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REIMEP-22 U Age Dating - Determination of the production date of a uranium certified test sample

Inter-laboratory comparison,
Report to participants

Célia Venchiarutti
Zsolt Varga
Stephan Richter
Adrian Nicholl
Judit Krajkó
Rožle Jakopič
Klaus Mayer
Yetunde Aregbe

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Contact information
Dr. Célia Venchiarutti
Address: Institute for Reference Materials and Measurements, Retieseweg 111, 2440 Geel, Belgium
E-mail: celia.venchiarutti@ec.europa.eu
Tel.: +32 (0)14 571 681

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Abstract
The REIMEP-22 inter-laboratory comparison (ILC) "U Age Dating - Determination of the production date of a uranium certified test sample" was organised by JRC-IRMM as support to the Nuclear Forensics International Technical Working Group (ITWG) This ILC was organised prior to the release of the candidate certified reference material IRMM-1000, produced in cooperation with JRC-ITU. The aim of REIMEP-22 was to determine the production date of the uranium certified test sample (i.e. the last chemical separation date of the material) using the disequilibrium between the ^{230}Th-^{234}U and ^{231}Pa-^{235}U nuclides as chronometers. The first was compulsory, the latter optional. Participants in REIMEP-22 received either a 20 mg or 50 mg low-enriched uranium sample of known age in solid uranyl nitrate form, depending on the type of analytical technique they used. Participating laboratories were asked to measure and report either the isotope amount ratio $n(^{230}\text{Th})/n(^{234}\text{U})$ for the 20 mg uranium samples or the activity ratio $A(^{230}\text{Th})/A(^{234}\text{U})$ for the 50 mg uranium samples and to report the calculated production date of the certified test samples. The participants were asked to apply their standard analytical procedures and report the results with the associated uncertainties. REIMEP-22 was announced to participants in June 2013 and fourteen laboratories registered for REIMEP-22 by October 2013. The shipment of the samples to the participants took place between December 2013 and late January 2014. Finally, by May 2014, nine laboratories reported results for the 20 mg uranium sample (using mass spectrometry and reporting amount ratios) and four laboratories for the 50 mg uranium sample (using α-spectrometry and reporting activity ratios). The reported measurement results have been evaluated against the certified reference value by means of zeta-scores in compliance with international guidelines. In general the REIMEP-22 participants' results were satisfactory. This report presents the REIMEP-22 participants' results; including the evaluation of the questionnaire.

REIMEP-22: "U Age Dating - Determination of the production date of a uranium certified test sample"

Inter-laboratory Comparison, Report to participants

January 2015

Célia Venchiarutti[1]
Zsolt Varga[2]
Stephan Richter[1]
Adrian Nicholl[2]
Judit Krajkó[2]
Rožle Jakopič[1]
Klaus Mayer[2]
Yetunde Aregbe[1]

[1] European Commission Joint Research Centre - Institute for Reference Materials and Measurements

[2] European Commission Joint Research Centre - Institute for Transuranium Elements

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Summary

Nuclear forensics is a key element of nuclear security aiming at the identification and characterisation of illicit nuclear material, such as uranium or plutonium, to re-establish the history of the nuclear material of unknown origin. By applying advanced analytical techniques to measure the isotopic compositions, elemental concentrations, chemical impurities and physical dimensions or microstructure of the nuclear material in question, the origin of an unknown material can be determined [1]. More recently, the determination of the "age" of the material has drawn increased interest, not only for nuclear security but also for nuclear safeguards [2].The "age" of a nuclear material refers to its production date, i.e. the time elapsed since the last chemical separation of the daughter nuclides from the mother radionuclide (typically U and Pu) [3,4].This specific signature allows to narrow the possible origins of the material in question and to provide valuable information on its history. In order to answer the emerging need of the nuclear forensic community for a suitable reference material, the European Commission - Joint Research Centre developed a unique uranium reference material (IRMM-1000) certified for the date of the last chemical separation. Certified reference materials, such as the new IRMM-1000, are a prerequisite for a successful validation of measurement procedures. Prior to the release of the IRMM-1000, the JRC organised in cooperation with the Nuclear Forensics International Technical Working Group (ITWG) the REIMEP-22 inter-laboratory comparison entitled "U Age Dating - Determination of the production date of a uranium certified test sample".

The Regular European Inter-laboratory Measurement Evaluation Programme (REIMEP) was established at the JRC Institute for Reference Materials and Measurements (JRC-IRMM) in 1982 to carry out external control of the quality of the measurements for materials characteristic for the nuclear fuel cycle. REIMEP-22 was aimed particularly at the ITWG members, as well as for the Network of Analytical laboratories of the International Atomic Energy Agency (IAEA-NWAL), laboratories from industry or experts in the fields of nuclear and environmental (geological) sciences.

Inter-Laboratory Comparisons (ILC), such as REIMEP-22, give participants the opportunity to benchmark their results against independent and traceable reference values, to identify possible problems, and to improve their measurement procedures. Participants in REIMEP-22 received a 20 mg or 50 mg uranium certified test sample, depending on the applied measurement technique (mass spectrometry or alpha spectrometry, respectively) with an undisclosed value for the production date. The participating laboratories were asked to apply their routine measurement procedures and to report the production date of the material with the associated measurement uncertainty. In addition participants reported the amount or activity ratios for $^{234}U/^{230}Th$ (compulsory) and $^{235}U/^{231}Pa$ (optional). The individual participant results were evaluated against the REIMEP-22 reference value established at JRC-IRMM by means of zeta-scores in compliance with international guidelines.

This report presents the REIMEP-22 participant results and a detailed evaluation of the questionnaire.

1. Introduction

Nuclear forensics supports nuclear security by providing tools for the identification and characterisation of illicit nuclear material, such as uranium or plutonium, to re-establish the history of the nuclear material of unknown origin [1]. Among the different parameters applied for the characterisation of an unknown radioactive or nuclear material, the "age" of the material, is now being determined regularly. The "age" of a nuclear material refers to its production date, i.e. the time elapsed since the last chemical separation of the daughter and parent radionuclides. Validated analytical procedures in combination with a proper estimation of measurement uncertainty [5] are required for a proper characterisation of an intercepted nuclear material to provide legally defendable measurement results. In addition, quality control tools for 'age-dating' have been recently identified also as a priority in nuclear safeguards [6].

The JRC-IRMM is an accredited provider of inter-laboratory comparisons according to ISO/IEC 17043:2010 [7] with a long time experience in organising quality control campaigns for measurements applied in nuclear safeguards and forensics. The Regular European Inter-laboratory Measurement Evaluation Programme (REIMEP) was established in 1982 as an external quality control tool for measurement of uranium and plutonium amount contents and isotope ratios in samples typically found in the nuclear fuel cycle. Previous REIMEP inter-laboratory comparisons involved uranium oxide, uranium in nitric acid solution, uranium in the form of UF_6, plutonium oxide, and others sample types [8].

Prior to the envisioned release of IRMM-1000 in 2015 [9], the JRC-IRMM as support to the Nuclear Forensics International Technical Working Group (ITWG) organised a REIMEP-22 inter-laboratory comparison entitled "U Age Dating - Determination of the production date of a uranium certified test sample" [10] using the prepared uranium age dating reference material. The ITWG is a group of nuclear forensics experts, including nuclear scientists, law enforcement and regulators, formed almost 20 years ago. The ITWG has contributed to the advancements in nuclear forensics through a variety of activities, such as comparative material analysis, table-top exercises (TTX), and providing guidelines for best practices.

The REIMEP-22 certified test samples were prepared at JRC-ITU from low-enriched uranium after a complete separation of thorium decay products at a well-defined time and by monitoring afterwards the ingrowth of the daughter nuclides in the purified material.

REIMEP-22 participating laboratories received a 20 mg or 50 mg uranium certified test sample, depending on the applied measurement technique (mass spectrometric or alpha spectrometry), with an undisclosed value for the production date. The participating laboratories were asked to apply their routine measurement procedures and to report the production date of the material with the associated measurement uncertainty. In addition participants reported the measured amount or activity ratios for $^{234}U/^{230}Th$ (compulsory) and $^{235}U/^{231}Pa$ (optional). Besides the measured and calculated results, participants were also asked to answer a specific questionnaire. The aim of this questionnaire was to obtain detailed information concerning the measurement protocols, the types of instrumentation used for the measurement and the evaluation of measurement uncertainty.

2. Scope and aim

Confidence in the integrity and quality of measurement results is essential in nuclear security, safeguards and forensics. In order to answer the emerging need of the nuclear forensic community for reference materials and validated methods to better characterise seized radioactive or nuclear materials, REIMEP-22 on "U Age Dating - Determination of the production date of a uranium certified test sample" was organised in cooperation with the ITWG, and in compliance with ISO/IEC 17043:2010 [7]. Besides expert laboratories in nuclear forensics, other laboratories that are considering acquiring capabilities in this field were particularly encouraged to participate in REIMEP-22.

The measurand of interest for REIMEP-22 was the production date of the certified test samples. In order to evaluate whether a discrepancy of reported results for the production date with the REIMEP-22 reference value originates from the measurements or from the calculation of the production date, participating laboratories were asked to report in addition either the $n(^{230}\text{Th})/n(^{234}\text{U})$ amount ratio for the 20 mg uranium test samples or the activity $A(^{230}\text{Th})/A(^{234}\text{U})$ ratio for the 50 mg uranium test samples applying their routine analytical procedures. Moreover, the participants had the possibility to report the production date of the sample by measuring the $n(^{231}\text{Pa})/n(^{235}\text{U})$ amount ratio or the $A(^{231}\text{Pa})/A(^{235}\text{U})$ activity ratio. The date of production of the sample had to be reported as dd/mm/yyyy with the associated expanded uncertainty in days.

Participants had also to answer a questionnaire in order to identify future needs for inter-laboratory comparisons. Participants' results were evaluated against the certified reference value established at JRC-IRMM by means of zeta-scores in compliance with ISO 13528:2005 [11].

3. Time frame

REIMEP-22 was announced for participation on June 19, 2013 (see Annex A). The deadline for registration was October 31, 2013. The confirmation of registration was sent to the participants (see Annex B) and subsequently the samples were delivered between December 2013 and January 2014. Due to delays in the shipment for some of the samples, the initial deadline for the reporting of results (March 31, 2014) was extended to May 1, 2014 (see Annex F). By the deadline, three participants could not report their results due to technical problems in their labs.

The characterisation of the uranium material, the homogeneity and short-term stability assessments were carried out as part of the IRMM-1000 certification between July 2012 and October 2013 [9]. The certification was finalised in November 2014 with the realisation of the long-term stability assessment. The REIMEP-22 reference value of the production date for the uranium certified test sample was communicated to the participants during the IAEA international conference on Advances in Nuclear Forensics (CN-218) and the ITWG Annual Meeting in July 2014 [12].

4. Test material

4.1. Preparation of REIMEP-22

The REIMEP-22 certified test samples were prepared in the framework of the production and certification of the IRMM-1000 reference material in compliance with ISO Guide 34 [13]. This material was produced at JRC-ITU from low-enriched uranium (with a relative mass fraction $m(^{235}U)/m(U)$ of 3.6 %) after complete removal of thorium decay products from the original material (i.e. zeroing the initial daughter nuclide concentration at a well-defined time). Afterwards, the ingrowth of the daughter nuclides in the purified material was monitored. The analytical method is described in detail in [9,14], and therefore it is only summarised here briefly.

The separation of the thorium from the uranium was done by extraction chromatography applying TEVA resin (Triskem International, France) and silica gel, in a "sandwiched-column" arrangement. This approach was chosen to allow the separation and removal of protactinium from the uranium material besides the separation of thorium, which was the principal objective of the work. However, the Pa/U separation was not monitored and this chronometer was not applied for the determination of the certified value for the production date. The purified uranium solution was dispensed into pre-cleaned PFA vials, evaporated to dryness and sealed. Finally, 161 units were produced containing 20 mg uranium (distributed as IRMM-1000a) or 50 mg uranium (distributed as IRMM-1000b) as dried uranyl-nitrate. The test samples for REIMEP-22 were selected from the batch of uranium material produced for the candidate reference materials for certification IRMM-1000a and IRMM-1000b (see Certification Report). Fig. 1 shows the dispensing of uranium solution into PFA vials.

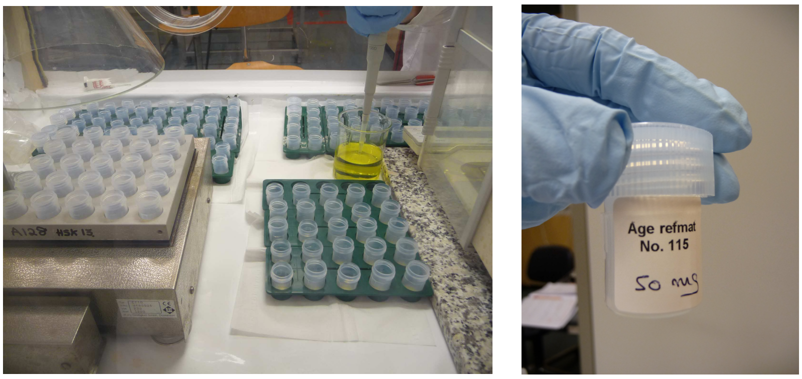

Fig. 1: Dispensing of the purified uranium solution into PFA vials (left) and a 50 mg REIMEP-22 test sample (right).

4.2. REIMEP-22 reference value assignment

4.2.1. Processing of the sample

The reference value is based on reading of the clock at the time of the last chemical separation. This corresponds to the complete removal of the thorium radionuclide from uranium in the original uranyl nitrate. In the case of the production of REIMEP-22, the last chemical separation took place on July 9, 2012 at 11:08 a.m. This production date, REIMEP-22 reference value, is expressed as 09/07/2012 (dd/mm/yyyy) with an expanded uncertainty ($k=2$) in days and is based on the measured $n(^{230}Th)/n(^{234}U)$ amount ratio in the purified sample. The uncertainty of the production date was established in accordance with the *'Guide to the Expression of Uncertainty in Measurement'* [15].

