



**NUSIMEP 5:**

**Uranium, plutonium and caesium  
isotopic abundances in a saline matrix**

**Report to participants**

L. Benedik, T. Altzitzoglou, R. van Ammel, S. Pomme, S. Richter,  
G. Sibbens, A. Stolarz, A. Verbruggen, R. Wellum

**NUSIMEP**  
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Institute for Reference Materials and Measurements

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## Summary

The Nuclear Signatures Inter-laboratory Measurement Evaluation Programme (NUSIMEP) was established to support the growing need to measure the isotopic abundances of elements characteristic of 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 Non-Proliferation Treaty (NPT). Through this and similar programmes, the degree of equivalence of measurements of individual laboratories can be ascertained.

It complements the two other long running programmes at the Institute for Reference Materials and Measurements (IRMM): the Regular European Inter-laboratory Measurement Evaluation Programme (REIMEP) in the nuclear field and the International Measurement Evaluation Programme (IMEP) for measurements of amounts of chemical substances.

The first three NUSIMEP campaigns were devoted to measurements of uranium in various media ranging from simple dried nitrate salts to a complex saline solution matrix. In NUSIMEP 4 the matrix was made somewhat more complex: a simulated urine solution was prepared from the saline solution used in NUSIMEP 3 with the addition of urea.

In the NUSIMEP 5 campaign reported here, essentially the same saline solution was chosen as matrix and participating laboratories were requested to measure uranium isotopic ratios, plutonium isotopic and alpha activity ratios, and the gamma activity ratio of  $^{134}\text{Cs}$  relative to  $^{137}\text{Cs}$ .

The same 1% saline material purified from traces of uranium was used for preparation of 4 solutions of different isotopic composition. This type of matrix was shown in previous campaigns to provide a good substitute for natural environment samples. Uranium (depleted, near-natural and low-enriched) was added to each sample at a concentration of  $5 \text{ ng}\cdot\text{g}^{-1}$  and Pu was added to the first two solutions and Cs to the second two. The activity of the Pu and Cs was kept to below 1 Bq per sample so that samples could be transported using standard transport carriers.

The uranium isotopic ratios were mainly measured by the participating laboratories by alpha-particle- and mass-spectrometry, for the most part by inductively-coupled-plasma (ICP-MS) but also by thermal ionisation mass-spectrometry (TIMS). Plutonium isotopic ratios were measured by mass-spectrometry (again mainly ICP-MS) and activity ratios by alpha spectrometry. The gamma-ray activities of the Cs isotopes were measured by semi-conductor gamma-ray spectrometry.

The measurement results are shown as a set of graphs in which each laboratory is specified only as a number. Details of the laboratories' experience, qualifications and of their measurement methods were requested in a questionnaire and these are summarised also in this report.

# 1. Introduction

The NUSIMEP campaigns originated as a tool for laboratories to demonstrate their abilities to measure isotopic ratios in small amounts of nuclear materials in environmental samples. This programme is built on the experience gained in similar external quality control campaigns over many years: the Regular European Inter-laboratory Measurement Evaluation Programme, REIMEP and the International Measurement Evaluation Programme, IMEP. These programmes are open for all laboratories active in the appropriate field and have in common the following:

- The samples are prepared and certified at IRMM and are sent to participating laboratories as 'blind' samples;
- Participating laboratories are requested to measure the parameters specified using their standard methods and send results with uncertainties to IRMM. In return the laboratory receives the certified values;
- A summary of the campaign results is published giving a 'snapshot' of the laboratory measurement capabilities for these samples.

The NUSIMEP 5 campaign follows earlier campaigns for the measurement of uranium isotopic ratios in various media. The previous campaigns were valuable in showing the measurement capabilities of laboratories active in this measurement field. It is a field characterized by very different methods. For measuring the isotopic ratios inductively-coupled-plasma mass-spectrometry (ICP-MS) is of increasing importance, although thermal-ionisation mass-spectrometry (TIMS) is still employed. For certain uranium isotopes, mainly  $^{234}\text{U}$ , measurements of radioactivity by alpha-particle spectrometry (AS) are also employed by some laboratories although earlier NUSIMEP campaigns showed that this method cannot compete with mass-spectrometry in sensitivity and in the final levels of uncertainty.

Each NUSIMEP campaign up to now has had a matrix more complicated than the previous round. This is partly to approach more closely the problems encountered in natural samples and to show how they affect the measurement results. In NUSIMEP 1 (the only campaign with restricted participation) the uranium was in a simple (aquatic) matrix, in NUSIMEP 2 dry uranium nitrate samples were sent to participants and in NUSIMEP 3 a saline solution was used as matrix. In NUSIMEP 2 and 3, four samples were provided, each with different isotopic abundances for the isotopes specified ( $^{234}\text{U}$ ,  $^{235}\text{U}$ ,  $^{236}\text{U}$ ,  $^{238}\text{U}$ ). The measured values were more scattered for NUSIMEP 3 than for NUSIMEP 2, which was the result of a more complex matrix (3 % saline solution). The matrix for NUSIMEP 4 was a saline solution containing urea, to simulate urine.

For the present campaign, NUSIMEP 5, it was decided to retain the saline matrix, add uranium at the same level as for NUSIMEP 3 and 4 (5 ppb) and to add low levels of Pu and Cs isotopes. The concentrations of Pu and Cs were chosen to provide a realistic test for laboratories



and were kept low enough so that the samples could be sent to laboratories using standard transport carriers.

Plutonium and caesium materials certified for activity concentration were made available by the Radionuclide Group at IRMM and personnel of this group prepared solutions and certified the materials for this campaign. Laboratories were asked to complete a questionnaire when submitting results. The aim of the questionnaire was to show the influence of the laboratories' experience, of the procedure applied for sample preparation prior to measurements and of the techniques used for measurements on the results.

## 2. Samples

The details of the preparation of the samples can be found in the preparation report [1] sent to the participants after receipt of their results.

For this campaign, 4 solutions were prepared labelled NUSIMEP 5A, 5B, 5C and 5D. One litre of each solution was prepared; all solutions contained uranium, solutions 5A and 5B contained plutonium and solutions 5C and 5D caesium radionuclides.

To eliminate problems of loss of materials during transport, the solutions were filled into polyethylene transfer pipettes of 10 mL capacity. The necks of the pipettes were heat-sealed. Two 10 mL ampoules of each sample were sent to each laboratory. This method solved the problem of sample integrity: no laboratory reported losses of material during delivery. The method also allowed the laboratories some flexibility in using the sample material and the individual ampoules were also an acceptable container for those laboratories that performed gamma-ray-counting directly.

The 4 uranium materials were prepared by hydrolysing samples of  $UF_6$  that were selected from the large library of  $UF_6$  materials certified at IRMM for isotopic abundances. The natural uranium in the salt mixture used for the base of the matrix solutions was removed by passing a solution of the salts through EICHROM U-TEVA resins as described in the previous reports for NUSIMEP 3 and NUSIMEP 4 [2, 3]. The amount of the residual natural uranium in the matrix solutions was measured by IDMS using a high-enriched  $^{233}U$  spike (IRMM-058) and measuring the  $^{233}U/^{238}U$  ratio on a Thermo Electron Triton TIMS. The amount concentration of the natural uranium measured this way was in agreement with the value measured by ICP-MS on the matrix solution used for NUSIMEP 4 campaign, which confirmed the use of this value for small corrections to the uranium ratios in the 4 samples.

The Pu materials were chosen for the values of the alpha ratio  $^{238}Pu/(^{239}Pu + ^{240}Pu)$  as well as for the isotope ratios  $n(^{240}Pu/^{239}Pu)$ ,  $n(^{241}Pu/^{239}Pu)$  and  $n(^{242}Pu/^{239}Pu)$ . Two samples that had been used for a EUROCHEM campaign [4] in 1986 and which were therefore well certified, were diluted by weighing to a concentration of ca.  $1 \text{ kBq}\cdot\text{g}^{-1}$  solution in 1M nitric acid.  $50 \mu\text{L}$

aliquots from these two solutions were pipetted directly into the 1 L recipients containing matrix solutions and uranium for NUSIMEP 5A and NUSIMEP 5B respectively. This led to approximately 1 Bq total Pu activity per 20 mL sample.

Two Cs mixtures were made by weighing certified solutions of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  together to achieve two different activity ratios. The final activity concentration of these solutions was 1 kBq and 50  $\mu\text{L}$  aliquots of each were pipetted into solutions NUSIMEP 5C and NUSIMEP 5D yielding Cs activities of ca. 1 Bq per 20 mL sample.

### 3. Participation

Invitations to participate were sent to a number of laboratories. Registrations were received from 43 laboratories in 25 countries (Table 1).

**Table 1:** Countries of origin of participating laboratories.

	COUNTRY	No of registered participants	No of submitted results
1	Australia	5	5
2	Bosnia and Herzegovina	1	1
3	Bulgaria	1	1
4	Denmark	1	1
5	France	2	1
6	Germany	1	0
7	Greece	1	1
8	Hungary	1	1
9	Ireland	1	1
10	Israel	1	1
11	Italy	3	3
12	Japan	2	4
13	Korea	1	1
14	Latvia	1	1
15	Lithuania	1	1
16	Malaysia	1	1
17	Norway	1	0
18	Poland	2	2
19	Russian Federation	1	1
20	Serbia and Montenegro	2	2
21	Spain	4	4
22	Sweden	3	3
23	Switzerland	1	1
24	United Kingdom	2	1
25	United States	3	1
<b>25</b>	<b>total</b>	<b>43</b>	<b>39</b>

These laboratories were active in the fields of research and development, measurement of radioactivity in the environment, monitoring of nuclear facilities, medical applications, measurement of radionuclides in professionally exposed workers and measurement for fissile material control or safeguards. Forty five sets of samples were sent (one laboratory measured the samples using 3 different techniques) and 39 sets of results were received together with the questionnaire. A list of the laboratories that registered and submitted results is given in Annex 1.

Unfortunately 2 of the registered laboratories reported problems with their mass-spectrometers that could not be resolved before the final deadline for reporting results.

Six of the participating laboratories had already participated in previous NUSIMEP campaigns. Twenty seven participating laboratories reported that they participated in other inter-laboratory campaigns organised internationally or nationally by IAEA, NIST, US/DOE or other national organisations. Six participating laboratories reported this was their first participation in an inter-laboratory comparison.

#### 4. Results and discussion

The participating laboratories were requested to measure and report the isotopic amount ratios  $n(^{234}\text{U})/n(^{238}\text{U})$ ,  $n(^{235}\text{U})/n(^{238}\text{U})$ ,  $n(^{236}\text{U})/n(^{238}\text{U})$ ,  $n(^{238}\text{Pu})/n(^{239}\text{Pu})$ ,  $n(^{240}\text{Pu})/n(^{239}\text{Pu})$ ,  $n(^{241}\text{Pu})/n(^{239}\text{Pu})$ ,  $n(^{242}\text{Pu})/n(^{239}\text{Pu})$  or alternatively the activity ratio  $^{238}\text{Pu}/(^{239}\text{Pu}+^{240}\text{Pu})$  as well as the activity ratio  $^{134}\text{Cs} / ^{137}\text{Cs}$ , using methods of their own choice.

The laboratories were free to measure any or all of the parameters. The numbers of submitted results per sample and ratio are shown in Table 2.

**Table 2:** Number of submitted results per sample and ratio.

Ratio	A	B	C	D
$n(^{234}\text{U})/n(^{238}\text{U})$	16 (4)	18 (12)	21 (11)	21 (14)
$n(^{235}\text{U})/n(^{238}\text{U})$	20 (17)	21 (19)	19 (17)	22 (19)
$n(^{236}\text{U})/n(^{238}\text{U})$	6 (1)	6 (0)	9 (4)	13 (6)
$n(^{238}\text{Pu})/n(^{239}\text{Pu})$	4 (4)	4 (4)		
$n(^{240}\text{Pu})/n(^{239}\text{Pu})$	13 (12)	12 (10)		
$n(^{241}\text{Pu})/n(^{239}\text{Pu})$	8 (4)	7 (4)		
$n(^{242}\text{Pu})/n(^{239}\text{Pu})$	8 (2)	9 (1)		
$^{238}\text{Pu}/(^{239}\text{Pu}+^{240}\text{Pu})$	21 (20)	21 (20)		
$^{134}\text{Cs}/^{137}\text{Cs}$			23 (14)	23 (10)

Values lying within  $\pm 10\%$  deviation from the reference value were considered to be a “good result” and the number of results which are in this range is given in brackets.

Laboratories applied alpha-particle spectrometry (AS), gamma-ray spectrometry (GS), inductively-coupled-plasma mass spectrometry (ICP-MS), thermal ionisation mass spectrometry (TIMS) and accelerator mass spectrometry (AMS) as measurement methods. A combination of AS and TIMS, a combination of AS and ICP-MS and a combination of liquid scintillation counting (LSC) and ICP-MS were used by laboratories to measure  $^{238}\text{Pu}$ . More detailed information is shown in Table 3:

**Table 3:** Techniques applied for NUSIMEP 5 sample measurements.

Technique	Uranium	Plutonium	Caesium
AS (activity ratio)		21	
AS (isotopic ratio)	8		
GS (activity ratio)			23
GS (isotopic ratio)	1		
AMS	1	1	
TIMS	2	2	
ICP-MS	17	10	
Quadrupole	6	2	
Sector field	1	1	
Multi collector	5	2	
High resolution	5	5	
AS + TIMS		1	
AS + HR-ICP-MS		3	
LSC + HR-ICP-MS		1	

Results from measurements performed at IRMM are not included in the statistics and discussions presented here.

The deviation of the submitted results from the reference values show a high degree of scattering for some ratios although the majority of the laboratories achieved acceptable agreement with the reference values. The measured values are plotted for each isotopic ratio and sample as well as for activity ratio for plutonium ( $^{238}\text{Pu}/^{239+240}\text{Pu}$ ) and caesium in Figs. 1- 24. A scale of  $\pm 50\%$  was chosen for most of the graphs, except for the  $n(^{235}\text{U})/n(^{238}\text{U})$ ,  $n(^{238}\text{Pu})/n(^{239}\text{Pu})$ ,  $n(^{240}\text{Pu})/n(^{239}\text{Pu})$ ,  $n(^{241}\text{Pu})/n(^{239}\text{Pu})$  and  $^{238}\text{Pu}/^{239+240}\text{Pu}$  ratios where a scale of  $\pm 25\%$  was found to be more appropriate and for  $^{234}\text{U}/^{238}\text{U}$  for sample A where a log plot is also shown.

The number of submitted results for each measurement technique for uranium isotope ratios is given in Table 4, for plutonium in Table 5 and for activity ratios of plutonium and caesium in Table 6. The number of results for each technique lying within the range of  $\pm 10\%$  deviation from the reference value is given in brackets.

Abbreviations used in the tables 4 - 6:

AS: alpha-particle spectrometry,

GS: gamma-ray spectrometry,

AMS: accelerator mass-spectrometry,

TIMS: thermal ionisation mass-spectrometry,

ICP-MS: inductively coupled plasma mass-spectrometry,

LSC: liquid-scintillation spectrometry.

