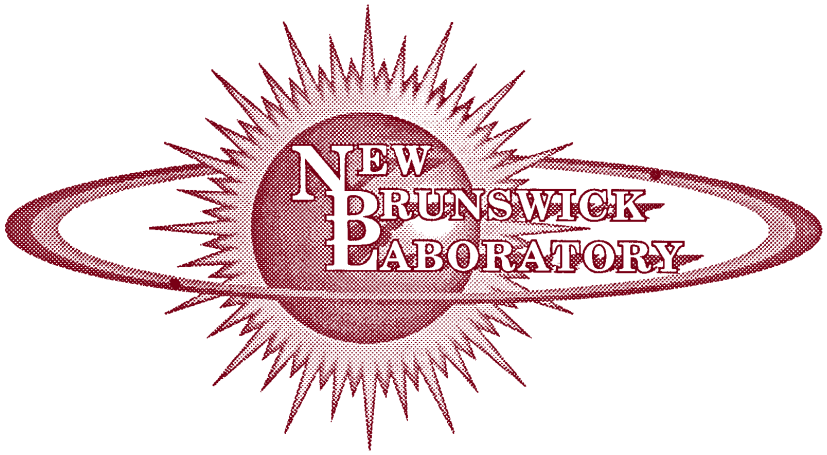
NBL-374
JULY 2003

MINUTES OF THE MEASUREMENT EVALUATION PROGRAM MEETING

URANIUM SAMPLE EXCHANGE
PLUTONIUM SAMPLE EXCHANGE
CALORIMETRY EXCHANGE



July 13, 2003

Edited by Jay M. Thompson

U.S. DEPARTMENT OF ENERGY · CHICAGO OPERATIONS OFFICE · ARGONNE, ILLINOIS

Department of Energy
New Brunswick Laboratory
Measurement Evaluation Program

Agenda

Morning of July 13, 2003
Pinnacle Room #2

Safeguards Measurement Evaluation Program
and
Calorimetry Exchange Program

8:30 AM	Sign in
9:00 AM	Welcome and Introductions (Jon Neuhoff, New Brunswick Laboratory)
9:15 AM	Summary of 2002 Safeguards Measurement Evaluation Program Results (Jay Thompson, New Brunswick Laboratory)
10:00 AM	Break
10:15 AM	Summary of 2002 Calorimetry Exchange Program Results (Jay Thompson, New Brunswick Laboratory)
10:45 AM	Status of the SO-13 Evaluation of Safeguards NDA Systems Project (Ray Dewberry/Saleem Salaymeh, Westinghouse Savannah River Company)
11:00 AM	Experiences with Reference Materials (Mike Michlik, Argonne National Laboratory – West)
11:15 AM	Status of Reference Material Production (Jon Neuhoff, New Brunswick Laboratory)
11:45AM	Discussion and session wrap-up

---------Break for Lunch--------

Department of Energy
New Brunswick Laboratory
Measurement Evaluation Program

Agenda

Afternoon of July 13, 2003
Pinnacle Room #2

Workshop on NDA Standards and Calibration – Part I*

This two-part workshop is being conducted in accordance with the Memorandum of Agreement on Nondestructive Assay Standards and Calibration Support between SO, NBL, LANL, and LLNL. Other facilities are welcome to attend.

12:45 PM	Calorimetry (Cliff Rudy, Los Alamos National Laboratory)
1:45 PM	Break
2:00 PM	Uranium Enrichment Measurement (Doug Reilly, Los Alamos National Laboratory)
3:00 PM	Break
3:15 PM	Portable In-Situ Gamma (Phyllis Russo, Los Alamos National Laboratory)
4:45 PM	Meeting wrap-up and closing remarks

*** Part II of this workshop will be held at Los Alamos on August 19-20, 2003. Contact Phyllis Russo (prusso@lanl.gov) or Bill Geist (wgeist@lanl.gov) for details.**

NEW BRUNSWICK LABORATORY

SAFEGUARDS MEASUREMENT EVALUATION PROGRAM

Annual Meeting
July 13, 2003

Jay Thompson

TABLE 1

URANIUM SAMPLE EXCHANGE PARTICIPATING FACILITIES
ARGONNE NATIONAL LABORATORY–WEST
LOS ALAMOS NATIONAL LABORATORY
NEW BRUNSWICK LABORATORY
SAVANNAH RIVER SITE
TOKAI SAFEGUARDS ANALYTICAL LABORATORY
Y-12 NATIONAL SECURITY COMPLEX

TABLE 2

PLUTONIUM ISOTOPIC EXCHANGE PARTICIPATING FACILITIES
NEW BRUNSWICK LABORATORY
TOKAI SAFEGUARDS ANALYTICAL LABORATORY

TABLE 3
LABORATORY PARTICIPATION FOR FISCAL YEAR 2002
BY MATERIAL AND MEASUREMENT METHOD

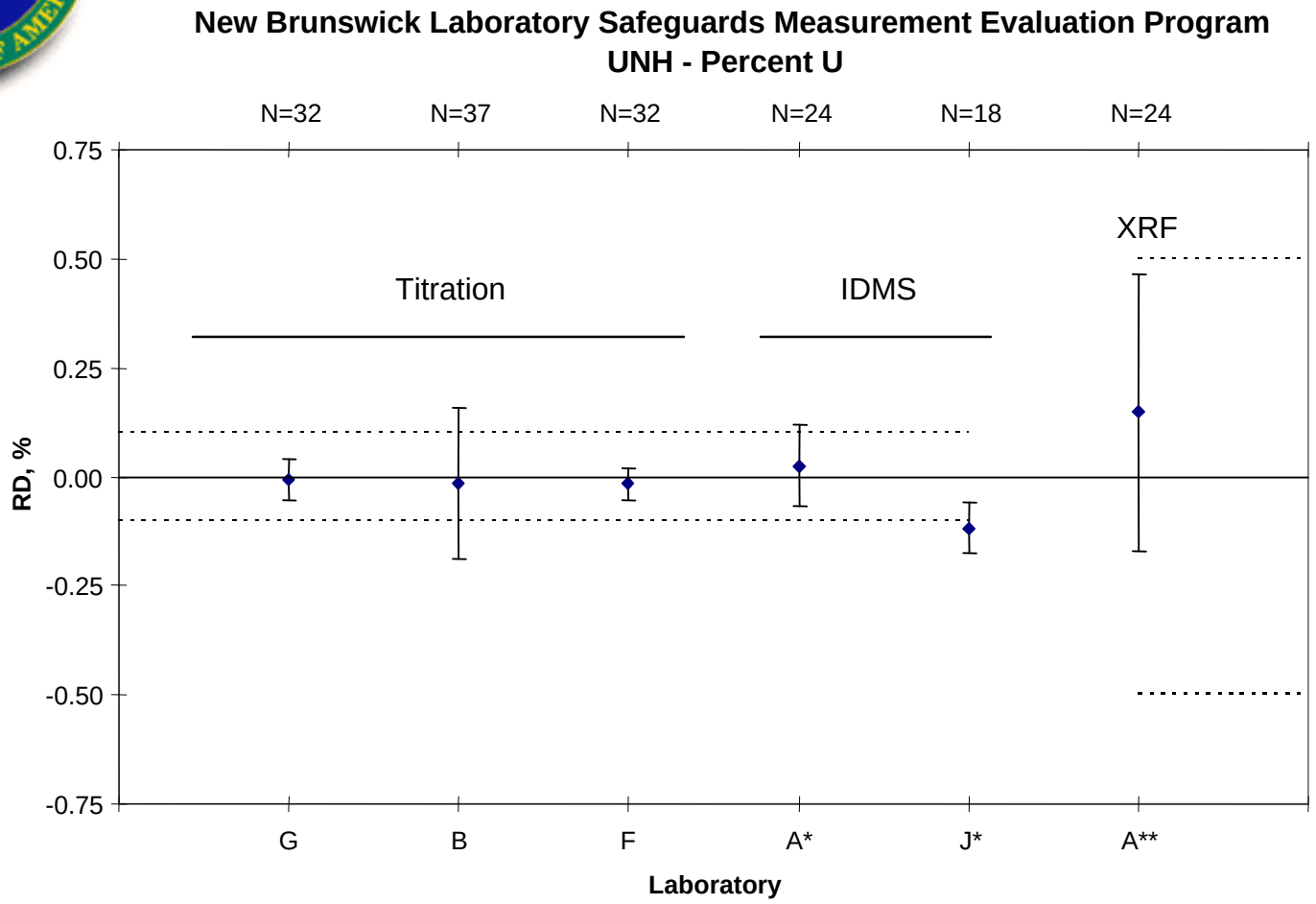
Table Entries are Facility Codes with the Number of Times Participated in Fiscal Year 2002

UPPER Portion of this Table Shows Methods and Materials for Assay Measurements
LOWER Portion of this Table Shows Methods and Materials for Isotopic Measurements

Method	UNH Solutions	UO_2 Pellets	UO_3 Powder	UF_6	Pu Sulfate
Dichromate Titration	B4 F2	F1 T2	F1		
Ceric Titration	G4				
U IDMS	A3 J1		A4		
X-Ray Fluorescence	A3		A8		
Pu IDMS					F1
TIMS LEU	A1	F1 T2	F1	F1	
HEU	A3 F1 J1				
Pu					F1 T2

Table 4
Interlaboratory Performance Summary
UNH - Percent U

Method	Lab code	Mean	Standard deviation	N
Ceric Titration	G	-0.006	0.048	32
Davies-Gray Titration	B	-0.014	0.174	37
	F	-0.016	0.037	32
IDMS	A*	0.025	0.094	24
	J*	-0.118	0.058	18
X-Ray Fluorescence	A**	0.147	0.319	24


New Brunswick Laboratory Safeguards Measurement Evaluation Program
UNH - Percent U
N=32
N=37
N=32
N=24
N=18
N=24
0.75
0.50
0.25
0.00
-0.25
-0.50
-0.75
RD, %
Titration
IDMS
XRF
G
B
F
A*
J*
A**
Laboratory
Laboratory Mean
Bias Target Values
Standard Deviation

New Brunswick Laboratory Safeguards Measurement Evaluation Program
UNH - Percent U

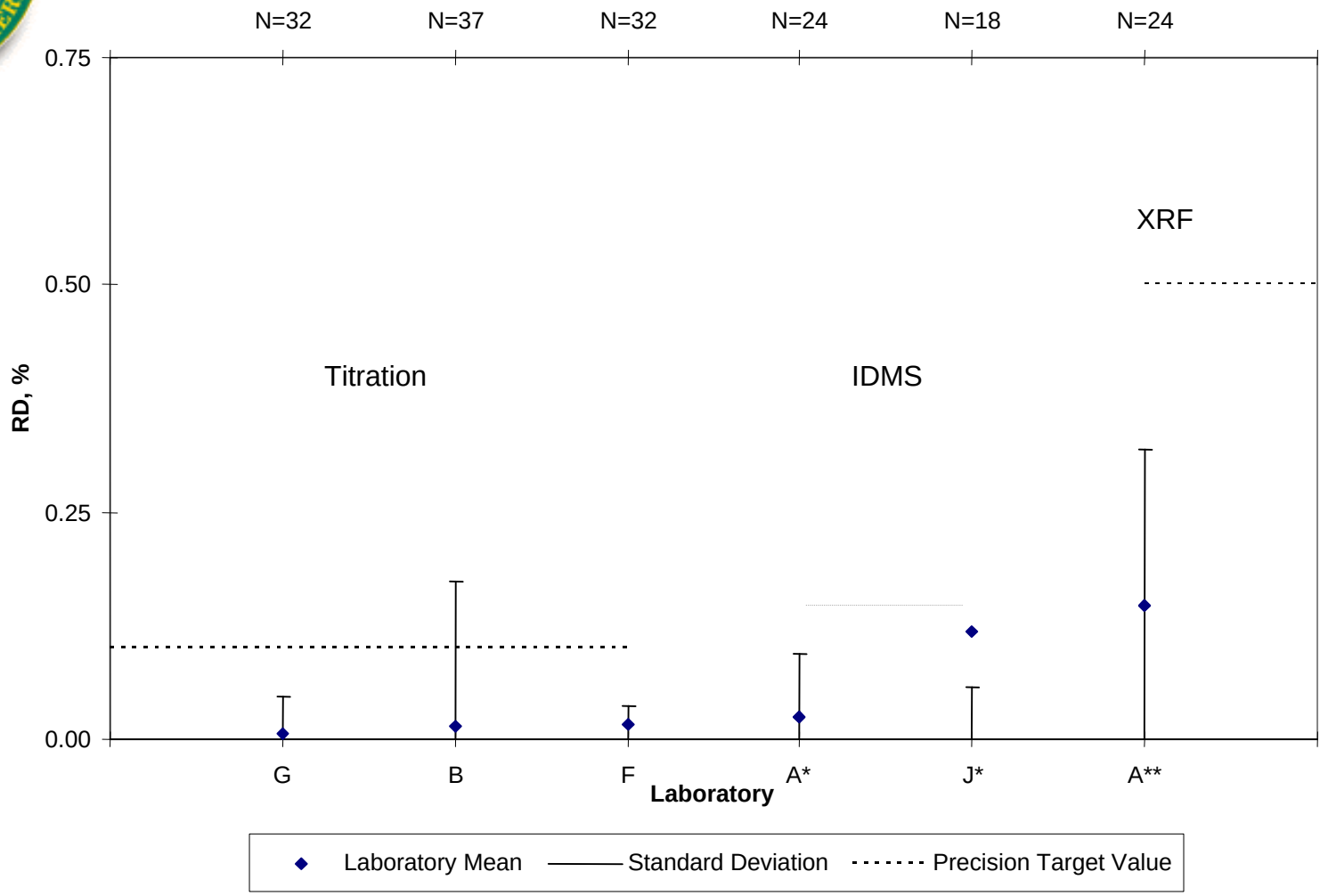
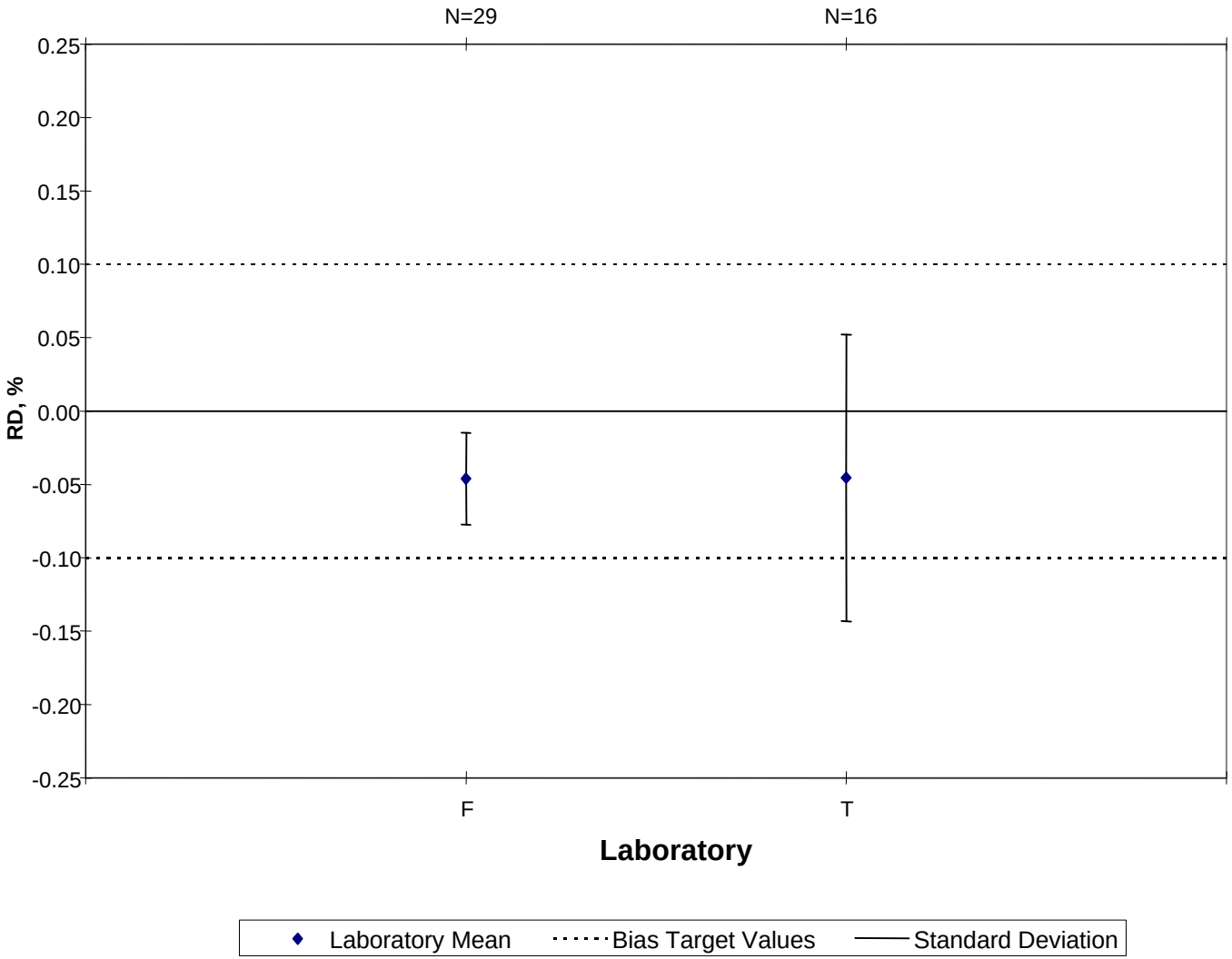


Table 5
Interlaboratory Performance Summary
UO_2 Pellets - Percent U

Method	Lab code	Mean	Standard deviation	N
Davies-Gray Titration	F	-0.046	0.031	29
	T	-0.046	0.098	16

New Brunswick Laboratory Safeguards Measurement Evaluation Program
UO2 Pellets - Percent U by Davies and Gray Titration



New Brunswick Laboratory Safeguards Measurement Evaluation Program
UO2 Pellets - Percent U by Davies and Gray Titration

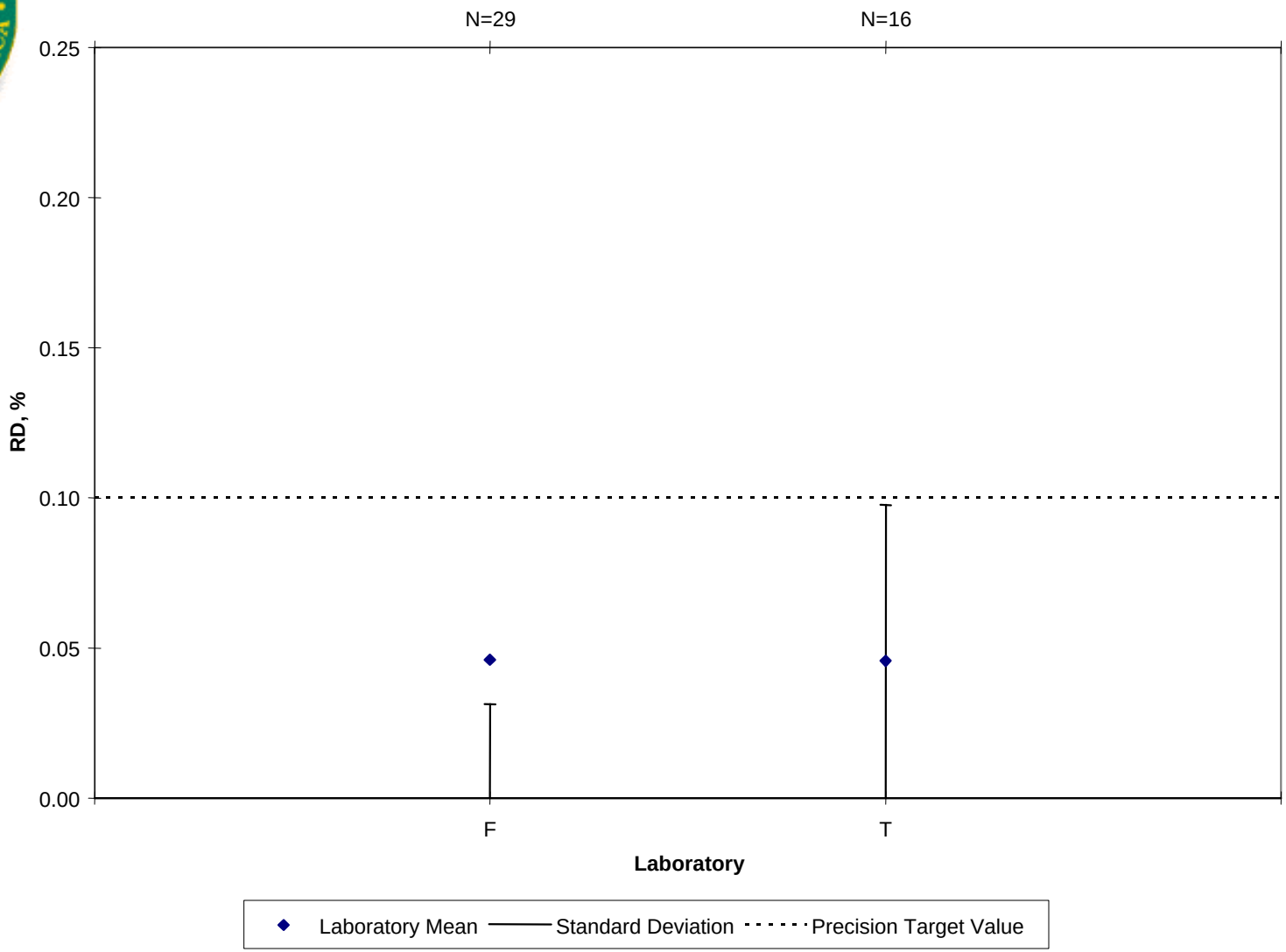
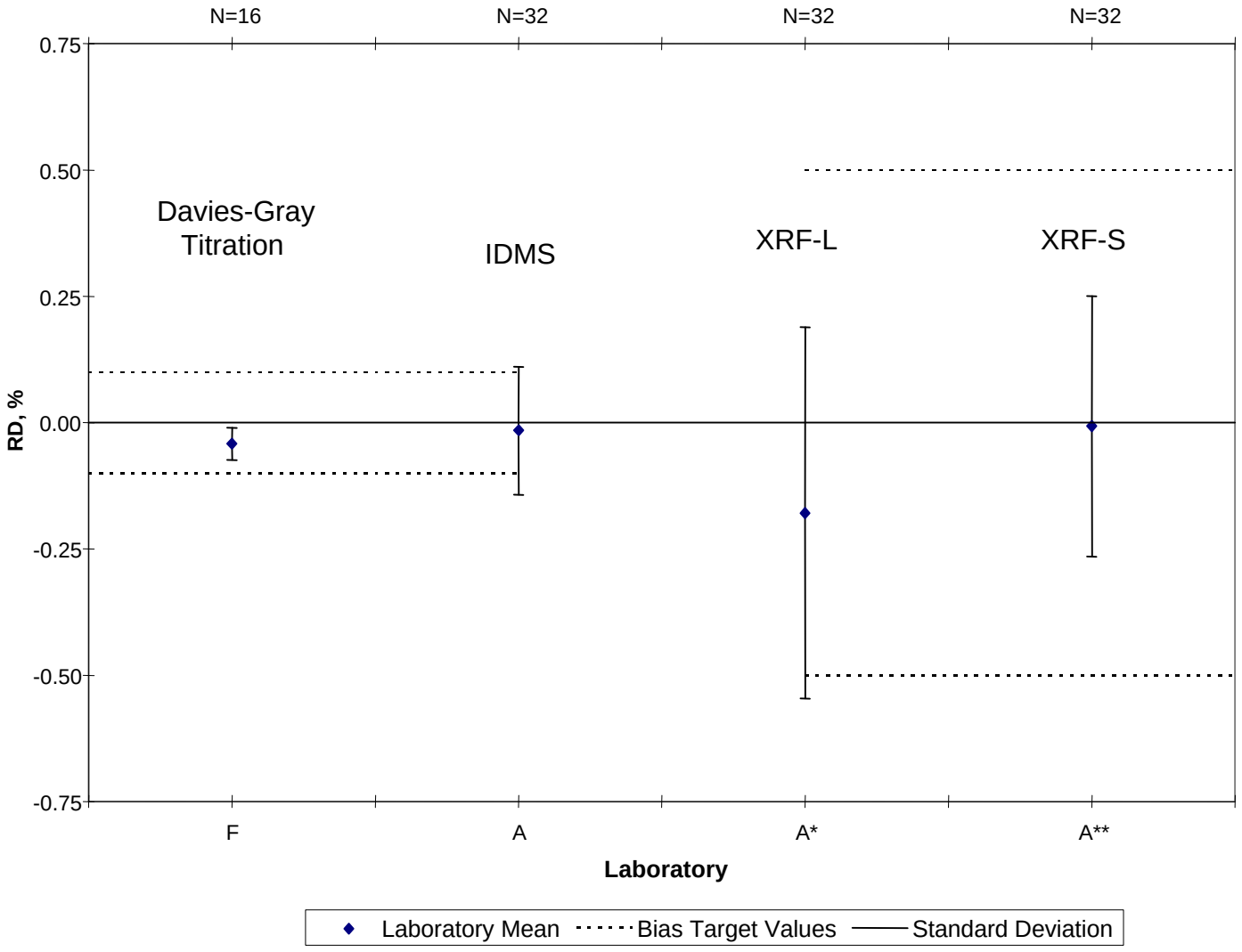


Table 6
Interlaboratory Performance Summary
UO_3 - Percent U

Method	Lab code	Mean	Standard deviation	N
Davies-Gray Titration	F	-0.042	0.032	16
IDMS	A	-0.016	0.126	32
X-Ray Fluorescence Liquid	A*	-0.179	0.368	32
X-Ray Fluorescence Solid	A**	-0.007	0.258	32

New Brunswick Laboratory Safeguards Measurement Evaluation Program

UO3 Powder - Percent U



New Brunswick Laboratory Safeguards Measurement Evaluation Program
UO3 Powder - Percent U

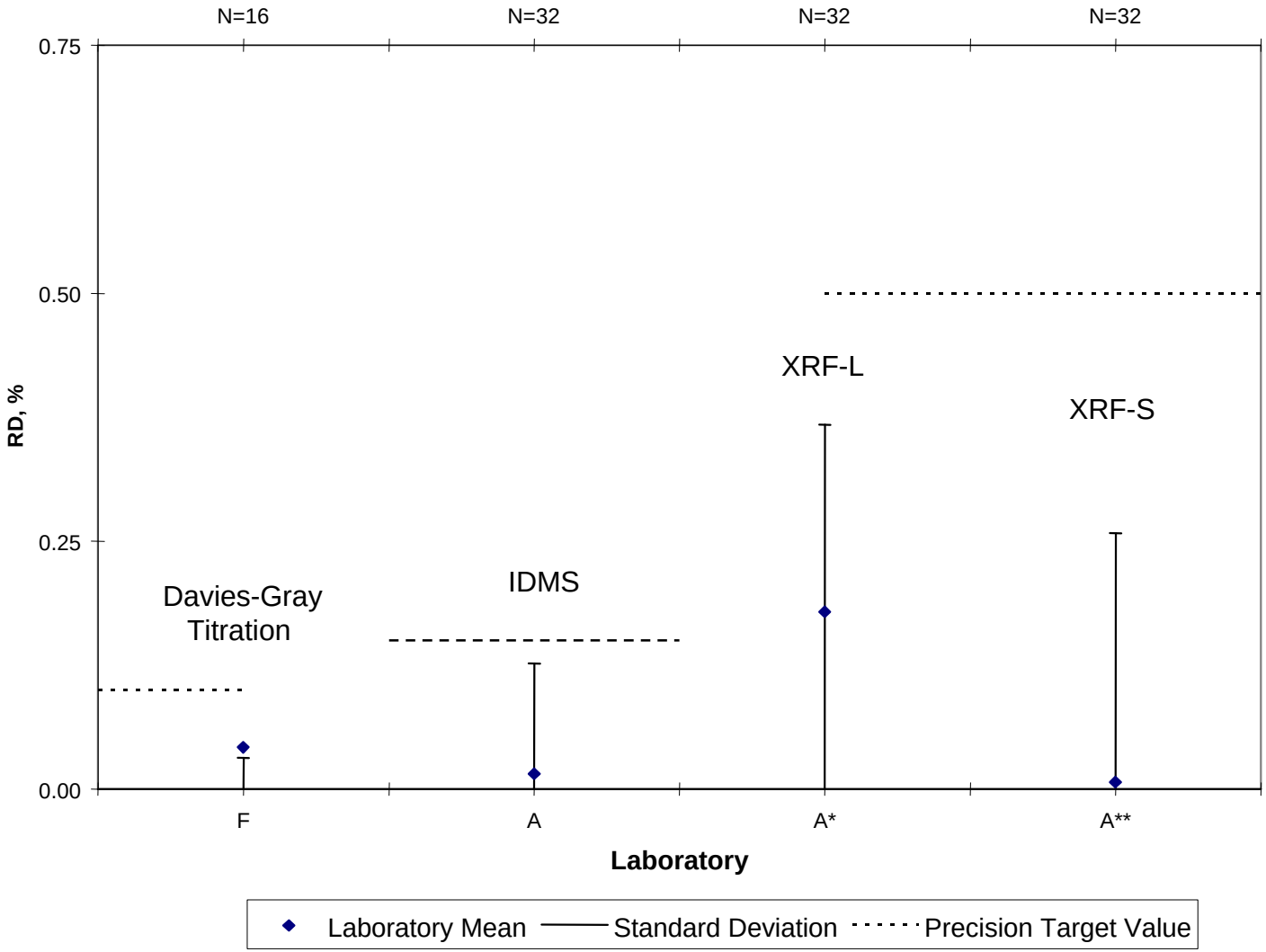
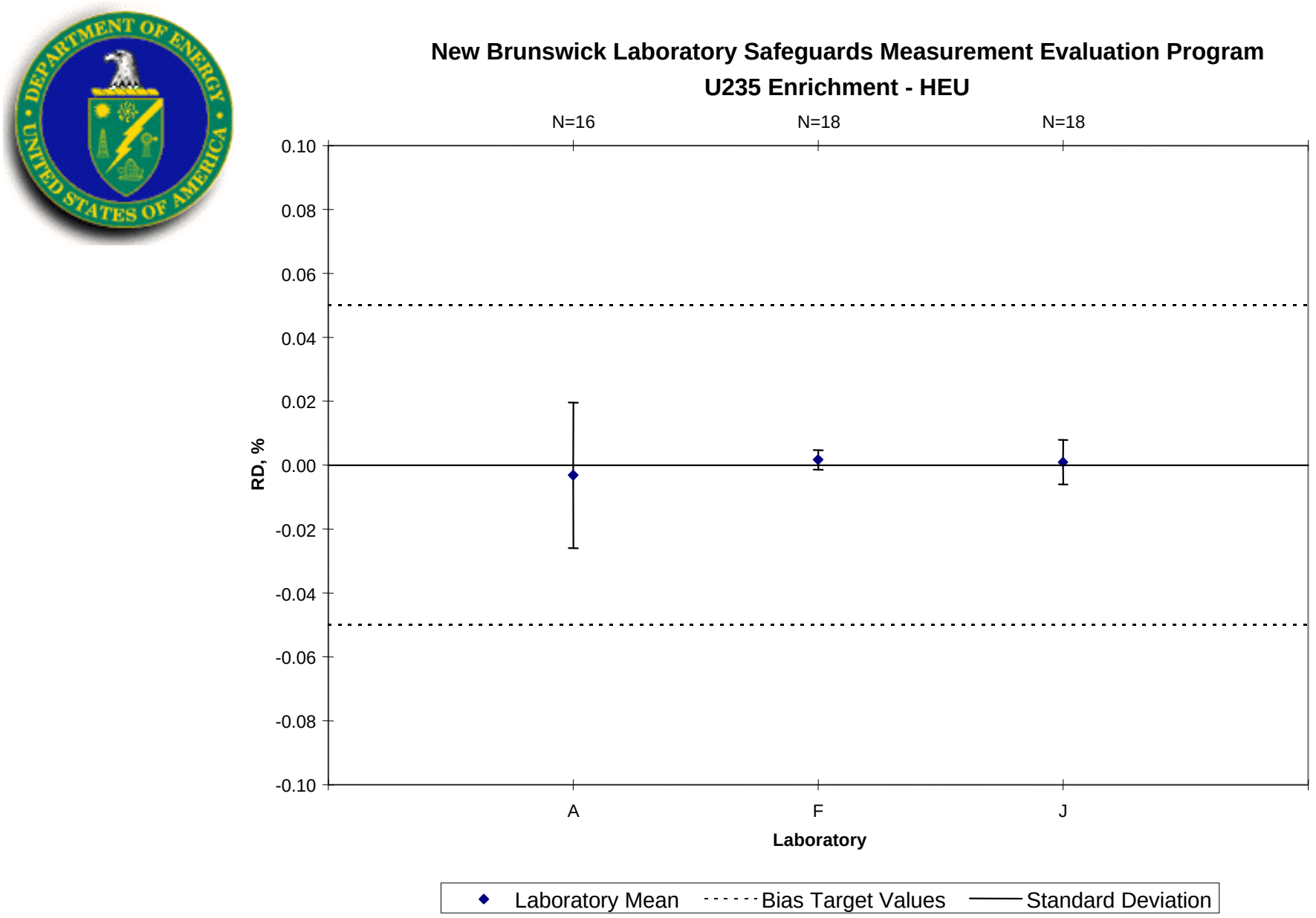


Table 7
Interlaboratory Performance Summary
^{235}U Enrichment - HEU

Method	Lab code	Mean	Standard deviation	N
TIMS	A	-0.003	0.023	16
TIMS	F	0.002	0.003	18
TIMS	J	0.001	0.007	18


New Brunswick Laboratory Safeguards Measurement Evaluation Program
U235 Enrichment - HEU
N=16
N=18
N=18
0.10
0.08
0.06
0.04
0.02
0.00
-0.02
-0.04
-0.06
-0.08
-0.10
RD, %
A
F
J
Laboratory
Laboratory Mean
Bias Target Values
Standard Deviation

New Brunswick Laboratory Safeguards Measurement Evaluation Program
U235 Enrichment - HEU

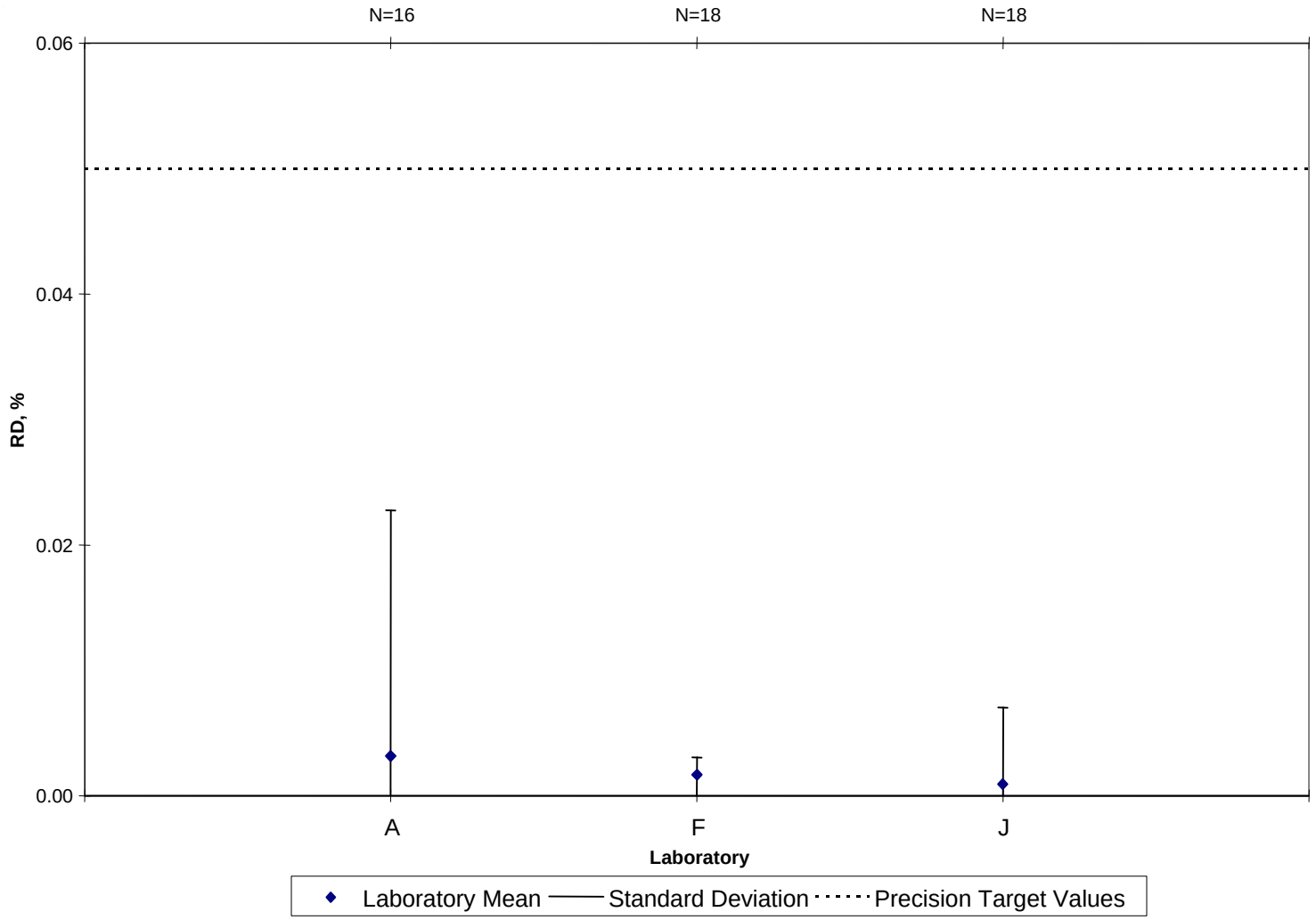
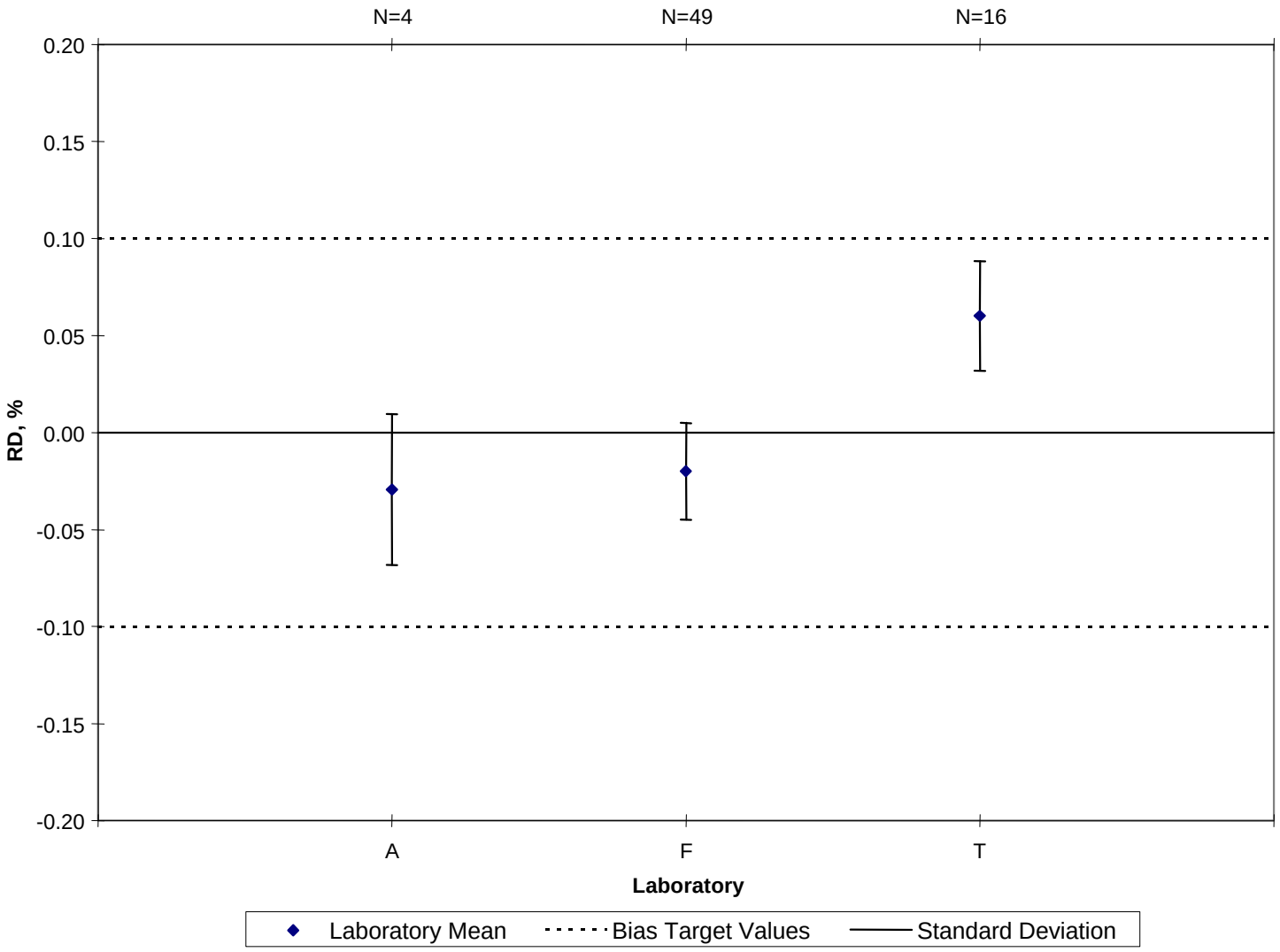


Table 8
Interlaboratory Performance Summary
^{235}U Enrichment - LEU

Method	Lab code	Mean	Standard deviation	N
TIMS	A	-0.029	0.039	4
TIMS	F	-0.020	0.025	49
TIMS	T	0.060	0.028	16

New Brunswick Laboratory Safeguards Measurement Evaluation Program

U235 Enrichment - LEU



New Brunswick Laboratory Safeguards Measurement Evaluation Program

U235 Enrichment - LEU

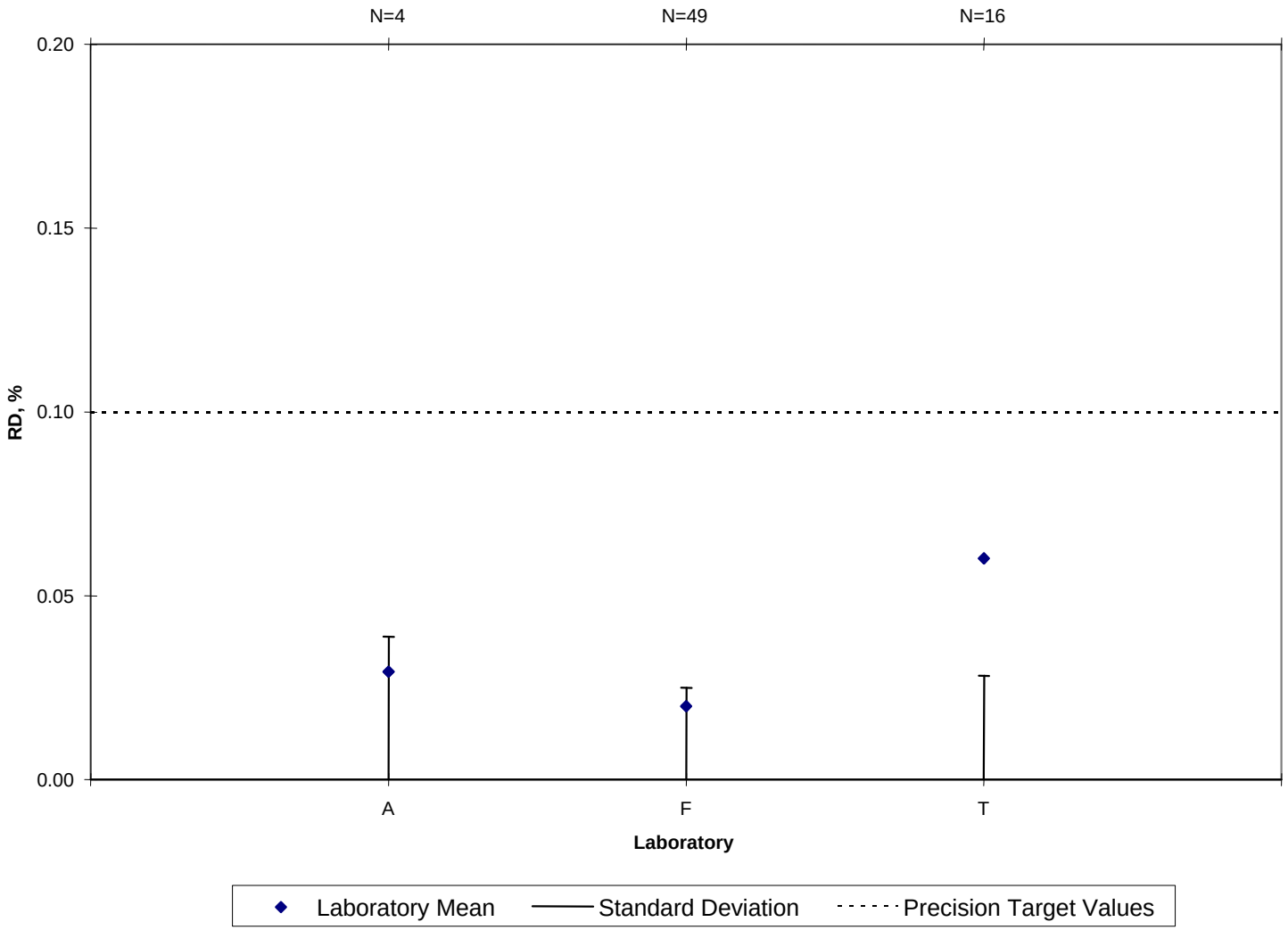
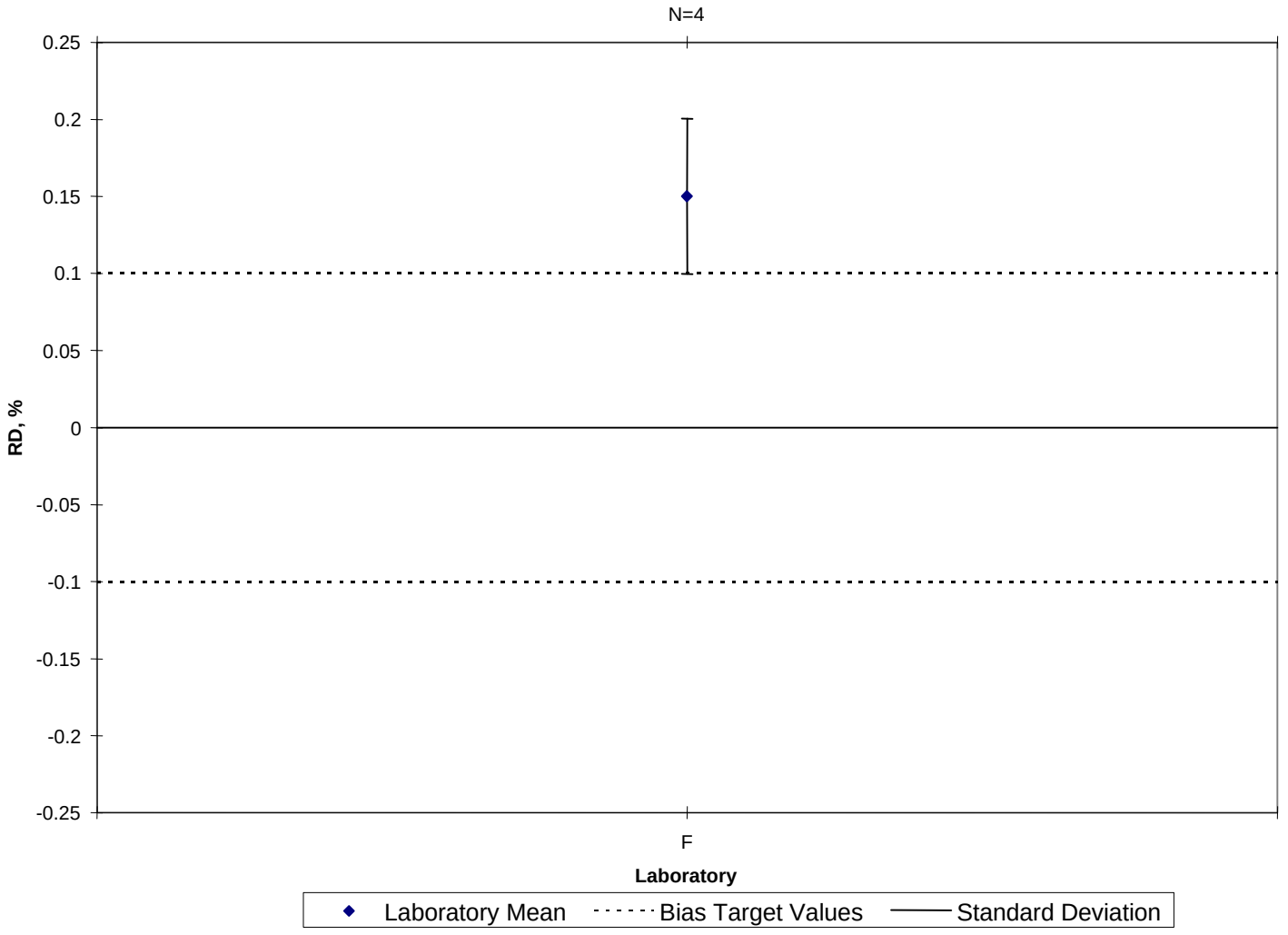


Table 9
Interlaboratory Performance Summary
Pu sulfate –Pu Mass

Method	Lab code	Mean	Standard deviation	N
IDMS	F	0.150	0.050	4

New Brunswick Laboratory Safeguards Measurement Evaluation Program
Pu Sulfate - Percent Pu



New Brunswick Laboratory Safeguards Measurement Evaluation Program
Pu Sulfate - Percent Pu