To assess the completeness of thorium removal from the uranium in the original sample, a U/Th separation factor (i.e. the ratio between the U and Th amount in the purified U fraction) of higher than $1{\times}10^7$ was set as target value. Gamma spectrometry measurements of the U fractions were performed for each separation step during the production of the certified test sample to determine the

U/Th separation factor and the effective recovery of uranium. Using the well-resolved γ-peaks of the short-lived ^{234}Th ($T_{1/2}$= 24.1 days) and the ^{235}U, a cumulative U/Th separation factor of (2.8± 0.9) ×10^7 and an overall U recovery of (83.7 ± 0.3) % were confirmed.
The completeness of the removal of thorium from the initial uranium material was additionally confirmed by the measurements of the Th amount content and isotope ratio by ICP-MS in the final purified product (^{232}Th tracer was added to the uranium fraction after the first separation). The final (cumulative) U/Th separation factor was found to be higher than 1.8×10^7. The residual Th concentration in the purified solution was less than 0.01 µg·g^{-1} uranium.

The uncertainty for the characterisation includes the contribution from the date of the last chemical separation (i.e. the time interval bracketing the exact time of the last elution of Th from U) and the contribution from the residual thorium in the final purified uranium material. The uncertainty on the last chemical separation of the Th from the U material was estimated to be 1.5 hours (0.063 days, $k=1$) accounting for the whole elution time for thorium. The uncertainty coming from the residual ^{230}Th was estimated to be less than 80 min or 0.056 days ($k=1$). Therefore the final uncertainty for the characterisation of the certified test sample was 0.17 days ($k=2$).

4.2.2. Confirmation study

Confirmation measurements were carried out after the production of the certified test sample to assess whether the measured age corresponded to the known production date. Six 20 mg units (referred hereafter as series A to F) were randomly selected from the 161 units of REIMEP-22 and dissolved in 2 mL concentrated nitric acid. Several aliquots were prepared for the measurement of the U isotopic composition by TIMS, and the uranium and thorium amount contents by ICP-MS. The chemical separation/purification of uranium from thorium was carried out on a single TEVA column as described in section 4.1 [4,14].
Four thorium aliquots per sample (numbered from 3 to 6) and two independent uranium aliquots were measured to determine the ^{230}Th and ^{234}U amount contents in the samples by IDMS to determine the $n(^{230}\mathrm{Th})/n(^{234}\mathrm{U})$ amount ratios for age confirmation. The 24 ages (six selected units, four aliquots each) and their associated expanded uncertainties ($k=2$) were determined using the GUM Workbench Software [16] and the following equation:

$$t = \frac{1}{\lambda^{234}U - \lambda^{230}Th} \times \ln\left(1 - \frac{n(^{230}Th)}{n(^{234}U)} \times \frac{\lambda^{230}Th - \lambda^{234}U}{\lambda^{234}U}\right)$$

Equation 1

where t is the age of the uranium sample (in years), $\lambda^{234}U$ and $\lambda^{230}Th$ are the decay constants of ^{234}U and ^{230}Th, respectively, calculated from the half-lives ($T_{1/2}$= (245.5 ± 1.2) ×10^3 a and $T_{1/2}$= (75.38 ± 0.3) ×10^3 a, $k = 2$ [17], respectively). The $n(^{230}\mathrm{Th})/n(^{234}\mathrm{U})$ is the measured amount ratio in the sample.

The measurements for all 24 samples were carried out over 3 days. In order to compare all the ages, they were normalised to March 6, 2013; i.e. the date of the separation of the first series A. The values were then converted into production dates. As can be seen from Fig. 2, a good agreement between the calculated production dates and the reference value was achieved for all 24 ages. This also confirmed the successful separation of the thorium from the uranium in the initial material during the production of the uranium reference material (see also sections 4.3 and 4.4).

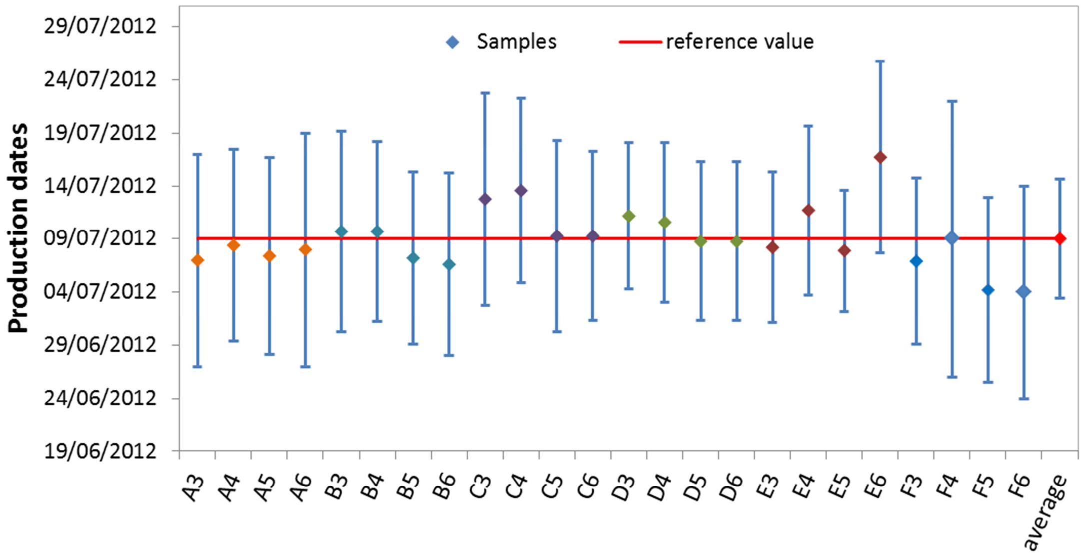

Fig. 2: Production dates with the associated expanded uncertainties (*k=2*) for the confirmation study of REIMEP-22. The reference value (09/07/2012) with its expanded uncertainty of 0.17 days, *k=2* is represented by the red line.

4.3. Homogeneity

4.3.1. Set-up of homogeneity study

The homogeneity assessment was carried out in compliance with ISO Guide 35:2006 [18] and the IUPAC International Harmonized Protocol for the Proficiency Testing of Analytical Chemistry Laboratories [19]. The number of selected units corresponds to approximately the cubic root of the total number of units produced. Five units of 20 mg uranium sample and five of 50 mg uranium sample were selected for the between-unit homogeneity assessment [11,18], using a random stratified sampling scheme covering the whole batch. The analytical procedure was the same as described in the section 4.2. Three thorium aliquots per sample were measured by ICP-MS in a randomised order. As the chemical separations were performed over consecutive days, the separation date for the first series (i.e. October 16, 2013) was chosen as the reference date for the comparison of the ages in all samples measured for the homogeneity assessment. These ages were then compared to the "known age", meaning the time elapsed between the production of the REIMEP-22 certified test samples and the date of the chemical separation for the first series carried out on 16 October 2013. Fig. 3 shows the average ages per unit in a chronological order of the ICP-MS measurements. The average age values for all ten samples agreed well with the known age, and therefore confirmed the homogeneity of the whole batch of REIMEP-22 certified test samples.

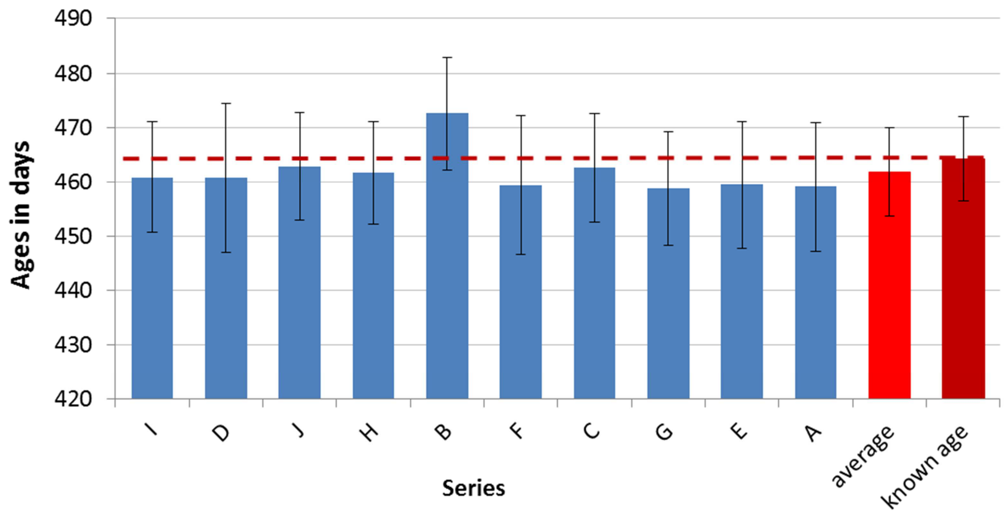

Fig. 3: Calculated average age for the individual series selected for the homogeneity study (blue), their average (red) and the known age based on the time elapsed since the production date with their respective expanded uncertainties (*k=2*).

4.3.2. ANOVA analysis and homogeneity results

The final evaluation of the homogeneity study was carried out using a one-way analysis of variance (ANOVA) as presented in Table 1.

The ANOVA analysis allows the separation of the method variation (s_{wb}) from the experimental averages over the replicates measured in one bottle and the determination of the real variation between bottles (s_{bb}). Moreover, it calculates u^*_{bb}, i.e. the lower limit of the between bottle variance which depends on the mean squares within bottles, the number of replicate measurements per bottle and the degrees of freedom of the mean squares within bottles. It can be understood as the "detection limit" of the homogeneity study. Consequently, the uncertainty of homogeneity, noted u_{bb}, can be estimated either as s_{bb} or as u^*_{bb} in case of $s_{bb} < u^*_{bb}$.

The method repeatability ($s_{\mathrm{wb,rel}}$), the between-unit standard deviation ($s_{\mathrm{bb,rel}}$) and $u^*_{\mathrm{bb,rel}}$ were calculated as:

$$s_{wb,rel} = \frac{\sqrt{MS_{within}}}{\bar{y}}$$ **Equation 2**

$$s_{bb,rel} = \frac{\sqrt{\dfrac{MS_{between} - MS_{within}}{n}}}{\bar{y}}$$ **Equation 3**

$$u^*_{bb,rel} = \frac{\sqrt{\dfrac{MS_{within}}{n}}\sqrt[4]{\dfrac{2}{\nu_{MSwithin}}}}{\bar{y}}$$ **Equation 4**

MS_{within} mean square within a unit from an ANOVA

MS_{between} mean squares between-unit from an ANOVA

$\bar{y}$	mean of all results of the homogeneity study
n	mean number of replicates per unit
$v_{MSwithin}$	degrees of freedom of MS_{within}

This approach, applying single factor ANOVA, as described in [20], is compliant with ISO Guide 35:2006 [18], the IUPAC Harmonized Protocol, and is similar to tests determining whether an ILC material is sufficiently homogeneous for its purpose as described in ISO 13528 [11]. In the end, these tests compare the unit heterogeneity with the standard deviation for proficiency assessment ($\hat{\sigma}$). In the case of REIMEP-22, the assessment criterion for the homogeneity check was defined as s_{bb} (or u^*_{bb}) ≤ $0.3 \cdot \hat{\sigma}$, where the criterion $\hat{\sigma}$ was set to 5 % of the known age at the time of the homogeneity study, i.e. 464.2 days on October 16, 2013 as described in section 4.3.1 and reported in Table 1.

Table 1: Results for the homogeneity assessment for REIMEP-22

REIMEP-22	Ages[a] [days]		
Selected Units	Aliquot 1	Aliquot 2	Aliquot 3
A	460.3	456.6	459.7
B	472.5	471.9	473.1
C	463.2	460.1	465.6
D	460.1	458.9	462.6
E	459.4	458.9	461.2
F	456.3	461.2	461.8
G	455.7	460.6	460.0
H	461.0	462.2	461.6
I	460.8	461.4	460.8
J	464.6	464.0	460.4
Mean or x_s[b] [days]	461.9		
Known age [c] [days]	464.2		
$\hat{\sigma}$ [days]	23.2		
$0.3 \cdot \hat{\sigma}$ [days]	7.0		
s_{bb} [days]	3.9		
s_{wb} [days]	2.0		
u^*_{bb} [days]	0.6		
u_{bb} [days]	3.9		
s_{bb}, (u_{bb}) $< 0.3 \cdot \hat{\sigma}$	YES		
y_s[b]	468		
$\lvert x_s - y_s \rvert \leq 0.3\,\hat{\sigma}$[b]	YES		

[a] Note that the results were presented as ages in days and not as production dates

[b] See section 4.4 on the stability assessment for definitions of x_s and y_s and stability assessment criteria.

[c] The known age corresponds to the elapsed time between the production date and the date of the homogeneity study (October 16, 2013).

As a result, the REIMEP-22 certified test samples were considered sufficiently homogeneous for the purpose of this inter-laboratory comparison.

4.4. Stability

The 'short-term' stability assessment result was combined with the homogeneity study (see section 4.2). The long-term stability study was carried out as part of the certification of IRMM-1000 two years after the production. Two 20 mg uranium samples were selected and analysed by TIMS and ICP-MS at JRC-ITU following the same analytical procedures as described in section 4.3.1. Methods to assess whether an ILC material is sufficiently stable for its purpose are described in ISO 13528 [11,21]. These tests compare the general average of the measurand (here, the age) obtained during the homogeneity check noted x_s (461.9 days as can be seen in Table 1) with that obtained during the stability check, noted y_s, and which corresponds to 468.0 days once normalised to the reference date of the homogeneity assessment (see Section 4.3.2). The absolute difference of these averages is then compared to the standard deviation for proficiency assessment $\hat{\sigma}$ (Section 4.3.2 and Table 1), using the assessment criterion for the stability check $|x_s-y_s| \leq 0.3\hat{\sigma}$, as defined in ISO 13528 [11].
For the long-term stability assessment, no significant difference was observed between the reference value and the results of the long-term stability measurements within their expanded uncertainties as can be seen in Fig. 4.

