R. Collé & L. Laureano-Perez
Radioactivity Group -- PML
March, 2018

Notes on Experimental Design, Data Reduction, and Uncertainty Evaluations for Certification of Radioactivity SRMs

These notes are intended to serve as informal documentation of the "template" used by the PML Radioactivity Group (PML Radiation Physics Division) for production of Radioactivity SRMs. It is largely based on the information provided to SED in October 2009 (attached ADDENDUM A), but has been substantially updated and revised based on further developments and interactions with SED (Stefan Leigh and Alan Heckert) over the following two years, particularly in providing select RG Group personnel (Collé, Laureano-Perez, and Fitzgerald) with a series of tutorials on statistical methods and in developing, at our request, some new on-line analysis tools. We presently and routinely use both the e-FITS and e-METROLOGY toolkits for our data analyses, particularly normality tests and 2-way ANOVA Refer to

http://stat.nist.gov/~heckert/sed/dlmf/prob_toolkit.htm

http://stat.nist.gov/heckert-bin/e-metrology/anova_2_prod.pl

These notes and our employed procedures are also fully compatible with the quality requirements in our Quality Manual.

https://www.nist.gov/sites/default/files/documents/2016/10/18/procedure15v400.pdf

APPLICABLE SRMS

This "template" outlined here applies to all 4000 series SRMs, excepting the "natural matrix" radioactivity SRMs. This exempted work is done under the direction of Jerry LaRosa as part of the low-level radiochemistry project, and is considerably separate from the production and standardization of the more routine radioactivity SRMs. As in the past, development of future natural matrix SRMs will require considerable collaboration with SED in terms of experimental design and data analyses.

This "template" is also applicable to the 4400 series of SRMs, which are developed as part of the NRMAP quality assurance program for radionuclides used in nuclear medicine. Although these SRMs are distributed to users before certification is completed because of the short half-lives, the experimental designs, data reduction, uncertainty evaluations, and certification is performed under the direct supervision of the Radioactivity SRM Co-Coordinator (Collé) using the identical procedures and principles outlined in this "template."

RADIOACTIVITY STANDARDIZATION PROGRAM ELEMENTS

These program elements (as promulgated in publications by R. Collé) provide, in part, an overview of the underlying philosophy for the development of Radioactivity SRMs.

- Choice of nuclides / standards based on identified user needs
- Standardization based on at least one primary method, whose certified value is traceable to the derived SI unit, becquerel (Bq)
- Validity of primary method supported & verified by one or more independent confirmatory methods
- Standardizations typically utilize many trials, with widely varying experimental conditions,
    - minimizing type-B uncertainty assessments
- Any new standardization linked back to all previous ones (when possible)
    - through stored solutions or calibration factors for secondary instruments
- Disseminated standards from primary methods used as SRM transfer standards
    - and/or employed as sources for quality assurance, & proficiency testing programs
- Primary standardization uncertainties ($k = 2$) are typically < 1 % (few tenths at $k = 1$)
- All uncertainty treatments follow GUM principles (as given in ISO and NIST documents)
- Comparisons are made with other metrology labs to demonstrate & ensure international consistency

The role of these elements as it applies to the "template" will become evident, as demonstrated in extant SRM certificates (examples given in ADDENDUM B).

EXPERIMENTAL DESIGN, DATA REDUCTION & UNCERTAINTY EVALUATION CONSIDERATIONS

The experimental design & uncertainty analyses need for the production and standardization of radioactivity SRMs are largely NOT statistical issues.

The experimental design (basic measurement model) is largely fixed and based on efficacious procedures developed over many years. Examples of typical experimental designs are given in many publications (see attached ADDENDUM C). An example of a typical experimental design for ^{239}Pu follows in ADDENDUM A. The analysis for this case was originally presented to SED in October 2009 as an example. It was subsequently used by them to not only verify the procedures used by us at the time, but also to help them understand our needs for toolkits with easy to use and efficient analysis tools. We now routinely use the e-FITS and e-METROLOGY tools as part of our data analyses.

The SRMS are homogeneous and chemically stable, pristine, inorganic solutions. Therefore, sampling considerations for the stable & homogeneous solutions are trivial, having negligible or non-existent inhomogeneity issues. Nevertheless, the SRM solutions or solution ampoules are generally tested for homogeneity in some way.

Data reduction and analyses are largely done "by hand", largely using EXCEL spreads sheets. This is done rather than employing automated computerized data reduction programs because we believe it is important to examine the data (and its pieces) very closely for trends and effects. Even before our interactions with SED in October 2009, we generally relied upon extensive graphical analyses – plotting every variable we could think of against every other. The value of this was also emphasized by SED, and they provided us with additional graphical tools, like GANOVA.

Uncertainty analyses for the type-A-method components are derived directly from the measurement data; *viz.*, statistical estimators. A nested uncertainty approach is employed and considers multiple within- and between-component uncertainties. This is outlined in many of our published papers. For example, it would be usual to (i) select perhaps three SRM ampoules for examination, and (ii) from each of these ampoules prepare six LS counting sources in three different cocktail compositions, and (iii) these 18 sources per ampoule would then be measured in at least two (sometimes three) different LS counters. Hence a 3 x 3 array of data would be created for each of the sampled ampoules, which is ideally suitable for ANOVA, looking at the within-source precision for the six sources in each box, at the between-composition variance for a given counter, and the between-counter variance for a given composition.

For a given sampled ampoule			
	Comp 1	Comp 2	Comp 3
Counter 1	6 sources	6 sources	6 sources
Counter 2	6 sources	6 sources	6 sources
Counter 3	6 sources	6 sources	6 sources

Then the array for each ampoule can be compared, resulting in 162 measurement results as obtained from 54 sources (18 x 3) in three counters. Such a case as this is considered in ADDENDUM A for the standardization of ^{239}Pu for SRM 4330c. In general, there is negligible difference between ampoules, but the data may exhibit between-source or between-instrument differences.

At times, the data can be thrown into the same "pot" to obtain precision estimators if the data are sufficiently consistent (on testing for normality, t- and F-tests, ANOVA, etc.). Otherwise, the uncertainty estimators are derived considering all of the identified within-and between-components of variance.

These analyses up to about 2010 - 2012 were entirely performed manually. With the advent of the SED Heckert toolkits, they are now largely done in conjunction with the on-line tools. Yet, the measurement data are still largely examined graphically and by hand.

Uncertainty analyses for the type-B-method components is obtained from an understanding of the METROLOGY and the underlying science and effects. These are largely judgment based (often canonically assumed) as obtained from experience, testing, and sensitivity analyses.

The magnitudes of both components (A & B methods) are largely driven and determined by the variables and parameters chosen for the primary standardization measurement (replications and trials varying instruments, times, counting source preparation, etc.). Compared to most standardizations by our sister laboratories (i.e., other National Metrology Institutes, like NPL in UK, LNHB in France, PTB in Germany, etc.) our laboratory is well known and distinguished for routinely conducting more experimental trials and varying more measurement variables for typical standardizations.

With rare exceptions, the type-B-method components are largely dominant; with statistical uncertainties making smaller contributions to the combined standard uncertainty.

By a wise policy decision made many years ago, the primary standardization for any of our radioactivity SRMs is based on only ONE METHOD (the best method available to us). Other standardization methods (invariably inferior with larger uncertainties) performed at the same time are used only for confirmatory purposes, to ensure the validity of the primary method.

Discrepant data in our SRM work is rarely encountered (unlike the situation for the Natural Matrix SRMs with multi-lab & multi-method results).

GUM principles (as given in ISO & NIST documents) are adhered to religiously (as interpreted by one of the original GUM founders). Uncertainty budget often consists of one component from type-A evaluation (maybe two or three at times) and up to a half-dozen or more components from type-B evaluations. Examples of such budgets are shown in the Certificates given in ADDENDUM B.

