European Commission

JRC SCIENCE FOR POLICY REPORT

Evaluation of the 2012 EC interlaboratory comparison on gross alpha/beta activity concentration in drinking water

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2016

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EUR 28351 EN

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JRC Science Hub
https://ec.europa.eu/jrc

JRC104692

EUR 28351 EN

PDF ISBN 978-92-79-64622-5 ISSN 1831-9424 doi: 10.2789/573272

Luxembourg: Publications Office of the European Union, 2016

How to cite this report: V. Jobbágy, J. Merešová, T. Altzitzoglou, M. Hult, P. Malo, U. Wätjen; *Evaluation of the 2012 EC interlaboratory comparison on gross alpha/beta activity concentration in drinking water*; EUR 28351 EN; 10.2789/573272

Evaluation of the 2012 EC interlaboratory comparison on gross alpha/beta activity concentration in drinking water

An interlaboratory comparison was organized among 71 environmental radioactivity monitoring laboratories for the determination of gross alpha/beta activity concentration in drinking water. The performance of participating laboratories was evaluated with respect to the reference values using relative deviations. Sample preparation and measurement methods used by the participating laboratories are detailed, in particular in view of method-dependency of the results. Many of the participants' results deviate by more than two orders of magnitude from the reference values. This clearly demonstrates gross methods as unreliable and inaccurate in its present form and suggests revising the written standards for gross methods and restricting their application under clearly defined rules. Repeating this interlaboratory comparison is considered.

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Acknowledgements[1]

Thanks are due to all the 71 participant laboratories because this work was possible only with their active participation (listed in Appendix 6)

The authors wish to thank Pieter Kwakman (RIVM, The Netherlands), Edmond Dupuis (SCK•CEN, Belgium) and Andrej Rožkov (EC-JRC-Geel, Belgium) for their contributions to the reference value determination.

Thanks are due to Radionuclide Metrology Sector at Nuclear Physics Unit (JRC-Geel). The following people at the Standards for Innovation and Sustainable Development Unit (EC-JRC-Geel) helped in preparing, mixing and filling the samples: Håkan Emteborg, Albert Oostra, Hanne Leys, Diana Vernelen and Irma Huybrechts.

Thanks are also due to Coca-Cola Belgium for providing one of the test materials for free of charge and the central storage staff at JRC-Geel for assisting in the smooth sample distribution.

([1]) Since the start of this exercise the following changes have taken place:
- JRC-IRMM became JRC-Geel.
- The Nuclear Physics Unit became the unit for Standards for Nuclear Safety, Security and Safeguards.
- The Reference Materials Unit became the unit for Standards for Innovation and Sustainable Development.

Executive summary

Policy context

On the basis of the EURATOM treaty (Article 35-36), monitoring and reporting of environmental radioactivity is one of the EU member states' obligations. To check quality and comparability of these measurement results the JRC-Geel was requested by the European Commission's Directorate-General for Energy to organize interlaboratory comparisons (ILCs). Anticipating and supporting the new EURATOM Drinking Water Directive (EC, 2013) which includes gross alpha/beta activity screening levels, JRC-Geel organised an interlaboratory comparison (ILC) to check the fitness for purpose of this method and the performance of European monitoring laboratories in 2012.

JRC-Geel is responsible for the coordination of ILCs in environmental radioactivity measurements since 2003. ILCs have been already organized on radioactivity measurements in different matrixes: air filter, soil, organic material and water. In anticipation of new European requirements for monitoring radioactivity concentration in drinking water, JRC-Geel organized an interlaboratory comparison on one of the most widespread radioanalytical monitoring methods – gross alpha/beta activity measurement in drinking water samples. This report was prepared for European Commission's Directorate-General for Energy, MS national radiation protection authorities, but it may be useful also for European Commission's Directorate-General for Health and Food Safety.

The report describes in detail all phases of the intercomparison exercise from the materials selection until the participants' performance evaluation. The participants, 71 European laboratories monitoring radioactivity in the environment and foodstuff, are proficient laboratories in determination of natural radionuclides in mineral waters. For the ILC exercise one spiked water sample and two commercially available mineral waters were used as test items since their activity concentration of natural radionuclides is usually higher than in most tap waters. Reference values were established in collaboration of the JRC-Geel, the Belgian Nuclear Research Centre (SCK-CEN) and the Dutch National Institute for Public Health and the Environment (RIVM). The homogeneity and short term stability of the batch of distributed samples were checked.

Key conclusions

The performance evaluation of individual laboratories was done on the basis of reported results and answers given to a questionnaire. The evaluation of the results was based on their relative deviations from the reference values. It shows that there are many highly discrepant measurement results for the gross alpha/beta activity concentrations. More than half of the laboratories have severe problems with the determination of gross alpha/beta activity concentration in drinking waters. Only one laboratory was able to determine gross alpha/beta activities within the reference range ($\pm$ 30% from the reference values) for all three waters and only 30 laboratories (42 % of the participants) could report at least half of the results within the reference range. It has to be noted that there were 8 laboratories (11 %) that were not able to report measurement data within the reference range at all. However, unsatisfactory comparison results for gross activities may have been expected for some reasons. For example, the initial radionuclide composition of the samples was not known beforehand the analysis, and one of these samples had rather low gross alpha activity concentration, below the detection limits required by the draft EC directive.

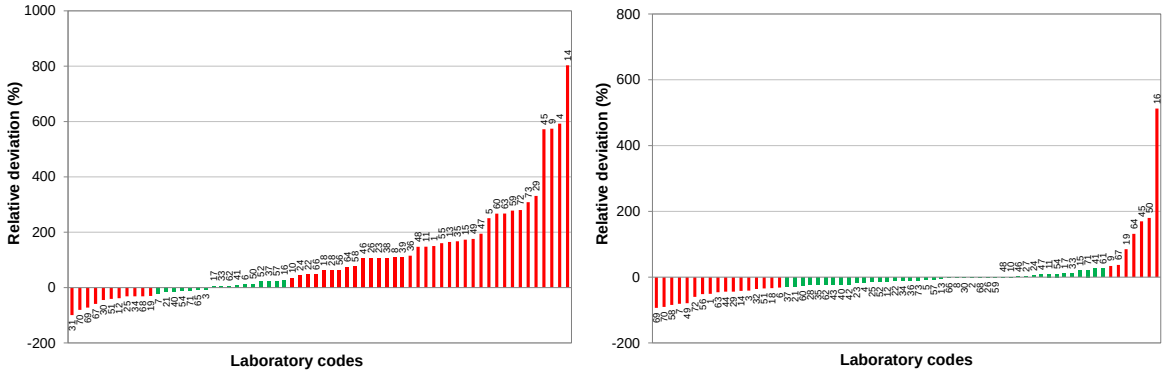


Fig. I. Deviation chart of the participants' results of gross beta activity concentration for Water B (left) and gross alpha activity concentration for Water C (right) plotted in ascending order. Green colour indicates results within the range ± 30 % from the reference value and red indicates results outside this range. Numbers indicate the laboratory code.

Many of the participants' results deviate by more than two orders of magnitude from the reference values regardless of the techniques used. The ratio of maximum to minimum reported gross activities and the percentage of compatible results are presented in Table 1 and 2 respectively. The number of compatible results together with the number of laboratories and their identification codes are presented in Table III.

Table I. Ratio of the reported maximum to minimum gross activities.

Parameter	**A_{max}/ A_{min}**		
	Water A	**Water B**	**Water C**
Gross alpha activity	1017	346	93
Gross beta activity	3050	2080	3150

Table II. Percentage of the reported results within ± 30 % from the reference value.

Parameter	**Results within ± 30 % deviation (%)**		
	Water A	**Water B**	**Water C**
Gross alpha activity	36	39	63
Gross beta activity	45	27	61

Table III. Number of laboratories and their ILC identification codes versus the number of reported compatible results.

Number of compatible results	Number of laboratories	Laboratory code
6	1	33
5	7	17, 21, 41, 54, 57, 62, 71
4	11	2, 18, 22, 30, 34, 35, 36, 37, 40, 48, 51
3	11	1, 5, 6, 8, 11, 13, 24, 25, 26, 52, 68
2	20	3, 7, 10, 15, 16, 19, 23, 27, 28, 39, 43, 46, 47, 49, 59, 60, 63, 64, 66, 73
1	13	4, 9, 12, 14, 29, 32, 42, 50, 55, 61, 65, 67, 72
0	8	31, 38, 44, 45, 56, 58, 69, 70

Main findings

After evaluating the results of the ILC, one can draw as a conclusion that the existing analytical procedures/standards need to be critically revised for gross alpha/beta measurement in order to obtain reliable and comparable measurement results. This is needed in regular monitoring to correctly identify the source of radioactivity in drinking waters and eventually decide if remedial action with respect to the natural radioactivity concentration needs to be taken. These findings have to be reported to the concerned laboratories, authorities and national representatives to be aware of the outcome of such a highly participated ILC. The possible interferences and corrective actions need to be addressed and discussed in different fora (conferences, standardization bodies and policy makers). Considering the high number of individual gross measurements, the ability of those laboratories to provide consistent and reliable results will directly influence the implementation of the drinking water directive. Furthermore, the decisions made on the basis of the measurement results have health and economic impacts as well.

There are numerous pitfalls and sources of interferences of the gross alpha/beta methods as discussed in a journal article by Jobbágy et al. (2014). Some of the influential parameters are listed here: sample preparation methods (possible loss of volatile radionuclides), time delay between sample preparation and measurement (ingrowth of radon and its progenies), detection technique, calibration source energy.

Because of the many variables playing a key role in gross measurement, it is important to fix as many parameters as possible via a true standardization of the analysis methods in use.

We propose some recommendations for the gross alpha/beta method applied to drinking water analysis:

(1) Gross measurement should be used for monitoring samples with known radionuclide composition is known (from radionuclide specific analysis of representative samples).

(2) Gross measurements could be considered as a complementary or substitute method for radionuclide-specific measurement only with important restrictions:

   (a) no temporary change is expected in the radiochemical composition of the monitored water,

   (b) no complex decay chains are present,

   (c) a true standardized method is used where the measurement parameters are fixed.

Radionuclide specific analysis should be repeated on a regular basis in accordance with the drinking water directive concerning check and audit monitoring (EC, 1998).

Related and future JRC work

The gross alpha/beta activity in drinking water ILC will be repeated as agreed among DG-ENER, the Euratom article 35-36 national representatives and JRC-Geel.

1 Introduction

Within the framework of the European Atomic Energy Community (Euratom) Treaty and derived European legislation, member states (MS) of the European Union are obliged to perform measurements of the radioactivity levels in their environment and to report the results to the European Commission (EC). In order to verify the performance of the monitoring laboratories and to ensure comparability of reported results, regular interlaboratory comparisons (ILCs) were introduced by the EC. Since 2003, the JRC Institute for Reference Materials and Measurements (JRC-GEEL) has had the responsibility for their organization. Since 1st July 2016, JRC-JRC-GEEL has changed name to JRC-Geel, which will be used throughout this report as it is launched after the name change although the work presented was performed before.

The metrological approach of JRC-Geel in conducting comparisons relies on its participation in key comparisons among national metrology institutes (Wätjen et al., 2008) as described in **Fig. 1.** This allows JRC-GEEL to work with intercomparison samples for which it determines reference values that are traceable to SI units and the International Reference System (SIR) for gamma-ray emitting radionuclides (Ratel, 2007). In terms of physical properties as well as radioactivity concentration levels, JRC-GEEL comparison samples are generally closer to the real samples measured in monitoring laboratories than calibration standards and, therefore, they give a realistic estimate of the performance of these laboratories in their monitoring tasks.

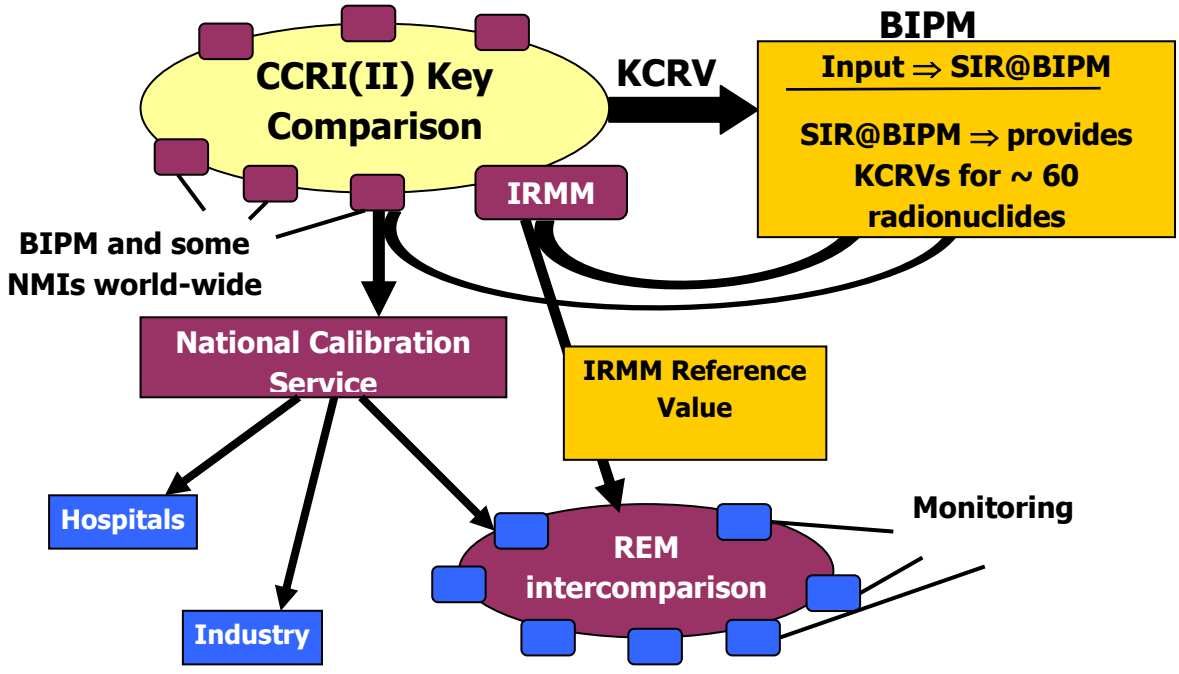


Fig. 1. Key comparisons of CCRI(II) and traceability of the reference values for samples provided by JRC-GEEL for the intercomparisons amongst monitoring laboratories (KCRV = key comparison reference value).

The aim of this ILC was to verify the performance of EU MS monitoring laboratories for the determination of gross alpha and beta activity in drinking waters. Two natural and one artificial spiked water samples were used as a test item.

This report describes in detail all phases of the ILC organized in 2012, from the description of the analytical methods used at the laboratories, the treatment of the data reported by the participants and, finally, the evaluation and comparison of the participants' results with the reference values. An evaluation of the performance of individual laboratories is performed using relative deviations, (ISO 13528, 2005).

The individual results of the participants were distributed shortly after the ILC in 2013. The results have also been presented in scientific articles (Jobbágy et al., 2014; Jobbágy et al., 2015) and discussed at the Article 35/36 Expert group meetings in Geel 2013 and 2016. This comprehensive report serves to provide the complete story of the exercise including the details of the preparation and certification of the reference materials used for this ILC. The report thus forms the basis for possible action by DG ENER in this field.

1.1 Reporting of the results

All results of activity concentrations were reported normalized to volume ($mBq \cdot L^{-1}$) with the associated expanded uncertainty U ($U = k \cdot u_c$, where U is determined from the combined standard uncertainty u_c with a coverage factor k = 2, corresponding to a level of confidence of about 95 %).

The deadline for results reporting was 31 October 2012.

The reporting of the results was realised via an online reporting system (MILC) which served also as a questionnaire. Participants were asked to answer all relevant questions regarding the used measurement procedures. Information given in the questionnaire is essential in order to evaluate the results of the intercomparison. Moreover, it allows us to find out the sources of possible pitfalls and to get an overview of the methods used among the laboratories.

Timetable of ILC:

17 July 2012	Invitation letter sent to the national representatives (**Appendix 1**)
15 August 2012	Laboratories are nominated by the national representatives
15 September 2012	Registration deadline for the nominated laboratories
September 2012	Water samples are sent to the participants via express mail (DHL) together with the information on the ILC (**Appendix 3**)
31 October 2012	Laboratories submit their results and questionnaire to JRC-GEEL
January 2013	Preliminary results sent to participants on Water-C sample (**Appendix 5**)
July 2013	Preliminary results sent to participants on Waters A and B (**Appendix 5**)

1.2 Participating laboratories

The participating laboratories were mainly national research institutes, authorities and monitoring laboratories. From MSs, 67 laboratories were nominated by the national representatives in the expert group according to the Euratom Treaty Art. 35/36. Traditionally, laboratories from the pre-accession countries (AC) and other European countries are also invited by JRC-GEEL to participate. This time, 9 laboratories from AC joined the ILC.

In total 71 laboratories (63 from MS and 8 from AC) from 31 countries reported results. The list of all participating laboratories is shown in Appendix 6. Since the anonymity is a

requirement in this programme, the identity of the laboratories is not shown in the compilation of the results. The order of the listing of participants in Appendix 6 is not the same as the laboratory number used throughout the data evaluation in this report.

Table 1. Participating laboratories' origin.

	MS	**AC**	**Total**
Nominated	67	9	76
Sample sent	64	9	73
Results submitted	63	8	71

Three nominated laboratories (one from Romania, one from Lithuania and one from Greece) did not respond to our letters. Laboratories 20 and 53 did not submit their results due to the technical problems with their equipment.

Table 2. Number of results submitted per sample.

Water sample	**Gross alpha activity**	**Gross beta activity**
Water A		
All submitted results	68	68
Results below LOD	24	4
Results over LOD	44	64
No measurements	3 (labs 27, 31, 63)	3 (labs 27, 42, 43)
Water B		
All submitted results	70	68
Results below LOD	0	4
Results over LOD	70	64
No measurements	1 (lab 31)	3 (labs 27, 42, 43)
Water C		
All submitted results	66	64
Results below LOD	0	0
Results over LOD	66	64
No measurements	5 (labs 31, 38, 39, 55, 65)	7 (27, 38, 39, 42, 43, 55, 65)

Laboratories 27 and 31 informed in advance that they are measuring only gross alpha and gross beta activity concentrations, respectively. Laboratory 39 did not report result of gross alpha for Water C due to the break of the equipment.

1.3 Questionnaire

Participants were asked to fill in a questionnaire (**Appendix 7**). It was composed of seven parts concerning the information on the laboratory, its routine measurements, sample treatment, measurement methods, instrumentation, uncertainty budgets and some additional information. Information in the questionnaire is essential in order to evaluate the results of the intercomparison. All of the laboratories submitted the questionnaire electronically which shows the good collaboration of the participants.

2 Reference values

2.1 Test materials

To run a representative ILC, the selection of test items is a crucial step. Therefore, our first objective was to select waters as realistic test items for an interlaboratory comparison on gross alpha/beta measurement in drinking waters. For this reason, a preliminary radioanalytical survey studying the naturally occurring alpha emitting radionuclides was carried out in 11 popular and regularly consumed mineral waters from the European market. The activity concentrations of the main naturally occurring alpha-emitting radionuclides (^{226}Ra, ^{210}Po, ^{234}U, ^{235}U, ^{238}U and ^{228}Th) were determined using alpha-particle spectrometry after separation from the matrix elements.

In order to find representative water samples of natural origin for the gross alpha/beta ILC, the following important parameters were taken into account during the material selection: activity concentration of the alpha-emitting radionuclides, salinity, chemical composition, directives and recommendations (WHO, 2011; EC, 2013).On the basis of the alpha-particle spectrometry results and the salinity, two candidates were selected as ILC materials. Additionally, one spiked sample was prepared as quality check sample at JRC-GEEL, thus three ILC samples were used in total. More details on the ILC sample selection are described elsewhere (Jobbágy et al., 2013).

The most important stages of the sample selection and the characterisation of the candidate waters are presented in the next paragraphs.

The codes of the analysed mineral waters are listed with their country of origin together with their total dissolved solids (TDS) in **Table 3**. The EU classification of mineral waters as a function of total dissolved solids (TDS) or salinity is presented in **Table 4**.

Table 3. List of candidate mineral waters.

Sample code	Country of origin	TDS (mg L^{-1})	Mineral water class (EC, 2009)
1	Belgium	2078	Rich in mineral salts
2	Belgium	33	Very low mineral content
5	Belgium	385	Low mineral content
4	France	3325	Rich in mineral salts
3	France	479	Low mineral content
7	France	1200	Intermediate mineral content
8	Italy	948	Intermediate mineral content
6	Poland	2193	Rich in mineral salts
10	Poland	821	Intermediate mineral content
11	Poland	3931	Rich in mineral salts
9	Poland	1370	Intermediate mineral content

Table 4. The EU classification of mineral waters as a function of total dissolved solids (TDS) (EC, 2009).

Mineral water type	TDS criteria
Very low mineral content	< 50 mg L^{-1}
Low mineral content	50 - 500 mg L^{-1}
Intermediate mineral content	500 - 1500 mg L^{-1}
Rich in mineral salts	> 1500 mg L^{-1}

On the basis of the salinity classification, one mineral water had very low mineral content, two were with low mineral content (+as the majority of the European tap waters), and four waters each had intermediate mineral content or were rich in mineral salts. The measured gross alpha activity concentrations of the mineral waters and the total alpha activities are summarized in **Table 5**. The total alpha activity values are calculated from the sum of the activity concentrations of ^{226}Ra, ^{210}Po, ^{234}U, ^{235}U, ^{238}U and ^{228}Th.

Table 5. The gross alpha activity-and the total alpha activity concentrations in mineral waters.

Sample code	Gross alpha activity (mBq L^{-1})	Total alpha activity (mBq L^{-1})
1	75 ± 23	68.8 ± 1.9
2	23 ± 9	4.1 ± 0.3
3	70 ± 15	87.5 ± 2.1
4	392 ± 77	248 ± 12
5	423 ± 37	358 ± 17
6	1073 ± 89	664 ± 30
7	111 ± 21	98.9 ± 2.4
8	518 ± 42	292 ± 11
9	422 ± 42	268 ± 14
10	26 ± 11	n.a.
11	489.4 ± 84.5	n.a.

The difference between the gross alpha (measured) and the total alpha (calculated from radionuclide specific analysis) is due to the interferences during sample preparation and measurement (e.g. self-absorption, ^{222}Rn ingrowth from ^{226}Ra) for the gross alpha method, therefore it is less accurate than alpha-particle spectrometry (Jobbágy et al., 2010). Since we had insufficient amount of sample 10-11, and they had either too low gross alpha activity (water 10) or too high salinity (water 11), we decided to exclude them from the later analysis. The measured activity concentrations of ^{210}Po, ^{226}Ra, ^{228}Th and uranium isotopes in mineral water samples are given in **Table 6**-**7**.

Table 6. The activity concentrations of ^{210}Po, ^{226}Ra and ^{228}Th.

Sample code	Activity concentration (mBq L^{-1})		
	^{210}Po	^{226}Ra	^{228}Th
1	1.5 ±0.2	11.1± 1.0	n.m.
2	2.6 ±0.2	1.5± 0.3	n.m.
3	3.2 ±0.3	12.6± 1.1	n.m.
4	9.0 ± 0.4	236± 12.	n.m.
5	2.7 ± 0.2	333± 17	5.4 ± 0.5
6	4.7 ± 0.3	632± 30	n.m.
7	10.8 ± 0.6	3.7± 0.5	0.4 ± 0.1
8	2.3 ±0.7	97± 5	n.m.
9	n.m.	268± 14	n.m.

n.m. – not measured; Thorium activity concentrations were measured in the waters selected for the ILC only.

Table 7. The activity concentrations of ^{234}U, ^{235}U, ^{238}U and the ratio of ^{234}U/^{238}U.

Sample Code	Activity concentration (mBq L^{-1})			^{234}U/^{238}U
	^{234}U	^{235}U	^{238}U	
1	38.9 ± 1.4	0.7 ± 0.1	16.6 ± 0.7	2.34 ± 0.13
2	< 0.8	< 0.44	< 0.44	-
3	30.3 ± 1.1	1.6 ± 0.1	39.8 ± 1.4	0.76 ± 0.04
4	1.7 ±0.1	< 0.44	0.8 ± 0.1	2.16 ± 0.33
5	11.9 ± 0.5	0.2 ± 0.1	5.0 ± 0.3	2.40 ± 0.16
6	19.8 ± 0.9	0.2 ± 0.1	7.1 ± 0.4	2.78 ± 0.21
7	58.5 ± 2.1	1.0 ± 0.1	24.4 ± 1.0	2.39 ± 0.13
8	98 ± 67	2.7 ± 0.5	92 ± 7	1.06 ± 0.11
9	n.m.	n.m.	n.m.	n.m.

n.m. – not measured

Measurement uncertainties are given as expanded uncertainty with a coverage factor of k = 2, corresponding to a ~95% confidence level.

Five water samples showed elevated levels of ^{226}Ra activity concentration (> 100 mBq L^{-1}) furthermore in one sample the ^{226}Ra activity concentration itself already exceeded the less strict WHO gross alpha screening level (500 mBq L^{-1}) (WHO, 2011). The ^{226}Ra activity concentration in four mineral water samples was well below 50 mBq L^{-1}.

As shown in **Table 7**, the uranium activity concentrations in these waters are in the range of 1.7 – 98 mBq L^{-1} for ^{234}U, < 0.44–92 mBq L^{-1} for ^{238}U and < 0.44–2.7 mBq L^{-1} for ^{235}U.

Activity concentrations of ^{210}Po are relatively low, in the range of 1.5–10.8 mBq L^{-1}, as compared to the activity concentrations of uranium and radium isotopes.

The total alpha activity concentration of the samples - which is the sum of ^{226}Ra, ^{210}Po, ^{234}U, ^{235}U, ^{238}U and ^{228}Th activity concentrations - is presented in **Fig. 2** together with the EC and WHO screening levels for drinking water concerning gross alpha activity.

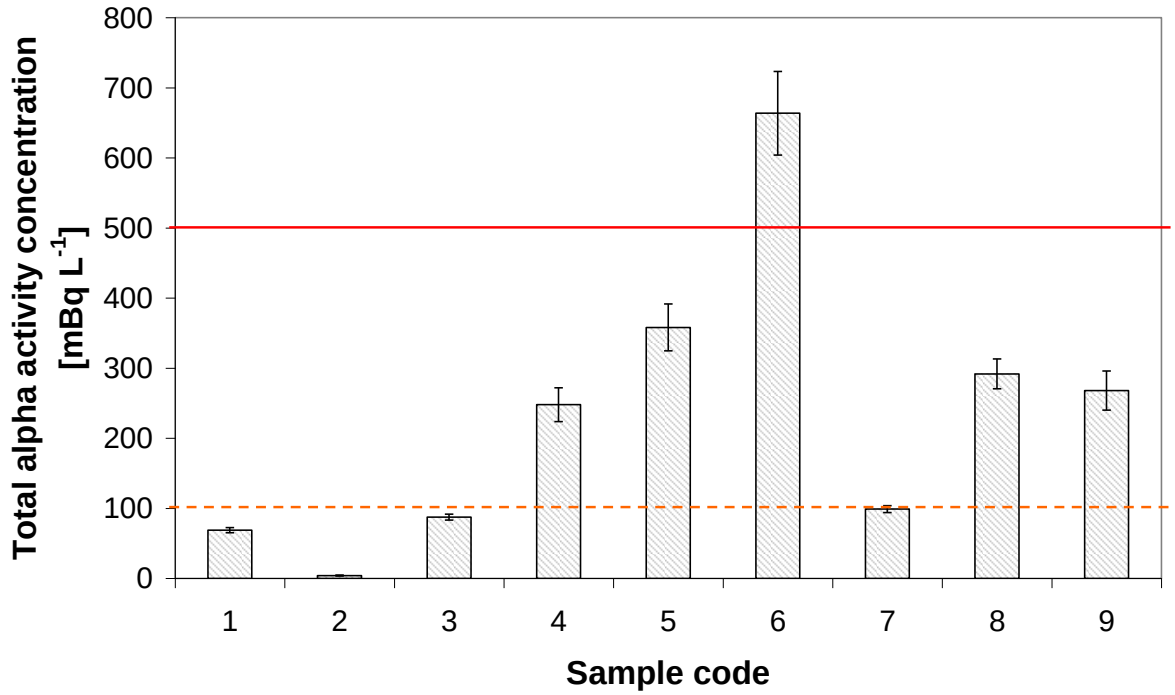


Fig. 2. Total alpha activities of mineral water samples compared with the EC [19] (dashed line) and the WHO [5] (solid line) gross alpha screening levels for drinking water.

Since the monitoring laboratories have to be confident in measuring activities near the screening level or close to the detection limit, but also at elevated levels or in the range of the recent WHO screening level (WHO, 2011), we selected the ILC water samples accordingly (**Table 8**).

Table 8. Parameters taken into account for ILC water selection.

Parameter	Activity concentration (Bq L^{-1})	References
Limit of detection	0.02-0.1	ISO 9696, 2007; ISO 9697, 2008
	$\Sigma\alpha = 0.04$; $\Sigma\beta = 0.4$	EC, 2013
Screening levels	$\Sigma\alpha = 0.5$; $\Sigma\beta = 1$	WHO, 2011
	$\Sigma\alpha = 0.1$; $\Sigma\beta = 1$	EC, 2013

To get an overview how the analysed waters meet our pre-set requirements on salinity and alpha activity concentration, the total alpha activity concentration as a function of total dissolved solids are presented in **Fig. 3**.

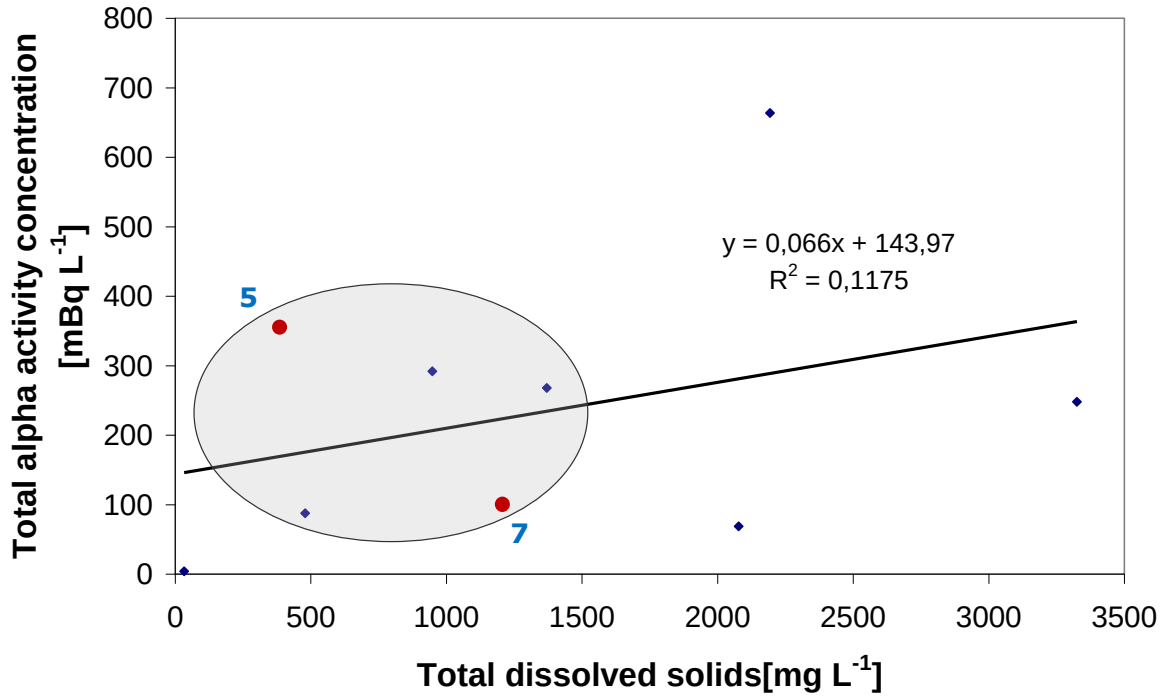


Fig. 3. Total alpha activity concentration as a function of total dissolved solids. The data points in the oval area represent waters that fulfil the ILC test requirements for salinity and alpha activity concentration. Red circles with the corresponding sample code indicate the selected waters.