**Table 4:** Number of results submitted per technique for the uranium isotopic ratios.

Ratio	Sample	AS	AMS	TIMS	ICP-MS	GS
$n(^{234}\text{U})/n(^{238}\text{U})$	A	5 (0)		2 (1)	9 (4)	
	B	6 (3)		2 (2)	10 (7)	
	C	7 (1)		2 (2)	11 (8)	1 (0)
	D	7 (2)		2 (2)	11 (9)	1 (0)
$n(^{235}\text{U})/n(^{238}\text{U})$	A	1 (1)		2 (2)	17 (13)	
	B	2 (2)		2 (2)	17 (15)	
	C			2 (2)	17 (15)	
	D	2 (1)		2 (2)	17 (16)	1(0)
$n(^{235}\text{U})/n(^{238}\text{U})$	A		1 (0)	1 (0)	4 (1)	
	B		1 (0)	2 (0)	4 (0)	
	C		1 (1)	2 (1)	6 (2)	
	D		1 (0)	2 (1)	10 (5)	

**Table 5:** Number of results submitted per technique for plutonium isotopic ratios.

Ratio	Sample	AMS	TIMS	ICPMS	AS + TIMS	AS + ICPMS	LSC + ICPMS
$n(^{238}\text{Pu})/n(^{239}\text{Pu})$	A				1 (1)	3 (3)	
	B				1 (1)	3 (3)	
$n(^{240}\text{Pu})/n(^{239}\text{Pu})$	A	1 (1)	2 (2)	10 (9)			
	B	1 (1)	2 (2)	9 (7)			
$n(^{241}\text{Pu})/n(^{239}\text{Pu})$	A	1 (1)	2 (1)	4 (2)			1 (0)
	B	1 (0)	2 (2)	3 (2)		1 (0)	
$n(^{242}\text{Pu})/n(^{239}\text{Pu})$	A		2 (1)	6 (1)			
	B	1 (0)	2 (0)	6 (1)			

**Table 6:** Number of results submitted for plutonium and caesium activity ratios.

<b>Sample</b>	$^{238}\text{Pu}/(^{239}\text{Pu}+^{240}\text{Pu})$ <b>AS</b>	$^{134}\text{Cs} / ^{137}\text{Cs}$ <b>GS</b>
A	21 (20)	
B	21 (20)	
C		23 (14)
D		23 (10)

All submitted values for a given measurement method are plotted with expanded uncertainty ( $k=2$ ) as declared by the laboratory (in some cases, following the laboratory's declaration, results with other  $k$  values were converted to  $k=2$  before plotting). A simple rule is that the measured value agrees with the certified value if this latter is included in the uncertainty of the measurement. Verifying measurements performed at IRMM are plotted as laboratory 20 on the graphs.

#### **4.1. Results of measurements of individual ratios**

##### **4.1.1. $n(^{234}\text{U})/n(^{238}\text{U})$ (Figures 1 – 4)**

The certified ratios for  $n(^{234}\text{U})/n(^{238}\text{U})$  lie between  $6.45 \text{ E-}6$  (sample A) and  $1.83 \text{ E-}4$  (sample D). Only four of the laboratories applying ICP-MS did not separate the uranium before measuring. The results for this ratio from these laboratories are characterised by significant biases relative to the certified values or larger uncertainties, especially for the first two samples with low abundances of  $^{234}\text{U}$ . Otherwise the power of the mass-spectrometry methods is clearly seen, with most of the laboratories supplying excellent results.

The measurements by alpha-particle-spectrometry are quite puzzling. For the low  $^{234}\text{U}$  abundances in samples A and B a strong positive bias is found generally for all the results obtained by alpha ratio measurements. In samples C and D, where the abundances are much higher (ratios relative to  $^{238}\text{U}$  are  $1.83 \cdot 10^{-4}$  for sample C and  $2.94 \cdot 10^{-4}$  for sample D) a negative bias is observed for the alpha-particle spectrometry measurements. The strong clustering in these measurement results implies that there is a common interference endemic to the measurements. However each laboratory applied its own favoured purification and measurement techniques and a common factor has not yet been found to explain the biases.

##### **4.1.2. $n(^{235}\text{U})/n(^{238}\text{U})$ (Figures 5 - 8)**

The ratio  $n(^{235}\text{U})/n(^{238}\text{U})$  is one of the most critical ratios for tracking the source of nuclear material in environmental samples and one that laboratories would expect to measure well. The 4 samples covered the range of depleted (0.2%) to enriched (3%) and included one sample with

a natural uranium abundance. Alpha-particle spectrometry was applied only in the case of the 3% enriched material (sample D) and for this sample the submitted results had comparatively high uncertainties because of the low alpha activity of the samples.

The mass-spectrometry measurements were generally well performed, showing little or no bias. Two laboratories did no separation before measurements. The results from these laboratories, however, showed high positive biases.

Noticeable in the case of several of the results is that the stated uncertainties are too low, simply because the certified values are not covered by the uncertainties of the submitted results. However, many laboratories clearly make good estimations of their measurement uncertainties and in general we can place a lot of confidence in the measurements of this ratio by all mass-spectrometry techniques applied in this campaign.

#### **4.1.3. $n(^{236}\text{U})/n(^{238}\text{U})$ (Figures 9 - 12)**

The  $^{236}\text{U}$  abundance for samples A and B was  $< 10^{-6}$  and for these two samples only 6 laboratories submitted results and 4 of these in each case were outside the plotted range (Figures 9, 10). The  $^{236}\text{U}$  abundance for sample C was higher:  $3 \cdot 10^{-6}$  and results of 7 laboratories from 9 that submitted results agreed with the certified values. For sample D,  $^{236}\text{U}$  abundance =  $3.3 \cdot 10^{-5}$ , 13 laboratories submitted results and 9 of those results agreed with the certified value. The values from three other laboratories were submitted with uncertainties that were clearly too low. If these had been more realistically estimated, these laboratories could easily have had results agreeing with the certified value

One laboratory measured using accelerator mass-spectrometry. This was one of only two laboratories that consistently measured  $^{236}\text{U}$  down to the lowest  $^{236}\text{U}/^{238}\text{U}$  ratio in these samples.

We can conclude from the results that with present mass-spectrometer techniques isotopic ratios as low as  $3 \cdot 10^{-6}$  can be measured with a good degree of reliability on samples of uranium of this size (maximum amount of uranium per sample = 100 ng). However the measurement of very small ratios seems to be a hard test and this is illustrated by the number of laboratories that submitted results decreased when the isotopic ratio was lower than  $1 \cdot 10^{-6}$ .

#### **4.1.4. Pu isotopic ratios (Figures 13 - 20)**

Samples A and B each had approximately 1 Bq plutonium added (~ 0.75 ng per sample). The measured plutonium isotopic ratios relative to  $^{239}\text{Pu}$  are shown in figures 13 - 20. Four laboratories submitted results for the ratio  $n(^{238}\text{Pu})/n(^{239}\text{Pu})$  for each sample: the results agree within 5 % of the certified value. In all cases the values were calculated after measuring the ratio  $^{238}\text{Pu}/(^{239}\text{Pu}+^{240}\text{Pu})$  by alpha-particle-spectrometry and the  $n(^{240}\text{Pu})/n(^{239}\text{Pu})$  ratio by TIMS or ICP-MS.

The isotopic ratio  $n(^{240}\text{Pu})/n(^{239}\text{Pu})$  is the most straightforward to measure by mass-spectrometry and at the same time the most used for identifying sources of Pu in the environment. Laboratories did not appear to have a problem measuring this ratio: only one laboratory submitted a value of the ratio that had a greater than 25 % deviation from the reference value. Mostly excellent measured results were submitted and it appears that for this level of Pu concentration laboratories have the measurement methods well under control.

It is an indication that a lower concentration of Pu would be better to test measurements capabilities in real samples. This is also borne out by the alpha ratio measurements reported below.

The isotopic ratio  $n(^{241}\text{Pu})/n(^{239}\text{Pu})$  was only measured by 9 laboratories for sample A and 8 for sample B. It is noticeable that the obvious outliers all had an extreme positive bias, which could indicate poor separation of  $^{241}\text{Am}$  before measurement.

Although the  $n(^{242}\text{Pu})/n(^{239}\text{Pu})$  ratios were the same order of magnitude as the  $n(^{241}\text{Pu})/n(^{239}\text{Pu})$  ratios of these samples, fewer laboratories managed to measure these ratios successfully and several submitted values with a very high positive bias relative to the certified values. No technical reason has been suggested for this bias. In all these measurements a chemical separation was performed before mass-spectrometry.

#### **4.1.5. Pu alpha ratio $^{238}\text{Pu}/(^{239}\text{Pu}+^{240}\text{Pu})$ (Figures 21 and 22)**

Twenty one laboratories submitted results for this ratio in the two samples A and B; only one of the results had a value with a deviation greater than 25 % of the certified value. The results are characterised in general by the excellent agreement, most of them lying within  $\pm 5\%$  of the certified values. This underlines the observation made in 4.1.4 above that a lower Pu concentration in samples in future campaigns would provide a more realistic test for laboratories.

#### **4.1.6. $^{134}\text{Cs}/^{137}\text{Cs}$ ratios (Figures 23 and 24)**

Solutions C and D both had a (different) mixture of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  nuclides added: about 1 Bq per sample. Twenty three laboratories submitted results. Nearly all laboratories measured samples activity/ratios directly by gamma-ray-spectrometry; only two laboratories separated Cs before measurement. The results agreed well with the certified values although there appears to be a slight negative bias for sample D. This could possibly be attributed to the coincidence summing correction applied to the  $^{134}\text{Cs}$  gamma-ray peaks or to the detector efficiency calibration. But it appears that laboratories could measure the gamma activity of these samples and correct for sum-peaks (for  $^{134}\text{Cs}$ ) at levels of the radionuclides that are not unusual in environmental samples and at activity ratios between 0.06 and 0.15. Again, as for the Pu concentration levels, it could be stated that lower activities would be a harder and more realistic test of measurement capabilities for laboratories.



## 5. Evaluation of questionnaire

A questionnaire (Annex 2) was a part of the result reporting page and participating laboratories were required to fill it in when sending results.

The questionnaire was divided into three sets of questions:

- about the laboratory, the types of samples normally measured and use of reference materials;
- about details of the uranium, plutonium and caesium separations;
- about measurement procedures and reporting results.

The questions and the answers of the 39 participating laboratories are summarised below.

As the mission of the laboratory most of the laboratories (26 out of 39) declared activity in more than one area; 29 laboratories reported that their mission is research and development, 28: measurements of the radioactivity in the environment, 9: monitoring of nuclear facilities, 8: measurements for fissile material control or safeguards, 3: medical applications. One laboratory reported that one of its main activities was the measurement of professionally exposed workers, and another that is responsible for calibration of radiation equipment.

Eleven participants reported that their laboratories are neither 'certified', 'accredited' nor 'authorised'. Two of the participating laboratories declared that they are 'certified', 'accredited' and 'authorised', and of the other laboratories 2 were 'certified', 5 'accredited' and 14 'authorised'. Three reported that they are 'certified' and 'accredited' and two are 'accredited' and 'authorised'.

Twenty laboratories stated they are working within a quality management system. Most of these laboratories are following ISO 17025 (16), and ISO 9000 (4). Two laboratories reported that they follow GLP procedures according to national standards and internally developed laboratory and specific quality assurance protocols.

Most laboratories (34) have already participated in inter-laboratory comparisons. Only 6 laboratories reported that the NUSIMEP 5 campaign was their first inter-laboratory comparison.

The number of samples of a similar type as the NUSIMEP samples that the laboratories process per year is shown in Table 7.

**Table 7:** Number of samples analysed per year.

Samples of this type per year	Number of laboratories		
	U	Pu	Cs
No response	4	5	12
< 25	12	14	4
25 – 50	10	10	7
50 – 100	3	2	5
< 100	10	8	11

The laboratories were asked which types of samples they regularly analysed. Soil, sediments, biota, marine samples, waters (drinking, surface, sea, underground), food, urine, faeces, hair, air filters as well as radioactive liquid waste, spent nuclear fuel and safeguard swipe samples were the normal types of sample analysed.

Indicating the level of experience for this type of analysis 30 laboratories judged themselves to be experienced, 7 to be less experienced and 2 of them judged themselves as not-experienced.

31 laboratories used one or more certified reference materials for calibration and validation of their instruments and methods, mainly supplied by IRMM, NIST and IAEA.

32 laboratories reported that the NUSIMEP 5 samples were treated according to the same analytical procedures routinely used for this sample types.

15 laboratories reported that sample preparation and/or analysis were carried out in the clean room or laboratory specifically designed. 24 laboratories reported that they performed separation and measurements under normal measurements conditions.

Radiochemical separations were carried out by all laboratories before measuring plutonium isotope ratios. The separations were done by liquid-liquid extraction, ion exchange chromatography (Dowex, BioRad, Diaion) as well as extraction chromatography (TEVA, U-TEVA, TRU Eichrom resins). Most laboratories separated uranium from matrix interferences, although a few described direct measurement after a dilution step.

The following measurements techniques were applied: direct gamma-ray spectrometry, alpha-particle spectrometry, AMS, TIMS, Q-ICPMS, MC-ICPMS, HR-ICPMS as well as combinations of AS or LSC and ICP-MS.

For the alpha-particle spectrometry measurements alpha sources were prepared by electro-deposition or micro-coprecipitation. Only one laboratory reported using a drop-deposition technique.

Direct gamma-ray spectrometry was used for the determination of caesium nuclide activities in most cases, although 2 laboratories reported that they performed a separation prior to measurement. Most of the laboratories adjusted the volume of the solutions to standard volumes for their counting systems and some used dried sources.

Twenty eight participants reported that they are familiar with the Guides for Quantifying Measurement Uncertainty issued by the International Organisation for Standardisation and EURACHEM [5, 6].

All laboratories, except one, reported results with measurement uncertainties. Nineteen participants reported expanded uncertainties as requested and 6 reported standard uncertainties. Fourteen laboratories based their uncertainty values on the statistical evaluation of repeated measurements, uncertainties on control samples and reference materials as well as counting statistics and relative standard determination of the instrument. In the result reporting page the uncertainty budget was not requested.

All laboratories expressed an interest in future NUSIMEP campaigns. A number of future materials or matrices for NUSIMEP rounds were proposed including:

- Environmental (soil, sediments, biota, water, foodstuffs, marine)
- Air filters, cotton swipes, particulates
- Urine, human hair
- Building rubble, geological samples
- Matrices with components that interfere with U and Pu measurements.

The isotopes suggested for future campaigns covered a wide range. One group included the gamma-ray emitters  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$ , Ra isotopes,  $^{110\text{m}}\text{Ag}$ ,  $^{210}\text{Pb}$  as well as  $^{89}\text{Sr}$  and  $^{90}\text{Sr}$ . Other participants suggested natural radioactive elements including tritium. Several participants requested the actinides – U, Pu, Th, Np, Am and Cm - and others specified isotopic ratios such as  $^{235}\text{U}/^{238}\text{U}$ ,  $^{241}\text{Am}/^{243}\text{Am}$ .

Participants were also asked if and which mixtures of radionuclides they would like to receive in future samples. A wide range of mixtures was specified in answers. In some cases simply U and Pu were mentioned, in others specific mixtures such as Th/U/Pu or Pu/Am/Np. Mixtures of gamma-ray-emitters were also requested. But many of the participants were not specific and stated they would be content with any mixture offered. The concentration levels requested were all low and typical of environmental samples: between mBq to Bq for gamma and alpha emitting isotopes, U below 1 ppb and Pu at levels down to 1 ppt.

## 6. Conclusions

This inter-laboratory comparison exercise was carried out successfully with a larger response from laboratories than expected. Last minute requests for participation had to be refused because the sample solutions were exhausted.

The possibility to measure alpha and gamma ratios brought in a number of laboratories that concentrate on the measurement of these parameters. However the exercise was designed so that non-destructive measurements of the Cs isotope ratios could be done first and then followed by separations and measurements by alpha-particle or mass-spectrometry for measuring other requested ratios.