ADDENDUM A :

An example of ^{239}Pu standardization for SRM 4330c

Attachments for this example follow and include:

- Schematic showing gravimetric and measurement links to previous ^{239}Pu SRMs
- Design / scheme for preparation and measurements of SRM 4330c
- Dispensing precision
- All LS data (n = 135) for three cocktail compositions on three instruments
- Summary of LS data averages
- The 9 LS means
- Normal probability plot
- SRM 4330c Certificate

ADDENDUM B

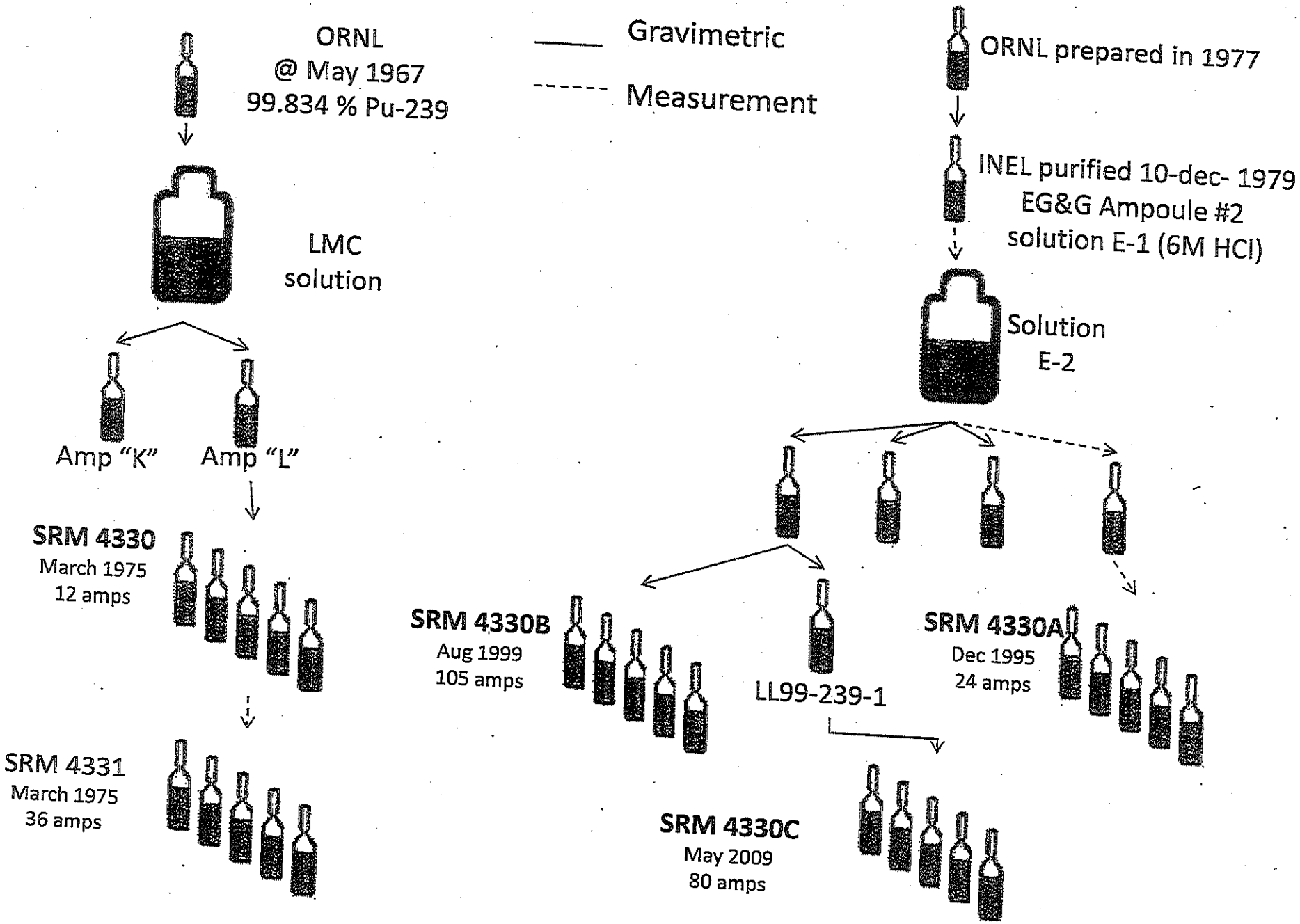
Two recent Radioactivity SRM Certificates

SRM 4323c, Plutonium-238 Radioactivity Standard (January 2017)

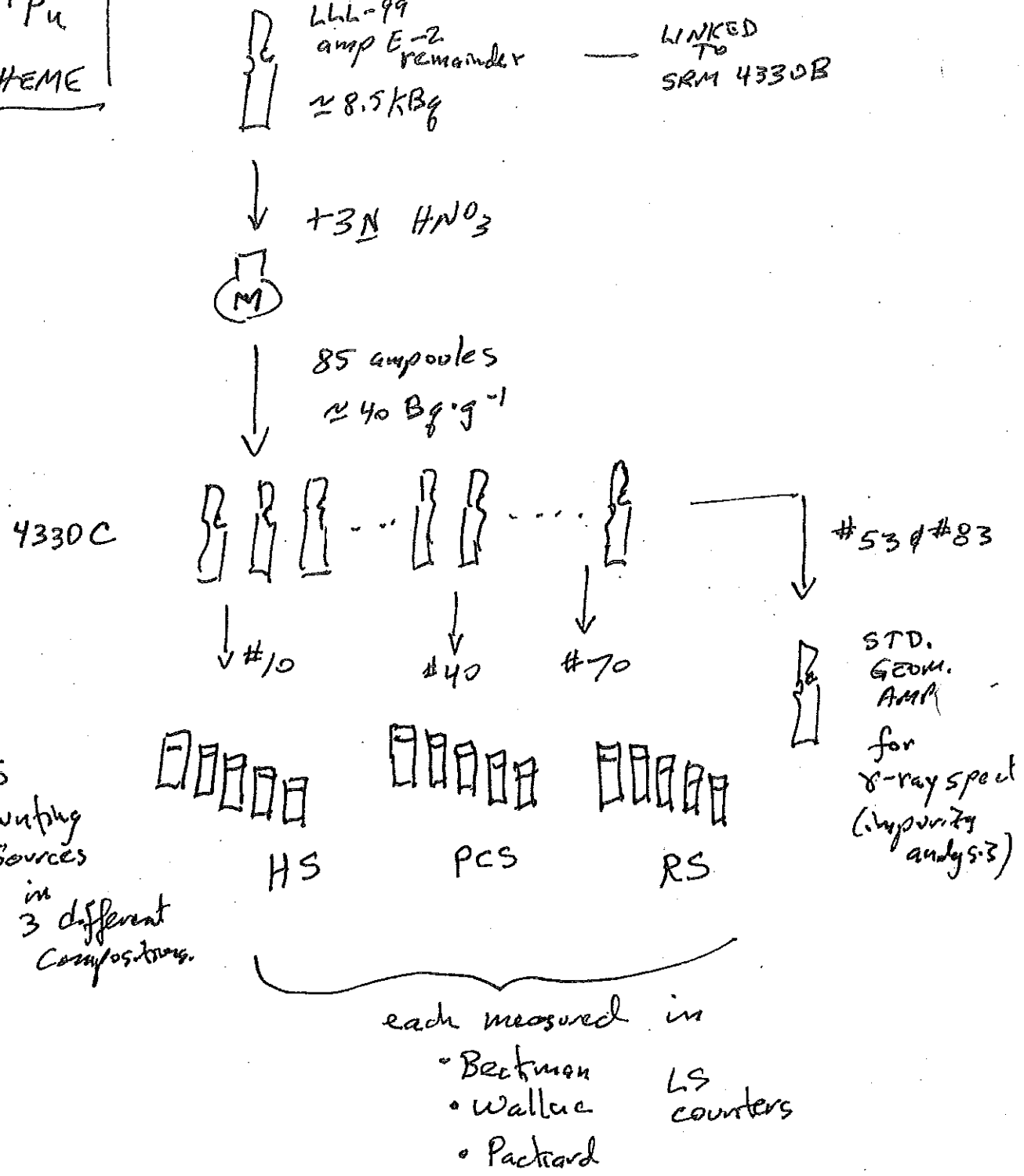
SRM 4334j, Plutonium-242 Radioactivity Standard (January 2018)

