (mBq·L^{-1})**	
	Act. conc.	u_c	**Act. conc.**	u_c	**Act. conc.**	u_c
W3-14	3.39	0.29	3.59	0.30	3.48	0.26
W3-15	2.65	0.24	2.62	0.24	2.63	0.20
W3-25	2.86	0.25	2.90	0.28	2.88	0.23
W3-26	2.47	0.22	2.65	0.24	2.55	0.20

W3-27	2.17	0.23	2.16	0.20	2.16	0.18
W3-28	2.47	0.22	2.39	0.22	2.43	0.19
W3-40	3.86	0.33	4.23	0.34	4.05	0.29
W3-41	3.50	0.30	3.16	0.27	3.32	0.25
W3-42	3.63	0.30	3.70	0.31	3.66	0.27
W3-43	3.58	0.31	3.76	0.30	3.68	0.27
Mean	3.06	0.20	3.12	0.20	3.09	0.20
Weighted mean	2.96	0.19	2.98	0.19	2.92	0.19
Minimum	2.17		2.16		2.16	
Maximum	3.86		4.23		4.05	
St. dev. (abs)	0.60		0.68		0.64	
Rel. st. dev. (%)	19.6		21.9		20.7	

Table 12: Measured activity concentrations of ^{226}Ra (mBq·L^{-1}) in sample batch 3 (W3 - IM-RN-2006-05-001450 / IM-RN-2006-07-001451). Sources prepared from a volume of 3.0 L.

Sample ID	**Measurement 1, (mBq·L^{-1})**		**Measurement 2 (mBq·L^{-1})**		**Weighted mean value (mBq·L^{-1})**	
	Act. conc.	u_c	**Act. conc.**	u_c	**Act. conc.**	u_c
W3-10	2.55	0.22	2.62	0.23	2.58	0.20
W3-11	3.18	0.26	3.11	0.25	3.14	0.23
W3-12	2.81	0.26	2.86	0.24	2.84	0.22
W3-13	2.94	0.23	2.95	0.23	2.95	0.21
W3-16	3.47	0.26	3.34	0.26	3.41	0.24
W3-17	3.18	0.24	3.27	0.25	3.22	0.22
W3-18	2.81	0.23	2.88	0.24	2.84	0.21
W3-19	2.71	0.22	2.84	0.24	2.76	0.20
W3-20	3.11	0.29	2.78	0.27	2.94	0.24
W3-21	2.72	0.23	2.74	0.24	2.73	0.20
W3-22	2.84	0.21	2.88	0.22	2.85	0.20
W3-23	3.20	0.24	3.44	0.26	3.31	0.23
W3-24	3.06	0.24	3.07	0.23	3.06	0.21
W3-39	3.04	0.23	3.28	0.25	3.15	0.22
W3-44	3.42	0.26	3.37	0.25	3.40	0.23
Mean	3.00	0.19	3.03	0.19	3.02	0.19
Weighted mean	3.01	0.19	3.05	0.20	3.00	0.19
Minimum	2.55		2.62		2.58	
Maximum	3.47		3.44		3.41	
St. dev. (abs)	0.27		0.26		0.25	
Rel. st. dev. (%)	8.89		8.54		8.41	

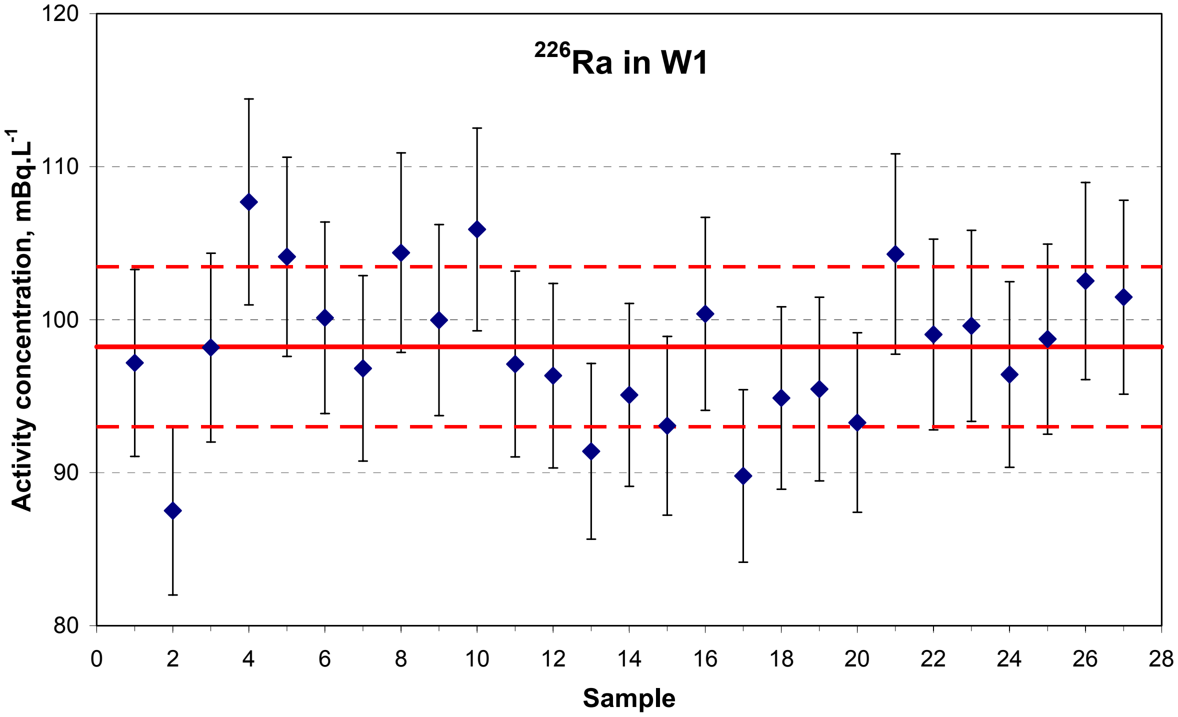

Fig. 3: ^{226}Ra activity concentration in mineral water - Batch 1 (W1 - IM-RN-2006-02-001446)

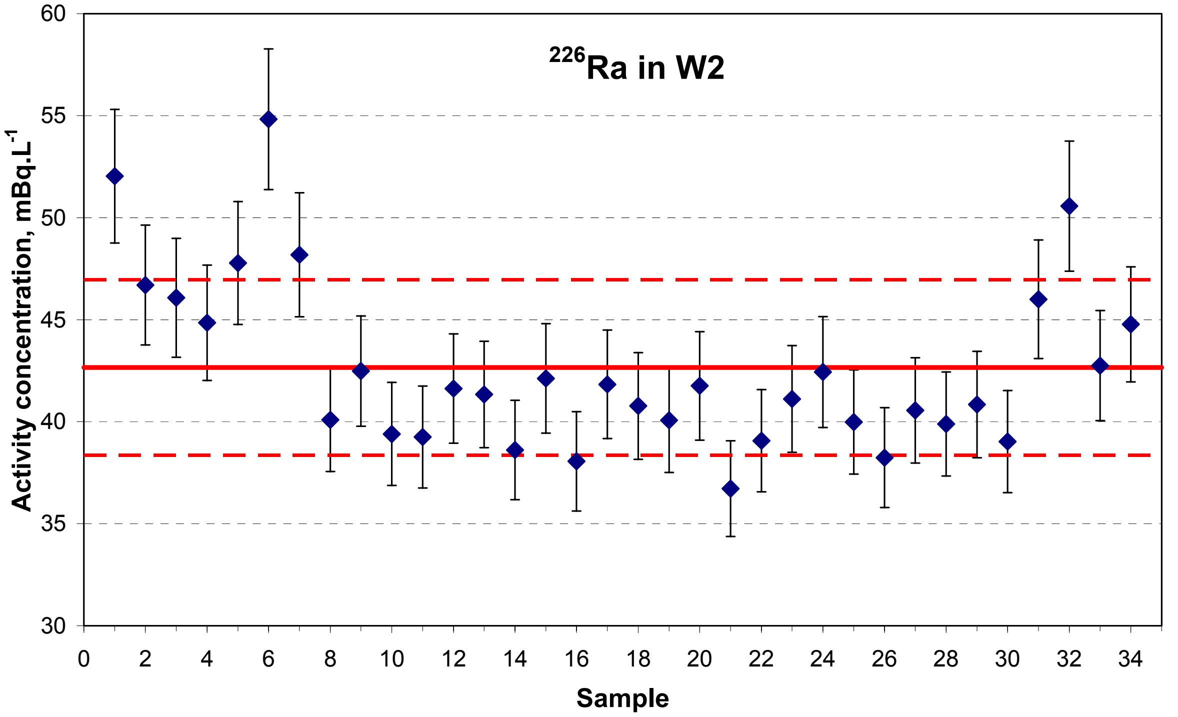

Fig. 4: ^{226}Ra activity concentration in mineral water – Batch 2 (W2 - IM-RN-2006-05-001448 / IM-RN-2006-07-001449)

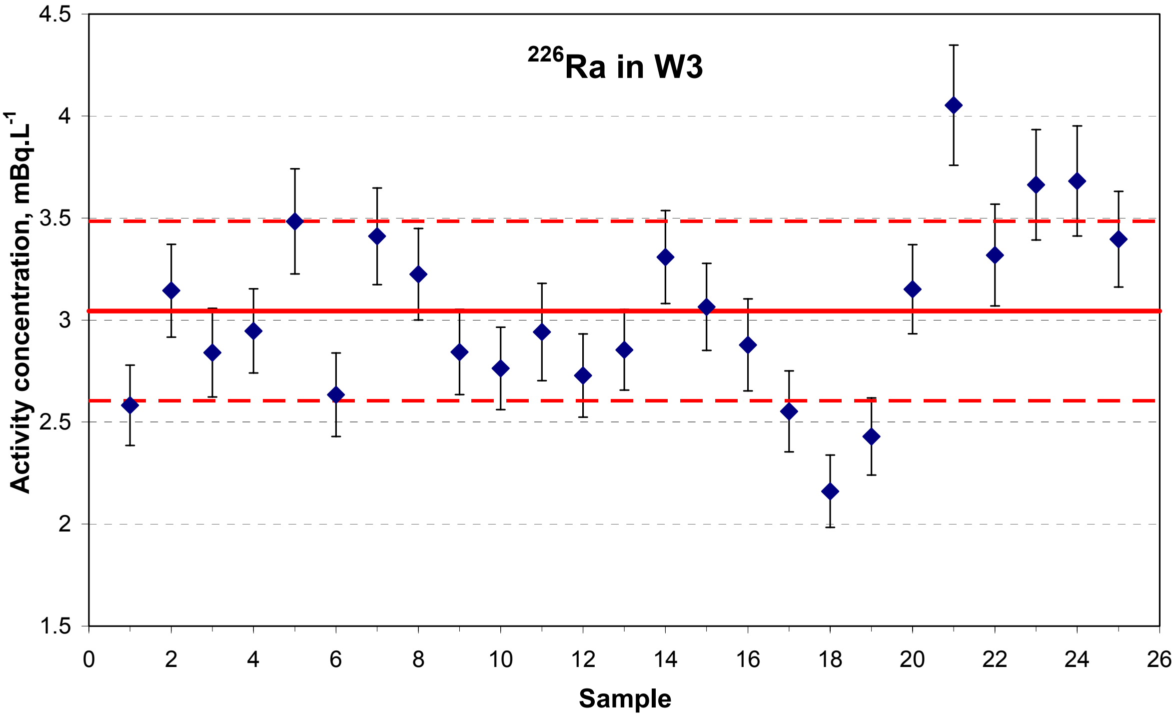

Fig. 5: ^{226}Ra activity concentration in mineral water – Batch 3 (W3 - IM-RN-2006-05-001450 / IM-RN-2006-07-001451)

Table 13: Activity concentration results for ^{226}Ra in mineral water (mBq·L^{-1})

Measurand	**W1**	**W2**	**W3***
Minimum value	85	36	2.50
Maximum value	112	56	3.50
Standard deviation	5.2	4.3	0.30
Mean value	**98.2**	**42.7**	**3.02**
Standard deviation of mean	0.7	0.5	0.05
Weighted mean value (mBq·L^{-1})	**97.7**	**42.0**	**3.00**
Expanded uncertainty *U* (*k*=2) (mBq·L^{-1})	**12.2**	**5.3**	**0.38**

Reference date - 1 May 2006 0:00 h UTC
*Results from 3L samples only

Table 14: Measured activity concentrations of ^{234}U and ^{238}U (mBq·L^{-1}) in sample batch 1 (W1 - IM-RN-2006-02-001446). Sources prepared from a volume of 1.5 L.

Sample ID	**^{234}U (mBq·L^{-1})**						**^{238}U (mBq·L^{-1})**					
	Measurement 1		**Measurement 2**		**Weighted mean value**		**Measurement 1**		**Measurement 2**		**Weighted mean value**	
	Act. conc.	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**
W1-16	13.36	0.53	13.39	0.53	13.38	0.43	9.85	0.43	9.51	0.42	9.68	0.33
W1-19	15.67	0.60	15.55	0.60	15.61	0.49	11.19	0.48	11.96	0.50	11.56	0.39
W1-22	13.26	0.52	14.56	0.55	13.89	0.43	9.85	0.42	10.14	0.43	9.99	0.34
W1-23	14.67	0.55	14.56	0.51	14.61	0.43	10.05	0.42	9.93	0.39	9.98	0.32
W1-25	16.75	0.57	15.27	0.52	15.96	0.45	10.80	0.41	10.07	0.39	10.42	0.32
W1-26	14.70	0.54	14.45	0.51	14.57	0.43	10.50	0.43	9.94	0.40	10.20	0.33
W1-28	16.82	0.56	16.29	0.55	16.55	0.46	11.71	0.43	11.83	0.43	11.77	0.35
W1-29	15.46	0.53	15.75	0.56	15.60	0.45	11.58	0.43	10.97	0.43	11.28	0.35
W1-35	15.24	0.58	16.10	0.59	15.67	0.48	11.36	0.47	10.61	0.44	10.96	0.36
W1-36	15.57	0.55	15.63	0.55	15.60	0.46	11.15	0.44	10.89	0.42	11.02	0.35
W1-38	16.23	0.58	16.43	0.58	16.33	0.48	11.53	0.46	11.25	0.44	11.38	0.36
W1-39	17.85	0.69	16.83	0.62	17.28	0.53	12.12	0.53	12.11	0.49	12.11	0.40
Mean	15.47	0.36	15.40	0.36	15.42	0.35	10.97	0.26	10.77	0.26	10.86	0.25
Weighted mean	15.38	0.36	15.34	0.36	15.36	0.34	10.94	0.26	10.68	0.25	10.80	0.24
Minimum	13.26		13.39		13.38		9.85		9.51		9.68	
Maximum	17.85		16.83		17.28		12.12		12.11		12.11	
St. dev. (abs)	1.36		1.00		1.12		0.76		0.88		0.79	
Rel. st. dev. (%)	8.79		6.48		7.29		6.94		8.17		7.30	

Table 15: Measured activity concentrations of ^{234}U and ^{238}U (mBq·L^{-1}) in sample batch 1 (W1 - IM-RN-2006-02-001446). Sources prepared from a volume of 3.0 L.

Sample ID	**^{234}U (mBq·L^{-1})**						**^{238}U (mBq·L^{-1})**					
	Measurement 1		**Measurement 2**		**Weighted mean value**		**Measurement 1**		**Measurement 2**		**Weighted mean value**	
	Act. conc.	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**
W1-10	13.09	0.38	12.88	0.38	12.98	0.33	9.60	0.30	9.10	0.29	9.34	0.25
W1-11	13.33	0.41	12.89	0.40	13.11	0.35	10.00	0.33	9.65	0.32	9.83	0.27
W1-12	13.49	0.41	13.10	0.40	13.29	0.35	9.88	0.33	9.87	0.32	9.88	0.27
W1-13	13.30	0.42	12.71	0.40	12.99	0.35	9.88	0.33	8.99	0.31	9.40	0.27
W1-14	13.54	0.40	12.84	0.39	13.18	0.34	9.64	0.31	9.23	0.30	9.43	0.26
W1-17	13.69	0.49	13.35	0.49	13.52	0.40	10.19	0.39	9.66	0.38	9.92	0.31
W1-18	14.23	0.55	14.22	0.56	14.22	0.45	9.91	0.43	9.98	0.43	9.94	0.34
W1-21	15.50	0.47	15.14	0.46	15.31	0.40	11.53	0.38	10.66	0.35	11.07	0.31
W1-24	15.26	0.47	14.06	0.45	14.66	0.39	10.82	0.36	9.81	0.34	10.32	0.29
W1-27	15.80	0.47	14.54	0.45	15.16	0.40	10.87	0.35	10.42	0.35	10.64	0.30
W1-30	15.73	0.54	15.73	0.52	15.73	0.44	11.32	0.42	11.94	0.42	11.64	0.35
Mean	14.27	0.33	13.77	0.32	14.01	0.31	10.33	0.24	9.94	0.23	10.13	0.23
Weighted mean	14.10	0.32	13.57	0.32	13.83	0.30	10.24	0.24	9.78	0.23	10.00	0.22
Minimum	13.09		12.71		12.98		9.60		8.99		9.34	
Maximum	15.80		15.73		15.73		11.53		11.94		11.64	
St. dev. (abs)	1.08		1.04		1.04		0.68		0.84		0.73	
Rel. st. dev. (%)	7.57		7.55		7.42		6.62		8.44		7.21	

Table 16: Measured activity concentrations of ^{234}U and ^{238}U (mBq·L^{-1}) in sample batch 2 (W2 - IM-RN-2006-05-001448 / IM-RN-2006-07-001449). Sources prepared from a volume of 1.5 L.

Sample ID	**^{234}U (mBq·L^{-1})**						**^{238}U (mBq·L^{-1})**					
	Measurement 1		**Measurement 2**		**Weighted mean value**		**Measurement 1**		**Measurement 2**		**Weighted mean value**	
	Act. conc.	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**
W2-12	4.58	0.26	4.13	0.24	4.51	0.17	1.04	0.12	1.17	0.13	1.17	0.08
W2-13	3.61	0.23	4.01	0.21	3.92	0.15	0.76	0.10	1.22	0.12	1.01	0.07
W2-22	4.48	0.34	3.37	0.26	3.84	0.19	0.75	0.14	0.67	0.13	0.85	0.09
W2-26	4.02	0.25	3.87	0.22	3.94	0.18	0.75	0.11	0.71	0.10	0.72	0.07
W2-27	3.96	0.25	3.77	0.21	3.92	0.15	0.91	0.11	1.07	0.12	0.97	0.07
W2-28	4.05	0.23	4.22	0.22	4.13	0.18	0.89	0.11	0.83	0.10	0.86	0.07
W2-30	4.19	0.24	4.18	0.24	4.18	0.19	1.03	0.11	0.95	0.11	0.99	0.08
W2-31	3.96	0.25	3.98	0.21	3.97	0.18	1.17	0.13	1.09	0.12	1.13	0.09
Mean	4.11	0.12	3.94	0.12	4.05	0.10	0.91	0.05	0.96	0.05	0.96	0.03
Weighted mean	4.07	0.12	3.96	0.15	4.05	0.10	0.90	0.05	0.95	0.05	0.96	0.03
Minimum	3.61		3.37		3.84		0.75		0.67		0.72	
Maximum	4.58		4.22		4.50		1.17		1.22		1.17	
St. dev. (abs)	0.31		0.27		0.22		0.16		0.21		0.15	
Rel. st. dev. (%)	7.55		6.96		5.35		17.25		21.88		15.35	

Table 17: Measured activity concentrations of ^{234}U and ^{238}U (mBq·L^{-1}) in sample batch 2 (W2 - IM-RN-2006-05-001448 / IM-RN-2006-07-001449). Sources prepared from a volume of 3.0 L.

Sample ID	**^{234}U (mBq·L^{-1})**						**^{238}U (mBq·L^{-1})**					
	Measurement 1		**Measurement 2**		**Weighted mean value**		**Measurement 1**		**Measurement 2**		**Weighted mean value**	
	Act. conc.	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**
W2-15	3.91	0.20	3.72	0.19	3.81	0.15	0.89	0.09	0.82	0.08	0.85	0.06
W2-16	4.16	0.21	4.36	0.21	4.13	0.14	0.83	0.08	1.06	0.09	0.93	0.05
W2-17	4.03	0.18	4.22	0.19	4.12	0.15	1.00	0.08	0.97	0.08	0.99	0.06
W2-19	3.87	0.20	3.85	0.19	3.93	0.13	0.97	0.09	1.18	0.10	1.07	0.06
W2-20	3.49	0.18	3.73	0.20	3.60	0.15	0.86	0.09	0.85	0.09	0.85	0.06
W2-21	4.25	0.20	4.19	0.21	4.34	0.16	1.10	0.10	1.38	0.11	1.16	0.07
W2-23	3.75	0.20	3.89	0.20	3.88	0.13	0.83	0.09	0.95	0.09	0.90	0.05
W2-24	3.69	0.17	3.89	0.16	3.79	0.13	0.85	0.07	0.77	0.06	0.81	0.05
W2-25	3.67	0.16	3.68	0.17	3.67	0.13	0.70	0.06	0.73	0.07	0.71	0.05
W2-29	3.84	0.17	3.94	0.18	3.77	0.12	1.01	0.08	0.87	0.08	0.88	0.05
Mean	3.86	0.10	3.95	0.11	3.90	0.09	0.90	0.03	0.96	0.04	0.92	0.03
Weighted mean	3.84	0.10	3.91	0.10	3.89	0.09	0.89	0.03	0.88	0.03	0.90	0.03
Minimum	3.49		3.68		3.60		0.70		0.73		0.71	
Maximum	4.25		4.36		4.34		1.10		1.38		1.16	
St. dev. (abs)	0.23		0.23		0.23		0.12		0.20		0.13	
Rel. st. dev. (%)	6.01		5.91		5.88		12.79		21.14		14.20	

Table 18: Measured activity concentrations of ^{234}U and ^{238}U (mBq·L^{-1}) in sample batch 3 (W3 - IM-RN-2006-05-001450). Sources prepared from a volume of 1.5 L.

Sample ID	**^{234}U (mBq·L^{-1})**						**^{238}U (mBq·L^{-1})**					
	Measurement 1		**Measurement 2**		**Weighted mean value**		**Measurement 1**		**Measurement 2**		**Weighted mean value**	
	Act. conc.	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**
W3-14	43.02	1.18	43.40	1.19	43.21	1.07	21.88	0.68	21.64	0.67	21.76	0.58
W3-15	44.05	1.36	42.90	1.29	43.43	1.15	21.38	0.78	21.49	0.75	21.44	0.63
W3-16	44.32	1.47	43.99	1.44	44.15	1.23	21.92	0.86	21.01	0.82	21.45	0.68
W3-17	42.41	1.58	43.05	1.61	42.72	1.30	21.18	0.96	21.57	0.98	21.37	0.76
W3-18	45.32	1.50	45.86	1.52	45.59	1.27	22.53	0.87	22.57	0.88	22.55	0.71
W3-19	43.04	1.73	43.56	1.76	43.30	1.40	21.54	1.04	22.49	1.07	22.00	0.82
W3-20	43.27	1.74	45.04	1.82	44.11	1.43	21.18	1.03	21.53	1.06	21.35	0.81
W3-50_4	47.28	1.36	45.63	1.32	46.44	1.18	23.23	0.77	23.24	0.77	23.24	0.65
W3-50_5	48.20	1.41	46.11	1.37	47.14	1.22	24.66	0.82	23.42	0.80	24.03	0.68
W3-50_6	44.29	1.56	46.67	1.63	45.43	1.32	21.93	0.94	23.99	1.00	22.90	0.77
W3-50_7	48.44	1.71	46.26	1.58	47.24	1.37	22.74	0.99	22.95	0.94	22.85	0.77
W3-50_8	45.22	1.64	47.49	1.74	46.28	1.39	22.06	0.98	22.61	1.02	22.32	0.78
Mean	44.90	1.03	45.00	1.04	44.92	0.99	22.18	0.54	22.38	0.55	22.27	0.50
Weighted mean	44.97	1.03	44.78	1.03	44.87	0.99	22.31	0.54	22.32	0.54	22.31	0.50
Minimum	42.41		42.90		42.72		21.18		21.01		21.35	
Maximum	48.44		47.49		47.24		24.66		23.99		24.03	
St. dev. (abs)	2.06		1.56		1.62		1.00		0.93		0.86	
Rel. st. dev. (%)	4.58		3.47		3.61		4.50		4.14		3.88	

Table 19: Measured activity concentrations of ^{234}U and ^{238}U (mBq·L^{-1}) in sample batch 3 (W3 - IM-RN-2006-05-001450). Sources prepared from a volume of 3.0 L.

Sample ID	**^{234}U (mBq·L^{-1})**						**^{238}U (mBq·L^{-1})**					
	Measurement 1		**Measurement 2**		**Weighted mean value**		**Measurement 1**		**Measurement 2**		**Weighted mean value**	
	Act. conc.	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**
W3-10	43.79	1.14	43.02	1.12	43.40	1.04	20.91	0.60	21.35	0.60	21.13	0.53
W3-11	42.67	1.11	43.40	1.13	43.03	1.03	21.48	0.61	22.00	0.62	21.74	0.55
W3-12	44.22	1.08	43.86	1.07	44.03	1.01	21.70	0.57	21.60	0.57	21.65	0.52
W3-13	44.08	1.12	42.05	1.07	43.00	1.01	21.76	0.60	20.68	0.57	21.19	0.52
W3-21	45.54	1.79	45.54	1.80	45.54	1.45	21.22	0.97	22.11	1.01	21.65	0.77
W3-22	43.90	1.30	45.18	1.34	44.51	1.15	21.51	0.70	21.65	0.71	21.58	0.60
W3-23	44.47	1.30	44.45	1.31	44.46	1.15	22.87	0.73	22.05	0.72	22.45	0.62
W3-24	45.22	1.39	44.18	1.36	44.69	1.19	21.40	0.73	21.85	0.75	21.62	0.62
W3-50-9	44.74	1.18	48.63	1.29	46.49	1.12	22.37	0.64	24.00	0.69	23.11	0.59
W3-50-10	46.81	1.23	47.39	1.26	47.08	1.13	23.85	0.68	23.79	0.69	23.82	0.60
Mean	44.54	0.99	44.77	0.99	44.62	0.97	21.91	0.52	22.11	0.52	21.99	0.48
Weighted mean	44.35	0.99	44.32	0.98	44.34	0.97	21.90	0.51	21.96	0.52	21.93	0.48
Minimum	42.67		42.05		43.00		20.91		20.68		21.13	
Maximum	46.81		48.63		47.08		23.85		24.00		23.82	
St. dev. (abs)	1.13		2.00		1.39		0.88		1.03		0.87	
Rel. st. dev. (%)	2.53		4.47		3.12		4.04		4.66		3.96	

Table 20: Measured activity concentrations of ^{234}U and ^{238}U (mBq·L^{-1}) in sample batch 3 (W3 - IM-RN-2006-07-001451). Sources prepared from a volume of 1.5 L.

Sample ID	^{234}U (mBq·L^{-1})						^{238}U (mBq·L^{-1})					
	Measurement 1		**Measurement 2**		**Weighted mean value**		**Measurement 1**		**Measurement 2**		**Weighted mean value**	
	Act. conc.	u_c	**Act. conc.**	u_c	**Act. conc.**	u_c	**Act. conc.**	u_c	**Act. conc.**	u_c	**Act. conc.**	u_c
W3-37	39.95	1.22	40.34	1.27	40.14	1.07	19.71	0.71	21.43	0.78	20.49	0.61
W3-38	41.33	1.28	39.59	1.21	40.41	1.07	19.78	0.73	19.20	0.71	19.48	0.59
W3-40	40.97	1.18	40.21	1.14	40.57	1.03	20.22	0.67	19.36	0.64	19.77	0.55
W3-41	41.16	1.18	40.82	1.15	40.98	1.03	20.04	0.68	19.66	0.65	19.84	0.56
W3-42	39.39	1.21	38.95	1.20	39.17	1.04	21.03	0.73	20.37	0.73	20.70	0.60
W3-45	41.22	1.21	41.64	1.22	41.42	1.06	21.77	0.71	20.87	0.71	21.34	0.60
W3-51_4	38.90	1.21	41.28	1.27	40.05	1.07	19.99	0.73	20.77	0.75	20.37	0.61
W3-51_5	39.13	1.17	40.96	1.27	39.94	1.05	19.99	0.69	19.98	0.73	19.98	0.59
W3-51_6	40.89	1.18	39.77	1.14	40.31	1.02	18.56	0.64	20.96	0.69	19.70	0.55
W3-51-8	39.16	1.22	41.07	1.25	40.10	1.06	20.42	0.72	20.84	0.73	20.63	0.60
W3-51-9	39.73	1.28	42.67	1.33	41.19	1.12	19.54	0.73	20.98	0.75	20.26	0.61
W3-51-10	39.99	1.18	41.82	1.26	40.82	1.06	19.65	0.67	21.15	0.73	20.32	0.58
W3-51-11	38.84	1.15	41.35	1.26	39.95	1.04	19.06	0.65	20.45	0.72	19.66	0.57
W3-51-12	42.29	1.28	41.66	1.27	41.98	1.10	21.82	0.75	21.50	0.74	21.66	0.62
W3-51-13	40.50	1.24	41.60	1.26	41.05	1.08	21.28	0.73	21.58	0.73	21.43	0.61
W3-51-14	40.62	1.28	43.38	1.37	41.91	1.13	21.46	0.77	20.66	0.75	21.05	0.63
Mean	40.26	0.92	41.07	0.94	40.62	0.89	20.27	0.47	20.61	0.48	20.42	0.45
Weighted mean	40.25	0.92	40.97	0.93	40.60	0.89	20.21	0.47	20.56	0.48	20.38	0.45
Minimum	38.84		38.95		39.17		18.56		19.20		19.48	
Maximum	42.29		43.38		41.98		21.82		21.58		21.66	
St.dev. (abs)	1.02		1.13		0.77		0.96		0.73		0.68	
Rel. st. dev. (%)	2.54		2.76		1.89		4.72		3.55		3.32	

Table 21: Measured activity concentrations of ^{234}U and ^{238}U (mBq·L^{-1}) in sample batch 3 (W3 - IM-RN-2006-07-001451). Sources prepared from a volume of 3.0 L.

Sample ID	**^{234}U (mBq·L^{-1})**						**^{238}U (mBq·L^{-1})**					
	Measurement 1		**Measurement 2**		**Weighted mean value**		**Measurement 1**		**Measurement 2**		**Weighted mean value**	
	Act. conc.	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**	**Act. conc.**	**u_c**
W3-26	38.37	1.47	39.84	1.49	39.10	1.20	19.52	0.83	19.96	0.83	19.74	0.66
W3-36	40.97	1.14	40.15	1.12	40.56	1.01	19.71	0.61	19.79	0.62	19.75	0.53
W3-39	39.77	1.14	40.51	1.16	40.14	1.02	20.22	0.65	19.36	0.63	19.78	0.54
W3-51-7	39.61	1.19	43.05	1.30	41.18	1.08	20.75	0.69	22.59	0.75	21.59	0.60
W3-51-15	43.16	1.27	42.79	1.24	42.97	1.10	21.49	0.70	20.89	0.68	21.17	0.58
W3-51-16	40.58	1.23	42.56	1.27	41.55	1.09	20.36	0.69	21.19	0.71	20.77	0.59
W3-51-17	41.50	1.22	41.65	1.18	41.58	1.06	19.87	0.65	20.89	0.65	20.41	0.56
W3-51-18	41.42	1.22	42.52	1.25	41.96	1.08	21.19	0.70	20.80	0.70	20.99	0.59
W3-51-19	40.53	1.29	41.27	1.31	40.90	1.11	21.41	0.76	21.08	0.75	21.24	0.62
W3-51-20	40.30	1.20	40.72	1.26	40.49	1.06	20.72	0.68	20.65	0.71	20.69	0.59
W3-51-21	43.05	1.32	41.67	1.28	42.34	1.12	21.14	0.73	20.51	0.71	20.82	0.60
Mean	40.84	0.95	41.52	0.97	41.16	0.90	20.58	0.49	20.70	0.49	20.63	0.46
Weighted mean	40.90	0.95	41.52	0.97	41.20	0.90	20.55	0.49	20.64	0.49	20.60	0.46
Minimum	38.37		39.84		39.10		19.52		19.36		19.74	
Maximum	43.16		43.05		42.97		21.49		22.59		21.59	
St. dev. (abs)	1.42		1.12		1.09		0.69		0.85		0.64	
Rel. st. dev. (%)	3.49		2.69		2.64		3.37		4.11		3.11	

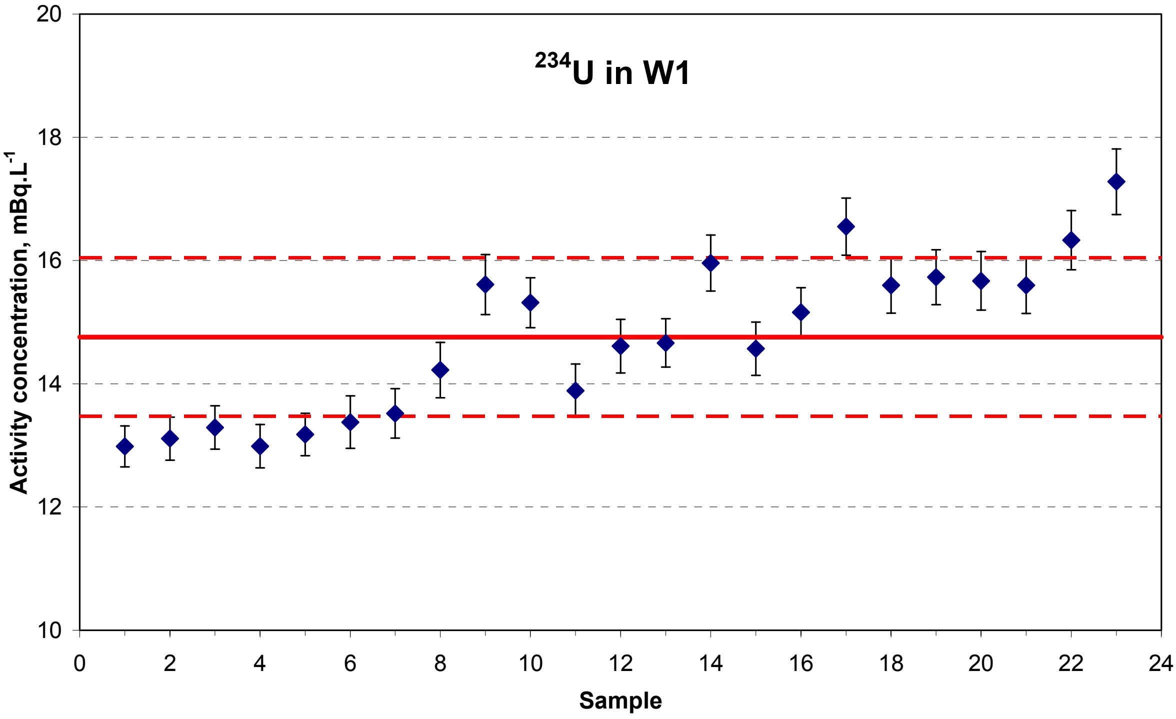

Fig. 6: ^{234}U activity concentration in mineral water - Batch 1 (W1 - IM-RN-2006-02-001446)

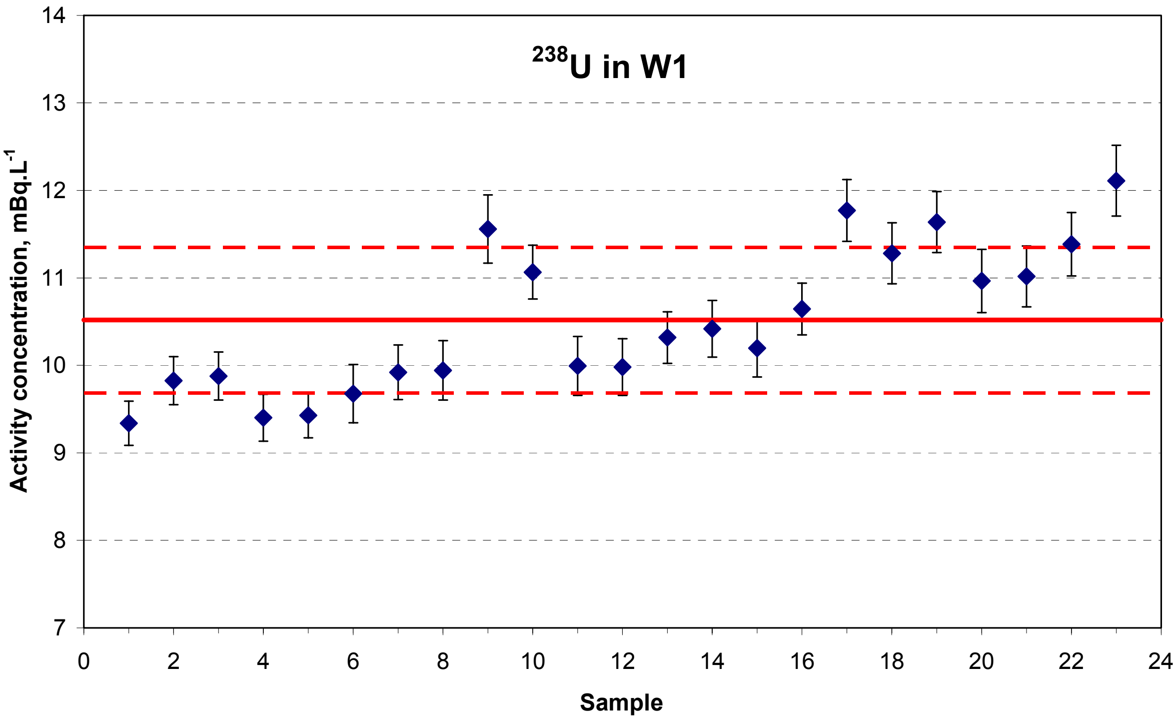

Fig. 7: ^{238}U activity concentration in mineral water - Batch 1 (W1 - IM-RN-2006-02-001446)

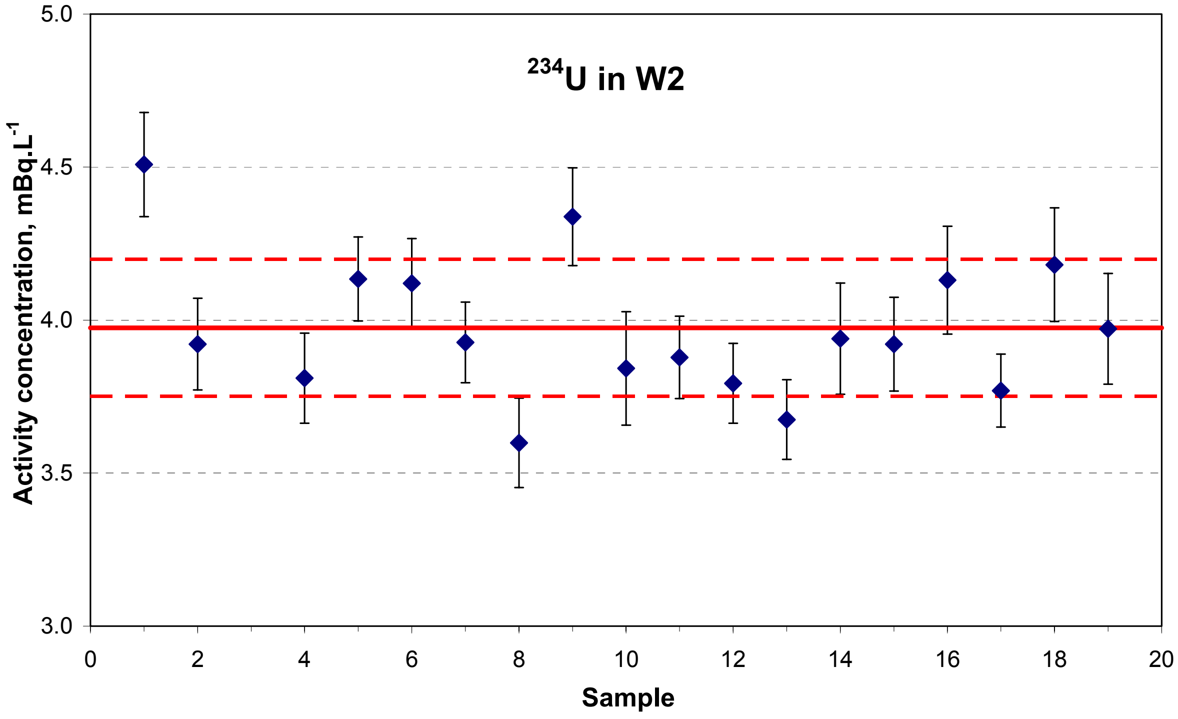

Fig. 8: ^{234}U activity concentration in mineral water - Batch 2
(W2 - IM-RN-2006-05-001448 / IM-RN-2006-07-001449)

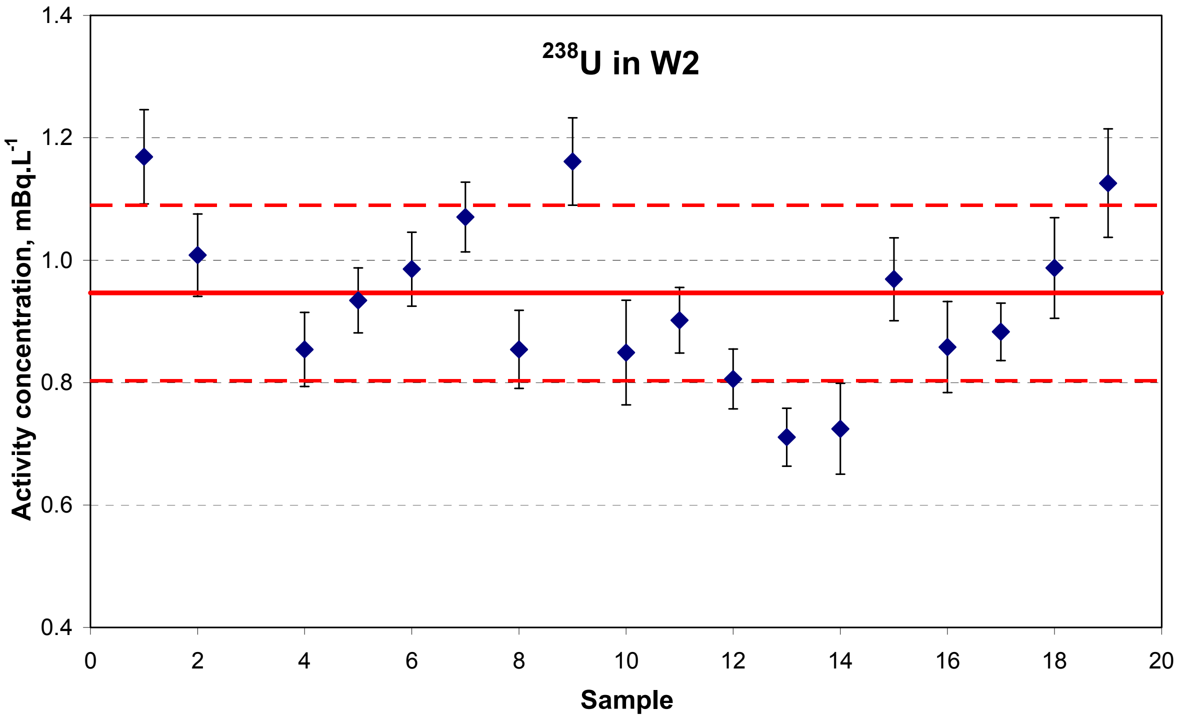

Fig. 9: ^{238}U activity concentration in mineral water - Batch 2
(W2 - IM-RN-2006-05-001448 / IM-RN-2006-07-001449)

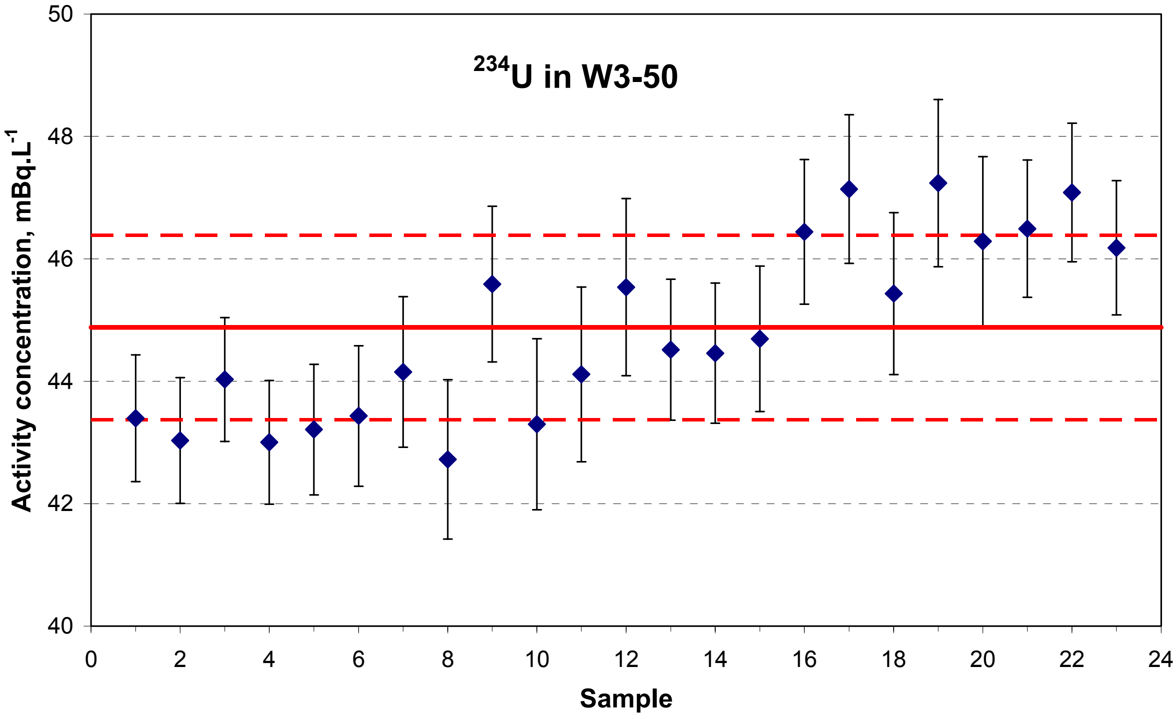

Fig. 10: ^{234}U activity concentration in mineral water - Batch 3-50 (W3 - IM-RN-2006-05-001450)

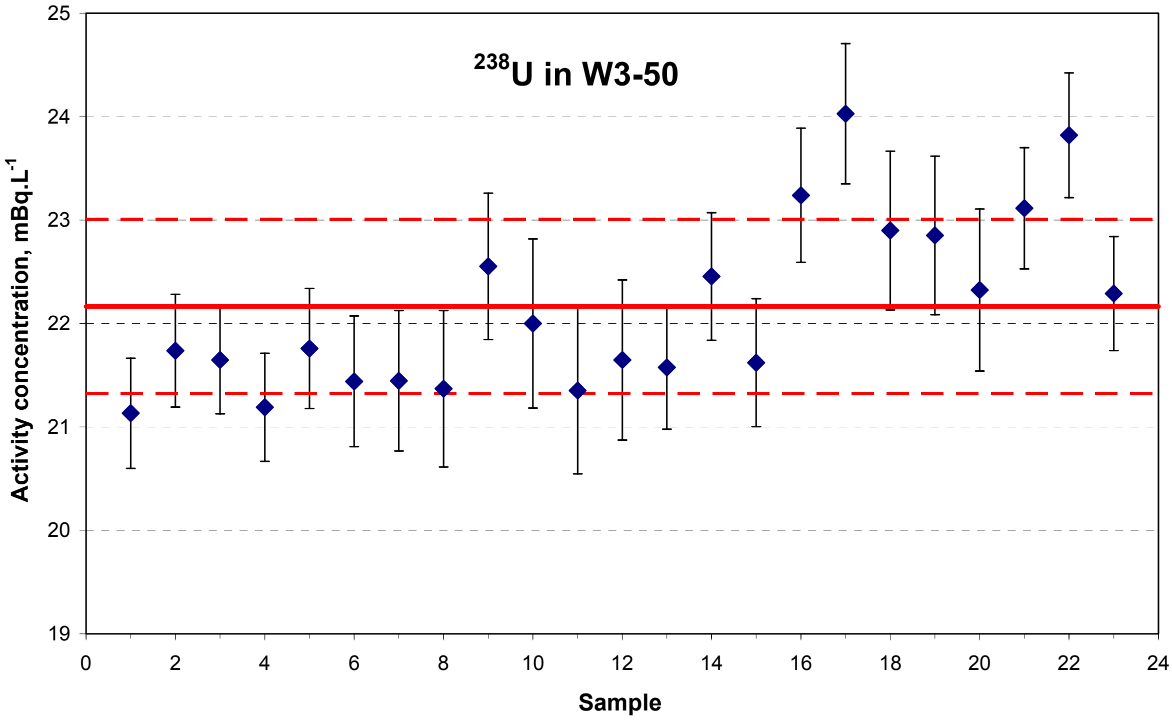

Fig. 11: ^{238}U activity concentration in mineral water - Batch 3-50 (W3 - IM-RN-2006-05-001450)

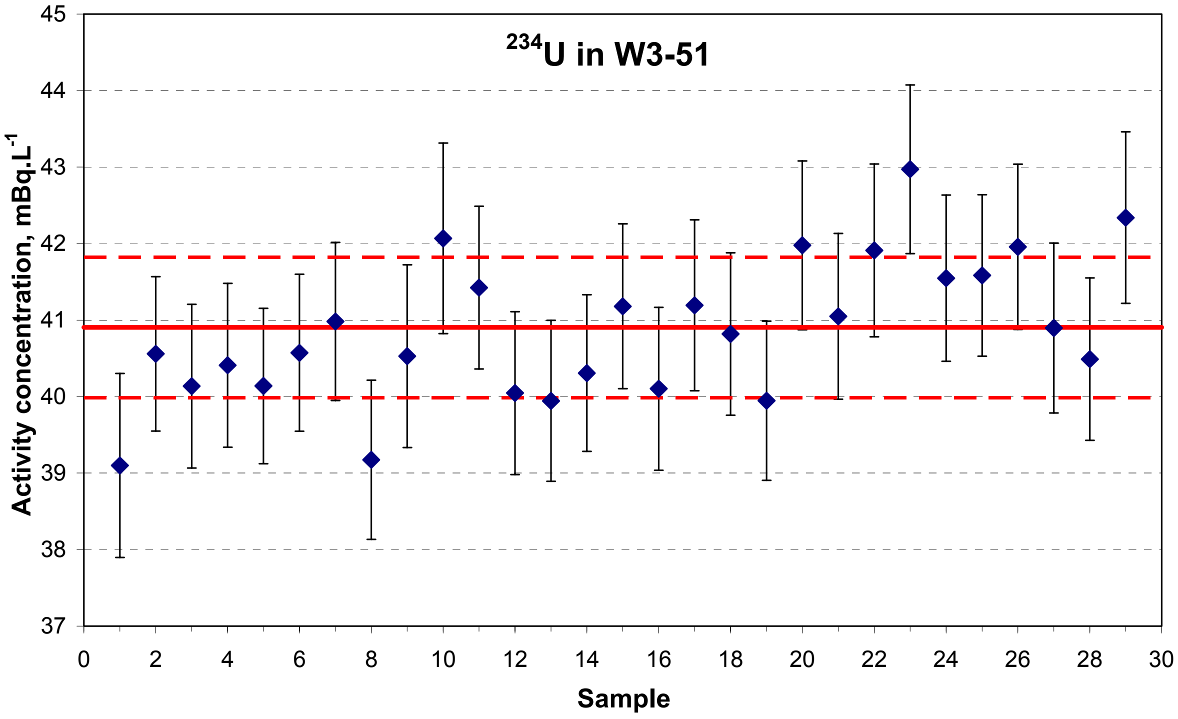

Fig. 12: ^{234}U activity concentration in mineral water - Batch 3-51 (W3 - IM-RN-2006-07-001451)

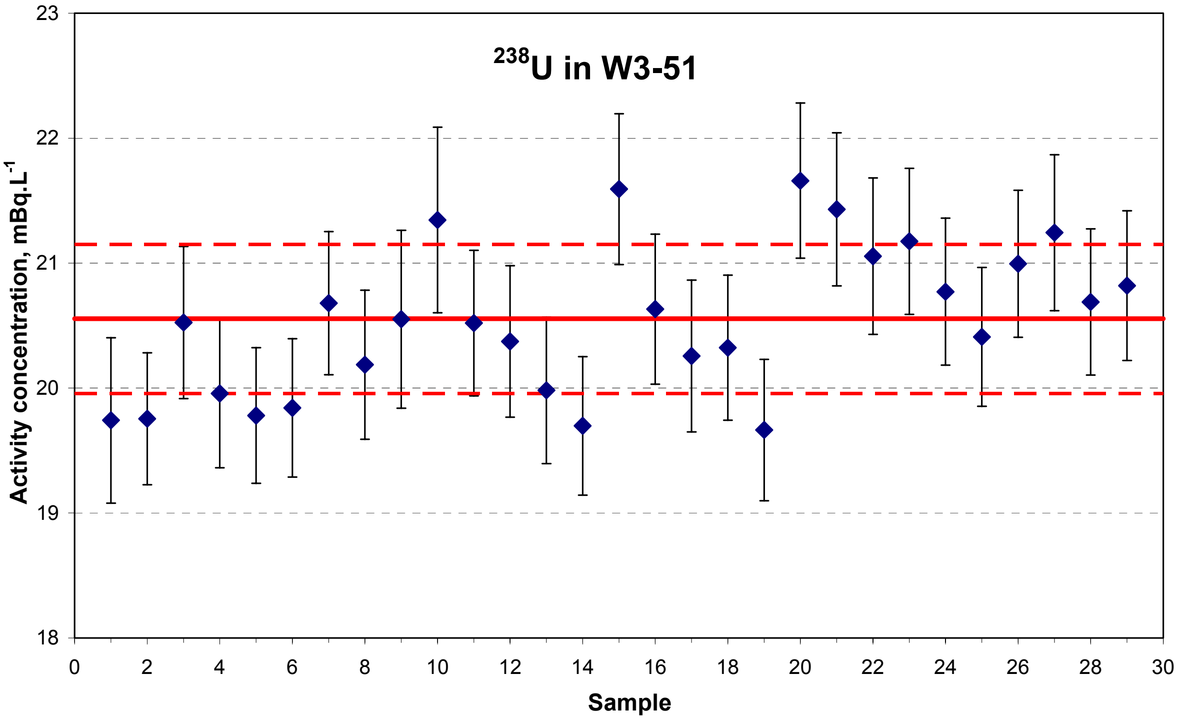

Fig. 13: ^{238}U activity concentration in mineral water - Batch 3-51 (W3 - IM-RN-2006-07-001451)

Table 22: Activity concentration results for ^{234}U in mineral water ($mBq \cdot L^{-1}$)

Measurand	**W1**	**W2**	**W3-50**	**W3-51**
Minimum value	13.0	3.60	42.8	39.1
Maximum value	17.3	4.50	47.4	43.0
Standard deviation	1.3	0.30	1.5	1.0
Mean value	**14.8**	**4.00**	**44.9**	**40.9**
Standard deviation of mean	0.3	0.05	0.3	0.2
Weighted mean value ($mBq \cdot L^{-1}$)	**14.4**	**3.90**	**44.8**	**40.9**
Expanded uncertainty *U* (*k*=2) ($mBq \cdot L^{-1}$)	**0.6**	**0.20**	**2.0**	**1.8**

Reference date - 1 May 2006 0:00 h UTC

Table 23. Activity concentration results for ^{238}U in mineral water ($mBq \cdot L^{-1}$)

Measurand	**W1**	**W2**	**W3-50**	**W3-51**
Minimum value	9.4	0.74	21.1	19.7
Maximum value	12.1	1.24	24.0	21.7
Standard deviation	0.8	0.14	0.9	0.6
Mean value	**10.5**	**0.95**	**22.2**	**20.6**
Standard deviation of mean	0.2	0.03	0.2	0.1
Weighted mean value ($mBq \cdot L^{-1}$)	**10.3**	**0.90**	**22.1**	**20.5**
Expanded uncertainty *U* (*k*=2) ($mBq \cdot L^{-1}$)	**0.4**	**0.04**	**1.0**	**0.9**

Reference date - 1 May 2006 0:00 h UTC

Measurements of ^{228}Ra at IRMM

The activity concentration of ^{228}Ra was determined via its daughter ^{228}Ac and measured by a liquid scintillation counter, Wallac Quantulus 1220^{TM}.