It was concluded that most laboratories found the measurements of radioactivity, either by alpha-particle- or gamma-ray-spectrometry, well within their capabilities and a conclusion is that in future campaigns we should reduce the levels of Pu and Cs, for instance. Laboratories generally measured the isotopic ratios of uranium and plutonium well. An exception was the measurements of the minor isotope ratios of both uranium and plutonium. Very few laboratories can measure the  $n(^{236}\text{U})/n(^{238}\text{U})$  ratio reliably for  $^{236}\text{U}$  abundances  $< 10^{-6}$ . Large deviations, sometimes of several hundred percent of the certified values, were observed in the results submitted by several laboratories for the minor isotopic ratios of plutonium:  $n(^{241}\text{Pu})/n(^{239}\text{Pu})$  and  $n(^{242}\text{Pu})/n(^{239}\text{Pu})$ . The reason for this is not clear.

The alpha activity ratio  $^{238}\text{Pu}/(^{239}\text{Pu}+^{240}\text{Pu})$  and the gamma-ray activity ratio  $^{134}\text{Cs}/^{137}\text{Cs}$  were also measured well by the great majority of laboratories. Many laboratories also measured the alpha activity ratios for uranium and converted these values into isotopic ratios for the report. Some puzzling behaviour was noted in particular with the alpha ratio  $^{234}\text{U}/^{238}\text{U}$ , where the results from the alpha activity measurements were consistently biased relative to the certified and the mass-spectrometry values. Alpha activity ratios are regularly measured on a routine basis in many countries and these results suggest a systematic effect – possibly background subtraction in the spectra - that is not properly taken into account.

It was gratifying to see how well this latest NUSIMEP campaign was received by the participating laboratories. There have been many requests to continue with similar samples. It appears that such campaigns can play an important role for laboratories in assuring their measurement capabilities and meeting the requirements of the quality management systems.

A common parameter, already remarked on above, was that for many laboratories the concentrations of all the elements were higher than they normally receive in routine samples. This was clear in the excellent agreements between laboratories and with the certified values in all measurement areas.

The possibilities of reducing the concentration of the elements to be measured are presently being investigated at IRMM. For the uranium the value of the blanks arising from solutions,

labware, handling and the saline matrix is a limiting factor because, although the correction for the blank is relatively small for the present samples, this could be the determining factor for the uncertainties of the certified isotope ratio values if the uranium concentrations are lowered by a factor of 10 or even 100. Blank corrections are not a problem for Pu and Cs and in subsequent campaigns it is envisaged to lower the concentrations of these elements towards values found in environmental samples. A greater problem is to obtain suitable, well certified starting materials in particular for plutonium.

## 7. References

- [1] L.Benedik, A. Alonso, T.Altizoglou, S.Richter, G.Sibbens, A.Stolarz, A.Verbruggen, R.Wellum *Preparation of samples for the NUSIMEP 5 campaign containing uranium, plutonium and caesium certified for isotopic abundances*; EUR Report 22180 EN
- [2] A. Held, A. Alonso, W. De Bolle, A. Verbruggen, R. Wellum, *NUSIMEP-3: Uranium isotopic abundances in saline solutions, Report to participants*, EUR Report 20377
- [3] A. Stolarz, A. Alonso, W. De Bolle, A. Moens, E. Ponzevera, C. Quétel, S. Richter, A. Verbruggen, R. Wellum; *NUSIMEP 4: Uranium isotopic abundances in simulated urine; Report to participants*; EUR Report 21839 EN
- [4] G. Bortels, A. Verbruggen, G. Sibbens, T. Altizoglou, 1996. *EUROMET Project No 325: Analysis of Plutonium Alpha-Particle Spectra*. Internal report GE/R/RN/01/96
- [5] International Organisation for Standardisation, "*Guide to the Expression of Uncertainty in Measurement*", ©ISO, ISBN 92-67-10188-9, Geneva, Switzerland, 1993.
- [6] EURACHEM, "*Quantifying Uncertainty in Analytical Measurement*", ISBN 0-948926-08-2, ©Crown copyright, 1995; LGC Information Services, Queens Road, Teddington, Middlesex, TW11 0LY, England.

## 8. Participants' results for all measured parameters

The results submitted by participating laboratories are presented in the form of graphs. Two different scales of  $\pm 25\%$  and  $\pm 50\%$  deviation from reference value are applied depending on the scatter of the results. A log plot is also shown for clarification for  $n(^{234}\text{U})/n(^{238}\text{U})$  in sample A to show the unexpected deviations of alpha spectrometry measured results.

The range of the certified reference value is shown on each graph by lines at 'certified value – uncertainty' and at 'certified value + uncertainty' (expanded uncertainties,  $k=2$ ). The dotted lines mark the range of  $+10\%$  and  $-10\%$  from reference value chosen in this report as the criterion of good result.

Results lying outside the plotted range are indicated in the figures. More detailed information about their values, deviation from reference value and technique used for measurements are given in table 8 at the end of the chapter.

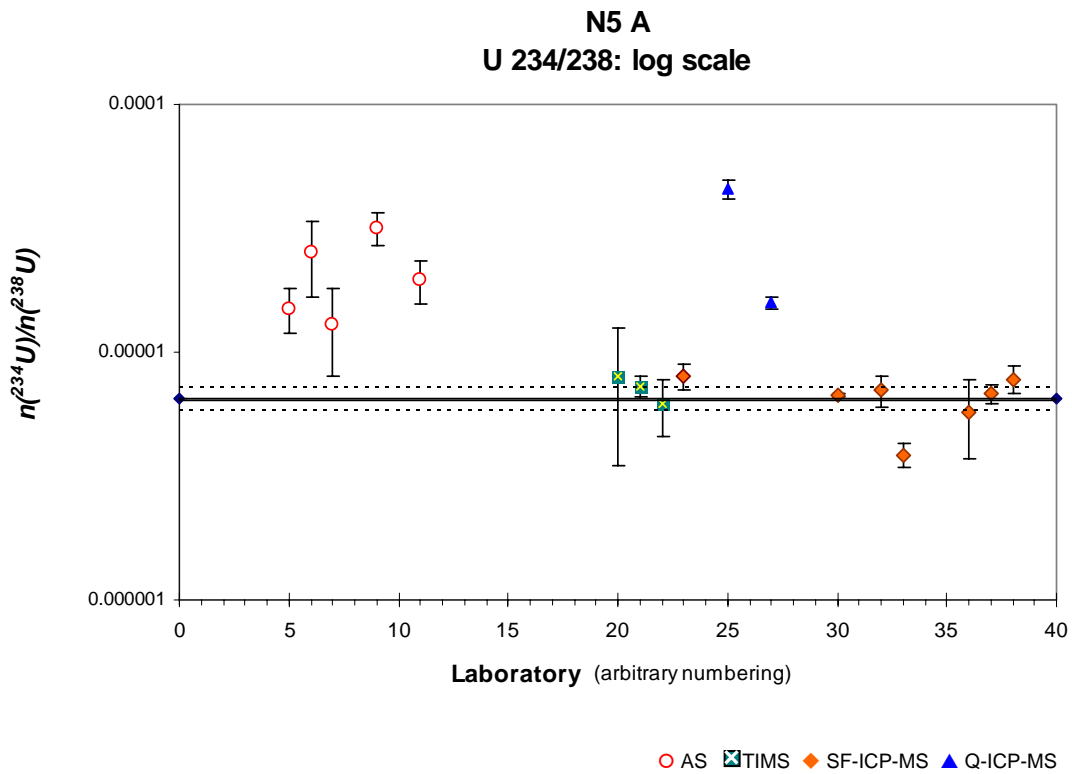
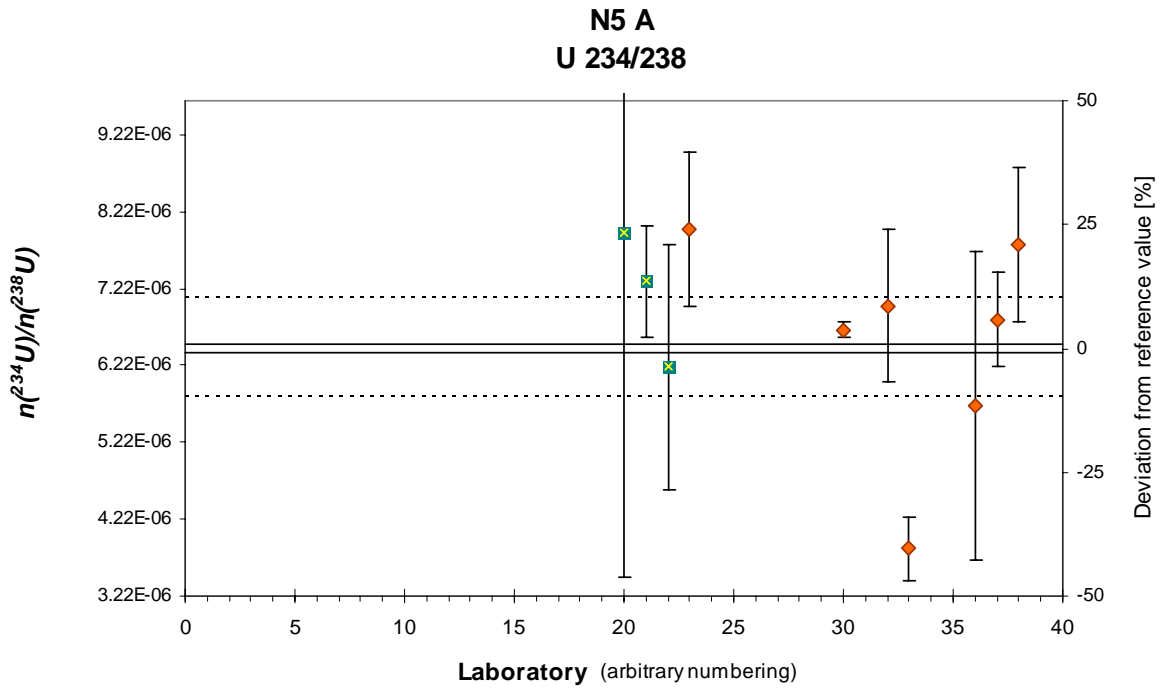
The results have been sorted so that the Laboratories which applied alpha-particle spectrometry (AS) and gamma-ray spectrometry (GS) have the lowest numbers, followed by laboratories using TIMS and then ICP-MS techniques.

The following abbreviations are used for the figures and in Table 8:

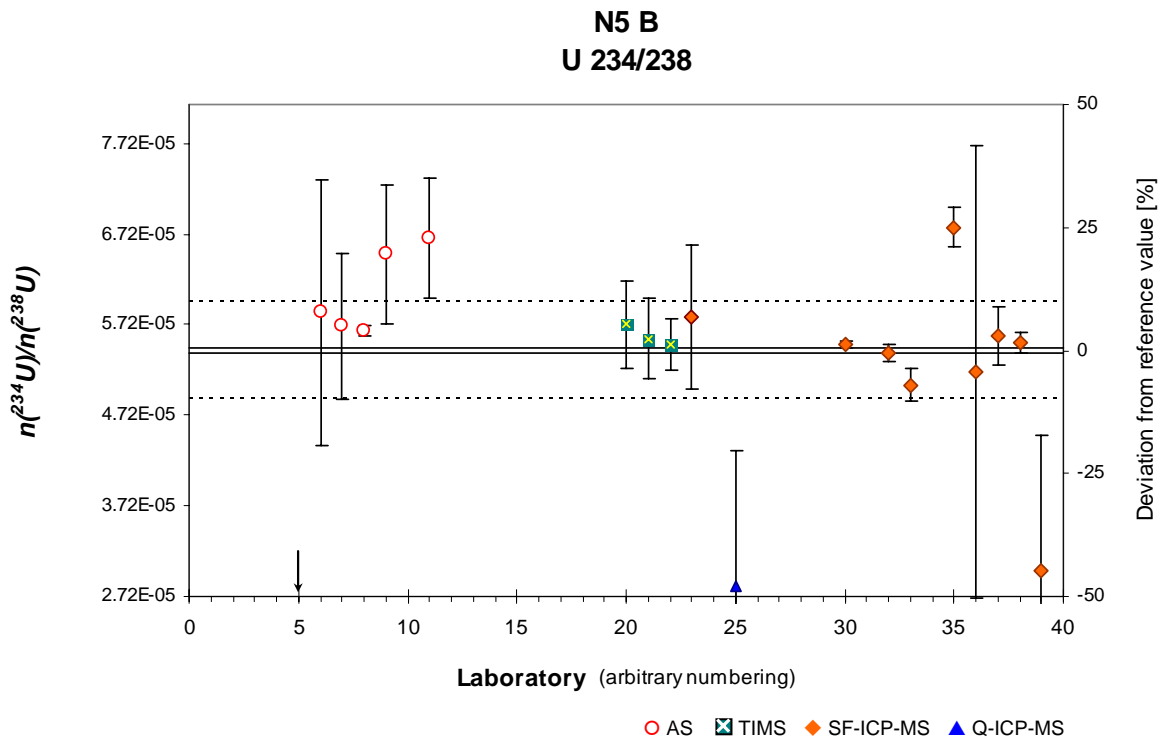
AS	alpha-particle spectrometry
GS	gamma-ray spectrometry
TIMS	thermal ionisation mass-spectrometry
Q-ICP-MS	quadrupole inductively-coupled-plasma mass-spectrometry
SF-ICP-MS	sector-field inductively-coupled-plasma mass-spectrometry
MC-ICP-MS	multi-collector inductively-coupled-plasma mass-spectrometry
AMS	accelerator mass-spectrometry
AS+TIMS	alpha spectrometry + thermal ionisation mass-spectrometry
AS+SF-ICP-MS	alpha spectrometry + sector-field inductively-coupled-plasma mass-spectrometry
LSC	liquid-scintillation spectroscopy

The results marked by  $\blacklozenge$  for SF-ICP-MS include the categories: Sector Field (SF-ICP-MS), Multi-collector (MC-ICP-MS) and High-resolution ICP-MS (HR-ICP-MS).