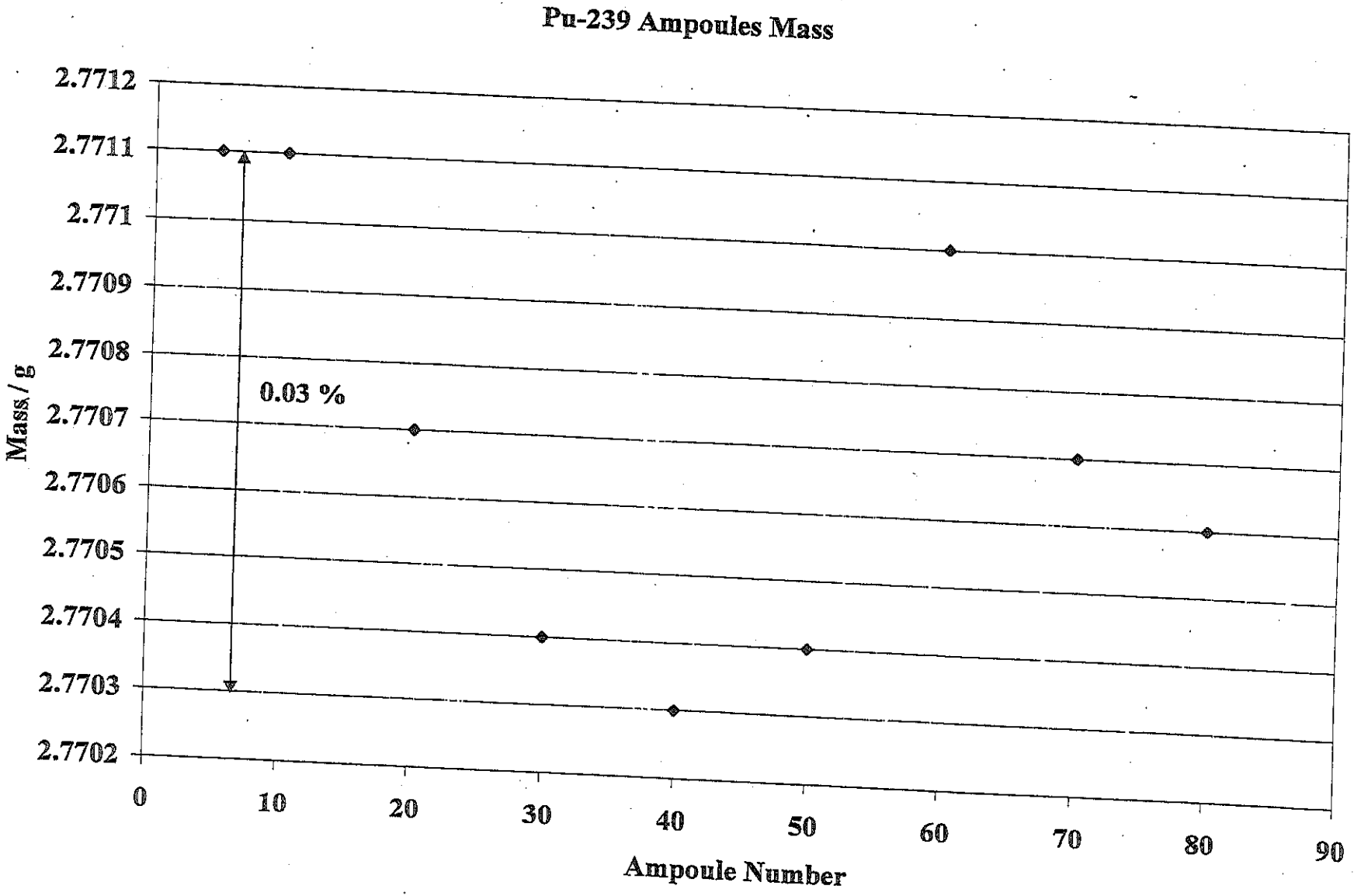
ADDENDUM C

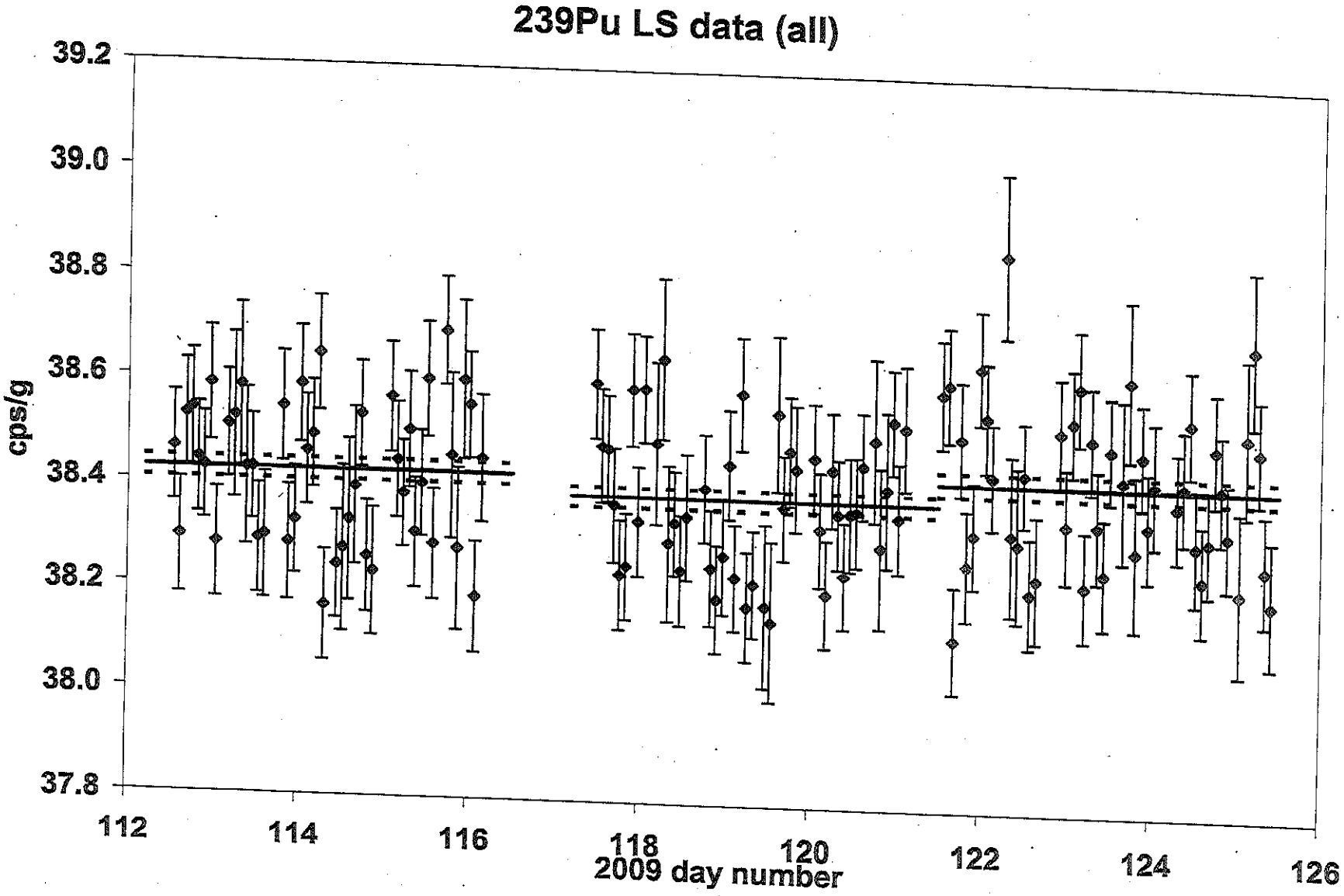
Published papers describing experimental designs, data reductions and uncertainty evaluations for Radioactivity SRMs


ORNL
@ May 1967
99.834 % Pu-239
Gravimetric
Measurement
ORNL prepared in 1977
INEL purified 10-dec- 1979
EG&G Ampoule #2
solution E-1 (6M HCl)
LMC
solution
Solution
E-2
Amp "K"
Amp "L"
SRM 4330
March 1975
12 amps
SRM 4330B
Aug 1999
105 amps
LL99-239-1
SRM 4330A
Dec 1995
24 amps
SRM 4331
March 1975
36 amps
SRM 4330C
May 2009
80 amps

^{239}Pu SCHEME




Pu-239 Ampoules Mass
2.7712
2.7711
2.771
2.7709
2.7708
2.7707
2.7706
2.7705
2.7704
2.7703
2.7702
Mass / g
0.03 %
0
10
20
30
40
50
60
70
80
90
Ampoule Number


239Pu LS data (all)
39.2
39.0
38.8
38.6
38.4
38.2
38.0
37.8
cps/g
112
114
116
118
120
122
124
126
2009 day number

^{239}Pu averages

	BECKMAN	WALLAC	PACKARD	(all)
HS	38.521 0.23% n=15	38.438 0.32% n=15	38.501 0.40% n=15	38.486 0.33% n=45
PCS	38.427 0.33% n=15	38.366 0.36% n=15	38.397 0.40% n=15	38.397 0.36% n=45
RS	38.325 0.29% n=15	38.347 0.34% n=15	38.387 0.37% n=15	38.353 0.34% n=45
(all)	38.424 0.35% n=45	38.384 0.35% n=45	38.428 0.40% n=45	38.412 0.37% sd n=135
				0.032% sdm

Mean was invariant of
- cocktail composition (scintillant used)
- instrument (with different detection thresholds, deadtime, etc.
- aliquant mass in cocktail
- sample quenching
- stability (time)

n=9 means w/ 15 per mean
38.412
0.17₃% sd

Colle, Ronald (Fed)

From: Laureano-Perez, Lizbeth (Fed)
Sent: Thursday, March 08, 2018 11:41 AM
To: Colle, Ronald (Fed)
Subject: FW: Radioactive Solutions SRMs

From: Possolo, Antonio (Fed)
Sent: Wednesday, March 07, 2018 1:11 PM
To: Laureano-Perez, Lizbeth (Fed) <lizbeth.laureano-perez@nist.gov>
Cc: Leber, Dennis D. (Fed) <dennis.leber@nist.gov>
Subject: Radioactive Solutions SRMs

Hello, Lizbeth,

I know that you follow a "template" for data reductions and uncertainty evaluations for the radioactive solutions SRMs, which I believe was developed jointly with Stefan Leigh.

