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Interlaboratory Comparison Report

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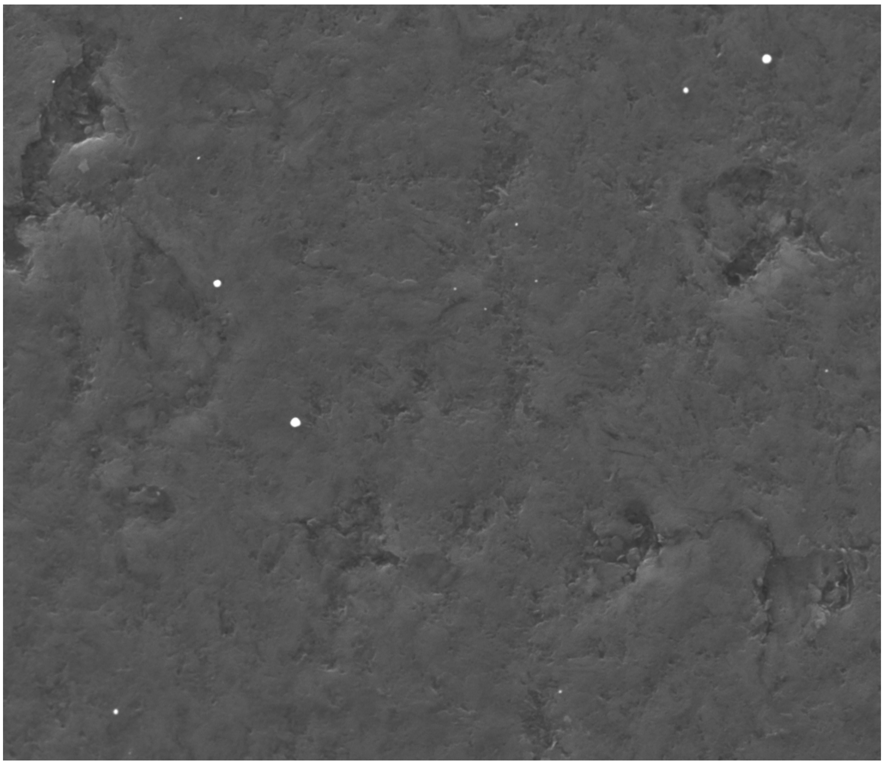

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European Commission
Joint Research Centre
Institute for Reference Materials and Measurements

Contact information
Mr. Jan Truyens
European Commission
Joint Research Centre
Institute for Reference Materials and Measurements
Retieseweg 111
2440 Geel, Belgium

E-mail: Jan.Truyens@ec.europa.eu

Tel.: +32 (0) 14 571 976
Fax: +32 (0) 14 571 548

http://irmm.jrc.ec.europa.eu/
http://www.jrc.ec.europa.eu/

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Elzbieta Stefaniak
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Contents

1 Summary

The Additional Protocol (AP) authorizes safeguards authorities to verify the absence of undeclared nuclear activities in all parts of a state's nuclear fuel cycle as well as any other location where nuclear material is or may be present. As a part of the Additional Protocol, environmental sampling has become an important tool for the detection of non-declared nuclear activities. In environmental sampling micrometer-sized uranium particles with an isotopic composition characteristic for the processes at the inspected facility need to be collected, identified and analysed. Considering the potential consequences of the analyses, these measurements need to be subjected to a rigorous quality management system.

NUSIMEP-7 focused on measurements of *uranium isotope amount ratios in uranium particles* aiming to support EURATOM safeguards (DG ENER), the IAEA network of analytical laboratories for particle analysis of environmental samples for safeguards (NWAL) and laboratories involved in uranium particle analysis. It was organised as a follow-up of NUSIMEP-6, which was the first NUSIMEP in 2008 on particle analysis coordinated by IRMM, intended as a pilot interlaboratory comparison in this field. The feedback from participants in NUSIMEP-6 was taken into account for optimisation and improvements resulting in the second NUSIMEP on particle analysis coordinated by IRMM. NUSIMEP-7 was open for participation to all laboratories in the field of particle analysis, and explicitly recommended by the IAEA to the NWAL for participation.

The NUSIMEP-7 test samples were prepared by controlled hydrolysis of certified uranium hexafluoride. Participating laboratories in NUSIMEP-7 received the test samples of uranium particles on two graphite planchets with undisclosed isotope amount ratio values $n(^{234}\mathrm{U})/n(^{238}\mathrm{U})$, $n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$ and $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$; one planchet with particles of single isotopic deposition and the other with particles of two different isotopic compositions. For both samples, the uranium isotope amount ratios had to be measured using their routine analytical procedures. Measurement of the major ratio $n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$ was obligatory; measurement of the minor ratios $n(^{234}\mathrm{U})/n(^{238}\mathrm{U})$ and $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ was optional. 24 institutes registered for NUSIMEP-7, whereof 17 have reported measurement results using different analytical methods, among those were 7 NWAL laboratories. The participants' measurement results have been evaluated against the certified reference values by means of z-scores and zeta-scores in compliance with ISO 13528:2005. The results of NUSIMEP-7 do not only confirm the capability of laboratories to measure $n(^{234}\mathrm{U})/n(^{238}\mathrm{U})$, $n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$ and $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ in uranium particles of <1 µm, but also to distinguish between groups of particles with different isotopic composition. Furthermore they underpin the recent advances in instrumental techniques in the field of particle analysis. In addition feedback from the measurement communities in nuclear safeguards, nuclear security and earth sciences was collected in view of identifying future needs for NUSIMEP interlaboratory comparisons.

The Director of the Division of Concepts and Planning at the IAEA Department of Safeguards expressed her appreciation and compliments to IRMM for NUSIMEP-7 for '*further improving the detection and analysis capability of the IAEA's NWAL*'

2 NUSIM EP

The IRMM Nuclear Signatures Interlaboratory Measurement Evaluation Programme (NUSIMEP) is an external quality control programme organised by the Joint Research Centre - Institute for Reference Materials and Measurements (IRMM). NUSIMEP was established in 1996 to support the growing need to detect and measure the isotopic abundances of elements characteristic for the nuclear fuel cycle present in trace amounts in the environment. Such measurements are required for safeguards applications as well as for the implementation of the Treaty on the Non-Proliferation of Nuclear Weapons (NPT) [1]. Measurements of the isotopic ratios of the elements uranium and plutonium in small amounts, such as typically found in environmental samples, are required for nuclear safeguards, for the control of environmental contamination and for the detection of nuclear proliferation.

Laboratories participating in NUSIMEP are requested to measure the parameters specified using their standard methods and invited to report measurement results with uncertainties to IRMM. Those reported measurement results are compared with independent external certified reference values with demonstrated traceability and uncertainty, as evaluated according to international guidelines. Laboratory performance evaluation is done according to the respective ISO standard on performance evaluation in proficiency testing by interlaboratory comparisons [2].

Laboratories analysing environmental samples are invited to participate in these external NUSIMEP quality control exercises to demonstrate and assess their ability to carry out accurate measurements in particular on trace amounts of uranium and plutonium. Through this and similar programmes, the degree of equivalence of measurements of individual laboratories can be ascertained.

Several NUSIMEP interlaboratory comparisons of measurements of uranium isotopic ratios were organised previously: for example NUSIMEP 2, uranium isotopic abundances in dry uranium nitrate samples; NUSIMEP 3, uranium isotopic abundances in saline media, NUSIMEP 4, uranium isotopic abundances in a simulated urine and NUSIMEP 5 uranium, plutonium and caesium isotopic ratios in saline medium. Reports on previous NUSIMEP interlaboratory comparisons can be found on the IRMM website [3].

The organisation of the interlaboratory comparison follows the standard procedures of the Interlaboratory Measurement Evaluation Programmes IMEP, REIMEP, NUSIMEP of the Institute for Reference Materials and Measurements (IRMM) of the Joint Research Centre, a Directorate-General of the European Commission. This programme is accredited according to ISO/IEC 17043:2010 [4]. The designation of this interlaboratory comparison is NUSIMEP-7.

3 Introduction

Nuclear safeguards arrangements exist on international level under the protocols of the International Atomic Energy Agency (IAEA) [1] on European Union level under the Euratom Treaty [5] and on regional levels. The INFCIRC/540 [6], also referred to as the Additional Protocol (AP), moved the focus from exclusively accounting for known quantities of fissile material towards a more qualitative system that is able to provide a comprehensive picture of a state’s nuclear activities. Through unannounced inspections and nuclear material balances, safeguards inspectors are able to verify that no nuclear material is diverted from its intended peaceful use. As part of the Additional Protocol, environmental sampling has become an important tool for the detection of non-declared nuclear activities. Analysis of environmental samples is carried out to detect the (unavoidable) traces in the environment originating from technological activities. One extensively developed technique in environmental sampling (ES) makes use of pieces of cotton cloth called swipes to wipe surfaces inside and around a nuclear facility. The dust collected on these swipes typically contains micrometer-sized uranium particles with an isotopic composition characteristic for the processes at the inspected facility. Measurements of minor isotope abundance ratios of uranium in those particles may provide additional information about equipment or plant design and about irradiation history, and may also help to evaluate mixing and decay scenarios. Major and minor uranium isotope ratios in environmental samples collected by inspectors are measured by the IAEA's Safeguards Analytical Laboratories in Seibersdorf, Austria and the IAEA's Network of Analytical Laboratories (NWAL) [7].

The ESARDA Working Group on Standards and Techniques for Destructive Analysis (WG DA) organised two dedicated workshops dealing amongst other topics with advances in instrumental techniques on measurements of major and minor isotopes in particle samples [8, 9]. Participants in these workshops came from the main European and international nuclear safeguards organisations, nuclear measurement laboratories as well as from geochemistry and environmental science institutes. During this workshop, it was stressed that considering the potential consequences of particle analyses in nuclear safeguards, bio- and earth sciences, these measurements need to be subjected to a rigorous quality management system. The reliability and comparability of measurement results of isotope ratios in uranium particles need to be guaranteed and monitored via the correct use of reference materials and quality tools. Currently it is clearly a significant drawback for laboratories involved in particle analysis that such materials are not available. Therefore special attention has been given at IRMM to the development of uranium particle reference materials for the analysis for environmental samples [10, 11, 12, 13]. To address the needs from European and international safeguards authorities and research institutions, IRMM organised the second NUSIMEP interlaboratory comparison on isotope ratio measurements in uranium particles, as a follow-up of NUSIMEP-6, which was the first NUSIMEP on particle analysis coordinated by IRMM in 2008 intended as a pilot interlaboratory comparison in this field [14].

4 Scope and aim

Measurements of the isotopic ratios of the elements uranium and plutonium in small amounts, such as typically found in environmental samples, are required for nuclear safeguards, for the control of environmental contamination and for the detection of nuclear proliferation. NUSIMEP-7 aims at laboratories carrying out particle analysis in these various application fields. Particular emphasis was given to participation of the IAEA network of analytical laboratories for environmental sampling (NWAL) in support to nuclear safeguards arrangements. Participation of the NWAL laboratories in this NUSIMEP interlaboratory comparison was formally recommended by the IAEA at the IAEA Technical Meeting on Particle Analysis of Environmental Samples for Safeguards. NUSIMEP-7 is an interlaboratory comparison that not only should picture the measurement capabilities of the participating laboratories at a certain point in time, but should also investigate advances in instrumental techniques and collect feedback from the participants towards future improvements and needs.

Measurands are the isotope amount ratio values $n(^{234}U)/n(^{238}U)$, $n(^{235}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$. The matrix is uranium particles on two graphite planchets. For both planchets, the uranium isotope amount ratios had to be measured. In addition the participants had to identify which of the two samples contained particles of a single isotopic composition and which contained particles of two different isotopic compositions. The participants received with the NUSIMEP-7 samples guidelines on result reporting. Contrary to NUSIMEP-6 they were asked to report the uranium isotope amount ratios of 10 different particles from the planchet that they identified being loaded with particles of single isotopic composition. On the planchet they identified being loaded with two different isotopic compositions, two times 10 different particles were to be measured (i.e. 10 particles for enrichment 1 and 10 particles for enrichment 2). For both planchets also the averages had to be reported. Measurement of the major ratio $n(^{235}U)/n(^{238}U)$ was obligatory; measurement of the minor ratios $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ were optional, but it was highly recommended to report also the minor ratios.

5 Time frame

NUSIMEP-7 was announced for participation in the beginning of March 2011. Registration was open until the end of March 2011. A confirmation of registration was sent to the participants and subsequently the samples were dispatched end of May 2011. The reporting deadline was 27 June 2011. This deadline was extended by almost 10 weeks for all participants and additionally extended by two extra weeks for participants using Fission Track Thermal Ionisation Mass Spectrometry due to unforeseeable limitations in access to a nuclear reactor and for other participants who faced technical problems in their laboratory. The homogeneity and short term stability studies were carried out between April and October 2011.

End of September 2011 the provisional certified reference values were sent to the participants.

6 Test material

6.1 General remarks

The process applied at IRMM to produce uranium particles from certified uranium hexafluoride (UF_6) is described in detail elsewhere [10]. The aerosol deposition chamber was developed for NUSIMEP-6 at IRMM to control the relative humidity and temperature during the production of uranium particles from the controlled hydrolysis of uranium hexafluoride (UF_6) aiming at the production of single uranium particles in the 1 µm range. The same aerosol deposition chamber was used to produce the reference particles for NUSIMEP-7, but with an improved and optimised analytical protocol.

6.2 Preparation

UF_6 reference material, stored in a monel (cupper-nickel alloy) ampoule, was used for NUSIMEP-7. Milligram amounts of this certified UF_6 was distilled into a glass vial. The set-up of the distillation unit is shown in Fig. 1.

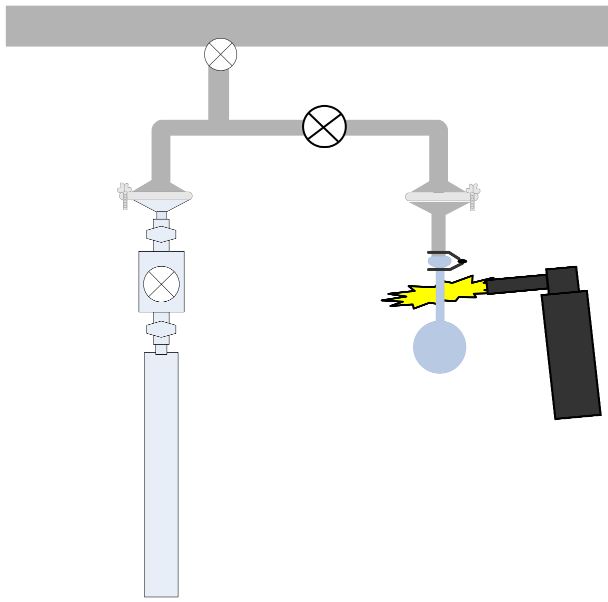

Fig. 1: The distillation unit to transfer milligram amounts of certified UF_6 from a reference material ampoule to a glass bulb

After transfer, the glass vial containing the gaseous UF_6 was placed into the upper part of the aerosol deposition chamber. The apparatus consists of an aluminium cylindrical reaction chamber with lids in Plexiglas (Fig. 2). The glass vial containing the UF_6 reference material was broken by a pin that was inserted by turning a screw from the top of the chamber. In this way, the UF_6 was released and subsequently hydrolyzed.

Fig. 2: Set-up of the aerosol deposition chamber

The humidity and the temperature of the air inside the chamber were monitored by a hygrometer (Rotronic). The relative humidity varied between 61.9 % and 81.1 %. The temperature of the air was about 21 ºC. The reaction between the released uranium hexafluoride and the atmospheric moisture in the deposition chamber proceeds very rapidly to form solid uranium oxyfluoride particles and hydrogen fluoride. The simplified overall equation is as follows:

$$UF_6 + 2H_2O \rightarrow UO_2F_2 + 4HF$$

At the base of the aerosol deposition chamber, a retractable platform containing 6 graphite discs of 25 mm diameter was used to collect the settling uranium oxyfluoride particles. This platform was inserted in the chamber after breaking the glass vial. In this way, the collection of glass shards from the UF_6 vial, that are generally much larger than the uranium-bearing particles, was avoided and the particle density was decreased. The retractable platform with the graphite discs was inserted after about 60 minutes. The uranium oxyfluoride particles were collected for about half a minute. After this time, the graphite discs could be removed from the aerosol deposition chamber. They were placed under a quartz lamp for 1 hour at full capacity, in order to remove excess water and other volatile elements. This heating procedure typically removed most of the fluorine in the particles, hereby changing the molecular structure to U_3O_8.

The particle morphology was then verified by scanning electron microscopy (SEM) for all of the NUSIMEP-7 samples.

One participant provided an estimation of the particle size distribution of one of his NUSIMEP-7 samples to IRMM (see Figure 3).

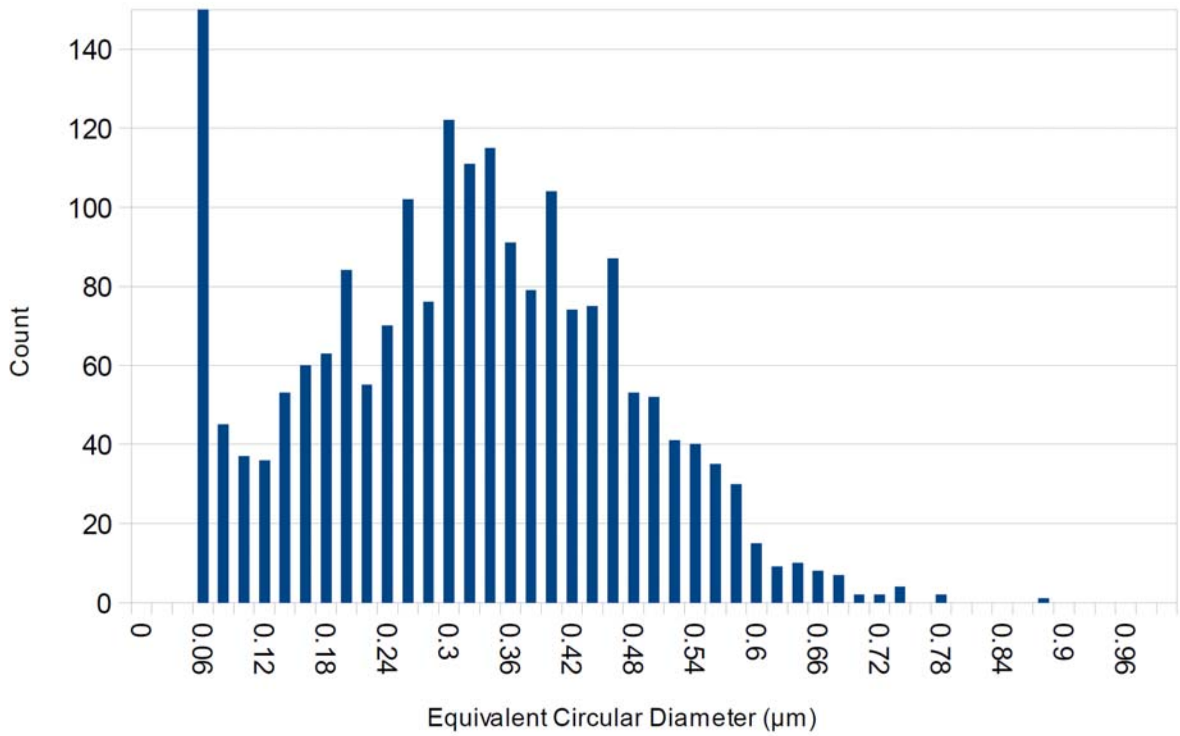

Figure 3: The histogram of equivalent circular diameter data collected from 250 field images

A circular area comprising of approximately the inner 50% of the area of the carbon planchet was tiled with approximately 24000 100 µm × 100 µm fields. 250 of these fields were selected at random and imaged with SED and BSED at an image dimension of 2048 pixel × 2048 pixels. The resulting BSE images were batch processed using ImageJ to threshold the high Z particles. The area of each particle was tabulated and transferred to a spreadsheet. The equivalent circular diameter ($ECD=2\cdot\sqrt{(A/\pi)}$) was calculated and plotted (see Figure 3)

The results for particle distribution on the NUSIMEP-7 sample were the following:

Particle count: 1851 in 250 fields = 7.4 particles/field (single pixels ignored)

Average diameter: 0.327 µm

Standard deviation: 0.139 µm

In Figure 3 a maximum at the equivalent circular diameter corresponding to a single pixel can be seen. This is likely due to noise pixels which creep in as the threshold approached the background. The particle count and statistics exclude the 297 single pixel events.

The NUSIMEP-7 samples were fixed in boxes using a commercial available glue roller, packed together with silica-gel in plastic bags and stored at room temperature until dispatch.

6.3 Verification

The NUSIMEP-7 uranium particles are produced from certified uranium hexafluoride reference materials. These reference materials were certified by gas mass spectrometry for the $n(^{235}U)/n(^{238}U)$ ratio and by thermal ionisation mass spectrometry for the $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ ratios. From previous studies it was known that no isotopic effects occur during aerosol deposition of uranium hexafluoride [10]. Nevertheless, isotope ratio measurements of particles on samples taken from each produced batch were performed. Due to the different approach in NUSIMEP-7 compared to NUSIMEP-6, where bulk analysis using Thermal Ionisation Mass Spectrometry (TIMS) was used for verification, this time an instrumental technique needed to be used that could provide average results on an exact number of single particles. For that reason Secondary Ion Mass Spectrometry (SIMS), being a mainstay technique for particle analysis, was applied in cooperation with JRC-ITU and IAEA-SGAS to verify the quality of the produced particles. The method repeatability using SIMS for single particle analysis cannot meet the requirements for method repeatability using TIMS (< 0.2% relative standard uncertainty) when performing bulk analysis of uranium particles, the approach that was used in NUSIMEP-6. Therefore in NUSIMEP-7 it was aimed at a relative standard uncertainty for method repeatability of about 0.5% for $n(^{235}U)/n(^{238}U)$, 5% for $n(^{234}U)/n(^{238}U)$ and 30% for $n(^{236}U)/n(^{238}U)$ using SIMS.

6.4 Homogeneity

Due to the limited number of test samples (6 planchets) that can be produced per deposition in the aerosol deposition chamber the strategy was adopted for the homogeneity study measuring one planchet for the isotopic composition of uranium per batch produced. 13 series (6 single isotopic depositions and 7 double isotopic depositions) of 6 planchets were prepared using the aerosol deposition chamber. All batches were investigated using Scanning Electron Microscopy (SEM) to check the particle density and subsequently 1 planchet per batch was measured according to the procedure described in Paragraph 6.3. For the samples with single isotopic composition a similar statistical approach as in NUSIMEP-6 was applied to verify homogeneity [14]. For the samples with two different isotopic compositions homogeneity assessment is not straightforward because the samples are inhomogeneous per definition. Results from the measurements of the 6 single isotopic depositions were evaluated by a one-way analysis of variance (ANOVA) [15, 16, 17]. This allows the separation of the method variation (s_{wb}) from the experimental averages over the particles measured on one planchet to obtain estimation for the real variation between planchets (s_{bb}), with u^*_{bb} being the lower limit to the between planchet variance which depends on the mean squares between planchets, the number of replicate measurements (= particles measured) per planchet and the degrees of freedom of the mean squares within planchets. It can be understood as the “detection limit” of the homogeneity study. The uncertainty of homogeneity is consequently estimated as s_{bb} or in case of $s_{bb}<u^*_{bb}$ as u^*_{bb}. This approach, applying single factor ANOVA as described in [15, 16, 17] was found to be comparable to tests

to determine whether an ILC material is sufficiently homogeneous for its purpose as described in ISO 13528 [2]. The variation between units (s_{bb}) for $n(^{234}U)/n(^{238}U)$ and $n(^{235}U)/n(^{238}U)$ was about 0.6%, and for $n(^{236}U)/n(^{238}U)$ about 11%. Essentially, these tests compare the unit heterogeneity with the standard deviation for proficiency assessment $\hat{\sigma}$. Assessment criterion for a homogeneity check is s_{bb} (or u^*_{bb}) ≤ 0.3$\hat{\sigma}$.

One of the aims of the previous NUSIMEP-6 pilot intercomparison was to picture the present measurement capabilities for uranium particle analysis. The results from NUSIMEP-6 served as valuable input to safeguards authorities to define assessment criteria for the future, particularly for the minor isotopes and in view of advances of instrumental techniques in the last couple of years. There was an agreement during respective technical meetings of the NUSIMEP organisers with the IAEA in the planning phase of NUSIMEP-7 that the safeguards requirements for particle analysis from swipe samples can be expressed as follows:

1) NWAL laboratories should report the true value for the major isotope ratio within 1% relative expanded uncertainty.

2) NWAL laboratories have to be able to measure the minor isotope amount ratios in case the isotope amount fraction of the respective isotope is above 10^{-6}; for isotope amount fractions below 10^{-6} an upper limit has to be reported to the safeguards authorities.

These requirements translated into more stringent performance criteria $\hat{\sigma}$ in NUSIMEP-7 compared to NUSIMEP-6. Therefore the standard deviation for proficiency assessment $\hat{\sigma}$ was set to 0.005X_{ref} for $n(^{235}U)/n(^{238}U)$, 0.025X_{ref} for $n(^{234}U)/n(^{238}U)$ and 0.5X_{ref} for $n(^{236}U)/n(^{238}U)$. The tests indicate that the uranium test material is sufficiently homogeneous for $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ in the frame of this ILC. For $n(^{235}U)/n(^{238}U)$ the test indicated that s_{bb} ≥ 0.3$\hat{\sigma}$, which is due to the fact that the method repeatability used for the homogeneity assessment was > 0.2%, see Table 1. It was not feasible to perform additional bulk measurements with TIMS, but due to the fact that the percentage difference of the average values $n(^{235}U)/n(^{238}U)$ from the certified value of the UF_6 reference material was ≤ 1% for all batches the uranium test material was considered sufficiently homogeneous for the purpose of this interlaboratory comparison and the assessment criterion for $n(^{235}U)/n(^{238}U)$ was not changed.

6.5 Stability

From previous interlaboratory comparisons and the production of isotopic reference materials it is well known that no isotopic effects occur over time when storing samples properly. Therefore after the homogeneity was assessed to be fit for purpose, the sample dispatch was started. Short term stability of uranium particles has been demonstrated in NUSIMEP-6 [14]. In addition a short term stability study was carried out with the aim of verifying the isotope ratios on one NUSIMEP-7 planchet. This planchet was stored at room temperature for 3 months and then measured using Large Geometry SIMS (LG-SIMS), at a time when all the participants had already reported their measurement results. Methods to assess whether an ILC material is sufficiently stable for its purpose are described in ISO 13528 [2]. Essentially, these tests compare the general averages of the measurand obtained in the homogeneity check (x_s) with those obtained in the stability check (y_s). The absolute difference of these averages is again compared to the standard deviation for proficiency assessment $\hat{\sigma}$. Assessment criterion for a stability check is $|x_s - y_s| \leq 0.3\hat{\sigma}$. The tests indicated that the uranium test material is sufficiently stable for all the ratios in the frame of this ILC, see Table 1. The homogeneity test results for $n(^{235}U)/n(^{238}U)$ and the stability test results for $n(^{234}U)/n(^{238}U)$ measurements on single particles were slightly > 0.3$\hat{\sigma}$, but the percentage difference between the average values and the certified values of the UF_6

reference material was ≤ 1%. The material was therefore found to be appropriate for the purpose of this interlaboratory comparison.