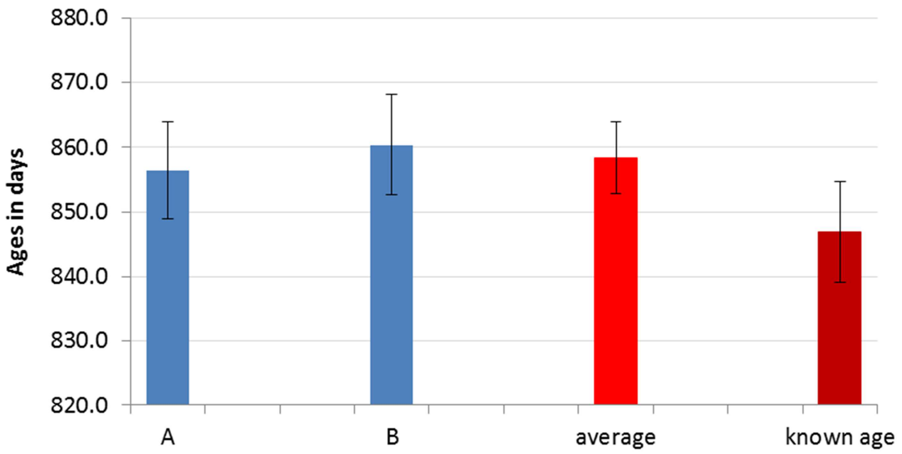

Fig. 4 : Calculated average age for the individual samples selected for the long-term stability study (blue), their average (red) and the known age based on the time elapsed since the production date with their respective expanded uncertainties (*k=2*)

Finally, long term stability of the REIMEP-22 samples was successfully proven, and the assessment criterion $|x_s-y_s| \leq 0.3\hat{\sigma}$ was met, as seen in Table 1.

5. Participant invitation, registration, distribution and information

REIMEP-22 was announced for participation in relevant conferences and meetings convened by international organisations (IAEA, ESARDA, INMM, CETAMA) and on the IRMM website (Annex A and [10]). Participants had to register electronically using the MILC online server, sign the confirmation form and send it to the organisers as pdf per email or fax (Annex A). Subsequently the REIMEP-22 coordinator confirmed their participation (Annex B). The REIMEP-22 certified test samples were shipped to the participants by JRC-IRMM between December 2013 and January 2014 as a nuclear material in exempted quantities. Participants had to provide the necessary documentations in order to obtain the license for the transport.

Participants received a package with either a 20 mg or 50 mg uranium certified test sample with accompanying letters on general instructions and their personal participation keys to access the result reporting page (Annex C). Upon receipt of the sample(s), participants had to return via email or fax the signed 'Confirmation of sample receipt' (Annex D). In addition, detailed guidelines were also enclosed with the sample to help participants with the online reporting tool (Annex E).

Fourteen laboratories registered for REIMEP-22, with two laboratories registering for both, the 20 mg and 50 mg uranium certified test samples. Ten laboratories registered for the 20 mg uranium sample and six laboratories for the 50 mg sample. The number of participants per country is shown in Fig. 5.

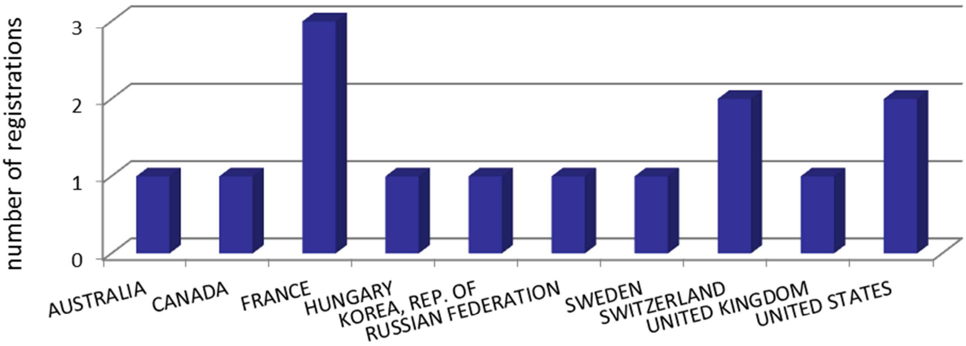

Fig. 5: Number of participants in REIMEP-22 per country

6. REIMEP-22 reference value

The REIMEP-22 reference value X_{ref} (i.e. the production date based on the $^{230}Th/^{234}U$ radiochronometer) and its associated expanded uncertainties U_{ref} (k=2) at the time of the REIMEP-22 ILC are given in Table 2:

Table 2: REIMEP-22 reference value for the production date with its uncertainty

REIMEP-22	Production date	
	X_{ref} [1)] [dd/mm/yyyy]	U_{ref} [2)] [day]
based on $n(^{230}Th)/n(^{234}U)$	09/07/2012	7.8

[1)] The reference value is the production date, i.e. the date of the last chemical separation between ^{230}Th and ^{234}U.

[2)] The uncertainty on the reference value is traceable to the International System of Units (SI). corresponds to the expanded uncertainty with a coverage factor *k=2*, i.e. to a level of confidence of about 95 % estimated in accordance ISO/IEC Guide 98-3, Guide to the Expression of Uncertainty in Measurement, ISO, 2008 [15]. Note that this is not the final uncertainty as it will appear on the certificates for the certified reference materials IRMM-1000a and IRMM-1000b. More details can be found in the certification report of the IRMM-1000a and IRMM-1000b.

7. Reported results

7.1. General observations

Among the fourteen laboratories who registered for REIMEP-22, three could not report their results because of technical problems. Finally, eleven different laboratories reported results; among those, two laboratories submitted results for 20 mg and 50 mg uranium certified test samples, making thirteen participant results in total. Nine participants reported results for the 20 mg sample and four participants reported results for the 50 mg sample. Additionally, two laboratories reported the production dates based on the $n(^{231}\text{Pa})/n(^{235}\text{U})$ amount ratios.

7.2. Measurement results

Participants in REIMEP-22 had to report the isotope amount ratios $n(^{230}\text{Th})/n(^{234}\text{U})$, $n(^{231}\text{Pa})/n(^{235}\text{U})$ or activity ratios $A(^{230}\text{Th})/A(^{234}\text{U})$, for three replicates, for the reference date of March 6, 2013 (this reference date for all reported ratios was compulsory in order to compare the measurement results of the participants without any data manipulation by the ILC organisers). Furthermore, they were requested to report the average of these three measured amount or activity ratios and the calculated production date with respective uncertainty. The participants' results are presented in Fig. 6- 8 and Tables 3- 4. All the results are displayed as reported by the participants.

From the Fig. 6, it can be seen that labs 10242 and 10243 may not have reported the average $n(^{230}\text{Th})/n(^{234}\text{U})$ amount ratio for the reference date of March 6, 2013, since the reported values are not following the correct trend/relationship between $n(^{230}\text{Th})/n(^{234}\text{U})$ amount ratio and production date. From Tables 3- 4, it can be seen that the reported uncertainties for the activity ratios measured with alpha-spectrometry are generally larger than those for amount ratios measured with mass spectrometry.

The reporting of the $n(^{231}\text{Pa})/n(^{235}\text{U})$ amount ratios or $A(^{231}\text{Pa})/A(^{235}\text{U})$ activity ratios was optional since the REIMEP-22 samples were not certified for the production date based on Pa-U chronometer. Therefore, the reference value of 09/07/2012 (July 9, 2012) in Fig. 8 is only given as indicative value to allow an evaluation of the participant performance based on the measurement of the $n(^{231}\text{Pa})/n(^{235}\text{U})$ amount ratios. The participant (lab code 10246) reported a single value for $n(^{231}\text{Pa})/n(^{235}\text{U})$ amount ratio due to analytical problems, therefore the reported production date is based only on one replicate measurement.

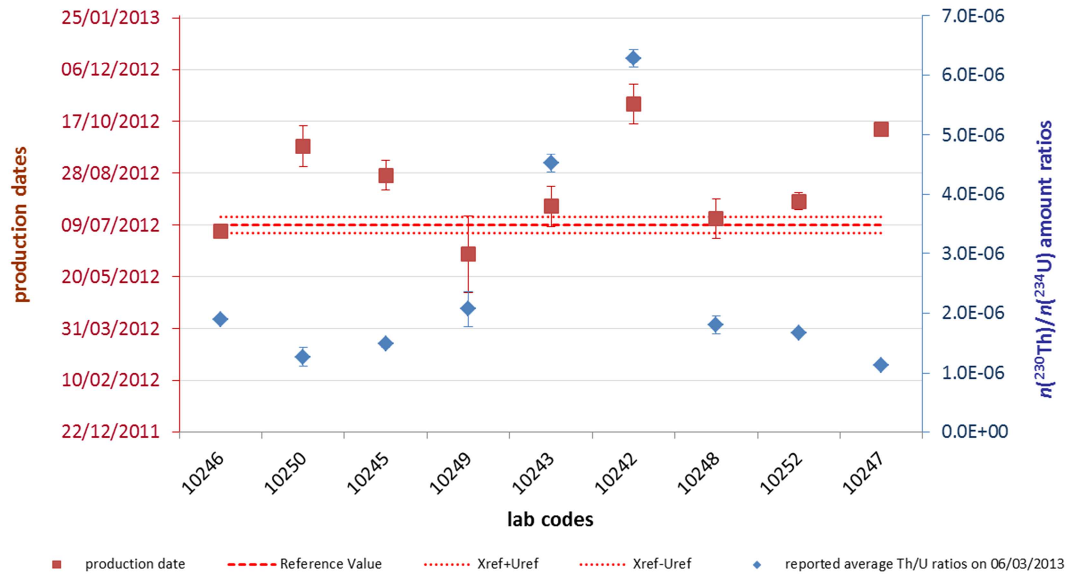

Fig. 6: Reported results for the 20 mg uranium certified sample with uncertainties for production date (red squares) and $n(^{230}Th)/n(^{234}U)$ amount ratios (blue diamonds) normalised to March 6, 2013 (reference date) The reference value and its uncertainty are shown by the dashed red lines.

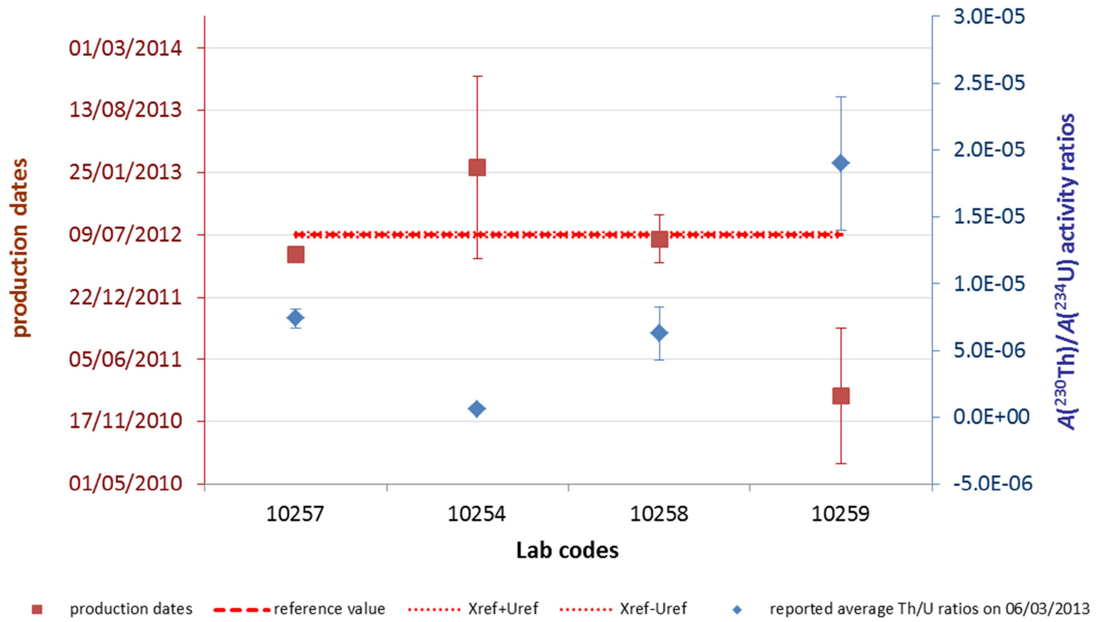

Fig. 7: Reported results for the 50 mg uranium certified sample with uncertainties for production date (red squares) and $A(^{230}Th)/A(^{234}U)$ activity ratios (blue diamonds) normalised to March 6, 2013 (reference date).The reference value and its uncertainty are shown by the dashed red lines. The average $A(^{230}Th)/A(^{234}U)$ activity ratio reported by lab 10254 is not plotted with its associated uncertainty since there was a mistake in the reported uncertainty.