We in SED are surveying the user's of approved templates to try to make everybody's life as simple as possible and still satisfy the requirements of the ORM, by providing ORM with a list of approved templates and associated documentation.

Do you have any document, however informal, that describes your template and states what SRMs it applies to?

If you do, I'd be grateful for an electronic copy or scan of it. If not then we'd better develop one. In such case, I'll be happy to work with you to get it done.

Much appreciated.

- Antonio

- Antonio Possolo
NIST Fellow and Chief Statistician
National Institute of Standards &Technology

1/16/2021

x2853

Colle, Ronald (Fed)

From: Possolo, Antonio (Fed)
Sent: Tuesday, March 20, 2018 1:59 PM
To: Colle, Ronald (Fed)
Cc: Laureano-Perez, Lizbeth (Fed); Zimmerman, Brian E. (Fed); Fitzgerald, Ryan P. (Fed); Regits, Willie (Assoc); Neal, Khyra A. (Assoc); Choquette, Steven J (Fed); Camara, William Dinis (Fed); Leber, Dennis D. (Fed)
Subject: Re: Radioactivity SRM "template"

Hello, Ron,

I have now read the outline of the procedures you and your colleagues use for value assignment and uncertainty evaluation for radioactive SRMs in the classes that you specify under "Applicable SRMs": series 4000 and 4400, except "natural matrix" SRMs.

All is in good order, as I expected.

I will confirm to ORM that your existing "template" is valid and acceptable, and I will continue to sign the corresponding blue sheets without the need for participation of a SED statistician.

However, we will remain at your service in case you'll like to discuss any unusual situations that may arise in relation with SRMs that are covered by this "template."

- Antonio

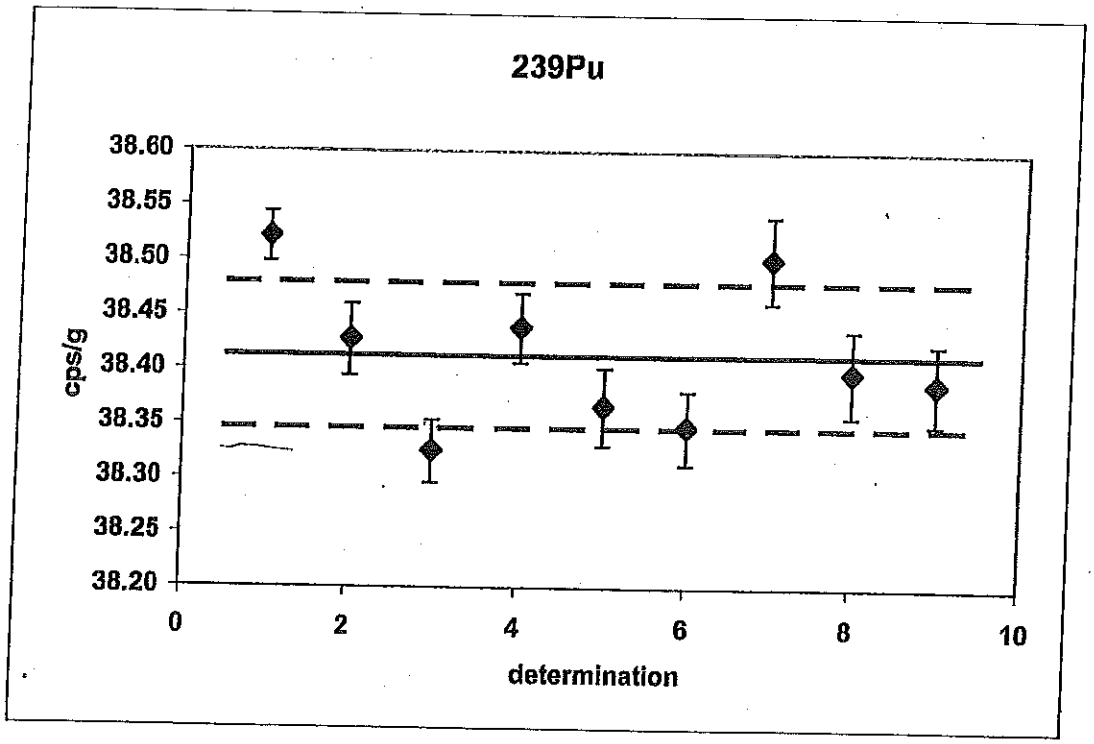
- Antonio Possolo
NIST Fellow and Chief Statistician
National Institute of Standards &Technology

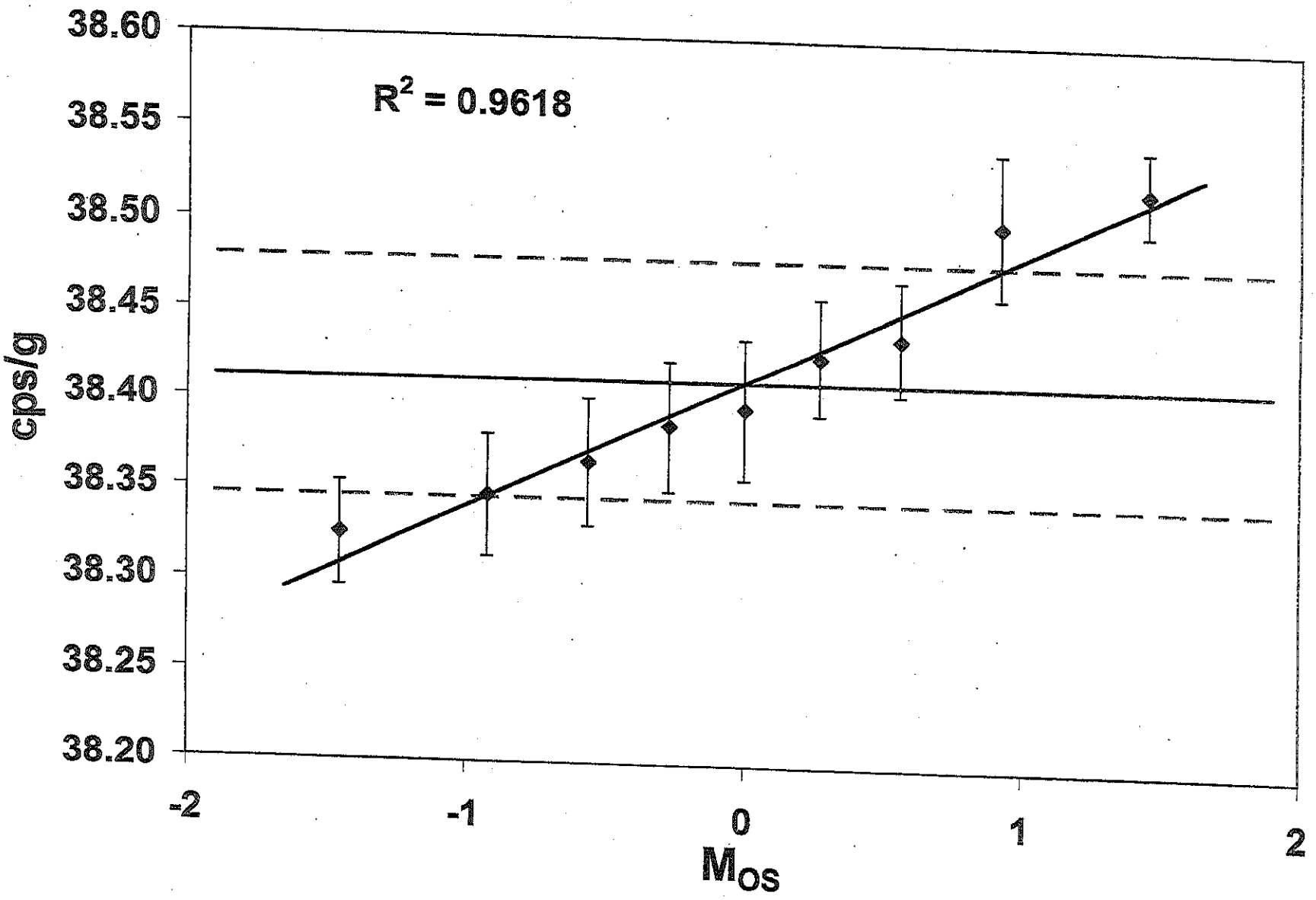
On: 20 March 2018 12:48,
"Colle, Ronald (Fed)" <ronald.colle@nist.gov> wrote:

Hello Antonio,
I have attached the requested "template" that explains our data reductions and uncertainty evaluations for the Radioactivity SRM Program.
Of necessity, it is not a template in the sense of a fill-in-the-blanks document, but rather (in a general sense) describes the pattern we use, in terms of the procedures, processes, and tools we employ for this work.
I hope it fulfills SED and ORM needs.
Sincerely,
Ron

	mean	%sd	%sdm	unc	
B-H	38.521	0.23	0.059386	0.022876	-1.45
B-P	38.427	0.33	0.085206	0.032742	-0.92
B-R	38.325	0.29	0.074878	0.028697	-0.56
W-H	38.438	0.32	0.082624	0.031759	-0.27
W-P	38.366	0.36	0.092952	0.035662	0
W-R	38.347	0.34	0.087788	0.033664	+0.27
P-H	38.501	0.397	0.102505	0.039465	0.56
P-P	38.397	0.399	0.103021	0.039557	0.92
P-R	38.387	0.37	0.095534	0.036672	1.45
ave	38.41211				
sd	0.06658				

0.5	38.41211	38.47869	38.34553
9.5	38.41211	38.47869	38.34553




38.60
38.55
38.50
38.45
38.40
38.35
38.30
38.25
38.20
cps/g
$R^2 = 0.9618$
-2
-1
0
1
2
M_{OS}

(7)

National Institute of Standards & Technology

Certificate

Standard Reference Material® 4330C

Plutonium-239 Radioactivity Standard

This Standard Reference Material (SRM) consists of a solution of a standardized and certified quantity of radioactive plutonium-239 in a suitably stable and homogeneous matrix. It is intended primarily for the calibration of instruments that are used to measure radioactivity and for the monitoring of radiochemical procedures. A unit consists of a solution, whose composition is specified in Table 1, contained in a flame-sealed 5 mL borosilicate-glass ampoule (see Note 1)*.

The certified **Plutonium-239** massic activity value, at a **Reference Time of 1200 EST, 1 May 2009**, is:

$$(38.41 \pm 0.18)\ \mathrm{Bq \cdot g^{-1}}$$

Additional physical, chemical, and radiological properties for this SRM, as well as details on the standardization method, are given in Table 1. Uncertainties for the certified quantities are expanded ($k = 2$). The uncertainties are calculated according to the ISO and NIST Guide (see Note 2). Table 2 contains a specification of the components that comprise the uncertainty analyses.

Expiration of Certification: The certification of **SRM 4330C** is valid indefinitely provided the SRM is handled and stored properly and no evaporation or change in composition has occurred. The solution matrix, in an unopened ampoule, is indefinitely homogeneous and stable within its half-life-dependent useful lifetime provided the SRM is handled in accordance with instructions given in this certificate (see "Instructions for Use"). The certification is nullified if the SRM is damaged, contaminated, or otherwise modified.

Maintenance of Certification: NIST will monitor this SRM over the period of its certification. If substantive technical changes occur that affect the certification before the expiration of this certificate, NIST will notify the purchaser. Registration (see attached sheet) will facilitate notification.

This SRM may represent a radiological hazard and a chemical hazard. Consult the Material Safety Data Sheet (MSDS), enclosed with the SRM shipment, for details (see Note 1).

This Standard Reference Material was prepared in the Physics Laboratory, Ionizing Radiation Division, Radioactivity Group, M.P. Unterweger, Group Leader. The overall technical direction and physical measurement leading to certification were provided by R. Collé and L. Laureano-Pérez of the NIST Radioactivity Group, with production assistance by D.B. Golas, Research Associate of the NRMAP, Inc., and photon-emitting impurity analyses by L. Pibida

Support aspects involved in the issuance of this SRM were coordinated through the NIST Measurement Services Division.

INSTRUCTIONS FOR USE

Storage: SRM 4330C should be stored and used at a temperature between 5 °C and 65 °C. The ampoule (or any subsequent container) should always be clearly marked as containing radioactive material.

Handling: If the ampoule is transported, it should be packed, marked, labeled, and shipped in accordance with the applicable national, international, and carrier regulations. The solution in the ampoule is a dangerous good (hazardous material) because of both the radioactivity and the strong acid. The ampoule should be opened only by persons qualified to handle both radioactive material and alkaline and/or acidic solutions. Appropriate shielding and/or distance should be used to minimize personnel exposure. Refer to MSDS for further information.

Lisa R. Karam, Chief
Ionizing Radiation Division

Gaithersburg, Maryland 20899
Certificate Issue Date: 17 July 2009
See Certificate Revision History on Last Page

Robert L. Watters, Jr., Chief
Measurement Services Division

* Notes and references may be found on page 4.

Table 1. Properties of SRM 4330C

Certified values

Radionuclide	**Plutonium-239**
Reference time	**1200 EST, 1 May 2009**
Massic activity of the solution	**38.41 Bq•g^{-1}**
Relative expanded uncertainty ($k = 2$)	**0.46 %** (see Note 2)*

Uncertified information

Source description	Liquid in a flame-sealed 5 mL borosilicate-glass ampoule (see Note 1)
Solution composition	3.4 mol•L^{-1} HNO_3
Solution density	(1.1082 ± 0.002) g•mL^{-1} at 23.9 °C (see Note 3)
Solution mass	(2.7707 ± 0.0003) g (see Note 3)
Photon-emitting impurities	None detected (see Note 4)
Half-lifes used	^{239}Pu: (24100 ± 11) a (see Note 5) [1]
Calibration methods (and instruments)	The certified massic activity for ^{239}Pu was obtained by 4πα liquid scintillation (LS) spectrometry with three commercial LS counters.

* Notes and references may be found on page 4.

Table 2. Uncertainty evaluation for the massic activity of SRM 4330C

	Uncertainty component	Assessment Type †	Relative standard uncertainty contribution to massic activity of ^{239}Pu (%)
1	LS measurement precision; standard deviation for $n = 9$ mean determinations as obtained with 3 different cocktail compositions (5 sources of each composition) measured 3 times in 3 different LS counters (135 determinations in all). See Note 6*. Data passes normality test at 95 % and 99 %. The typical internal relative standard deviation of the mean (n =15 for the 5 sources measured 3 times) for each of the 9 determinations ranged from 0.06 % to 0.10%.	A	0.17
2	Background LS measurement variability; wholly embodied in component 1	A	---
3	Gravimetric (mass) determinations for LS sources; estimated from calibration data and tests.	B	0.07
4	Decay correction due to the decay time interval and 0.046 % uncertainty in half-life.	B	5×10^{-8}
5	Live time determinations for LS counting time intervals, includes uncorrected dead time effects; assumed from specified tolerance limits of counters' gated oscillators	B	0.1
6	LS detection efficiency, including wall effects loss and extrapolation to zero energy; estimated from nuclear data and tests.	B	0.07
7	Alpha decay probability; assumed from nuclear decay data based on % of spontaneous fission	B	3×10^{-10}
8	LS non-detection of 26 min $^{235}U^{m}$; estimated from nuclear data and tests of LS counter detection threshold	B	< 0.001
9	Correction for ingrowth of ^{235}U; estimated from nuclear data	B	3×10^{-8}
10	Alpha-particle emitting impurities; based on nuclear data and mass spectrometric measurements of supplier. (See Note 7)	B	0.06
11	Photon-emitting impurities (non-detected). See Note 4.	B	---
Relative combined standard uncertainty			**0.23**
Relative expanded uncertainty ($k = 2$)			**0.46**

† = (A) denotes evaluation by statistical methods; (B) denotes evaluation by other methods.

* Notes and references may be found on page 4.

Note 1. Refer to http://physics.nist.gov/Divisions/Div846/srm.html for assistance and instructions on how to properly open an ampoule. Information on additional storage and handling requirements is also included on the website. This SRM is contained in a generic borosilicate-glass ampoule and not in the standard NIST ampoule.

Note 2. The uncertainties on certified values are expanded uncertainties, $U = ku_c$. The quantity u_c is the combined standard uncertainty calculated according to the ISO and NIST Guides [2-3]. The combined standard uncertainty is multiplied by a coverage factor of $k = 2$ and was chosen to obtain an approximate 95 % level of confidence.