Table 1: Homogeneity and stability tests for the uranium isotope amount ratios according to ISO 13528 [2]

	s_{bb}	standard deviation for proficiency assessment $\hat{\sigma}$	$0.3\hat{\sigma}$	Homogeneity check $s_{bb} \leq 0.3\hat{\sigma}$	Stability check $\lvert x_s - y_s \rvert \leq 0.3\hat{\sigma}$
$n(^{234}U)/n(^{238}U)$	0.61%	$0.025X_{ref}$	$5.58 \cdot 10^{-7}$	YES	NO**
$n(^{235}U)/n(^{238}U)$	0.64%	$0.005X_{ref}$	$1.36 \cdot 10^{-5}$	NO*	YES
$n(^{236}U)/n(^{238}U)$	10.81%	$0.5X_{ref}$	$4.01 \cdot 10^{-6}$	YES	YES

*Since the percentage difference between the average value $n(^{235}U)/n(^{238}U)$ and the certified value of the UF_6 reference material was ≤ 1% for all batches the uranium test material was considered sufficiently homogeneous for the purpose of this interlaboratory comparison.
**Since the percentage difference between the average value from short term stability testing of $n(^{234}U)/n(^{238}U)$ and the certified value of the UF_6 reference material was < 0.1% the uranium test material was considered sufficiently stable for the purpose of this interlaboratory comparison.

It was recommended to the participants to store the sample in a dry environment after receipt.

6.6 Distribution

The ILC samples were dispatched to the participants by IRMM on 31 May 2011. Each participant received a package with two graphite planchets, a letter with information on particle density, sample handling, result reporting and a form to confirm receipt of the package.

7 Participant invitation, registration and information

Participation of the NWAL laboratories in this NUSIMEP interlaboratory comparison was formally recommended by the IAEA. Furthermore NUSIMEP-7 was announced in relevant conferences and meetings. Invitations were sent to the NWAL laboratories and other participants who expressed their interest in participation. Measurement of the major ratio $n(^{235}U)/n(^{238}U)$ was obligatory, measurement of the minor ratios $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ was optional. Participants were invited to follow their routine procedures.
Participants were also informed that their measurement results will be evaluated against the certified reference values and on the confidentiality of results. The call for participation was also announced on the IRMM website and confirmation of registration was sent to those participants who had registered (cf. annex 1 and annex 2 respectively). This confirmation contained further details on the envisaged time frame. Instructions on measurands, sample storage, and measurements were sent to the participants together with the samples. The instructions also contained the individual code for access to the result reporting and the related questionnaire website (cf. annex 3, annex 4). After closure of the result reporting the participants received the NUSIMEP-7 reference values. Table 2 lists the number of participants per country.

Table 2: Number of participants per country

Country	**Number of participants**
Australia	1
Austria	1
Belgium	1
China	1
Denmark	1
European Commission	2
France	2
Germany	1
Hungary	1
Lithuania	1
Republic of Korea	1
Russian Federation	1
Serbia	1
Sweden	1
Switzerland	1
United Kingdom	3
United Nations	1
United States	3

8 NUSIMEP-7 reference values

The NUSIMEP-7 uranium particles are produced from certified uranium hexafluoride materials. The certificate is attached in annex 14.

Table 3 lists the NUSIMEP-7 reference values X_{ref} and their associated expanded uncertainties U_{ref} (k=2).

Table 3: NUSIMEP-7 reference values

SINGLE DEPOSITION

Isotope Amount Ratio	**Certified Value**	**Expanded Uncertainty *U*, *k=2***
$n(^{234}U)/n(^{238}U)$	0.000 074 365	0.000 000 060
$n(^{235}U)/n(^{238}U)$	0.009 072 6	0.000 004 5
$n(^{236}U)/n(^{238}U)$	0.000 008 020 5	0.000 000 007 1

DOUBLE DEPOSITION

Enrichment 1:

Isotope Amount Ratio	Certified Value	Expanded Uncertainty *U*, *k=2*
$n(^{234}U)/n(^{238}U)$	0.000 074 365	0.000 000 060
$n(^{235}U)/n(^{238}U)$	0.009 072 6	0.000 004 5
$n(^{236}U)/n(^{238}U)$	0.000 008 020 5	0.000 000 007 1

Enrichment 2:

Isotope Amount Ratio	Certified Value	Expanded Uncertainty *U*, *k=2*
$n(^{234}U)/n(^{238}U)$	0.000 345 14	0.000 000 24
$n(^{235}U)/n(^{238}U)$	0.034 148	0.000 017
$n(^{236}U)/n(^{238}U)$	0.000 103 268	0.000 000 070

9 Reported results

9.1 General observations

Fourteen institutes reported measurement results, among those 7 NWAL laboratories. Participants from the same institute applying more than one analytical method had to register separately. 17 out of 24 registered participants submitted results in NUSIMEP-7 for the single and double deposition samples and completed the associated questionnaire. The laboratories were asked to apply their routine measurement procedure and to report on the sample with a single isotopic deposition 10 different particles with their average. On the sample with two isotopic depositions, the participants were asked to report two times 10 different particles (i.e. 10 particles per enrichment) and their respective averages. For every result also the uncertainty and coverage factor had to be reported. For both samples, the uranium isotope amount ratios were to be measured. Measurement of the major ratio $n(^{235}U)/n(^{238}U)$ was obligatory; measurement of the minor ratios $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ were optional, but it was highly recommended to report also the minor ratios.
All laboratories, that submitted results, reported the major ratio $n(^{235}U)/n(^{238}U)$. 12 participants reported the minor ratios $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$. For the $n(^{236}U)/n(^{238}U)$, one participant reported an upper limit. One participant reported results for the uranium isotope ratios that were a factor 10-100000 higher than the reference values. All results in NUSIMEP-7 are displayed/listed as reported by the participants.

Table 4: Reported results per participant

Country	$n(^{234}U)/n(^{238}U)$			$n(^{235}U)/n(^{238}U)$			$n(^{236}U)/n(^{238}U)$		
	Single	Double		Single	Double		Single	Double	
		E1	E2		E1	E2		E1	E2
Australia	✓	✓	✓	✓	✓	✓	✓	✓	✓
Austria	✓	✓	✓	✓	✓	✓	✓	✓	✓
Belgium									
China	✓	✓	✓	✓	✓	✓	✓	✓	✓
Denmark									
European Commission	✓	✓	✓	✓	✓	✓	✓	✓	✓
European Commission									
France				✓	✓	✓			
France	✓	✓	✓	✓	✓	✓	✓	✓	✓
Germany									
Hungary									
Lithuania									
Republic of Korea	✓	✓	✓	✓	✓	✓	✓	✓	✓
Russian Federation	✓	✓	✓	✓	✓	✓	✓	✓	✓
Serbia	✓	✓	✓	✓	✓	✓	✓	✓	✓
Sweden	✓			✓	✓	✓	✓		
Switzerland				✓	✓	✓			
United Kingdom				✓	✓	✓			
United Kingdom	✓	✓	✓	✓	✓	✓	✓	✓	✓
United Kingdom				✓	✓	✓			
United Nations	✓	✓	✓	✓	✓	✓	✓	✓	✓
United States	✓	✓	✓	✓	✓	✓	✓	✓	✓
United States				✓	✓	✓			
United States									

9.2 Measurement results

Annexes 5-13 list the individual measurement results and display overview graphs. The graphs for $n(^{234}U)/n(^{238}U)$, $n(^{235}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ of both the single and double deposition samples show a roughly normal distribution with no irregularities. One participant reported an upper limit for $n(^{236}U)/n(^{238}U)$. According to safeguards requirements NWAL laboratories can report an upper limit in case the isotope amount fraction of the minor isotopes is below 10^{-6}, which is not the case in NUSIMEP-7; $n(^{236}U)/n(^{238}U) > 10^{-6}$ (see also paragraph 10.1). However the participant concerned was not part of the IAEA NWAL.

Annexes 5-13 display the results from the NWAL laboratories and results according to participant's replies to the questionnaire.

10 Scoring of results

10.1 The scores and their settings

Individual laboratory performance is expressed in terms of z and zeta scores in accordance with ISO 13528 [2]:

$$z = \frac{x_{lab} - X_{ref}}{\hat{\sigma}} \qquad \text{and} \qquad zeta = \frac{x_{lab} - X_{ref}}{\sqrt{u_{ref}^2 + u_{lab}^2}}$$

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

$\hat{\sigma}$ is the standard deviation for proficiency assessment

Both scores can be interpreted as: satisfactory result for |score| ≤ 2, questionable result for 2 < |score| ≤ 3 and unsatisfactory result for |score| > 3.

z score

The NUSIMEP-7 *z* score indicates whether a laboratory is able to perform the measurement in accordance with what can be considered as good practice for NWAL laboratories. The standard deviation for proficiency assessment $\hat{\sigma}$ is accordingly based on present safeguards requirements for the measurements of $n(^{235}U)/n(^{238}U)$ in environmental samples and on the ILC organiser's assessment after discussions with experts from SAL in the field for the $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ ratios. The IUPAC International Harmonised Protocol [18] suggests that participants can apply their own scoring settings and recalculate the scores if the purpose of their measurements is different. In this ILC, $\hat{\sigma}$ is $0.005X_{ref}$ for $n(^{235}U)/n(^{238}U)$, $0.025X_{ref}$ for $n(^{234}U)/n(^{238}U)$ and $0.5X_{ref}$ for $n(^{236}U)/n(^{238}U)$.

zeta score

The zeta score provides an indication of whether the estimate of uncertainty is consistent with the laboratory's deviation from the reference value [2]. It is calculated only for those results that were accompanied by an uncertainty statement. The interpretation is similar to the interpretation of the *z* score. An unsatisfactory zeta score may be caused by an underestimated uncertainty or by a large deviation from the reference value.

The standard uncertainty of the laboratory (u_{lab}) was calculated as follows: if an uncertainty was reported, it was divided by the coverage factor *k*. If no coverage factor was provided, the reported uncertainty was considered as the half-width of a rectangular distribution. The reported uncertainty was then divided by √3, in accordance with recommendations issued by Eurachem and CITAC [19].

10.2 Scoring the reported measurement results

A *z* score was calculated for all participants except for those who reported no value or an upper limit, "<" value. A zeta score was calculated for results that were accompanied by an uncertainty statement. Annexes 5-13 list the scores per ratio and participant in detail, and annex 17 summarises the scores per participant.

Table 5 summarises the scores per isotope amount ratio.

Due to the more stringent performance criteria and the complexity of the certified test samples in NUSIMEP-7, the distribution of scores for the major ratio was different to the one in NUSIMEP-6. Expressing laboratory performance by means of the *z* score, about half of the participants reported satisfactory measurement results, the other half unsatisfactory results for $n(^{235}U)/n(^{238}U)$, around 10% reported "questionable" results, which would still have been satisfactory when applying the performance criteria from NUSIMEP-6. Concerning the minor isotope ratios the performance of laboratories participating in NUSIMEP-7 was about to be comparable with NUSIMEP-6, although more laboratories were able to measure $n(^{236}U)/n(^{238}U)$ due to the higher ^{236}U abundance in the UF_6 base materials. It has to be kept in mind that participants can apply their own scoring settings and recalculate the scores if the purpose of their measurements is different [18]. Considering the challenging performance criteria as set for the IAEA-NWAL, it can be concluded that the participants in general performed quite well in NUSIMEP-7.

Table 5: Overview of scores: S(atisfactory), Q(uestionable), U(nsatisfactory)

Single deposition

	z score				zeta score				both z and zeta scores
	S	Q	U	n (*)	S	Q	U	n (*)	S
$n(^{234}U)/n(^{238}U)$	67%	-	33%	12	83%	-	17%	12	8
$n(^{235}U)/n(^{238}U)$	47%	6%	47%	17	53%	18%	29%	17	5
$n(^{236}U)/n(^{238}U)$	73%	-	27%	11	73%	-	27%	11	8

(*) n is the number of results for which a score was given.
The total number of participants (with and without a score) is 17.

Double deposition enrichment 1

	z score				zeta score				both z and zeta scores
	S	Q	U	n (*)	S	Q	U	n (*)	S
$n(^{234}U)/n(^{238}U)$	55%	18%	27%	11	91%	-	9%	11	6
$n(^{235}U)/n(^{238}U)$	41%	18%	41%	17	65%	-	35%	17	5
$n(^{236}U)/n(^{238}U)$	80%	-	20%	10	70%	10%	20%	10	7

(*) n is the number of results for which a score was given.

The total number of participants (with and without a score) is 17.

Double deposition enrichment 2

	z score				zeta score				both z and zeta scores
	S	Q	U	n (*)	S	Q	U	n (*)	S
$n(^{234}U)/n(^{238}U)$	82%	-	18%	11	82%	-	18%	11	8
$n(^{235}U)/n(^{238}U)$	35%	-	65%	17	35%	18%	47%	17	3
$n(^{236}U)/n(^{238}U)$	82%	-	18%	11	73%	9%	18%	11	7

(*) n is the number of results for which a score was given.
The total number of participants (with and without a score) is 17.

11 Further information extracted from the results

In addition to submission of the results, the participants were asked to answer a number of questions relating to the measurements. All participants completed the questionnaire. Issues that may be relevant to the outcome of the intercomparison are discussed below.

11.1 Methods of analysis

The methods of analysis applied were TIMS by 3 participants, SIMS by 9 participants, ICP-MS by 4 participants, and alpha spectrometry by 1 participant. 1 participant selected the particle with fission track, the others used either SIMS (9) for particle selection, SEM-EDX (3) or Laser Ablation (4). 4 participants involved a particle transfer step prior to measurements by using a micromanipulator or by swiping the samples.

For the single deposition sample satisfactory zeta scores were achieved for $n(^{235}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ by participants with all the different analytical methods, except alpha spectrometry, where an upper limit was reported. For the $n(^{234}U)/n(^{238}U)$ isotope amount ratio all the different analytical methods led to a satisfactory zeta-score. Not all the techniques resulted in satisfactory *z*-scores but this is, at least for the $n(^{235}U)/n(^{238}U)$ ratio, also due to the stringent performance requirement of $\hat{\sigma} = 0.005X_{ref}$ for $n(^{235}U)/n(^{238}U)$ in NUSIMEP-7 compared to NUSIMEP-6, where the performance criteria was $\hat{\sigma} = 0.01X_{ref}$ for this parameter. On the other hand NUSIMEP-7 confirmed that all the mass spectrometry techniques (TIMS, SIMS and ICP-MS) are advanced enough from an instrumental point of view to meet this performance requirement. In addition it confirms the technical expertise available at institutes world-wide using these techniques reliably for nuclear safeguards purposes. Remarkable is the excellent performance for major and minor ratios of the three laboratories that measured the NUSIMEP-7 certified test samples with LG-SIMS, demonstrating that this new generation of secondary ion mass spectrometers is indeed a step forward in environmental swipe sample analysis. Secondly, the good performance of some of the laboratories using LA-ICP-MS in NUSIMEP-7 shows clearly the potential of this analytical method for nuclear safeguards applications [[20]].

Annex 15 summarises the information given by the participants on instrument parameters and measurement conditions for SIMS, TIMS and LA-ICP-MS.

94% of the participants applied a correction for mass fractionation / mass bias to their measurement results. Most of the participants used a uranium standard to apply a correction to their measurement results.

11.2 A representative study

11 of the 17 participants indicated that the measurements were carried out according to the same analytical procedure routinely used for this kind of samples. 10 participants reported that they are experienced in this type of measurement. 65% of the participants indicated to analyse at least 11-50 samples per year, 5 participants analyse more than 50 samples per year. The mission of the majority of the laboratories participating in NUSIMEP-7 is to carry out measurements for fissile material control or safeguards but also for environmental sciences, including the 7 NWAL laboratories. The remaining 3 other participants are from the fields of nuclear forensics, biological applications, cosmochemistry, research & development and metrology. One of the participants is currently in qualification to become part of the IAEA NWAL. 9 participants indicated that their laboratories are either certified, accredited and/or authorised for this type of measurements. This confirms that NUSIMEP-7 is a useful and representative study for the current capability of laboratories in the field of uranium particle analysis.

11.3 Quality system and use of standards

All laboratories but four indicated that they are working according to a quality management system; 4 participants according to ISO 17025; 4 according to ISO 9000 series, 1 participant according to both ISO 17025 and ISO9000 series and 1 participant according to a ISO 17025 compliant system [21]. 82% of the participants confirmed participation in interlaboratory comparisons. The ILC schemes listed were REIMEP, NUSIMEP, CETAMA, NBL, ITWG, SRR-2001, SME, IRSN and IAEA ILCs [3, 22, 23, 24]. All participants but one routinely use certified reference materials mostly for instrument calibration but also for method validation. The certified reference materials used by the NUSIMEP-7 participants are listed in Annex 16.

11.4 Determination of measurement uncertainty

All of the participants who reported results for ratios and not upper limits provided an uncertainty estimate with a coverage factor. All the participants stated that they routinely report uncertainties on chemical measurements to their customers. 65% of the participants stated that their reported uncertainties in NUSIMEP-7 are calculated according to the Guide for Quantifying Measurement Uncertainty (GUM) issued by the International Organization for Standardization (ISO, 2005) and/or EURACHEM/CITAC (2000) [19, 25]. The other participants indicated that they evaluated their measurement uncertainty either via replicate measurements only, which causes that they are likely to underestimate their uncertainty, or somewhat similar to the GUM approach via performing replicate measurements of sample and measurement standards, including relevant correction factors for mass fractionation, hydrogen correction, baseline correction, dead time correction, linear drift correction etc. As already observed in NUSIMEP-6 there were also in NUSIMEP-7 sometimes large differences in the reported uncertainties even among participants using the same instrumental technique. This topic has been taken up in the ESARDA WGDA "Workshop on uncertainties in nuclear measurements", held at the IAEA-SGAS in November 2011 and will be further discussed in relevant publications [26].

11.5 Future NUSIMEP ILCs on particles

NUSIMEP-7 is an interlaboratory comparison that should not only picture the measurement capabilities of the participating laboratories in uranium particle analysis, but is also collecting feedback from the participants regarding future improvements and needs. Participants expressed interest in future NUSIMEP ILCs on particles, particularly on uranium, plutonium and uranium/plutonium mixed samples. The feedback on the desired particle density for future NUSIMEP samples was somewhat depending on the instrumental technique applied, but ranged from a particle density like the present NUSIMEP-7 sample to a smaller particle density starting from a known number e.g. 20 particles per sample up to 10 000 per mm² for LG-SIMS analysis. Participants using TIMS preferred a lower density than what was offered in NUSIMEP-7. One of the TIMS laboratories suggested 2 particles per mm². For half of the LA-ICP-MS labs the density of the provided samples was acceptable. The other half preferred a lower density with a range from 100 up to 10000 particles per mm².
Although the participants acknowledged that the NUSIMEP-7 samples were representative for real-life swipe samples taken by inspectors, most of the participants would appreciate that the particle density of the samples for the next NUSIMEP on uranium particles could even resemble more real inspector samples. The "wish–list" from participants concerning isotopic composition of future NUSIMEP particles ranged from depleted uranium via a 'low' enriched to a mixed sample with variable enrichment. A few participants expressed also the need for larger particles (> 1μm). The majority of the participants confirmed that they would like to analyse uranium oxyfluoride particles in a future NUSIMEP ILC.

12 Feedback

The questionnaire invited laboratories to provide feedback of any kind to the ILC coordinators. One participant had difficulties with taking the sample out of the plastic box. The glue appeared to be too sticky. One participant suggested it would be significantly easier if the results could be submitted via a standard spreadsheet format to eliminate any risk of transcription errors. Another participant suggested it would be good for a future ILC to have a bit more challenging samples in terms of background interfering elements and to have the material on cotton swipes. 25 mm mounts were preferred by one of the labs having a Cameca instrument. Three participants noted the heavy loading of the samples. One of these labs estimated the number of particles to be > 50 000 and the other estimated the particle size in both the micron and sub-micron ranges. Another participant who made an estimate of the number of particles per planchet reported 170 000 for the single deposition and 150 000 for the double deposition. The deposition of particles on carbon planchets is not very suitable for the FT-TIMS method because the particles need to be removed first before undergoing the FT-TIMS process. Therefore the removed particles were not representative for their initial distribution on the planchet. One participant reported contamination with "alien particles" on both planchets (single and double deposition). The NUSIMEP-7 planchets produced during the same deposition in the aerosol deposition chamber were part of the homogeneity study and were re-measured for confirmation of the absence of any "alien" particle after the result reporting deadline. There was no trace of any particle that did not stem from one of the two UF_6 base materials. The contamination of the NUSIMEP-7 samples observed by the participant must have occurred at the participating laboratory. Additional measurements carried out using LG-SIMS on a sample from the same batch where the planchet of the participant originated from confirmed the excellent quality of the NUSIMEP-7 samples. Contamination, as observed by the participant, could definitely be excluded to have occurred at IRMM. During the

technical meeting on Particle Analysis of Environmental Samples for Safeguards, held at the IAEA, 4-6 October 2011 the following was recommended towards future NUSIMEP ILCs on uranium particles:
- NUSIMEP with samples of mono-dispersed uranium particles,
- Addition of environmental dust to NUSIMEP certified test samples,
- Adding particles of a third isotopic composition with a significantly lower abundance,
- Performance evaluation for multiple depositions (i.e. particles of different isotopic composition present on one sample),
- The NWAL, and laboratories that are considering qualification to become part of the NWAL, are encouraged to participate in NUSIMEP ILCs on uranium particles.

13 Conclusion

Isotopic "fingerprinting" is a powerful tool needed in nuclear safeguards for the verification of the correctness and completeness of a State's declarations and for attribution of intercepted materials, particularly in view of the detection of any undeclared material or activities. To this end, techniques like particle analysis have been implemented in safeguards laboratories and improvements on instrumentation and methods are ongoing. Conclusions must be based on reliable measurement results ensured with appropriate reference materials and conformity assessment tools. The fundamental importance of measurements of major and minor uranium isotopes in environmental samples has been stressed by the IAEA during technical expert meetings and, for instance, by the IAEA Director General during the inauguration ceremony of the Safeguards Clean Laboratory Extension at the IAEA-SGAS, in Seibersdorf, September 2011 [27]. During the joint workshop of the Institute of Nuclear Materials Management (INMM) and the European Safeguards R&D Association (ESARDA) on "Future Directions for Nuclear Safeguards and Verification", held in October 2011, a main focus was put on environmental sample analysis and in expanding IAEA analytical capabilities and the NWAL [28, 29].
NUSIMEP-7 was the first IRMM interlaboratory comparison providing a sample with uranium particles of two different isotopic compositions. Uranium particles with a single isotope composition were deposited on the other sample, as it was in NUSIMEP-6. Furthermore, compared to NUSIMEP-6, the particle density was optimised and the $n(^{236}U)/n(^{238}U)$ was > 10^{-6}. The major and minor isotope amount ratios in uranium were the measurands under investigation in NUSIMEP-7. The performance evaluation criteria were more stringent in NUSIMEP-7 than in NUSIMEP-6, especially for the major ratio $n(^{235}U)/n(^{238}U)$. This was clearly visible in the distribution of z and zeta scores for this ratio in comparison to NUSIMEP-6. Nevertheless, the measurement capabilities of the participants in the analysis of uranium particles for major and minor ratios were in general satisfactory. Differences were observed in the uncertainty estimates provided by the participants, even when using the same instrumental techniques. Remarkable is the excellent performance for major and minor ratios of the three laboratories that measured the NUSIMEP-7 certified test samples with LG-SIMS. In that sense NUSIMEP-7 was very beneficial to the SIMS measurement community in confirming the advances in particle analysis achievable with this powerful instrumentation. Furthermore NUSIMEP-7 confirmed that techniques that were currently investigated for their potential and application range to safeguards, such as LA-ICP-MS, demonstrated that they can meet performance requirements which have only been met by SIMS or TIMS until now. Last but not least the satisfactory measurement results achieved by leading experts with different instrumental techniques confirmed the high quality of the NUSIMEP-7 uranium reference particles provided as certified test sample by IRMM to the participants. The outcome of NUSIMEP-7 underlines once more that the definition of performance standards for

measurements in particle analysis is highly recommended and extremely useful for laboratories that are in the qualification process to become a part of the IAEA NWAL. The ESARDA Working Group on Techniques and Standards for Destructive Analysis (WGDA) undertakes establishing such performance standards as guidance for measurement laboratories, similar to the concept of "Target Values for Uncertainty Components" for element and isotope assay of nuclear materials [30]. One objective of NUSIMEP-7 was to collect feedback from the participants in view of optimisation of uranium reference particle production, particularly towards samples with a double or triple deposition. The feedback from NUSIMEP-7 participants was very positive and the efforts made by IRMM in providing this interlaboratory comparison to the measurement community were highly appreciated. The identified needs for reference particles for future NUSIMEP ILCs are manifold, but re-occurring requests are a lower particle density, a sample with a triple isotopic composition and a sample with uranium oxyfluoride particles and dust. In addition to the particles produced at IRMM simulating the real-life aerosol deposition process occurring in a nuclear facility, the need for mono-disperse uranium reference particles was stressed again.