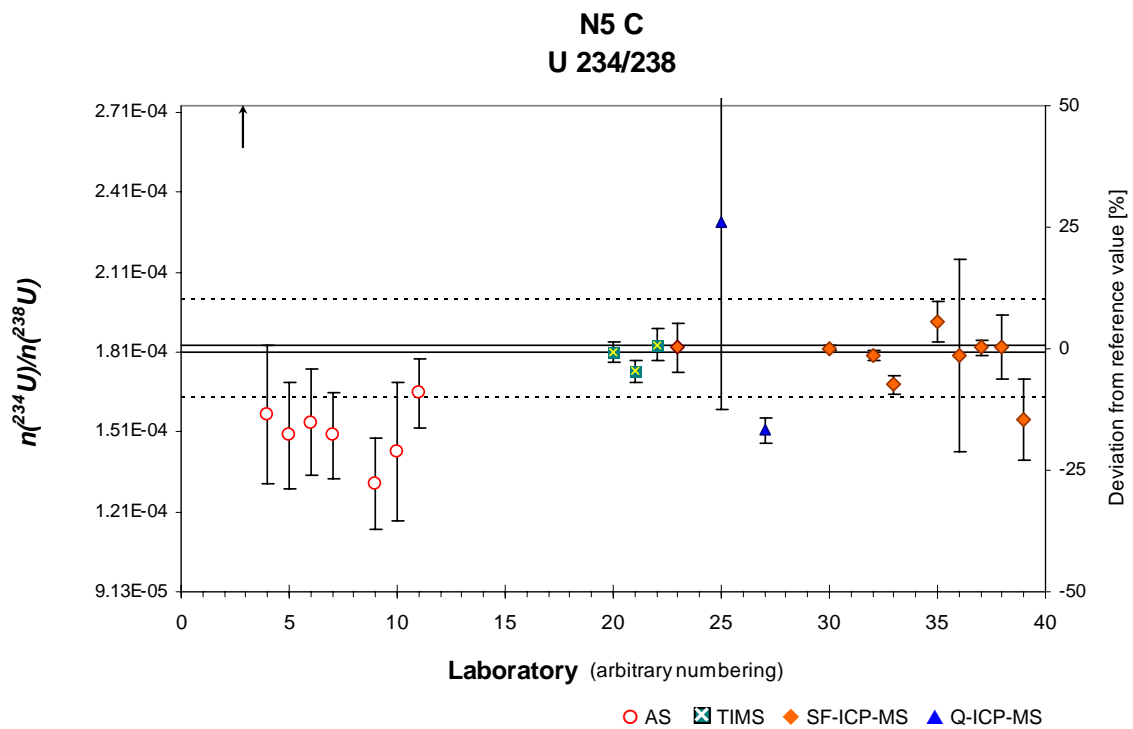
The arrow  $\uparrow$  or  $\downarrow$  on the plot indicates a result whose value lies outside the plotted area.



**Figure 1:** Submitted results for  $n(^{234}\text{U})/n(^{238}\text{U})$  in sample A normal scale (top) and logarithmic scale (bottom)



**Figure 2:** Submitted results for  $n(^{234}\text{U})/n(^{238}\text{U})$  in sample B



**Figure 3:** Submitted results for  $n(^{234}\text{U})/n(^{238}\text{U})$  in sample C



N5 D  
U 234/238

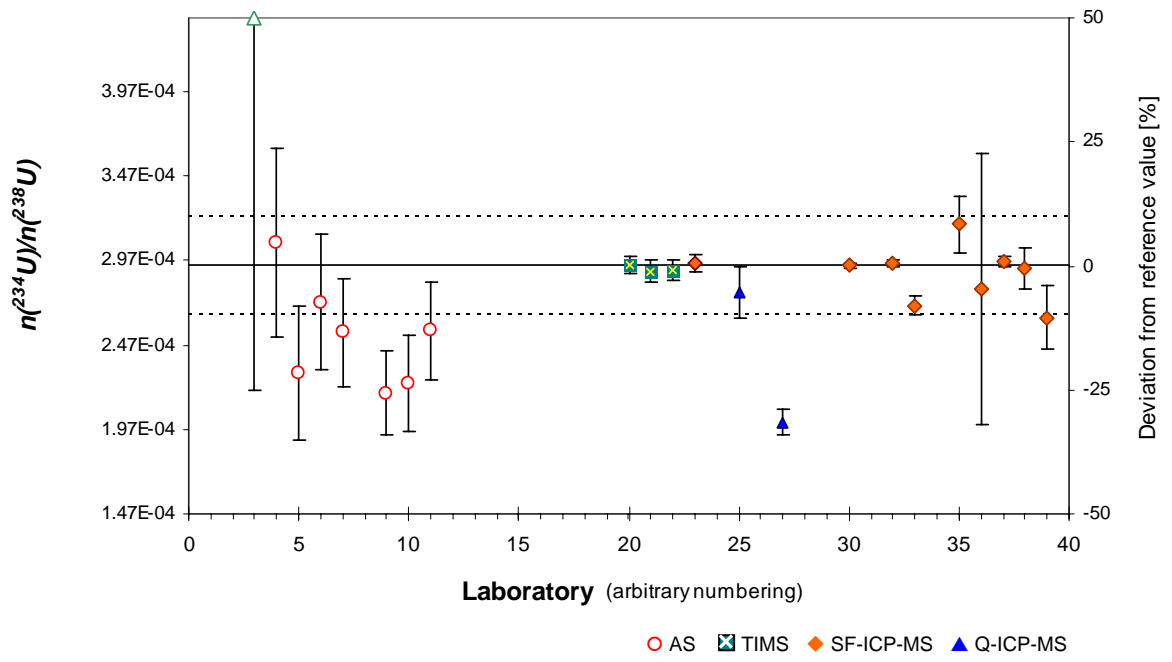
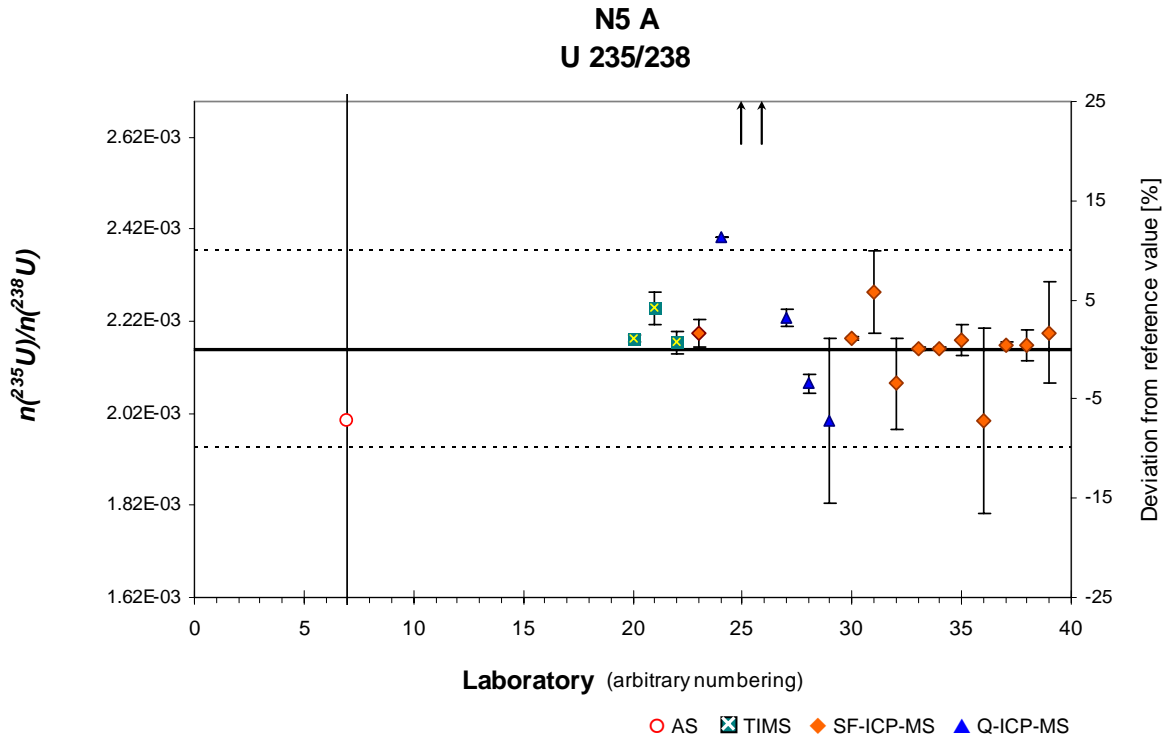
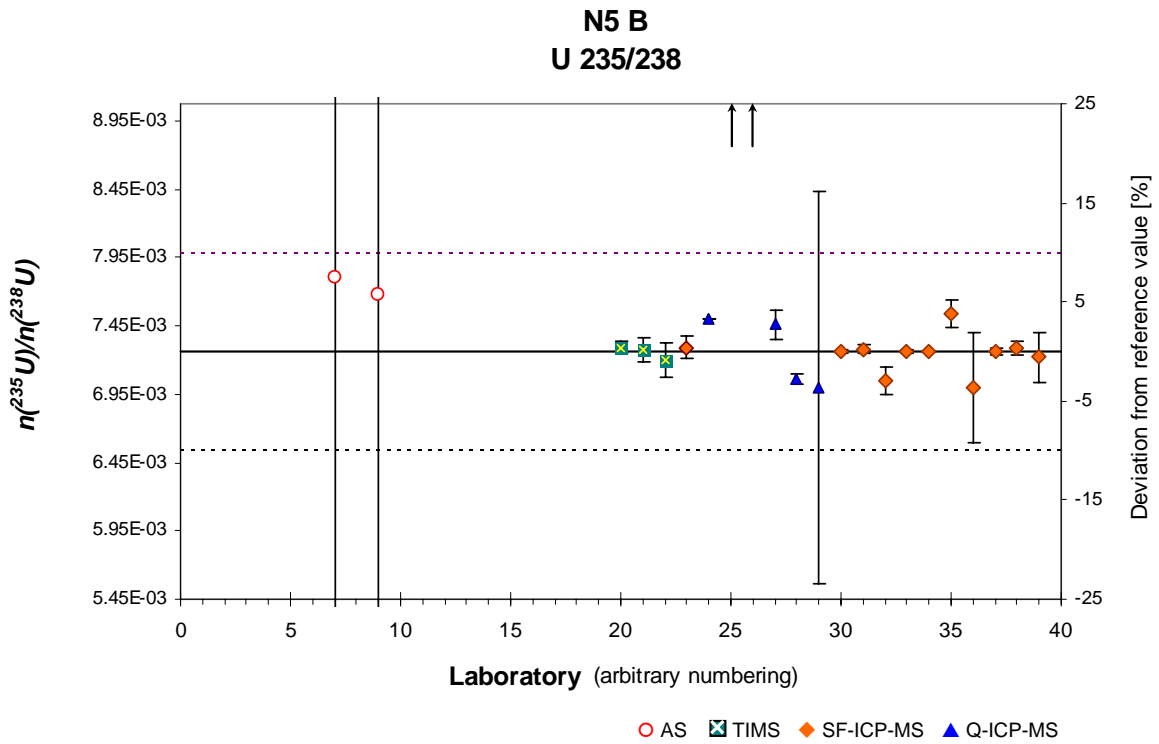


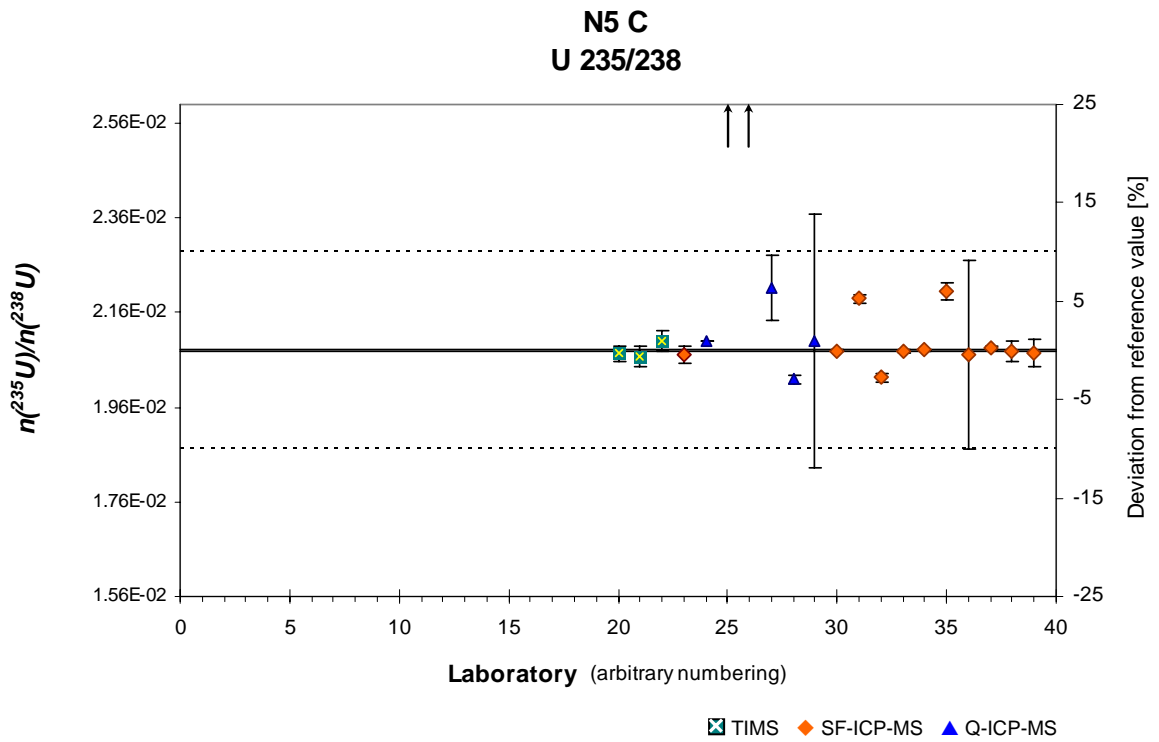
Figure 4: Submitted results for  $n(^{234}\text{U})/n(^{238}\text{U})$  in sample D



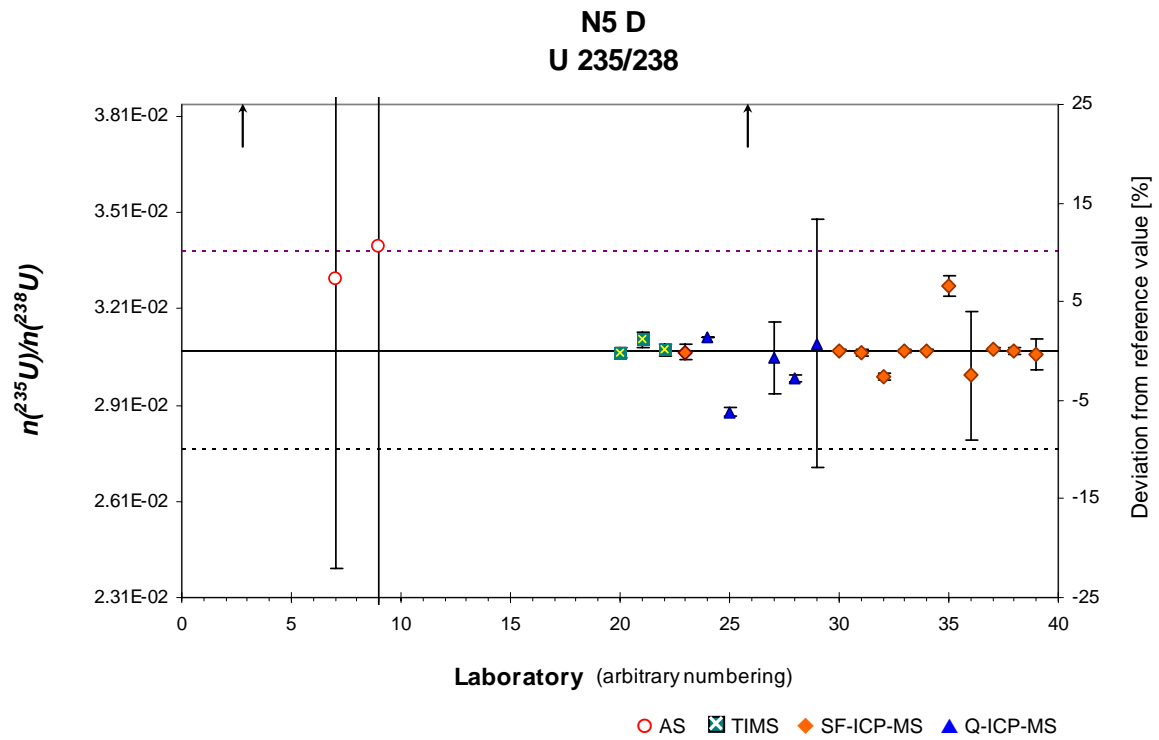
**Figure 5:** Submitted results for  $n(^{235}\text{U})/n(^{238}\text{U})$  in sample A