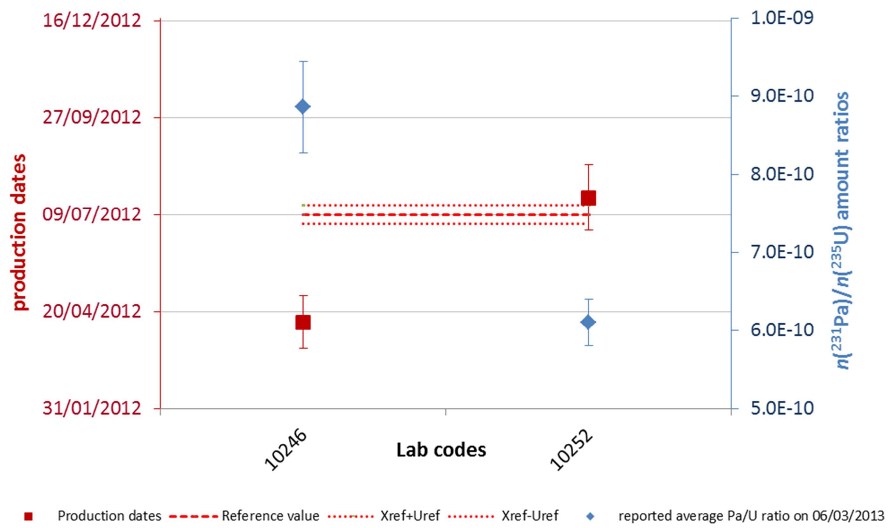

Fig. 8: Reported results for the 20 mg uranium certified sample with uncertainties for production date (red squares) and $n(^{231}Pa)/n(^{235}U)$ amount ratios (blue diamonds) normalised to March 6, 2013 (reference date). The reference value and its uncertainty are shown by the dashed red lines.

Table 3: REIMEP-22 reported results for the 20 mg uranium test sample analysis

REIMEP-22	Average ratios (of 3 replicates)			Production dates		
Lab codes	$n(^{230}Th)/n(^{234}U)\cdot 10^{-6}$	U (10^{-7})	*k*	*dd/mm/yyyy*	U days	*k*
10246	1.90	0.42	2	03/07/2012	5.5	2
10250	1.27	1.53	2	23/09/2012	20	2
10245	1.48	0.55	2	26/08/2012	14	2
10249	2.07	2.9	2	11/06/2012	37	2
10243	4.53	1.50	2	27/07/2012	19	2
10242	6.28	1.46	1	03/11/2012	19	1
10248	1.81	1.5	2	15/07/2012	19	2
10252	1.68	0.23	1	01/08/2012	8	1
10247	1.135	0.37	2	10/10/2012	4.8	2

Table 4: REIMEP-22 reported results for the 50 mg uranium test sample analysis

REIMEP-22	Average ratios (of 3 replicates)			Production dates		
Lab codes	$A(^{230}Th)/A(^{234}U)\cdot 10^{-6}$	U (10^{-6})	*k*	*dd/mm/yyyy*	U days	*k*
10257	7.40	0.74	2	07/05/2012	25	1
10254	0.58	X	2	11/02/2013	292	2
10258	6.3	2.0	2	26/06/2012	77	2
10259	19	5	2	08/02/2011	217	2

Note that compared to the uncertainty reported for the $n(^{230}Th)/n(^{234}U)$ in Table 3, the uncertainty for the $A(^{230}Th)/A(^{234}U)$ in the table above is at 10^{-6}. The X indicates that there is a mistake in the reported uncertainty.

8. Scoring of results

8.1. The scores and their settings

The evaluation of the laboratory performance was done by means of zeta scores in accordance with ISO 13528 [11]:

$$zeta = \frac{x_{lab} - X_{ref}}{\sqrt{u_{ref}^2 + u_{lab}^2}}$$

Where

x_{lab} is the measurement result reported by a participant

X_{ref} is the certified reference value (assigned value)

u_{ref} is the standard uncertainty of the reference value

u_{lab} is the standard uncertainty reported by a participant

The laboratory performance expressed as zeta scores can be interpreted as: satisfactory for zeta score ≤ 2 (green), questionable for 2 < zeta score ≤ 3 (yellow) and unsatisfactory for zeta score > 3 (red), see Table 5 and Table 6. This score provides an indication of whether the estimate of the uncertainty is consistent with the laboratory's deviation from the reference value as given in section 6. It is calculated only for the results that were accompanied by an uncertainty statement. An unsatisfactory laboratory performance may be caused by an underestimated uncertainty or by a large deviation from the reference value. Since all the laboratories participating in REIMEP-22 reported uncertainties with a coverage factor (*k*), the standard uncertainty of the laboratory (u_{lab}) was calculated as the reported uncertainty divided by the coverage factor.

8.2. Scoring the reported measurement results

Table 5 and Table 6 list in detail the zeta scores per participant as described in Section 8.1.

Table 5: Overview of the zeta scores for REIMEP-22 20 mg uranium certified test sample

	zeta scores	
Lab codes	$n(^{230}Th)/n(^{234}U)$	$n(^{231}Pa)/n(^{235}U)$ [(a)]
10246	**1.3**	**7.6**
10250	**-7.1**	-
10245	**-6.0**	-
10249	**1.5**	-
10243	**-1.8**	-
10242	**-6.0**	-
10248	**-0.6**	-
10252	**-2.6**	**-0.5**
10247	**-20.4**	-

(a) Note that to calculate zeta scores for the evaluation of the $n(^{231}Pa)/n(^{235}U)$ reporting, the REIMEP-22 reference value for the production date based on $n(^{230}Th)/n(^{234}U)$ was used, although it is only an indicative value in the case of production dates based on $n(^{231}Pa)/n(^{235}U)$.

Table 6: Overview of the zeta scores for REIMEP-22 50 mg uranium certified test sample

Lab codes	Zeta scores $A(^{230}Th)/A(^{234}U)$
10257	**2.5**
10254	**-1.5**
10258	**0.3**
10259	**4.8**

Six out of the thirteen participants obtained zeta scores ≤ 2 for the measurements of the $^{230}Th/^{234}U$ ratios. Two participants obtained 2 < zeta score ≤ 3 and five zeta scores > 3.
There is no reference value based on Pa/U chronometer for the evaluation of the $n(^{231}Pa)/n(^{235}U)$ ratio results, however the reference value for the production date based on the $n(^{230}Th)/n(^{234}U)$ can be used as indicative value, especially when one takes into account, see also Fig. 8, the good agreement of the reported production date based on the $n(^{231}Pa)/n(^{235}U)$ ratio with the REIMEP-22 reference value based on the $n(^{230}Th)/n(^{234}U)$ ratio. Applying this approach to the two participants who reported $n(^{231}Pa)/n(^{235}U)$ results, one participant obtained a satisfactory zeta-score.
However, one has to bear in mind when evaluating the measurement performance that REIMEP-22 was the first REIMEP of its kind on the determination of production date. It was particularly challenging for some participants, because of the very low Th content in the young certified test material. In general, most of the seized materials analysed by nuclear forensics laboratories are older samples compared to the REIMEP-22 samples. Therefore, participants had to adapt, and sometimes develop new analytical procedures to analyse the REIMEP-22 samples. Moreover, the age determination based on the Pa/U ratio measurement is not routinely performed on nuclear samples, since it is most often based on the Th/U chronometer [22]. In this context, it can be concluded that REIMEP-22 participants performed reasonably well for the measurements of $n(^{230}Th)/n(^{234}U)$,$A(^{230}Th)/A(^{234}U)$ and of $n(^{231}Pa)/n(^{235}U)$ ratios.

9. Further information extracted from the results

The participants were asked to answer questions (see Annex GAnnex H) related to the analytical and measurement protocols applied for the analysis of the REIMEP-22 samples. The answers to the questionnaire are discussed in the sections 9.1 to 9.6.

9.1. A representative study

The mission of most of the laboratories is to carry out measurements for fissile material control or safeguards (36.4 %) or they are from the field of research and development in Nuclear and Earth sciences (54.6 %). Many of these laboratories also perform regularly measurements of radioactivity in the environment (45.5 %). A few of them carry out measurements for the regular monitoring of nuclear facilities (9.1 %). Among the participating laboratories, six are part of the ITWG and are involved in the analysis of nuclear forensics samples.

9.2. Method of analysis

Four participants indicated that the REIMEP-22 sample was not treated according to their routine analytical procedures due to the low amount of thorium present in the sample or because they did not yet have an analytical procedure for this kind of samples.

Three laboratories using mass spectrometry technique did not perform chemical separation prior to measurements. Others applied a chemical treatment for the thorium analysis by dissolving the samples in nitric acid or hydrochloric acid, followed by a separation using TEVA extraction chromatography, anion exchange or by co-precipitation. In most cases the uranium was measured without prior separation.

All the participants applied Isotope Dilution Mass Spectrometry (IDMS) for the determination of the Th and U amount in the samples. Seven of the participants used Multi- Collector Inductively Coupled Plasma Mass Spectrometry (MC-ICP-MS) and two labs used Sector Field Inductively Coupled Plasma Mass Spectrometry (SF-ICP-MS). For the measurement of the 50 mg uranium certified samples, all the participants used alpha spectrometry. However, two labs applied a combination of alpha spectrometry and Thermal Ionisation Mass Spectrometry (TIMS).

Four laboratories stated that they are experienced in the Th/U measurement and perform between 11 and 50 measurements a year; analysing mainly forensics, safeguards, environmental samples and reference materials. One participant did not have any experience in the analysis of Th/U samples.

Only one participant (lab code 10246) of the two who reported results based on the Pa/U analysis indicated to be experienced in such type of analysis mostly for forensics samples and reference materials, performing between 50-100 sample measurements per year. Most of the other participants did not report the results for Pa/U analysis because of the lack of appropriate (validated) methods, the unavailability of a ^{233}Pa spike for IDMS or because of time constraints.

The amounts of sample analysed per replicate measurement are listed in Annex I 1and Annex J 1.

9.3. Quality system

Some participating laboratories are appointed by governmental authorities to act as reference laboratory for a specific topic, these laboratories are authorised. Others document their working approaches according to ISO 9001: 2008 [23], they undergo external audits to check the compliance, and therefore they are certified. Seven laboratories reported that they work according to a quality management system: three participants according to ISO 17025 [24] (they are therefore accredited), three according to ISO 9000 series [25] (they are therefore certified) and one according to both. One participant stated to work according to an internal quality control system and three others reported that quality systems were not applicable to their laboratories.

Nine participants confirmed that they participate in various inter-laboratory comparisons, among them, eight participate regularly in ILCs organised by JRC-IRMM such as REIMEP and NUSIMEP. The other ILC schemes mentioned were those organised by the IAEA, the CEA (EQRAIN, CETAMA), NATO, the DOE or NBL ILCs

9.4. Use of standards

All the participants routinely use certified reference materials mostly for instrument calibration and for method validation. Six out of the eleven participating laboratories use regularly IRMM certified reference materials. The specific certified reference materials used by the participants for the analysis of the REIMEP-22 samples are given in Annex I 2 and Annex J 2.

For mass spectrometry techniques, CRMs and in-house standards are used for instrument calibration, monitoring of mass fractionation and abundance sensitivity, and for IDMS. It seems that CRMs are only used for the quantification of analytes in the spikes (e.g. certification of in-house spikes). For α-spectrometry, two participants reported using CRMs and in-house standards for instrument calibration, for the isotope dilution and for the quantification of the analytes in the spikes.

9.5. Determination of measurement uncertainty

All participants except one stated that they routinely report measurement uncertainties to their customers.

Nine out of the eleven participating laboratories estimated the uncertainties according to the Guide for Quantifying Measurement Uncertainty (GUM) [15] issued by the International Organization for Standardization (ISO, 2005) and/or EURACHEM/CITAC (2000) [26]. Five participants reported expanded uncertainties with a coverage factor k of 2, and four participants reported standard uncertainties.

Two participants estimated their measurement uncertainty using another standard than GUM for the quantification of uncertainty (State Standard R-ISO-5725-2-2002) or by propagating the analytical uncertainties with $k=2$ and using a Student's factor for the average.

The detailed lists of the major uncertainty contributors to the uncertainty for the participant results are given in Annex I 3 and Annex J 3.

9.6. Half-lives and molar masses used for REIMEP-22 age determination

REIMEP-22 participants were asked to report the half-lives (in years) and molar masses ($g \cdot mol^{-1}$) with associated uncertainties that they applied in their calculations for the production date. The half-lives and molar masses are presented in Annex I 4 - Annex J 4 and Annex I 5 -Annex J 5, respectively.

From the reported half-lives for the 20 mg uranium certified test sample (Annex I 1), it can be seen that lab 10243 did not report half-lives, but rather the respective decay constants λ.

Moreover, in Annex I 4 - Annex J 4, it can be seen that for similar half-lives, different expanded uncertainties were reported by the participants. Moreover, molar masses ($g \cdot mol^{-1}$) which were used mainly for the production date calculation based on the $A(^{230}Th)/A(^{234}U)$ activity ratios were also reported (see Annex I 5 - Annex J 5) with different uncertainties for the same molar mass value. These differences in the reported uncertainties for the half-lives and molar masses may indicate possible sources of errors in the values used by the participants for the calculation of the production dates.

10. Feedback and Outlook on future REIMEP ILCs

Some REIMEP-22 participants stated that the amount of thorium in REIMEP-22 samples was much lower compared to their routine samples.

All the participants expressed interest in future REIMEP ILCs dedicated to age dating. Some participants expressed that they would be interested in samples similar to REIMEP-22 but preferably older samples, i.e. with higher ^{230}Th amount content.

Most of the participants are interested in analysing uranium, plutonium or thorium samples, some are interested in protactinium and americium as well; and in different matrixes: similar to real samples, oxides (U_3O_8), reprocessed and environmental samples. They would like to participate in ILCs on age dating using different clocks such as Th/U, Pu/Am, Pu/U and Pa/U.

Participants who have not reported results for the production date based on the ^{235}U/^{231}Pa were encouraged to do so using the remaining amount of REIMEP-22 sample and communicate their results to JRC-IRMM.

11. Conclusion

Accurate determination of the production date of a radioactive or nuclear material, with uncertainties preferably within days, is of utmost importance for establishing the origin of illicit nuclear material. Therefore, measurements have to be reliable, with demonstrated uncertainty and traceability to the SI and within uncertainties fit for intended purpose. Strict quality controls need to be applied to ensure confidence in those measurement results. The provision of quality control tools for conformity assessment directly contributes to the effectiveness of nuclear forensics and safeguards systems.