The Director of the Division of Concepts and Planning at the IAEA Department of Safeguards expressed her appreciation and compliments to IRMM for NUSIMEP-7 for further improving the detection and analysis capability of the IAEA's NWAL. - *"The difficult-to-produce IRMM particle standards were of exceptionally high quality and proved to be very valuable for the IAEA"*

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Abbreviations

ANOVA	Analysis of Variance
AP	Additional Protocol
BSE	Backscattered Electron
BSED	Backscattered Electron Detector
CETAMA	Commission d'Etablissement des Méthodes d'Analyse
CITAC	Co-operation for International Traceability in Analytical Chemistry
CRM	Certified Reference Material
DG ENER	Directorate General for Energy
ECD	Equivalent Circular Diameter
ES	Environmental Sampling
ESARDA	European Safeguards Research and Development Association
ESARDA WG DA	ESARDA Working Group on Standards and Techniques for Destructive Analysis
EU	European Union
EURACHEM	A focus for Analytical Chemistry in Europe
FT-TIMS	Fission Track Thermal Ionisation Mass Spectrometry
GUM	Guide to the Expression of Uncertainty in Measurement
IAEA	International Atomic Energy Agency
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
ILC	Interlaboratory Comparison
IMEP	Interlaboratory Measurement Evaluation Programme
IRMM	Institute for Reference Materials and Measurements, JRC, European Commission
IRSN	Institut de Radioprotection et de Sûreté Nucléaire
ISO	International Organization for Standardization
ITU	Institute for Transuranium Elements, JRC, European Commission
ITWG	International Technical Working Group
IUPAC	International Union for Pure and Applied Chemistry
JRC	Joint Research Centre
LA	Laser Ablation
LA-ICP-MS	Laser Ablation Inductively Coupled Plasma Mass Spectrometry
LG-SIMS	Large Geometry Secondary Ion Mass Spectrometry
NBL	New Brunswick Laboratory
NPT	Treaty on the Non-Proliferation of Nuclear Weapons
NUSIMEP	Nuclear Signatures Interlaboratory Measurement Evaluation Programme
NWAL	Network of Analytical Laboratories
REIMEP	Regular European Interlaboratory Measurement Evaluation Programme
SAL	Safeguards Analytical Laboratory
SED	Secondary Electron Detector
SEM	Scanning Electron Microscopy
SEM-EDX	Scanning Electron Microscopy Energy Dispersive X-ray Spectroscopy
SEM-TIMS	Secondary Electron Multiplier Thermal Ionisation Mass Spectrometry
SGAS	Safeguards Analytical Services
SIMS	Secondary Ion Mass Spectrometry
SME	Safeguard Measurement Evaluation Programme
TIMS	Thermal Ionisation Mass Spectrometry

References

[1] http://www.iaea.org/Publications/Documents/Treaties/npt.html
[2] ISO 13528:2005: Statistical methods for use in proficiency testing by interlaboratory comparisons
[3] http://irmm.jrc.ec.europa.eu/interlaboratory_comparisons/nusimep/Pages/index.aspx
[4] Conformity assessment - General requirements for proficiency testing, ISO/IEC 17043:2010
[5] http://europa.eu/legislation_summaries/institutional_affairs/treaties/treaties_euratom_en.htm
[6] http://www.iaea.org/Publications/Documents/Conventions/index.html
[7] http://www.iaea.org/OurWork/SV/Safeguards/analytical-services.html
[8] Y. Aregbe, K. Mayer, M. Hedberg, S. Richter, J Poths, T. Prohaska, R. Kips, ESARDA BULLETIN, No. 40, December 2008, http://esarda2.jrc.it/bulletin/bulletin_40/index.html
[9] Y. Aregbe, T. Prohaska, Z. Stefanka, É. Széles, A Hubert, S Boulyga, ESARDA BULLETIN, No. 46, December 2011, http://esarda2.jrc.it/bulletin/bulletin_46/index.html
[10] R. Kips, R. Wellum, P.D.P. Taylor, M. Betti, G. Tamborini, A. Leenaers, S. Van Den Berghe, Characterisation of Particles Produced by Hydrolysis of UF6 Using SEM and SIMS, Microscopy and Microanalysis 13 (2007) 156-164
[11] R. Kips, A.J. Pidduck, M.R. Houlton, A. Leenaers, J.D. Mace, O. Marie, F. Pointurier, E. A. Stefaniak, P.D.P. Taylor, S. Van Den Berghe, P. Van Espen, R. Van Grieken, R. Wellum, Spectrochimica Acta Part B.
[12] M. Kraiem, S. Richter, H. Kühn, E.A. Stefaniak, G. Kerckhove, J. Truyens, and Y. Aregbe; Anal. Chem., 2011, 83 (8), pp 3011–3016
[13] M. Kraiem, S. Richter, H. Kühn, Y. Aregbe, Analytica Chimica Acta Vol. 688, Issue 1, 2011, 1–7
[14] NUSIMEP-6: Uranium isotope amount ratios in uranium particles; EUR 23702 EN - 2008
[15] T.P.J. Linsinger et al., Accred. Qual. Assur (2001) 6:20-25
[16] A.M.H. Van Der Veen, T.P.J. Linsinger and J. Pauwels, Accred. Qual. Assur. (2001) 6:26-30
[17] A.M.H. Van Der Veen, T.P.J. Linsinger, A. Lamberty, J. Pauwels, Accred. Qual. Assur. (2001) 6, 257-263
[18] M. Thompson, M.L.R. Ellison, R. Wood (2006), Pure Appl Chem 78:145–196
[19] Guide to the Expression of Uncertainty in Measurement, ISO (2005)
[20] Y. Aregbe, T. Prohaska, Z. Stefanka, É. Széles, A. Hubert, S. Boulyga, Report on the Workshop on LA-ICP-MS organized by the Destructive Analysis Working Group, ESARDA BULLETIN, No. 46, December 2011, http://esarda2.jrc.it/bulletin/bulletin_46/index.html
[21] General requirements for the competence of testing and calibration laboratories, ISO/IEC 17025:2005
[22] http://www.bipm.org
[23] http://www-cetama.cea.fr
[24] http://www.iaea.org
[25] Quantifying Uncertainty in Analytical Measurement, Eurachem/CITAC (2000), http://www.eurachem.org
[26] http://esarda2.jrc.it/internal_activities/WG-DA/index.html
[27] http://www.iaea.org/newscenter/news/2011/cleanlabext.html
[28] http://www.inmm.org/Content/NavigationMenu/CommunicatorWinter2012/ISD2011Roundup/default.htm
[29] http://esarda2.jrc.it/events/other_meetings/2011-Aix-en-Provence/index.html
[30] International Target Values 2000 for Measurement Uncertainties in Safeguarding Nuclear Materials, ESARDA BULLETIN, NO. 31, http://esarda2.jrc.it/bulletin/bulletin_31/08.pdf

Annexes

Annex 1: Invitation to nominate laboratories

JRC
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NUSIMEP

Geel, 10 March 2011
JRC.DDG.D.2/TL/JTr/acs/ARES(2011) 11-038/270001

The IRMM Nuclear Signatures Interlaboratory Measurement Evaluation Programme

NUSIMEP-7: Uranium isotope amount ratios in uranium particles

NUSIMEP is an external quality control programme organised by IRMM with the object of providing materials for measurements of trace amounts of nuclear materials in environmental matrices. Measurements of the isotopic ratios of the elements uranium and plutonium in small amounts, such as typically found in environmental samples, are required for nuclear safeguards, for the control of environmental contamination and for the detection of nuclear proliferation. Several NUSIMEP comparison campaigns of measurements of uranium isotopic ratios were organised previously: for example NUSIMEP 2, uranium isotopic abundances in dry uranium nitrate samples; NUSIMEP 3, uranium isotopic abundances in saline media; NUSIMEP 4, uranium isotopic abundances in a simulated urine; NUSIMEP 5 uranium, plutonium and caesium isotopic ratios in saline medium and NUSIMEP-6, uranium isotope amount ratios in uranium particles.

We would like to announce the forthcoming NUSIMEP-7 interlaboratory comparison: "Uranium isotope amount ratios in uranium particles" and invite laboratories to participate.

Participating laboratories in NUSIMEP-7 receive two test samples of uranium particles on a graphite planchet with undisclosed isotope amount ratio values $n(^{234}U)/n(^{238}U)$, $n(^{235}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$. One sample will have a single isotopic deposition and the other sample will consist of particles with two different isotopic compositions. For both samples, the uranium isotope amount ratios are to be measured by participating laboratories on a prescribed number of particles using their routine analytical procedures. Measurement of the major ratio $n(^{235}U)/n(^{238}U)$ is obligatory; measurement of the minor ratios $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ are optional. Although, it is highly recommended that participants from the IAEA network of analytical laboratories in the field of particle analysis submit results for major and minor isotope ratios. The participants' measurement results will be evaluated against the certified reference values. Full confidentiality is guaranteed with respect to the link between measurement results and the participants' identity.

Participation fee is € 200, including dispatch. 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 NUSIMEP-7 samples is exhausted.

Please register electronically for this interlaboratory comparison using the following link: https://irmm.jrc.ec.europa.eu/ilc/ilcRegistration.do?selComparison=640

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

The deadline for registration is **31 March 2011**. Samples will be sent to participants end of April 2011. The deadline for submission of results is 27 June 2011. Please do not hesitate to contact us in case you need more information.

Yours sincerely,

Jan Truyens
NUSIMEP-7 Co-ordinator

Yetunde Aregbe
IRMM Safeguards Co-ordinator

irmm Institute for Reference Materials and Measurements
Retieseweg 111, 2440 Geel, Belgium
Tel.: +32-(0)14-571 976 • Fax: +32-(0)14-571 865
jrc-irmm-nusimep@ec.europa.eu • http://www.irmm.jrc.be

Annex 2: Confirmation of registration

TRUYENS Jan (JRC-GEEL)

From: TRUYENS Jan (JRC-GEEL)
Sent: maandag 6 juni 2011 8:46
To: [redacted]
Subject: NUSIMEP-7: shipment of samples

Dear [redacted],

You were registered successfully for the Nuclear Signatures Interlaboratory Measurement Evaluation Programme: Uranium isotope amount ratios in uranium particles and now we're pleased to inform you that your NUSIMEP-7 sample has been sent. Within a couple of days it should arrive at your laboratory.

The sample consists of 2 graphite planchets covered with uranium particles with undisclosed isotope amount ratio values $n(^{234}U)/n(^{238}U)$, $n(^{235}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$. The uranium isotope amount ratios are to be measured using your routine analytical procedures. Measurement of the major ratio $n(^{235}U)/n(^{238}U)$ is obligatory; measurements of the minor ratios $n(^{234}U)/n(^{238}U)$ and $n(^{236}U)/n(^{238}U)$ are optional. Nevertheless, it is highly recommended that particularly the participants from the IAEA Network of Analytical Laboratories (NWAL) measure the major as well as the minor uranium ratios.

Your measurement results will be evaluated against the certified reference values. Full confidentiality is guaranteed with respect to the link between results and the participants' identity.

To login to the result reporting page you need a participant key, indicated on the accompanying letter to the sample. Please read the accompanying letter carefully and store the participant key ([redacted]) safely.

Also the guidelines for reporting your results online are sent together with the sample.

Please be aware of the **reporting deadline**, which is **01 September 2011**.

After we've received your "Confirmation of receipt" form, we will send an invoice to pay the participation fee.

We wish a lot of success with your measurements and please do not hesitate to contact us in case of any questions or problems.

Kind regards,

Jan Truyens
NUSIMEP-7 Co-ordinator

Yetunde Aregbe
IRMM Safeguards Co-ordinator

European Commission (EC) / Joint Research Centre (JRC)
Institute for Reference Materials and Measurements (IRMM)
Reference Materials Unit
Retieseweg 111
B-2440 Geel
Belgium

Tel: +32 14 571 976
Fax: +32 14 571 548

jrc-irmm-nusimep@ec.europa.eu
www.irmm.jrc.be

Annex 3: Instructions for measurement and reporting

EUROPEAN COMMISSION
JOINT RESEARCH CENTRE

Institute for Reference Materials and Measurements
Reference Materials Unit

NUSIMEP

Geel, 31 May 2011
JRC.DDG.D.2/YAr/JTr/FvV/ARES(2011)
11-071/578551

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

NUSIMEP-7

Dear «TITLE» «SURNAME»,

Thank you very much for your participation in NUSIMEP-7.
Together with this letter we are sending to you two NUSIMEP-7 graphite planchet samples for particle analysis as specified in the NUSIMEP-7 announcement:

http://www.irmm.jrc.be/interlaboratory_comparisons/nusimep/Nusimep-7/Pages/index2.aspx

Please check whether the test material remained undamaged during transport. Then sign the "Confirmation of receipt" form and send it by email or fax it to us (Fax: +32 14 571 548).
The particles are separated by at least a few micrometers on average, although small agglomerates may also be present on the graphite planchet substrate. It is recommended to store the sample in a dry environment.

Participants in NUSIMEP are asked to apply the same measurement procedure as used in routine sample analysis of this kind.

For both samples, the uranium isotope amount ratios are to be measured. On the sample with a single isotopic deposition, 10 different particles are to be measured. On the sample with two isotopic depositions, two times 10 different particles are to be measured (i.e. 10 particles for each enrichment). For both samples also the averages are to be reported. Measurement of the major ratio $n(^{235}\text{U})/n(^{238}\text{U})$ is obligatory; measurement of the minor ratios $n(^{234}\text{U})/n(^{238}\text{U})$ and $n(^{236}\text{U})/n(^{238}\text{U})$ are optional, but it is highly recommended to report also the minor ratios.

You can find the reporting website at:

http://intranet.irmm.jrc.be/ilcexport

To access this webpage you need a personal password key, which is:

«Part_key»

IRMM - Retieseweg 111, B-2440 Geel - Belgium. Telephone: +32 (0)14 571 211. http://irmm.jrc.ec.europa.eu
Telephone: direct line +32 (0)14 571 976. Fax: +32 (0)14 571 548.

E-mail: JRC-IRMM-NUSIMEP@ec.europa.eu

The system will guide you through the reporting procedure. The result reporting page will be active from the end of May on. After entering your results, please also complete the questionnaire. Do not forget to submit and confirm always when required. Directly after submitting your results and after having filled out the questionnaire online, you will be prompted to print the completed report form. Please do so, sign the paper version and return it to IRMM by fax (at +32 14 571 548) or by e-mail. Check your results carefully for any errors before submission, since this is your definitive confirmation.

The deadline for submission of results is 01 September 2011.

Although we have no doubts about your professionalism, we would like to mention that collusion nullify the benefits of interlaboratory comparisons to customers, analysts and accreditation bodies.

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

Yours sincerely,

Jan Truyens
NUSIMEP-7 Co-ordinator

Yetunde Aregbe
IRMM Safeguards Co-ordinator

IRMM - Retieseweg 111, B-2440 Geel - Belgium. Telephone: +32 (0)14 571 211. http://irmm.jrc.ec.europa.eu
Telephone: direct line +32 (0)14 571 976. Fax: +32 (0)14 571 548.

E-mail: JRC-IRMM-NUSIMEP@ec.europa.eu

EUROPEAN COMMISSION
JOINT RESEARCH CENTRE

Institute for Reference Materials and Measurements
Isotope measurements

NUSIMEP

Geel, 31 May 2011
JRC.DDG.D.2/YAr/JTr/FvV/ARES(2011)
11-071/578551

«TITLE» «FIRSTNAME» «SURNAME»
«ORGANISATION»
«DEPARTMENT»
«ADDRESS»
«ADDRESS1»
«ADDRESS2»
«ADDRESS3»
«ZIP» «TOWN»
«COUNTRY»

NUSIMEP-7

Confirmation of receipt of two graphite planchet samples

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 CODES ....-................

ANY REMARKS ....

....

Date of package arrival ....

Signature ....

Please return this form to:

Jan Truyens
NUSIMEP-7 Co-ordinator
EC-JRC-IRMM
Retieseweg 111
B-2440 GEEL
BELGIUM
Fax : +32 14 571 548
e-mail : jrc-irmm-nusimep@ec.europa.eu

IRMM - Retieseweg 111, B-2440 Geel - Belgium. Telephone: +32 (0)14 571 211. http://irmm.jrc.ec.europa.eu
Telephone (direct line): +32 (0)14 571 976. Fax: +32 (0)14 571 548

Annex 4: Questionnaire

Milc questionnaire

Comparison for NUSIMEP-7

PLEASE COMPLETE THIS FORM TOGETHER WITH THE RESULT REPORTING FORM. ALL ANSWERS WILL BE TREATED CONFIDENTIALLY (non-disclosure of the identity of the laboratories).

Submission Form

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

☐ a) Environmental sciences

☐ b) Measurements for fissile material control or safeguards

☐ c) Network of Analytical Laboratories (NWAL)

☐ d) Other

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

[ ]

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

☐ a) Accredited

☐ b) Authorised

☐ c) Certified

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

○ a) Yes

○ b) No

3.1. If 'Yes' please specify:

☐ a) ISO 17025

☐ b) ISO 9000 series

☐ c) Other

3.1.1. If 'Other' please specify:

[ ]

4. **Does your laboratory participate in inter-laboratory comparisons?** *

- a) Yes
- b) No

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

5. **How many measurement of this type does your laboratory routinely perform per year?** *

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

6. **How does your laboratory rate itself for these types of measurement?** *

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

7. **Was the NUSIMEP-7 sample treated according to the same analytical procedure routinely used for this sample type?** *

- a) Yes
- b) No

7.1. If 'No' please specify why not: *

8. **Was a chemical treatment applied to the NUSIMEP-7 sample?** *

- a) Yes
- b) No

8.1. If 'Yes' please specify *

9. **How did you select (scan) the particle(s) for your measurements?** *

- a) FT
- b) SEM-EDX
- c) SIMS

9.1. Please give a brief description on the particle selection: *

10. **Was a particle transfer needed?** *

- a) Yes
- b) No

10.1. If 'Yes' please describe the particle transfer: *

11. **Did you make an estimate of the number of particles per planchet?** *

- a) Yes
- b) No

11.1. If 'Yes', please indicate your estimation of the number of particles for the SINGLE DEPOSITION. *

(number)

11.2. If 'Yes', please indicate your estimation of the number of particles for the DOUBLE DEPOSITION *

(number)

12. **Did you make an estimate of the particle size distribution?** *

- a) Yes
- b) no

12.1. If 'Yes', please indicate your estimation of the particle size distribution. *

(number)

13. **Does your laboratory routinely use Certified Reference Materials?** *

- a) Yes
- b) No

13.1. If 'Yes' please specify which CRMs and suppliers: *

13.2. How are the CRMs applied? *

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

13.2.1. If 'Other' please specify:

14. **Did you apply a correction for mass fractionation / mass bias to your measurement results?** *

- a) Yes
- b) No

14.1. If "Yes", how was the mass fractionation determined? *

15. **Did you use Secondary Ion Mass Spectrometry (SIMS) to analyse the uranium particles?** *

- a) Yes
- b) No

15.1. If 'Yes', please provide the following information on the instrument parameters:

15.2. Type of SIMS used (brand, model,..): *

15.3. Did you use automatic particle search software? *

- a) Yes
- b) No

15.3.1. If 'Yes', which software? *

15.4. Primary ion beam and primary ion beam accelerating energy: *

15.5. Secondary ion extraction kV: *

15.6. Mass resolution used: *

15.7. Secondary ion energy window: *

15.8. EM detector type: *

15.9. Dead time correction applied? *

- a) Yes
- b) No

15.9.1. If 'Yes' report the dead time: *

16. **Did you use Secondary Ion Mass Spectrometry (SIMS) to analyse the uranium particles?** *

- a) Yes
- b) No

16.1. If 'Yes', please provide the following information on the measurement conditions:

16.2. Presputter conditions (before microprobe measurements) *

16.3. Microprobe measurement conditions: *

16.4. Spot size (estimated diameter): *

16.5. Raster size (if used): *

16.6. Masses cycled & cycle times: *

16.7. Total sputter time per measurement: *

17. **Did you use Secondary Ion Mass Spectrometry (SIMS) to analyse the uranium particles?** *

- a) Yes
- b) No

17.1. If 'Yes', please provide the following information for each measurement:

17.2. 238U signal level from particle at start (cps): *

17.3. 238U signal level from particle at end (cps) *

17.4. Comment on stability of signal levels during microprobe: stable, decreasing, increasing, variable? *

17.5. Comments on appearance of U ion image field (if possible): clear particles?, diffuse spots?, uniform signal? Did this change during the analysis? *

18. **Did you use Thermal Ionisation Mass Spectrometry (TIMS) to analyse the uranium particles?** *

- a) Yes
- b) No

18.1. If 'Yes', please provide the following information on the instrument parameters:

18.2. Type of TIMS used (brand, model,..): *

18.3. Detector used (brand): *

18.4. Please provide information on detector used: *

- a) Analogue detector operation mode
- b) Channeltron (contiuous dynode SEM)
- c) Discrete dynode SEM
- d) Ion counting detector operation mode
- e) multiple detectors used
- f) Single detector used

18.5. Dead time correction applied? *

- a) Yes
- b) No

18.5.1. If 'Yes' report the dead time: *

18.6. Did you apply any other correction to the detector output (e.g. SEM non-linearity correction)? *

- a) Yes
- b) No

18.6.1. If 'Yes' please specify: *

18.7. Specify filament type used: *

- [ ] a) Single filament technique
- [ ] b) Double filament technique
- [ ] c) Zone refined Re
- [ ] d) Not zone refined Re
- [ ] e) Tungsten
- [ ] f) Other

18.7.1. If 'other' please specify:

19. **Did you use Thermal Ionisation Mass Spectrometry (TIMS) to analyse the uranium particles?** *

- ○ a) Yes
- ○ b) No

19.1. If 'Yes', please provide the following information for each measurement:

19.2. Measurement of single/multiple particles? *

- ○ a) Single particles
- ○ b) Multiple particles

19.3. 238U average signal level *

19.4. Overall ionisation efficiency (estimate) *

20. **Did you use Laser Ablation Inductively Coupled Plasma Mass Spectrometry (LA-ICP-MS) to analyse the uranium particles?** *

- ○ a) Yes
- ○ b) No

20.1. If 'Yes', please provide the following information on the instrument parameters:

20.2. Type of carrier gas used? (He (%), Ar (%), other) *

20.3. Combination with nebulizer? *

- ◯ a) Yes
- ◯ b) No

20.3.1. If 'Yes' please specify type of nebulizer: *

[ ]

20.4. LA carrier introduced into nebulizer or additional mixing of nebulizer? *

[ ]

20.5. Gas flow after LA cell: *

[ ]

20.6. Type of laser: *

[ ]

20.7. Laser wavelength (nm): *

[ ]

20.8. Energy flux (J/cm2): *

[ ]

20.9. Spot size (micrometer): *

[ ]

20.10. Repetition rate (Hz): *

[ ]

20.11. Ablation type (single spot, line scan, other…): *

[ ]

20.12. Type of instrument: *

- ☐ a) ICP-HR-MS (single collector, high resolution)
- ☐ b) ICP-Q-MS (single collector, quadrupole)
- ☐ c) ICP-SF-MS (single coll., sector field, using low resolution (<1000))
- ☐ d) MC-ICP-MS (multi collector)

20.13. Type of detector used for each isotope (SEM, channeltron, Faraday cup): *

[ ]

21. **Are your reported uncertainties in NUSIMEP-7 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

21.1. If 'No', how was the measurement uncertainty evaluated? *

[ ]

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

- ○ a) Yes
- ○ b) No

23. **Would you be interested in participating in future NUSIMEP Inter-laboratory comparisons on particle analysis?** *

- ○ a) Yes
- ○ b) No

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

[ ]

23.2. If 'Yes', what should be the particle density on the sample? *

[ ]

23.3. If 'Yes', Which isotopic composition (enrichment)? *

[ ]

23.4. If 'Yes', would it be of interest to you to analyse uranium oxyfluoride particles? *

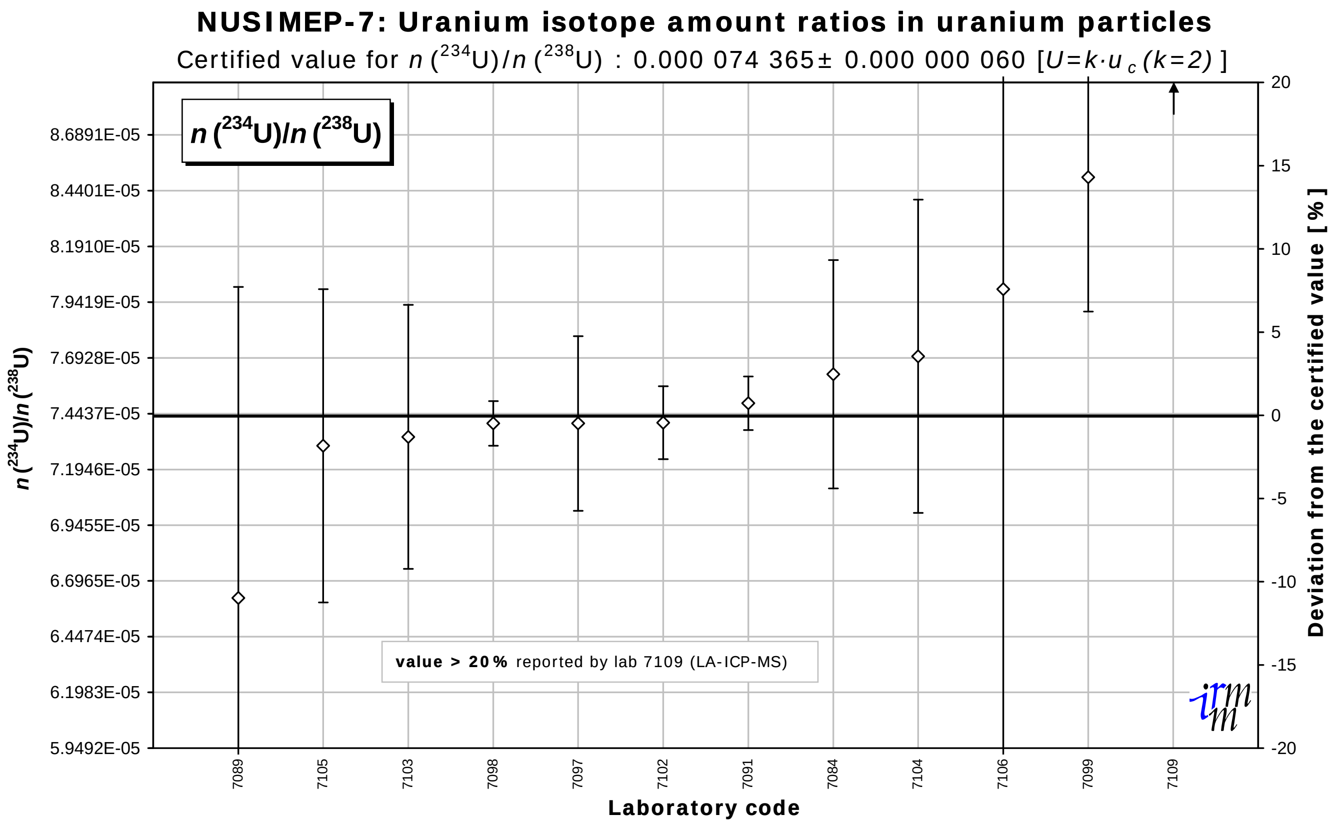
[ ]

24. **Do you have any other feedback/comments on NUSIMEP-7?**

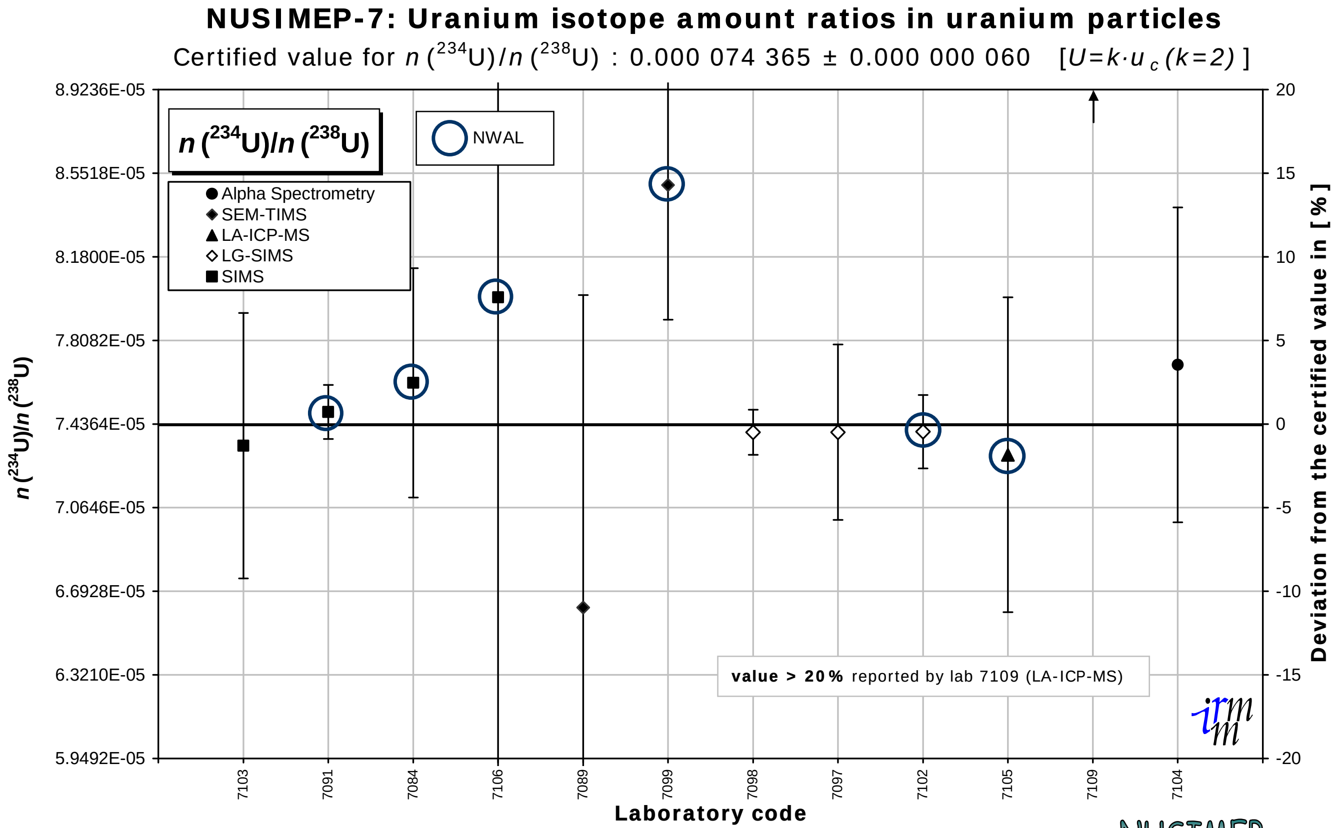
[ ]

Annex 5: Results for *n*(^{234}U)/*n*(^{238}U) of the sample with a single isotopic deposition