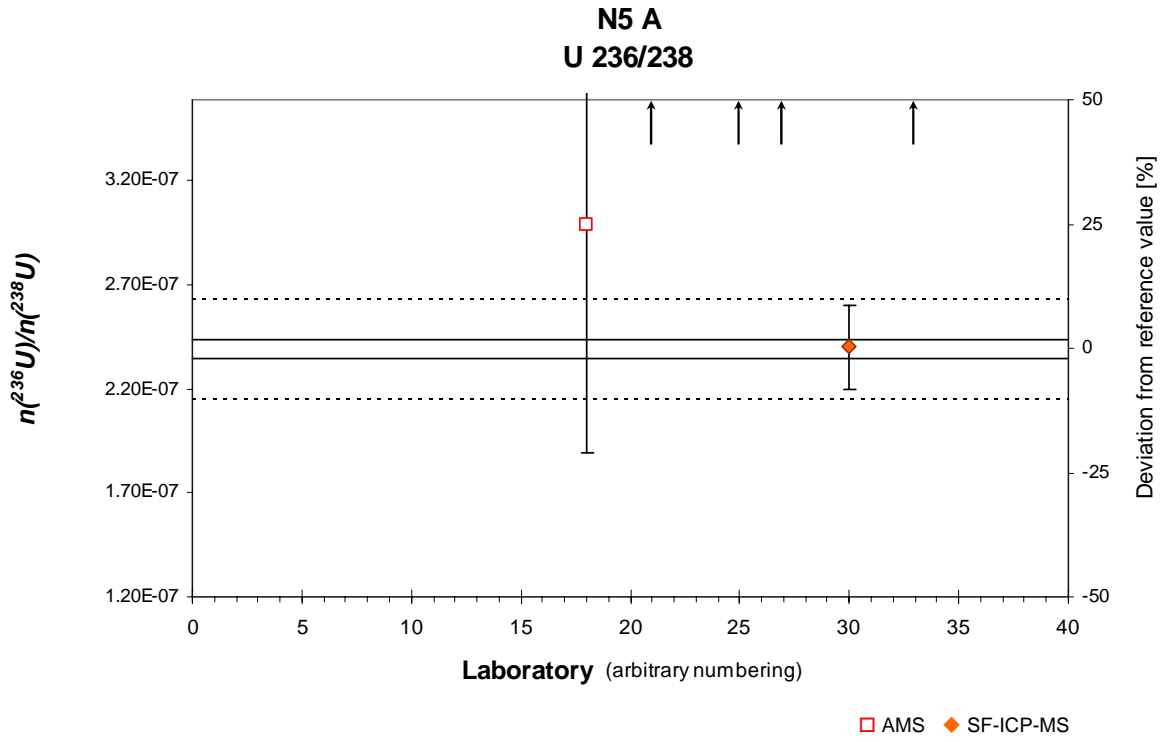
**Figure 6:** Submitted results for  $n(^{235}\text{U})/n(^{238}\text{U})$  in sample B



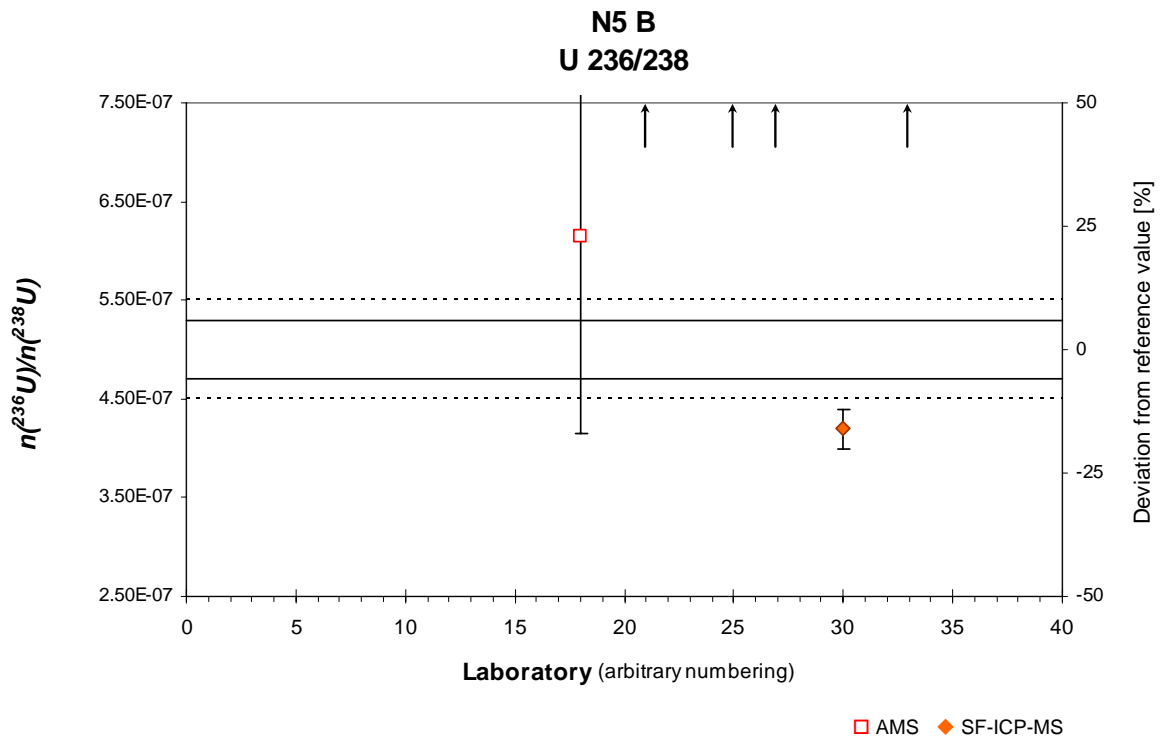
**Figure 7:** Submitted results for  $n(^{235}\text{U})/n(^{238}\text{U})$  in sample C



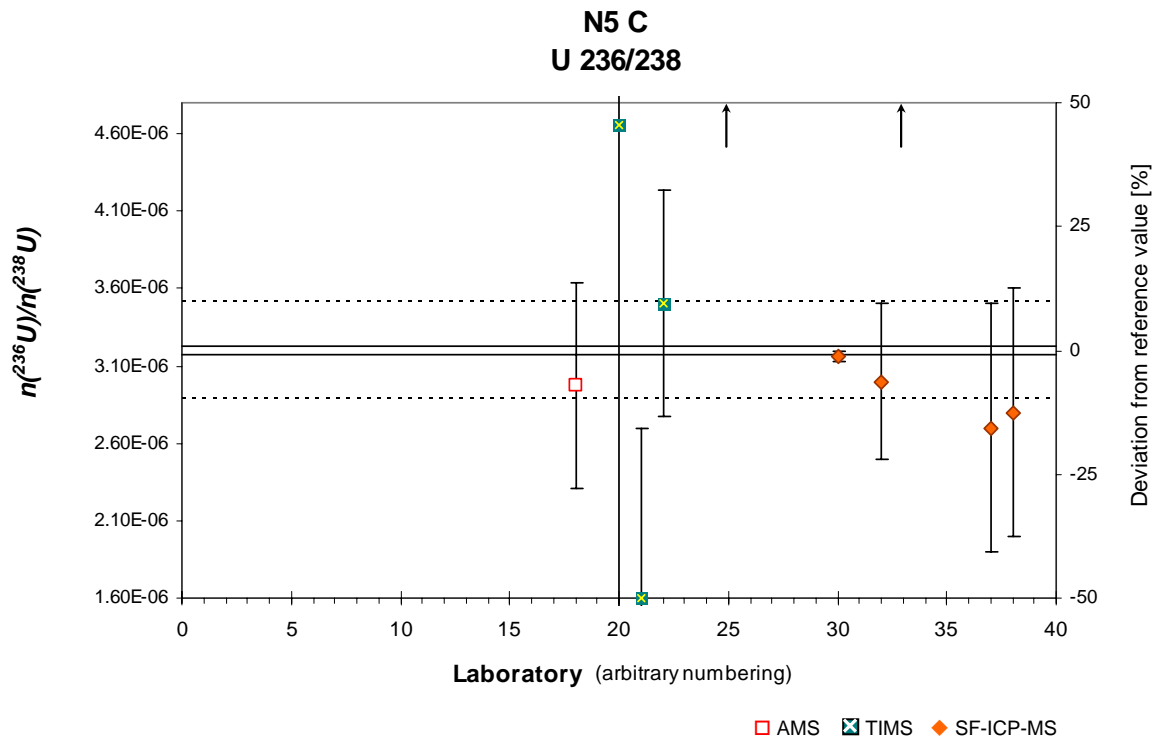
**Figure 8:** Submitted results for  $n(^{235}\text{U})/n(^{238}\text{U})$  in sample D



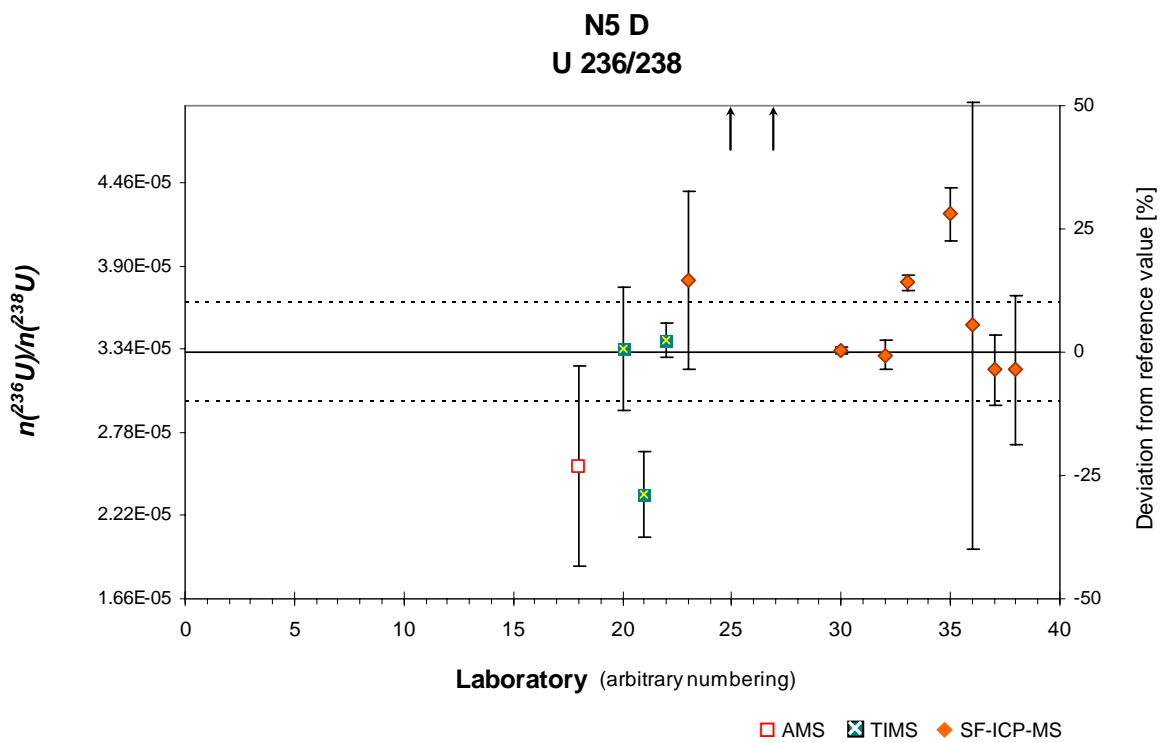
**Figure 9:** Submitted results for  $n(^{236}\text{U})/n(^{238}\text{U})$  in sample A



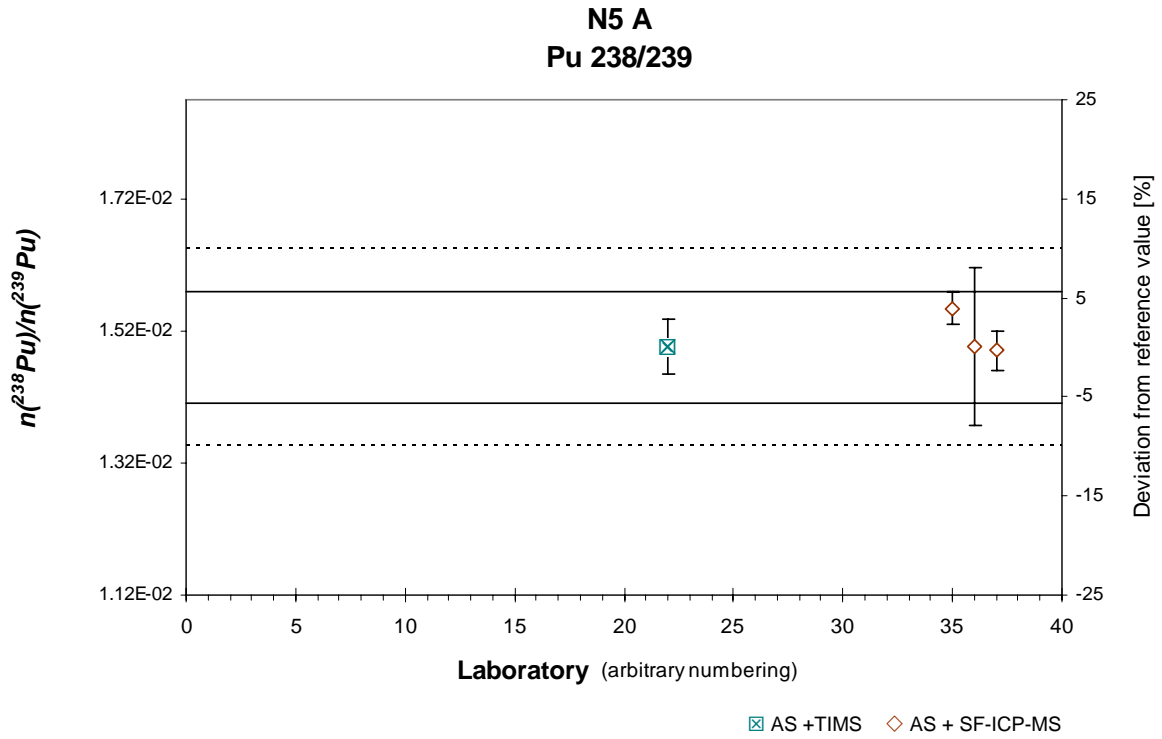
**Figure 10:** Submitted results for  $n(^{236}\text{U})/n(^{238}\text{U})$  in sample B