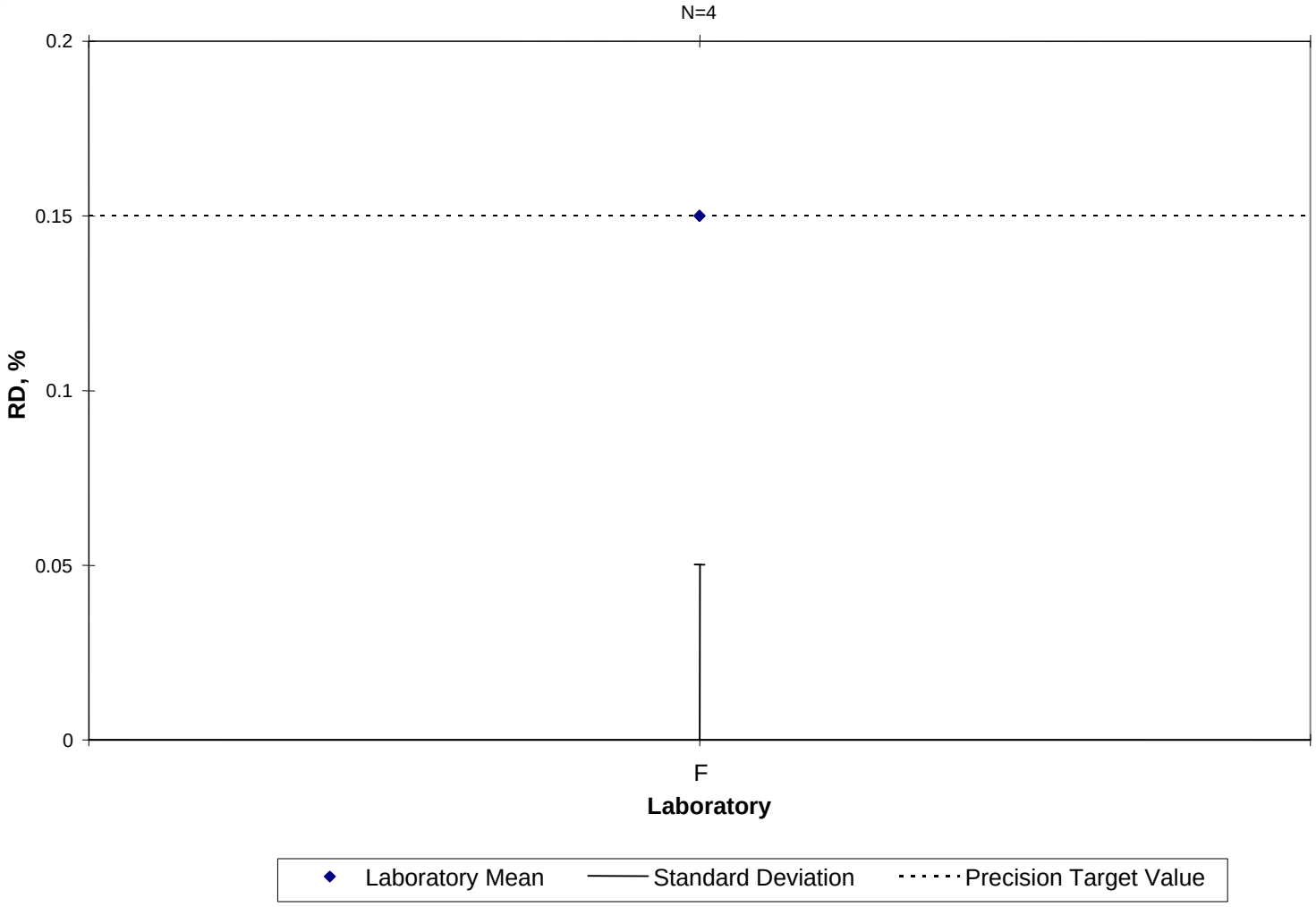
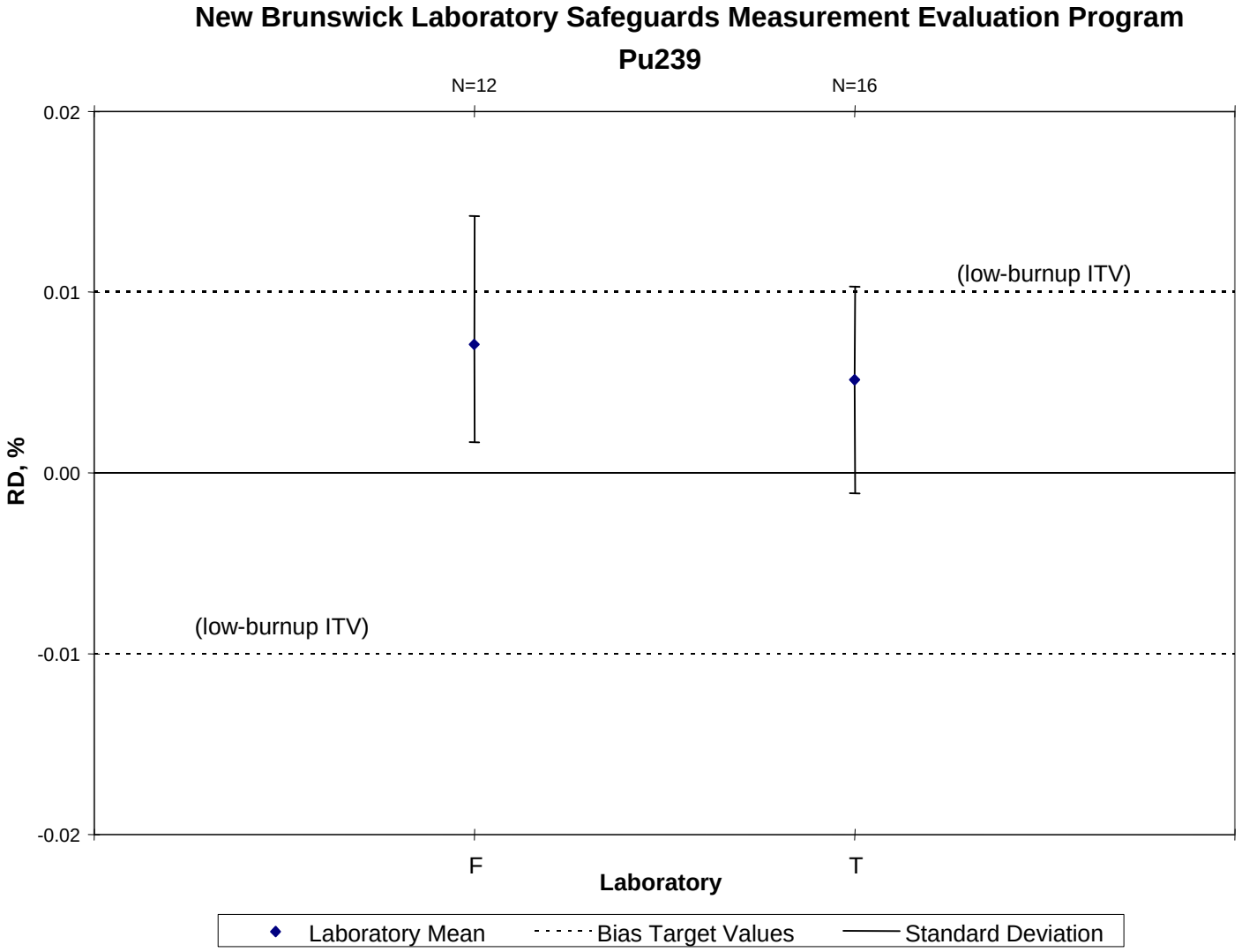


Table 10
Interlaboratory Performance Summary
^{239}Pu Abundance

Method	Lab code	Mean	Standard deviation	N
TIMS	F	0.007	0.005	12
TIMS	T	0.005	0.006	16


New Brunswick Laboratory Safeguards Measurement Evaluation Program
Pu239
N=12
N=16
0.02
0.01
0.00
-0.01
-0.02
RD, %
(low-burnup ITV)
(low-burnup ITV)
F
T
Laboratory
Laboratory Mean
Bias Target Values
Standard Deviation

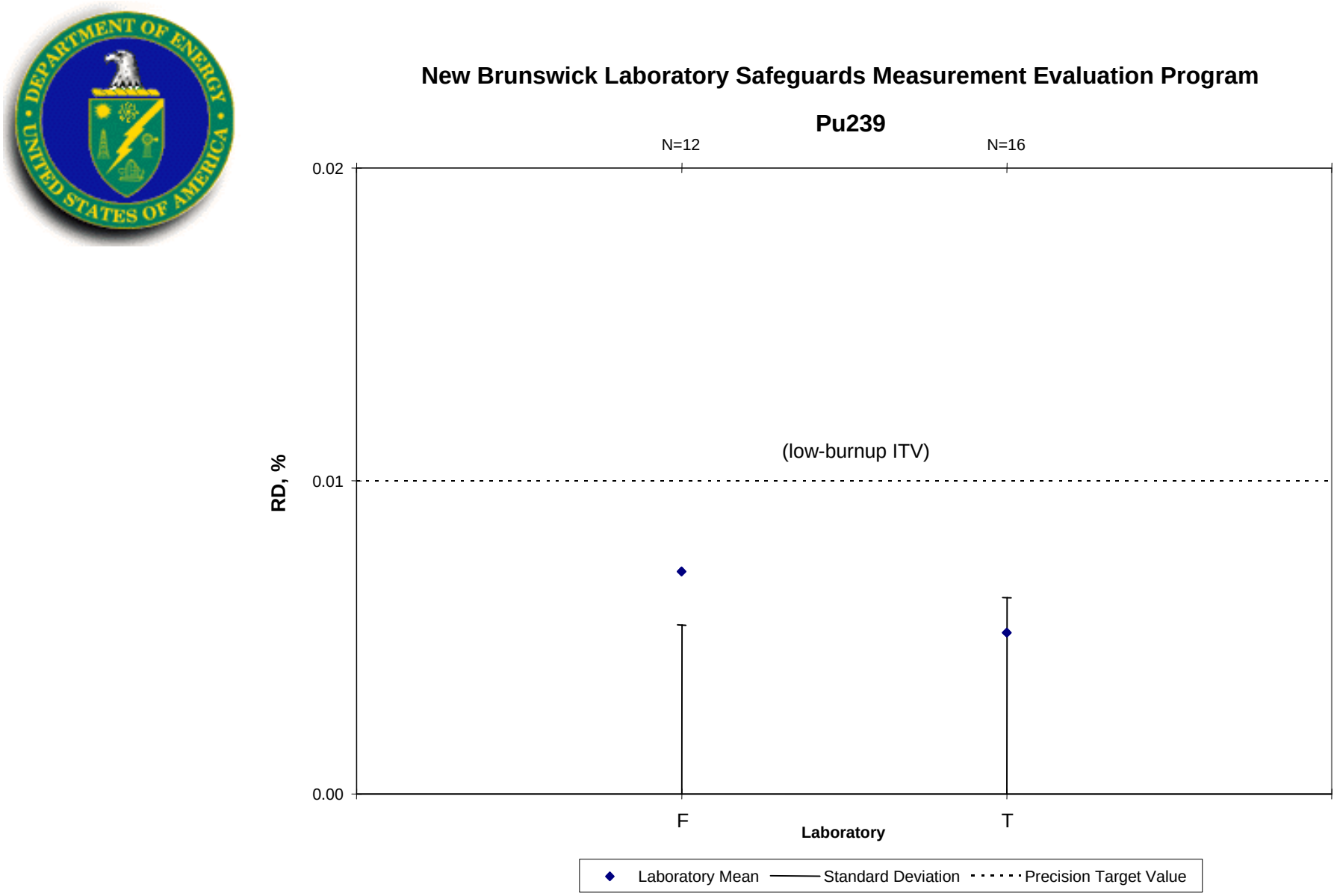
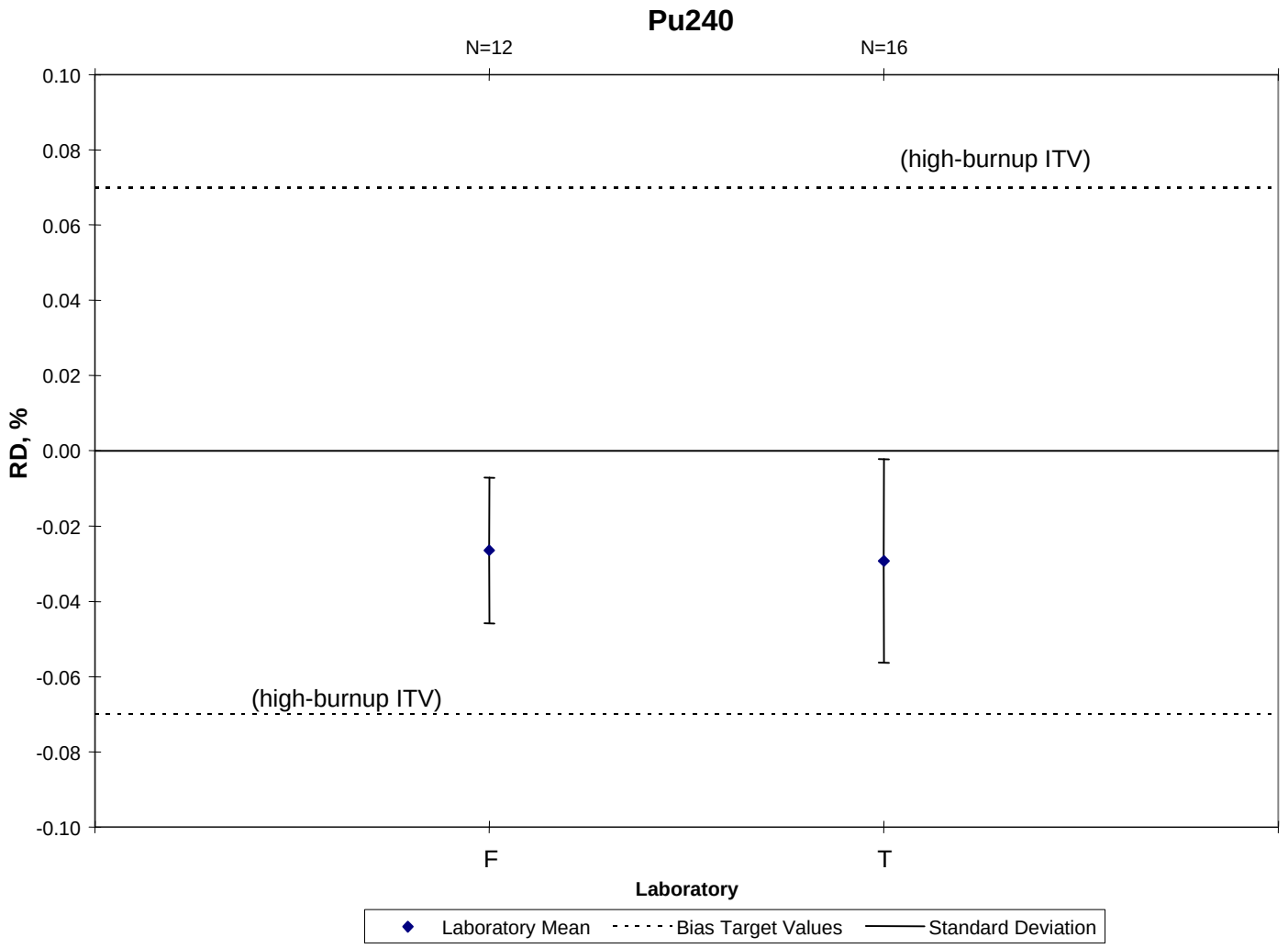

New Brunswick Laboratory Safeguards Measurement Evaluation Program
Pu239
N=12
N=16
0.02
0.01
0.00
RD, %
(low-burnup ITV)
F
T
Laboratory
Laboratory Mean
Standard Deviation
Precision Target Value

Table 11
Interlaboratory Performance Summary
^{240}Pu Abundance

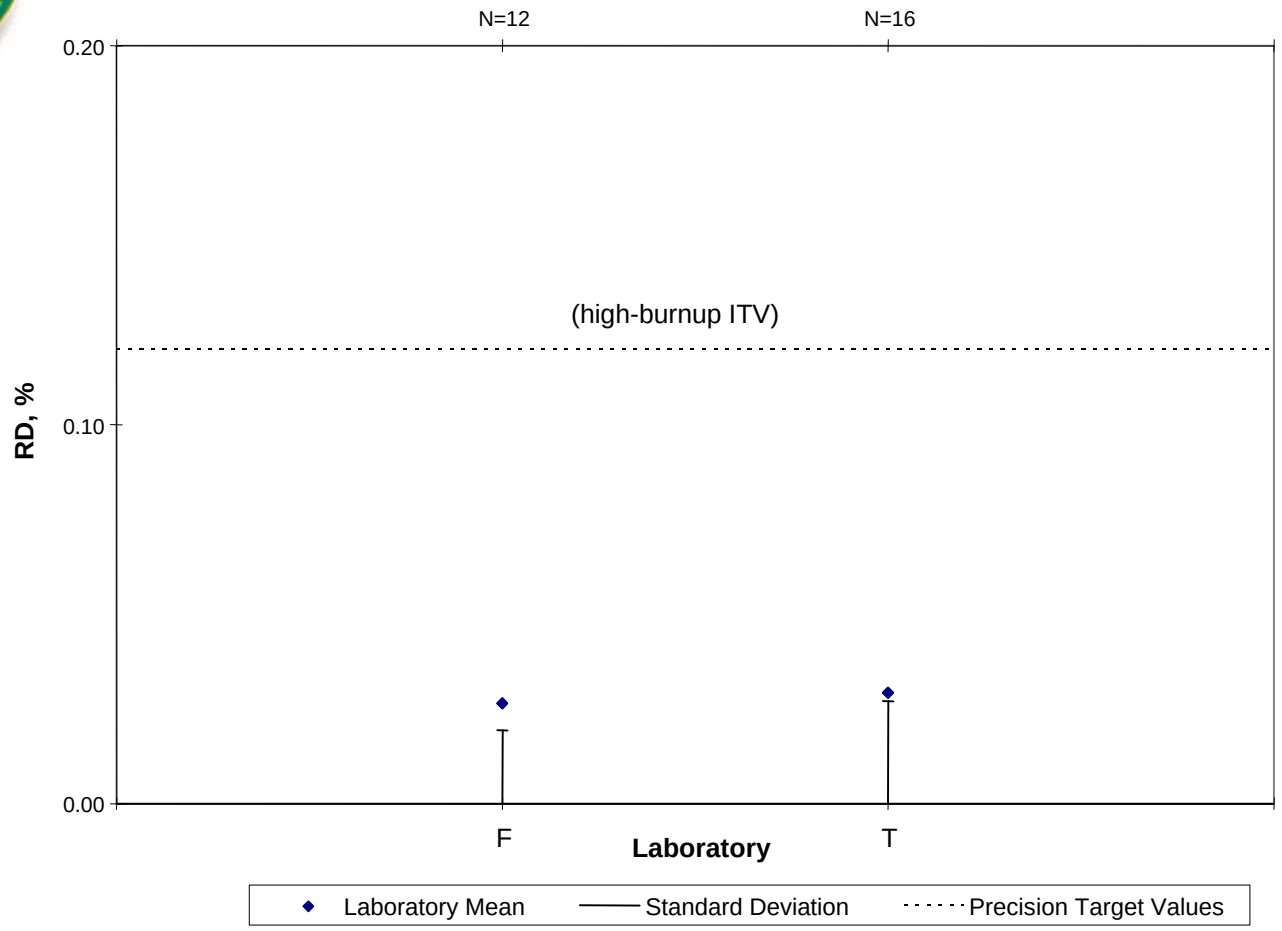
Method	Lab code	Mean	Standard deviation	N
TIMS	F	-0.026	0.019	12
TIMS	T	-0.029	0.027	16

New Brunswick Laboratory Safeguards Measurement Evaluation Program



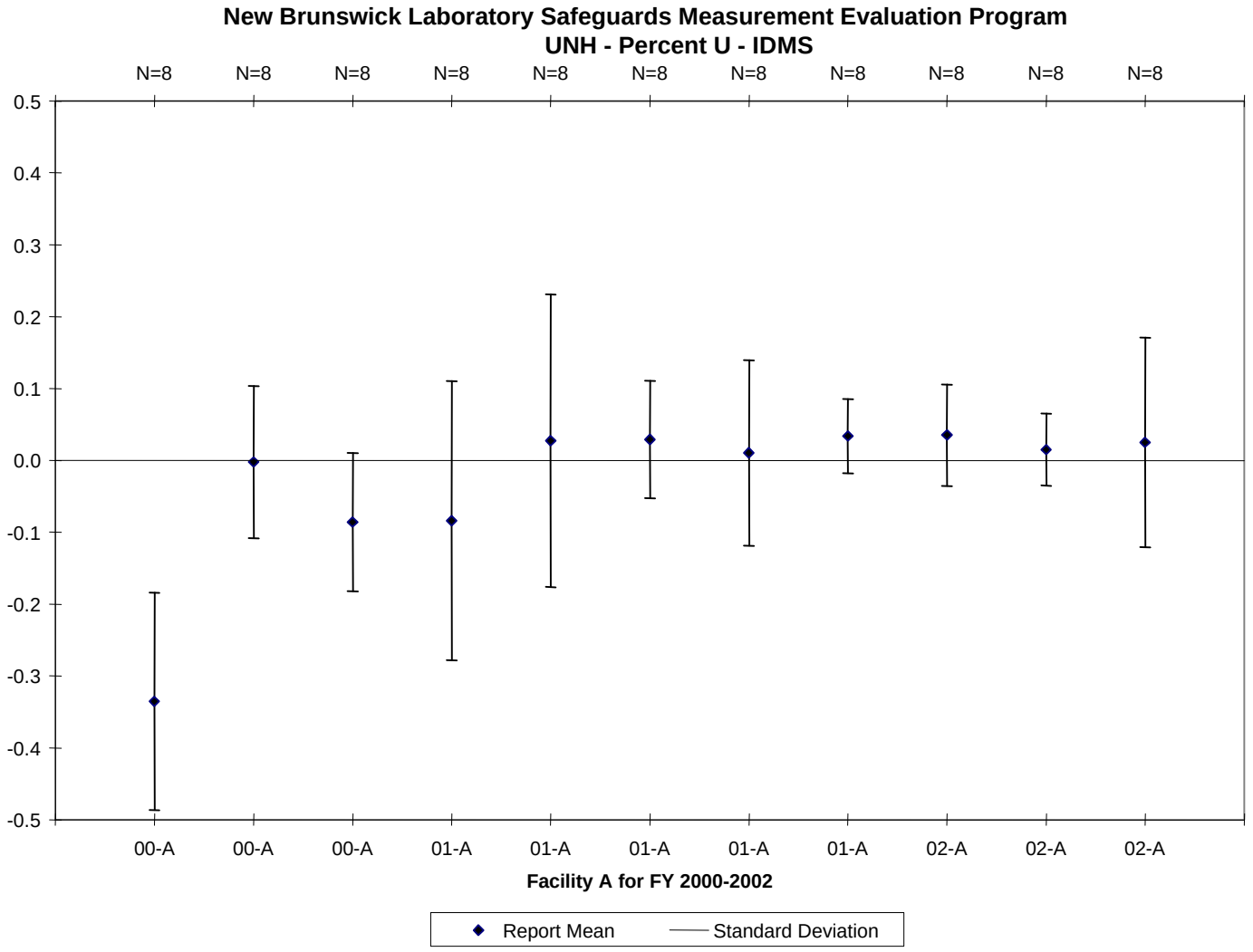
New Brunswick Laboratory Safeguards Measurement Evaluation Program

Pu240

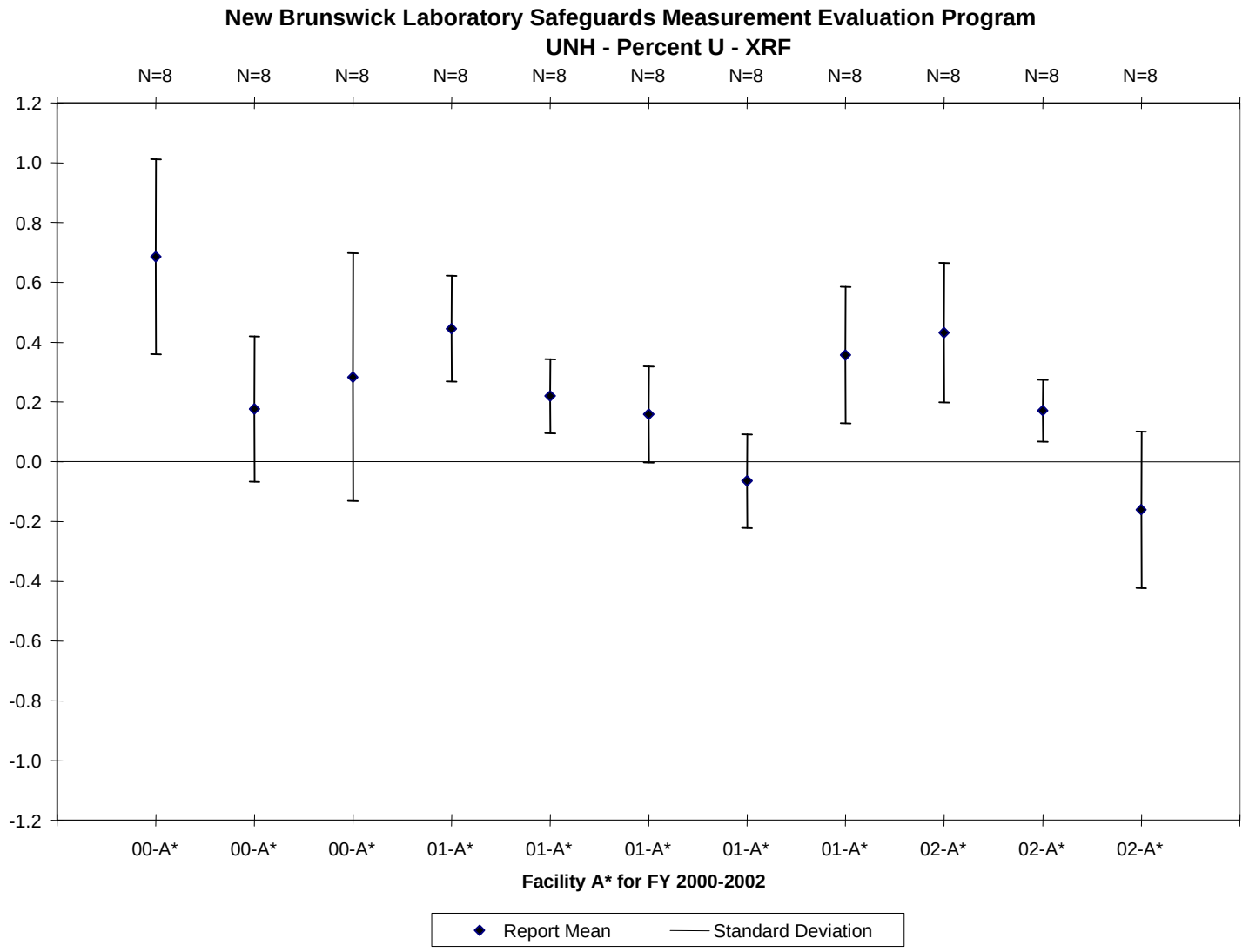


3-yr summaries

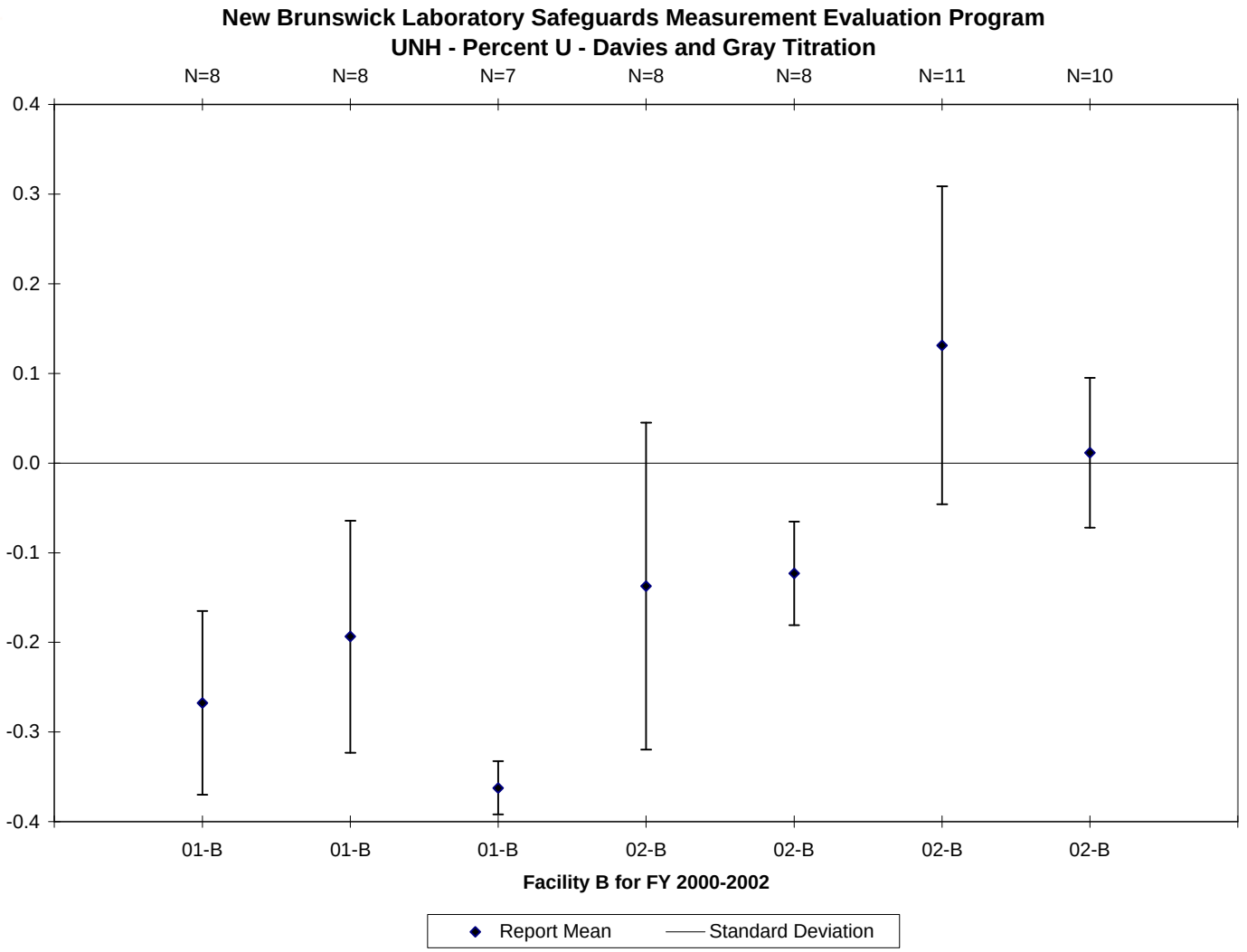
[u(s) = 0.1]


New Brunswick Laboratory Safeguards Measurement Evaluation Program
UNH - Percent U - IDMS
N=8
0.5
0.4
0.3
0.2
0.1
0.0
-0.1
-0.2
-0.3
-0.4
-0.5
00-A
01-A
02-A
Facility A for FY 2000-2002
Report Mean
Standard Deviation

[u(s) = 0.5]


New Brunswick Laboratory Safeguards Measurement Evaluation Program
UNH - Percent U - XRF
N=8
1.2
1.0
0.8
0.6
0.4
0.2
0.0
-0.2
-0.4
-0.6
-0.8
-1.0
-1.2
00-A*
01-A*
02-A*
Facility A* for FY 2000-2002
Report Mean
Standard Deviation

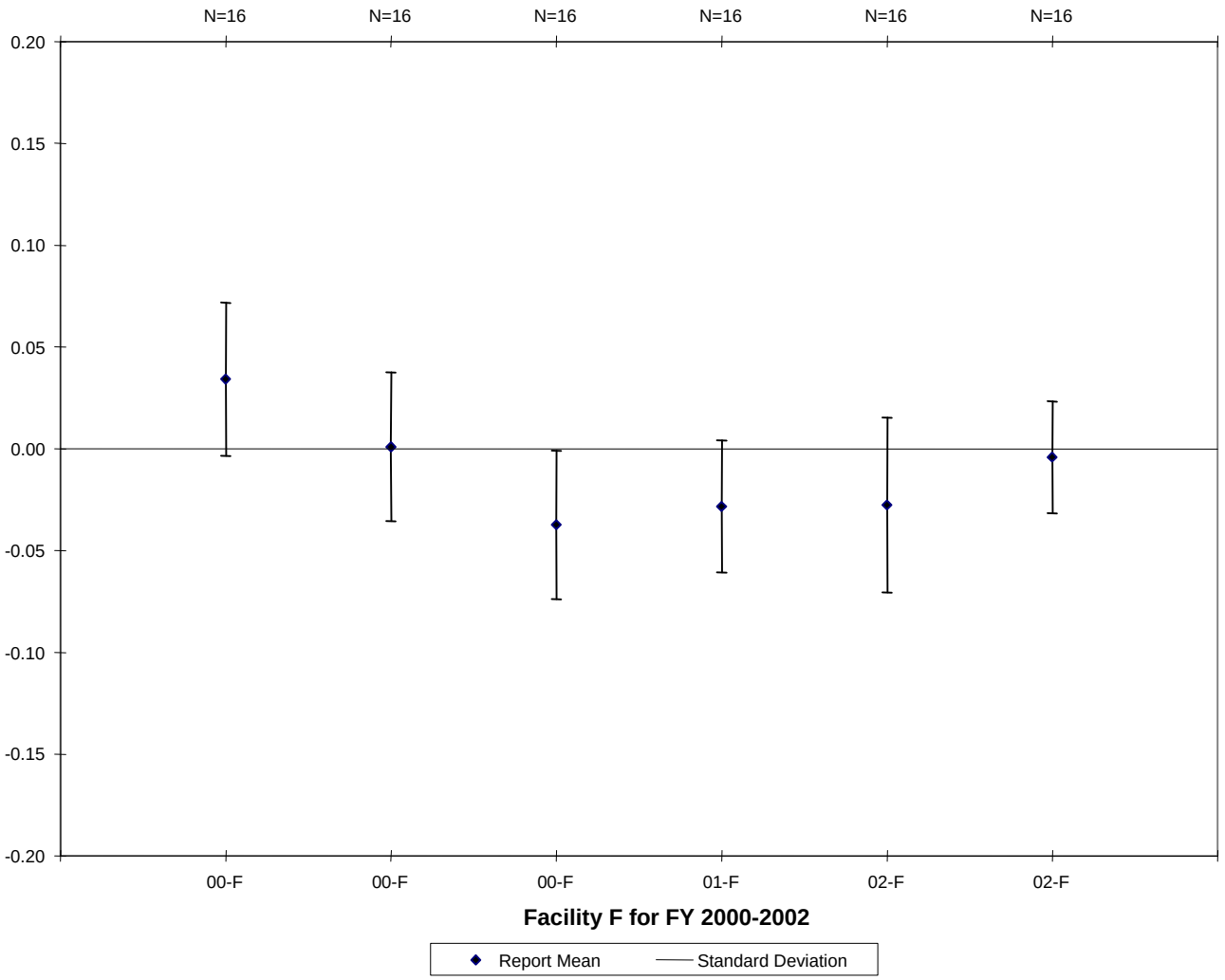
[u(s) = 0.1]


New Brunswick Laboratory Safeguards Measurement Evaluation Program
UNH - Percent U - Davies and Gray Titration
N=8
N=8
N=7
N=8
N=8
N=11
N=10
0.4
0.3
0.2
0.1
0.0
-0.1
-0.2
-0.3
-0.4
01-B
01-B
01-B
02-B
02-B
02-B
02-B
Facility B for FY 2000-2002
Report Mean
Standard Deviation

New Brunswick Laboratory Safeguards Measurement Evaluation Program

UNH - Percent U –Titration

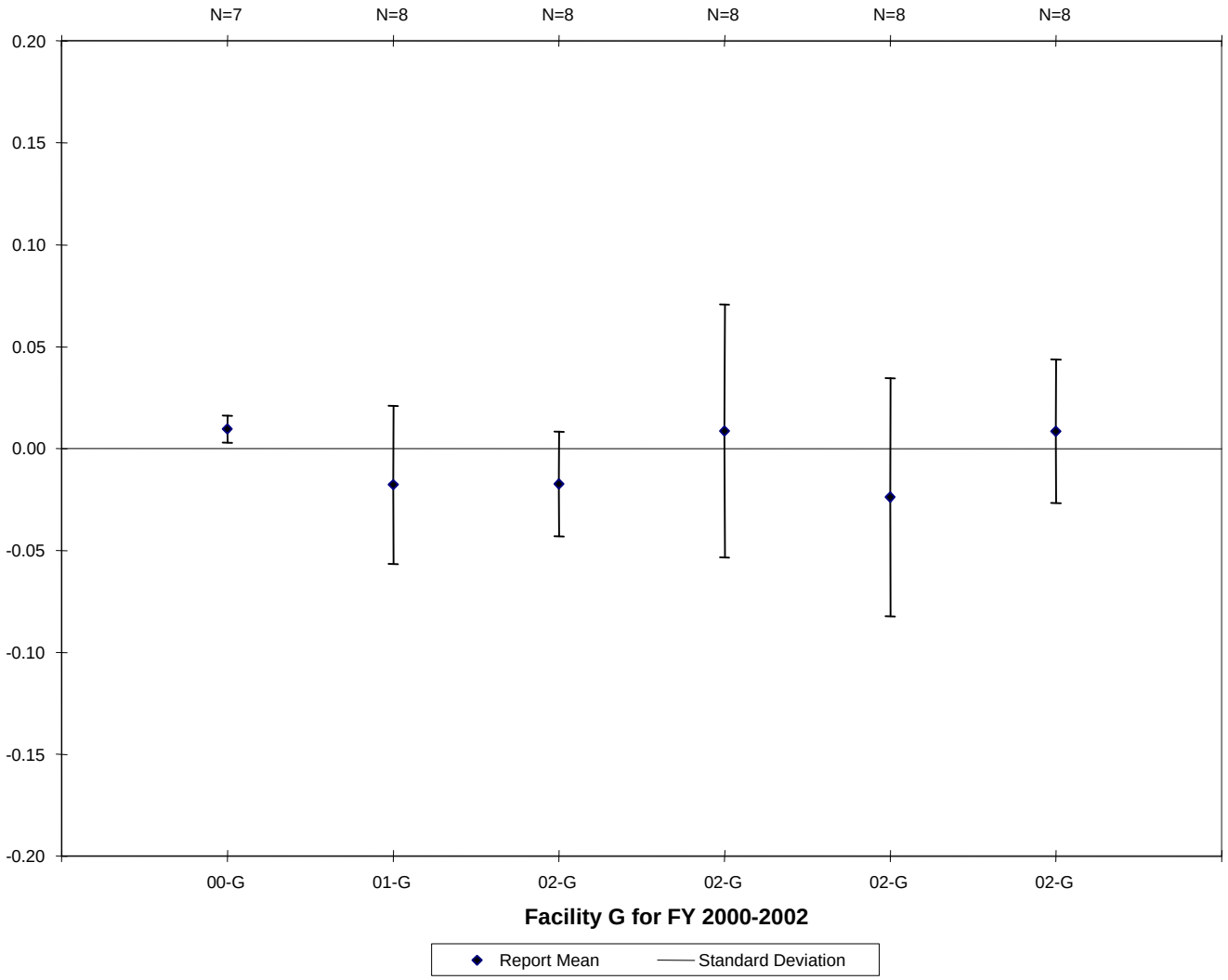
[u(s) = 0.1]



New Brunswick Laboratory Safeguards Measurement Evaluation Program

UNH - Percent U - Titration

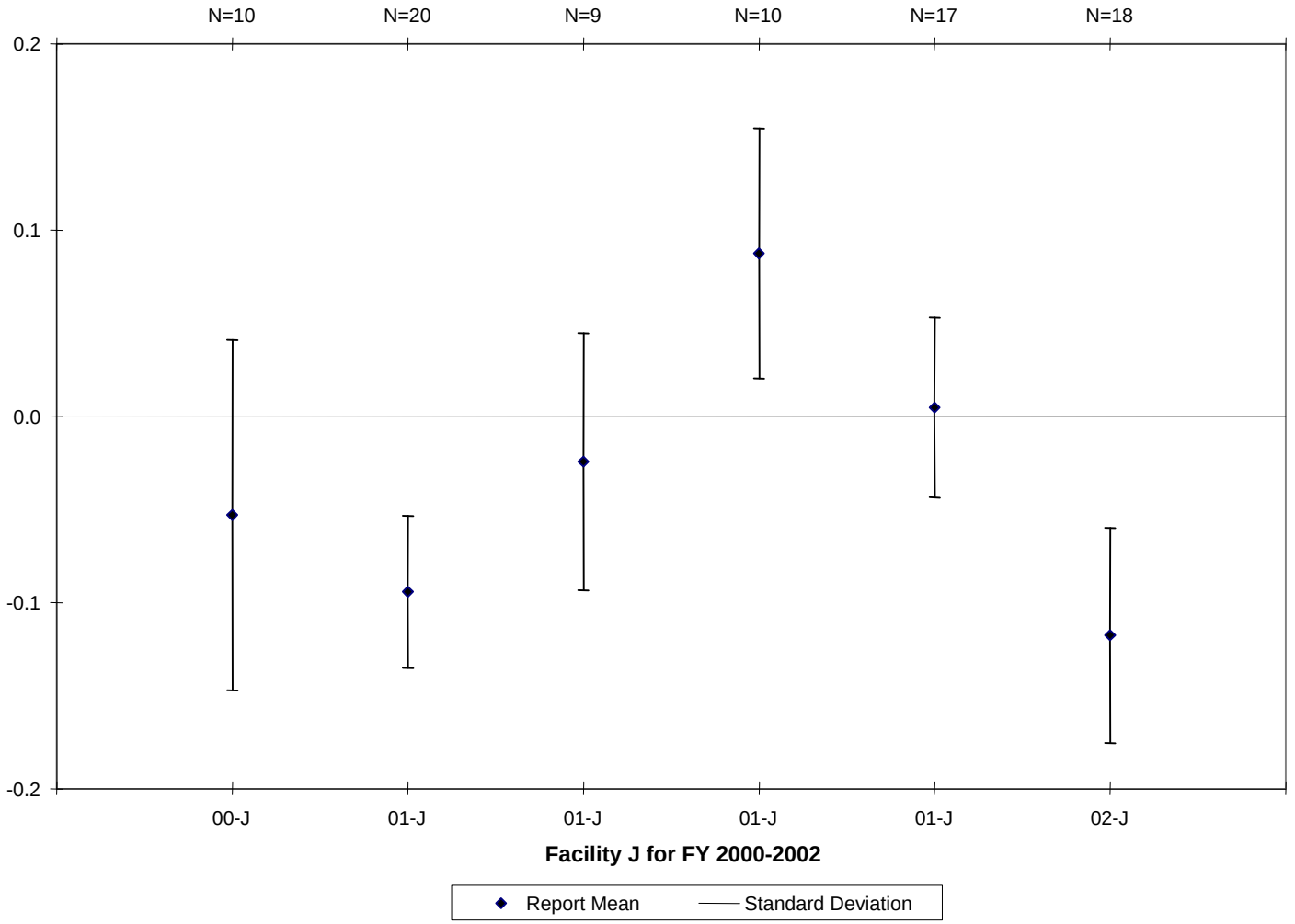
[u(s) = 0.1]

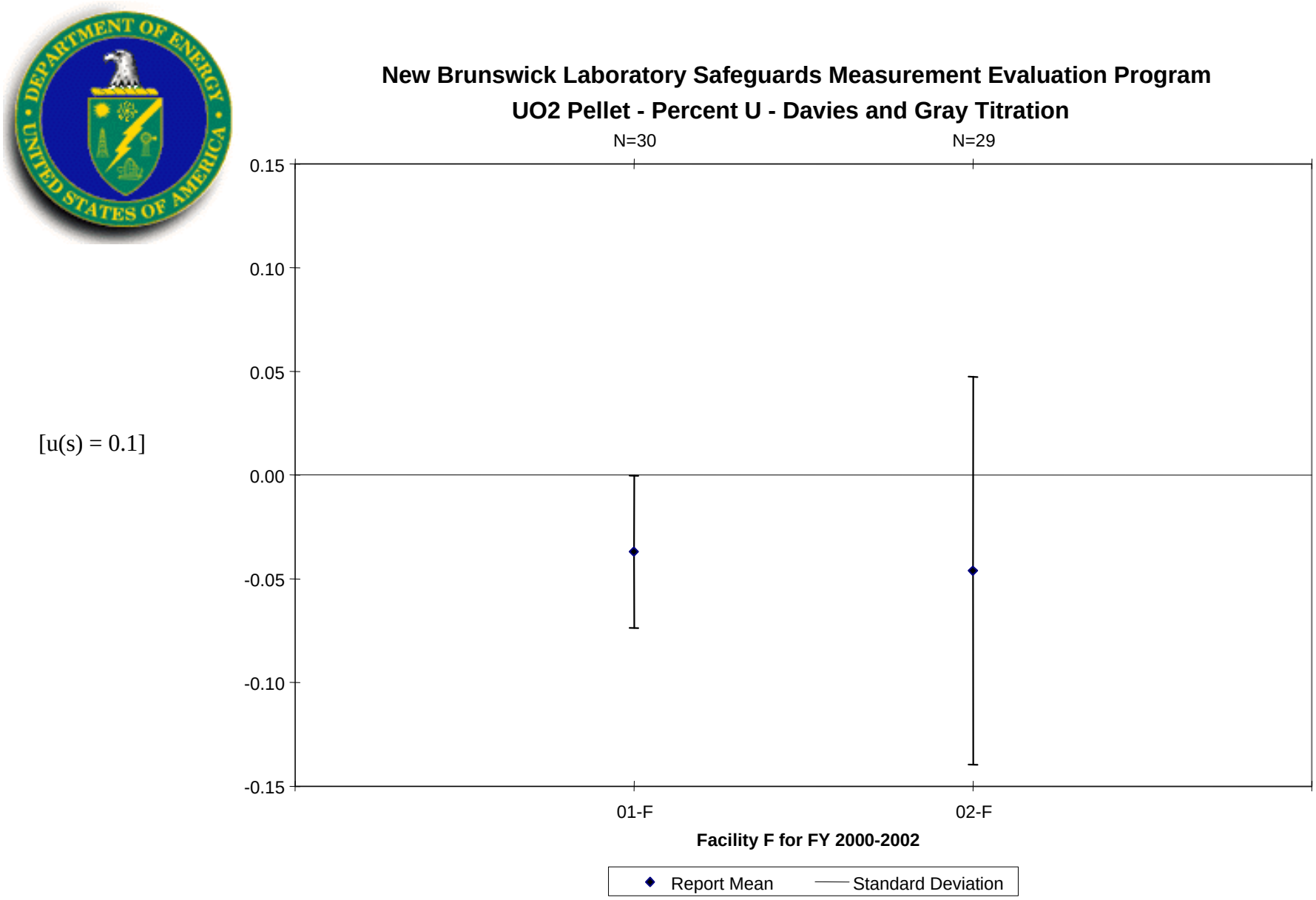


New Brunswick Laboratory Safeguards Measurement Evaluation Program

UNH - Percent U - IDMS

[u(s) = 0.1]



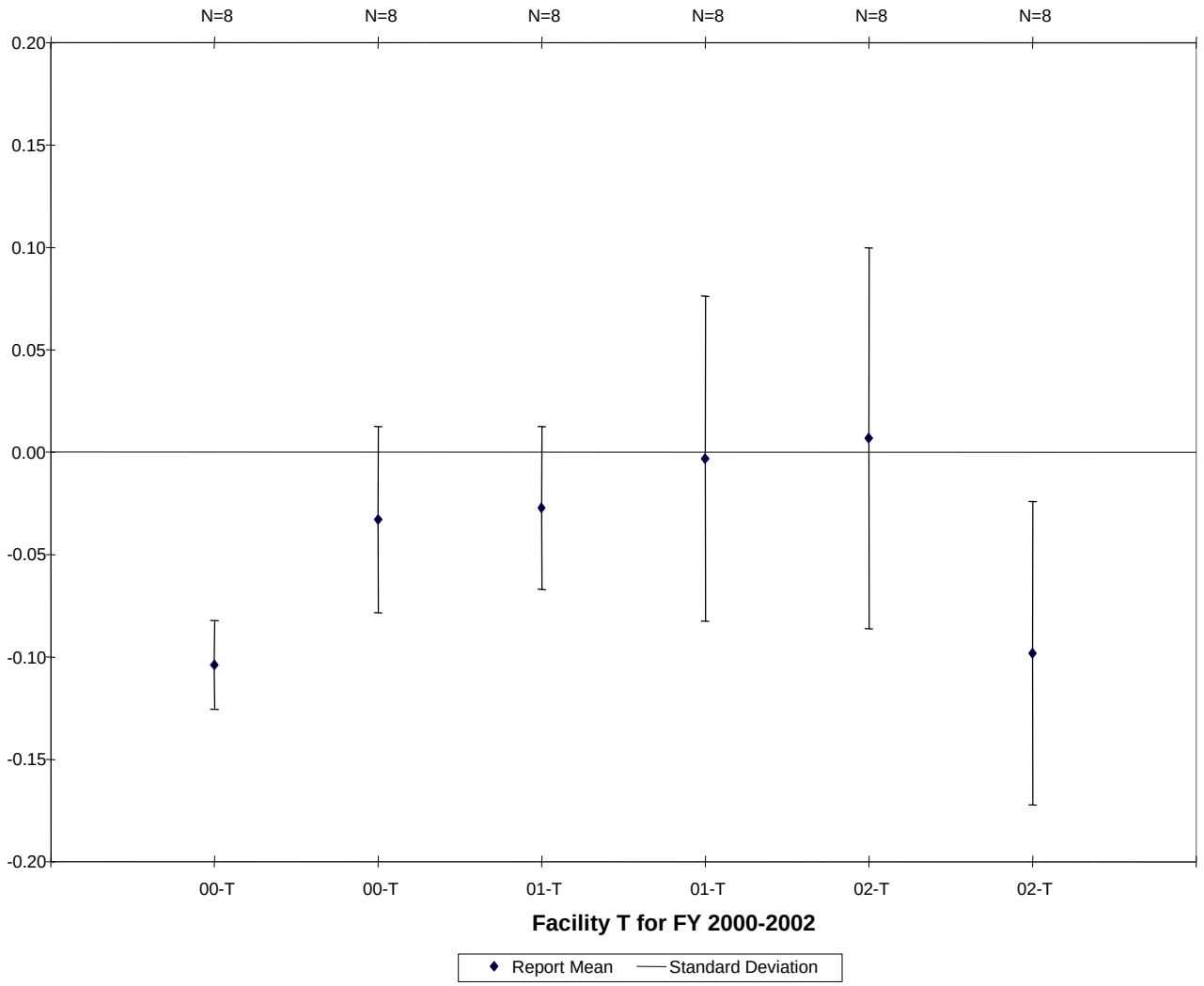

New Brunswick Laboratory Safeguards Measurement Evaluation Program
UO2 Pellet - Percent U - Davies and Gray Titration
N=30
N=29
0.15
0.10
0.05
0.00
-0.05
-0.10
-0.15
01-F
02-F
Facility F for FY 2000-2002
Report Mean
Standard Deviation

[u(s) = 0.1]

New Brunswick Laboratory Safeguards Measurement Evaluation Program

UO2 Pellet - Percent U by Titration

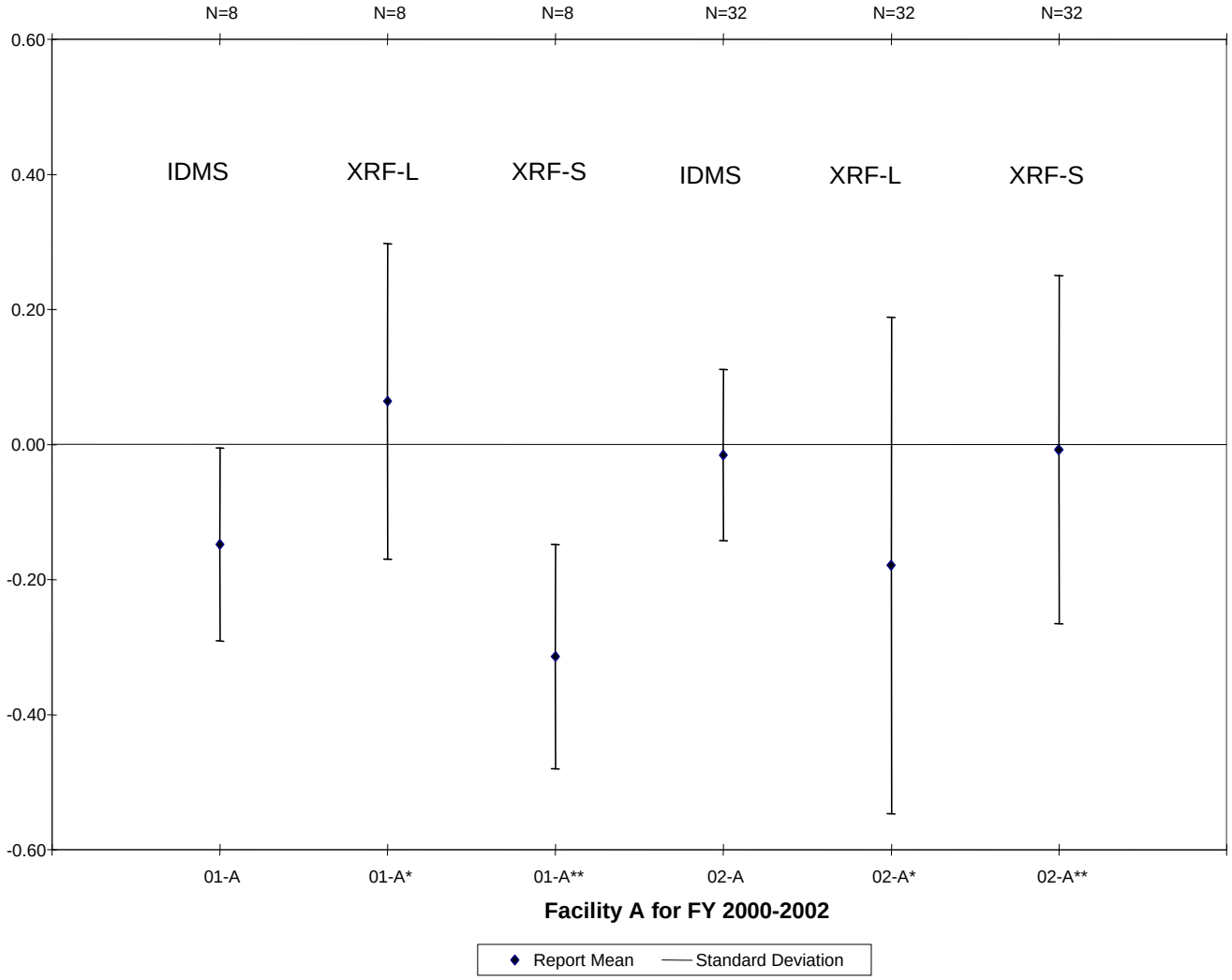
[u(s) = 0.1]