The shaded area in **Fig. 3** represents the region where waters fulfil the pre-set salinity and alpha activity requirements for ILC test items. After applying this filter, five waters were considered as potential candidates for our interlaboratory comparison. Finally we decided to select two waters as ILC test items, where the first one is close to the gross alpha detection limit. While the other has an elevated total alpha activity in the range of the recent WHO gross alpha screening level. In terms of salinity, the selected ILC test items fall into the range where the majority of drinking waters are (~50-1500 mg L^{-1}).Besides the two natural origin water samples (Water A, Water B) a third one (Water C) was also included. The latter water was deionised water that was spiked.

Water C is, in principle, the easiest sample to measure since its gross alpha/beta activity concentration is the highest among the ILC samples. However, from a measurement point of view, the gross alpha activity is not the only key factor, but the alpha/beta emitting radionuclides and the total dissolved solid content have to be considered as well. Taking into account all three factors one can make an order of difficulty in terms of measurement as follows: Water C < Water A ≤ Water B.

2.2 Processing and homogeneity

One of the water samples was provided by a mineral water supplier in anonymous 1.5 L polyethylene terephthalate (PET) bottles. The other two samples were homogenised and bottled into 1 L polypropylene bottles as described below. Only the spiked sample was acidified to pH = 1.2 ± 0.1 with concentrated HNO_3. Samples were stored in a dark and dry place at a basement ambient temperature.

Each vessel used in this project was custom made and fulfils the requirements for trace elements in water reference materials since they can be rigorously cleaned with a sequence of strong acid and Type 1 ultrapure water. The vessel wall is a sandwich construction and consists of glass fibre reinforced plastic (GRP) as outer liner and Teflon® PFA (perfluoroalkoxy copolymer resin) as an inner liner. The dimensions of these vessels are such that the Dyna-mixer CM500 (WAB, Basel - Switzerland) be used for easy cleaning of these vessels between projects. Consequently before filling with the water and the Type 2 pure water the vessels were rinsed with >50 L Type 2 pure water and placed in the Dyna-mixer CM500.

The whole system, when comprising of four inter-connected vessels, allows homogenisation of up to 2 m^3 of water at the same time. Other combinations of for example two or three vessels are also possible. The pneumatically driven bellow- pumps (Iwaki FS-30-HT2) are made so that all parts in contact with the water are made of PFA or PTFE. The vessels are also equipped with a level sensor and via a feedback circuit the pumping speed is individually controlled so that the level stays the same in all vessels during recirculation. A full re-circulation of 2 m^3 can be achieved in approximately one hour with a flow of about 30 L/min per pump. As an example, the one of the mineral waters was emptied in two of these vessels and homogenised by circulating them in the vessels for three days. After the complete homogenisation water samples were bottled into 1L polypropylene bottles (Nalgene type) and transported to their interim storage room into within JRC-GEEL premises.

Water B was bottled into anonymous 1.5 L polyethylene terephthalate bottles by a mineral water producer company. Water A and Water C were prepared as follows.

Water A was produced from a commercial mineral water (Badoit, SaintGalmier) from France. Two perfluoroalkoxy polymers (PFA)-lined drums of 550 L were filled with the mineral water and the water was thereafter re-circulated for 24 h at 15 L/min using two inert Iwaki bellow pumps. During filling an intermediate polycarbonate buffer tank of 20 L (Nalgene, Rochester, NY, USA) was used and the water was pumped simultaneously from the two tanks into the buffer tank. The buffer tank was placed in a clean bench and the water bottles were filled manually by opening and closing the tap of the buffer tank. Prior to filling, the buffer tank was rinsed with 2 x 10 L of Type 1 water (18.2 MΩ cm, 0.056 µS/cm at 25 °C and TOC < 5 ng/mL) from a Milli-Q Advantage system (Millipore, Billerica, MA, USA) and 20 L of mineral water. In this manner 777 bottles were filled. The 1-L bottles were made of high density polypropylene (HDPE) with a leak-proof HDPE-screw cap (Nalgene).

Water C was a spiked Type 2 water from a Millipore ELIX-35 system (>5 MΩ cm, 0.2 µS/cm at 25 °C and TOC < 30 ng/mL) with added inorganic salt mixture composed by NaCl, $CaCl_2$ and $Sr(NO_3)_2$. During several days 500 L of Type 2 water was collected in portions into one PFA-lined drum of 550 L. The relative standard deviation on the total

water mass was 0.5 %. Subsequently preliminary weighed NaCl, $CaCl_2$ and $Sr(NO_3)_2$ salt mixture was added. Thereafter 2 L of concentrated nitric acid was added (pH = 1.2 ± 0.1) followed by $^{90}Sr/^{90}Y$ and ^{241}Am spikes from standardized solutions. The contents were thereafter mixed using the Iwaki inert bellows pump of the water handling system for 16 hours at 15 L/min. Subsequently 482 of the 1-L HDPE bottles (Nalgene) were filled as described above. Samples were stored in a dark and dry underground storage place at ambient temperature. Reference values were determined by using gravimetric approach, where the standardized solutions were weighed on a calibrated balance which is traceable to the JRC-GEEL standard kilogram.

The $^{90}Sr/^{90}Y$ (A = 17.5 ± 0.7 kBq/g) and ^{241}Am (A = 1980 ± 80 Bq/g) radioactive solutions were standardized at JRC-Geel using liquid scintillation counting where the efficiency calibration was done by following CIEMAT/NIST method. These standardized solutions were traceable to the SI. The activity values were calculated for the reference date 25 April 2012. From the above mentioned original standardized solutions 0.02978 g $^{90}Sr/^{90}Y$ and 0.24275 g ^{241}Am solutions were taken respectively to produce the spiked water sample. The uncertainty on the weighing was approximately 0.1 %.

Fig.4. Interlaboratory comparison materials: Water A, Water B and Water C from left to right.

2.2.1 Homogeneity study

Since inhomogeneity may occur within a batch and can lead to discrepant results, it had to be demonstrated that these samples are identical within the whole batch such that each laboratory receives samples with the same parameters. Therefore, a homogeneity study between bottles was necessary to establish its contribution to the uncertainty budget of the reference values. The uncertainty budget was built with respect to all

contributing parameters like weighing, volumetric measurements, counting statistics and homogeneity. Adsorption tests were performed to check the loss of radionuclides due to adsorption to the container wall.

The manufacturer provided Water B in the same bottles and caps as they are available in supermarkets but without labels (anonymous bottles). These bottles were from the same batch, thus for these reasons they were considered homogeneous and were not homogenised at JRC-GEEL. However, their between bottles homogeneity was checked. Water B samples were sent to the participant laboratories in the same package as arrived from the bottling site.

Only Water A and Water C have been homogenised at JRC-GEEL. Water A was homogenised and re-bottled on 10 May 2012. Water C was filled in a 500 L tank on 25 May 2012, homogenised for several days and bottled on 30 May 2012.

For the homogeneity study a random stratified method was used to avoid systematic errors within the batch. Bottles were selected with the help of SNAP excel application developed at Reference Materials unit at JRC-GEEL. From each batch of water eight to ten bottles were randomly selected and analysed using gross measurements and radionuclide specific analysis of the natural origin alpha emitting radionuclides (Water A and B) as presented in **Table 9**. This included the activity concentration determination of the main contributing alpha-emitting radionuclides to the gross alpha activity concentration (e.g. in case of Water B the activity concentration of ^{226}Ra was determined).

Table 9. Parameters used for the homogeneity study.

ILC test item	*Parameters checked*
Water A	Gross alpha/beta activity; uranium activity
Water B	Gross alpha/beta activity; ^{226}Ra activity
Water C	Gross alpha/beta activity

The homogeneity and the short term stability of the radionuclides in the matrix was evaluated using the SoftCRM version 2.0.10 software following the certification principles for reference materials as given in ISO Guide 35 (2006). Grubbs' test was performed to detect potentially outlying individual results. No outliers were detected for gross alpha and beta activities.

A priori requirement on the uncertainty from between bottle homogeneity (u_{bb}) was set to < 10 %.

In the case of bottled waters, the main contribution to their instability was the adsorption of radionuclides to the container wall. Short term stability analysis was done when a small aliquot of water sample was taken from the 1 L bottle and analysed using gross measurements and the aforementioned radionuclide specific analysis (Water A and B). The first stability measurements were done already before the beginning of the ILC and the last one month after the submission of the last result.

Table 10. Contribution of uncertainties to the expanded uncertainty of the reference values (%).

Sample	u_{bb}	u_{sts}
	Gross alpha activity	
Water A	13.1	4.8
Water B	1.5	2.0
Water C	3.4	6.1
	Gross beta activity	
Water A	2.0	5.9
Water B	1.5	2.0
Water C	2.8	4.7

Only Water A gross alpha activity exceeded the target uncertainty of inhomogeneity. However, the nuclide specific analysis confirmed that u_{bb} is below 10 %, thus there was no technical reason to exclude this water from the ILC. For the combined uncertainty this higher value (13.1 %) was considered as it was derived from the gross alpha measurements.

Homogeneity plots are presented in **Fig. 5-10**. The numerical values are collected in **Appendix 9**.

Water A

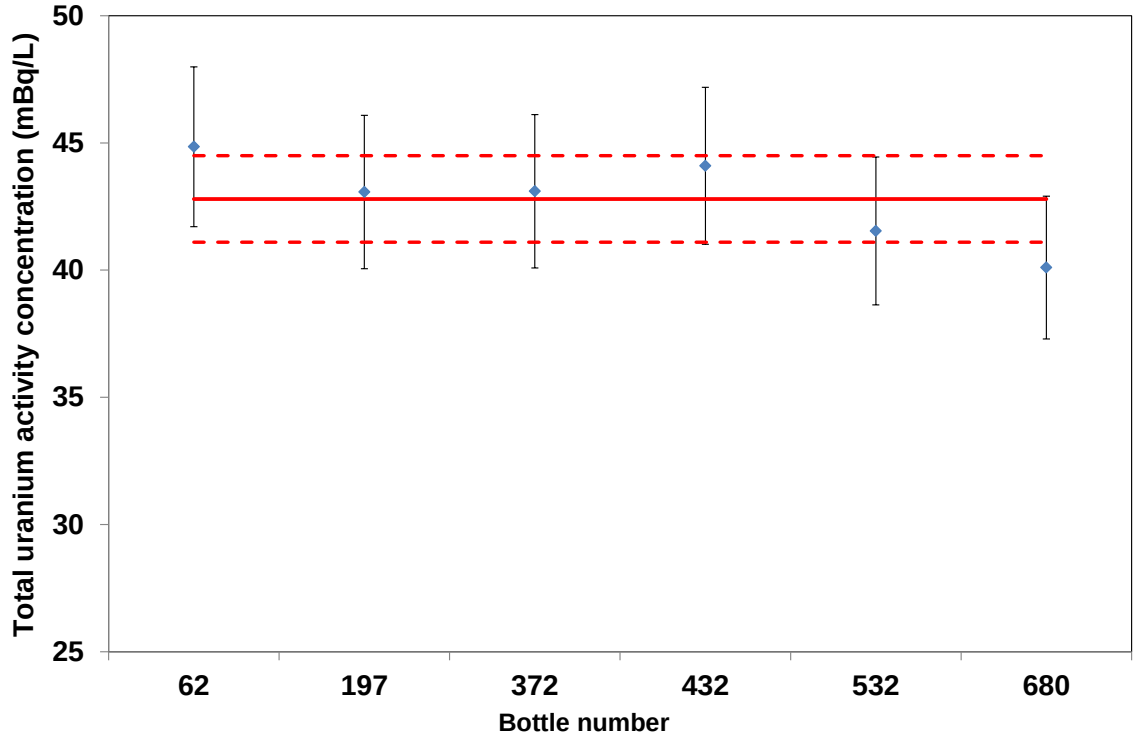


Fig. 5. Total uranium (sum of ^{238}U and ^{234}U) activity concentration in Water A. All uncertainties are combined standard uncertainties at the 1 sigma level (k=1). The red solid line indicates the average and the dashed lines indicate the ± 1sigma (k=1).

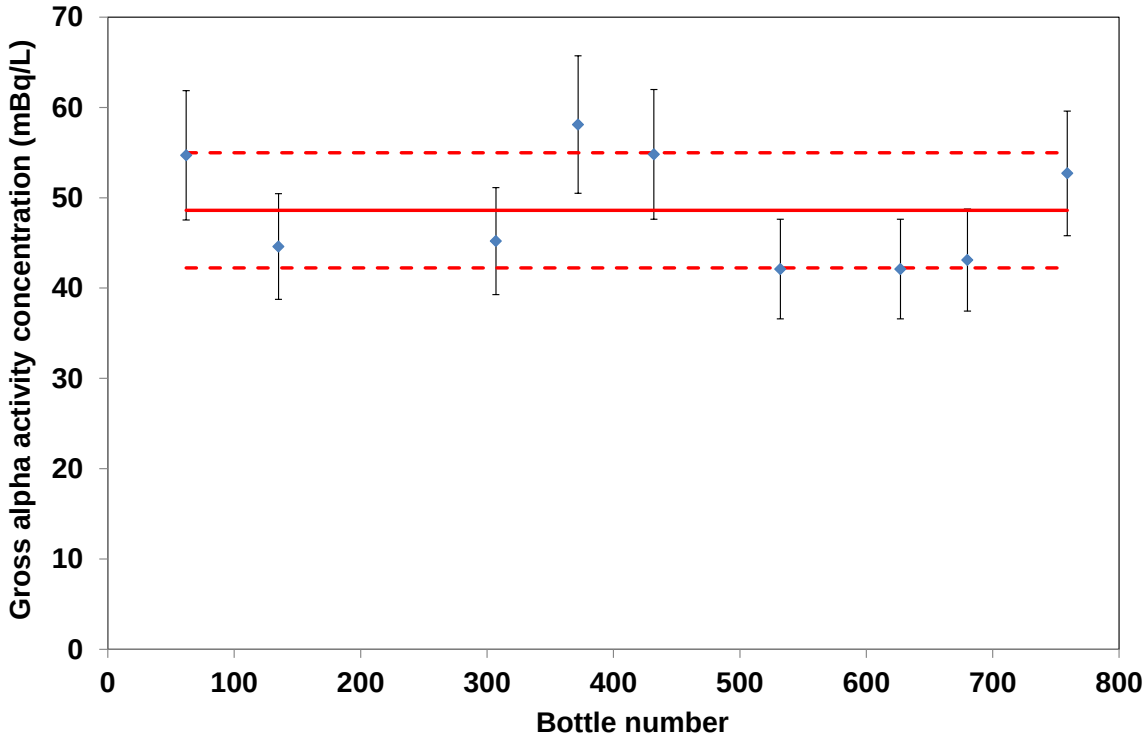


Fig. 6. Gross alpha activity concentration in Water A. All uncertainties are combined standard uncertainties at the 1 sigma level (k=1). The red solid line indicates the average and the dashed lines indicate the ± 1sigma (k=1).

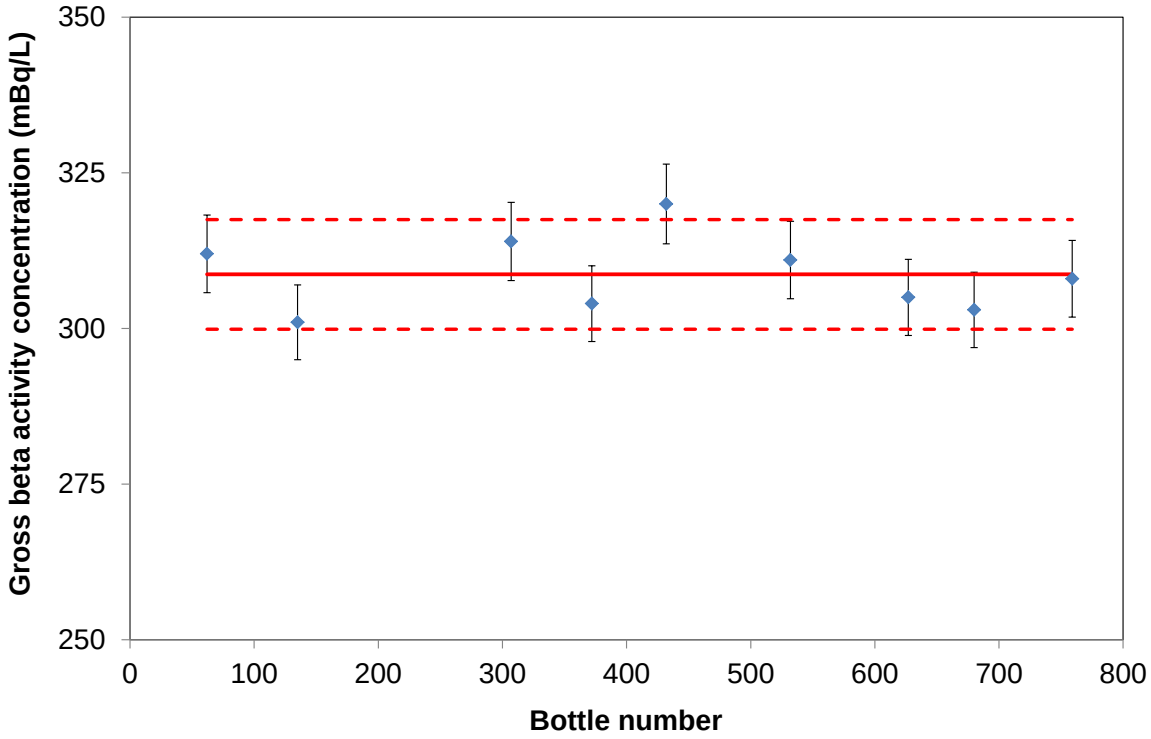


Fig. 7. Gross beta activity concentration in Water A. All uncertainties are combined standard uncertainties at the 1 sigma level (k=1). The red solid line indicates the average and the dashed lines indicate the ± 1sigma (k=1).

Water B

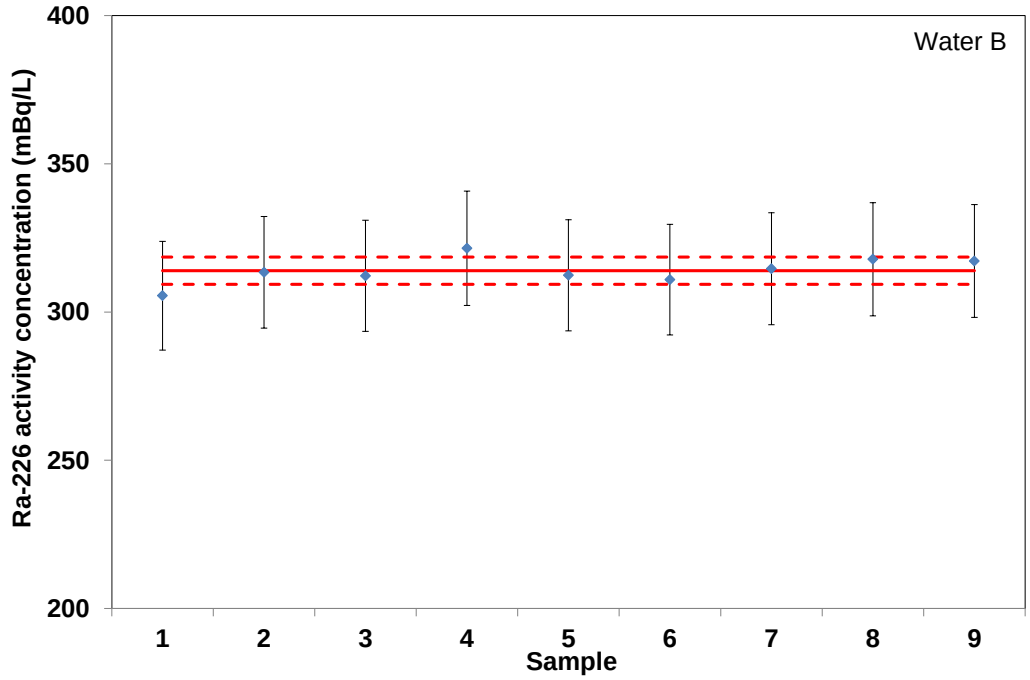


Fig. 8. ^{226}Ra activity concentration in Water B. All uncertainties are combined standard uncertainties at the 1 sigma level (k=1). The red solid line indicates the average and the dashed lines indicate the ± 1sigma (k=1).

Water C

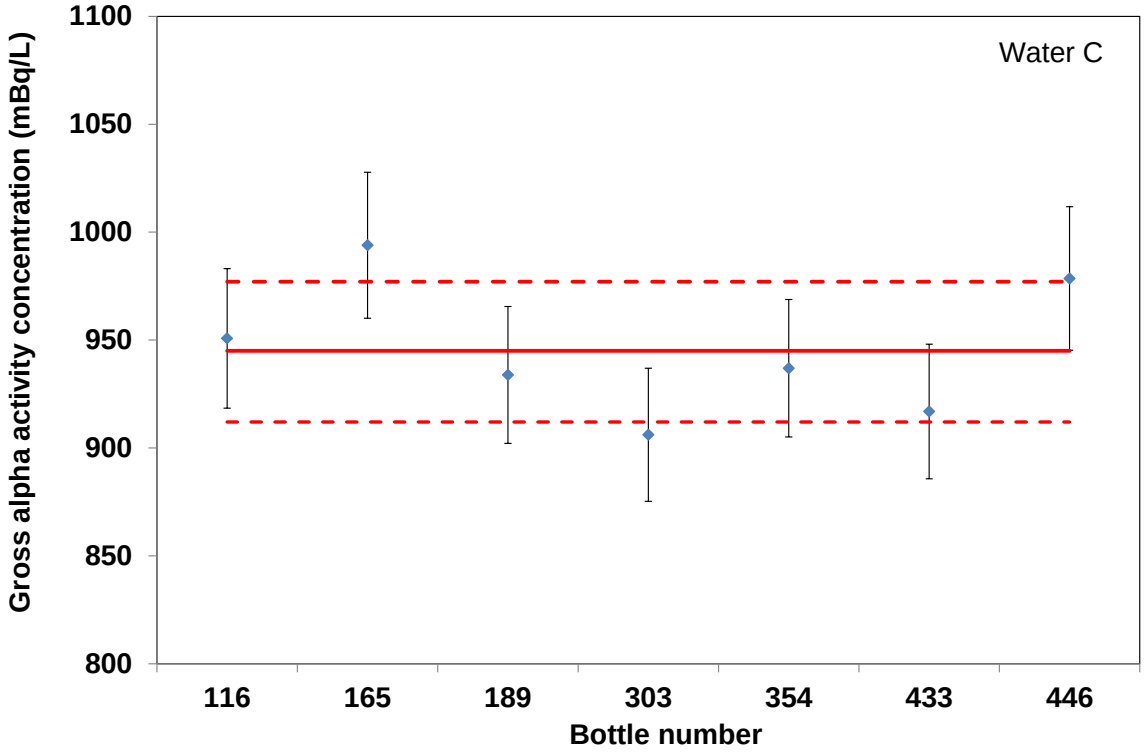


Fig. 9. Gross alpha activity concentration in Water C. All uncertainties are combined standard uncertainties at the 1 sigma level (k=1). The red solid line indicates the average and the dashed lines indicate the ± 1sigma (k=1).

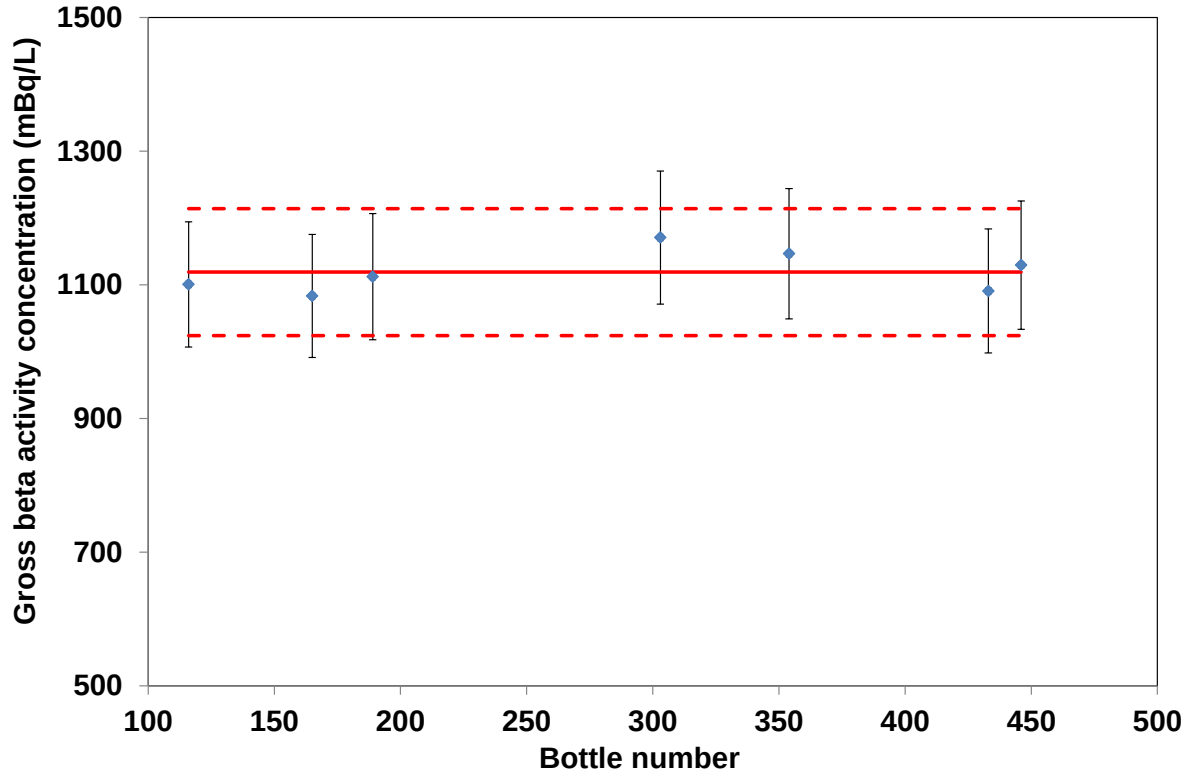


Fig. 10. Gross beta activity concentration in Water C. All uncertainties are combined standard uncertainties at the 1 sigma level (k=1). The red solid line indicates the average and the dashed lines indicate the ± 1sigma (k=1).

2.2.2 Stability study

The stability of the intercomparison samples was also checked via the adsorption of radionuclides on the walls of the bottles previously containing the ILC samples. This was done by filling empty bottles with 0.1 M nitric acid, the bottles were stored for a period of two months. Then uranium, ^{226}Ra and gross alpha/beta analyses were performed for Water A, B and C respectively. The measured activity concentrations were below the detection limit. The samples were considered stable over time of the complete comparison exercise. However, from the direct water analysis higher variations were observed, therefore those values were considered for the evaluation of combined uncertainty of the reference value (u_{sts}> 0).

The homogeneity and stability values (u_{bb},u_{sts}) mentioned above were taken into account in determining the uncertainty of the reference values (see Section 2.4, Page. 31).The scatter of the results from the gross measurements was larger than that from the homogeneity values from the radionuclide specific analysis, so the former were used in the uncertainty budget.

Stability plots are presented in **Fig. 11-13**. The numerical values are presented in **Appendix 10**.

Water A

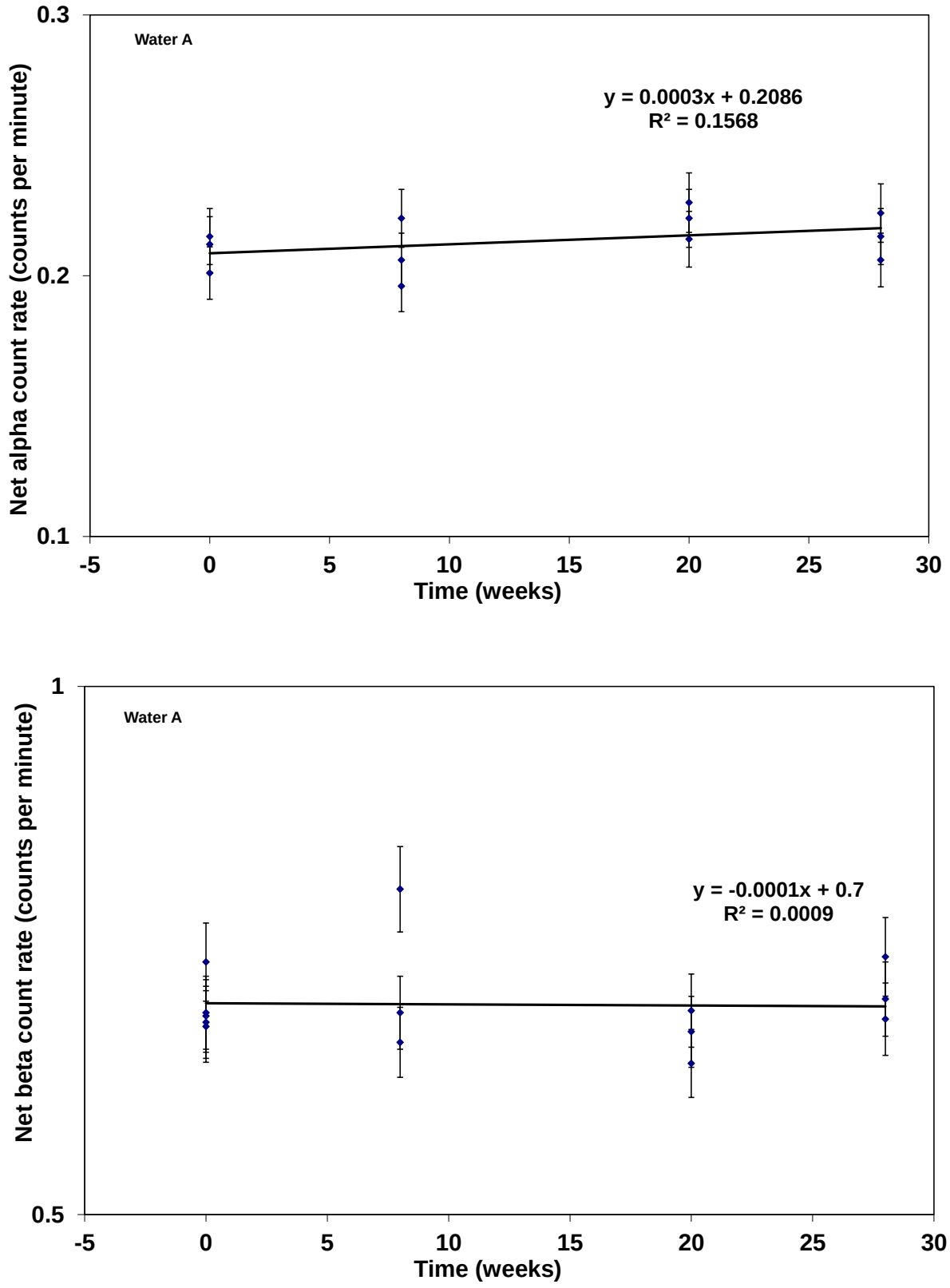


Fig. 11. Gross alpha and beta results displayed as corrected and normalised net counting rate with regression line for Water A sample stored ambient temperature. Regression parameters are given in the figure. Error bars indicate combined measurement uncertainty, u_{meas}.