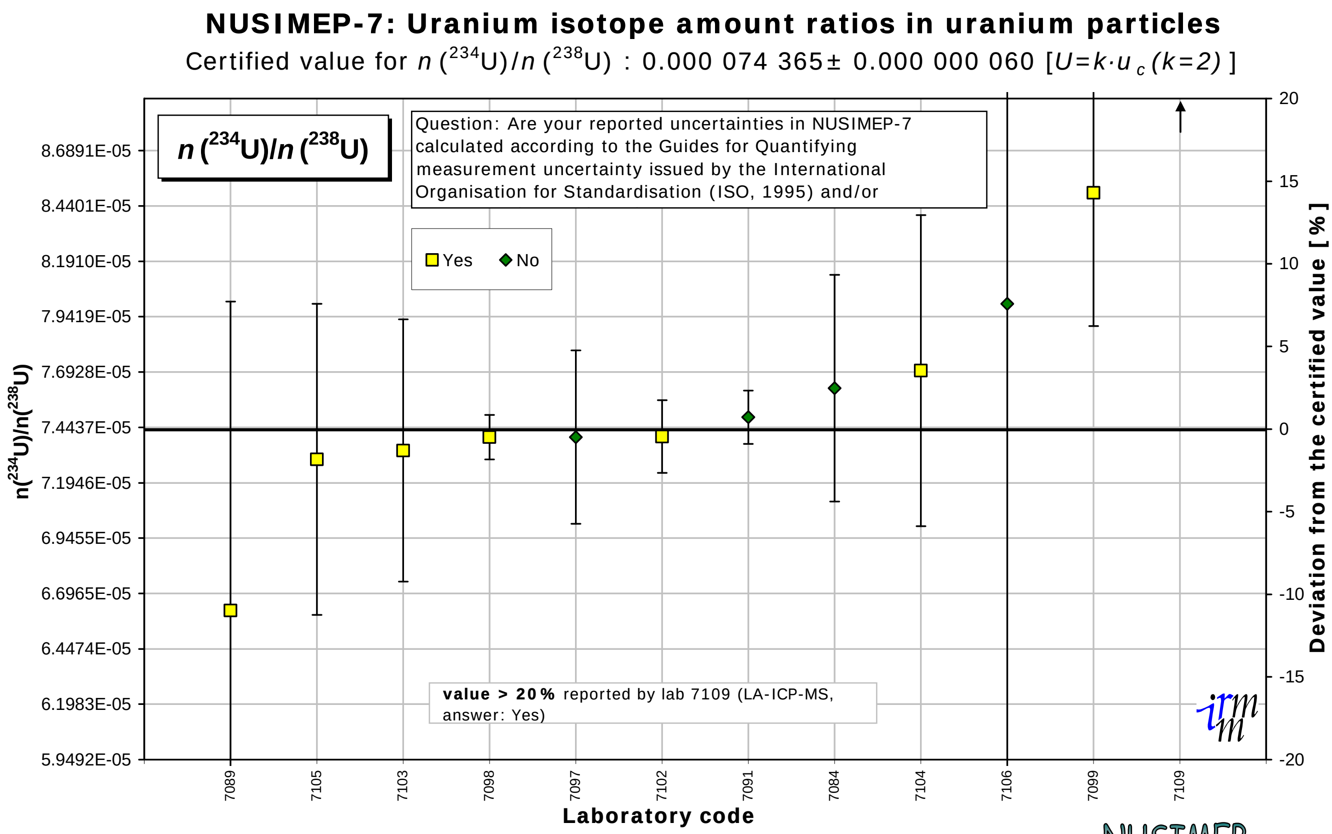
Laboratory	Analytical method	Reported *n*(^{234}U)/*n*(^{238}U)	Reported uncertainty *n*(^{234}U)/*n*(^{238}U)	Coverage factor *k*	z score	zeta score
7084	SIMS	0.0000762	0.0000051	1	0.99	0.36
7089	SEM-TIMS	0.0000662	0.0000139	1	4.39	0.59
7090						
7091	SIMS	0.0000749	0.0000012	1	0.29	0.45
7092						
7093						
7097	LG-SIMS	0.000074	0.0000039	2	0.20	0.19
7098	LG-SIMS	0.000074	0.000001	2	0.20	0.73
7099	SEM-TIMS	0.000085	0.000006	2	5.72	3.54
7101						
7102	LG-SIMS	0.00007403	0.00000163	1	0.18	0.21
7103	SIMS	0.0000734	0.0000059	1.96	0.52	0.32
7104	Alpha spectrometry	0.000077	0.000007	1	1.42	0.38
7105	LA-ICP-MS	0.000073	0.000007	2	0.73	0.39
7106	SIMS	0.00008	0.00003	1	3.03	0.19
7107						
7109	LA-ICP-MS	0.000354	0.000149	2	150.41	3.75



This graph displays all measurement results and their associated uncertainties.
These uncertainties are shown as reported, with various coverage factors and levels of confidence.
The grey band represents the reference interval (X_{ref} ± $2u_{ref}$).



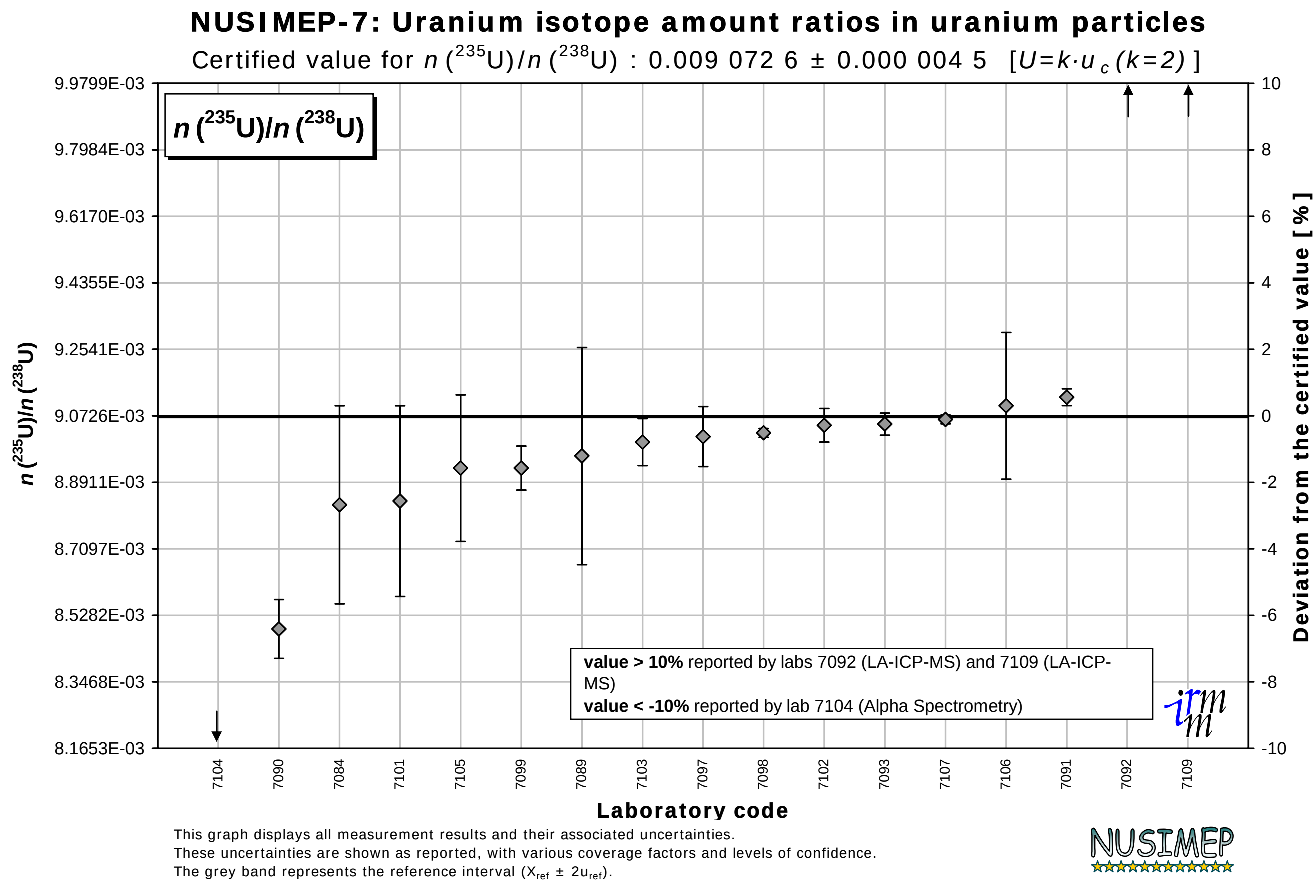
This graph displays all measurement results and their associated uncertainties.
These uncertainties are shown as reported, with various coverage factors and levels of confidence.
The grey band represents the reference interval (X_{ref} ± $2u_{ref}$).



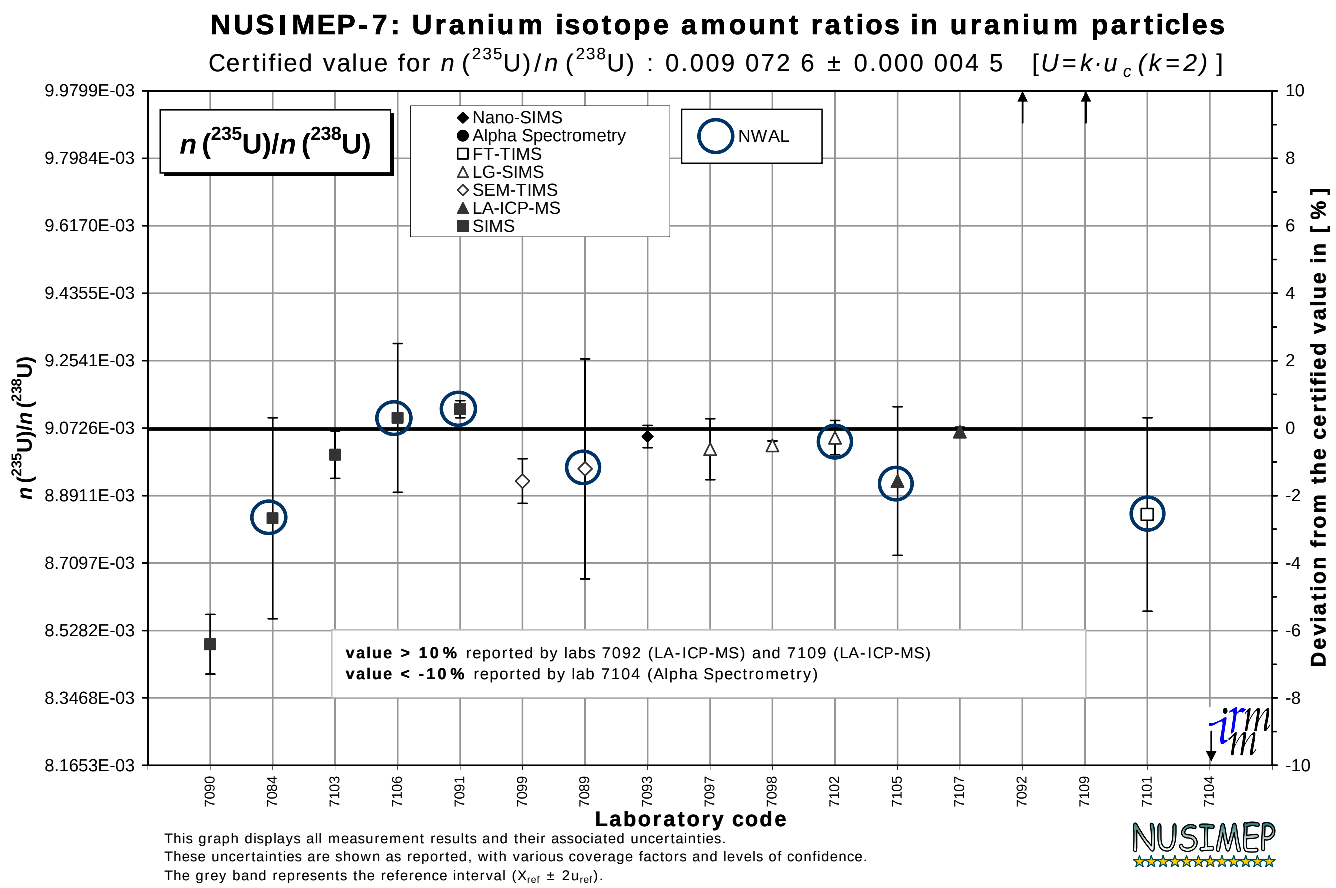
This graph displays all measurement results and their associated uncertainties.
These uncertainties are shown as reported, with various coverage factors and levels of confidence.
The grey band represents the reference interval (X_{ref} ± $2u_{ref}$).

Annex 6: Results for $n(^{235}U)/n(^{238}U)$ of the sample with a single isotopic deposition

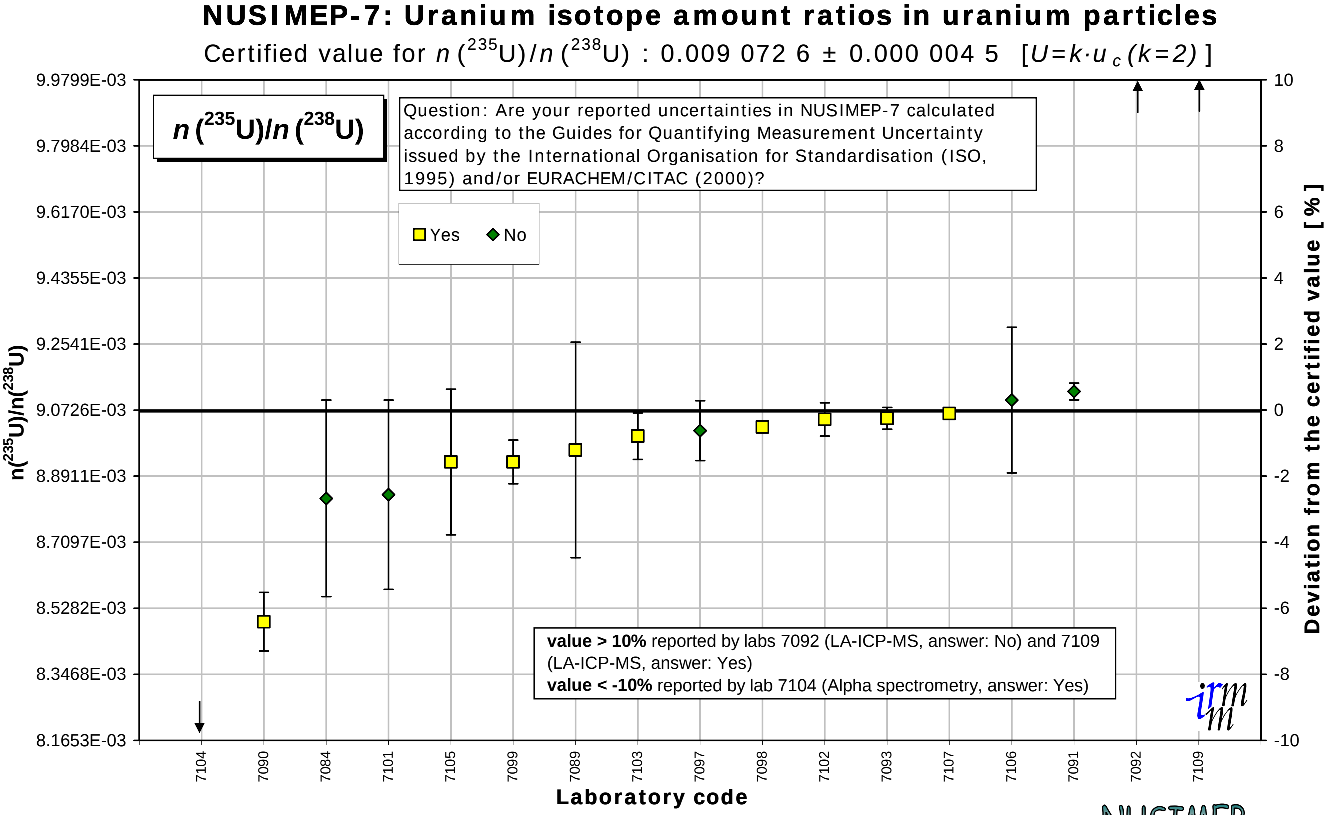
Laboratory	Analytical method	Reported $n(^{235}U)/n(^{238}U)$	Reported uncertainty $n(^{235}U)/n(^{238}U)$	Coverage factor k	z score	zeta score
7084	SIMS	0.00883	0.00027	1	5.35	0.90
7089	SEM-TIMS	0.008963	0.000296	1	2.42	0.37
7090	SIMS	0.008491	0.0000803	2	12.82	14.46
7091	SIMS	0.0091234	0.000023	1	1.12	2.20
7092	LA-ICP-MS	0.0129	0.0028	2	84.37	2.73
7093	NanoSIMS	0.00905	0.00003	2	0.50	1.49
7097	LG-SIMS	0.009016	0.000082	2	1.25	1.38
7098	LG-SIMS	0.009026	0.000012	2	1.03	7.27
7099	SEM-TIMS	0.00893	0.00006	2	3.14	4.74
7101	FT-TIMS	0.00884	0.00026	1	5.13	0.89
7102	LG-SIMS	0.00904675	0.00004594	1	0.57	0.56
7103	SIMS	0.009001	0.000064	1.96	1.58	2.19
7104	Alpha spectrometry	0.003	0.001	1	133.87	6.07
7105	LA-ICP-MS	0.00893	0.0002	2	3.14	1.43
7106	SIMS	0.0091	0.0002	1	0.60	0.14
7107	LA-ICP-MS	0.009063	0.000012	1	0.21	0.79
7109	LA-ICP-MS	0.0331	0.00292	2	529.67	16.46



NUSIMEP-7: Uranium isotope amount ratios in uranium particles
Certified value for n (235U)/n (238U) : 0.009 072 6 ± 0.000 004 5 [U=k·u c (k=2)]
n (235U)/n (238U)
value > 10% reported by labs 7092 (LA-ICP-MS) and 7109 (LA-ICP-MS)
value < -10% reported by lab 7104 (Alpha Spectrometry)
Laboratory code
Deviation from the certified value [%]
This graph displays all measurement results and their associated uncertainties.
These uncertainties are shown as reported, with various coverage factors and levels of confidence.
The grey band represents the reference interval (Xref ± 2uref).
NUSIMEP



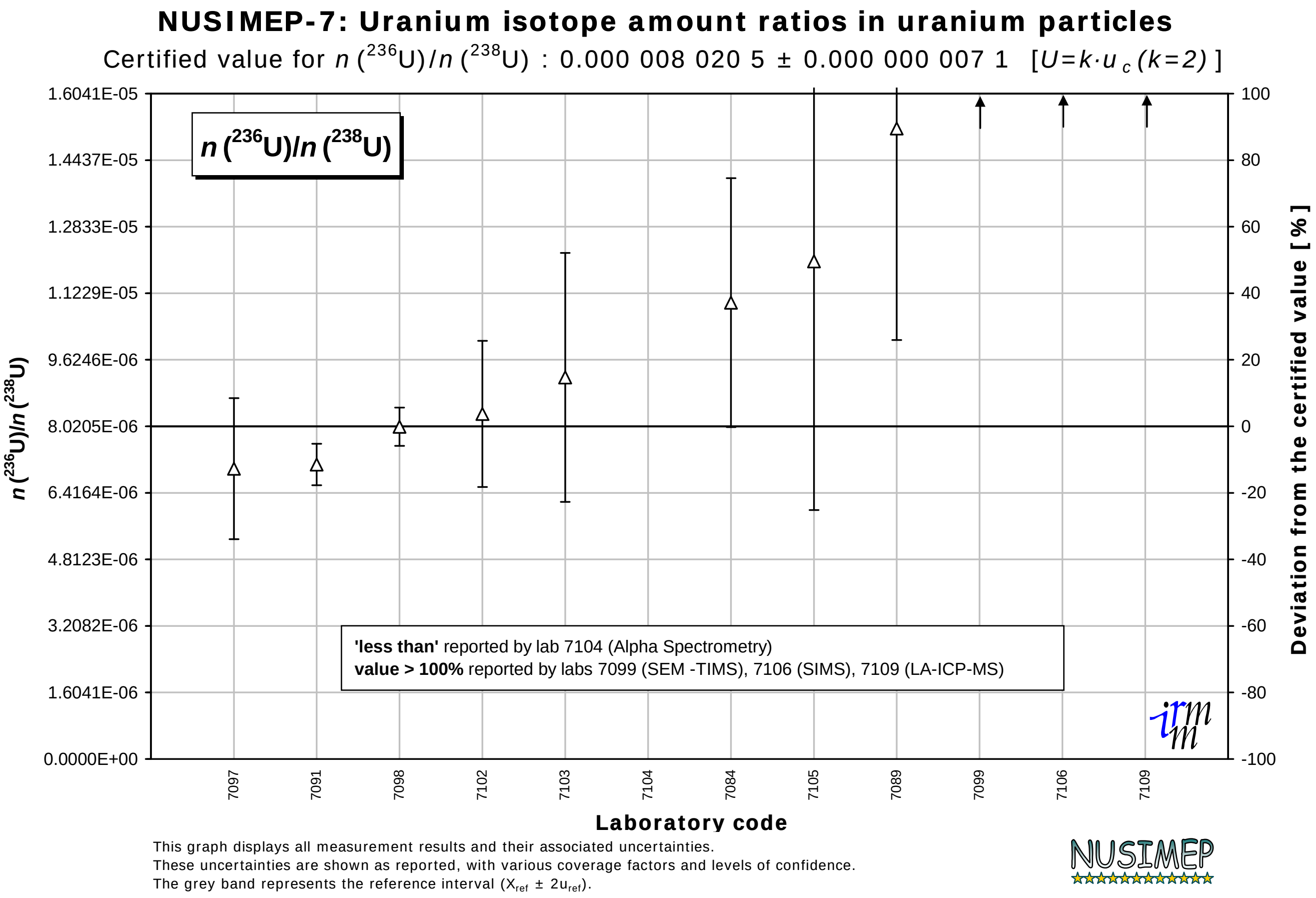
NUSIMEP-7: Uranium isotope amount ratios in uranium particles
Certified value for n (235U)/n (238U) : 0.009 072 6 ± 0.000 004 5 [U=k·u c (k=2)]
n (235U)/n (238U)
Nano-SIMS
Alpha Spectrometry
FT-TIMS
LG-SIMS
SEM-TIMS
LA-ICP-MS
SIMS
NWAL
value > 10% reported by labs 7092 (LA-ICP-MS) and 7109 (LA-ICP-MS)
value < -10% reported by lab 7104 (Alpha Spectrometry)
Laboratory code
Deviation from the certified value in [%]
This graph displays all measurement results and their associated uncertainties.
These uncertainties are shown as reported, with various coverage factors and levels of confidence.
The grey band represents the reference interval (Xref ± 2uref).
NUSIMEP



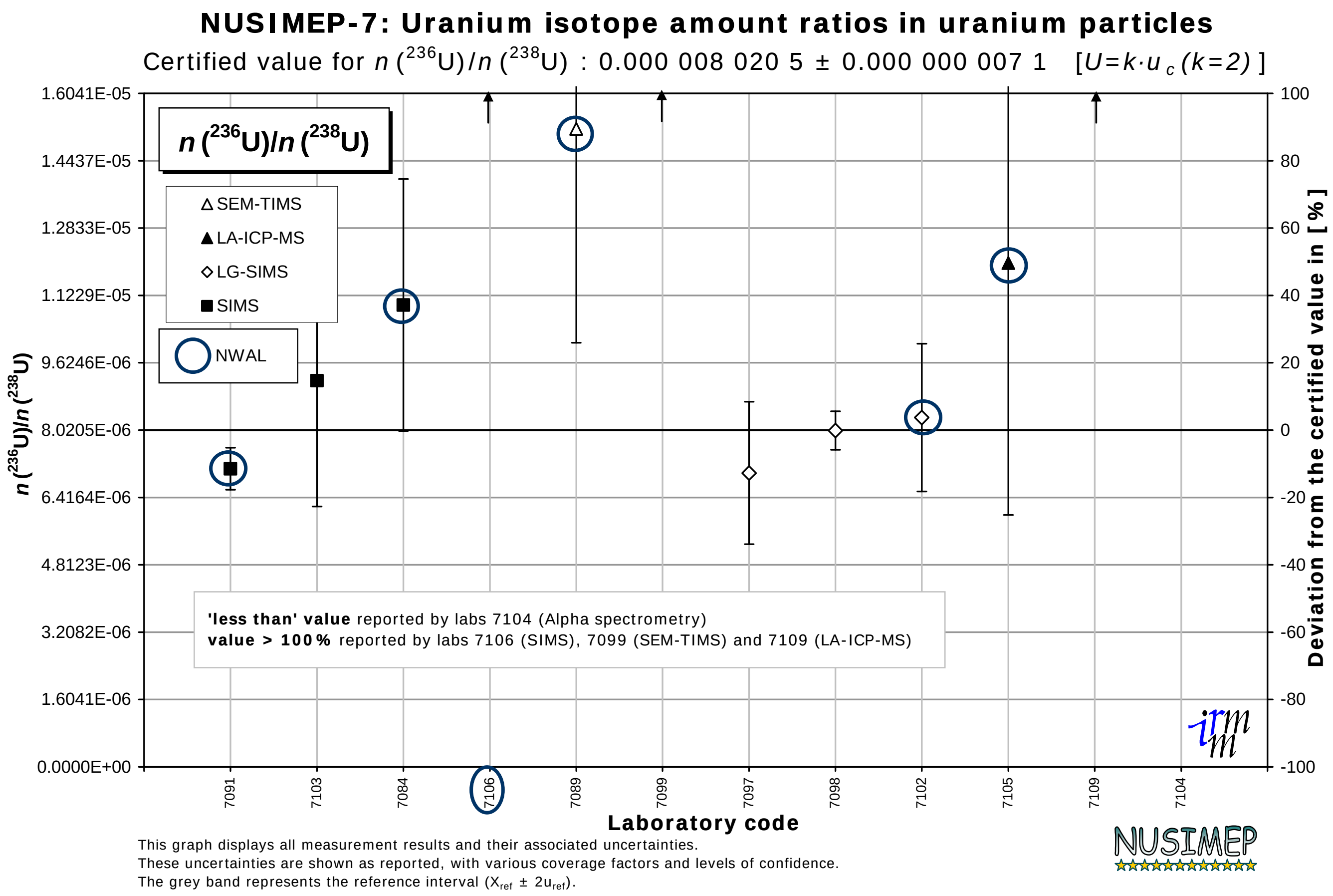
This graph displays all measurement results and their associated uncertainties.
These uncertainties are shown as reported, with various coverage factors and levels of confidence.
The grey band represents the reference interval ($X_{ref} \pm 2u_{ref}$).