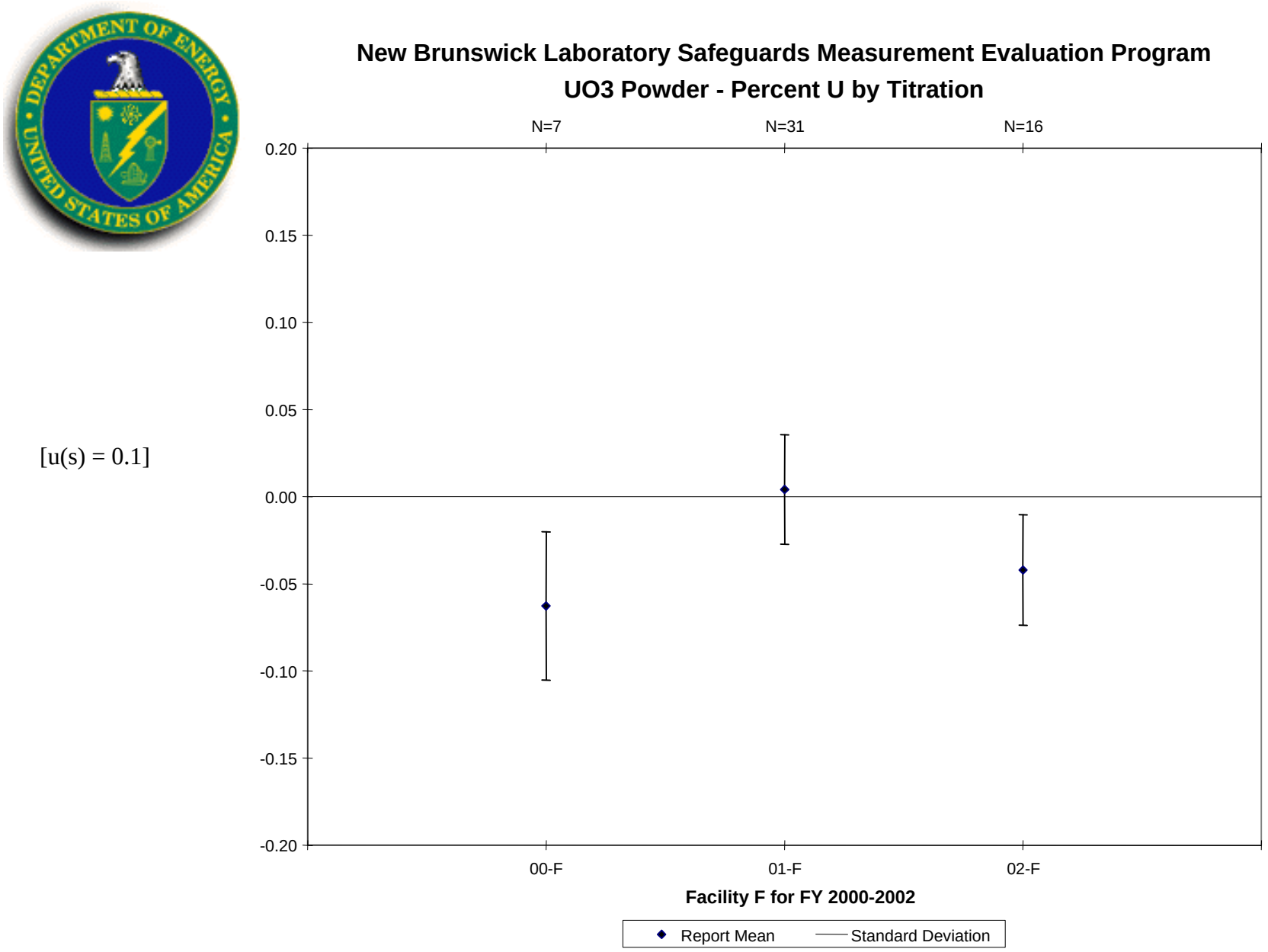


New Brunswick Laboratory Safeguards Measurement Evaluation Program

UO3 Powder - Percent U

[u(s) = 0.1 (IDMS)]
[u(s) = 0.5 (XRF)]



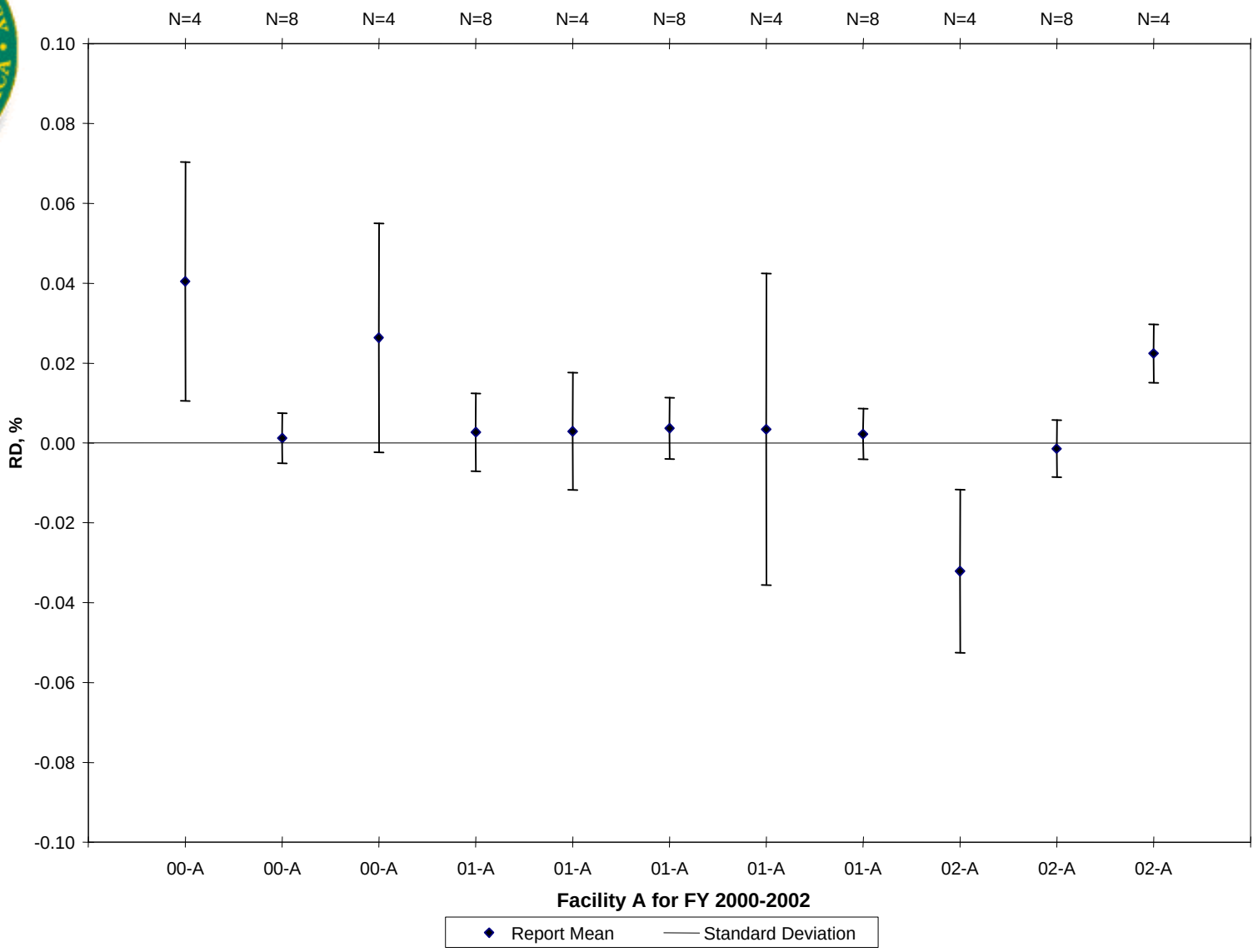

New Brunswick Laboratory Safeguards Measurement Evaluation Program
UO3 Powder - Percent U by Titration
N=7
N=31
N=16
0.20
0.15
0.10
0.05
0.00
-0.05
-0.10
-0.15
-0.20
00-F
01-F
02-F
Facility F for FY 2000-2002
Report Mean
Standard Deviation

[u(s) = 0.1]

New Brunswick Laboratory Safeguards Measurement Evaluation Program

U235 Enrichment - HEU

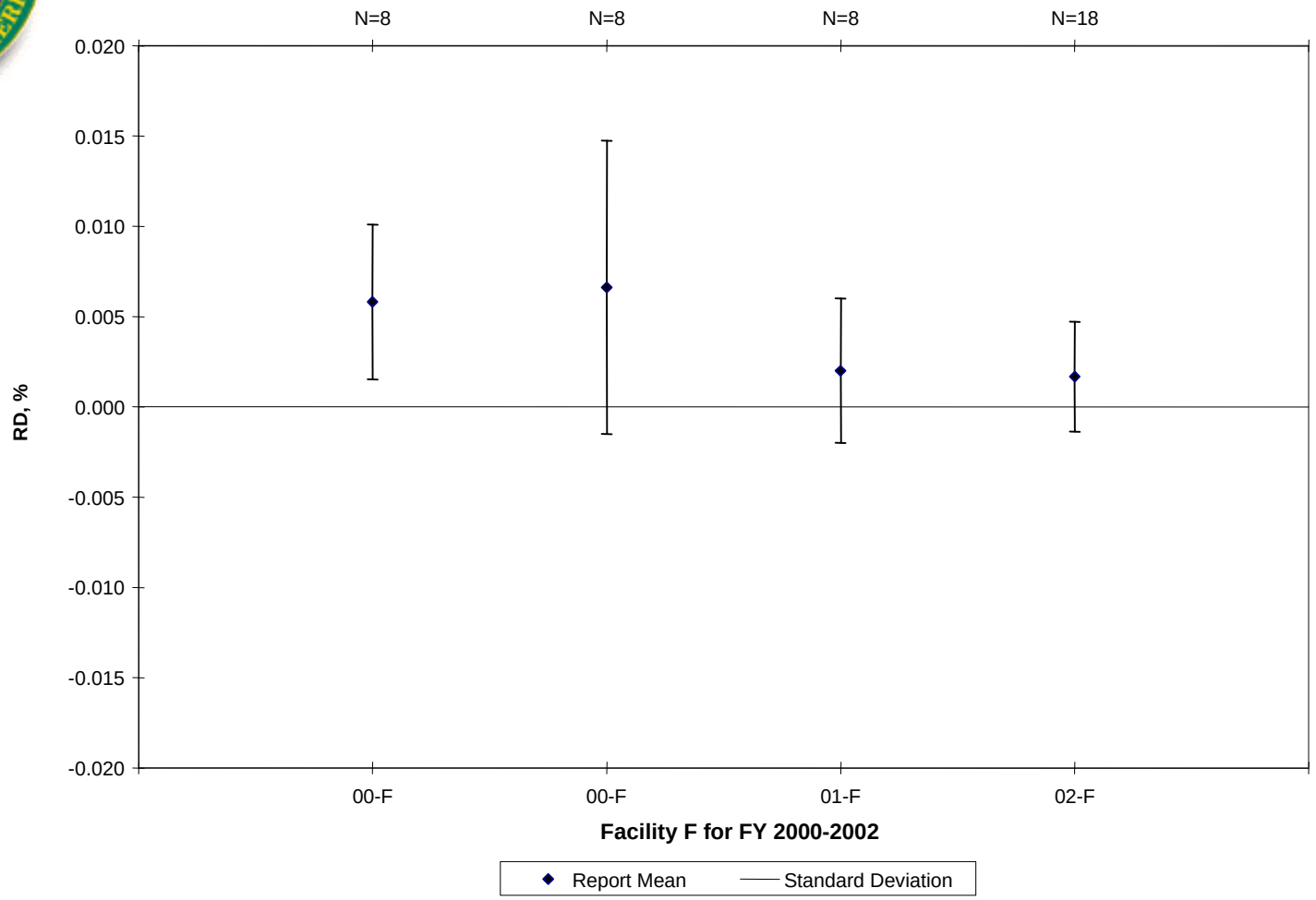
[u(s) = 0.05]



New Brunswick Laboratory Safeguards Measurement Evaluation Program

U235 Enrichment - HEU

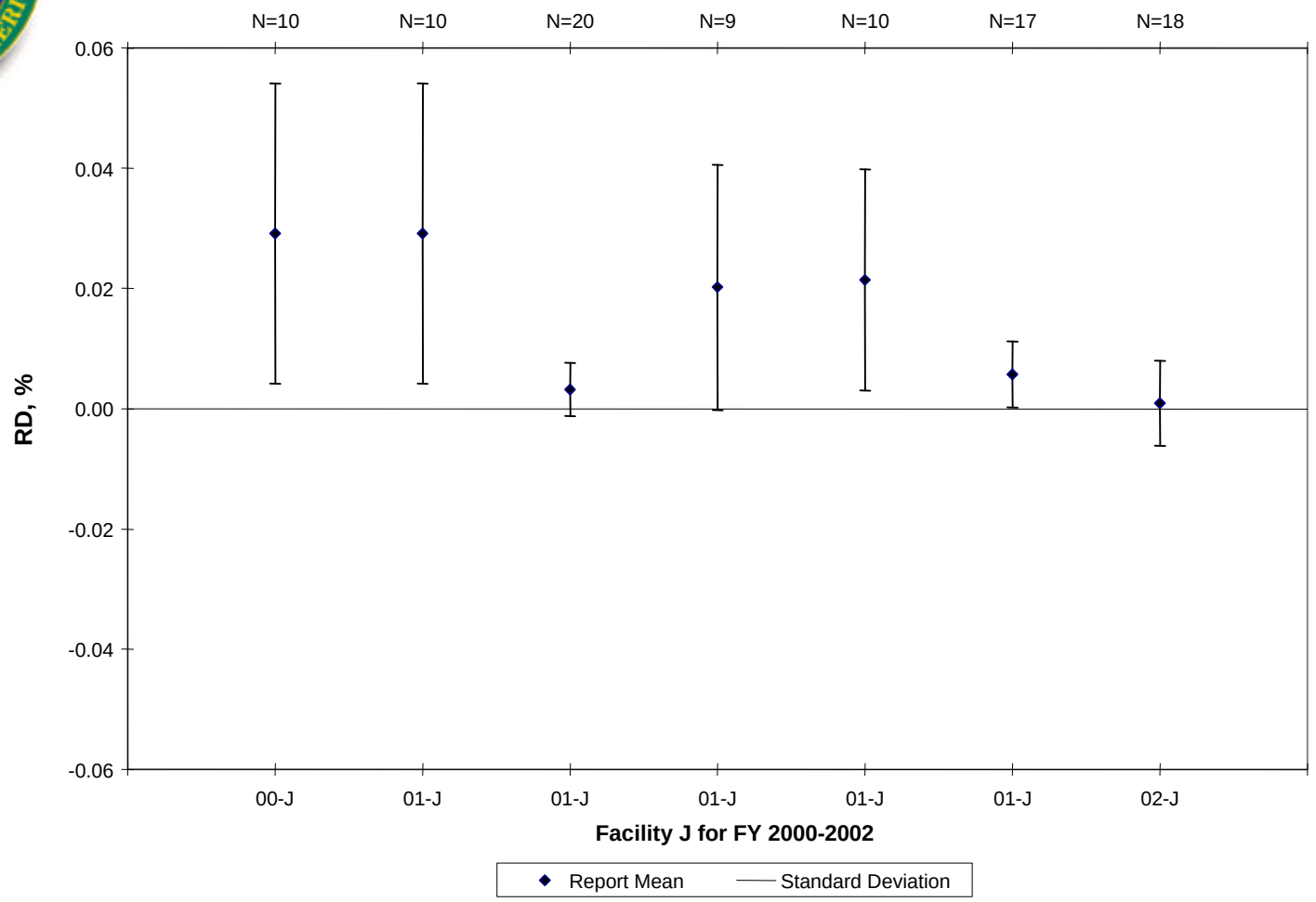
[u(s) = 0.05]



New Brunswick Laboratory Safeguards Measurement Evaluation Program

U235 Enrichment - HEU

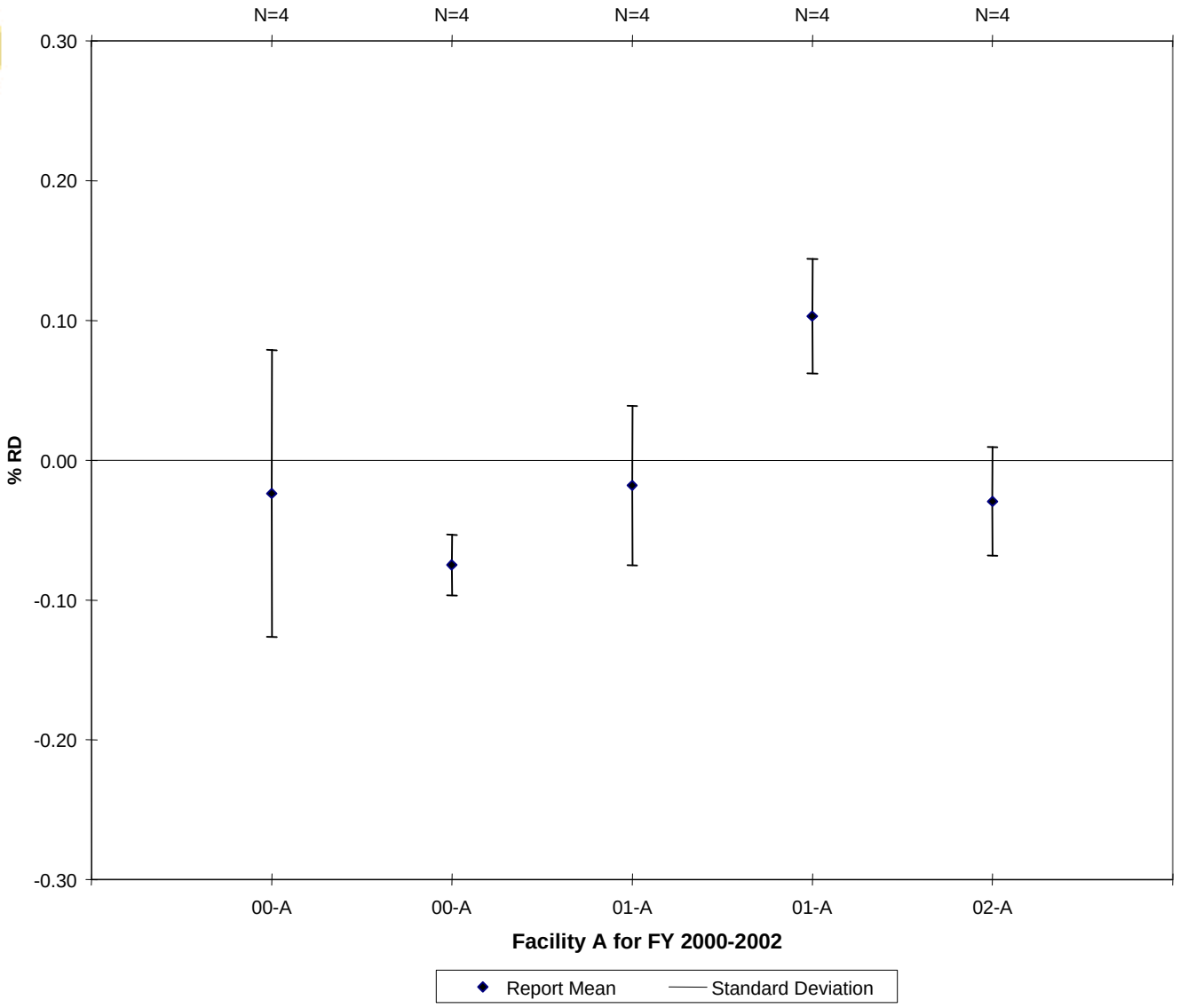
[u(s) = 0.05]



New Brunswick Laboratory Safeguards Measurement Evaluation Program

U235 Enrichment - LEU

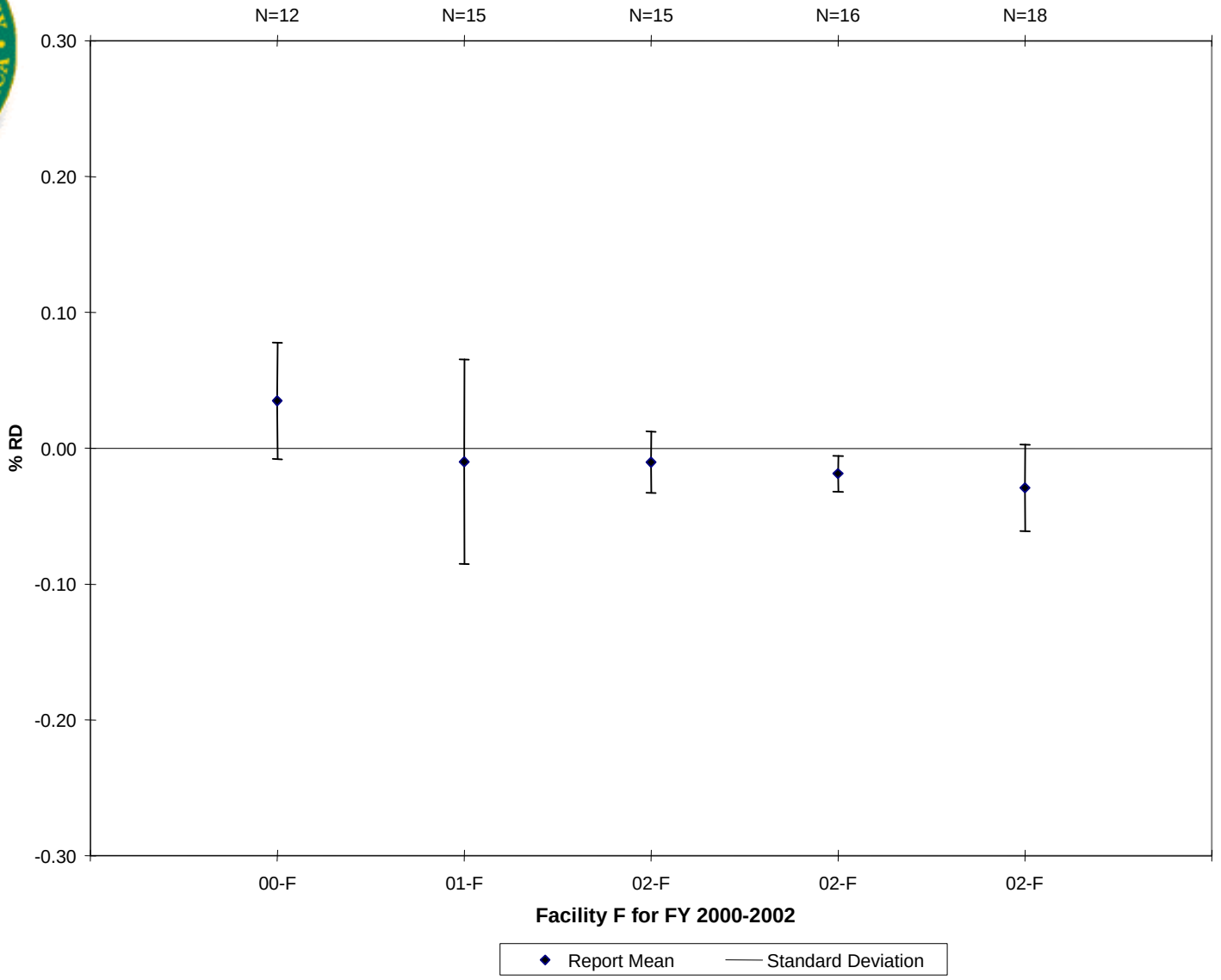
[u(s) = 0.1]



New Brunswick Laboratory Safeguards Measurement Evaluation Program

U235 Enrichment - LEU

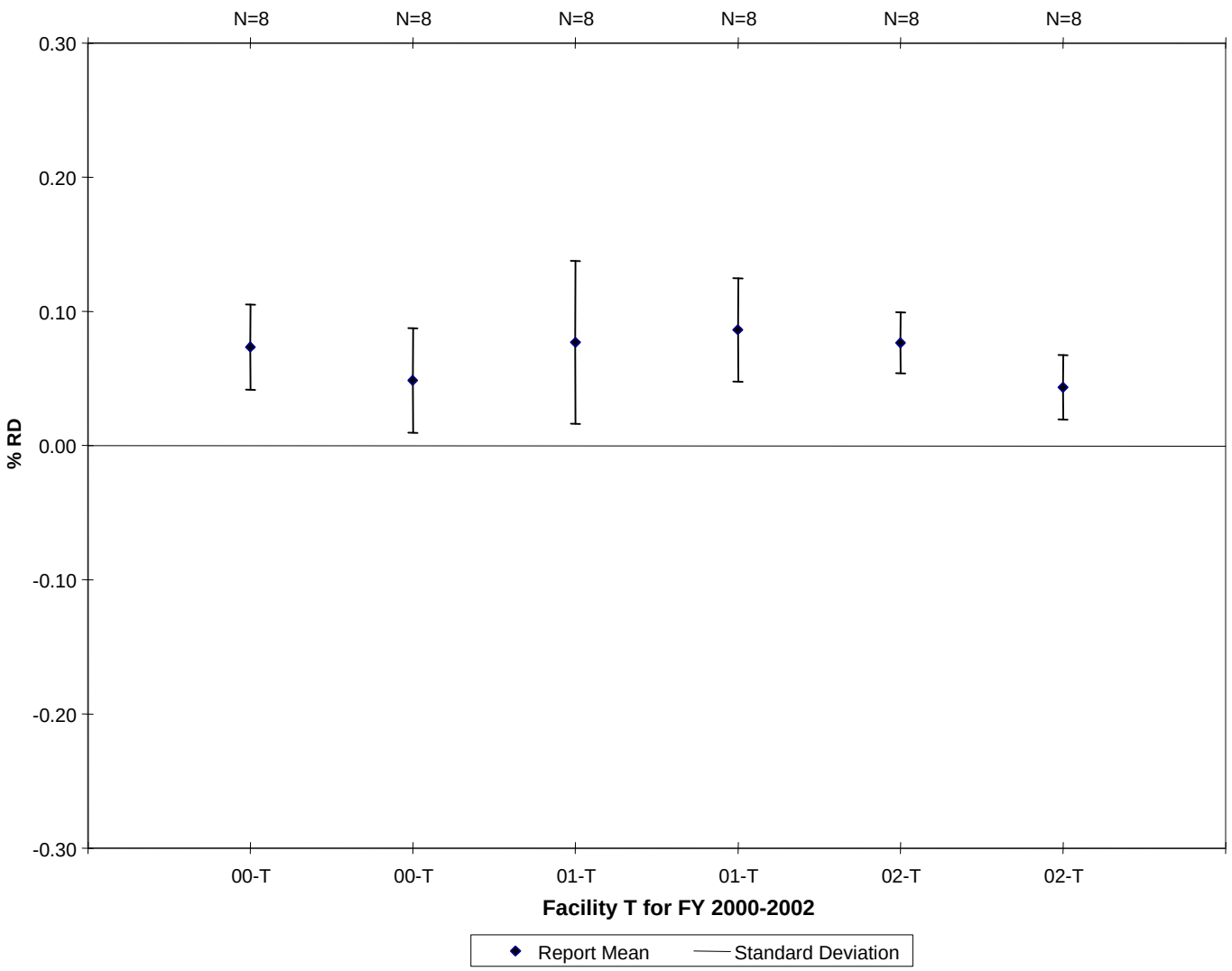
[u(s) = 0.1]



New Brunswick Laboratory Safeguards Measurement Evaluation Program

U235 Enrichment - LEU

[u(s) = 0.1]



Safeguards Measurement Evaluation Program Initiatives

- Several sites submit data electronically via email
- All sites receive data evaluation reports via email in Adobe Acrobat pdf format
- Annual report distributed in pdf format on CD at the annual meeting

NEW BRUNSWICK LABORATORY

CALORIMETRY EXCHANGE PROGRAM

CY2002 Summary

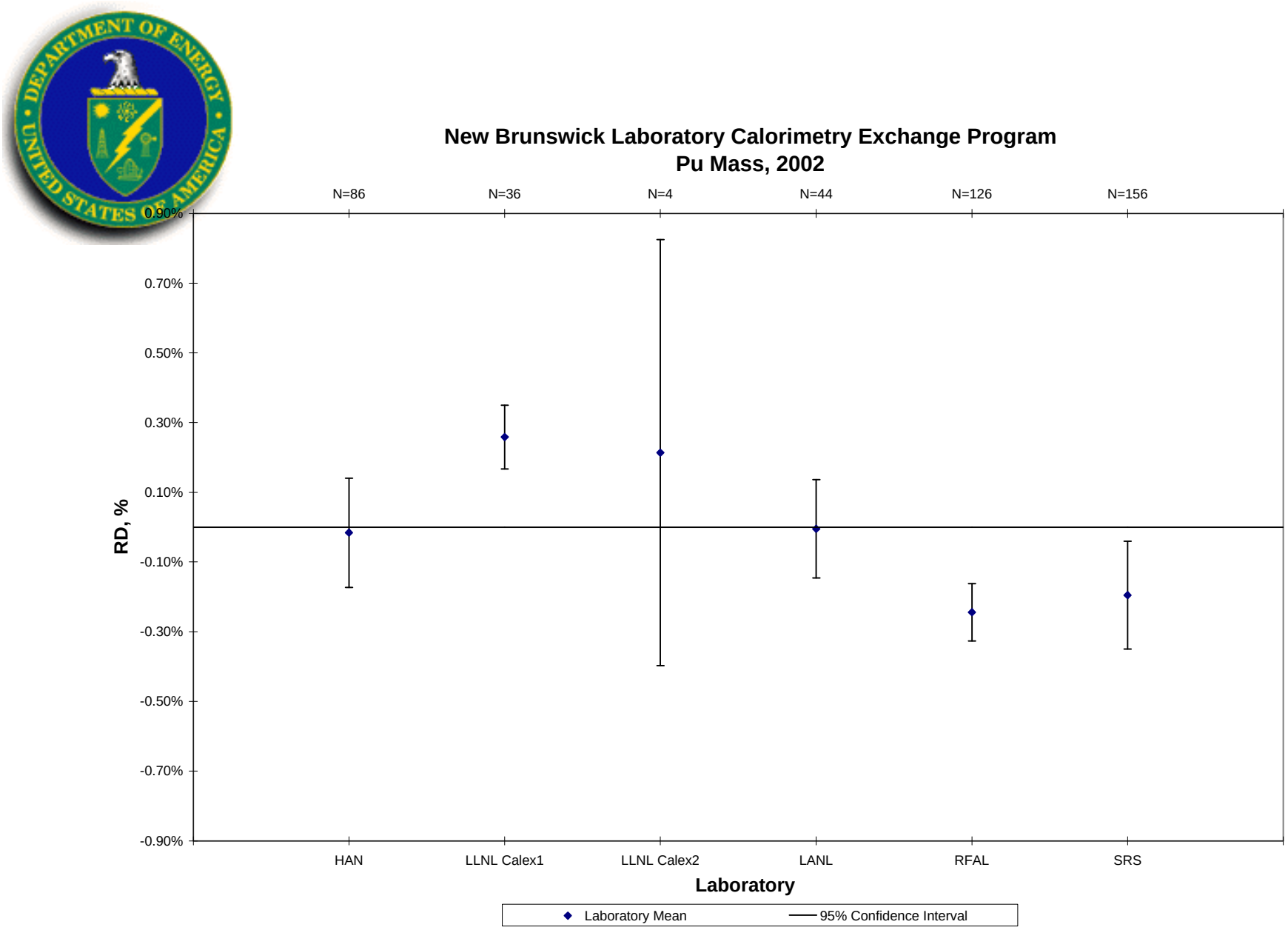
Jay Thompson

International Target Values for Calorimetry

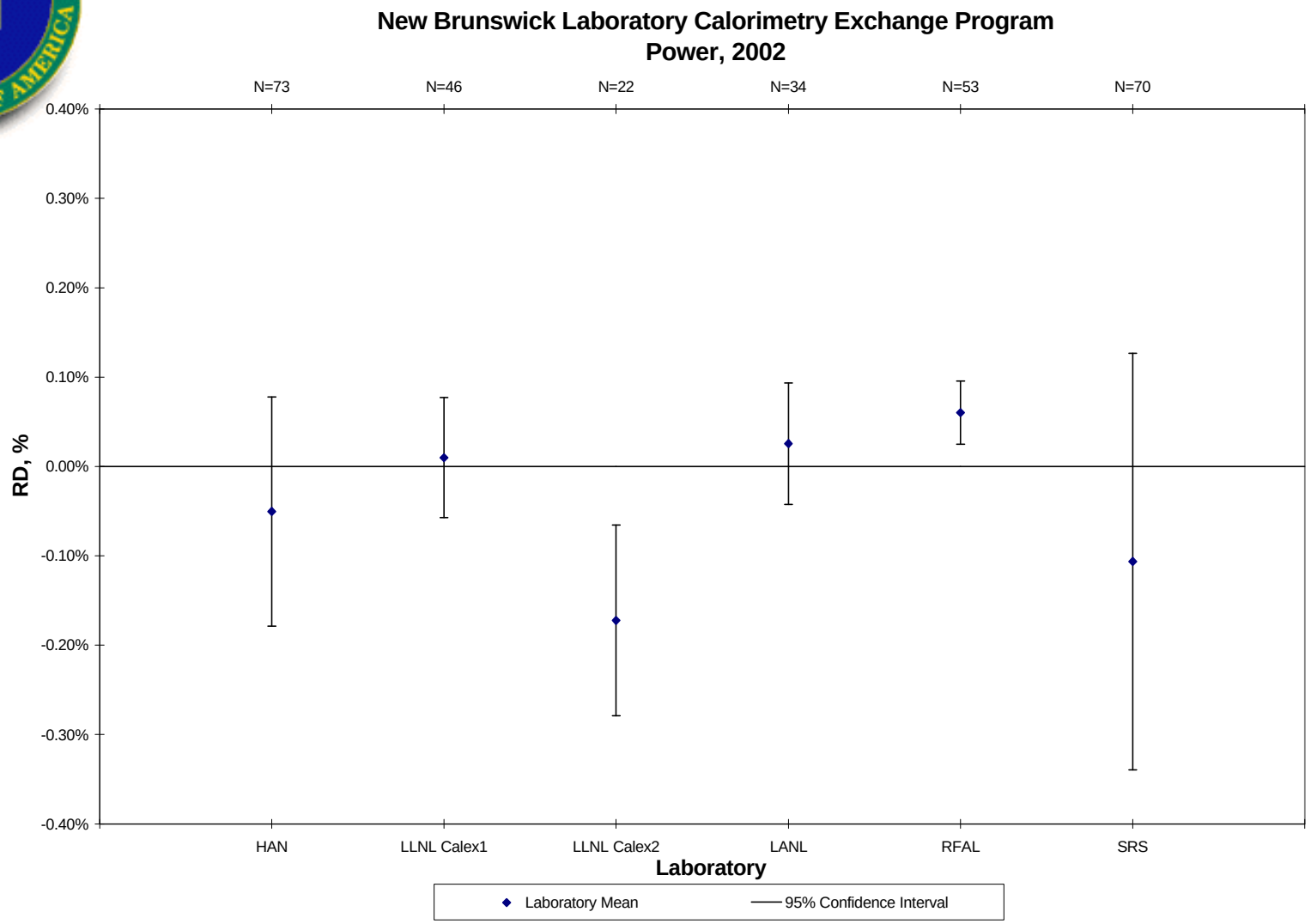
$u(r) = 0.4\%$

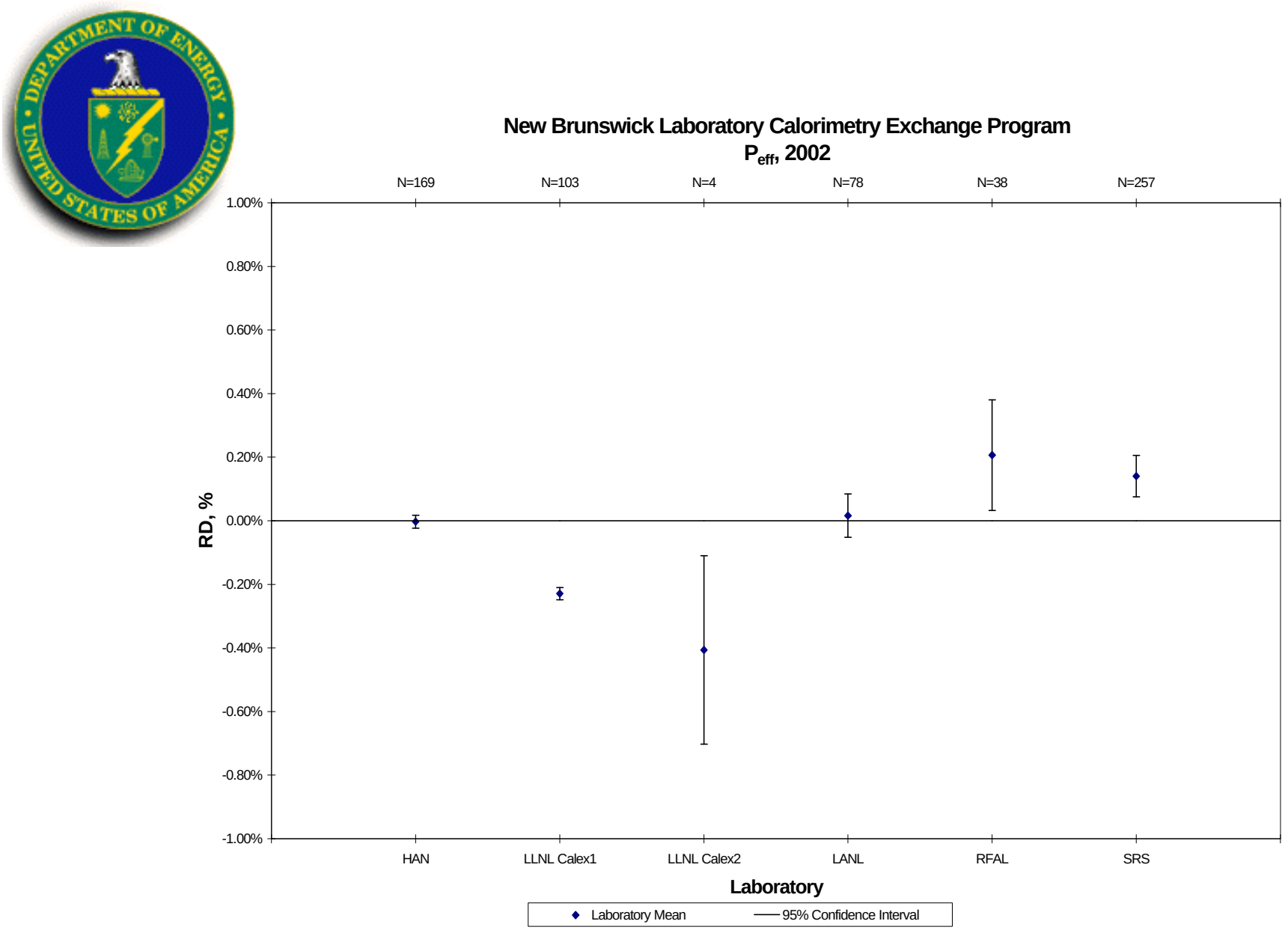
$u(s) = 0.4\%$

- Isotopic determination by mass spectrometry and alpha spectrometry
- ^{241}Am content determined by gamma spectrometry or alpha spectrometry
- Lower uncertainties are achievable for materials containing low burnup Pu



New Brunswick Laboratory/Office of Security/U.S. Department of Energy


New Brunswick Laboratory Calorimetry Exchange Program
Power, 2002
N=73
N=46
N=22
N=34
N=53
N=70
0.40%
0.30%
0.20%
0.10%
0.00%
-0.10%
-0.20%
-0.30%
-0.40%
RD, %
HAN
LLNL Calex1
LLNL Calex2
LANL
RFAL
SRS
Laboratory
Laboratory Mean
95% Confidence Interval



New Brunswick Laboratory/Office of Security/U.S. Department of Energy

2000 ITVs

Table 6: Plutonium Isotope Assay of Pu Oxide and MOX

(% Relative Standard Uncertainties)

Material Type	Isotope Ratio	Typical Value for Ratio (*100)	Method					
			TIMS[1/]		HRGS[2/]		LMCA[3/]	
			u(r)	u(s)	u(r)	u(s)	u(r)	u(s)
High-Burnup Pu	$^{238}Pu/^{239}Pu$	1.7	1.5	1	2	2	1	1
	$^{240}Pu/^{239}Pu$	43	0.1	0.05	1	1	0.7	0.7
	$^{241}Pu/^{239}Pu$	13	0.2	0.2	1	1	0.7	0.7
	$^{242}Pu/^{239}Pu$	8	0.2	0.3				
Low-Burnup Pu	$^{238}Pu/^{239}Pu$	0.02	10	10	10	10	5	5
	$^{240}Pu/^{239}Pu$	6	0.15	0.1	2	2	1.5	1.5
	$^{241}Pu/^{239}Pu$	0.2	1	1	2	2	1	1
	$^{242}Pu/^{239}Pu$	0.05	2	2				

1.) $^{238}Pu/^{239}Pu$ by alpha spec./TIMS combination

2.) Measurement time 3 x 100 sec.

3.) Measurement time 3 x 1000 sec.; 0.5 g Pu.

Isotopic Assay of Pu Oxide

(Derived % Relative Standard Uncertainties)

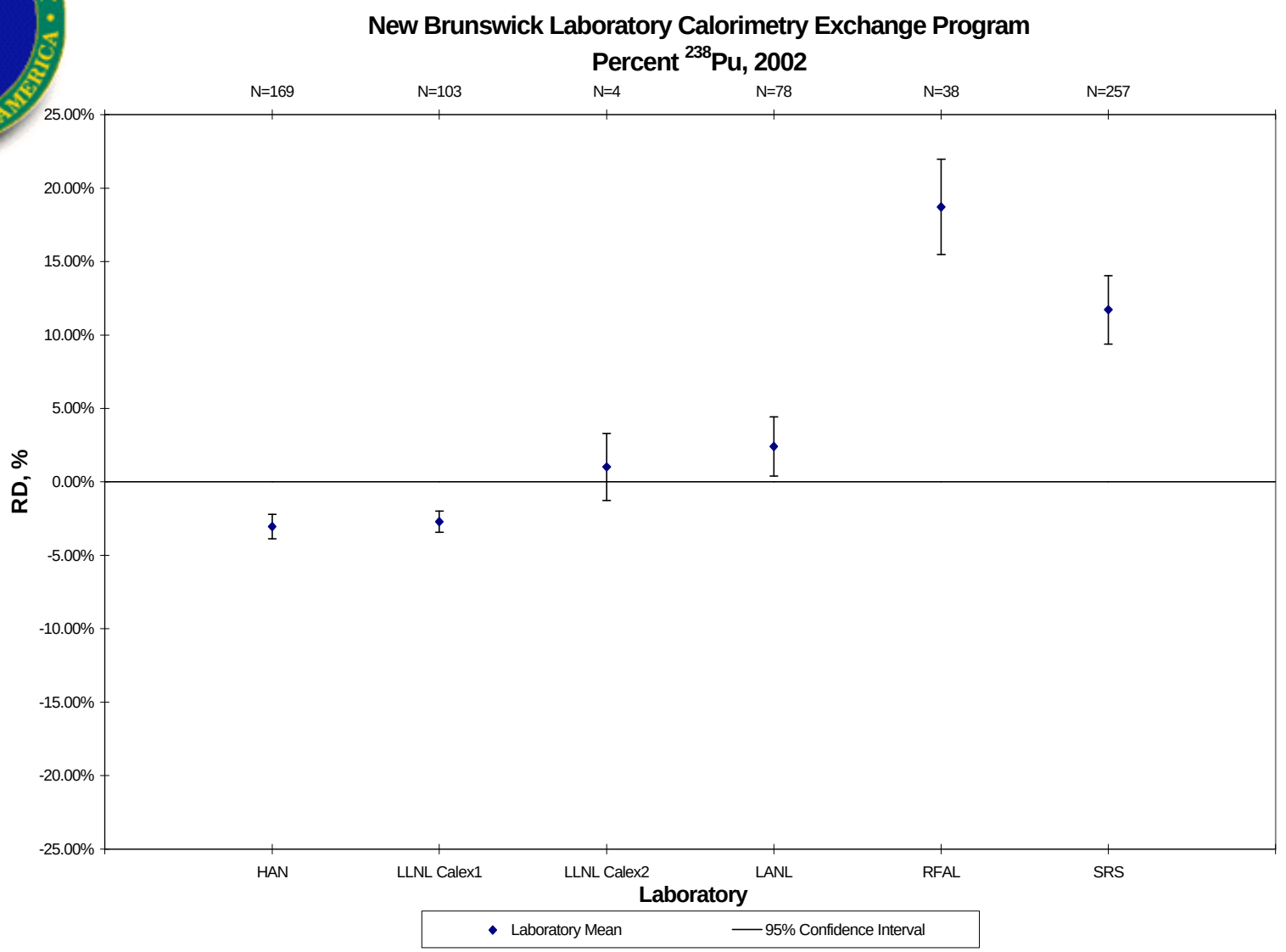
Material Type	Isotope	Typical Weight Percent	Method			
			TIMS[1/]		HRGS[2/]	
			u(r)	u(s)	u(r)	u(s)
High-Burnup Pu	^{238}Pu	1	1.5	1	2	2
	^{239}Pu	60	0.06	0.04	0.45	0.45
	^{240}Pu	26	0.12	0.07	1	1
	^{241}Pu	8	0.2	0.2	1	1
	^{242}Pu	5	0.2	0.3		
Low-Burnup Pu	^{238}Pu	0.02	10	10	10	10
	^{239}Pu	94	0.01	0.01	0.12	0.12
	^{240}Pu	6	0.15	0.1	2	2
	^{241}Pu	0.2	1	1	2	2
	^{242}Pu	0.05	2	2		

1.) ^{238}Pu/^{239}Pu by alpha spec./TIMS combination

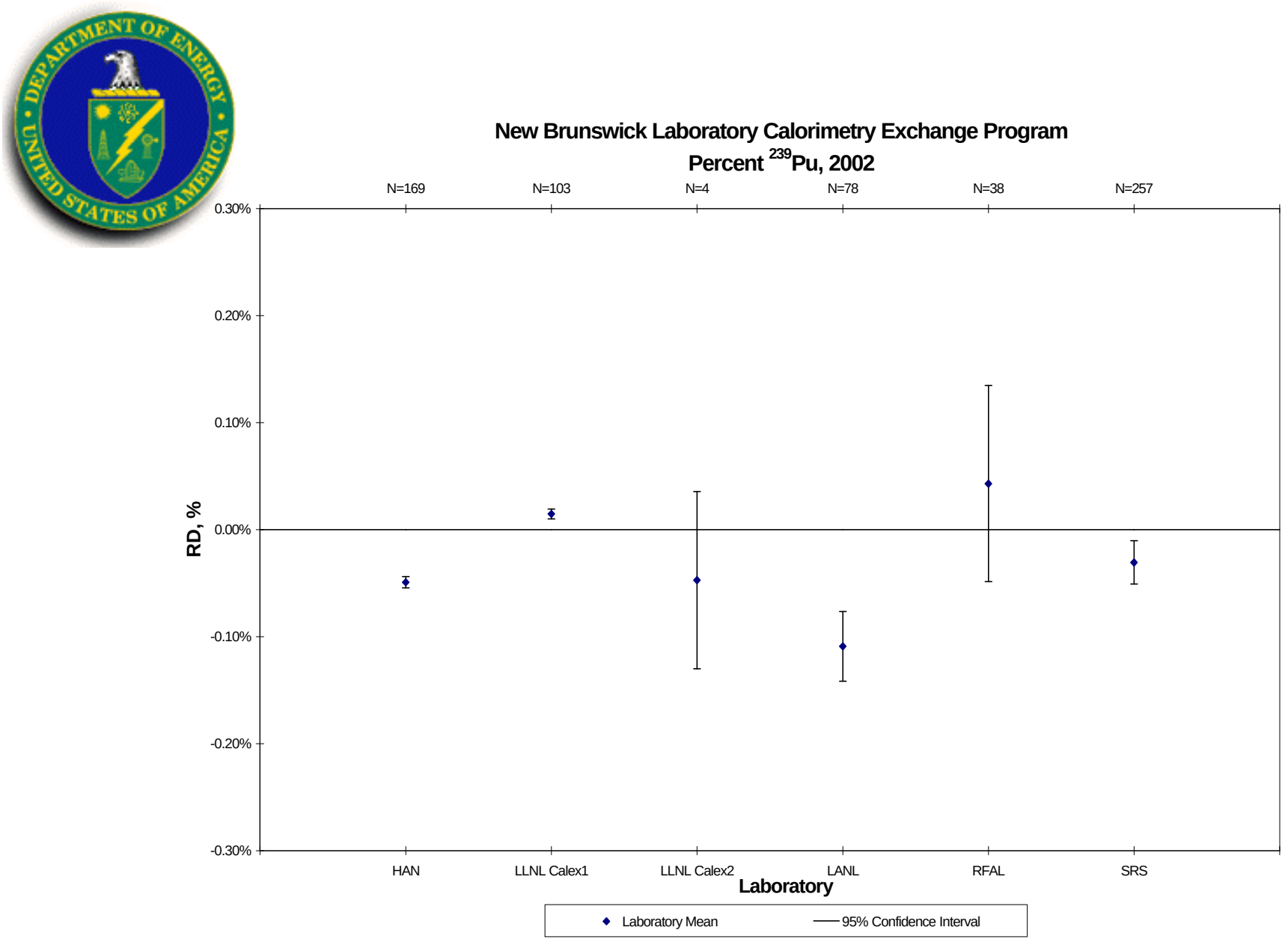
2.) Measurement time 3 x 100 sec.

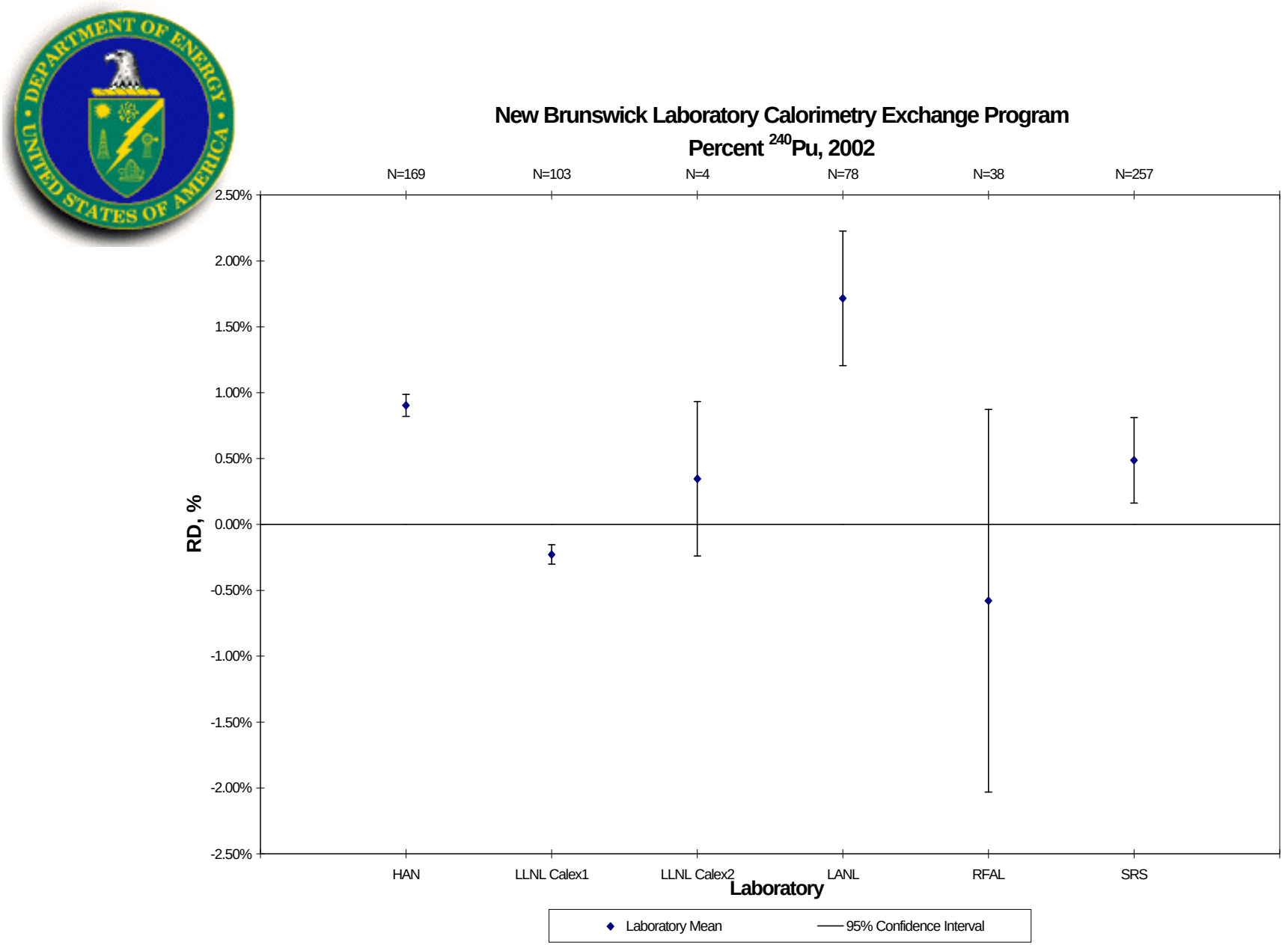
3.) Measurement time 3 x 1000 sec.; 0.5 g Pu.