Water B

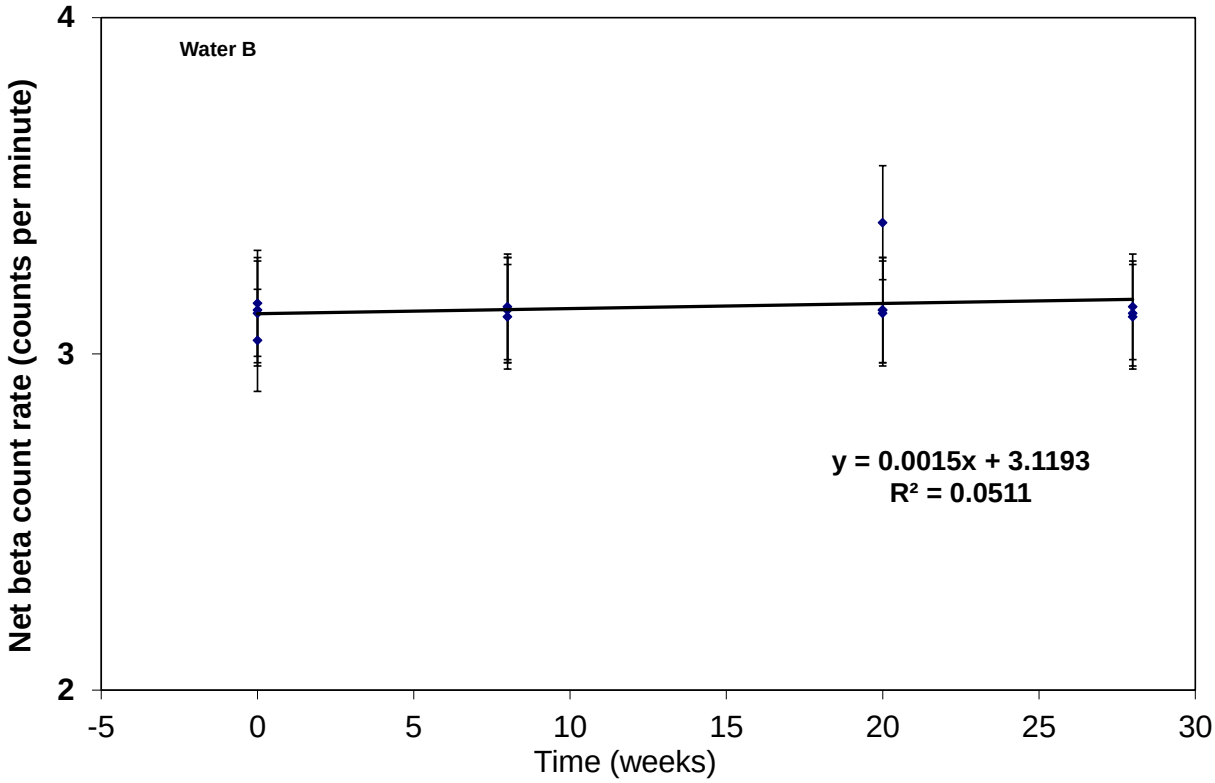


Fig. 12. Gross beta results displayed as corrected and normalised net counting rate with regression line for Water B sample stored ambient temperature. Regression parameters are given in the figure. Error bars indicate combined measurement uncertainty, u_{meas}.

Water C

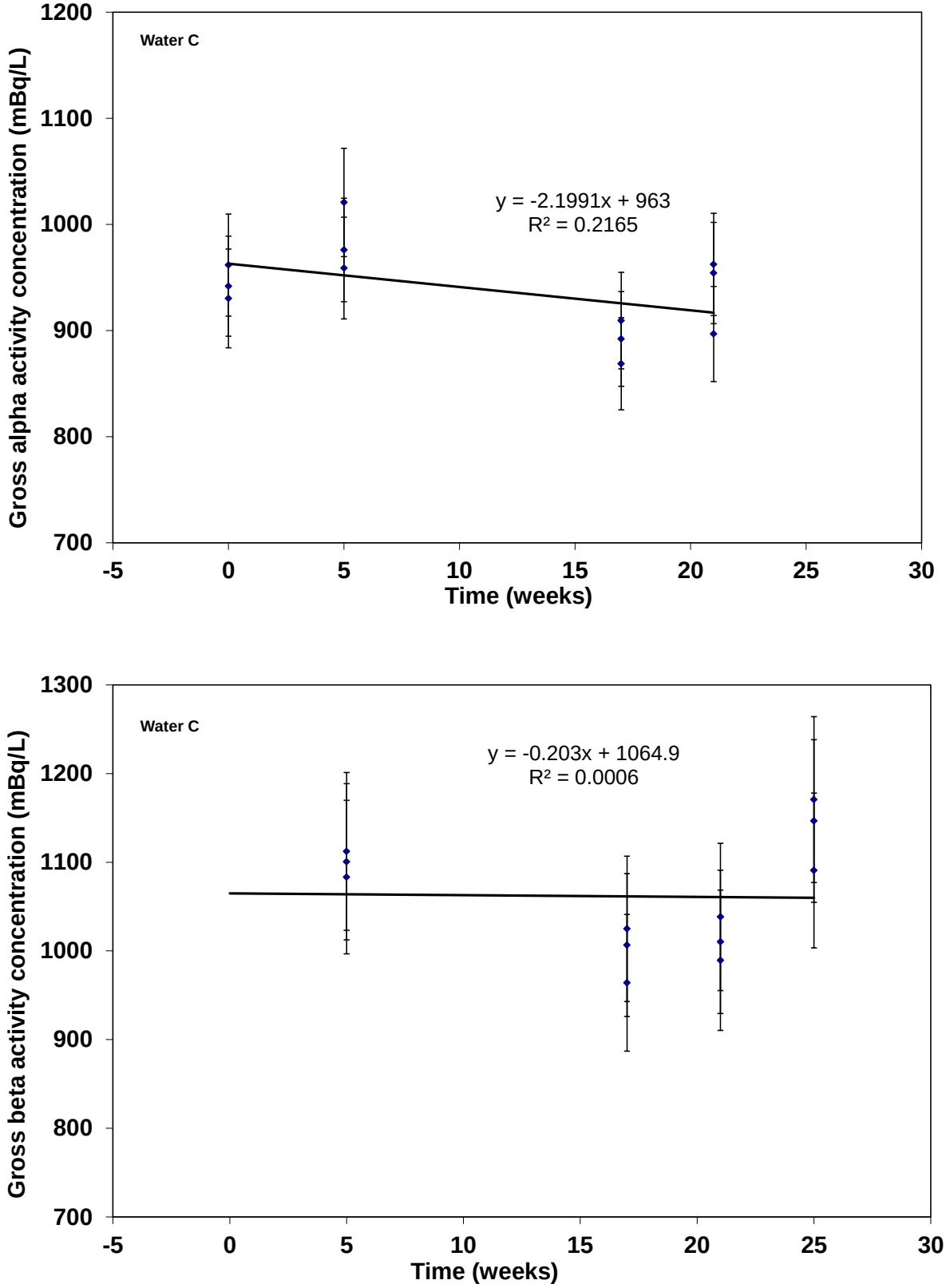


Fig. 13. Gross alpha and beta results displayed as corrected and normalised net counting rate with regression line for Water C sample stored ambient temperature. Regression parameters are given in the figure. Error bars indicate combined measurement uncertainty, u_{meas}.

No correlation can be observed for the three water samples. However, a decreasing but not statistically significant trend can be discerned for Water C gross alpha activities. There is no reasonable explanation for this phenomenon since water sample was acidified and no microbial activity was observed. Analysing Water C gross beta activity results the stability of water C can be confirmed for 25 weeks.

No significant change in the activity concentration of the uranium, ^{226}Ra and gross alpha/beta activity at ambient temperature was observed in samples kept for > 20 weeks, when a decrease was observed this was only within overlapping uncertainties. Taking into consideration that the shipping period of the material did not take more than 1 week and samples were not exposed to extreme temperatures the material was dispatched without further precautions under ambient conditions. The gross alpha analysis had to be performed within six weeks (sample shipment mid-September2016, result submission 31 October 2016).

2.3 Establishing reference values

Reference value determination was done in three independent laboratories where the most common routine methods were used as listed below (**Table 11**).

Table 11. Methods used for gross alpha/beta reference values.

Lab	Method
SCK•CEN	Evaporation, solid scintillation counting
RIVM	Spike addition, evaporation and gas flow proportional counting (ISO 9696/9697)
JRC-GEEL	Co-precipitation, gas flow proportional counting (ISO 10704)
JRC-GEEL	Thermal pre-treatment, liquid scintillation counting (ISO 11704)
JRC-GEEL	Ultra low level gamma-ray spectrometry for ^{40}K analysis

2.3.1 Evaporation method (SCK•CEN and RIVM)

SCK•CEN

Sample preparation started with evaporation of 250 mL sample. To keep all the soluble materials in solution, 5 mL of 10 % acetic acid are added and evaporated under vacuum in a BuchiSyncore Analyst system with a flush back option. With this system all the activity and salt are concentrated in a little volume of about 5 mL. This sample volume is transferred into a stainless steel planchet and the water is dried under infrared lamp until complete dryness. The residue is weighed and measured with the gross alpha/beta system.

Detector system for gross alpha counting: 5 inch (1 inch = 2.54 cm) ZnS(Ag) low background detector. To reduce the background of the counter, the counting cell is flushed with a low flow of dry nitrogen gas. Typical measurement time: 5 × 10000 sec. Alpha background: 0.04 – 0.09 cpm.

Detector system for gross beta counting: the samples were counted in a proportional counter 5 inch low background Canberra Tennelec LB 5500 with sample changer. Typical measurement time: 6 × 3000 sec and 10 × 6000 sec. Beta background: < 2.5 cpm.

For quality check purposes background is measured before each sample measurement. The efficiency of all the counters is controlled each month with a certified source made at SCK·CEN. Radionuclides used for calibration: ^{239}Pu for alpha, ^{204}Tl and ^{90}Sr/^{90}Y for beta. Self-absorption factor is determined by using $NaNO_3$.

RIVM

RIVM followed thick source method from the ISO 9696/9697standards where the surface density has to be kept > 10 mg/cm^2. These methods are based on the direct evaporation of the sample together with radioactive spike. Measurement of the dried residue was done by gas flow proportional counter using P10 counting gas (Ar/CH_4). The most important features of the gross counting system at RIVM are summarised in **Table 12**.

Table 12. Parameters of gross alpha/beta activity measurements at RIVM.

Detector:	Tennelec LB 4100	Gas flow counter using Ar/CH_4 counting gas
Background alpha	0.02 cpm	Typical 0.02 - 0.03 cpm
Efficiency alpha	3.20 %	Following ISO-9696 with thick layer method
Background beta	0.7 cpm	Typical 0.7 - 0.8 cpm
Efficiency beta	44 %	Following ISO-9697 with thick layer method

Measurements were started usually after drying the sample to complete dryness. For gross alpha counting efficiency calibration^{241}Am for gross beta counting efficiency calibration^{40}K in KCl were used respectively.

It has to be mentioned here that RIVM had problems during the evaporation due to the higher salinity of Water A and high relative ambient air humidity during Water B sample preparation (samples looked hygroscopic).

2.3.2 Co-precipitation method (JRC-GEEL)

The gross alpha/beta analysis is based on the co-precipitation approach of the ISO 10704 standard, which consists of a co-precipitation pre-concentration step, filtration and measurement step. The pH of the filtered water sample is set with sulfuric acid and is heated to purge radon and CO_2. Then the radium isotopes are co-precipitated with barium as $Ba(Ra)SO_4$, whereas uranium, thorium and polonium isotopes can be co-precipitated with $Fe(OH)_3$ by adding Fe^{3+} carrier while NH_4OH is used to adjust the pH ≈ 7-8. In the next step the co-precipitates are filtered through a membrane filter and the filters with the precipitate are dried. The dried residue-bearing filters are covered with a co-polymer foil (6 % VYNS: vinyl-acetate/vinyl chloride) to fix the precipitate on the filter and to prevent from contamination of the instrument.

Gross alpha/beta measurements were carried out with a 10-detector, low-background gas-flow proportional counting system (Berthold, model LB790). The high voltage was

set to 1450 V and the counting gas (P10: Ar/CH_4, 90/10) flow was kept stable with a flow rate of ~ 25mL min^{-1}. The gross alpha/beta activity of the filtered and dried precipitate was measured for 3 × 300 min. Alpha and beta counting efficiencies were determined by checking the degree of self-absorption for this geometry. Sources for self-absorption were prepared by using standard solutions – ^{241}Am for alpha,$^{90}Sy/^{90}Y$ in equilibrium for beta-, deionized water and the ISO 10704 standard was followed as for the routine water analysis. Self-absorption factor was determined for a wide range of surface density by varying the $Fe(OH)_3$ and $BaSO_4$ co-precipitate. For background determination the blank samples were prepared identically to the routine water samples by using deionized water and analytical grade reagents.

2.3.3 Liquid scintillation measurements (JRC-GEEL)

The method of the determination of gross alpha/beta activity concentrations in water samples was elaborated on the basis of the ISO 11704-2010 standard and consists of a water sample concentration step, counting sample preparation and a measurement step (**Fig. 14**).

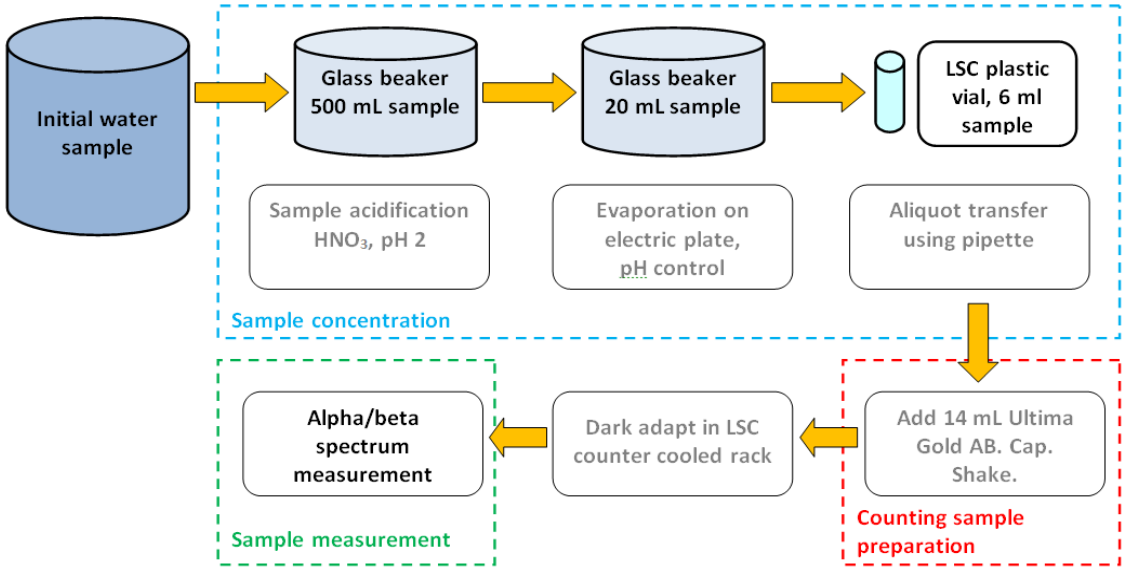


Fig. 14. Scheme of a water sample concentration, preparation of a counting sample and a measurement steps used in the work.

In a sample concentration step 500 g of water were weighted into a glass beaker and a sample was acidified to pH 2 using the nitric acid solution. Water was slowly evaporated up to 20 mL on an electrical plate. The beaker was cooled down and remaining water was weighed. Next, in a counting sample preparation step, an aliquot of 6 mL of water was dispensed into a 20 mL plastic vial containing 14 mL of the scintillator cocktail Ultima Gold AB. A vial was capped and shaken 30 min at 300 rpm. A vial was placed into a cooled tray of the liquid scintillation (LS) counter for 4 hours. Finally, an alpha/beta spectrum of a counting sample was measured 8 hours using the low background LS counter Quantulus. Alpha particle registration events were counted in a window of 500 - 1000 channels, and beta particles were registered in a window of 50 - 900 channels.

Prior to a measurement of a counting sample the pulse shape analysis (PSA) value of the alpha/beta discriminator of the LS counter was determined by dispensing known activities of alpha emitting (^{241}Am) and beta emitting ($^{90}Sr/^{90}Y$) radionuclide standard solution to a concentrated water sample and measuring alpha and beta spectra. The PSA

values were different for a different type of water to be analysed: PSA 80 for samples of water type A and C and PSA 40 for samples of water type B, respectively.

Similarly, a gross alpha/beta counting efficiency was determined by dispensing known activity of alpha or beta emitting radionuclide standard solution to a concentrated water sample. Alpha counting efficiency (referred to ^{241}Am) was 100 %, and beta counting efficiency (referred to ^{90}Sr/^{90}Y) was 98.4 ± 0.9 %.

Blank samples were prepared by dispensing an aliquot of 6 ml of a pre-concentrated de-ionized water sample into a 20 mL plastic vial containing 14 mL of scintillator cocktail Ultima Gold AB. Blank samples were measured 8 hours before and after a measurement of a counting sample. The detailed method optimization procedure is presented in a JRC report (Rožkov, 2014)

2.3.4 ^{40}K measurement with gamma-ray spectrometry

The water-samples were transferred to Marinelli beakers (GaMa 541 G) and measured on detector Ge-4 in the 225 m underground laboratory HADES (Andreotti et al., 2011). Ge-4 is 106% relative efficiency coaxial detector with a thin top dead layer (so-called XtRa). The background count-rate in the 1460 keV peak from ^{40}K is only 4 counts per day (cpd). The count rate for ^{40}K in Water A and B were 35 cpd and 10 cpd, respectively. Clearly, long measurement times are needed with this technique but it is very robust as it requires no pre-treatment of the sample (water). The full Energy Peak efficiency was derived from a reference sample containing KCl dissolved in water contained in a similar Marinelli beaker as for the other samples. The efficiency transfer to correct for small differences in filling height was performed using the Monte Carlo code EGS4. The reported uncertainties are the combined standard uncertainties (k = 1) with major components being counting statistics and the natural isotopic abundance of ^{40}K. The massic activities of ^{40}K are reported in **Table 13**.

Table 13. Massic activities of ^{40}K for the two samples.

Sample	Massic Activity wet mass [mBq·kg^{-1}]
Water A	284 ± 16
Water B	79 ± 9

Table 14. Summary of the gross alpha/beta methods used for ILC 2012 reference value determination.

	SCK	RIVM	JRC-GEEL-1	JRC-GEEL-2
Related standard	ISO 10704	ISO 9696/9697	ISO 10704	ISO 11704
Sample preparation	Direct evaporation	direct evaporation of the sample; sulfation and evaporation; Ignition at 350 ^{o}C for 1 h	Co-precipitation as $BaSO_4$, $Fe(OH)_3$, filtration	Pre-concentration by evaporation
Counting system	Alpha: 5 inch ZnS(Ag); Beta: low background proportional counter	Gas flow proportional counter Ar/CH_4 counting gas	Gas flow proportional counter Ar/CH_4 counting gas	Quantulus: low background liquid scintillation counter
Efficiency calibration	Alpha: ^{239}Pu ; Beta: ^{204}Tl and ^{90}Sr/^{90}Y	Alpha: ^{241}Am; Beta: ^{40}K in KCl	Alpha: ^{241}Am, ^{238}U Beta: ^{40}K, ^{90}Sr/^{90}Y	Alpha: ^{241}Am, Beta: ^{90}Sr/^{90}Y
Counting efficiency	Alpha: ~ 40 % Beta: ~ 50 %	Alpha: 3.2 %; Beta: 44%	Alpha: 20 % Beta: 38 %	Alpha: 100 % Beta: 98 %
Self-absorption correction	Yes, with $NaNO_3$	Yes, spiking the sample	Yes, with the co-precipitate	Not applicable
Background	Alpha: 0.04 - 0.09 cpm; Beta: < 2.5 cpm	Alpha: 0.02 - 0.03 cpm; Beta: 0.7 - 0.8 cpm	Alpha: 0.02 cpm; Beta: 0.8 cpm	Alpha: 0.3-1.5 cpm; Beta: 3.2-4.6cpm
Time delay	No	Few days, but for Water-B ~ 30days.	12-15 hours (drying the precipitate)	4 hours

2.4 Calculation of the reference values

The calculation of reference values was done by taking the arithmetic means from the reference measurements and the uncertainty of the mean from the corresponding uncertainties with this formula:

$$\frac{\sqrt{u_1 + u_2 + \cdots u_n}}{\sqrt{n}} \qquad (1)$$

-$u_{1\ldots n}$: uncertainties from the reference measurements,

-n: number of results considered.

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

-$u_{c,i}$ is the combined standard uncertainty of the laboratory's (or method's) result,

-n: number of laboratories considered.

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

-k: coverage factor (k=2) at ~ 95 % confidence interval

-u_{char}: is the combined standard uncertainty of the mean of the measurement results from the laboratories contributing to the reference value,

-u_{bb}: uncertainty from the activity concentration between bottles of the same batch,

-u_{sts}: uncertainty due to the short-term stability of the samples (longer than the duration of the comparison-exercise).

The contribution of uncertainties from the characterization of the material (u_{char}), homogeneity between bottles (u_{bb}), and the short term stability ($u_{sts,}$) are presented in **Table 15**.

As shown, the largest part of the uncertainty comes from the characterisation followed by the short term stability, except in the case of the gross alpha activity in Water A, where the uncertainty contribution from homogeneity is much higher than that from the short term stability.

Table 15. Contribution of uncertainties for the expanded uncertainty of the reference values (%). (The two rightmost columns are identical to Table 10).

Sample	u_{char}	u_{bb}	u_{sts}
	Gross alpha calculation		
Water A	19.6	13.1	4.8
Water B	6.0	1.5	2.0
Water C	7.5	3.4	6.1
	Gross beta calculation		
Water A	6.0	2.0	5.9
Water B	8.5	1.5	2.0
Water C	7.4	2.8	4.7

Table 16. Uncertainty budget for ISO 10704 gross alpha/beta activity concentration measurements in water samples (Water A-B-C), giving the standard uncertainties (1 s) for a single measurement (5 h counting time).

Uncertainty component	Gross alpha [%]	Gross beta [%]
Counting statistics (min-max)	2.5 - 12.4	1.8 - 6.5
Counting efficiency/Self absorption	4.5	3.1
Volume of the test sample	0.5	0.5
Calibration source	1	1
Weighing	0.2	0.2
Chemical yield	not known	not known
Combined standard uncertainty (u_c) (min-max)	5.3-13.2	3.8-7.3

As expected, the major contributions are counting statistics and self-absorption. The uncertainty due to sample preparation (sample volume, weighing) contributes only 1.1 % to the combined standard uncertainty. There can be a significant bias due to the variation of counting efficiency as a function of alpha particle energies and from the fitting of the self-absorption curve. The bias from the counting efficiency can be up to 75 % (ISO 9696), making the appropriate calibration and determination of self-absorption absolutely crucial. We obtained approximately 20 % bias using different electrodeposited and drop deposited calibration sources (for alpha energies of 4 to 7 MeV). However, positioning of these sources is also important. The uncertainty from chemical yield cannot be quantified, since the determination of the chemical yield itself is difficult in the case of the co-precipitation approach of ISO 10704. Due to chemical manipulations, the yield can never be assumed to be 100 %. Loss of precipitate occurs during the filtration step and some precipitate can adsorb to the walls of the glassware. The mean values of the gross alpha and gross beta activity concentration values from the four measurement methods for each sample are presented in **Fig. 15-17**.

Water A

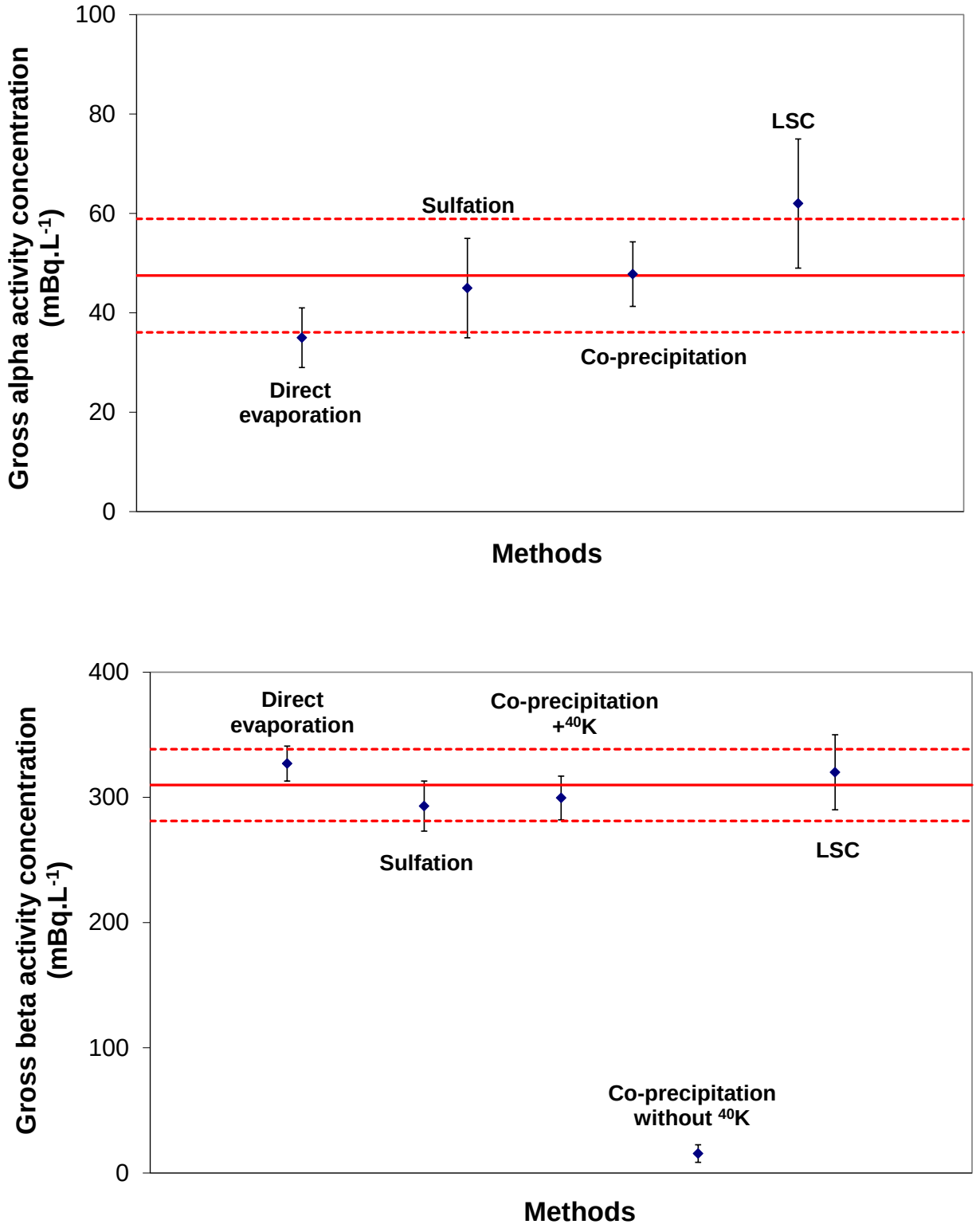
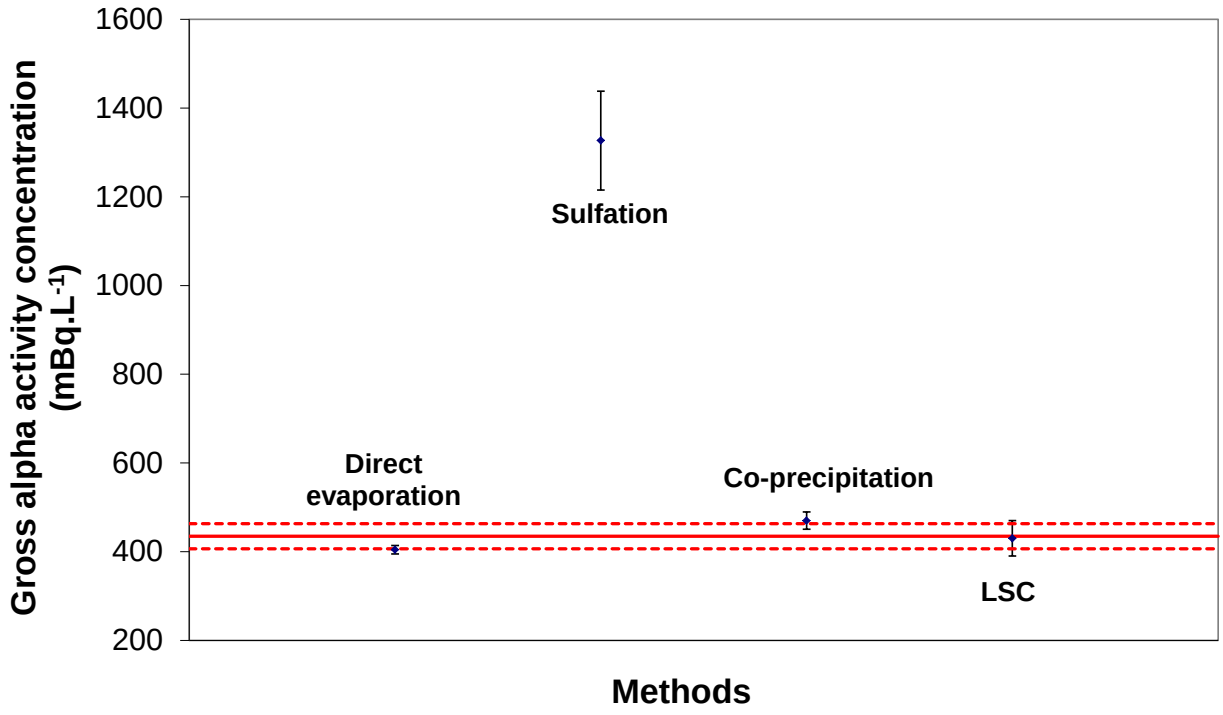


Fig. 15. Gross alpha and gross beta activity concentrations in water A samples, obtained using the radioanalytical techniques from **Table 12**. Activity concentration values are given with their combined uncertainties (k=1). Red solid line shows the mean activity concentration, calculated using measurement results obtained by all techniques. Red dashed lines show the upper and lower values of combined measurement uncertainty of mean activity concentration (k=1).

Water B



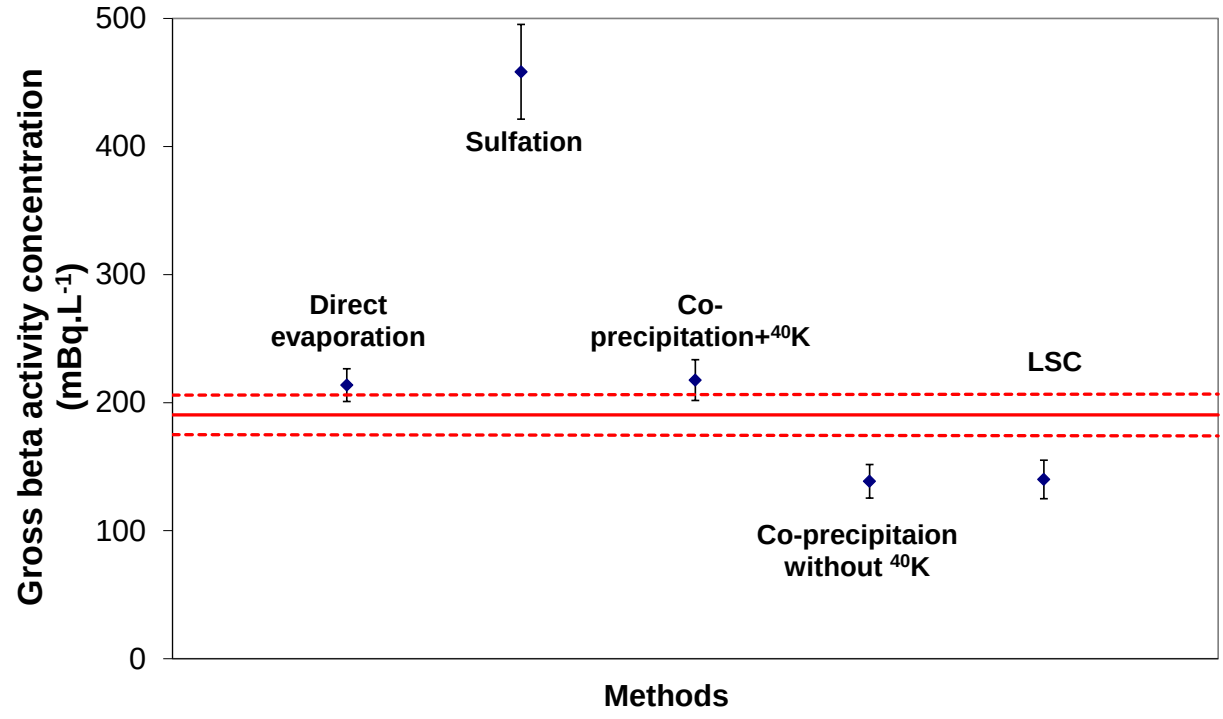


Fig. 16. Gross alpha and gross beta activity concentrations in water B samples, obtained using the radioanalytical techniques from **Table 12**. Activity concentration values are given with their combined uncertainties (k=1). Red solid line shows the mean activity concentration, calculated using measurement results obtained by all techniques excluding sulfation and co-precipitation without ^{40}K methods for gross alpha/beta and gross beta respectively. Red dashed lines show the upper and lower values of combined measurement uncertainty of mean activity concentration (k=1).

Since the sulfation method gave an outlier result for Water B due to the approximately 1 month of delay between sample preparation and measurement, it was not included in the reference value calculation. For gross beta reference value, the results from co-precipitation were excluded as well for the reference value calculation for Water A and B. Nevertheless, the sum results from the co-precipitation and gamma spectrometry ^{40}K measurement are considered. The LSC results were also included, since after optimisation of the system and measuring quality check samples we did not find any technical reason not to do so for Water C.