Note 3. The stated uncertainty is two times the standard uncertainty. See reference [3].

Note 4. The estimated lower limit of detection for photon-emitting impurities, expressed as massic photon emission rate, on 1 May 2009 is:

$0.64\ s^{-1} \cdot g^{-1}$ for energies between 40 keV and 50 keV,
$0.17\ s^{-1} \cdot g^{-1}$ for energies between 50 keV and 250 keV, and
$0.13\ s^{-1} \cdot g^{-1}$ for energies between 250 keV and 2600 keV.

provided that the photons are separated in energy by 4 keV or more from photons emitted in the decay of ^{239}Pu or progeny.

Note 5. The stated uncertainty is the standard uncertainty. See reference [3].

Note 6. The mean was found to be invariant of: (i) cocktail composition based on scintillation fluid used and the aqueous fraction, (ii) aliquant mass in cocktails, (iii) sample quenching; and (iv) instrument (with different detection thresholds).

Note 7. From mass spectrometric measurements performed by the supplier, the non-certified massic activities of other detected radionuclides (in $Bq \cdot g^{-1}$ as of 1200 EST, 15 November 1999) are: $^{240}Pu = 0.002$, $^{241}Pu = 0.02$, $^{241}Am = 0.001$. Solution was purified 10 December 1979.

REFERENCES

[1] E. Browne, *Nuclear Data Sheets* 98, 665 (2003), Evaluated Nuclear Structure Data File (ENSDF), online database, National Nuclear Center, Brookhaven National Laboratory (Upton, NY), accessed May 2009. Refer to http://www.nndc.bnl.gov/ensdf/.

[2] International Organization for Standardization (ISO), *Guide to the Expression of Uncertainty in Measurement*; 1993 (corrected and reprinted, 1995). Ordering and purchasing information available at http://physics.nist.gov/cuu/Uncertainty/isoorder.html.

[3] B.N. Taylor and C.E. Kuyatt; *Guidelines for Evaluating and Expressing the Uncertainty of NIST Measurement Results,* NIST Technical Note 1297, 1994. Available at http://physics.nist.gov/Pubs/guidelines/contents.html.

Certificate Revision History: 17 July 2009 (Original certificate date).

Users of this SRM should ensure that the certificate in their possession is current. This can be accomplished by contacting the SRM Program at: telephone (301) 975-2200; fax (301) 926-4751; e-mail srminfo@nist.gov; or via the Internet at http://www.nist.gov/srm.

National Institute of Standards & Technology

Certificate

Standard Reference Material® 4323c

Plutonium-238 Radioactivity Standard

This Standard Reference Material (SRM) consists of a solution of a standardized and certified quantity of radioactive plutonium-238 in a suitably stable and homogeneous matrix. It is intended primarily for the calibration of instruments that are used to measure radioactivity and for the monitoring of radiochemical procedures. A unit of SRM 4323c consists of approximately 5 mL of a solution, whose composition is specified in Tables 1 and 2, contained in a flame-sealed borosilicate-glass ampoule [1].

The certified **plutonium-238** massic activity, at a **Reference Time of 1200 EST, 11 October 2016**, is:

(22.73 ± 0.11) Bq•g^{-1}

A NIST certified value, as used within the context of this certificate, is a value for which NIST has the highest confidence in its uncertainty assessment. It is a "measurement result" [2] obtained directly or indirectly from a "primary reference measurement procedure" [3]. The certified value is traceable to the derived SI unit, becquerel (Bq).

Additional physical, chemical, and radiological properties for this SRM, as well as details on the standardization method, are given in Tables 1 and 2. Uncertainties for the certified quantities are expanded ($k = 2$). The uncertainties are calculated according to the ISO/JCGM and NIST Guides [4,5]. Table 3 contains a specification of the components that comprise the uncertainty analysis.

Expiration of Certification: The certification of **SRM 4323c** is valid indefinitely, within the measurement uncertainty specified, provided that the SRM is handled and stored properly and that no evaporation or change in composition has occurred. The solution matrix, in an unopened ampoule, is homogeneous and stable within its half-life-dependent useful lifetime provided the SRM is handled in accordance with instructions given in this certificate (see "Instructions for Handling and Storage"). Periodic recertification of this SRM is not required. The certification is nullified if the SRM is damaged, contaminated, or otherwise modified.

Maintenance of Certification: NIST will monitor this SRM over the period of its certification. If substantive technical changes occur that affect the certification, NIST will notify the purchaser. Registration (see attached sheet or register online) will facilitate notification.

Radiological and chemical hazard: Consult the Safety Data Sheet (SDS), enclosed with the SRM shipment, for radiological and chemical hazard information.

This SRM was prepared in the NIST Physical Measurement Laboratory, Radiation Physics Division, under the direction of M.P. Unterweger, Group Leader of the Radioactivity Group. The overall technical direction and physical measurement leading to certification were provided by R. Collé and L. Laureano-Perez of the NIST Radiation Physics Division, Radioactivity Group. All additional technical support was provided by members of the NIST Radiation Physics Division, Radioactivity Group. Confirmatory measurements by J. LaRosa and S. Nour were performed by isotope dilution analysis with alpha-particle spectrometry. Homogeneity evaluations were assisted by D. Bergeron. Alpha-emitting-impurity and photon-emitting-impurity analyses were provided by J. LaRosa and S. Nour, and L. Pibida, respectively.

Support aspects involved in the issuance of this SRM were coordinated through the NIST Office of Reference Materials.

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Certificate Issue Date: 1 January 2017

Steven J. Choquette, Director
Office of Reference Materials

Table 1. Certified Massic Activity of SRM 4323c

Radionuclide	**Plutonium-238**
Reference time	**1200 EST, 11 October 2016**
Massic activity of the solution	**22.73 Bq•g^{-1}**
Relative expanded uncertainty ($k = 2$)	**0.48 %**[a]

[a] The uncertainties on certified values are expanded uncertainties, $U = ku_c$. The quantity u_c is the combined standard uncertainty calculated according to the ISO/JCGM and NIST Guides [4,5]. The combined standard uncertainty is multiplied by a coverage factor of $k = 2$ and was chosen to obtain an approximate 95 % level of confidence.

Table 2. Uncertified Information of SRM 4323c

Source description	Liquid in a flame-sealed 5 mL borosilicate-glass ampoule [1]
Solution composition	(2.85 ± 0.04) mol•L^{-1} HNO_3[a]
Solution density	(1.099 ± 0.001) g•mL^{-1} at 21 °C[a]
Solution mass	(5.511 ± 0.003) g[a]
Photon-Emitting Impurities	None detected[b]
Alpha-emitting Impurities	None detected[c]
Half-lives used	^{238}Pu: (87.74 ± 0.03) a [d] [6]
Calibration methods (and instruments)	The certified value was determined by liquid scintillation (LS) spectrometry with two LS commercial counters using six cocktail compositions (each with three counting sources) in sixteen separate measurement trials. Confirmatory measurements were performed by isotope dilution analysis with alpha-particle spectrometry. [e]

[a] The stated uncertainty is two times the standard uncertainty. See reference 5.

[b] The estimated lower limits of detection for photon-emitting impurities, expressed as massic photon emission rate, in May 2016, were:

0.24 s^{-1}•g^{-1} in the region 15 keV ≤ E ≤ 30 keV;
0.04 s^{-1}•g^{-1} in the region 35 keV ≤ E ≤ 1430 keV;
0.05 s^{-1}•g^{-1} in the region 1440 keV ≤ E ≤ 1480 keV, and
0.03 s^{-1}•g^{-1} in the region 1490 keV ≤ E ≤ 2000 keV;

[c] The massic activity of ^{239}Pu at the Reference Time is estimated to be 0.00012 Bq•g^{-1}, based on a ^{239}Pu/^{238}Pu atom ratio of 0.00107 as obtained from mass spectrometry by Oak Ridge National Laboratory in 1981 for the stock material used to prepare SRM 4323c. The estimated lower limits of detection for alpha -emitting impurities, expressed as massic alpha emission rate, in June 2016, were:

0.003 s^{-1}•g^{-1} in the region $E \leq 4580$ keV;
0.007 s^{-1}•g^{-1} in the region 5020 keV ≤ E ≤ 5280 keV; and
0.0001 s^{-1}•g^{-1} in the region $E \geq 5630$ keV.

[d] The stated uncertainty is the standard uncertainty. See reference 5.