The activity concentration of ^{228}Ra is calculated from equation (9):

$$A = \frac{N}{Eff \times R_{chem} \times V \times e^{-\lambda t_1}} \times \frac{\lambda \times t_2}{1 - e^{-\lambda t_2}} \qquad (9)$$

where, N = net counting rate (cps)

Eff = counting efficiency (%)

R_{chem} = chemical recovery (%)

V = volume of the sample (L)

t_1 = time from the first rinse of the column till start of measurement of ^{228}Ac (min)

t_2 = measurement time (min)

λ = decay constant of ^{228}Ac

The activity concentration of ^{228}Ra and, therefore, that of the resulting ^{228}Ac in the studied mineral waters is very low. In addition, the decay of ^{228}Ac occurs very fast ($T_{1/2}$ = 6.13 h), too fast to allow for lengthy measurements. The background contribution is very significant and any means to reduce it must be employed. A volume of 7 mL of Insta-Gel Plus cocktail was enough to assure a homogeneous single phase mixture with the sample and at the same time obtain a low background in the measurement window from 350 to 850 channels. To compensate for fluctuations, the blank was measured for 120 minutes each time before and after each sample.

The chemical recovery of radium was determined using the 356 keV gamma-ray line of ^{133}Ba. The chemical recovery R_{chem} was calculated according to equation (10):

$$R_{chem} = \left(\frac{A_{sample}}{t_{sample} \cdot m_{sample}} \right) \cdot \left(\frac{t_{Std} \cdot m_{Std}}{A_{Std}} \right),$$

(10)

where:

A_{sample}, A_{Std}	are the net areas of ^{133}Ba peak from the sample and the standard sample, respectively;
t_{sample}, t_{Std}	are the counting times of sample and standard sample, respectively;
m_{sample}, m_{Std}	are the masses of added ^{133}Ba in the sample and of ^{133}Ba in the standard sample respectively.

Separate experiments were performed for the determination of the chemical recovery of ^{228}Ac from the second Diphonix column. A 1 mg·mL^{-1} natural thorium solution in equilibrium was used and the ^{228}Ac was isolated. The latter was determined by measuring its 911 keV gamma-ray line using a HPGe detector and it was used to evaluate the chemical recovery. The mean value for the chemical recovery obtained from three replicate experiments was (83 ± 5) %.

To determine the liquid scintillation counting efficiency, ^{228}Ac was separated from 1mg·mL^{-1} natural thorium standard solution in equilibrium applying the same radiochemical procedure as described above. A detection efficiency of (80.3 ± 6.3) % in the window from 350 - 850 channels was obtained.

Measurement results and plots of the obtained activity concentrations of ^{228}Ra in W1 (1.5 L and 3 L) and W2 (1.5 L and 3 L), together with their combined standard uncertainties u_c, are presented in Tables 24 to 29 and Figures 14 and 15. The solid and the dashed red lines in the figures indicate the mean values and the standard deviations. The error bars represent combined standard uncertainties u_c of the individual measurements.

Table 24: Measured activity concentrations of ^{228}Ra in sample batch 1(W1 - IM-RN-2006-02-001446). Sources prepared from a volume of 1.5 L.

Sample ID	Activity concentration (mBq·L^{-1})	Uncertainty u_c (mBq·L^{-1})
W1-10	20.36	2.58
W1-11	26.21	3.24
W1-12	28.52	3.79
W1-13	26.06	3.46
W1-14	24.75	3.28
W1-15	22.42	3.08
W1-16	20.38	2.70
W1-18	27.44	3.76
W1-19	21.11	2.85
W1-22	23.31	3.18
Mean	**24.06**	
Weighted mean	**23.40**	
Minimum	**20.36**	
Maximum	**28.52**	
Standard deviation	**2.97**	
Relative standard deviation (%)	**12.35**	

Table 25: Measured activity concentrations of ^{228}Ra in sample batch 1 (W1 - IM-RN-2006-02-001446). Sources prepared from a volume of 3.0 L.

Sample ID	Activity concentration (mBq·L^{-1})	Uncertainty u_c (mBq·L^{-1})
W1-24	22.76	2.97
W1-25	22.95	3.00
W1-29	21.08	2.54
W1-30	24.47	2.92
W1-31	24.81	2.97
W1-33	21.64	2.62
W1-34	20.59	2.50
W1-35	25.04	3.00
W1-37	26.42	3.16
W1-38	20.24	2.43
Mean	**23.00**	
Weighted mean	**22.69**	
Minimum	**20.24**	
Maximum	**26.42**	
Standard deviation	**2.12**	
Relative standard deviation (%)	**9.20**	

Table 26: Measured activity concentrations of ^{228}Ra in sample batch 2 (W2 - IM-RN-2006-05-001448 / IM-RN-2006-07-001449). Sources prepared from a volume of 1.5 L.

Sample ID	Activity concentration (mBq·L^{-1})	Uncertainty u_c (mBq·L^{-1})
W2-29	69.78	8.37
W2-30	69.13	8.32
W2-27	72.23	8.60
W2-31	75.17	9.00
W2-25	63.19	7.70
W2-33	70.00	8.42
W2-37	65.98	7.98
W2-35	61.52	7.52
W2-26	63.93	7.92
W2-38	69.51	8.18
Mean	**68.04**	
Weighted mean	**67.80**	
Minimum	**61.52**	
Maximum	**75.17**	
Standard deviation	**4.29**	
Relative standard deviation (%)	**6.31**	

Table 27: Measured activity concentrations of ^{228}Ra in sample batch 2 (W2 - IM-RN-2006-05-001448 / IM-RN-2006-07-001449). Sources prepared from a volume of 3.0 L.

Sample ID	**Activity concentration (mBq·L^{-1})**	**Uncertainty u_c (mBq·L^{-1})**
W2-40	69.88	9.18
W2-41	60.54	7.09
W2-45	61.04	7.21
W2-47	70.92	8.38
W2-51	66.93	7.66
W2-52	66.19	7.66
W2-53	70.74	8.13
W2-54	61.66	7.06
W2-55	66.99	7.69
W2-56	61.22	7.11
Mean	**65.61**	
Weighted mean	**64.97**	
Minimum	**60.54**	
Maximum	**70.92**	
Standard deviation	**4.19**	
Relative standard deviation (%)	**6.39**	

Table 28: Measured activity concentrations of ^{228}Ra in sample batch 1 (W1 - IM-RN-2006-02-001446). Sources prepared from volumes of 1.5 L and 3.0 L.

Sample ID	**Activity concentration (mBq·L^{-1})**	**Uncertainty u_c (mBq·L^{-1})**
W1-10	20.36	2.58
W1-11	26.21	3.24
W1-12	28.52	3.79
W1-13	26.06	3.46
W1-14	24.75	3.28
W1-15	22.42	3.08
W1-16	20.38	2.70
W1-18	27.44	3.76
W1-19	21.11	2.85
W1-22	23.31	3.18
W1-24	22.76	2.97
W1-25	22.95	3.00
W1-29	21.08	2.54
W1-30	24.47	2.92
W1-31	24.81	2.97
W1-33	21.64	2.63
W1-34	20.59	2.50
W1-35	25.04	3.00
W1-37	26.42	3.16

W1-38	20.24	2.43
Mean	**23.53**	
Weighted mean	**22.96.**	
Minimum	**20.24**	
Maximum	**28.52**	
Standard deviation	**2.57**	
Relative standard deviation (%)	**10.92**	

Table 29: Measured activity concentrations of ^{228}Ra in sample batch 2 (W2 - IM-RN-2006-05-001448 / IM-RN-2006-07-001449). Sources prepared from volumes of 1.5 L and 3.0 L.

Sample ID	Activity concentration ($mBq \cdot L^{-1}$)	Uncertainty u_c ($mBq \cdot L^{-1}$)
W2-29	69.78	8.37
W2-30	69.13	8.32
W2-27	72.23	8.60
W2-31	75.17	9.00
W2-25	63.19	7.70
W2-33	70.00	8.42
W2-37	65.98	7.98
W2-35	61.52	7.52
W2-26	63.93	7.92
W2-38	69.51	8.18
W2-40	69.88	9.18
W2-41	60.54	7.09
W2-45	61.04	7.21
W2-47	70.92	8.38
W2-51	66.93	7.66
W2-52	66.19	7.66
W2-53	70.74	8.13
W2-54	61.66	7.06
W2-55	66.99	7.69
W2-56	61.22	7.11
Mean	**66.83**	
Weighted mean	**66.26**	
Minimum	**60.54**	
Maximum	**75.17**	
Standard deviation	**4.31**	
Relative standard deviation (%)	**6.46**	

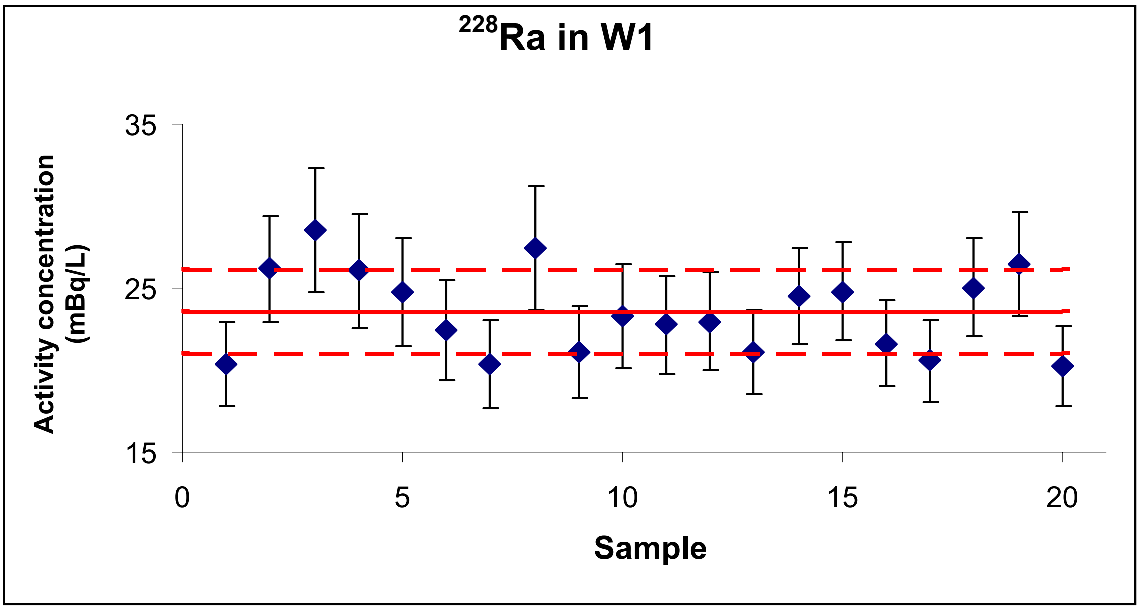

Fig. 14: ^{228}Ra activity concentration in mineral water - Batch 1 (W1 - IM-RN-2006-02-001446). Sources prepared from volumes of 1.5 L and 3.0 L.

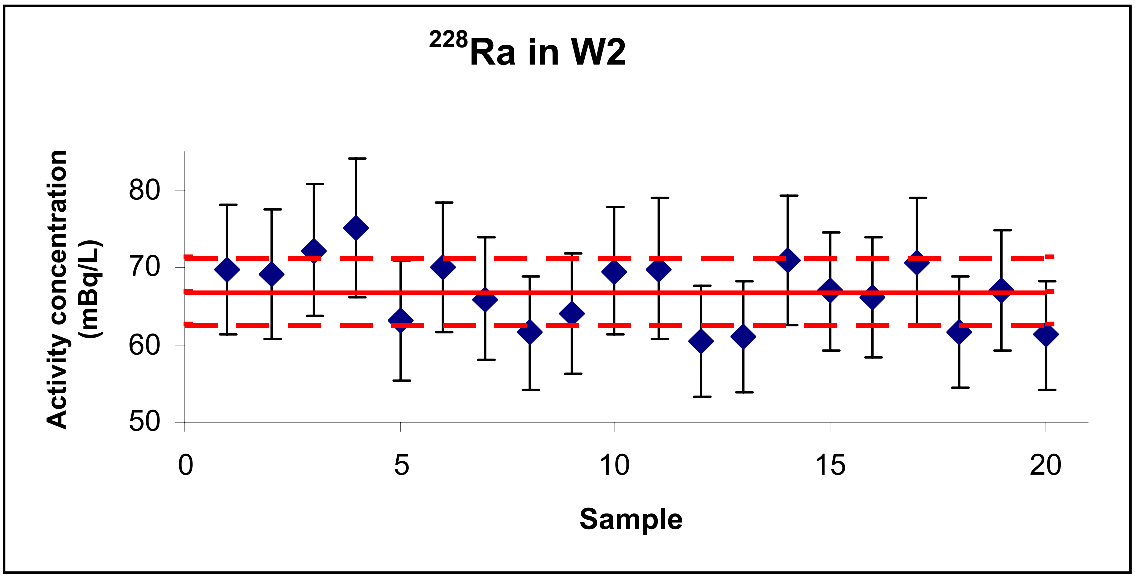

Fig. 15: ^{228}Ra activity concentration in mineral water - Batch 2 (W2 - IM-RN-2006-05-001448 / IM-RN-2006-07-001449). Sources prepared from volumes of 1.5 L and 3.0 L.

Uncertainty budget for ^{228}Ra (IRMM)

The combined standard uncertainty is calculated from equation (11):

$$u(A_{228_{Ra}}) = A_{228_{Ra}} \times \sqrt{\left(\frac{u(N)}{N}\right)^2 + \left(\frac{u(R_{chem})}{R_{chem}}\right)^2 + \left(\frac{u(V)}{V}\right)^2 + \left(\frac{u(Eff)}{Eff}\right)^2} \tag{11}$$

The uncertainty associated with the *count rate* taking into account the standard deviation of the count rate for the sample and standard deviation for the count rate of the background.

Net count = counts (sample) – counts (background)

The uncertainty associated to the *net* count rate is then the sum of the squares of the two uncertainties.

$$u(N) = \sqrt{\left(\frac{counts(sample)}{time(sample)} + \frac{counts(bkg)}{time(bkg)}\right)} \tag{12}$$

The uncertainty associated with *weighing* of the tracer was estimated using the data from the repeatability of the balance on standard deviation as 0.02 mg. The uncertainty associated with the weighing of the tracer represents 0.2 %.

The uncertainty of the *radiochemical recovery* of the method is determined according to equation (13):

$$u(R_{chem}) = \sqrt{\left(u(R_{228_{Ra}})\right)^2 + \left(u(R_{228_{Ac}})\right)^2} \tag{13}$$

The relative standard uncertainty of the chemical yield of ^{228}Ra is calculated with equation (14):

$$u(R_{Ra-228}) = R_{Ra-228}\sqrt{\left(\frac{u\left(A_{133_{Ba-sample}}\right)}{A_{133_{Ba-sample}}}\right)^2 + \left(\frac{u\left(m_{133_{Ba-sample}}\right)}{m_{133_{Ba-sample}}}\right)^2 + \left(\frac{u\left(A_{133_{Ba-std}}\right)}{A_{133_{Ba-std}}}\right)^2 + \left(\frac{u\left(m_{133_{Ba-std}}\right)}{m_{133_{Ba-std}}}\right)^2} \tag{14}$$

Uncertainty of the chemical recovery of ^{228}Ac is taken as standard deviation of three replicate experiments.

The uncertainty associated with the *volume of the sample* is estimated using data from the manufacturer: for a flask of 1000 mL the uncertainty is ± 5 mL measured at a temperature of 20°C. The value of the uncertainty is given without confidence level or distribution information; therefore, it is necessary to make an assumption and the standard uncertainty is calculated assuming a triangular distribution (Eurachem, 2000). A negligible contribution of 0.1% to the uncertainty budget of ^{228}Ra is coming from the sample volume

The efficiency of the Wallac Quantulus 1220™ is calculated using equation (15):

$$\varepsilon = \frac{N_{Ac-228Std}}{m_{Th} \times A_{Th} \times R_{Ac-228}} \tag{15}$$

where: ε efficiency of beta-particle detector

R_{Ac-228} recovery of the ^{228}Ac standard

$N_{Ac\text{-}228}$ net counting rate of ^{228}Ac standard
m_{Th} mass of added Th natural standard solution (g)
A_{Th} activity of Th natural standard solution (Bq/g)

and yields the uncertainty expression

$$u(\varepsilon)=\varepsilon\sqrt{\left(\frac{u(A_{Th})}{A_{Th}}\right)^2+\left(\frac{u(m_{Th})}{m_{Th}}\right)^2+\left(\frac{uN_{Ac-228\,Std}}{N_{Ac-228\,Std}}\right)^2+\left(\frac{u(R_{Ac-228\,Std})}{R_{Ac-228\,Std}}\right)^2} \qquad (16)$$

The uncertainty on the measurement time is negligible.

Table 30: Uncertainty budget for the efficiency determination of ^{228}Ac in the Quantulus 1220^{TM}. The combined standard uncertainty is the quadratic sum of all components.

Uncertainty component	%
Chemical recovery of Ac-228	**5.3**
Counting statistics	**0.8**
Weighing	**0.2**
Th activity	**5.6**
Half-life Ac-228 (T1/2 = 6.13 h)	**1**
Combined standard uncertainty	**7.8**

Table 31: Uncertainty budget for ^{228}Ra in mineral water sample W1. The uncertainty budget shows the standard uncertainty components (1 *s*) for a single measurement.

Uncertainty component	%
Chemical recovery	6.1
Counting statistics (incl. background)	8.0
Efficiency	7.8
Volume	0.1
Half-life Ra-228 ($T_{1/2}$ = 6.13 h)	0.15
Half-life Ac-228 ($T_{1/2}$ = 6.13 h)	0.5
Combined standard uncertainty	**12.7**

Table 32: Uncertainty budget for ^{228}Ra in mineral water sample W2. The uncertainty budget shows the standard uncertainty components (1 *s*) for a single measurement.

Uncertainty component	%
Chemical recovery	6.01
Counting statistics (incl. background)	6.7
Efficiency	7.8
Volume	0.1
Half-life Ra-228 ($T_{1/2}$ = 6.13 h)	0.15
Half-life Ac-228 ($T_{1/2}$ = 6.13 h)	0.5
Combined standard uncertainty	**11.9**

For the second method of IRMM, use was made of sources prepared a year earlier for the determination of ^{226}Ra by α-particle spectrometry as described above. The basic idea is to separate and determine by α-particle spectrometry the ^{228}Th grown in as a granddaughter nuclide of ^{228}Ra after more than a year. The procedure is described in detail in Vasile et al. (2009).

Reference values for ^{226}Ra, ^{228}Ra, ^{234}U and ^{238}U

As mentioned in the beginning of chapter 3, the reference values were determined by consensus between the results of IRMM and those of the Bundesamt für Strahlenschutz (BfS), Department for Radioprotection and the Environment, Berlin. Two largely different, independent methods were applied by the two laboratories, three in the case of ^{228}Ra. The summary of laboratory results of IRMM and BfS and of the derived **comparison reference values A_{ref}** is given in Table 33.

As is outlined in chapter 2, the uncertainty of the reference values has to take a possible instability of the samples and inhomogeneity among them into account. This is expressed by the formula for the expanded uncertainty, U_{ref} (k = 2), of a reference value

$$U_{\text{ref}} = k \cdot \sqrt{u_{\text{char}}^2 + s_{\text{bb}}^2 + u_{\text{sts}}^2} \quad , \qquad (17)$$

where s_{bb} is the standard deviation in activity concentration between bottles of the same batch and u_{sts} is the short-term stability (for the duration of the comparison exercise) of the samples; u_{char} is the combined standard uncertainty of the mean of the two (or three, ^{228}Ra) measurement results from IRMM and BfS contributing to the reference value:

$$u_{\mathrm{char}} = \frac{\sqrt{\sum_{i=1}^{n} (u_{\mathrm{c},i})^2}}{n} \quad , \qquad (18)$$

where $u_{\mathrm{c,i}}$ is the combined standard uncertainty of the laboratory's (or method's) result (Pauwels et al., 1998).

As no wall adsorption could be found as source of a potential short-term instability, u_{sts} = 0 (compare chapter 2), only u_{char} and s_{bb} are contributing to the expanded uncertainty U_{ref} of the reference values A_{ref} given in Table 33.

Table 33: Activity concentrations in mineral waters determined by the two laboratories, IRMM and BfS, and **comparison reference values A_{ref}** calculated as arithmetic mean of these results (reference date 1 May 2006). Values A_{ref} given in brackets were not required to be determined by comparison participants. Uncertainties of laboratory results given as combined standard uncertainty u_{c}, uncertainty of reference values as expanded uncertainty $U_{\mathrm{ref}} = u \cdot k$ with a coverage factor k = 2, corresponding to a level of confidence of about 95 %.

Water	Nuclide	laboratory mean value with combined standard uncertainty u_{c}			**reference value A_{ref}** with expanded uncertainty U_{ref}
		activity concentration / mBq·L^{-1}			activity concentration / mBq·L^{-1}
		IRMM [1]	**IRMM** second method [2]	**BfS**	
W1	U-238	10.3 ± 0.2		12.1 ± 0.4	11.2 ± 1.7
	U-234	14.4 ± 0.3		15.7 ± 0.5	15.0 ± 2.2
	Ra-226	98 ± 6		97 ± 11	98 ± 15
	Ra-228	23.0 ± 2.3	29.1 ± 1.6	32 ± 5	28 ± 6
W2	U-238	0.91 ± 0.02		0.85 ± 0.06	(0.88 ± 0.29)
	U-234	3.94 ± 0.09		4.0 ± 0.2	(4.0 ± 0.5)
	Ra-226	42.0 ± 2.6		44 ± 5	43 ± 11
	Ra-228	66 ± 7	69.4 ± 3.7	87 ± 13	74 ± 12
W3-50	U-238	22.1 ± 0.5		21.2 ± 0.5	21.6 ± 1.9
	U-234	44.7 ± 1.0		42.3 ± 0.9	43.5 ± 3.0
	Ra-226	3.0 ± 0.2		4.3 ± 0.5	(3.6 ± 0.8)
	Ra-228	< 9	6.5 ± 0.6	6.9 ± 1.4	(6.7 ± 1.2)
W3-51	U-238	20.5 ± 0.8		---	20.5 ± 1.6
	U-234	40.9 ± 1.2		---	40.9 ± 2.4
	Ra-226	3.0 ± 0.2		4.3 ± 0.5	(3.6 ± 0.8)
	Ra-228	< 9	6.5 ± 0.6	6.9 ± 1.4	(6.7 ± 1.2)

[1] ^{228}Ra via liquid-scintillation counting of ^{228}Ac

[2] via ^{228}Th ingrowth and alpha-spectrometry

4. Methods used by the participating laboratories

Participants were free to use separation procedures and measurement methods of their own choice. Of the 45 laboratories, which reported results, not all determined all four radionuclides. Table 34 gives an overview of the measurement techniques used.

^{234}U and ^{238}U:

Thirty two laboratories (2, 3, 4, 8, 12, 13, 14, 17, 18, 22, 23, 25, 26, 27, 31, 32, 33, 34, 35 (W1 only), 36, 37, 38, 40, 41, 42, 43, 45, 47, 48, 49, 50, 51) used **α-particle spectrometry** for the determination of ^{234}U as well as ^{238}U (except for lab 38). All laboratories added isotopic tracer (^{232}U, in one case ^{236}U) at the beginning of the chemical procedure in order to determine the radiochemical yield of the procedure.

Except for one (lab 49), all laboratories preconcentrated uranium from the water samples and carried out radiochemical separation from the matrix components. For preconcentration from water volumes between 0.2 L and 6 L, nine laboratories applied evaporation (12, 14, 25, 26, 35, 36, 37, 41 and 51) and 17 laboratories carried out co-precipitation, using phosphates (2, 34, 40, 43, 45 and 47), $Fe(OH)_3$ (3, 13, 18, 23, 27, 33, 38, 48 and 50) and MnO_2 (22 and 31), respectively. Several laboratories did not report the correponding details. The radiochemical purification was performed with ion exchange chromatography, extraction chromatography and solvent extraction, respectively. Some laboratories used a combination of the mentioned techniques. Sources for alpha-particle spectroscopic measurement were prepared by electrodeposition (17 labs: 4, 17, 18, 23, 25, 26, 31, 33, 35, 37, 41, 42, 43, 45, 48, 50 and 51), micro-coprecipitation with rare earth fluorides (9 labs: 2, 12, 13, 14, 34, 36, 38, 40 and 47) and micro-coprecipitation with cadmium chloride (lab 22), respectively. Again, several laboratories did not report the details of source preparation.

The four laboratories (5, 20, 21 and 28) using **γ-ray spectrometry** for ^{238}U evaporated the water and measured the dry residue directly. The activity concentration of ^{238}U was determined via ^{234}Th (at 63 keV and 93 keV) assuming equilibrium between ^{238}U and its progeny ^{234}Th.

Lab 38 determined ^{238}U by **radiochemical neutron activation analysis**. After irradiation, the induced ^{239}U ($t_{1/2}$ = 23 min, E_γ = 74.5 keV) was separated with tributylphosphate in toluene from 5 M HNO_3. Addition of natural uranium to the sample after irradiation allowed determination of the radiochemical yield via ^{235}U at 185.7 keV.

Laboratory 7 determined the uranium isotopes with **inductively-coupled plasma mass spectrometry** (ICP-MS) from sample aliquots of 1 mL without sample preparation.

Three laboratories used **liquid scintillation counting** (LSC, labs 11, 29 and 35 (W3 only)) in α/β-discrimination mode. The ^{234}U and ^{238}U activity concentrations were evaluated applying spectral deconvolution of the uranium α-peaks.

Laboratories 16 and 46 determined total uranium using **fluorimetry** after evaporating the water samples and fusing the residues with a mixture of NaF and $NaCO_3$ at 900°C. The activity concentration of ^{238}U was calculated assuming the average natural isotopic composition and that of ^{234}U based on the assumption of equilibrium.

Table 34: Number of laboratories reporting results and measurement techniques used for determining the four radionuclides.

radionuclide	U-234	U-238	U tot	Ra-226	Ra-228
reported results (all methods)	36	41	2	41	32
reported results by method:					
α-particle spectr.	31 [1]	30 [1]		11	1 (via ^{224}Ra)
gross-α counting				3	
ICP-MS	1	1			
γ-ray spectr.		4		6	24
radiochemical NAA		1			
LSC	1 [2]	2 [2]		12	3
fluorimetry	2	2	2		
gross-β counting					4
sorption emanation technique				9	

[1] Plus one additional laboratory (Water-1 only)
[2] Plus one additional laboratory (Water-3 only)

^{226}Ra:

α-particle spectrometry was applied in eleven laboratories (2, 13, 18, 23, 26, 31, 36, 38, 41, 47 and 49), all of them including the use of tracers. One laboratory (no. 49) determined ^{226}Ra directly from a Ra-adsorbing disc (MnO_2) which had been immersed into the sample for 6 hours. The radiochemical recovery of 95 % was determined with an external ^{226}Ra standard. Laboratory 47 purified ^{226}Ra from unconcentrated water samples of 100 mL with ion exchange resins and prepared α-sources by micro-coprecipitation with $BaSO_4$. Lab 26 coprecipitated similar sources with $BaSO_4$ carrier from 100 mL samples merely evaporated from volumes of 1 L.

Eight laboratories used a large variety of coprecipitation techniques for preconcentration: three labs with MnO_2 (no. 13, 18, 31), two as $Pb(Ra)(Ba)SO_4$ (labs 2, 38), one lab each with $Pb(NO_3)_2$ (no. 41), with $BaSO_4$ and $Fe(OH)_3$ (no. 23) and with $Pb(CrO_4)_4$ (no. 36). Labs 2, 13 and 38 coprecipitated radium sources with $BaSO_4$ directly after the preconcentration step. Laboratories 31 and 41 reported purification of radium from the other precipitated radionuclides by ion exchange chromatography. Sources for α-spectrometric measurement were prepared by electrodeposition on stainless steel discs, and the recovery of the radiochemical procedure was determined with the tracer ^{225}Ra (via its progeny ^{217}At) in equilibrium with ^{229}Th (lab 41, 68 %). Lab 31 (tracer ^{224}Ra) obtained a recovery of 30 % for its radiochemical procedure. Lab 23 purified radium from coprecipitated uranium and thorium by extraction chromatography on a Microthene TOPO column and subsequently separated radium from barium with an ion exchange resin. Sources were prepared by electrodeposition, a recovery of 75 % was obtained (tracer ^{225}Ra in equilibrium with ^{229}Th). Labs 18 and 36 separated radium from interfering radionuclides in the precipitates by solvent extraction using TBP or DTPA complexation, respectively, followed by precipitation with $BaCl_2$ or $BaSO_4$ for source preparation.

Where the radium sources for α-spectrometry were coprecipitated with $BaSO_4$ or $BaCl_2$ (labs 26, 47, 2, 13, 38, 18 and 36), ^{133}Ba added to the sample as tracer before preconcentration enabled determination of the chemical recovery with γ-ray spectrometry. The yields obtained varied for the different procedures between 25 % and 90 %.

Six laboratories applied **γ-ray spectrometry** (labs 5, 19, 20, 21, 24 and 28), five of them evaporating the water samples (between 3 L and 12 L) to dryness. Only one lab (no. 19) preconcentrated radium by coprecipitation with $BaSO_4$. The $Ba(Ra)SO_4$ precipitate or residues after evaporation were sealed and ^{226}Ra was determined by the 351 keV and 609 keV γ-ray lines of its ^{214}Pb and ^{214}Bi progenies after an ingrowth period of at least 3 weeks. Lab 19 assumed a radiochemical recovery of the radium coprecipitation of 100 %.

Nine laboratories (4, 25, 27, 29, 33, 34, 39, 43 and 46) applied the **sorption emanation technique** similar to the procedure described above for BfS (chapter 3 – ^{226}Ra). One laboratory (no. 43), however, did not carry out a preconcentration step, and lab 33 coprecipitated radium with calcium as phosphomolybdate instead of using $BaSO_4$. Chemical yields obtained varied from 65 % to 100 %.

Twelve laboratories (3, 7, 8, 11, 12, 15, 22, 32, 35, 42, 45 and 48) used the **liquid scintillation technique** for measurement after different preconcentration procedures had been applied. Laboratories 7, 8, 15, 22, 32 and 42 coprecipitated radium with MnO_2 or as $Ba(Ra)SO_4$, respectivelly. The precipitate was purified in order to remove interfering radionuclides, dissolved and mixed with scintillation cocktail. The scintillation vial was tightly closed and stored up to 1 month (labs 15, 32 and 42) to allow ingrowth of ^{222}Rn and its daughters. Lab 22 measured radium as soon as possible after sample preparation. Laboratories 3 and 11 preconcentrated with freeze drying, mixed the sample with a liquid scintillation cocktail, and stored for 1 month for radon ingrowth. Laboratories 35 and 45, on the other hand, thermally preconcentrated the sample desorbing at the same time all dissolved radon, followed by the usual waiting time in scintillation vials. Laboratory 12 mixed the sample with a mineral oil scintillator and measured directly by LSC. Laboratory 48 used the Empore Radium Rad disc which selectively extracts radium from an acidic solution, here with 0.01 M EDTA.

Laboratories 16, 17 and 37 applied one of the preconcentration varieties of coprecipitating radium with lead sulfate and/or $BaSO_4$, and the determination of ^{226}Ra was carried out by measuring the **gross-α activity** of the $Ba(Ra)SO_4$ precipitate on a filter using a low- background gas proportional counter.

^{228}Ra:

About half of the laboratories used evaporation to a certain reduced volume or to dryness as preconcentration step. A bit less than half performed coprecipitation, where $Ba(Ra)SO_4$ was the most often formed precipitate. Some of the labs applied a combination of preconcentration procedures or added purification steps; Figure 16 gives an overview of the methods used.

Not all laboratories determined the recovery of their concentration and purification procedures. Some used ^{133}Ba as tracer or stable Ba in form of added carrier, determined by photometry or ICP-MS. The laboratory which determined ^{228}Ra via α-particle spectrometry of ^{224}Ra used ^{225}Ra in equilibrium with ^{229}Th as a tracer. A few laboratories used external standards (^{226}Ra, ^{228}Ra) or gravimetry to determine the chemical yield.

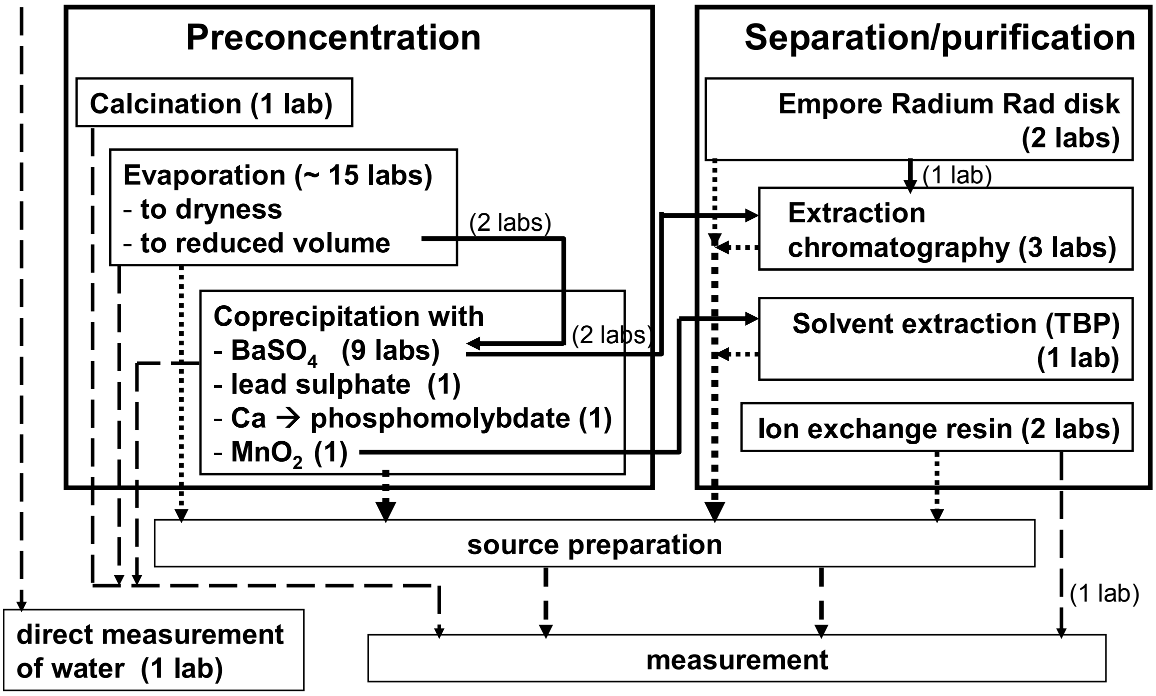

Fig. 16: Summary of preconcentration and radiochemical separation procedures used for the determination of ^{228}Ra by the comparison participants.

5. Reported results

In total, 45 laboratories reported results in this comparison exercise, although not all of them measured all four radionuclides requested. The participants were asked to perform duplicate analysis of all radionuclides. Therefore, it was possible to report up to 2 individual results per radionuclide and per sample in the reporting form (Annex 2 – Result reporting form). If more than one result was submitted, the unweighted laboratory mean A_{lab} of the reported values was calculated by us and taken into account in the evaluations below (starting with Figure 27).

The number of laboratories that have reported results and the number of individual values submitted for a certain radionuclide and per sample is given in Table 35.

Table 35: Reported results in the comparison exercise.

Nuclide	Nr. of results	W1	W2	W3-50	W3-51
^{226}Ra	Labs	41	41	–	–
	Individual values	77	77 (1<DL)*	–	–
^{228}Ra	Labs	32	32	–	–
	Individual values	56 (6<DL)*	56 (6<DL)*	–	–
^{234}U	Labs	36	–	20	15
	Individual values	67 (1<DL)*	–	36	28
^{238}U	Labs	41	–	21	19
	Individual values	74 (1<DL)*	–	37	35

*Results below the detection limit (DL)

Figures 17 to 26 show the individual activity concentrations (decay-corrected to the reference date 1 May 2006) and expanded uncertainties (with a coverage factor k = 2) as they were reported by the participants. If a coverage factor different from 2 was reported, we recalculated the expanded uncertainty for k = 2. The solid red lines indicate the reference activity concentration and the dashed lines the expanded uncertainty ± U_{ref} (k = 2) of the reference value. Results below the detection limit are represented with symbols on the abscissa.

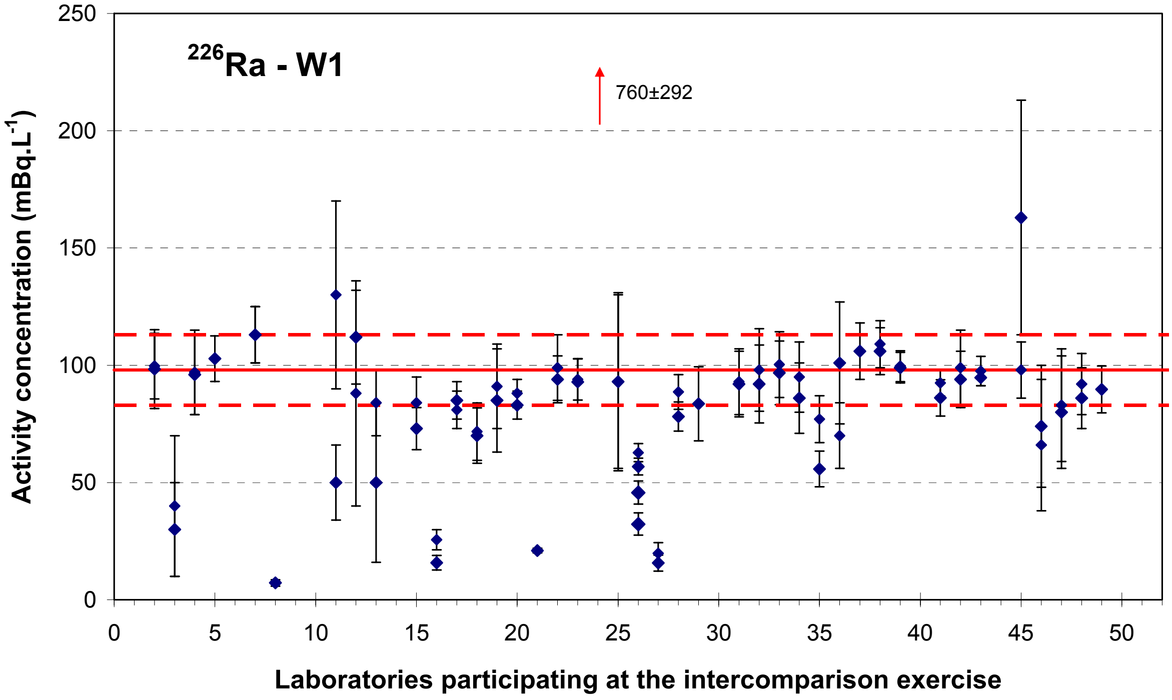

Fig. 17: Measured ^{226}Ra activity concentration in Water 1 (W1). Error bars represent expanded uncertainty (with k = 2) of individual measurements. Red lines are reference value A_{ref} ± expanded uncertainty U_{ref} (k = 2).

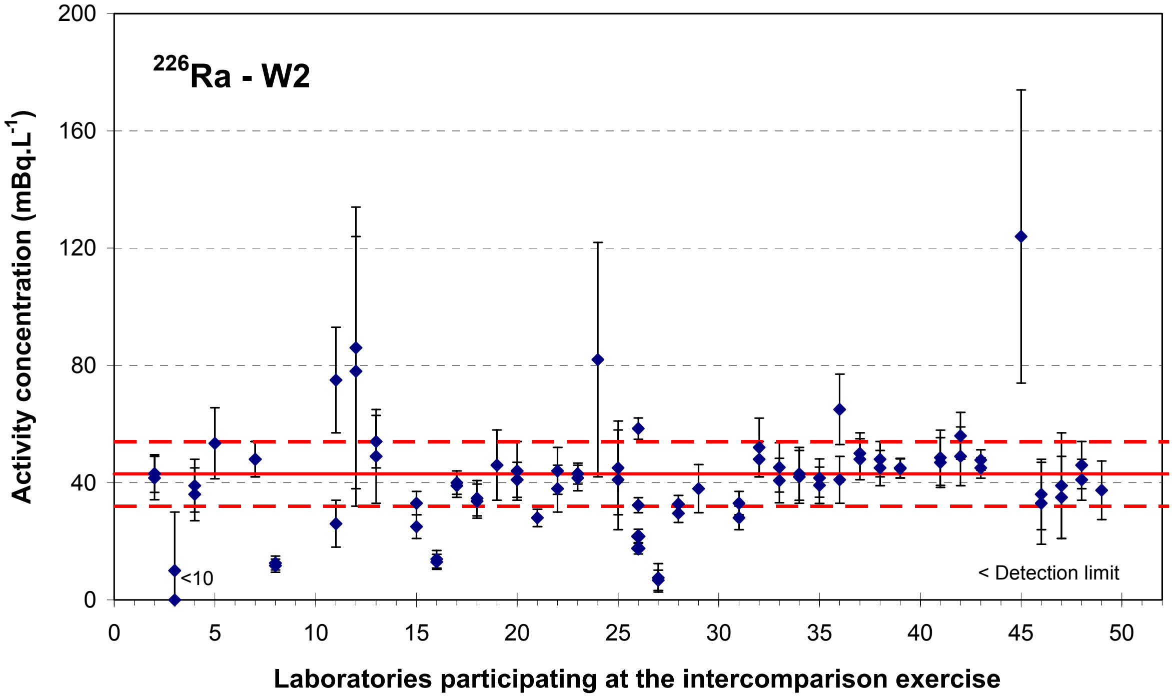

Fig. 18: Measured ^{226}Ra activity concentration in Water 2 (W2).
Error bars, reference value and its uncertainty as in Fig. 17.

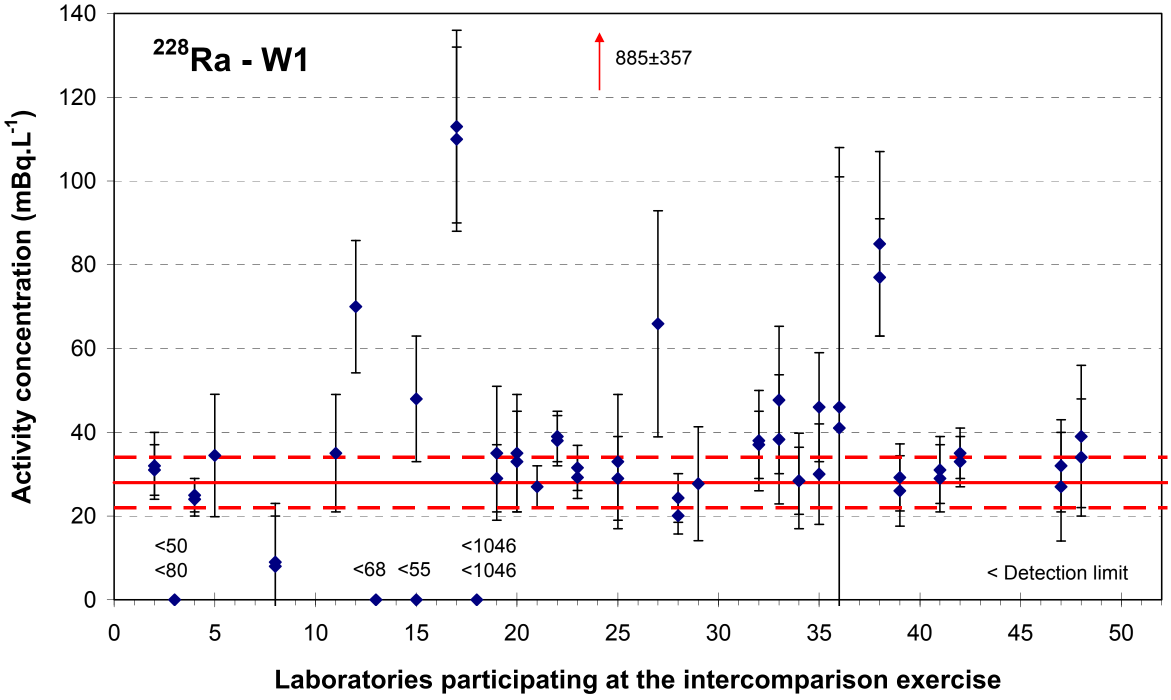

Fig. 19: Measured ^{228}Ra activity concentration in Water 1 (W1).
Error bars, reference value and its uncertainty as in Fig. 17.

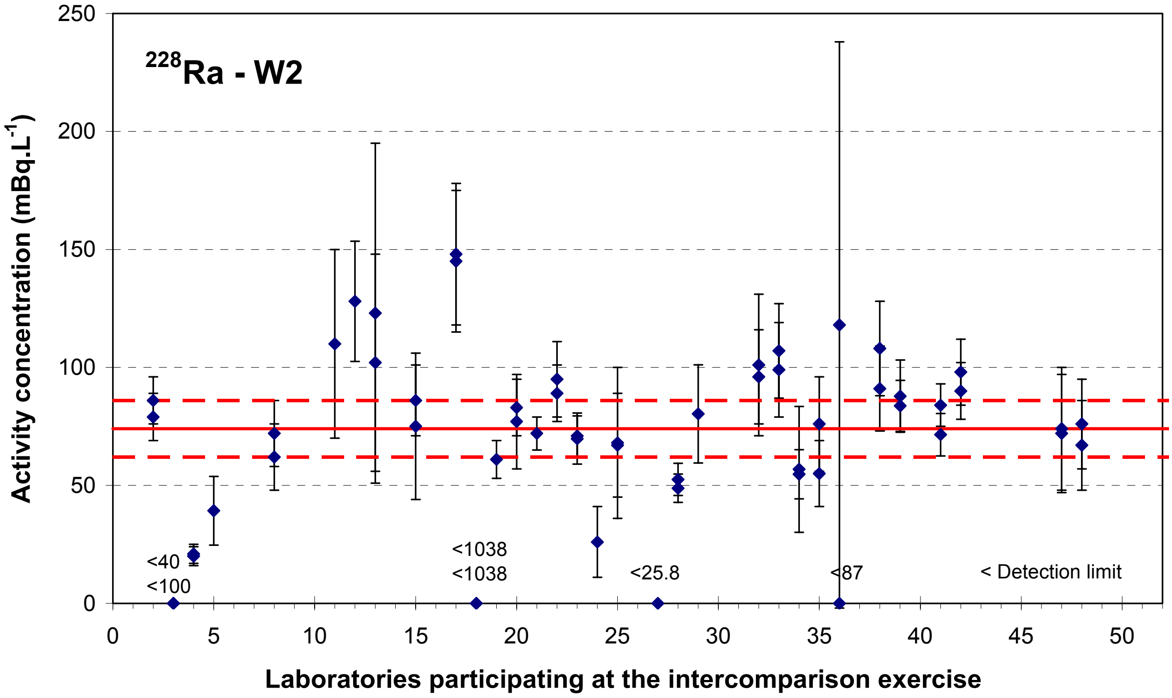

Fig. 20: Measured ^{228}Ra activity concentration in Water 2 (W2).
Error bars, reference value and its uncertainty as in Fig. 17.

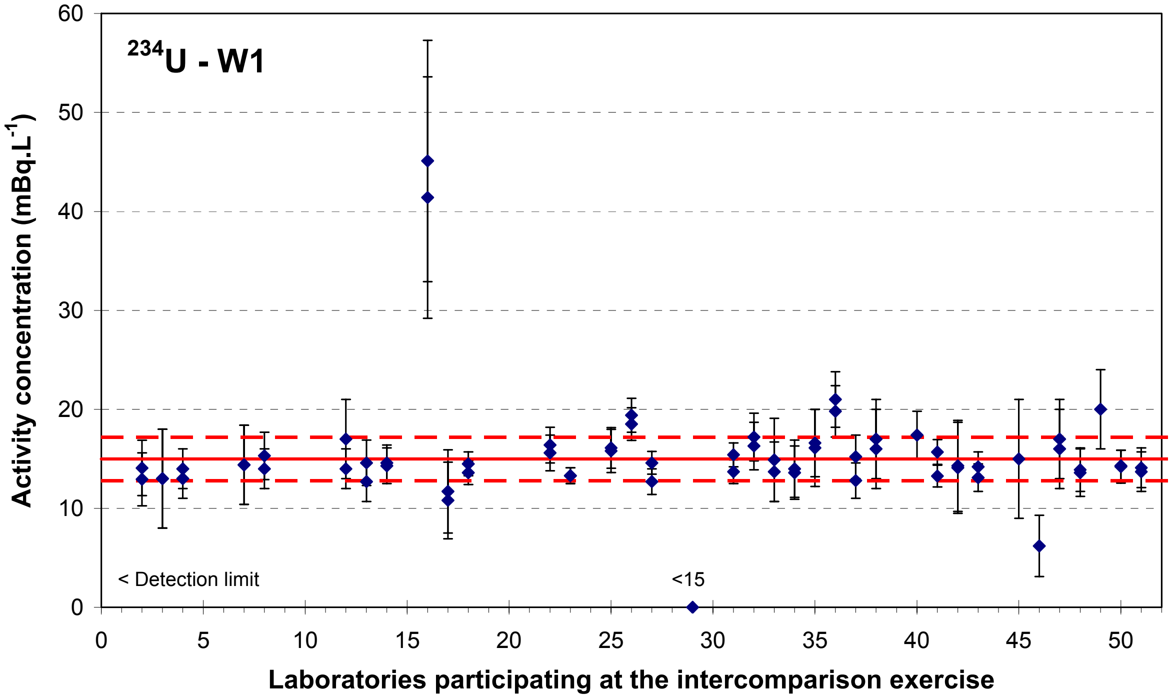

Fig. 21: Measured ^{234}U activity concentration in Water 1 (W1).
Error bars, reference value and its uncertainty as in Fig. 17.