Water C

Reference values for this spiked water was established by weighing standardized solutions using ^{241}Am for alpha, $^{90}Sr/^{90}Y$ for beta activity. The standardization was done at JRC-GEEL with LSC using high purity solutions free from radiochemical impurities. However, Water C was also measured by the reference laboratories whose results are plotted in the next figures.

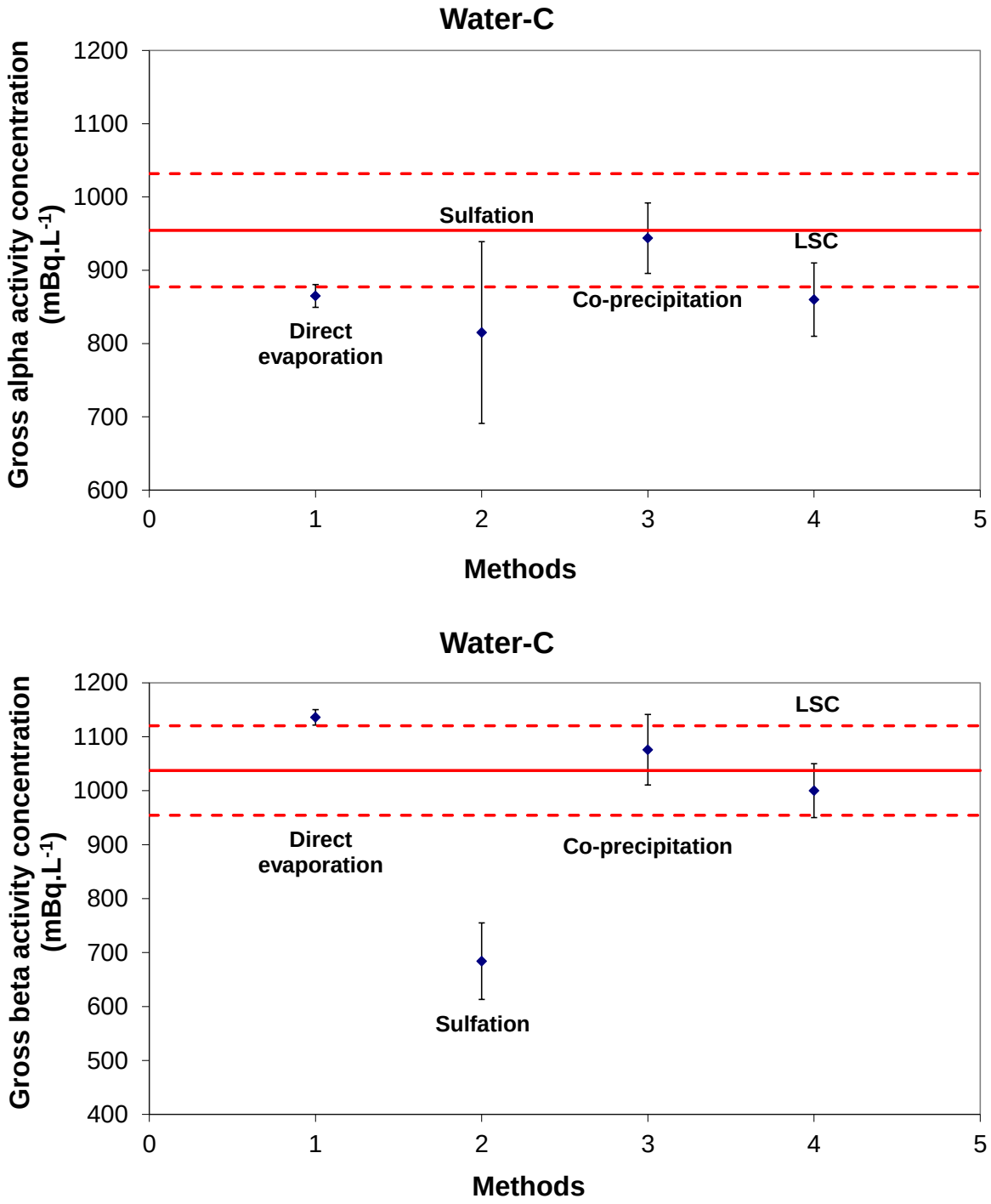


Fig. 17. Gross alpha and gross beta activity concentrations in Water C samples, obtained from spiking. Activity concentration values are given with their combined uncertainties (k=1). Red solid line shows the mean activity concentration and red dashed line shows the upper and lower values of combined measurement uncertainty of mean activity concentration (k=1).

As it is seen, all the four methods are within the reference range for gross alpha activity, while for gross beta three of them provided results within the reference range.

One of the main reasons of the deviation of the sulfation method can be the different behaviour (solubility and precipitation procedure) of americium from calcium and the alkali earth metals. Americium may not precipitate in the same way as calcium; it may stay on the surface of the $CaSO_4$ particles, so alpha particles are attenuated but not completely. However, for strontium, since it is a member of the same group as calcium (alkali earth metals) it follows the same precipitation route so strontium forms with calcium a sulfate co-precipitate. This phenomenon may result in a situation when a considerable fraction of strontium is inside the crystal lattice. Therefore, the emitted beta particles are absorbed in the crystal itself meaning that higher degree of self-absorption may occur.

The reference activity concentrations summarized in Table 17 are the average of the considered laboratory results except Water C, where the activity concentrations were calculated on the basis of weighing the spikes.

Table 17. The reference activity concentration values (A_{ref}) in the ILC water samples with their expanded uncertainties (U_{ref}) with a coverage factor k = 2.

	Reference values with expanded uncertainty (A_{ref} ± U_{ref}; mBq/L)		
	Water A	Water B	Water C
Gross α activity	48 ± 23	435 ± 57	955 ± 77
Gross β activity	310 ± 57	190 ± 33	1037 ± 83

The total dissolved solid (TDS) content of the two natural mineral waters (Water A and B) was indicated by the manufacturer without uncertainties, while in case of water C it was calculated from weighing of inorganic salts and the water (see chapter 2.2).

However, we also measured the total dissolved solid (TDS) content of the ILC water samples by direct evaporation of10-100 mL aliquot of water sample. The JRC-Geel experimental results and the manufacturers` values are presented in **Table 18**.

Table 18. Indicative values of the total dissolved solid (TDS) content of the ILC water samples.

	Water A	Water B	Water C
JRC-Geel values	955 ± 44	364 ± 27	10.2 ± 0.1 10.5 (weighing)
Manufacturers` value	1200	385	

3 Evaluation of results

Initially the results from the ILC participants were tested for outliers. However, the outlying values were not discarded and were included in the further evaluations. The presence of statistical outliers among the reported results was investigated using Grubb's test at a level of significance α = 1 %, as suggested in ISO/IEC 5725-2 (1994).

Water A

In the dataset of gross alpha activities, the first run of the Grubb's test identified one outlier (lab 66). In the second and third run laboratories 57, 36 and 53, respectively, were tagged as outliers. All four extreme results were overestimated in comparison to the average of reported values.

For gross beta activity, three results were indicated as outliers by the Grubb's test. The first run of the Grubb's test identified one outlier (lab 2). In the second and third run laboratories 3 and 60, respectively, were tagged as outliers.

Water B

In the dataset of gross alpha activities no outliers were detected. While four results were identified as outliers among the gross beta activity results: lab 2 after the first run and laboratories 34, 63 and 58 after the second and third runs.

Water C

In the dataset of gross alpha activities, the first run of the Grubb's test identified one outlier (lab 43). In the second and third run laboratories 66, 58 and 30, respectively, were tagged as outliers. All four extreme results were overestimated in comparison to the average of reported values.

For gross beta activity, also four results were indicated as outliers by the Grubb's test. The first run of the Grubb's test identified two outliers (lab 15, 53). In the second and third run laboratories 58 and 2, respectively, were tagged as outliers.

The next evaluation of the ILC was based on the raw results including outlier results where the arithmetic average was calculated together with median and standard deviation of the results. The reported minimum and maximum results together with the number of reported results are presented in Table 19.

Table 19. EC interlaboratory comparison on gross alpha/beta - raw results.

	Activity concentration (mBq/L)					Number of results
Water A	average	median	SD	minimum	maximum	
Gross alpha	88	57	131	0.81	824	44
Gross beta	370	318	291	0.633	1930	64
Water B						
Gross alpha	571	463	386	5.0	1729	70
Gross beta	392	296	331	0.828	1720	64
Water C						
Gross alpha	954	855	768	63.2	5846	66
Gross beta	1073	976	684	1.379	4331	64

3.1 Scores and evaluation criteria

The results of the participating laboratories were evaluated against the reference values using relative deviation only. One of the reasons for not using other scoring criteria is that the analyte is unclear and difficult to define in case of gross alpha/beta activities. In most cases, this includes groups of radionuclides and not well defined single radionuclide.

3.1.1 Relative deviations

The evaluation of the measurement results is based on their relative deviation from the reference value (Formula 4). Relative deviations (percentage differences in ISO/IEC 13528 (ISO13528, 2005) are calculated as

$$D_{\%} = 100 \times \frac{A_{lab} - A_{ref}}{A_{ref}} \qquad (4)$$

where,

- A_{lab} is the participant's result;
- A_{ref} is the reference value.

These values are plotted in ascending order in deviation charts and the laboratories reporting too low or too high values become more visible. The criterion $|D_{\%}| < 30$ % is used for acceptance. In principle, this is an arbitrarily chosen level, but based on the perception that, at least, routine gross alpha/beta analysis is achievable within this level of deviation.

4 Methods used by the participating laboratories

Besides sending the measurement results, laboratories submitted answers to a questionnaire giving details of their laboratory and routine procedures.

The participating laboratories perform numerous gross alpha/beta measurements per year. Out of the 71 laboratories, 17 laboratories perform less than 25 measurements per year, 22 laboratories perform 25-100 measurements and 32 perform more than 100 measurements per year. The total number of gross alpha/beta measurements performed by the participant laboratories is estimated to be between 4000 and 10000 individual measurements.

From the questionnaire it turned out that 65 laboratories work according to a quality system (mainly ISO 9000 and ISO 17025) and 58 laboratories are either accredited, authorised, certified or have a combination of these three. In 65 laboratories, the same routine analytical procedure was used for the ILC samples as for their regular routine samples.

Table 20. Quality system in the participating laboratories.

Quality system	Number of labs
ISO 17025	45
ISO 9000	2
ISO 9000 and ISO 17025	10
ISO 17025 and other	5
Other	2
No	6

The amount of water used for the preparation of a single measurement sample ranged from 5 mL up to 5 L. Details on the sample preparation and measurement techniques are presented in **Table 21**. The measurement time ranged from 1800 s to 3 days. For the counting efficiency calibration the following radionuclides were used: ^{241}Am, U_{nat}, ^{239}Pu, ^{226}Ra, ^{210}Po, ^{236}U for alpha; and ^{40}K, ^{90}Sr/^{90}Y, ^{36}Cl, ^{137}Cs, ^{210}Pb, ^{14}C, ^{3}H for beta. These radionuclides cover a wide alpha/beta energy range (18.6 keV – 1175.6 keV). Furthermore, one laboratory reported to use ^{226}Ra for beta calibration.

Table 21. Number of laboratories for sample preparation and measurement techniques used for determining the gross activities.

Sample preparation method	Number of laboratories
Evaporation to complete dryness	36
Evaporation and mixing with LSC cocktail	16
Evaporation to complete dryness, Coprecipitation	7
Other*	4
Coprecipitation	3
Evaporation to complete dryness, Other*	3
Measurement technique	**Number of laboratories**
Proportional counter	42
Liquid scintillation counter	22
Scintillation counter (solid)	10
Semiconductor Si detector	2
i-Matic Si-det	1
Grid ionization chamber	1

*Category "other" not specified by the participants.

The most used sample preparation method was evaporation to dryness with no further sample treatment. The second most used method was by evaporation (thermal pre-concentration) of an aliquot of the sample to a smaller volume and by mixing it with LSC cocktail. Co-precipitation was applied in ten cases and other techniques were used by seven laboratories.

Among the 49 participants who used other techniques than LSC, 20 laboratories answered "yes" and 29 "no" to the question if they have a procedure for hygroscopic residue. These 49 laboratories deposit the residue onto the planchet in many different ways as listed, like automatic evaporation, residue homogenisation with a solvent, evaporation of the last few mL on the planchet, direct evaporation on filter paper, direct evaporation and mechanical homogenisation.

The most popular measurement techniques were proportional counting, LSC and solid state scintillation counting. Few laboratories applied some non-conventional gross counting like semiconductor Si detector, i-Matic Si-det and grid ionization chamber.

In the case of LSC, the following sample to cocktail ratios was used: 1:4, 2:3, 1:21, 1:3 and 2:1. Only five out of the 21 laboratories using LSC applied quench correction. The type of LSC vials used were: polyethylene (used by 10 laboratories), Teflon coated (9), low potassium glass (1), glass (1) and other (1). One of the laboratories used two different vials. The procedures for the determination of background used by the participant laboratories are summarised in **Table 22**.

Table 22. Procedures used for gross alpha/beta background determination by the participants.

Background determination procedure	Number of laboratories
Empty planchet	35
Blank samples	7
Acidified water + LS cocktail	5
Distilled water + LS cocktail	5
ZnS(Ag) powder	3
Background sample in nearly the same chemical composition as the water sample	2
$CaSO_4$ spread on planchet	1
Filter paper on a planchet	1
Acidified water + Radon removal + LS cocktail	1
No definite answer	11

As seen, there are nine different approaches for the background determination which may be a reason for biased results. Moreover, there were 11 laboratories that did not provide definite answers but we assume that they might have used one of the nine background determination approaches. Comparing the gross alpha/beta detection limits with the detection limits given in the new drinking water directive (**Table 23**) one can see there are laboratories not complying with the requirements.

Table 23. Limit of detection of gross alpha/beta activity concentrations reported by the participant laboratories in mBq L^{-1}.

Limit of detection reported by the participants (mBq L^{-1})	
Gross alpha	Gross beta
1.4 - 340	0 - 424
Limit of detection (mBq L^{-1}) from the new drinking water directive (EC, 2013)	
40	400

5 Reported results

In total, 404 results were reported including values below the detection limit. Most results were reported as single results with expanded uncertainties which were directly taken into account for further evaluation. For the evaluation expanded uncertainty with coverage factor k = 2 was used. If a laboratory reported its result with different coverage factor, then the result was corrected for k = 2.

The 71 registered participant laboratories were requested to determine the gross alpha and beta activity concentration of three different water samples. This means that each participant could submit maximum six independent measurement results with their corresponding expanded uncertainties.

The reported results of gross alpha and beta activity concentration are sorted in ascending order in **Fig. 18, 20, 22, 24, 26** and **28**. The error bars represent expanded uncertainties (k = 2) and solid red line represents the reference value, while dashed red lines represent the corresponding expanded uncertainties. Laboratories' codes are indicated with the results. In **Appendix 8** tables with all reported and averaged values are presented. In the odd numbered figures (**Fig. 19, 21, 23, 25, 27, 29**), the relative deviation is plotted in ascending order. Green colour means less than 30% deviation from the reference value.

Water A

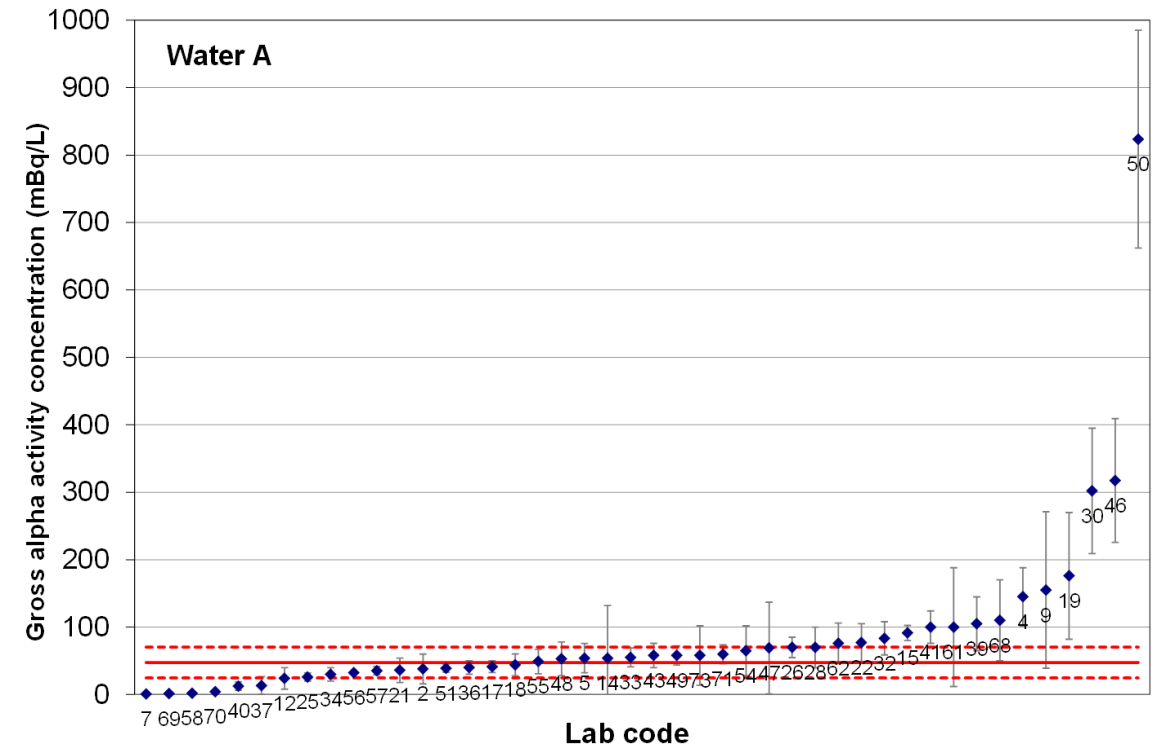


Fig. 18. Laboratory results for gross alpha activity concentration for Water A. The solid red lines indicate the reference activity concentrations (A_{ref}) of gross alpha activity. Their corresponding expanded uncertainties ± U_{ref} (k = 2) are plotted in dashed red lines.

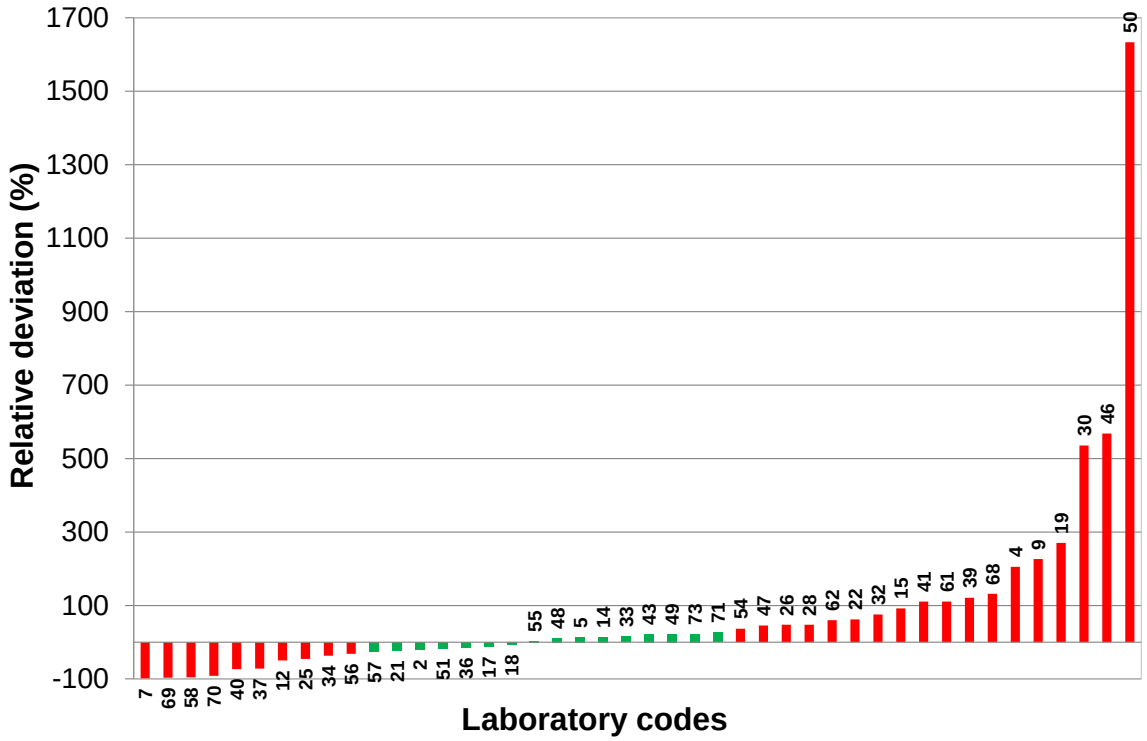


Fig. 19. Deviation chart of the participants' results of gross alpha activity concentration for Water A plotted in ascending order. Green colour indicates results within the range ± 30 % from the reference value and red indicates results outside this range. Numbers indicate the laboratory code.

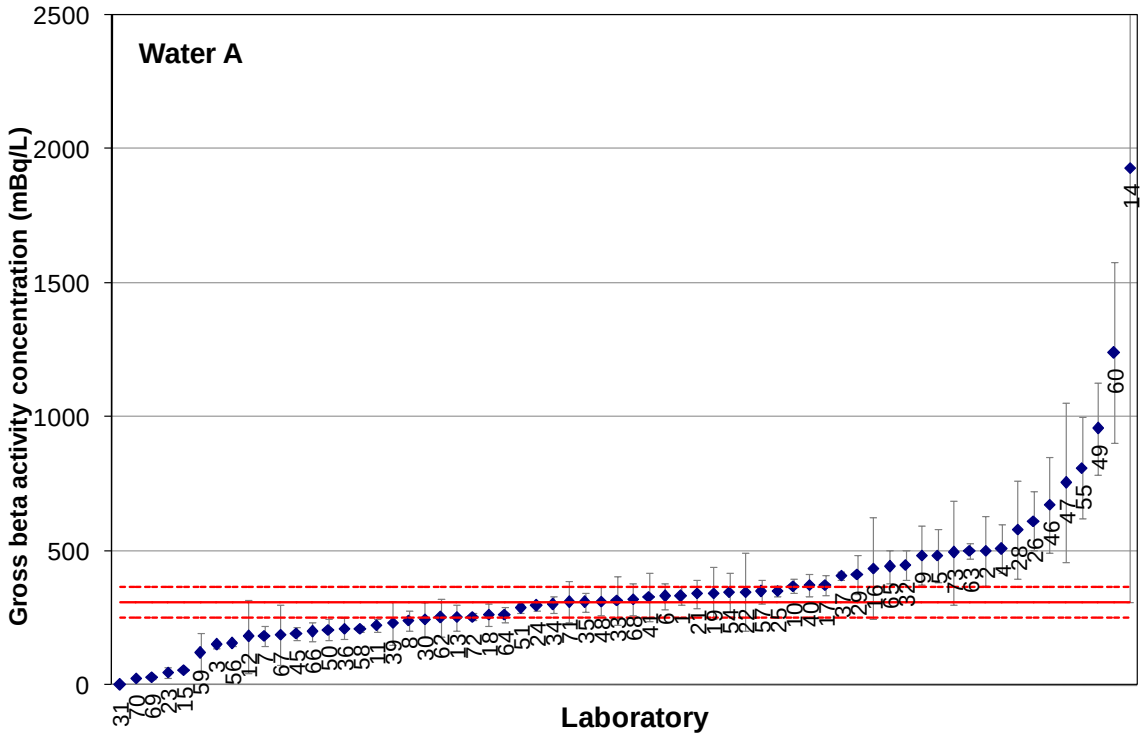


Fig. 20. Laboratory results for gross beta activity concentration for Water A. The solid red lines indicate the reference activity concentrations (A_{ref}) of gross beta activity. Their corresponding expanded uncertainties ± U_{ref} (k = 2) are plotted in dashed red lines.

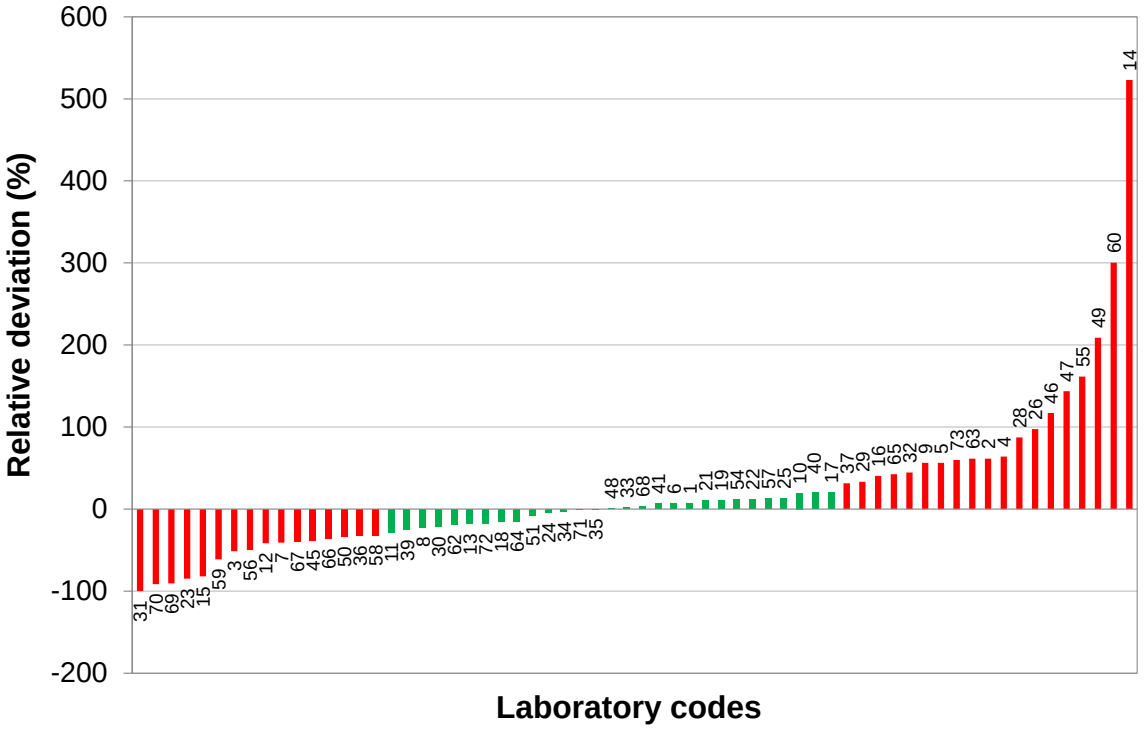


Fig. 21. Deviation chart of the participants' results of gross beta activity concentration for Water A plotted in ascending order. Green colour indicates results within the range ± 30 % from the reference value and red indicates results outside this range. Numbers indicate the laboratory code.

Water B

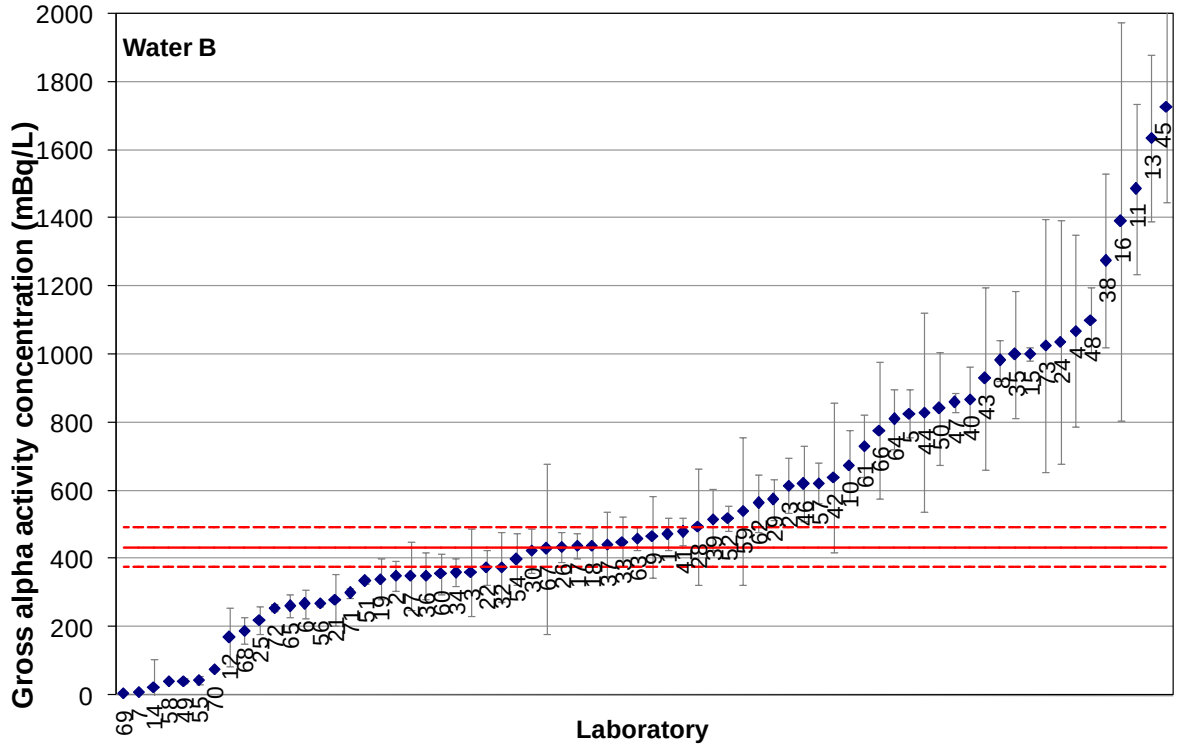


Fig. 22. Laboratory results for gross alpha activity concentration for Water B. The solid red lines indicate the reference activity concentrations (A_{ref}) of gross alpha activity. Their corresponding expanded uncertainties ± U_{ref} (k = 2) are plotted in dashed red lines.

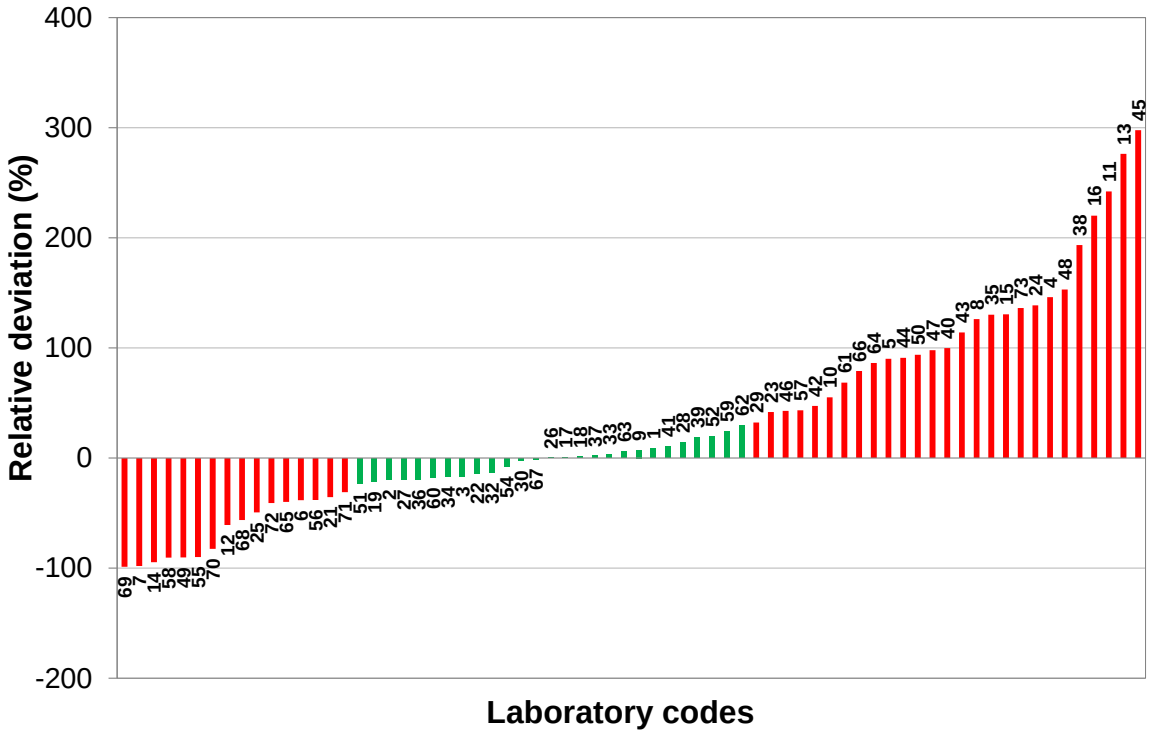


Fig. 23. Deviation chart of the participants' results of gross alpha activity concentration for Water B plotted in ascending order. Green colour indicates results within the range ± 30 % from the reference value and red indicates results outside this range. Numbers indicate the laboratory code.