For REIMEP-22, two sizes of the uranium certified test samples were provided to the participants, 20 mg and/or 50 mg with an undisclosed value for the production date. The challenge in REIMEP-22 was to successfully separate ^{230}Th and ^{234}U (and optionally ^{231}Pa and ^{235}U) in the samples with a high chemical recovery in order to determine the date of the last separation of the daughter from the parent radionuclide.

Considering the relatively young age of the certified test sample, it can be concluded that the participants in REIMEP-22 performed well for the measurements of amount and activity ratios; however, the spread of results was larger for the activity ratio results measured by alpha spectrometry. Finally, out of thirteen reported results, six participants obtained satisfactory zeta scores and two participants obtained questionable zeta scores. The results confirmed the analytical capabilities of laboratories for this type of measurements. However, it also showed that more care still needs to be brought in the estimation of measurement uncertainties, which were generally underestimated for REIMEP-22.

Moreover, two participants reported $n(^{231}\text{Pa})/n(^{235}\text{U})$ amount ratios and the associated production dates. In one case the reported values agreed well with the reference value based on the ^{230}Th/^{234}U within its uncertainty. This is already a good indication that the certified production date of IRMM-1000 can also be used as indicative value for the Pa/U chronometer, although IRMM-1000 will not be certified for this specific chronometer.

Different half-lives and molar masses were used by the participants for the calculations. Moreover, there seems to be two different groups of reported half-lives based on mainly two bibliographic references [17,27]. These could possibly be sources of error in the evaluation of the uncertainty calculation on some of the production dates.

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Abbreviations

ANOVA	Analysis of Variance
CETAMA	Commission d'Etablissement des Méthodes d'Analyse
CITAC	Co-operation for International Traceability in Analytical Chemistry
CRM	Certified Reference Material
DOE	US Department of Energy
EQRAIN	Evaluation de la Qualité des Résultats d'Analyse dans l'Industrie Nucléaire
ESARDA	European Safeguards Research and Development Association
EU	European Union
EURACHEM	A focus for Analytical Chemistry in Europe
EURATOM	European Atomic Energy Community
GUM	Guide to the Expression of Uncertainty in Measurement
HEU	High Enriched Uranium
IAEA	International Atomic Energy Agency
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
IDMS	Isotope Dilution Mass Spectrometry
ILC	Inter-Laboratory Comparison
ISO	International Organization for Standardization
ITV	International Target Value
ITWG	Nuclear Forensics International Technical Working Group
IUPAC	International Union for Pure and Applied Chemistry
JRC	Joint Research Centre, European Commission
JRC-IRMM	Institute for Reference Materials and Measurements, JRC, European Commission
JRC-ITU	Institute for Transuranium Elements, JRC, European Commission
LEU	Low enriched uranium
MC-ICP-MS	Multi-Collection Inductively Coupled Plasma Mass Spectrometry
NATO	North Atlantic Treaty Organisation
NBL	New Brunswick Laboratory
NUSIMEP	Nuclear Signatures Inter-laboratory Measurement Evaluation Programme
REIMEP	Regular European Inter-laboratory Measurement Evaluation Programme
SF-ICP-MS	Sector Field Inductively Coupled Plasma Mass Spectrometry
TIMS	Thermal Ionisation Mass Spectrometry

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Annexes

Annex A Announcement letter

EUROPEAN COMMISSION
JOINT RESEARCH CENTRE

Institute for Reference Materials and Measurements

REIMEP

Geel, 19 June 2013
JRC.D.4/SR/CV/ccp/D-ARES(2013) 2326503

The IRMM Regular European Interlaboratory Measurement Evaluation Programme

REIMEP-22: Interlaboratory Comparison on U Age Dating

The determination of the production date of a uranium certified test sample

The Regular European Interlaboratory Measurement Evaluation Programme (REIMEP) was started by IRMM in 1982 to carry out external control of the quality of the measurements of the nuclear fuel cycle materials. In REIMEP campaigns, samples matching materials analysed routinely in the nuclear fuel cycle are sent throughout the world to participating laboratories for measurements.

We would like to announce the forthcoming REIMEP-22 interlaboratory comparison: "U Age Dating - Determination of the production date of a uranium certified test sample" and invite laboratories to participate.

The determination of the production date of a uranium material (i.e. the last chemical separation date of this material) can be based either on the disequilibrium between the two nuclides ^{230}Th-^{234}U or between the two nuclides ^{231}Pa-^{235}U.

Participants in REIMEP-22 will receive one low-enriched uranium sample (~ 4%) containing either 20 mg or 50 mg of uranium. The sample is in solid uranyl-nitrate form.

Depending on the type of technique used by the participating laboratory to measure the sample, either a 20 mg or a 50 mg U sample will be dispatched to the laboratory. The participants planning to use mass spectrometric methods will receive a 20 mg U sample, whereas laboratories planning to use radiometric methods (e.g. alpha-spectrometry) will receive a 50 mg U sample.

Therefore, in order to determine the production date of the material, participating laboratories will be asked to measure and report either the isotope amount ratio $n(^{230}\text{Th})/n(^{234}\text{U})$ for the 20 mg uranium samples or the activity ratio $A(^{230}\text{Th})/A(^{234}\text{U})$ for the 50 mg uranium samples and to report the calculated production date of the certified tests sample (compulsory).

Moreover, the participants will have the possibility as well (optional) to report the date of production of the sample by measuring the isotope amount ratio $n(^{231}\text{Pa})/n(^{235}\text{U})$ for the 20 mg uranium sample or the activity ratio $A(^{231}\text{Pa})/A(^{235}\text{U})$ for the 50 mg uranium sample.

The isotope amount ratio/activity ratios, $n(^{230}\text{Th})/n(^{234}\text{U})$, $A(^{230}\text{Th})/A(^{234}\text{U})$ or $n(^{231}\text{Pa})/n(^{235}\text{U})$, $A(^{231}\text{Pa})/A(^{235}\text{U})$, are to be measured by participating laboratories using their routine analytical procedures. The date of production of the sample should be reported as follows: dd/mm/yyyy and the associated expanded uncertainty should be reported in ± days.

The measurement results will be evaluated against the reference value for the production date of the certified test sample. Full confidentiality is guaranteed with respect to the link between measurement results and the participants' identity.

Due to the nature of this comparison only a limited number of samples are available. Samples will be allocated to participants in order of registration until the stock of REIMEP-22 samples is used up.

…/…

Retieseweg 111, 2440 Geel, Belgium
Tel.: +32-(0)14-571 681 • Fax: +32-(0)14-571 863
jrc-irmm-reimep@ec.europa.eu • http://www.irmm.jrc.be

The participation fee for REIMEP-22 is **€ 1000 per sample** (including sample dispatch), which has to be paid upon reception of the sample.

The REIMEP-22 samples will be shipped from the EC-JRC-IRMM, Geel to the participants. Each participant has to request the Import Licence in time to enable shipment. The handling or operation licence will be organized by the IRMM Transport officers.

We ask each participant to provide the following information:

1) Contact person (full name, e-mail address and telephone number)
2) Contact person for nuclear transport licensing
3) Contact person for dangerous goods
4) Nuclear accountancy area
5) Delivery address (not a PO box, but a real address)

By registering in REIMEP-22, the participants agree to the following *Transfer of Title and Risks*: "Title and risks associated with the samples provided by IRMM shall pass to the participants upon delivery of the samples to their premises. Participants will be responsible as well for "the sample disposal and costs involved".

Please register electronically for this interlaboratory comparison using the following links:

- To register for the comparison on a 20 mg uranium sample (spectrometric techniques):

https://web.jrc.ec.europa.eu/ilcRegistrationWeb/registration/registration.do?selComparison=1121

- To register for the comparison on a 50 mg uranium sample (radiometric techniques):

https://web.jrc.ec.europa.eu/ilcRegistrationWeb/registration/registration.do?selComparison=1122

Participating laboratories willing to use the two kinds of techniques can ask for a 20 mg and a 50 mg U sample and will have then to report results for two distinct samples. Note that in this case, the participants will have to register for two samples (using the two links above) and consequently will have to pay for two distinct participations in REIMEP-22.

Once you will have submitted your registration electronically, please follow the procedure indicated: a) print your registration form; b) sign it; and c) fax or email it to us. Your fax/email will be then the confirmation of your participation.

The deadline for registration is 1st October 2013. After this deadline, the participants in REIMEP-22 will be contacted by the IRMM transport officer regarding shipment and transport. The samples will be sent to participants between October and December 2013.

The deadline for submission of results is 31st March 2014.

Please do not hesitate to contact us in case you need more information.

Yours sincerely,

Célia Venchiarutti
REIMEP-22 Co-ordinator

Stephan Richter
REIMEP-22 Co-ordinator

Retieseweg 111, 2440 Geel, Belgium
Tel.: +32-(0)14-571 681 • Fax: +32-(0)14-571 863
jrc-irmm-reimep@ec.europa.eu • http://www.irmm.jrc.be

Annex B Email to confirm participation in REIMEP-22

VENCHIARUTTI Celia (JRC-GEEL)

From:	VENCHIARUTTI Celia (JRC-GEEL) on behalf of JRC IRMM REIMEP
Sent:	04 October 2013 17:42
To:	JRC IRMM REIMEP
Cc:	AREGBE Yetunde (JRC-GEEL); RICHTER Stephan (JRC-GEEL); MAYER Klaus (JRC-KARLSRUHE); VARGA Zsolt (JRC-KARLSRUHE)
Subject:	Confirmation of participation to REIMEP-22

Dear colleagues,

Thank you very much for your interest in participating in REIMEP-22. We received your registration forms that you sent by fax or email, and you are therefore now considered as a REIMEP-22 participant. You should soon receive further information regarding the shipment of the sample(s), your personal participation key (for the reporting of the results) and detailed guidelines on the reporting of the results for REIMEP-22 and the following steps.

However, before creating you as participant in our system, we would like to check that your registration has been saved for the correct comparison(s), that is either comparison 1121 (20 mg U sample, spectrometric techniques) or comparison 1122 (50mg U sample, radiometric techniques) or for both comparisons.

Could you please send an email to this address (jrc-irmm-reimep@ec.europa.eu) in order to confirm the comparison(s) in which you wished to participate?

Thank you very much in advance for your reply.

Do not hesitate to contact us if you have any questions.

Kind regards,

Dr. Célia Venchiarutti
REIMEP-22 co-ordinator

****
REIMEP
European Commission - Joint Research Centre
Institute for Reference Materials and Measurements (IRMM)
Retieseweg 111, B-2440 GEEL, BELGIUM
Tel: +32 14 571 681
Fax: +32 14 571 863
Email: jrc-irmm-reimep@ec.europa.eu

Web: http://irmm.jrc.ec.europa.eu; http://irmm.jrc.ec.europa.eu/catalogue

Help save paper - do you need to print this email?

Annex C Accompanying letter

EUROPEAN COMMISSION
DIRECTORATE-GENERAL
JOINT RESEARCH CENTRE
Directorate D - Institute for Reference Materials and Measurements
Standards for Nuclear Safety, Security and Safeguards

REIMEP

Geel, 21 November 2013
JRC.D.4/SR/CV/ccp/D-ARES(2013) 3532033

«TITLE» «FIRSTNAME» «SURNAME»
«ORGANISATION»
«DEPARTMENT»
«ADDRESS»
«ADDRESS2»
«ADDRESS3»
«Address4»
«ZIP» «TOWN»
«COUNTRY»

REIMEP-22: Interlaboratory Comparison on U Age Dating/ The determination of the production date of a uranium certified test sample

Dear «TITLE» «SURNAME»,

Thank you for your participation in REIMEP-22.

Together with this letter, we are sending you a 20 mg or 50 mg uranium certified test sample depending on the type of technique that your laboratory is planning to use for the determination of the production date of this sample. The sample is slightly-enriched uranium, in solid uranyl-nitrate form.

Please check whether the sample (Savillex® beaker and packaging) remained intact during the transport and then sign the "Confirmation of receipt" form and return it to us by email or fax (Fax: +32 14 571 863).

As described in the REIMEP-22 announcement letter, the determination of the production date of the material, has to be based on the compulsory measurements of either isotope amount ratio $n(^{230}Th)/n(^{234}U)$ for the 20 mg uranium samples or the activity ratio $A(^{230}Th)/A(^{234}U)$ for the 50 mg uranium samples.

We remind you that in addition to the $^{230}Th/^{234}U$ measurements, participants have the possibility to measure as well the $n(^{231}Pa)/n(^{235}U)$ amount ratio for the 20 mg uranium sample or the $A(^{231}Pa)/A(^{235}U)$ activity ratio for the 50 mg uranium sample and to report the production date based on these measurements.

Note that participants analysing a 50 mg uranium sample and using both techniques (spectrometry and radiometry) should report only activity ratios in the result reporting system.

http://irmm.jrc.ec.europa.eu/interlaboratory_comparisons/reimep/reimep-22/Pages/index.aspx

The whole analysis and measurement procedures should be carried out using the existing routine analytical procedures in your laboratory for this kind of sample.

Participants are asked to measure 3 **replicates** per sample for each isotope amount ratio/activity ratio.

In the final reporting of results, the measurement values of these 3 replicates and their average value and its associated uncertainty will have to be reported.

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jrc-irmm-reimep@ec.europa.eu • http://www.irmm.jrc.be

Please be aware that the reference date for your measurements results is **6 March 2013!**

The production date of the sample should be reported as follows: dd/mm/yyyy and the associated expanded uncertainty should be reported in ± days.

You can report your results via the following website:

https://web.jrc.ec.europa.eu/ilcReportingWeb

To access this website you will need your personal password key, which is:

«Part_key»

The system will guide you through the reporting procedure. The result-reporting page will be active from **1 January 2014**.

More information about the result reporting and the questionnaire can be found in the participant's guidelines enclosed with this letter.