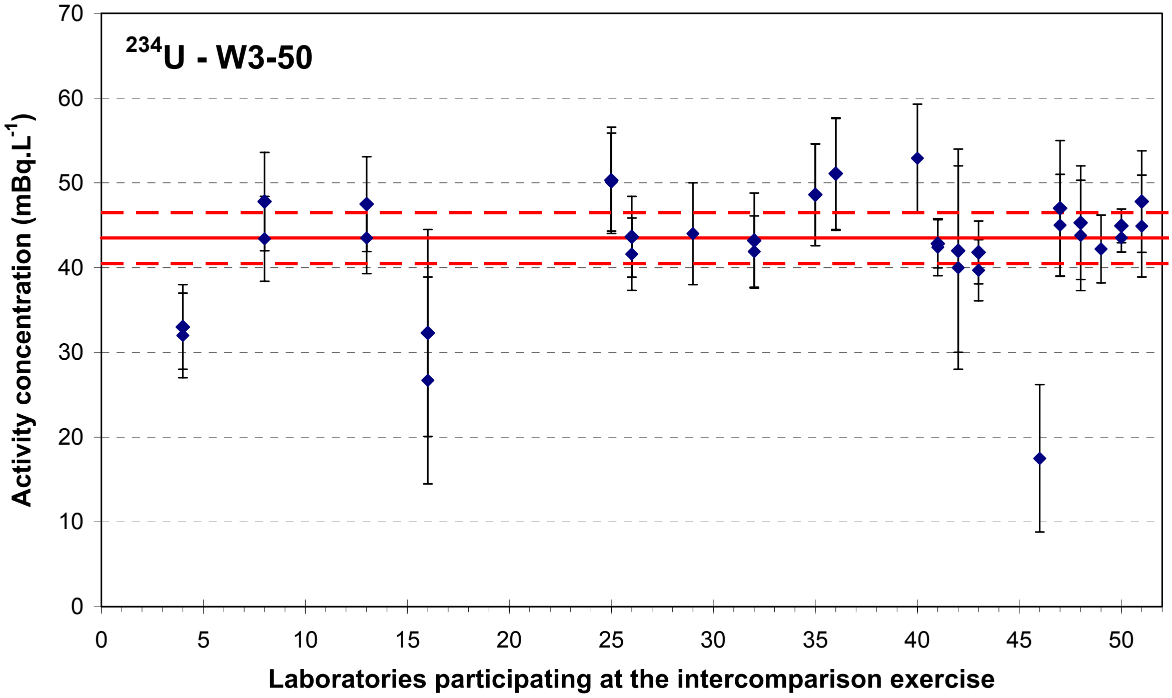

Fig. 22: Measured ^{234}U activity concentration in Water 3-50 (W3-50).
Error bars, reference value and its uncertainty as in Fig. 17.

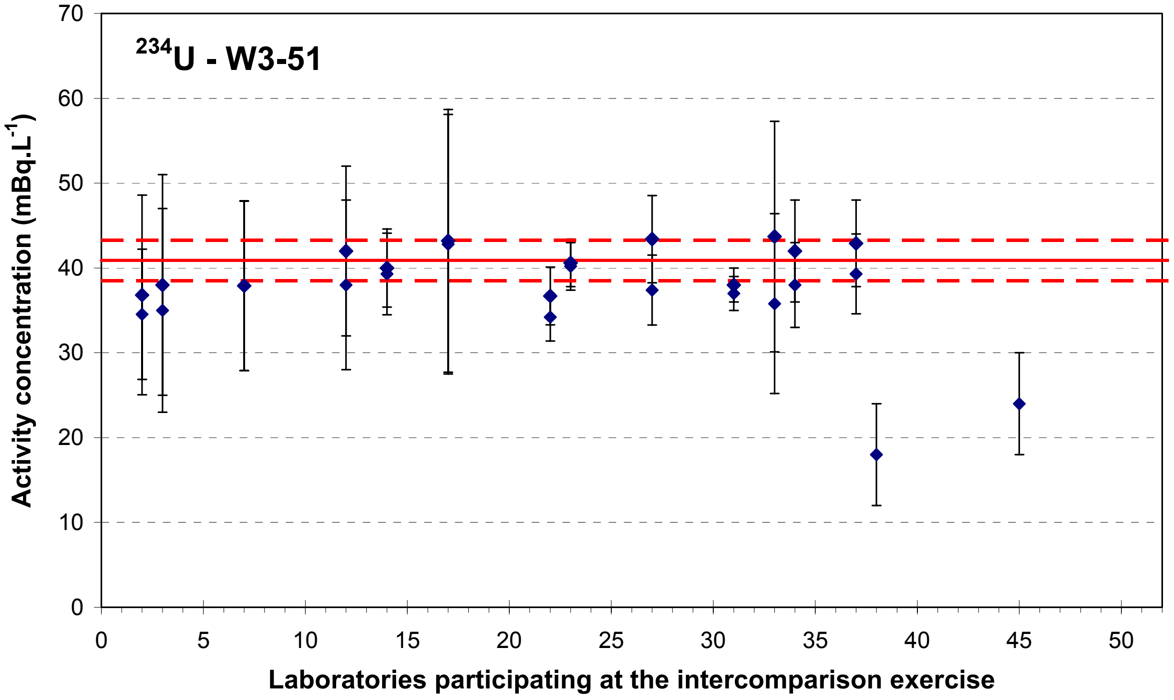

Fig. 23: Measured ^{234}U activity concentration in Water 3-51 (W3-51). Error bars, reference value and its uncertainty as in Fig. 17.

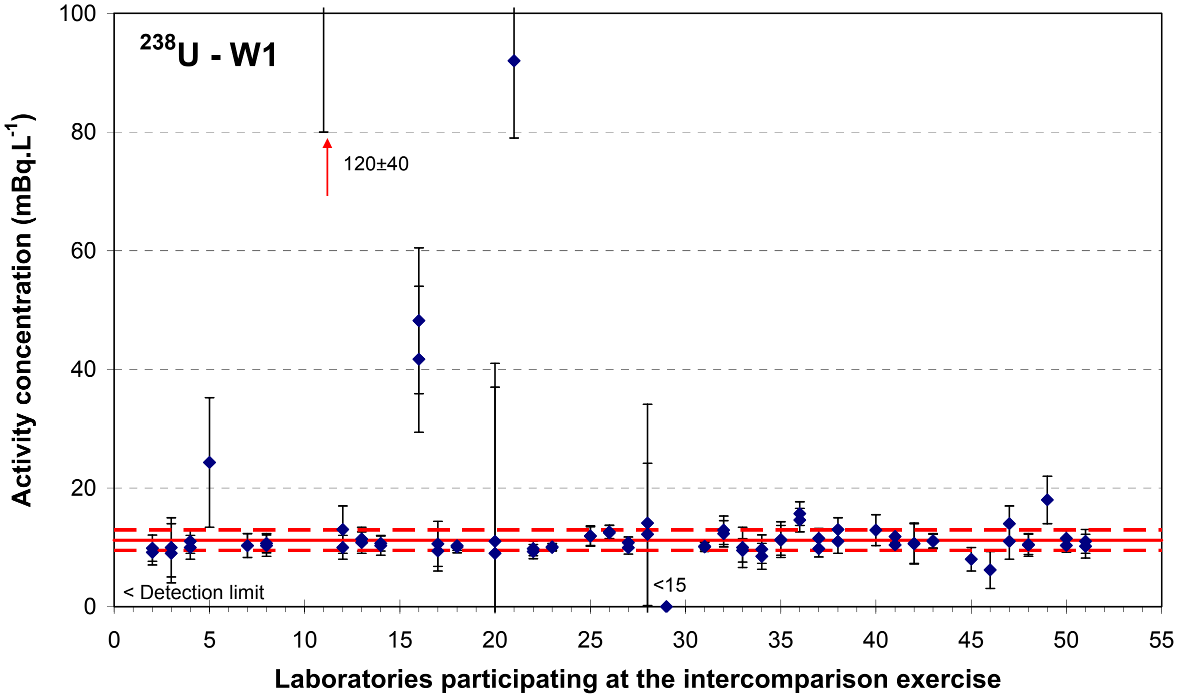

Fig. 24: Measured ^{238}U activity concentration in Water 1 (W1). Error bars, reference value and its uncertainty as in Fig. 17.

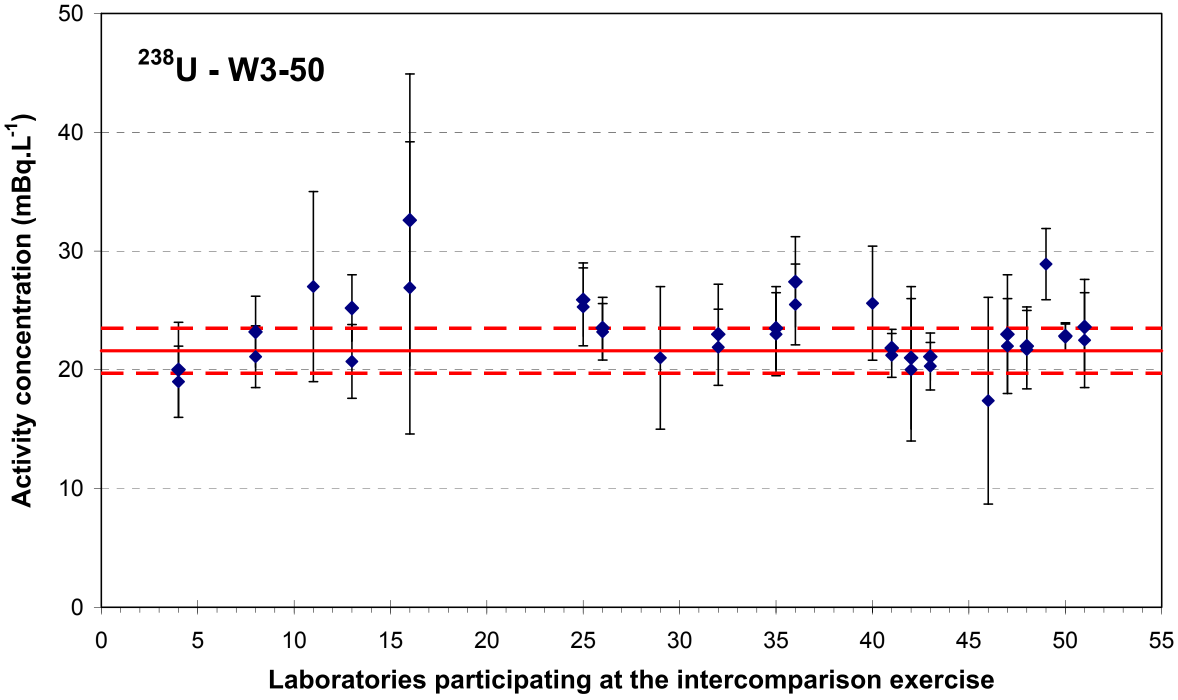

Fig. 25: Measured ^{238}U activity concentration in Water 3-50 (W3-50). Error bars, reference value and its uncertainty as in Fig. 17.

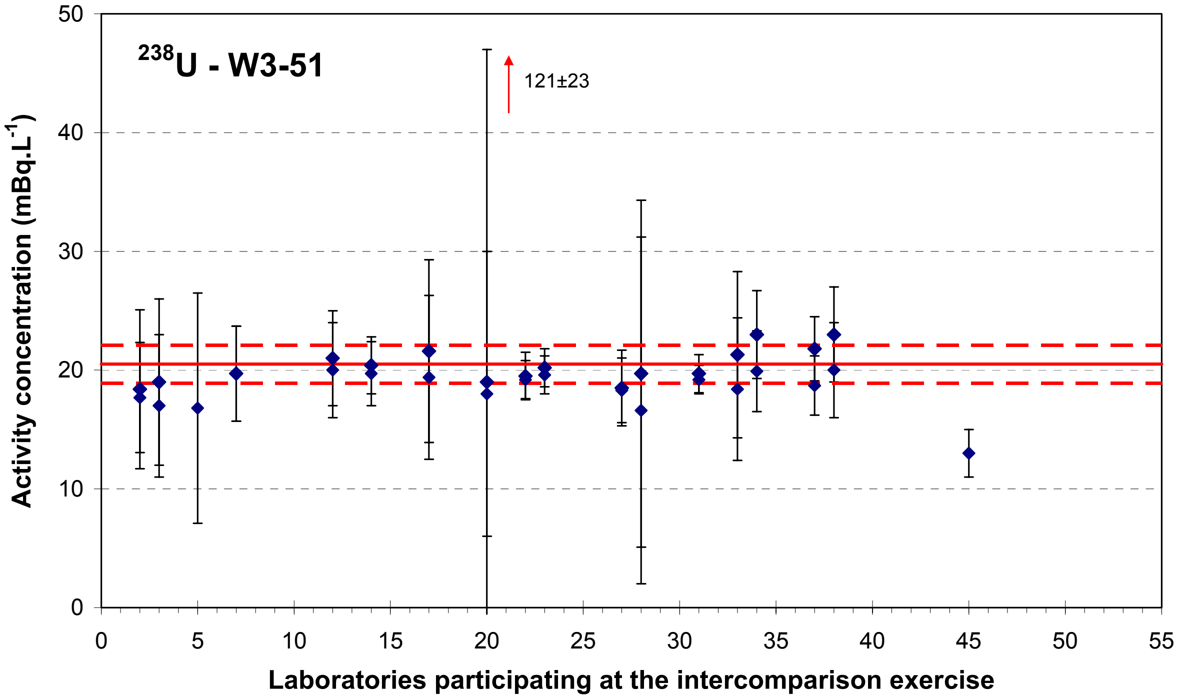

Fig. 26: Measured ^{238}U activity concentration in Water 3-51 (W3-51). Error bars, reference value and its uncertainty as in Fig. 17.

As seen from Figs. 17 to 20, many discrepant results for ^{226}Ra and ^{228}Ra were obtained. In the case of Water 1 (W1) the ^{226}Ra results were often lower than the reference value. One laboratory (no. 24) reported far too high values for ^{226}Ra in W1 (indicated with an arrow on Fig. 17), while their result for sample W2 agrees with the reference value within the expanded uncertainty (Fig. 18). Laboratories 8 and 21 reported results for W1 much lower than the reference value with very small uncertainties (error bars invisible on the graph, Fig. 17). Laboratory 26 reported four results for ^{226}Ra in both water samples (W1 and W2). It is interesting to note that their results are discrepant and do not agree within their expanded uncertainties (Figs. 17 and 18). Laboratory 3 reported two results for ^{226}Ra in W2 as one of them is below the detection limit of their method (< 10 mBq·L^{-1}) (Fig. 18, symbol on the abscissa).

The reported ^{228}Ra activity concentrations were often too high in both water samples (W1 and W2). Similar to the ^{226}Ra results, laboratory 24 reported far too high value for ^{228}Ra in W1 (indicated with an arrow on Fig. 19), while their result for sample W2 is lower than the reference value (Fig. 20). A large number of laboratories reported results deviating from the reference value by more than the sum of its expanded uncertainty U_{ref} and the expanded uncertainty of the individual measurement results. In total, six laboratories (no. 3, 13, 15, 18, 27, 36) reported results below the detection limit of the method used. The reported values are indicated with symbols on the abscissa and the limit of detection is given on top of it (Figs. 19 and 20).

In the case of both uranium isotopes, better agreement of the reported results with the reference values was observed. Only one laboratory (no.16) reported ^{234}U activity concentrations in W1 much higher than the reference value (Fig. 21). Laboratory 46 reported ^{234}U results lower than the reference value in both water samples (Figs. 21 and 22). Another two participants (no. 38 and 45) also determined ^{234}U activity levels deviating from the reference value by more than the sum of its expanded uncertainty U_{ref} and the expanded uncertainty of the individual measurement results (Fig. 23). Laboratory 29 reported only a detection limit below 15 mBq·L^{-1} (Fig. 21, symbol on the abscissa).

Nearly all reported ^{238}U results for Water 3 (W3-50 and W3-51) agreed with the reference value within the expanded uncertainties (Figs. 25 and 26). Laboratory 21 with far too high results asked to withdraw their results, albeit after having been informed about a large deviation of its results. The participant reported that the samples have not been treated according to the routinely used analytical procedure. One laboratory (no. 45) determined ^{238}U activity level in W3-51 lower than the reference value (Fig. 26). In the case of Water 1 (W1), three of the participants (no. 11, 16 and 21) determined much higher activity concentrations (Fig. 23). Again, laboratory 29 reported that the activity concentration of ^{238}U in the sample is below the detection limit of their method (< 15 mBq·L^{-1}) (Fig. 24, symbol on the abscissa).

The estimation of the expanded uncertainty U_{lab} of the mean laboratory result was based on the information given by the participants in the result reporting form (Annex 2) and in the questionnaire (Annex 3). Laboratories were requested to give the individual expanded uncertainty (and the coverage factor applied) together with the measurement results in the reporting form, and to provide the full uncertainty budget for one measurement of each nuclide and for one sample in the questionnaire. A non-exhaustive list of uncertainty contributions served as budget template for information. Moreover, participants were invited to send their complete uncertainty budget in a free format of their own choice.

The submitted uncertainty budgets were analysed and compared with the numerical values reported with the measurement results. When discrepancies between the individual uncertainty values and the uncertainty budget were observed (or no budget at all was presented), the concerned laboratories were contacted to review their budgets (or send such if not submitted earlier). Not all of the contacted laboratories responded. Every two of three laboratories reported uncertainty values which were consistent with the given budget. Still, in about 30 % discrepancies between the given uncertainty budget and the reported uncertainty values were observed. The final number of uncertainty budgets not submitted, inconsistent with the reported uncertainties as well as the total number of budgets submitted per radionuclide and per sample is given in Table 36. Admittedly, the budget template provided by IRMM in the questionnaire was not of great help to some of the laboratories for estimating correctly the uncertainty of the activity determination.

Table 36: Overview of the uncertainty budgets provided in the comparison exercise.

Nuclide	**^{226}Ra**	**^{228}Ra**	**^{234}U**	**^{238}U**
Submitted	38	29	32	36
Consistent	26	20	21	24
Inconsistent	12	8	11	12
Not submitted	3	3#	4*	5*

#2 of the laboratories did not submit an uncertainty budget as their result is below MDA for all samples
*1 of the laboratories did not submit an uncertainty budget as their result is below MDA for all samples

When estimating the expanded uncertainties U_{lab} of the laboratory mean values, two cases had to be distinguished. For the majority of the participants the expanded uncertainty U_{lab} (with k = 2) of the mean activity concentration A_{lab} was calculated based on the information given in the uncertainty budget (approach 1). The following formula was applied:

$$U_{lab} = k \cdot \sqrt{\frac{u_{count}^{2}}{n} + u_{other}^{2}} \qquad (19)$$

where

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

It is obvious from Eq. (19) that only the counting uncertainty was considered as a random component and, consequently, is divided by √n in the combined uncertainty.

For all radionuclides u_{other} is determined from:

$$u_{other} = \sqrt{u_{sample\,prep}^{2} + u_{tracer}^{2} + u_{efficiency}^{2} + u_{acts\tan dards}^{2} + u_{blank}^{2} + u_{other\,contrib}^{2}} \qquad (20)$$

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

$u_{sample\ prep}$	propagated uncertainty due to uncertainty in sample preparation, separation etc.;
u_{tracer}	propagated uncertainty due to uncertainty in tracer activity;
$u_{efficiency}$	propagated uncertainty due to uncertainty in efficiency of the detection system;

$u_{act.\ standards}$	propagated uncertainty due to uncertainty in the activity efficiency standards;
u_{blank}	propagated uncertainty due to uncertainty in blank and background measurement;
$u_{other\ contrib}$	other propagated uncertainty contributions.

Equation (20) may lead to an overestimation of u_{other} and U_{lab} in cases where a propagated contribution, e.g. $u_{sample\ prep}$, represents a random component if this is not taken care of by dividing by $\sqrt{n}$.

As the laboratories were asked to fill in an uncertainty budget only for one sample, the u_{other} was considered as a systematic component for all water samples. Hence, the counting uncertainty u_{count} for the sample for which no information was available was calculated from the submitted combined uncertainty u_{lab} and the systematic uncertainty u_{other}:

$$u_{count} = \sqrt{u_{lab}^2 - u_{other}^2} \qquad (21)$$

The expanded uncertainties U_{lab} of the laboratory mean values were calculated from the uncertainty budgets following the approach described in the cases where the uncertainties reported with the individual results were found to be consistent with the budget and also in the cases when discrepancies between the individual uncertainty values and the uncertainty budget were observed.

In the case of three laboratories, the expanded uncertainty U_{lab} was calculated by us using the uncertainty budgets submitted although only one result was reported (lab 19 for ^{226}Ra and ^{228}Ra results in W2; lab 11 – ^{228}Ra and ^{238}U results; lab 36 - ^{228}Ra in W2). In the case of lab 11, the reported combined uncertainty was calculated as an arithmetic mean of the individual uncertainty contributions. Lab 19 reported the values of the counting uncertainty as combined uncertainty. Finally, lab 36 submitted an example budget for ^{228}Ra in W2 in which the calculated combined uncertainty was by about a factor of two lower than the reported value.

Only in very few cases (laboratories 3, 16, 25 and 47), where the observed discrepancies between the individual uncertainty values and the uncertainty budget were too big (no. 25 and 47) or no budget at all was presented (labs 3 and 16), a second approach was used*. In that case, U_{lab} was calculated by us as an arithmetic mean (if n > 1) of the reported individual expanded uncertainties (setting the coverage factor, if not given like that, to k = 2).

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[*] Lab 25 did not want to make any changes to the budgets already reported. Lab 47 did not reply to the request for revision. Labs 3 and 16 did not submit any uncertainty budget after being contacted to send such.

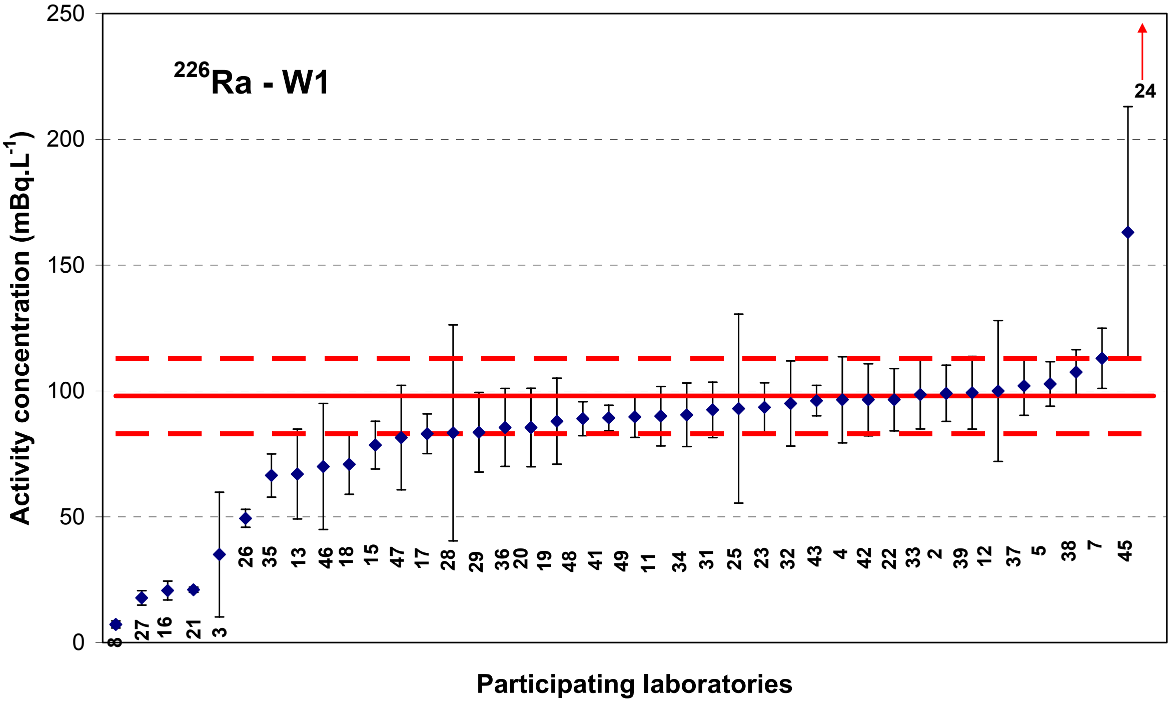

Fig. 27: Mean laboratory results A_{lab} for ^{226}Ra activity concentration in Water 1 (W1). Error bars indicate expanded uncertainty U_{lab} (k = 2) of laboratory mean, red lines are reference value A_{ref} ± expanded uncertainty U_{ref} (k = 2). Laboratory numbers are indicated.

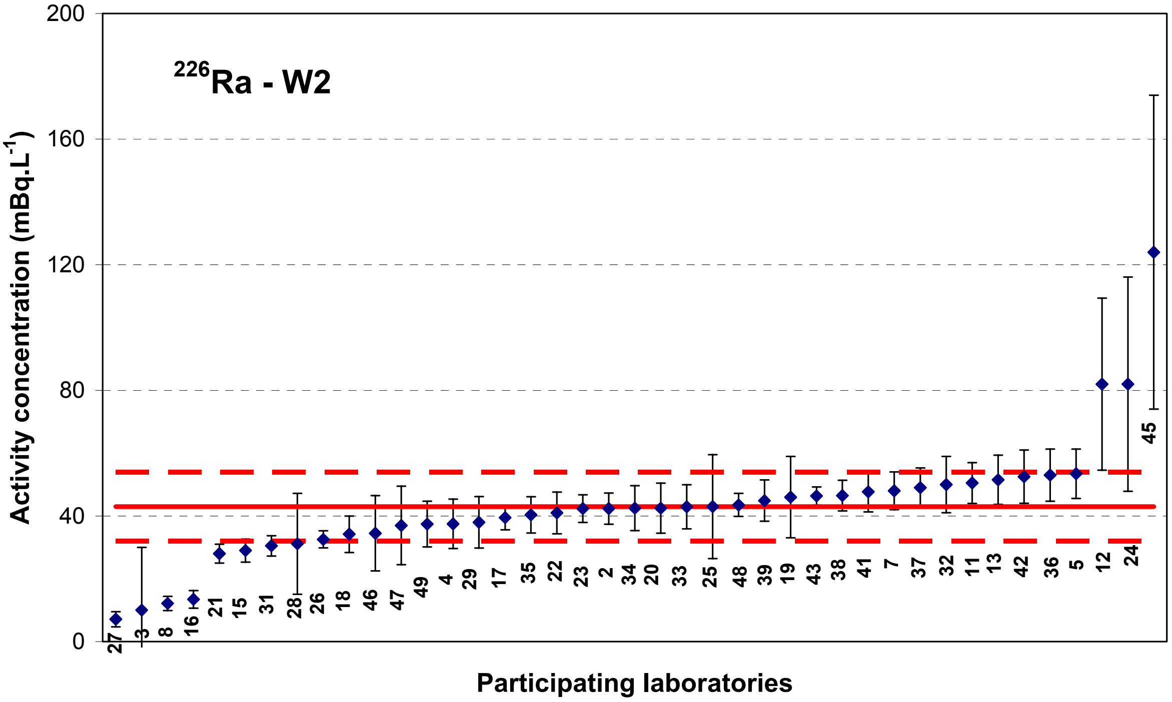

Fig. 28: Mean laboratory results A_{lab} for ^{226}Ra activity concentration in Water 2. Error bars, reference value, its uncertainty and lab numbers as in Fig. 27.

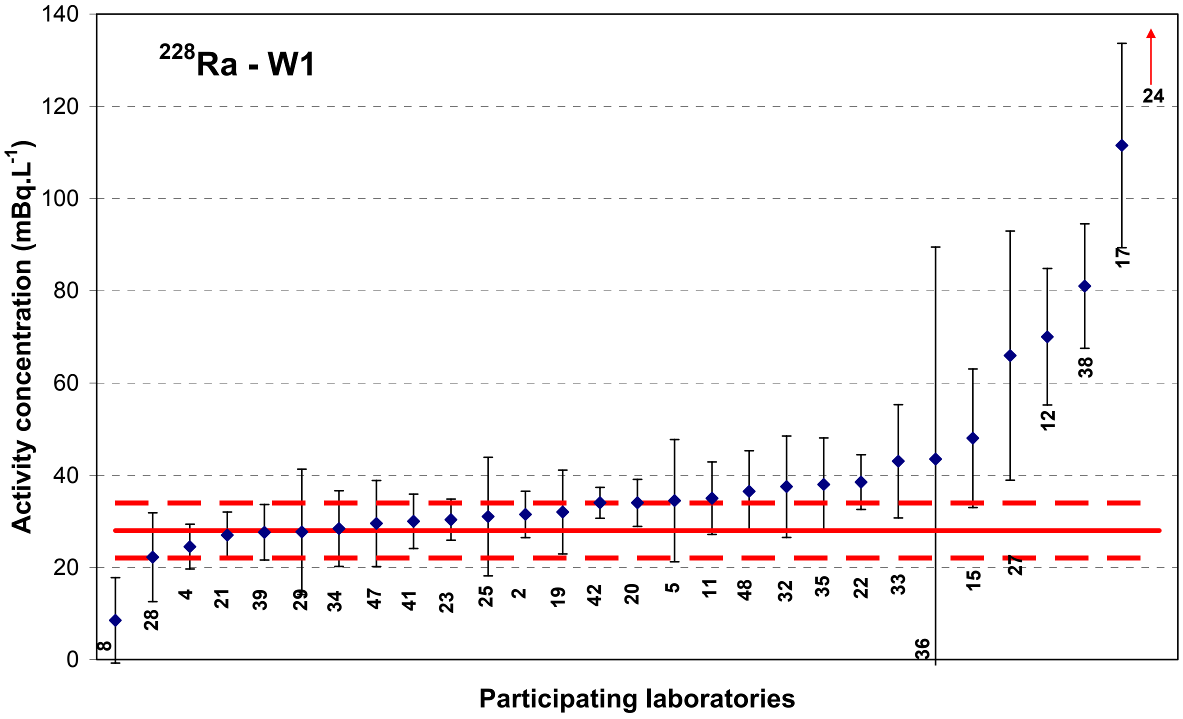

Fig. 29: Mean laboratory results A_{lab} for ^{228}Ra activity concentration in Water 1. Error bars, reference value, its uncertainty and lab numbers as in Fig. 27.

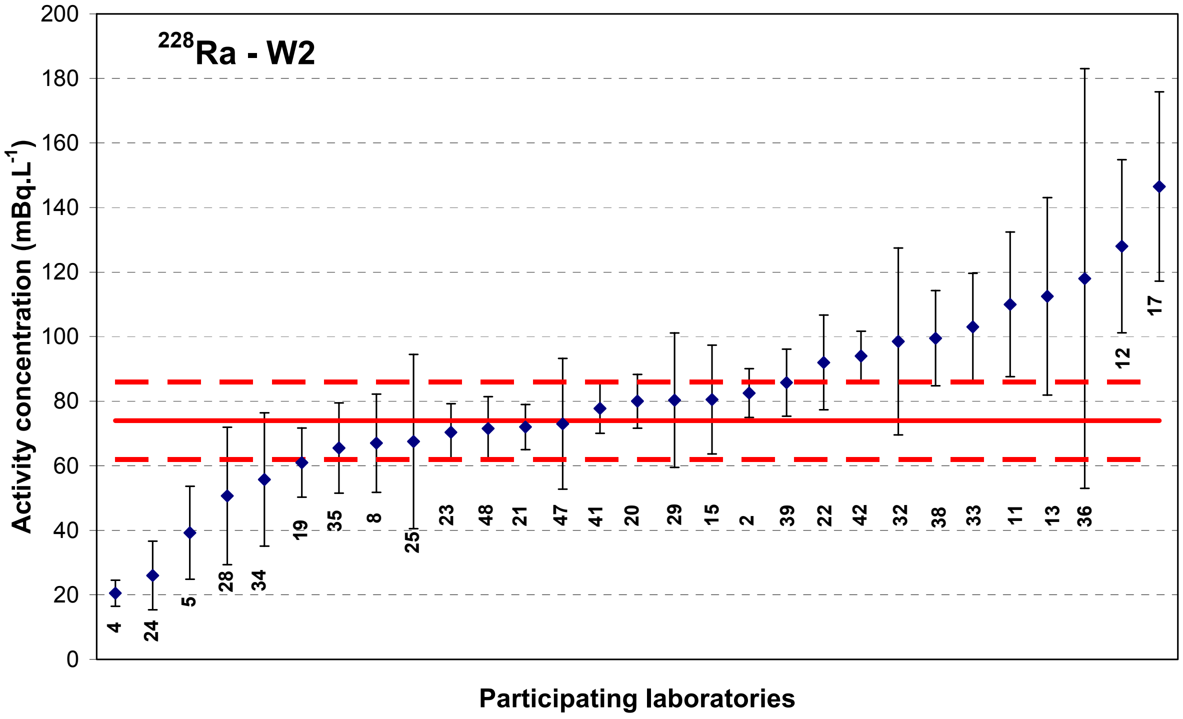

Fig. 30: Mean laboratory results A_{lab} for ^{228}Ra activity concentration in Water 2. Error bars, reference value, its uncertainty and lab numbers as in Fig. 27.

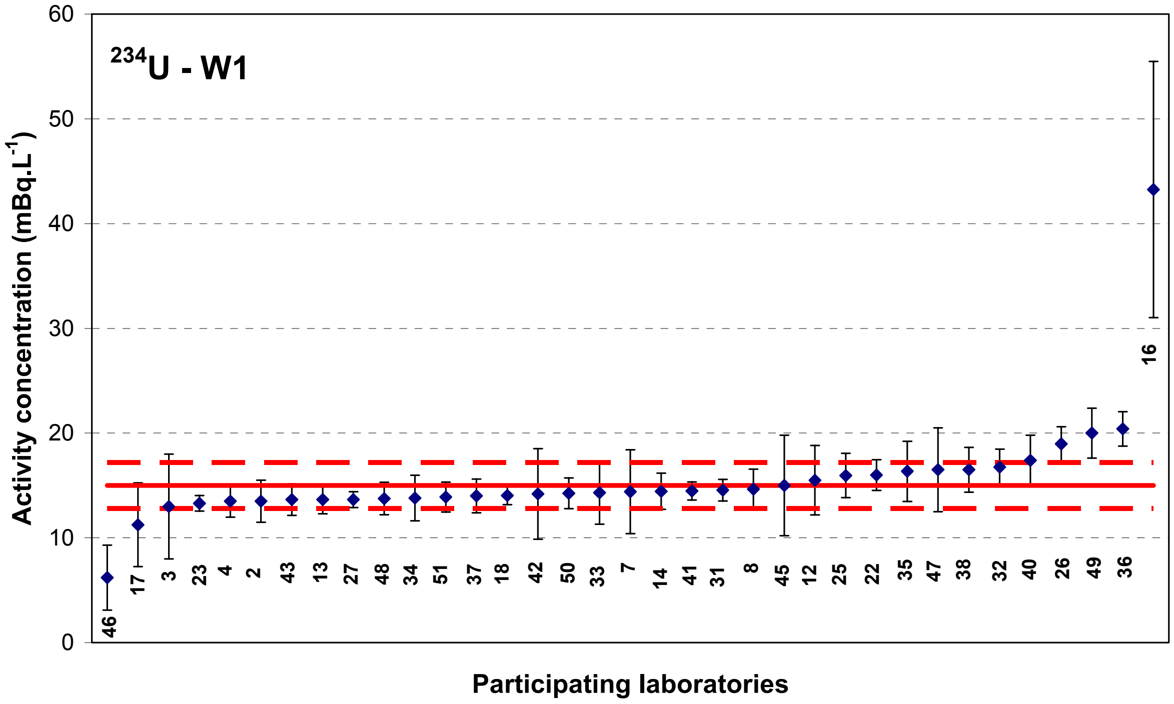

Fig. 31: Mean laboratory results A_{lab} for ^{234}U activity concentration in Water 1. Error bars, reference value, its uncertainty and lab numbers as in Fig. 27.

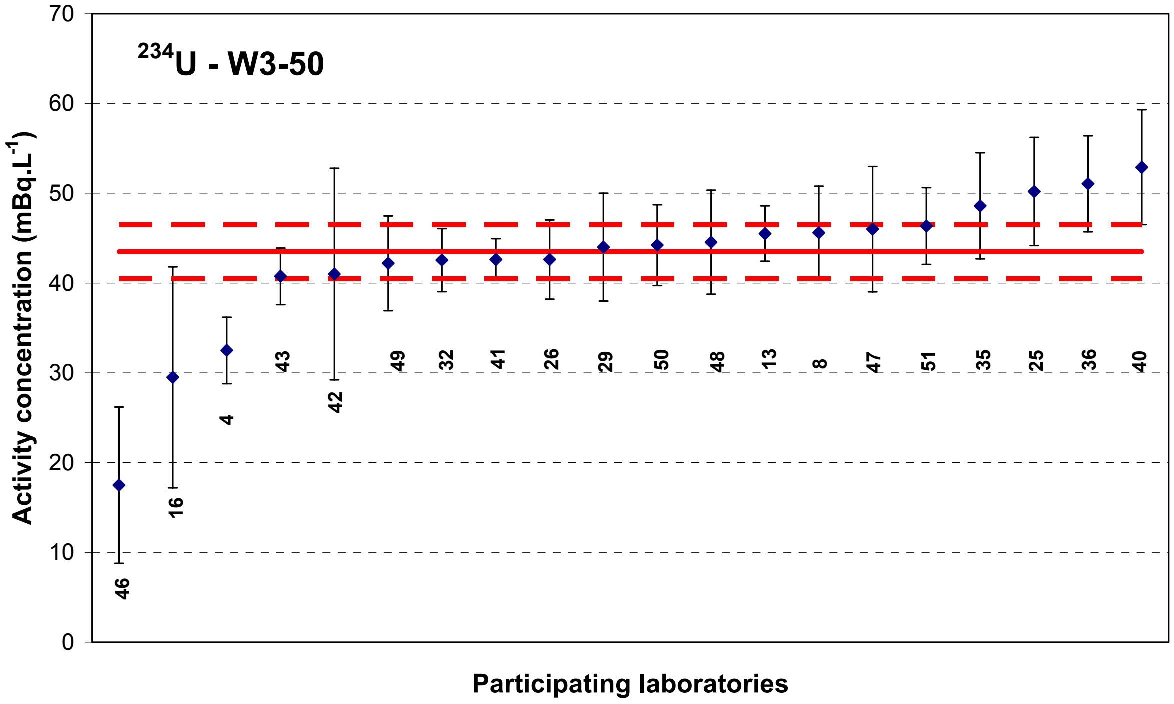

Fig. 32: Mean laboratory results A_{lab} for ^{234}U activity concentration in Water 3-50. Error bars, reference value, its uncertainty and lab numbers as in Fig. 27.

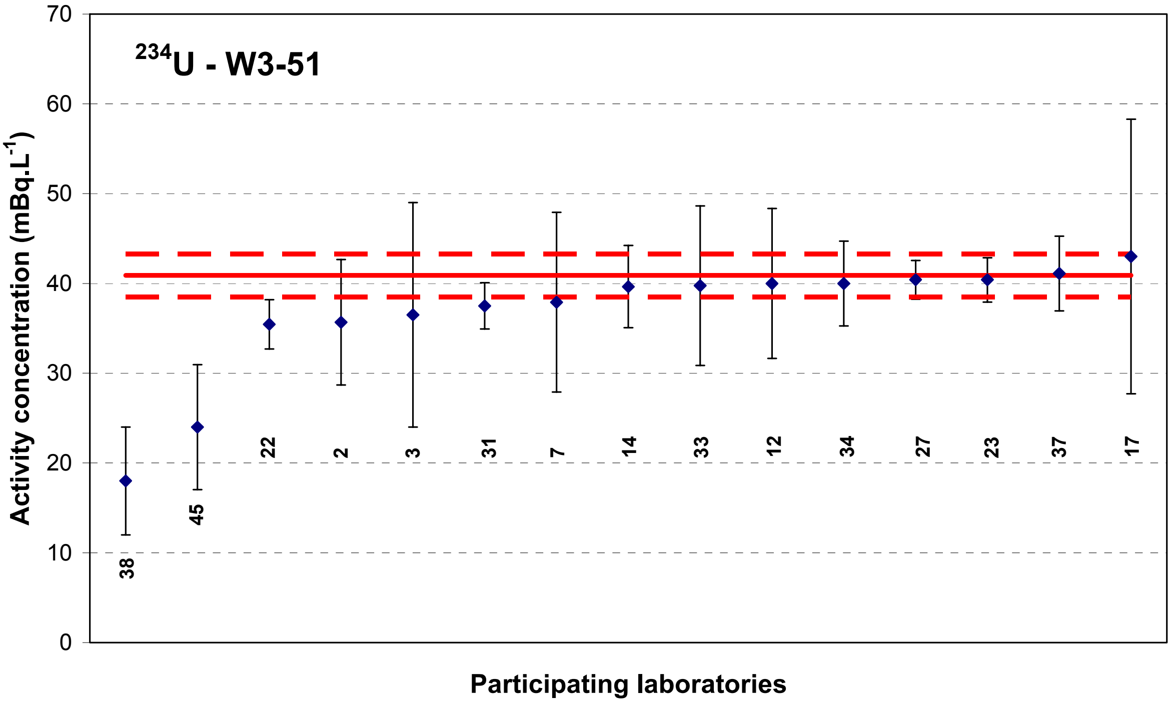

Fig. 33: Mean laboratory results A_{lab} for ^{234}U activity concentration in Water 3-51. Error bars, reference value, its uncertainty and lab numbers as in Fig. 27.

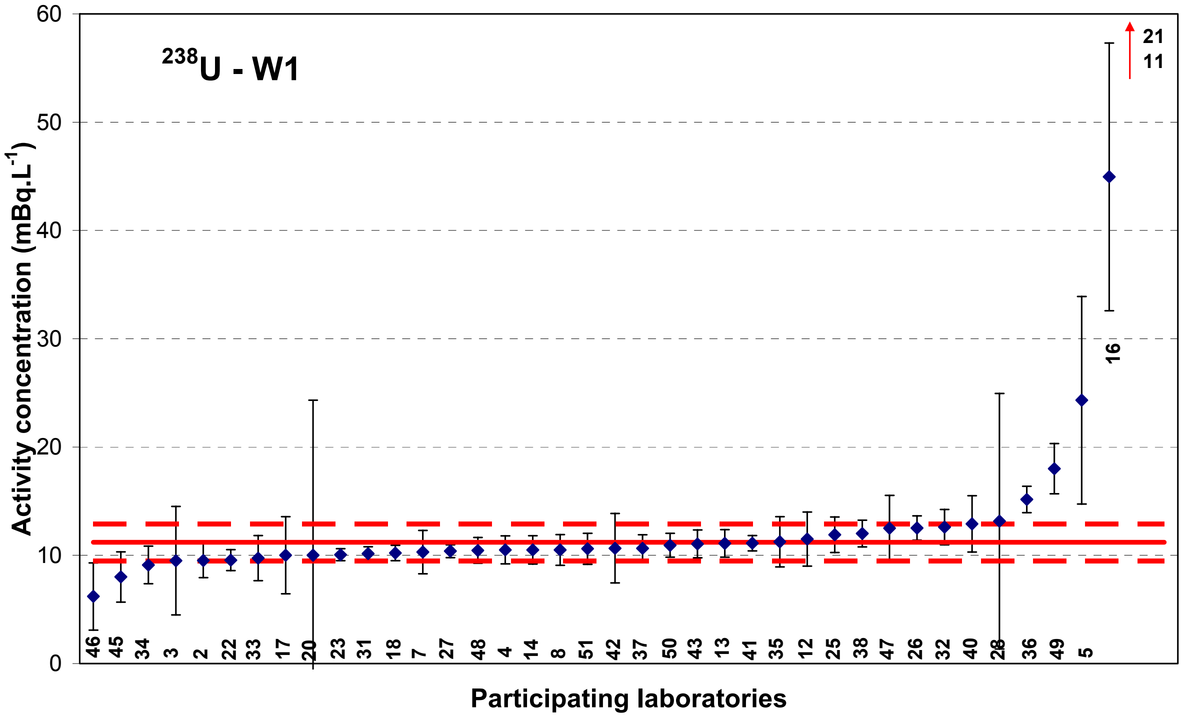

Fig. 34: Mean laboratory results A_{lab} for ^{238}U activity concentration in Water 1. Error bars, reference value, its uncertainty and lab numbers as in Fig. 27.

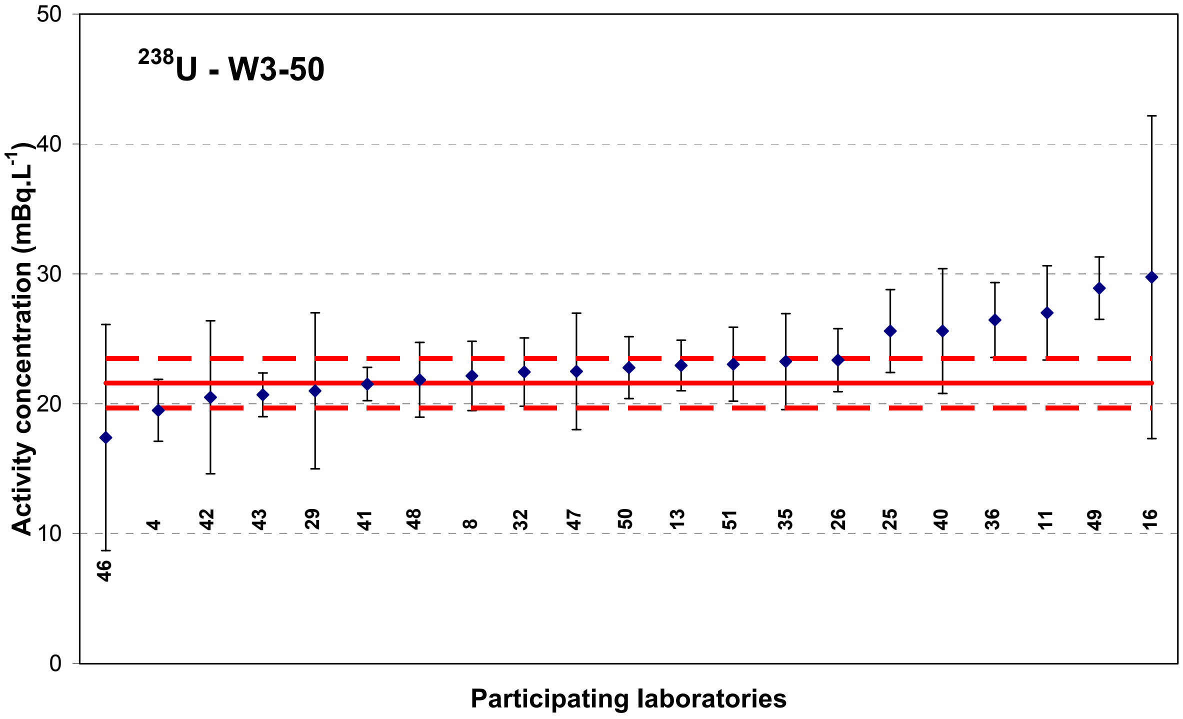

Fig. 35: Mean laboratory results A_{lab} for ^{238}U activity concentration in Water 3-50. Error bars, reference value, its uncertainty and lab numbers as in Fig. 27.

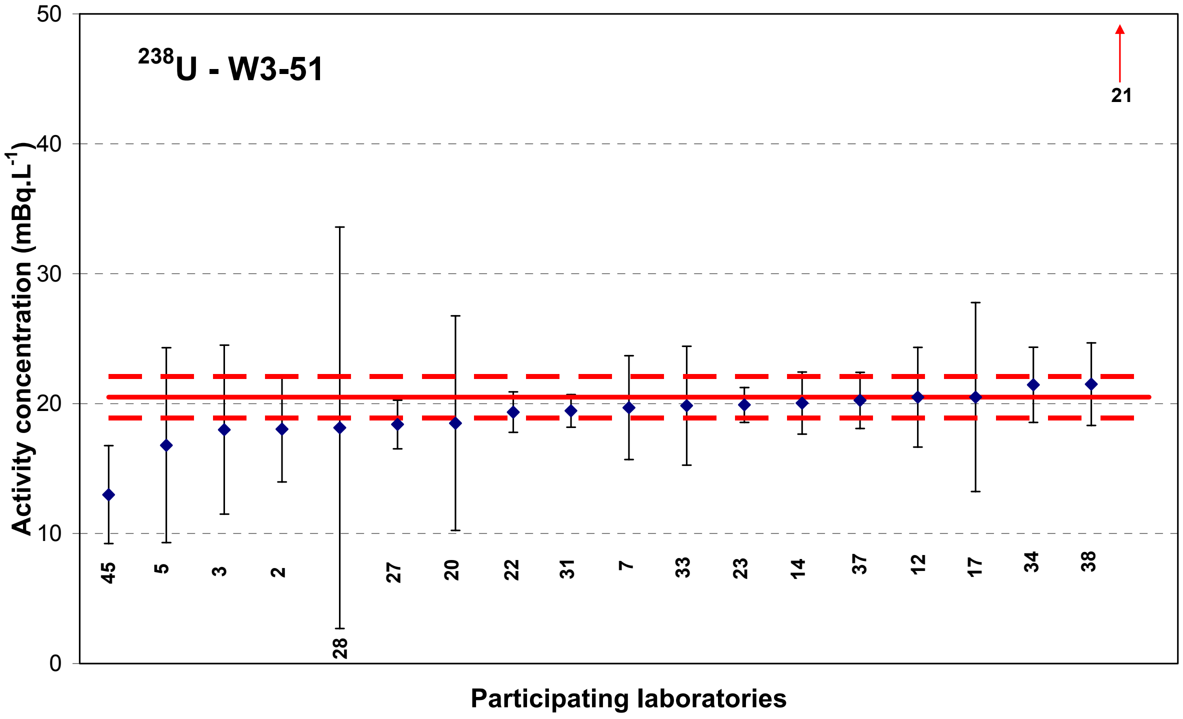

Fig. 36: Mean laboratory results A_{lab} for ^{238}U activity concentration in Water 3-51. Error bars, reference value, its uncertainty and lab numbers as in Fig. 27.

Tables 37 to 44 give in detail for each participating laboratory the number of measurements n, the unweighted laboratory mean A_{lab} of the reported values and its standard deviation s. Furthermore, the expanded uncertainty U_{lab} and its relative value U_{lab}/A_{lab} are given. The uncertainty values U_{lab} printed normally or in bold were calculated by us based on the reported uncertainty budget (approach 1 above). The values in bold indicate uncertainty budgets not consistent with the reported uncertainties. Values of U_{lab} printed in grey were generated using approach 2.

In Figures 27 to 36 the mean activity concentration A_{lab} with its corresponding expanded uncertainty U_{lab} (k = 2) obtained by one of the approaches described above are plotted in ascending order. Again, the solid line indicates the reference activity concentration A_{ref}, and its expanded uncertainty ± U_{ref} (k = 2) is plotted with dashed lines. Laboratory numbers are indicated with the results.

Apart from clearly wrong results (e.g. labs 8, 16, 21, 24 and 27 for ^{226}Ra in W1 and several for the other radionuclides and samples), it is obvious from Figs. 27 to 36 that some laboratories underestimate their uncertainties for one or more radionuclides (e.g. lab 26 for ^{226}Ra in W2).