Annex 7: Results for $n(^{236}U)/n(^{238}U)$ of the sample with a single isotopic deposition

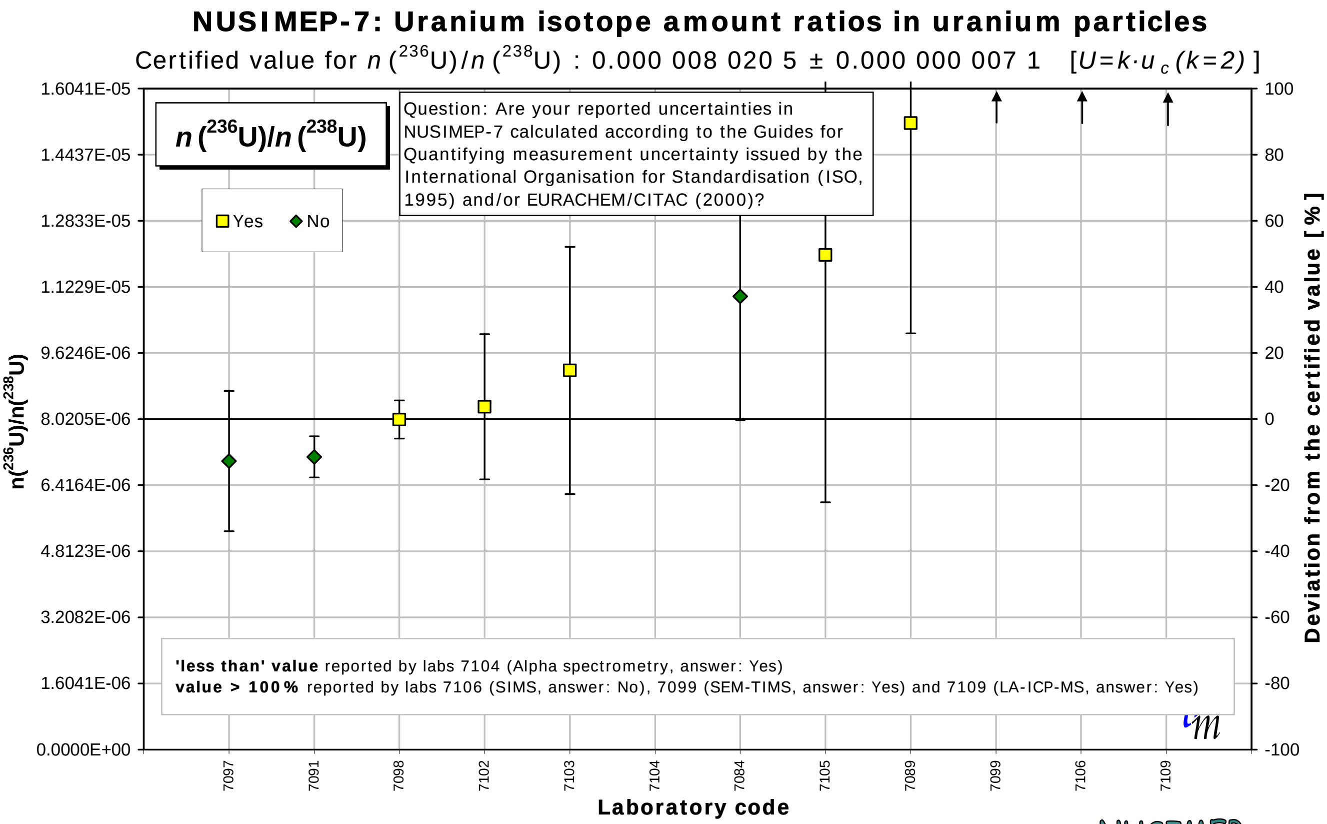
Laboratory	Analytical method	Reported $n(^{236}U)/n(^{238}U)$	Reported uncertainty $n(^{236}U)/n(^{238}U)$	Coverage factor *k*	z score	zeta score
7084	SIMS	0.000011	0.000003	1	0.74	0.99
7089	SEM-TIMS	0.0000152	0.0000051	1	1.79	1.41
7090						
7091	SIMS	0.0000071	0.0000005	1	0.23	1.84
7092						
7093						
7097	LG-SIMS	0.000007	0.0000017	2	0.25	1.20
7098	LG-SIMS	0.00000801	0.00000046	2	0.003	0.05
7099	SEM-TIMS	0.000036	0.000004	2	6.98	13.99
7101						
7102	LG-SIMS	0.00000832	0.00000176	1	0.07	0.17
7103	SIMS	0.0000092	0.000003	1.96	0.29	0.77
7104	Alpha spectrometry	<1E-5				
7105	LA-ICP-MS	0.000012	0.000006	2	0.99	1.33
7106	SIMS	0.00007	0.00002	1	15.46	3.10
7107						
7109	LA-ICP-MS	0.0000959	0.0000521	2	21.91	3.37



NUSIMEP-7: Uranium isotope amount ratios in uranium particles
Certified value for n (236U)/n (238U) : 0.000 008 020 5 ± 0.000 000 007 1 [U=k·u c (k=2)]
n (236U)/n (238U)
'less than' reported by lab 7104 (Alpha Spectrometry)
value > 100% reported by labs 7099 (SEM -TIMS), 7106 (SIMS), 7109 (LA-ICP-MS)
Laboratory code
Deviation from the certified value [%]
This graph displays all measurement results and their associated uncertainties.
These uncertainties are shown as reported, with various coverage factors and levels of confidence.
The grey band represents the reference interval (Xref ± 2uref).



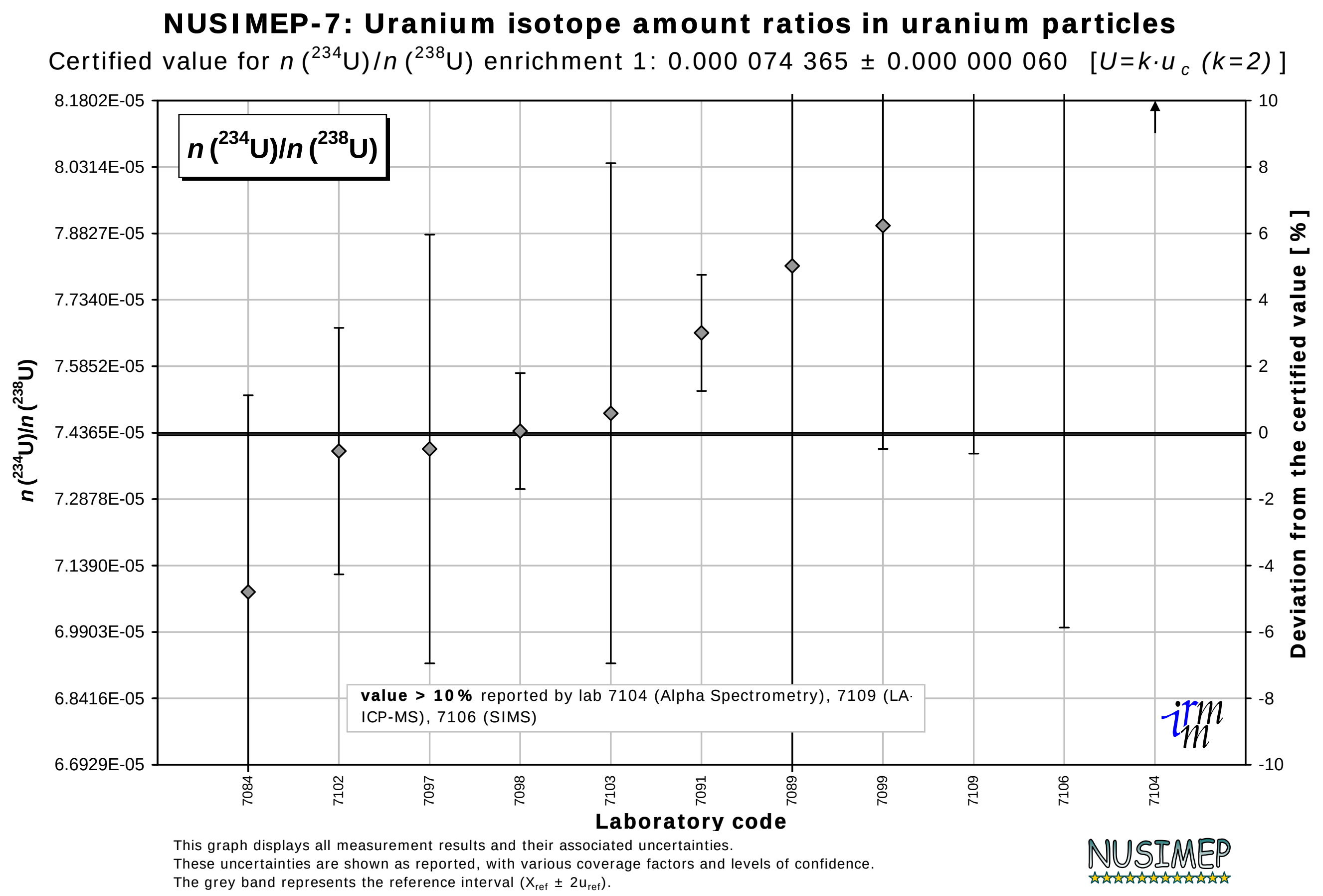
NUSIMEP-7: Uranium isotope amount ratios in uranium particles
Certified value for n (236U)/n (238U) : 0.000 008 020 5 ± 0.000 000 007 1 [U=k·u c (k=2)]
n (236U)/n (238U)
SEM-TIMS
LA-ICP-MS
LG-SIMS
SIMS
NWAL
'less than' value reported by labs 7104 (Alpha spectrometry)
value > 100% reported by labs 7106 (SIMS), 7099 (SEM-TIMS) and 7109 (LA-ICP-MS)
Laboratory code
Deviation from the certified value in [%]
This graph displays all measurement results and their associated uncertainties.
These uncertainties are shown as reported, with various coverage factors and levels of confidence.
The grey band represents the reference interval (Xref ± 2uref).



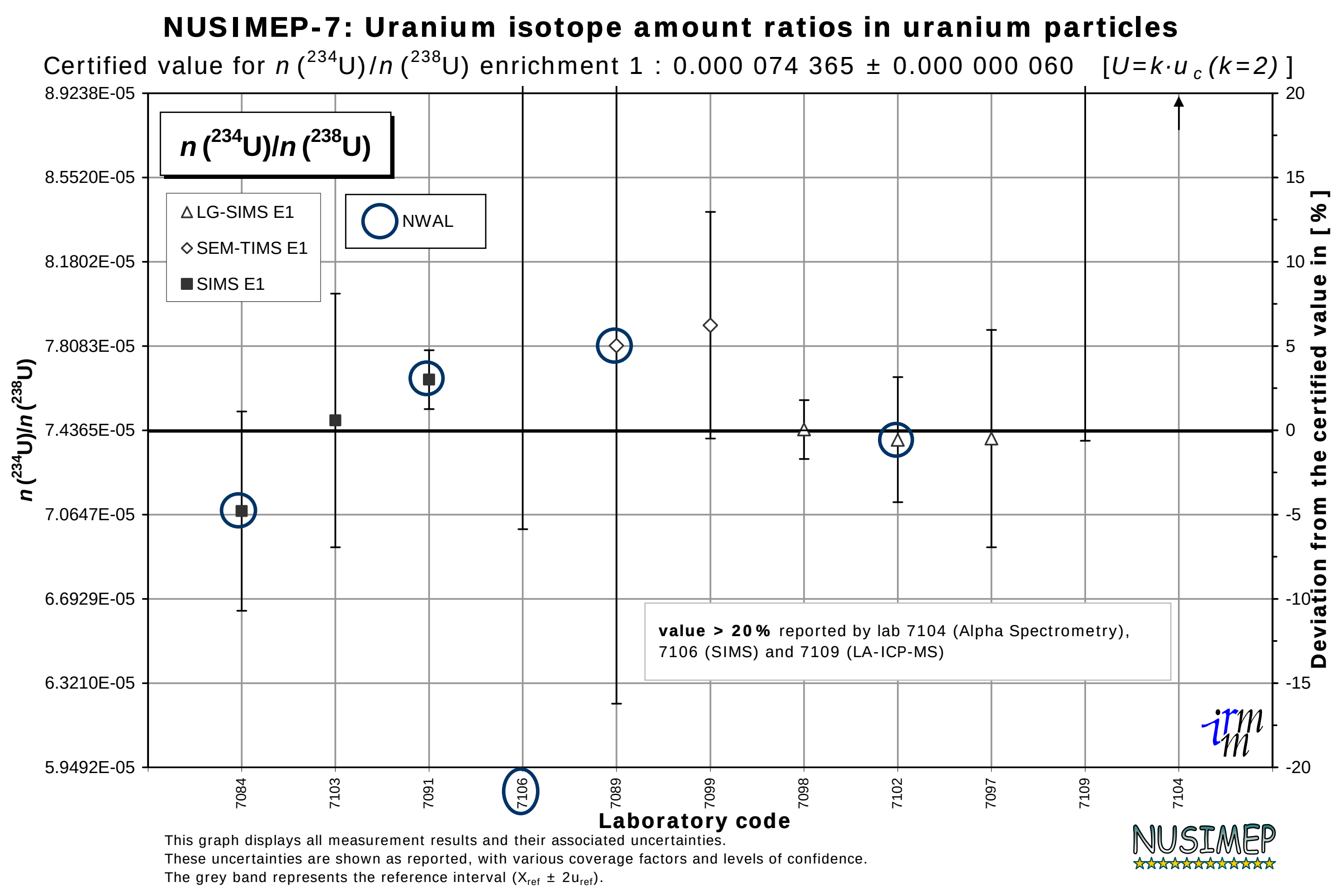
This graph displays all measurement results and their associated uncertainties.
These uncertainties are shown as reported, with various coverage factors and levels of confidence.
The grey band represents the reference interval ($X_{ref} \pm 2u_{ref}$).

Annex 8: Results for $n(^{234}U)/n(^{238}U)$ of the first enrichment of the sample with a double isotopic deposition

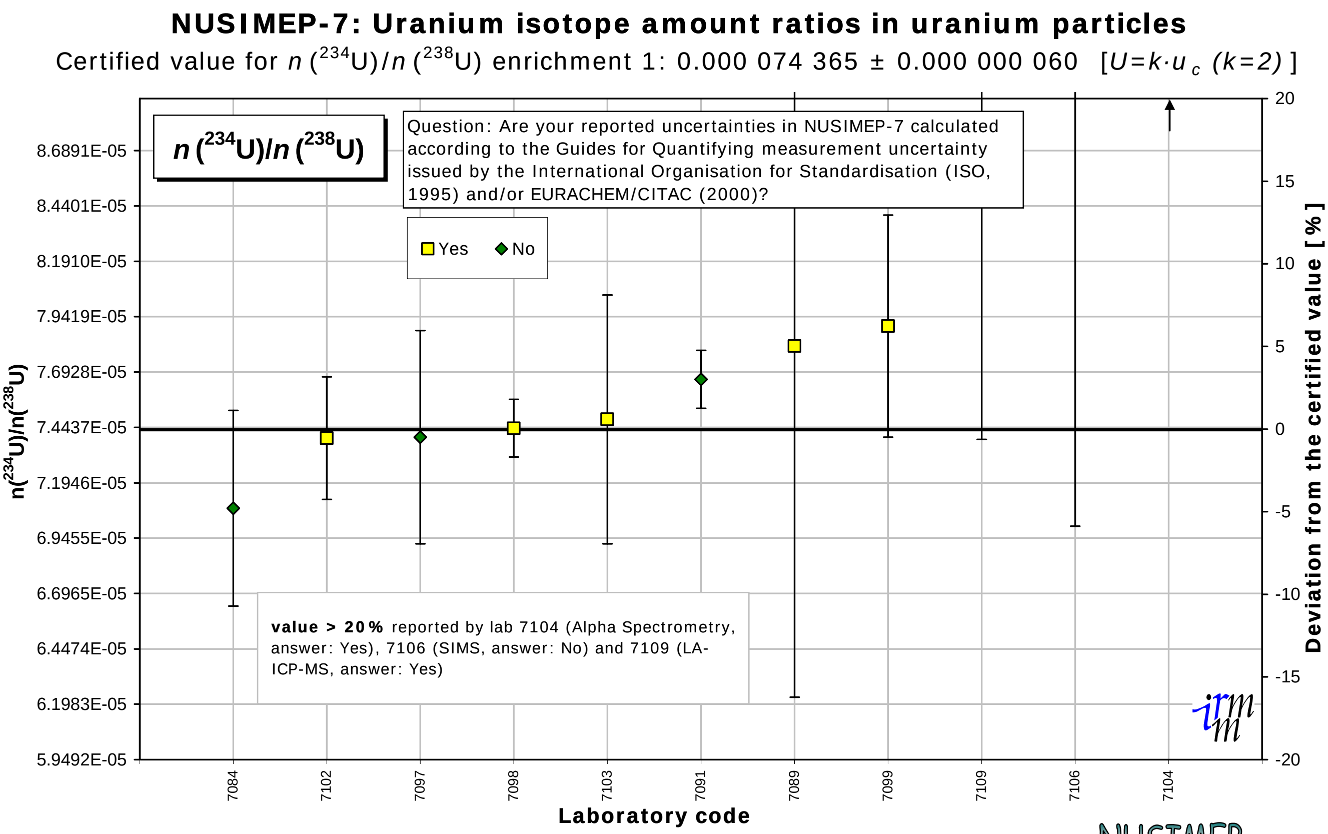
Laboratory	Analytical method	Reported $n(^{234}U)/n(^{238}U)$	Reported uncertainty $n(^{234}U)/n(^{238}U)$	Coverage factor *k*	z score	zeta score
7084	SIMS	0.0000708	0.0000044	1	1.92	0.81
7089	SEM-TIMS	0.0000781	0.0000158	1	2.01	0.24
7090						
7091	SIMS	0.0000766	0.0000013	1	1.20	1.72
7092						
7093						
7097	LG-SIMS	0.000074	0.0000048	2	0.20	0.15
7098	LG-SIMS	0.0000744	0.0000013	2	0.02	0.05
7099	SEM-TIMS	0.000079	0.000005	2	2.49	1.85
7101						
7102	LG-SIMS	0.00007395	0.00000276	1	0.22	0.15
7103	SIMS	0.0000748	0.0000056	1.96	0.23	0.15
7104	Alpha spectrometry	0.00024	0.00001	1	89.09	16.56
7105						
7106	SIMS	0.0001	0.00003	1	13.79	0.85
7107						
7109	LA-ICP-MS	0.000096	0.0000221	2	11.64	1.96



NUSIMEP-7: Uranium isotope amount ratios in uranium particles
Certified value for n (234U)/n (238U) enrichment 1: 0.000 074 365 ± 0.000 000 060 [U=k·u c (k=2)]
n (234U)/n (238U)
value > 10 % reported by lab 7104 (Alpha Spectrometry), 7109 (LA-ICP-MS), 7106 (SIMS)
8.1802E-05
8.0314E-05
7.8827E-05
7.7340E-05
7.5852E-05
7.4365E-05
7.2878E-05
7.1390E-05
6.9903E-05
6.8416E-05
6.6929E-05
7084
7102
7097
7098
7103
7091
7089
7099
7109
7106
7104
Laboratory code
Deviation from the certified value [%]
This graph displays all measurement results and their associated uncertainties.
These uncertainties are shown as reported, with various coverage factors and levels of confidence.
The grey band represents the reference interval (Xref ± 2uref).
NUSIMEP



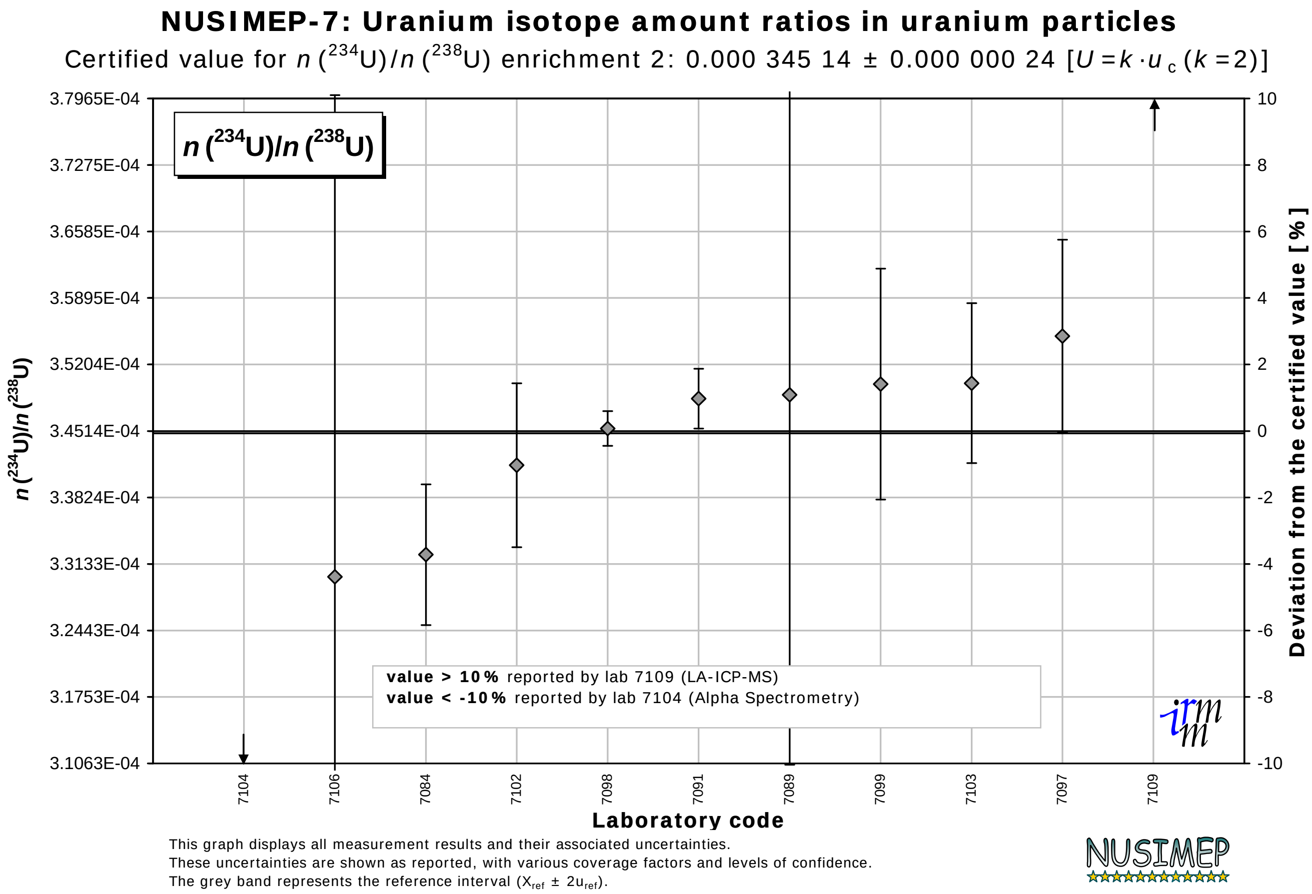
NUSIMEP-7: Uranium isotope amount ratios in uranium particles
Certified value for n (234U)/n (238U) enrichment 1 : 0.000 074 365 ± 0.000 000 060 [U=k·u c (k=2)]
n (234U)/n (238U)
LG-SIMS E1
SEM-TIMS E1
SIMS E1
NWAL
value > 20 % reported by lab 7104 (Alpha Spectrometry), 7106 (SIMS) and 7109 (LA-ICP-MS)
8.9238E-05
8.5520E-05
8.1802E-05
7.8083E-05
7.4365E-05
7.0647E-05
6.6929E-05
6.3210E-05
5.9492E-05
7084
7103
7091
7106
7089
7099
7098
7102
7097
7109
7104
Laboratory code
Deviation from the certified value in [%]
This graph displays all measurement results and their associated uncertainties.
These uncertainties are shown as reported, with various coverage factors and levels of confidence.
The grey band represents the reference interval (Xref ± 2uref).
NUSIMEP

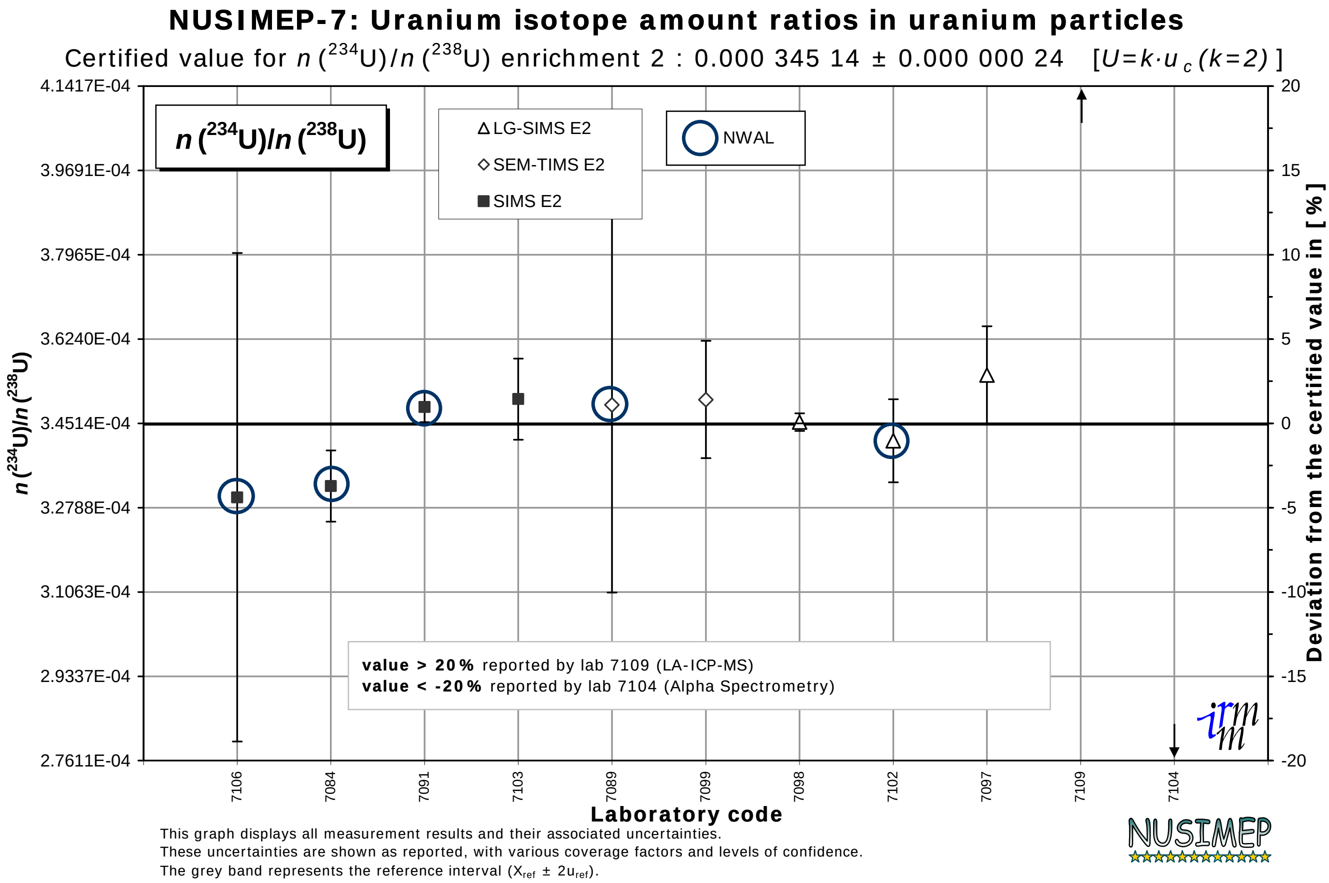


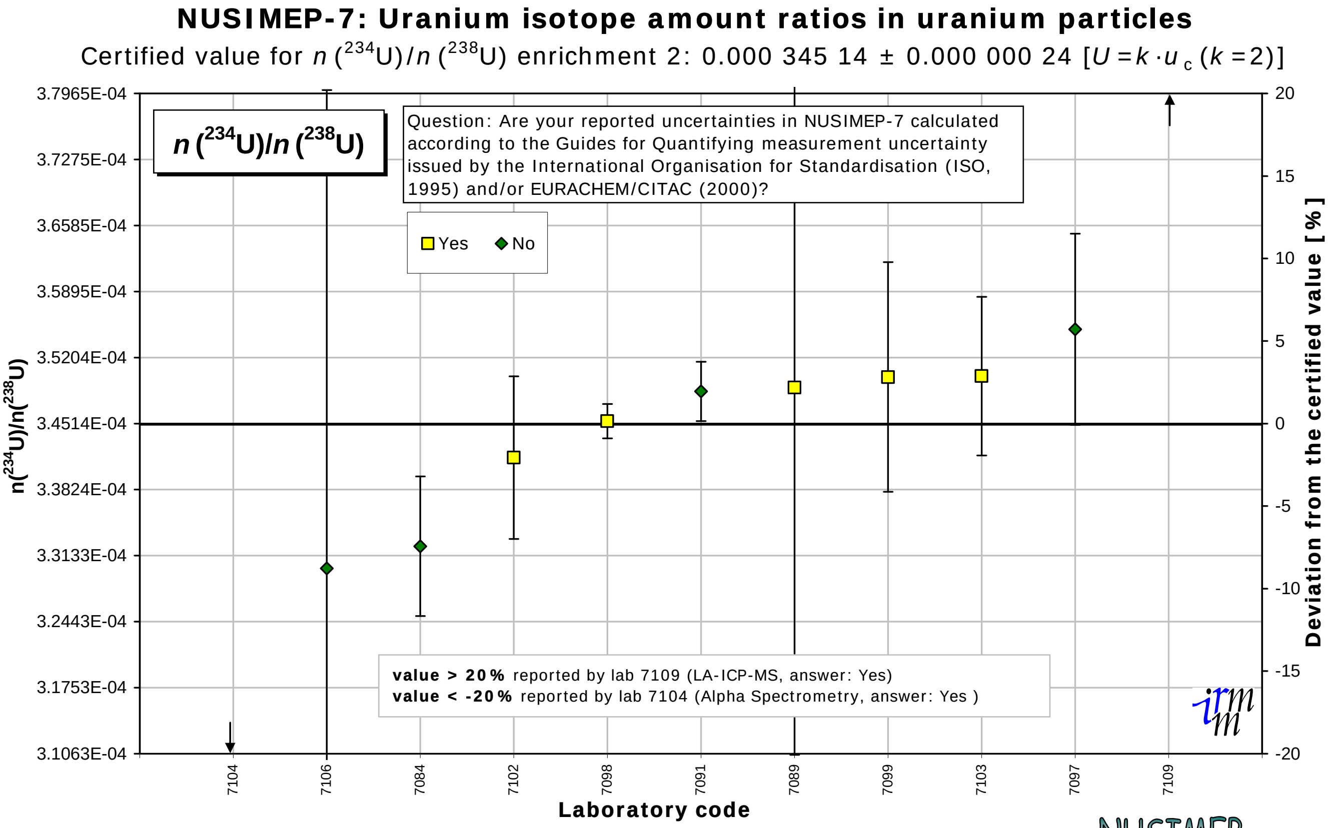
This graph displays all measurement results and their associated uncertainties.
These uncertainties are shown as reported, with various coverage factors and levels of confidence.
The grey band represents the reference interval ($X_{ref} \pm 2u_{ref}$).