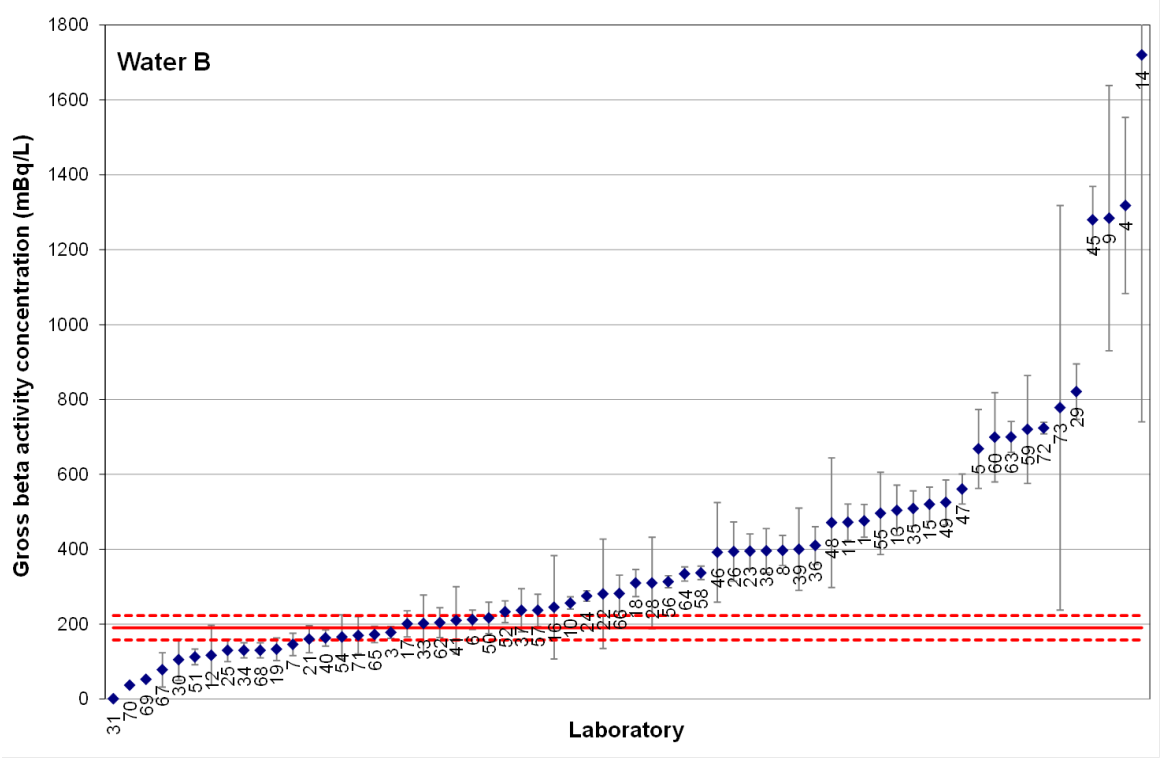


Fig. 24. Laboratory results for gross beta activity concentration for Water B. The solid red lines indicate the reference activity concentrations (A_{ref}) of gross beta activity. Their corresponding expanded uncertainties ± U_{ref} (k = 2) are plotted in dashed red lines.

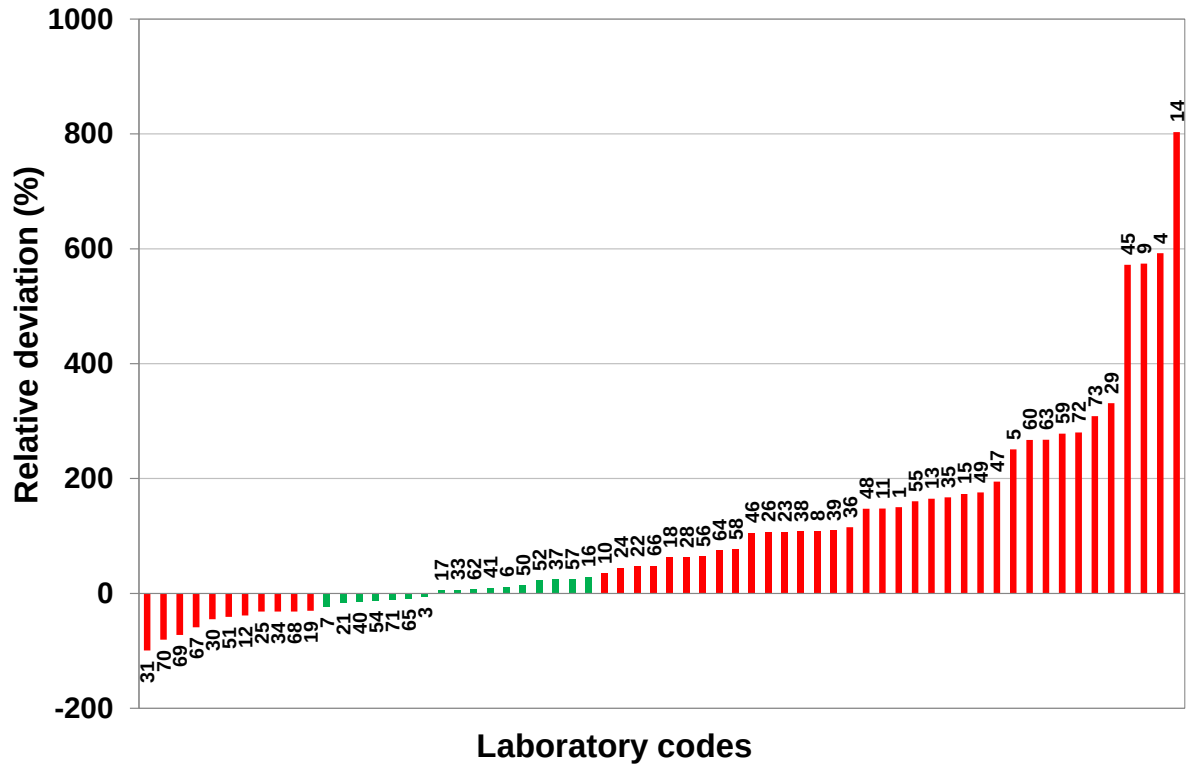


Fig. 25. Deviation chart of the participants' results of gross beta activity concentration for Water B plotted in ascending order. Green colour indicates results within the range ± 30 % from the reference value and red indicates results outside this range. Numbers indicate the laboratory code.

Water C

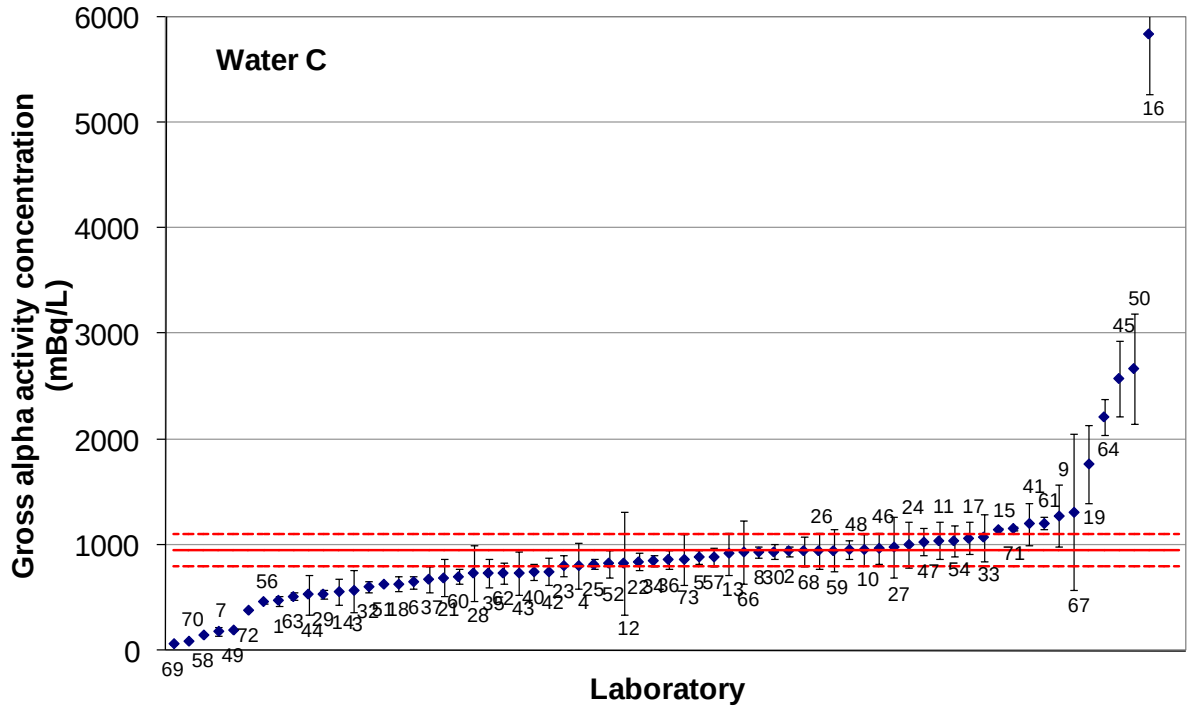


Fig. 26. Laboratory results for gross alpha activity concentration for Water C. The solid red lines indicate the reference activity concentrations (A_{ref}) of gross alpha activity. Their corresponding expanded uncertainties ± U_{ref} (k = 2) are plotted in dashed red lines.

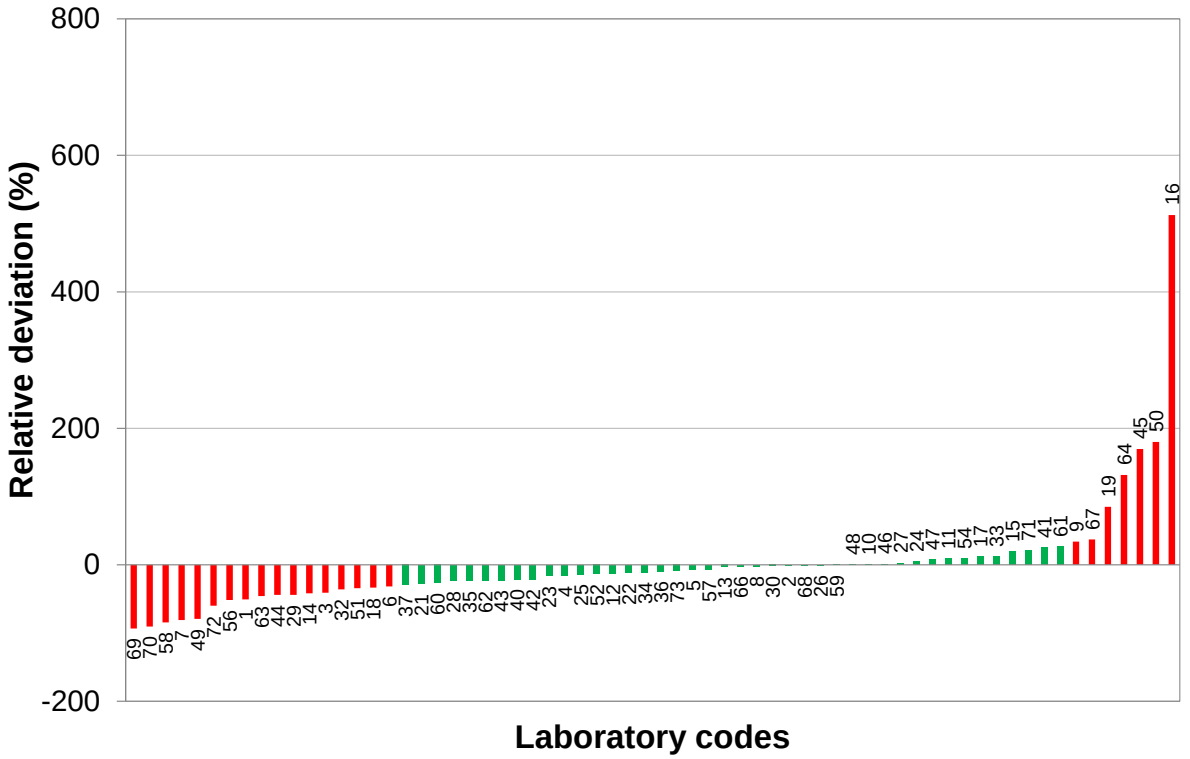


Fig. 27. Deviation chart of the participants' results of gross alpha activity concentration for Water C plotted in ascending order. Green colour indicates results within the range ± 30 % from the reference value and red indicates results outside this range. Numbers indicate the laboratory code.

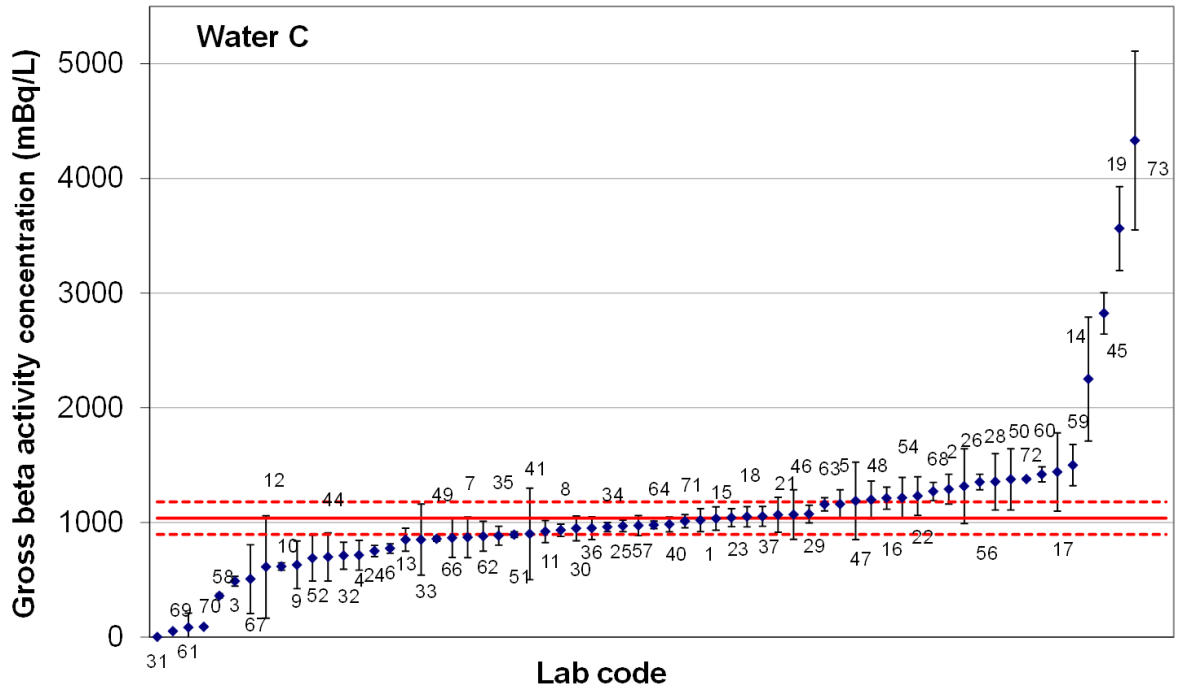


Fig. 28. Laboratory results for gross beta activity concentration for Water C. The solid red lines indicate the reference activity concentrations (A_{ref}) of gross beta activity. Their corresponding expanded uncertainties ± U_{ref} (k = 2) are plotted in dashed red lines.

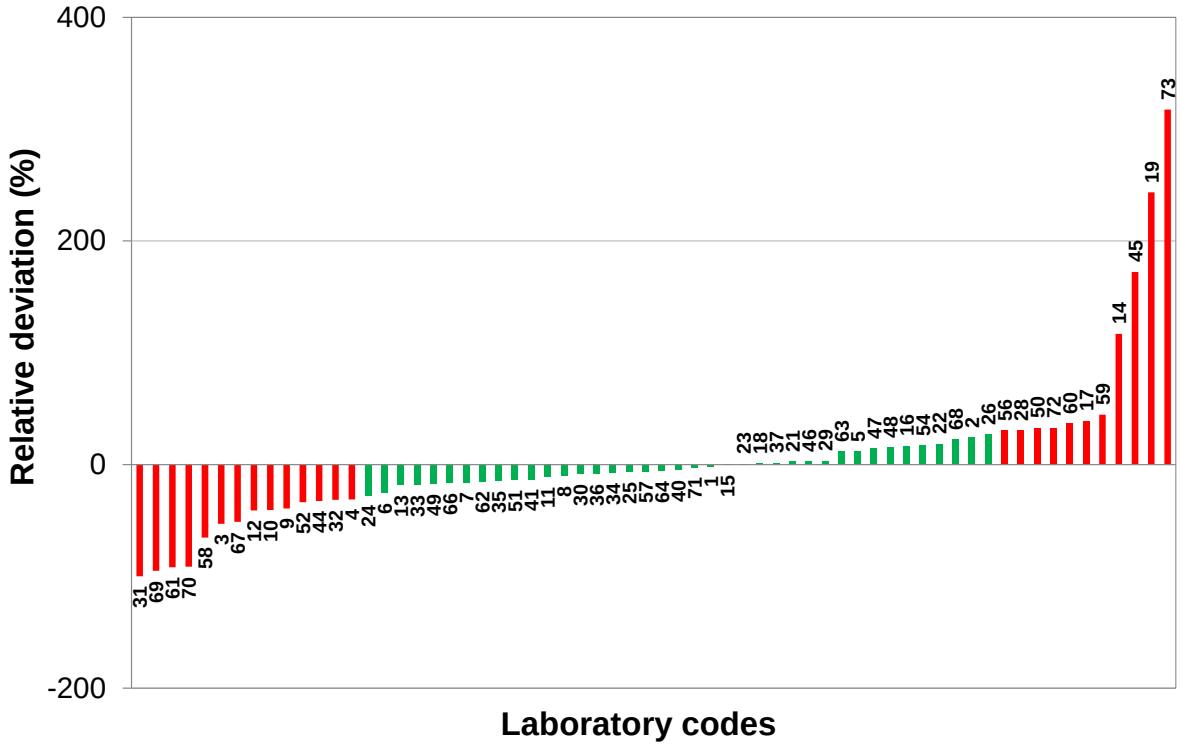


Fig. 29. Deviation chart of the participants' results of gross beta activity concentration for Water C plotted in ascending order. Green colour indicates results within the range ± 30 % from the reference value and red indicates results outside this range. Numbers indicate the laboratory code.

Many of the participants' results deviate by more than two orders of magnitude from the reference values regardless of the techniques used. It is interesting to evaluate the ratio of maximum to minimum reported gross activities (**Table 24**) and the percentage of compatible results (**Table 25**). The number of compatible results together with the number of laboratories and their identification codes are presented in **Table 26**.

Table 24. Ratio of the reported maximum to minimum gross activities.

Parameter	A_{max}/ A_{min}		
	Water A	Water B	Water C
Gross alpha activity	1017	346	93
Gross beta activity	3050	2080	3150

Table 25. Percentage of the reported results within ± 30 % from the reference value.

Parameter	Results within ± 30 % deviation (%)		
	Water A	Water B	Water C
Gross alpha activity	36	39	63
Gross beta activity	45	27	61

Table 26. Number of laboratories and their ILC identification codes versus the number of reported compatible results.

Number of compatible results	Number of laboratories	Laboratory code
6	1	33
5	7	17, 21, 41, 54, 57, 62, 71
4	11	2, 18, 22, 30, 34, 35, 36, 37, 40, 48, 51
3	11	1, 5, 6, 8, 11, 13, 24, 25, 26, 52, 68
2	20	3, 7, 10, 15, 16, 19, 23, 27, 28, 39, 43, 46, 47, 49, 59, 60, 63, 64, 66, 73
1	13	4, 9, 12, 14, 29, 32, 42, 50, 55, 61, 65, 67, 72
0	8	31, 38, 44, 45, 56, 58, 69, 70

As shown in **Table 26**, only 30 laboratories (42 %) out of 71 reported at least half of the results within the reference range and only one laboratory (Lab code: 33, LSC technique) could succeed to be within this 30% in each of the six cases. Furthermore, 8 laboratories (11 %) did not report compatible result at all. Of the 30 laboratories with at least 3 compatible results, only six laboratories used solid scintillation counting (21, 57, 62, 34, 48, 51) and 21 laboratories applied proportional counting (21, 54, 57, 62, 71, 18, 30, 34, 35, 36, 37, 40, 48, 51, 1, 6, 8, 11, 13, 24, 25) and nine laboratories used liquid scintillation counting (33, 17, 41, 2, 22, 5, 26, 52, 68).

It has to be mentioned that six laboratories used multiple techniques: scintillation counting and proportional counting technique for gross alpha and gross beta measurement respectively. Among the 30 best performing laboratories one could say that proportional counting technique was the most popular. However, we cannot find any of the methods to be superior to the other methods.

As is evident from the reported results (**Figures 18** to **29**), the outcome of the laboratory comparison exercise is far from satisfactory. The measurement results span a wide range, e.g. for Water C the maximum reported gross beta activity was more than 3000 times higher than the minimum reported gross beta activity.

Furthermore, several laboratories (for example laboratories 49 and 50) present for one type of sample a measurement result several times higher than the reference, whilst for another type of sample the same laboratory has a result several times lower.

5.1 Sorted results

During the evaluation of the ILC, results were sorted by counting technique, sample preparation, radionuclides used for calibration and the time delay between sample preparation and counting. Some of the evaluations are given in graphical form in **Fig. 30-33**.

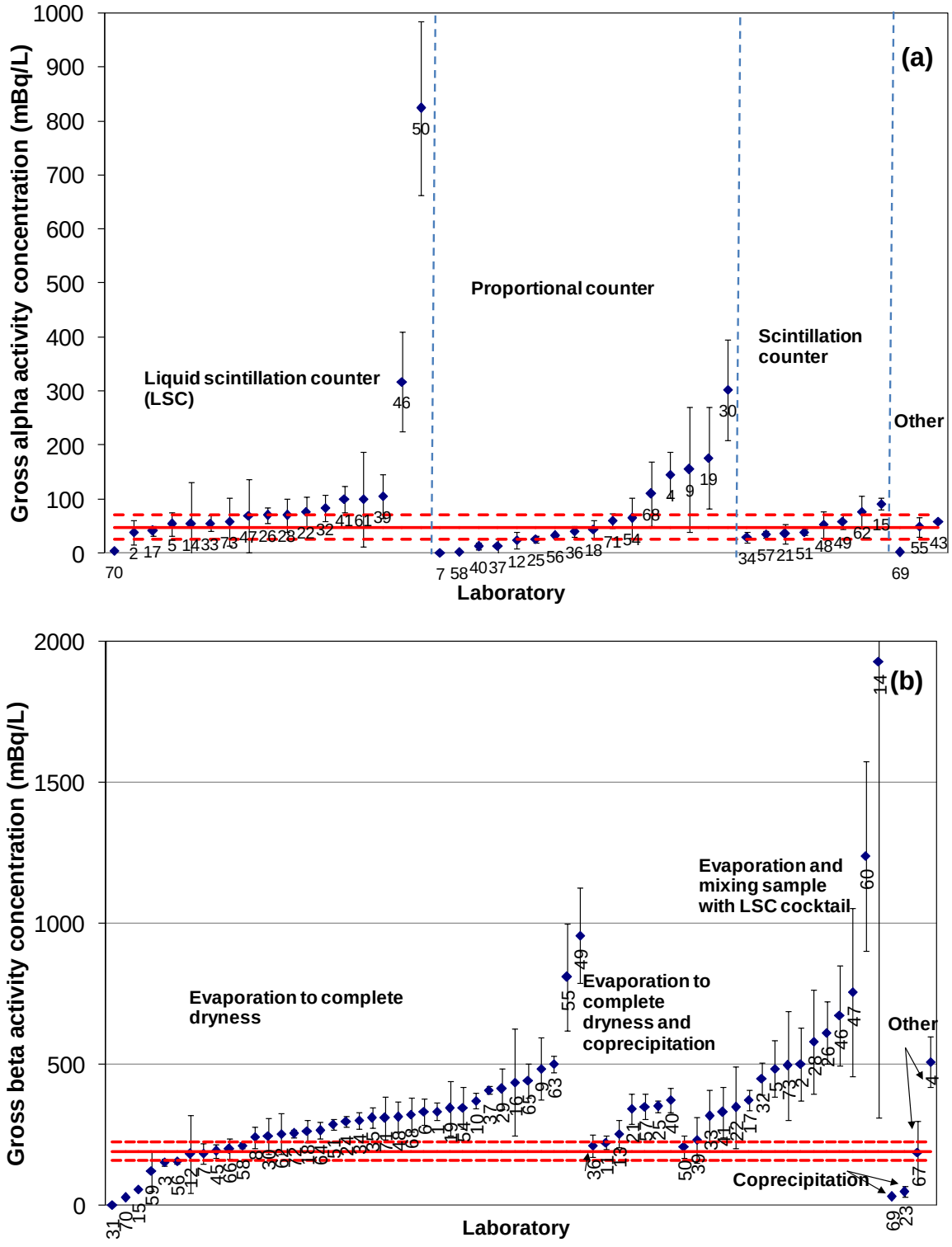


Fig. 30. Water A results sorted on the basis of (a) measurement techniques, (b) sample preparation used. Numbers on the plots indicate the laboratory code.

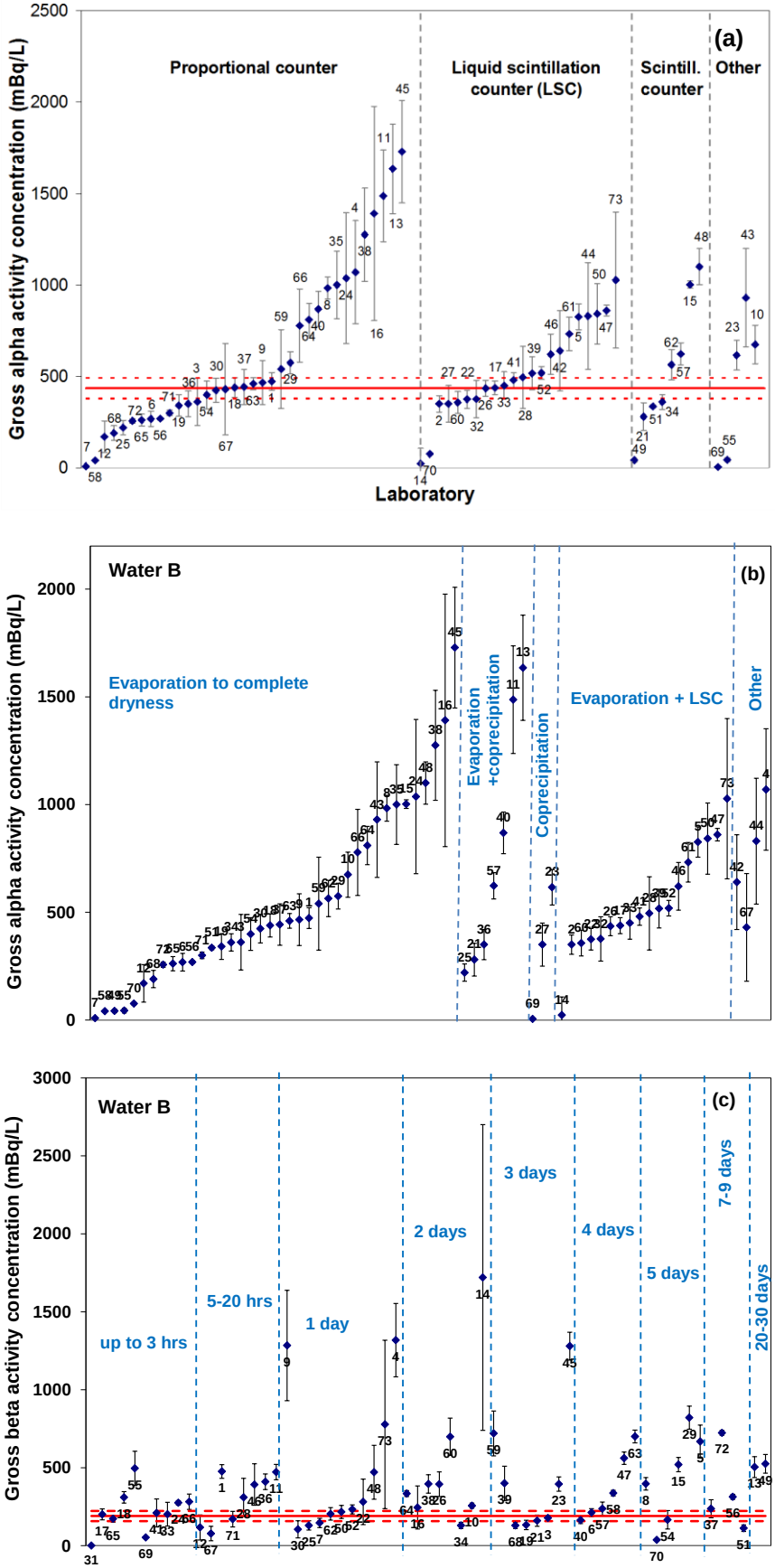


Fig. 31. Water B results sorted on the basis of (a) measurement techniques, (b) sample preparation used and (c) time delay between sample preparation and measurement. Numbers on the plots indicate the laboratory code.

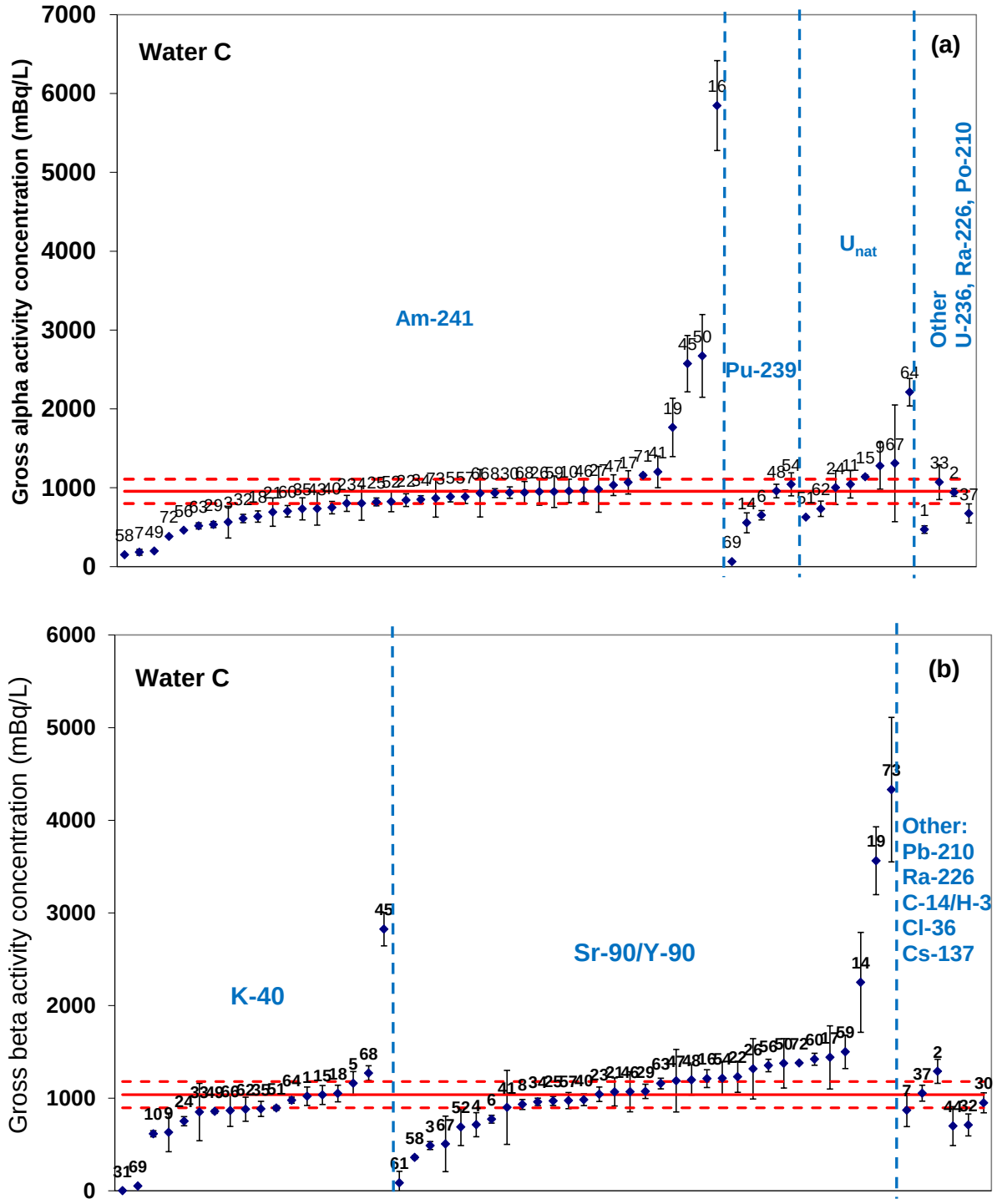


Fig. 32. Water C results sorted on the basis of the radionuclides used for (a) alpha and (b) beta counting efficiency calibration. Numbers on the plots indicate the laboratory code.