Furthermore, an attempt is made to distinguish results obtained with the different methods of separation and measurement. Figs. 37 to 46 show the results plotted in groups of counting methods. It is obvious that in each group there are outlying values but almost always also laboratories which manage to obtain results within the uncertainty limits of the reference value. The group of laboratories using fluorimetry for the determination of the uranium isotopes renders the relatively largest proportion of deviating results. The two laboratories (no. 16 and 46) applying this method measured natural uranium and determined the activity concentrations of ^{234}U and ^{238}U by calculations, supposing an equilibrium. The participants (no. 16 and 46) applying this method are too small a group to allow reliable conclusions to be drawn. Nevertheless, a critical review of the implementation of the method should be done by these two laboratories. The results of this comparison, however, do not justify earmarking a particular counting method as being superior or inferior to others.

Although difficult due to the variety and complex nature of separation methods, an attempt is made to group the results by separation procedures (Figs. 47 to 56). The most important conclusion is similar to that for counting methods: For each separation procedure there is a reasonably large number of laboratories which succeed to obtain results close to the reference value, even though some labs in most groups produce largely deviating results. Again, no particular separation procedure can be identified as the source for deviating results, but the reason must be sought in the individual laboratories concerned. Only a small number of results could not be attributed to a particular separation method (“no info”).

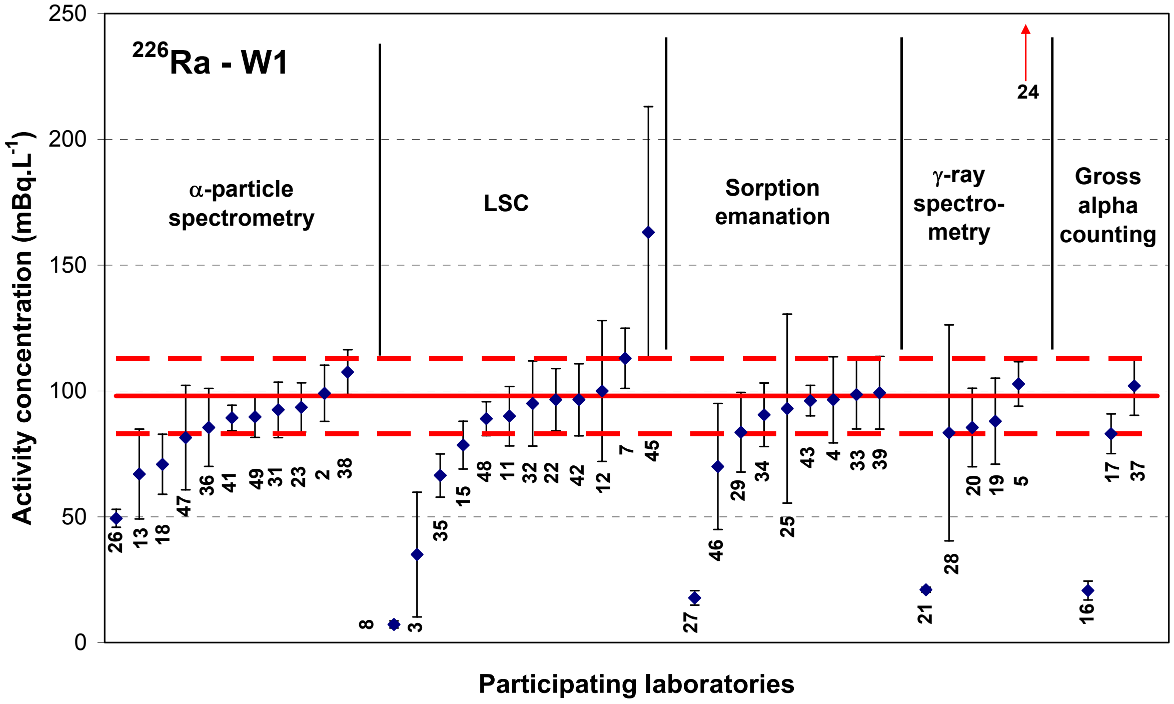

Fig. 37: Same as Fig. 27 but mean laboratory results A_{lab} for ^{226}Ra activity concentration sorted according to counting method

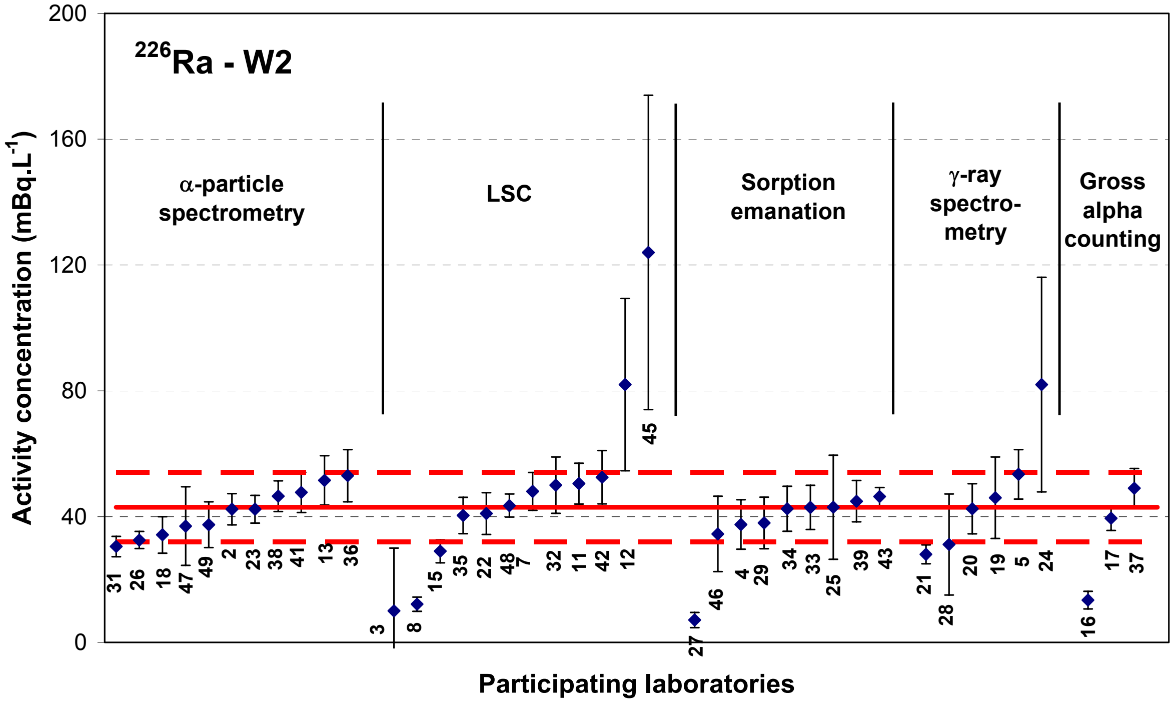

Fig. 38: Same as Fig. 28 but mean laboratory results A_{lab} for ^{226}Ra activity concentration sorted according to counting method

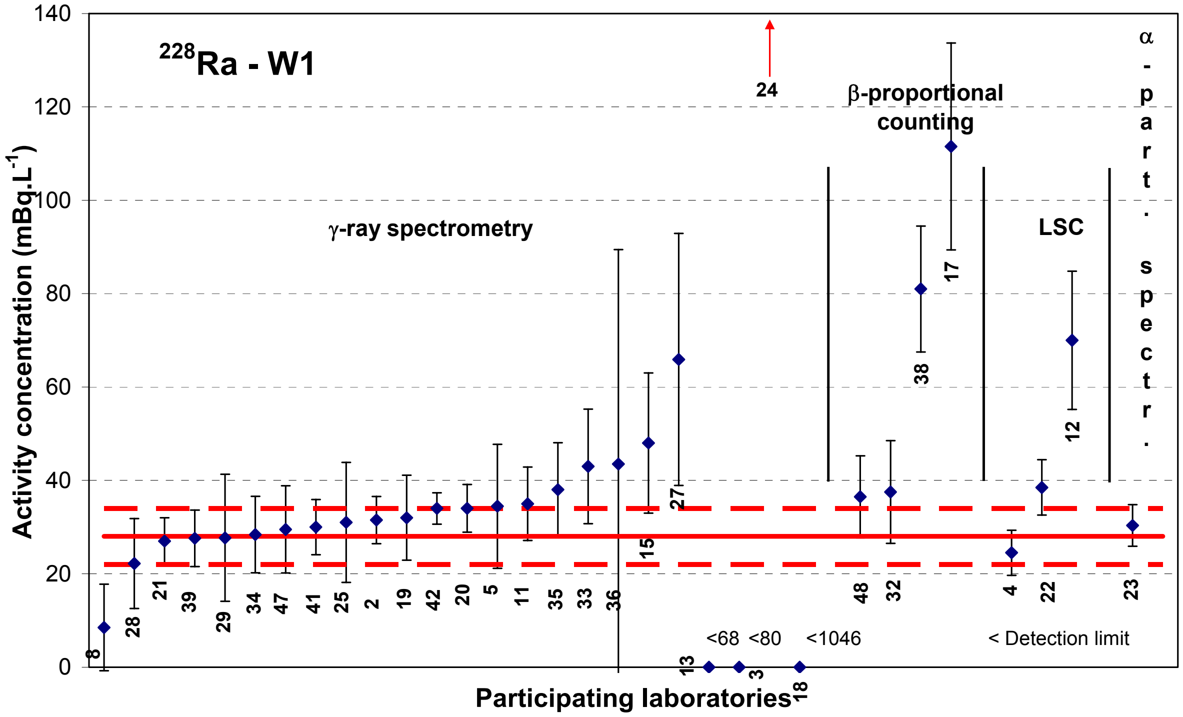

Fig. 39: Same as Fig. 29 but mean laboratory results A_{lab} for ^{228}Ra activity concentration sorted according to counting method

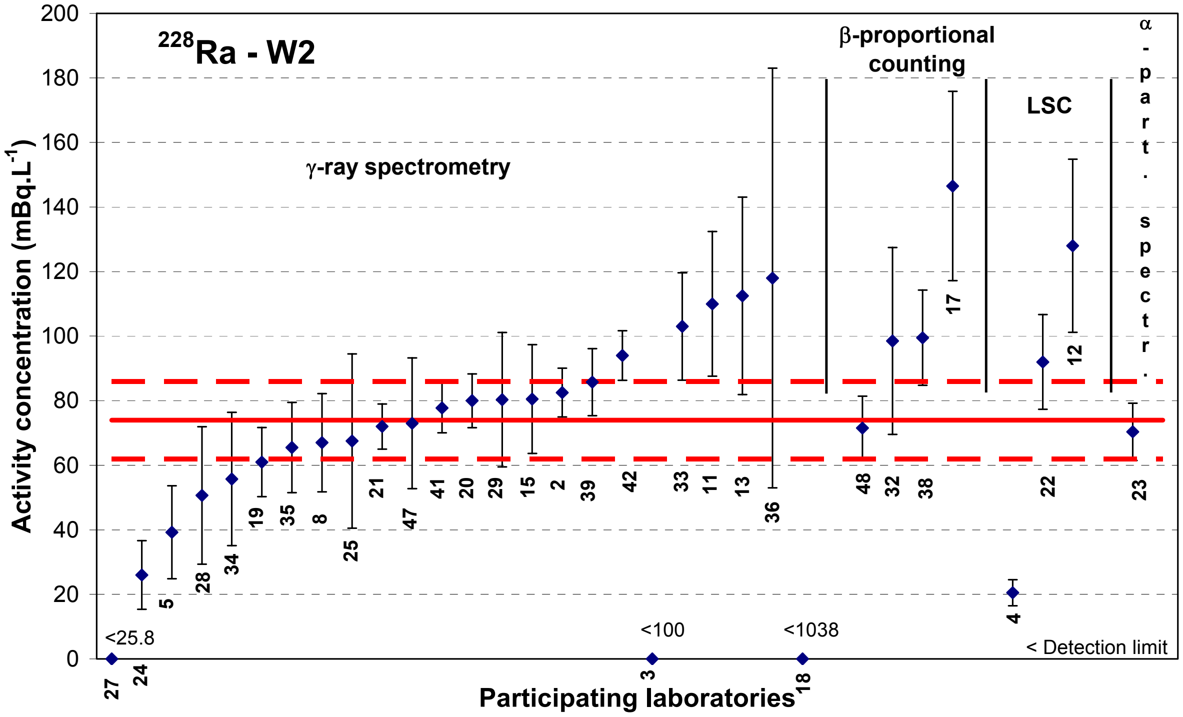

Fig. 40: Same as Fig. 30 but mean laboratory results A_{lab} for ^{228}Ra activity concentration sorted according to counting method

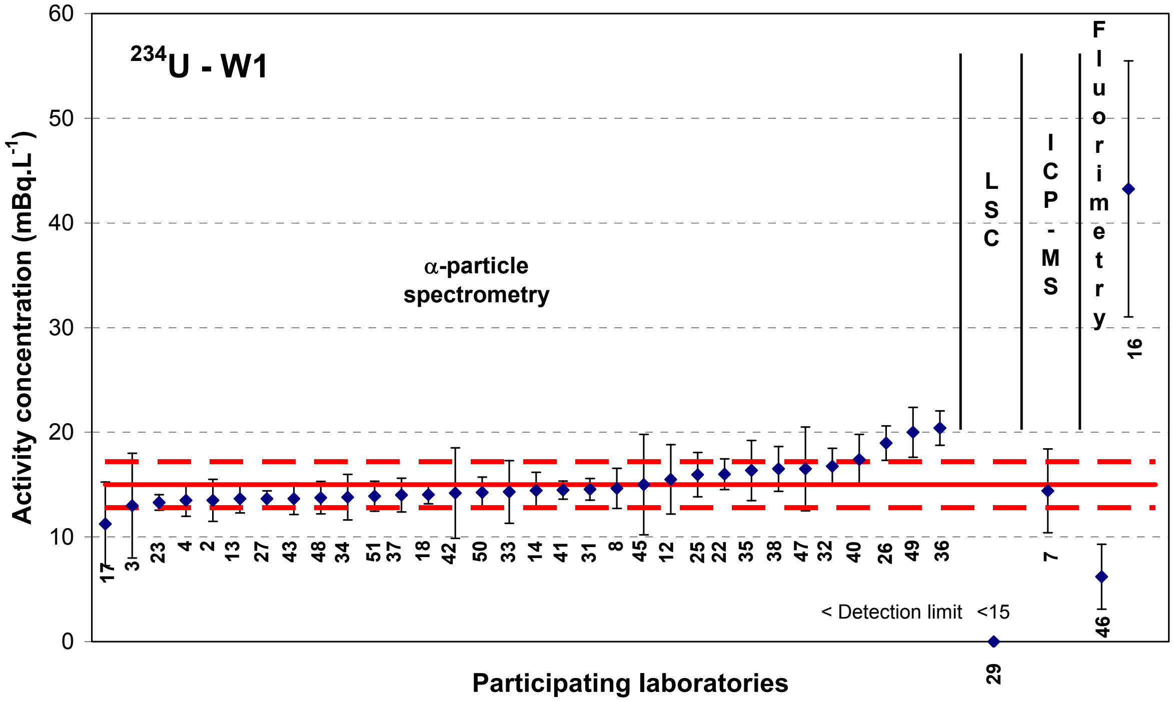

Fig. 41: Same as Fig. 31 but mean laboratory results A_{lab} for ^{234}U activity concentration sorted according to counting method

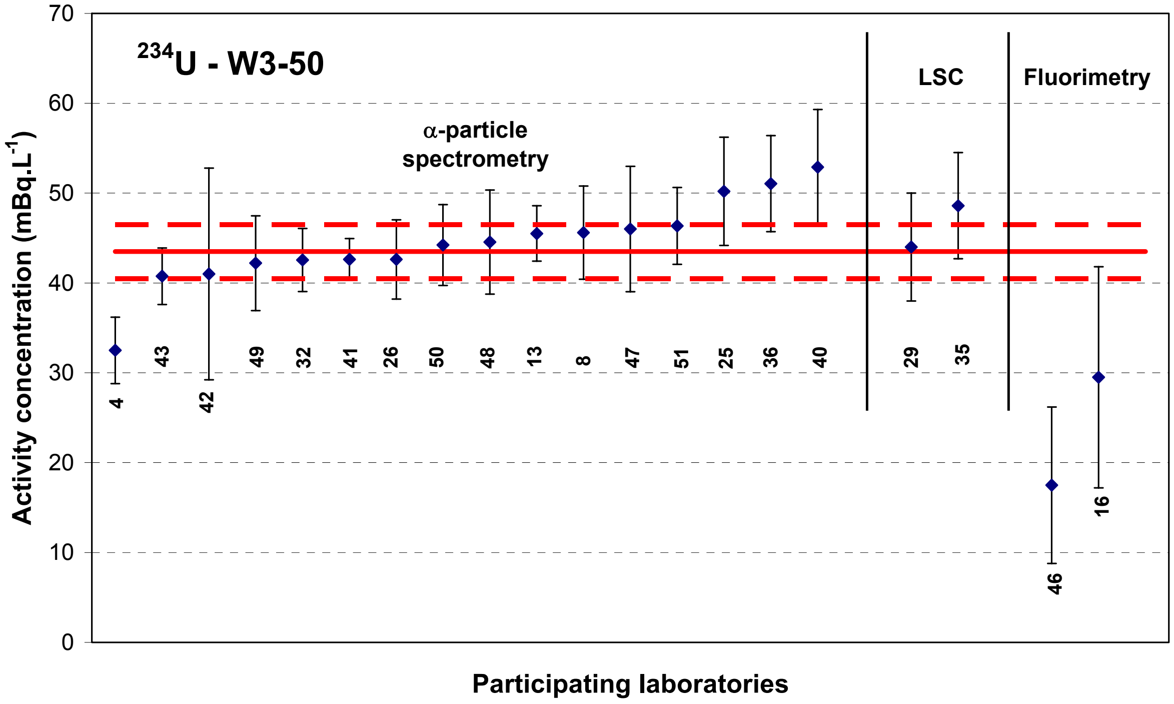

Fig. 42: Same as Fig. 32 but mean laboratory results A_{lab} for ^{234}U activity concentration sorted according to counting method

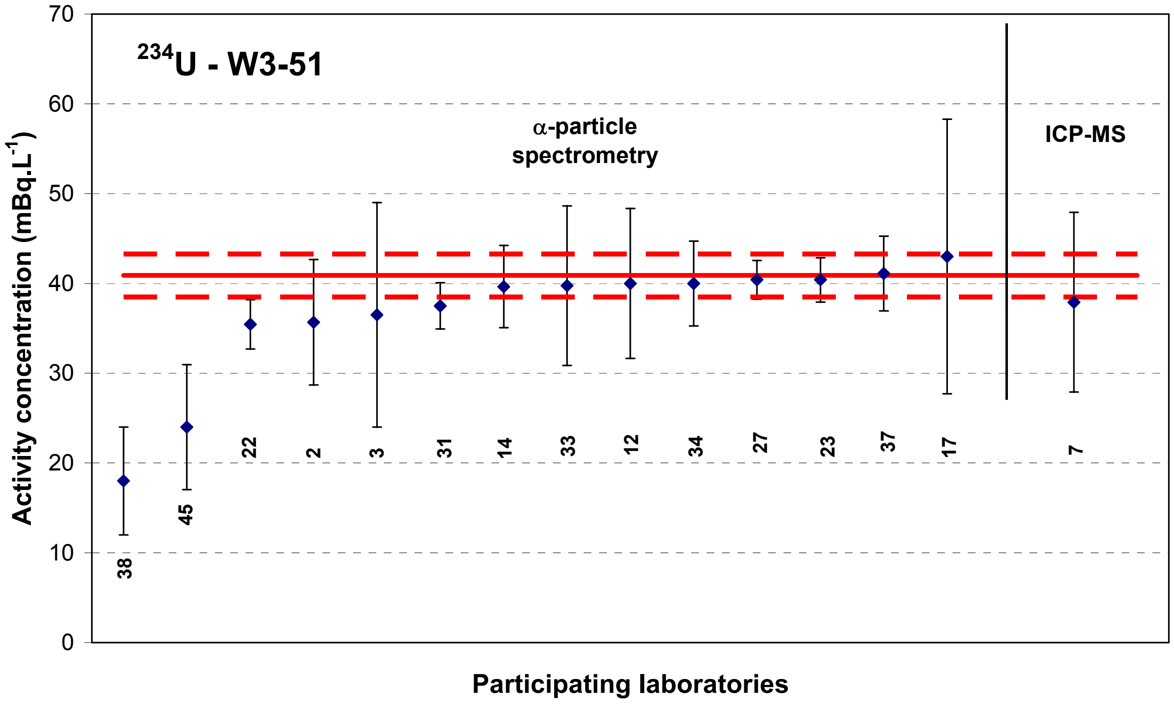

Fig. 43: Same as Fig. 33 but mean laboratory results A_{lab} for ^{234}U activity concentration sorted according to counting method

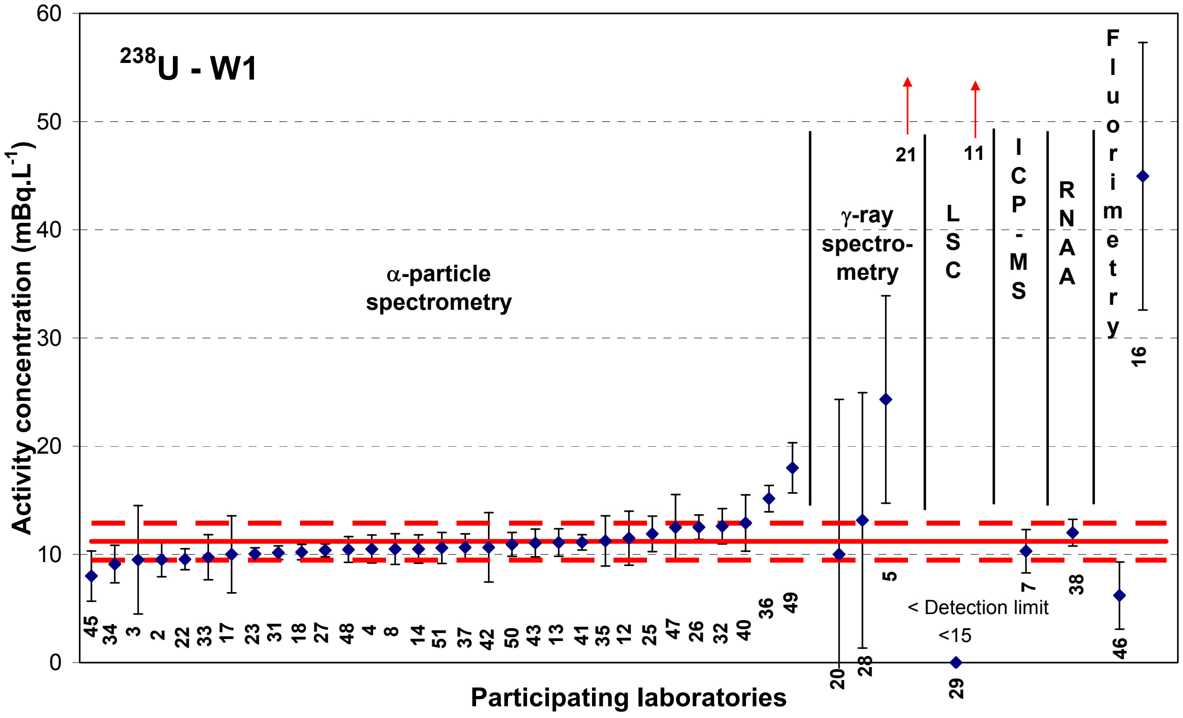

Fig. 44: Same as Fig. 34 but mean laboratory results A_{lab} for ^{238}U activity concentration sorted according to counting method

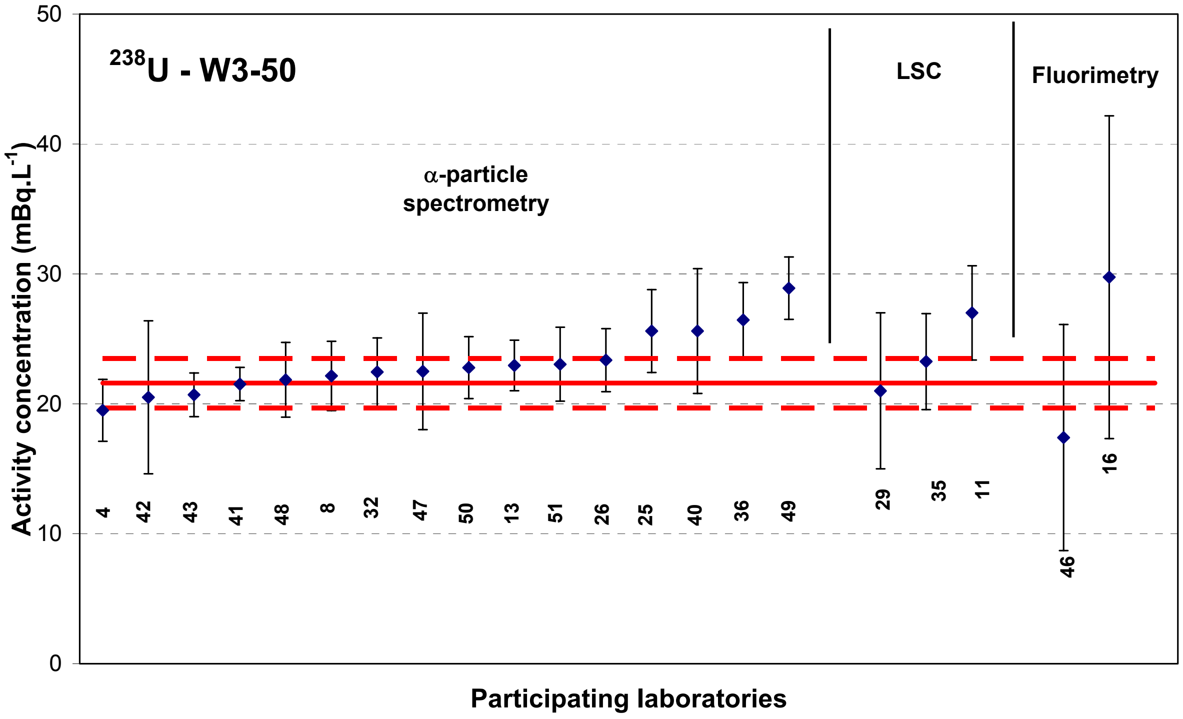

Fig. 45: Same as Fig. 35 but mean laboratory results A_{lab} for ^{238}U activity concentration sorted according to counting method

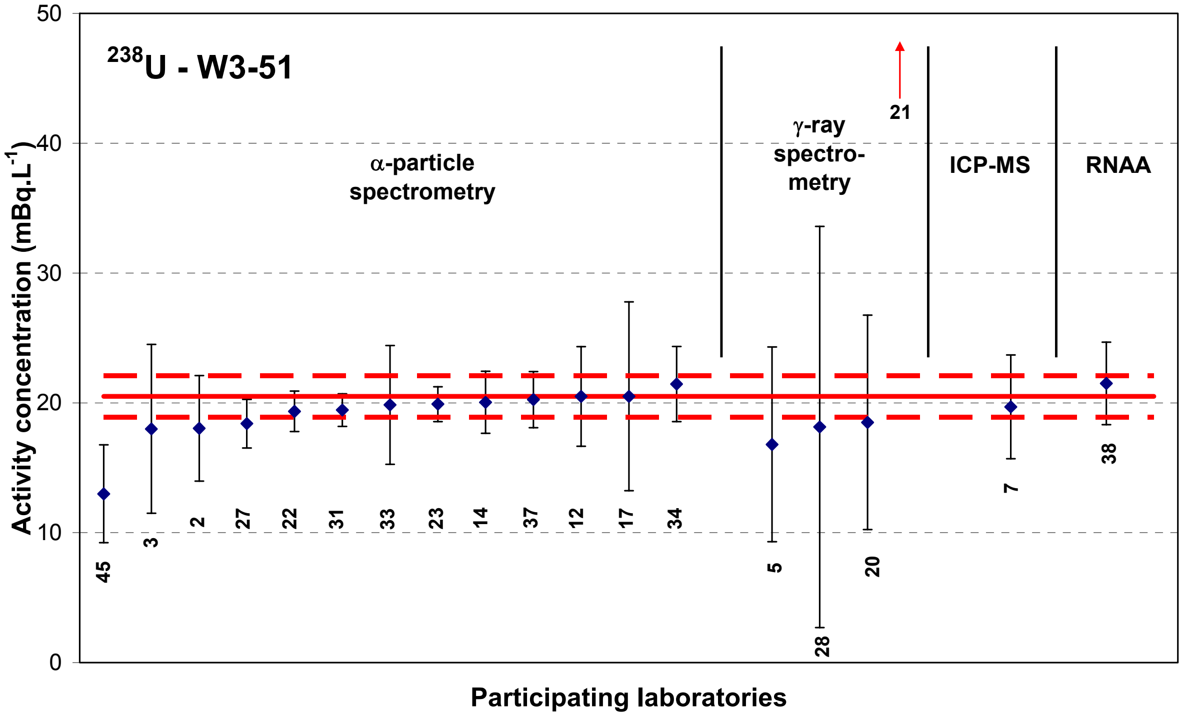

Fig. 46: Same as Fig. 36 but mean laboratory results A_{lab} for ^{238}U activity concentration sorted according to counting method

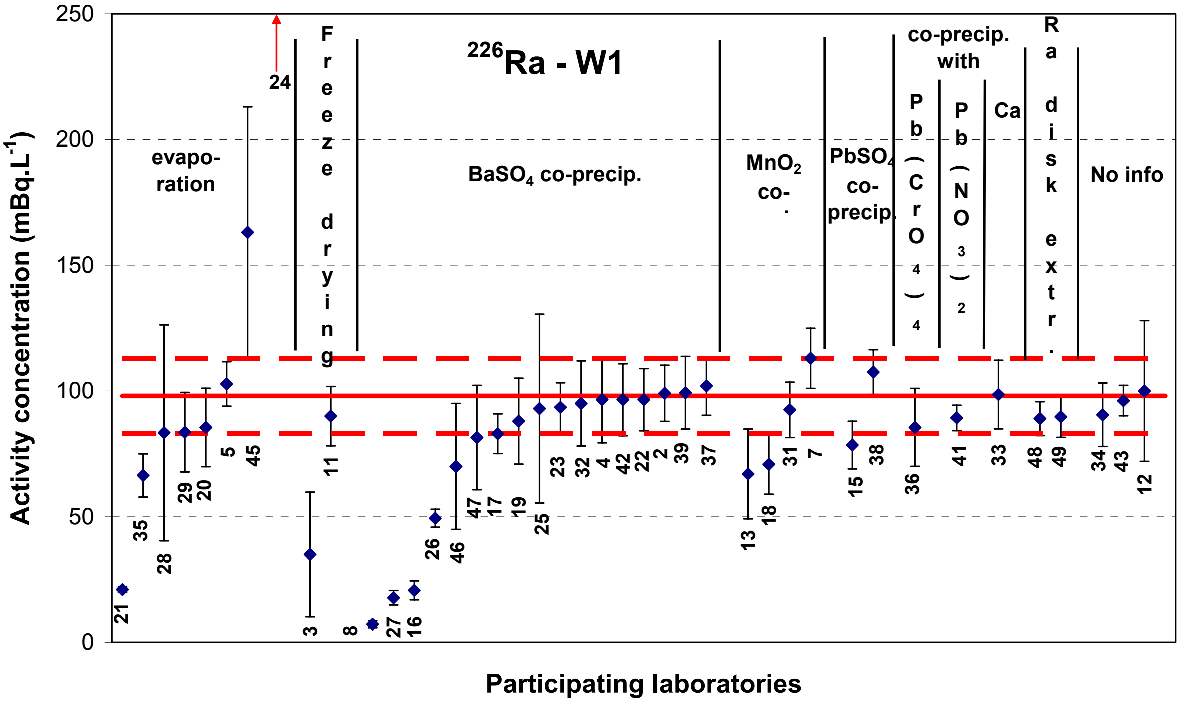

Fig. 47: Same as Fig. 27 but mean laboratory results A_{lab} for ^{226}Ra activity concentration sorted according to separation method

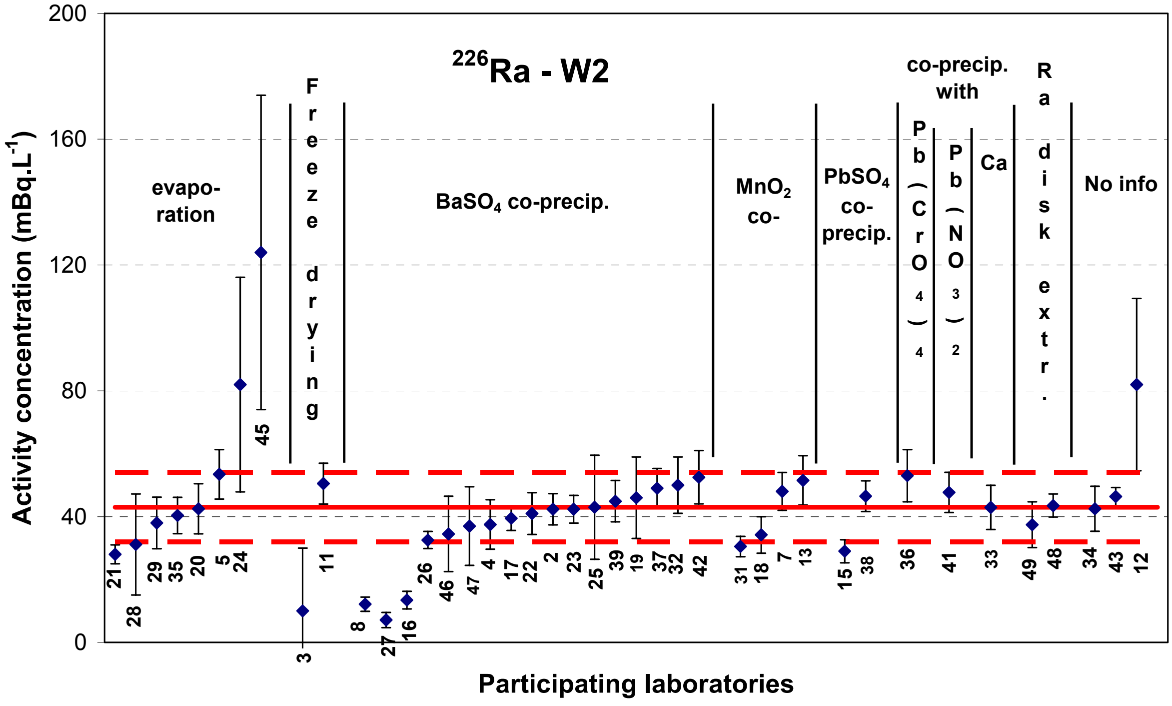

Fig. 48: Same as Fig. 28 but mean laboratory results A_{lab} for ^{226}Ra activity concentration sorted according to separation method

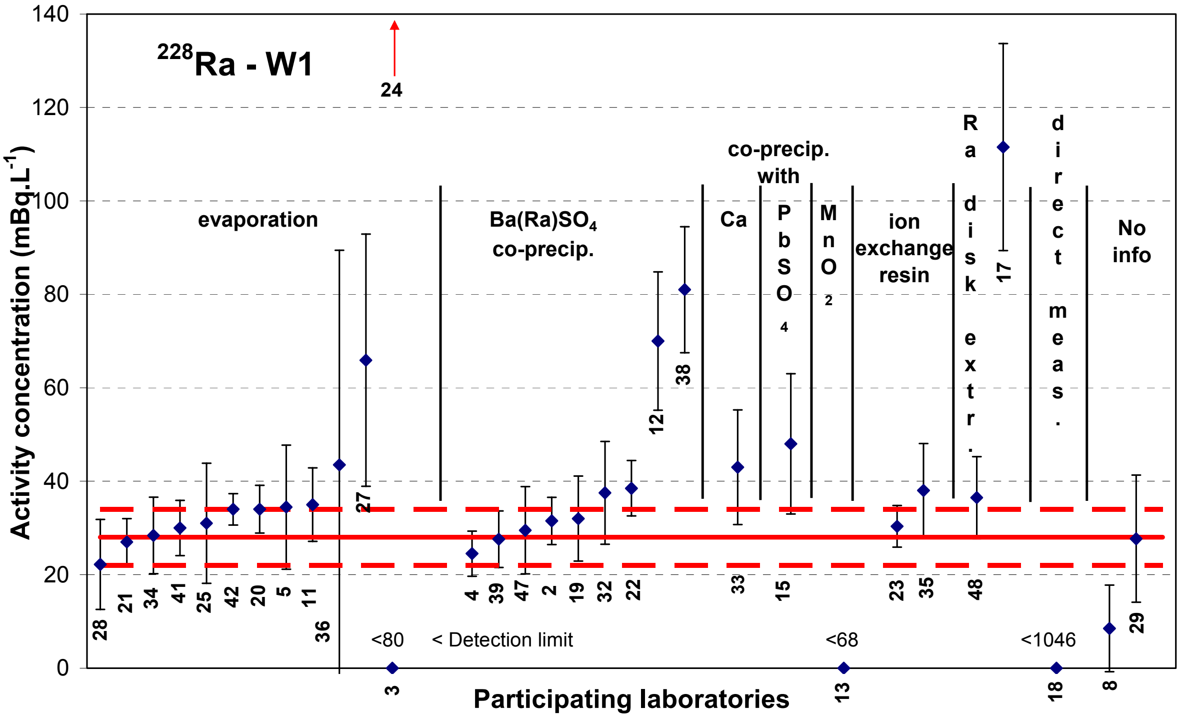

Fig. 49: Same as Fig. 29 but mean laboratory results A_{lab} for ^{228}Ra activity concentration sorted according to separation method

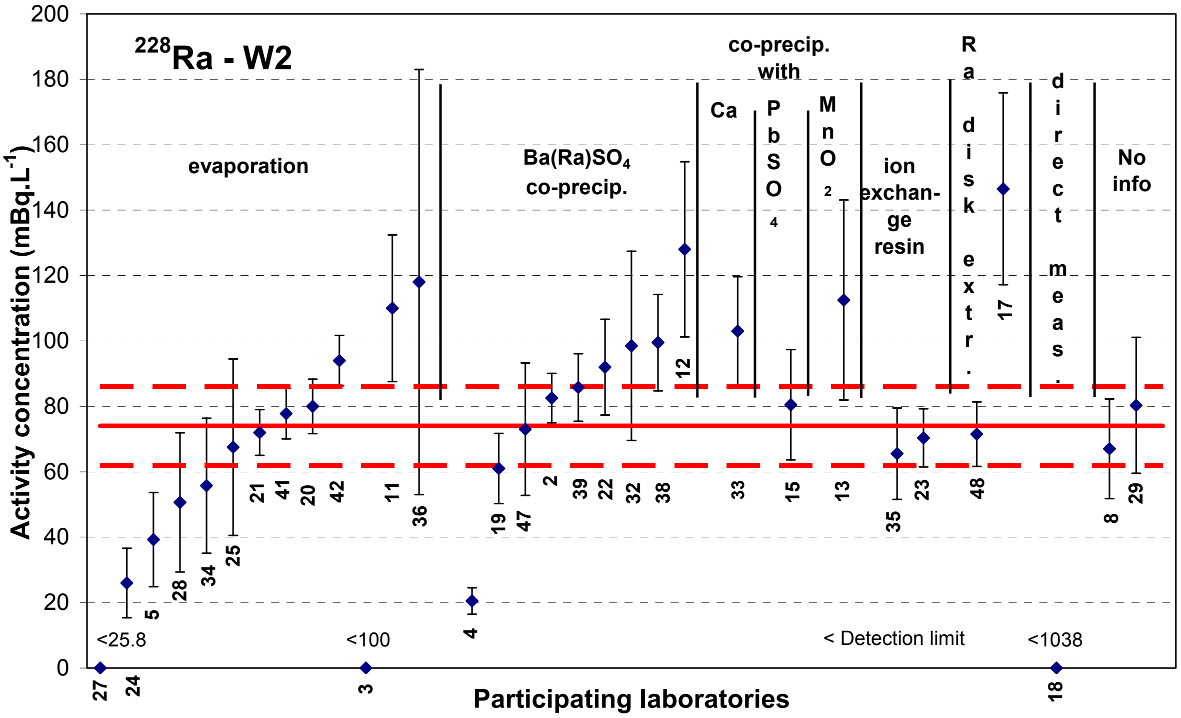

Fig. 50: Same as Fig. 30 but mean laboratory results A_{lab} for ^{228}Ra activity concentration sorted according to separation method

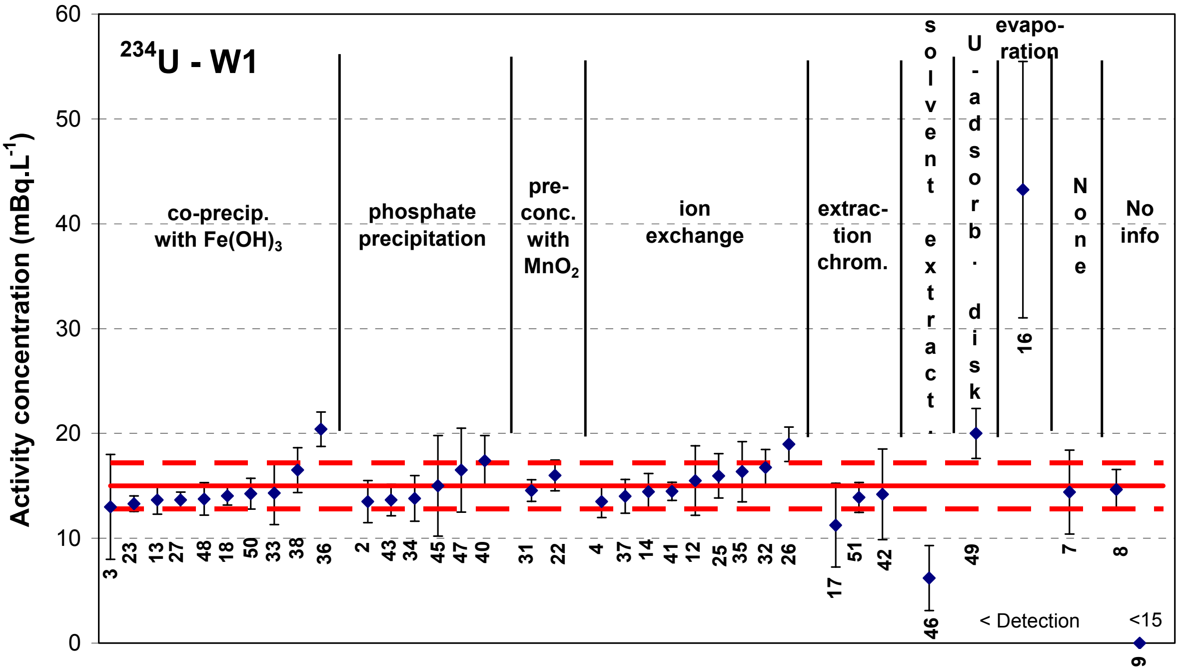

Fig. 51: Same as Fig. 31 but mean laboratory results A_{lab} for ^{234}U activity concentration sorted according to separation method

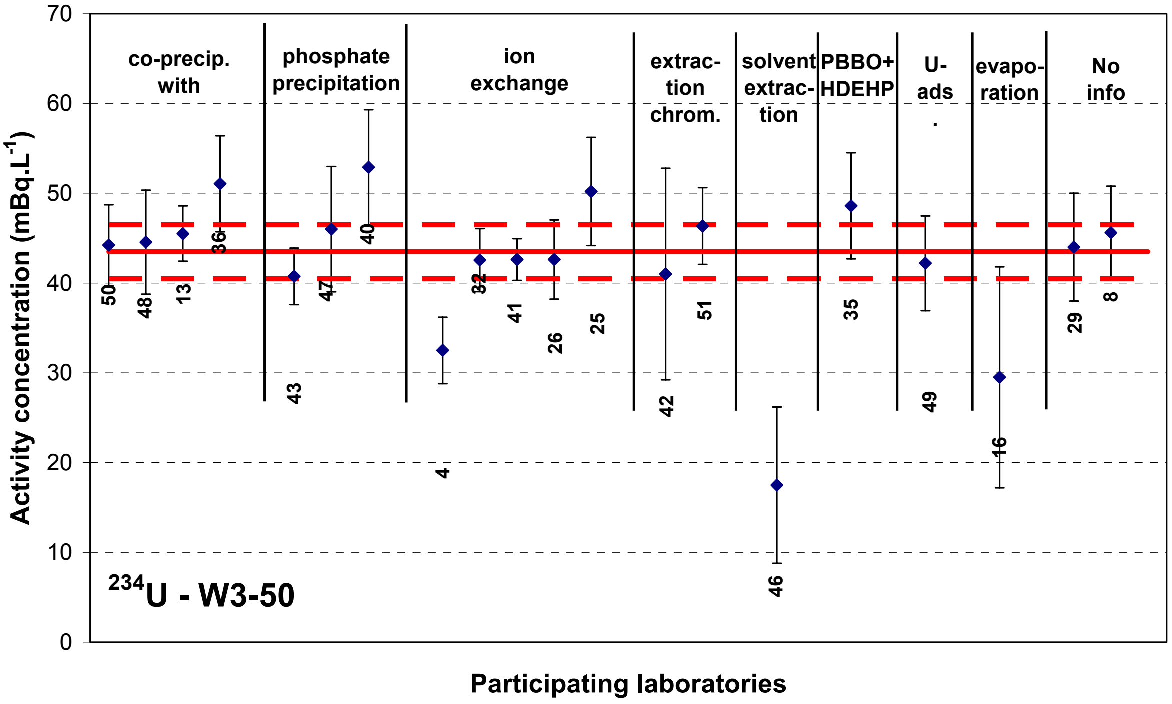

Fig. 52: Same as Fig. 32 but mean laboratory results A_{lab} for ^{234}U activity concentration sorted according to separation method

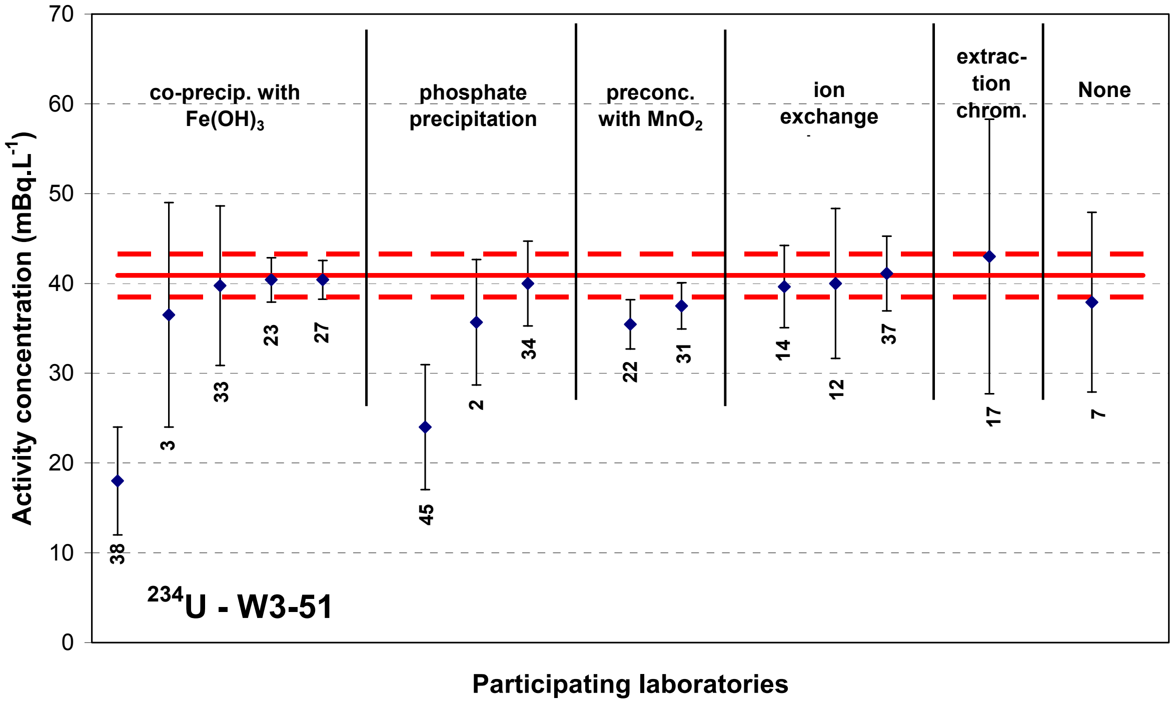

Fig. 53: Same as Fig. 33 but mean laboratory results A_{lab} for ^{234}U activity concentration sorted according to separation method

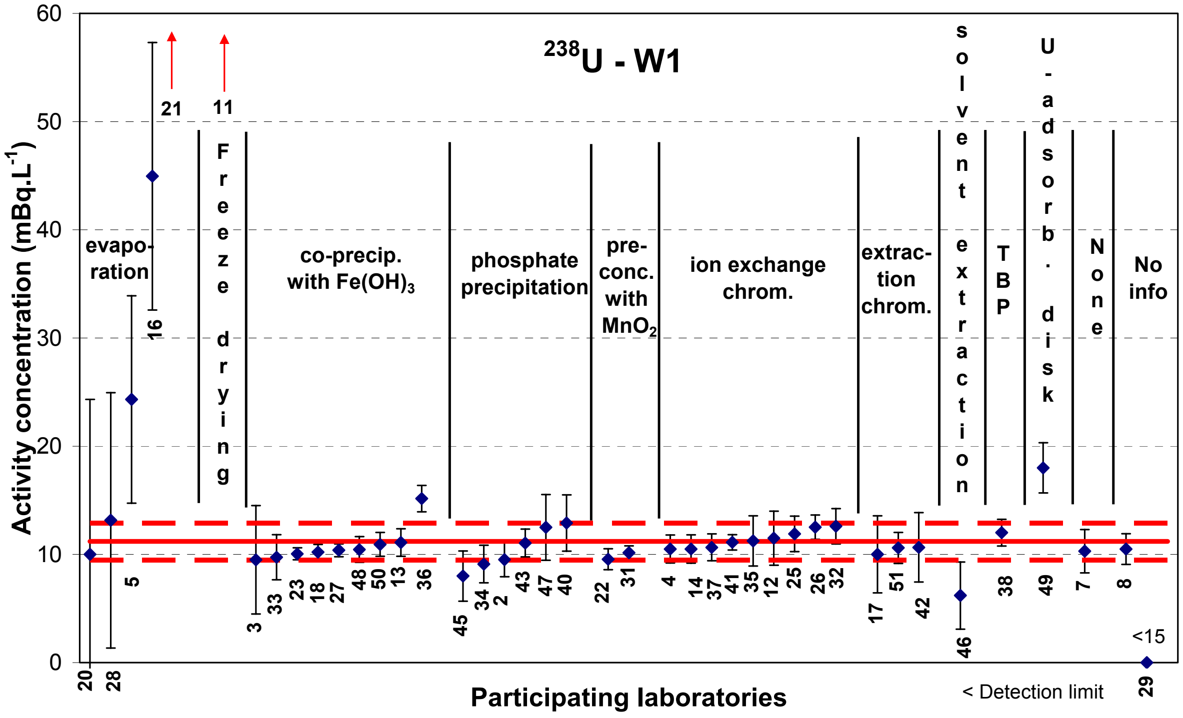

Fig. 54: Same as Fig. 34 but mean laboratory results A_{lab} for ^{238}U activity concentration sorted according to separation method

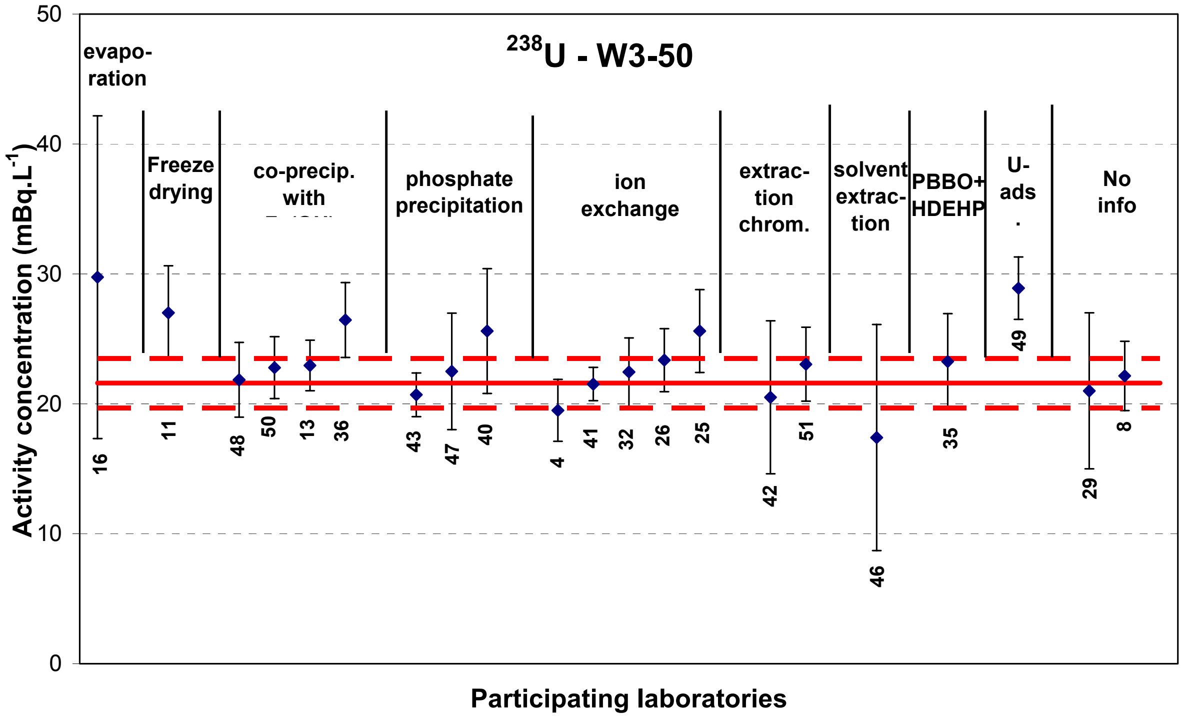

Fig. 55: Same as Fig. 35 but mean laboratory results A_{lab} for ^{238}U activity concentration sorted according to separation method