[e] Aliquants of SRM 4323c were taken and spiked with known activities of ^{242}Pu from SRM 4334h to radiochemically prepare thin counting sources for the alpha spectrometry measurements. The result for the ^{238}Pu massic activity in SRM 4323c from this isotope dilution analysis was (22.71 ± 0.27) Bq•g^{-1} at $k = 2$, and agreed with the LS-based certified value to within 0.06 %.

Table 3. Uncertainty Evaluation for the Massic Activity of SRM 4323c

	Uncertainty component	Assessment Type[a]	Relative standard uncertainty contribution on massic activity of ^{238}Pu (%)
1	LS measurement precision: Relative standard deviation of the mean on the great-grand mean for 16 LS measurement trails, considering all of the within-trial and between-trial components of variance. Each of the 16 grand mean values was based on 5 replicate measurements on each of 3 LS counting sources. The typical within-trial relative standard deviation of the mean (considering the variations for the between 5 measurements and the between 3 sources) for each trial was 0.12 %. The between-trial relative standard deviation across the 16 trials was 0.17 %. The 16 data values fit a Normal distribution.	A	0.21
2	Background; LS measurement variability and cocktail composition stability effects; wholly embodied in component 1.	A	--
3	LS counters dependencies; wholly embodied in components 1 & 2	A	--
4	Live time determinations for LS counting time intervals, includes uncorrected dead time effects	B	0.07
5	Aliquant mass determinations by gravimetric measurements for preparation of counting sources; includes mass measurement precision partially embodied in component 1.	B	0.05
6	LS detection inefficiency, includes wall effect; partially embodied in component 1.	B	0.01
7	^{238}Pu decay corrections for half-life uncertainty of 0.034 %.	B	< 0.0002
8	Potential alpha- and photon-emitting impurities	B	0.1
Relative combined standard uncertainty			**0.25**
Relative expanded uncertainty ($k = 2$)			**0.50**

[a] Letter A, denotes evaluation by statistical methods; B denotes evaluation by other methods [4,5].

INSTRUCTIONS FOR HANDLING AND STORAGE

Handling: If the ampoule is transported, it should be packed, marked, labeled, and shipped in accordance with the applicable national, international, and carrier regulations. The solution in the ampoule is a dangerous good (hazardous material) because of the radioactivity. Only persons qualified to handle radioactive material should open the ampoule. To minimize personnel exposure, appropriate shielding and/or distance should be used. Refer to the SDS for further information.

Storage: SRM 4323c should be stored and used at a temperature between 5 °C and 65 °C. The ampoule (or any subsequent container) should always be clearly marked as containing radioactive material.

REFERENCES

[1] NIST Physical Measurement Laboratory; *Storage and Handling of Radioactive Standard Reference Materials, Ampoule Specifications and Opening Procedure*; available at https://www.nist.gov/pml/radiation-physics/ampoule-specifications-and-opening-procedure (accessed Jan2017). Note: This SRM is contained in a generic borosilicate-glass ampoule and not in the standard NIST ampoule.

[2] JCGM 200:2012; *International Vocabulary of Metrology - Basic and General Concepts and Associated Terms (VIM)*; (2008 version with Minor Corrections), 3rd edition; Joint Committee for Guides in Metrology (JCGM): BIPM, Sevres Cedex, France; p. 19 (2012); available at http://www.bipm.org/utils/common/documents/jcgm/JCGM_200_2012.pdf (accessed Sep 2015).

[3] JCGM 200:2012; *International Vocabulary of Metrology - Basic and General Concepts and Associated Terms (VIM)*; (2008 version with Minor Corrections), 3rd edition; JCGM: BIPM, Sevres Cedex, France; p. 18 (2012); available at http://www.bipm.org/utils/common/documents/jcgm/JCGM_200_2012.pdf (accessed Sep 2015).

[4] JCGM 100:2008; *Guide to the Expression of Uncertainty in Measurement*; (GUM 1995 with Minor Corrections), JCGM: BIPM, Sevres Cedex, France (2008); available at http://www.bipm.org/utils/common/documents/jcgm/JCGM_100_2008_E.pdf (accessed Jan2017)

[5] Taylor, B.N.; Kuyatt, C.E.; *Guidelines for Evaluating and Expressing the Uncertainty of NIST Measurement Results*; NIST Technical Note 1297, U.S. Government Printing Office: Washington, DC (1994); available at https://www.nist.gov/pml/productsservices/special-publications-tutorials (accessed Jan2017).

[6] Chechev, V.P.; *LNE-LNHB/CEA Table of Radionuclides, ^{238}Pu*; (June 2009); available at http://www.nucleide.org/DDEP_WG/Nuclides/Pu-238_com.pdf (accessed June 2016).

Users of this SRM should ensure that the Certificate in their possession is current. This can be accomplished by contacting the SRM Program: telephone (301) 975-2200; fax (301) 948-3730; e-mail srminfo@nist.gov; or via the Internet at http://www.nist.gov/srm.

National Institute of Standards & Technology

Certificate

Standard Reference Material® 4334j

Plutonium-242 Radioactivity Standard

This Standard Reference Material (SRM) consists of a solution of a standardized and certified quantity of radioactive plutonium-242 in a suitably stable and homogeneous matrix. It is intended primarily for the calibration of instruments that are used to measure radioactivity and for the monitoring of radiochemical procedures. A unit of SRM 4334j consists of approximately 5 mL of a solution, whose composition is specified in Tables 1 and 2, contained in a flame-sealed borosilicate-glass ampoule [1].

The certified **Plutonium-242** massic activity, at a **Reference Time of 1200 EST, 9 August 2017**, is:

(26.08 ± 0.13) Bq•g^{-1}.

A NIST certified value, as used within the context of this certificate, is a value for which NIST has the highest confidence in its uncertainty assessment. It is a "measurement result" [2] obtained directly or indirectly from a "primary reference measurement procedure" [3]. The certified value is traceable to the derived SI unit, becquerel (Bq).

Additional physical, chemical, and radiological properties for this SRM, as well as details on the standardization method, are given in Tables 1 and 2. Uncertainties for the certified quantities are expanded ($k = 2$). The uncertainties are calculated according to the ISO/JCGM and NIST Guides [4,5]. Table 3 contains a specification of the components that comprise the uncertainty analysis.

Expiration of Certification: The certification of **SRM 4334j** is valid indefinitely, within the measurement uncertainty specified, provided that the SRM is handled and stored properly and that no evaporation or change in composition has occurred. The solution matrix, in an unopened ampoule, is homogeneous and stable within its half-life-dependent useful lifetime provided the SRM is handled in accordance with instructions given in this certificate (see "Instructions for Handling and Storage"). Periodic recertification of this SRM is not required. The certification is nullified if the SRM is damaged, contaminated, or otherwise modified.

Maintenance of Certification: NIST will monitor this SRM over the period of its certification. If substantive technical changes occur that affect the certification before the expiration of this certificate, NIST will notify the purchaser. Registration (see attached sheet) will facilitate notification.

Radiological and chemical hazard: Consult the Safety Data Sheet (SDS), enclosed with the SRM shipment, for radiological and chemical hazard information.

This SRM was prepared in the NIST Physical Measurement Laboratory, Radiation Physics Division, under the direction of M.P. Unterweger, Group Leader of the Radioactivity Group. Overall technical direction and physical measurement leading to certification were provided by R. Collé and L. Laureano-Perez of the NIST Radiation Physics Division, Radioactivity Group. Photon-emitting-impurity analyses were provided by L. Pibida.

Support aspects involved in the issuance of this SRM were coordinated through the NIST Office of Reference Materials.

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Gaithersburg, Maryland 20899
Certificate Issue Date: 24 January 2018

Steven J. Choquette, Director
Office of Reference Materials

Table 1. Certified Massic Activity of SRM 4334j

Radionuclide	**Plutonium-242**
Reference time	**1200 EST, 9 August 2017**
Massic activity of the solution	**26.08 $Bq \cdot g^{-1}$ [(a)]**
Relative expanded uncertainty ($k = 2$)	**0.51 % [(b)]**

(a) Both SRM 4334j and 4334i (a previous issue of ^{242}Pu) were derived from gravimetric dilution of the identical standard master solution. The massic activity of SRM 4334j is in agreement with the decay corrected massic activity of SRM 4334i to ± 0.12 %.