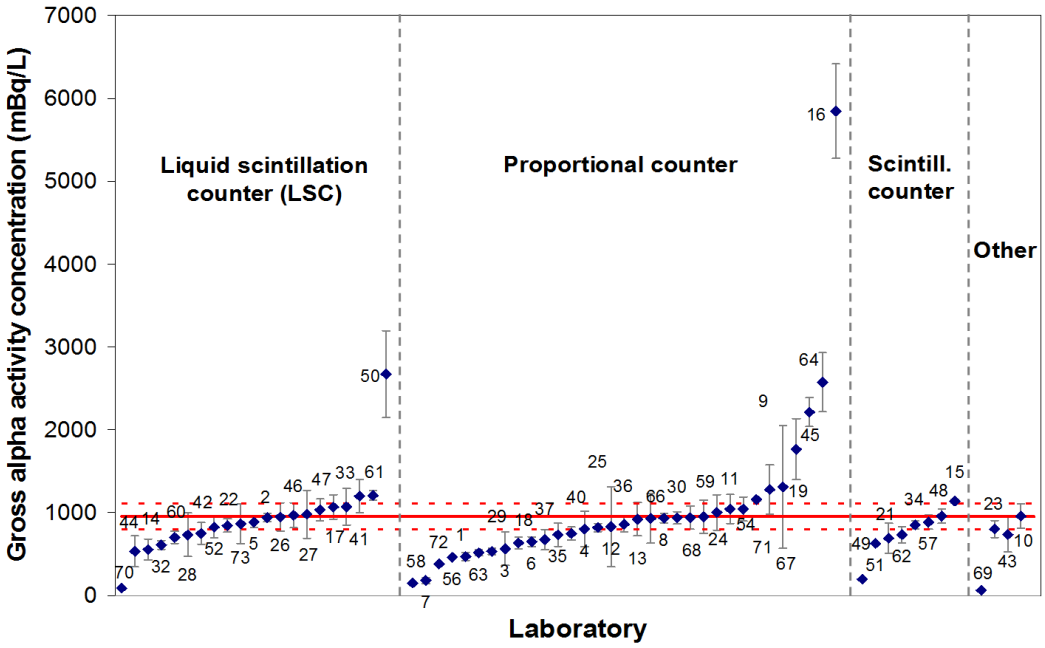


Fig. 33. Water C results sorted on the basis of measurement techniques. Numbers on the plots indicate the laboratory code.

Comparing the different groups of sorted results, no significant differences between those groups are observed. However, for some groups the available data are limited (e.g. for group "Other"). It is worth to mention that laboratories using the same radionuclide for calibration, added as spike in Water C, did not perform better than the laboratories using other radionuclides. Additionally, laboratories were requested to determine the total dissolved solid (TDS) content of each water samples. The reported results showed also large scatter of data just like the gross measurement results. The most likely reasons for this can be the insufficient sample size (maximum 50 mL was instructed by the organizers due to sample availability), hygroscopic residues and the incorrect use of decimals. The full list of TDS results are presented in **Appendix 8**.

5.2 Participants' feedback

Within the questionnaire and the EURATOM workshop held in October 2013 participants had the opportunity to comment the ILC, share experience and express their difficulties with the measurements.

One comment was about the total dissolved solid content of Water C which labs found too low. The reason for such a low dissolved solid content was to avoid the self-absorption from the sample itself if one used direct evaporation.

Several participants expressed their interest to participate also in future water ILCs. Furthermore, they would welcome samples with various types of matrices. They expressed appreciation for this kind of exercises with the emphasis on the possible future improvement and improvement of their measurement routines. Other laboratories mentioned that workshops on ILCs should be organized more often or very soon after the ILC and be combined with a short training event. There were comments about too many questions in the questionnaire but we believe that all those questions were necessary for the ILC evaluation and to enable an understanding of the problems that can be encountered. The overall aim is to improve measurements in the EU and the participants' feedback is very important to achieve that goal.

6 Conclusions

As presented above, only 42 % of the labs reported at least three of the six results within the reference range, while 11 % reported incompatible results only. None of the methods used by participants was proven to be superior to the others. Even application of the same method in different laboratories does not guarantee comparable results. Considering the number of individual gross measurements in a year, the ability of those laboratories to provide consistent and reliable results will directly influence the implementation of the drinking water directive. Furthermore, the decisions made on the basis of the measurement results have health and economic impacts as well.

The present situation is far from satisfactory knowing that these screening methods are very likely to be used for testing drinking water as is foreseen by the drinking water directive (EC, 2013) and will lead to different decisions seen the large spread in the data. The large spread of the results may be due to influencing factors during both the sample preparation and the measurement process. These influences cannot generally be predicted and it is already difficult to define the measurand for gross activity analysis since the radionuclide composition of the sample is *a priory* not known.

Additionally, the activity of the sample may substantially change with time as some radionuclides decay and others grow in during the measurement time. Furthermore, for drinking waters a few decay processes are very likely to occur and should be considered in the measurement and data analysis process.

For these reasons, revision of the written standards for gross methods is needed as proposed by Jobbágy et al. (2014). We recommend (i) following strictly accepted common procedures for sample preparation and measurement, (ii) to be aware of all decay processes that may affect the measurement, (iii) to test procedures for robustness and (iv) to establish realistic uncertainty budgets.

The outcome of the analysis is certainly also influenced by the proficiency and skills of laboratory personnel. At least in two European countries (Austria and Switzerland) no gross methods are used for drinking water qualification due to their drawbacks and unreliability. As long as gross activity parameters are included in the European drinking water directive, this interlaboratory comparison should be repeated with pre-defined guideline procedures to be followed by the participants.

6.1 Sources of interferences

The most probable reason for such spread of the reported results is that there are many variables to control from the sample preparation until the sample counting. There are numerous pitfalls and sources of interferences of the gross alpha/beta methods as discussed in a journal article by Jobbágy et al. (2014). A few examples of influential parameters are listed here: (i) sample preparation methods (loss of volatile radionuclides), (ii) time delay between sample preparation and measurement (ingrowth of radon and its progenies), (iii) detection technique and (iv) calibration source energy distribution.

There are also method specific pitfalls such as in LSC, if inappropriate quench correction or alpha/beta discrimination is used. Since ^{40}K is not co-precipitated it is excluded from the gross beta results, whereas if direct evaporation or LSC is used then ^{40}K contributes to the gross beta activity. Issues with co-precipitation are related to the uncertainties with chemical yield and ^{40}K. During the direct evaporation approach source matrix and uniformity, surface density (i.e. self absorption) and hygroscopic sample play important roles.

In the light of the many combinations of parameters that are possible and that might vary from one laboratory to another, it is not unexpected that results show such a

spread. These findings underpin the importance of fixing as many parameters as possible and making stricter gross alpha/beta measurement standards.

Human factors also contributed to the non-compatible results but to a lesser extent. Namely, there were some discrepancies with reporting results: some labs gave higher limit of detection values than the reported activities. It turned out that some laboratories introduced decimals to the wrong place which led to incompatible results and some just might have not paid attention to the requested result format. Attention has to be paid on the decimals and units when results are submitted.

6.2 True standardization of methods

The main reason for unreliability of gross methods and the non-comparability of gross alpha/beta measurement results is the lack of knowledge of the real radionuclide composition of the water. Since there are many variables playing a key role in gross measurement (Jobbágy et al., 2014), it is important to fix as many parameters as possible. The radionuclides used in the calibration, the geometry of the source, quenching parameters, the chemical form and any time delay must be clearly defined. Acceptable time delays for each step (e.g. between sampling and sample preparation, source preparation and start of measurement) must be set in the written measurement standards, which is particularly important when ^{226}Ra- and ^{224}Ra-containing waters are analysed.

Replacing gross methods of drinking water analysis with radionuclide-specific methods would not take a lot of effort or expense. Radionuclide measurements can be done using the same instrumentation (gas-flow proportional counter, liquid-and solid scintillation counter) that are used for gross alpha/beta analysis. However, more expertise/proficiency and validated methods are needed, but in a routine radiochemistry laboratory they should already exist.

6.3 Recommendations for the gross alpha/beta method applied to drinking water analysis

Gross methods are far from accurate and in some cases they fail to determine certain radionuclides, they give only an "activity index" rather than an approximate activity concentration, as explained by Schönhofer (2012) and confirmed by the data spread from this laboratory comparison. The difference between laboratory results in this ILC is sometimes two or three orders of magnitude, which is far beyond the measurement uncertainties. These findings lead us to conclude that gross alpha/beta methods are not fit to be used as an independent method to assess activity concentration. Gross measurement should be used for monitoring only after the radionuclide composition is known from radionuclide specific analysis of representative samples. It can be used as a complementary or substitute method for radionuclide-specific measurement only with important restrictions:

1) no temporary change is expected in the radiochemical composition (no significant ingrowth of progenies during the measurement and changes in the monitored water itself during short periods of time),
2) no complex decay chains are present,
3) a true standardized method is used, and
4) the measurement parameters are fixed.

Radionuclide specific analysis should be repeated on a regular basis in accordance with the drinking water directive concerning check and audit monitoring (EC, 1998). Any suspected change in parameters requires more frequent nuclide specific analysis.

6.4 Future actions

After the evaluation of the 2012 gross alpha/beta activity in water ILC, DG-ENER and JRC-Geel discussed the repetition possibilities of the interlaboratory comparison. Furthermore, the proposal was supported by the participants during the 2013 ILC workshop. In the course of the EURATOM article 35-36 meeting in March 2016 a decision was taken on the repetition of this exercise which would be announced in the near future. Taking into account the other requests from the Article 35 Expert group on JRC-Geel to perform proficiency tests of member states' capacity to measure various radionuclides in environmental samples ("emergency radionuclides" in feed, radon in water), it is most probable that the next gross alpha/beta activity in water will take place in 2019 or 2020.

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List of abbreviations and definitions

AC	accession countries
A_{lab}	mean laboratory result of activity concentration
A_{ref}	reference value of activity concentration
BIPM	Bureau International des Poids et Mesures
CCRI(II)	Comité Consultatif des Rayonnements Ionisants, Section 2
D	Relative deviation between the reported and the reference activity concentration
EURATOM	European Atomic Energy Community
GUM	Guide to the Expression of Uncertainty in Measurement
HPGe	high-purity germanium detector
IRMM	Institute for Reference Materials and Measurements (since 1 July 2016 JRC-Geel)
ILC	interlaboratory comparison
ISO	International Organization for Standardization
k	coverage factor according to GUM
LOD	limit of detection
LSC	liquid scintillation counting
MS	member states of the European Union
RIVM	Rijksinstituut voor Volksgezondheid en Milieu (Laboratory for Radiation Research)
SCK•CEN	Studiecentrum voor Kernenergie Centre d'Etudes Nucleaire (Belgian Nuclear Research Centre)
SI	Système International d'Unités, International System of Units
SIR	Système International de Référence, International Reference System for radionuclides
u	standard uncertainty according to GUM
u_c	combined standard uncertainty according to GUM
U	expanded uncertainty according to GUM
U_{lab}	expanded uncertainty of average laboratory result
U_{ref}	expanded uncertainty of reference value

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upper and lower values of combined measurement uncertainty of mean activity concentration (k=1).

Fig. 17. Gross alpha and gross beta activity concentrations in Water C samples, obtained from spiking. Activity concentration values are given with their combined uncertainties (k=1). Red solid line shows the mean activity concentration and red dashed line shows the upper and lower values of combined measurement uncertainty of mean activity concentration (k=1).

Fig. 18. Laboratory results for gross alpha activity concentration for Water A. The solid red lines indicate the reference activity concentrations (A_{ref}) of gross alpha activity. Their corresponding expanded uncertainties ± U_{ref} (k = 2) are plotted in dashed red lines.

Fig. 19. Deviation chart of the participants' results of gross alpha activity concentration for Water A plotted in ascending order. Green colour indicates results within the range ± 30 % from the reference value and red indicates results outside this range. Numbers indicate the laboratory code.

Fig. 20. Laboratory results for gross beta activity concentration for Water A. The solid red lines indicate the reference activity concentrations (A_{ref}) of gross beta activity. Their corresponding expanded uncertainties ± U_{ref} (k = 2) are plotted in dashed red lines.

Fig. 21. Deviation chart of the participants' results of gross beta activity concentration for Water A plotted in ascending order. Green colour indicates results within the range ± 30 % from the reference value and red indicates results outside this range. Numbers indicate the laboratory code.

Fig. 22. Laboratory results for gross alpha activity concentration for Water B. The solid red lines indicate the reference activity concentrations (A_{ref}) of gross alpha activity. Their corresponding expanded uncertainties ± U_{ref} (k = 2) are plotted in dashed red lines.

Fig. 23. Deviation chart of the participants' results of gross alpha activity concentration for Water B plotted in ascending order. Green colour indicates results within the range ± 30 % from the reference value and red indicates results outside this range. Numbers indicate the laboratory code.

Fig. 24. Laboratory results for gross beta activity concentration for Water B. The solid red lines indicate the reference activity concentrations (A_{ref}) of gross beta activity. Their corresponding expanded uncertainties ± U_{ref} (k = 2) are plotted in dashed red lines.

Fig. 25. Deviation chart of the participants' results of gross beta activity concentration for Water B plotted in ascending order. Green colour indicates results within the range ± 30 % from the reference value and red indicates results outside this range. Numbers indicate the laboratory code.

Fig. 26. Laboratory results for gross alpha activity concentration for Water C. The solid red lines indicate the reference activity concentrations (A_{ref}) of gross alpha activity. Their corresponding expanded uncertainties ± U_{ref} (k = 2) are plotted in dashed red lines.

Fig. 27. Deviation chart of the participants' results of gross alpha activity concentration for Water C plotted in ascending order. Green colour indicates results within the range ± 30 % from the reference value and red indicates results outside this range. Numbers indicate the laboratory code.

Fig. 28. Laboratory results for gross beta activity concentration for Water C. The solid red lines indicate the reference activity concentrations (A_{ref}) of gross beta activity. Their corresponding expanded uncertainties ± U_{ref} (k = 2) are plotted in dashed red lines.

Fig. 29. Deviation chart of the participants' results of gross beta activity concentration for Water C plotted in ascending order. Green colour indicates results within the range ± 30 % from the reference value and red indicates results outside this range. Numbers indicate the laboratory code.

Fig. 30. Water A results sorted on the basis of (a) measurement techniques,(b) sample preparation used. Numbers on the plots indicate the laboratory code.

Fig. 31. Water B results sorted on the basis of (a) measurement techniques,(b) sample preparation used and (c) time delay between sample preparation and measurement. Numbers on the plots indicate the laboratory code.

Fig. 32. Water C results sorted on the basis of the radionuclides used for (a) alpha and (b) beta counting efficiency calibration. Numbers on the plots indicate the laboratory code.

Fig. 33. Water C results sorted on the basis of measurement techniques. Numbers on the plots indicate the laboratory code.

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Appendix 1: Invitation letter

Ref. Ares(2012)870955 - 17/07/2012

EUROPEAN COMMISSION
JOINT RESEARCH CENTRE

Institute for Reference Materials and Measurements (Geel)
Nuclear physics

Geel, 17 July 2012
JRC.D.04/UW/mvdl/ARES

Mrs	V. Borbala (RO)	M. Skotniczna (PL)	Mr	K. Andersson (DK)	J. Lahtinen (FI)
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	U. Portius (DE)			P. Demetriades (CY)	G. Torri (IT)
	Ch. Poynton (UK)			C. Katzlberger (AT)	A. Vincze (HU)
	E. Simion (RO)			J. Kuhlen (DE)	M. Yeomans (MT)
				P. Kwakman (NL)	

Subject: Articles 35-36 of the Euratom Treaty
Nomination of laboratories for the EC interlaboratory comparison on gross alpha/gross beta activity measurement in drinking water

Dear colleagues,

As you know, EU Member States are obliged under Art. 35 and 36 of the Euratom Treaty (and as further specified in the Commission Recommendation 2000/473/Euratom) to inform the European Commission (EC) on a regular basis of the radioactivity levels in their environment. In order to obtain more information on the measurement methods and on the quality of the values reported by the Member States, the EC (DG JRC on request of DG ENER) organises regularly a European interlaboratory comparison exercise. The draft of the new EC directive presented under Article 31 of the Euratom Treaty – with regard to radioactive substances in water intended for human consumption - incorporates screening radioactivity measurements in drinking water. Therefore, JRC IRMM is preparing to organise an interlaboratory comparison exercise on the gross alpha/gross beta activity measurement in drinking water. The samples are planned to be sent to the participating laboratories during August and September 2012. Next, the participating laboratories are requested to send their results to JRC IRMM by 31 October 2012. The preliminary report is foreseen to be available by the end of November 2012.

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E-mail: uwe.waetjen@ec.europa.eu; jana.meresova@ec.europa.eu • Internet: http://www.irmm.jrc.be

It would be appreciated if you could investigate which laboratories in your country would be interested in participating in this exercise or which laboratories you would like to see participating. To proceed according to the plans, we require your (nationally coordinated) response **by 15 August 2012**. If this should be impossible due to the holiday period, then please send your response as soon as possible after that date.

Due to the limited availability of the sample material, the exercise must be curtailed to two laboratories per country. If you should wish to nominate more than two laboratories, exceptions might be possible depending on the total number of participants.
We request you to provide us with your nationally coordinated answer, containing the contact data (responsible person, complete mail address, telephone, telefax and e-mail) for the nominated institutes with an indication of priority for your country, if you should nominate more than 2 laboratories.

As on the occasion of this comparison, DG ENER.D.4 would like to get an overview of sampling concepts in the member states for gross alpha/beta analysis of drinking waters, we would like to announce already now that we will send a small questionnaire to you at a later time.

Looking forward to hearing from you with the laboratory nominations, I remain,

Yours sincerely,

Uwe Wätjen

c.c.: Mr. Marc De Cort (JRC ITU-Ispra)
Mr. A. Janssens, C. Gitzinger (DG ENER.D.4, Luxembourg)

Appendix 2: Registration and information letter

EUROPEAN COMMISSION
JOINT RESEARCH CENTRE

Institute for Reference Materials and Measurements (Geel)
Nuclear physics

Geel, 4 September 2012
JRC.D.4/UW/JM/mvdl/ARES

Subject: EC interlaboratory comparison on gross alpha/beta activity measurement in drinking waters

Dear colleague,

Your laboratory has been nominated by your national representative(s) or authority to participate in above mentioned interlaboratory comparison organised by JRC-IRMM. In order to provide you with an appropriate volume of water for the requested analysis, we need to know your requirements. We kindly ask you to give us the following information:

a) **the typical sample volume** (L) used for gross alpha/beta activity measurement in your laboratory,
b) **contact details** (e-mail) of the personnel involved in the gross alpha/beta analysis.

Please reply as soon as possible but no later than **10 September 2012** (by e-mail to jana.meresova@ec.europa.eu with copy to viktor.jobbagy@ec.europa.eu and uwe.waetjen@ec.europa.eu).

In the present interlaboratory comparison, the participating laboratories are requested to analyse three different water samples. It is mandatory to report the results for all three samples. Each of them contains alpha and beta emitting radionuclides with different activity concentrations (close to the limits of detection or close to the screening levels). These performance characteristics are quantified in the table below.

Parameter	Activity concentration [Bq/L]	Reference
Limit of detection	0.02-0.1	ISO 9696, ISO 9697, ISO 10704
	$\Sigma\alpha = 0.04$; $\Sigma\beta = 0.4$	EC COM/2011/0385; 83/98/EC
Screening level	$\Sigma\alpha = 0.5$; $\Sigma\beta = 1$	WHO 2011

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

The reporting of laboratory results will be done via Internet. Therefore, we kindly ask you to register your laboratory via the following WEB link:

https://web.jrc.ec.europa.eu/ilcRegistrationWeb/registration/registration.do?selComparison=960

After your registration, we will send you the password key in a separate e-mail. You will need this password key for the online reporting of your results. The description of your analytical procedures will be collected via questionnaire. We kindly ask that you answer all relevant questions regarding the procedures that you have used. Please be aware that the deadline for the registration is **15 September 2012**.

Registration instructions:

> Please fill out all your organisation and contact details. The fields with an asterisk indicator (*) are mandatory. Data must be entered in these fields; otherwise, the final registration information will not be submitted. Do not forget to click the checkbox to confirm that you have read the specific privacy policy statement and agree with the terms. The full text of the privacy statement will be shown after click on the link. When all data are entered, press the Register button. The screen on the next page summarises the information. If the data are correct, then press the Confirm button. The final screen concludes that the data have been input into the system. A new window will appear with the printout form. This screen is common to all campaigns at IRMM. You may print out the registration form for your own use. However, we do not require a copy of a signed printout of this form. If you notice after the final confirmation that your entries are wrong, you may correct them by sending to us a fax or e-mail with the corrected entries.

As soon as you are registered for the interlaboratory comparison, the samples with further instructions will be sent to the address specified in the registration form. The deadline for the results reporting and questionnaire submission is **31 October 2012**. It will be possible to report values below the detection limit. The reported results will be evaluated on the basis of the E_n numbers and the relative deviations from the reference values. Therefore, always give your measurement results with their associated uncertainties and the coverage factors.

The preliminary report of this exercise is foreseen to be available by the end of 2012. If you have further questions, please contact us. We will be happy to help you. Looking forward to your participation in this comparison.

Sincerely yours,

Uwe Wätjen
Sector Head Radionuclide Metrology

Appendix 3: Information letter and e-mail

EUROPEAN COMMISSION
JOINT RESEARCH CENTRE

Institute for Reference Materials and Measurements (Geel)
Nuclear physics

Geel, 6 September 2012
JRC.D.4/UW/JM/ARES

EC interlaboratory comparison on gross alpha/beta activity in drinking waters

Background information

EU Member States are obliged under Art. 35 and 36 of the Euratom Treaty (and as further specified in Commission Recommendation 2000/473/Euratom) to inform the European Commission (EC) on a regular basis on the radioactivity levels in their environment, in some food products and in drinking water. In order to obtain more information on the quality of the values reported by the Member States, the EC (through its Directorate General JRC) organises regularly a European interlaboratory comparison exercise.

Your laboratory has been nominated by your national representative(s) or authority to participate in the interlaboratory comparison on gross alpha/beta activity in drinking waters organised by JRC-IRMM. During the past few days, three different water samples were sent by DHL courier services to your laboratory.

Water A	in 1 L bottles
Water B (blue bottle with green cap)	in 1.5 L bottles
Water C	in 1 L bottles

Each bottle contains either 1 L or 1.5 L of the material, which is expected to be sufficient for the requested analyses. If this should not be the case, we have some additional samples of the material which we could distribute on request.

Material information

The samples contain environmental levels of alpha and beta emitting radionuclides such that the material can be transported freely and handled in the laboratory without any radiological restrictions. However, some of these waters may have gross alpha/beta activity above the screening levels. Only one of the water samples (Water C) was acidified by cc. HNO_3 where its acidity was set to pH ~ 1.2 and spiked with alpha and beta emitting isotopes.

Water A and Water C were bottled in polypropylene bottles at IRMM. The total dissolved solid (TDS) content of the material after bottling was determined but needs to be remeasured in each laboratory in order to get information on their salinity.

Reference values of the comparison samples have been established by using different preconcentration and measuring methods. The comparison will be evaluated with respect to these reference values using the E_n number and relative deviation. Therefore, a well-founded estimate of the uncertainty of the reported results is required from each participating laboratory.

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E-mail: uwe.waetjen@ec.europa.eu

Protocol for the comparison

1. Each participant should receive requested number of bottles of drinking water samples. An additional sample may be requested, if required by the routine procedures.

2. The gross alpha/beta activity concentration (volumetric activity) should be determined and reported as *mBq/L*. Also the uncertainty should be reported in *mBq/L*.

3. The laboratory may use a measurement procedure of its choice, which preferably is consistent with routine procedures used in the laboratory.

4. Total dissolved solid (TDS) content is to be determined by the participant on small aliquots – max. 50 mL is sufficiently enough – that will NOT be used for the gross alpha/beta determination.

5. TDS content should be reported in *mg/L* as dry residue at 180 °C.

6. The minimum sample intake for gross alpha/beta analysis depends on the TDS content. It should be adjusted to the applied method.

7. Questionnaire is a part of this exercise and participants should answer all relevant questions regarding the procedures that they have used.

8. Timing and deadlines:

    i. Material distribution: September 2012

    ii. **Deadline for results reporting: 31 October 2012**

9. Preliminary information on the individual laboratory performance will be sent by e-mail in November 2012. The preliminary report of this comparison exercise is foreseen to be available by the end of 2012.

10. The results and performance of each laboratory will be made available to its national representative(s) (the nominating authority) and to the relevant services of the European Commission at DG ENER-D.4. Apart from informing these authorities, each laboratory's results will be treated anonymously.

Uwe Wätjen
Sector Head Radionuclide Metrology

Subject: EC interlaboratory comparison on gross alpha/beta activities in drinking waters – sample dispatch

Dear colleague,

Today, the parcel containing water samples was dispatched to your laboratory by DHL courier from our site (IRMM). Once arrived, **please confirm the receipt of the sample** by e-mail to jana.meresova@ec.europa.eu with copy to viktor.jobbagy@ec.europa.eu. Thank you.

Attached to this e-mail you will find a letter containing information on the testing material as well as the protocol for the comparison:

Your unique password key with the link for the results reporting will be sent to you later.

If you have any further questions, please contact us. We would be happy to help you. Thank you very much for your participation in the comparison.

Best regards,

Appendix 4: Reporting (e-mail)

EUROPEAN COMMISSION
JOINT RESEARCH CENTRE

Institute for Reference Materials and Measurements (Geel)
Standards for Nuclear Safety, Security and Safeguards

Geel, 9 October 2012
JRC.D.4/JM/Ares(2012)1185558

Dear «Title» «Surname»,

We are pleased to inform you that the online reporting system is now operational. The results' reporting is done, via the login page using the following URL:

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

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

Your password key is: **«Part_key»**

Please note that only **submitted** results will be taken into account, therefore, do not only **Save** your results but also select the **Submit** button. Once you have submitted your results and questionnaire, please remember to send us a pdf-file by e-mail or to print the results report form and to fax us a signed copy (Fax no. +32 14 584 273).

As you have been informed, the description of your analytical and measurement procedures will be collected via questionnaire using the same URL link as for the results reporting. We kindly ask that you answer all relevant questions regarding the procedures that you have used.

We would like to inform you that all fields and questions marked with an asterisk (*) are mandatory. The uncertainty of the result must be reported in the same units as the activity concentration (mBq/L). During the reporting of your results and questionnaire the **Cancel** button serves as an exit or return button.

The deadline for reporting results and completing questionnaire is **31 October 2012**.

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

Kind regards,

Jana Meresova

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Appendix 5: Communication on preliminary results

EUROPEAN COMMISSION
JOINT RESEARCH CENTRE

Institute for Reference Materials and Measurements (Geel)
Standards for Nuclear Safety, Security and Safeguards

Geel, 9 January 2013
JRC.D.4/UW/ARES

«Firstname»«Surname»
«Organisation»
«Address»
«Address2»
«Zip» «Town»
«Country»

Subject: Preliminary results of the EC interlaboratory comparison on gross alpha/beta activity measurement in drinking waters

Dear «Title» «Surname»,

First of all, thank you for your participation in the EC interlaboratory comparison on gross alpha/beta activity measurement in drinking waters. At the moment we are working on the evaluation of the results. However, for your early information we are sending you a first of the preliminary evaluation.

Since the anonymity is a requirement in this programme of measurement comparisons, the identity of the laboratories is not shown in the compilation of the results. Each laboratory was assigned a code number.

The code number for your laboratory is **«LCode»**.

In Figures 1 and 2 the reported gross alpha and gross beta activity concentrations, respectively, with their corresponding expanded uncertainties ($k = 2$) are plotted in ascending order for the Water C sample. The solid red lines indicate the reference activity concentrations of ^{241}Am and ^{90}Sr, which were used for spiking to represent gross alpha and gross beta activity, respectively. Their corresponding standard deviations are plotted in dashed lines. The values of activity concentrations in the sample Water C are shown in Table 1. Laboratory codes are indicated with the results. The total dissolved solid (TDS) content of the Water C is 10.2 ± 0.1 mg/L.

Table 1. The activity concentrations in the sample Water C with their expanded uncertainties ($k = 2$).

Radionuclide used for spiking	Activity concentration in the Water C (mBq/L)
^{241}Am	954.5 ± 77.3
^{90}Sr/^{90}Y	1037.3 ± 83.0

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E-mail: uwe.waetjen@ec.europa.eu

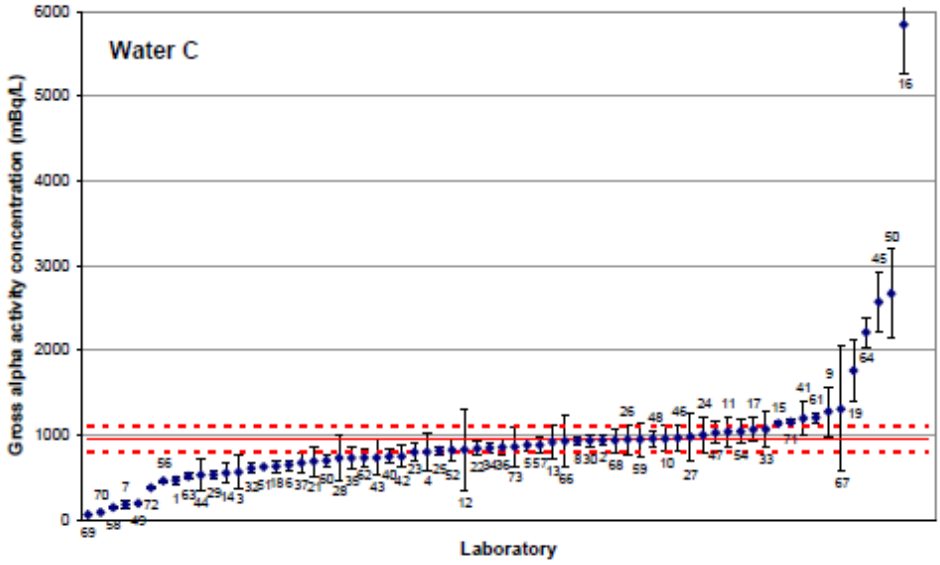

Fig. 1: Laboratory results for gross alpha activity concentration.

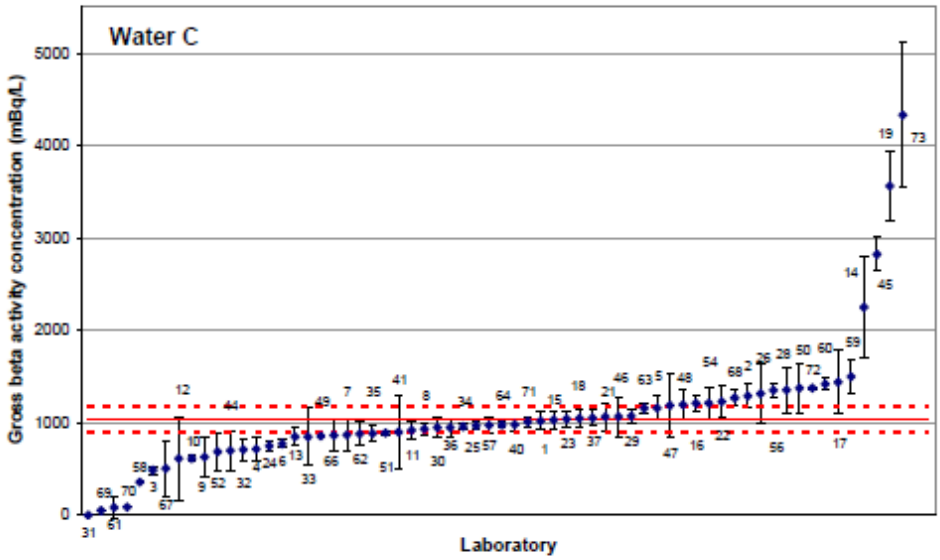

Fig. 2: Laboratory results for gross beta activity concentration

The information on other samples will follow later. If you have any further questions with respect to this interlaboratory comparison, please feel free to contact us.