Together with the reporting of your results, you will have to fill out the questionnaire. Do not forget to submit and always confirm when required. Please do check your results carefully for any errors before submission, since this is your definitive confirmation.

Please keep in mind that collusion is contrary to professional scientific conduct and serves only to nullify the benefits of proficiency tests to customers, accreditation bodies and analysts alike.

Directly after submitting your results and the questionnaire online, you will be prompted to print the completed report form (pdf file). Please do so, sign the paper version and return it to us at IRMM by fax (+32 14 571 863) or by e-mail jrc-irmm-reimep@ec.europa.eu.

The deadline for the submission of results is 31 March 2014

Please do not hesitate to contact us in case you need more information.

Yours sincerely,

Célia Venchiarutti
REIMEP-22 Co-ordinator

Stephan Richter
REIMEP-22 Co-ordinator

Retieseweg 111, 2440 Geel, Belgium
Tel.: +32-(0)14-571 681 • Fax: +32-(0)14-571 863
jrc-irmm-reimep@ec.europa.eu • http://www.irmm.jrc.be

Annex D Confirmation of receipt of the sample

EUROPEAN COMMISSION
DIRECTORATE-GENERAL
JOINT RESEARCH CENTRE
Directorate D - Institute for Reference Materials and Measurements
Standards for Nuclear Safety, Security and Safeguards

Geel, 21 November 2013
JRC.D.4/SR/CV/ccp/D-ARES(2013) 3532033

«TITLE» «FIRSTNAME» «SURNAME»
«ORGANISATION»
«DEPARTMENT»
«ADDRESS»
«ADDRESS2»
«ADDRESS3»
«Address4»
«ZIP» «TOWN»
«COUNTRY»

REIMEP-22

Interlaboratory Comparison on U Age Dating/ The determination of the production date of a uranium certified test sample

Confirmation of receipt of the REIMEP-22 sample

Please return this form at your earliest convenience.
This confirms that the sample package arrived.
In case the package is damaged,
please state this on the form and contact us immediately.

SAMPLE CODE	................-................
ANY REMARKS	....
	....
Date of package arrival	....
Signature	....

Please return this form to:

Dr. Celia Venchiarutti
REIMEP-22 Co-ordinator
EC-JRC-IRMM
Retieseweg 111
B-2440 GEEL
BELGIUM
Fax : +32 14 571 548
e-mail : jrc-irmm-reimep@ec.europa.eu

Retieseweg 111, 2440 Geel, Belgium
Tel.: +32-(0)14-571 681 • Fax: +32-(0)14-571 863
jrc-irmm-reimep@ec.europa.eu • http://www.irmm.jrc.be

Annex E Guidelines for the result reporting

EUROPEAN COMMISSION
DIRECTORATE-GENERAL
JOINT RESEARCH CENTRE
Directorate D - Institute for Reference Materials and Measurements
Standards for Nuclear Safety, Security and Safeguards

Geel, 21 November 2013
JRC.D.4/SR/CV/ccp/D-ARES(2013) 3532033

REIMEP-22: U Age Dating

The determination of the production date of a uranium certified test sample

Participant's guidelines for the reporting of the results via the IRMM's online registration and reporting tool

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1 Sample mailing to participants

Together with the sample you should find the following:

- An accompanying letter, which includes your **participation key** with the following format: your initials (e.g. VC), followed by a 9 characters with letters and numbers. **This key is the password** you need to access our online reporting tool to enter your results. Each key is a unique code generated per participant.

- A confirmation of receipt, which needs **to be returned** to us upon reception of the sample.

2 Reporting of results (the ILC Reporting page)

2.1 Reporting results:
The link for reporting results is https://web.jrc.ec.europa.eu/ilcReportingWeb
As soon as this link has been opened, the login screen below is displayed.

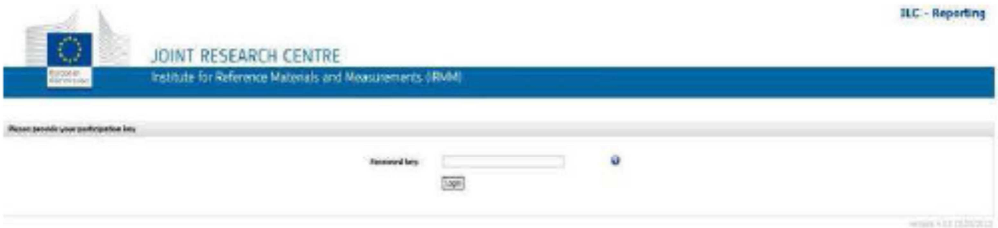

Figure 1 Interface to enter your participation key

2.2 Enter your participation key:
Now, you should enter your unique password/participation key in order to get access to the online reporting of results.

2.3 ILC reporting page:

Once done, you are automatically redirected to the ILC reporting screen of REIMEP-22:

- If for instance, you have asked for "a 20 mg U sample" (same interface for a 50 mg uranium sample), then the following screen will be displayed:

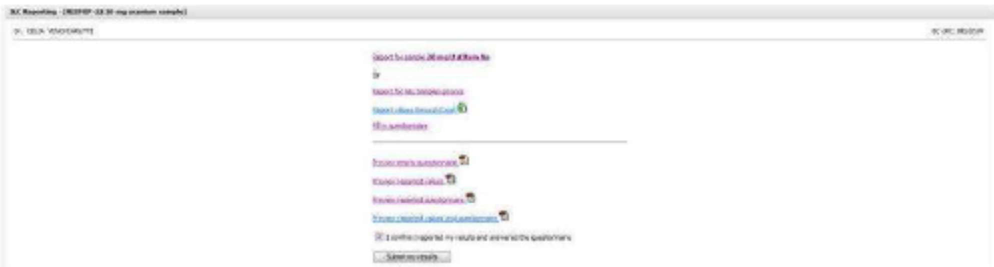

Figure 2 ILC reporting page

- The first part is to report the results or fill the questionnaire and the second part corresponds to the different previews of the reported results or questionnaire.

2.4 Result input page:

In order to report your results, please select "Report for sample 20 mg U" or "Report for sample 50 mg U". Once selected, the following result input screen will be displayed:

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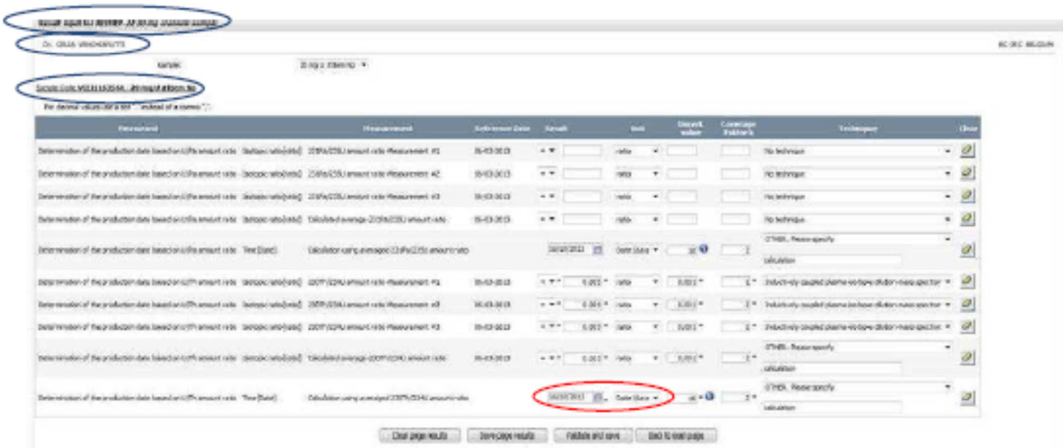

Figure 3 Result input interface/screen

On top of the result input screen you can see:

- ➢ The name of the comparison for which you are about to enter results (1)
- ➢ The name of the participant (2)
- ➢ The code of the sample for which results are reported (3)

Then, the Table displayed in *Figure 3* consists of 8 columns:

- ➢ **Measurand** (i.e. the aim of the ILC; here, the determination of the production date).
- ➢ **Measurement** (i.e. the parameter for which you have to report a value).
- ➢ **Reference date** (i.e. the date for which you should report/calculate your measurement). This will be explained in more details.
- ➢ Results that consists of two parts in the data grid. This will be explained in more details.
- ➢ **Unit**
- ➢ **Uncert. Value** (the uncertainty of the reported result/measurement).
- ➢ **Coverage factor k** (value of the coverage factor).
- ➢ **Technique** (the technique, used for the measurement or to obtain the reported value, has to be selected from the dropdown menu).

NOTE: Measurands for which it is mandatory to enter/report a value, are marked with a red asterix *.

2.5 The Reference date:

For REIMEP-22, the **reference date** was fixed at: **6 March 2013**. The reported ratios should then be **reported** as if **measured at this date**, whenever you performed the chemical separation on your uranium certified test sample or whenever you measured the ratios in the sample. That means that you will have to calculate from your measured ratios what they would be if you had measured them on the **6 March 2013**.

2.6 Results:

They can be entered in 3 possible ways:

- ➢ Either = a certain value, in this case, the uncertainty value and coverage factor must be entered.
- ➢ Or > a certain value, no uncertainty value nor coverage factor can be entered.
- ➢ Or < a certain value, no uncertainty value nor coverage factor can be entered.

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Except for the production date, which you have to enter via the calendar and that will have the default format dd/mm/yyyy, all the other reported values have to be **integers**!

Decimal values can only be entered **using a dot** (.) instead of a comma (,).

Important!! Note that in order to "**save and validate**" your results, you have to fill the fields for the production date based on the $^{231}Pa/^{235}U$ ratio, even if you haven't measured it (since not mandatory). In this case (if not measured), fill the fields with the values (date, uncertainty and coverage factor) found for the production date based on the compulsory measurement of the $^{230}Th/^{234}U$ ratio.

2.7 Unit:

For each measurement, you will have to enter a result (and can select </=/>) and to choose a unit from the dropdown menu. Note that for amount or activity ratios, the unit should be **ratio**.

NOTE: That when reporting the production date via the calendar, the default unit is dd/mm/yyyy, therefore the dropdown menu for the production date **unit** corresponds to the unit chosen **to report the uncertainty associated with the production date**, that is here in **Date (days)** (*Figure 3*).

2.8 Uncertainty value:

To enter the uncertainty value and the coverage factor (k =) associated with each measurement. Note that for amount or activity ratios, the uncertainty value unit should also be a **ratio**.

However, as mentioned above in 2.6, for **the production date**, the reported uncertainty unit has to be selected using the dropdown menu in the column "Unit": **Date (days)**. Remember that reported values have to be integers!
If you report a decimal value for this uncertainty (remember to use "dot", not "coma").

Example: if the reported uncertainty for the production date is *2 days and 12 hours*, then please enter *2.5* for the uncertainty Date (days).

2.9 Managing your results in the result input grid:

You can delete the reported **Uncert. Value** and the selected **Technique** using the *eraser* at the end of each row (next to "Technique").

At the bottom of the screen, you can either:

1) *Clear* the entire reported results.

2) *Save* your results (this will temporarily keep them available).

3) *Validate and save* your results, this will create a draft pdf document, which you can then see in the ILC Reporting page by clicking on "Preview reported values".

4) Once you saved your results by either option 2) or 3), use the *Back to main page* button to go back to the ILC reporting page.

3 Filling the questionnaire

Now that you entered your results, you can start to fill the questionnaire. For this, in the ILC reporting page/screen, click on "**Fill in questionnaire**". The following screen with the questionnaire will be displayed:

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Figure 4 Questionnaire page

This screen shows the name of the comparison (1) as well as the participant's name (2)

- Questions that are mandatory are marked with a red asterix.
- A question mark at the end of the line of a question indicates additional information/help, which is displayed when moving the mouse on .

Please do answer to the given questions as thoroughly as possible.

At the bottom of the screen, you can either:

1) *Clear* the entire answers to the questionnaire.

2) *Save* your answers (this will temporarily keep them available).

3) *Validate* all your answers in the questionnaire and create a draft pdf document, which you can then see in the ILC Reporting page by clicking on "Preview reported questionnaire".

Once you saved your results by either option 2) or 3), use the *Cancel* button to go back to the ILC reporting page.

Attention! Unlike for the *result reporting interface*, there is no reminder message set for the questionnaire when you click on the *Cancel* button! So do save your data before clicking!

4 Preview of your reported results and completed questionnaire

Once you reported and validated both results/values and filled the questionnaire, you can see them as a pdf file by clicking on "Preview reported values and questionnaire" in the ILC Reporting page.

Attention! After opening the pdf, use the back button ("back arrow" in the menu bar) of your web browser (Internet Explorer or Mozilla Firefox) to go back to the ILC result reporting interface. If you close the tab, you will logout automatically from the result reporting interface.

5 Submission of the results and questionnaire

Note that you cannot submit your results alone, but must fill and submit the questionnaire as well.

As long as you **do not click** on "Submit my results" in the ILC reporting page, you can:

1) Change the reported values and uncertainties = go back to the *Results Input page*

2) Change your answers to the questionnaire = go back the *Questionnaire page*

Retieseweg 111, 2440 Geel, Belgium
Tel.: +32-(0)14-571 681 • Fax: +32-(0)14-571 863
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To submit your results and questionnaire, first you have to tick "I confirm I reported my results and answered the questionnaire" (*but will be usually ticked by default*) to acknowledge that you reported your data (and all of them) and your personal answers to the questionnaire.