Annex 9: Results for $n(^{234}U)/n(^{238}U)$ of the second enrichment of the sample with a double isotopic deposition

Laboratory	Analytical method	Reported $n(^{234}U)/n(^{238}U)$	Reported uncertainty $n(^{234}U)/n(^{238}U)$	Coverage factor *k*	z score	zeta score
7084	SIMS	0.0003323	0.0000073	1	1.49	1.76
7089	SEM-TIMS	0.0003489	0.0000384	1	0.44	0.10
7090						
7091	SIMS	0.0003485	0.0000031	1	0.39	1.08
7092						
7093						
7097	LG-SIMS	0.000355	0.00001	2	1.14	1.97
7098	LG-SIMS	0.0003454	0.0000018	2	0.03	0.29
7099	SEM-TIMS	0.00035	0.000012	2	0.56	0.81
7101						
7102	LG-SIMS	0.00034158	0.00000851	1	0.41	0.42
7103	SIMS	0.0003501	0.0000083	1.96	0.57	1.17
7104	Alpha spectrometry	0.00024	0.00001	1	12.19	10.51
7105						
7106	SIMS	0.00033	0.00005	1	1.75	0.30
7107						
7109	LA-ICP-MS	0.00315	0.000551	2	325.07	10.18


NUSIMEP-7: Uranium isotope amount ratios in uranium particles
Certified value for n (234U)/n (238U) enrichment 2: 0.000 345 14 ± 0.000 000 24 [U = k · u c (k = 2)]
n (234U)/n (238U)
3.7965E-04
3.7275E-04
3.6585E-04
3.5895E-04
3.5204E-04
3.4514E-04
3.3824E-04
3.3133E-04
3.2443E-04
3.1753E-04
3.1063E-04
10
8
6
4
2
0
-2
-4
-6
-8
-10
Deviation from the certified value [%]
value > 10 % reported by lab 7109 (LA-ICP-MS)
value < -10 % reported by lab 7104 (Alpha Spectrometry)
7104
7106
7084
7102
7098
7091
7089
7099
7103
7097
7109
Laboratory code
This graph displays all measurement results and their associated uncertainties.
These uncertainties are shown as reported, with various coverage factors and levels of confidence.
The grey band represents the reference interval (Xref ± 2uref).
NUSIMEP

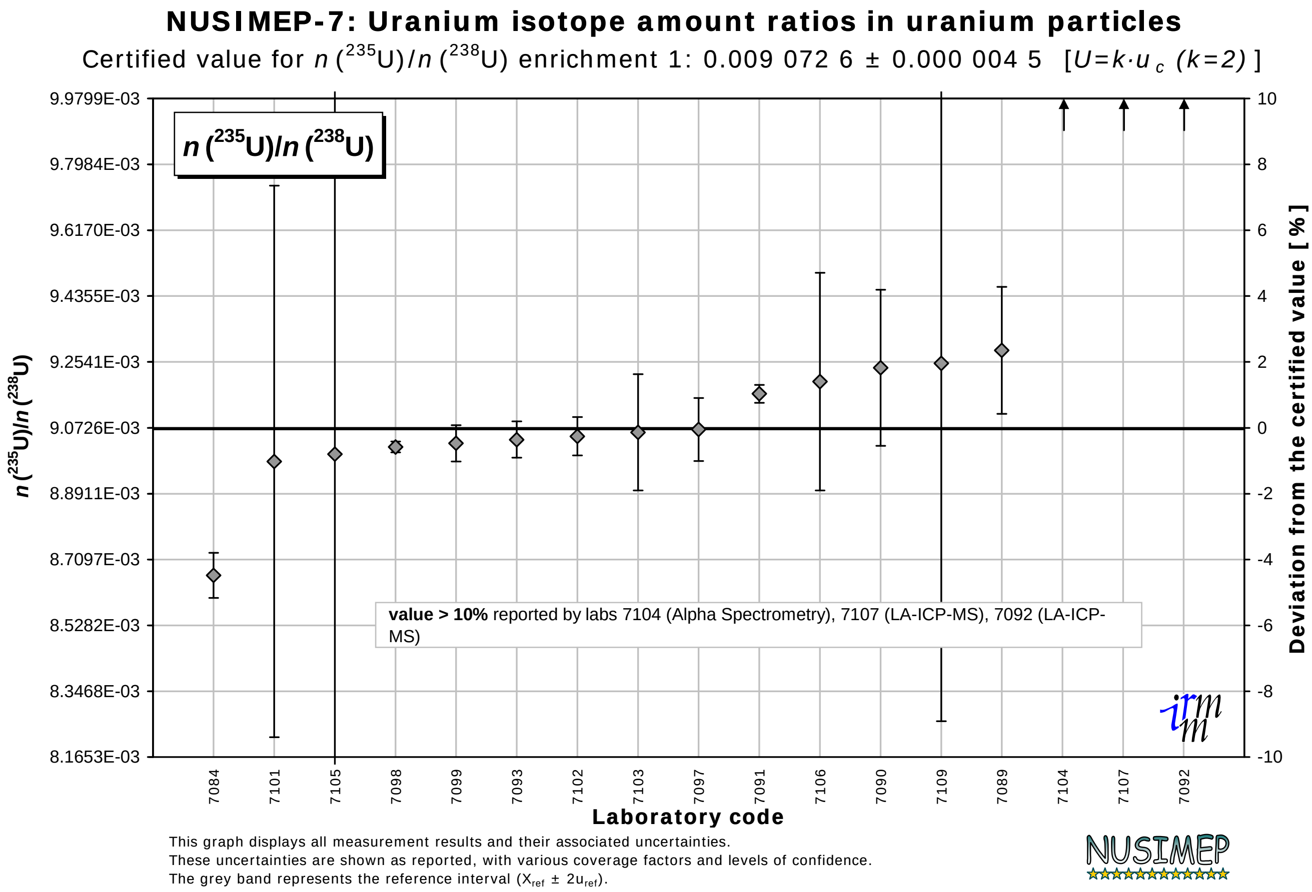

NUSIMEP-7: Uranium isotope amount ratios in uranium particles
Certified value for n (234U)/n (238U) enrichment 2 : 0.000 345 14 ± 0.000 000 24 [U=k·u c (k=2)]
n (234U)/n (238U)
LG-SIMS E2
SEM-TIMS E2
SIMS E2
NWAL
4.1417E-04
3.9691E-04
3.7965E-04
3.6240E-04
3.4514E-04
3.2788E-04
3.1063E-04
2.9337E-04
2.7611E-04
20
15
10
5
0
-5
-10
-15
-20
Deviation from the certified value in [%]
value > 20 % reported by lab 7109 (LA-ICP-MS)
value < -20 % reported by lab 7104 (Alpha Spectrometry)
7106
7084
7091
7103
7089
7099
7098
7102
7097
7109
7104
Laboratory code
This graph displays all measurement results and their associated uncertainties.
These uncertainties are shown as reported, with various coverage factors and levels of confidence.
The grey band represents the reference interval (Xref ± 2uref).
NUSIMEP

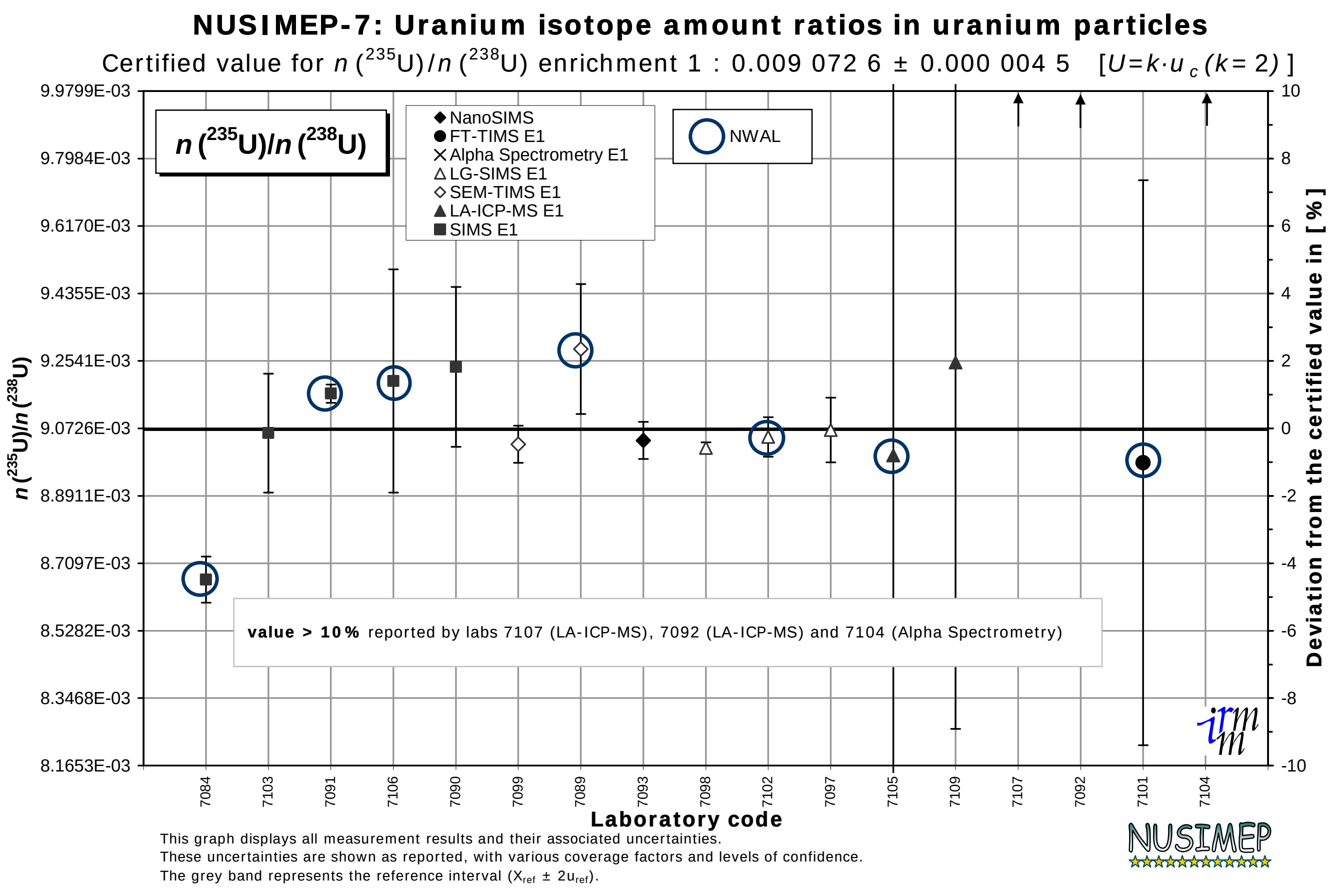


This graph displays all measurement results and their associated uncertainties.
These uncertainties are shown as reported, with various coverage factors and levels of confidence.
The grey band represents the reference interval (X_{ref} ± $2u_{ref}$).

Annex 10: Results for $n(^{235}U)/n(^{238}U)$ of the first enrichment of the sample with a double isotopic deposition

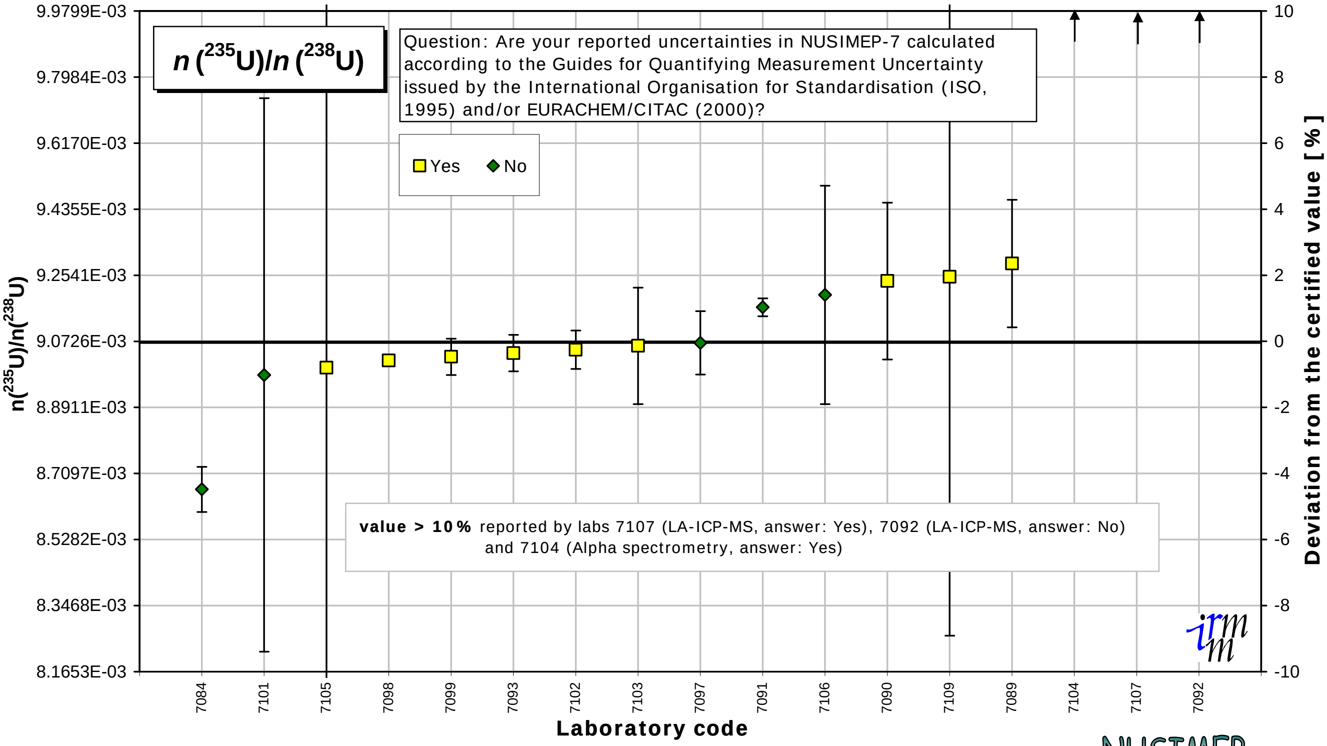
Laboratory	Analytical method	Reported $n(^{235}U)/n(^{238}U)$	Reported uncertainty $n(^{235}U)/n(^{238}U)$	Coverage factor k	z score	zeta score
7084	SIMS	0.008666	0.000062	1	8.96	6.55
7089	SEM-TIMS	0.009286	0.000175	1	4.70	1.22
7090	SIMS	0.009238	0.000215	2	3.65	1.54
7091	SIMS	0.0091662	0.0000245	1	2.06	3.80
7092	LA-ICP-MS	0.0534	0.0085	2	977.17	10.43
7093	NanoSIMS	0.00904	0.00005	2	0.72	1.30
7097	LG-SIMS	0.009068	0.000087	2	0.10	0.11
7098	LG-SIMS	0.00902	0.000015	2	1.16	6.72
7099	SEM-TIMS	0.00903	0.00005	2	0.94	1.70
7101	FT-TIMS	0.00898	0.00076	1	2.04	0.12
7102	LG-SIMS	0.00904939	0.00005285	1	0.51	0.44
7103	SIMS	0.00906	0.00016	1.96	0.28	0.15
7104	Alpha spectrometry	0.021	0.003	1	262.93	3.98
7105	LA-ICP-MS	0.009	0.005	2	1.60	0.03
7106	SIMS	0.0092	0.0003	1	2.81	0.42
7107	LA-ICP-MS	0.0286	0.0022	1	430.47	8.88
7109	LA-ICP-MS	0.00925	0.000986	2	3.91	0.36


NUSIMEP-7: Uranium isotope amount ratios in uranium particles
Certified value for n (235U)/n (238U) enrichment 1: 0.009 072 6 ± 0.000 004 5 [U=k·u c (k=2)]
n (235U)/n (238U)
value > 10% reported by labs 7104 (Alpha Spectrometry), 7107 (LA-ICP-MS), 7092 (LA-ICP-MS)
Laboratory code
Deviation from the certified value [%]
This graph displays all measurement results and their associated uncertainties.
These uncertainties are shown as reported, with various coverage factors and levels of confidence.
The grey band represents the reference interval (Xref ± 2uref).


NUSIMEP-7: Uranium isotope amount ratios in uranium particles
Certified value for n (235U)/n (238U) enrichment 1 : 0.009 072 6 ± 0.000 004 5 [U=k·u c (k= 2)]
n (235U)/n (238U)
NanoSIMS
FT-TIMS E1
Alpha Spectrometry E1
LG-SIMS E1
SEM-TIMS E1
LA-ICP-MS E1
SIMS E1
NWAL
value > 10% reported by labs 7107 (LA-ICP-MS), 7092 (LA-ICP-MS) and 7104 (Alpha Spectrometry)
Laboratory code
Deviation from the certified value in [%]
This graph displays all measurement results and their associated uncertainties.
These uncertainties are shown as reported, with various coverage factors and levels of confidence.
The grey band represents the reference interval (Xref ± 2uref).

NUSIMEP-7: Uranium isotope amount ratios in uranium particles

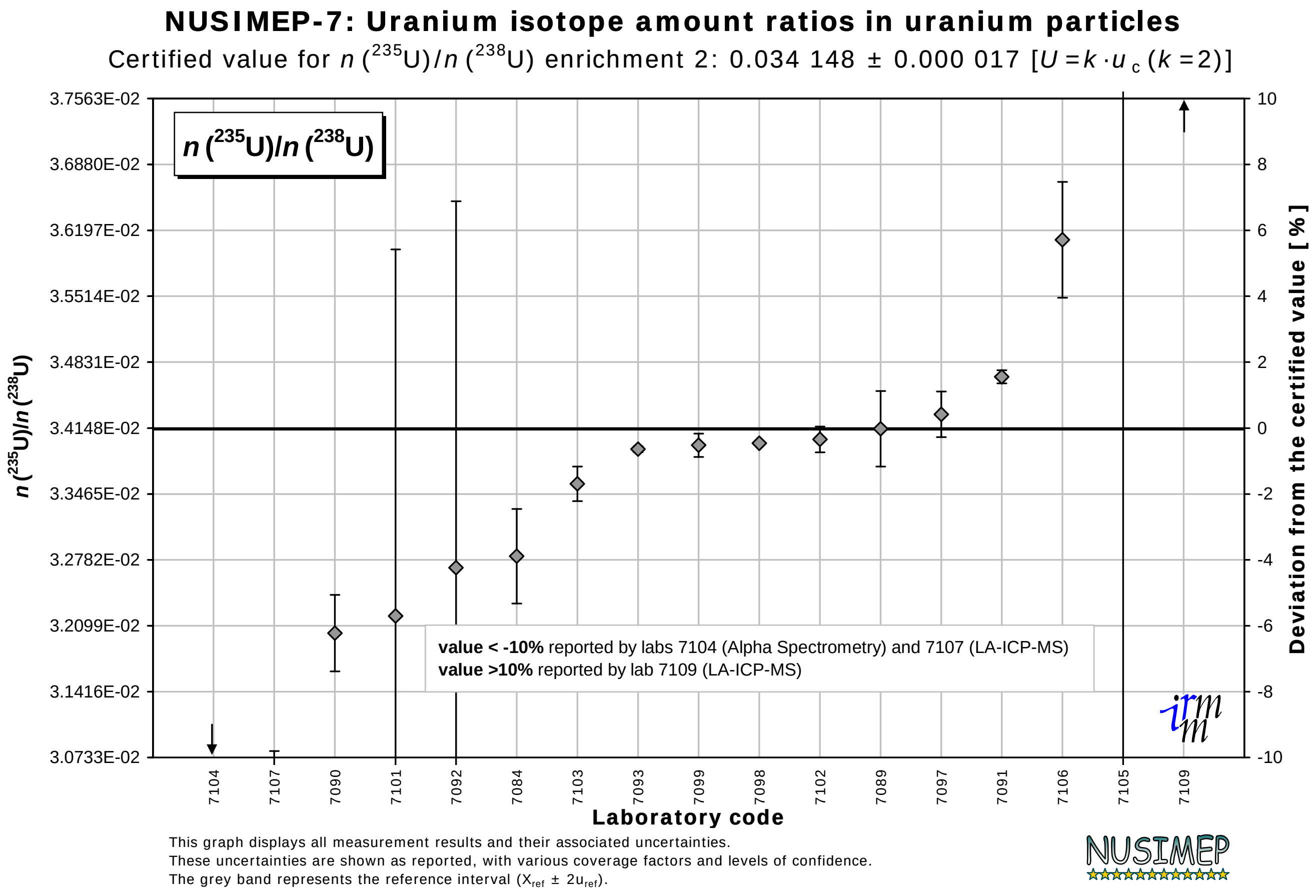
Certified value for $n(^{235}U)/n(^{238}U)$ enrichment 1 : 0.009 072 6 ± 0.000 004 5 [$U = k \cdot u_c (k = 2)$]

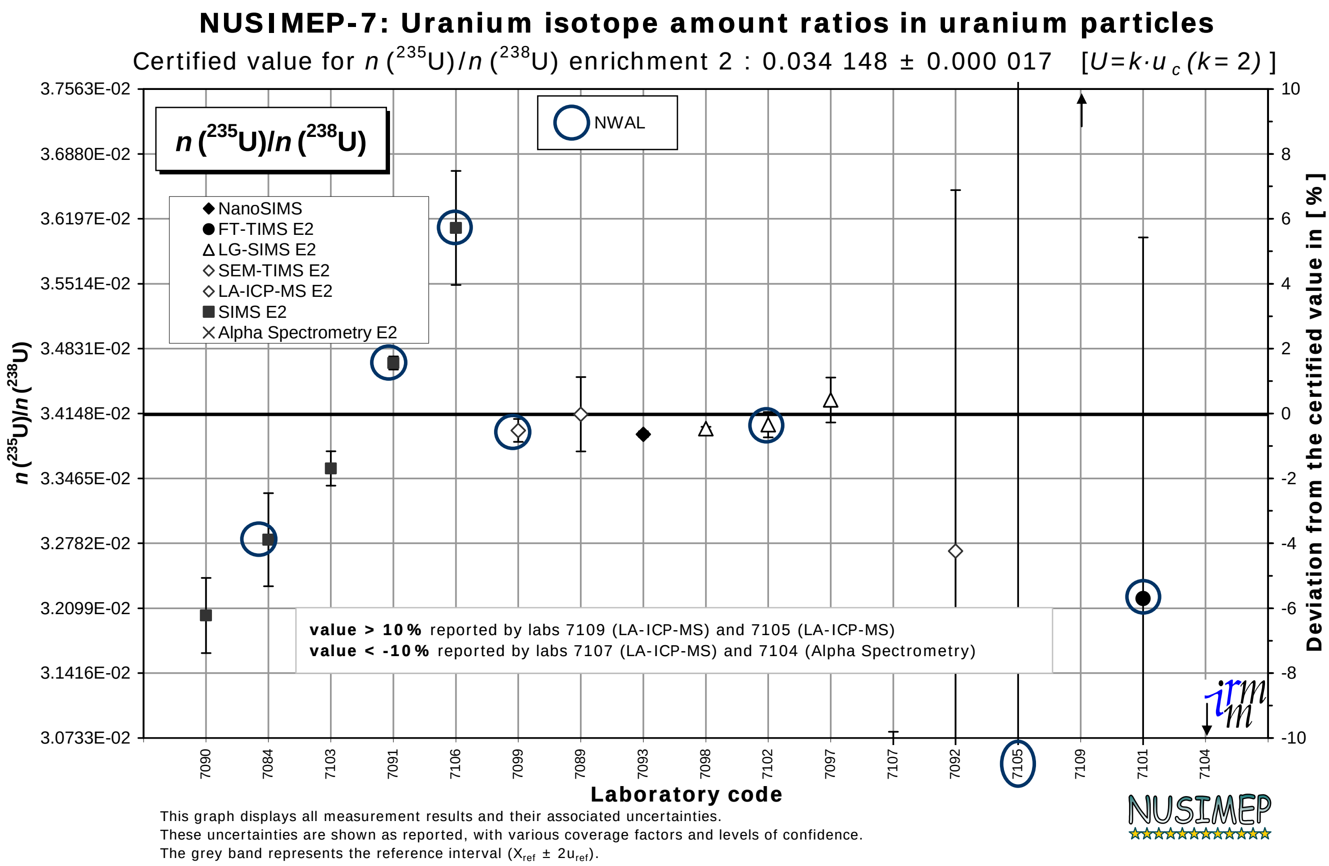


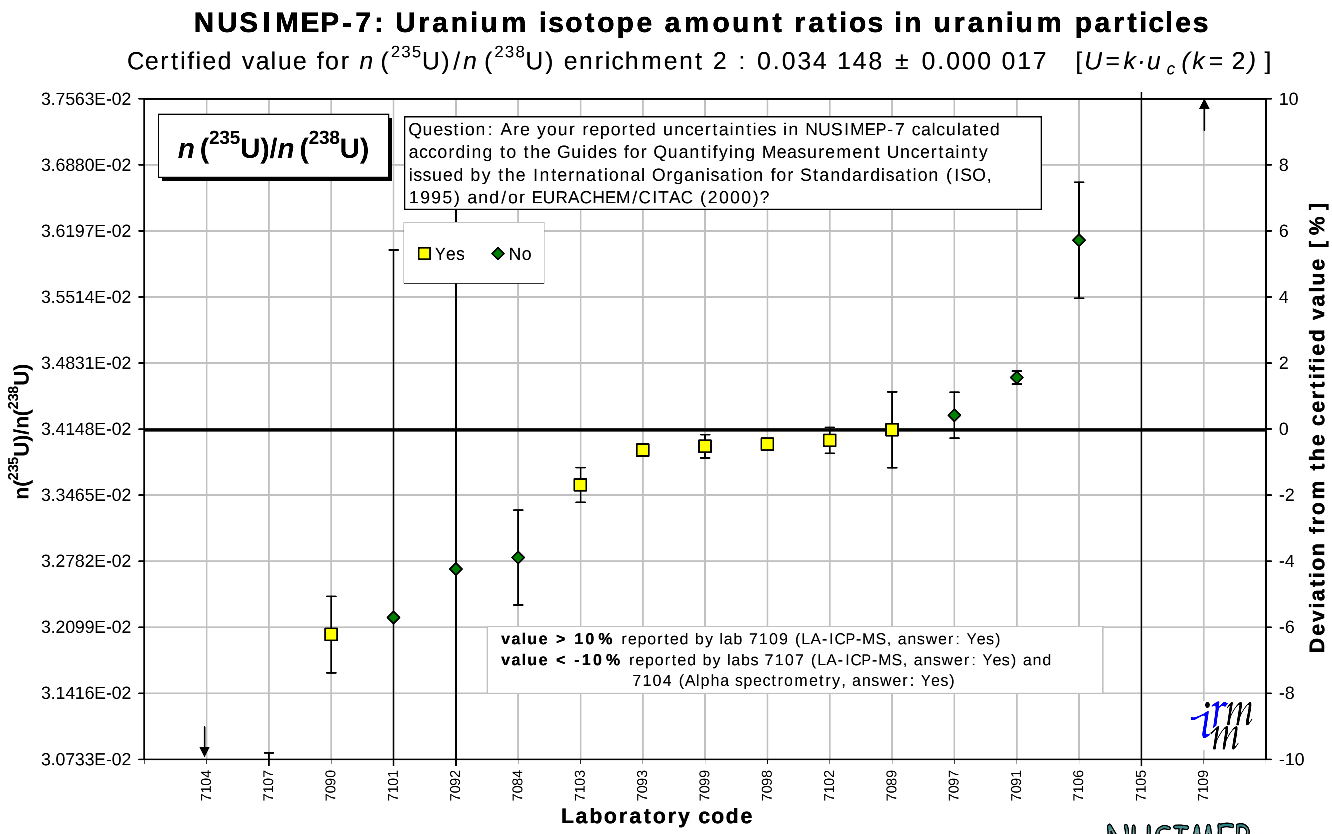
This graph displays all measurement results and their associated uncertainties.
These uncertainties are shown as reported, with various coverage factors and levels of confidence.
The grey band represents the reference interval ($X_{ref} \pm 2u_{ref}$).