New Brunswick Laboratory/Office of Security/U.S. Department of Energy

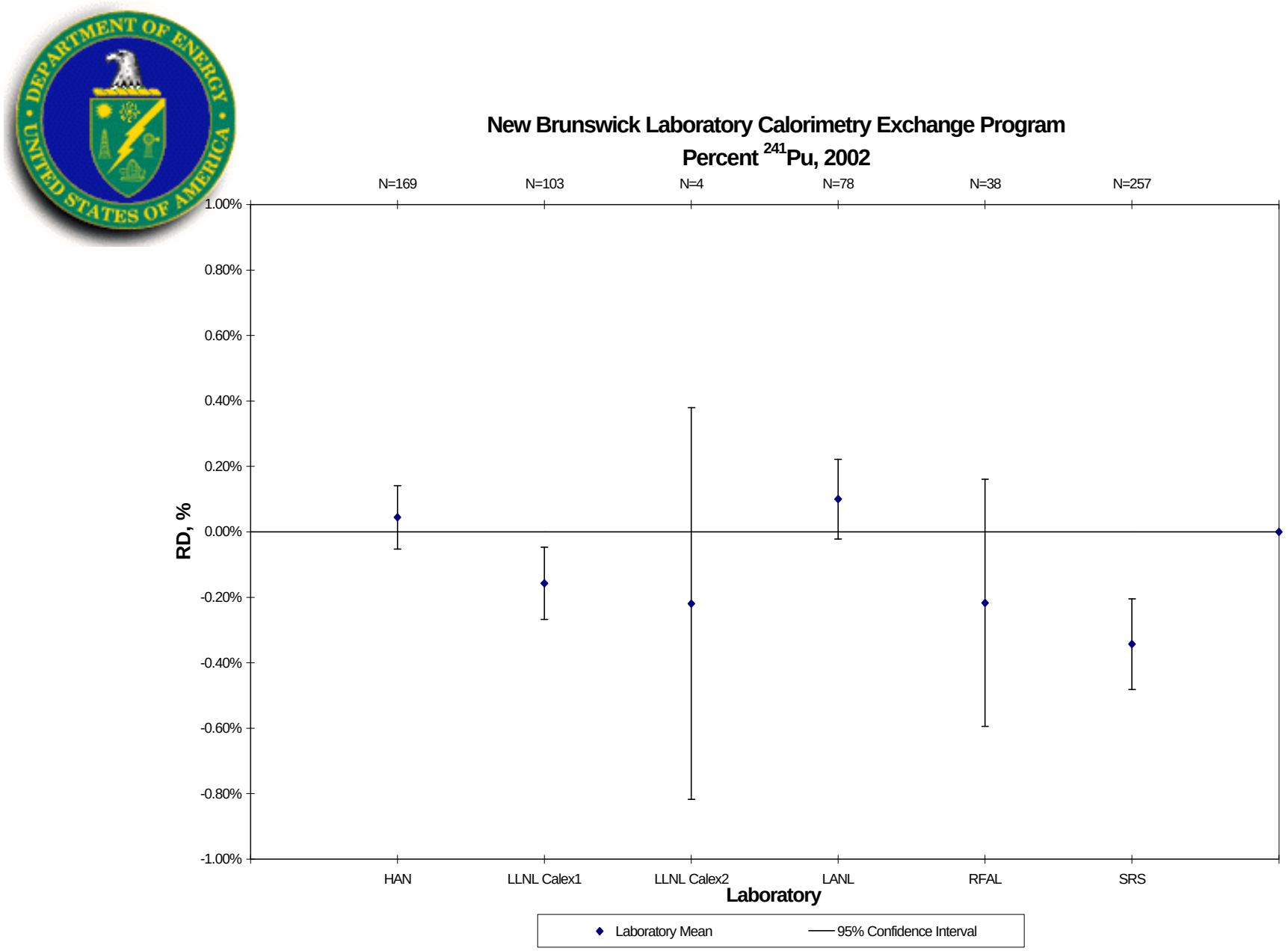


New Brunswick Laboratory/Office of Security/U.S. Department of Energy

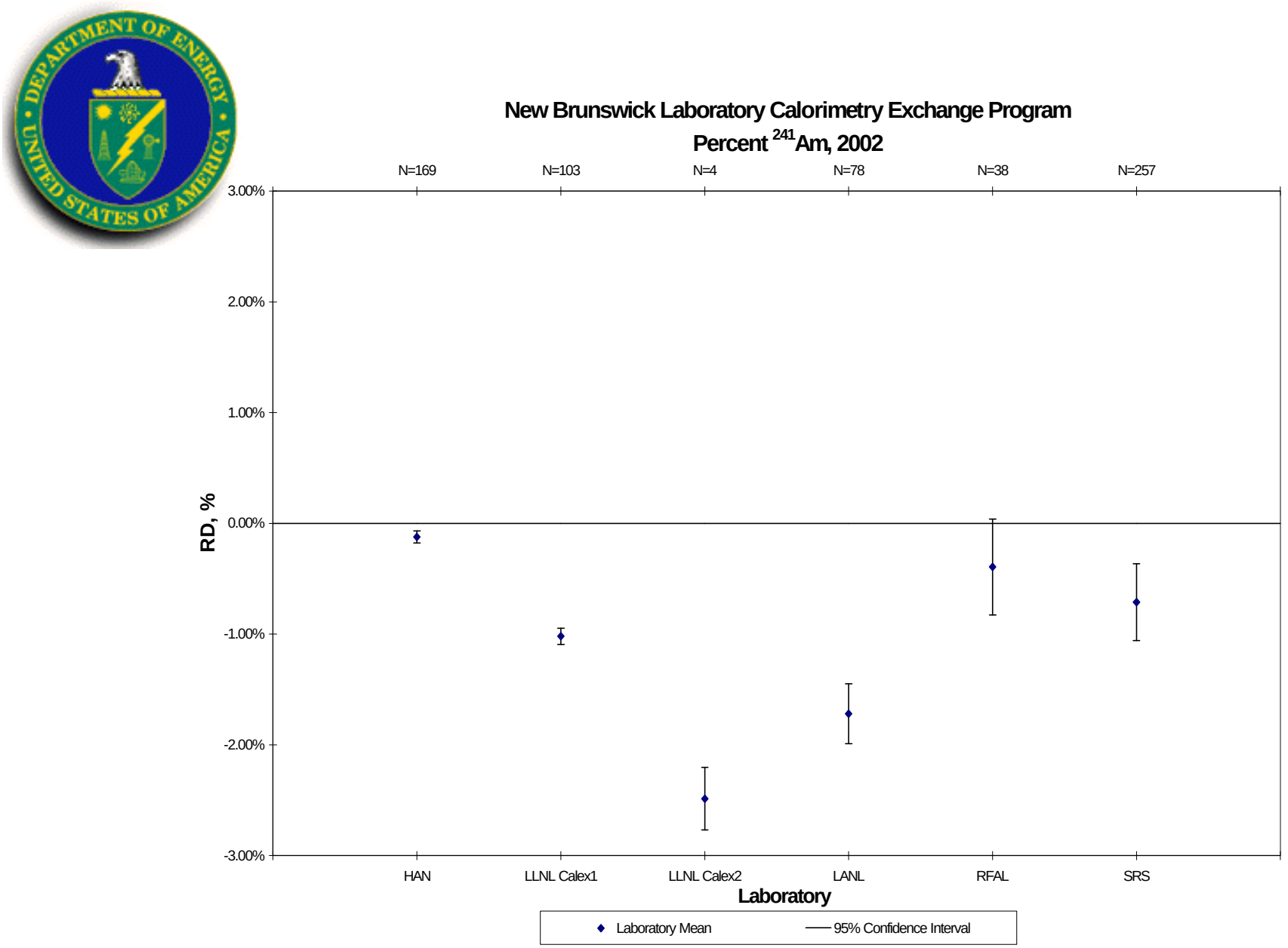

New Brunswick Laboratory Calorimetry Exchange Program
Percent 239Pu, 2002
N=169
N=103
N=4
N=78
N=38
N=257
0.30%
0.20%
0.10%
0.00%
-0.10%
-0.20%
-0.30%
RD, %
HAN
LLNL Calex1
LLNL Calex2
LANL
RFAL
SRS
Laboratory
Laboratory Mean
95% Confidence Interval



New Brunswick Laboratory/Office of Security/U.S. Department of Energy



New Brunswick Laboratory/Office of Security/U.S. Department of Energy


New Brunswick Laboratory Calorimetry Exchange Program
Percent 241Am, 2002
N=169
N=103
N=4
N=78
N=38
N=257
3.00%
2.00%
1.00%
0.00%
-1.00%
-2.00%
-3.00%
RD, %
HAN
LLNL Calex1
LLNL Calex2
LANL
RFAL
SRS
Laboratory
Laboratory Mean
95% Confidence Interval

Calorimetry Exchange 2002 Performance Summary

	HAN	LLNL1	LLNL2	LANL	RFAL	SRS
Mean Pu Mass	-0.02	0.26	0.21	0.00	-0.24	-0.20
sd	0.73	0.27	0.38	0.46	0.47	0.98
95% CI	0.16	0.09	0.61	0.14	0.08	0.15
Mean Power	-0.05	0.01	-0.17	0.03	0.06	-0.11
sd	0.55	0.23	0.24	0.19	0.13	0.98
95% CI	0.13	0.07	0.11	0.07	0.04	0.23
MeanPeff	0.00	-0.23	-0.41	0.02	0.21	0.14
sd	0.13	0.10	0.19	0.30	0.53	0.53
95% CI	0.02	0.02	0.30	0.07	0.17	0.07
Mean238	-3.03	-2.71	1.02	2.41	18.71	11.71
sd	5.48	3.66	1.43	8.92	9.87	18.89
95% CI	0.83	0.72	2.28	2.01	3.24	2.32
Mean239	-0.05	0.01	-0.05	-0.11	0.04	-0.03
sd	0.03	0.02	0.05	0.14	0.28	0.16
95% CI	0.01	0.00	0.08	0.03	0.09	0.02
Mean240	0.90	-0.23	0.35	1.72	-0.58	0.49
sd	0.55	0.38	0.37	2.26	4.42	2.64
95% CI	0.08	0.07	0.59	0.51	1.45	0.32
Mean241	0.04	-0.16	-0.22	0.10	-0.22	-0.34
sd	0.64	0.56	0.38	0.54	1.15	1.13
95% CI	0.10	0.11	0.60	0.12	0.38	0.14
MeanAm241	-0.12	-1.02	-2.49	-1.72	-0.39	-0.71
sd	0.35	0.37	0.18	1.20	1.32	2.81
95% CI	0.05	0.07	0.28	0.27	0.43	0.35

Calorimetry Exchange 2002 Performance Summary

	HAN	LLNL1	LLNL2	LANL	RFAL	SRS	ITV %
Mean Pu Mass	-0.02	0.26	0.21	0.00	-0.24	-0.20	0.4*
sd	0.73	0.27	0.38	0.46	0.47	0.98	0.4*
Mean Power	-0.05	0.01	-0.17	0.03	0.06	-0.11	0.4
sd	0.55	0.23	0.24	0.19	0.13	0.98	0.4
MeanPeff	0.00	-0.23	-0.41	0.02	0.21	0.14	
sd	0.13	0.10	0.19	0.30	0.53	0.53	
Mean238	-3.03	-2.71	1.02	2.41	18.71	11.71	10.00
sd	5.48	3.66	1.43	8.92	9.87	18.89	10.00
Mean239	-0.05	0.01	-0.05	-0.11	0.04	-0.03	0.12
sd	0.03	0.02	0.05	0.14	0.28	0.16	0.12
Mean240	0.90	-0.23	0.35	1.72	-0.58	0.49	2
sd	0.55	0.38	0.37	2.26	4.42	2.64	2
Mean241	0.04	-0.16	-0.22	0.10	-0.22	-0.34	2
sd	0.64	0.56	0.38	0.54	1.15	1.13	2
MeanAm241	-0.12	-1.02	-2.49	-1.72	-0.39	-0.71	
sd	0.35	0.37	0.18	1.20	1.32	2.81	

Calorimetry Exchange Program Updates

- Annual report will be published electronically; will appear on new NBL website
- Participating facilities will change
- CalEx-2 standard reference values will be improved

Savannah River Technology Center
SRTC
We Put Science to Work

Status of the Performance Demonstration Project at SRS

Raymond Dewberry and Saleem Salaymeh
Savannah River Technology Center

Linda Baker and Don Faison
Central Laboratory Facility

David Eisele and Don McCurry
KAMS Facility

Savannah River Site
Aiken, SC 29808

Performance Demonstration Program Measurements

William H. Geist and Norbert Ensslin
Los Alamos National Laboratory

Larry Kayler and Michelle Cameron
Rocky Flats Environmental Technology Site

Wendy Rhodes
OS-13 DOE-HQ

Performance Demonstration Program

Savannah River Technology Center
SRTC
We Put Science to Work

Objectives:

- To ensure that consistent results are obtained from various NDA techniques.
- To provide greater confidence in inventory values.
- To identify causes of biases which contribute to shipper/receiver differences.

Performance Demonstration Program

Savannah River Technology Center
SRTC
We Put Science to Work

Scope:

- Evaluate calorimeter, isotopic, and neutron data in different facilities.
- Evaluate Likely Biases Between shipper/receiver NDA Measurements for Shipping RFETS Pu Material to SRS K-Area Material Storage facility.
- Determine applicable correction factors.

Performance Demonstration Program

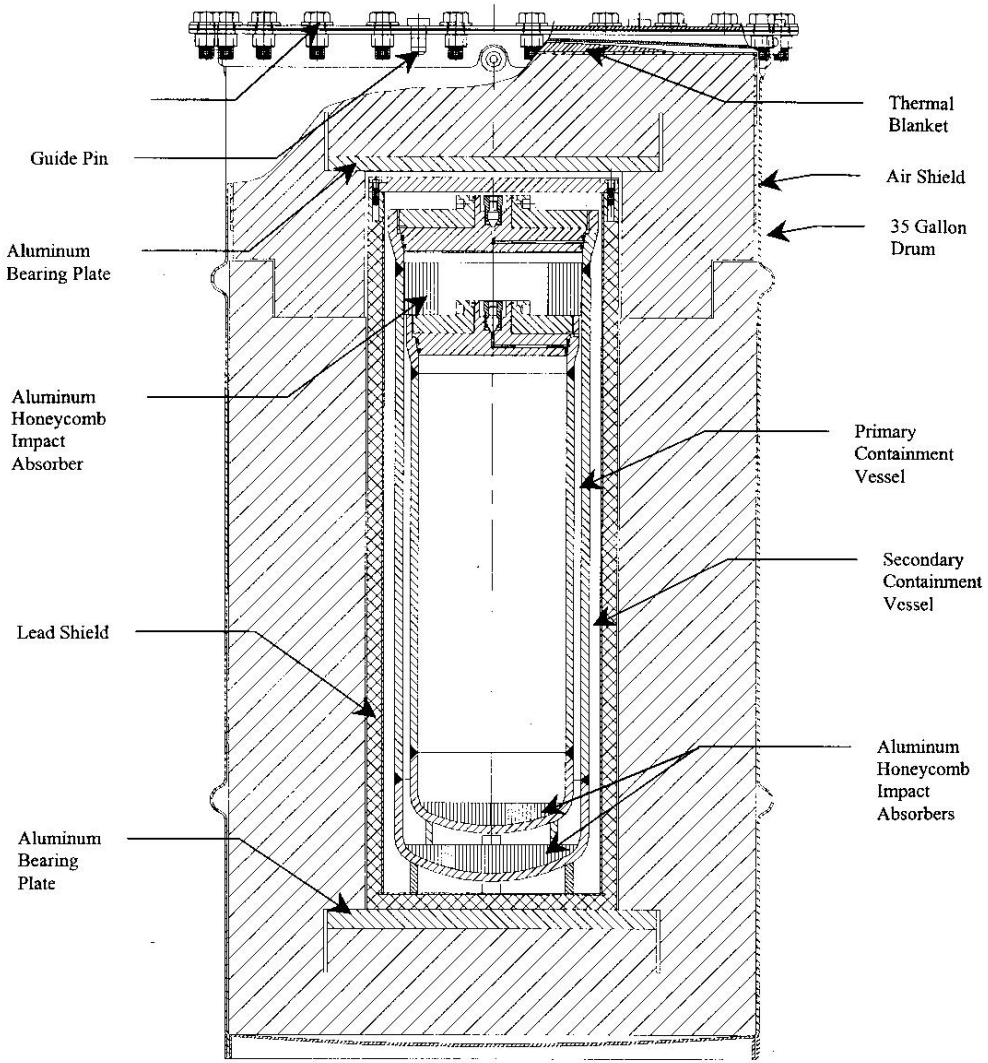
Savannah River Technology Center
SRTC
We Put Science to Work

Scope: (continued)

- RFETS shipper measurements are performed by calorimetry inside 3013 containers.
- SRS KAMS receipt measurements are performed by neutron multiplicity counting inside 9975 shipping container.

PDP 9975 Measurements

PDP 9975 Measurements



Performance Demonstration Program

Savannah River Technology Center
SRTC
We Put Science to Work

Phase 1:

- Designated Six working standards for characterization at RFETS.
- Calormetry, isotopic measurements, and neutron in the 3013 and the 9975 shipping container at Rocky.
- Isotopic and neutron measurements in the 9975 container at SRS.
- Benchmark SRS KAMS NMC using ^{252}Cf and Calex standards measurements versus FB-Line.

Performance Demonstration Program

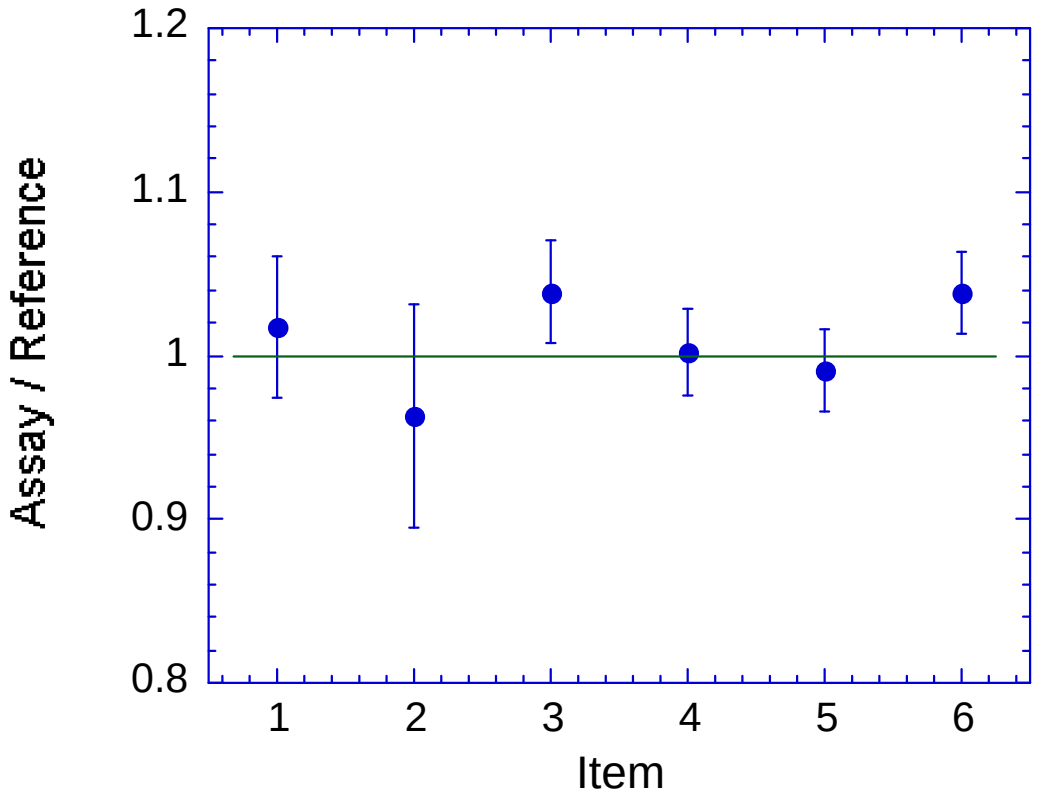
Savannah River Technology Center
SRTC
We Put Science to Work

3013 Initial Characterization:

- In support of the shipper receiver agreement between SRS and RFETS, 3 Oxide and 3 Metal 3013 samples were prepared and characterized by cal/gamma measurements and NMC. These samples provided the basis for the PDP study.
- The results of this characterization study were provided to SRS and LANL for further analysis.

PDP 3013 Results

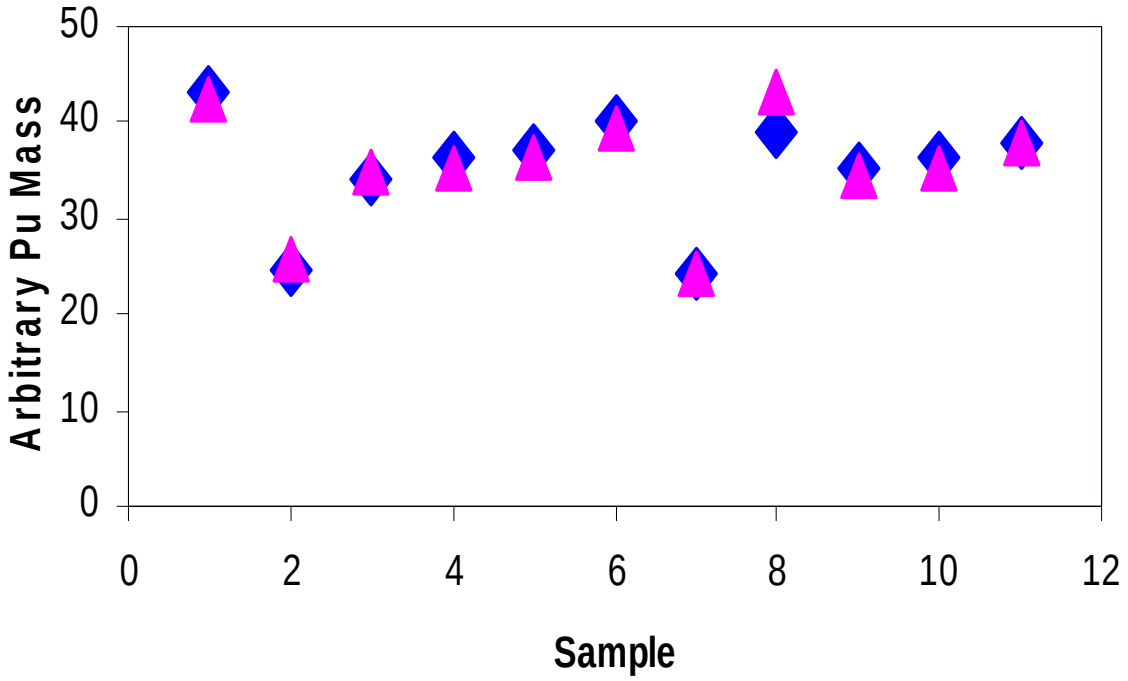
- Reference values were determined from calorimetry (R).
- The mass values determined from the neutron Assay (A).



Average A/R = 1.009

1 σ rsd = 0.030

3013 Initial Characterization:



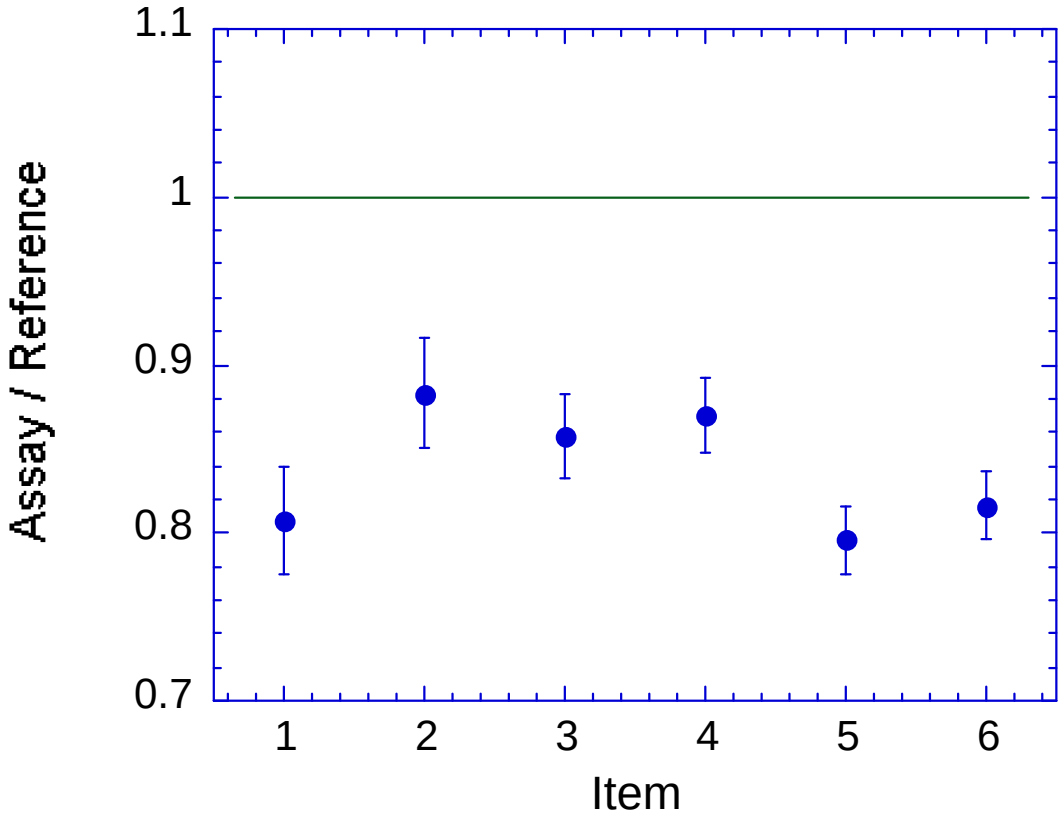
◊ Pu arbitrary mass by NMC
Δ Pu arbitrary mass by calorimetry

RFETS PDP 9975 Results

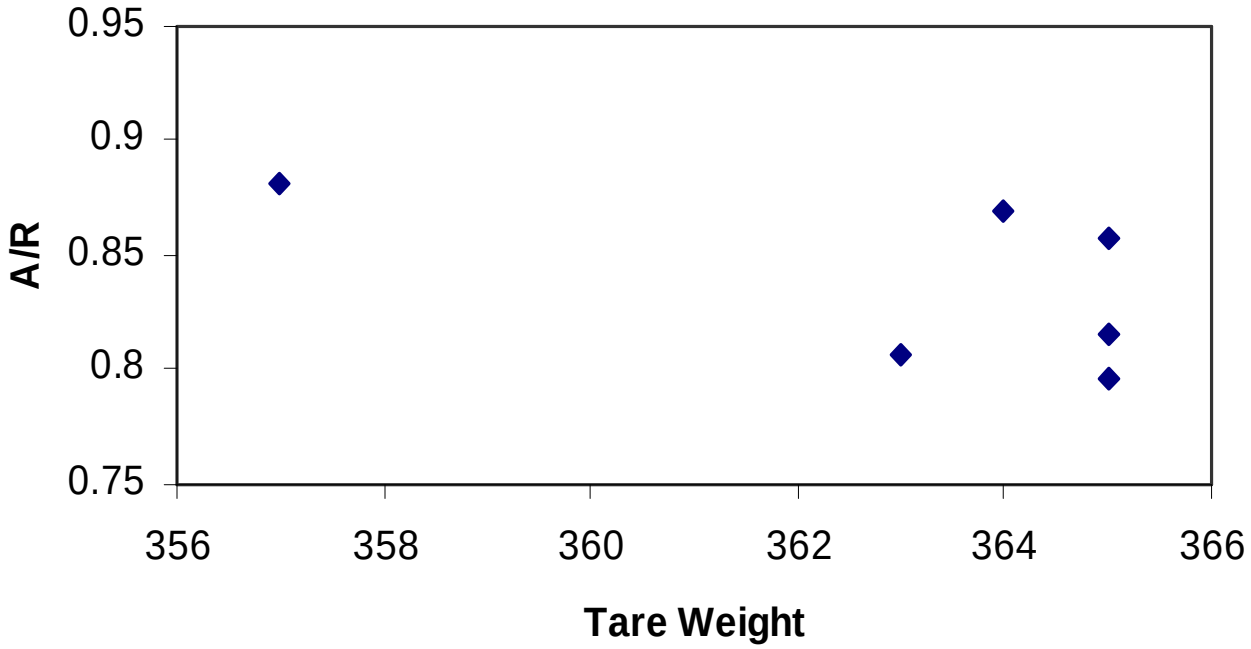
Savannah River Technology Center
SRTC
We Put Science to Work

10% to 20% bias caused by:

- 9975 shipping container.
- variations in Celotex and other components of the 9975.



RFETS PDP 9975 Results

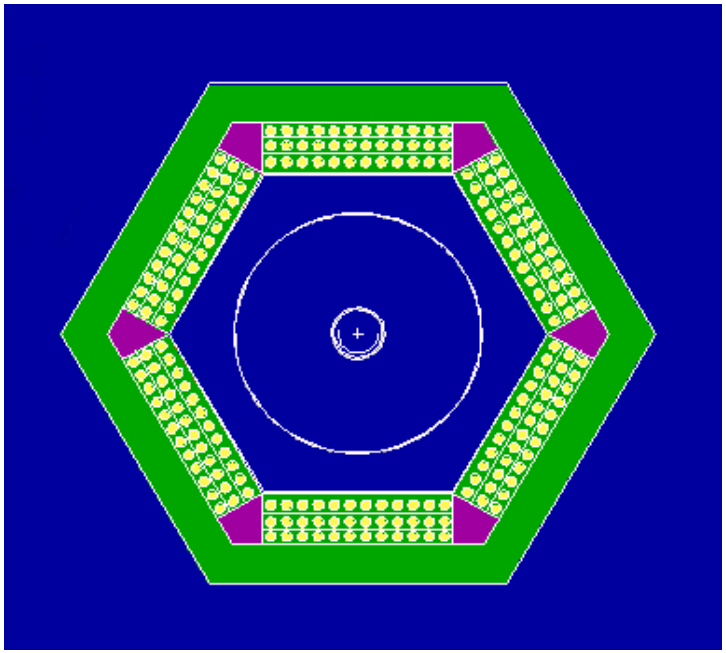


PDP FY02 Conclusions

Savannah River Technology Center
SRTC
We Put Science to Work

- Completed all measurements.
- Completed data analysis of the 3013 and 9975.
- Good agreement for cal/gamma and NMC assay in 3013.
- 10% to 20% bias caused by the 9975 containers.
- Issued a technical report on results of phase I.
- Future work:
  - •Complete phase I measurements.
  - • Send results to NBL for a statistical analysis.
  - • Determine the cause of the bias in the 9975 data.
  - •Complete KAMS measurements inside 9975.

Drum Neutron Multiplicity Counter

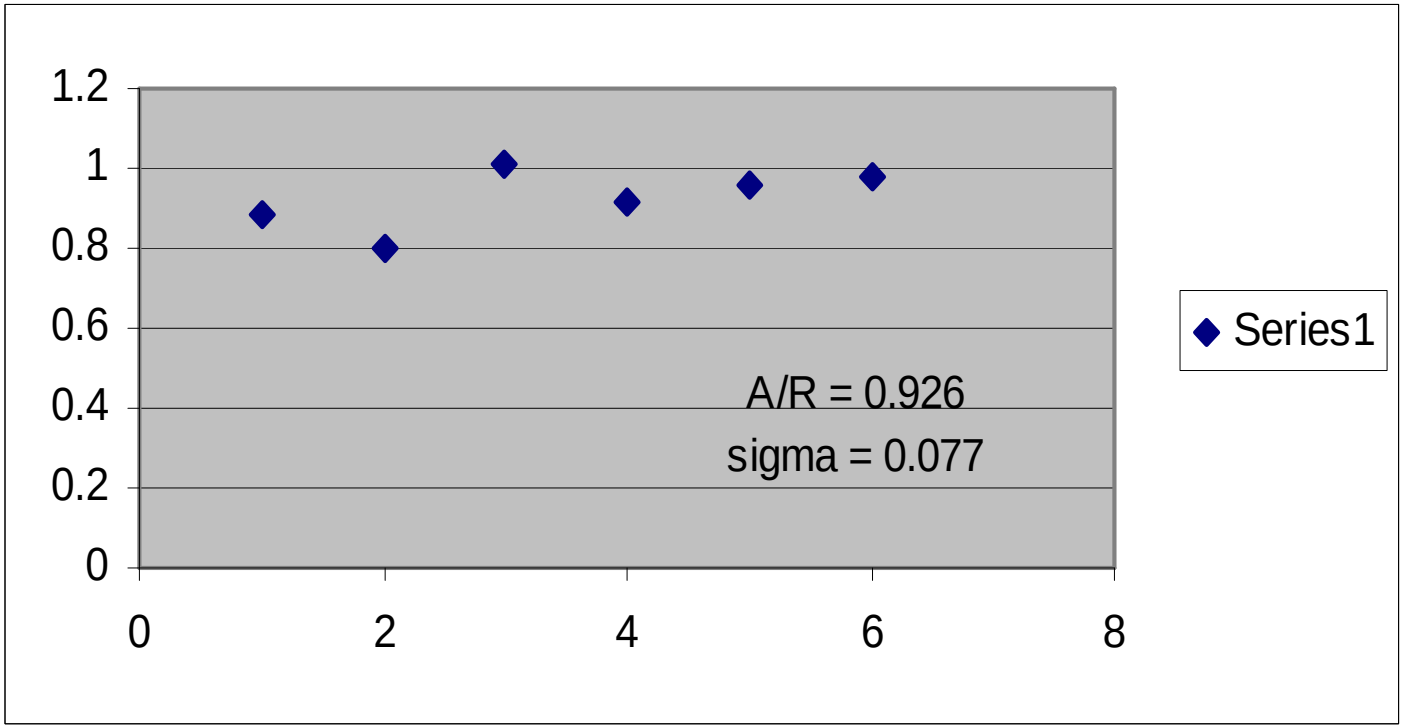


- Final Counter
  - 198 tubes
  - 3 rings
  - 10 atm

SRS KAMS Characterization

High Voltage	1740 V
Pre-delay time	2.5 usec
Die-Away time	37.3 ± 0.87 usec
Gate Width	35 usec
Dead time Parameters (NCC)	a = $71.56 \times 10^{-9} \pm 1.4 \times 10^{-9}$ sec
	b = 0 $usec^2$
Dead time parameters (multiplicity)	c = $15.63 \times 10^{-9} \pm 0.76 \times 10^{-9}$ sec
	d = $15.75 \times 10^{-9} \pm 2.48 \times 10^{-9}$ sec
Dead time parameter (τ)	19.15 ± 0.45 nsec
Doubles Gate Fraction	0.5633 ± 0.0005
Triples Gate Fraction	0.3340 ± 0.0007
Efficiency (Cf-252 point source)	0.516 ± 0.008
Cf-252 ρ_0	0.5346 ± 0.0005
Cf-252 a	674.0 ± 8.4 cps/ nanogram Cf-252
Efficiency (Pu-240 estimated)	0.526 ± 0.008
Pu-240 ρ_0	0.2554 ± 0.0010
Pu-240 a	139.9 ± 0.9 cps / g Pu-240 effective

SRS KAMS Characterization



KAMS GIS Qualification

Savannah River Technology Center
SRTC
We Put Science to Work

Item id		547	
Spectrum	declared	60	67
Pu-238	0.01%	0.008%	0.010%
Pu-239	93.71%	94.59%	94.78%
Pu-240	6.08%	5.36%	5.17%
Pu-241	0.18%	0.017%	0.017%
Pu-242	0.02%	0.019%	0.018%
Pu-240eff	6.14%	5.41%	5.22%
Average	5.32	Std. Dev.	0.13

KAMS GIS Qualification

Savannah River Technology Center
SRTC
We Put Science to Work

Standard		Declared Pu-240eff		Measured Pu-240 eff	
1		6.14%		(5.32 +/- 0.13)%	
2		5.99%		(5.81 +/- 0.23)%	
3		5.91%		(5.34 +/- 0.24)%	
4		5.77%		(5.58 +/ -0.16)%	
5		5.79%		(5.75 +/- 0.10)%	
6		6.02%		(5.98 +/- 0.08)%	

Conclusion

- We have succeeded to reproduce the measurement bias in the RFETS acquisitions using the MCNPX code.
- KAMS NMC results are better than RFETS NMC measurements in 9975 and have adequate precision to distinguish oxide standards from metal standards.
- KAMS measurements by NMC are biased low by (7% +/- 8%) for six working standards versus (16% +/- 4%) at RFETS.
- KAMS GIS measurements are attainable with 4 hours acquisitions, but 9975 lead shielding introduces significant difficulties.
- PDP measurements were an important contribution toward recognizing problems and generating corrective action.

Experiences with Reference Materials

Prepared by

Jeffrey Berg, Andrew Maddison,

and Michael Michlik

Analytical Laboratory

Nuclear Technology Division

Argonne National Laboratory

Certified Reference Materials

- **CRM 126 (^{239}Pu metal) to calibrate ^{244}Pu spikes**
- **CRM 135 (^{235}U Uranyl Nitrate solution) to calibrate ^{233}U spikes**

Combination Spikes

- **Samples requiring U and Pu analyses**
- **Decontamination (^{233}U oxide contains 3900 µg/g ^{239}Pu and 82 µg/g ^{241}Am with a balance of ^{232}Th and ^{233}U daughters).**

^{233}U Spike Preparation

U_3O_8 (1.2 to 1.8g) Dissolved in 8M HNO_3

↓

Convert to 9M HBr matrix (10mL)

↓

Load on AG1-X2 (20cc)

↓

Rinsed with 60mL 9M HBr
Am/Pu/Np/Th eluted

↑

Elute Uranium with 100mL 0.1M HCl

↓

Convert to 8M HNO3 matrix

↓

Diluted to 1000mL volume

MC&A Requirements for Spike Calibration

- **At least annually**
- **Suspect problems, e.g. Control Standard indication**
- **New Spike**

^{233}U Spike History

- **Spike dissolved and decontaminated in 1997 and stored in glass bottles**
- **Concentration History (mg/g)**
  - 1997 – 0.79426
  - 1998 – 0.79553
  - 1999 – 0.79566
  - 2000 – 0.79481
  - 2001 – 0.79343
  - 2002 – 0.79560
  - 2003 – 0.79531 & 0.79416

Spread of 0.28%, eliminating 2001 – spread of 0.176%

CRM 135 Certificate of Analysis

U. S. Department of Energy
New Brunswick Laboratory

New Brunswick Laboratory
Certified Reference Materials
Certificate of Analysis

CRM 135

Uranium-235 Spike Assay and Isotopic Solution Standard

Uranium Concentration	28.270 ± 0.051 μmoles/gram
Uranium-234	0.0442 ± 0.0004 At. %
Uranium-235	99.8195 ± 0.0013 At. %
Uranium-236	0.0574 ± 0.0004 At. %
Uranium-238	0.0789 ± 0.0004 At. %

This Certified Reference Material (CRM) is an assay and isotopic standard for use as a spike in the analysis of uranium materials by isotope dilution mass spectrometry (IDMS). Each unit of CRM 135 contains approximately 100 milligrams of uranium, dissolved in 0.8 N HNO_3, sealed in a glass ampoule.

The indicated uncertainty for the concentration is the tolerance limit for at least 99% coverage with a probability level of 0.95. In brief, at least 99% of the measured values on all ampoules should fall within the indicated interval with a probability of 0.95. This statistical approach is necessary due to the concentration variability between ampoules. (See page 14 of "The Role of Standard Reference Materials in Measurement Systems," NBS Monograph 148, 1975, for a more detailed explanation of the tolerance limit concept.) Since isotopic composition shows little variability between ampoules, the indicated uncertainties for the isotopic composition are 95% confidence limits for the mean.

This CRM was originally issued in 1975 by the National Bureau of Standards (NBS) as Standard Reference Material (SRM) 993. The measurements made at NBS leading to the certification were performed by E. L. Garner and L. A. Machlan, under the direction of I. L. Barnes. In 1987, the technical and administrative transfer of NBS Special Nuclear SRMs into the NBL CRM Program was coordinated by the NBS Office of Standard Reference Materials and N. M. Trahey, NBL.

October 1, 1987
Argonne, Illinois
(Revision of NBS Certificate dated June 30, 1975)

Carleton D. Bingham
Director

(Over)

Uranium Concentration . . . . . .
. . . . 28.270 ± 0.051 μmoles/gram
(0.180% rsd)

In brief, at least 99% of the measured values on all ampoules should fall within the indicated interval with a probability of 0.95. This statistical approach is necessary due to the concentration variability between ampoules.

2001 SME Report

- **"The 2000 International Target Value (ITV) of 0.1% for systematic error is exceeded for both enrichment levels." Note: Prior to this date, a DOE systematic error of 0.5% was used.**
- **Recalculation of SME samples using 2002 concentration gave results within the 0.1% ITV.**

2003 Spike Calibration

- **^{233}U spike calibration using separate sealed ampoules of CRM 135.**

2003 Spike Calibration Data

Pioneering Science and Technology

Ampoule #144			Ampoule #200	
Date	Conc (mg/g)		Date	Conc (mg/g)
3/28/2003	0.79602		4/4/2003	0.79522
"	0.79528		"	0.79359
"	0.79576		"	0.79434
"	0.79533		"	0.79441
"	0.79542		4/10/2003	0.79328
"	0.79554		"	0.79331
3/31/2003	0.79471		"	0.79457
"	0.79439		"	0.79461
4/21/2003	0.79528		4/21/2003	0.79320
"	0.79610		"	0.79492
"	0.79501		"	0.79435
4/23/2003	0.79475		"	0.79454
"	0.79552		"	0.79395
"	0.79484		"	0.79456
"	0.79528		"	0.79357
Average	0.79528		Average	0.79416
% rsd 1s	0.061		% rsd 1s	0.080
Difference between sets				
0.141%				

Spike Calibration Options

- **Use NBL's ^{233}U spike (CRM 111) – more difficult to tailor to samples and presently have a large inventory of ORNL ^{233}U.**
- **Use a solid sample such as CRM 116 – currently not available**
- **Continue using CRM 135 to calibrate spike**

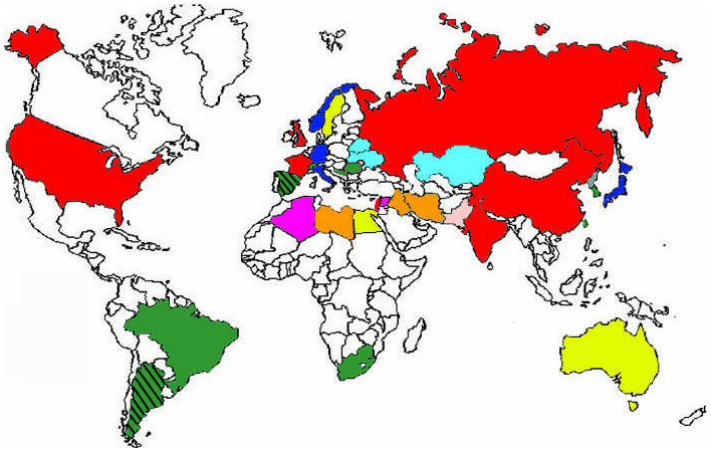
Status of Certified Reference Material Production

Jon W. Neuhoff, NSND Director
New Brunswick Laboratory
MEP Annual Meeting - Phoenix, Arizona
July 13, 2003

NBL is the U.S. Government's Certifying Authority for Nuclear Reference Materials

- We produce, certify, and distribute Certified Reference Materials (CRMs) for nuclear material accountability and verification measurements
- We provide CRMs to facilities in order to ensure traceability of their nuclear material accountability and verification measurements to a national and international measurements database
- We assist facilities in the preparation and characterization of Working Reference Materials (WRM) to ensure their traceability to a higher tier CRM
- We customize our CRMs based upon customer needs (e.g., dilution, splitting)

NBL is Focused on Meeting the Needs of our Domestic Customers



- Our primary focus continues to be the provision of CRMs to cover the full range of nuclear material processing
- We prioritize meeting the needs of U.S. DOE and NRC-licensed facilities
- However, these needs are balanced with urgent needs coming from our international customers and U.S. government threat reduction programs

NBL CRMs - Current Availability

- 51 NBL CRMs are currently available for purchase within the following categories:
  - Uranium and Plutonium Assay CRMs (7)
  - Uranium and Thorium Impurity CRMs (3)
  - Uranium and Plutonium Isotopic CRMs (28)
  - Uranium NDA CRMs (3)
  - Uranium and Thorium Ore CRMs (14)
- 4 NBL CRMs are in two categories (Assay and Isotopic):
  - CRMs 113-B, 115, 122, and 125-A
- NBL also produces CRM 99 (Potassium Dichromate) for our NBL-Modified Davies and Gray titrimetric method

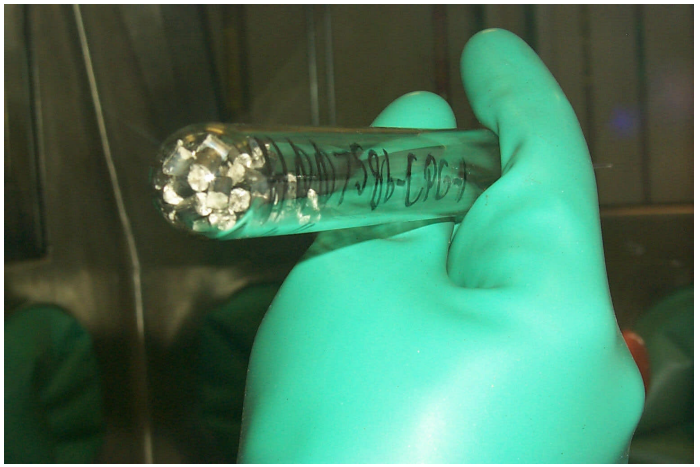
NBL CRMs - Recently-Issued

- CRM U930-D (Uranium Isotopic Standard) - 09/97
- CRM 125-A (Enriched Uranium Oxide Assay and Isotopic Standard) - 12/97
- CRM U010 (Uranium Isotopic Standard) - 09/98
- CRM 112-A (Uranium Metal Assay Standard) - 09/98
- CRM 113-B (Enriched Uranium Hexafluoride (Solid Form) Assay and Isotopic Standard) - 12/98
- CRM 146 (Enriched Uranium Gamma Spectrometry Standard) - 07/99
- CRM 149 (Uranium NDA Standard for AWCC) - 11/99
- CRM 42A(1-4) (Normal Uranium Counting Standard) - 03/01
- CRM 115 (Depleted Uranium Metal Assay Standard) – 09/02
- CRM U005-A (Uranium Isotopic Standard, 0.5% Enriched) – 09/02

NBL CRMs - Active Projects

- CRM 113-B (Enriched Uranium Hexafluoride Assay and Isotopic Standard) (re-verification of assay and isotopic abundance) **(August, 2003)** – final stage of completion
- CRM U045 (Uranium Isotopic Standard, 4.5% Enriched) **(August, 2003)** – final stage of completion
- CRM U630 (Uranium Isotopic Standard, 63% Enriched) **(August, 2003)** – final stage of completion
- CRM 116 (Enriched Uranium Metal Standard, 93% Enriched) **(March, 2004)** – issues need to be resolved
  - Oxidation and degradation of bulk material received from Y-12
  - Looking into other bulk material at Y-12 and ANL-W
  - Characterizing and qualifying received material

NBL CRMs - Active Projects



- CRM 129-A (Normal Uranium Assay and Isotopic Standard) **(September, 2003)** – highest priority CRM project at NBL; analysis is progressing
- CRM 126-A (Plutonium Metal Assay and Isotopic Standard) **(December, 2003)** – second highest priority CRM project at NBL; analysis is progressing
- These CRMs represent NBL's most in-demand reference materials and most of our efforts are focused upon completion of these two important projects in FY 2003 and early FY 2004

NBL CRMs – Near-Term Plans for U CRMs

- CRM U500 (Uranium Isotopic Standard, 50% Enriched) – repackaging bulk material for new units
- CRM U970 (Uranium Isotopic Standard, 97% Enriched) – repackaging bulk material for new units
- Californium Shuffler NDA Standard – Performance Demonstration Project (March, 2004)
- CRM U0002-A (Uranium Isotopic Standard, 0.2% Enriched) – USEC providing base material
- CRM U010-A (Uranium Isotopic Standard, 1% Enriched) – USEC providing base material
- CRM U005-B (Uranium Isotopic Standard, 0.5% Enriched) – USEC providing base material
- Uranium Isotope Calibration Mixes
- CRM 17-B (Normal Uranium Tetrafluoride Assay Standard) – repackaging bulk material for new units
- WRM support to Y-12 for several DA and NDA standards
- UF_6 standards for international safeguards – USEC providing base material
- International suite of uranium isotopic CRMs – jointly certified by NBL and IRMM

NBL CRMs – Near-Term Plans for Pu CRMs

- CRM 138-A (Plutonium Isotopic Standard)
  - Will be made from CRM 126-A to remove CRM 138 from stock (apparent 0.1% bias in major ratio in CRM 138)
- CRM 147 (Plutonium NDA Standard) – 3013 standard
- CRM 122-A (Plutonium Oxide Assay and Isotopic Standard)
- CRM 144 (Plutonium Triple Atom Spike) – awaiting Pu-244 acquisition from either Russia (Arzamas-16) or U.S. source (Mark 18-A)
- CRMs 140-143 (Plutonium Isotopic Standards) – most important will be to replenish Pu-244 once available
- Plutonium Double Atom Spike – awaiting Pu-244 acquisition
- Plutonium Impurity Standard – Pu metal matrix containing metallic/non-metallic impurities
- Plutonium NDA Standards – Weapons grade and reactor grade Pu standards in metal and oxide form
- Pu standards for international safeguards

NBL CRMs – Near-Term Plans for Other CRMs

- Mixed Oxide (MOX) Standard (U and Pu assay and isotopics with five levels of impurities)
- CRM 66 (Thorium Oxide Impurity Standard)
- Np standards for international safeguards
- WRM support to SRS for a Np standard

Calorimetry
Working Standards and Verification

Clifford Rudy, Los Alamos National Laboratory, NIS-5

LA-UR-03-4067

NBL Workshop on NDA Calibration and Standards

Phoenix, Arizona, July 13, 2003

Abstract

This presentation describes calorimetry heat standard calibration, preparation of physical standards for non-destructive assay, the use and performance of calorimetric assay for nuclear material verification, and potential problems with the two components of calorimetric assay: the gamma-ray measurement and the calorimeter thermal power measurement.

Topics

- Calibration of Pu-238 Heat Standards
- NDA Standards
- Verification with Calorimetry
- Cal/Iso Assay Caveats

Pu-238 heat standards

- Pu-238 heat standards generate a known amount of thermal power

- Pu-238 Heat Standards are used to calibrate calorimeters.

Pu-238 Heat Standard Certificate

Los Alamos
National Laboratory

Safeguards Science & Technology Group, NIS-5

Certificate of Calibration and Traceability

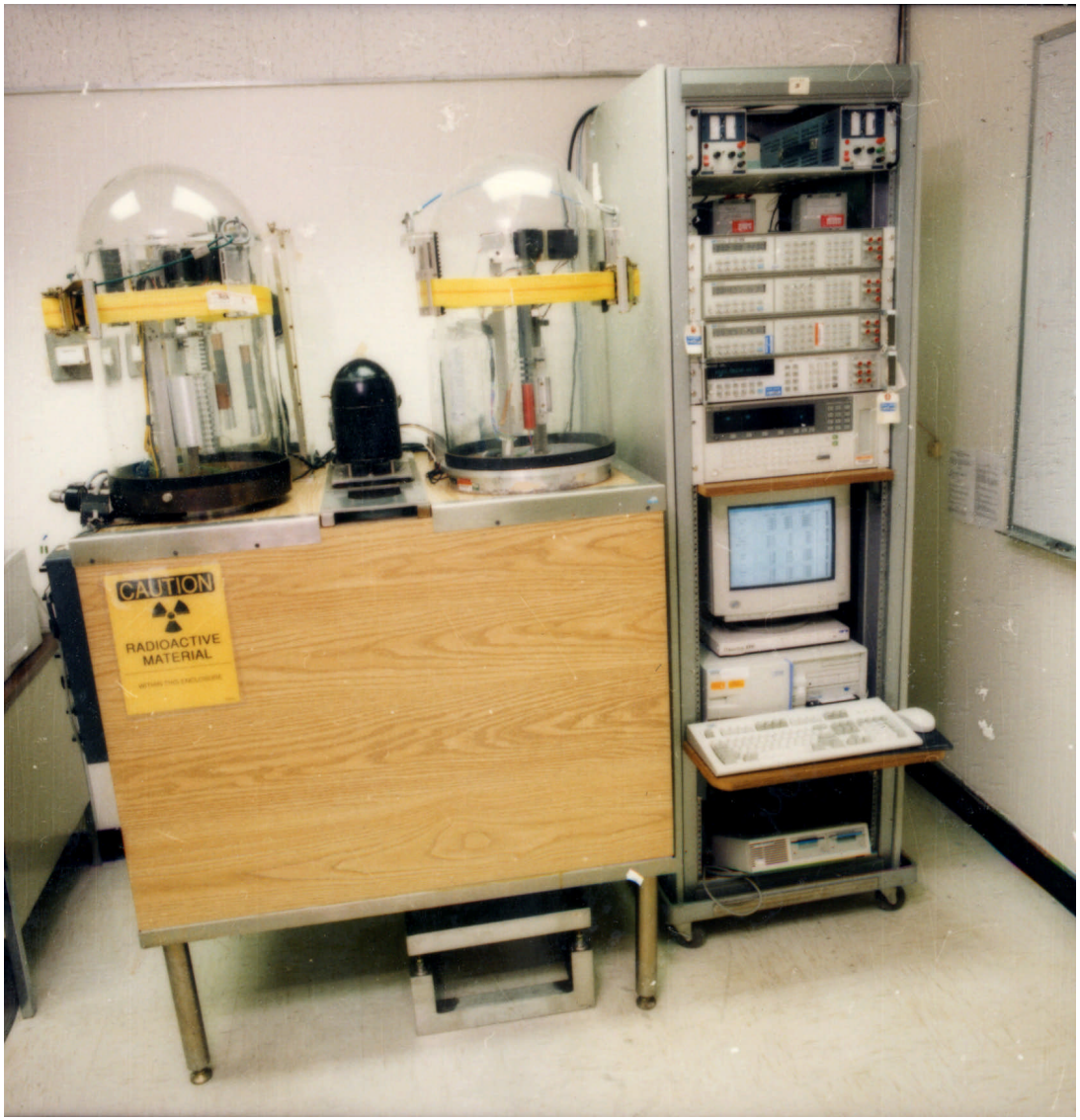
Pu-238 Heat Standard 1.5WF

The source designated as 1.5WF has been calibrated by the LANL Heat Standards Laboratory. This Calibration expires on February 1, 2003.