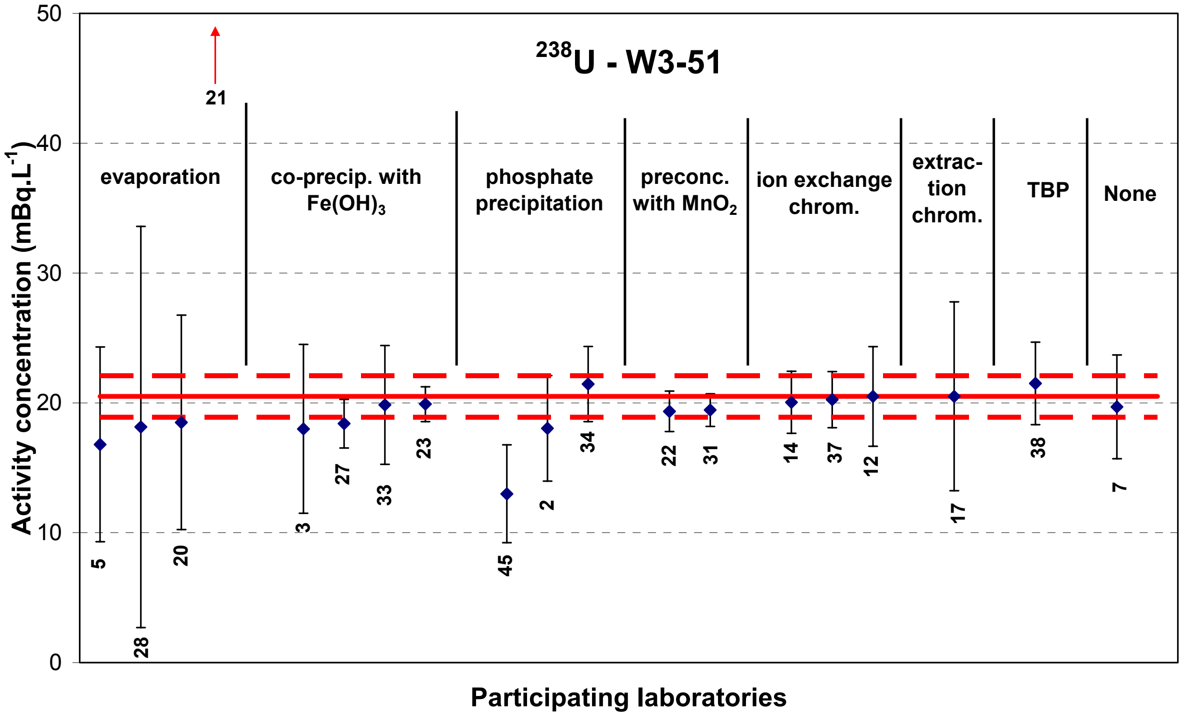

Fig. 56: Same as Fig. 36 but mean laboratory results A_{lab} for ^{238}U activity concentration sorted according to separation method

Table 37: Mean laboratory results A_{lab} for ^{226}**Ra** activity concentrations in **Water 1**, their standard deviations s and estimated expanded uncertainties U_{lab}

lab no.	***n***	**mean activity concentration A_{lab} / (mBq·L^{-1})**	**standard deviation s / (mBq·L^{-1})**	**expanded uncertainty U_{lab} / (mBq·L^{-1})**	**rel. expanded uncertainty U_{lab}/A_{lab} / (%)**
2	2	99.1	0.92	11.15	11.3
3	2	35.0	7.07	24.79	70.8
4	2	96.5	0.71	17.12	17.7
5	3	102.8	-	8.84	8.6
7	4	113.0	0.00	12.00	10.6
8	1	7.2	-	1.40	19.4
9	-	-	-	-	-
10	-	-	-	-	-
11	2	90.0	56.57	**11.78**	13.1
12	6	100.0	16.97	**27.99**	28.0
13	2	67.0	24.04	**17.86**	26.7
14	-	-	-	-	-
15	2	78.5	7.78	9.47	12.1
16	2	20.7	6.93	3.77	18.2
17	2	83.0	2.83	7.90	9.5
18	2	70.9	1.20	11.94	16.8
19	2	88.0	4.24	**17.08**	19.4
20	9	85.5	3.54	**15.58**	18.2
21	1	21.0	-	1.00	4.8
22	2	96.5	3.54	12.37	12.8
23	2	93.5	0.78	9.79	10.5
24	4	760.0	-	295.77	38.9
25	2	93.0	0.00	37.50	40.3
26	4	49.4	13.40	3.60	7.3
27	2	17.8	2.90	**2.83**	15.9
28	4	83.4	7.42	**42.92**	51.5
29	1	83.6	-	15.80	18.9
31	2	92.5	0.71	10.98	11.9
32	2	95.0	4.24	16.92	17.8
33	2	98.6	2.47	13.62	13.8
34	2	90.5	6.36	12.66	14.0
35	2	66.4	14.99	8.59	12.9
36	2	85.5	21.92	15.45	18.1
37	2	102.0	5.66	11.67	11.4
38	2	107.5	2.12	8.95	8.3
39	2	99.3	0.42	**14.47**	14.6
40	-	-	-	-	-
41	2	89.3	4.53	5.06	5.7
42	4	96.5	3.54	14.28	14.8
43	2	96.2	1.91	**6.07**	6.3
45	1	163.0	-	50.00	30.7
46	2	70.0	5.66	25.00	35.7
47	2	81.5	2.12	20.75	25.5
48	2	89.0	4.24	**6.72**	7.5
49	2	89.7	-	8.15	9.1
50	-	-	-	-	-
51	-	-	-	-	-

Table 38: Mean laboratory results A_{lab} for ^{226}**Ra** activity concentrations in **Water 2**, their standard deviations *s* and estimated expanded uncertainties U_{lab}

lab no.	***n***	**mean activity concentration A_{lab} / (mBq·L^{-1})**	**standard deviation *s* / (mBq·L^{-1})**	**expanded uncertainty U_{lab} / (mBq·L^{-1})**	**rel. expanded uncertainty U_{lab}/A_{lab} / (%)**
2	2	42.4	1.06	4.99	11.8
3	1	10.0	-	20.00	200.0
4	2	37.5	2.12	7.87	21.0
5	3	53.5	-	7.87	14.7
7	4	48.0	0.00	6.00	12.5
8	2	12.2	0.64	2.24	18.4
9	-	-	-	-	-
10	-	-	-	-	-
11	2	50.5	34.6	**6.48**	12.8
12	6	82.0	5.66	**27.39**	33.4
13	2	51.5	3.54	**7.84**	15.2
14	-	-	-	-	-
15	2	29.0	5.66	3.66	12.6
16	2	13.5	0.64	2.80	20.8
17	2	39.5	0.71	3.87	9.8
18	2	34.2	0.71	5.82	17.0
19	1	46.0	-	**12.98**	28.2
20	9	42.5	2.12	**7.95**	18.7
21	1	28.0	-	3.00	10.7
22	2	41.0	4.24	6.64	16.2
23	2	42.4	1.06	4.44	10.5
24	4	82.0	-	34.10	41.6
25	2	43.0	2.83	16.56	38.5
26	4	32.5	18.37	2.72	8.4
27	2	7.1	0.62	**2.43**	34.0
28	4	31.2	2.19	**16.07**	51.6
29	1	38.0	-	8.20	21.6
31	2	30.5	3.54	3.25	10.6
32	2	50.0	2.83	8.97	17.9
33	2	43.0	3.18	7.01	16.3
34	2	42.5	0.71	7.14	16.8
35	2	40.4	1.77	5.75	14.3
36	2	53.0	16.97	8.32	15.7
37	2	49.0	1.41	6.29	12.8
38	2	46.5	2.12	4.86	10.5
39	2	44.9	0.14	**6.54**	14.6
40	-	-	-	-	-
41	2	47.7	1.13	6.40	13.4
42	4	52.5	4.95	8.47	16.1
43	2	46.4	1.91	**2.93**	6.3
45	1	124.0	-	50.00	40.3
46	2	34.5	2.12	12.01	34.8
47	2	37.0	2.83	12.50	33.8
48	2	43.5	3.54	**3.67**	8.4
49	2	37.4	-	7.27	19.4
50	-	-	-	-	-
51	-	-	-	-	-

Table 39: Mean laboratory results A_{lab} for ^{228}**Ra** activity concentrations in **Water 1**, their standard deviations s and estimated expanded uncertainties U_{lab}

lab no.	***n***	**mean activity concentration A_{lab} / (mBq·L^{-1})**	**standard deviation s / (mBq·L^{-1})**	**expanded uncertainty U_{lab} / (mBq·L^{-1})**	**rel. expanded uncertainty U_{lab}/A_{lab} / (%)**
2	2	31.5	0.71	5.04	16.0
3	2	< 80	-	-	-
4	2	24.5	0.71	4.84	19.7
5	3	34.5	-	13.26	38.5
7	-	-	-	-	-
8	2	8.5	0.71	9.27	109.1
9	-	-	-	-	-
10	-	-	-	-	-
11	1	35.0	-	**7.86**	22.4
12	6	70.0	-	14.79	21.1
13	1	< 68	-	-	-
14	-	-	-	-	-
15	1	48.0	-	15.00	31.3
16	-	-	-	-	-
17	2	111.5	2.12	22.15	19.9
18	2	< 1046	-	-	-
19	2	32.0	4.24	**9.10**	28.4
20	9	34.0	1.41	5.11	15.0
21	1	27.0	-	5.00	18.5
22	2	38.5	0.71	5.93	15.4
23	2	30.4	1.63	4.46	14.7
24	4	885.0	-	343.41	38.8
25	2	31.0	2.83	12.86	41.5
26	-	-	-	-	-
27	1	65.9	-	27.00	41.0
28	4	22.2	2.97	**9.63**	43.4
29	1	27.7	-	13.60	49.1
31	-	-	-	-	-
32	2	37.5	0.71	11.01	29.4
33	2	43.0	6.65	12.28	28.5
34	2	28.4	0.00	8.20	28.9
35	2	38.0	11.31	10.05	26.4
36	2	43.5	3.54	45.94	105.6
37	-	-	-	-	-
38	2	81.0	5.66	13.49	16.6
39	2	27.6	2.26	6.03	21.8
40	-	-	-	-	-
41	2	30.0	1.41	5.90	19.7
42	4	34.0	1.41	3.36	9.9
43	-	-	-	-	-
45	-	-	-	-	-
46	-	-	-	-	-
47	2	29.5	3.54	9.35	31.7
48	2	36.5	3.54	**8.77**	24.0
49	-	-	-	-	-
50	-	-	-	-	-
51	-	-	-	-	-

Table 40: Mean laboratory results A_{lab} for **^{228}Ra** activity concentrations in **Water 2**, their standard deviations s and estimated expanded uncertainties U_{lab}

lab no.	***n***	**mean activity concentration A_{lab} / (mBq·L^{-1})**	**standard deviation s / (mBq·L^{-1})**	**expanded uncertainty U_{lab} / (mBq·L^{-1})**	**rel. expanded uncertainty U_{lab}/A_{lab} / (%)**
2	2	82.5	4.95	7.57	9.2
3	2	< 100	-	-	-
4	2	20.5	0.71	4.04	19.7
5	3	39.3	-	14.39	36.7
7	-	-	-	-	-
8	2	67.0	7.07	15.24	22.7
9	-	-	-	-	-
10	-	-	-	-	-
11	1	110.0	-	**22.44**	20.4
12	6	128.0	-	26.81	20.9
13	2	112.5	14.85	**30.59**	30.6
14	-	-	-	-	-
15	2	80.5	7.78	16.86	20.9
16	-	-	-	-	-
17	2	146.5	2.12	29.33	20.0
18	2	< 1038	-	-	-
19	1	61.0	-	**10.72**	17.6
20	9	80.0	4.24	8.33	10.4
21	1	72.0	-	7.00	9.7
22	2	92.0	4.24	14.65	15.9
23	2	70.4	0.78	8.89	12.6
24	4	26.0	-	**10.62**	40.8
25	2	67.5	0.71	26.96	40.0
26	-	-	-	-	-
27	1	< 25.8	-	-	-
28	4	50.7	2.62	**21.29**	42.0
29	1	80.3	-	20.80	25.9
31	-	-	-	-	-
32	2	98.5	3.54	28.92	29.4
33	2	103.0	5.66	16.64	16.2
34	2	55.8	1.48	20.65	37.0
35	2	65.5	14.85	13.99	21.4
36	1	118.0	-	**65.03**	55.1
37	-	-	-	-	-
38	2	99.5	12.02	14.73	14.8
39	2	85.8	2.90	10.36	12.1
40	-	-	-	-	-
41	2	77.8	8.84	7.71	9.9
42	4	94.0	5.66	7.70	8.2
43	-	-	-	-	-
45	-	-	-	-	-
46	-	-	-	-	-
47	2	73.0	1.41	20.25	27.7
48	2	71.5	6.36	**9.89**	13.8
49	-	-	-	-	-
50	-	-	-	-	-
51	-	-	-	-	-

Table 41: Mean laboratory results A_{lab} for ^{234}**U** activity concentrations in **Water 1**, their standard deviations *s* and estimated expanded uncertainties U_{lab}

lab no.	***n***	**mean activity concentration A_{lab} / (mBq·L^{-1})**	**standard deviation *s* / (mBq·L^{-1})**	**expanded uncertainty U_{lab} / (mBq·L^{-1})**	**rel. expanded uncertainty U_{lab}/A_{lab} / (%)**
2	2	13.5	0.81	2.02	14.9
3	2	13.0	0.00	5.00	38.5
4	6	13.5	0.71	1.52	11.3
5	-	-	-	-	-
7	2	14.4	0.00	4.00	27.8
8	2	14.7	0.92	1.92	13.1
9	-	-	-	-	-
10	-	-	-	-	-
11	-	-	-	-	-
12	2	15.5	2.12	**3.32**	21.4
13	2	13.7	1.34	**1.35**	9.9
14	2	14.5	0.21	1.72	11.9
15	-	-	-	-	-
16	2	43.3	2.62	12.22	28.3
17	2	11.3	0.64	4.00	35.6
18	2	14.1	0.64	0.90	6.4
19	-	-	-	-	-
20	-	-	-	-	-
21	-	-	-	-	-
22	2	16.0	0.57	1.47	9.2
23	2	13.3	0.00	0.74	5.6
24	-	-	-	-	-
25	2	16.0	0.21	2.12	13.3
26	2	19.0	0.63	1.65	8.7
27	2	13.7	1.34	**0.75**	5.5
28	-	-	-	-	-
29	1	<15	-	-	-
31	2	14.6	1.20	1.03	7.1
32	2	16.8	0.64	1.71	10.2
33	4	14.3	0.85	**3.00**	21.0
34	2	13.8	0.28	2.18	15.8
35	2	16.4	0.35	2.87	17.5
36	2	20.4	0.85	**1.64**	8.0
37	2	14.0	1.70	1.61	11.5
38	2	16.5	0.71	**2.13**	12.9
39	-	-	-	-	-
40	1	17.4	-	2.40	13.8
41	2	14.5	1.72	0.87	6.0
42	2	14.2	0.14	4.32	30.4
43	2	13.7	0.78	**1.50**	11.0
45	4	15.0	-	4.80	32.0
46	1	6.2	-	3.10	50.0
47	2	16.5	0.71	4.00	24.3
48	2	13.8	0.21	**1.55**	11.3
49	4	20.0	-	2.38	11.9
50	2	14.3	0.06	1.47	10.3
51	2	13.9	0.28	1.42	10.2

Table 42: Mean laboratory results A_{lab} for **^{234}U** activity concentrations in **Waters 3-50 and 3-51**, their std. deviations *s* and estimated expanded uncertainties U_{lab}

lab no.	***n***	**mean activity concentration A_{lab} / (mBq·L^{-1})**	**standard deviation *s* / (mBq·L^{-1})**	**expanded uncertainty U_{lab} / (mBq·L^{-1})**	**rel. expanded uncertainty U_{lab}/A_{lab} / (%)**
W3-50					
4	6	32.5	0.71	3.70	11.4
8	2	45.6	3.11	5.19	11.4
10	-	-	-	-	-
11	-	-	-	-	-
13	2	45.5	2.83	**3.08**	6.8
16	2	29.5	3.96	12.31	41.7
19	-	-	-	-	-
25	2	50.2	0.14	6.02	12.0
26	2	42.6	1.44	4.41	10.3
29	1	44.0	-	6.00	13.6
32	2	42.6	0.92	3.51	8.2
35	2	48.6	0.00	5.91	12.2
36	2	51.1	0.07	5.34	10.5
39	-	-	-	-	-
40	1	52.9	-	6.40	12.1
41	2	42.6	0.29	2.32	5.5
42	2	41.0	1.41	11.78	28.7
43	2	40.8	1.48	3.14	7.7
46	1	17.5	-	8.70	49.7
47	2	46.0	1.41	6.98	15.2
48	2	44.6	1.06	5.79	13.0
49	4	42.2	-	**5.27**	12.5
50	2	44.2	1.00	4.49	10.2
51	2	46.4	2.05	4.28	9.2
W3-51					
2	2	35.7	1.61	6.98	19.6
3	2	36.5	2.12	12.50	34.2
5	-	-	-	-	-
7	2	37.9	0.00	10.00	26.4
9	-	-	-	-	-
12	2	40.0	2.83	**8.35**	20.9
14	2	39.7	0.49	4.57	11.5
15	-	-	-	-	-
17	2	43.0	0.28	15.29	35.6
18	-	-	-	-	-
20	-	-	-	-	-
21	-	-	-	-	-
22	2	35.5	1.77	2.73	7.7
23	2	40.4	0.28	2.47	6.1
24	-	-	-	-	-
27	2	40.4	4.24	**2.16**	5.3
28	-	-	-	-	-
31	2	37.5	0.71	**2.57**	6.9
33	4	39.8	5.59	8.89	22.4
34	2	40.0	2.83	4.71	11.8
37	2	41.1	2.55	4.16	10.1
38	1	18.0	-	6.00	33.3
45	4	24.0	-	6.95	29.0

Table 43: Mean laboratory results A_{lab} for ^{238}**U** activity concentrations in **Water 1**, their standard deviations *s* and estimated expanded uncertainties U_{lab}

lab no.	***n***	**mean activity concentration A_{lab} / (mBq·L^{-1})**	**standard deviation *s* / (mBq·L^{-1})**	**expanded uncertainty U_{lab} / (mBq·L^{-1})**	**rel. expanded uncertainty U_{lab}/A_{lab} / (%)**
2	2	9.5	0.50	1.58	16.6
3	2	9.5	0.71	5.01	52.8
4	6	10.5	0.71	1.29	12.3
5	3	24.3	-	9.58	39.4
7	2	10.3	0.00	2.00	19.4
8	2	10.5	0.28	1.42	13.5
9	-	-	-	-	-
10	-	-	-	-	-
11	1	120.0	-	**18.29**	15.2
12	2	11.5	2.12	2.49	21.6
13	2	11.1	0.42	**1.27**	11.4
14	2	10.5	0.28	1.31	12.5
15	-	-	-	-	-
16	2	45.0	4.60	12.36	27.5
17	2	10.0	0.85	3.57	35.7
18	2	10.2	0.14	0.71	7.0
19	-	-	-	-	-
20	6	10.0	1.41	**14.32**	143.2
21	1	92.0	-	13.00	14.1
22	2	9.6	0.35	0.96	10.0
23	2	10.1	0.07	0.56	5.6
24	-	-	-	-	-
25	2	11.9	0.00	1.64	13.8
26	2	12.5	0.13	1.12	9.0
27	2	10.4	0.59	**0.59**	5.7
28	2	13.2	1.34	11.80	89.7
29	1	< 15	-	-	-
31	2	10.2	0.07	0.63	6.2
32	2	12.6	0.42	1.64	13.0
33	4	9.7	0.37	**2.08**	21.4
34	2	9.1	0.85	1.74	19.1
35	2	11.3	0.07	2.31	20.6
36	2	15.2	0.78	**1.22**	8.0
37	2	10.7	1.20	1.25	11.7
38	2	12.0	1.41	**1.24**	10.3
39	-	-	-	-	-
40	1	12.9	-	2.60	20.2
41	2	11.1	1.00	0.71	6.4
42	2	10.7	0.07	3.22	30.2
43	2	11.1	0.07	**1.28**	11.6
45	4	8.0	-	2.32	29.0
46	1	6.2	-	3.10	50.0
47	2	12.5	2.12	3.04	24.4
48	2	10.5	0.07	**1.19**	11.4
49	4	18.0	-	2.32	12.9
50	2	10.9	0.85	1.10	10.1
51	2	10.6	0.57	1.42	13.4

Table 44: Mean laboratory results A_{lab} for ^{238}**U** activity concentrations in **Waters 3-50 and 3-51**, their std. deviations *s* and estimated expanded uncertainties U_{lab}

lab no.	***n***	**mean activity concentration A_{lab} / (mBq·L^{-1})**	**standard deviation *s* / (mBq·L^{-1})**	**expanded uncertainty U_{lab} / (mBq·L^{-1})**	**rel. expanded uncertainty U_{lab}/A_{lab} / (%)**
W3-50					
4	6	19.5	0.71	2.38	12.2
8	2	22.2	1.48	2.66	12.0
10	-	-	-	-	-
11	1	27.0	-	**3.62**	13.4
13	2	23.0	3.18	**1.94**	8.5
16	2	29.8	4.03	12.41	41.7
19	-	-	-	-	-
25	2	25.6	0.42	3.20	12.5
26	2	23.4	0.23	2.43	10.4
29	1	21.0	-	6.00	28.6
32	2	22.5	0.78	2.63	11.7
35	2	23.3	0.35	**3.69**	15.9
36	2	26.5	1.34	2.88	10.9
39	0			-	
40	1	25.6	-	4.80	18.8
41	2	21.5	0.44	1.28	5.9
42	2	20.5	0.71	5.89	28.7
43	2	20.7	0.57	1.68	8.1
46	1	17.4	-	8.70	50.0
47	2	22.5	0.71	4.49	20.0
48	2	21.9	0.21	2.88	13.2
49	4	28.9	-	2.40	8.3
50	2	22.8	0.07	2.38	10.4
51	2	23.1	0.78	2.84	12.3
W3-51					
2	2	18.0	0.49	4.06	22.5
3	2	18.0	1.41	6.49	36.1
5	3	16.8	-	7.50	44.6
7	2	19.7	0.00	4.00	20.3
9	-	-	-	-	-
12	2	20.5	0.71	3.83	18.7
14	2	20.1	0.49	2.39	11.9
15	-	-	-	-	-
17	2	20.5	1.56	7.27	35.5
18	-	-	-	-	-
20	6	18.5	0.71	8.25	44.6
21	1	121.0	-	23.00	19.0
22	2	19.4	0.21	1.55	8.0
23	2	19.9	0.42	1.34	6.8
24	-	-	-	-	-
27	2	18.4	0.14	**1.87**	10.2
28	2	18.2	2.19	**15.45**	85.1
31	2	19.5	0.35	1.25	6.4
33	4	19.9	2.05	4.58	23.1
34	2	21.5	2.19	2.90	13.5
37	2	20.3	2.19	2.16	10.7
38	2	21.5	2.12	3.18	14.8
45	4	13.0	-	3.77	29.0

Eleven laboratories also reported results for the activity concentration of additional radionuclides, other than the predefined measurands. Laboratories 11, 28, 32 and 42 measured ^{40}K in samples W1 and W2. ^{228}Th (labs 20, 32, 42, 49) and ^{235}U (labs 14, 23, 41, 43) were also determined in the samples. The reported individual results and their expanded uncertainties are plotted in Figs. 57 to 59. For these radionuclides no reference value was determined in the frame of this intercomparison. The overall mean and standard deviation *s* of these measurement results for the additional radionuclides are given in Table 45 and are indicated in the figures as solid and dashed red lines. In the case of ^{235}U two laboratories (no. 23 and 41) reported more than one measurement per sample and on the graph (Fig. 57) the individual results are presented.

Table 45: Mean activity concentrations of ^{235}U, ^{228}Th and ^{40}K in the mineral water samples and their standard deviations *s* in mBq·L^{-1}, as measured by some laboratories.

Sample	W1	W2	W3
^{235}U	0.53 ± 0.07	-	0.98 ± 0.08
^{228}Th	17.4 ± 7.4	44.3 ± 16.5	-
^{40}K	76 ± 20	327 ± 69	-

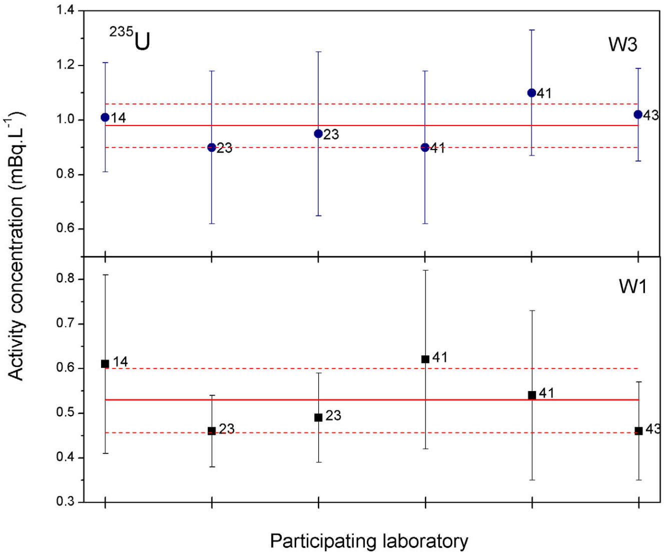

Fig. 57: ^{235}U activity concentration determined additionally by some laboratories. Error bars represent expanded uncertainty (*k* = 2) of individual measurements, mean value and standard deviation *s* are indicated in red. Laboratory numbers are indicated.

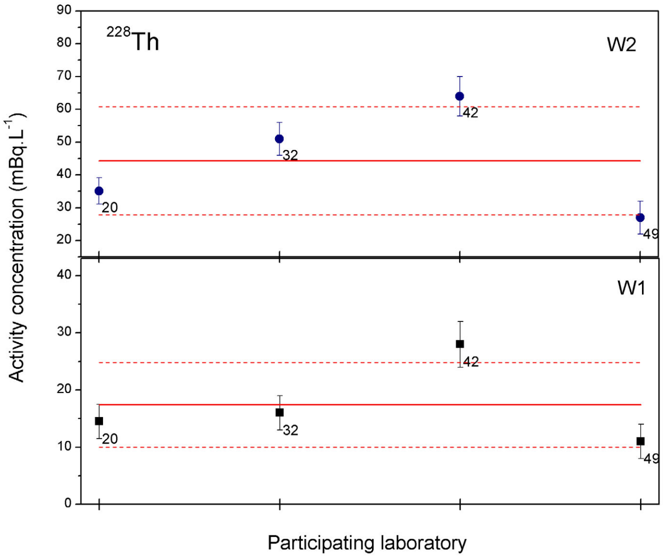

Fig. 58: ^{228}Th activity concentration determined additionally by some laboratories. Error bars represent expanded uncertainty (*k* = 2) of individual measurements, mean value and standard deviation *s* are indicated in red. Laboratory numbers are indicated.

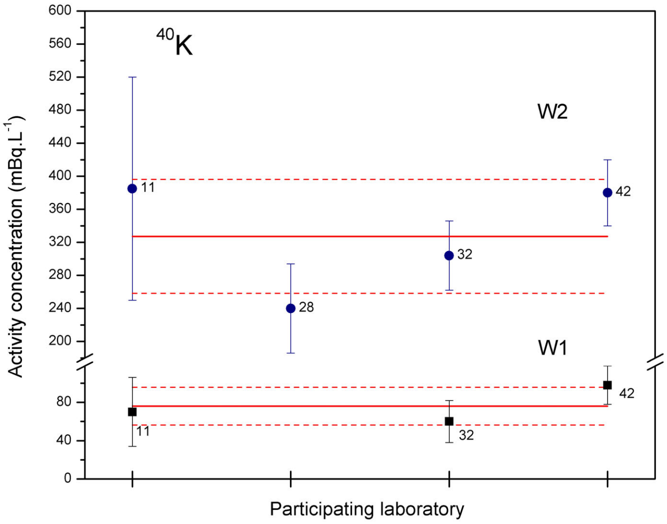

Fig. 59: ^{40}K activity concentration determined additionally by some laboratories. Error bars represent expanded uncertainty (*k* = 2) of individual measurements, mean value and standard deviation *s* are indicated in red. Laboratory numbers are indicated.

Furthermore, laboratories 19 and 28 reported values for the ^{210}Pb activity concentration Laboratory 28 reported values of (33 ± 20) mBq·L^{-1} for W1 and (26 ± 18) mBq·L^{-1} for W3, while the values submitted by lab 19 for all three water samples were below the detection limit of their method (< 17500 mBq·L^{-1}). The same participant (no.19) measured activity concentrations of ^{222}Rn in all samples of less than 1000 mBq·L^{-1}.

Results for ^{224}Ra activity concentration in all intercomparison samples were submitted by one participant (no. 23). The reported values (two per sample) are listed in Table 46. The results are decay corrected to the measurement date 10-Jan-2008.

Table 46: Activity concentrations of ^{224}Ra in the mineral water samples and their expanded uncertainties (k = 2) in $mBq \cdot L^{-1}$ as reported by laboratory 23.

Sample	**W1**	**W2**	**W3**
Measurement 1	13.4 ± 1.6	36.0 ± 5.2	2.62 ± 0.70
Measurement 2	14.5 ± 1.6	32.4 ± 4.6	2.22 ± 0.94

6. Data evaluation and comparison of data

The most intuitive way to display the compliance of measurement results with reference values is the deviation chart. Tables 47 - 54 contain the relative deviations of the laboratory mean values A_{lab} from the reference activity concentration A_{ref}:

$$Rel.dev. = \frac{A_{lab} - A_{ref}}{A_{ref}} \cdot 100 = \frac{D}{A_{ref}} \cdot 100 \qquad (22)$$

These values are plotted in ascending order in a deviation chart, and the laboratories reporting too low or too high values become more visible (Figs. 60 - 69). Dashed red lines indicate a deliberately chosen range of ± 20 % in the case of ^{234}U and ^{238}U and ± 30 % in the case of ^{226}Ra and ^{228}Ra. Laboratories which have reported values outside of the specified range were considered to deviate considerably from the reference value. These laboratories were contacted after the deadline for reporting results and were asked to critically review their analysis and measurement procedures and calculations, and to report back if the review would lead to changed values. Some of the contacted laboratories corrected their results and the newly submitted results were used for the final evaluation of the laboratory performance as presented here.

For completeness, the expanded uncertainty U_{ref} of the reference value is plotted as dotted red lines in Figs. 60 - 69. In the figures, as well as in Tables 47 - 54, large relative deviations are highlighted by colours: orange for $D/A_{ref} > 15$ % (> 25 % in case of ^{226}Ra and ^{228}Ra), red for $D/A_{ref} > 20$ % (> 30 % in case of ^{226}Ra and ^{228}Ra).

As far as the determination of radium is concerned, the results of the comparison are far from satisfactory. Figure 60 shows that the values determined by many laboratories for ^{226}Ra in Water 1 were too low (up to a factor of 14 – lab 8), while two laboratories (no. 24 and 45) determine values too high (as much as up to 8 times the reference value). By contrast, the reference value determined by IRMM and BfS has a relative expanded uncertainty, U_{ref}, of 15 %. Laboratory 45 followed its routine analytical procedure including sample pre-concentration to reduce the volume and counting by liquid scintillation (LSC) of the ingrowth of ^{222}Rn. The ^{226}Ra activity concentration in the comparison samples, however, was lower than the activities measured routinely (~150 mBq·L^{-1}). Another laboratory (no. 24) used gamma-ray spectrometry and reported a far too high activity concentration (> 670 % too high) for ^{226}Ra in Water 1. This laboratory declared that they do not have much experience with such low activities and routinely do not perform evaporation of the sample which was done in this case. Laboratory 8 used liquid scintillation counting of the ^{222}Rn ingrowth for the determination of ^{226}Ra activity concentration in the samples after pre-concentration of the radium from the water with $BaSO_4$. Also, another seven laboratories reported results deviating by more than 30 % from the reference value (no. 3, 13, 16, 21, 26, 27 and 35). Laboratories 13 and 16 used alpha-particle spectrometry and labs 3 and 35 liquid scintillation counting for the ^{226}Ra determination. Sorption emanation technique, gamma-ray spectrometry and gross alpha counting were applied in lab 27, 21 and 16, respectively. In the case of lab 21, due to a misunderstanding, the routine analytical procedure was not used and wrong acid was added for conservation. The other six participants followed their routine analysis procedures.

One should note that a ^{226}Ra activity concentration of 100 mBq·L^{-1} in water is more than twice the limit of detection required by the Proposal for a Council Directive, COM(2011)385 (European Communities, 2011), thus a non-negligible amount, and measurement results five times too high or too low would result in unnecessary or omitted remedial action, respectively. It is not satisfactory that only 31 laboratories (out of 41) are able to determine the ^{226}Ra activity concentration within 30 % from the reference value.

The results for ^{226}Ra in Water 2, although at lower activity concentration, reveal a similar, although slightly better performance (Fig. 61). Again, 32 laboratories show results spread around the reference value to within ± 30 %. The extreme deviations are in this case 6 times too low (lab 27) and a factor of 3 too high (lab 45). Laboratory 27 applied its routine procedure - sorption emanation technique after radium pre-concentration from the water with barium sulphate. Comparing laboratory numbers in Figures 60 and 61, it appears that there is a group of laboratories – with labs 12 and 26 as exceptions – which implement the chosen ^{226}Ra analysis procedures systematically incorrect.

Even more problematic is laboratory performance in the determination of ^{228}Ra. In Water 2, there are only 17 laboratories (of 29 submitting results) that are within ± 30 % of the reference value without forming a distinct plateau of measurement results around the reference value (Fig. 63). Four laboratories (no. 4, 5, 24, 28) determine values too low, and 8 submit results too high (by > 30 %) (no. 11, 12, 13, 17, 32, 33, 36, 38). The situation for ^{228}Ra in Water 1 is similar (Fig. 62); 13 laboratories, i.e. about half, obtain results that are too high by > 30 %. One of the laboratories (no. 24) determines by gamma-ray spectrometry a far too high activity concentration of 885 mBq·L^{-1} (A_{ref} = (28 ± 6) mBq·L^{-1}) in Water 1, whereas its result for Water 2 is a factor of 2.8 too low. This lab does not routinely determine this radionuclide. Laboratory 12, which reported too high activity concentration for both water samples (> 150 % for W1 and > 70 % for W2), did not follow their routine analytical procedure. The sample was first pre-concentrated by evaporation and then the radium was co-precipitated as $BaSO_4$. The precipitate was dissolved in alkaline EDTA and measured with Hisafe 3 by LSC. In order to enhance the chemical recovery the sulphide precipitation was done separately. All other laboratories with results deviating more than 30 % from the reference value for both water samples have followed their routine analytical procedures. Some of the participants (labs 3, 13, 15, 18 and 24) reported difficulties encountered during the determination of ^{228}Ra in the samples due to the low activity levels.

In general, the analysis of uranium activity concentration in mineral water is under much better control by the vast majority of participating laboratories (Figs. 64 - 69). For example, Fig. 67 depicts the results for the determination of ^{238}U in Water 1. In this case, 32 of 40 laboratories obtain results within ± 20 % of the reference value, only two laboratories (no. 45 and 46) obtain results that are too low (maximum by 45 %), and six laboratories (labs 5, 11, 16, 21, 36, 49) report values > 20 % too high (in the two worst cases – no. 21 and 11 - by a factor of about 8 and 11, though). Laboratories 16 and 46 used fluorimetry for the determination of natural uranium and determined the activity concentrations of ^{234}U and ^{238}U by calculations, under the assumption of equilibrium. In the case of laboratories 5 and 21, gamma-ray spectrometry was used and in lab 11 liquid scintillation counting. Alpha-particle spectrometry was applied in laboratories 36, 45 and 49.

The comparison results for the determination of ^{234}U in both waters and ^{238}U in Water 3-50 and Water 3-51 are similarly acceptable. The majority of the participants followed their routine analytical procedures. Only few participants reported that they have applied other procedures (no. 20, 21, and 22). Laboratory 22 does not perform routine analysis of uranium in water. Laboratory 20 reported that the sample quantity was too small for application of their routine gamma-ray spectrometry counting and spectrum analysis procedure. Therefore, to increase the amount of the residue after evaporation, oxalic acid was added. In the case of lab 21, because of a misunderstanding, wrong acid was added for conservation. The uranium was pre-concentrated from the sample by evaporation and measured by gamma-ray spectrometry. One laboratory (no. 3) reported difficulties encountered during the determination of uranium in the mineral water samples due to the low activity levels.

As seen from the discussion above, just as all counting methods used for the determination of the different radionuclides are represented among the largely deviating results, there is not a single sample preparation or radiochemical separation procedure which could be pin-pointed as being particularly frequent among or even responsible for largely deviating results. An exception is the group of laboratories using fluorimetry for the determination of the uranium isotopes (no. 16 and 46). The assumption in their calculations for equilibrium between ^{234}U and ^{238}U in the mineral water samples was incorrect. The ratio of ^{234}U and ^{238}U in mineral water is different than one (usually $^{234}U/^{238}U > 1$) and depends on the (geo)chemical conditions within the aquifer (Ivanovich and Harmon, 1992). Therefore, a critical review of the implementation of the method should be done by these two laboratories.

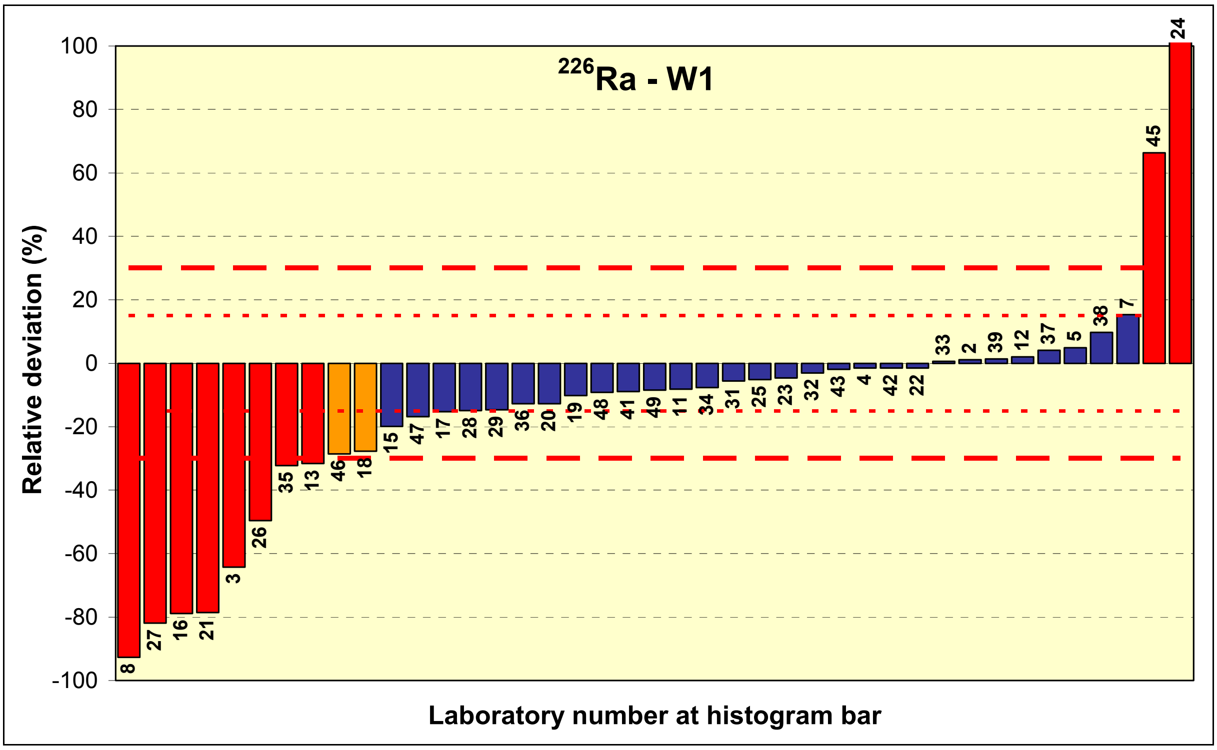

Fig. 60: Deviation chart of the 41 measurement results for ^{226}Ra in Water 1 plotted as D/A_{ref} in ascending order. Range of ± 30 % (dashed) and expanded uncertainty U_{ref} (dotted) are indicated

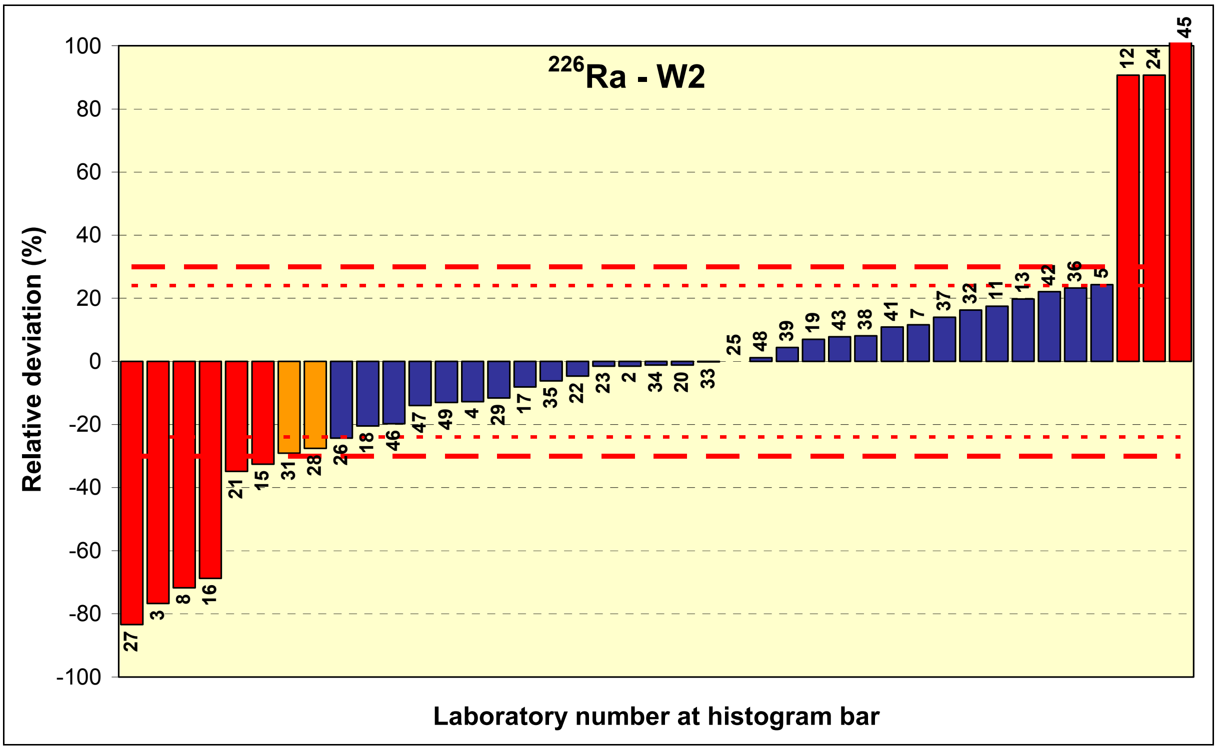

Fig. 61: Deviation chart of the 41 measurement results for ^{226}Ra in Water 2 plotted as D/A_{ref} in ascending order. Range of ± 30 % (dashed) and expanded uncertainty U_{ref} (dotted) are indicated

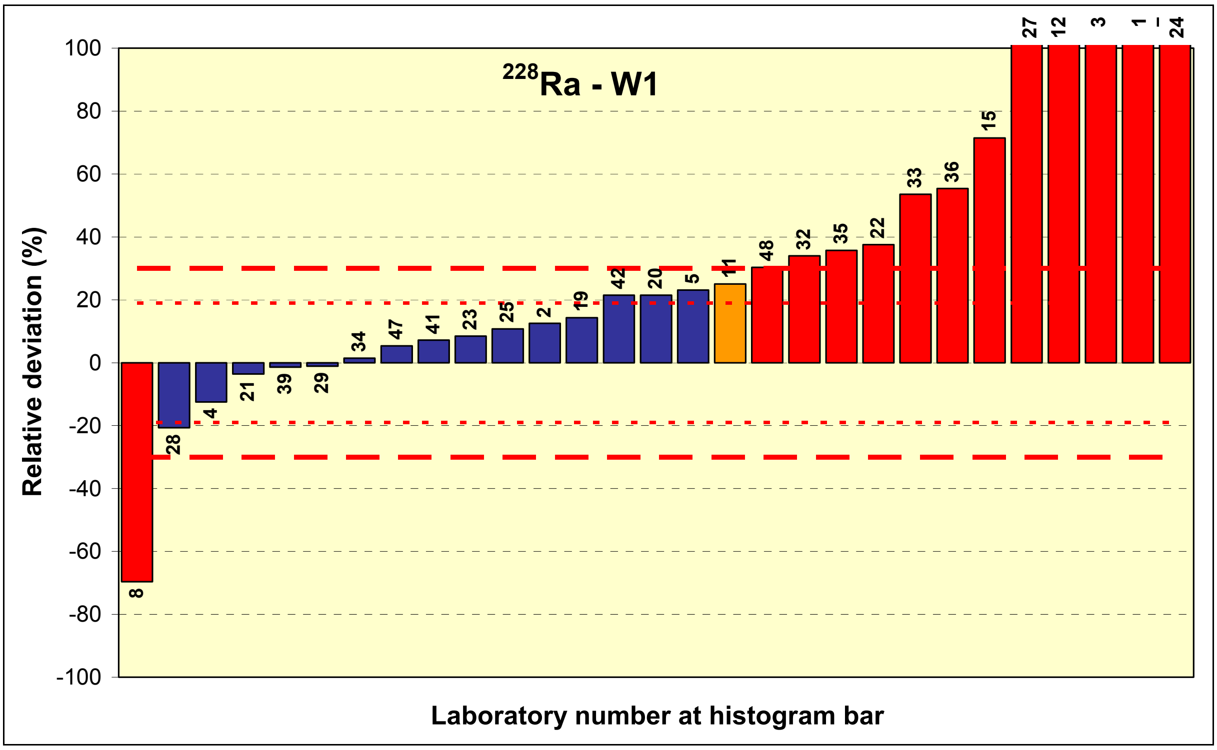

Fig. 62: Deviation chart of the 29 measurement results for ^{228}Ra in Water 1 plotted as D/A_{ref} in ascending order. Range of ± 30 % (dashed) and expanded uncertainty U_{ref} (dotted) are indicated

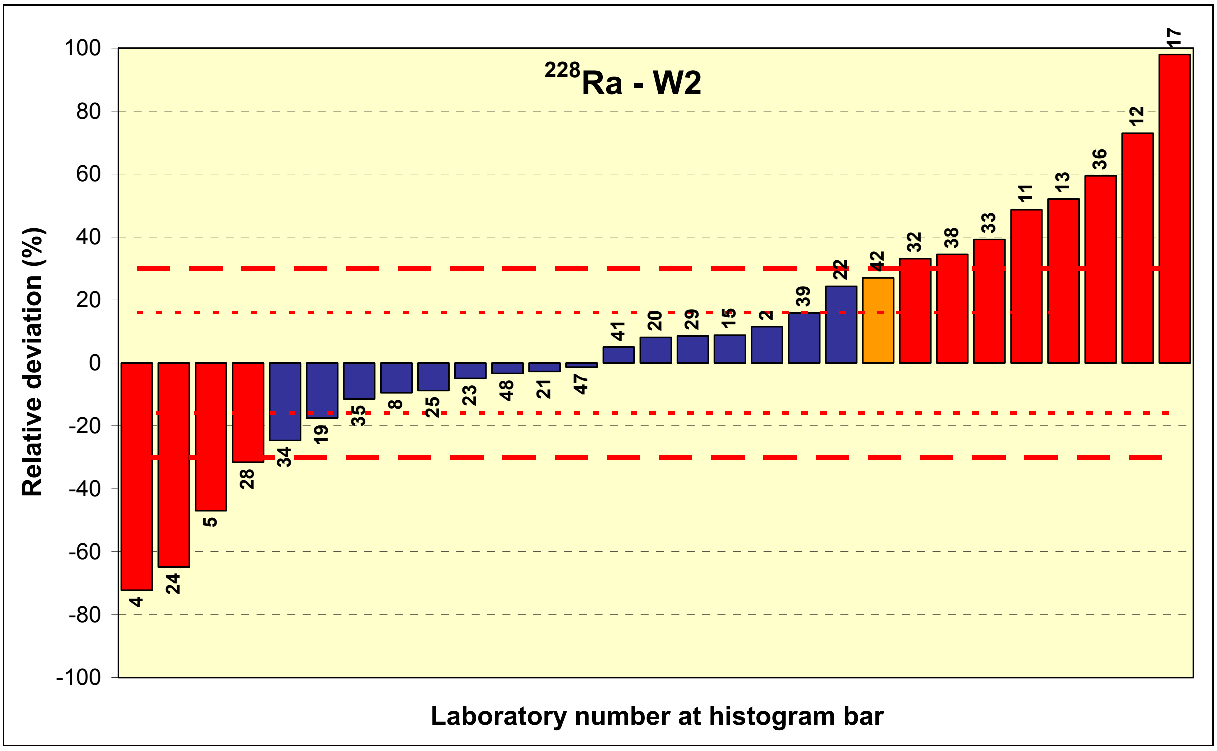

Fig. 63: Deviation chart of the 29 measurement results for ^{228}Ra in Water 2 plotted as D/A_{ref} in ascending order. Range of ± 30 % (dashed) and expanded uncertainty U_{ref} (dotted) are indicated

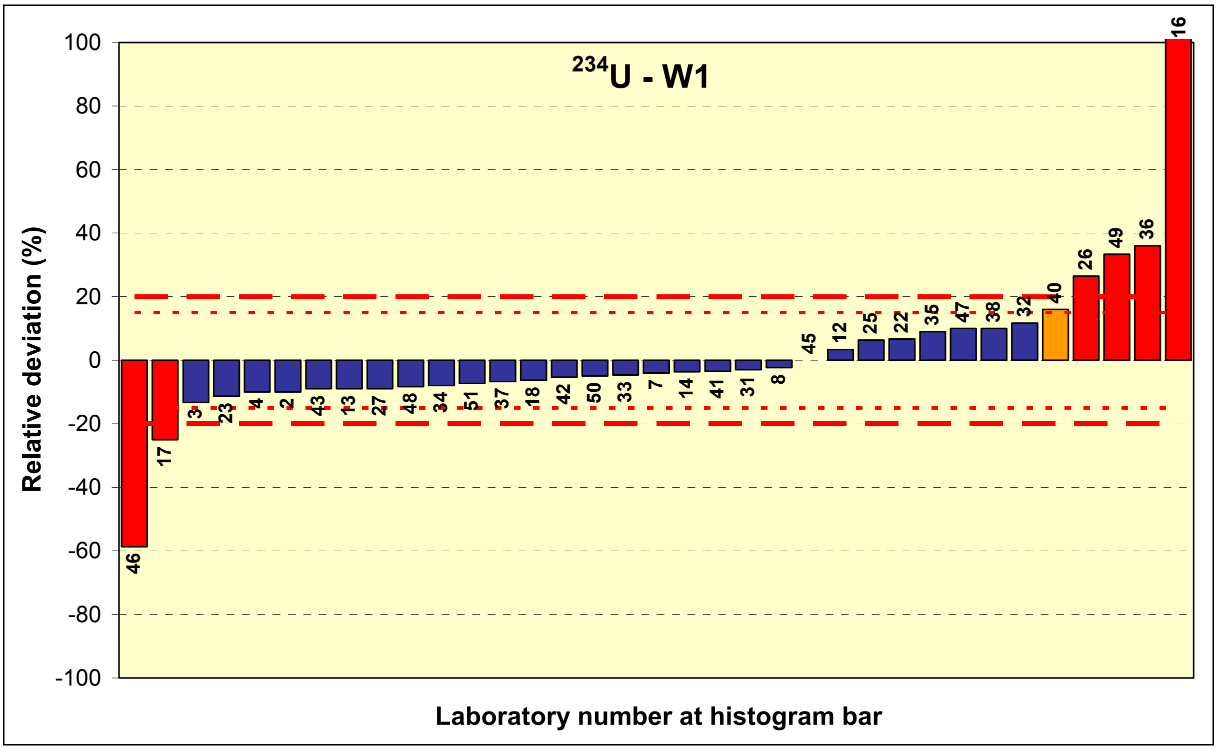

Fig. 64: Deviation chart of the 35 measurement results for ^{234}U in Water 1 plotted as D/A_{ref} in ascending order. Range of ± 20 % (dashed) and expanded uncertainty U_{ref} (dotted) are indicated