Annex 1 1: Results for $n(^{235}U)/n(^{238}U)$ of the second enrichment of the sample with a double isotopic deposition

Laboratory	Analytical method	Reported $n(^{235}U)/n(^{238}U)$	Reported uncertainty $n(^{235}U)/n(^{238}U)$	Coverage factor *k*	z score	zeta score
7084	SIMS	0.03282	0.00049	1	7.78	2.71
7089	SEM-TIMS	0.03414	0.000392	1	0.05	0.02
7090	SIMS	0.032023	0.000396	2	12.45	10.72
7091	SIMS	0.0346798	0.0000676	1	3.11	7.81
7092	LA-ICP-MS	0.0327	0.0038	2	8.48	0.76
7093	NanoSIMS	0.03393	0.00003	2	1.28	12.64
7097	LG-SIMS	0.034291	0.000237	2	0.84	1.20
7098	LG-SIMS	0.03399	0.00002	2	0.93	12.04
7099	SEM-TIMS	0.03397	0.00012	2	1.04	2.94
7101	FT-TIMS	0.0322	0.0038	1	11.41	0.51
7102	LG-SIMS	0.03403086	0.00013330	1	0.69	0.88
7103	SIMS	0.03357	0.00018	1.96	3.39	6.27
7104	Alpha spectrometry	0.021	0.003	1	77.01	4.38
7105	LA-ICP-MS	0.04	0.04	2	34.27	0.29
7106	SIMS	0.0361	0.0006	1	11.43	3.25
7107	LA-ICP-MS	0.0286	0.0022	1	32.49	2.52
7109	LA-ICP-MS	0.214	0.0000983	2	1053.37	3605.72


NUSIMEP-7: Uranium isotope amount ratios in uranium particles
Certified value for n (235U)/n (238U) enrichment 2: 0.034 148 ± 0.000 017 [U = k · u c (k = 2)]
n (235U)/n (238U)
value < -10% reported by labs 7104 (Alpha Spectrometry) and 7107 (LA-ICP-MS)
value >10% reported by lab 7109 (LA-ICP-MS)
Deviation from the certified value [%]
Laboratory code
7104 7107 7090 7101 7092 7084 7103 7093 7099 7098 7102 7089 7097 7091 7106 7105 7109
3.7563E-02 3.6880E-02 3.6197E-02 3.5514E-02 3.4831E-02 3.4148E-02 3.3465E-02 3.2782E-02 3.2099E-02 3.1416E-02 3.0733E-02
10 8 6 4 2 0 -2 -4 -6 -8 -10
This graph displays all measurement results and their associated uncertainties.
These uncertainties are shown as reported, with various coverage factors and levels of confidence.
The grey band represents the reference interval (Xref ± 2uref).
NUSIMEP

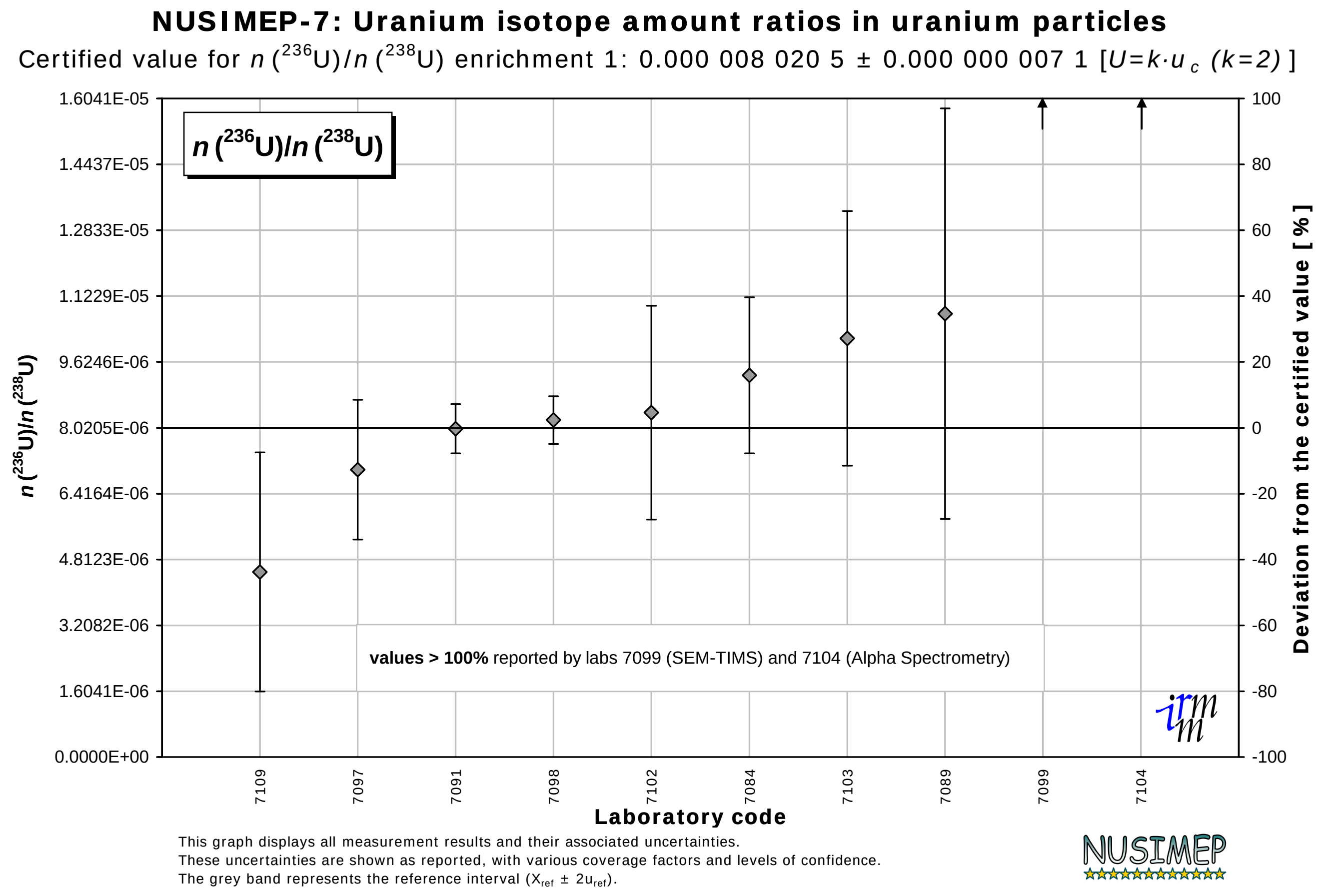

NUSIMEP-7: Uranium isotope amount ratios in uranium particles
Certified value for n (235U)/n (238U) enrichment 2 : 0.034 148 ± 0.000 017 [U=k·u c (k= 2)]
n (235U)/n (238U)
NWAL
NanoSIMS
FT-TIMS E2
LG-SIMS E2
SEM-TIMS E2
LA-ICP-MS E2
SIMS E2
Alpha Spectrometry E2
value > 10% reported by labs 7109 (LA-ICP-MS) and 7105 (LA-ICP-MS)
value < -10% reported by labs 7107 (LA-ICP-MS) and 7104 (Alpha Spectrometry)
Deviation from the certified value in [%]
Laboratory code
7090 7084 7103 7091 7106 7099 7089 7093 7098 7102 7097 7107 7092 7105 7109 7101 7104
3.7563E-02 3.6880E-02 3.6197E-02 3.5514E-02 3.4831E-02 3.4148E-02 3.3465E-02 3.2782E-02 3.2099E-02 3.1416E-02 3.0733E-02
10 8 6 4 2 0 -2 -4 -6 -8 -10
This graph displays all measurement results and their associated uncertainties.
These uncertainties are shown as reported, with various coverage factors and levels of confidence.
The grey band represents the reference interval (Xref ± 2uref).
NUSIMEP



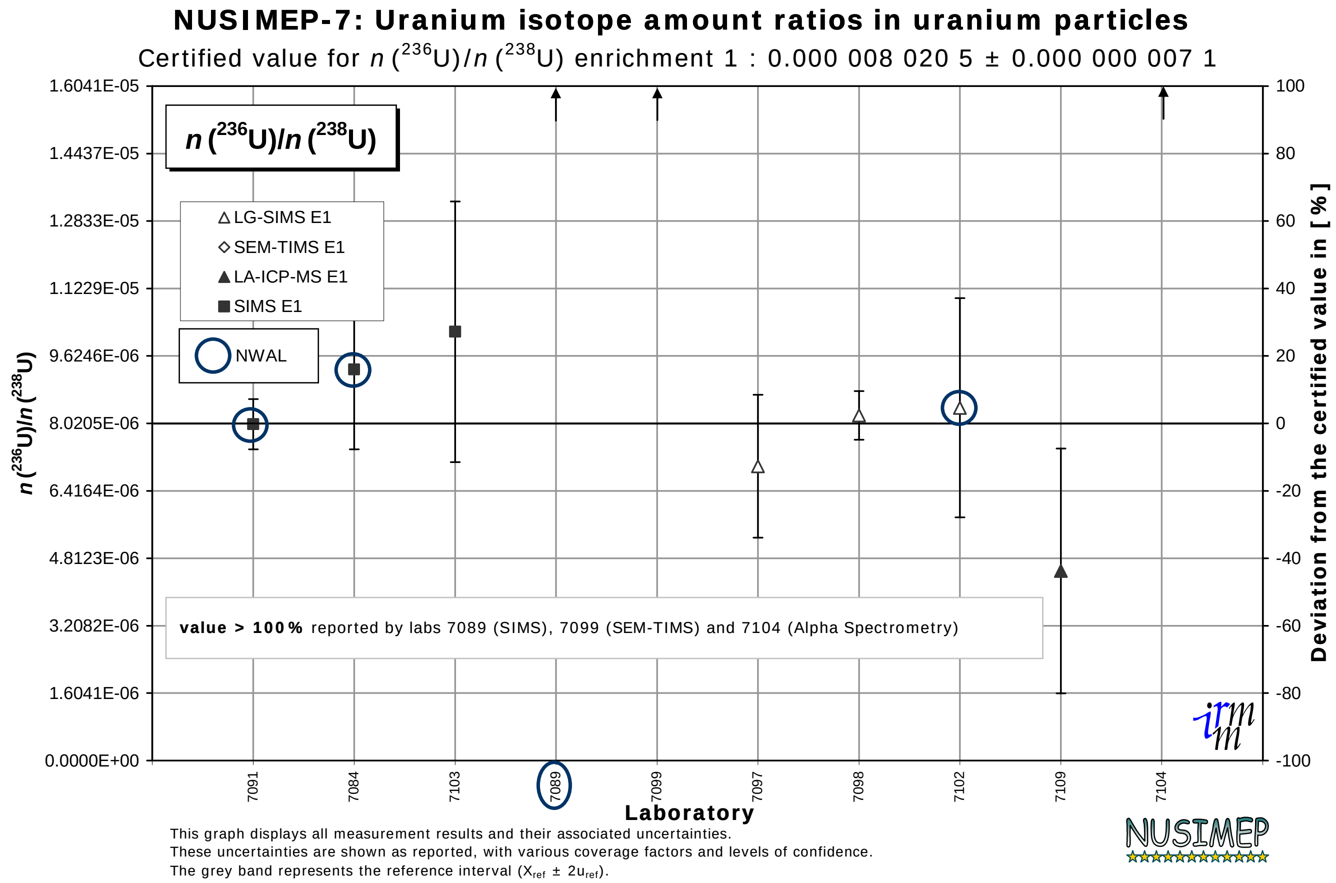
This graph displays all measurement results and their associated uncertainties.
These uncertainties are shown as reported, with various coverage factors and levels of confidence.
The grey band represents the reference interval ($X_{ref} \pm 2u_{ref}$).

Annex 12: Results for $n(^{236}U)/n(^{238}U)$ of the first enrichment of the sample with a double isotopic deposition

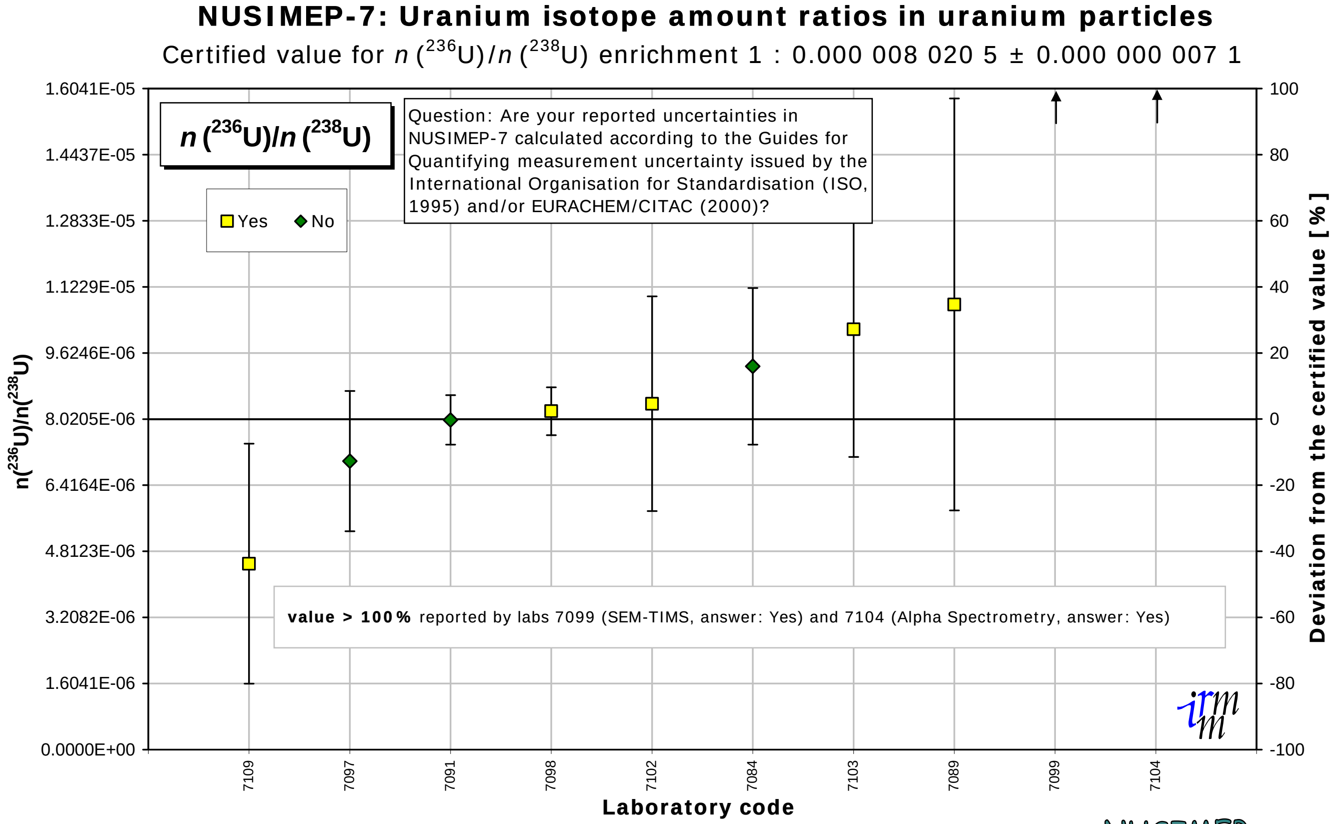
Laboratory	Analytical method	Reported $n(^{236}U)/n(^{238}U)$	Reported uncertainty $n(^{236}U)/n(^{238}U)$	Coverage factor *k*	z score	zeta score
7084	SIMS	0.0000093	0.0000019	1	0.32	0.67
7089	SEM-TIMS	0.0000108	0.000005	1	0.69	0.56
7090						
7091	SIMS	0.000008	0.0000006	1	0.01	0.03
7092						
7093						
7097	LG-SIMS	0.000007	0.0000017	2	0.25	1.20
7098	LG-SIMS	0.00000821	0.00000058	2	0.05	0.65
7099	SEM-TIMS	0.000029	0.000003	2	5.23	13.99
7101						
7102	LG-SIMS	0.00000839	0.00000261	1	0.09	0.14
7103	SIMS	0.0000102	0.0000031	1.96	0.54	1.38
7104	Alpha spectrometry	0.00026	0.00006	1	62.83	4.20
7105						
7106						
7107						
7109	LA-ICP-MS	0.00000451	0.00000291	2	0.88	2.41



NUSIMEP-7: Uranium isotope amount ratios in uranium particles
Certified value for n (236U)/n (238U) enrichment 1: 0.000 008 020 5 ± 0.000 000 007 1 [U=k·u c (k=2)]
n (236U)/n (238U)
values > 100% reported by labs 7099 (SEM-TIMS) and 7104 (Alpha Spectrometry)
Laboratory code
Deviation from the certified value [%]
7109
7097
7091
7098
7102
7084
7103
7089
7099
7104
This graph displays all measurement results and their associated uncertainties.
These uncertainties are shown as reported, with various coverage factors and levels of confidence.
The grey band represents the reference interval (Xref ± 2uref).



NUSIMEP-7: Uranium isotope amount ratios in uranium particles
Certified value for n (236U)/n (238U) enrichment 1 : 0.000 008 020 5 ± 0.000 000 007 1
n (236U)/n (238U)
LG-SIMS E1
SEM-TIMS E1
LA-ICP-MS E1
SIMS E1
NWAL
value > 100% reported by labs 7089 (SIMS), 7099 (SEM-TIMS) and 7104 (Alpha Spectrometry)
Laboratory
Deviation from the certified value in [%]
7091
7084
7103
7089
7099
7097
7098
7102
7109
7104
This graph displays all measurement results and their associated uncertainties.
These uncertainties are shown as reported, with various coverage factors and levels of confidence.
The grey band represents the reference interval (Xref ± 2uref).



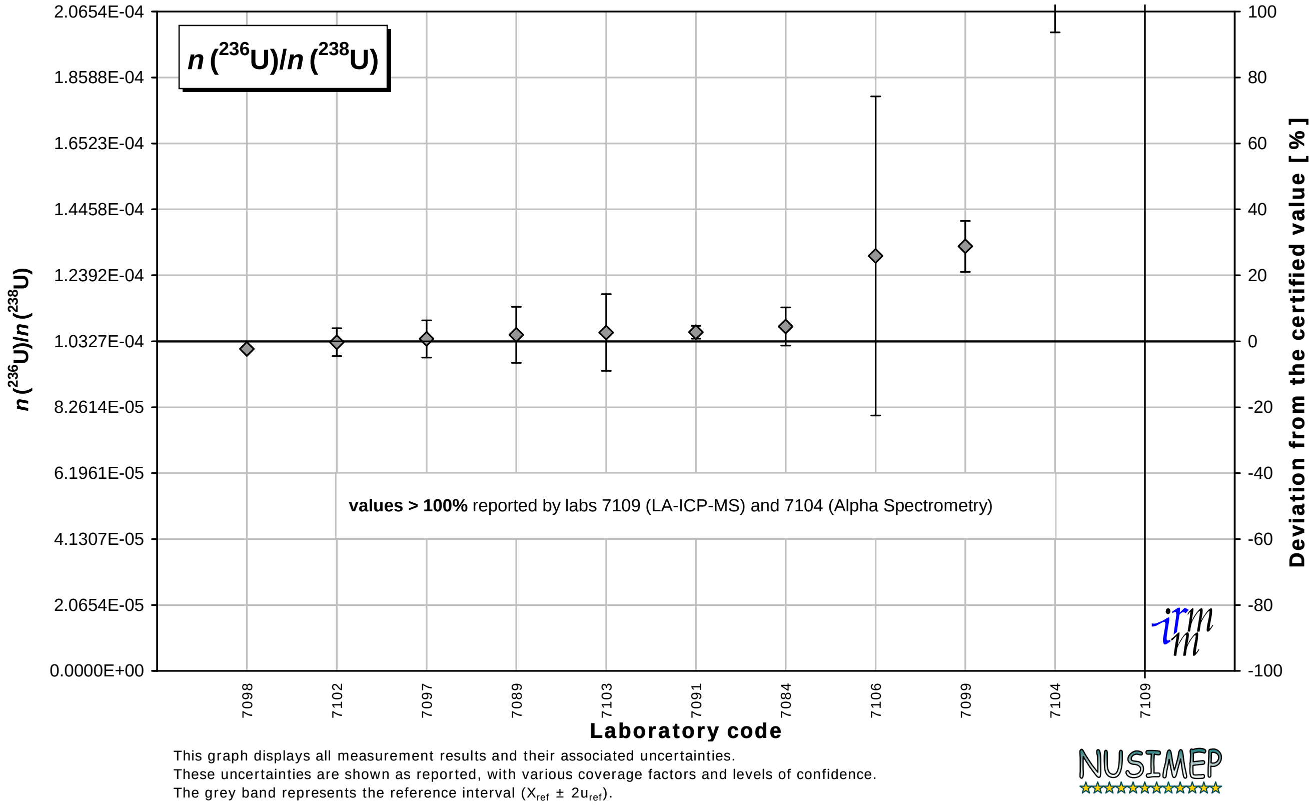
This graph displays all measurement results and their associated uncertainties.
These uncertainties are shown as reported, with various coverage factors and levels of confidence.
The grey band represents the reference interval ($X_{ref} \pm 2u_{ref}$).

Annex 13: Results for $n(^{236}U)/n(^{238}U)$ of the second enrichment of the sample with a double isotopic deposition

Laboratory	Analytical method	Reported $n(^{236}U)/n(^{238}U)$	Reported uncertainty $n(^{236}U)/n(^{238}U)$	Coverage factor k	z score	zeta score
7084	SIMS	0.0001079	0.000006	1	0.09	0.77
7089	SEM-TIMS	0.0001053	0.0000088	1	0.04	0.23
7090						
7091	SIMS	0.0001061	0.000002	1	0.05	1.42
7092						
7093						
7097	LG-SIMS	0.000104	0.0000058	2	0.01	0.25
7098	LG-SIMS	0.00010088	0.00000079	2	0.05	6.02
7099	SEM-TIMS	0.000133	0.000008	2	0.58	7.43
7101						
7102	LG-SIMS	0.00010300	0.00000437	1	0.01	0.06
7103	SIMS	0.000106	0.000012	1.96	0.05	0.45
7104	Alpha spectrometry	0.00026	0.00006	1	3.04	2.61
7105						
7106	SIMS	0.00013	0.00005	1	0.52	0.53
7107						
7109	LA-ICP-MS	0.000313	0.0239	2	4.06	0.02

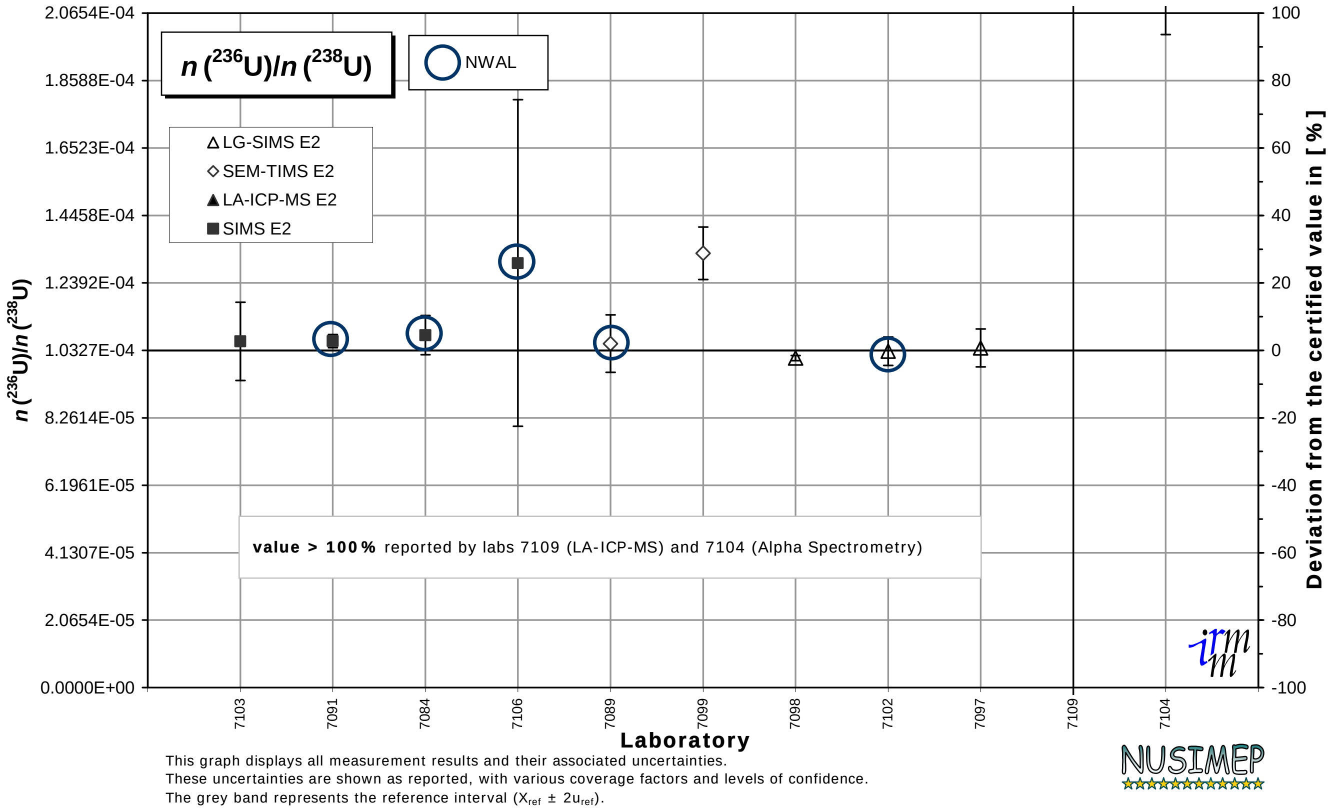
NUSIMEP-7: Uranium isotope amount ratios in uranium particles