After submitting your results and questionnaire, you should see the following screen:

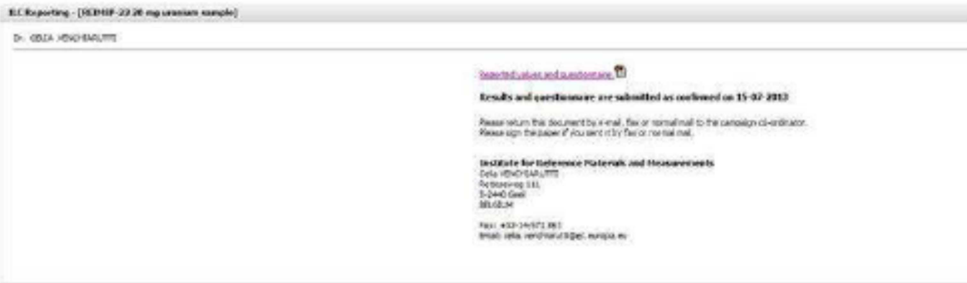

Figure 5 Message displayed once results and questionnaire have been submitted

The final pdf has to be sent (via email or fax) to the ILC coordinator as mentioned on the screen, duly signed in the signature box at the bottom the document!!

6 Assistance required

We hope that these guidelines will be of help. However, if you experience any problems during the reporting of your results, filling the questionnaire or any malfunction of the system, do not hesitate to contact us via our functional mailbox:

jrc-irmm-reimep@ec.europa.eu

Yours sincerely,

Célia Venchiarutti
REIMEP-22 Co-ordinator

Stephan Richter
REIMEP-22 Co-ordinator

Retieseweg 111, 2440 Geel, Belgium
Tel.: +32-(0)14-571 681 • Fax: +32-(0)14-571 863
jrc-irmm-reimep@ec.europa.eu • http://www.irmm.jrc.be

Annex F Letter for extension of result reporting deadline

EUROPEAN COMMISSION
JOINT RESEARCH CENTRE

Institute for Reference Materials and Measurements

Ref. Ares(2014)817663 - 20/03/2014

REIMEP

Geel, 20 March 2014

The IRMM Regular European Interlaboratory Measurement Evaluation Programme

REIMEP-22: Interlaboratory Comparison on U Age Dating

The determination of the production date of a uranium certified test sample

Subject: Deadline extension for submission of results

Dear REIMEP-22 participants,

Due to some delays in the sample shipments, we would like to announce that the deadline for the reporting of REIMEP-22 results is extended.

The extended deadline for online submission of your results is now: **30th April 2014**.

This is the definite deadline for submission of results and there will be no further extension.

To report your results online please use the following link:

https://web.jrc.ec.europa.eu/ilcReportingWeb

Please follow carefully the instructions about the analysis of the sample(s) as indicated in the accompanying letter with your REIMEP-22 sample: about the number of replicates (3 replicates per sample), the reference date (6th March 2013) to report the ratios $^{230}Th/^{234}U$ and optionally $^{231}Pa/^{235}U$, etc.

To guide you through the reporting of your results, please follow the guidelines that were enclosed with your sample(s).

Do not forget to fill in the associated questionnaire. Your results cannot be submitted without the completed questionnaire.

After online submission of your results, we remind you that the final pdf has to be sent (via email or fax) to the ILC coordinator as mentioned on the screen, duly signed in the signature box at the bottom the document.

Please do not hesitate to contact us in case you need more information.

Yours sincerely,

Célia Venchiarutti
REIMEP-22 Co-ordinator

Stephan Richter
REIMEP-22 Co-ordinator

Retieseweg 111, 2440 Geel, Belgium
Tel.: +32-(0)14-571 681 • Fax: +32-(0)14-571 863
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Annex G Questionnaire on 20 mg uranium certified test sample

Milc questionnaire

Comparison for REIMEP-22 20 mg uranium sample

Please fill in your results and answer the corresponding questionnaire.

Submission Form

1. What is the mission of your laboratory (more than one choice possible)? *

- [ ] a) Environmental sciences
- [ ] b) Measurement of radioactivity in the environment
- [ ] c) Monitoring of nuclear facilities
- [ ] d) Measurements for fissile material control or safeguards
- [ ] e) Other

1.1. If you have selected 'Other', please specify:

1.2. Are you taking part in the ITWG Network? *

- ○ a) Yes
- ○ b) No

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

- [ ] a) Certified
- [ ] b) Accredited
- [ ] c) Authorised
- [ ] d) Not applicable

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

- ○ a) Yes
- ○ b) No

3.1. If 'Yes', please specify (more than one choice possible): *

- [ ] a) ISO 17025
- [ ] b) ISO 9000 series
- [ ] c) Other

- Page 1 of 9 -

3.1.1. If you have selected 'Other', please specify:

4. **Has your laboratory already participated in inter-laboratory comparisons?** *

- a) Yes
- b) No

4.1. If 'Yes', please list the name(s) of the comparison(s) and the organizer(s): *

5. **Measurements of Th and U:**

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

- a) 0-10
- b) 11-50
- c) 51-100
- d) > 100

5.2. How does your laboratory rate itself for this kind of measurement? *

- a) Experienced
- b) Less experienced
- c) Not experienced

5.3. On what type of samples do you routinely perform uranium and thorium assay analysis (more than one choice possible)? *

- a) Safeguards samples
- b) Forensic samples
- c) Samples from reprocessing facilities
- d) Reference materials
- e) Environmental samples
- f) Other

5.3.1. If 'Other', please specify:

5.3.2. If 'Environmental samples', please specify the matrices (soil, sediments, water, etc):

6. **Measurements of Pa and U:**

6.1. How many measurements of Pa/U does your laboratory routinely perform per year?

- a) 0-10
- b) 11-50
- c) 51-100
- d) > 100

6.2. How does your laboratory rate itself for this kind of measurement?

- a) Experienced
- b) Less experienced
- c) Not experienced

6.3. On what type of samples do you routinely perform uranium and protactinium assay analysis (more than one choice possible)?

- a) Safeguards samples
- b) Forensic samples
- c) Samples from reprocessing facilities
- d) Reference materials
- e) Environmental samples
- f) Other

6.3.1. If 'Other', please specify:

6.3.2. If 'Environmental samples', please specify the matrices (soil, sediments, water, etc):

7. Was the REIMEP-22 sample treated according to the same analytical procedure as routinely used in your laboratory for this type of sample? *

- a) Yes
- b) No

7.1. If 'No', please describe why it was not performed according to your routine analytical procedure *

8. Did you report on the Pa/U amount isotopic ratios and the associated production date? *

- a) Yes
- b) No

8.1. If 'No', please specify the reason(s) why you did not carry out this type of measurement *

9. **Did you apply any kind of chemical treatment to the REIMEP-22 sample prior to the measurement?** *

☐ a) Yes

☐ b) No

9.1. If 'Yes', please describe in details the analytical procedure for the fraction Th/U:

9.2. If you measured the Pa/U fraction and used a different procedure from that of the Th/U, please describe in details the analytical procedure for the fraction Pa/U:

9.3. What amount of material in mg did you use for a single analysis (one replicate)? *

10. **Does your laboratory routinely use Certified Reference Materials (CRMs)?** *

○ a) Yes

○ b) No

10.1. If 'Yes', please specify which CRM(s) and supplier(s) *

10.2. What are the CRM(s) usually applied for (more than one choice possible)? *

☐ a) Validation of procedure

☐ b) Calibration of instrument

☐ c) Other

10.2.1. If 'Other', please specify:

11. **More specifically for REIMEP-22, what are the CRMs or reference materials that you used?** *

See table **CRMs used for REIMEP-22 sample** at bottom

12. Are your reported uncertainties for REIMEP-22 calculated according to the Guides for Quantifying Measurement Uncertainty issued by the International Organisation for Standardisation (ISO, 1995) and/or EURACHEM/CITAC (2000)? *

- a) Yes
- b) No

12.1. If 'No', how was the measurement uncertainties estimated? *

12.2. If 'Yes', what did you report as an uncertainty? *

- a) Standard uncertainty
- b) Expanded uncertainty with a coverage factor, k:_____

12.3. Please list here the major uncertainty contributions for the reported ratios and production date: *

13. Do you routinely report uncertainties on measurements to your customers? *

- a) Yes
- b) No

14. Did you report the values for the isotope ratios relative to 6 March 2013 when submitting the results? *

- a) Yes
- b) No

15. Please give the half-lives used for the calculation of the production date *

See table **Half-lives** at bottom

15.1. Please quote here the references from the literature (Authors, Journal, Year) that you used for the half-lives: *

16. If applicable, please give the molar masses used for the calculation of the production date:

See table **Molar masses** at bottom

16.1. Please quote here the references from the literature (Authors, Journal, Year) that you used for the molar masses: *

17. How did you learn about REIMEP-22 (more than one choice possible)?

- ☐ a) IRMM website
- ☐ b) e-mail
- ☐ c) From other participants
- ☐ d) Other

17.1. If 'Other', please specify:

18. Would you be interested in participating in future REIMEP inter-laboratory comparisons on age dating?

- ○ a) Yes
- ○ b) No

18.1. If 'Yes', in what type of samples would you be interested (U,Th,Pu)? * *

18.2. If 'Yes', which radionuclide disequilibrium/clock would you like to use (Th/U, Pa/U, Pu/Am, other)? *

18.3. What type of matrices? *

19. Do you have any feedback/comments on REIMEP-22?

19.1. Please rate this questionnaire *

See table **Evaluation of questionnaire** at bottom

CRMs used for REIMEP-22 sample

Please give the name or reference of the CRM used when applicable, or describe the isotopes used in the case of an "in-house" standard.

Questions/Response table	*CRMs*	*In-house standards*
For calibration		
For quantification of the analyte (spikes)		
For isotope dilution		
For mass bias/ mass fractionation		
For abundance sensitivity		

Evaluation of questionnaire

Rate 1= not satisfying/difficult Rate 5= satisfying/easy

Questions/Response table	*Rate 1 to 5*
Difficulty to fill in the questionnaire	

Half-lives

Please fill this Table with the half-lives that were used for the determination of the production date.

Questions/Response table	*Half-life (years)*	*Expanded uncertainty (k=2)*
230Th		
234U		
231Pa		
235U		

Molar masses

Please fill this Table with themolar masses that were used for the determination of the production date.

Questions/Response table	*Molar masses (g/mol)*	*Expanded uncertainty (k=2)*
230Th		
234U		
231Pa		
235U		

Annex H Questionnaire on 50 mg uranium certified test sample

Milc questionnaire

Comparison for REIMEP-22 50 mg uranium sample

Please fill in your results and answer the corresponding questionnaire.

Submission Form

1. What is the mission of your laboratory (more than one choice possible)? *

- [ ] a) Environmental sciences
- [ ] b) Measurement of radioactivity in the environment
- [ ] c) Monitoring of nuclear facilities
- [ ] d) Measurements for fissile material control or safeguards
- [ ] e) Other

1.1. If you have selected 'Other', please specify:

1.2. Are you taking part in the ITWG network?

- ( ) a) Yes
- ( ) b) No

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

- [ ] a) Certified
- [ ] b) Accredited
- [ ] c) Authorised
- [ ] d) Not applicable

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

- ( ) a) Yes
- ( ) b) No

3.1. If 'Yes', please specify (more than one choice possible): *

- [ ] a) ISO 17025
- [ ] b) ISO 9000 series
- [ ] c) Other

- Page 1 of 10 -

3.1.1. If you have selected 'Other', please specify:

4. **Has your laboratory already participated in inter-laboratory comparisons?** *

- a) Yes
- b) No

4.1. If 'Yes', please list the name(s) of the comparison(s) and the organizer(s): *

5. **Measurements of Th and U:**

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

- a) 0-10
- b) 11-50
- c) 51-100
- d) > 100

5.2. How does your laboratory rate itself for this kind of measurement? *

- a) Experienced
- b) Less experienced
- c) Not experienced

5.3. On what type of samples do you routinely perform uranium and thorium assay analysis (more than one choice possible)? *

- a) Safeguards samples
- b) Forensic samples
- c) Samples from reprocessing facilities
- d) Reference materials
- e) Environmental samples
- f) Other

5.3.1. If 'Environmental samples', please specify the matrices (soil, sediments, water, etc):

5.3.2. If 'Other', please specify:

6. **Measurements of Pa and U:**

- Page 2 of 10 -

6.1. How many measurements of Pa/U does your laboratory routinely perform per year?

- a) 0-10
- b) 11-50
- c) 51-100
- d) > 100

6.2. How does your laboratory rate itself for this kind of measurement?

- a) Experienced
- b) Less experienced
- c) Not experienced

6.3. On what type of samples do you routinely perform uranium and protactinium assay analysis (more than one choice possible)?