Yours sincerely,

Uwe Wätjen and Viktor Jobbágy

EUROPEAN COMMISSION
JOINT RESEARCH CENTRE

Institute for Reference Materials and Measurements (Geel)
Standards for Nuclear Safety, Security and Safeguards

Geel, 15 July 2013

«Firstname» «Surname»
«Organisation»
«Address»
«Address2»
«Zip» «Town»
«Country»

Subject: **Preliminary results of the EC interlaboratory comparison on gross alpha/beta activity measurement in drinking waters (Water A)**

Dear «Title» «Surname»,

First of all, thank you for your participation in the EC interlaboratory comparison on gross alpha/beta activity measurement in drinking waters. At the moment we are working on the final evaluation of the results. However, we are sending you now a first, preliminary evaluation.

Since the anonymity is a requirement in this programme of measurement comparisons, the identity of the laboratories is not shown in the compilation of the results. Each laboratory was assigned a code number.

The code number for your laboratory is **«LCode»**.

In Figures 1 and 2 the reported gross alpha and gross beta activity concentrations, respectively, with their corresponding expanded uncertainties ($k = 2$) are plotted in ascending order for the Water A sample. The solid red lines indicate the reference activity concentrations (A_{ref}) of gross alpha and gross beta activity, respectively. Their corresponding expanded uncertainties ± U_{ref} (k = 2) are plotted in dashed red lines. The reference values of activity concentrations in the sample Water A are collected in Table 1. Laboratory codes are indicated with the results.

An additional sample info: the Water A is a commercially available mineral water from the intermediate mineral content class (Total dissolved solid-TDS: 500-1500 mg/L). The total dissolved solid (TDS) content of the Water A is 955 ± 44 mg/L.

Table 1. The reference activity concentration values (A_{ref}) in the sample Water A with their expanded uncertainties (U_{ref}) with a coverage factor $k = 2$.

Parameter	Activity concentration in the Water A(mBq/L)
Gross alpha activity	47.5 ± 22.8
Gross beta activity	309.8 ± 57.4

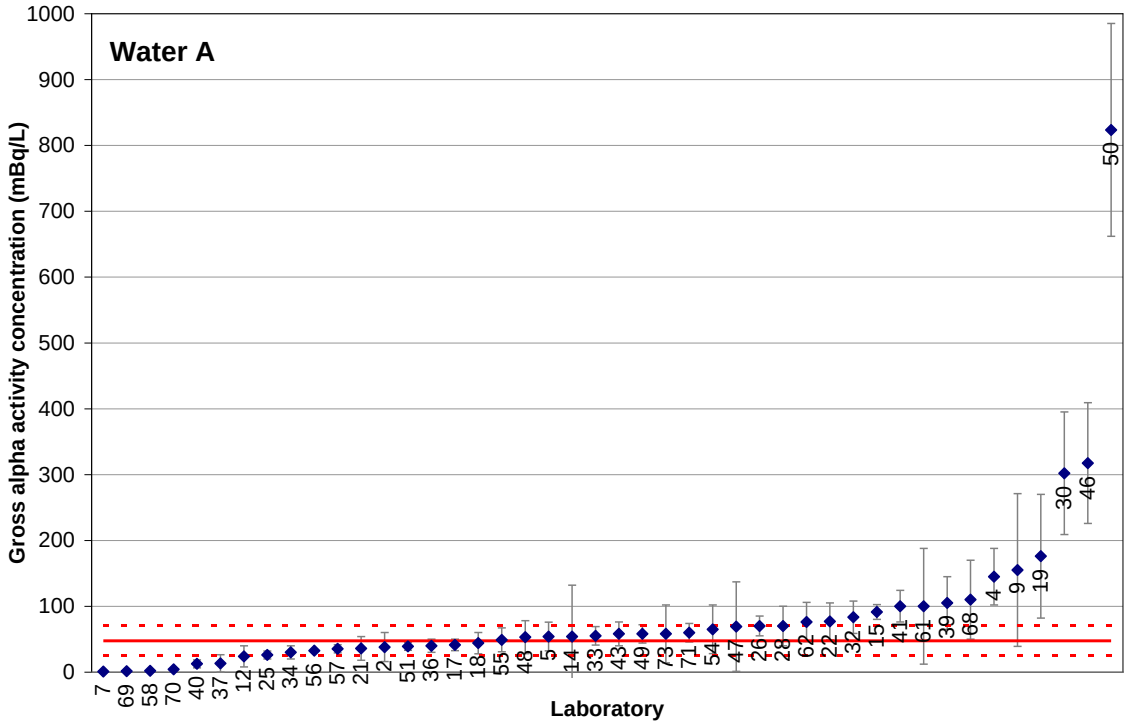

Fig. 1: Laboratory results for gross alpha activity concentration.

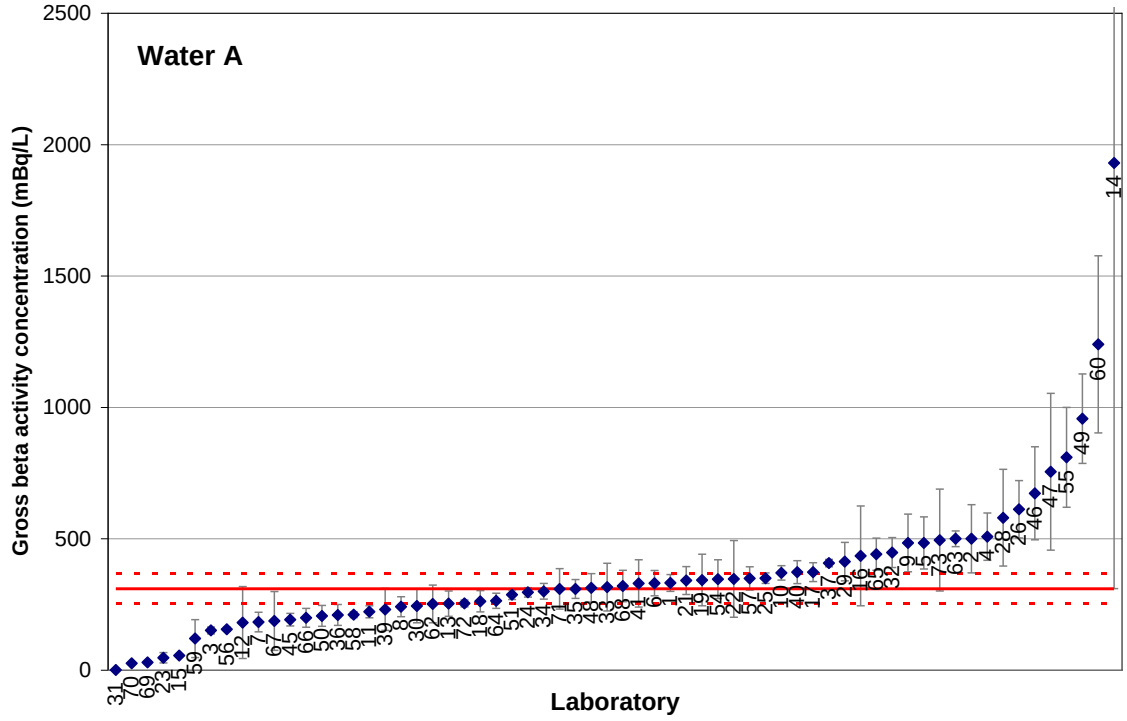

Fig. 2: Laboratory results for gross beta activity concentration

The detailed final report on the gross alpha/beta interlaboratory comparison will follow later this year (2013). If you have any further questions with respect to this interlaboratory comparison, please feel free to contact us.

Yours sincerely,

Uwe Wätjen

EUROPEAN COMMISSION
JOINT RESEARCH CENTRE

Institute for Reference Materials and Measurements (Geel)
Standards for Nuclear Safety, Security and Safeguards

Geel, 15 July 2013

«Firstname» «Surname»
«Organisation»
«Address»
«Address2»
«Zip» «Town»
«Country»

Subject: Preliminary results of the EC interlaboratory comparison on gross alpha/beta activity measurement in drinking waters (Water B)

Dear «Title» «Surname»,

First of all, thank you for your participation in the EC interlaboratory comparison on gross alpha/beta activity measurement in drinking waters. At the moment we are working on the final evaluation of the results. However, we are sending you now a first, preliminary evaluation.

Since the anonymity is a requirement in this programme of measurement comparisons, the identity of the laboratories is not shown in the compilation of the results. Each laboratory was assigned a code number.

The code number for your laboratory is **«LCode»**.

In Figures 1 and 2 the reported gross alpha and gross beta activity concentrations, respectively, with their corresponding expanded uncertainties ($k = 2$) are plotted in ascending order for the Water B sample. The solid red lines indicate the reference activity concentrations (A_{ref}) of gross alpha and gross beta activity, respectively. Their corresponding expanded uncertainties ± U_{ref} (k = 2) are plotted in dashed red lines. The reference values of activity concentrations in the sample Water B are collected in Table 1. Laboratory codes are indicated with the results.

An additional sample info: the Water B is a commercially available mineral water from the low mineral content class (Total dissolved solid-TDS: 50 - 500 mg/L). The total dissolved solid (TDS) content of the Water B is 364 ± 27 mg/L.

Table 1. The reference activity concentration values (A_{ref}) in the sample Water B with their expanded uncertainties (U_{ref}) with a coverage factor $k = 2$.

Parameter	Activity concentration in the Water B (mBq/L)
Gross alpha activity	434.7 ± 56.6
Gross beta activity	190.4 ± 32.6

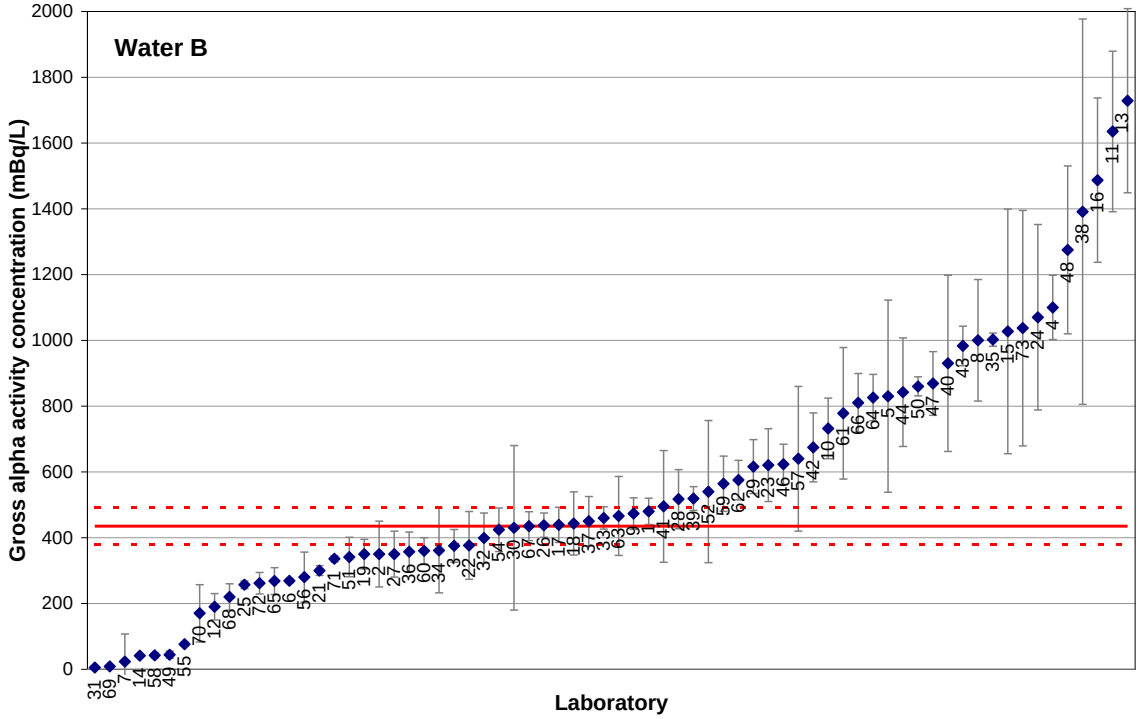

Fig. 1: Laboratory results for gross alpha activity concentration.

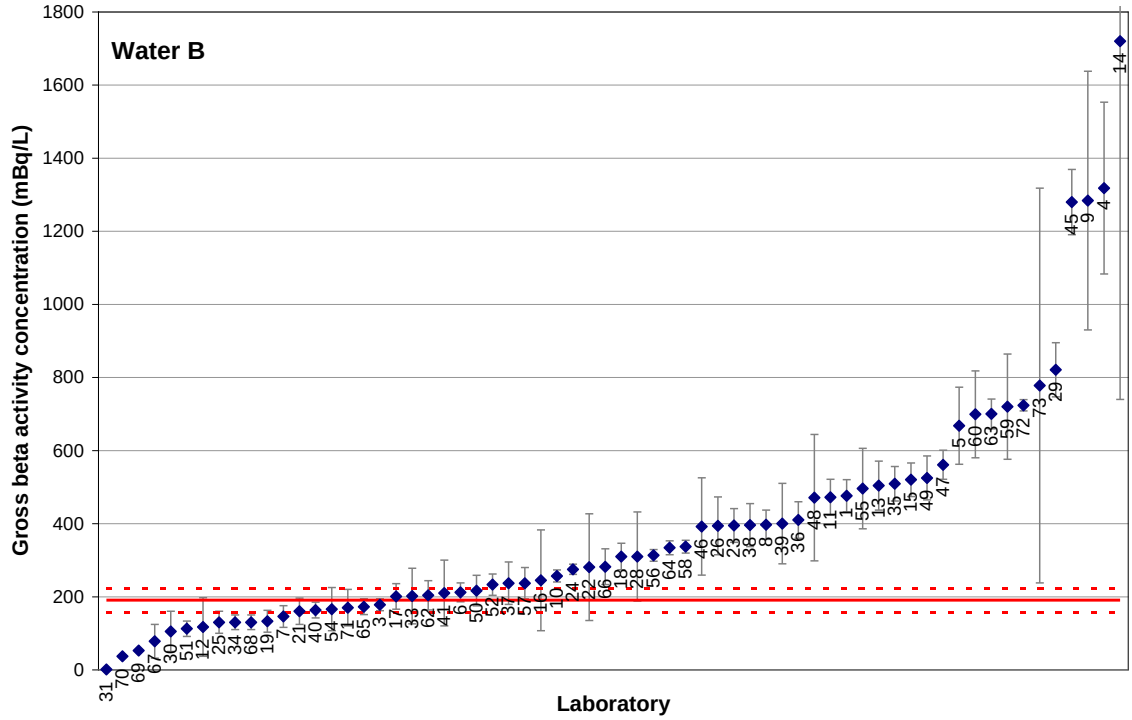

Fig. 2: Laboratory results for gross beta activity concentration

The detailed final report on the gross alpha/beta interlaboratory comparison will follow later this year (2013). If you have any further questions with respect to this interlaboratory comparison, please feel free to contact us.

Yours sincerely,
Uwe Wätjen

Appendix 6: List of participating laboratories (countries in alphabetical order)

AUSTRIA

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BELGIUM

Mr Benoit Deconninck
IRE ELIT
SEM
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BOSNIA-HERZEGOVINA

Mrs Zorana Ilic
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Radiation Protection Centre
Marsala Tita 9
71000 Sarajevo

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Mrs Tsveta Ivanova
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Radiation Control
Perushtica 1
4002 Plovdiv

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Georgi Sofiiski Blvd. 3
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3000 Vratza

Mr Mihail Shishenkov
Executive Environment Agency
Radioactivity Measurements Laboratory
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1618 Sofia

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10000 Zagreb

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Radioactivity Lab (09)
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1451 Nicosia

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10416 Tallinn

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University of Tartu
Institute of Physics
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51014 Tartu

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STUK - Radiation and Nuclear Safety Authority
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Nuclear Physical Chemistry
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Radiation Protection
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Turkish Atomic Energy Authority/Sarayköy Nuclear Research and Training Center (TAEK-SANAEM)
Saray Mahallesi, Atom Caddesi No: 27, Kazan
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Mr Graham Coe
Thames Water
Radiochemistry
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RG2 0JN Reading

Mr Kevin Snaddon
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Organics and Radiochemistry
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EH144AP Edinburgh

Appendix 7: Questionnaire

Milc questionnaire

Comparison for ILC Water 2012

As you have been informed, the description of your analytical and measurement procedures will be collected via questionnaire. We kindly ask that you answer all relevant questions regarding the procedures that you have used. Disregard questions which are not related to the methods used in your laboratory. Several questions are mandatory to answer. These are marked with an asterisk indicator (*). Please be aware that the deadline for the results reporting and submission of the questionnaire is 31 October 2012. Thank you very much for your cooperation.

Submission Form

1. **General information on laboratory**

1.1. What is the type of your laboratory? (More than one choice is possible) *

- ☐ a) Research and development
- ☐ b) Monitoring of radioactivity in the environment
- ☐ c) Monitoring of nuclear facilities
- ☐ d) Measurements for fissile material control or safeguards
- ☐ e) University
- ☐ f) Other

1.1.1. If other, please specify here:

1.2. Is your laboratory certified, accredited or authorised for this type of analysis? (More than one choice is possible) *

- ☐ a) Certified
- ☐ b) Accredited
- ☐ c) Authorised
- ☐ d) No

1.3. Is your laboratory working according to a quality management system? (More than one choice is possible) *

- ☐ a) ISO 9000 series
- ☐ b) ISO 17025
- ☐ c) No quality management system
- ☐ d) Other

1.3.1. If other, please specify here:

2. **General information on measurement**

2.1. Does your country have a legislation on gross alpha/beta activity in water? *

- a) Yes
- b) No

2.1.1. If yes, what are the screening levels? *
See table **Screening levels** at bottom

2.2. Do you have routine procedures for gross alpha/beta activity measurement in your laboratory? *

- a) Yes
- b) No

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

- a) < 25
- b) 25 - 100
- c) > 100

2.4. What are the typical activity concentrations measured in your laboratory? *
See table **Typical activity concentrations** at bottom

3. **Sample treatment**

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

- a) Yes
- b) No

3.2. What was the date of sample treatment? *
See table **Date of treatment** at bottom

3.3. What was the volume of sample used? *
See table **Sample volume** at bottom

3.4. Did you check the amount of total dissolved solids prior to the sample preparation? *

- ◯ a) Yes
- ◯ b) No

3.4.1. If yes, what was the content of total dissolved solids determined in the intercomparison samples? *
See table **Total dissolved solids** at bottom

3.5. Which method have you used for the sample preparation? (More than one choice is possible) *

- ☐ a) Evaporation to complete dryness
- ☐ b) Coprecipitation
- ☐ c) Evaporation and mixing sample with LSC cocktail
- ☐ d) Other

3.5.1. If other, please specify here:

3.6. How do you deposit the residue onto the planchet?

3.7. Do you check the surface density of the prepared source/sample on the planchet?

- ◯ a) Yes
- ◯ b) No

3.7.1. If yes, briefly describe the procedure: *

3.8. Do you have a procedure how to treat sample if it is hygroscopic?

- ◯ a) Yes
- ◯ b) No

3.8.1. If yes, briefly describe the procedure: *

4. **Measurement**

4.1. What type of detector was used? (More than one choice is available) *

- ☐ a) Proportional counter
- ☐ b) LSC counter
- ☐ c) Solid scintillation detector
- ☐ d) Other

4.1.1. If solid scintillation detector, please specify the type:

4.1.2. If other, please specify here:

4.2. What was the date of sample measurement? *
See table **Date of measurement** at bottom

4.3. What was the time between the sample treatment and measurement? *
See table **Time between sample treatment and measurement** at bottom

4.4. Do you measure simultaneously alpha and beta gross activities with your instrument? *

- ☐ a) Yes
- ☐ b) No

4.5. What was the counting time? *
See table **Counting time** at bottom

4.6. What are the detection limits of your routine method? *
See table **Limits of detection** at bottom

4.7. How do you determine the background level of your instrument? *

4.8. What is the typical background count rate of your instrument? *
See table **Background counts** at bottom

4.9. How did you determine the counting efficiency of your instrument? What kind of standard sources did you use? *

5. **Proportional counter**

5.1. What type of proportional counter do you use in your laboratory?

- ◯ a) Gas flow
- ◯ b) Permanently sealed

5.2. What is the counting efficiency of the proportional counter used?
See table **PC counting efficiency** at bottom

6. **Liquid scintillation counting**

6.1. What kind of scintillation cocktail did you use?

6.2. What was the ratio sample to LSC cocktail?

6.3. Did you apply quenching correction?

- ◯ a) Yes
- ◯ b) No

6.3.1. If yes, briefly describe the procedure: *

6.4. Did you apply alpha/beta discrimination?

- ◯ a) Yes
- ◯ b) No

6.4.1. If yes, briefly describe the procedure: *

6.5. What is the counting efficiency of the LSC counter used?
See table **LSC counting efficiency** at bottom

6.6. In case you used LSC method, what type of vials did you use?

- ○ a) Glass
- ○ b) Polyethylene
- ○ c) Teflon coated
- ○ d) Low potassium glass
- ○ e) Other

6.6.1. If other, please specify: *

7. **Uncertainty budget**

7.1. Please give an example of uncertainty budget for one single measurement of the gross alpha activity concentration. In the table are listed possible contributions, which should be accounted for in the estimation of the combined uncertainty: *

See table **Uncertainty budget - Gross alpha** at bottom

7.1.1. Combined relative standard uncertainty of gross alpha activity concentration (%) *

(number)

7.1.2. If you added other uncertainty contribution 1, please specify:

7.1.3. If you added other uncertainty contribution 2, please specify:

7.2. Please give an example of uncertainty budget for one single measurement of the gross beta activity concentration. In the table are listed possible contributions, which should be accounted for in the estimation of the combined uncertainty: *

See table **Uncertainty budget - Gross beta** at bottom

7.2.1. Combined relative standard uncertainty of gross beta activity concentration (%) *

(number)

7.2.2. If you added other uncertainty contribution 1, please specify:

7.2.3. If you added other uncertainty contribution 2, please specify:

Background counts

Questions/Response table	*counts per second*
Gross alpha activity	
Gross beta activity	

Counting time

Questions/Response table	*seconds*
Water A	
Water B	
Water C	

Date of measurement

Questions/Response table	*Date*
Water A	
Water B	

Questions/Response table	*Date*
Water C	

Date of treatment

Questions/Response table	*Date*
Water A	
Water B	
Water C	

LSC counting efficiency

Questions/Response table	*%*
Gross alpha activity	
Gross beta activity	

Limits of detection

Questions/Response table	mBq/L
Gross alpha activity	
Gross beta activity	

PC counting efficiency

Questions/Response table	%
Gross alpha activity	
Gross beta activity	

Sample volume

Questions/Response table	L
Water A	
Water B	
Water C	

Screening levels

Questions/Response table	mBq/L
Gross alpha activity	
Gross beta activity	

Time between sample treatment and measurement

Questions/Response table	Hours
Water A	
Water B	
Water C	

Total dissolved solids

Questions/Response table	mg/L
Water A	
Water B	
Water C	

Typical activity concentrations

Questions/Response table	*mBq/L*
Gross alpha activity	
Gross beta activity	

Uncertainty budget - Gross alpha

Contribution to relative uncertainty of activity concentration is product of relative uncertainty of the component and its sensitivity coefficient (or partial derivative df/dx).

Questions/Response table	*Relative uncertainty of the component (%)*	*Contribution to rel. unc. of act. conc. (%)*
Counting statistics		
Backround measurement		
Efficiency calibration		
Self-absorption		
Activity of the calibration source		
Sample volume		
Weighing		
Other uncertainty contribution 1		
Other uncertainty contribution 2		

Uncertainty budget - Gross beta

Contribution to relative uncertainty of activity concentration is product of relative uncertainty of the component and its sensitivity coefficient (or partial derivative df/dx).

Questions/Response table	*Relative uncertainty of component (%)*	*Contribution to rel. unc. of act. conc. (%)*
Counting statistics		
Backround measurement		
Efficiency calibration		
Self-absorption		
Activity of the calibration source		
Sample volume		
Weighing		
Other uncertainty contribution 1		
Other uncertainty contribution 2		

Appendix 8: Results, methods and scores of laboratories

Water A

Lab code	Laboratory result Water A alpha		D% (%)	Laboratory result Water A beta		D% (%)	Measurement technique
	$A_{lab} \pm U_{lab}$ (mBq/L) (k=2)	U% (%)		$A_{lab} \pm U_{lab}$ (mBq/L) (k=2)	U% (%)		
1	< 35	-	**-**	332 ± 32	10	**7.2**	Proportional counter
2	38 ± 22	58	**-20.0**	500 ± 130	26	**61.4**	Liquid-scint. counting
3	< 40	-	**-**	151 ± 13	9	**-51.3**	Proportional counter
4	145 ± 43	30	**205.3**	508 ± 90	18	**64.0**	Proportional counter
5	54 ± 21.818	40	**13.7**	484 ± 99	21	**56.2**	Liquid-scint. counting
6	< 50	-	**-**	331 ± 48	15	**6.8**	Proportional counter
7	0.81 ± 0.16	20	**-98.3**	182.99 ± 37	20	**-40.9**	Proportional counter
8	< 43	-	**-**	241 ± 38	16	**-22.2**	Proportional counter
9	155 ± 116	75	**226.3**	484 ± 110	23	**56.2**	(α) Proportional counter (β) Scintillation counter
10	< 15	-	**-**	370.3 ± 27	7	**19.5**	-
11	< 40	-	**-**	222 ± 23	10	**-28.3**	Proportional counter
12	24 ± 16	67	**-49.5**	181 ± 137	76	**-41.6**	Proportional counter
13	< 40	-	**-**	253 ± 48	19	**-18.3**	Proportional counter
14	54 ± 78	144	**13.7**	1930 ± 1620	84	**523.0**	Liquid-scint. counting
15	91.3 ± 11.2	12	**92.2**	55.8 ± 3	6	**-82.0**	Scintillation counter
16	< 143	-	**-**	435 ± 190	44	**40.4**	Proportional counter
17	41.3 ± 8.8	21	**-13.1**	373 ± 36	10	**20.4**	Liquid-scint. counting
18	44 ± 16.1	37	**-7.4**	262 ± 41	16	**-15.4**	Proportional counter
19	176 ± 94	53	**270.5**	343 ± 98	29	**10.7**	Proportional counter
20	did not report	-	**-**	did not report	-	**-**	-
21	36 ± 18	50	**-24.2**	341 ± 53	16	**10.1**	(α) Scintillation counter

										(β) Proportional counter
22		77 ± 28	36	**62.1**		347 ± 146	42	**12.0**		Liquid-scint. counting
23		< 37	-	**-**		47 ± 20	42	**-84.8**		i-Matic Si-det
24		< 39	-	**-**		296 ± 18	6	**-4.5**		Proportional counter
25		26 ± 6	23	**-45.3**		350 ± 20	6	**13.0**		Proportional counter
26		70 ± 15	21	**47.4**		612 ± 109	18	**97.5**		Liquid-scint. counting
27		0	-	**-**		0	-	**-**		-
28		70 ± 30	43	**47.4**		580 ± 184	32	**87.2**		Liquid-scint. counting
29		< 120	-	**-**		413 ± 73	18	**33.3**		(β) Proportional counter
30		302 ± 93	31	**535.8**		244 ± 64	26	**-21.2**		Proportional counter
31		0	-	**-**		0.633 ± 0.072	11	**-99.8**		(β) Proportional counter
32		83.3 ± 24.6	30	**75.4**		447.8 ± 57	13	**44.5**		Liquid-scint. counting
33		55 ± 14	25	**15.8**		316 ± 91	29	**2.0**		Liquid-scint. counting
34		30 ± 10	33	**-36.8**		300 ± 30	10	**-3.2**		(α) Scintillation counter (β) Proportional counter
35		< 69	-	**-**		309 ± 36	12	**-0.3**		Proportional counter
36		40 ± 10	25	**-15.8**		210 ± 40	19	**-32.2**		Proportional counter
37		13.3 ± 13.2	99	**-72.0**		407.2 ± 14	3	**31.4**		Proportional counter
38		< 92	-	**-**		< 114	-	**-**		Proportional counter
39		105 ± 40	38	**121.1**		231 ± 80	35	**-25.4**		Liquid-scint. counting
40		12.51 ± 6.135	49	**-73.7**		372.7 ± 44	12	**20.3**		Proportional counter
41		100 ± 24	24	**110.5**		330 ± 90	27	**6.5**		Liquid-scint. counting
42		< 74	-	**-**		> 0	-	**-**		(α) Liquid-scint. counting
43		58 ± 18	31	**22.1**		0	-	**-**		(α) grid ionization chamber
44		< 102	-	**-**		< 423	-	**-**		Liquid-scint. counting
45		< 225.2	-	**-**		192.1 ± 24	12	**-38.0**		Proportional counter
46		317.4 ± 91.73	29	**568.2**		672.8 ± 177	26	**117.2**		Liquid-scint. counting
47		69 ± 68	99	**45.3**		755 ± 298	39	**143.7**		LSC

48		53 ± 25	47	**11.6**		313 ± 54	17	**1.0**		(α) Scintillation counter (β) Proportional counter
49		58 ± 14	24	**22.1**		957 ± 170	18	**208.9**		Scintillation counter
50		823.5 ± 161.5	20	**1633.7**		206.5 ± 40	19	**-33.3**		Liquid-scint. counting
51		39 ± 6	15	**-17.9**		287 ± 18	6	**-7.4**		(α) Analyzer MC 2256, mix of powder sample and ZnS(Ag) powder with direct contact with PMT (β) Proportional counter
52		< 169	-	**-**		< 263	-	**-**		Liquid-scint. counting
53		did not report	-	**-**		did not report	-	**-**		-
54		65 ± 37	57	**36.8**		346 ± 74	21	**11.7**		Proportional counter
55		49 ± 18	37	**3.2**		810 ± 190	23	**161.5**		(α) silicon detector (β) Scintillation counter
56		32.507 ± 0.812244898	2	**-31.6**		155.404 ± 8	5	**-49.8**		Proportional counter
57		35.3 ± 6.2	18	**-25.7**		349 ± 44	13	**12.7**		(α) Scintillation counter (β) Proportional counter
58		2.06 ± 0.11	5	**-95.7**		211 ± 11	5	**-31.9**		Proportional counter
59		< 40	-	**-**		120 ± 72	60	**-61.3**		Proportional counter
60		< 174	-	**-**		1240 ± 337	27	**300.3**		Liquid-scint. counting
61		100 ± 88	88	**110.5**		< 260	-	**-**		Liquid-scint. counting
62		76 ± 30	39	**60.0**		252 ± 72	29	**-18.7**		(α) Scintillation counter (β) Proportional counter
63		0	-	**-**		500 ± 30	6	**61.4**		Proportional counter
64		< 40	-	**-**		264 ± 29	11	**-14.8**		Proportional counter
65		< 99.7	-	**-**		441.2 ± 61	14	**42.4**		Proportional counter
66		< 20	-	**-**		199 ± 36	18	**-35.8**		Proportional counter
67		< 20	-	**-**		187 ± 112	60	**-39.6**		Proportional counter
68		110 ± 60	55	**131.6**		320 ± 60	19	**3.3**		Proportional counter
69		1.47 ± 0.41	28	**-96.9**		29.3 ± 5	18	**-90.5**		semiconductor Si detector
70		4.1 ± 0.6	15	**-91.4**		26 ± 2	8	**-91.6**		Liquid-scint. counting

71		59.9 ± 14	23	**26.1**		309 ± 77	25	**-0.3**		Proportional counter
72		< 10	-	**-**		253.8 ± 13	5	**-18.1**		Proportional counter
73		58 ± 44	76	**22.1**		495 ± 194	39	**59.8**		Liquid-scint. counting