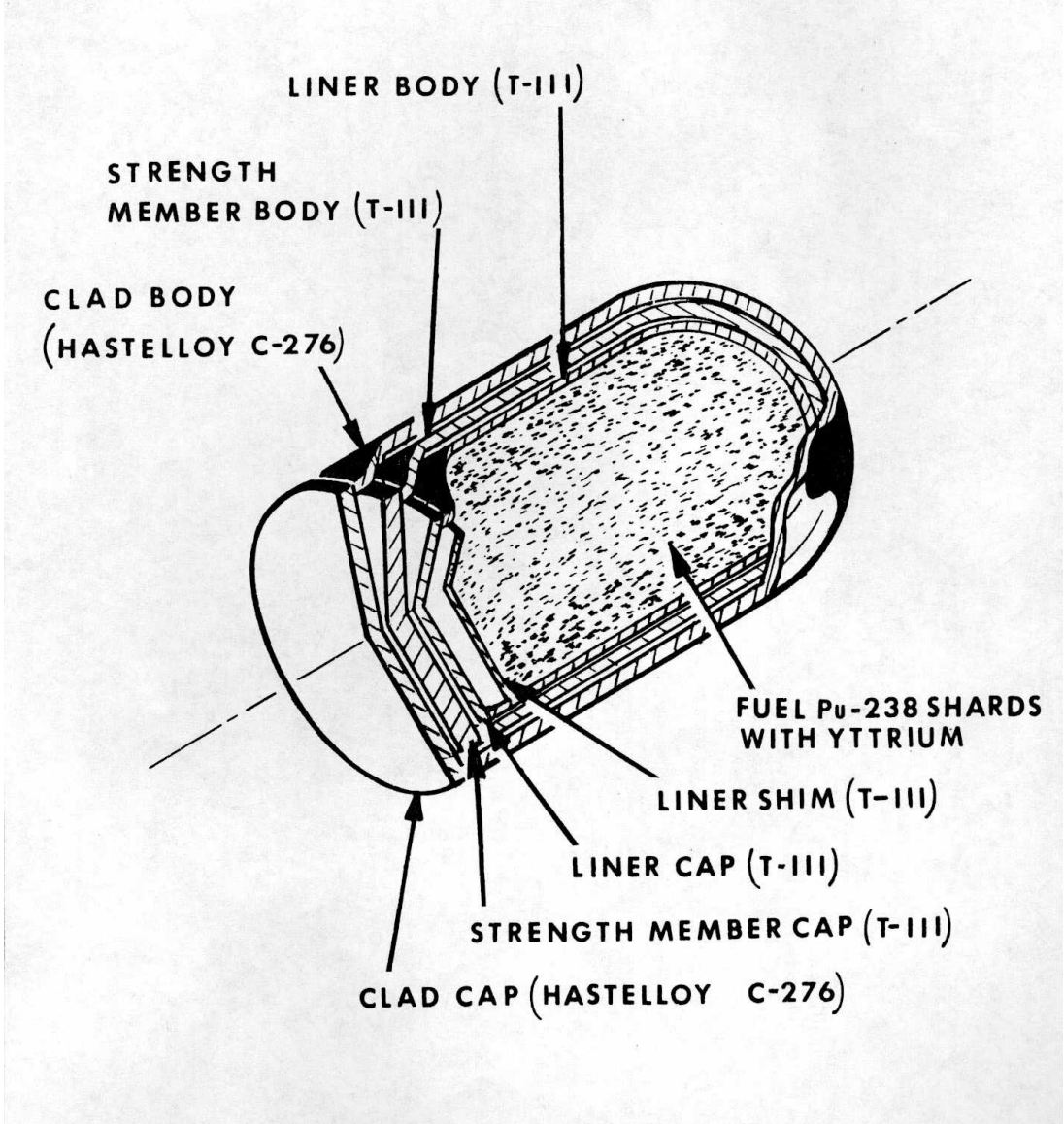
A power output of 1.275948 watts as of 12 noon MST February 1, 1998 was determined calorimetrically for this source. The uncertainty of this measurement is estimated to be no more than 0.019% at the 95% confidence level. Possible errors in the isotopic composition can contribute another 13 PPM for each year listed in the decay table supplied with this document. CAUTION: This source should be returned to the LANL Heat Standards Laboratory at the end of the calibration period for recalibration and reevaluation of the integrity of the encapsulation.

To insure capsule integrity, this source was nondestructively tested by radiography on March 26,1998 and helium-leak tested on March 26, 1998. Dose equivalent rate measurements were performed on March 26, 1998 to aid in the safe handling and storage of the source. Gamma-ray dose equivalent rate was 6.5 mrem/hr at 15 cm. Neutron dose equivalent was 0.7 mrem/hr at 15 cm. CAUTION: Exposure to sources should be kept as low as reasonably achievable.

Heat Standard Calorimeters



Pu-238 Heat Standard



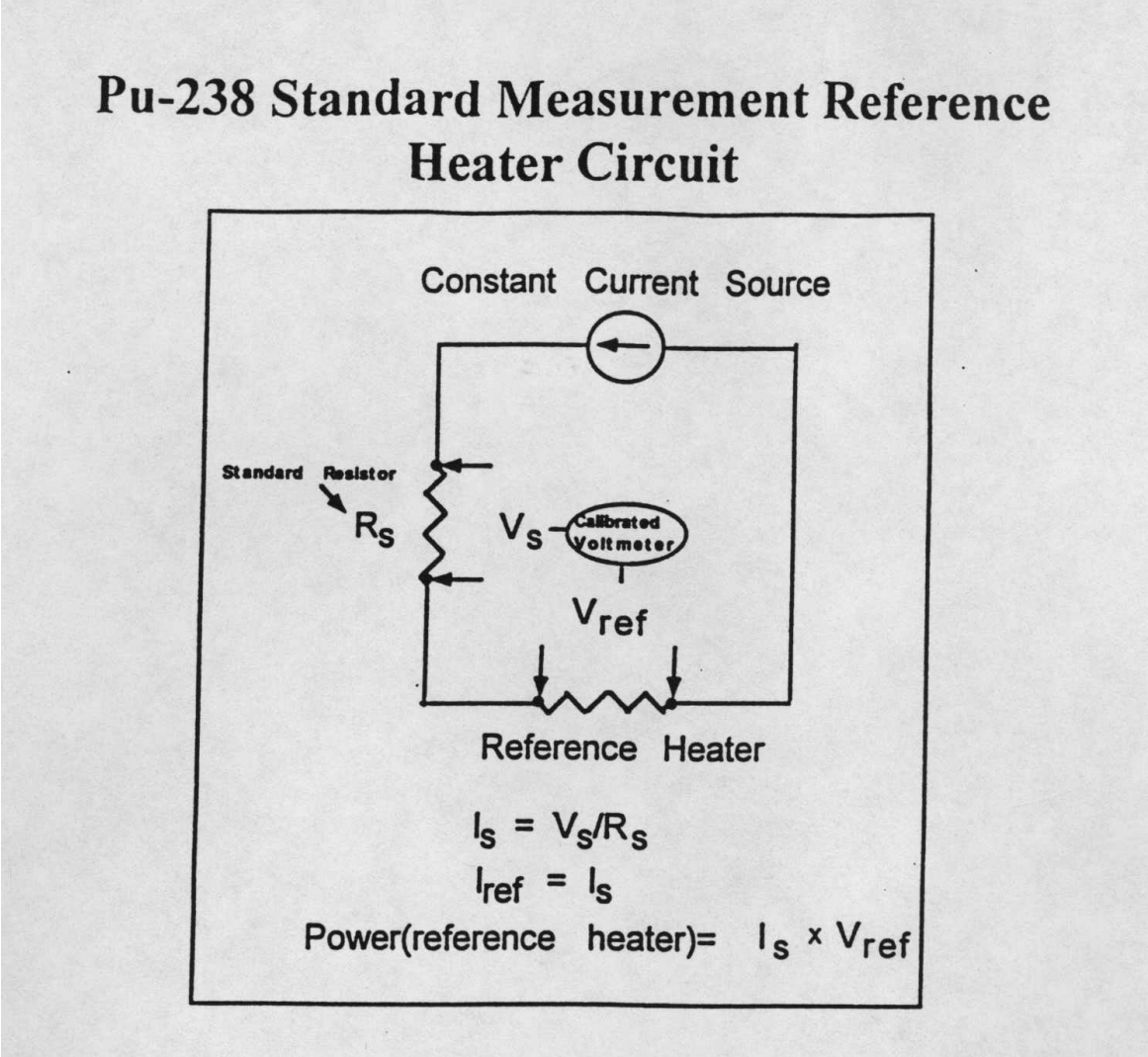
Heat Standards Inventory (1996)

Type	Dimensions (diameter x height)	N	Median power (Watts)	Range (Watts)
Eraser	1/4" x 1/4"	35	0.005	0.0008 - 0.043
Pencil	3/8" x 3/4"- 2 3/4"	118	0.94	0.0015 - 2.8
MWG	3/4" x 3/4", 0.91" x 0.94"	93	3.8	0.10 - 7.0
NAVY	1" x 2"	5	9.5	8 - 26
Other	various	45	---	0.06 - 139

Twin Bridge, Water Bath Calorimeters are used for Pu-238 Calibration

Power Range (Watts)	Method of Operation	Calorimeters: ID
0 - 2	sample vs. heater	#132, #325
2 - 20	sample vs. calibrated Pu-238 standards	#58C, #326
> 20	Replacement mode vs. calibrated Pu-238 heat standards	125 Watt

Pu-238 Power Output Compared to Electric Resistance Heat



Los Alamos
NATIONAL LABORATORY

Standards & Calibration Labor
Mail Stop D478, 667-4864

Calibration Certificate

Digital Multimeter
Hewlett Packard Model 3458A
Serial No. 2823A18158

File No. 018393

Certified: September 16, 1997
Expires: December 16, 1997

Calibration of the above item(s) was achieved in a controlled environment through the use of equipr
traceable to national standards. It is expected that for the duration of the calibration interval and und
normal operational conditions, this item will remain within the tolerance limits specified.

Calibration Information

Unit Received

[✓] In tolerance
[ ] Initial test out of tolerance
[ ] Initial test near out of tolerance
[ ] Previous limitation still exist
[ ] Returned from Repair

Unit Returned

[✓] In tolerance
[ ] Limited
[ ] Adjusted
[✓] Not adjusted
[ ] Adjustment not possible

Los Alamos
NATIONAL LABORATORY

Standards & Calibration Laboratory
Mail Stop D478, 667-4864

Calibration Certificate

Ten Ohm Resistor
Fluke Model 742A-10
Serial No. 5545008

File No. 017114

Certified: October 22, 1996
Expires: April 23, 1997

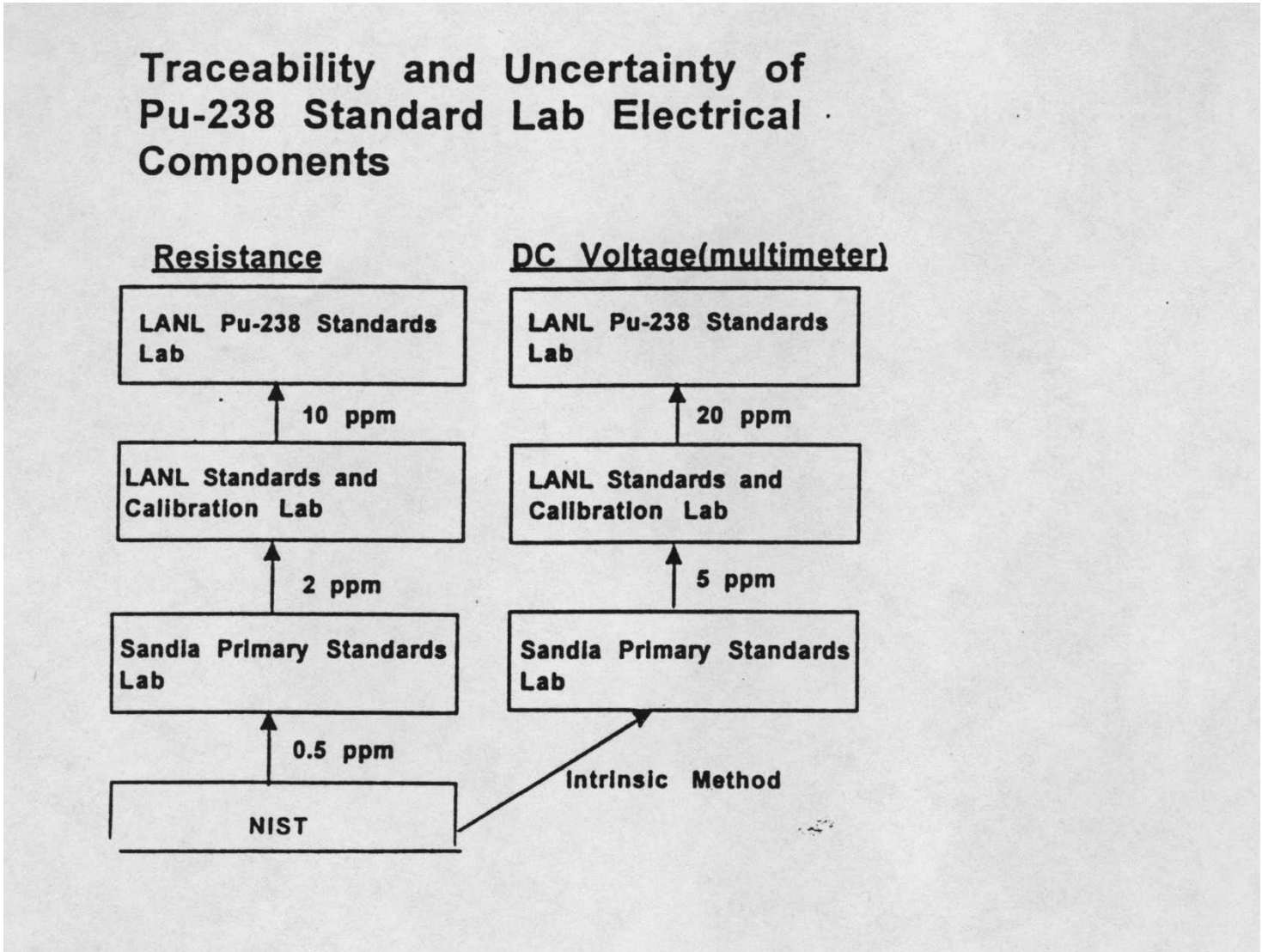
Calibration of the above item(s) was achieved in a controlled environment through the use of equipment traceable to national standards. It is expected that for the duration of the calibration interval and under normal operational conditions, this item is expected to remain within the tolerance limits specified.

Calibration Information

[✓] Received within tolerance

[✓] Returned with same tolerance

Traceability Chain for Resistance and Voltage measurement



National Institute of Standards and Technology **NVLAP®** National Voluntary Laboratory Accreditation Program

ISO/IEC GUIDE 25:1990
ANSI/NCSL Z540-1-1994
ISO 9002:1987

Scope of Accreditation

CALIBRATION LABORATORIES — NVLAP LAB CODE 105002-0

SANDIA NATIONAL LABORATORIES

NVLAP Code: 20/E05
DC Resistance

Range in ohms	*Best Uncertainty (±) in ppm*[note 1]	*Remarks*
0.0001 to 0.001	11	Low Resistance
0.001 to 0.01	4	Low Resistance
0.01 to 0.1	2.5	Low Resistance
0.1 to 1	2	Low Resistance
1	0.057	Thomas
1 to 10	1	
10 to 10^4	0.5	
10 k	0.15	SR104
10^5	2	
10^6	3	
10^7	5	
10^8	10	
10^8	240	with Teraohmeter

December 31, 2001
Effective through

David F. Alderman
For the National Institute of Standards and Technology

Calibration Period for Standards is up to 5 years, extendable to 10 years

NOTE: When measurement standards and M&TE are found to be out-of-tolerance, the as-found and-left data must be recorded.

e. Identification. Methods shall be established in order to readily identify the certification status of measurement standards and M&TE. The following is the minimum required identification information:

(1) standard or M&TE description or identification number;

(2) expiration date or criteria;

(3) indication of any special limitations of use, when applicable; and

(4) identification of the person responsible for the calibration.

f. Intervals. Calibration intervals for measurement standards and M&TE shall be established in such a manner as to minimize the occurrence of out-of-tolerance conditions. Periodic evaluations shall be performed on measurement standards and M&TE to ensure adequate calibration requirements and intervals are instituted. Calibration intervals shall be limited to a maximum of five years unless formal documentation is made to justify either longer intervals or exemption from calibration activities. Calibration intervals may be extended only after review of the calibration history for the device in question and similar devices, and review of cross-check data. Justification for not performing a cross-check must be documented. Calibration interval extensions shall not exceed one calibration interval. When measurement standards or M&TE are either removed from service, removed from periodic calibration, or placed in storage for a period of time that exceeds its calibration expiration date, and they have been used since their last calibration, a calibration or cross-check must be performed.

Calibration Runs for 1.5WF

SUMMARY

CALORIMETRY DATE 11653.5 -11/27/97
SENSITIVITY = 38817.8

RUN#	KCJ DATE	DEL BP	DEL W	W HEAT	W SAMP	W NMST	STATUS
1	11650.87	5.27	.0001385	1.2779007	1.2780391	1.2779672	ok ok ok
2	11651.29	1.10	.0000288	1.2779063	1.2779351	1.2778745	ok ok ok
3	11651.71	-.33	-.0000087	1.2779040	1.2778953	1.2778463	ok ok ok
4	11652.13	.42	.0000111	1.2779026	1.2779137	1.2778761	ok ok ok
5	11652.55	1.23	.0000323	1.2779058	1.2779381	1.2779120	ok ok ok
6	11652.96	-.34	-.0000088	1.2779256	1.2779168	1.2779021	ok ok ok
7	11653.38	1.09	.0000287	1.2779230	1.2779517	1.2779485	ok ok ok
8	11653.80	-2.03	-.0000533	1.2779123	1.2778590	1.2778672	ok ok ok
9	11654.22	-1.75	-.0000461	1.2779131	1.2778670	1.2778867	ok ok ok
10	11654.64	-.63	-.0000165	1.2779122	1.2778956	1.2779268	ok ok ok
11	11655.05	-.95	-.0000250	1.2779104	1.2778854	1.2779280	ok ok ok
12	11655.47	-1.14	-.0000299	1.2779038	1.2778738	1.2779279	ok ok ok

AVERAGE = 1.2779053
STD DEV = .0000361
OF PTS = 12.0000000

Latest Calibration Result Combined with Historical Data to get New Value

Pu-238 Heat Standard 1.5WF Historical Data + Latest Measurement

USER	Cal ID	n	date	Av power (Watts)	Power on day 4747* (Watts)
MD	90	8	1/14/75	1.528594	1.481946
MD	91	5	1/14/75	1.528579	1.481931
MD	90	5	8/11/77	1.498146	1.481962
MD	58C	13	6/17/78	1.488093	1.481823
MD	91	7	9/25/78	1.485084	1.482000
MD	58C	52	2/22/79	1.480254	1.481936
MD	91	6	9/13/80	1.462391	1.482020
MD	90	12	5/16/84	1.420946	1.482041
MD	91	11	9/7/90	1.352283	1.481975
LANL	132	12	11/21/97	1.277905	1.482164

* Day 1 starts 0000 hrs. 1/1/66

Heat Standard Power Uncertainty Calculation

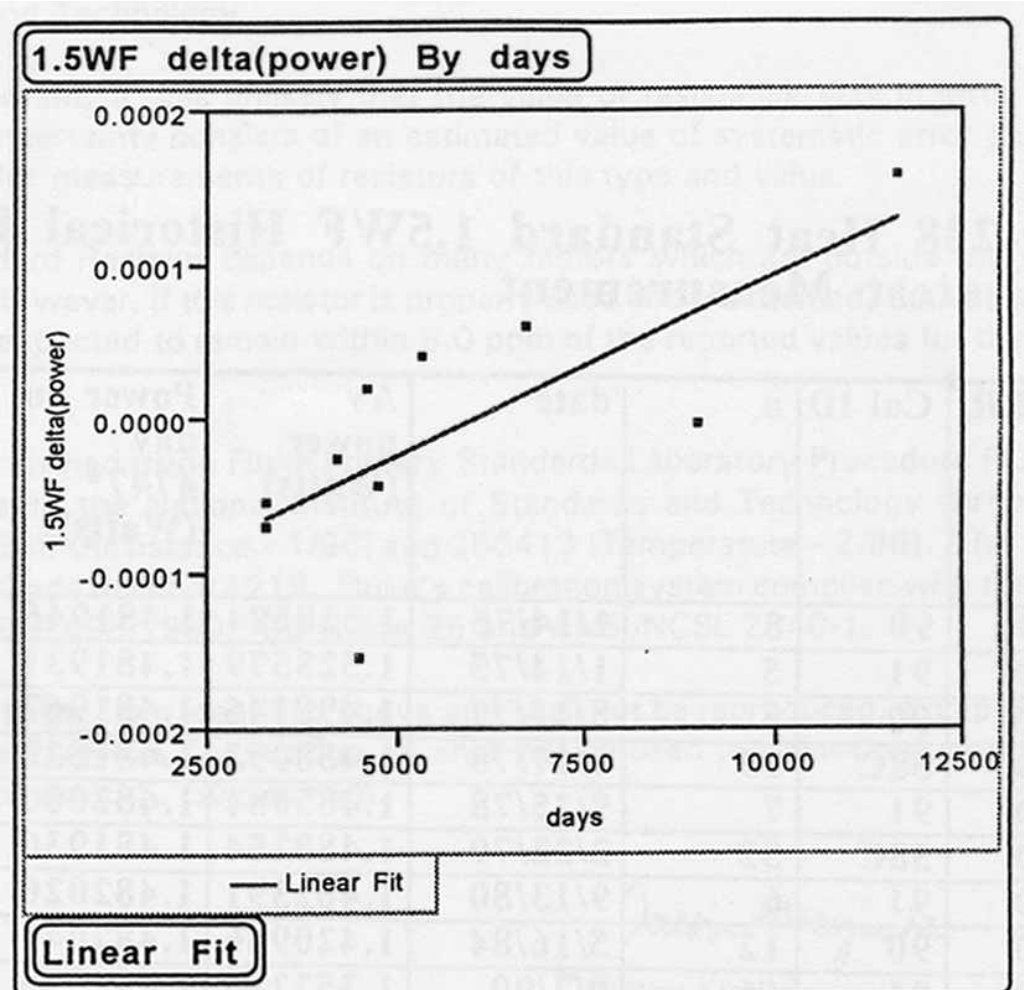
Latest set of data combined with historical data to get uncertainty

Historical Data Compared to Data Calculated from Grand Average for 1.5WF

1.5WF (delta power)

Rows	Power	days	Calculated	1.5WF delta(power
1	1.528594	3300	1.52864800	-0.00005
2	1.528579	3300	1.52864800	-0.00006
3	1.498146	4240	1.49817100	-0.00002
4	1.488098	4550	1.48825100	-0.00015
5	1.485084	4650	1.48506400	0.0000
6	1.480254	4800	1.48029700	-0.00004
7	1.462391	5369	1.46235000	0.00004
8	1.420946	6710	1.42088600	0.0000
9	1.352283	9015	1.35228500	-0.00000
10	1.277905	11653.5	1.27774700	0.00015

Plot of difference between Calculated Value and True values. The greater the variability and drift the larger the uncertainty.



Extrapolation of Least Square Fit Uncertainty + Drift is Used to Specify Heat Standard Uncertainty

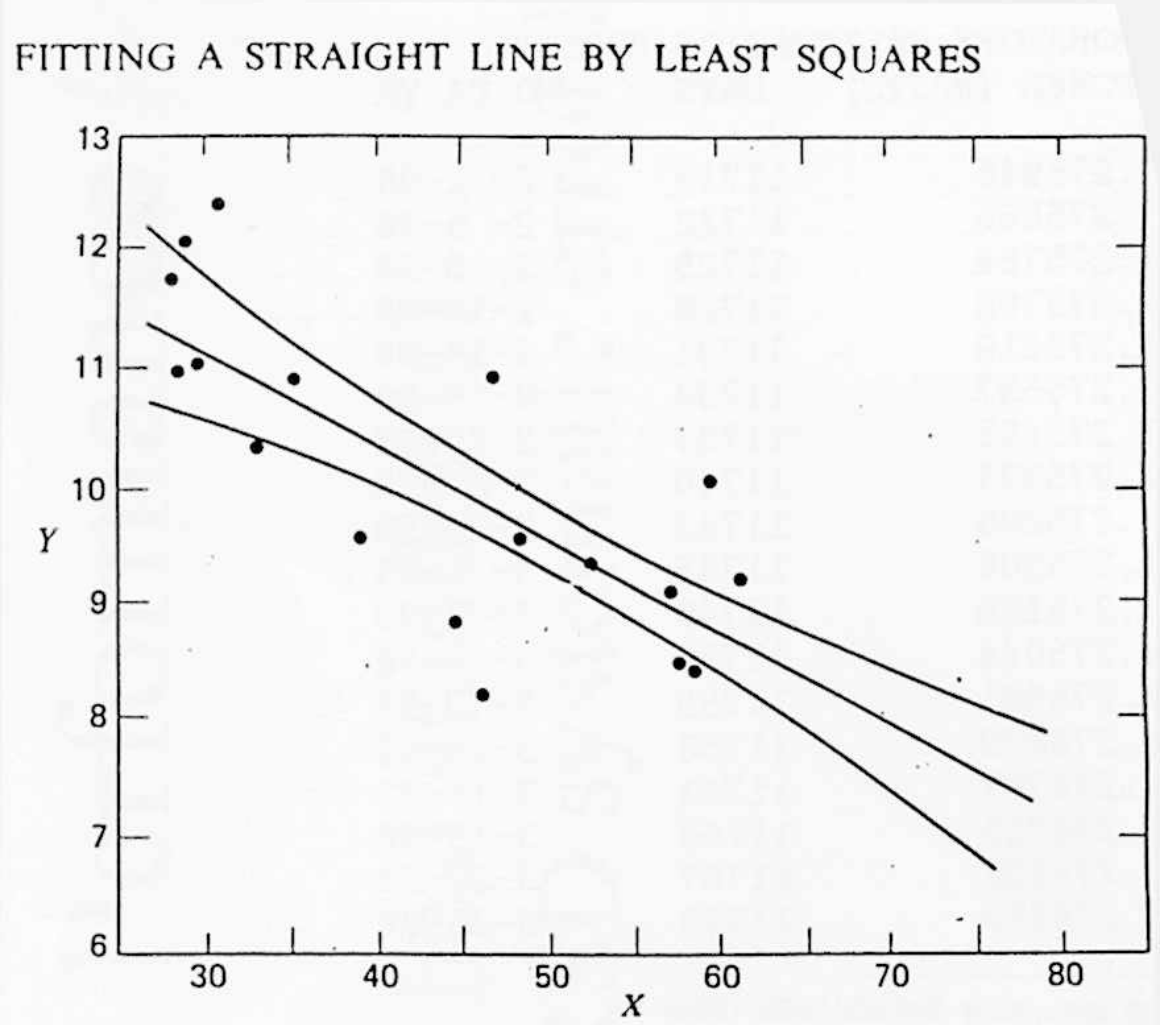


Figure 1.8 95% confidence bands for the true mean value of Y.

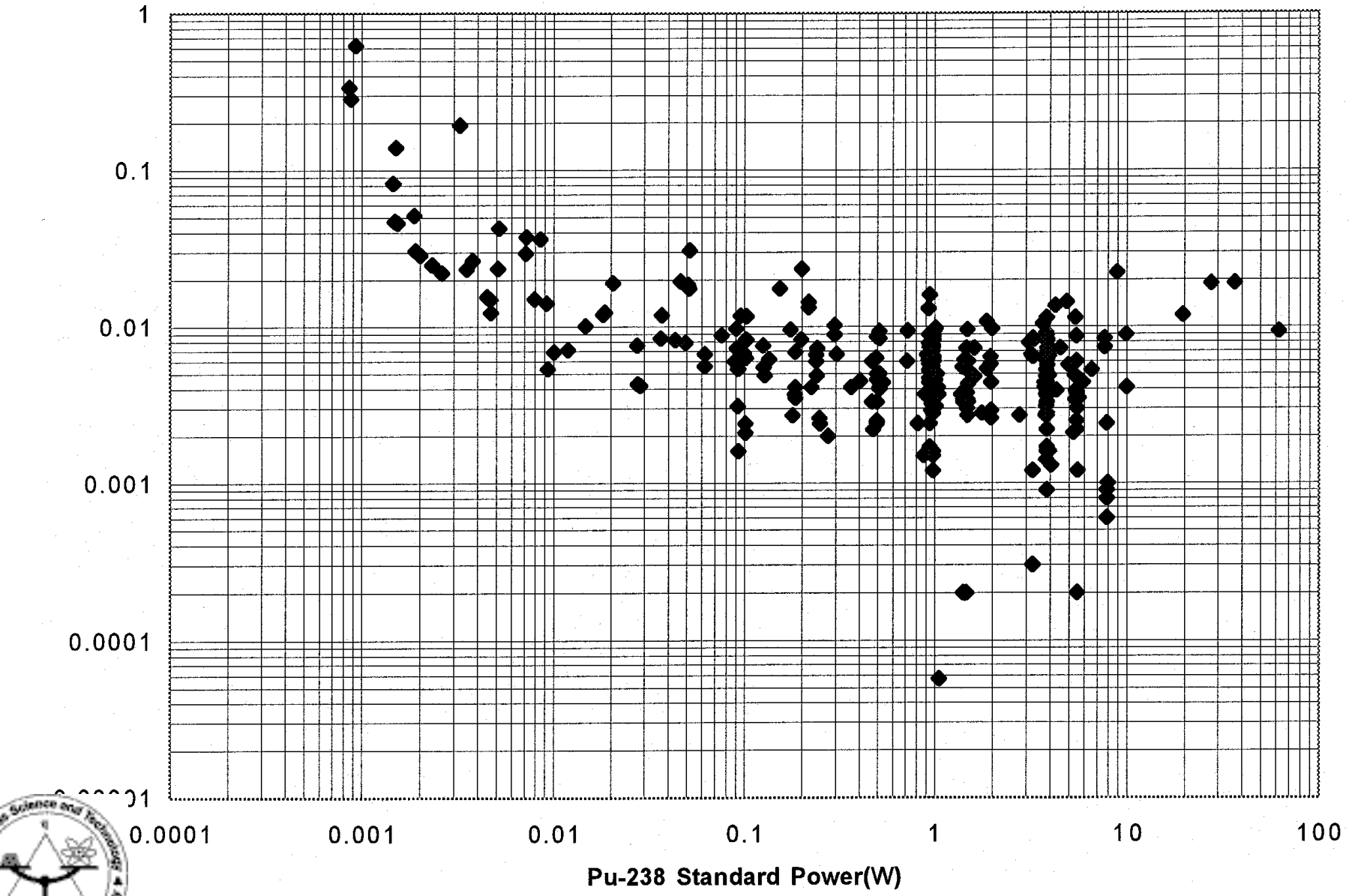
Power Uncertainty for 1.5 WF

Pu-238 Heat Standard 1.5WF Uncertainty Calculation, 5 yr calibration period

Days*	Predicted Power (Watts)	Delta + 95% CL (Watts)	(Delta + 95% CL)/ Predicted Power
11700	1.27647	0.000243	0.000190
11719	1.275948	0.000244	0.000191
12000	1.268264	0.000255	0.000201
12300	1.26011	0.000267	0.000212
12600	1.252007	0.000279	0.000222
12900	1.243957	0.000290	0.000233
13200	1.235957	0.000302	0.000245
13500	1.228009	0.000314	0.000256
13545	1.226821	0.000316	0.000258

How Well do the Replicate Standard Calibration Average Results Measured over Decades with Different Instruments/Analysts Agree?

Pu-238 Standard %RSD(1965-1996)



Based on Historical Data
Uncertainties in Pu-238 Heat Standard Values better than 0.01% RSD for Thermal power Greater Than than 0.1 Watt

NDA Standards

NDA Standards Fabrication Approaches

- Synthetic standard approaches
  - Disperse carefully measured SNM in representative matrix
  - Carefully sample/analyze representative matrix material, prepare set of standards, NBL, LANL
- Production material standard
  - Select items from material category to serve as working standards

Physical standards used in US facilities(~ 1981)

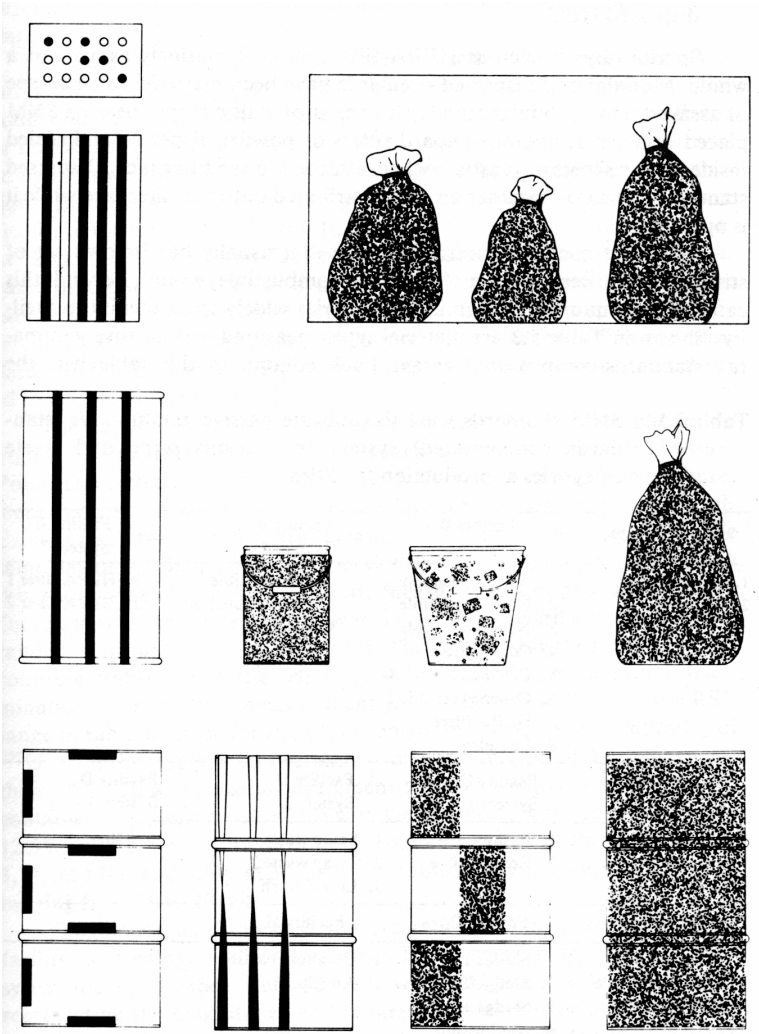


Figure 5.2 Physical standard types.

Facilities have large number of different material categories

RFETS residue categories

Magnesia Crucible		Unleached Resin
Grit		leached resin
Firebrick Fines		graphite
Unpulverized slag		firebrick
Unpulverized sand and crucible		coarse firebrick
Unpulverized sand, slag and crucible		raschig ring
SS&C Heel		MSE salt(11 categories)
Pulverized SS&C		Chloride salt
Sand		filter sludge
Ash heel		glovebox filter
Soot		grease oxide
Soot Heels		plastics
filter		peroxide cake
Oily Sludge		fluoride
Wet Combustibles		sludge

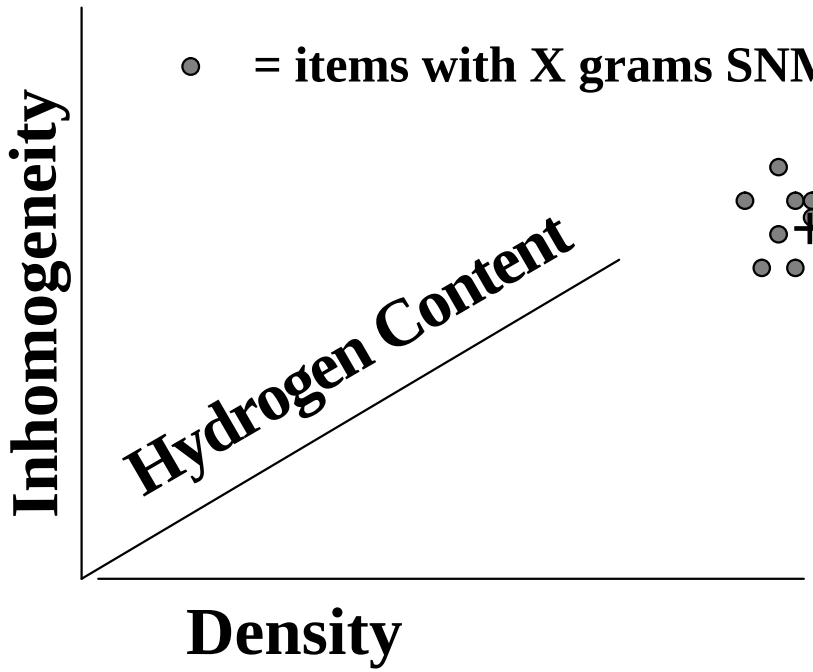
+100 MOR

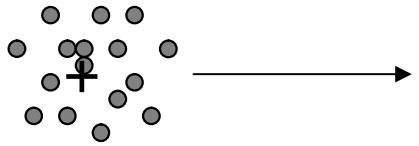
Physical standards should be representative of items being assayed

Matrix Space of particular material category

+ = standard with X grams SNM

= items with X grams SNM

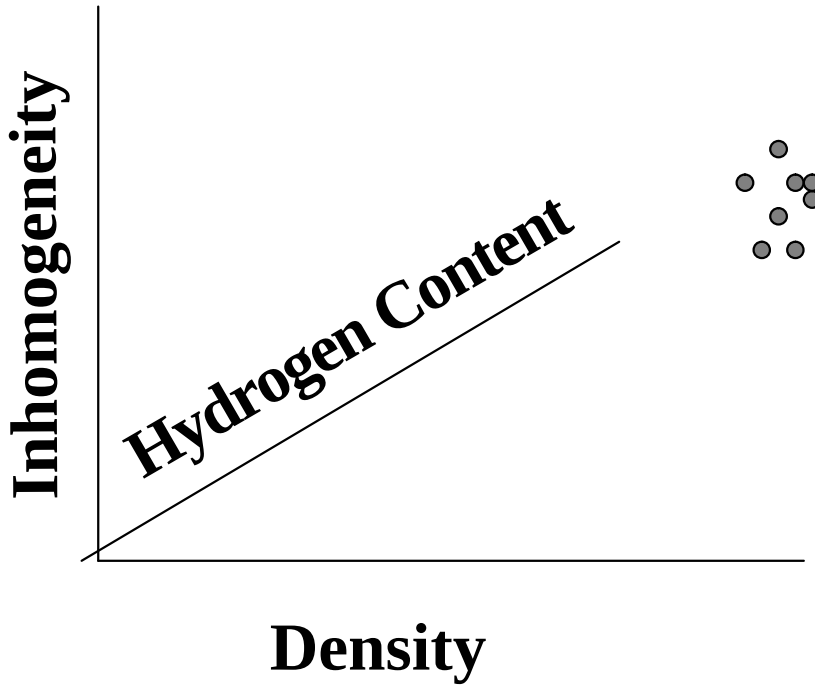


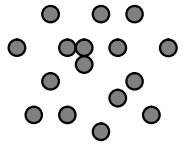


Representative

Axes are possible matrix variables that can affect the assay result

Non-representative standard is one that does not have matrix properties matching the items in that category





+ ← Non-representative

Non-representative standard can lead to biased measurement result

Representative standard

Non-representative standard

CAL/ISO uses production items as standards

- CAL/ISO technique uses its matrix independence, accuracy, and traceability
- Much less expensive, more practical, to produce than chemical standards.
- Calorimetry measurement traceable to national measurement system (NIST in US) through electrical standards
- Gamma-ray measurement based on physical constants of isotopes(half life, specific power, branching ratio).

Calorimeter sensor output at equilibrium is independent of matrix type

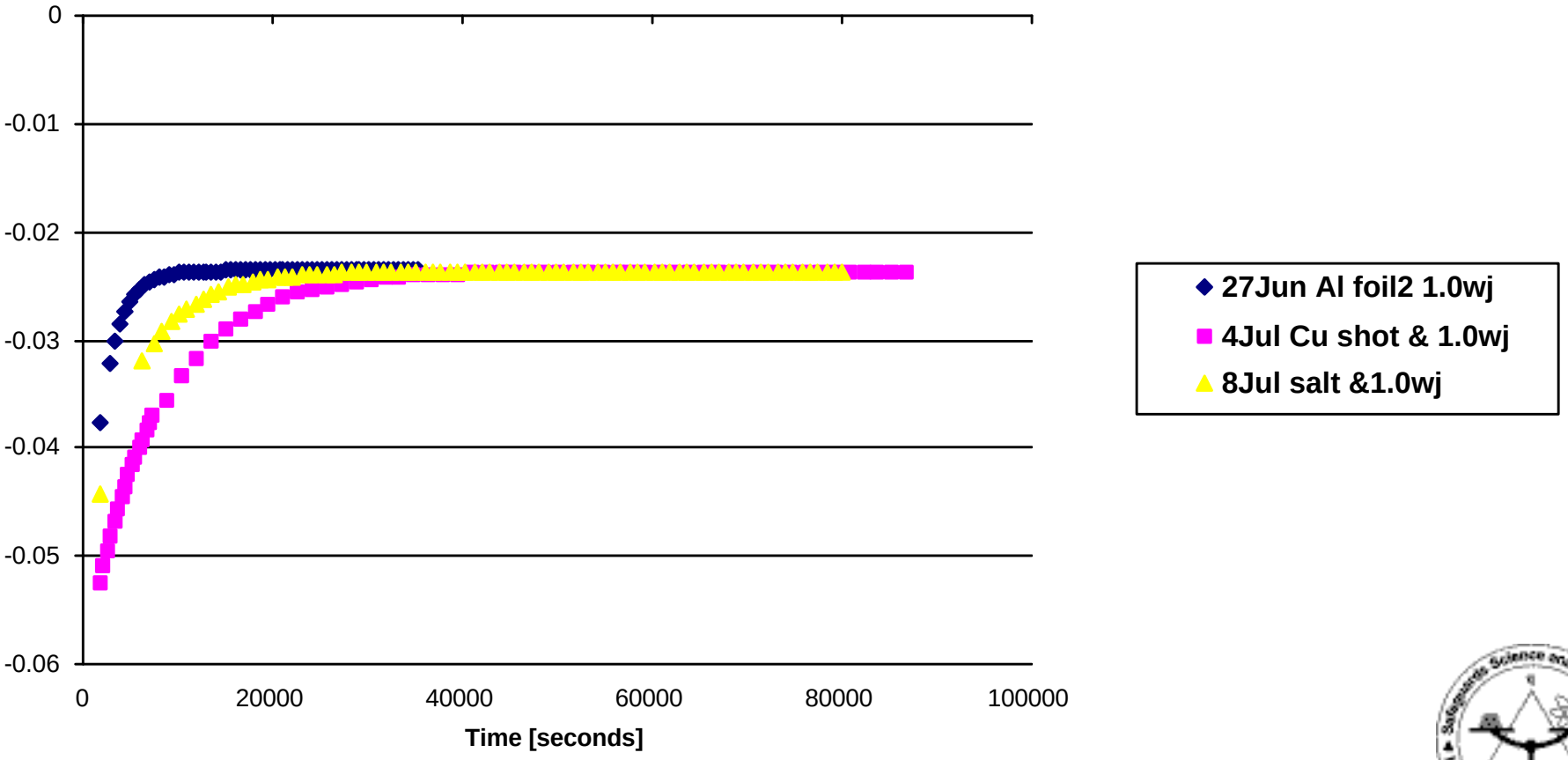
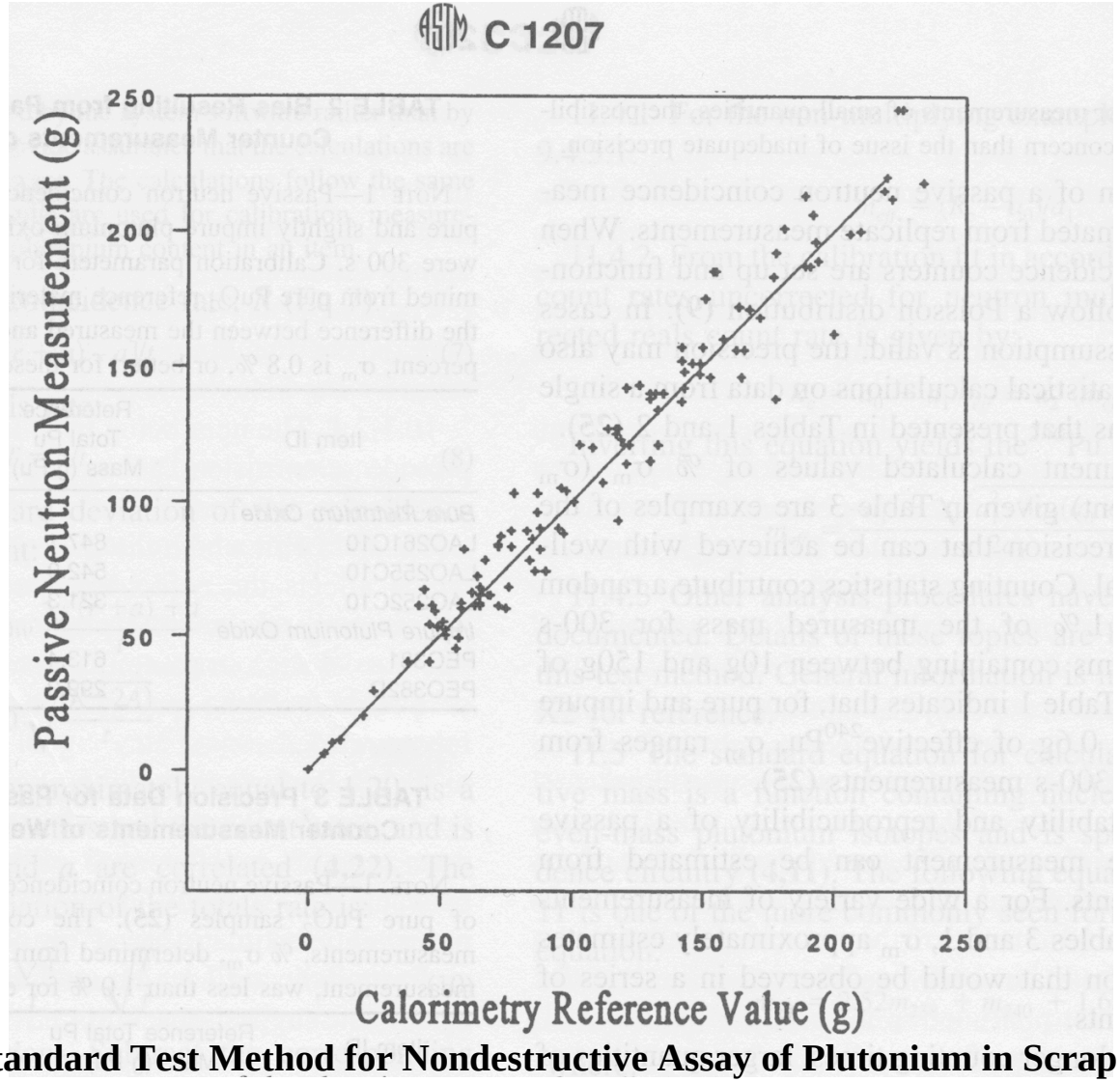


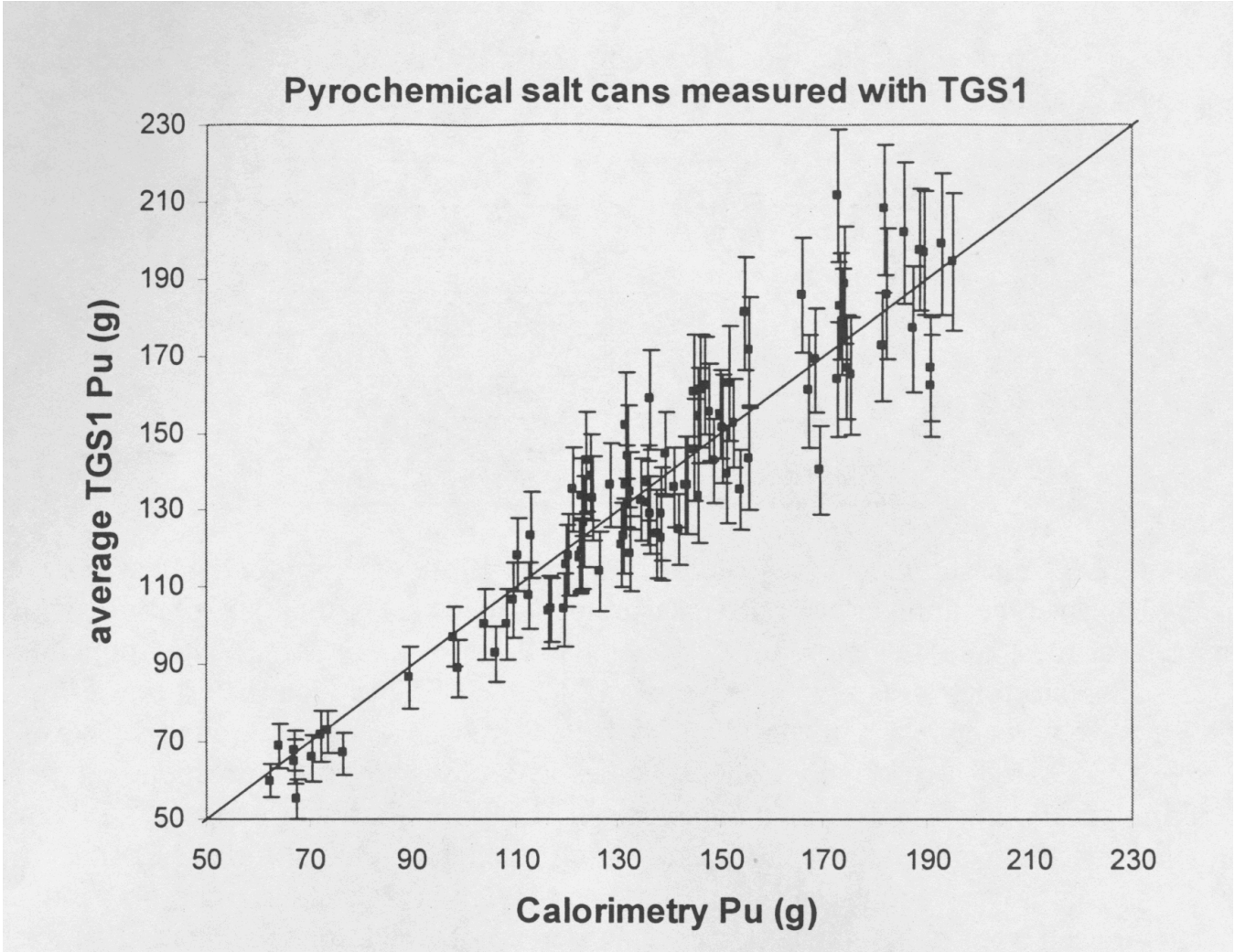
Figure courtesy of M. Smith and P. Hypes, NIS-5, LANL

Use of cal/iso for verification of neutron coincidence counting of Pu in non-hydrogenous matrices at LANL



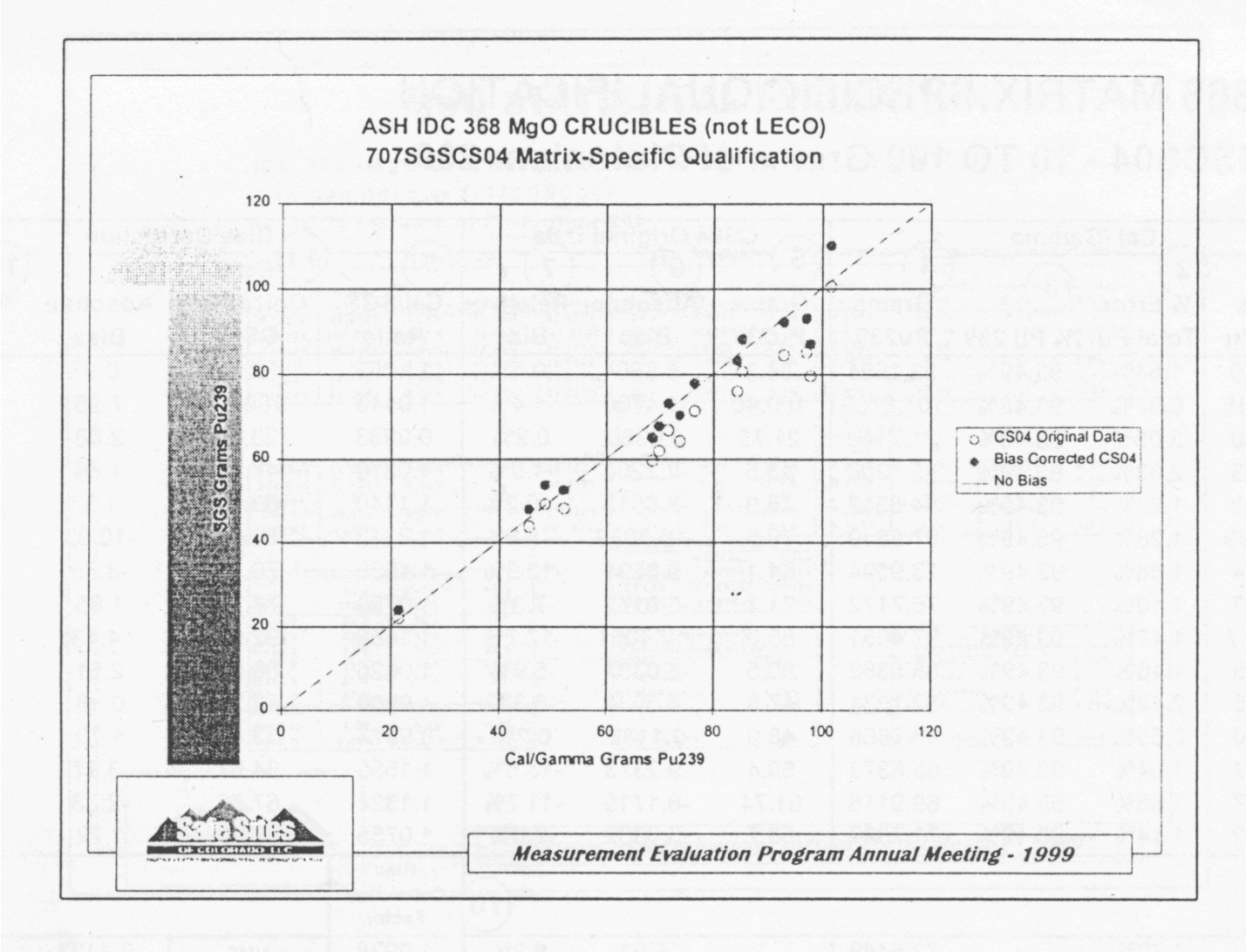
From "Standard Test Method for Nondestructive Assay of Plutonium in Scrap and Waste by Passive Neutron Coincidence Counting," C1207, Vol 12.01, 2000

Use of cal/iso for verification of tomographic assay of Pu in molten salt residues at RFETS



From, "Determination of the Total Medasaurement Uncertainty for the RFETS Skid

Use of cal/iso for bias correction of segmented gamma scan assay of Pu in crucible residues at RFETS



rom " Nuclear Materials Safeguards Matrix-Specific Qualification and Continuous Bias Correction rograms", V. Gupta, P. Hyman, and D. Sullivan NBL-356, 10/99

Verification

MOUND APPLIED TECHNOLOGIES

In-Field Calibration of a Neutron Correlation Counter via Calorimetric Assay

L.J. Satkowiak, J.A. McDaniel, and D.P. Renz

Presented to:

Second International Workshop on Calorimetric Assay
Santa Fe, New Mexico, USA
27 October 1994

This work was supported by: US DOE Office of Arms Control and Nonproliferation and the Office of Safeguards and Security.