- [ ] a) Safeguards samples
- [ ] b) Forensic samples
- [ ] c) Samples from reprocessing facilities
- [ ] d) Reference materials
- [ ] e) Environmental samples
- [ ] f) Other

6.3.1. If 'Environmental samples', please specify the matrices (soil, sediments, water, etc):

6.3.2. If 'Other', please specify:

7. Was the REIMEP-22 sample treated according to the same analytical procedure as routinely used in your laboratory for this type of sample? *

- a) Yes
- b) No

7.1. If 'No', please describe why it was not performed according to your routine analytical procedure: *

8. Did you report on the Pa/U amount isotopic ratios and the associated production date? *

- a) Yes
- b) No

8.1. If 'No', please specify the reason(s) why you did not carry out this type of measurement *

9. Did you apply any kind of chemical treatment to the REIMEP-22 sample prior to the measurement? *

- [ ] a) Yes
- [ ] b) No

9.1. If 'Yes', please describe in details the analytical procedure for the fraction Th/U:

9.2. If you measured the Pa/U fraction and used a different procedure from that of Th/U, please describe in details the analytical procedure for the fraction Pa/U:

9.3. What amount of material in mg did you use for a single analysis (one replicate)? *

10. If you measured REIMEP-22 sample using both spectrometric and radiometric techniques, did your results agree within the uncertainties? *

- ○ a) Yes
- ○ b) No
- ○ c) Not applicable

10.1. If 'No', could you please explain briefly what could be the reasons for the discrepancies in your results: *

10.2. If 'Yes', please give the production date dd/mm/yyyy and its uncertainty: *

See table **Production date and uncertainty based on spectrometric measurement results** at bottom

11. Does your laboratory routinely use Certified Reference Materials (CRMs)? *

- ○ a) Yes
- ○ b) No

11.1. If 'Yes', please specify which CRM(s) and supplier(s): *

11.2. What are the CRM(s) usually applied for (more than one choice possible)?

- [ ] a) Validation of procedure
- [ ] b) Calibration of instrument
- [ ] c) Other

11.2.1. If 'Other', please specify:

12. More specifically for REIMEP-22, what are the CRMs or reference materials that you used? *

See table **CRMs used for REIMEP-22 sample** at bottom

13. Are your reported uncertainties for REIMEP-22 calculated according to the Guides for Quantifying Measurement Uncertainty issued by the International Organisation for Standardisation (ISO, 1995) and/or EURACHEM/CITAC (2000)? *

- ○ a) Yes
- ○ b) No

13.1. If 'No', how was the measurement uncertainties estimated? *

13.2. If 'Yes', what did you report as an uncertainty? *

- ○ a) Standard uncertainty
- ○ b) Expanded uncertainty with a coverage factor, k:_____

13.3. Please list here the major uncertainty contributions to the reported ratios and production date: *

14. Do you routinely report uncertainties on measurements to your customers? *

- ○ a) Yes
- ○ b) No

15. Did you report the values for the isotope ratios relative to 6 March 2013 when submitting the results? *

- ○ a) Yes
- ○ b) No

16. Please give the half-lives and references (literature) used for the calculation of the production date *

See table **Half-lives** at bottom

16.1. Please quote here the references from the literature (Authors, Journal, Year) that you used for the half-lives: *

17. If applicable, please give the molar masses and references (literature) used for the calculation of the production date:

See table **Molar masses** at bottom

17.1. Please quote here the references from the literature (Authors, Journal, Year) that you used for the molar masses: *

18. How did you learn about REIMEP-22 (more than one choice possible)?

- ☐ a) IRMM website
- ☐ b) e-mail
- ☐ c) From other participants
- ☐ d) Other

18.1. If 'Other', please specify:

19. Would you be interested in participating in future REIMEP inter-laboratory comparisons on age dating?

- ○ a) Yes
- ○ b) No

19.1. If 'Yes', in what type of samples would you be interested (U,Th,Pu)? *

19.2. If 'Yes', which radionuclide disequilibrium/clock would you like to use (Th/U, Pa/U, Pu/Am, other)? *

19.3. What type of matrices? *

20. Do you have any feedback/comments on REIMEP-22?

20.1. Please rate this questionnaire *

Evaluation of questionnaire

Rate 1= not satisfying/difficult Rate 5= satisfying/easy

Questions/Response table	*Rate 1 to 5*
Difficulty to fill in the questionnaire	

CRMs used for REIMEP-22 sample

Questions/Response table	*CRMs*	*In-house standards*
For calibration		
For quantification of analyte (spikes)		
For isotope dilution		
For mass bias/ mass fractionation		
For abundance sensitivity		

Half-lives

Please fill this Table with the half-lives that were used for the determination of the production date.

Questions/Response table	*Half-life (years)*	*Expanded uncertainty (k=2)*
230Th		
234U		
231Pa		
235U		

Molar masses

Please fill this Table with the molar masses that were used for the determination of the production date.

Questions/Response table	*Molar masses (g/mol)*	*Expanded uncertainty (k=2)*
230Th		
234U		
231Pa		
235U		

Production date and uncertainty based on spectrometric measurement results

Please give the production date in dd/mm/yyyy and its associated uncertainty as deduced from the spectrometric measurement results (atom ratio). Please mention as well if the uncertainty corresponds to standard or expanded uncertainty.

Questions/Response table	*Production date (dd/mm/yyyy)*	*Uncertainty (standard uncertainty)*	*Uncertainty (expanded unceratinty)*
Values (based on measured atom ratio)			

Annex I Results from questionnaire on 20 mg uranium certified test sample

Annex I 1 Amount of sample used per replicate for a 20 mg sample

Lab codes	Amounts (mg)
10246	2.3
10250	5.7
10245	2.5
10249	2.0
10243	0.1
10242	5
10248	0.7
10252	45(*)
10247	0.2

(*)The participant reported a sample amount of 45 mg used for the 20 mg certified test sample analysis. We suppose that it must be a mistake in the reporting of the amount and that it should be understood as 4.5 mg.

Annex I 2 CRMs used for REIMEP-22 mass spectrometry analysis

	For abundance sensitivity		For calibration		For mass fractionation		For isotope dilution		For quantification of the analyte (spikes)
Lab codes	CRMs	In-house	CRMs	In-house	CRMs	In-house	CRMs	In-house	CRMs
10246	U005A, CRM129A	-	NBL U010	-	U010	-	-	^{233}U, ^{229}Th, ^{233}Pa	NIST 4342, CRM-145, 112A
10250	HPS	-	Merck	-	-	-	-	-	-
10245	-	-	-	-	IRMM standards	-	NBL 111a and NIST Th-229	-	NBL 111a and NIST Th-229
10249	IRMM184	-	-	-	IRMM183	-	IRMM057	^{229}Th	-
10243	U 015	Th 105 (^{232}Th, ^{230}Th)	U 015	Th 105 (^{232}Th, ^{230}Th)	-	T2U5 (^{230}Th, ^{234}U)	-	-	-
10242	-	-	U-010	-	IRMM-184, IRMM-185	-	-	-	-
10248	IRMM0731	-	IRMM0731, IRMM184	-	IRMM0731	-	IRMM040a	Th-229 (from IRMM040a)	-
10252	Alfa Aesar Specpure	-	Alfa Aesar Specpure	-	-	natural U	-	-	Alfa Aesar Specpure
10247	-	-	-	-	NIST SRM U-030	-	IRMM-040a	-	-

Note that an empty field "-" in the Table means that the participants did not report any CRMs or standards for this field. No in-house standards were reported (or used) for the quantification of the analyte (spike calibration).

Annex I 3 Uncertainty budget for mass spectrometry analysis

Lab codes	Major uncertainty contribution to REIMEP-22 results
10246	^{230}Th measurement, ^{229}Th spike calibration, ^{231}Pa measurement, ^{233}Pa spike calibration
10250	^{230}Th measurement
10245	230/229 measured ratio, ^{230}Th half-life, Th229 NIST standard, 234/233 measured ratio
10249	Uncertainty on ^{229}Th concentration in the tracer, counting statistics on ^{230}Th
10243	Primarily abundance sensitivity
10242	Noise on the ^{230}Th signal
10248	Measurement of intensity at m/z230 for determination of ^{230}Th, concentration of ^{233}U in IRMM040a for determination of ^{234}U
10252	Separation yield of Pa, ^{234}U and ^{230}Th measurements
10247	Amounts of n(^{230}Th) and n(^{234}U)

Annex I 4 Half-lives (in years) and uncertainties (with *k=2*) as used by participants

Lab codes	^{234}U	^{230}Th	^{235}U	^{231}Pa
10246	$2.4525 \cdot 10^{5}$ ±490 a	$7.569 \cdot 10^{4}$ ±230 a	$7.0381 \cdot 10^{8}$ ±960000 a	$3.276 \cdot 10^{4}$ ±220 a
10250	$2.4550 \cdot 10^{5}$ ±600 a	$7.538 \cdot 10^{4}$ ±300 a	-	-
10245	$2.4540 \cdot 10^{5}$ ±600 a	$7.540 \cdot 10^{4}$ ±300 a	-	-
10249	$2.4525 \cdot 10^{5}$ ±490 a	$7.569 \cdot 10^{4}$ ±230 a	-	-
10243	*2.83*$\cdot 10^{-7}$(*)	*9.16*$\cdot 10^{-7}$(*)	-	-
10242	$2.4525 \cdot 10^{5}$ ±980 a	$7.569 \cdot 10^{4}$ ±460 a	-	-
10248	$2.4550 \cdot 10^{5}$ ±1200 a	$7.538 \cdot 10^{4}$ ±600 a	-	-
10252	$2.4500 \cdot 10^{5}$	$7.540 \cdot 10^{4}$	$7.0400 \cdot 10^{8}$	$3.276 \cdot 10^{4}$
10247	$2.4525 \cdot 10^{5}$ ±490 a	$7.569 \cdot 10^{4}$ ±230 a	-	-

(*) Note that this participant likely reported decay constants and not the half-lives as described in Section 9.6.

Bibliographic references and sources used by participants for half-lives

Lab codes	References
10246	Cheng et al. (2000) Chemical Geology; Jaffey et al. (1971) Physical Reviews C; Robert et al. (1969) Radiochimica Acta
10250	G. Audi, O. Bersillon, J, Blachot and A.H. Wapstra Nuclear Physics 2003
10245	Brown and Firestone, Table of Radioactive Isotopes, 1986
10249	Cheng H., Edwards R.L., Hoff J., Gallup C.D., Richards D.A. and Asmerom Y., Chemical Geology 169, 17-33, 2000
10243	Bourdon et al., Reviews in Mineralogy and Geochemistry 52, 1-19 and references therein
10242	nucleonica
10248	http://www.nucleide.org/DDEP_WG/DDEPdata.htm, http://www.nndc.bnl.gov/ensdf/
10252	IAEA Safety Standards, Advisory Material for the IAEA Regulations, Safety Guide
10247	Zsolt Varga et.al, Analytica Chimica Acta, 2012

Annex I 5 Molar masses (in g·mol^{-1}) and uncertainties (with $k=2$) as used by participants

Lab codes	^{234}U	^{230}Th	^{235}U	^{231}Pa
10246	234.040945	230.033126	235.043923	231.035878
10250	234.040952 ± $2.0 \cdot 10^{-6}$	230.033134 ± $1.9 \cdot 10^{-6}$	-	-
10245	234.040946	230.033127	-	-
10249	234.041000	230.033127	-	-
10243	234.040946	230.033127	-	-
10242	-	-	-	-
10248	234.040952 ± $4.0 \cdot 10^{-6}$	230.033134 ± $3.8 \cdot 10^{-6}$	-	-
10252	234.040000	230.030000	235.040000	231.040000
10247	234.040945 ± $4.4 \cdot 10^{-6}$	230.033131 ± $1.6 \cdot 10^{-6}$	-	-

Bibliographic references and sources used by participants for molar masses

Lab codes	References
10246	-
10250	G. Audi, A.H. Wapstra and C. Thibault Nuclear Physics 2003
10245	Baum et al, 16th edition Chart of the Nuclides, 2002
10249	Handbook of chemistry and physics, 86th edition 2005-2006, CRC Press
10243	Handbook of Chemistry and Physics, 72nd edition
10242	-
10248	G. Audi, A.H. Wapstra and C. Thibault, Nuclear Physics A 729 (2003) 337-676.
10252	KAERI, Nuclear Data Center, 2000
10247	IRMM certificate

Annex J Results from questionnaire on 50 mg uranium certified test sample

Annex J 1 Amount of sample used per replicate for a 50 mg sample

Lab codes	Amounts (mg)
10257	15
10254	10.8
10258	6
10259	4.5 (Th) 0.000001 (U)

Annex J 2 CRMs used for REIMEP-22 α-spectrometry analysis

	For calibration		For isotope dilution		For quantification of the analyte (spikes)
Lab codes	CRMs	In-house	CRMs	In-house	CRMs
10257	-	-	-	-	-
10254	-	-	-	-	spike passport № 364/1
10258	IRMM184	-	NIST SRM4324A for U232	^{229}Th (from IAEA040a)	-
10259	-	-	-	-	-

Note that an empty field "-" in the Table means that the participants did not report any CRMs or standards for this field. No in-house standards were reported (or used) for the quantification of the analyte (spike calibration).

Annex J 3 Uncertainty budget for α-spectrometry analysis

Lab codes	Major uncertainty contribution to REIMEP-22 results
10257	Th quantification
10254	Uncertainty of measurement of ^{230}Th Activity
10258	Alpha spectrometry measurement, number of counts for ^{230}Th in sample and background at ^{230}Th
10259	Measurement Technique Efficiencies, Measurement Uncertainties, Tracer Uncertainties, Balance Uncertainties, Error Propagation.

Annex J 4 Half-lives (in years) and uncertainties (with *k=2*) as used by participants

Lab codes	^{234}U	^{230}Th
10257	$2.455 \cdot 10^{5}$	$7.538 \cdot 10^{4}$
10254	$2.4550 \cdot 10^{5}$ ±600 a	$7.538 \cdot 10^{4}$ ±300 a
10258	$2.4550 \cdot 10^{5}$ ±1200 a	$7.538 \cdot 10^{4}$ ±600 a
10259	$2.450 \cdot 10^{5}$	$7.540 \cdot 10^{4}$

Bibliographic references and sources used by participants for half-lives

Lab codes	References
10257	LARA
10254	G. Audi, O. Bersillon, J, Blachot and A.H. Wapstra Nuclear Physics 2003
10258	http://www.nucleide.org/DDEP_WG/DDEPdata.htm, http://www.nndc.bnl.gov/ensdf/
10259	-

Annex J 5 Molar masses (in g·mol^{-1}) and uncertainties (with $k=2$) as used by participants

Lab codes	**^{234}U**	**^{230}Th**
10257	-	-
10254	234.040952 ± 2.0·10^{-6}	230.033134 ± 1.9·10^{-6}
10258	234.040952 ± 4.0·10^{-6}	230.033134 ± 3.8·10^{-6}
10259	-	-

Bibliographic references and sources used by participants for molar masses

Lab codes	**References**
10257	nucleids LARA tables
10254	G. Audi, A.H. Wapstra and C. Thibault Nuclear Physics 2003
10258	G. Audi, A.H. Wapstra and C. Thibault, Nuclear Physics A 729 (2003) 337-676.
10259	-

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