Water B

Lab code	Laboratory result Water B alpha		$D_{\%}$ (%)	Laboratory result Water B beta		$D_{\%}$ (%)	Measurement technique
	$A_{lab} \pm U_{lab}$ (mBq/L) (k=2)	$U_{\%}$ (%)		$A_{lab} \pm U_{lab}$ (mBq/L) (k=2)	$U_{\%}$ (%)		
1	473 ± 48	10	**8.8**	476 ± 44	9	**150.0**	Proportional counter
2	350 ± 45	13	**-19.5**	< 200	-	**-**	(α) Liquid-scint. counting
3	361 ± 129	36	**-17.0**	178 ± 16	9	**-6.5**	Proportional counter
4	1070 ± 282	26	**146.1**	1318 ± 235	18	**592.2**	Proportional counter
5	826 ± 70	8	**90.0**	668 ± 105.5	16	**250.8**	Liquid-scint. counting
6	268 ± 41	15	**-38.3**	212 ± 26	12	**11.3**	Proportional counter
7	8.42 ± 2	24	**-98.1**	145.89 ± 29.8	20	**-23.4**	Proportional counter
8	983 ± 60	6	**126.1**	397 ± 40	10	**108.5**	Proportional counter
9	466 ± 120	26	**7.2**	1284 ± 354	28	**574.4**	(α) Proportional counter (β) Scintillation counter
10	674.5 ± 105	16	**55.2**	256.6 ± 16.5	6	**34.8**	-
11	1487 ± 250	17	**242.1**	472 ± 49	10	**147.9**	Proportional counter
12	170 ± 87	51	**-60.9**	117 ± 80	68	**-38.6**	Proportional counter
13	1635 ± 244	15	**276.1**	504 ± 67	13	**164.7**	Proportional counter
14	23 ± 84	365	**-94.7**	1720 ± 980	57	**803.4**	Liquid-scint. counting
15	1002 ± 20	2	**130.5**	520 ± 46	9	**173.1**	Scintillation counter
16	1391 ± 586	42	**220.0**	245 ± 138	56	**28.7**	Proportional counter
17	438 ± 37	8	**0.8**	201 ± 35	17	**5.6**	Liquid-scint. counting
18	439 ± 54	12	**1.0**	310 ± 36.4	12	**62.8**	Proportional counter
19	341 ± 60	18	**-21.6**	133 ± 30	23	**-30.1**	Proportional counter
20	did not report	-	**-**	did not report	-	**-**	-
21	280 ± 76	27	**-35.6**	160 ± 36	23	**-16.0**	(α) Scintillation counter (β) Proportional counter
22	375 ± 50	13	**-13.7**	281 ± 73	52	**47.6**	Liquid-scint. counting

23	616 ± 82	13	**41.7**	395 ± 46	12	**107.5**	i-Matic Si-det
24	1037 ± 358	35	**138.6**	275 ± 14	5	**44.4**	Proportional counter
25	220 ± 40	18	**-49.4**	130 ± 30	23	**-31.7**	Proportional counter
26	435 ± 44	10	**0.1**	394 ± 79	20	**106.9**	Liquid-scint. counting
27	350 ± 100	29	**-19.5**	0	-	**-**	(α) Liquid-scint. counting
28	495 ± 170	34	**13.9**	310 ± 122	39	**62.8**	Liquid-scint. counting
29	575 ± 60	10	**32.3**	821 ± 74	9	**331.2**	Proportional counter
30	424 ± 66	16	**-2.5**	105 ± 55	52	**-44.9**	Proportional counter
31	0	-	**-**	0.828 ± 0.288	35	**-99.6**	(β) Proportional counter
32	376.3 ± 103	27	**-13.4**	< 0.3	-	**-**	Liquid-scint. counting
33	450 ± 75	17	**3.5**	202 ± 76	38	**6.1**	Liquid-scint. counting
34	360 ± 40	11	**-17.2**	130 ± 20	15	**-31.7**	(α) Scintillation counter (β) Proportional counter
35	1000 ± 185	19	**130.0**	509 ± 47	9	**167.3**	Proportional counter
36	350 ± 70	20	**-19.5**	410 ± 50	12	**115.3**	Proportional counter
37	443 ± 96	22	**1.9**	237 ± 58	24	**24.5**	Proportional counter
38	1275 ± 255	20	**193.3**	396 ± 59	15	**108.0**	Proportional counter
39	517 ± 90	17	**18.9**	400 ± 110	28	**110.1**	Liquid-scint. counting
40	868.6 ± 97	11	**99.8**	163.33 ± 21.56	13	**-14.2**	Proportional counter
41	480 ± 40	8	**10.4**	210 ± 90	43	**10.3**	Liquid-scint. counting
42	640 ± 220	34	**47.2**	> 0	-	**-**	(α) Liquid-scint. counting
43	930 ± 268	29	**113.9**	0	-	**-**	(α) grid ionization chamber
44	830 ± 292	35	**90.9**	< 423	-	**-**	Liquid-scint. counting
45	1728.5 ± 280	16	**297.6**	1279.8 ± 89.2	7	**572.2**	Proportional counter
46	620.5 ± 111	18	**42.7**	392 ± 133.09	34	**105.9**	Liquid-scint. counting
47	860 ± 29	3	**97.8**	561 ± 40	7	**194.6**	LSC
48	1100 ± 98	9	**153.0**	471 ± 173	37	**147.4**	(α) Scintillation counter

									(β) Proportional counter
49		42 ± 8	19	**-90.3**		525 ± 60	11	**175.7**	Scintillation counter
50		842.1 ± 165	20	**93.7**		216.7 ± 41.97	19	**13.8**	Liquid-scint. counting
51		335.5 ± 9	3	**-22.8**		112.5 ± 21	19	**-40.9**	(α) Analyzer MC 2256, mix of powder sample and ZnS(Ag) powder with direct contact with PMT (β) Proportional counter
52		519 ± 36	7	**19.4**		233 ± 29	12	**22.4**	Liquid-scint. counting
53		did not report	-	**-**		did not report	-	**-**	-
54		399 ± 76	19	**-8.2**		166 ± 59	36	**-12.8**	Proportional counter
55		44 ± 11	25	**-89.9**		496 ± 110	22	**160.5**	(α) silicon detector (β) Scintillation counter
56		268.934 ± 7	3	**-38.1**		313.268 ± 15.91428571	5	**64.5**	Proportional counter
57		623 ± 61	10	**43.3**		237 ± 43	18	**24.5**	(α) Scintillation counter (β) Proportional counter
58		41.1 ± 2	5	**-90.5**		337 ± 17.66	5	**77.0**	Proportional counter
59		540 ± 216	40	**24.2**		720 ± 144	20	**278.2**	Proportional counter
60		357 ± 60	17	**-17.9**		699 ± 119	17	**267.1**	Liquid-scint. counting
61		732 ± 92	13	**68.4**		< 242	-	**-**	Liquid-scint. counting
62		564 ± 84	15	**29.7**		204 ± 40	20	**7.1**	(α) Scintillation counter (β) Proportional counter
63		460 ± 34	7	**5.8**		700 ± 41	6	**267.6**	Proportional counter
64		810 ± 89	11	**86.3**		334 ± 19	6	**75.4**	Proportional counter
65		261.4 ± 33	13	**-39.9**		172.6 ± 21.7	13	**-9.3**	Proportional counter
66		778 ± 200	26	**79.0**		282 ± 49	17	**48.1**	Proportional counter
67		430 ± 250	58	**-1.1**		78 ± 46	59	**-59.0**	Proportional counter
68		190 ± 40	21	**-56.3**		130 ± 20	15	**-31.7**	Proportional counter
69		5 ± 0	0	**-98.8**		52.5 ± 0.71	1	**-72.4**	semiconductor Si detector
70		76 ± 6	8	**-82.5**		37 ± 3	8	**-80.6**	Liquid-scint. counting

71		300 ± 15	5	**-31.0**		170 ± 50	29	**-10.7**		Proportional counter
72		256.67 ± 12	5	**-41.0**		723.8 ± 15.62	2	**280.1**		Proportional counter
73		1027 ± 372	36	**136.3**		778 ± 540	69	**308.6**		Liquid-scint. counting

Water C

Lab code	Laboratory result Water C alpha		$D_{\%}$ (%)	Laboratory result Water C beta		$D_{\%}$ (%)	Measurement technique
	$A_{lab} \pm U_{lab}$ (mBq/L) (k=2)	$U_{\%}$ (%)		$A_{lab} \pm U_{lab}$ (mBq/L) (k=2)	$U_{\%}$ (%)		
1	470 ± 48	10	**-50.8**	1020 ± 100	10	**-1.7**	Proportional counter
2	940 ± 50	5	**-1.5**	1290 ± 130	10	**24.4**	Liquid-scint. counting
3	564 ± 201	36	**-40.9**	488 ± 43	9	**-53.0**	Proportional counter
4	803 ± 214	27	**-15.9**	714 ± 130	18	**-31.2**	Proportional counter
5	885 ± 64	7	**-7.3**	1160 ± 126.1	11	**11.8**	Liquid-scint. counting
6	651 ± 59	9	**-31.8**	773 ± 42	5	**-25.5**	Proportional counter
7	181.9 ± 37.12	20	**-80.9**	870.4 ± 177.6	20	**-16.1**	Proportional counter
8	933 ± 56	6	**-2.3**	932 ± 54	6	**-10.2**	Proportional counter
9	1278 ± 296	23	**33.9**	630 ± 208	33	**-39.3**	(α) Proportional counter (β) Scintillation counter
10	959 ± 148.6	15	**0.5**	615.5 ± 31.9	5	**-40.7**	-
11	1044 ± 175	17	**9.4**	920 ± 96	10	**-11.3**	Proportional counter
12	830 ± 484	58	**-13.0**	612 ± 447	73	**-41.0**	Proportional counter
13	920 ± 200	22	**-3.6**	850 ± 100	12	**-18.1**	Proportional counter
14	555 ± 126	23	**-41.9**	2250 ± 540	24	**116.9**	Liquid-scint. counting
15	1139 ± 24	2	**19.3**	1034 ± 102	10	**-0.3**	Scintillation counter
16	5846 ± 570	10	**512.5**	1211 ± 96	8	**16.7**	Proportional counter
17	1068 ± 148	14	**11.9**	1440 ± 341	24	**38.8**	Liquid-scint. counting
18	634 ± 71.6	11	**-33.6**	1050 ± 88.9	8	**1.2**	Proportional counter
19	1765 ± 370	21	**84.9**	3563 ± 366	10	**243.5**	Proportional counter
20	did not report	-	**-**	did not report	-	**-**	-
21	691 ± 180	26	**-27.6**	1067 ± 152	14	**2.9**	(α) Scintillation counter (β) Proportional counter

22		843 ± 80	9	**-11.7**		1230 ± 168	14	**18.6**		Liquid-scint. counting
23		802 ± 102	13	**-16.0**		1042 ± 78	7	**0.5**		i- Matic Si-det
24		1002 ± 214	21	**5.0**		751 ± 50	7	**-27.6**		Proportional counter
25		820 ± 50	6	**-14.1**		970 ± 50	5	**-6.5**		Proportional counter
26		949 ± 170	18	**-0.6**		1316 ± 326	25	**26.9**		Liquid-scint. counting
27		980 ± 290	30	**2.7**		0	-	-		(α) Liquid-scint. counting
28		732 ± 262	36	**-23.3**		1355 ± 246	18	**30.6**		Liquid-scint. counting
29		533 ± 39	7	**-44.2**		1073 ± 77	7	**3.4**		Proportional counter
30		938 ± 72	8	**-1.7**		949 ± 108	11	**-8.5**		Proportional counter
31		0	-	**-**		1.379 ± 0.028	2	**-99.9**		(β) Proportional counter
32		609.8 ± 53.2	9	**-36.1**		710.3 ± 118.2	17	**-31.5**		Liquid-scint. counting
33		1070 ± 220	21	**12.1**		850 ± 310	36	**-18.1**		Liquid-scint. counting
34		850 ± 50	6	**-10.9**		960 ± 40	4	**-7.5**		(α) Scintillation counter (β) Proportional counter
35		733 ± 140	19	**-23.2**		884 ± 82	9	**-14.8**		Proportional counter
36		860 ± 90	10	**-9.9**		950 ± 100	11	**-8.4**		Proportional counter
37		674 ± 122	18	**-29.4**		1053 ± 86	8	**1.5**		Proportional counter
38		0	-	**-**		0	-	-		-
39		0	-	**-**		0	-	-		breaking lsc equipment
40		748.5 ± 78.7	11	**-21.6**		983.03 ± 64.18	7	**-5.2**		Proportional counter
41		1200 ± 200	17	**25.7**		900 ± 400	44	**-13.2**		Liquid-scint. counting
42		750 ± 130	17	**-21.4**		> 0	-	-		(α) Liquid-scint. counting
43		735 ± 210	29	**-23.0**		0	-	-		(α) grid ionization chamber
44		532 ± 187	35	**-44.3**		699 ± 210	30	**-32.6**		Liquid-scint. counting
45		2573.5 ± 357.6	14	**169.6**		2823.7 ± 179.8	6	**172.2**		Proportional counter
46		968.3 ± 149.87	15	**1.4**		1068.3 ± 216.65	20	**3.0**		Liquid-scint. counting
47		1033 ± 131	13	**8.2**		1188 ± 338	28	**14.5**		LSC

48		958 ± 86	9	**0.4**		1197 ± 164	14	**15.4**	(α) Scintillation counter (β) Proportional counter
49		197 ± 7	4	**-79.4**		857 ± 21	2	**-17.4**	Scintillation counter
50		2671.9 ± 523.9	20	**179.9**		1375.9 ± 266.5	19	**32.6**	Liquid-scint. counting
51		627.3 ± 1.4	0	**-34.3**		893.2 ± 24.4	3	**-13.9**	(α) Analyzer MC 2256, mix of powder sample and ZnS(Ag) powder with direct contact with PMT (β) Proportional counter
52		824 ± 129	16	**-13.7**		688 ± 200	29	**-33.7**	Liquid-scint. counting
53		did not report	-	**-**		did not report	-	**-**	-
54		1044 ± 146	14	**9.4**		1215 ± 178	15	**17.1**	Proportional counter
55		0	-	**-**		0	-	-	(α) silicon detector (β) Scintillation counter
56		461.24 ± 11.532	3	**-51.7**		1352.5 ± 67.62	5	**30.4**	Proportional counter
57		886 ± 86	10	**-7.2**		973 ± 87	9	**-6.2**	(α) Scintillation counter (β) Proportional counter
58		149 ± 7.87	5	**-84.4**		360 ± 18.86	5	**-65.3**	Proportional counter
59		950 ± 200	21	**-0.5**		1500 ± 180	12	**44.6**	Proportional counter
60		702 ± 73	10	**-26.5**		1420 ± 65	5	**36.9**	Liquid-scint. counting
61		1208 ± 58	5	**26.6**		85 ± 124	146	**-91.8**	Liquid-scint. counting
62		733 ± 98	13	**-23.2**		880 ± 130	15	**-15.2**	(α) Scintillation counter (β) Proportional counter
63		516 ± 38	7	**-45.9**		1158 ± 58	5	**11.6**	Proportional counter
64		2212 ± 174	8	**131.7**		978 ± 33	3	**-5.7**	Proportional counter
65		0	-	**-**		0	-	-	Proportional counter
66		930 ± 300	32	**-2.6**		866 ± 171	20	**-16.5**	Proportional counter
67		1310 ± 740	56	**37.2**		506 ± 300	59	**-51.2**	Proportional counter
68		940 ± 140	15	**-1.5**		1270 ± 80	6	**22.4**	Proportional counter
69		63.2 ± 0.42	1	**-93.4**		51 ± 1.41	3	**-95.1**	semiconductor Si detector
70		90 ± 7	8	**-90.6**		90 ± 5	6	**-91.3**	Liquid-scint. counting

71		1158 ± 20	2	**21.3**		1010 ± 58	6	**-2.6**		Proportional counter
72		381.67 ± 10.88	3	**-60.0**		1378.7 ± 10.16	1	**32.9**		Proportional counter
73		866 ± 238	27	**-9.3**		4331 ± 780	18	**317.5**		Liquid-scint. counting

Methods used

1) Which method have you used for the sample preparation? (More than one choice is possible)

1a) If other, please specify here.

2) How do you deposit the residue onto the planchet?

3) Do you check the surface density of the prepared source/sample on the planchet? If yes, briefly describe the procedure.

4) Do you have a procedure how to treat sample if it is hygroscopic? If yes, briefly describe the procedure.

Lab code	Measurement technique	Used method
1	Proportional counter	1) Evaporation to complete dryness; 2) The residue is transferred onto the planchet with alcohol; 3) The surface density is checked by the weight of total residue dividing the disk surface; 4) The residue is transformed to sulfate form
2	Liquid-scint. counting	1) Evaporation and mixing sample with LSC cocktail
3	Proportional counter	1) Evaporation to complete dryness; 2) Mix with Acetone and allow solvent to evaporate; 4) sample stored in a dessicator with silca gel
4	Proportional counter	1a) Concentration, sulfation, evaporation and ignition; 2) I transfer certain amount of the residue after ignition onto the planchet and fix it with acetone
5	Liquid-scint. counting	1) Evaporation and mixing sample with LSC cocktail.
6	Proportional counter	1) Evaporation to complete dryness, 1a) treating with sulfuric acid; 2) by grinding and dispersing in acetone; 3) by measuring the mass of the sample and dividing it by the surface area of the planchet
7	Proportional counter	1) Evaporation to complete dryness; 2) evaporate on the planchet; 3) weighing before and after

8	Proportional counter	1) Evaporation to complete dryness, 1a) Evaporation under IR lamps then drying at 180 C in oven and burn up to red dull using an electrical burner (Hydroscopic samples); 2) Adding small portions of the water in pre-weighted planchet under IR lamps; 3) No but we try to distribute uniformly the residue on the planchet(not more than 100mg); 4) We convert the salts to their oxide form burning the planchet with the residue up to red dull using un electrical burner. This is a disadvantage of the method for volatile radionuclides.
9	(α) Proportional counter (β) Scintillation counter	1) Evaporation to complete dryness, 1a) CaSO4 was added to obtain residue for sample C; 2) Deposit aliquot after scraping, mashing and homogenization; 3) Weighting of residue, used for measurement in order to obtain |mg/cm2|; 4) Ashing or/and heating with UV lamp and temperate in dessicator
10	-	1) Evaporation to complete dryness; 2) dry residue sample weight was placed in a planchety and add 2 ml of ethyl alcohol
11	Proportional counter	1) Evaporation to complete dryness, Coprecipitation; 2) with acetone; 4) according to ISO 9696:2007
12	Proportional counter	1) Evaporation to complete dryness; 2) An aliquot of the water is evaporated to a small volume and then transferred with a pasteur pipette to a 60mm diameter stainless steel counting planchet; 3) by weighting; 4) We measure as quickly as possible. We let the planchets on the heating block till measurements
13	Proportional counter	1) Evaporation to complete dryness, Coprecipitation; 2) with aceton; 3) 150mg using an analytic balance 4) using ISO 9696:2010 - treatmant with sulphyric acid, than ignition to 350 gradus
14	Liquid-scint. counting	1) Evaporation and mixing sample with LSC cocktail
15	Scintillation counter	1) Evaporation to complete dryness; 2) Aliquot of the residue is transferred to the map planchet so as to provide mass density 2.5 mg / mm ^ 2; 3) Evaporated 50 ml of each sample

16	Proportional counter	1) Evaporation to complete dryness; 2) By homogenization of residue and weighing of 0.2 g of residue onto the planchet.; 3) By homogenization of 0.2 residue onto planchet with 3 ml of water and evaporation to complete dryness
17	Liquid-scint. counting	1) Evaporation and mixing sample with LSC cocktail
18	Proportional counter	1) Evaporation to complete dryness; 2) Automatic evaporation system
19	Proportional counter	1) Evaporation to complete dryness; 2) By adding the sample containing residue in very small amounts onto the planchet and then leaving the planchet for drying under infrared lamp; 4) The sample is leaved to drying in oven for about 2 hours at 105C,then the sample is taken into a descicator and is waited for about 30 minutes.
20	did not report	-
21	(α) Scintillation counter (β) Proportional counter	1) Evaporation to complete dryness, Coprecipitation; 2) Transfering the aliquot concentrate in small portions to a tared planchet, evaporating each portion to dryness; 3) With a intervals of surface density for gross alpha and gross beta
22	Liquid-scint. counting	1) Evaporation and mixing sample with LSC cocktail
23	i-Matic Si-det	1) Coprecipitation, 1a) Procedure from ISO 10704:2009(E) was used; 2) on filter paper (millipore 45 micron); 3) gravimetrically by weighing
24	Proportional counter	1) Evaporation to complete dryness; 2) manualy
25	Proportional counter	1) Evaporation to complete dryness, Coprecipitation; 2) Direct deposition and mechanical homogenization; 4) keep in desiccator until measurement
26	Liquid-scint. counting	1) Evaporation and mixing sample with LSC cocktail
27	(α) Liquid-scint. counting	1) Coprecipitation
28	Liquid-scint. counting	1) Evaporation and mixing sample with LSC cocktail
29	Proportional counter	1) Evaporation to complete dryness; 2) Part of the sample is transfered to the planchet; 3) Measuring the mass of sample in the planchet
30	Proportional counter	1) Evaporation to complete dryness;

		2) Evaporation to complete dryness
31	(β) Proportional counter	1) Evaporation to complete dryness; 2) By scraping and transfer with a special stanless steel spoon; 3) 14.12 - 294.70 mg/cm2 (planchet surface = 4.52 cm2)
32	Liquid-scint. counting	1) Evaporation and mixing sample with LSC cocktail
33	Liquid-scint. counting	1) Evaporation and mixing sample with LSC cocktail
34	(α) Scintillation counter (β) Proportional counter	1) Evaporation to complete dryness
35	Proportional counter	1) Evaporation to complete dryness; 2) 0.28 g
36	Proportional counter	1) Evaporation to complete dryness, Coprecipitation; 2) by using evaporation method
37	Proportional counter	1) Evaporation to complete dryness; 2) After evaporation up to 1 ml, this volume is swept by using between 2-4 ml of HNO3 1M in steps.
38	Proportional counter	1) Evaporation to complete dryness; 2) We transfer exact amount of solid to planchet, then add ethylalcohol and spread homogeneously on plachet; 4) by using annealing before transfering to planchet
39	Liquid-scint. counting	1) Evaporation and mixing sample with LSC cocktail
40	Proportional counter	1) Evaporation to complete dryness, Coprecipitation; 3) by weigh; 4) Sampler are preserved in one desiccator and weighted before and after measuring and the self-absorption correction factor used corresponds to the main weight.
41	Liquid-scint. counting	1) Evaporation and mixing sample with LSC cocktail
42	(α) Liquid-scint. counting	1) Coprecipitation, Evaporation and mixing sample with LSC cocktail
43	(α) grid ionization chamber	1) Evaporation to complete dryness; 2) The residue is mixed with demineralized water and the suspension is coated on the planchet; 3) Usage of a defined amount of the solid from evaporation (900 mg for a 20 cm planchet); 4) Heating up to 450°C and storage in a desicator.

44	Liquid-scint. counting	1a) tal quale
45	Proportional counter	1) Evaporation to complete dryness; 4) according to ISO 9697 and ISO 9696
46	Liquid-scint. counting	1) Evaporation and mixing sample with LSC cocktail
47	LSC	1) Evaporation and mixing sample with LSC cocktail, 1a) Evaporation is applied only to samples with Total Dissolved Solids < 500mg/l
48	(α) Scintillation counter (β) Proportional counter	1) Evaporation to complete dryness; 2) Complete slow evaporation do dryness; 3) weighting the planchet before and after evaporation
49	Scintillation counter	1) Evaporation to complete dryness; 2) in a desiccator; 3) Acording to ISO9696 / 2007, ISO9697/2008 and acording to procedures developed from ISO 17025 / 2005; 4) Acording to ISO9696 / 2007, ISO9697/2009 and acording to procedures developed from ISO 17025 / 2005
50	Liquid-scint. counting	1) Evaporation and mixing sample with LSC cocktail
51	(α) Analyzer MC 2256, mix of powder sample and ZnS(Ag) powder with direct contact with PMT (β) Proportional counter	1) Evaporation to complete dryness
52	Liquid-scint. counting	1) Evaporation and mixing sample with LSC cocktail
53	did not report	-
54	Proportional counter	1) Evaporation to complete dryness; 2) manual drop by drop; 3) measurement of dry deposit mass; 4) standard addition
55	(α) silicon detector (β) Scintillation counter	1) Evaporation to complete dryness; 2) we deposit with microspatula and spread it with TDI spatula

56	Proportional counter	1) Evaporation to complete dryness; 2) we scrape the residue from the capsule, put it on the planchet, make it uniform and fix with acetone; 3) for gross alpha measurement: aprox. 0.015 mg/mm2 ; for gross beta measurement: aprox. 0.115 mg/mm2; 4) beside the tray sulfation and calcination we apply the tray warming plate with adjustable temperature heating
57	(α) Scintillation counter (β) Proportional counter	1) Evaporation to complete dryness, Coprecipitation; 2) The last few mL of sample are evaporated onto the planchet; 4) Routinely, before measurement, planchets are dried at 180ºC 12 hours and keeped in a desiccator during two days.
58	Proportional counter	1) Evaporation to complete dryness; 2) with ethylic alcohol
59	Proportional counter	1) Evaporation to complete dryness; 2) The sample was evaporated directly onto the 200 mm diameter planchet; 3) The planchet was weighed during evaporation.
60	Liquid-scint. counting	1) Evaporation and mixing sample with LSC cocktail
61	Liquid-scint. counting	1a) evaporation to dryness, dissolution and mixing with LSC coctail
62	(α) Scintillation counter (β) Proportional counter	1) Evaporation to complete dryness, 1a) Evaporation and mixing sample with ZnS(Ag); 2) alpha - using water, beta - using acetone; 3) weighting
63	Proportional counter	1) Evaporation to complete dryness; 2) water sample is evaporated to a small volume and transferred quantitatively to stainless steel counting planchet; 3) from the difference im mass of empty planchet and planchet with sample divided by area of planchet; 4) flamed to a red heat if dried solids appear to be noticeably hygroscopic
64	Proportional counter	1) Evaporation to complete dryness
65	Proportional counter	1) Evaporation to complete dryness; 2) By Evaporation; 4) Sample is dried by heating under an infrared lamp until it glows with a characteristic dull red colour to stabilize the mass
66	Proportional counter	1) Evaporation to complete dryness; 2) gravimetrically with methanol distribution; 4) addition of sulfuric acid
67	Proportional counter	1a) according to ISO 9696 and 9697; 2) with ethanol (96%);

		3) according to ISO 9696 and 9697
68	Proportional counter	1) Evaporation to complete dryness
69	semiconductor Si detector	1) Coprecipitation
70	Liquid-scint. counting	1) Evaporation to complete dryness
71	Proportional counter	1) Evaporation to complete dryness
72	Proportional counter	1) Evaporation to complete dryness
73	Liquid-scint. counting	1) Evaporation and mixing sample with LSC cocktail

Reported total dissolved solid contents (TDS)

	Indicative values	Reported total dissolved solids (mg L^{-1})			
		Average	Standard deviation	Minimum	Maximum
Water A	955 ± 44	422	485	0.00304	2100
Water B	364 ± 27	194	202	0.00085	768
Water C	10.1 ± 0.1	35.1	41.8	0.000036	16940

Note: The total dissolved solid content was requested to be reported in mg L^{-1}. Reported total dissolved solids values (mg L^{-1}) are presented as given by the participants.

Water A	
Laboratory code	**TDS (mg L^{-1})**
73	0.00304
32	0.01
2	0.97
57	1.2
40	1.45
5	5.18
3	508
63	567
7	610
65	752
4	770
24	777
41	792
37	794
56	825.211
49	834.8
18	840
61	852
1	863
58	866.67
43	870
21	890
64	900
9	902
46	905
71	908
67	910
27	913
50	919
55	922
36	925
30	930
6	931

35	936
31	942.15
38	952
8	955
17	956.5
11	974
62	979
34	984
48	1001
33	1003
13	1020
29	1025
16	1028
15	1031
45	1033
51	1033.4
47	1070
70	1074
69	1080
60	1092
68	1156
28	1190
10	1242
54	1974
25	2100
42	2100

Water B	
Laboratory code	**TDS mg/L**
73	0.00085
32	0.01
2	0.38
57	0.61
40	0.66
5	4.45
21	32
8	281
65	302
46	310
67	320
41	334
18	340
43	340
24	350
61	350

27	352
9	360
4	360
64	360
31	363.04
50	372
56	374.068
49	376.5
35	378
48	383
38	384
58	386.67
55	388
6	389
30	390
11	392
71	393
1	395
3	398
15	400
17	402.6
16	409
62	416
68	416
33	419
13	420
63	420
51	420.41
70	423
7	429
36	440
45	448
47	452
37	452
34	468
10	498
60	536
29	550
69	560
25	584
42	650
54	688
28	768

Water C	
Laboratory code	**TDS mg/L**
24	0
28	0
73	3.60E-05
2	0.01
57	0.019
40	0.028
60	2
5	2.47
43	3
9	4
27	9
10	9
67	12
65	14.82
7	15
3	18
51	18.14
56	19.56
18	20
4	20
64	20
11	22
6	24
55	25
63	25
8	26
25	26
50	27
70	28
38	28
61	29
33	30
16	30
48	32
49	34
1	34.4
30	35
13	40
68	42
54	43
15	48
36	50
45	50

62	54
58	73.33
17	90.5
34	100
31	117.85
71	123
29	125
42	180
21	366
37	380
69	820
46	1065
47	16940

Appendix 9: Homogeneity study

Water A uranium

Sample ID	**Total uranium activity concentration (mBq/L)**
62	44.6
197	43.1
372	43.1
432	44.1
532	41.5
680	40.1
mean	**42.8**
S_{bb}	**1.7**
U_{bb} (%)	**4**

Water A gross alpha activity concentration.

Sample ID	**Gross alpha activity concentration (mBq/L)**
62	54.7
135	44.6
307	45.2
372	58.1
432	54.8
532	42.1
627	42.1
680	43.1
759	52.7
mean	**48.6**
S_{bb}	**6.4**
U_{bb} (%)	**13.1**

Water A gross beta activity concentration.

Sample ID	**Gross beta activity concentration (mBq/L)**
62	312
135	301
307	314
372	304
432	320
532	311
627	305
680	303
759	308
mean	**308.7**
S_{bb}	**6.1**
U_{bb} (%)	**2.0**

Water B ^{226}Ra activity concentration.

Sample ID	**^{226}Ra activity concentration (mBq/L)**
1	305.5
2	313.4
3	312.2
4	321.5
5	312.4
6	310.9
7	314.6
8	317.8
9	317.2
mean	**313.9**
S_{bb}	**4.6**
U_{bb} (%)	**1.5**

Water C gross alpha activity concentration.

Sample ID	**Gross alpha activity concentration (mBq/L)**
116	950.8
165	993.9
189	933.8
303	906.1
354	936.9
433	916.9
mean	**945.3**
S_{bb}	**32.1**
U_{bb} (%)	**3.40**

Water C gross beta activity concentration.

Sample ID	**Gross beta activity concentration (mBq/L)**
116	1100.6
165	1083.3
189	1112.2
303	1170.7
354	1146.6
433	1090.8
446	1129.5
mean	**1119.1**
S_{bb}	**31.6**
U_{bb} (%)	**2.80**

Appendix 10: Short term stability analysis

Water A	weeks	Gross alpha counts per minute	weeks	Gross beta counts per minute
	0	0.212	0	0.691
	0	0.215	0	0.682
	0	0.201	0	0.739
	8	0.206	0	0.688
	8	0.222	0	0.678
	8	0.196	8	0.808
	20	0.222	8	0.663
	20	0.214	8	0.691
	20	0.228	20	0.643
	28	0.206	20	0.673
	28	0.215	20	0.693
	28	0.224	28	0.704
			28	0.685
			28	0.744

Water B	weeks	Gross alpha counts per minute	weeks	Gross beta counts per minute
	0	3.04		Not determined
	0	3.13		
	0	3.12		
	0	3.15		
	8	3.11		
	8	3.13		
	8	3.14		
	8	3.13		
	20	3.39		

	20	3.13
	20	3.13
	20	3.12
	28	3.11
	28	3.12
	28	3.14

Water C	weeks	Gross alpha activity concentration (mBq/L)	weeks	Gross beta activity concentration (mBq/L)
	0	930.2	5	1100.6
	0	961.7	5	1083.3
	0	941.7	5	1112.2
	5	975.9	17	964.1
	5	1020.7	17	1024.9
	5	958.8	17	1006.6
	17	868.7	21	989.4
	17	909.3	21	1038.4
	17	892.1	21	1010.2
	21	954.2	25	1170.7
	21	896.7	25	1146.6
	21	962.3	25	1090.8

LC-NA-28351-EN-N

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doi:10.2789/573272
ISBN 978-92-79-64622-5