EG&G

MOUND'S AUDIT VERIFICATION SYSTEM

Since 1972, the Safeguards R&D section at Mound has performed calorimetric assay measurements at other DOE sites in support of safeguards audits.

Purpose:

Independent verification of SNM content

Independent evaluation of facility measurement systems

Main Challenge:

Throughput

Advances in calorimetry

Addition of PNNC

EG&G

MOUND'S AUDIT VERIFICATION SYSTEM

Audit Details:

Measurements done in support of DOE safeguards audit

Currently, LANL, in past also RF and Hanford

Length of exercise - two weeks

Sample measurement time - 4 to 8 hours

Number of samples - 15 to 31

Sample set usually contains 1 or 2 material categories

Categories defined by process, batch, waste stream, etc.

Need to measure enough samples such that a defensible conclusion concerning the category can be reached

EG&G

MOUND'S AUDIT VERIFICATION SYSTEM

Increasing calorimetric assay throughput:

Optimize sample chamber for sample size

Maximize heat transfer, use of sleeves, sample packaging, etc.

Implementation of servo-control

Sample pre-conditioning

Still unable to measure all the samples!!!!

EG&G

MOUND'S AUDIT VERIFICATION SYSTEM

Increasing throughput via PNCC:

In the mid-1980's neutron correlation counting was added as a third measurement technique

By combining the accuracy, precision, and matrix insensitivity of calorimetric assay with the speed of neutron correlation counting, a synergistic union was formed.

EG&G

MOUND'S AUDIT VERIFICATION SYSTEM

Measurement/calibration overview:

For a given material category a PNCC calibration is performed
This consists of:

- Calorimetric assay on a subset of samples (gamma & cal)
  (This subset should span the range of Pu content)
- Neutron correlation measurements made on subset samples
- A calibration curve for the category is constructed
- Gamma & neutron measurements made on rest of samples

This is not how it usually happens!!!

EG&G

MOUND'S AUDIT VERIFICATION SYSTEM

Measurement/calibration overview (cont):

The gamma-ray and neutron measurements results are monitored for:

Unusual isotopics

May indicate item not in 'category'

Large variations in R/T ratio

May be due to matrix effects, material type, etc.

May indicate item not in 'category', calibration not valid

These items should be calorimetered!!!

Selections from

VERIFICATION REPORT

LOS ALAMOS NATIONAL LABORATORY

MAY, 1992

EG&G MOUND APPLIED TECHNOLOGIES

P.O. BOX 3000 MIAMISBURG, OHIO 45343-3000 513-865-4020

operated for the UNITED STATES DEPARTMENT OF ENERGY

Contract No. DE-AC04-88-DP43495

VERIFICATION REPORT

CALORIMETRIC AND NEUTRON ASSAY OF SELECTED SAMPLES FROM SPECIAL NUCLEAR MATERIAL CATEGORIES XBLC AND XBSOX AT LOS ALAMOS NATIONAL LABORATORY, 1992

Twenty-nine samples from two categories of special nuclear material were measured with the Mound transportable large-volume calorimeters (LV1 and LV2), Verification Gamma-ray Assay System (VGAS), and High-Level Neutron Coincidence Counter (HLNCC) during the period 11 to 29 May 1992. The categories represented were XBLC (crucible pieces with plutonium metal droplets in a salt matrix), and XBSOX (plutonium-rich chloride salt). Seven XBLC items and eight XBSOX items were assayed by calorimetry, gamma-ray spectroscopy, and neutron coincidence counting. Nine additional XBLC items and four XBSOX samples were measured with the HLNCC and gamma-ray isotopic systems. One XBSOX item was assayed by calorimetry and gamma-ray spectroscopy, but was too large in diameter to fit inside the HLNCC.

As a measurement-control check of the assay systems' performance, the calorimetry exchange sample STDCALEX1 was measured before data from the verification samples were collected. STDCALEX1 was measured in both calorimeters. The results of these measurements are summarized in Table 1.

The results of the calorimetric assay measurements of the XBLC samples are shown in Table 2, and those for the XBSOX items are given in Table 3. The uncertainties given in the tables are at the one-sigma level. The Los Alamos measurement codes, which indicate the assay method used to assign the inventory value for each item, are also listed in the tables. A description of these codes is given in Table 4.

The differences, in percent, between the Mound calorimetric assay values and the Los Alamos inventory values are plotted in Figure 1 for the XBLC samples, and in Figure 2 for the XBSOX samples. The uncertainties depicted in the figures are at the one-sigma level, and include the uncertainties in the inventory values. The inventory-value relative uncertainties quoted by LANL are 4.95% for the N01 items and 2.83% for the KFnn items. While the N01 relative uncertainty seems reasonable for this measurement method, the KFnn value is greater than twice the relative uncertainty typically expected from calorimetric assay. However, these relative uncertainties were reportedly determined from the actual standard deviations observed in repeated measurements of standards with these systems. On average, the values measured by Mound are 3.1% less than the inventory values for the XBLC items, and 0.6% greater than the inventory values for the XBSOX items. None of the observed differences is statistically significant at the 95% confidence level.

The gamma-ray measurement live times ranged from 1 to 12 hours. The neutron-coincidence rates were determined by averaging three 1000-second counts of each item. All thermal powers reported are equilibrated values.

Attached to this report are summary sheets for each calorimetric assay measurement. These summaries list the measured thermal power of the sample, its uncertainty, the measured isotopic composition in atom ratios on the date of the gamma-ray measurement and in mass ratios on the date of the calorimetry measurement, the effective specific power of the sample, its uncertainty, and the plutonium content of the sample and its uncertainty. The LANL stream-average value was used for the $^{242}Pu/^{239}Pu$ ratio.

Corrections were made to the thermal-power data to compensate for drift in the calorimeter baselines. These corrections were determined for each calorimeter from a linear regression of the baseline data acquired during the LANL measurement period versus time. Uncertainties in the thermal-power values were estimated by combining in quadrature the uncertainties due to random error, systematic error, and a component related to the baseline drift. The random and systematic error components were determined from measurements made at Mound during March and April, 1992, on ^{238}Pu heat standards. The drift component of the error for each calorimeter is the standard error of the baseline estimate from the linear regressions.

TABLE 1: RESULTS OF CALORIMETRIC ASSAY OF THE CALORIMETRY EXCHANGE SAMPLE

Calo-rimeter	Date of Assay	Measured Pu (g)	Uncertainty (%)	Accepted Value (g)	Difference (%)
LV1	05/19/92	403	0.46	398	1.26
LV2	05/20/92	397	0.51	398	-0.25

Difference (%) = 100*(Measured - Accepted)/Accepted

TABLE 2: RESULTS OF CALORIMETRIC ASSAY OF XBLC SAMPLES

Sample ID	Date of Assay	Cal. Used	Measured Pu (g)	Uncertainty (%)	Inventory Value (g)	Difference (%)	LANL Code
XBLC2322	05/21/92	LV1	214	0.80	215	-0.47	KF65
XBLC6312	05/21/92	LV1	177	0.65	183	-3.39	N01
XBLC7334	05/21/92	LV2	192	0.74	207	-7.81	N01
XBLC8355	05/19/92	LV2	342	0.49	347	-1.46	KF45
XBLC9367	05/21/92	LV1	149	0.77	147	1.34	N01
XBLC9386	05/21/92	LV2	164	0.93	172	-4.88	N01
XBLC9390	05/20/92	LV1	199	0.61	204	-2.51	N01

Difference (%) = 100*(Measured - Inventory)/Measured

TABLE 3: RESULTS OF CALORIMETRIC ASSAY OF XBSOX SAMPLES

Sample ID	Date of Assay	Cal. Used	Measured Pu (g)	Uncertainty (%)	Inventory Value (g)	Difference (%)	LANL Code
XBSOX11	05/22/92	LV2	843	0.62	845	-0.24	KF25
XBSOX63	05/28/92	LV1	837	0.37	827	1.19	KF25
XBSOX98	05/27/92	LV2	846	0.61	849	-0.35	KF45
XBSOX99	05/27/92	LV1	846	0.67	841	0.59	KF15
XBSOX103	05/27/92	LV1	866	0.67	863	0.35	KF15
XBSOX114	05/29/92	LV2	736	0.75	732	0.54	KF25
XBSOX117	05/28/92	LV2	825	0.72	829	-0.48	KF45
XBSOX141	05/29/92	LV1	599	0.82	598	0.17	KF36
XBSOX147	05/27/92	LV2	540	0.44	525	2.78	KF27

Difference (%) = 100*(Measured - Inventory)/Measured

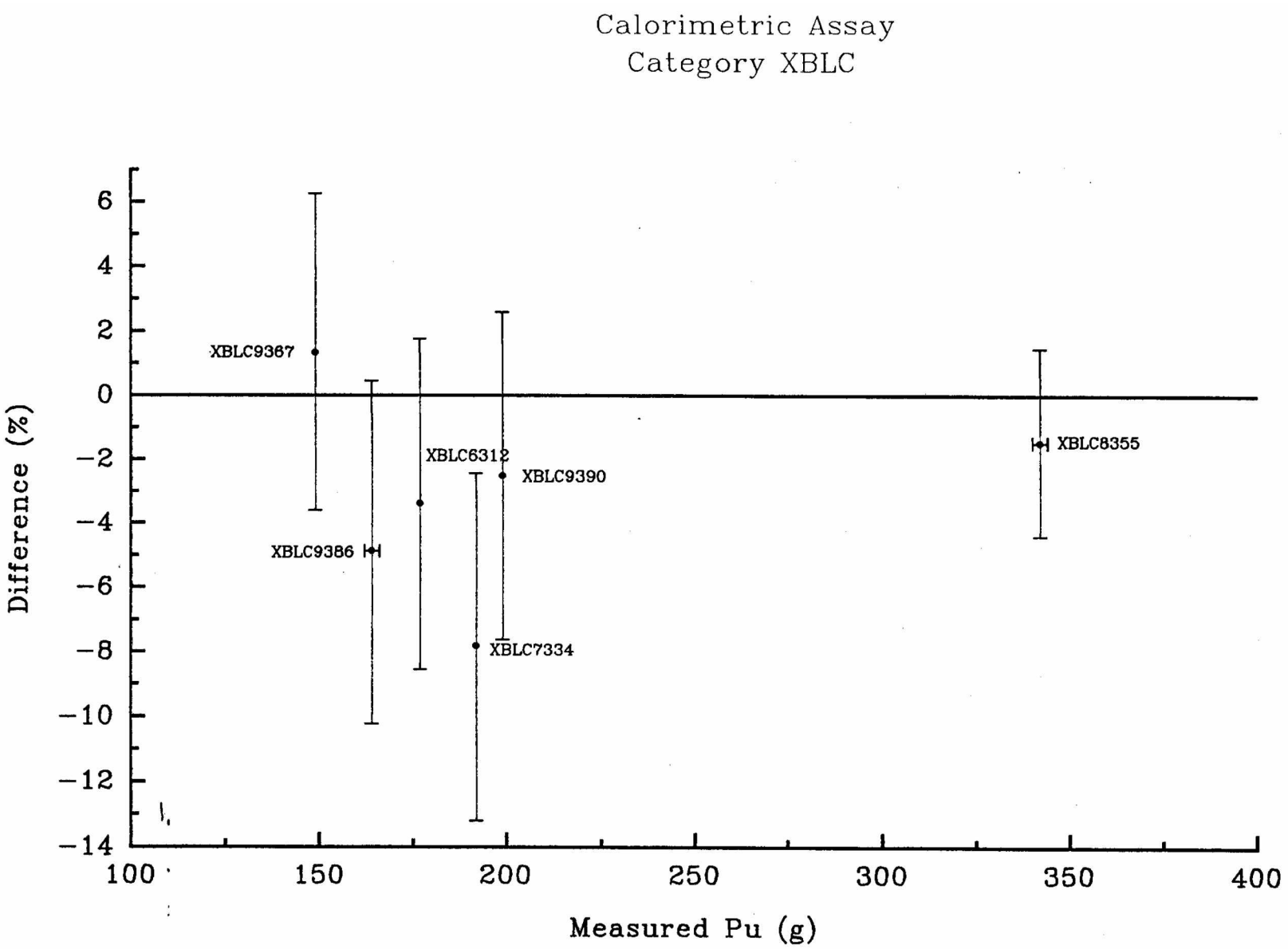


Figure 1. The per-cent difference between the inventory and measured values for the XBLC samples plotted as a function of the measured plutonium content of the sample. The data are taken from Table 2. The uncertainties are shown at the one-sigma level.

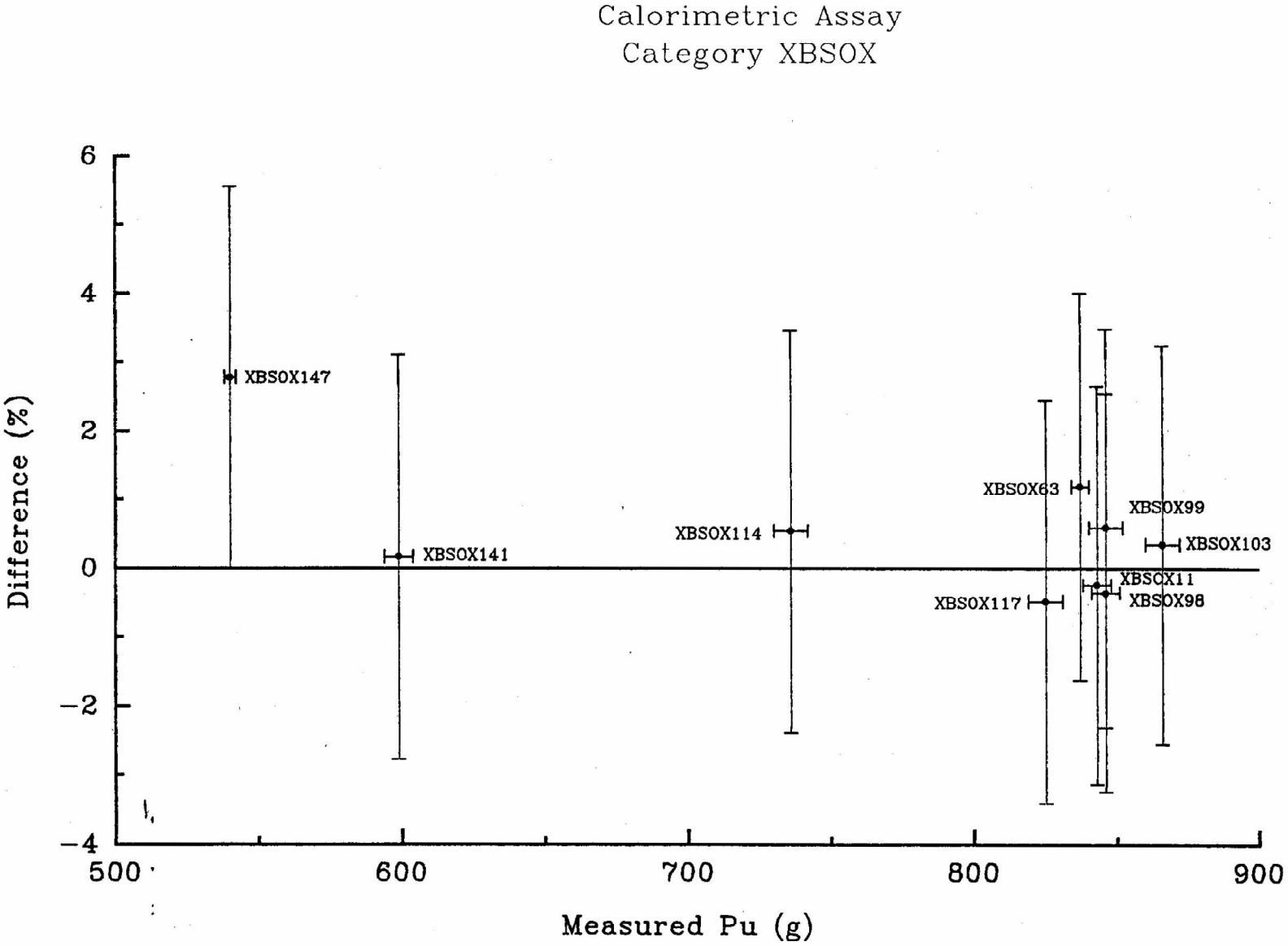


Figure 2. The per-cent difference between the inventory and measured values for the XBSOX samples plotted as a function of the measured plutonium content of the sample. The data are taken from Table 3. The uncertainties are shown at the one-sigma level.

HLNCC MEASUREMENTS OF XBLC AND XBSOX SAMPLES

High Level Neutron Coincidence Counter (HLNCC) and gamma-ray isotopic measurements were obatined for all sixteen of the XBLC items and for twelve of the XBSOX samples selected for verification. One XBSOX item, XBSOX11, was not measured because it was too large in diameter to fit in the HLNCC.

When the average real coincidence rates (counts/second) observed for six of the XBLC samples measured by calorimetric assay are plotted versus their total plutonium masses, the results depicted in Figure 3 are obtained. Item XBLC2322 was excluded from this data set because its 15% ^{240}Pu content, and resultant material type 56, rendered it unrepresentative of the other XBLC items.

Similarly, when four of the XBSOX items measured by calorimetric assay are plotted versus their effective ^{240}Pu masses, the results depicted in Figure 4 are obtained. The greater variability of both the ^{240}Pu and ^{238}Pu compositions of the XBSOX items, compared to the XBLC samples, resulted in a poor fit of the observed neutron-coincidence rates to the total plutonium masses. Hence, the effective ^{240}Pu masses, to which the HLNCC actually responds, were used. The other four XBSOX items (XBSOX63, XBSOX98, XBSOX99, and XBSOX117) were excluded from the calibration data set because their observed neutron-coincidence rates were inconsistent with their calculated effective ^{240}Pu masses. An examination of their isotopic compositions shows that XBSOX63, XBSOX98, and XBSOX99 have significantly higher ^{238}Pu concentrations than the other XBSOX items, while XBSOX117 has a significantly lower ^{240}Pu concentration than the others. Hence, these four items were not isotopically representative of the four XBSOX items remaining to be assayed.

The solid line in each figure is the "best fit" to a linear function of the form

$$y = a*x + b$$

where x is the average real coincidence rate and y is the total plutonium mass for the XBLC items and the effective ^{240}Pu mass for the XBSOX samples. The effective ^{240}Pu mass is given by

$$\text{Effective } ^{240}\text{Pu} = (2.49*f_{238} + f_{240} + 1.57*f_{242})*\text{Pu}$$

where f_{238}, f_{240}, and f_{242} are the ^{238}Pu, ^{240}Pu, and ^{242}Pu mass fractions, respectively, and Pu is the total plutonium mass. The coefficients a and b were calculated using the York-Deming least-squares algorithm, which takes uncertainties in both dependent and independent variables into account when determining the "best fit" to the input data. The two dotted lines in each figure indicate the 95% confidence intervals for the fitted calibration lines.

The calibration lines and the calibration data are given in Tables 5 and 6. These tables show the average measured neutron-coincidence rates, the ^{238}Pu, ^{240}Pu, and ^{242}Pu mass fractions, the plutonium masses from both calorimetric and HLNCC assays, and the differences between the two assay values. The larger relative uncertainties in the HLNCC assay values for the XBSOX samples, compared with the XBLC samples, results from the fewer number of data points used (degrees of freedom) in the fit.

The results of the HLNCC assay of the nine XBLC and four XBSOX items not measured by calorimetric assay are shown in Tables 7 and 8, respectively. Only one XBSOX item, XBSOX174, shows a discrepancy with its inventory value of more than three times the uncertainty in the difference, taking the uncertainty in the inventory value into account. However, three XBLC items (XBLC2347, XBLC3355, and XBLC9376) differ by more than three times the uncertainty from their inventory values. Curiously, these three items were assigned inventory values on the basis of calorimetric assay measurements.

For completeness, there are also summary sheets for these thirteen items. The summaries list the measured neutron-coincidence count rate of the sample, its uncertainty, the measured isotopic composition in atom ratios on the date of the gamma-ray assay and in mass ratios on the date of the HLNCC assay, the effective specific power of the sample, its uncertainty, and the plutonium content of the sample and its uncertainty.

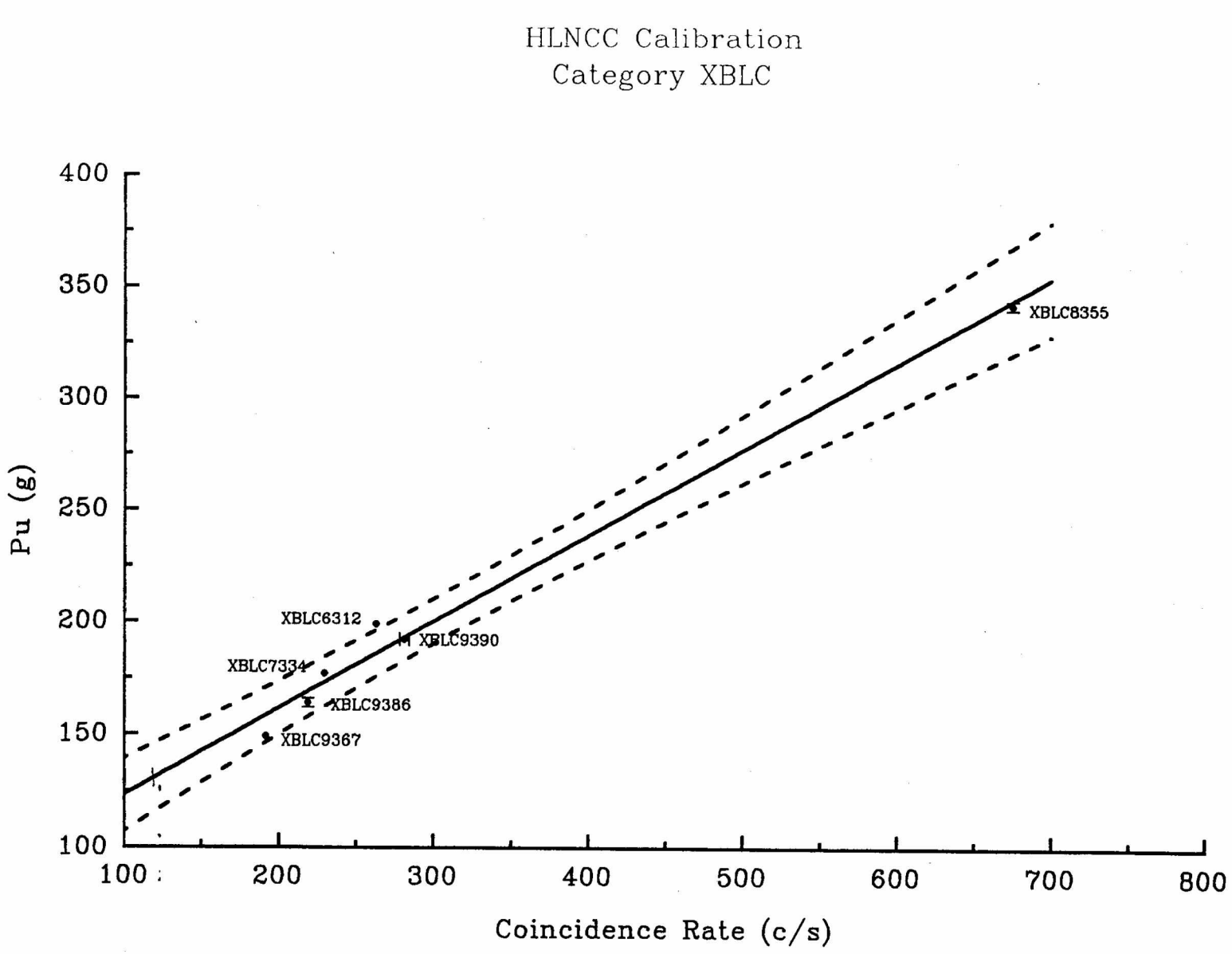


Figure 3. HLNCC calibration data for the XBLC samples. The data are taken from Table 5. The uncertainties are shown at the one-sigma level.

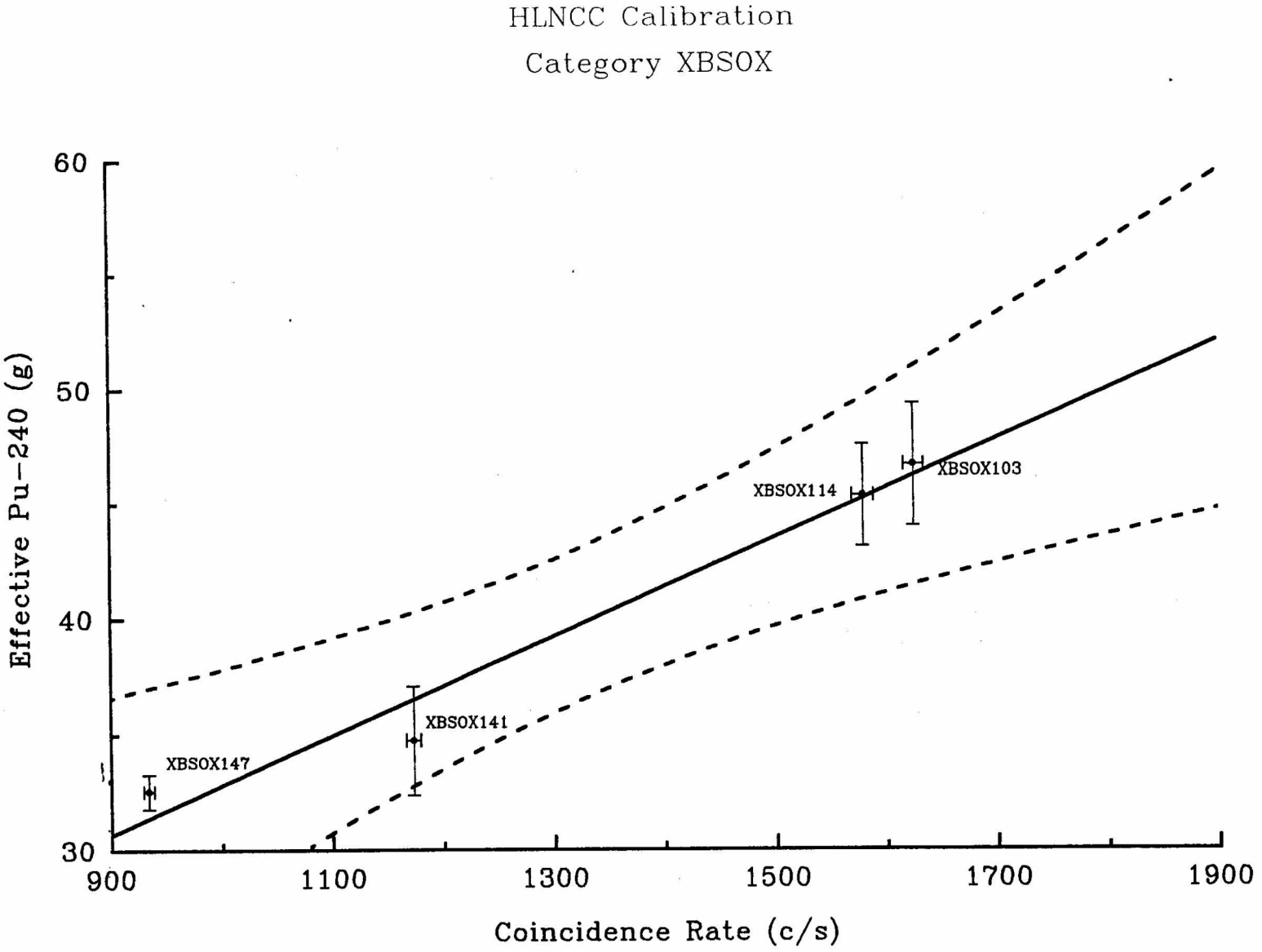


Figure 4. HLNCC calibration data for the XBSOX samples. Th data are taken from Table 6. The uncertainties are shown at the one-sigma level.

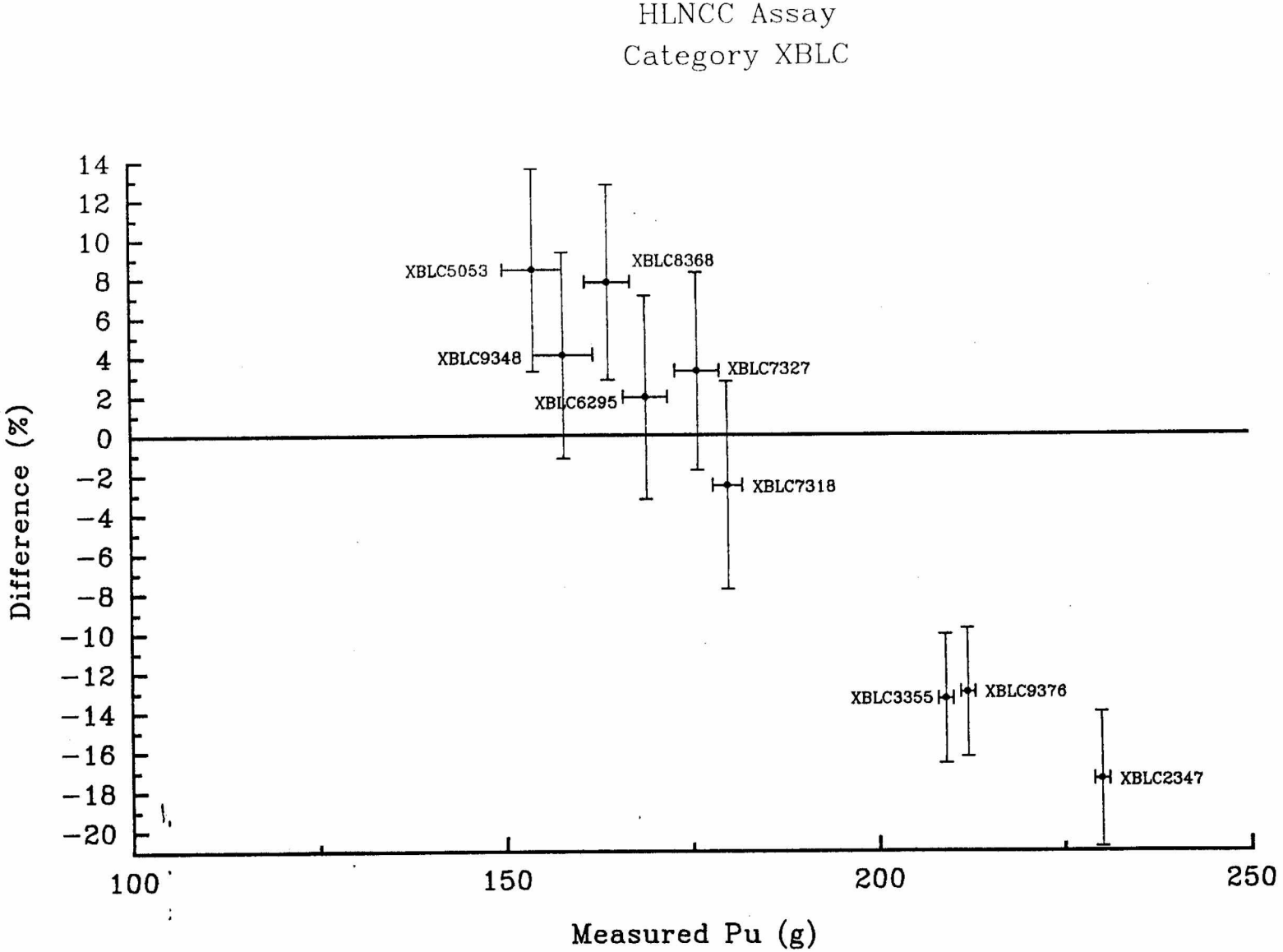


Figure 5. The per-cent difference between the inventory and measured values for the XBLC samples plotted as a function of the measured plutonium content of the sample. The data are taken from Table 7. The uncertainties are shown at the one-sigma level.

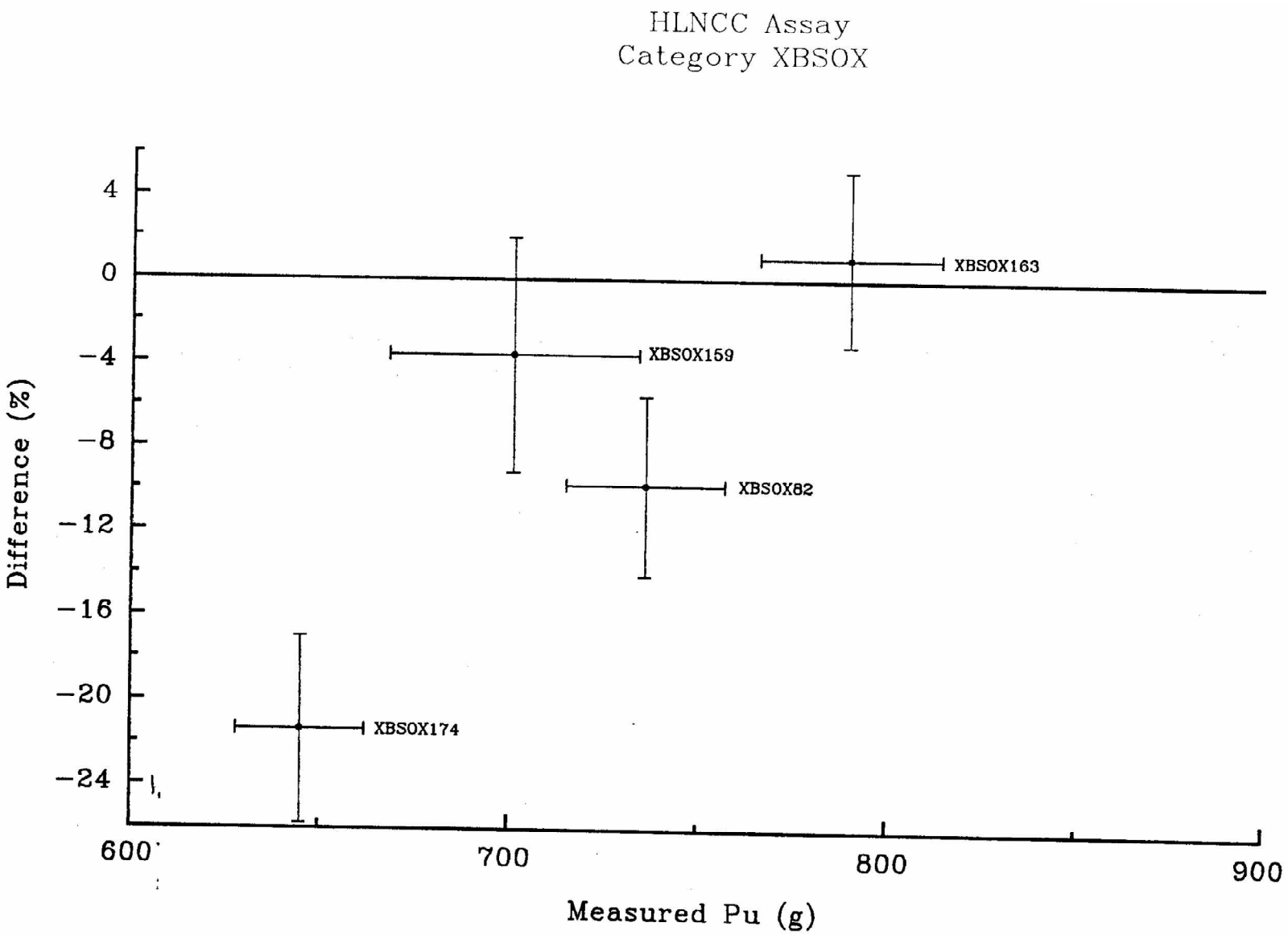


Figure 6. The per-cent difference between the inventory and measured values for the XBSOX samples plotted as a function of the measured plutonium content of the sample. The data are taken from Table 8. The uncertainties are shown at the one-sigma level.

*** CALORIMETRIC ASSAY RESULTS *** 10-JUN-92

INPUT DATA :

SAMPLE ID : XBLC6312
DATE CALORIMETERED : 21-MAY-92
DAYS SINCE ISOTOPIC MEASUREMENT : 8.
WATTS SAMPLE : 0.446000 +/- 0.590 %

ISOTOPIC COMPOSITION :

ATOM RATIOS (PPM) ON DATE OF GAMMA ASSAY

238-PU/239-PU = 138. +/- 3.41%
240-PU/239-PU = 59664. +/- 1.85%
241-PU/239-PU = 2397. +/- 0.71%
242-PU/239-PU = 475. +/- 10.00%
241-AM/239-PU = 2131. +/- 1.83%

RESULTS :

MASS RATIOS (NORMALIZED TO TOTAL PU) ON DATE OF CALORIMETRIC ASSAY

238-PU/PU = 0.000129 +/- 3.41%
239-PU/PU = 0.940781 +/- 0.10%
240-PU/PU = 0.056366 +/- 1.75%
241-PU/PU = 0.002272 +/- 0.72%
242-PU/PU = 0.000452 +/- 10.00%
241-AM/PU = 0.002024 +/- 1.83%

```
**********************************************************
*                                                        *
* SAMPLE ID: XBLC6312                                    *
*                                                        *
* WATTS/GRAM  =  0.0025261    +/-   0.27 %               *
* GRAMS OF PU =   176.558     +/-   1.15     (  0.65 %)  *
* GRAMS OF AM =  0.357345     +/-  0.639E-02 ( -1.79 %)  *
*                                                        *
* ON DATE : 21-MAY-92                                    *
*                                                        *
**********************************************************
```

Cal/Iso Assay Caveats

Situations that degrade cal/iso measurement performance

- **Gamma-ray Isotopic Assay**
- **Calorimetry**

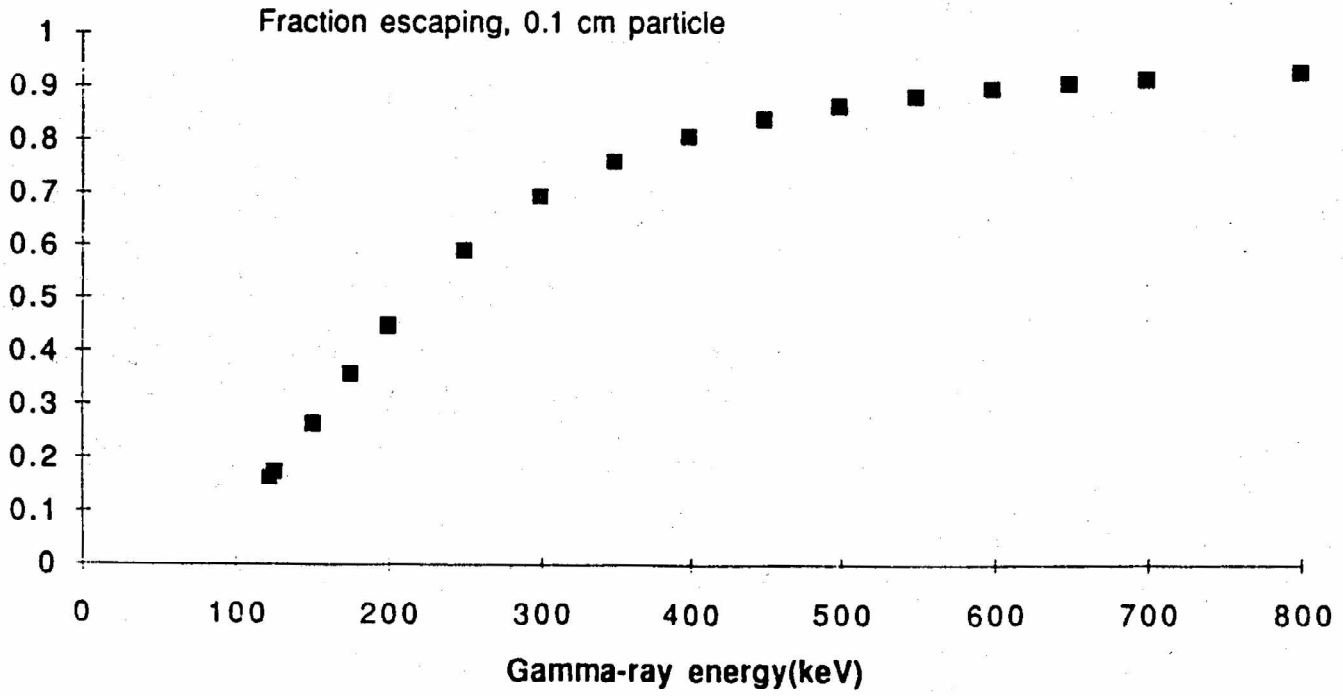
Situations that Degrade Gamma-ray Isotopic Measurement Performance

- **Gamma-ray Isotopic assay**
  - **Inhomogeneous isotopic distribution**
  - **Separated Am-241 and Pu, each in different matrices**
  - **Gamma-ray interferences**

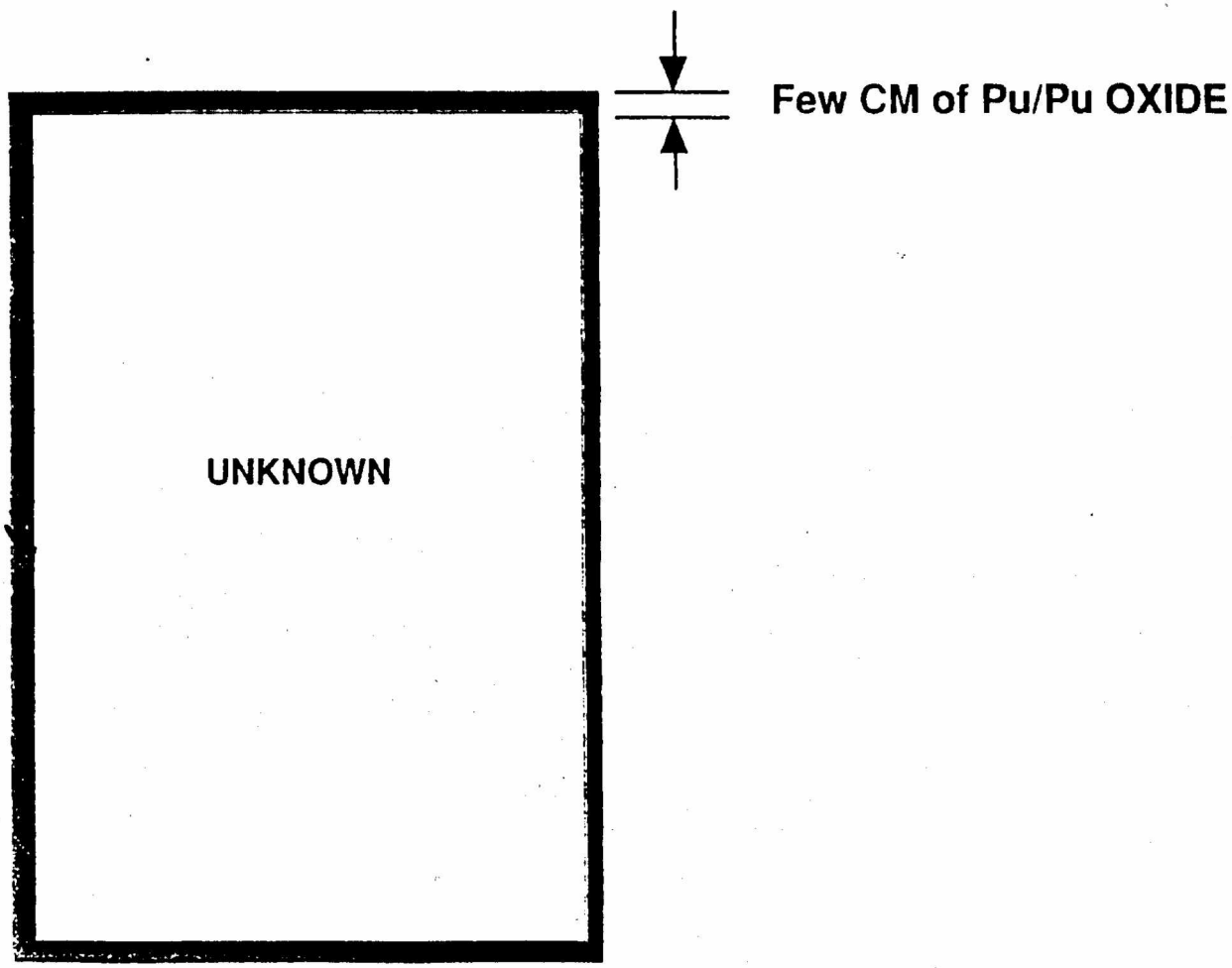
Gamma rays are strongly absorbed in plutonium

125 kev **mfp = 0.012 cm**

400 kev **mfp = 0.17 cm**



THE ISOTOPIC COMPOSITION OF PLUTONIUM AND ASSOCIATED AMERICIUM MUST BE HOMOGENEOUS



Heterogeneous Matrix Example

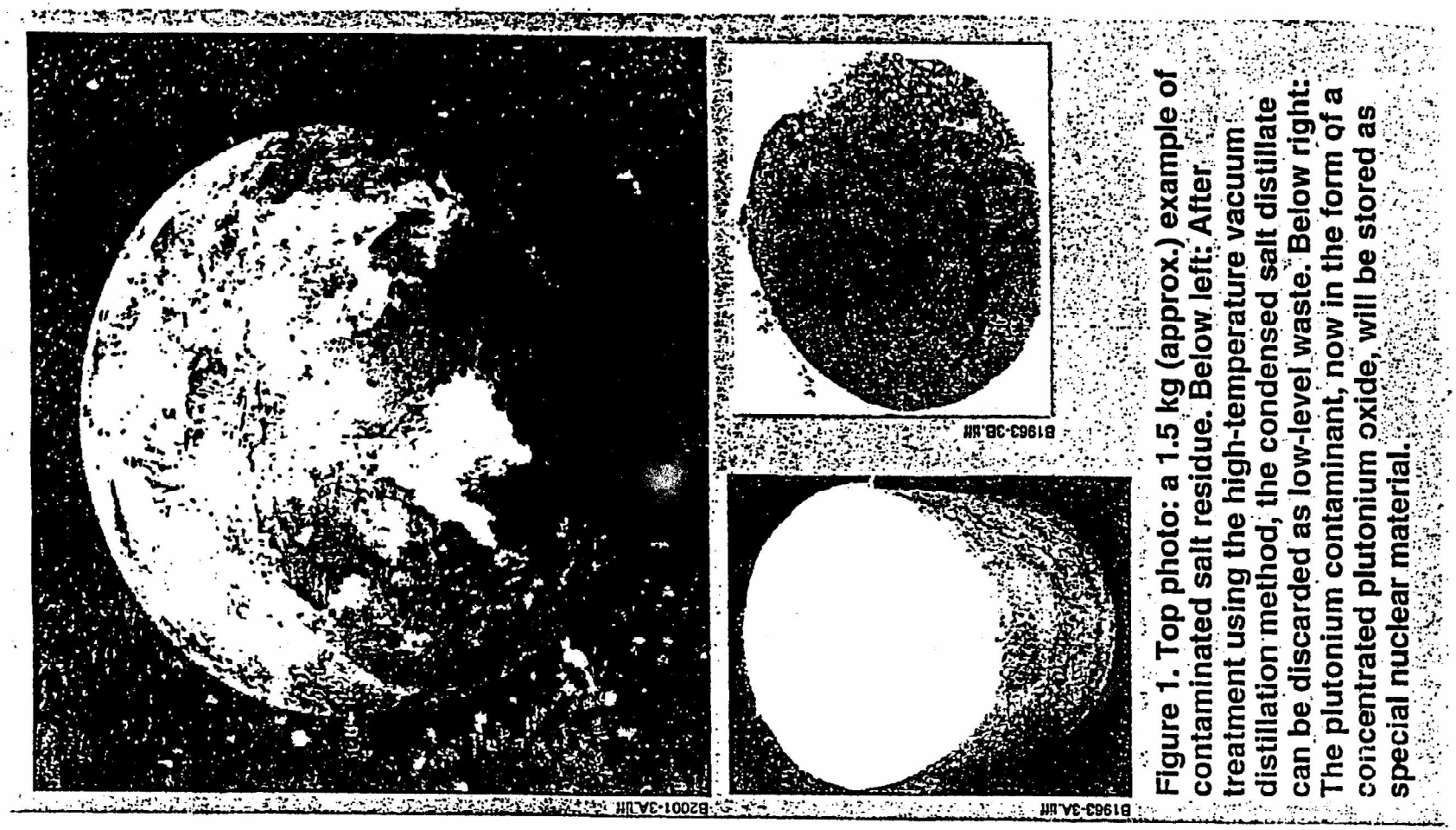
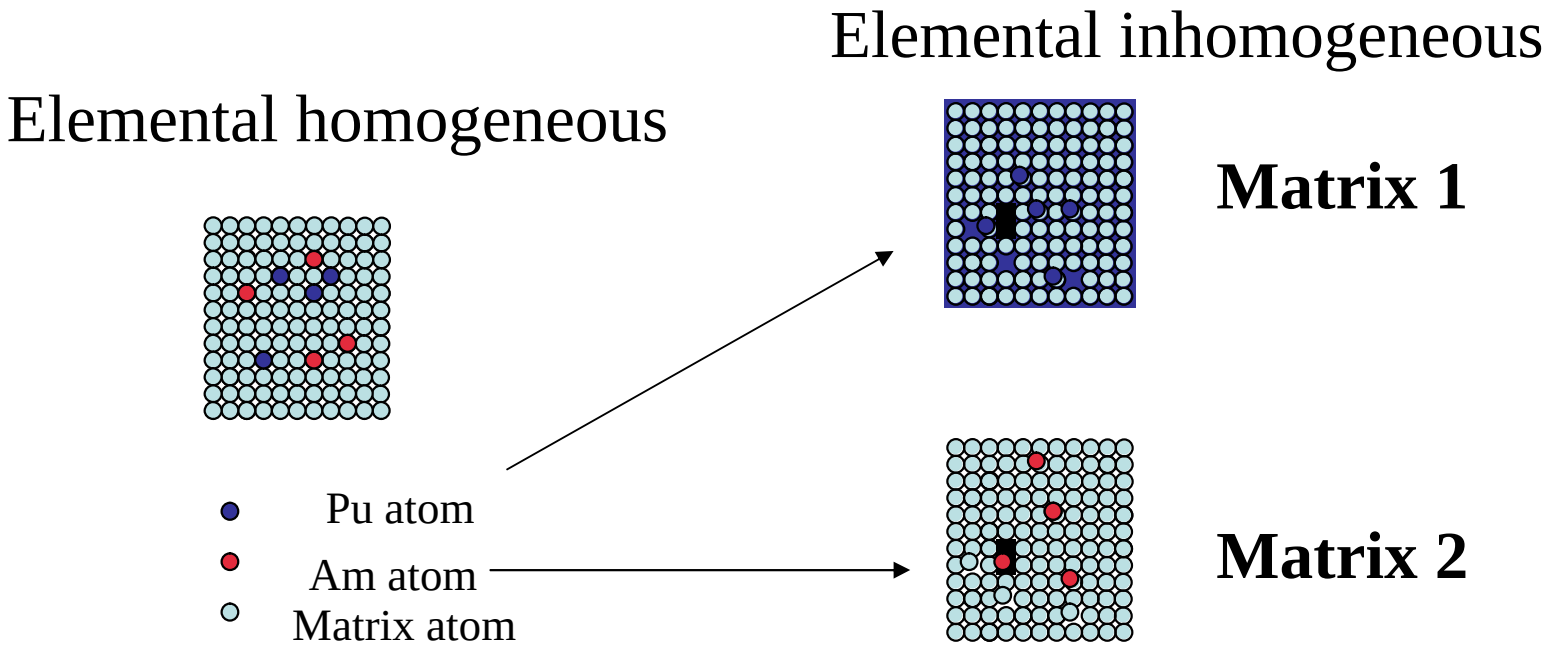


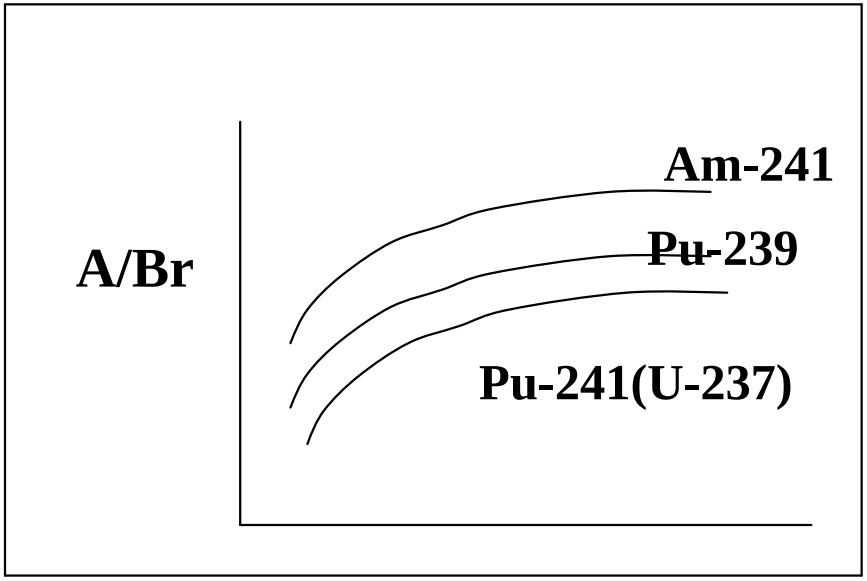
Figure 1. Top photo: a 1.5 kg (approx.) example of contaminated salt residue. Below left: After treatment using the high-temperature vacuum distillation method, the condensed salt distillate can be discarded as low-level waste. Below right: The plutonium contaminant, now in the form of a concentrated plutonium oxide, will be stored as special nuclear material.