(b) The uncertainties on certified values are expanded uncertainties, $U = ku_c$. The quantity u_c is the combined standard uncertainty calculated according to the ISO and NIST Guides [4-5]. The combined standard uncertainty is multiplied by a coverage factor of $k = 2$ and was chosen to obtain an approximate 95 % level of confidence

Table 2. Uncertified Information of SRM 4334j

Source description	Liquid in a flame-sealed 5 mL borosilicate-glass ampoule [1]
Solution composition	3.1 mol•L^{-1} HNO_3
Solution density	(1.099 ± 0.002) g•mL^{-1} at 24.4 °C [(a)]
Solution mass	5.499 ± 0.003 g
Alpha-particle- emitting impurities	^{241}Am: (0.0021 ± 0.0003) Bq•g^{-1} [(a, b, c, d)]
Beta-particle- emitting impurities	^{241}Pu: (0.039 ± 0.009) Bq•g^{-1} [(a, d)]
Photon-emitting impurities	None detected [(e)]
Half-lifes used [6]	^{242}Pu: (3.73 ± 0.03) × 10^5 a [(f)] ^{241}Pu: (14.33 ± 0.04) a ^{240}Pu: (6 561 ± 7) a ^{239}Pu: (24 100 ± 11) a ^{238}Pu: (87.74 ± 0.03) a ^{241}Am: (432.6 ± 0.6) a
Calibration methods (and instruments)	The certified massic activity for ^{242}Pu was obtained by 4πα liquid scintillation (LS) spectrometry with two commercial LS counters. Four separate measurement trials using nine LS cocktails prepared directly from the SRM solution and six cocktails prepared from a master stock solution with known gravimetric dilution factor.

(a) The stated uncertainty is two times the standard uncertainty. [5].

(b) The ^{242}Pu was chemically purified 7 June 1994 at the Lawrence Livermore National Laboratory (LLNL). Americium-241, the daughter of ^{241}Pu, was removed but has been growing in since that time. Photonic emission measurements of the ^{241}Am ingrowth were made at NIST in 1998-1999 and 2017.

(c) The estimated limits of detection for alpha-particle-emitting impurities, expressed as massic alpha-particle emission rates (number of alpha-particles emission rates per second per gram), are:

0.003 s^{-1}•g^{-1} for energies less than 3.1 MeV,
0.03 s^{-1}•g^{-1} for energies between 3.1 MeV and 4.4 keV, and
0.003 s^{-1}•g^{-1} for energies greater than 5.0 MeV

(d) The ^{242}Pu was chemically purified 7 June 1994. The relative massic activities of radionuclidic impurities follow;

Radionuclide	Relative Activity at Purification Time (7 June 1994) As Measured By		
	LLNL in 1994	NIST in 1998-1999	NIST in 2017
^{242}Pu	1	1	1
^{241}Pu	--	(3.5 ± 0.4) × 10^{-3} [(1,2)]	(3.6 ± 1.1) × 10^{-3} [(1,5)]
^{240}Pu + ^{239}Pu	< 10^{-6} [(3)]	(2.0 ± 2.1) × 10^{-5} [(1,4)]	--
^{238}Pu + ^{241}Am	< 1.6 × 10^{-5} [(3)]	(9 ± 16) × 10^{-6} [(1,4)]	--
^{241}Am	--	assumed 0 [(2)]	assumed 0 [(5)]

1) The stated uncertainty is the standard uncertainty.
2) The ^{241}Pu activity was calculated from a gamma-ray measurement of the ^{241}Am ingrowth as of 25 November 1998, assuming that ^{241}Am was completely removed at the time of chemical purification.
3) Using alpha-particle spectrometry. The value shown is an estimated upper limit based upon background and counting statistics. Measurements were made at the Lawrence Livermore National Laboratory (LLNL) in July of 1994.
4) Alpha-particle spectrometry measurements were made at the National Institute of Standards and Technology (NIST) in June and July 1999.
5) The ^{241}Pu activity was calculated from a gamma-ray measurement of the ^{241}Am ingrowth as of 01 September 2017, assuming that ^{241}Am was completely removed at the time of chemical purification.

(e) The estimated limits of detection for photon-emitting impurities, expressed as massic photon emission rates (numbers of photons per second per gram), are:

1 × 10^{-3} s^{-1}•g^{-1} for energies between 20 keV and 35 keV,
6 × 10^{-4} s^{-1}•g^{-1} for energies between 40 keV and 50 keV,
5 × 10^{-4} s^{-1}•g^{-1} for energies between 55 keV and 95 keV,
4 × 10^{-4} s^{-1}•g^{-1} for energies between 100 keV and 600 keV
4 × 10^{-4} s^{-1}•g^{-1} for energies between 610 keV and 1440 keV
6 × 10^{-4} s^{-1}•g^{-1} for energies between 1450 keV and 1480 keV, and
3 × 10^{-4} s^{-1}•g^{-1} for energies between 1490 keV and 2000 keV.

provided that the photons are separated in energy by 4 keV or more from photons emitted in the decay of ^{242}Pu, ^{241}Pu, or ^{241}Am.

(f) The stated uncertainty is the standard uncertainty. See reference [6].

Table 2. Uncertainty evaluation for the massic activity of SRM 4334j

	Uncertainty component	Assessment Type[(a)]	Relative standard uncertainty contribution on massic activity of ^{242}Pu (%)
1	LS measurement precision: Relative standard deviation of the mean on the great-grand mean for 4 LS measurement trials, considering all of the within-trial and between-trial components of variance. Each of the 4 grand mean values was based on 5 replicate measurements on each of either 6 or 9 LS counting sources. The typical within-trial relative standard deviation of the mean (considering the variations for the between 5 measurements and the between 6 to 9 sources) for each trial was 0.08 %. The between-trial relative standard deviation across the 4 trials was 0.18 %.	A	0.22
2	Background; LS measurement variability and cocktail composition stability effects; wholly embodied in component 1.	A	--
3	LS counters dependencies; wholly embodied in components 1 & 2	A	--
4	Live time determinations for LS counting time intervals, includes uncorrected dead time effects	B	0.07
5	Aliquant mass determinations by gravimetric measurements for preparation of counting sources; includes mass measurement precision partially embodied in component 1.	B	0.05
6	LS detection inefficiency, includes wall effect; partially embodied in component 1.	B	0.01
7	^{242}Pu decay corrections for half-life uncertainty of 0.22 %.	B	$< 10^{-10}$
8	Potential alpha- and photon-emitting impurities	B	0.1
Relative combined standard uncertainty			**0.26**
Relative expanded uncertainty ($k = 2$)			**0.51**

(a) = (A) denotes evaluation by statistical methods; (B) denotes evaluation by other methods.

INSTRUCTIONS FOR USE AND HANDLING

Storage: SRM 4334j should be stored and used at a temperature between 5 °C and 65 °C. The ampoule (or any subsequent container) should always be clearly marked as containing radioactive material.

Handling: If the ampoule is transported, it should be packed, marked, labeled, and shipped in accordance with the applicable national, international, and carrier regulations. The solution in the ampoule is a dangerous good (hazardous material) because of both the radioactivity and the strong acid. The ampoule should be opened only by persons qualified to handle both radioactive material and alkaline and/or acidic solutions. Appropriate shielding and/or distance should be used to minimize personnel exposure. Refer to MSDS for further information.

REFERENCES

[1] NIST Physical Measurement Laboratory; *Storage and Handling of Radioactive Standard Reference Materials, Ampoule Specifications and Opening Procedure*; available at https://www.nist.gov/pml/radiation-physics/ampoule-specifications-and-opening-procedure (accessed Nov 2017). Note: This SRM is contained in a generic borosilicate-glass ampoule and not in the standard NIST ampoule.

[2] JCGM 200:2012; *International Vocabulary of Metrology - Basic and General Concepts and Associated Terms (VIM)*; (2008 version with Minor Corrections), 3rd edition; Joint Committee for Guides in Metrology (JCGM): BIPM, Sevres Cedex, France; p. 19 (2012); available at http://www.bipm.org/utils/common/documents/jcgm/JCGM_200_2012.pdf (accessed Nov 2017).

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[4] JCGM 100:2008; *Guide to the Expression of Uncertainty in Measurement*; (GUM 1995 with Minor Corrections), JCGM: BIPM, Sevres Cedex, France (2008); available at http://www.bipm.org/utils/common/documents/jcgm/JCGM_100_2008_E.pdf (accessed Nov 2017).

[5] Taylor, B.N.; Kuyatt, C.E.; *Guidelines for Evaluating and Expressing the Uncertainty of NIST Measurement Results*; NIST Technical Note 1297, U.S. Government Printing Office: Washington, DC (1994); available at https://www.nist.gov/pml/productsservices/special-publications-tutorials (accessed Nov 2017).

[6] Chechev, V.P.; *LNE-LNHB/CEA Table of Radionuclides*, ^{242}Pu; (June 2009); available at http://www.nucleide.org/DDEP_WG/Nuclides/Pu-242_tables.pdf (accessed Nov 2017).

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Published papers describing experimental designs, data reductions and uncertainty evaluations for Radioactivity SRMs

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