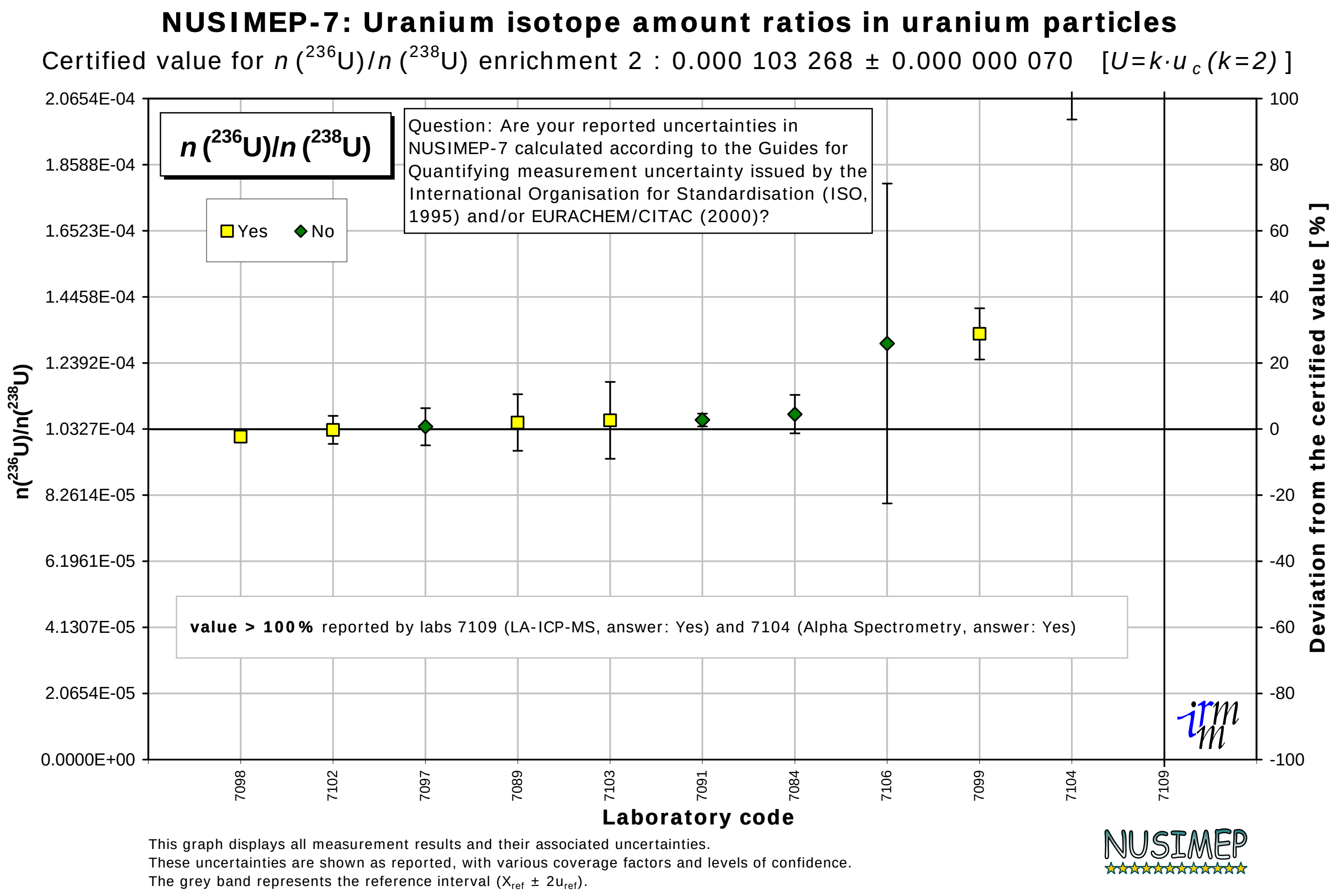
Certified value for $n(^{236}U)/n(^{238}U)$ enrichment 2: 0.000 103 268 ± 0.000 0000 070 [$U = k \cdot u_c$ (k = 2)]



NUSIMEP-7: Uranium isotope amount ratios in uranium particles

Certified value for $n(^{236}U)/n(^{238}U)$ enrichment 2 : 0.000 103 268 ± 0.000 000 070 [$U = k \cdot u_c$ (k=2)]




NUSIMEP-7: Uranium isotope amount ratios in uranium particles
Certified value for n (236U)/n (238U) enrichment 2 : 0.000 103 268 ± 0.000 000 070 [U=k·u c (k=2)]
n (236U)/n (238U)
Yes
No
Question: Are your reported uncertainties in NUSIMEP-7 calculated according to the Guides for Quantifying measurement uncertainty issued by the International Organisation for Standardisation (ISO, 1995) and/or EURACHEM/CITAC (2000)?
value > 100% reported by labs 7109 (LA-ICP-MS, answer: Yes) and 7104 (Alpha Spectrometry, answer: Yes)
n(236U)/n(238U)
2.0654E-04
1.8588E-04
1.6523E-04
1.4458E-04
1.2392E-04
1.0327E-04
8.2614E-05
6.1961E-05
4.1307E-05
2.0654E-05
0.0000E+00
Deviation from the certified value [%]
100
80
60
40
20
0
-20
-40
-60
-80
-100
7098
7102
7097
7089
7103
7091
7084
7106
7099
7104
7109
Laboratory code
This graph displays all measurement results and their associated uncertainties.
These uncertainties are shown as reported, with various coverage factors and levels of confidence.
The grey band represents the reference interval (Xref ± 2uref).

Annex 14: Certificates

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

CERTIFICATE of a reference measurement

IM/MeaC/07/04-IMN-10124
2 December 2004

1. Applicant: R Wellum, IRMM

2. Sample Identification:
   - LOT 2436
   - Chemical form: UF_6, and Uranium Nitrate
   - IM sample registration number: IMN-10124

3. Measurands:
   - Isotopic composition

isotope amount ratio(s)	
$n(^{234}U)/n(^{238}U)$	0.000 074 365(60)
$n(^{235}U)/n(^{238}U)$	0.009 072 6(45)
$n(^{236}U)/n(^{238}U)$	0.000 008 0205(71)

amount fraction (·100)		mass fraction (·100)	
$n(^{234}U)/n(U)$	0.007 369 0(59)	$m(^{234}U)/m(U)$	0.007 245 7(58)
$n(^{235}U)/n(U)$	0.899 03(45)	$m(^{235}U)/m(U)$	0.887 78(44)
$n(^{236}U)/n(U)$	0.000 794 78(70)	$m(^{236}U)/m(U)$	0.000 788 17(69)
$n(^{238}U)/n(U)$	99.092 81(45)	$m(^{238}U)/m(U)$	99.104 19(44)

molar mass: 238.023 440(14) $g \cdot mol^{-1}$

4. Date of receipt of sample : 1 April 2004
   Date of completion of measurement : 26 May 2004

5. Uncertainty:
All uncertainties indicated are expanded uncertainties $U = k \cdot u_c$ where u_c is the combined standard uncertainty calculated according to the ISO/BIPM guide. They are given in parentheses and include a coverage factor k=2. They apply to the last two digits of the value. The values certified are traceable to the SI.
The primary certified values are the isotope amount ratios; other values are derived from them. Reproducing the derived values may result in differences due to rounding errors.

6. The traceability to SI is established through UF_6-standards as IRMM 2400 and IRMM 071.

7. Analytical measurement procedure

- Mass spectrometric measurements were performed by W De Bolle for the $[n(^{235}U)/n(^{238}U)]$ isotope ratio using the MAT511 mass spectrometer on UF_6 samples prepared by A Moens and W De Bolle. TIMS measurements on $[n(^{234}U)/n(^{238}U)]$ and $[n(^{236}U)/n(^{238}U)]$ were performed by S Richter, H Kuehn and A Alonso using the TRITON on samples chemically prepared by A Alonso. A Verbruggen was responsible for the preparation and issuance of the certificate.
- The atomic masses, used in the calculations, are from G. Audi and A.H. Wapstra, The 1993 atomic mass evaluation , Nucl Phys A565 (1993) 1-65.
- Reference number of the measurement data: measurement number T4504, logged in S:\Im UNIT\Secure Data\Archive MS Measurements data files\TRITON\data.

Stephan Richter
Task leader

Copies:
P Taylor, IM Unit Head
R Wellum
A Alonso
H Kühn
A Moens
Archive

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

CERTIFICATE of a reference measurement

IM/MeaC/22/04-IMN-10004/REIMEP 15D
2 December 2004

1. Applicant: R Wellum

2. Sample Identification:
   - REIMEP 15 D, LOT 2392
   - chemical form: UF_6, and Uranium Nitrate
   - IM sample registration number: IMN 10004

3. Measurands:
   - Isotopic composition

isotope amount ratio(s)	
$n(^{234}U)/n(^{238}U)$	0.000 345 14(24)
$n(^{235}U)/n(^{238}U)$	0.034 148(17)
$n(^{236}U)/n(^{238}U)$	0.000 103 268(70)

amount fraction (·100)		mass fraction (·100)	
$n(^{234}U)/n(U)$	0.033 360(22)	$m(^{234}U)/m(U)$	0.032 812(22)
$n(^{235}U)/n(U)$	3.300 6(16)	$m(^{235}U)/m(U)$	3.260 3(16)
$n(^{236}U)/n(U)$	0.009 981 4(66)	$m(^{236}U)/m(U)$	0.009 901 5(66)
$n(^{238}U)/n(U)$	96.656 0(16)	$m(^{238}U)/m(U)$	96.697 0(16)

molar mass: 237.950 001(49) $g \cdot mol^{-1}$

4. Date of receipt of sample : 27 July 1999
   Date of completion of re-measurement : 3 May 2004

5. Uncertainty:
All uncertainties indicated are expanded uncertainties $U = k \cdot u_c$ where u_c is the combined standard uncertainty calculated according to the ISO/BIPM guide. They are given in parentheses and include a coverage factor k=2. They apply to the last two digits of the value. The values certified are traceable to the SI.
The primary certified values are the isotope amount ratios; other values are derived from them. Reproducing the derived values may result in differences due to rounding errors.

6. The traceability to SI is established through UF_6-standards as IRMM-028 and IRMM-295.

7. Analytical measurement procedure

- Mass spectrometric measurements were performed by W De Bolle for the [$n(^{235}U)/n(^{238}U)$] isotope ratio using the MAT511 mass spectrometer on UF_6 samples prepared by W De Bolle. TIMS re-measurements on [$n(^{234}U)/n(^{238}U)$] and [$n(^{236}U)/n(^{238}U)$] were performed by S Richter using the TRITON on samples chemically prepared by A Alonso. A Verbruggen was responsible for the preparation and issuance of the certificate.
- The atomic masses, used in the calculations, are from G. Audi and A.H. Wapstra, The 1993 atomic mass evaluation , Nucl Phys A565 (1993) 1-65.
- Reference number of the measurement data: measurement number T4429, logged in S:\Im UNIT\Secure Data\Archive MS Measurements data files\TRITON\data.

Stephan Richter
Task leader

Copies:
P Taylor, IM Unit Head
A Alonso
R Wellum
Archive

Annex 15: Summary of the information given by the participants on instrument parameters and measurement conditions for SIMS, TIMS and LA-ICP-MS

SIMS: parameters	
Type of SIMS used (brand, model,..):	
CAMECA IMS 3F	
CAMECA IMS 7F	
CAMECA IMS 6f	
CAMECA 4F	
CAMECA IMS-1280	
CAMECA IMS-4F	
Cameca NanoSIMS 50	
CAMECA IMS 1280	
IMS 1280	
Did you use automatic particle search software? If 'Yes', which software?	
b) No	
a) Yes	APM
b) No	
a) Yes	Yes and No. We have an automated particle software but there was no need to search for particles in this case as there were so many.
a) Yes	APM
b) No	
a) Yes	Cameca
b) No	
a) Yes	APM from Cameca, but the planchettes were too heavily loaded for APM to be useful
Primary ion beam and primary ion beam accelerating energy:	
Oyxgen (O2+) 10 keV	
O2+, +15KV	
15 kV.	
0.5nA, 15kV	
O2+,	
positive 15 kV and negative 10 kV	
O-, 16 keV	
O- 13 keV	
O-, 13 keV	

Secondary ion extraction kV:
4.5keV
+5kV
7 kV.
4.5 kV
8
4.44
8 kV
10 kV
10keV
Mass resolution used:
around 300
350
300
360 at 10%.
2400
300
~3500
M/deltaM at 10% of peak height about 2500
About 2200-2400
Secondary ion energy window:
Fully open with one turn in
75eV
150 µm.
45eV
50 eV
25 keV
Not measured, wide open
50 eV
55eV
EM detector type:
Balzers SEV217
Discrete dynode Em (ETP)
Jalousie type.
ETP
CAMECA/MassCon
dinod
Hamamatsu small multiplier
ETP AF133H
Hamamamatsu

Dead time correction applied? If 'Yes' report the dead time:	
a) Yes	75 ns
a) Yes	23ns
a) Yes	25 ns.
a) Yes	28nS
a) Yes	65 nsec (electronically fixed and measured)
a) Yes	0,3 s for each mass
b) No	
a) Yes	27.5 ns
a) Yes	60 ns
Presputter conditions (before microprobe measurements)	
Small amount of sputtering while finding and centering particles	
no presputtering (but an APM has been performed before the microprobe measurement)	
About 10s.	
A few seconds	
None	
For positive ions - 500 um x 500 um, 300 nA, 15 s; for negative ions - 500 um x 500 um, 100 nA, 15 s	
none	
No intentional presputter, only enough to find and align a particle	
As little as possible. Dose acquired while running automated particle search and while manually centering particle for microbeam analysis..	
Microprobe measurement conditions:	
focussed beam rastered over 200 microns	
250pA focused beam	
A little unfocus is arranged.	
O2+	
30 - 50 pico-amp primary beam	
For positive - 3 nA; for negative - 1 nA	
~200 pA	
focused beam, small raster, 50 um image field, typically 0.8 nA current	
Kohler illumination	
Spot size (estimated diameter):	
50 microns	
3µm	
More than 10 µm.	
2-3 um	
2 micron	
5 um	
~600 nm	
estimate 5 micrometers (not measured)	
15 x 30 micron	

Raster size (if used):
200 microns
0
0
5-6 um
2 to 5 micron
no used
3 x 3 micron
10 x 10 micrometer
none
Masses cycled & cycle times:
233U (2sec) 234U(4 sec) 235U(4sec) 236U(4sec) 238U(2 sec) 238U+H(2sec) over 20 cycles
234,235,236,238,239 12s per cycle
12;9.7s.
234-4s, 235-2s, 236-4s, 238-1s, 239-2s
multi-detector, 8 sec x 42
2 cycles - 7 s and 5 cycles - about 15 s
U-235 + UO-251; U-238 + UO-254 - approx. 4s per B field
232, 234, 235, 236, 238, 239 for 3, 4, 2, 4, 2, 4 s per cycle, wait times 2, 1, 1, 1, 1, 1 s per cycle, 20 cycles
Multicollector mode, simultaneous 234,235,236,238,239, 10 second acquisition cycles
Total sputter time per measurement:
450 seconds
about 7min
120s.
5-8 minutes
336 sec
96 s
< 10 min
520 s
varies, but usually about 8 minutes
238U signal level from particle at start (cps)
1E4 < cps < 2e4
for the single deposit: 40000-100000cps, for the double deposit : 40000-400000cps
About 100000.
Typically at about 20000-40000 counts/second
150000
from 3000 to 10000
~2.10E3 - 2.10E4 cps depending on particle size
ranged from 1.5E4 to 2.8E4 cps
About 90,000 cps for NA less for enriched particles

238U signal level from particle at end (cps)
5E3 < cps < 2E4
for the single deposit:100-1000, for the double deposit: 500-10000
About 10000 to 100000.
Typically at about 5000 counts
20000
from 0 to 7000
~2.10E3 - 2.10E4 cps depending on particle size
ranged from 0.5E4 to 2.3E4 cps
varies, but we stop analyses when minor isotopes drop below 0.1 cps
Comment on stability of signal levels during microprobe: stable, decreasing, increasing, variable?
Linear or decreasing
decreasing
Decreasing.
First increasing, followed by decrease
typically decrease by factor of 5; dependant on particle size; makes more sense to ask integrated counts range (3E6 to 6E7)
stable - sometimes, decreasing - as a rule
stable
typically increasing to max, then decreasing to half of max over 20 cycles; time interpolation used to compensate for signal changes during run
Depends on size of the particle. Most particles decreased slowly.
Comments on appearance of U ion image field (if possible): clear particles?, diffuse spots?, uniform signal? Did this change during the analysis?
clear particles
clear particles but particles are very close each other. The spots tend to spread during the course of the analysis
The clear particles are selected and the appearance of them will be changed with the measurement.
Clear particle image but single deposit was very heavily loaded
fields very crowded with particles, but possible to find individual particles separated from others to target for MP, usually mix of large and quite small particles, imaging and discrimination of adjacent particles would be very difficult with 4f SIMS for these overloaded planchets. We found very little sign of mixing for the two composition planchets with careful particle selection.
clear particles
clear particles
Only sputtered enough to detect particles above U background, not enough info to assess image quality
Diffuse spots with high background. Spots got more diffuse after an analysis.

TIMS: parameters	
Type of TIMS used (brand, model...):	
Finnegan MAT 262	
modified VG 54-38	
TRITON	
Detector used (Brand):	
n/a	c) Discrete dynode SEM
Daly detector	d) Ion counting detector operation mode
Multiple Ion Counter (Thermo)	d) Ion counting detector operation mode, e) multiple detectors used
Dead time correction applied? If 'Yes' report the dead time:	
a) Yes	n/a
a) Yes	13
b) No	
Any other correction to the detector output ? If 'Yes' please specify:	
a) Yes	n/a
b) No	
b) No	
Specify filament type used:	
a) Single filament technique, c) Zone refined Re	
a) Single filament technique, c) Zone refined Re	
b) Double filament technique, c) Zone refined Re	
Measurement of single/multiple particles ?	
a) Single particles	
a) Single particles	
a) Single particles	
^{238}U average signal level:	
n/a	
For the single deposition: 1500cps For the double deposition: 100cps for the enriched isotopy, 1000cps for the other isotopy	
20,000 cps	

Overall ionisation efficiency (estimate):
0.1%
0.01%
1.0%

LA-ICP-MS: parameters	
Type of carrier gas used ? (He (%), Ar (%), other)	
Ar	
100% He in the cell, mixed with Ar shortly after the cell exit.	
He (100%)	
He	
Combinaison with nebulizer ? If 'Yes' specify the type of nebulizer?	
a) Yes	Meinhard
a) Yes	DSN-100 (Nu Instruments) desolvating nebuliser but aspirated air for analysis after initial tuning
b) No	
a) Yes	PFA nebulizer in combination with membrane desolvation (DSN-100, Nu Instruments)
LA carrier introduced into nebulizer or additional mixing of nebulizer?	
Additional mixing	
additional mixing	
no	
additional mixing	
Gas flow after LA cell:	
100 mL/min	
0.8L/min He mixed with 0.7L/min Ar	
2 L min-1	
1700 mL /min	
Type of laser:	
LaserProbe	
UP193FX (193nm excimer, New Wave Research/ESI)	
Nd:YAG	
New Wave UP-193 ArF excimer laser	
Laser wavelength (nm):	
266	
193	
266	
193 nm	
Energy flux (J/cm2):	
0.02	
10-12	
10	
20.12	

Spot size (µm):
40
5
200
5
Repetition rate (Hz):
10
1
1
15
Ablation type (single spot, line scan, other...):
line scan
single pulse single spot
raster
line scan
Type of instrument:
a) ICP-HR-MS (single collector, high resolution)
c) ICP-SF-MS (single coll., sector field, using low resolution (<1000))
d) MC-ICP-MS (multi collector)
d) MC-ICP-MS (multi collector)
Type of detector:
SEM
MasCom SEM
Faraday cup
234: IC, 235:FC, 236:IC, 238:FC

Annex 16: Certified reference materials used by the NUSIMEP-7 participants

Laboratory	Does your laboratory use CRMs?	CRMs and suppliers
7084	Yes	NIST CRMs U-005, U-010, U-020, U-500
7089	Yes	NIST Uranium
7090	Yes	U005a, U010, U050 and CRM129-A
7091	Yes	NBL and IRMM
7092	Yes	NIST 610-612-614 glasses, a number of IRMM spikes and CRM's (e.g. 3636 spike and 074 series)
7093	Yes	CRM129a and others (NIST)
7097	Yes	SRM U900 from NBS
7098	Yes	CRM U010, NBL
7099	Yes	NIST U010, U030, U005
7101	Yes	NIST CRMs U-005, U-500, natural uranium
7102	Yes	NBS U010, U030A, U200, U500, U930, IRMM SMS 7686 series, IRMM SMS 7259 series
7103	Yes	The CRMs used in our lab are manufactured by New Brunswick Lab.
7104	No	
7105	Yes	hundreds of CRMs
7106	Yes	Several CRM from IAEA and Russian sources
7107	Yes	U and Pu from IRMM and NBL, Glass from NIST for Laser Ablation
7109	Yes	IRMM-187 (IRMM), IRMM-184 (IRMM), CRM U030 A , CRM U500, CRM 112-A, CRM U005 (New Brunswick Laboratory), S1 and S3 glass particles (IRMM)

Annex 17: Summary of lab scores

Summary of lab scores for single deposition

	$n(^{234}U)/n(^{238}U)$		$n(^{235}U)/n(^{238}U)$		$n(^{236}U)/n(^{238}U)$	
Laboratory	**z score**	**zeta score**	**z score**	**zeta score**	**z score**	**zeta score**
7084	0.99	0.36	5.35	0.90	0.74	0.99
7089	4.39	0.59	2.42	0.37	1.79	1.41
7090			12.82	14.46		
7091	0.29	0.45	1.12	2.20	0.23	1.84
7092			84.37	2.73		
7093			0.50	1.49		
7097	0.20	0.19	1.25	1.38	0.25	1.20
7098	0.20	0.73	1.03	7.27	0.003	0.05
7099	5.72	3.54	3.14	4.74	6.98	13.99
7101			5.13	0.89		
7102	0.18	0.21	0.57	0.56	0.07	0.17
7103	0.52	0.32	1.58	2.19	0.29	0.77
7104	1.42	0.38	133.87	6.07		
7105	0.73	0.39	3.14	1.43	0.99	1.33
7106	3.03	0.19	0.60	0.14	15.46	3.10
7107			0.21	0.79		
7109	150.41	3.75	529.67	16.46	21.91	3.37

Summary of lab scores for double deposition enrichment 1

	$n(^{234}U)/n(^{238}U)$		$n(^{235}U)/n(^{238}U)$		$n(^{236}U)/n(^{238}U)$	
Laboratory	**z score**	**zeta score**	**z score**	**zeta score**	**z score**	**zeta score**
7084	1.92	0.81	8.96	6.55	0.32	0.67
7089	2.01	0.24	4.70	1.22	0.69	0.56
7090			3.65	1.54		
7091	1.20	1.72	2.06	3.80	0.01	0.03
7092			977.17	10.43		
7093			0.72	1.30		
7097	0.20	0.15	0.10	0.11	0.25	1.20
7098	0.02	0.05	1.16	6.72	0.05	0.65
7099	2.49	1.85	0.94	1.70	5.23	13.99
7101			2.04	0.12		
7102	0.22	0.15	0.51	0.44	0.09	0.14
7103	0.23	0.15	0.28	0.15	0.54	1.38
7104	89.09	16.56	262.93	3.98	62.83	4.20
7105			1.60	0.03		
7106	13.79	0.85	2.81	0.42		
7107			430.47	8.88		
7109	11.64	1.96	3.91	0.36	0.88	2.41

Summary of lab scores for double deposition enrichment 2

	$n(^{234}U)/n(^{238}U)$		$n(^{235}U)/n(^{238}U)$		$n(^{236}U)/n(^{238}U)$	
Laboratory	**z score**	**zeta score**	**z score**	**zeta score**	**z score**	**zeta score**
7084	1.49	1.76	7.78	2.71	0.09	0.77
7089	0.44	0.10	0.05	0.02	0.04	0.23
7090			12.45	10.72		
7091	0.39	1.08	3.11	7.81	0.05	1.42
7092			8.48	0.76		
7093			1.28	12.64		
7097	1.14	1.97	0.84	1.20	0.01	0.25
7098	0.03	0.29	0.93	12.04	0.05	6.02
7099	0.56	0.81	1.04	2.94	0.58	7.43
7101			11.41	0.51		
7102	0.41	0.42	0.69	0.88	0.01	0.06
7103	0.57	1.17	3.39	6.27	0.05	0.45
7104	12.19	10.51	77.01	4.38	3.04	2.61
7105			34.27	0.29		
7106	1.75	0.30	11.43	3.25	0.52	0.53
7107			32.49	2.52		
7109	325.07	10.18	1053.37	3605.72	4.06	0.02

European Commission

EUR 25179 EN – Joint Research Centre – Institute for Reference Materials and Measurements
Title: NUSIMEP-7: Uranium isotope amount ratios in uranium particles
Authors: Jan Truyens, Elzbieta Stefaniak, Sébastien Mialle, Yetunde Aregbe
Luxembourg: Publications Office of the European Union
2011 – 84 pp. – 21.0 x 29.7 cm
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Abstract

The Additional Protocol (AP) authorizes safeguards authorities to verify the absence of undeclared nuclear activities in all parts of a state's nuclear fuel cycle as well as any other location where nuclear material is or may be present. As a part of the Additional Protocol, environmental sampling has become an important tool for the detection of non-declared nuclear activities. In environmental sampling micrometer-sized uranium particles with an isotopic composition characteristic for the processes at the inspected facility need to be collected, identified and analysed. Considering the potential consequences of the analyses, these measurements need to be subjected to a rigorous quality management system.

NUSIMEP-7 focused on measurements of *uranium isotope amount ratios in uranium particles* aiming to support EURATOM safeguards (DGENER), the IAEA network of analytical laboratories for particle analysis of environmental samples for safeguards (NWAL) and laboratories involved in uranium particle analysis. It was organised as a follow-up of NUSIMEP-6, which was the first NUSIMEP in 2008 on particle analysis coordinated by IRMM, intended as a pilot interlaboratory comparison in this field. The feedback from participants in NUSIMEP-6 was taken into account for optimisation and improvements resulting in the second NUSIMEP on particle analysis coordinated by IRMM. NUSIMEP-7 was open for participation to all laboratories in the field of particle analysis, and explicitly recommended by the IAEA to the NWAL for participation.
The NUSIMEP-7 test samples were prepared by controlled hydrolysis of certified uranium hexafluoride. Participating laboratories in NUSIMEP-7 received the test samples of uranium particles on two graphite planchets with undisclosed isotope amount ratio values $n(^{234}\mathrm{U})/n(^{238}\mathrm{U})$, $n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$ and $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$; one planchet with particles of single isotopic deposition and the other with particles of two different isotopic compositions. For both samples, the uranium isotope amount ratios had to be measured using their routine analytical procedures. Measurement of the major ratio $n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$ was obligatory; measurement of the minor ratios $n(^{234}\mathrm{U})/n(^{238}\mathrm{U})$ and $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ was optional. 24 institutes registered for NUSIMEP-7, whereof 17 have reported measurement results using different analytical methods, among those were 7 NWAL laboratories. The participants' measurement results have been evaluated against the certified reference values by means of z-scores and zeta-scores in compliance with ISO 13528:2005. The results of NUSIMEP-7 do not only confirm the capability of laboratories to measure $n(^{234}\mathrm{U})/n(^{238}\mathrm{U})$, $n(^{235}\mathrm{U})/n(^{238}\mathrm{U})$ and $n(^{236}\mathrm{U})/n(^{238}\mathrm{U})$ in uranium particles of <1 µm, but also to distinguish between groups of particles with different isotopic composition. Furthermore they underpin the recent advances in instrumental techniques in the field of particle analysis. In addition feedback from the measurement communities in nuclear safeguards, nuclear security and earth sciences was collected in view of identifying future needs for NUSIMEP interlaboratory comparisons.

The Director of the Division of Concepts and Planning at the IAEA Department of Safeguards expressed her appreciation and compliments to IRMM for NUSIMEP-7 for '*further improving the detection and analysis capability of the IAEA's NWAL*'

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