Assumed that Am-241 gamma-rays interact with the same matrix as the Pu gamma-rays, but in certain categories chemical processing separates Am and Pu

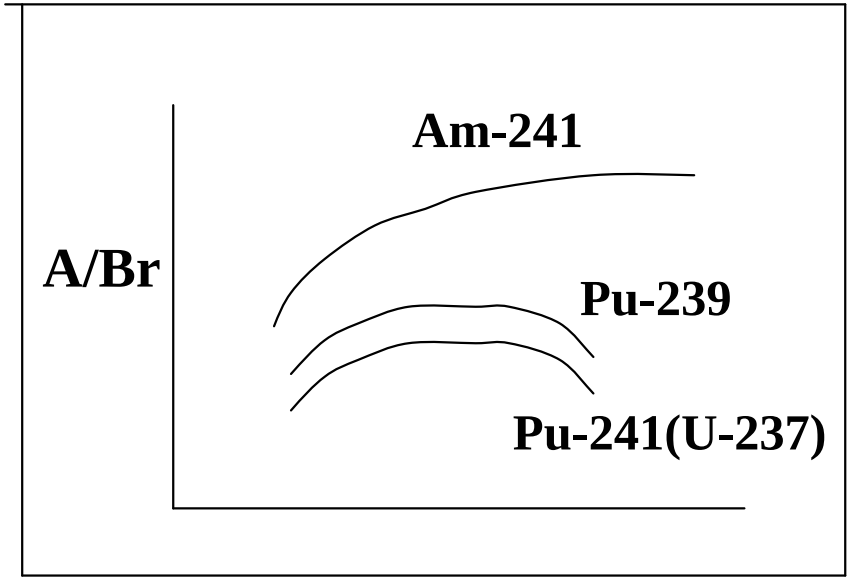


Different matrix environments for Pu and Am-241 lead to different gamma-ray relative efficiency curves

One matrix



Two matrices



Some gamma-ray, X-ray interferences

- **Fission products, Zr-95, 724.2 keV: Cs-137 661.7 keV**
- **10 uCi/g swamps 640 kev region**
- **Pa-233(from Np-237), 312.0 keV**
- **Np-239(from Am-243), 99.5, 103.7, 209.8, 334.3 keV**

Situations that degrade cal/iso measurement performance

- **<u>Calorimetry</u>**
  - **Thermal Power generating isotopes with nonmeasurable gamma ray (e.g., Pu-242)**
  - **Chemical reactions**
  - **Radiolysis of H_2O; radiolytic "heat poison"**

Chemical Reactions

- Heat from chemical reactions is indistinguishable from heat generated by radioactive decay.
- Endothermic or Exothermic reactions will bias calorimeter result,

Chemical Reaction Example

- Exothermic chemical reaction involving Pu occurring in calorimeter measurement chamber.
- Reaction generates ~30 kJ/mole of heat
- MW of Pu compound ~ 300 gms/mole
- Reaction takes place at constant rate over 1 month period, then stops

Chemical Reaction Example (continued)

Energy generation/gm Pu = 30,000 J/300 g
= 100J/g

Average Power/g ~ 100J/(2.5 x 10^{6} sec)
= 40 x 10^{-6} W/g or 40 x 10^{-3} W/kg

equivalent to ~ 17g Pu (6% Pu-240, 2.3 mW/g)
or +1.7 % bias

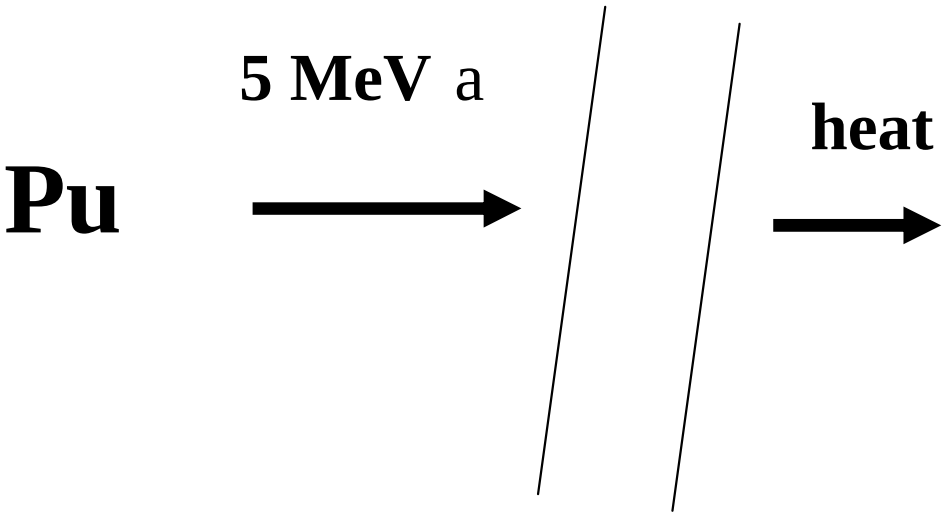
Chemical Reaction Example (continued)

Freshly-prepared compounds may have residual chemical reactions that will bias the cal assay result.

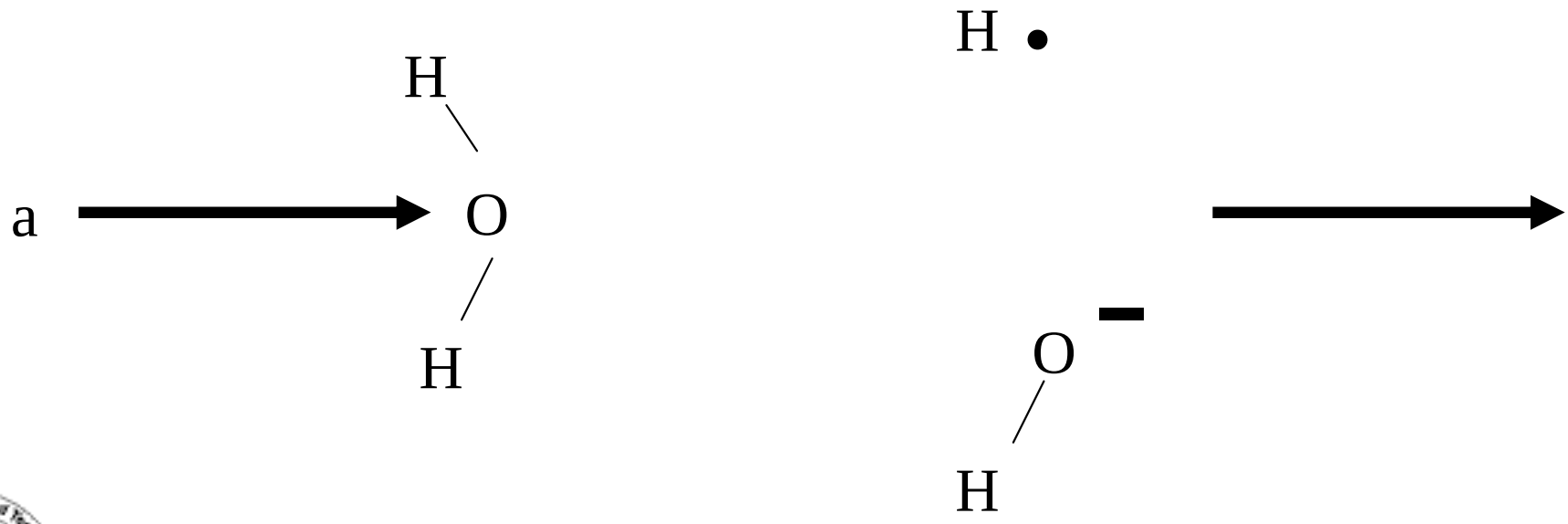
Radiolysis of H_2O

- Calorimetry of Pu solutions will be biased low.
- This is because new chemical compounds produced by radiolysis use up energy from radioactive decay.

Transformation of Radioactive Decay Energy Heat Assumed to be 100% Efficient



For Water Some of the Decay Energy Splits Molecular Bonds, Irreversibly. This Absorbs Energy



The Hydrogen Radical and OH^- Ion React to Form Compounds One of Which is H_2

A plausible reaction is

$2\ H_20 + \text{energy} \rightarrow H_2O_2 + H_2$ (1)

The Heat of Reaction(DH) for this reaction is 383 kJ/mole, an endothermic reaction

H_2 is a Common Radiolysis Product

It has been estimated that 1.6 molecules of H_2 are produced per 100 eV of energy absorbed in water.[1]

[1] R. R. Livingston, "Gas Generation Test Support for Transportation and Storage of Plutonium Residue Materials - Part 1: Rocky Flats sand, Slag, and Crucible Residues, WSRC-TR-99-00223, July 1999.

One of the Reaction Products, H_2O_2, is a Reactive Compound and Will Oxidize Another Material

- For example, assume elemental carbon is present and the H_2O_2 reacts with carbon.

H_2O_2 + C --> H_2O + CO **(2)**

D**H = - 208 kJ/mole, an exothermic reaction**

Summing up the Heat of Reactions for Reactions 1 and 2

Net DH = 175 kJ/Mole, overall endothermic reaction pair: This energy is not transformed into heat.

This is the net amount of energy needed to produce one Mole of H_2.

To produce one molecule of H_2 requires

$175\ kJ/\ N_{avogadro} = 175000/6.022 \times 10^{23} = 0.29 \times 10^{-18}$ J = 1.8 eV

Radiolysis in Water Removes a Significant Fraction of the Decay Heat

1.6 atoms of H_2 production require 1.6 x 1.8 = 2.9 eV of energy

2.9 eV/100 eV ==> 2.9% loss of energy for the assumed chemical reaction pair

Water is a ‘heat poison’ for calorimetry measurements. Calorimetry of aqueous solutions not recommended.

Conclusion

- Reviewed calibration procedure for Pu-238 Heat Standards.
- Calorimetric assay is a suitable technique for
  - Characterization of representative working standards selected from site inventories
  - Verification of nuclear material content.
- Discussed calorimetric assay limitations.

DOE Standards and Calibration Workshop

^{235}U Enrichment Measurement

T. D. Reilly

LA-UR-03-3800

Los Alamos NATIONAL LABORATORY

Contents

- ^{235}U gamma rays
- NaI, CdZnTe and Ge Detectors
- Enrichment Meter Principle
- Standards and Calibration Procedures
- Corrections and Uncertainties
- Response-Function Fitting (MGAU)

Los Alamos NATIONAL LABORATORY

^{235}U Enrichment (atom % vs. weight %)

relationship between weight % and atom %

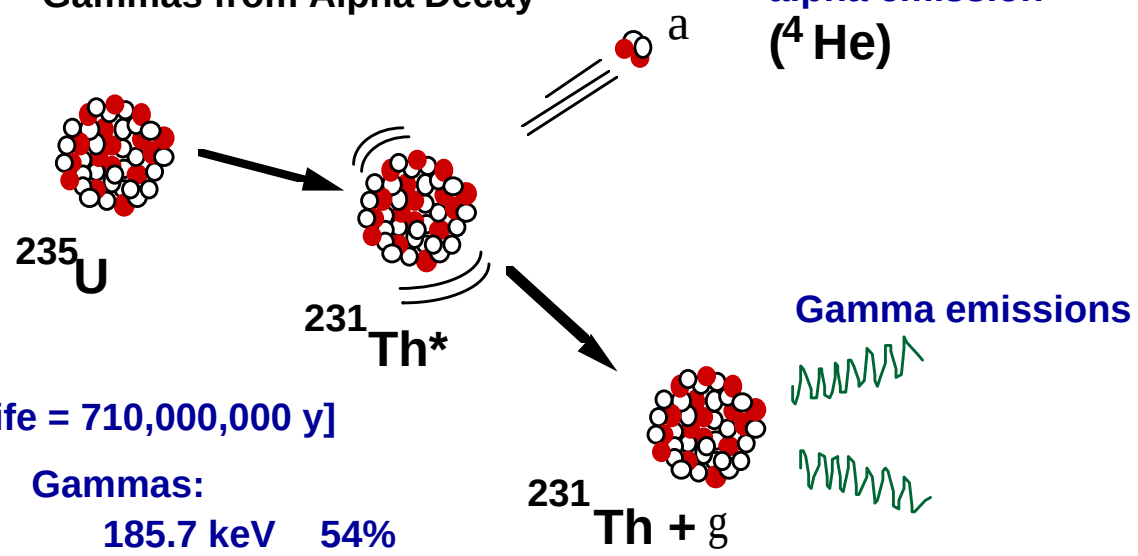
$$E_{wt\%} = \frac{235E_{at\%}}{238 - 0.03E_{at\%}} \approx \frac{235}{238} \times E_{at\%}$$

Los Alamos NATIONAL LABORATORY

^{235}U Decay

GAMMA-RAY SIGNATURE

Gammas from Alpha Decay



[Half-life = 710,000,000 y]

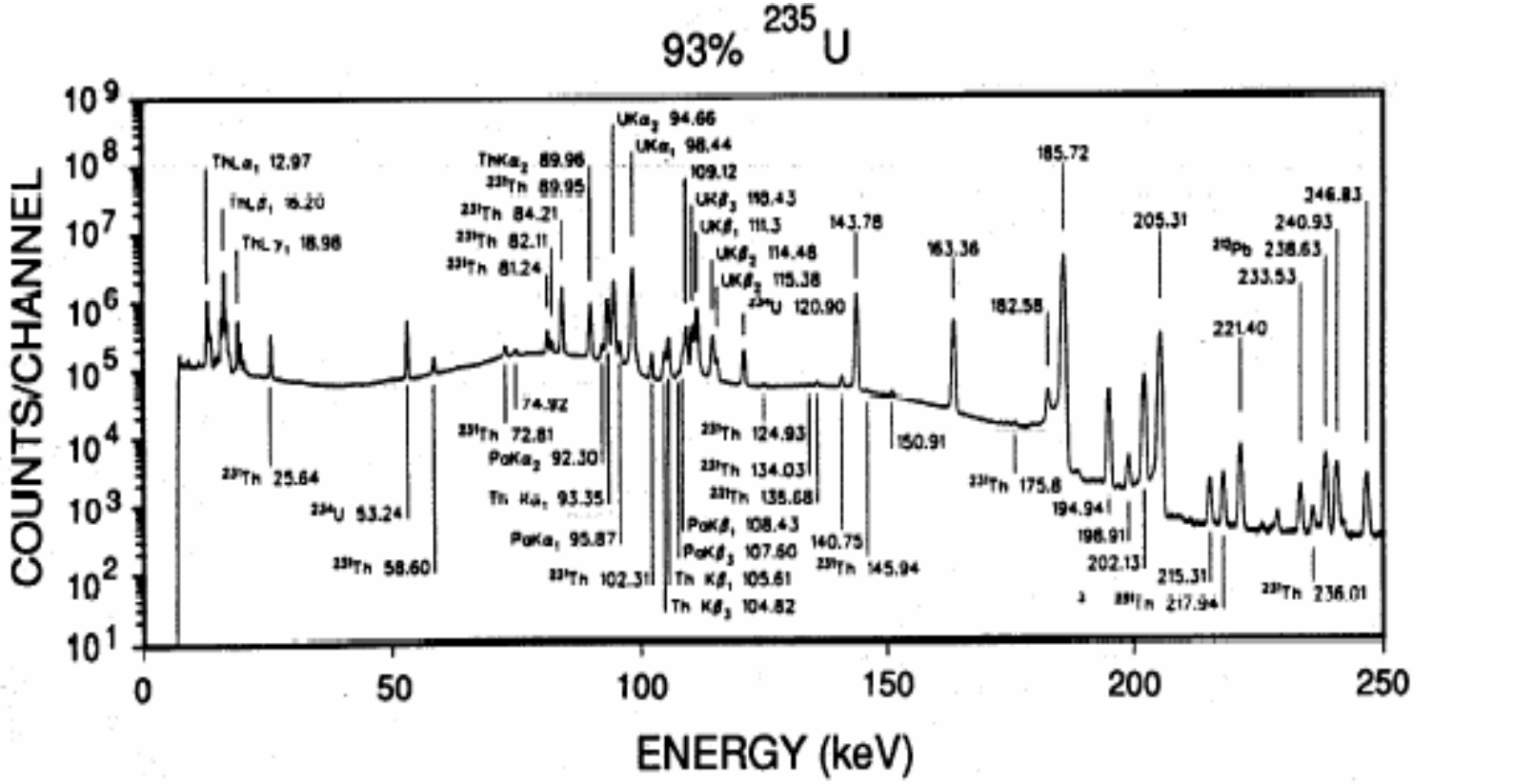
Gammas:

185.7 keV	54%
143.8 keV	11%
163.4 keV	5%
205.3 keV	3%

Los Alamos NATIONAL LABORATORY

Safeguards Science and Technology Los Alamos National Laboratory

Low-Energy Uranium Spectrum



Los Alamos NATIONAL LABORATORY

Principal Uranium g Rays

Element	Energy (keV)	Activity (g/g·s)
^{235}U	143.8	8400
	185.7	43200
^{238}U	766.4	25.7
	1001	73.4

Los Alamos NATIONAL LABORATORY

Detectors to Measure Enrichment

Low resolution

- **Sodium Iodide (NaI)**

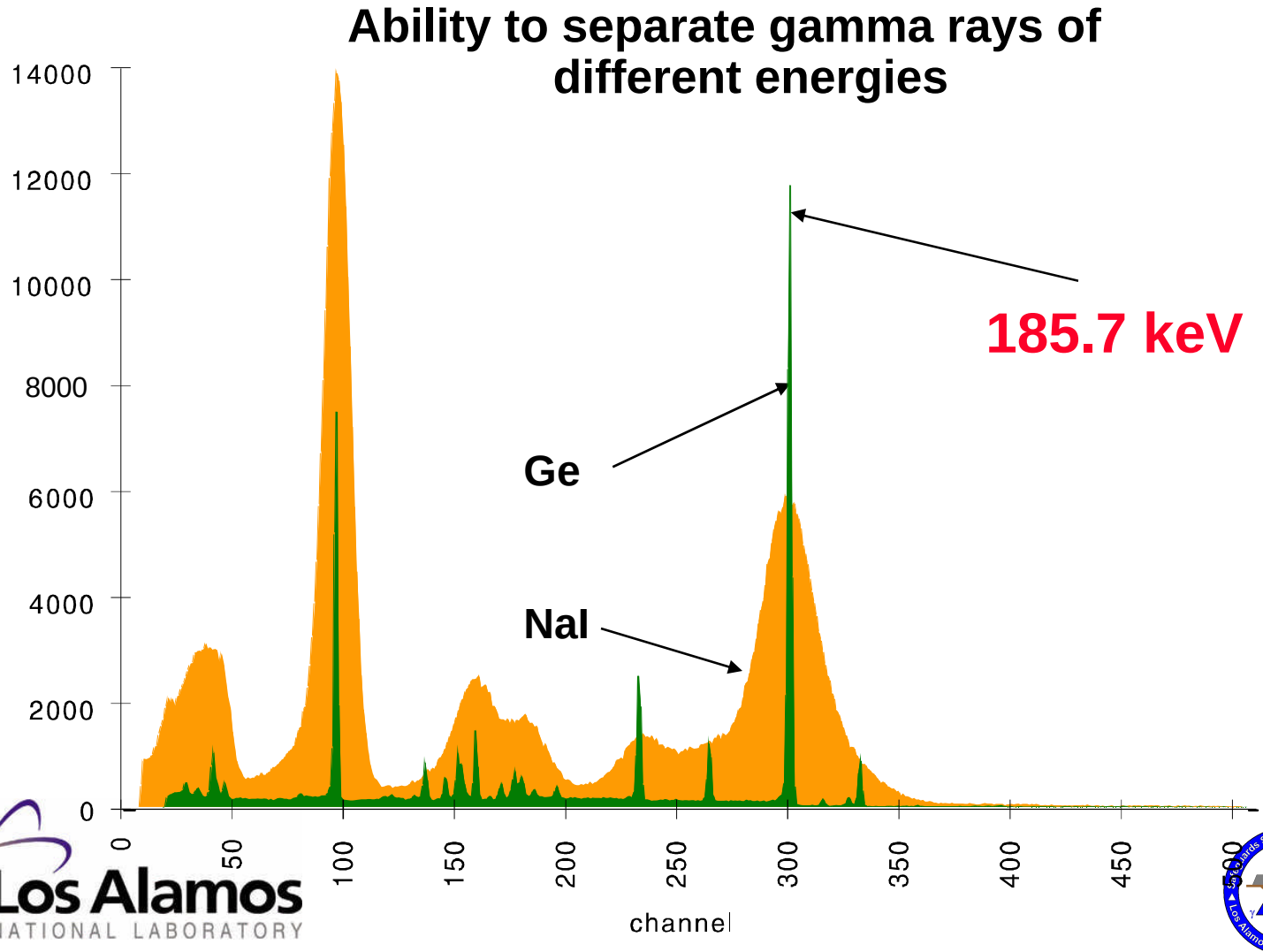
High resolution

- **Germanium (Ge)**
- **CdTe, CdZnTe**

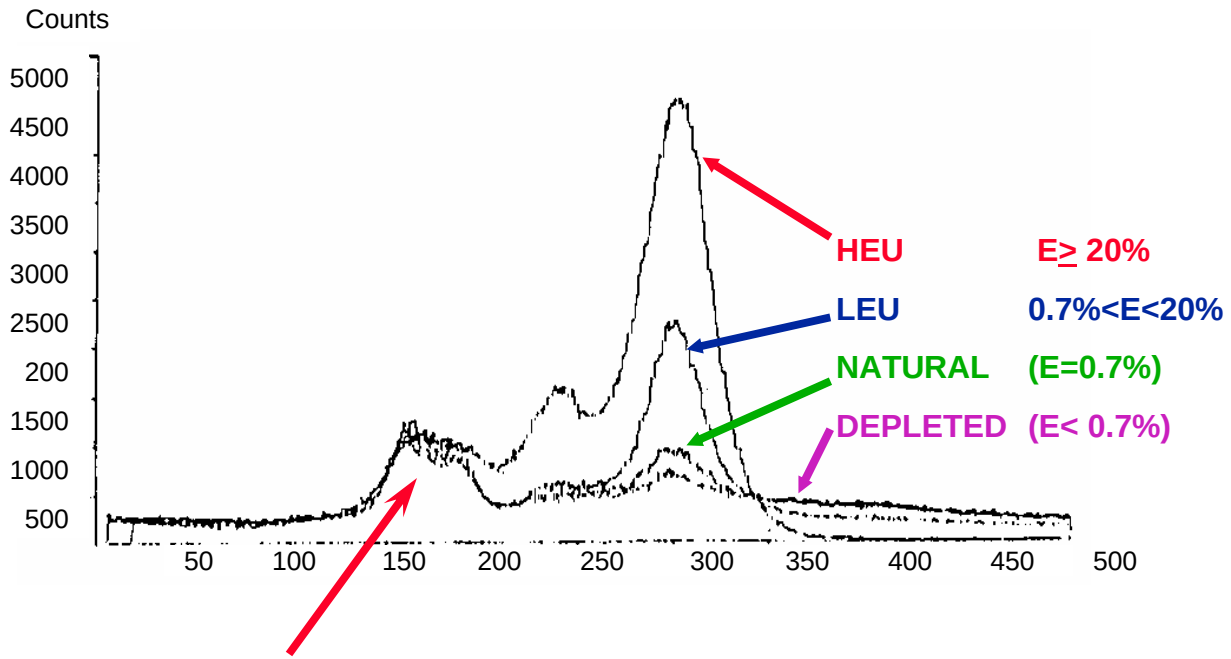
All can provide accurate measurements

Los Alamos NATIONAL LABORATORY

Resolution


Ability to separate gamma rays of different energies
14000
12000
10000
8000
6000
4000
2000
0
0
50
100
150
200
250
300
350
400
450
500
channel
185.7 keV
Ge
NaI

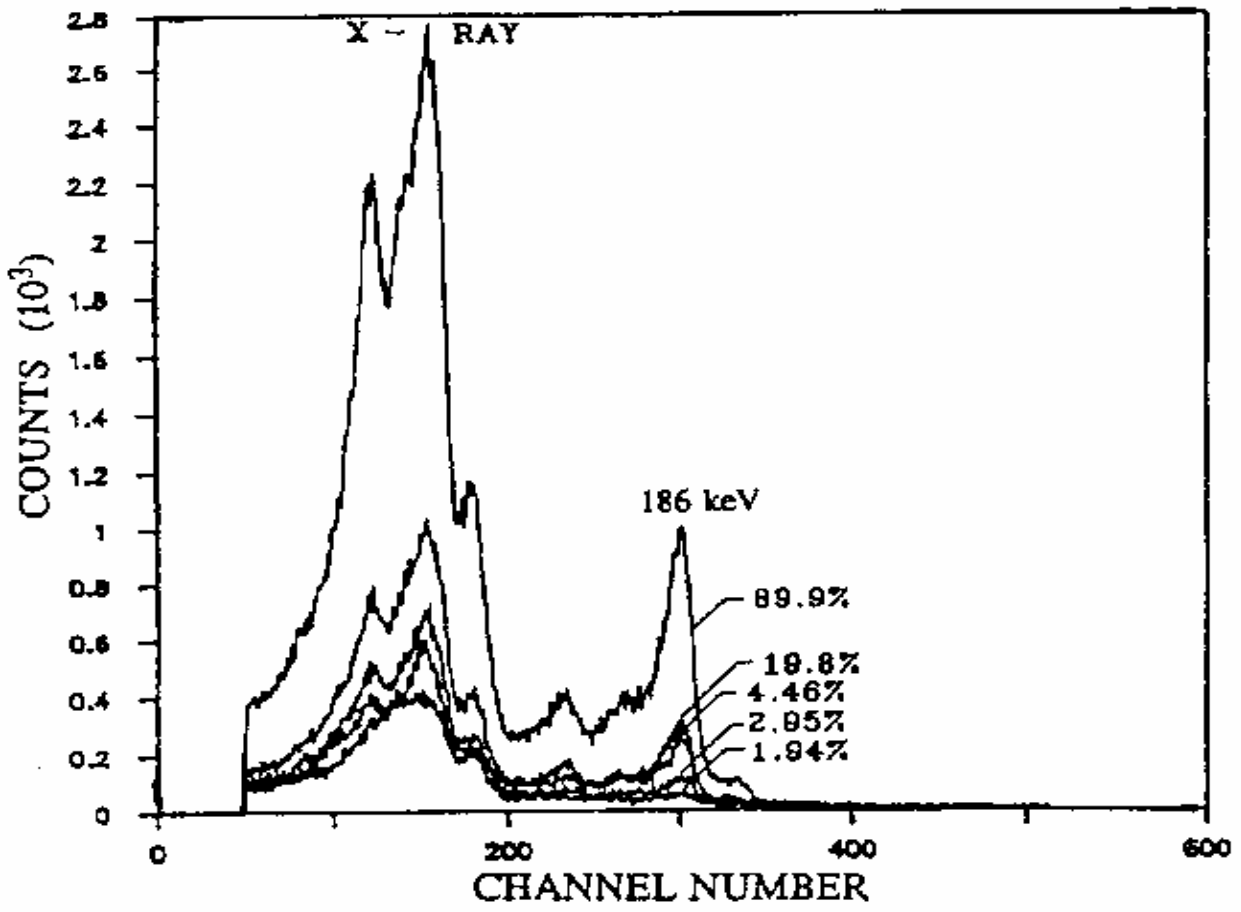
^{235}U Spectra with a NaI Detector



Cadmium preferentially attenuates 100 keV region

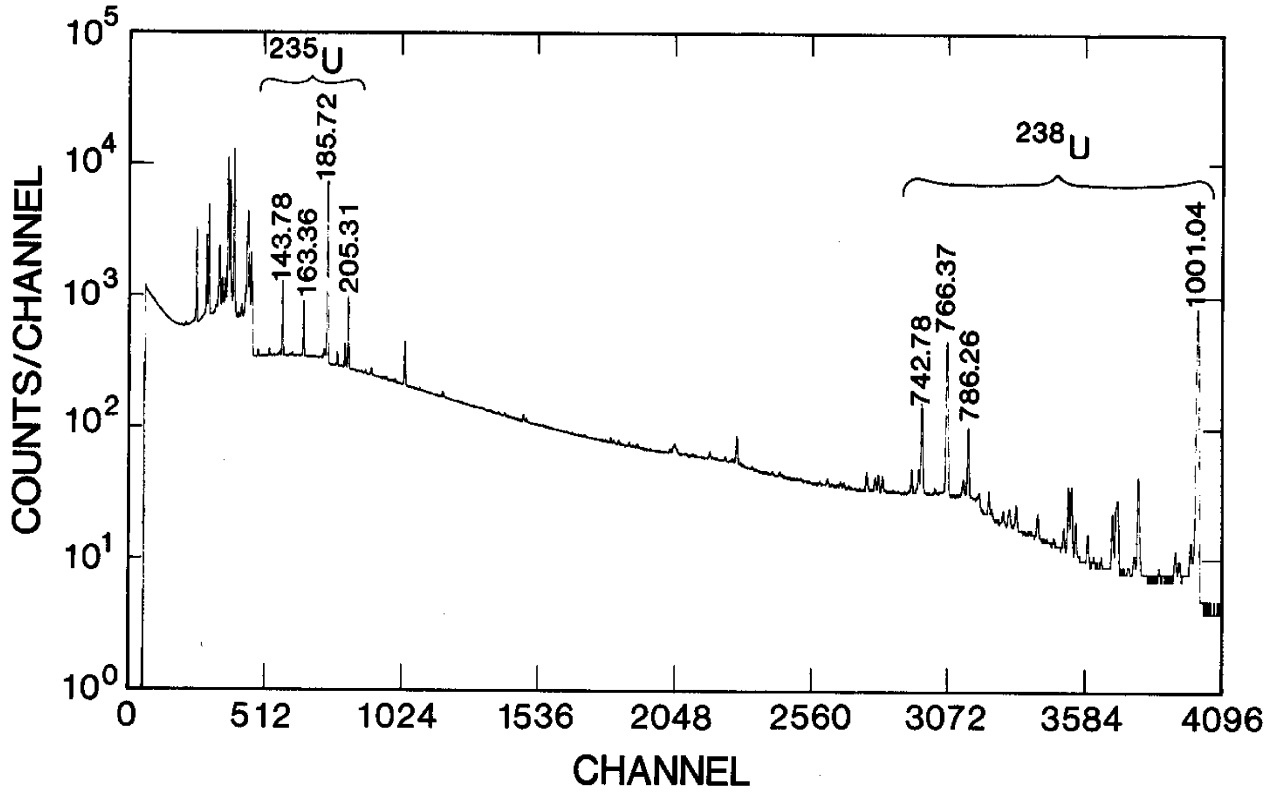
Los Alamos NATIONAL LABORATORY

^{235}U Spectra with a CdZnTe Detector



Los Alamos NATIONAL LABORATORY

^{235}U Spectrum with Ge Detector



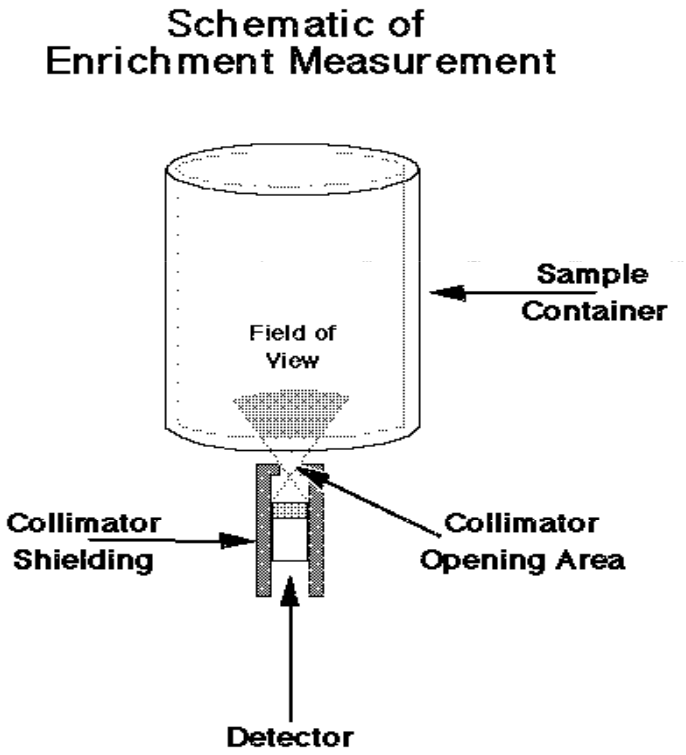
Los Alamos NATIONAL LABORATORY

“Enrichment Meter Technique”

also called

Infinite-thickness Method

Infinite-Thickness Enrichment Assay



Detector views the sample through a collimator.

Visible volume determined by collimator size and the absorption coefficient of U.

The measured 186-keV intensity is proportional to the ^{235}U in the visible volume.

Los Alamos

Los Alamos
NATIONAL LABORATORY

Infinite Thickness for 186-keV γ Rays in Uranium

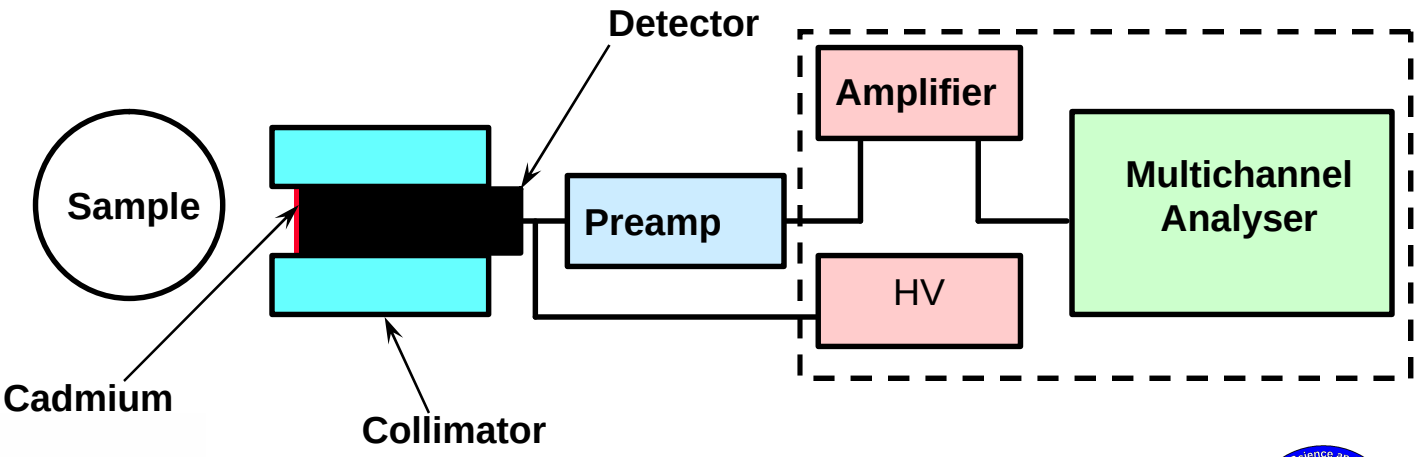
Material	Density(ρ) (g/cm^3)	"Infinite thickness" (cm)[a]
Metal	18.7	0.26
UF_6 solid	4.7	1.43
UO_2 (sintered)	10.9	0.49
UO_2 (powder)	2.0	2.75
U_3O_8 (powder)	7.3[b]	0.74
Uranyl nitrate	2.8	3.01

[a] 7 mean free paths

[b] Highly packed powder

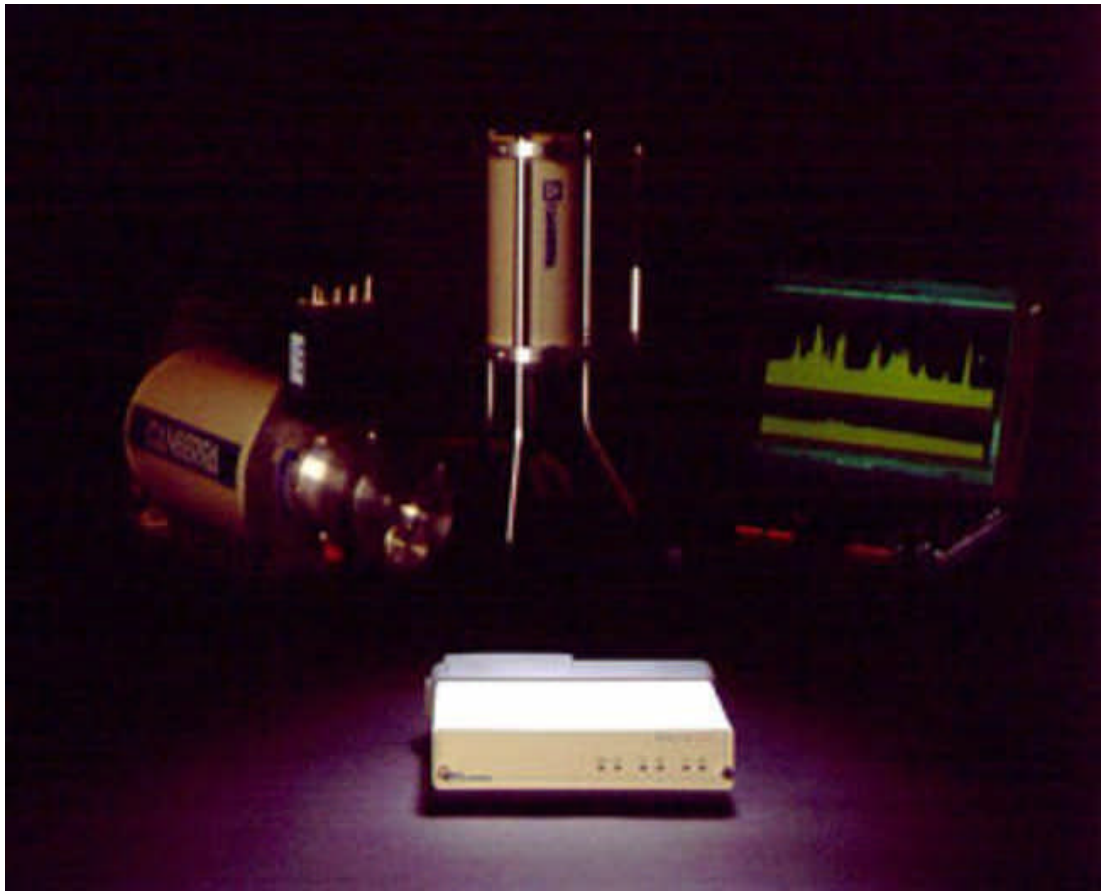
Los Alamos NATIONAL LABORATORY

Enrichment Measurement System



Los Alamos NATIONAL LABORATORY

InSpector - 2000



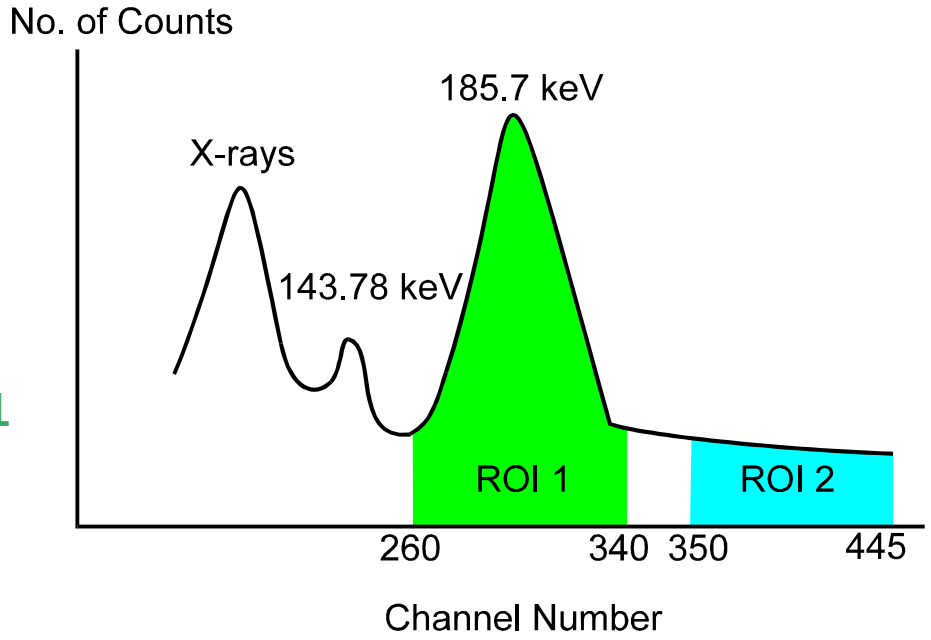
Los Alamos NATIONAL LABORATORY

Two-Region Enrichment Equation
(NaI detector)

$$E = AR_1 + BR_2 \quad (B<0)$$

where:

- **E** is the % ^{235}U
- **A, B** are calibration constants
- R_1, R_2 are count rates in **ROI 1** and **ROI 2**



Los Alamos NATIONAL LABORATORY

Container Wall Thickness Correction, K1

$$E = [A R_1 + B R_2] * K_1 * K_2$$

$$K_1 = e^{\mu T}$$

μ= linear attenuation coefficient (cm^{-1})

T= wall thickness (cm)

example:

μ = 1.21 cm^{-1} (steel)

T= 0.2 cm

$K_1 = e^{\mu T}$ = 1.274

Los Alamos NATIONAL LABORATORY

Calibration Correction for Material Type, K_2

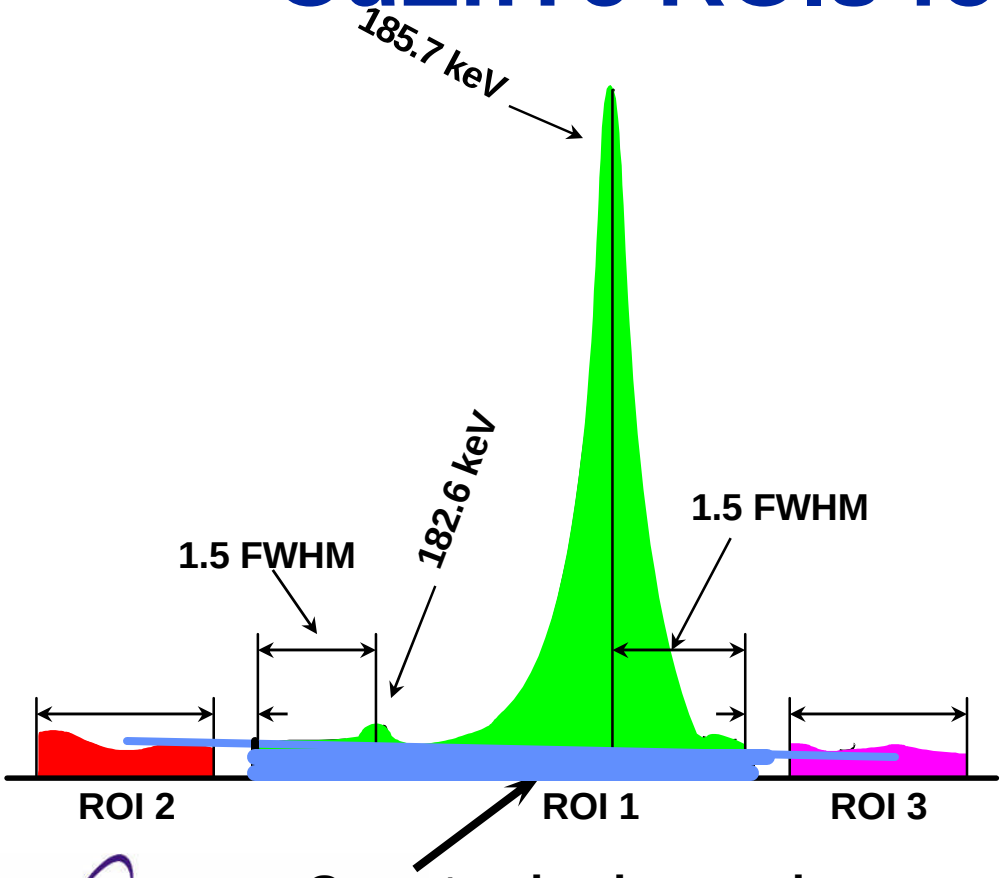
Calibration Standards	Measured Material: U	UC	UO_2	U_3O_8	UF_6	U-Nitrate
U (100% U)	1.000	1.004	1.011	1.014	1.038	1.090
UC (95% U)	0.996	1.000	1.007	1.010	1.033	1.086
UO_2 (88% U)	0.989	0.993	1.000	1.003	1.026	1.078
U_3O_8 (85% U)	0.986	0.990	0.997	1.000	1.023	1.075
UF_6 (68% U)	0.964	0.968	0.975	0.978	1.000	1.051
U-Nitrate (47% U)	0.917	0.921	0.927	0.930	0.952	1.000

These corrections reflect the lower attenuation of the element bound to uranium.

Los Alamos NATIONAL LABORATORY

Infinite-thickness ^{235}U enrichment measurements with high resolution detectors (Ge, CdTe, CdZnTe)

Ge, CdTe, and CdZnTe ROIs for ^{235}U



- Peak ROI 1 includes 185.7- and 182.6-keV γ rays. Both are from ^{235}U
- ROI 2 & 3 measure the Compton background under the ^{235}U peaks
- An ROI of 3 FWHM will include 99.96% of the total area

Los Alamos NATIONAL LABORATORY

Enrichment Equation

$$E = K * (C_{186}) \exp (\mu T)$$

K = calibration constant from a single standard

C_{186} = net count rate of the 186-keV gamma

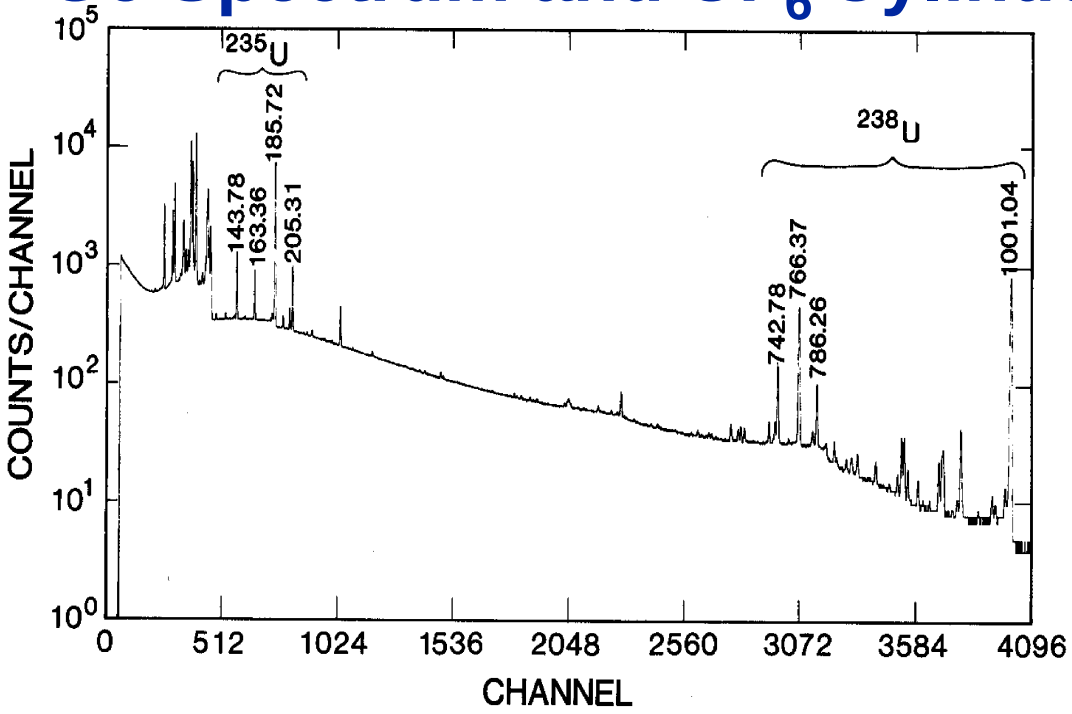
μ = linear attenuation coefficient of container

T = container wall thickness (ultra-sonic thickness gauge)

E = ^{235}U enrichment

Los Alamos NATIONAL LABORATORY

Ge Spectrum and UF_6 Cylinders



UF_6 cylinders cannot be cleaned completely and a heel always remains. This has a high concentration of non-volatile uranium daughters that interfere with the ^{235}U spectrum. *The use of a Ge detector reduces this problem.*

Los Alamos NATIONAL LABORATORY

A High-Quality MC&A System

- **Must be calibrated with standards whose <u>mass and isotopic composition are</u>**
  - **traceable to the national measurement system, and**
  - **determined 3-5 times more accurately than unknowns.**

Los Alamos NATIONAL LABORATORY

Availability of NDA Standards

From US DOE New Brunswick Laboratory (NBL), or

Institute of Reference Materials and Measurements (IRMM - Geel, Belgium)

- **Certified Reference Material (CRM)**
- **Isotopic standards**

Working standards are not available commercially and should be fabricated by each facility.