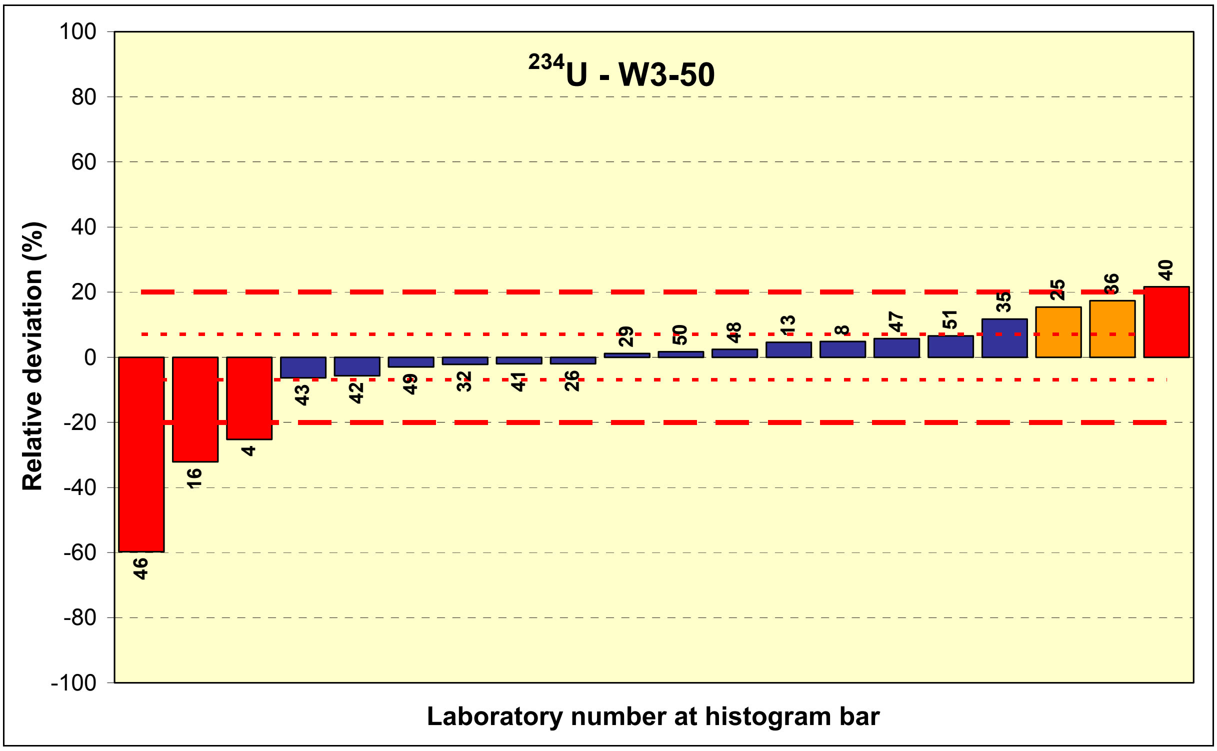

Fig. 65: Deviation chart of the 20 measurement results for ^{234}U in Water 3-50 plotted as D/A_{ref} in ascending order. Range of ± 20 % (dashed) and expanded uncertainty U_{ref} (dotted) are indicated

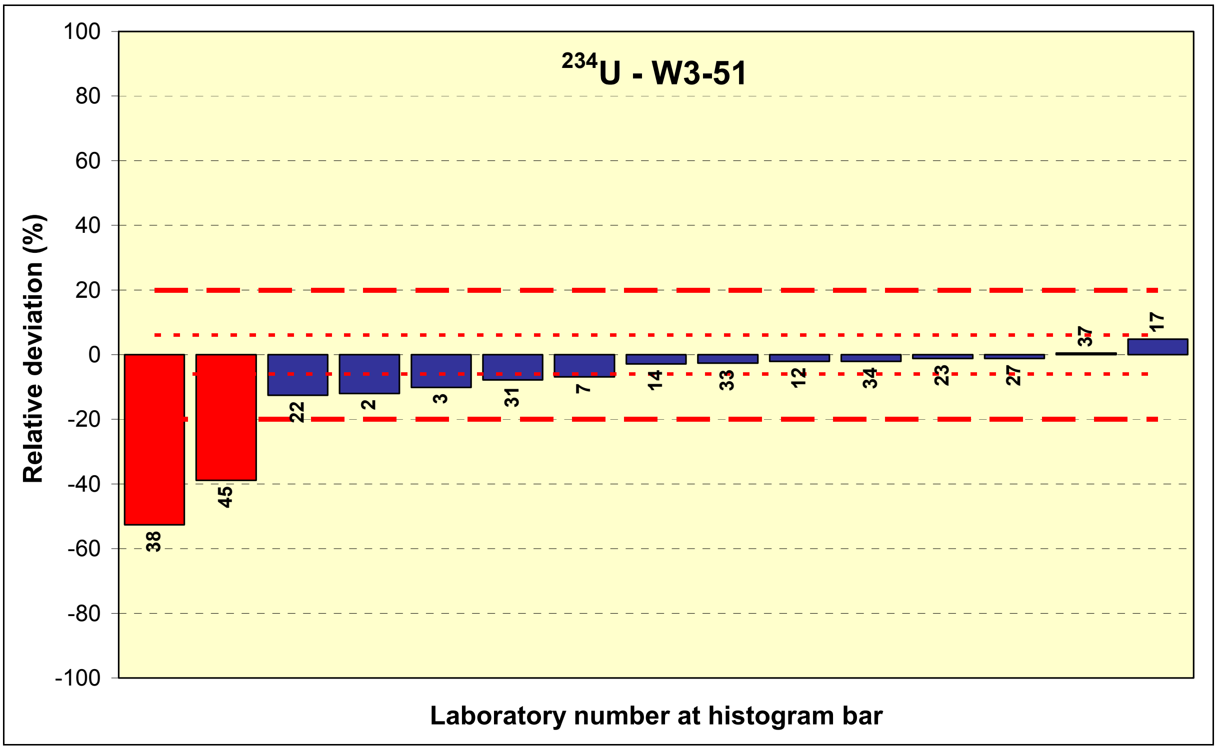

Fig. 66: Deviation chart of the 15 measurement results for ^{234}U in Water 3-51 plotted as D/A_{ref} in ascending order. Range of ± 20 % (dashed) and expanded uncertainty U_{ref} (dotted) are indicated

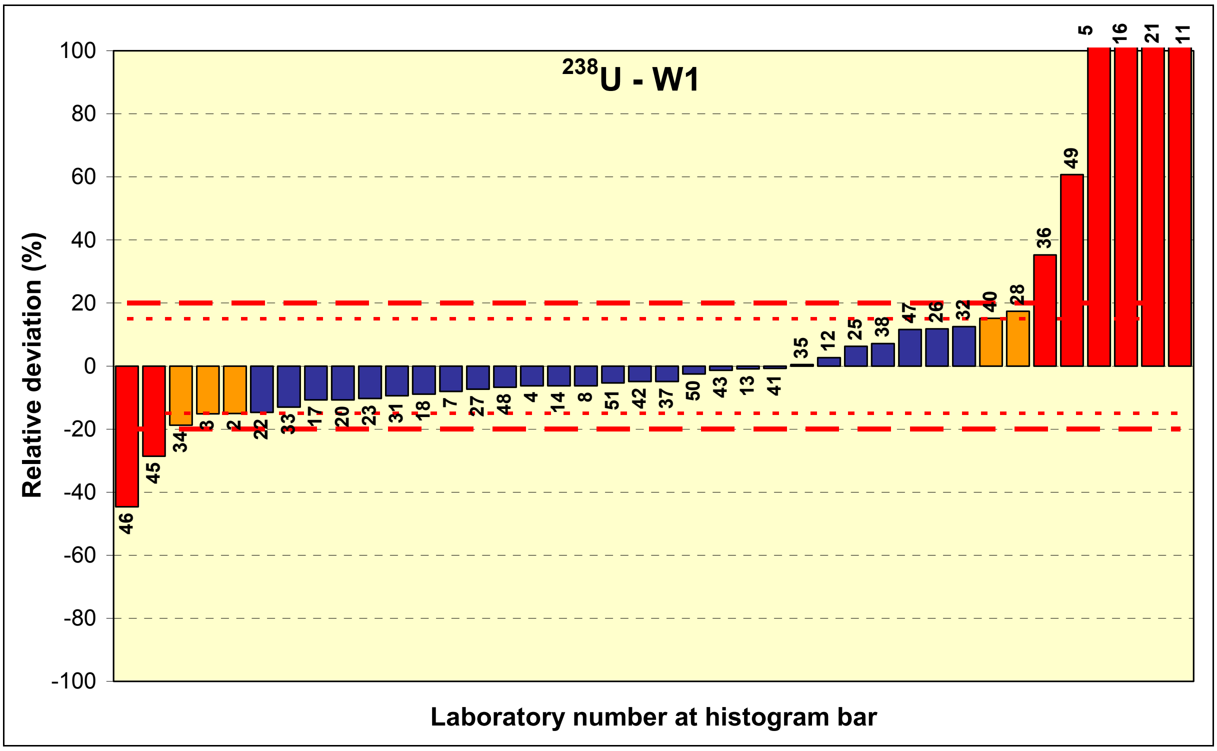

Fig. 67: Deviation chart of the 40 measurement results for ^{238}U in Water 1 plotted as D/A_{ref} in ascending order. Range of ± 20 % (dashed) and expanded uncertainty U_{ref} (dotted) are indicated

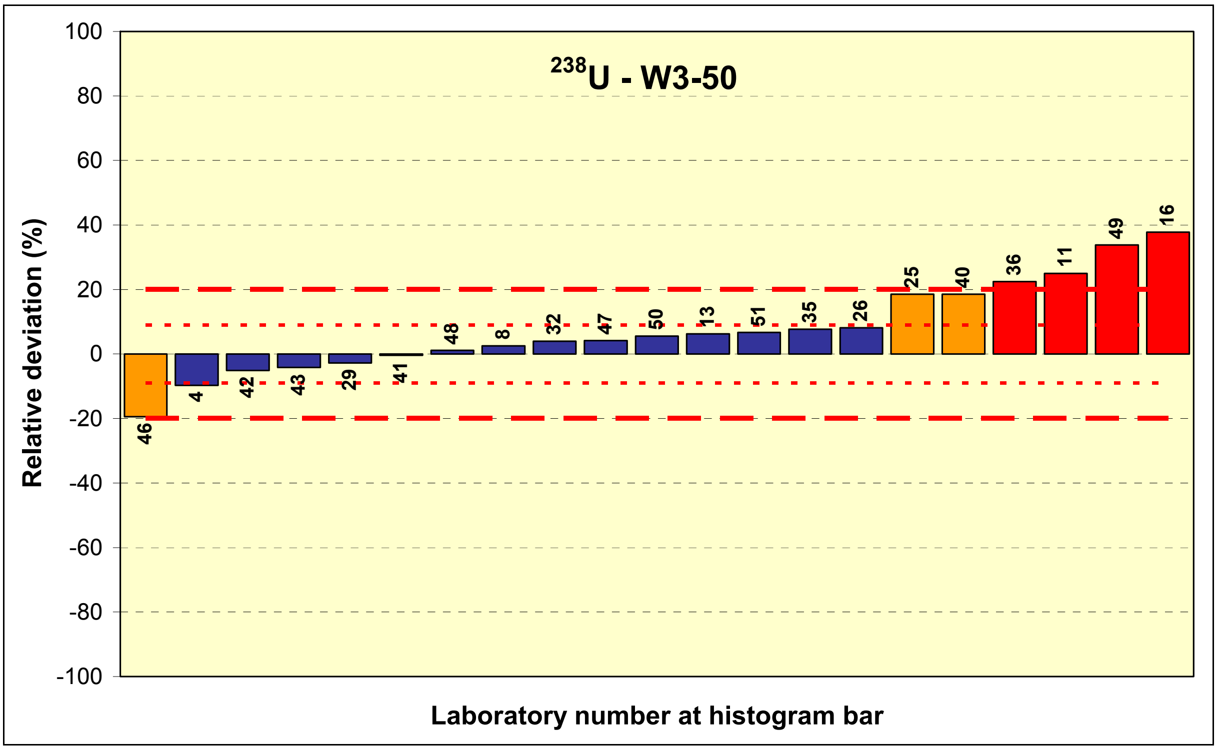

Fig. 68: Deviation chart of the 21 measurement results for ^{238}U in Water 3-50 plotted as D/A_{ref} in ascending order. Range of ± 20 % (dashed) and expanded uncertainty U_{ref} (dotted) are indicated

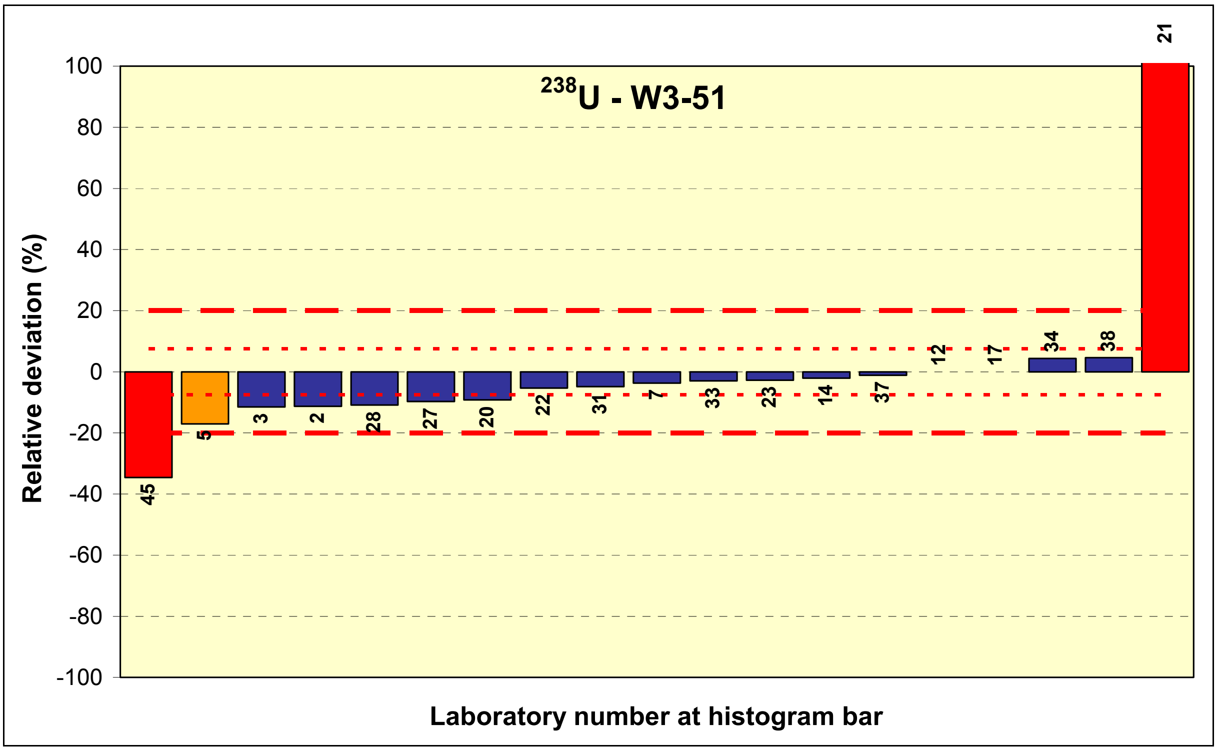

Fig. 69: Deviation chart of the 19 measurement results for ^{238}U in Water 3-51 plotted as D/A_{ref} in ascending order. Range of ± 20 % (dashed) and expanded uncertainty U_{ref} (dotted) are indicated

Table 47: Relative deviation of mean laboratory results A_{lab} from reference value A_{ref} and compatibility test (E_n numbers) for **^{226}Ra in Water 1**

lab no.	**mean activity concentration A_{lab} [mBq·L^{-1}]**	**expanded uncertainty U_{lab} [mBq·L^{-1}]**	**relative deviation D/A_{ref} [%]**	**E_n**	**compatibility**
2	99.1	11.15	1.1	0.06	YES
3	35.0	24.79	-64.3	-2.17	NO
4	96.5	17.12	-1.5	-0.07	YES
5	102.8	8.84	4.9	0.28	YES
7	113.0	12.00	15.3	0.78	YES
8	7.2	1.40	-92.7	-6.03	NO
9	-	-	-	-	-
10	-	-	-	-	-
11	90.0	**11.78**	-8.2	-0.42	YES
12	100.0	**27.99**	2.0	0.06	YES
13	67.0	**17.86**	-31.6	-1.33	NO
14	-	-	-	-	-
15	78.5	9.47	-19.9	-1.10	NO
16	20.7	3.77	-78.9	-5.00	NO
17	83.0	7.90	-15.3	-0.88	YES
18	70.9	11.94	-27.7	-1.42	NO
19	88.0	**17.08**	-10.2	-0.44	YES
20	85.5	**15.58**	-12.8	-0.58	YES
21	21.0	1.00	-78.6	-5.12	NO
22	96.5	12.37	-1.5	-0.08	YES
23	93.5	9.79	-4.6	-0.25	YES
24	760.0	295.77	675.5	2.24	NO
25	93.0	37.50	-5.1	-0.12	YES
26	49.4	3.60	-49.6	-3.15	NO
27	17.8	**2.83**	-81.9	-5.26	NO
28	83.4	**42.92**	-14.9	-0.32	YES
29	83.6	15.80	-14.7	-0.66	YES
31	92.5	10.98	-5.6	-0.30	YES
32	95.0	16.92	-3.1	-0.13	YES
33	98.6	13.62	0.6	0.03	YES
34	90.5	12.66	-7.7	-0.38	YES
35	66.4	8.59	-32.2	-1.83	NO
36	85.5	15.45	-12.8	-0.58	YES
37	102.0	11.67	4.1	0.21	YES
38	107.5	8.95	9.7	0.54	YES
39	99.3	**14.47**	1.3	0.06	YES
40	-	-	-	-	-
41	89.3	5.06	-8.9	-0.55	YES
42	96.5	14.28	-1.5	-0.07	YES
43	96.2	**6.07**	-1.9	-0.11	YES
45	163.0	50.00	66.3	1.25	NO
46	70.0	25.00	-28.6	-0.96	YES
47	81.5	20.75	-16.8	-0.64	YES
48	89.0	**6.72**	-9.2	-0.55	YES
49	89.7	8.15	-8.5	-0.49	YES
50	-	-	-	-	-
51	-	-	-	-	-

Table 48: Relative deviation of mean laboratory results A_{lab} from reference value A_{ref} and compatibility test (E_n numbers) for ^{226}**Ra in Water 2**

lab no.	**mean activity concentration A_{lab} [mBq·L^{-1}]**	**expanded uncertainty U_{lab} [mBq·L^{-1}]**	**relative deviation D/A_{ref} [%]**	**E_n**	**compatibility**
2	42.4	4.99	-1.5	-0.05	YES
3	10.0	20.00	-76.7	-1.45	NO
4	37.5	7.87	-12.8	-0.41	YES
5	53.5	7.87	24.3	0.77	YES
7	48.0	6.00	11.6	0.40	YES
8	12.2	2.24	-71.7	-2.75	NO
9	-	-	-	-	-
10	-	-	-	-	-
11	50.5	**6.48**	17.4	0.59	YES
12	82.0	**27.39**	90.7	1.32	NO
13	51.5	**7.84**	19.8	0.63	YES
14	-	-	-	-	-
15	29.0	3.66	-32.6	-1.21	NO
16	13.5	2.80	-68.7	-2.60	NO
17	39.5	3.87	-8.1	-0.30	YES
18	34.2	5.82	-20.5	-0.71	YES
19	46.0	**12.98**	7.0	0.18	YES
20	42.5	**7.95**	-1.2	-0.04	YES
21	28.0	3.00	-34.9	-1.32	NO
22	41.0	6.64	-4.7	-0.16	YES
23	42.4	4.44	-1.5	-0.05	YES
24	82.0	34.10	90.7	1.09	NO
25	43.0	16.56	0.0	0.00	YES
26	32.5	2.72	-24.3	-0.92	YES
27	7.1	**2.43**	-83.4	-3.18	NO
28	31.2	**16.07**	-27.6	-0.61	YES
29	38.0	8.20	-11.6	-0.36	YES
31	30.5	3.25	-29.1	-1.09	NO
32	50.0	8.97	16.3	0.49	YES
33	43.0	7.01	-0.1	0.00	YES
34	42.5	7.14	-1.2	-0.04	YES
35	40.4	5.75	-6.2	-0.21	YES
36	53.0	8.32	23.3	0.72	YES
37	49.0	6.29	14.0	0.47	YES
38	46.5	4.86	8.1	0.29	YES
39	44.9	**6.54**	4.4	0.15	YES
40	-	-	-	-	-
41	47.7	6.40	10.9	0.37	YES
42	52.5	8.47	22.1	0.68	YES
43	46.4	**2.93**	7.8	0.29	YES
45	124.0	50.00	188.4	1.58	NO
46	34.5	12.01	-19.8	-0.52	YES
47	37.0	12.50	-14.0	-0.36	YES
48	43.5	**3.67**	1.2	0.04	YES
49	37.4	7.27	-13.0	-0.42	YES
50	-	-	-	-	-
51	-	-	-	-	-

Table 49: Relative deviation of mean laboratory results A_{lab} from reference value A_{ref} and compatibility test (E_n numbers) for ^{228}**Ra in Water 1**

lab no.	**mean activity concentration A_{lab} [mBq·L^{-1}]**	**expanded uncertainty U_{lab} [mBq·L^{-1}]**	**relative deviation D/A_{ref} [%]**	**E_n**	**compatibility**
2	31.5	5.04	12.5	0.45	YES
3	< 80	-	-	-	-
4	24.5	4.84	-12.5	-0.45	YES
5	34.5	13.26	23.1	0.44	YES
7	-	-	-	-	-
8	8.5	9.27	-69.6	-1.77	NO
9	-	-	-	-	-
10	-	-	-	-	-
11	35.0	**7.86**	25.0	0.71	YES
12	70.0	14.79	150.0	2.63	NO
13	< 68	-	-	-	-
14	-	-	-	-	-
15	48.0	15.00	71.4	1.24	NO
16	-	-	-	-	-
17	111.5	22.15	298.2	3.64	NO
18	< 1046	-	-	-	-
19	32.0	**9.10**	14.3	0.37	YES
20	34.0	5.11	21.4	0.76	YES
21	27.0	5.00	-3.6	-0.13	YES
22	38.5	5.93	37.5	1.24	NO
23	30.4	4.46	8.4	0.31	YES
24	885.0	343.41	3060.7	2.50	NO
25	31.0	12.86	10.7	0.21	YES
26	-	-	-	-	-
27	65.9	27.00	135.4	1.37	NO
28	22.2	**9.63**	-20.7	-0.51	YES
29	27.7	13.60	-1.1	-0.02	YES
31	-	-	-	-	-
32	37.5	11.01	33.9	0.76	YES
33	43.0	12.28	53.6	1.10	NO
34	28.4	8.20	1.4	0.04	YES
35	38.0	10.05	35.7	0.85	YES
36	43.5	45.94	55.4	0.33	YES
37	-	-	-	-	-
38	81.0	13.49	189.3	3.59	NO
39	27.6	6.03	-1.4	-0.05	YES
40	-	-	-	-	-
41	30.0	5.90	7.1	0.24	YES
42	34.0	3.36	21.4	0.87	YES
43	-	-	-	-	-
45	-	-	-	-	-
46	-	-	-	-	-
47	29.5	9.35	5.4	0.14	YES
48	36.5	**8.77**	30.4	0.80	YES
49	-	-	-	-	-
50	-	-	-	-	-
51	-	-	-	-	-

Table 50: Relative deviation of mean laboratory results A_{lab} from reference value A_{ref} and compatibility test (E_n numbers) for ^{228}**Ra in Water 2**

lab no.	**mean activity concentration A_{lab} [mBq·L^{-1}]**	**expanded uncertainty U_{lab} [mBq·L^{-1}]**	**relative deviation D/A_{ref} [%]**	**E_n**	**compatibility**
2	82.5	7.57	11.5	0.60	YES
3	< 100	-	-	-	-
4	20.5	4.04	-72.3	-4.23	NO
5	39.3	14.39	-47.0	-1.85	NO
7	-	-	-	-	-
8	67.0	15.24	-9.5	-0.36	YES
9	-	-	-	-	-
10	-	-	-	-	-
11	110.0	**22.44**	48.6	1.41	NO
12	128.0	26.81	73.0	1.84	NO
13	112.5	**30.59**	52.0	1.17	NO
14	-	-	-	-	-
15	80.5	16.86	8.8	0.31	YES
16	-	-	-	-	-
17	146.5	29.33	98.0	2.29	NO
18	< 1038	-	-	-	-
19	61.0	**10.72**	-17.6	-0.81	YES
20	80.0	8.33	8.1	0.41	YES
21	72.0	7.00	-2.7	-0.14	YES
22	92.0	14.65	24.3	0.95	YES
23	70.4	8.89	-4.9	-0.24	YES
24	26.0	**10.62**	-64.9	-3.00	NO
25	67.5	26.96	-8.8	-0.22	YES
26	-	-	-	-	-
27	< 25.8	-	-	-	-
28	50.7	**21.29**	-31.6	-0.96	YES
29	80.3	20.80	8.5	0.26	YES
31	-	-	-	-	-
32	98.5	28.92	33.1	0.78	YES
33	103.0	16.64	39.2	1.41	NO
34	55.8	20.65	-24.7	-0.76	YES
35	65.5	13.99	-11.5	-0.46	YES
36	118.0	**65.03**	59.5	0.67	YES
37	-	-	-	-	-
38	99.5	14.73	34.5	1.34	NO
39	85.8	10.36	15.9	0.74	YES
40	-	-	-	-	-
41	77.8	7.71	5.1	0.26	YES
42	94.0	7.70	27.0	1.40	NO
43	-	-	-	-	-
45	-	-	-	-	-
46	-	-	-	-	-
47	73.0	20.25	-1.4	-0.04	YES
48	71.5	**9.89**	-3.4	-0.16	YES
49	-	-	-	-	-
50	-	-	-	-	-
51	-	-	-	-	-

Table 51: Relative deviation of mean laboratory results A_{lab} from reference value A_{ref} and compatibility test (E_n numbers) for **^{234}U in Water 1**

lab no.	**mean activity concentration A_{lab} [mBq·L^{-1}]**	**expanded uncertainty U_{lab} [mBq·L^{-1}]**	**relative deviation D/A_{ref} [%]**	**E_n**	**compatibility**
2	13.5	2.02	-10.0	-0.50	YES
3	13.0	5.00	-13.3	-0.37	YES
4	13.5	1.52	-10.0	-0.56	YES
5	-	-	-	-	-
7	14.4	4.00	-4.0	-0.13	YES
8	14.7	1.92	-2.3	-0.12	YES
9	-	-	-	-	-
10	-	-	-	-	-
11	-	-	-	-	-
12	15.5	**3.32**	3.3	0.13	YES
13	13.7	**1.35**	-9.0	-0.52	YES
14	14.5	1.72	-3.7	-0.20	YES
15	-	-	-	-	-
16	43.3	12.22	188.3	2.27	NO
17	11.3	4.00	-25.0	-0.82	YES
18	14.1	0.90	-6.3	-0.40	YES
19	-	-	-	-	-
20	-	-	-	-	-
21	-	-	-	-	-
22	16.0	1.47	6.7	0.38	YES
23	13.3	0.74	-11.3	-0.73	YES
24	-	-	-	-	-
25	16.0	2.12	6.3	0.31	YES
26	19.0	1.65	26.4	1.44	NO
27	13.7	**0.75**	-9.0	-0.58	YES
28	-	-	-	-	-
29	<15	-	-	-	-
31	14.6	1.03	-3.0	-0.19	YES
32	16.8	1.71	11.7	0.63	YES
33	14.3	**3.00**	-4.7	-0.19	YES
34	13.8	2.18	-8.0	-0.39	YES
35	16.4	2.87	9.0	0.37	YES
36	20.4	**1.64**	36.0	1.97	NO
37	14.0	1.61	-6.7	-0.37	YES
38	16.5	**2.13**	10.0	0.49	YES
39	-	-	-	-	-
40	17.4	2.40	16.0	0.74	YES
41	14.5	0.87	-3.5	-0.22	YES
42	14.2	4.32	-5.3	-0.16	YES
43	13.7	**1.50**	-9.0	-0.51	YES
45	15.0	4.80	0.0	0.00	YES
46	6.2	3.10	-58.7	-2.31	NO
47	16.5	4.00	10.0	0.33	YES
48	13.8	**1.55**	-8.3	-0.46	YES
49	20.0	2.38	33.3	1.54	NO
50	14.3	1.47	-4.9	-0.28	YES
51	13.9	1.42	-7.3	-0.42	YES

Table 52: Relative deviation of mean laboratory results A_{lab} from reference value A_{ref} and compatibility test (E_n numbers) for **^{234}U in Waters 3-50 and 3-51**

lab no.	**mean activity concentration A_{lab} [mBq·L^{-1}]**	**expanded uncertainty U_{lab} [mBq·L^{-1}]**	**relative deviation D/A_{ref} [%]**	**E_n**	**compatibility**
W3-50					
4	32.5	3.70	-25.3	-2.31	NO
8	45.6	5.19	4.8	0.35	YES
10	-	-	-	-	-
11	-	-	-	-	-
13	45.5	**3.08**	4.6	0.46	YES
16	29.5	12.31	-32.2	-1.10	NO
19	-	-	-	-	-
25	50.2	6.02	15.4	1.00	YES
26	42.6	4.41	-2.0	-0.17	YES
29	44.0	6.00	1.1	0.07	YES
32	42.6	3.51	-2.2	-0.21	YES
35	48.6	5.91	11.7	0.77	YES
36	51.1	5.34	17.4	1.23	NO
39	-	-	-	-	-
40	52.9	6.40	21.6	1.33	NO
41	42.6	2.32	-2.0	-0.23	YES
42	41.0	11.78	-5.7	-0.21	YES
43	40.8	3.14	-6.3	-0.63	YES
46	17.5	8.70	-59.8	-2.83	NO
47	46.0	6.98	5.7	0.33	YES
48	44.6	5.79	2.4	0.16	YES
49	42.2	**5.27**	-3.0	-0.21	YES
50	44.2	4.49	1.7	0.13	YES
51	46.4	4.28	6.6	0.54	YES
W3-51					
2	35.7	6.98	-12.0	-0.71	YES
3	36.5	12.50	-10.1	-0.35	YES
5	-	-	-	-	-
7	37.9	10.00	-6.9	-0.29	YES
9	-	-	-	-	-
12	40.0	**8.35**	-2.1	-0.10	YES
14	39.7	4.57	-2.9	-0.24	YES
15	-	-	-	-	-
17	43.0	15.29	4.8	0.14	YES
18	-	-	-	-	-
20	-	-	-	-	-
21	-	-	-	-	-
22	35.5	2.73	-12.5	-1.50	NO
23	40.4	2.47	-1.1	-0.15	YES
24	-	-	-	-	-
27	40.4	**2.16**	-1.1	-0.15	YES
28	-	-	-	-	-
31	37.5	**2.57**	-7.8	-0.97	YES
33	39.8	8.89	-2.6	-0.12	YES
34	40.0	4.71	-2.1	-0.17	YES
37	41.1	4.16	0.5	0.04	YES
38	18.0	6.00	-52.6	-3.54	NO
45	24.0	6.95	-38.9	-2.30	NO

Table 53: Relative deviation of mean laboratory results A_{lab} from reference value A_{ref} and compatibility test (E_n numbers) for **^{238}U in Water 1**

lab no.	**mean activity concentration A_{lab} [mBq·L^{-1}]**	**expanded uncertainty U_{lab} [mBq·L^{-1}]**	**relative deviation D/A_{ref} [%]**	**E_n**	**compatibility**
2	9.5	1.58	-15.0	-0.73	YES
3	9.5	5.01	-15.2	-0.32	YES
4	10.5	1.29	-6.2	-0.33	YES
5	24.3	9.58	117.1	1.35	NO
7	10.3	2.00	-8.0	-0.34	YES
8	10.5	1.42	-6.2	-0.32	YES
9	-	-	-	-	-
10	-	-	-	-	-
11	120.0	**18.29**	971.4	5.92	NO
12	11.5	2.49	2.7	0.10	YES
13	11.1	**1.27**	-0.9	-0.05	YES
14	10.5	1.31	-6.2	-0.33	YES
15	-	-	-	-	-
16	45.0	12.36	301.3	2.70	NO
17	10.0	3.57	-10.7	-0.30	YES
18	10.2	0.71	-8.9	-0.54	YES
19	-	-	-	-	-
20	10.0	**14.32**	-10.7	-0.08	YES
21	92.0	13.00	721.4	6.16	NO
22	9.6	0.96	-14.7	-0.85	YES
23	10.1	0.56	-10.3	-0.64	YES
24	-	-	-	-	-
25	11.9	1.64	6.3	0.30	YES
26	12.5	1.12	11.8	0.65	YES
27	10.4	**0.59**	-7.3	-0.46	YES
28	13.2	11.80	17.4	0.16	YES
29	< 15	-	-	-	-
31	10.2	0.63	-9.4	-0.58	YES
32	12.6	1.64	12.5	0.59	YES
33	9.7	**2.08**	-13.0	-0.54	YES
34	9.1	1.74	-18.8	-0.86	YES
35	11.3	2.31	0.4	0.02	YES
36	15.2	**1.22**	35.3	1.89	NO
37	10.7	1.25	-4.9	-0.26	YES
38	12.0	**1.24**	7.1	0.38	YES
39	-	-	-	-	-
40	12.9	2.60	15.2	0.55	YES
41	11.1	0.71	-0.7	-0.04	YES
42	10.7	3.22	-4.9	-0.15	YES
43	11.1	**1.28**	-1.3	-0.07	YES
45	8.0	2.32	-28.6	-1.11	NO
46	6.2	3.10	-44.6	-1.41	NO
47	12.5	3.04	11.6	0.37	YES
48	10.5	**1.19**	-6.7	-0.36	YES
49	18.0	2.32	60.7	2.37	NO
50	10.9	1.10	-2.5	-0.14	YES
51	10.6	1.42	-5.4	-0.27	YES

Table 54: Relative deviation of mean laboratory results A_{lab} from reference value A_{ref} and compatibility test (E_n numbers) for ^{238}**U in Waters 3-50 and 3-51**

lab no.	**mean activity concentration A_{lab} [mBq·L^{-1}]**	**expanded uncertainty U_{lab} [mBq·L^{-1}]**	**relative deviation D/A_{ref} [%]**	**E_n**	**compatibility**
W3-50					
4	19.5	2.38	-9.7	-0.69	YES
8	22.2	2.66	2.5	0.17	YES
10	-	-	-	-	-
11	27.0	**3.62**	25.0	1.32	NO
13	23.0	**1.94**	6.2	0.50	YES
16	29.8	12.41	37.7	0.65	YES
19	-	-	-	-	-
25	25.6	3.20	18.5	1.08	NO
26	23.4	2.43	8.2	0.57	YES
29	21.0	6.00	-2.8	-0.10	YES
32	22.5	2.63	3.9	0.26	YES
35	23.3	**3.69**	7.6	0.40	YES
36	26.5	2.88	22.5	1.41	NO
39		-	-	-	-
40	25.6	4.80	18.5	0.77	YES
41	21.5	1.28	-0.4	-0.03	YES
42	20.5	5.89	-5.1	-0.18	YES
43	20.7	1.68	-4.2	-0.35	YES
46	17.4	8.70	-19.4	-0.47	YES
47	22.5	4.49	4.2	0.18	YES
48	21.9	2.88	1.2	0.07	YES
49	28.9	2.40	33.8	2.39	NO
50	22.8	2.38	5.5	0.39	YES
51	23.1	2.84	6.7	0.42	YES
W3-51					
2	18.0	4.06	-11.4	-0.56	YES
3	18.0	6.49	-11.6	-0.37	YES
5	16.8	7.50	-17.1	-0.48	YES
7	19.7	4.00	-3.7	-0.19	YES
9	-	-	-	-	-
12	20.5	3.83	0.0	0.00	YES
14	20.1	2.39	-2.1	-0.16	YES
15	-	-	-	-	-
17	20.5	7.27	0.0	0.00	YES
18	-	-	-	-	-
20	18.5	8.25	-9.3	-0.24	YES
21	121.0	23.00	465.3	4.36	NO
22	19.4	1.55	-5.3	-0.52	YES
23	19.9	1.34	-2.8	-0.29	YES
24	-	-	-	-	-
27	18.4	**1.87**	-9.7	-0.85	YES
28	18.2	**15.45**	-10.9	-0.15	YES
31	19.5	1.25	-4.9	-0.52	YES
33	19.9	4.58	-3.0	-0.13	YES
34	21.5	2.90	4.4	0.29	YES
37	20.3	2.16	-1.2	-0.09	YES
38	21.5	3.18	4.6	0.28	YES
45	13.0	3.77	-34.7	-1.83	NO

In order to allow a more detailed analysis, several statistical tests – taking the measurement uncertainty and that of the reference values into account – have been applied. Strictly speaking, tests including measurement uncertainty must be used with caution when the uncertainty estimation is poorly understood. We have already observed, in chapter 5 ("Reported results"), that this holds for about one third of the participating laboratories. Nevertheless, the selected performance test using the E_n criterion (ISO, 2005 and 1997) of the activity concentrations proves to be robust enough justifying its use in this evaluation.* The E_n number takes into account the absolute deviation of the activity concentration value reported by each laboratory (A_{lab}) from the reference value (A_{ref}) and the combination of expanded uncertainties associated to them (U_{lab} and U_{ref}). It will become clear below that the conclusions from the previous discussion (which ignored uncertainty) will in general be confirmed and some additional insight will be gained.

The performance statistic "E_n number" is calculated as:

$$E_n = \frac{A_{lab} - A_{ref}}{\sqrt{U_{lab}^2 + U_{ref}^2}} \qquad (23)$$

where

A_{lab} the participant's result, mean activity concentration;

A_{ref} the reference value;

U_{lab} the expanded uncertainty of the participant's result;

U_{ref} the expanded uncertainty of the reference value.

When the estimation of uncertainties is consistent with the Guide to the Expression of Uncertainty in Measurement (GUM, 1995), a measurement result with its uncertainty interval giving a level of confidence of 95 % (corresponding to $A_{lab} \pm U_{lab}$ with an expanded uncertainty $U_{lab} = k \cdot u_c$ with a coverage factor of $k \approx 2$) will overlap with the reference value A_{ref} (and its expanded uncertainty U_{ref}).

Therefore, E_n numbers are interpreted in the following way:

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

If $|E_n| > 1$, "warning signal", the laboratory values differ significantly from the reference value, sources of deviation should be investigated and corrected (orange colour in Tables 47-54);

In analogy to the interpretation of z-scores (ISO, 2005), a second level of critical value can be defined:

If $|E_n| > 1.5$, "action signal", there is urgent need to investigate and find the sources of the large deviation (red colour in Tables 47-54).

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

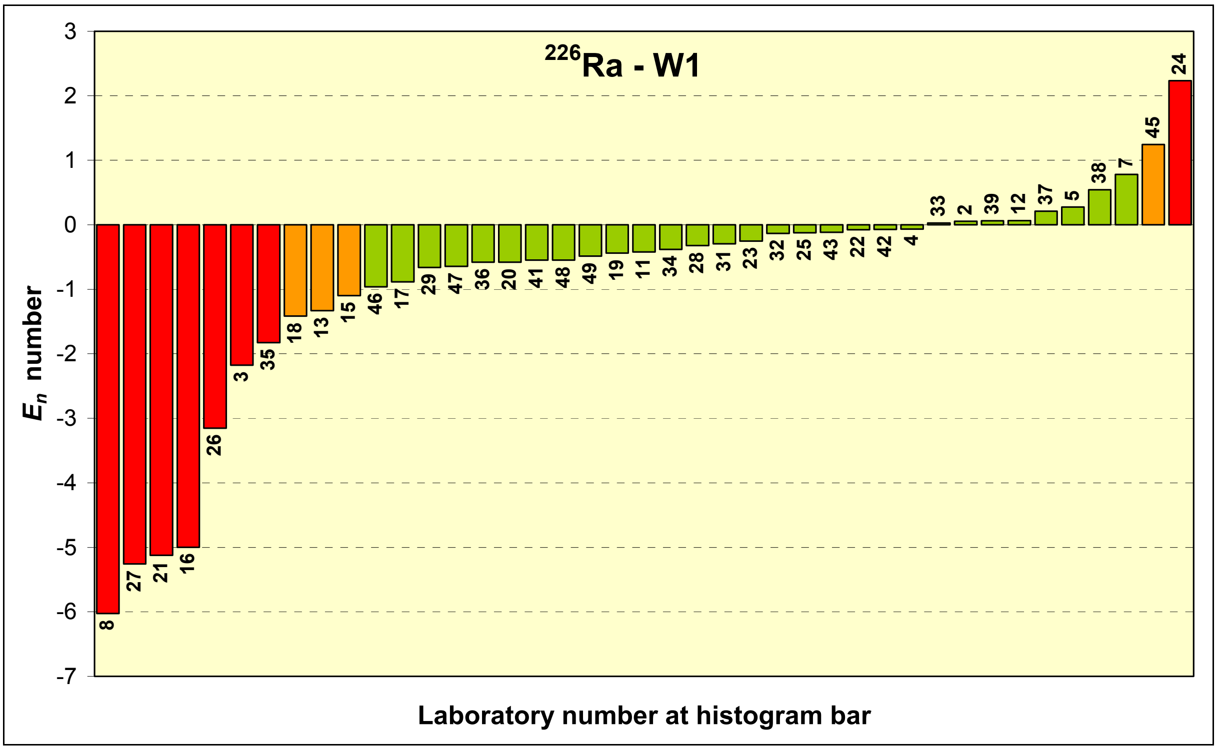

Fig. 70: E_n number for the determination of ^{226}Ra activity concentration in Water 1 (W1) obtained by 41 laboratories, sorted in ascending order

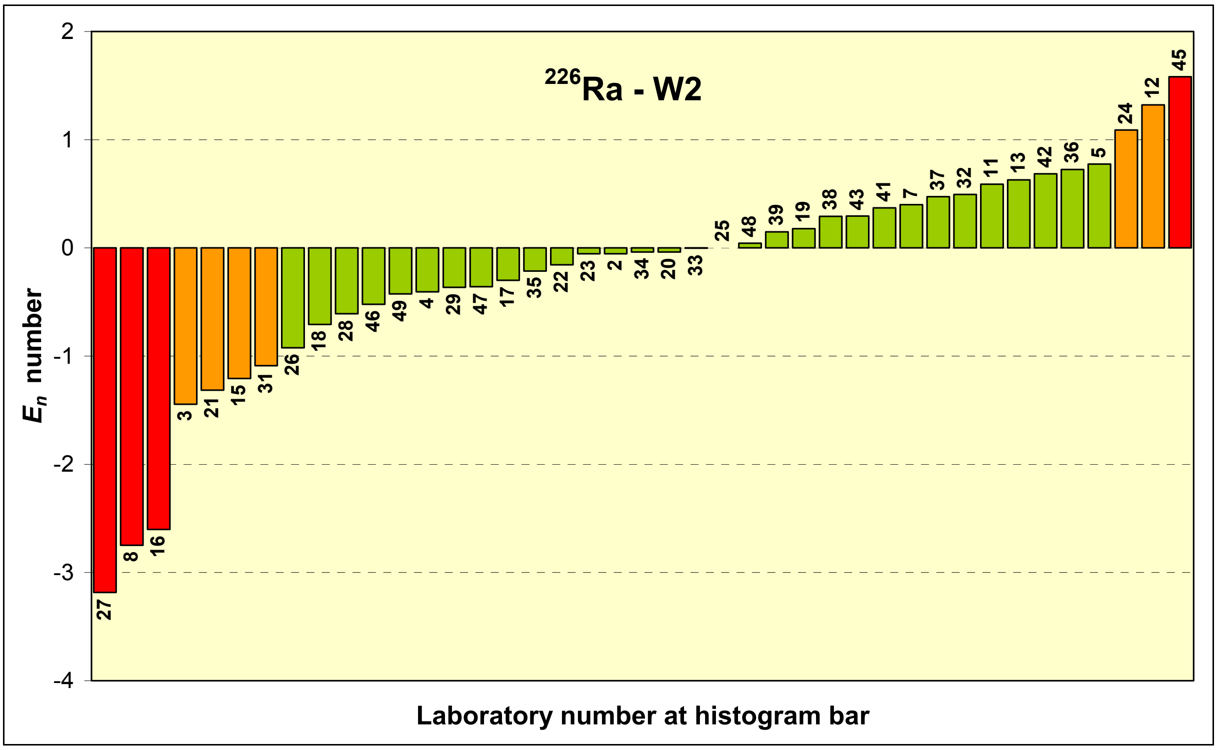

Fig. 71: E_n number for the determination of ^{226}Ra activity concentration in Water 2 obtained by 41 laboratories, sorted in ascending order

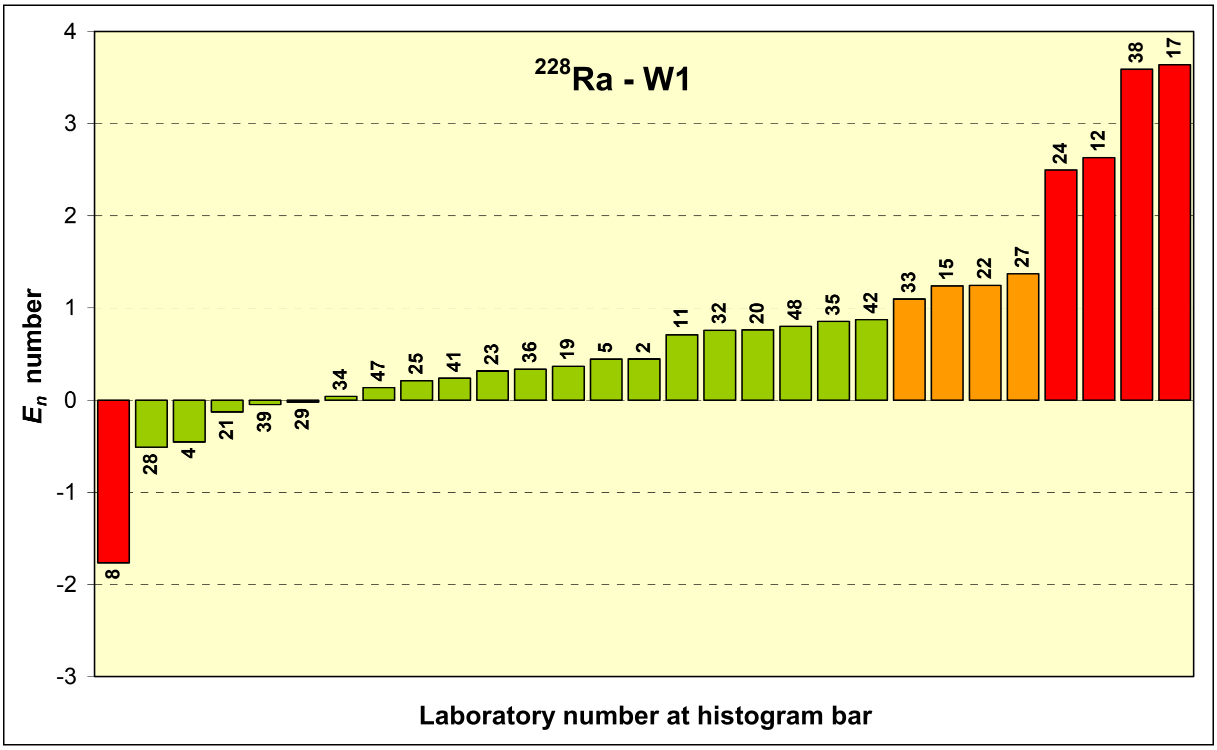

Fig. 72: E_n number for the determination of ^{228}Ra activity concentration in Water 1 obtained by 29 laboratories, sorted in ascending order

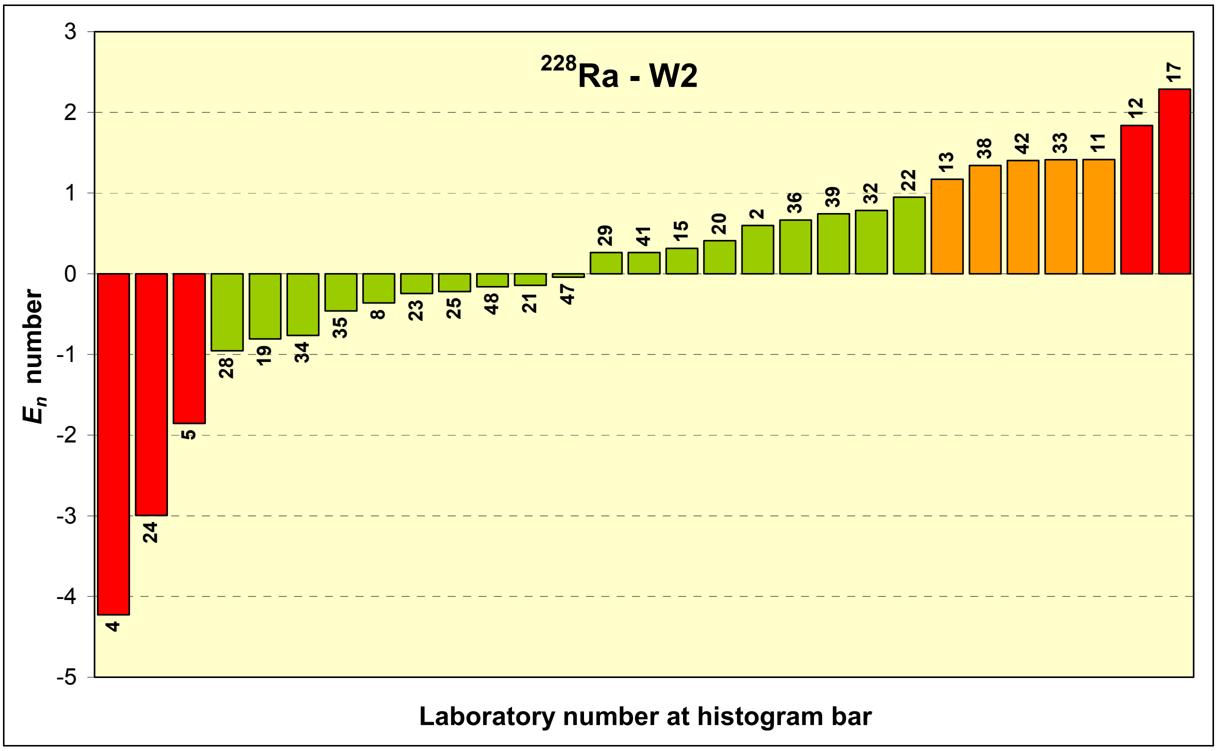

Fig. 73: E_n number for the determination of ^{228}Ra activity concentration in Water 2 obtained by 29 laboratories, sorted in ascending order