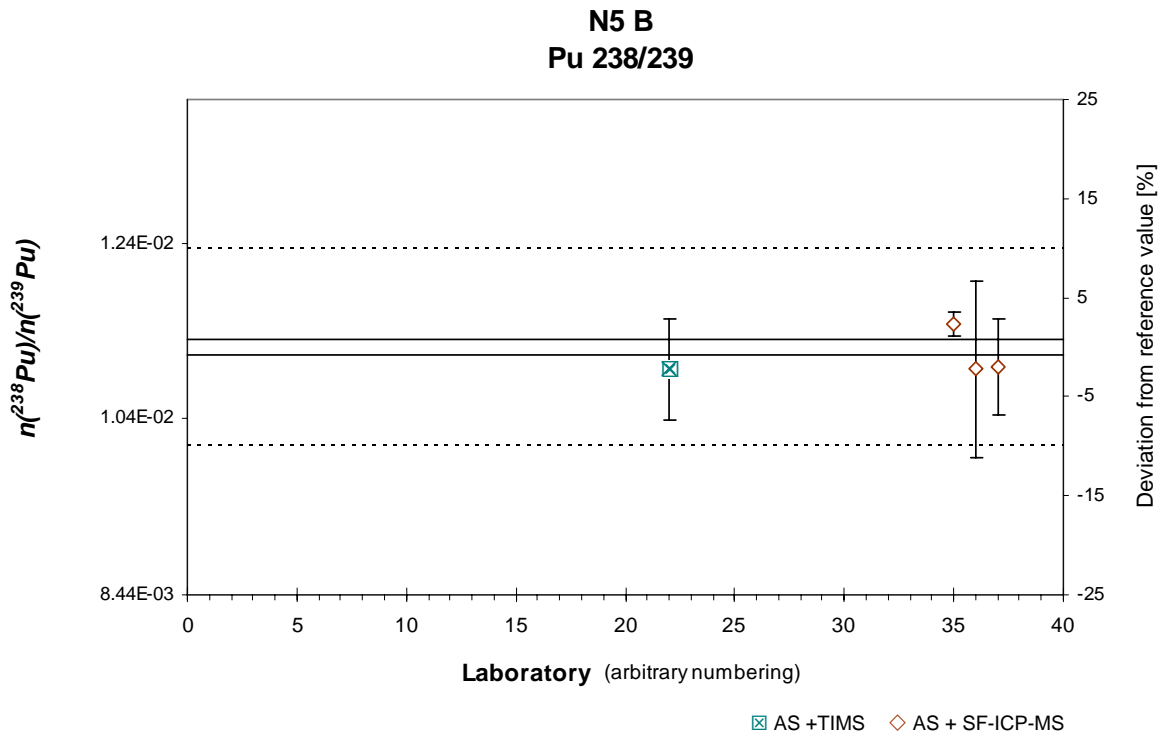
**Figure 11:** Submitted results for  $n(^{236}\text{U})/n(^{238}\text{U})$  in sample C



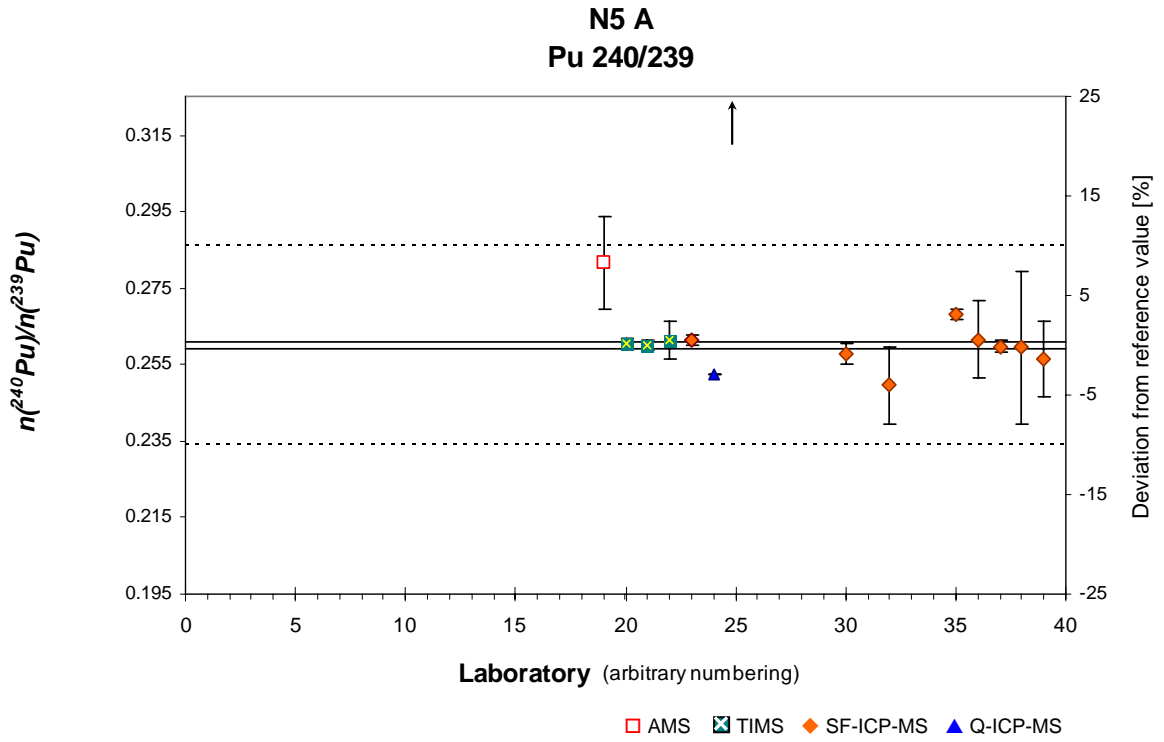
**Figure 12:** Submitted results for  $n(^{236}\text{U})/n(^{238}\text{U})$  in sample D



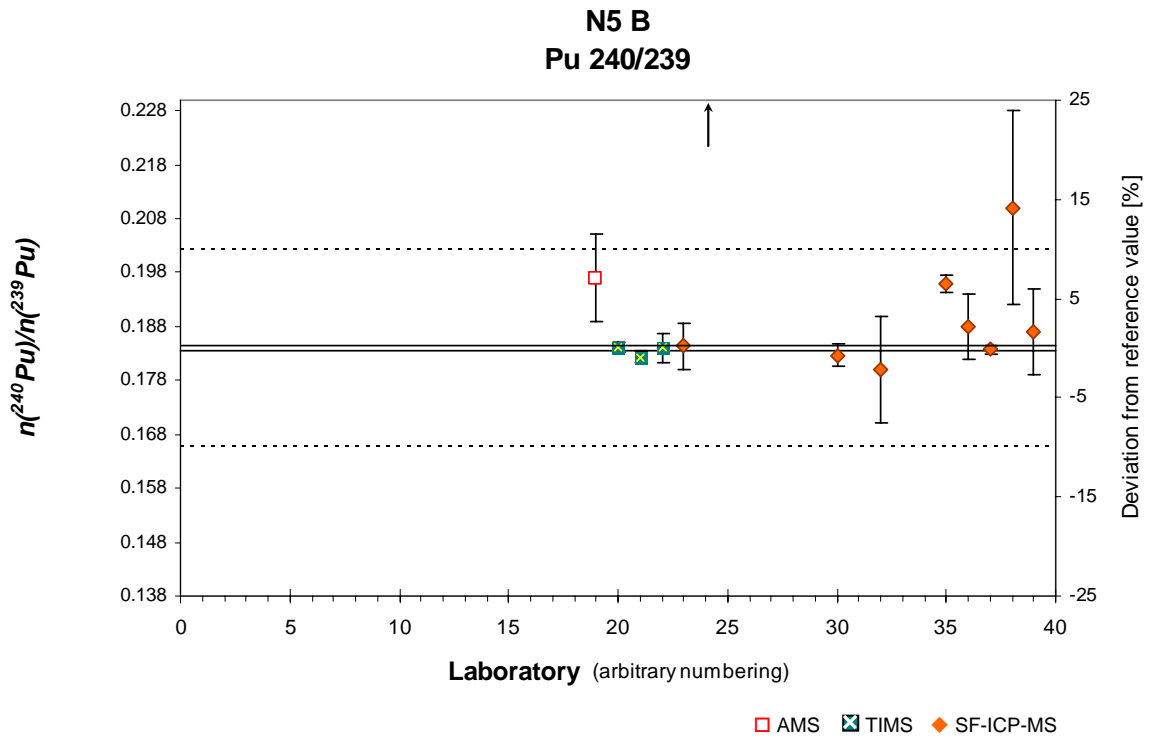
**Figure 13:** Submitted results for  $n(^{238}\text{Pu})/n(^{239}\text{Pu})$  in sample A



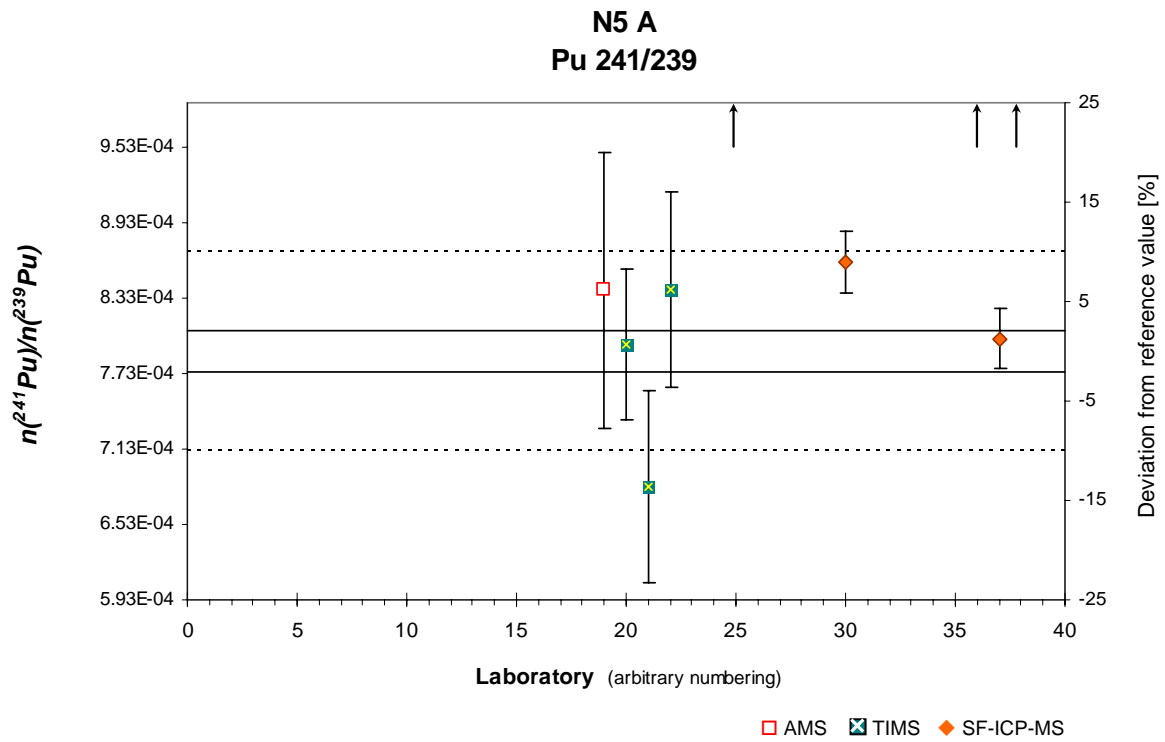
**Figure 14:** Submitted results for  $n(^{238}\text{Pu})/n(^{239}\text{Pu})$  in sample B



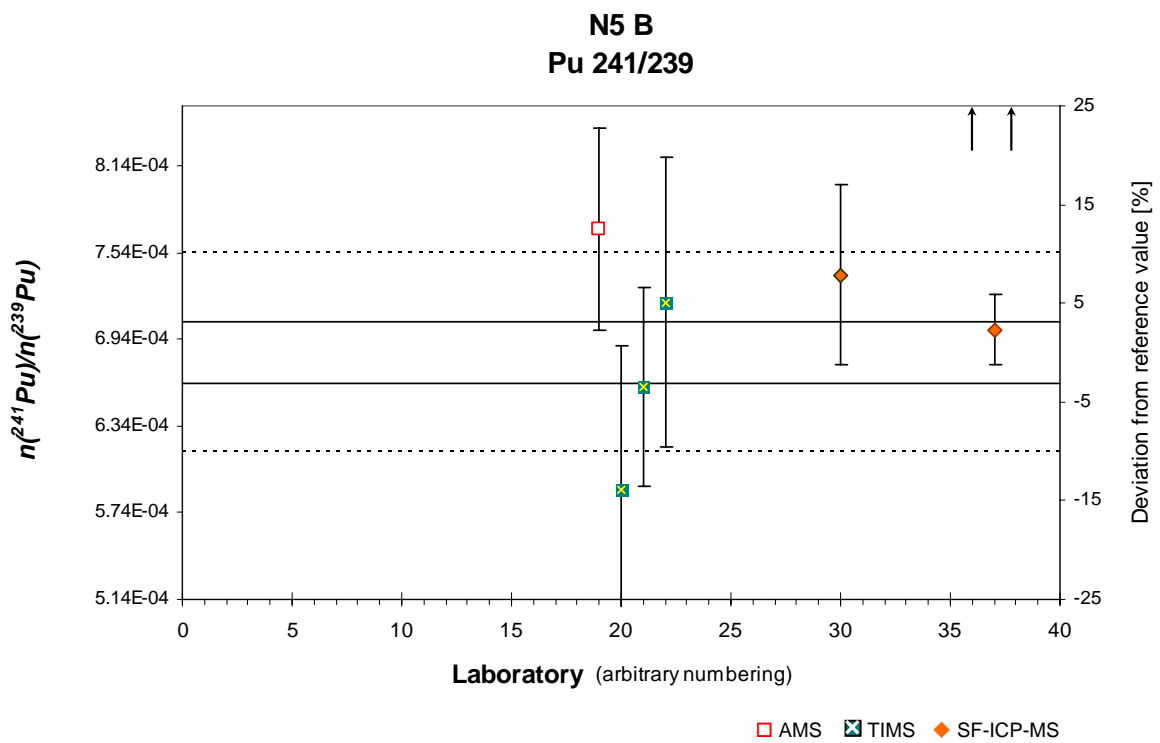
**Figure 15:** Submitted results for  $n(^{240}\text{Pu})/n(^{239}\text{Pu})$  in sample A



**Figure 16:** Submitted results for  $n(^{240}\text{Pu})/n(^{239}\text{Pu})$  in sample B

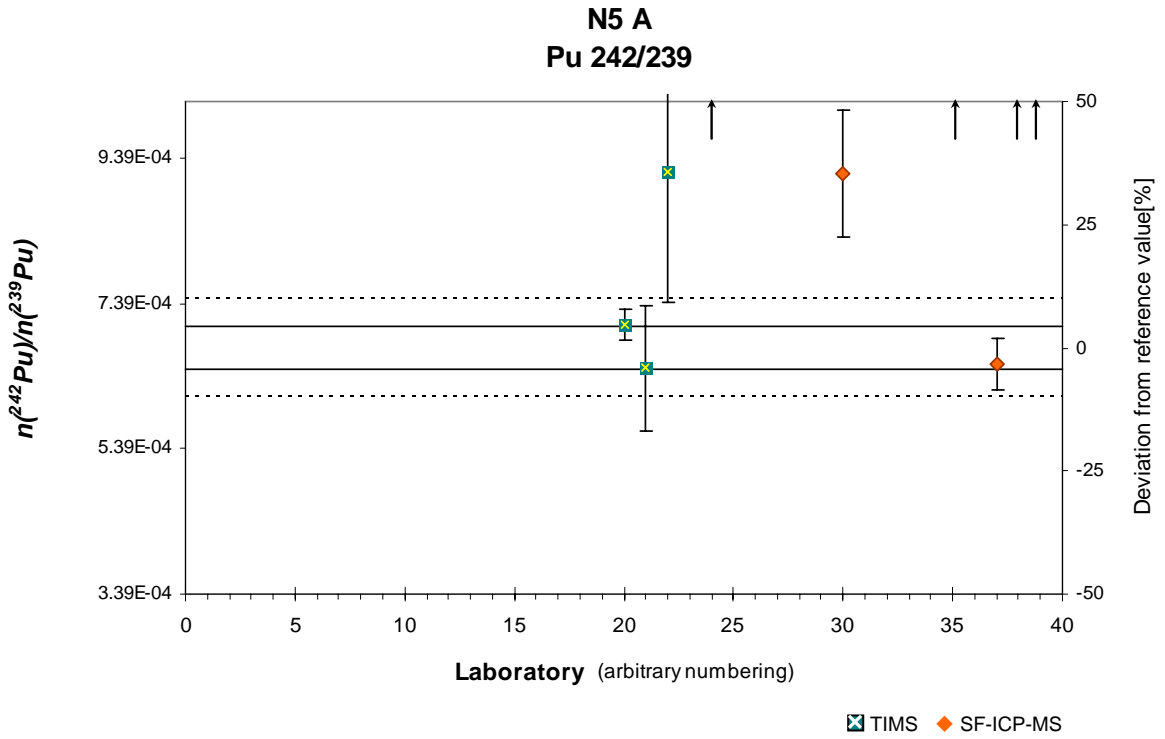


**Figure 17:** Submitted results for  $n(^{241}\text{Pu})/n(^{239}\text{Pu})$  in sample A

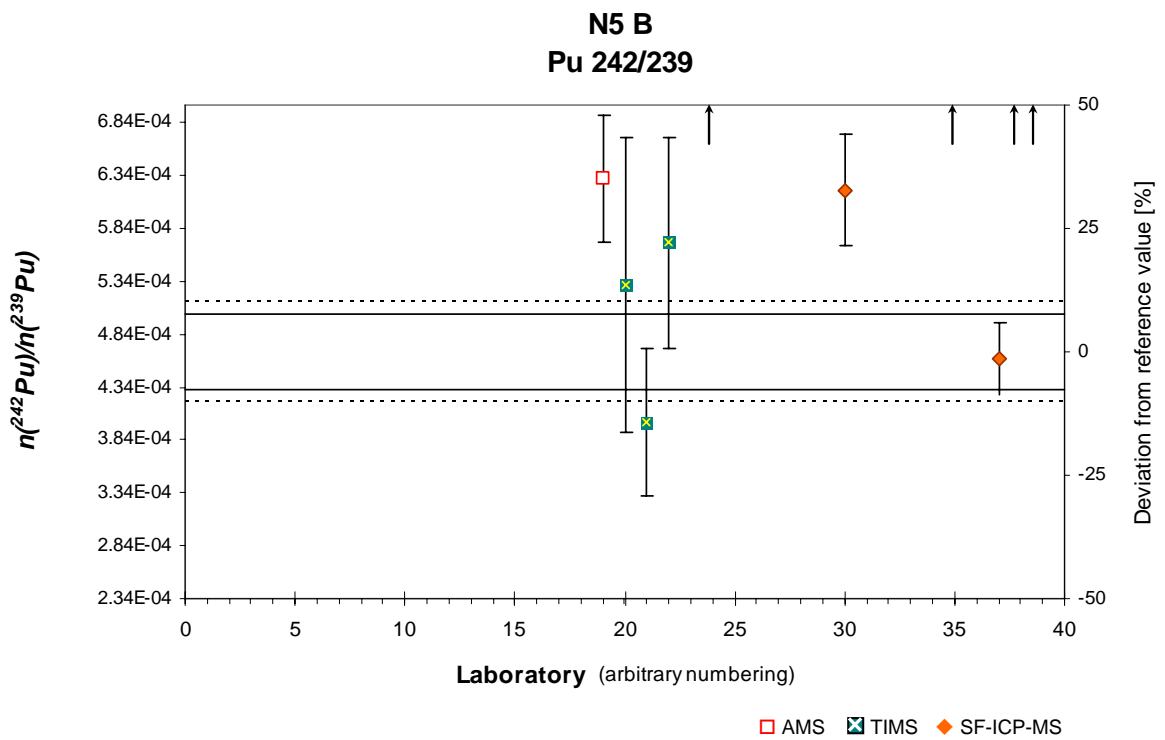


**Figure 18:** Submitted results for  $n(^{241}\text{Pu})/n(^{239}\text{Pu})$  in sample B

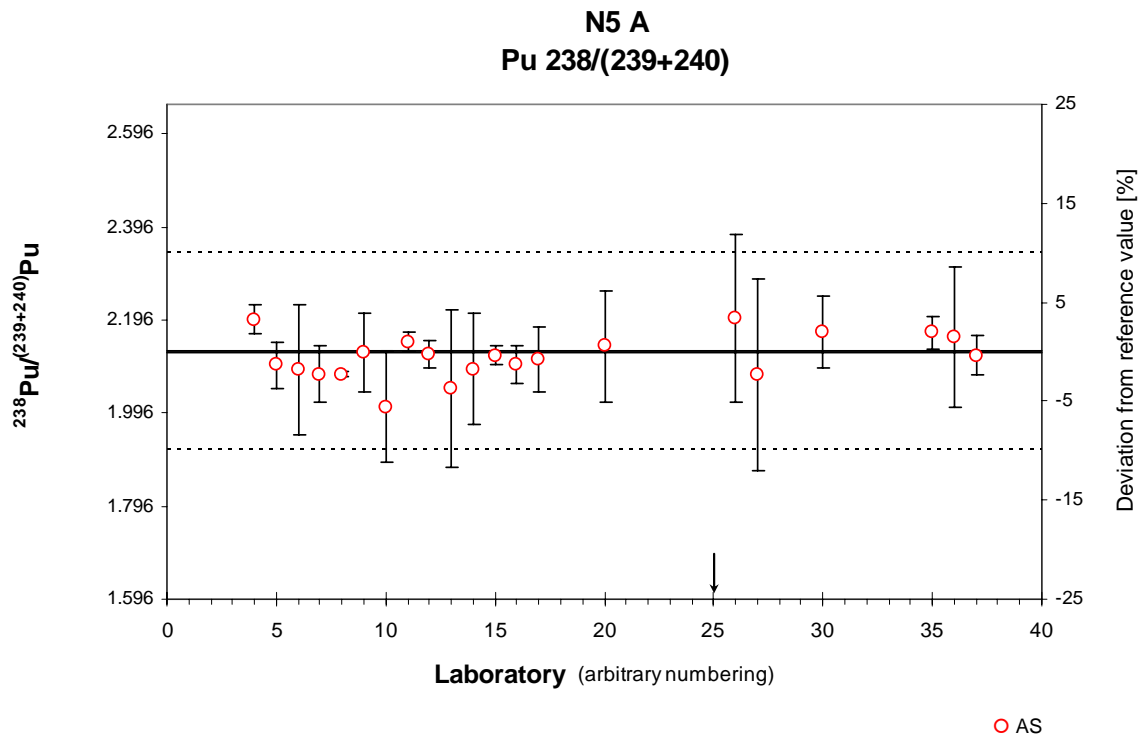




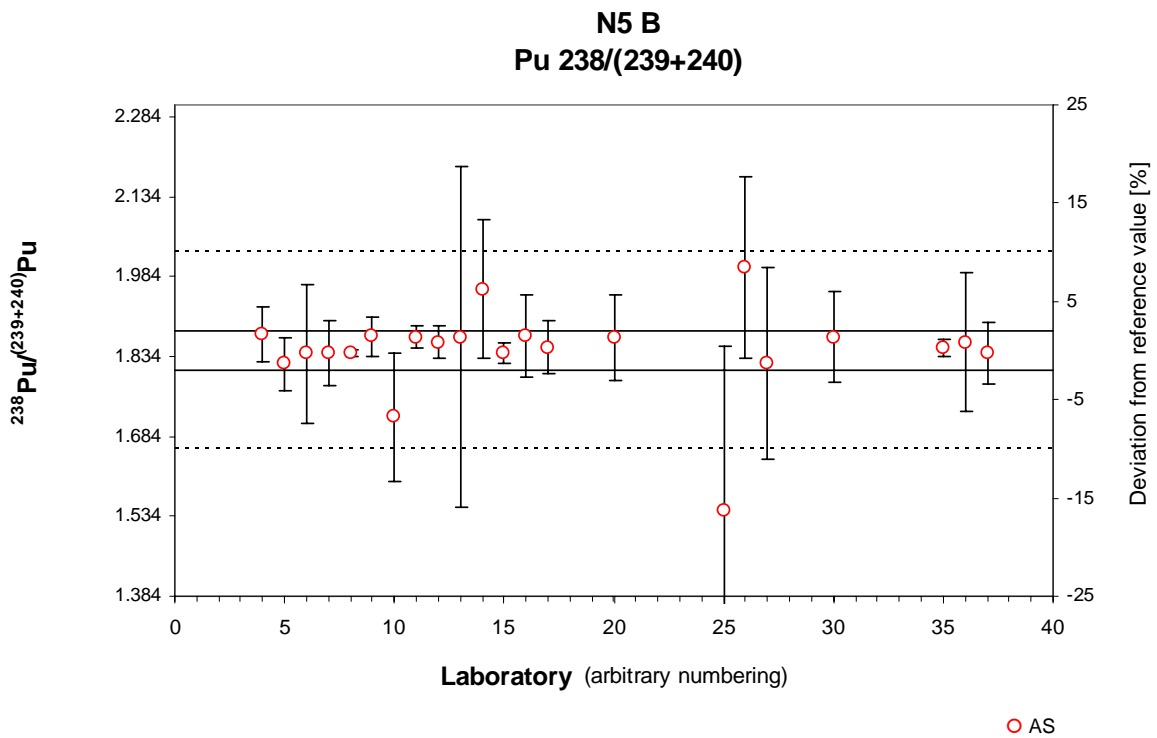
**Figure 19:** Submitted results for  $n(^{242}\text{Pu})/n(^{239}\text{Pu})$  in sample A



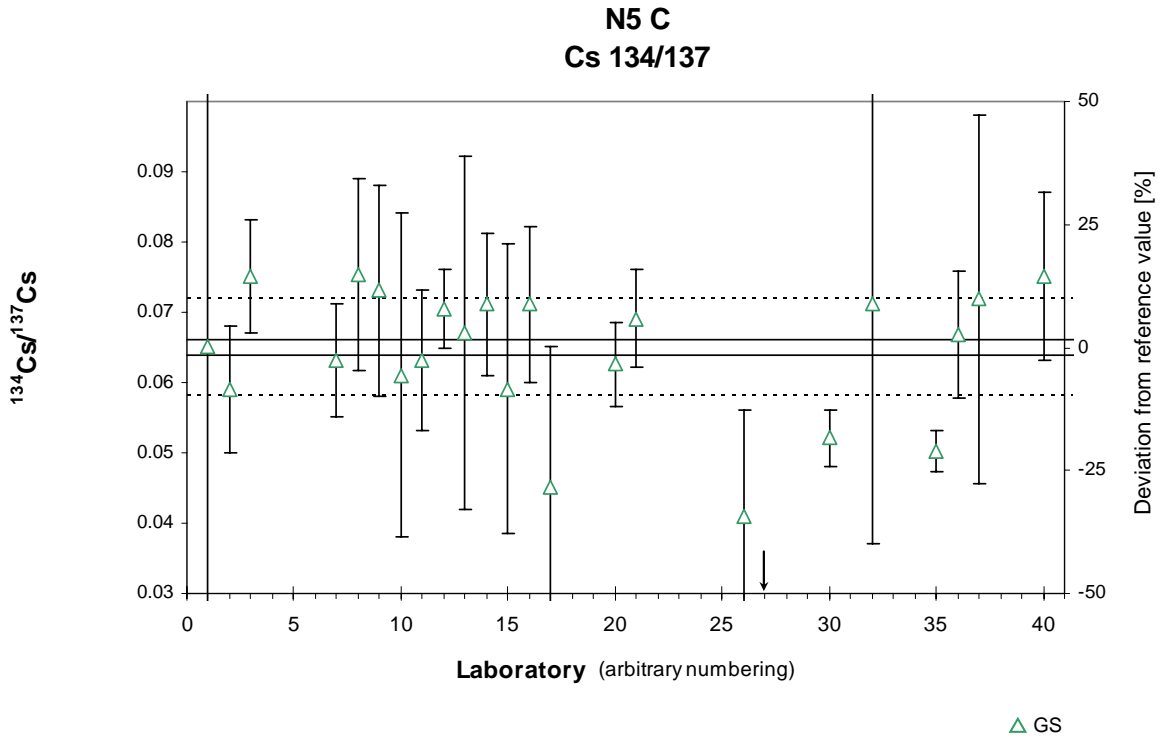
**Figure 20:** Submitted results for  $n(^{242}\text{Pu})/n(^{239}\text{Pu})$  in sample B



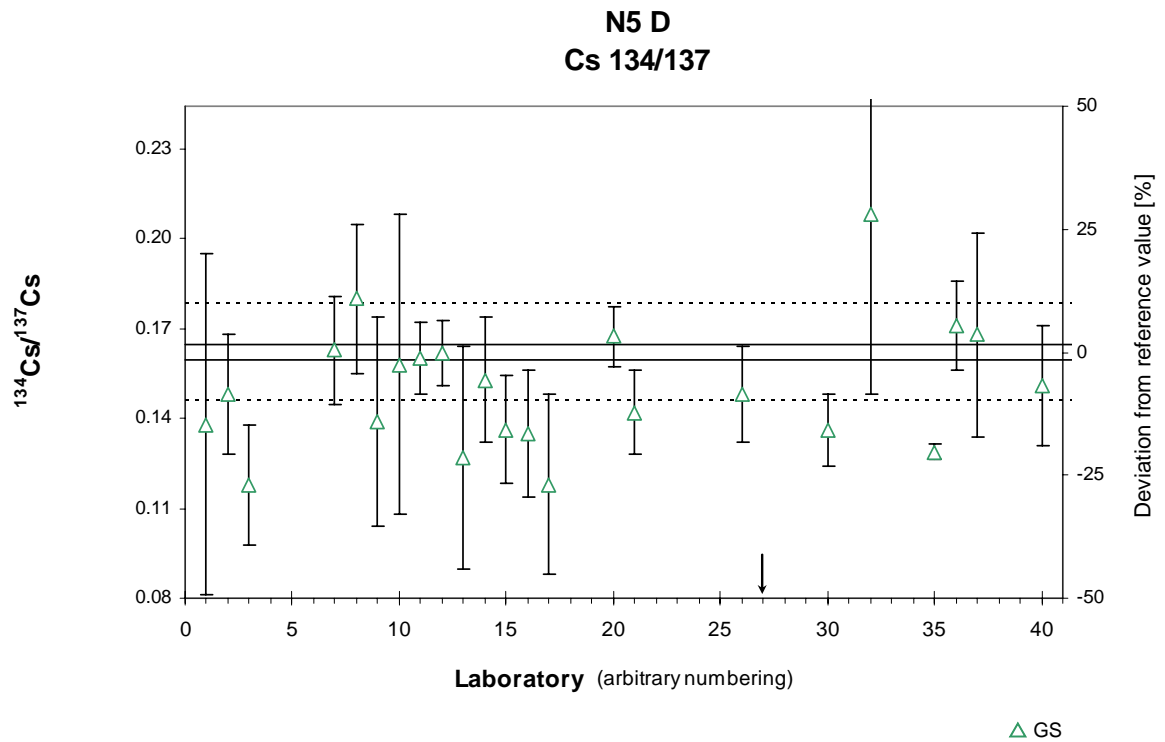
**Figure 21:** Submitted results for  $^{238}\text{Pu}/^{239+240}\text{Pu}$  activity ratio in sample A