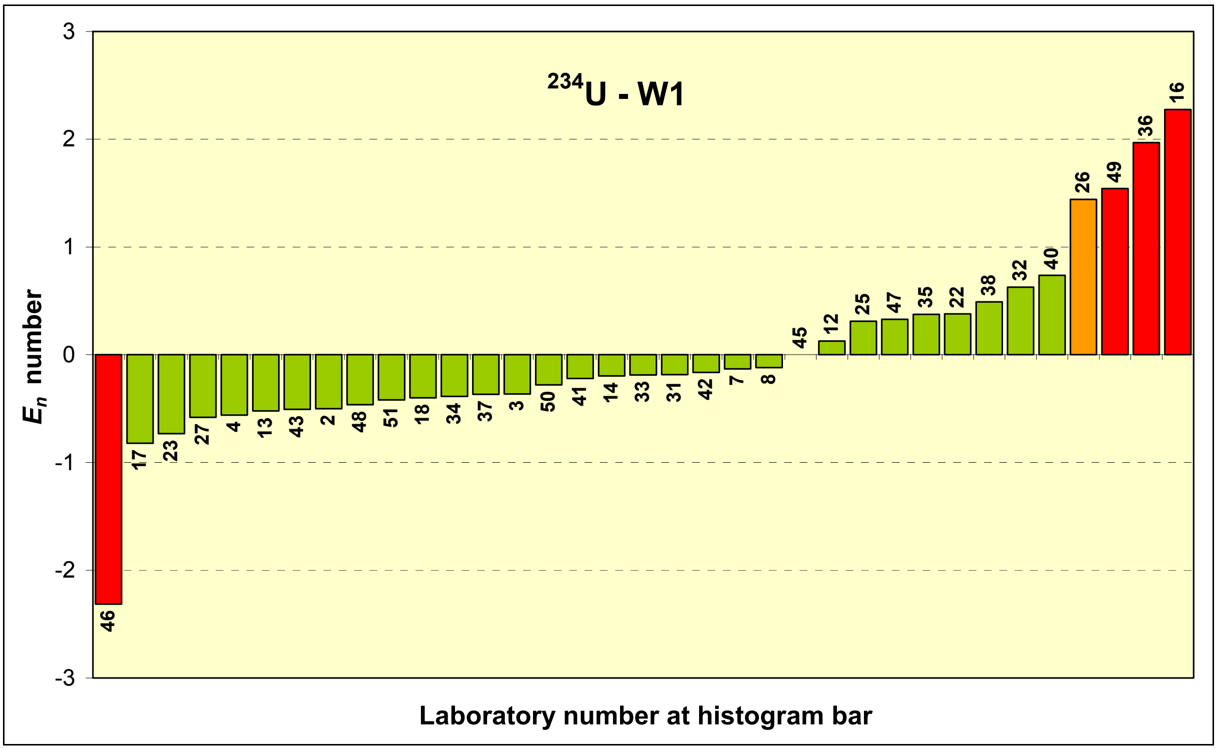

Fig. 74: E_n number for the determination of ^{234}U activity concentration in Water 1 obtained by 35 laboratories, sorted in ascending order

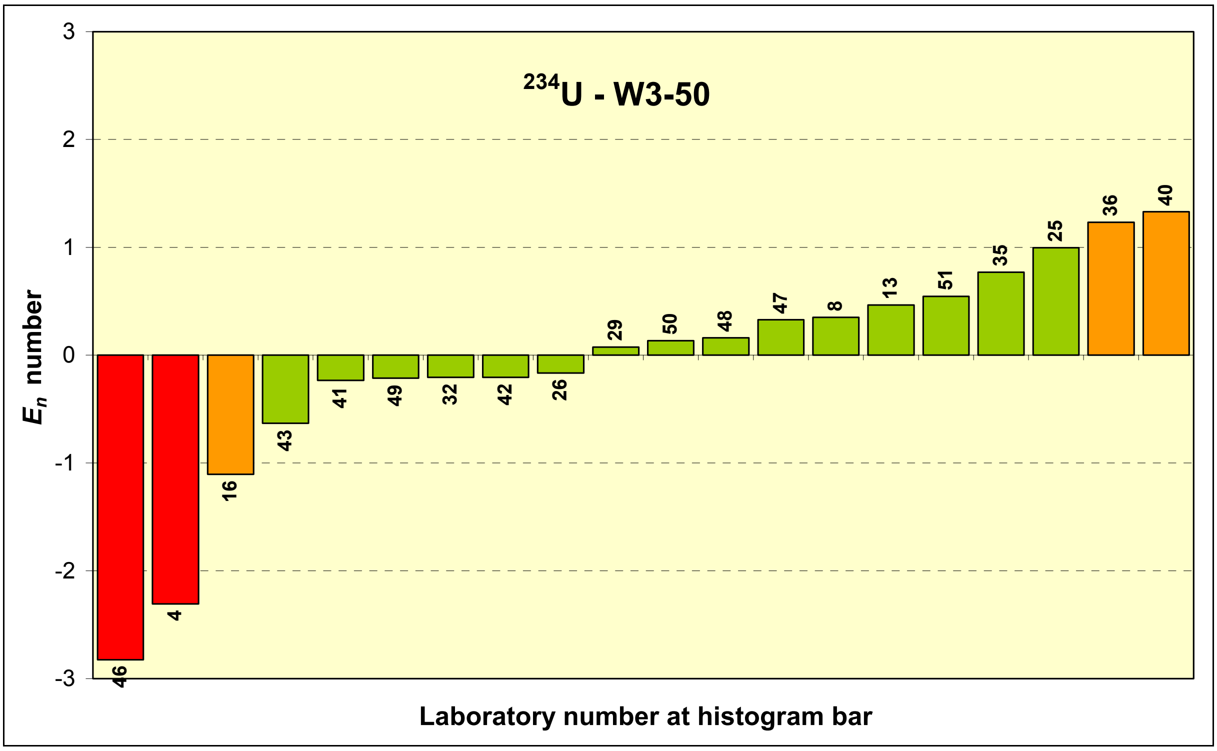

Fig. 75: E_n number for the determination of ^{234}U activity concentration in Water 3-50 obtained by 20 laboratories, sorted in ascending order

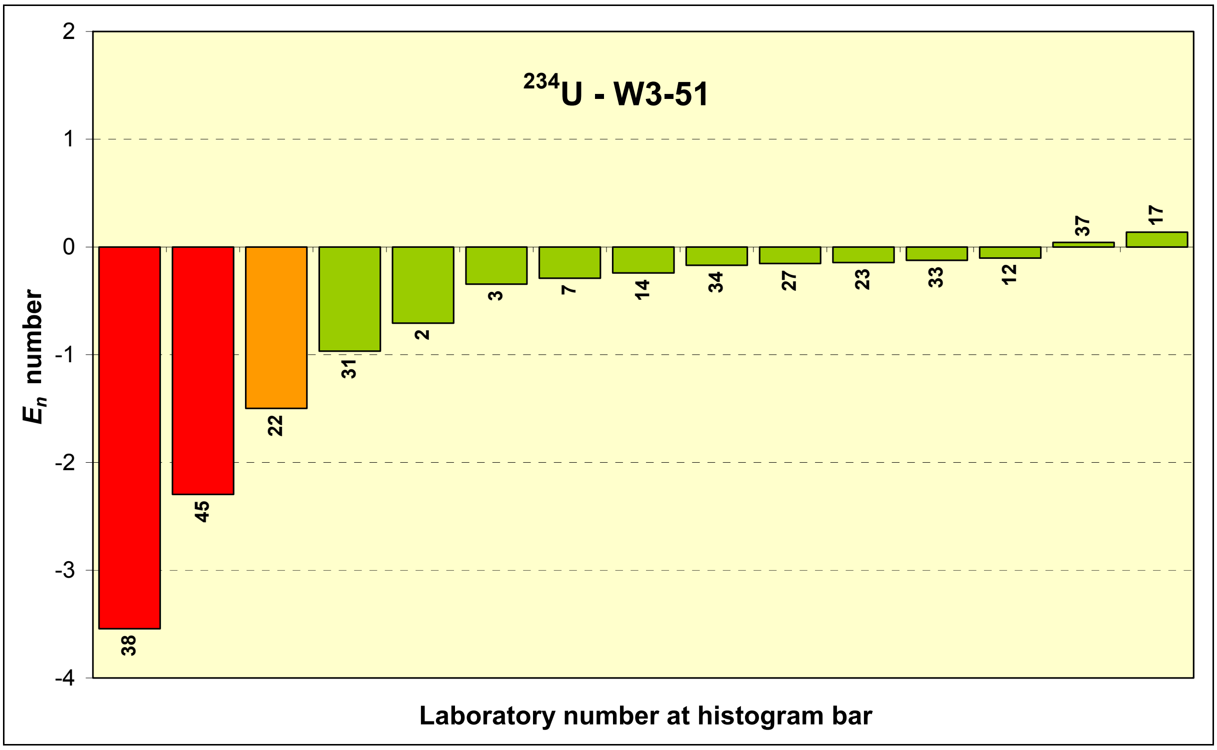

Fig. 76: E_n number for the determination of ^{234}U activity concentration in Water 3-51 obtained by 15 laboratories, sorted in ascending order

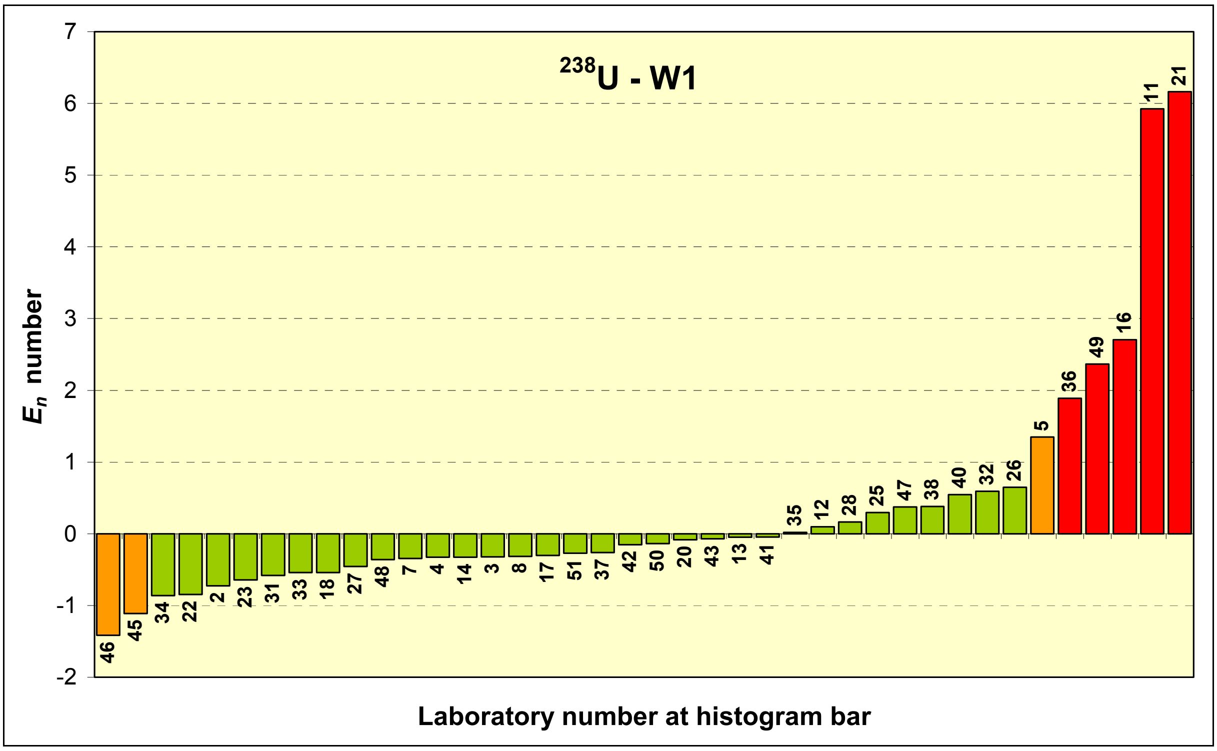

Fig. 77: E_n number for the determination of ^{238}U activity concentration in Water 1 obtained by 40 laboratories, sorted in ascending order

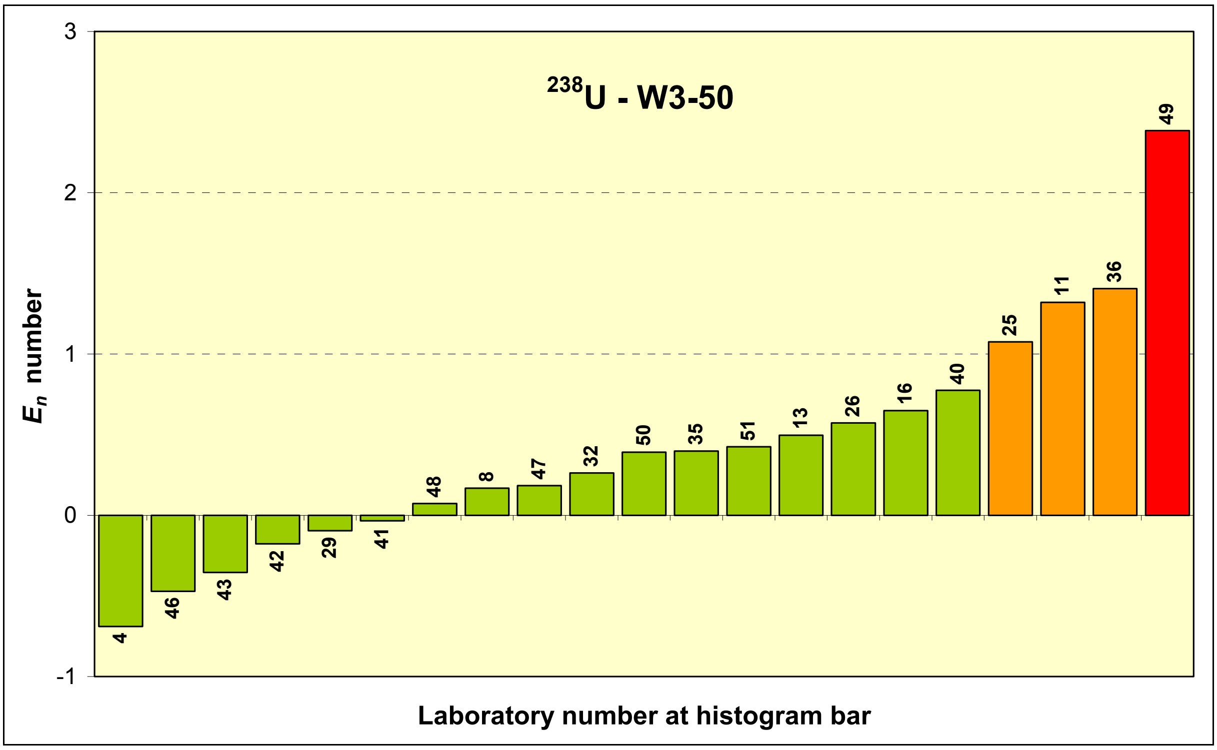

Fig. 78: E_n number for the determination of ^{238}U activity concentration in Water 3-50 obtained by 21 laboratories, sorted in ascending order

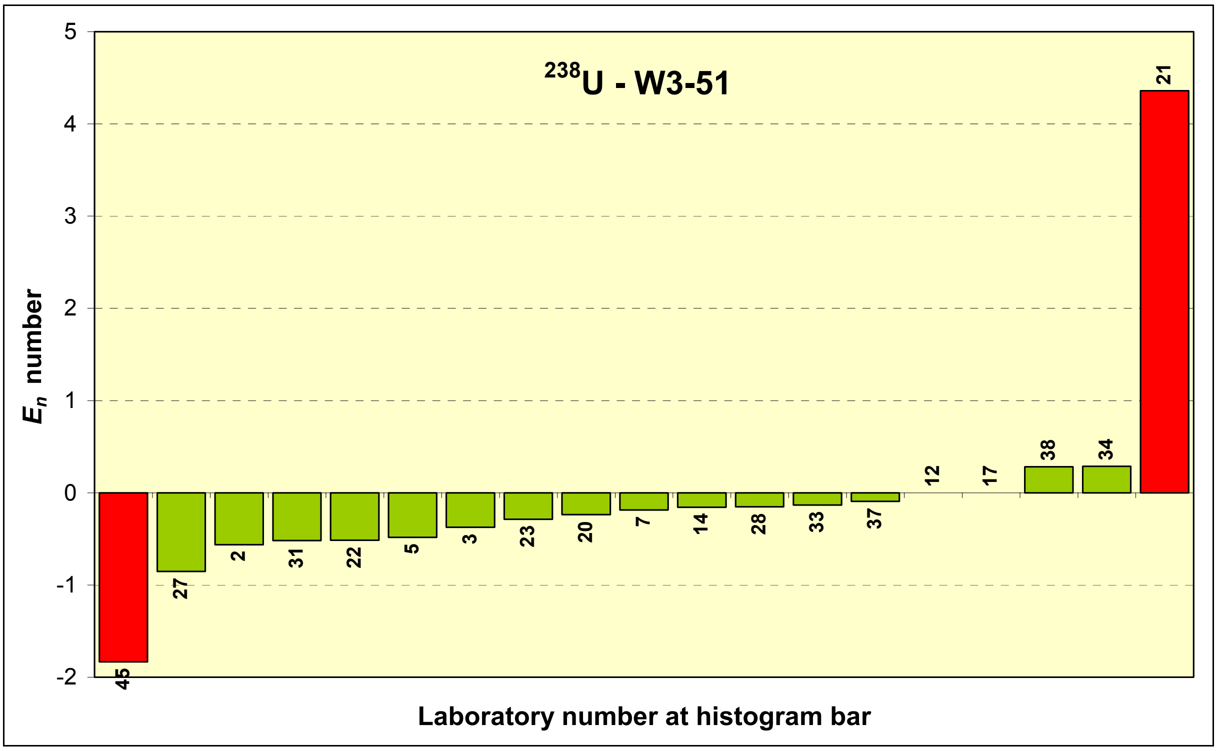

Fig. 79: E_n number for the determination of ^{238}U activity concentration in Water 3-51 obtained by 19 laboratories, sorted in ascending order

The E_n numbers and test results are given for each laboratory in the last two columns of Tables 47-54. Sorted in ascending order, the E_n numbers are graphically presented in Figures 70 - 79. The laboratories, which reported values below the detection limit of their methods (no. 3, 13, 18, 27 in the case of ^{228}Ra, lab 29 – ^{234}U and ^{238}U), could not be considered for the E_n criterion. Based on the E_n number criterion, 29 results out of 41 (71 %) are compatible with the reference activity concentrations for ^{226}Ra in Water 1 under the conditions of this test, 12 are not. Among those 12, eight laboratories (No.3, 8, 16, 21, 24, 26, 27 and 35) report largely incompatible results ($|E_n| > 1.5$). When comparing Figure 70 with 60, it is clear that results having large deviations from the reference value are scoring badly with E_n numbers as well. If a laboratory controls the measurement process well with only very small deviations *and* obtains a realistic estimate of uncertainty as well*, the E_n number is close to 0. Table 47 and Figure 70 depict several laboratories of this arbitrarily chosen category of $|E_n| < 0.1$, much narrower than requested for compliance: No. 2, 4, 12, 22, 33, 39 and 42.

Similarly for ^{226}Ra in Water 2, 31 results (76 %) are compatible with the reference value and 10 are not. Four laboratories (labs 8, 16, 27 and 45) of the incompatible ones have an $|E_n| > 1.5$ and all of them deviate more than 30 % from the reference value (Fig. 71).

In the case of ^{228}Ra, 20 and 19 values (~ 67 %) for Water 1 and Water 2, respectively, are satisfactory and 9/10 (for W1/W2) results are not. All of the laboratories with $|E_n| > 1.0$ for Water 1 (labs 8, 12, 15, 17, 22, 24, 27, 33, 38) show deviations larger than 30 % from A_{ref} (cf. Fig. 62). Similarly for Water 2 with one exception - laboratory 42, which deviates from the reference value < 30 % (~ 27 %) and its assigned E_n number is 1.4 (Fig. 63).

Table 55: Brief summary of the evaluation of laboratory results for the determination of ^{226}Ra, ^{228}Ra, ^{234}U and ^{238}U in mineral water

Measurand	**Sample**	**Number of compatible labs |En| ≤ 1**	**Number of incompatible labs |En| > 1**	**Number of largely incompatible labs |En| > 1.5**	**Number of labs with "large deviation"**
^{226}Ra	**W1**	29	4	8	10 **(>30%)**
	W2	31	6	4	9 **(>30%)**
^{228}Ra	**W1**	20	4	5	13 **(>30%)**
	W2	19	5	5	12 **(>30%)**
^{234}U	**W1**	30	1	4	6 **(>20%)**
	W3-50	15	3	2	4 **(>20%)**
	W3-51	12	1	2	2 **(>20%)**
^{238}U	**W1**	32	3	5	8 **(>20%)**
	W3-50	17	3	1	4 **(>20%)**
	W3-51	17	-	2	2 **(>20%)**

---

[*] Unfortunately, "guessing" a too large uncertainty would reduce the $|E_n|$ number without justification.

In terms of the E_n criterion for both uranium isotopes, between 11 % and 25 % of the scores are not compatible with the reference values. Again, in the majority of cases laboratories with $|E_n| > 1.0$ deviate more than 30 % from the reference value (cf. Figs. 74 - 79 and 64 - 69). The comparison between the relative deviations of the laboratory results and their E_n numbers, however, also shows the effect of estimating uncertainty *too small*: For example in the case of ^{234}U in Water 3-51, laboratory 22 with a still acceptable deviation of < 15 % is assigned a critical E_n number, because *not all contributions to uncertainty are correctly estimated* (e.g. an uncertainty contribution due to sample preparation was not taken into account).

The preceding discussion is summarised in Table 55.

Another modern type of plot, PomPlot, can be applied as well in order to compare the results. It also underlines the importance of the assigned uncertainties. The 'PomPlot' is used here for producing a summary overview of the participants' results (Pommé, 2006b). It displays relative deviations, *D/MAD*, of the mean results A_{lab} from the reference value on the horizontal axis and relative uncertainties, *u/MAD*, on the vertical axis. For both axes, the variables are expressed as multiples of *MAD*, which is defined as the median absolute deviation from the reference value:

$$MAD = \text{Median } |D_i|,\ (i=1,\ldots,n), \tag{24}$$

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

$$D_i = A_{lab,i} - A_{ref} \tag{25}$$

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

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

$$u_i^2 = u_c^2(A_{lab,i}) + u_c^2(A_{ref}) \tag{26}$$

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

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

Figure 80 shows the PomPlot for the ^{226}Ra results in Water 1. The reference value is indicated by a horizontal red dash. Outliers are indicated in gray and with an arrow (labs 8, 24, 27).

This is studied in more detail in Figure 81. Here the data (excluding labs 8, 24 and 27 indicated as outliers on the graph) are grouped according to the *u/MAD*-value and for each group the mean values of the normalised deviation $|D|/MAD$ is calculated, as well as the corresponding ratio of $|D|/u$. In the group of laboratories claiming the smallest uncertainty (*u/MAD*<0.8), the mean deviation is among the highest of all results, hence the uncertainty clearly underestimated ($<|D|/u>$ is higher than 1).

The best results concerning the activity measurement as well as uncertainty assessment can be found in the groups with $0.9 \leq u/MAD < 1.2$. Some laboratories seem to assign conservative uncertainty values (e.g labs 12, 25 and 28), even though their results are not worse than those of others. Indeed, a majority of the results can be found between $-2 < D/MAD < +2$, irrespective of the claimed uncertainty.

From the PomPlot for ^{226}Ra in Water 2 (Fig. 82) it is obvious that the reference value corresponds well with the mean and the median of lab results. Again, its relative uncertainty is not insignificant in the total uncertainty u (hence, the dots cannot reach the top). Similarly, the best results can be found in the groups with $1.0 \leq u/MAD < 1.1$ (Fig. 83).

Similar plots have been made for ^{228}Ra, ^{234}U and ^{238}U in the different water samples (Figs. 84 - 99). In the case of ^{228}Ra, the uncertainties on the reference values are relatively smaller and the lab results are closer to the top of the pyramid. Assuming that uncertainties would be assessed correctly by the laboratories, one should have an equal distribution of points above and below the $u/MAD = 1$ line. This is not the case for ^{228}Ra, showing that for many laboratories the estimation of uncertainty is not under control.

The ^{234}U and ^{238}U results for Water 1 show more outliers than the results for Water 3-50 and Water 3-51 (Figs. 88 - 99).Still, from a statistical point of view, there are too many points outside the $|\zeta| = 1$, 2 and 3 levels, which indicates that a significant fraction of the laboratories underestimates the uncertainties involved. Again, when analyzing this in more detail in Figures 89 to 99 one finds the best and the most realistic results around $0.9 \leq u/MAD < 1.1$. It seems that there is a group of laboratories that manages to provide accurate results together with a comprehensive uncertainty assessment.

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

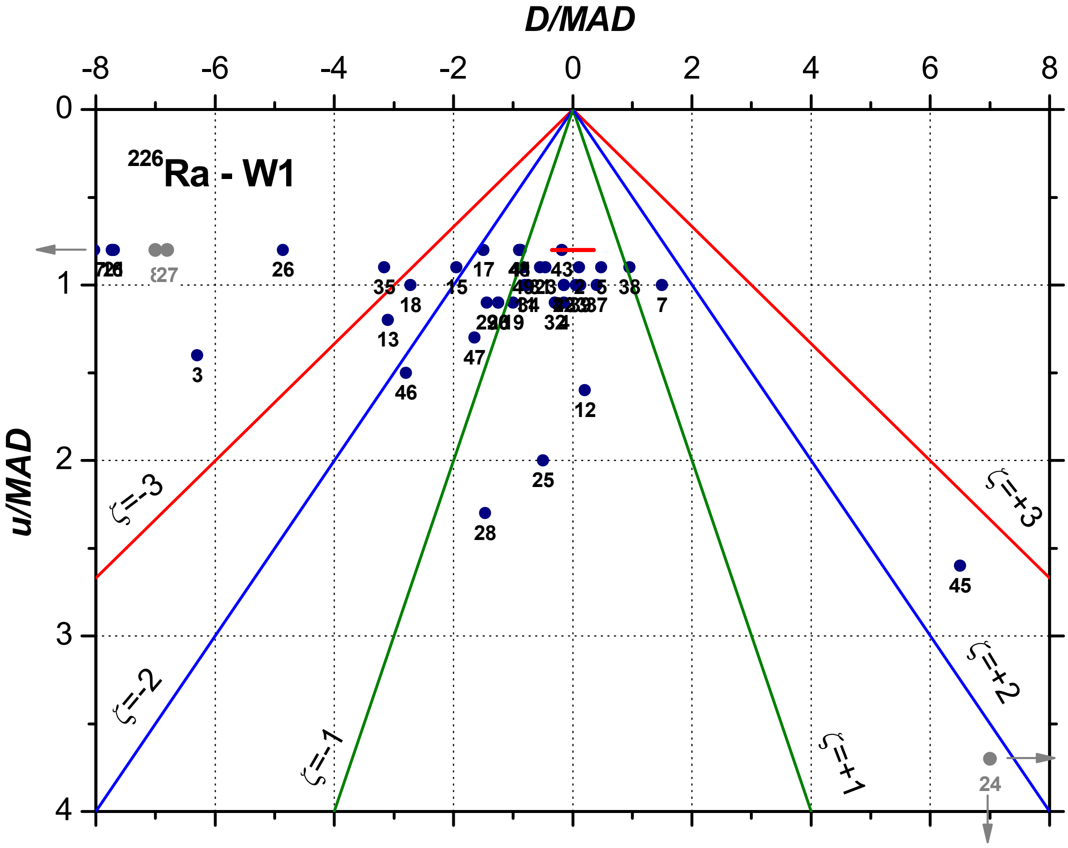

Fig. 80: PomPlot of the ^{226}Ra data for Water 1 (W1)

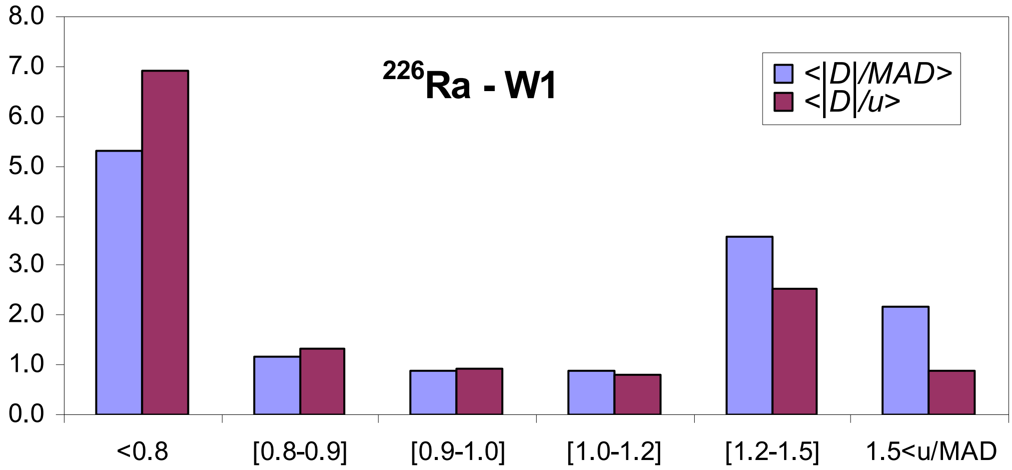

Fig. 81: Measures of average deviation <|*D*|/*MAD*>, <|*D*|/*u*> for different intervals of *u/MAD* in the case of ^{226}Ra in Water 1 (W1)

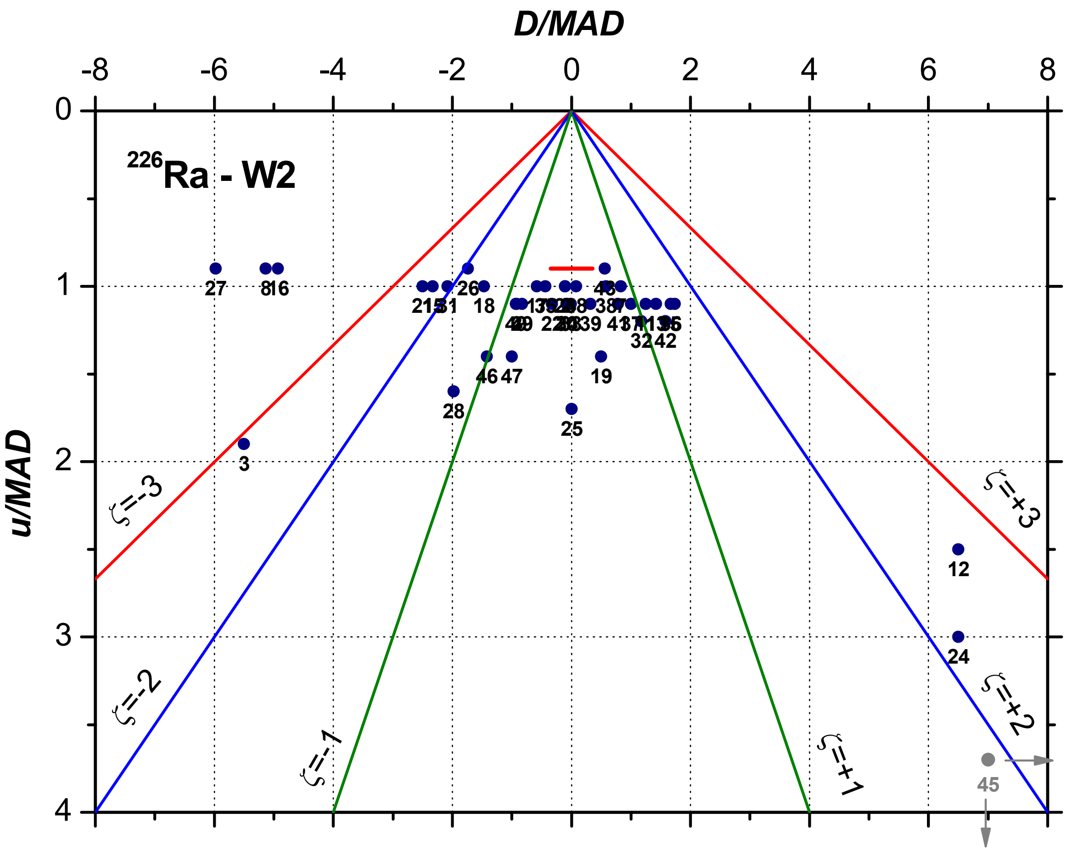

Fig. 82: PomPlot of the ^{226}Ra data for Water 2

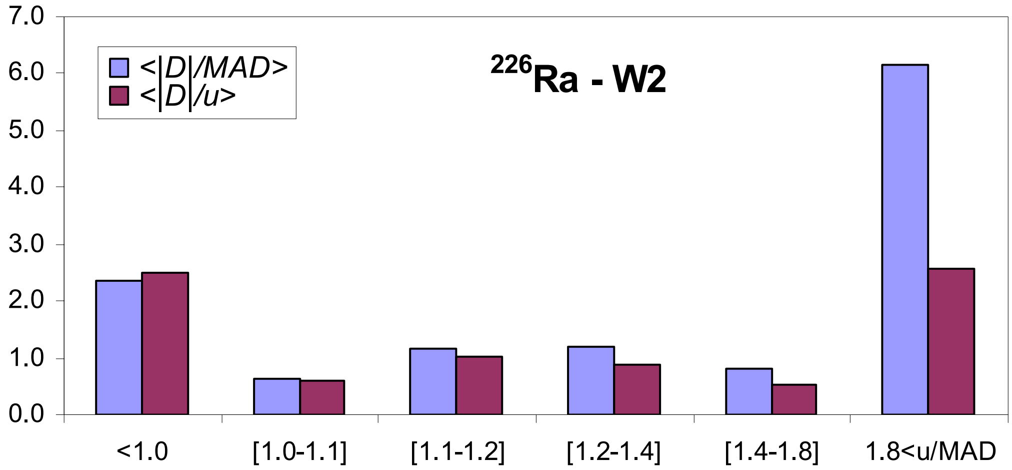

Fig. 83: Measures of average deviation <|*D*|/*MAD*>, <|*D*|/*u*> for different intervals of *u/MAD* in the case of ^{226}Ra in Water 2

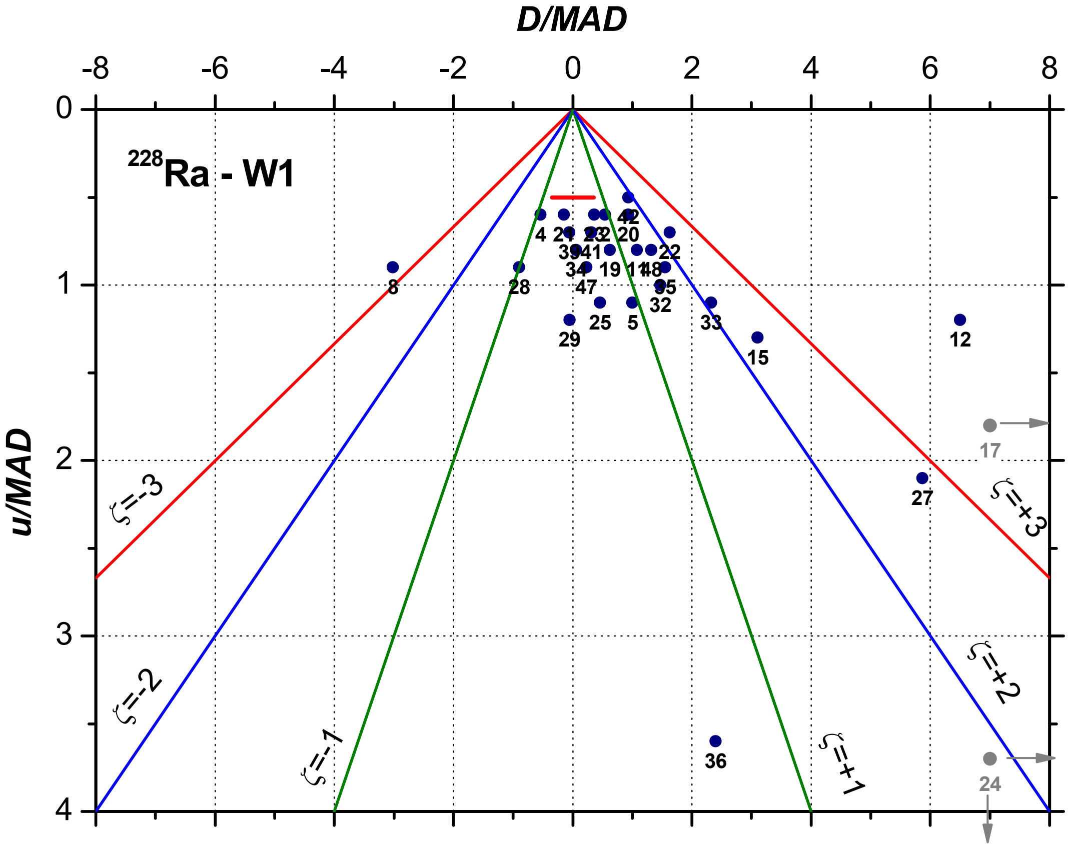

Fig. 84: PomPlot of the ^{228}Ra data for Water 1

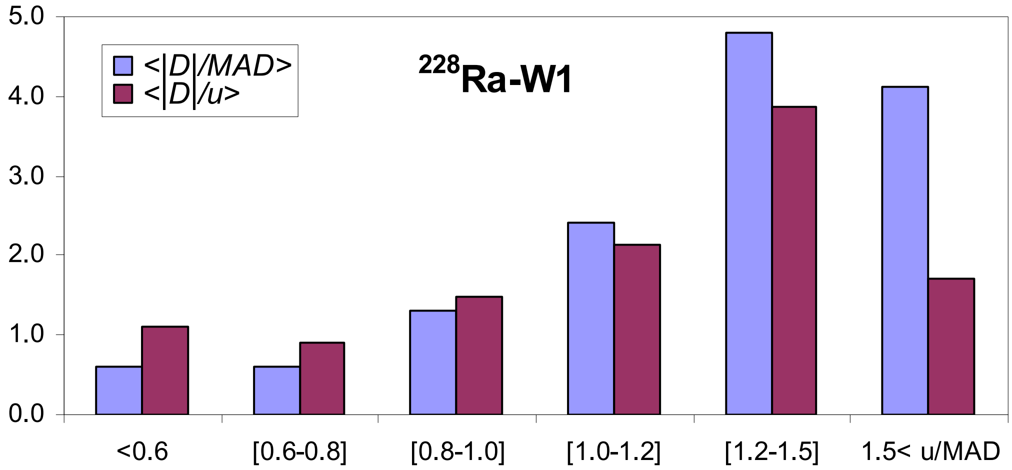

Fig. 85: Measures of average deviation <|*D*|/*MAD*>, <|*D*|/*u*> for different intervals of *u/MAD* in the case of ^{228}Ra in Water 1

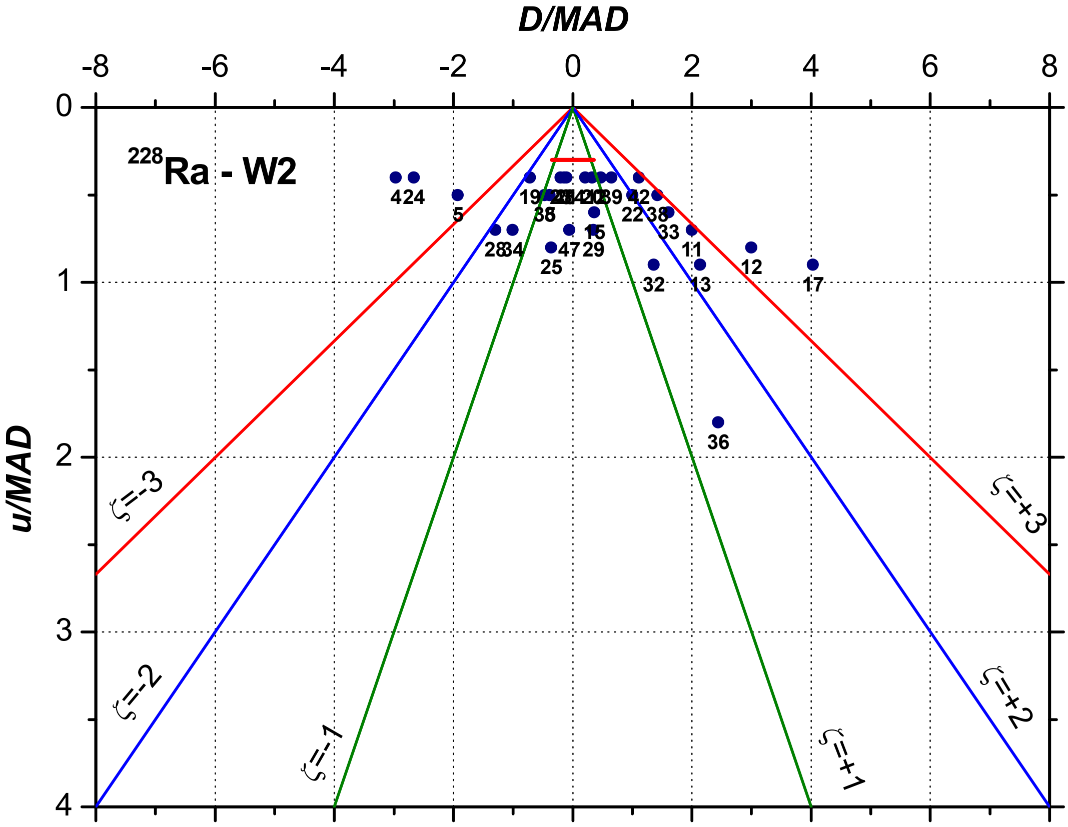

Fig. 86: PomPlot of the ^{228}Ra data for Water 2

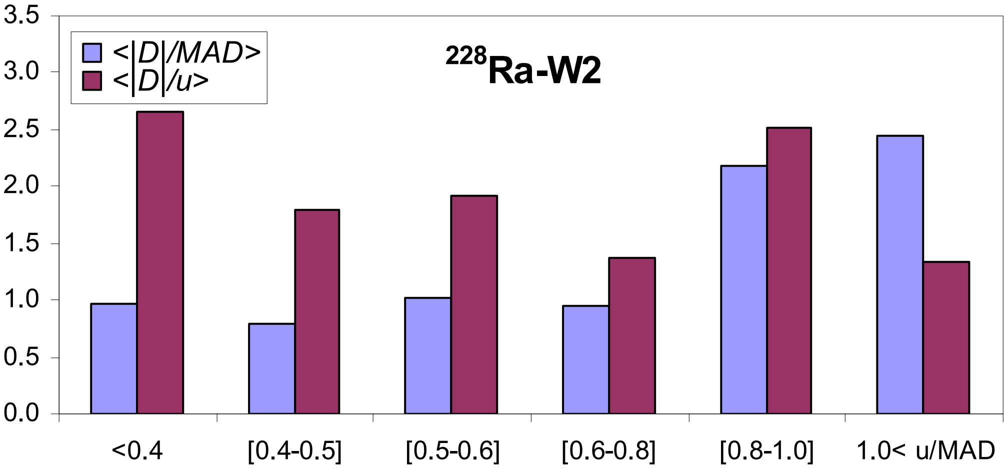

Fig. 87: Measures of average deviation <|*D*|/*MAD*>, <|*D*|/*u*> for different intervals of *u/MAD* in the case of ^{228}Ra in Water 2

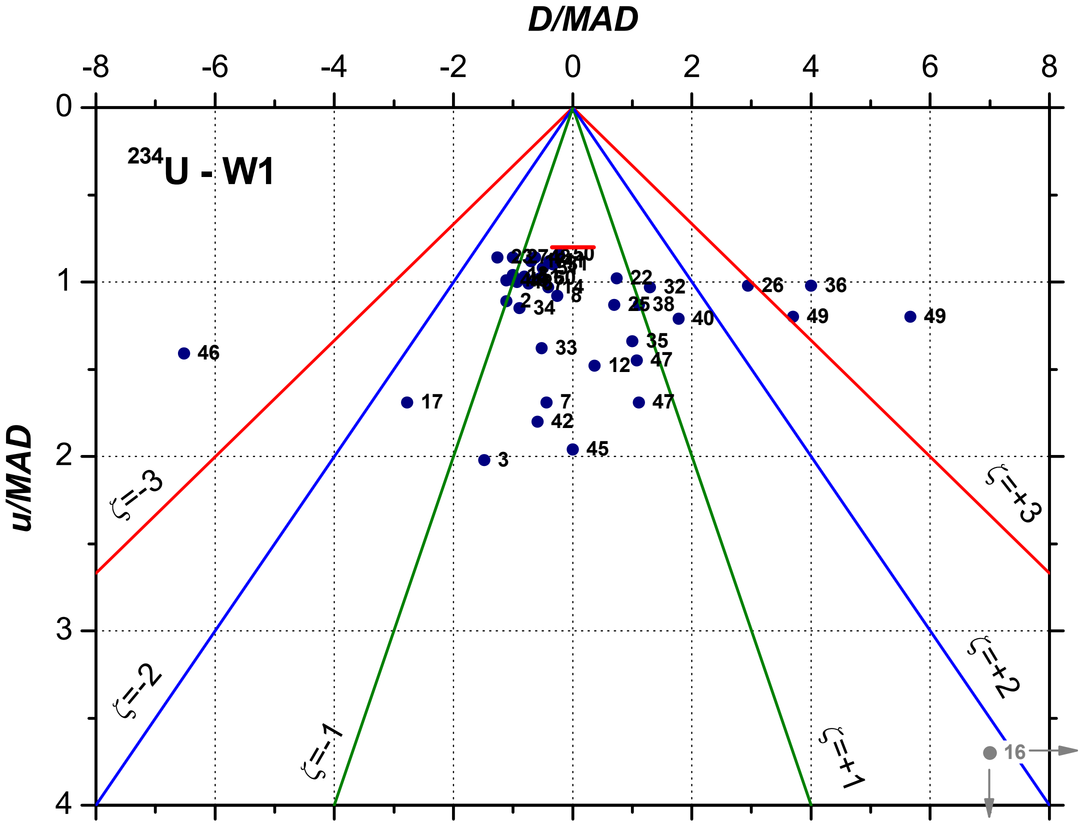

Fig. 88: PomPlot of the ^{234}U data for Water 1

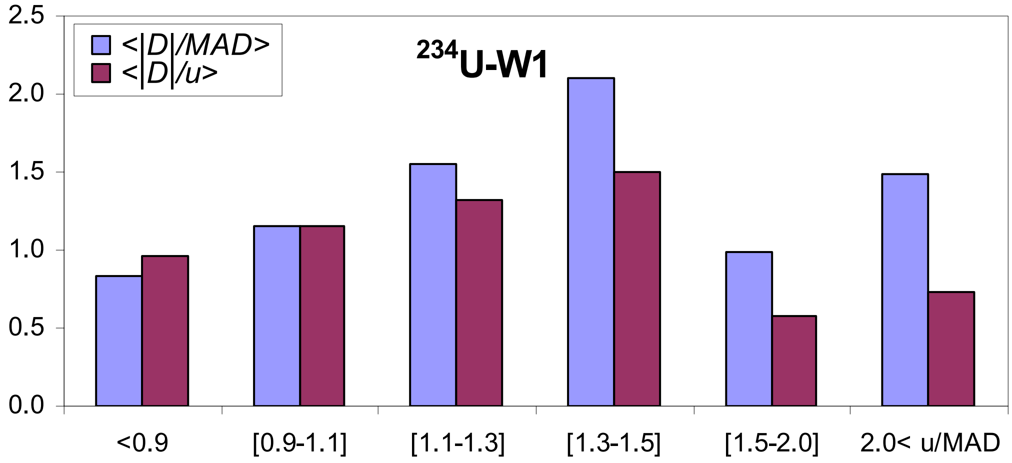

Fig. 89: Measures of average deviation <|*D*|/*MAD*>, <|*D*|/*u*> for different intervals of *u/MAD* in the case of ^{234}U in Water 1

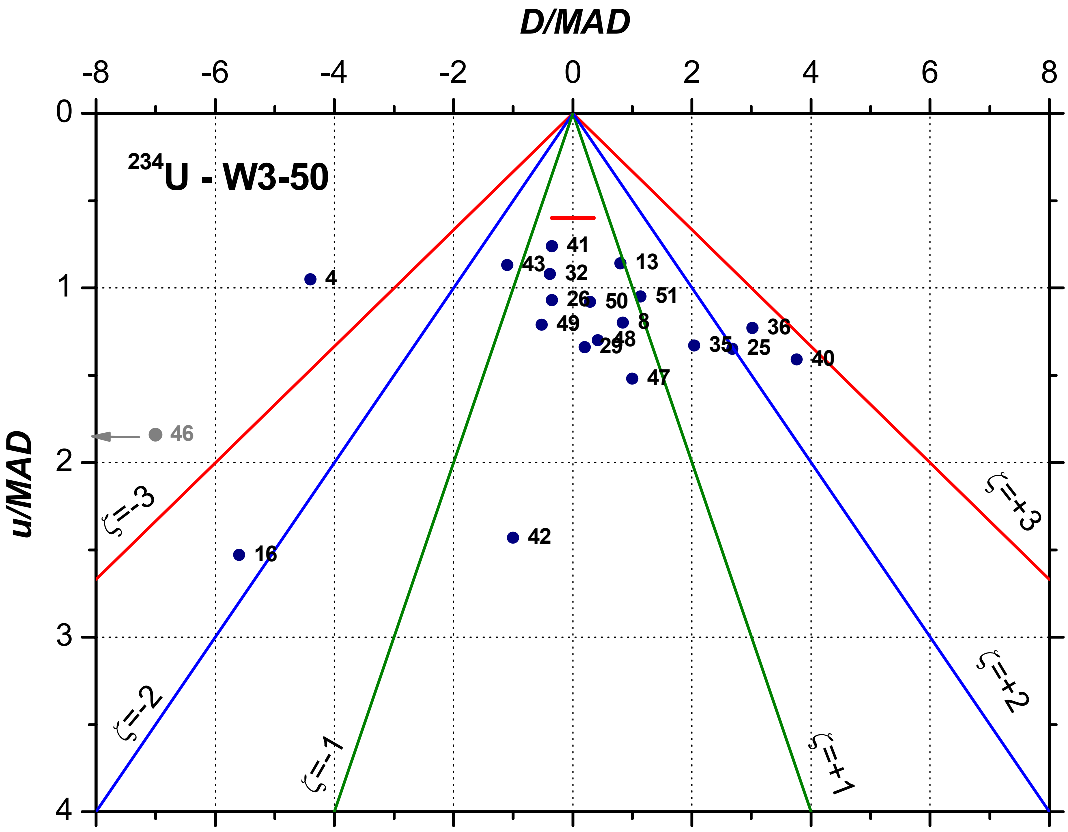

Fig. 90: PomPlot of the ^{234}U data for Water 3-50

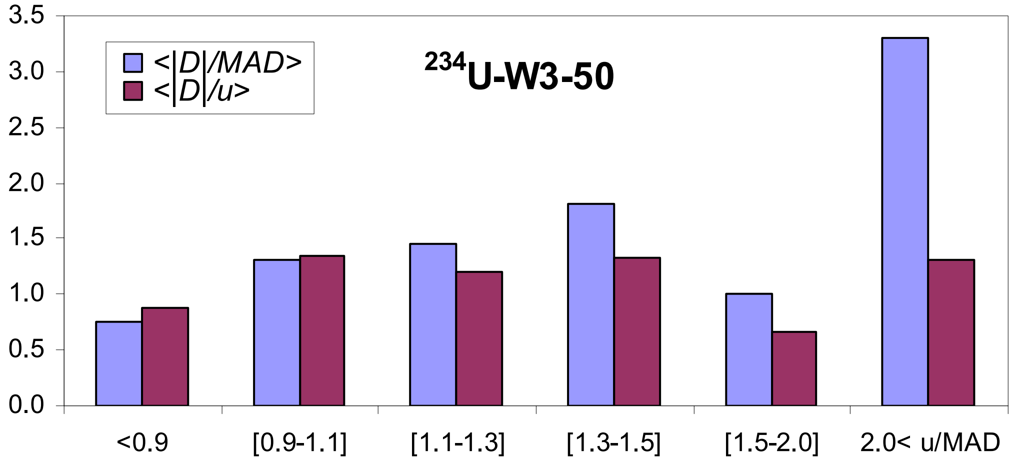

Fig. 91: Measures of average deviation <|*D*|/*MAD*>, <|*D*|/*u*> for different intervals of *u/MAD* in the case of ^{234}U in Water 3-50

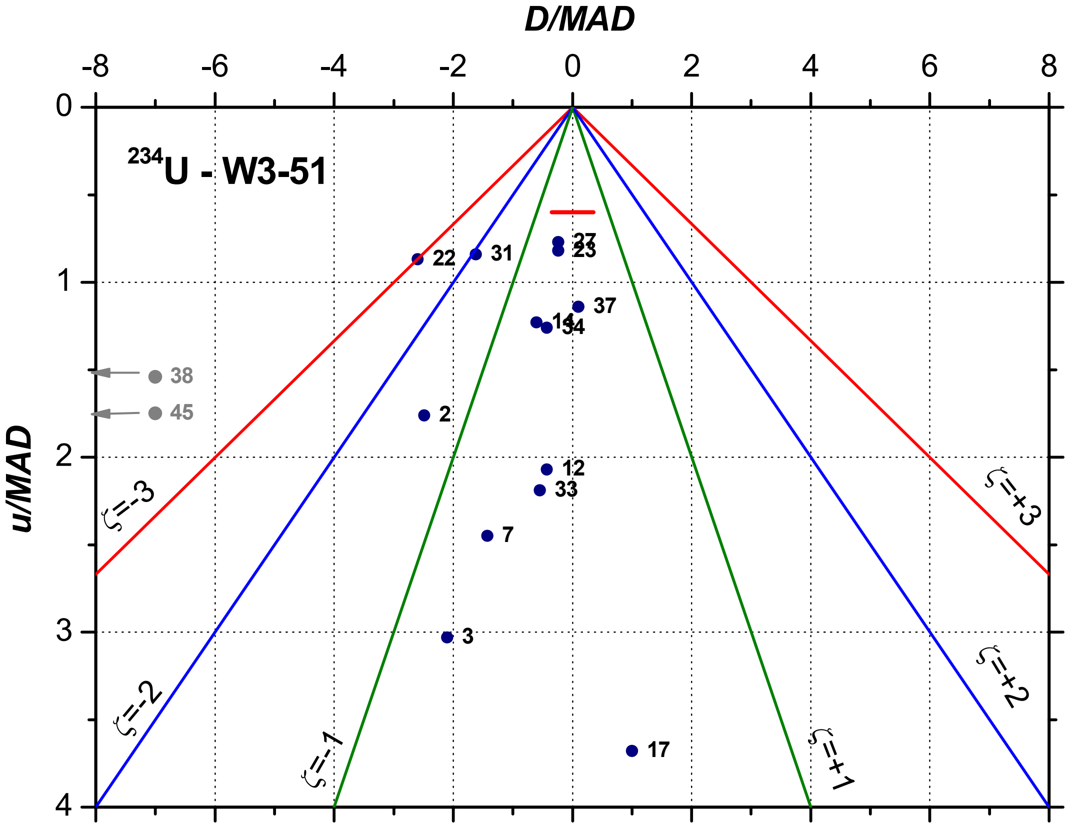

Fig. 92: PomPlot of the ^{234}U data for Water 3-51

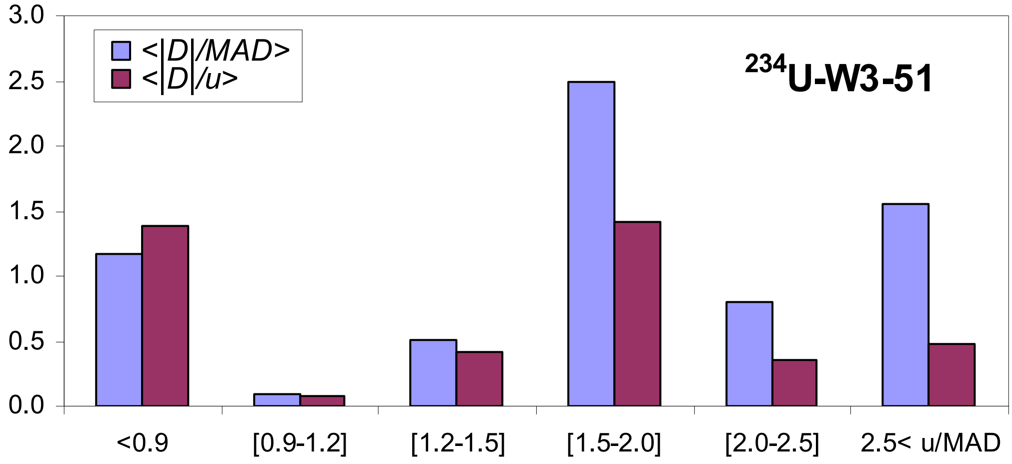

Fig. 93: Measures of average deviation <|*D*|/*MAD*>, <|*D*|/*u*> for different intervals of *u/MAD* in the case of ^{234}U in Water 3-51

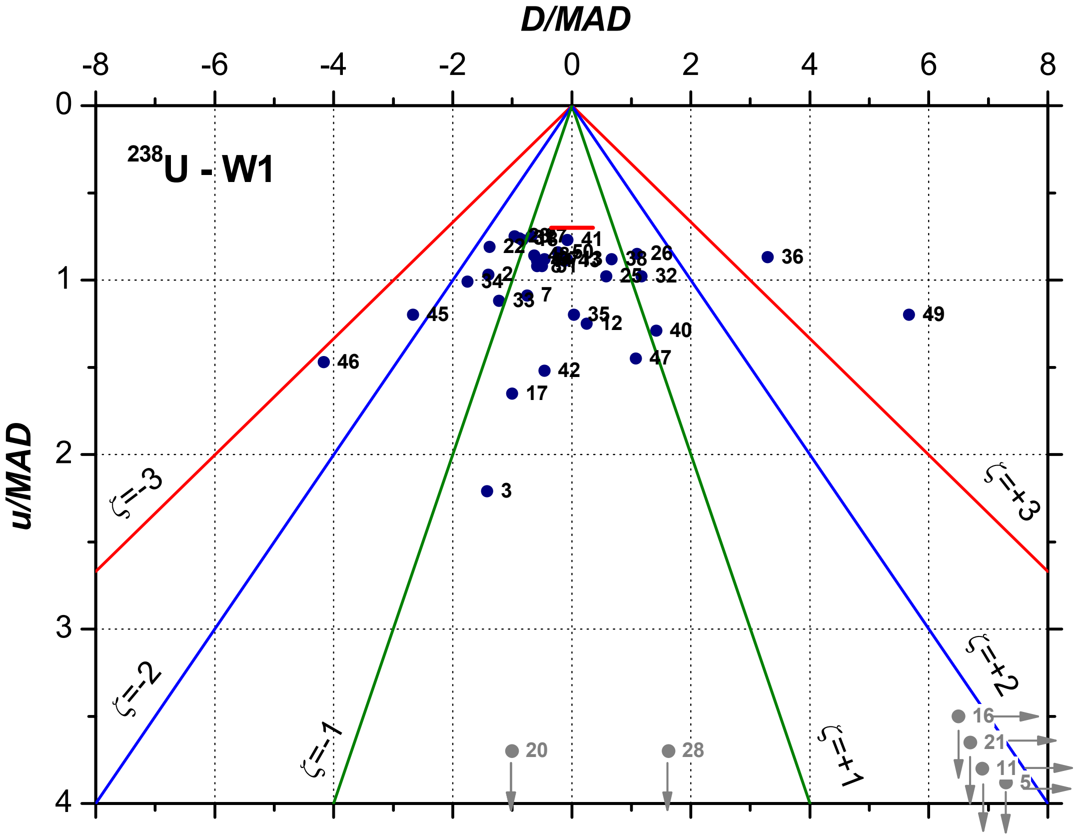

Fig. 94: PomPlot of the ^{238}U data for Water 1

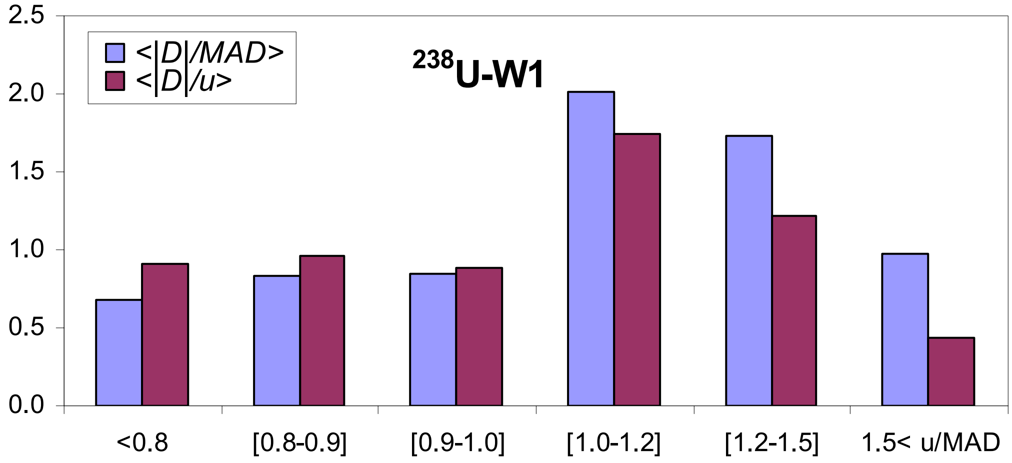

Fig. 95: Measures of average deviation <|*D*|/*MAD*>, <|*D*|/*u*> for different intervals of *u/MAD* in the case of ^{238}U in Water 1

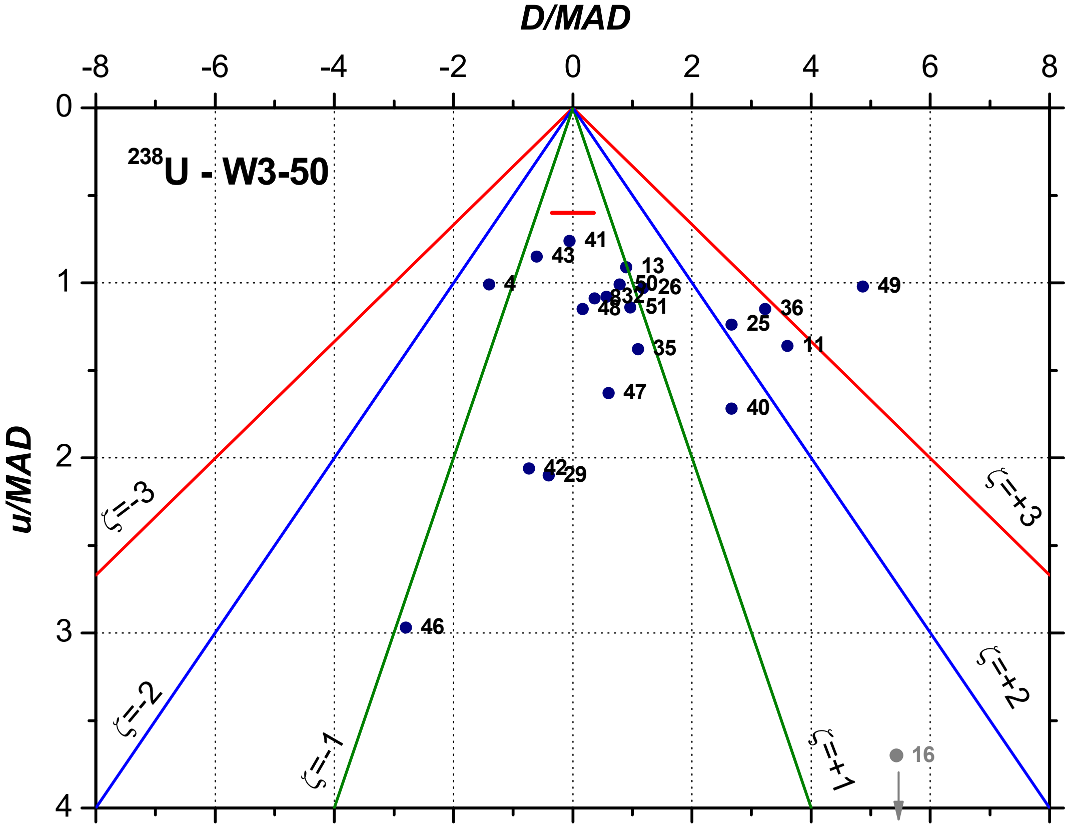

Fig. 96: PomPlot of the ^{238}U data for Water 3-50

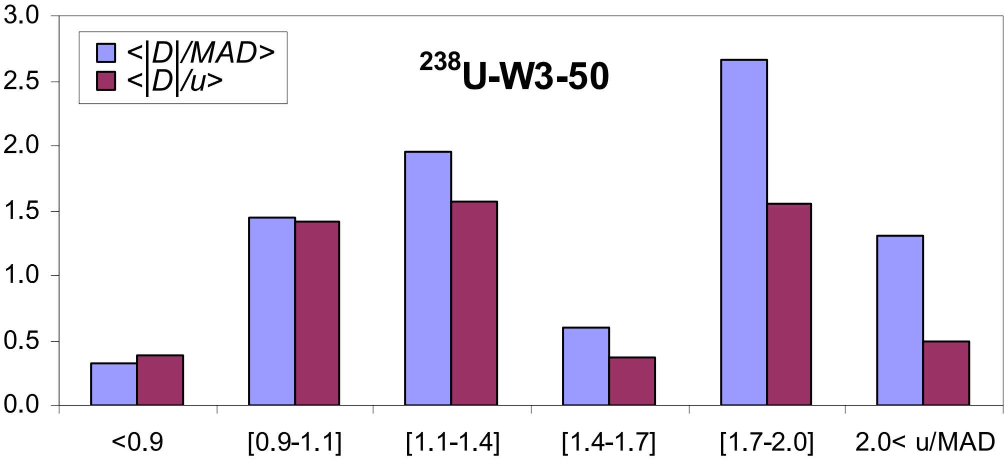

Fig. 97: Measures of average deviation <|*D*|/*MAD*>, <|*D*|/*u*> for different intervals of *u/MAD* in the case of ^{238}U in Water 3-50

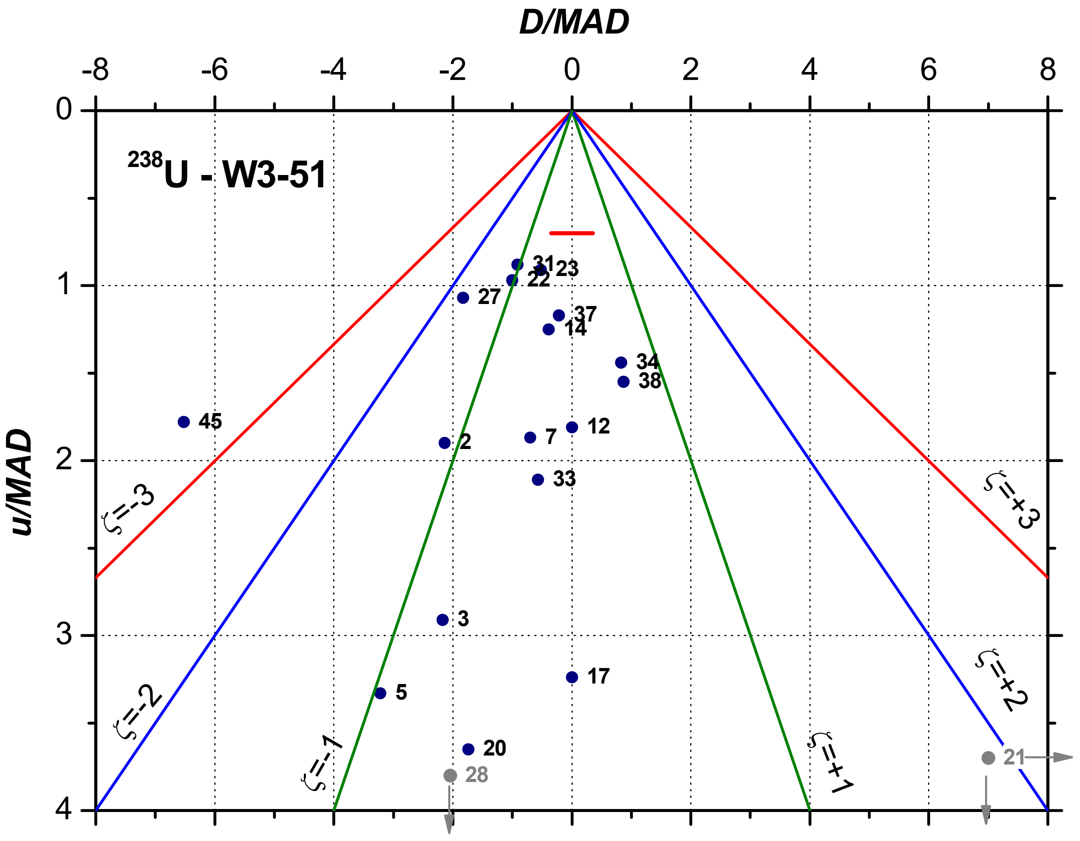

Fig. 98: PomPlot of the ^{238}U data for Water 3-51

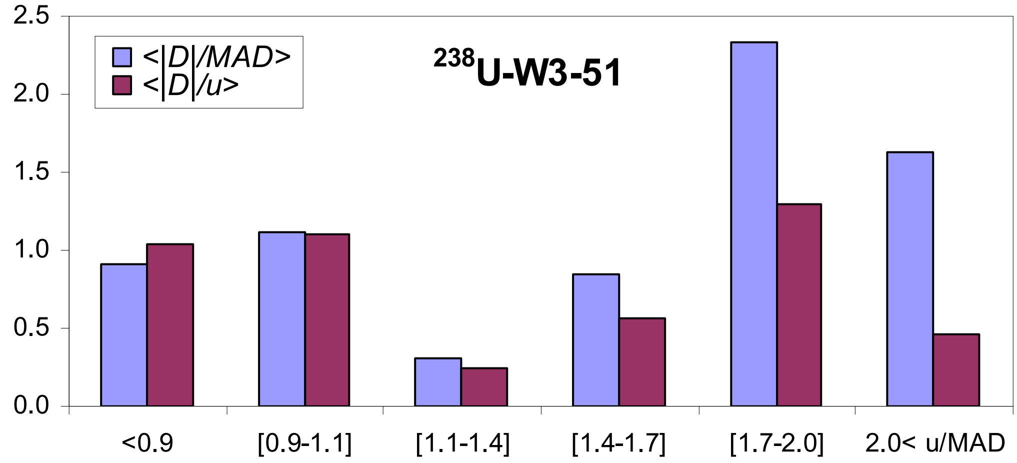

Fig. 99: Measures of average deviation <|*D*|/*MAD*>, <|*D*|/*u*> for different intervals of *u/MAD* in the case of ^{238}U in Water 3-51

7. Conclusions

Three commercially available mineral waters were provided as comparison samples. The reference values for the natural radioactivity concentration of ^{226}Ra, ^{228}Ra, ^{234}U and ^{238}U in these waters were established using independent determination methods at the IRMM and the Bundesamt für Strahlenschutz (BfS). Various radiochemical methods were applied by the 45 laboratories participating in the comparison. The comparison results show that there are many discrepant measurement results for the radium isotopes; about one fourth and more than 40 % of the laboratories have severe problems with the determination of ^{226}Ra and ^{228}Ra, respectively. For radium, 19 results, corresponding to 14 % of all, are even off by a factor of two or more. For uranium, this proportion is a bit better, but still far from satisfactory: 6 % (9 results out of 150) are off by a factor of two or more. In terms of the E_n criterion, 14 % to 23 % of the uranium results are not compliant, but as many as 25 % to 35 % of all radium results are incompatible.

As these samples had rather low activity concentrations – around the detection limits required by the draft EC directive – and not all laboratories are routinely analysing water for these radionuclides yet, unsatisfactory comparison results for ^{226}Ra and ^{228}Ra may not be unexpected. The comparison clearly demonstrates, however, that a number of monitoring laboratories need to improve their analysis procedures for radium in order to correctly identify drinking water sources for which remedial action (with respect to their natural radioactivity concentration) needs to be taken. This is a matter of ensuring radiological protection on one hand (minimising false low results) and of avoiding unjustified and unnecessary economic losses on the other hand (minimising false high results).

8. Acknowledgements

This work was possible only with the active participation of 45 laboratories in 27 countries (listed in chapter 10 below), and the support of A. Janssens and C. Gitzinger from the Directorate-General for Energy of the European Commission.

9. References

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10. List of participating laboratories (in alphabetical order, incl. nominations without actual participation)

ALBANIA

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AUSTRIA

Austrian Agency for Health and Food Safety (AGES)
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BELGIUM

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Studiecentrum Kernenergie · Centre d’Etudes Nucléaire (SCK · CEN)
Low Level Radioactivity Measurements (EHS / LRM)
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BULGARIA

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136, Tzar Boris III Bvld.
1618 Sofia
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CZECH REPUBLIC

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[1] Laboratory did not report results.

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P.O. Box 14
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GERMANY

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HUNGARY

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IRELAND

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ITALY

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Dipartimento Nucleare,Rischio Tecnologico e Industriale
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National Institute for Public Health and the Environment (RIVM)
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8224 AD Lelystad
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AGH University of Science and Technology
Faculty of Physics and Applied Computer Science
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ROMANIA

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---

[2] Laboratory did not report results due to delayed renovation works in the laboratory.

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Facultad de Veterinaria
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Universidad de Extremadura
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Elvas Avenue, s/n
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SWEDEN

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SWITZERLAND

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TURKEY

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Responsible: Abdullah Dirican

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Addlestone
Surrey KT15 3NB
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Annex 1. Questionnaire for participants sent prior to sample distribution

International Comparison Scheme for Radioactivity Environmental Monitoring ^{226}Ra, ^{228}Ra, ^{238}U and ^{234}U, radioisotopes in drinking water

QUESTIONNAIRE FOR PARTICIPANTS

I. LABORATORTY

1. Contact details

Name of the contact person:	
Position:	
Name of organisation/laboratory:	
Complete address (for courrier service delivery such as DHL):	
Country:	
Tel:	
E-mail:	

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

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

3. Is your laboratory certified, accredited or authorised for this type of analysis?

	YES	NO
Certified		
Accredited		
Authorised		

4. Is your laboratory working according to a quality management system?

YES NO

If YES, please state which system:

EN 45000 series / ISO 25	
ISO 9000 series	
ISO 17025	
OTHER (please specify)	

II. EXPERIENCE

5. How many measurements of this type does your laboratory perform per year?

	< 25	25 - 100	>100
Ra-226			
Ra-228			
U-234			
U-238			

6. Typical activity concentrations measured:

	Bq/L
Ra-226	
Ra-228	
U-234	
U-238	

III. SAMPLE PREPARATION

7. Brief description of sample preparation:

for Ra-226 determination:	
for Ra-228 determination:	
for U-234 determination:	
for U-238 determination:	

8. Total water volume needed for duplicate analysis of all radionuclides listed above in L **per water** sample? (indicate approximate volumes by X)

	1.5 L	3 L	4.5 L	6 L
Ra-226				
Ra-228				
U-234				
U-238				

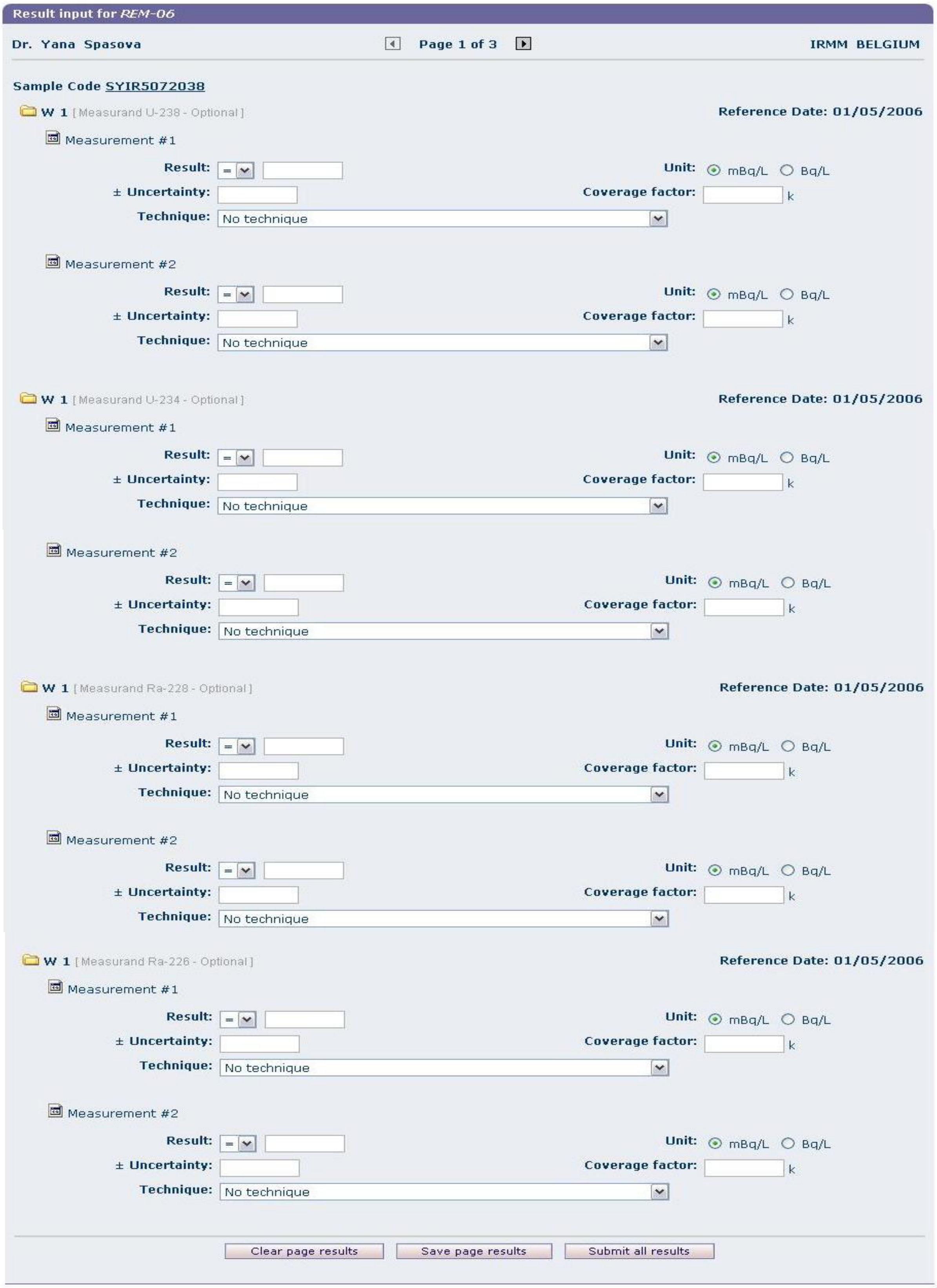
IV. MEASUREMENT

9. Brief description of measurement method:

Ra-226	
Ra-228	
U-234	
U-238	

10. Further comments:

Annex 2: Result reporting form



Result input for *REM-06*

Dr. Yana Spasova Page 1 of 3 **IRMM BELGIUM**

Sample Code **SYIR5072038**

W 1 [Measurand U-238 - Optional] **Reference Date: 01/05/2006**

Measurement #1

Result: = **Unit:** ● mBq/L ○ Bq/L

± Uncertainty: **Coverage factor:** k

Technique: No technique

Measurement #2

Result: = **Unit:** ● mBq/L ○ Bq/L

± Uncertainty: **Coverage factor:** k

Technique: No technique

W 1 [Measurand U-234 - Optional] **Reference Date: 01/05/2006**

Measurement #1

Result: = **Unit:** ● mBq/L ○ Bq/L

± Uncertainty: **Coverage factor:** k

Technique: No technique

Measurement #2

Result: = **Unit:** ● mBq/L ○ Bq/L

± Uncertainty: **Coverage factor:** k

Technique: No technique

W 1 [Measurand Ra-228 - Optional] **Reference Date: 01/05/2006**

Measurement #1

Result: = **Unit:** ● mBq/L ○ Bq/L

± Uncertainty: **Coverage factor:** k

Technique: No technique

Measurement #2

Result: = **Unit:** ● mBq/L ○ Bq/L

± Uncertainty: **Coverage factor:** k

Technique: No technique

W 1 [Measurand Ra-226 - Optional] **Reference Date: 01/05/2006**

Measurement #1

Result: = **Unit:** ● mBq/L ○ Bq/L

± Uncertainty: **Coverage factor:** k

Technique: No technique

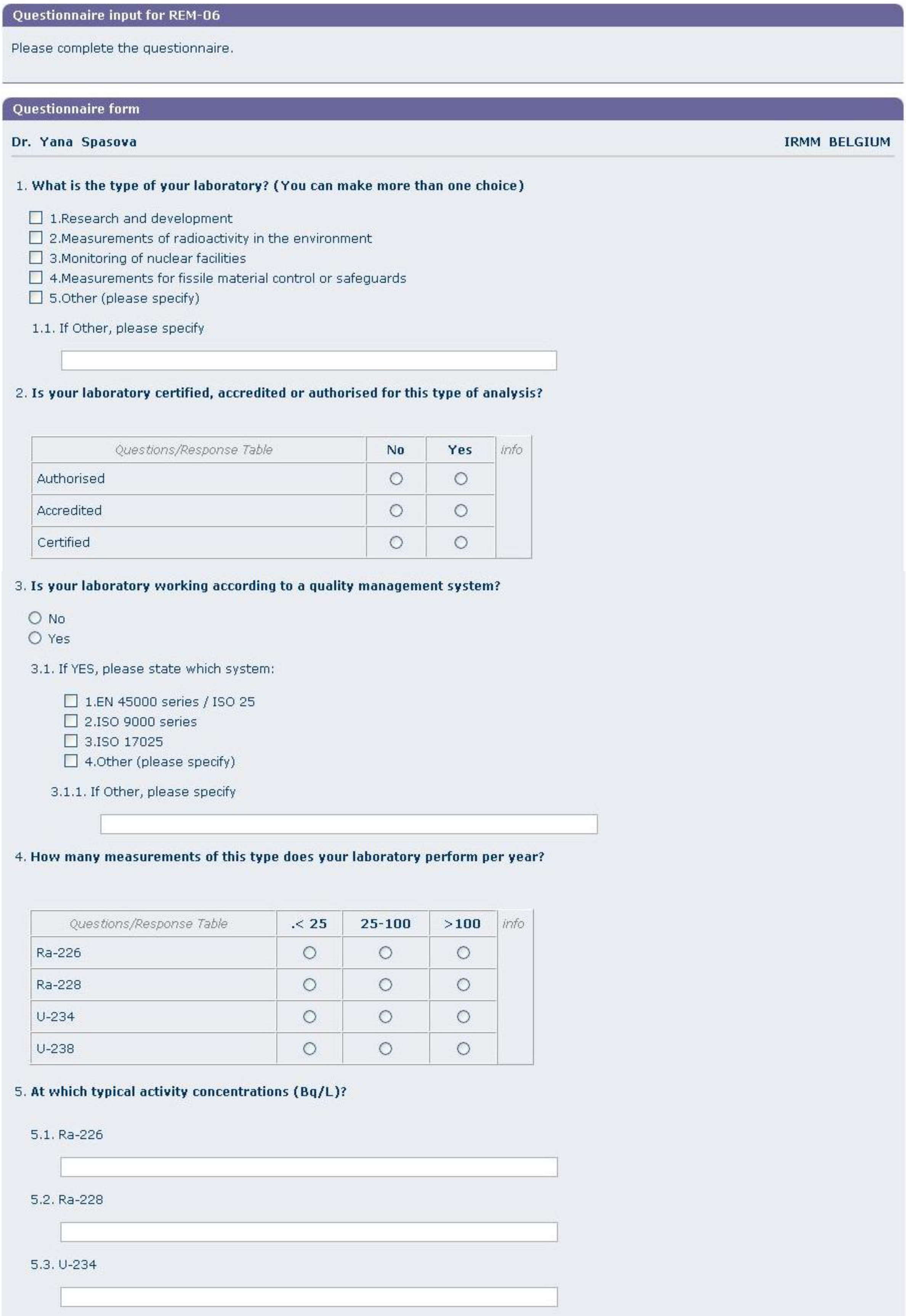
Measurement #2

Result: = **Unit:** ● mBq/L ○ Bq/L

± Uncertainty: **Coverage factor:** k

Technique: No technique

Annex 3: Questionnaire



Questionnaire input for REM-06

Please complete the questionnaire.

Questionnaire form

Dr. Yana Spasova **IRMM BELGIUM**

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

☐ 1.Research and development
☐ 2.Measurements of radioactivity in the environment
☐ 3.Monitoring of nuclear facilities
☐ 4.Measurements for fissile material control or safeguards
☐ 5.Other (please specify)

1.1. If Other, please specify

2. **Is your laboratory certified, accredited or authorised for this type of analysis?**

Questions/Response Table	**No**	**Yes**	*info*
Authorised	○	○	
Accredited	○	○	
Certified	○	○	

3. **Is your laboratory working according to a quality management system?**

○ No
○ Yes

3.1. If YES, please state which system:

☐ 1.EN 45000 series / ISO 25
☐ 2.ISO 9000 series
☐ 3.ISO 17025
☐ 4.Other (please specify)

3.1.1. If Other, please specify

4. **How many measurements of this type does your laboratory perform per year?**

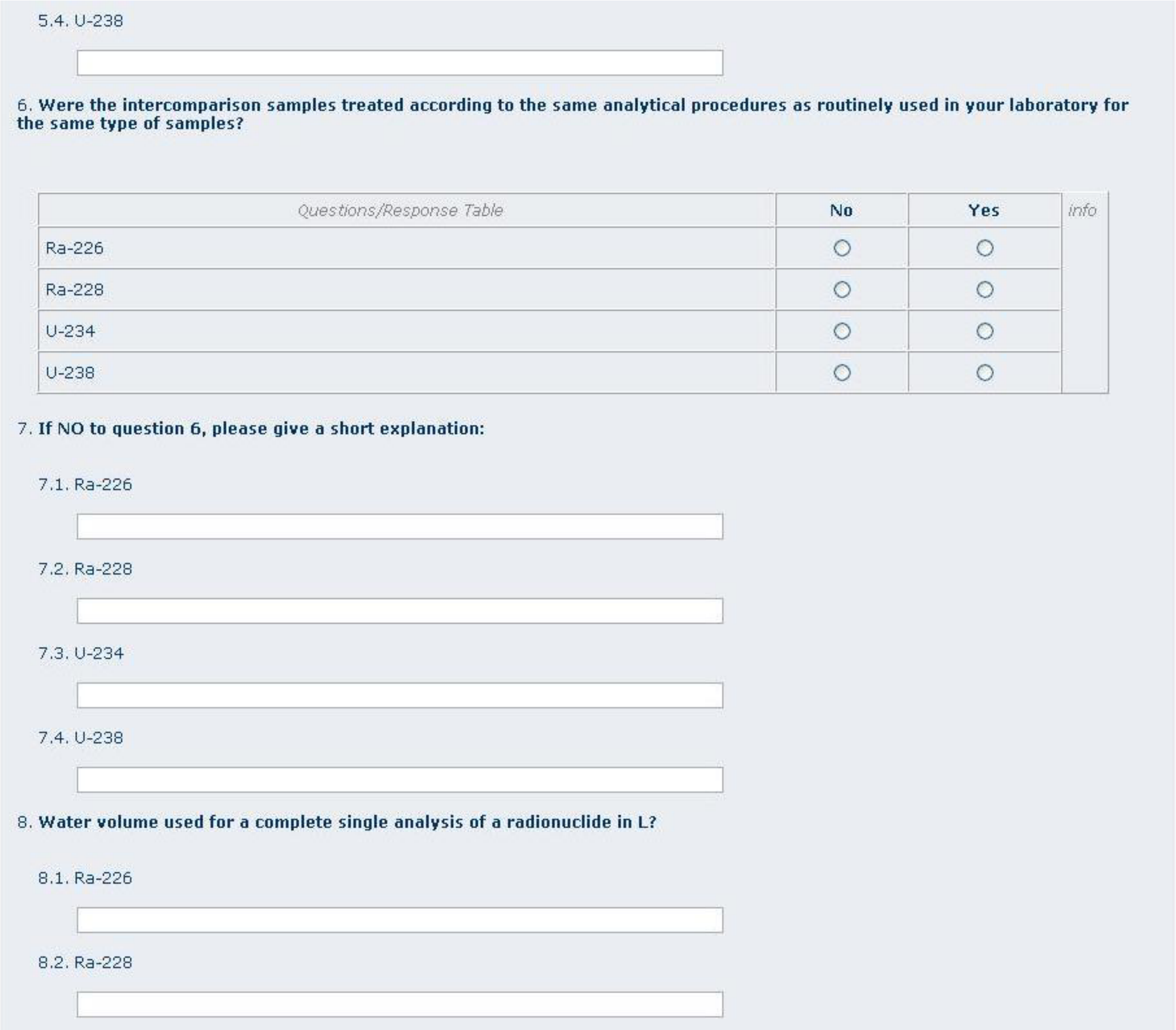
Questions/Response Table	**.< 25**	**25-100**	**>100**	*info*
Ra-226	○	○	○	
Ra-228	○	○	○	
U-234	○	○	○	
U-238	○	○	○	

5. **At which typical activity concentrations (Bq/L)?**

5.1. Ra-226

5.2. Ra-228

5.3. U-234



5.4. U-238

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

Questions/Response Table	**No**	**Yes**	*info*
Ra-226	○	○	
Ra-228	○	○	
U-234	○	○	
U-238	○	○	

7. **If NO to question 6, please give a short explanation:**

7.1. Ra-226

7.2. Ra-228

7.3. U-234

7.4. U-238

8. **Water volume used for a complete single analysis of a radionuclide in L?**

8.1. Ra-226

8.2. Ra-228

8.3. U-234

8.4. U-238

9. **Brief description of sample preparation (please, send a detailed description as an e-mail attachment to Yana.SPASOVA@ec.europa.eu).**

9.1. Ra-226

9.2. Ra-228

9.3. U-234

9.4. U-238

10. **Chemical recovery method used.**

10.1. Ra-226

10.2. Ra-228

10.3. U-234

10.4. U-238

11. **Average chemical recovery obtained (%).**

11.1. Ra-226

11.2. Ra-228

11.3. U-234

11.4. U-238

12. **Brief description of measurement method (please, send a detailed description as e-mail attachment to Yana.SPASOVA@ec.europa.eu).**

12.1. Ra-226

12.2. Ra-228

12.3. U-234

12.4. U-238

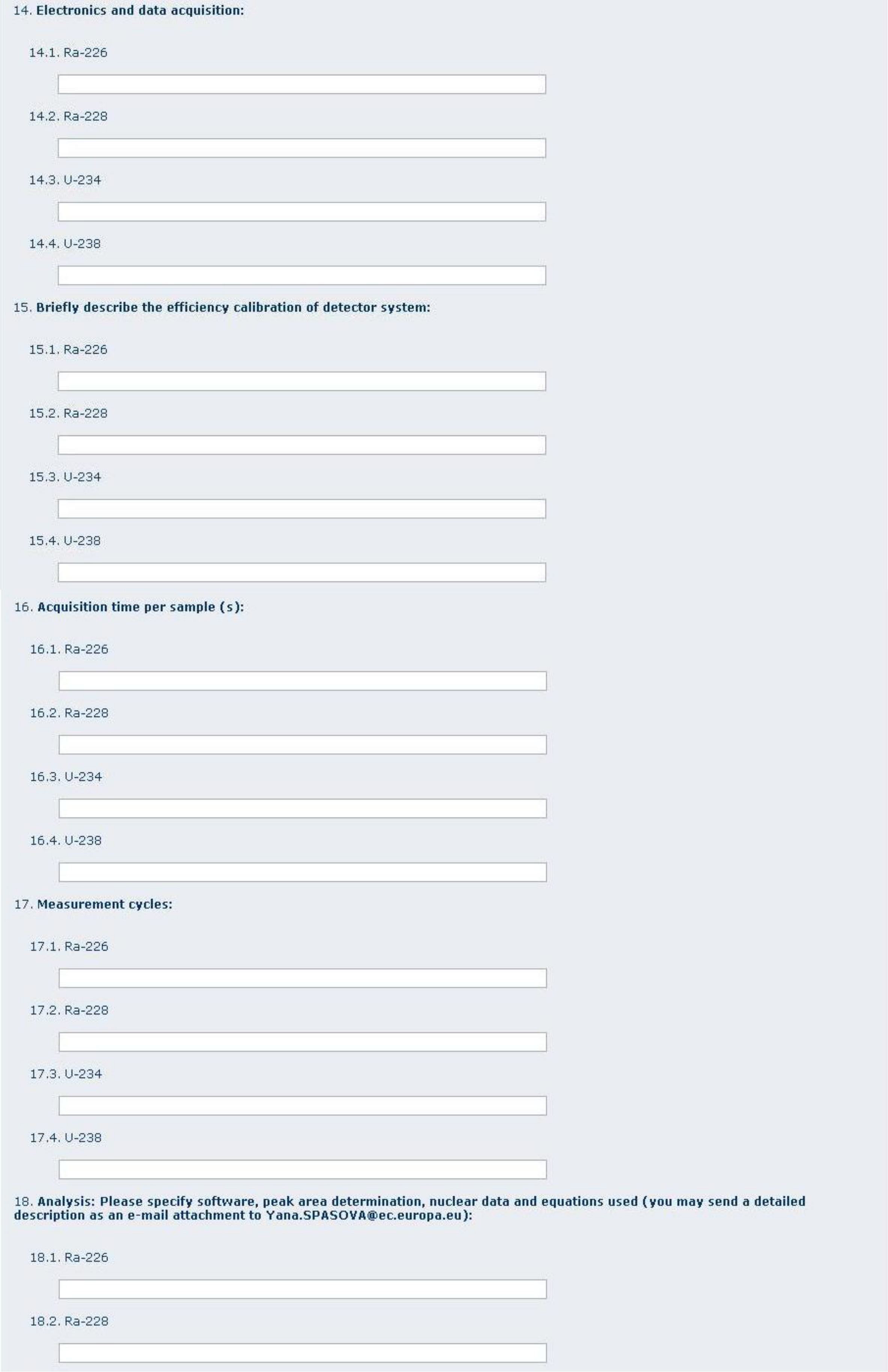
13. **Type of detector:**

13.1. Ra-226

13.2. Ra-228

13.3. U-234

13.4. U-238



14. **Electronics and data acquisition:**

14.1. Ra-226

14.2. Ra-228

14.3. U-234

14.4. U-238

15. **Briefly describe the efficiency calibration of detector system:**

15.1. Ra-226

15.2. Ra-228

15.3. U-234

15.4. U-238

16. **Acquisition time per sample (s):**

16.1. Ra-226

16.2. Ra-228

16.3. U-234

16.4. U-238

17. **Measurement cycles:**

17.1. Ra-226

17.2. Ra-228

17.3. U-234

17.4. U-238

18. **Analysis: Please specify software, peak area determination, nuclear data and equations used (you may send a detailed description as an e-mail attachment to Yana.SPASOVA@ec.europa.eu):**

18.1. Ra-226

18.2. Ra-228

18.3. U-234

18.4. U-238

19. **Uncertainty budget: Ra-226. Feel free to send the complete uncertainty budget as an e-mail attachment to Yana.SPASOVA@ec.europa.eu. If a contribution is not applicable enter 0 (zero).**

19.1. Please, state for one measurement result and indicate:

19.2. Sample (W1, W2 or W3): #

19.3. Measurement number: #

19.4. Number of measurements performed: #

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

19.6. Relative uncertainty on the tracer activity (%):

19.7. Relative counting uncertainty (%):

19.8. Relative uncertainty in efficiency (%):

19.9. Relative uncertainty due to activity efficiency standards (%):

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

19.11. Possibly other uncertainty contributions (%):

19.12. Relative combined standard uncertainty u*C (%):

19.13. Combined standard uncertainty uC (mBq/L):

19.14. Expanded uncertainty U (mBq/L, k=2):

20. **Uncertainty budget: Ra-228. Feel free to send the complete uncertainty budget as an e-mail attachment to Yana.SPASOVA@ec.europa.eu. If a contribution is not applicable enter 0 (zero).**

20.1. Please, state for one measurement result and indicate:

20.2. Sample (W1, W2 or W3): #

20.3. Measurement number: #

20.4. Number of measurements performed: #

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

20.6. Relative uncertainty on the tracer activity (%):

20.7. Relative counting uncertainty (%):

20.8. Relative uncertainty in efficiency (%):

20.9. Relative uncertainty due to activity efficiency standards (%):

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

20.11. Possibly other uncertainty contributions (%):

20.12. Relative combined standard uncertainty u*C (%):

20.13. Combined standard uncertainty uC (mBq/L):

20.14. Expanded uncertainty U (mBq/L, k=2):

21. **Uncertainty budget: U-234. Feel free to send the complete uncertainty budget as an e-mail attachment to Yana.SPASOVA@ec.europa.eu. If a contribution is not applicable enter 0 (zero).**

21.1. Please, state for one measurement result and indicate:

21.2. Sample (W1, W2 or W3): #

21.3. Measurement number: #

21.4. Number of measurements performed: #

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

21.6. Relative uncertainty on the tracer activity (%):

21.7. Relative counting uncertainty (%):

21.8. Relative uncertainty in efficiency (%):

21.9. Relative uncertainty due to activity efficiency standards (%):

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

21.11. Possibly other uncertainty contributions (%):

21.12. Relative combined standard uncertainty u*C (%):

21.13. Combined standard uncertainty uC (mBq/L):

21.14. Expanded uncertainty U (mBq/L, k=2):

22. **Uncertainty budget: U-238. Feel free to send the complete uncertainty budget as an e-mail attachment to Yana.SPASOVA@ec.europa.eu. If a contribution is not applicable enter 0.**

22.1. Please, state for one measurement result and indicate:

22.2. Sample (W1, W2 or W3): #

22.3. Measurement number: #

22.4. Number of measurements performed: #

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

22.6. Relative uncertainty on the tracer activity (%):

22.7. Relative counting uncertainty (%):

22.8. Relative uncertainty in efficiency (%):

22.9. Relative uncertainty due to activity efficiency standards (%):

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

22.11. Possibly other uncertainty contributions (%):

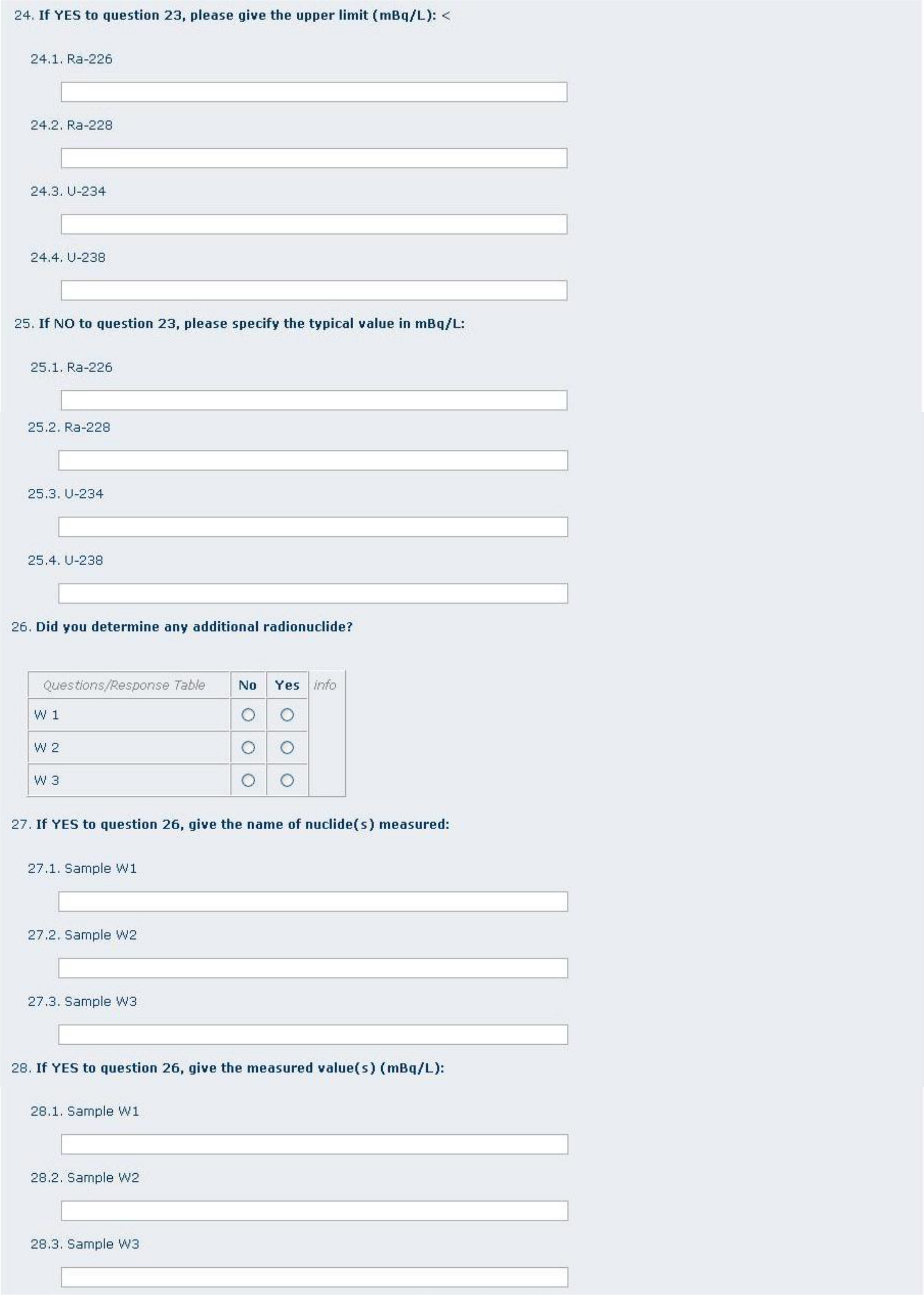
22.12. Relative combined standard uncertainty u*C (%):

22.13. Combined standard uncertainty uC (mBq/L):

22.14. Expanded uncertainty U (mBq/L, k=2):

23. **Routine measurements: Is the typical activity in drinking water below the detection limits/decision thresholds?**

Questions/Response Table	**No**	**Yes**	*info*
Ra-226	○	○	
Ra-228	○	○	
U-234	○	○	
U-238	○	○	



24. If YES to question 23, please give the upper limit (mBq/L): <

24.1. Ra-226

24.2. Ra-228

24.3. U-234

24.4. U-238

25. If NO to question 23, please specify the typical value in mBq/L:

25.1. Ra-226

25.2. Ra-228

25.3. U-234

25.4. U-238

26. Did you determine any additional radionuclide?

Questions/Response Table	**No**	**Yes**	*info*
W 1	○	○	
W 2	○	○	
W 3	○	○	

27. If YES to question 26, give the name of nuclide(s) measured:

27.1. Sample W1

27.2. Sample W2

27.3. Sample W3

28. If YES to question 26, give the measured value(s) (mBq/L):

28.1. Sample W1

28.2. Sample W2

28.3. Sample W3

29. **If YES to question 26, give the expanded uncertainty(ies) U (mBq/L, k=2):**

29.1. Sample W1

29.2. Sample W2

29.3. Sample W3

30. **Difficulties encountered.**

31. **Other comments on this intercomparison exercise.**

32. **Questionnaire completed by:**

32.1. Name

32.2. Position

European Commission

EUR 24694 EN – Joint Research Centre – Institute for Reference Materials and Measurements
Title: Evaluation of EC comparison on the determination of ^{226}Ra, ^{228}Ra, ^{234}U and ^{238}U in mineral waters
Authors: Y. Spasova, U. Wätjen, L. Benedik, M. Vasile, T. Altzitzoglou and M. Beyermann[2]
Luxembourg: Publications Office of the European Union
2011 – 149 pp. – 21.0 x 29.7 cm
EUR – Scientific and Technical Research series – ISSN 1018-5593
ISBN 978-92-79-19069-8
DOI 10.2787/36764

Abstract

This report describes all details of the comparison for the determination of ^{226}Ra, ^{228}Ra, ^{234}U and ^{238}U in mineral waters among 45 European laboratories monitoring radioactivity in food and the environment. Three commercially available mineral waters were provided as comparison samples. Reference values of the four radionuclides under study in this comparison were determined in collaborative work of IRMM and the Bundesamt für Strahlenschutz (BfS), using tracer techniques and standardised radionuclide solutions. The reference values are thus traceable to the SI units. The sample preparation and measurement processes applied in the participating laboratories are described and the results of the comparison are presented and discussed in detail. Whereas, in general, the measurement results for the uranium isotopes show a relatively favourable agreement with the reference value, the results of this comparison point at severe problems of ^{226}Ra and ^{228}Ra determination in about one fourth and more than one third of the laboratories, respectively. For radium, 19 results corresponding to 14 % of all are even off by a factor of two or more. By comparison, for uranium, this number amounts to 6 % (9 results out of 150). Nevertheless, also for the determination of uranium, 14 % to 23 % of the laboratories report results not compliant with the E_n evaluation criterion. The corresponding participants are urgently requested to investigate and revise their analytical methods.

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[2] Bundesamt für Strahlenschutz (BfS), Köpenicker Allee 120-130, D-10318 Berlin, Germany

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EUROPEAN COMMISSION

LA-NA-24694-EN-C

Publications Office

ISBN 978-92-79-19069-8

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