**Figure 22:** Submitted results for  $^{238}\text{Pu}/^{239+240}\text{Pu}$  activity ratio in sample B



**Figure 23:** Submitted results for  $^{134}\text{Cs}/^{137}\text{Cs}$  activity ratio in sample C



**Figure 24:** Submitted results for  $^{134}\text{Cs}/^{137}\text{Cs}$  activity ratio in sample D

**Table 8:** Submitted result outside plotted area.

Fig.	Sample	Ratio	Lab	value	% Deviation	Technique
2	B	$n(^{234}\text{U})/n(^{238}\text{U})$	5	0.000015	- 72	AS
3	C	$n(^{234}\text{U})/n(^{238}\text{U})$	3	0.00052	185	GS
5	A	$n(^{235}\text{U})/n(^{238}\text{U})$	25 26	0.006035 0.0059	180 174	Q-ICP-MS Q-ICP-MS
6	B	$n(^{235}\text{U})/n(^{238}\text{U})$	25 26	0.0033 0.019	55 162	Q-ICP-MS Q-ICP-MS
7	C	$n(^{235}\text{U})/n(^{238}\text{U})$	25 26	0.03574 0.055	72 165	Q-ICP-MS Q-ICP-MS
8	D	$n(^{235}\text{U})/n(^{238}\text{U})$	3 26	0.0035 0.079	89 157	GS Q-ICP-MS
9	A	$n(^{236}\text{U})/n(^{238}\text{U})$	21 25 27 33	0.00000079 0.00000179 0.000048 0.00000114	231 649 19983 377	TIMS Q-ICP-MS ICP-MS MC-ICP-MS
10	B	$n(^{236}\text{U})/n(^{238}\text{U})$	21 25 27 33	0.0000042 0.00000321 0.0000082 0.00000248	740 542 1540 396	TIMS Q-ICP-MS ICP-MS MC-ICP-MS
11	C	$n(^{236}\text{U})/n(^{238}\text{U})$	25 33	0.0000069 0.0000023	116 125	Q-ICP-MS MC-ICP-MS
12	D	$n(^{236}\text{U})/n(^{238}\text{U})$	25 27	0.000064 0.0001816	93 447	Q-ICP-MS MC-ICP-MS
15	A	$n(^{240}\text{Pu})/n(^{239}\text{Pu})$	25	0.010663	96	Q-ICP-MS
16	B	$n(^{240}\text{Pu})/n(^{239}\text{Pu})$	24	0.255	39	Q-ICP-MS
17	A	$n(^{241}\text{Pu})/n(^{239}\text{Pu})$	25 36 38	0.008491 0.002 0.0026	974 153 229	Q-ICP-MS LSC + HR-ICP-MS HR-ICP-MS
18	B	$n(^{241}\text{Pu})/n(^{239}\text{Pu})$	36 38	0.0024 0.002	250 192	AS + HR-ICP-MS HR-ICP-MS
19	A	$n(^{242}\text{Pu})/n(^{239}\text{Pu})$	24 35 38 39	0.021 0.004827 0.0025 0.0262	2997 612 269 3764	Q-ICP-MS HR-ICP-MS HR-ICP-MS HR-ICP-MS
20	B	$n(^{242}\text{Pu})/n(^{239}\text{Pu})$	24 35 38 39	0.747 0.0048 0.0088 0.0173	159857 928 1784 3604	Q-ICP-MS HR-ICP-MS HR-ICP-MS HR-ICP-MS
21	A	$^{238}\text{Pu}/^{239+240}\text{Pu}$ activity ratio	25	1.391	- 35	AS
23	C	$^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio	27	0.02218	- 68	GS
24	D	$^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio	27	0.02742	- 83	GS

## 9. Annex 1: List of laboratories which submitted results

COUNTRY	CITY	ORGANISATION NAME
Australia	Lucas Heights	ANSTO
	Canberra	Australian National University
	Melbourne	The University of Melbourne
Bosnia and Herzegovina	Sarajevo	Public Health Institute FB&H
Bulgaria	Plovdiv	NIMH - BAS
Denmark	Roskilde	Risø National Laboratory
France	Brujères le Châtel	CEA
Greece	Agia Paraskevi	Greek Atomic Energy Commission
Hungary	Budapest	HAS CRC Institute of Isotopes
Ireland	Dublin	RPII
Israel	Jerusalem	Geological Survey of Israel
Italy	Rome	ENEA
	Milano	ARPA LOMBARDIA
	Ispra	EC JRC
Japan	Naka-gun	JNC Tokai Works
	Tokai-mura	JAERI
Korea	Taejon	Korean Institute of Nuclear Safety
Latvia	Selaspils	RMTC
Lithuania	Vilnius	Institute of Physics
Malaysia	Kajang, Selangor	MINT
Poland	Sopot	Institute of Oceanology P.A.S.
	Kraków	IFJ PAN
Russian Federation	St. Petersburg	V.G. Khlopin Radium Institute
Serbia - Montenegro	Belgrade	VINCA Institute of Nuclear Sciences
Spain	Madrid	CIEMAT
	Barcelona	Universitat de Barcelona
	Bilbao	Universidad del Pais Vasco
Sweden	Luleå	Analytica AB
	Orebro	Orebro University
	Linköping	Linköping University
Switzerland	Villigen	Paul Scherrer Institute
United Kingdom	Glasgow	Health Protection Agency
United States	Livermore	Lawrence Livermore National Laboratory

# 10. Annex 2: Results reporting form and questionnaire

## 10.1. Result reporting page for samples A and B

IMPORTANT: Disclaimer, Confidentiality Notice and rules on Privacy Protection

European Commission  
Joint Research Centre  
Institute for Reference Materials and Measurements

IRMM Interlaboratory Comparison

> Login > Results

**Functions** Result input for NUSIMEP 5

Results

Dr. Ljudmila Benedik EUROPEAN COMMISSION BELGIUM Page 1 of 4

Sample Code **BLEI3340634**

■ **Isotopic ratio** n(234U)/n(238U) Optional

■ Measurement #1

Select measurement unit from the allowable units:

ratio

Result value  ± Uncertainty value  Coverage factor  k

Technique used

OTHER. Please specify,

▲

■ **Isotopic ratio** n(235U)/n(238U) Optional

■ Measurement #1

Select measurement unit from the allowable units:

ratio

Result value  ± Uncertainty value  Coverage factor  k

Technique used

OTHER. Please specify,

▲

■ **Isotopic ratio** n(236U)/n(238U) Optional

■ Measurement #1

Select measurement unit from the allowable units:

ratio

Result value  ± Uncertainty value  Coverage factor  k

Technique used

OTHER. Please specify,

▲

■ **Activity ratio** 238Pu/ (239Pu + 240Pu) Optional

■ Measurement #1

Select measurement unit from the allowable units:

ratio

Result value  ± Uncertainty value  Coverage factor  k

Technique used

OTHER. Please specify,

▲

■ **Isotopic ratio** n(238Pu)/n(239Pu) Optional

■ Measurement #1

Select measurement unit from the allowable units:

ratio

Result value  ± Uncertainty value  Coverage factor  k

Technique used

OTHER. Please specify,

▲

■ **Isotopic ratio** n(240Pu)/n(239Pu) Optional

■ Measurement #1

Select measurement unit from the allowable units:

ratio

Result value  ± Uncertainty value  Coverage factor  k

Technique used

OTHER. Please specify,

■ **Isotopic ratio** n(241Pu)/n(239Pu) **Optional**

■ **Measurement #1**

Select measurement unit from the allowable units:

ratio

Result value  ± Uncertainty value  Coverage factor  k

Technique used

OTHER. Please specify,

■ **Isotopic ratio** n(242Pu)/n(239Pu) **Optional**

■ **Measurement #1**

Select measurement unit from the allowable units:

ratio

Result value  ± Uncertainty value  Coverage factor  k

Technique used

OTHER. Please specify,

Save  Clear  Next

## 10.2. Result reporting page for samples C and D

IMPORTANT : Disclaimer, Confidentiality Notice and rules on Privacy Protection

European Commission  
Joint Research Centre  
Institute for Reference Materials and Measurements

IRMM Interlaboratory Comparison

> Login > Results

**Functions** Result input for NUSIMEP 5

Results

Dr. Ljudmila Benedik EUROPEAN COMMISSION BELGIUM Page 3 of 4

Sample Code **BLE1814452**

■ **Activity ratio** 134Cs/137Cs Optional

■ Measurement #1

Select measurement unit from the allowable units:

ratio

Result value  ± Uncertainty value  Coverage factor  k

Technique used

OTHER. Please specify,

▲

■ **Isotopic ratio** n(234U)/n(238U) Optional

■ Measurement #1

Select measurement unit from the allowable units:

ratio

Result value  ± Uncertainty value  Coverage factor  k

Technique used

OTHER. Please specify,

▲

■ **Isotopic ratio** n(235U)/n(238U) Optional

■ Measurement #1

Select measurement unit from the allowable units:

ratio

Result value  ± Uncertainty value  Coverage factor  k

Technique used

OTHER. Please specify,

▲

■ **Isotopic ratio** n(236U)/n(238U) Optional

■ Measurement #1

Select measurement unit from the allowable units:

ratio

Result value  ± Uncertainty value  Coverage factor  k

Technique used

OTHER. Please specify,

▲

Previous   
  Save   
  Clear   
  Next



## 10.3. Questionnaire

IMPORTANT : Disclaimer, Confidentiality Notice and rules on Privacy Protection



European Commission  
Joint Research Centre  
Institute for Reference Materials and Measurements

IRMM Interlaboratory Comparison

> Login > Results > Questionnaire

Functions

Questionnaire for NUSIMEP 5

Results

Dr. Ljudmila Benedik

EUROPEAN COMMISSION BELGIUM

1. What is the mission of your laboratory? (you can make more than one choice)

- research and development
- measurements of radioactivity in the environment
- monitoring of nuclear facilities
- medical applications
- measurements for fissile material control or safeguards
- Other

If Other, please specify.

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

- Yes No
- Certified
  - Accredited
  - Authorised

6.  Plutonium applicable measurement  Plutonium non applicable measurement

- < 25 25-50 50-100 > 100
- 

7.  Caesium applicable measurement  Caesium non applicable measurement

- < 25 25-50 50-100 > 100
- 

8. In what type of matrices do you routinely measure small amounts of uranium? (you can make more than one choice)

- applicable  not applicable

In what type of matrices do you routinely measure small amounts of plutonium? (you can make more than one choice)

- Soil
- sediments
- biota
- drinking water
- surface water
- sea water
- urine
- Other

If Other, please specify.

9. In what type of matrices do you routinely measure small amounts of plutonium? (you can make more than one choice)

- applicable  not applicable

- Soil
- sediments
- biota
- drinking water
- surface water
- sea water
- urine
- Other

If Other, please specify.

10. In what type of matrices do you routinely measure small amounts of caesium? (you can make more than one choice)

applicable  not applicable

In what type of matrices do you routinely measure small amounts of plutonium? (you can make more than one choice)

- Soil
- sediments
- biota
- drinking water
- surface water
- sea water
- urine
- Other

If Other, please specify.

11. For these types of measurements how does your laboratory rate itself ?

Experienced Less experienced Not experienced

12. Does your laboratory routinely use certified reference materials?

Yes  No

If Yes, please state.

Which CRM(s) and supplier(s)

How are the CRM(s) applied?

Validation of procedure

Callibration of instrument

Other

If Other, please supply additional information.

13. Were the NUSIMEP 5 samples treated according to the same analytical procedure routinely used for this sample type?

Yes No

If No, please give the reason?

14. Did you carry out the treatment of the samples in a clean laboratory?

Yes No Does not apply

If Yes please give details of the type of facility.

15.

Please give a brief overview of the analysis scheme leading to the measurements (more detailed questions specific to each element analysed are asked below)

16. Was a chemical separation done prior the uranium isotopic ratios measurements?

applicable  not applicable

Yes No

If YES, please specify and give details of the resins and reagents used:

17. If you used alpha spectrometry to measure uranium isotopic ratios, which source preparation technique did you apply?

applicable  not applicable

electrodeposition

drop deposition

rare earth coprecipitation

Other

If Other, please specify.

18. If you used alpha-spectrometry to measure uranium isotopic ratios please describe the equipment and detector:

applicable  not applicable

Equipment

Detector

19. If you used a mass-spectrometric technique to measure uranium isotopic ratios, did you apply a correction for mass fractionation / mass bias?

applicable  not applicable

Yes

No

If YES how was the mass fractionation / mass bias determined?

20. If you used a mass-spectrometric technique to measure uranium isotopic ratios, please describe:

applicable  not applicable

mass-spectrometer

any particular experimental parameters

21. Was a chemical separation done prior the plutonium isotopic ratios measurements?

applicable  not applicable

Yes

No

If YES, please specify and give details of the resins and reagents used:

22. If you used alpha spectrometry to measure plutonium isotopic ratios, which source preparation technique did you apply?

applicable  not applicable

electrodeposition

drop deposition

rare earth coprecipitation

Other

If Other, please specify.

23. If you used alpha-spectrometry to measure plutonium isotopic ratios please describe the equipment and detector:

applicable  not applicable

Equipment

Detector

24. If you used a mass-spectrometric technique to measure plutonium isotopic ratios, did you apply a correction for mass fractionation / mass bias?

applicable  not applicable

Yes

No

If YES how was the mass fractionation / mass bias determined?

25. If you used a mass-spectrometric technique to measure plutonium isotopic ratios, please describe:

applicable  not applicable

mass-spectrometer

any particular experimental parameters

26. Please specify in detail the method of caesium isotope activity ratio measurement including:

applicable  not applicable

Sample preparation:

Counting set-up:

Detector parameters:

Other details:

27. Did you correct for the detector efficiency for caesium isotope activity ratios measurement?  
 Yes  No
28. Did you correct for the gamma emission probabilities for caesium isotope activity ratios measurement?  
 Yes  No
29. Did you correct for gamma peak summing for caesium isotope activity ratios measurement?  
 Yes  No
30. Are you familiar with the Guides for Quantifying Measurement Uncertainty issued by the International Organisation for Standardisation (ISO, 1993) and/or EURACHEM (1995)?  
 Yes  No

31. Were the reported uncertainties calculated following the above-mentioned guides?

Yes  No

If YES, what did you report as an uncertainty?

Standard uncertainty

Expanded uncertainties with a coverage factor

k=

32. If NO, how were the measurement uncertainties evaluated?

33. Do you report uncertainties on measurements to your usual customer?

Yes  No

34. Would you be interested in participating in future NUSIMEP rounds?

Yes  No

If YES,

what type of samples would you be interested?

what type of matrices?

which isotopes?

any mixture of radionuclides?

concentration level?

Other (specify)

35. How did you learn about this NUSIMEP campaign? (you can make more than one choice)

IRMM web site

e-mail from us

from other participants

other (specify)

If Other, please specify.

36. Questionnaire completed by:

Name

Position:

 Clear

Submit questionnaire

European Commission

**EUR 22286 EN – DG Joint Research Centre, Institute for Reference Materials and Measurements -**

NUSIMEP 5: Uranium, plutonium and caesium isotopic abundances in a saline matrix

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**Abstract**

Four different samples were provided in the NUSIMEP 5 campaign, each consisting of two 10 mL saline solutions containing 5 ppb uranium, in the range depleted to low enriched, and with the addition of 1 Bq plutonium or caesium ( $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ ) in some samples. Thirty nine laboratories submitted results and received certified values in return. The results are presented in a series of graphs showing measurement methods and results relative to the certified values. The campaign has been successful in showing the ability of a variety of laboratories to measure the isotopic abundances or activity ratios in trace amounts of these elements in environmental samples.

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