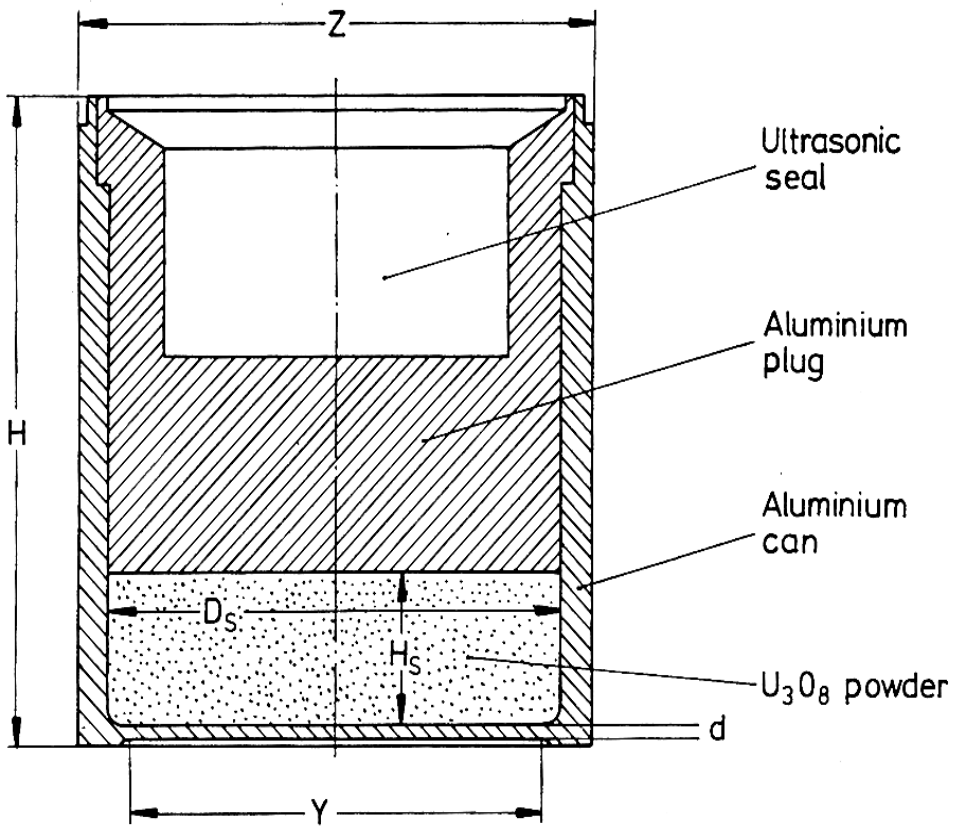
Los Alamos NATIONAL LABORATORY

Examples of NDA Standards

Los Alamos NATIONAL LABORATORY

U Enrichment Standards

EC-NRM-171/NBL-CRM-969



Los Alamos NATIONAL LABORATORY

Enrichments Available

$\%^{235}U$
• 0.27
• 0.72
• 1.99
• 2.99
• 4.49
• 20.0
• 50.0
• 93.3

only available from NBL

Los Alamos NATIONAL LABORATORY

Factors that affect the measurement of U enrichment

- **Sample Material [uniformity and interfering g rays]**
- **Container [wall thickness]**
- **Electronics [stability, pile-up]**
- **Sample size (infinite thickness)**
- **Collimator geometry [diameter, depth, distance to detector]**
- **Shielding against background radiation**
- **Gamma detector [efficiency, energy resolution]**

Los Alamos NATIONAL LABORATORY

Accuracy of Infinite-Thickness Method

- 1 – 5% typical depending on sample.
- NaI, CdZnTe, and Ge can provide similar accuracy.
- In special applications, involving installed systems, 0.1 – 0.2% is possible.
- ^{235}U enrichment measurement is the most accurate NDA technique.

Los Alamos NATIONAL LABORATORY

Comments on Standards

- **Standards should satisfy the fundamental assumption of uniformity**
- **SNM form and amount should be stable over time**
- **Standards should be similar in size & shape to unknowns, but a good deal of reasonable extrapolation is possible**
- **Standards DO NOT have to be of the same chemical composition as the unknowns*!!***

"A highly skilled measurement technician who can apply the proper measurement physics is far more valuable than a comprehensive set of standards."*— J.L. Parker*

Los Alamos
NATIONAL LABORATORY

Producing WNDA Standards

Facilities must:

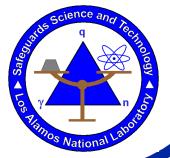
- produce the standards
- characterize each standard [i.e., define mass and/or isotopic composition]
- maintain and document traceability
- perform and document measurement control on
  - **the characterization of the WRM**
  - **the use of standards in NDA calibrations**

Los Alamos NATIONAL LABORATORY

Producing WNDA Standards
Continued

- **Use existing nuclear materials at facility**
- **Characterize mass and/or isotopic values using traceable analytical measurement techniques**
- **Monitor quality of standards through reciprocal measurement exercises with other laboratories**

Los Alamos
NATIONAL LABORATORY



Production of WNDA standards begins with Certified Reference Material (CRM)

- Generally highly pure U or Pu metal
- Characterized by certified laboratory
- Used in small quantities to make working standards for destructive analysis
- Mass value established gravimetrically (thus, directly traceable to international system of weights and measures)

Los Alamos
NATIONAL LABORATORY

Working Enrichment Standards

1 kg U_3O_8 enriched

0.72 at. %

1.96

3.07

10.20

11.91

13.09

17.43

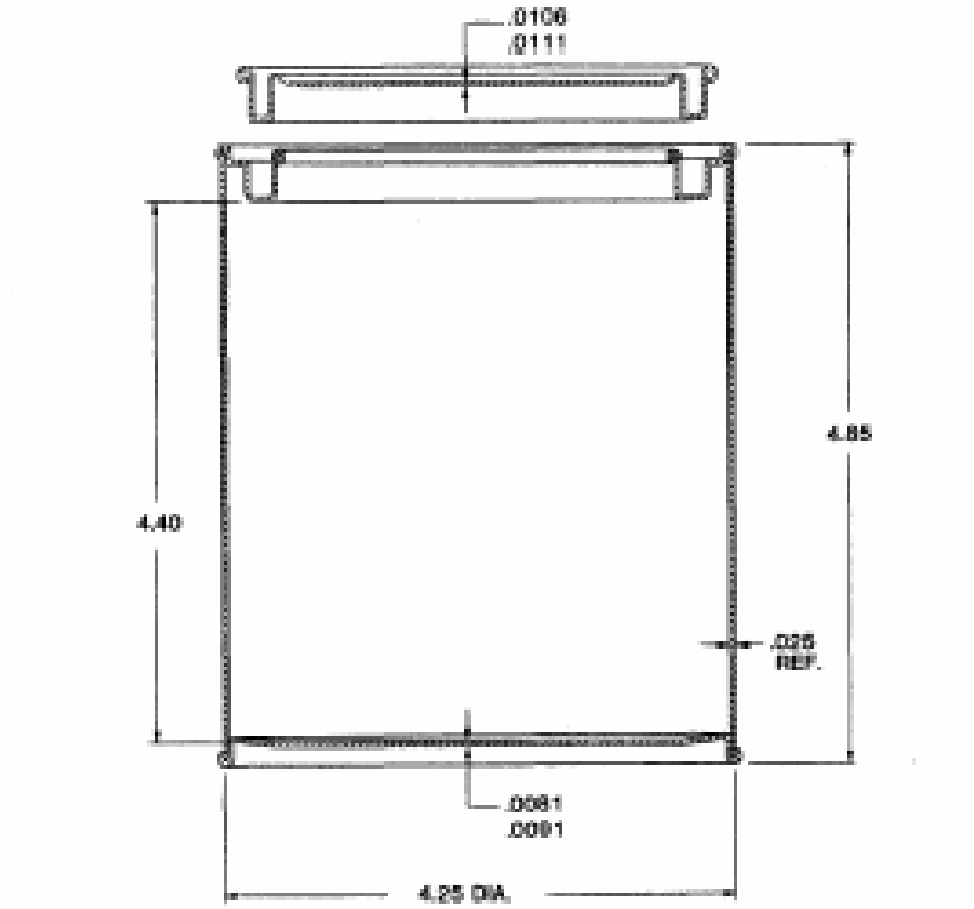
27.04

37.83

52.43

66.31

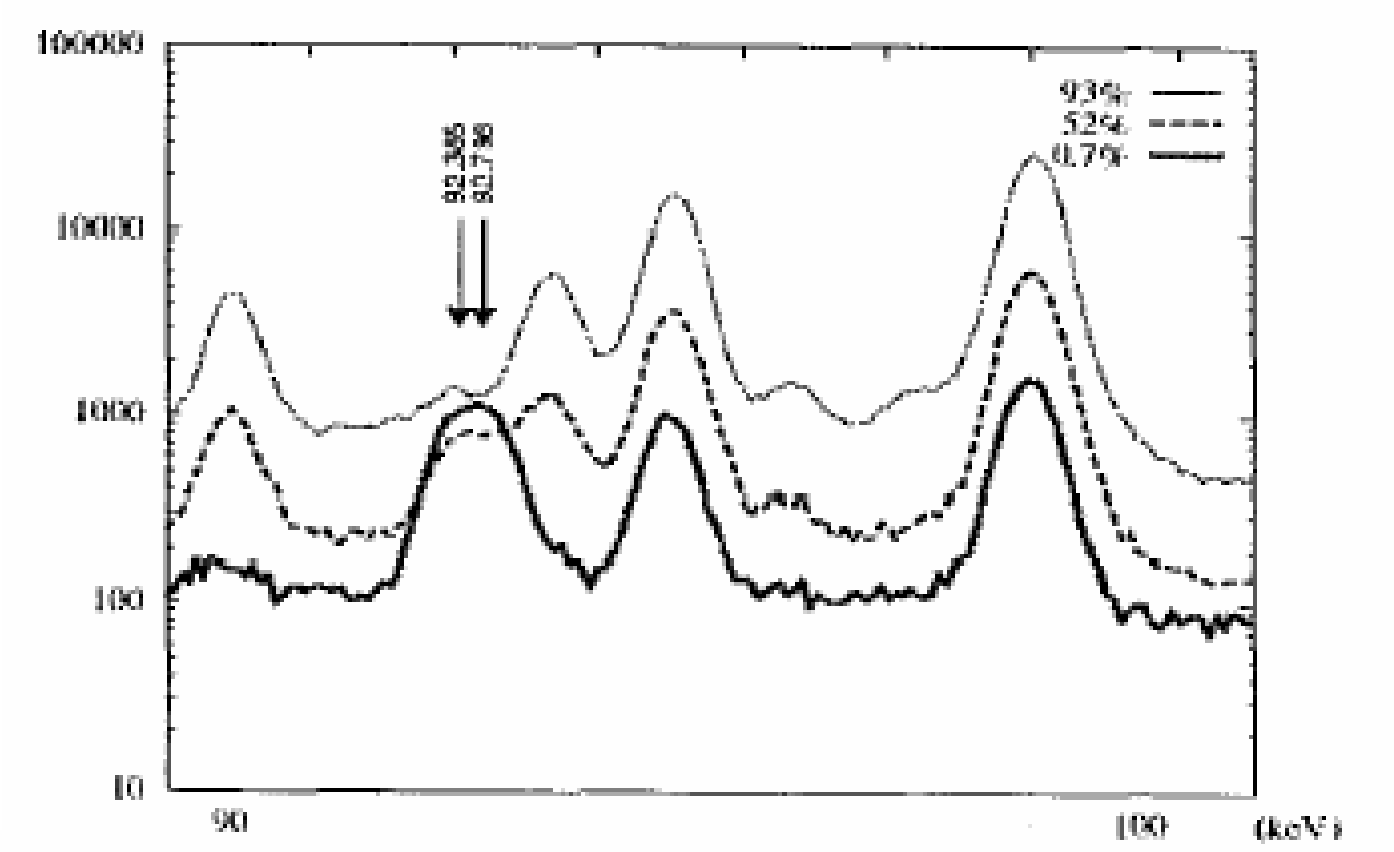
91.42



Los Alamos NATIONAL LABORATORY

Response function fitting, MGAU, for U isotopic composition

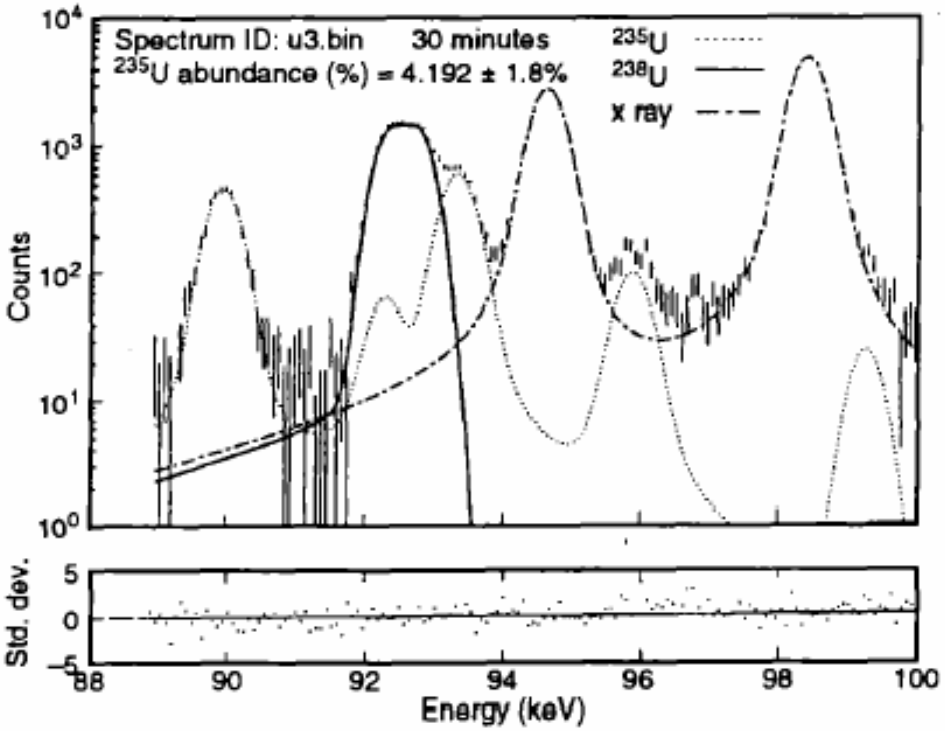
Energy Region Used by MGAU



Los Alamos
NATIONAL LABORATORY

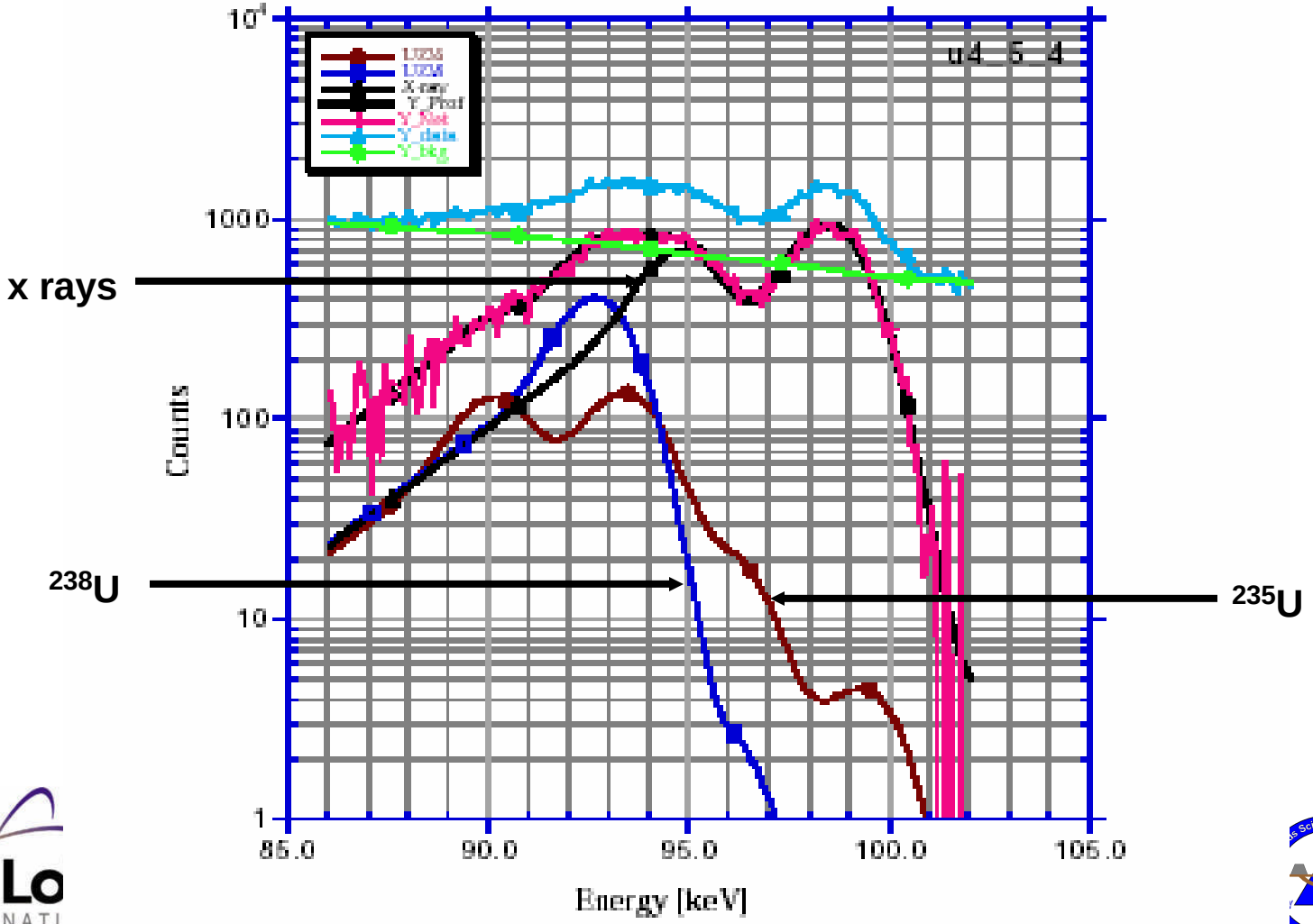
Uranium Gamma-Ray Spectrum

MGAU analysis region



Los Alamos NATIONAL LABORATORY

Response Function Fitting with CdZnTe



MGAU and Ge

- **No calibration or standards required**
- **Analyses 89 - 100 keV** for **^{234}U, ^{235}U, and ^{238}U**
- Measures **from depleted to fully enriched**
- **Typical precision: ^{235}U to ± 2% in 300 s**
- Container wall thickness must be less than 10mm steel

Los Alamos NATIONAL LABORATORY

Conclusions

Infinite-Thickness Method

- Any detector (NaI, CdZnTe, Ge)
- Requires stable geometry, good standards, and careful calibration.
- Typical accuracy 1 – 5%.
- In fixed installations, 0.1 – 0.2% possible.

Response-Function Fitting Method (MGAU)

- Requires high-resolution detector (Ge, CdZnTe).
- No standards or calibration required.
- Typical accuracy 2% in 300s.

Los Alamos NATIONAL LABORATORY

Standards and Calibration for Portable *In-Situ* Gamma-Ray Measurements

LA-UR-03-3799

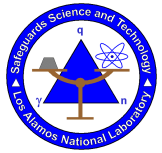
P. A. Russo, T. R. Wenz

ABSTRACT

The materials and methods for calibrating quantitative *in-situ* measurements of plutonium and uranium are presented. Because the standards do not (typically can not) match the composition and distribution of *in-situ* deposits, analysis methods rely on models. Models for geometry, attenuation, *etc. are* described for rapid plant-wide measurements of solid deposits and solutions. Measurement results are presented.

Portable *In-Situ* Gamma-Ray Measurements: Holdup

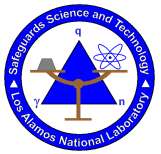
I. Unknowns *vs.* Standards
II. Calibration
    - Models
    - Assumptions
III. Revised models
IV. Results
V. Solution Measurements
VI. Discussion, Conclusions



I. Unknowns *vs.* Standards

***In-situ* SNM deposit (holdup) characteristics**

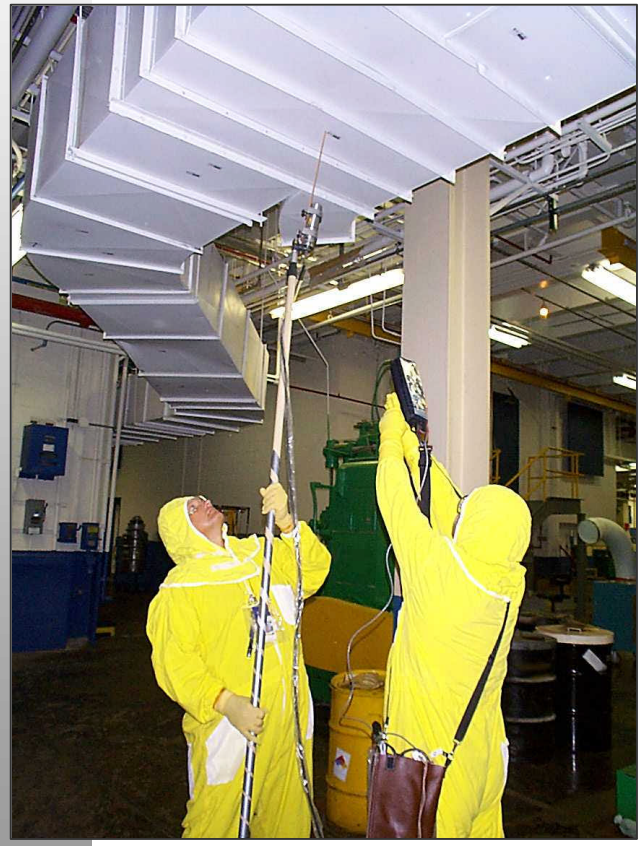
- Can consist of metals, compounds, mixtures...
- Can have mixed SNM.
- Can have other radionuclides.
- Is varied in shape with vast dimensions.
- Thickness is nonuniformly distributed.*
- Is widely distributed throughout the plant.
- SNM (not matrix) dominates the attenuation.
- Self attenuation is relatively small nonetheless.
- Sums to large SNM quantities.



* \ **thousands of very short measurements are needed.**

I. Unknowns *vs.* Standards

Examples of equipment with SNM holdup



Overhead ducts and piping at the Y-12 plant

I. Unknowns *vs.* Standards

Examples of equipment with SNM holdup

Vertical ducts and piping at the Y-12 plant

I. Unknowns *vs.* Standards

Examples of equipment with SNM holdup

Arrays of piping at the Y-12 plant

I. Unknowns *vs.* Standards

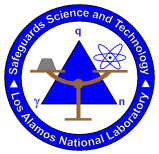
Examples of equipment with SNM holdup

Glove boxes and overhead ducts at Los Alamos TA-55

I. Unknowns *vs.* Standards

***In-situ* SNM solution characteristics**

- Is typically uniform chemically.
- Can have mixed SNM.
- Can have other radionuclides.
- Is varied in shape with vast dimensions.
- Thickness is (usually) uniformly distributed.**
- Is widely distributed throughout the plant.
- Has attenuation from both SNM & matrix.*
- Has relatively large self attenuation. *
- Sums to large SNM quantities.

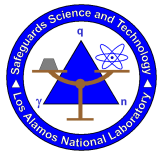


* **Applies to "sludge" as well.** ** \ **fewer measurement locations.**

I. Unknowns *vs.* Standards

Small, well characterized calibration standards are valid nonetheless:

- γ-ray response is independent of SNM form.
- Calibrate with standards for multiple SNM types.
- Select γ rays/detectors to avoid interferences.
- Model response for simple geometric shapes; correct for deviations from model assumptions.
- Use distance to “sample nonuniformities”.
- Analytical algorithms enable automation.
- Correct for self-attenuation with general self-consistent models.
- Measure both specific and total SNM mass.



I. Unknowns *vs.* Standards

The role of standards in a multi-part calibration

- Small well-characterized "point" standards determine only the γ-ray point response (counts/s/unit mass), independent of other effects (geometry, attenuation…).

- Additional measurements made at calibration determine parameters for geometric models of holdup deposits that supplant needs for representative standards.

- Other characterized* materials verify calibrations applied to materials of varying 1.) geometry (point, line, area, large point, wide line…) and 2.) attenuation.

* by i) sampling, ii) measurements using NDA reference techniques, or iii) calculations

I. Unknowns *vs.* Standards

Y-12 Standards for Uranium Holdup Measurements (Discs)

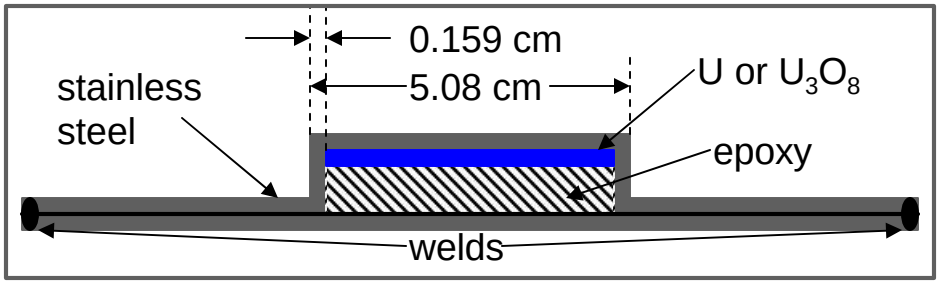
Source Material

U_x : U_3O_8 NBL CRM(U930)

U_{assay} = 0.8445 gU/g
^{234}U = 1.0759 (wt %)
^{235}U = 93.276 "
^{236}U = 0.2034 "
^{238}U = 5.445 "

U_m: U OR Alloy Metal

U_{assay} = 0.999133 gU/g
^{234}U = 1.016 (wt. %)
^{235}U = 93.162 "
^{236}U = 0.400 "
^{238}U = 5.421 "



Container Properties

Stainless Steel Holder: inside radius = 2.381 cm, μ_{steel} = 0.1459 cm^2/g
wall thickness = 0.159 cm, ρ_{steel} = 8.02 g/cm^3

CF_{steel} = 1.2045

Nuclear Properties

^{235}U g-ray Energy = 185.72 keV

U_x: μ = 1.2638 cm^2/g
ρ = 8.313 g/cm^3
radius = 2.381 cm
area = 17.810 cm^2

U_m: μ = 1.4679 cm^2/g
ρ = 18.759 g/cm^3
radius = 1.6828 cm
area = 8.896 cm^2

Set	std. #	mass	U	^{235}U	thickness (x)	T	CF_{self}
#02	001	0.052g U_x	0.044g	0.041g	0.000351 cm	0.9969	1.0016
#02	002	0.102g "	0.086g	0.080g	0.000689 cm	0.9939	1.0031
#02	003	0.253g "	0.214g	0.199g	0.001709 cm	0.9849	1.0076
#02	004	0.500g "	0.422g	0.394g	0.003377 cm	0.9705	1.0150
#00	005	1.000g "	0.845g	0.788g	0.006754 cm	0.9418	1.0303
----	006	11.358g U_m	11.348g	10.572g	0.068061 cm	0.1537	2.2127
----	nmca	11.704g "	11.694g	10.894g	0.070134 cm	0.1452	2.2574

CF Equations (slab)

$\rho x = U / \pi r^2$
$T = e^{-\mu\rho x}$
$CF_{slab} = - \ln (T) / (1-T)$

I. Unknowns *vs.* Standards

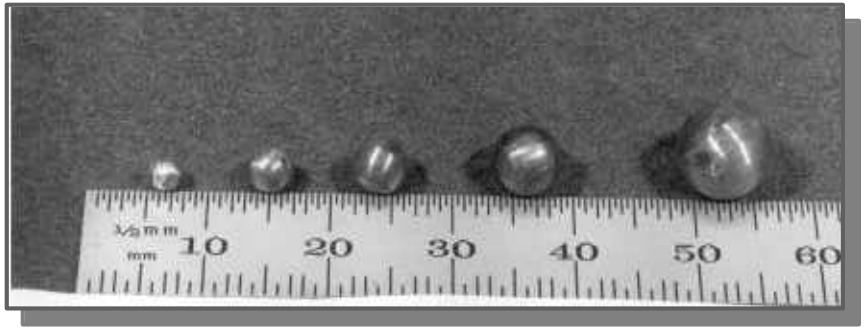
Los Alamos Standards for Plutonium Holdup Measurements (Metal Spheres)

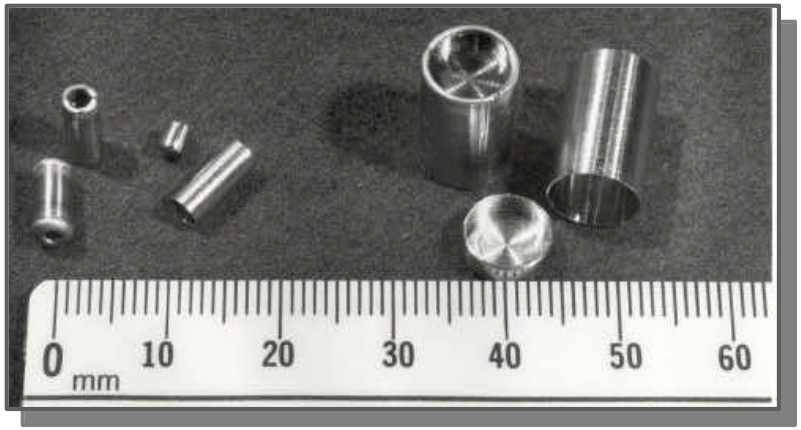
Source Material (7/1974)

Density: r_{metal} = 19.56 g/cm^3

Pu metal (spheres): Pu mass fraction = 99.89%

Impurity mass fractions (mg/g)		**Isotope atom %**	
Fe	100	^{238}Pu	0.016
F	5	^{239}Pu	93.56
C	20	^{240}Pu	5.92
O	210	^{241}Pu	0.462
Sc	<20	^{242}Pu	0.033
Ga	240		
Am-241	189		





Container Properties

Welded stainless steel: thickness = 0.0254 cm

ρ_{Fe} = 7.9 g/cm^3

$\mu_{414\ keV}$ = 0.091 cm^2/g

Container attenuation: **$CF_{414\text{-}keV}$ = $e^{+0.091 \cdot 7.9 \cdot 0.0254}$ = 1.018**

Self-Attenuation Algorithms (414-keV γ rays)

$CF_{sphere} = 1 / \{(3/2Z)[1 - 2/Z^2 + e^{-Z}(2/Z + 2/Z^2)]\}$

where $Z = \mu\rho D$,

D = sphere diameter,

ρ = density of plutonium (in sphere) = 19.56 g/cm^3,

μ = Pu mass attenuation coefficient = 0.29 cm^2/g .

Sphere Data (^{239}Pu mass fraction = 0.9354 g ^{239}Pu / g Pu)

ID	**Pu Mass (g)**	**^{239}Pu* Mass (g)**	**D_{min} / D_{max} (mm)**	**CF_{sphere}**
SPH1	5.3487	5.0032	8.04 / 8.05	3.345
SPH2	2.1177	1.9809	5.90 / 5.91	2.630
SPH3	1.0656	0.9968	4.71 / 4.72	2.253
SPH4	0.5383	0.5035	3.73 / 3.75	1.958
SPH5	0.2155	0.2016	2.74 / 2.76	1.675

II. Calibration Models/GG Holdup

Generalized-Geometry (GG)
Calibrate the quantitative g-ray spectroscopic assay for generalized (point, line, area) source geometries. First, measure absolute response.

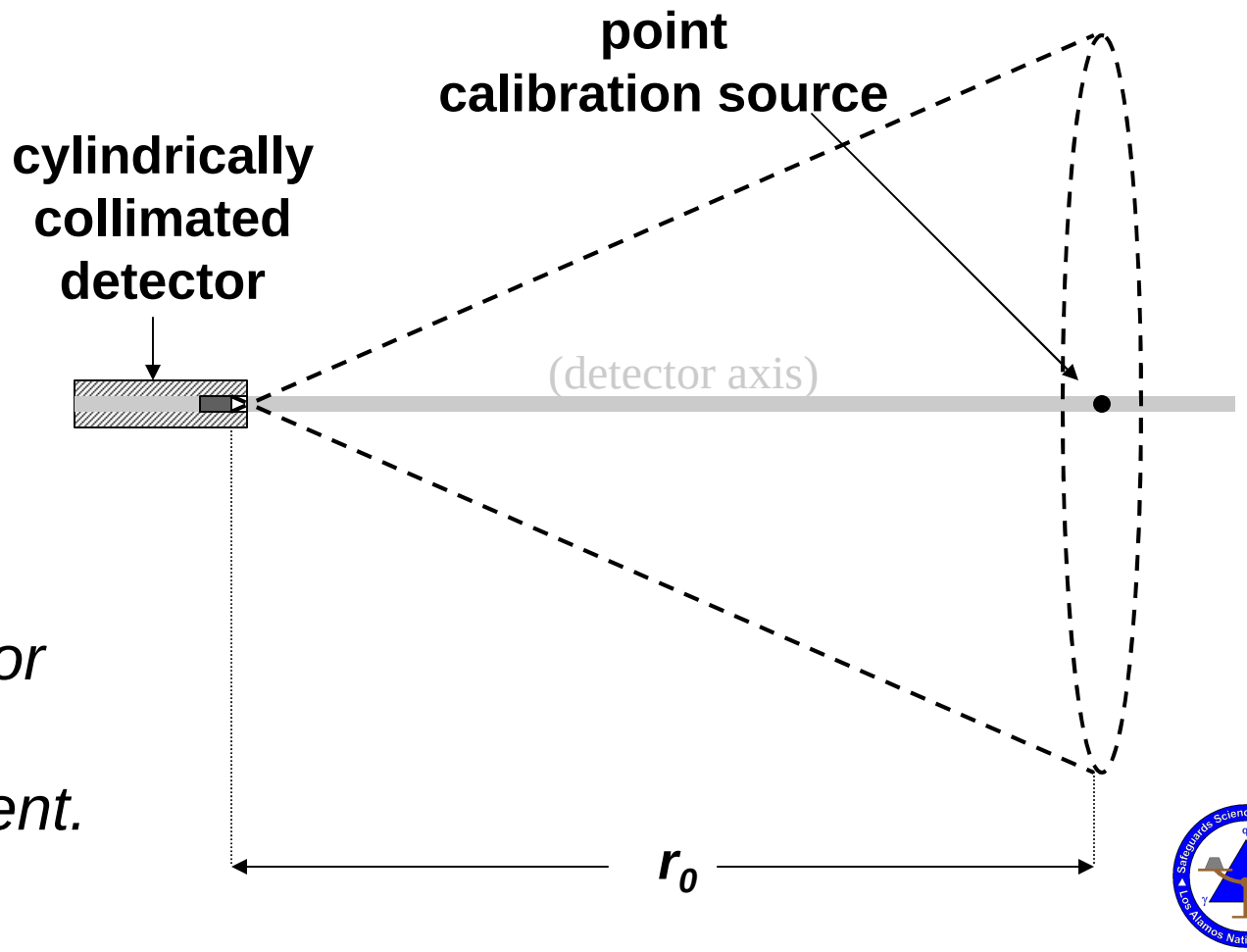
Use:

- **a cylindrically collimated g-ray detector.**
- **measured source-to-detector distance, r_o.**
- **point standard g-ray source and rotational symmetry to measure calibration response.**

Get response (C_0 = counts/s/g_{SNM}) for the ideal* on-axis point source at r_0.

*** very small point**

II. Calibration Models/GG Holdup



Calibration geometry illustrated for response measurement.

II. Calibration Models/GG Holdup

Move point source off axis on r_0 locus (line perpendicular to detector axis) across detector's field of view FOV. Measure geometric parameters.

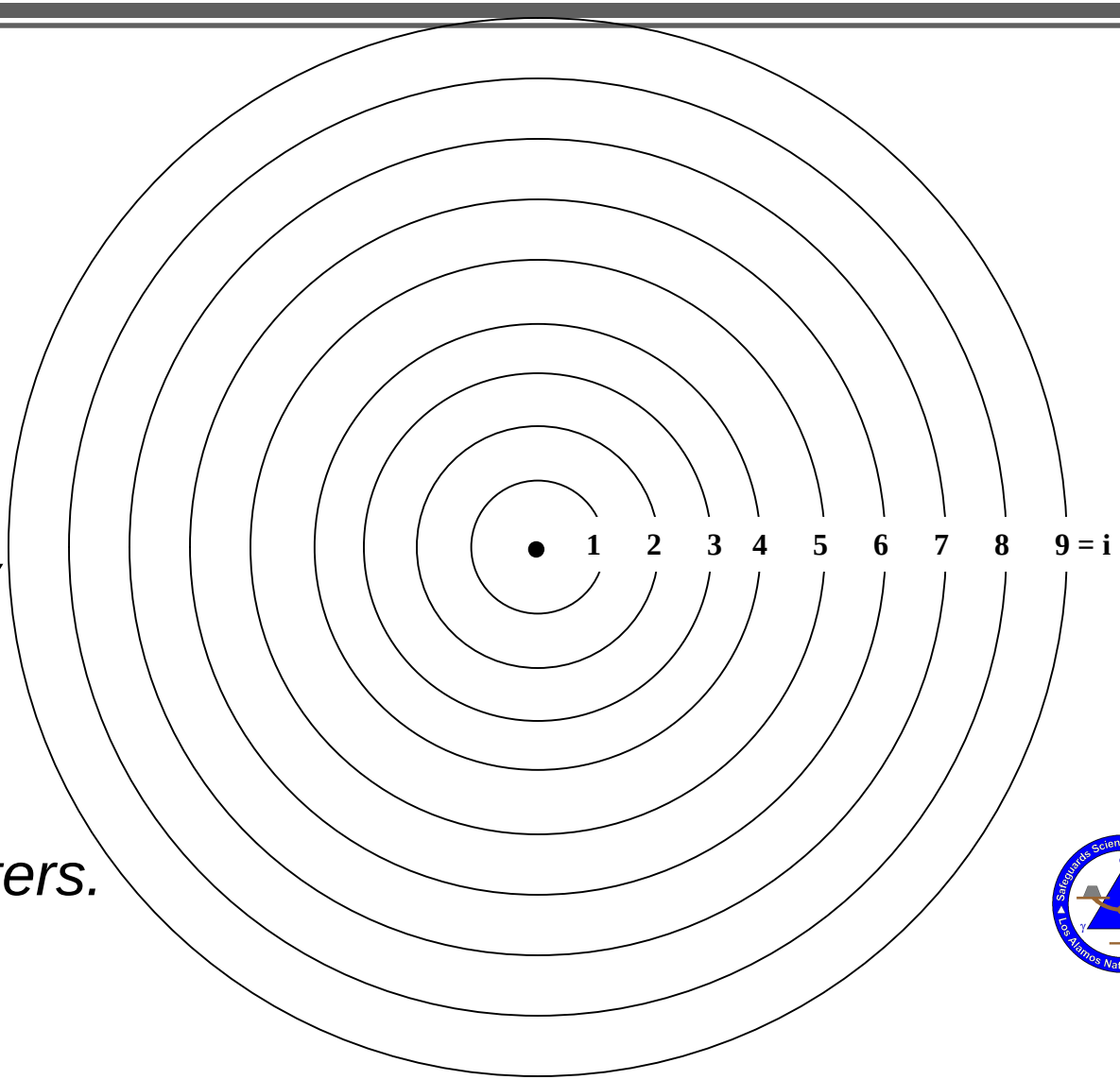
- **Measure response at each position.**
- **Integrate response over FOV <u>width</u> for "line" parameter.**
- **Integrate response over FOV *area* for "area" parameter.**

Get <u>effective length</u> and <u>effective area</u> parameters (*L* and *A*) for ideal* line and area sources that fill the circular FOV.

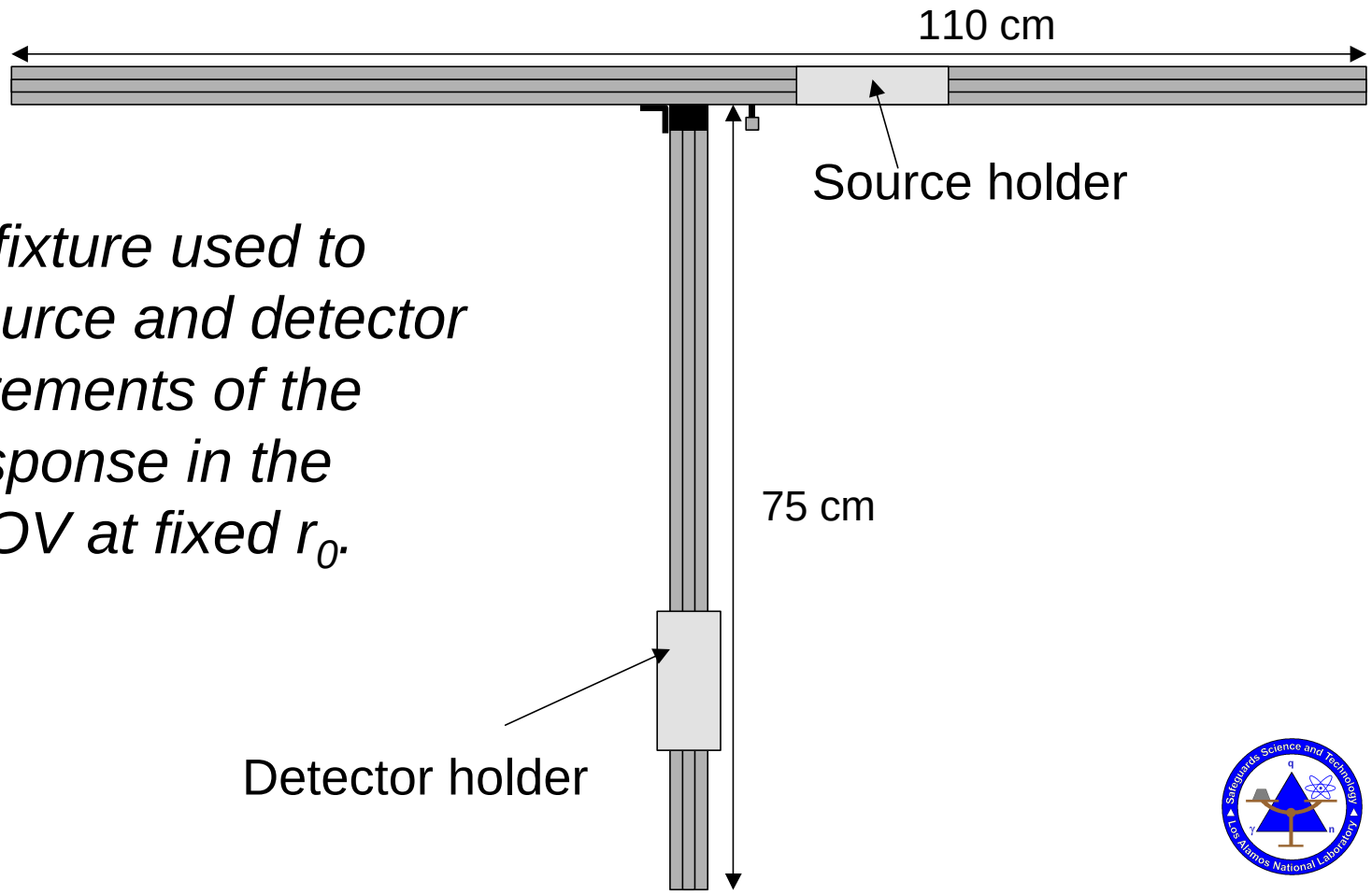
*** uniform sources, very narrow line**

II. Calibration Models/GG Holdup

Measurements at $\boldsymbol{r_0}$ *of nine off-axis source positions in the circular FOV give count rates* C_i *(i = 1-9) used to determine the geometric parameters.*



II. Calibration Models/GG Holdup

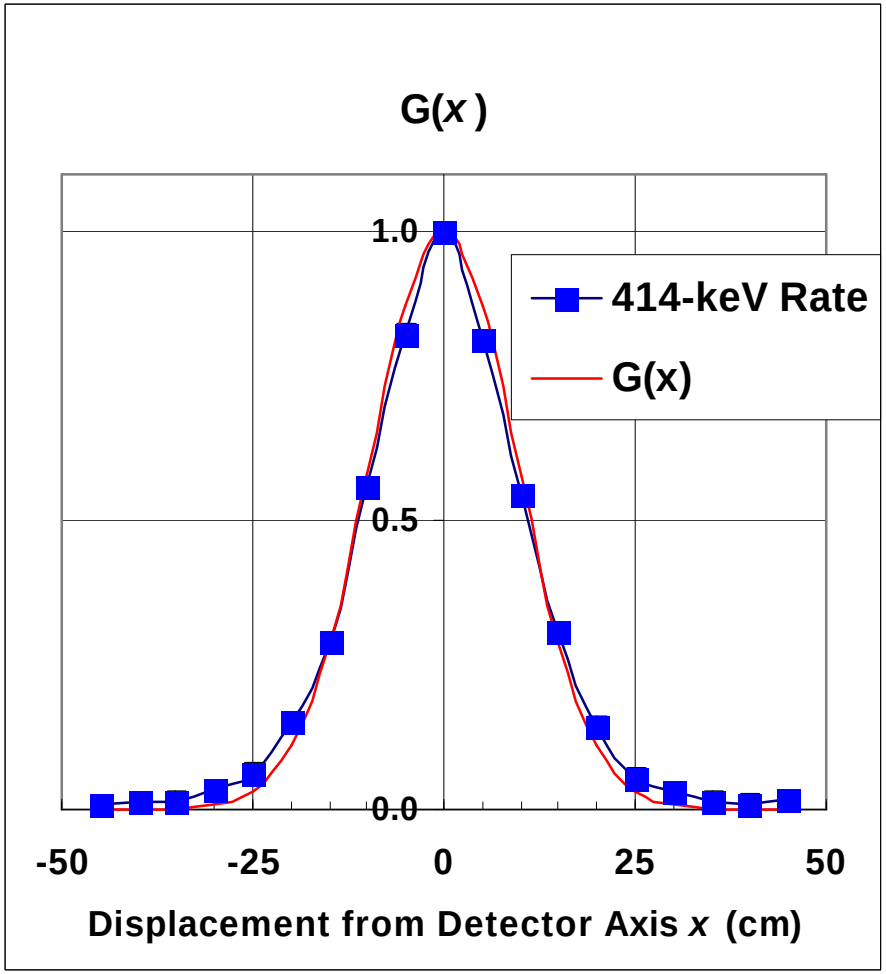


Hardware fixture used to position source and detector for measurements of the off-axis response in the detector FOV at fixed r_0.

.

II. Calibration Models/GG Holdup

The measured responses at nine positive and negative off-axis plutonium source positions are normalized to the on-axis response.



II. Calibration Models/GG Holdup

Calibration equations for ideal deposit geometries give the specific SNM mass for

- **POINT (P):** $g_{SNM} = C(K_P r^2)$
- **LINE (L):** g_{SNM}/unit length $= C(K_L r)$
- **AREA (A):** g_{SNM}/unit area $= C(K_A)$

deposits.

C = count rate for a P, L or A deposit at distance *r*
***r* = deposit-to-detector measurement distance**
$K_P = m_0/C_0 r_0^2$
$K_L = m_0/C_0 r_0 L$
$K_A = m_0/C_0 A$

II. Calibration Assumptions/GG Holdup

Requirements for ideal holdup deposits:

1. **Specific mass of L or A deposits is uniform across FOV.**
   Failure to meet requirement contributes to random uncertainty (not bias) in measured holdup.

2. **Width, *w*, of P or L deposits is very small compared to FOV.**
   Failure to meet requirement contributes to negative bias in measured holdup.

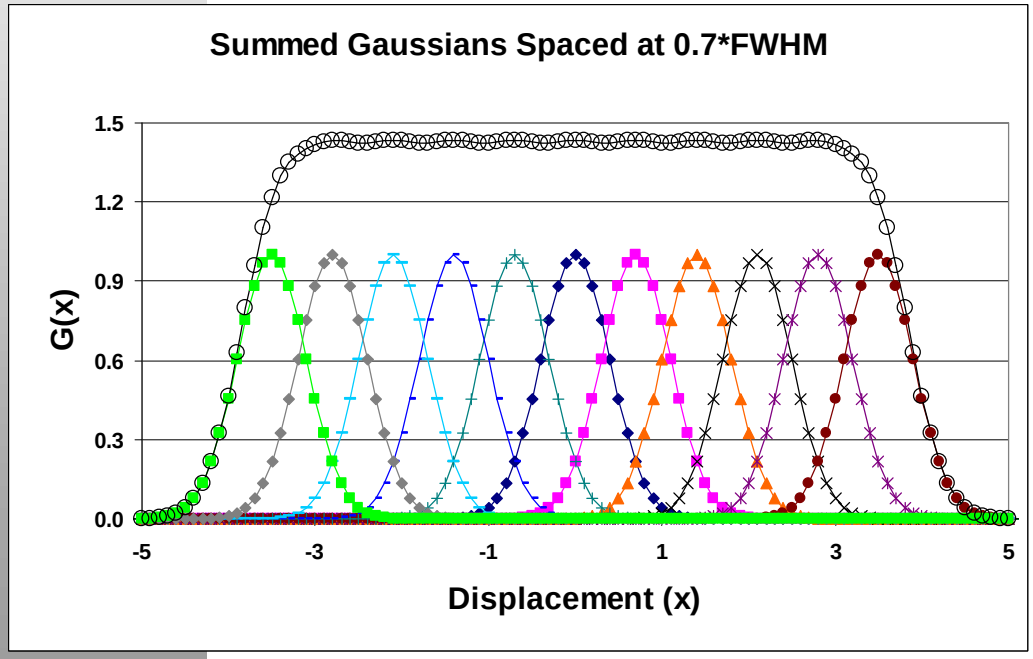
3. **g-ray self-attenuation in deposits is very small.**
   Failure to meet requirement contributes to negative bias in measured holdup.

Holdup meets none of these requirements.

II. Calibration Assumptions/GG Holdup

Reduce random uncertainty from

(1.) Nonuniform specific mass of L or A deposit.



Make "sampling" uniform with spacings of ~FWHM (of detector's radial response) between measurements. Increasing ***r****, the measurement distance, helps achieve this.*
NB: Nonuniformities do not contribute to bias.

II. Calibration Assumptions/GG Holdup

Eliminate negative bias from

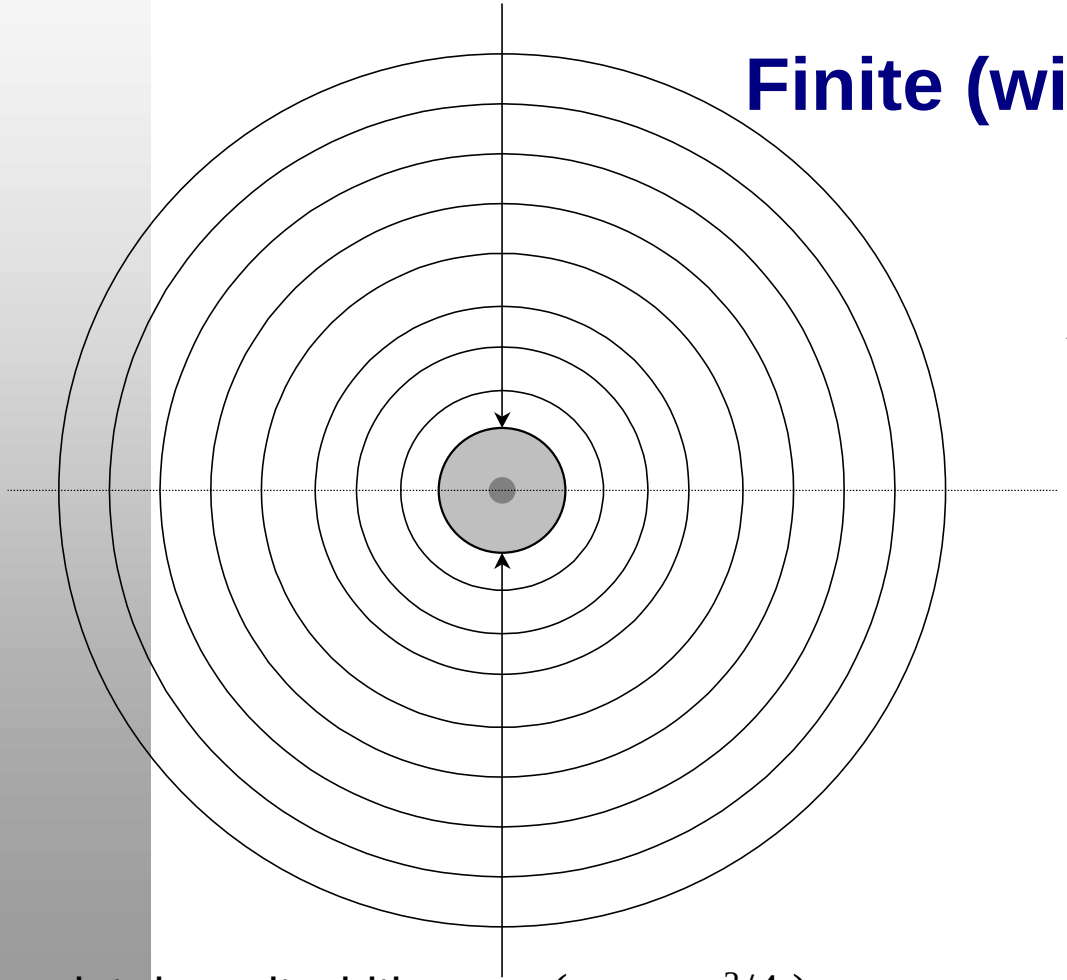
(2.) finite width, w, of P and L holdup deposits.

(3.) significant g-ray self-attenuation by holdup deposits.

This requires revising models but retaining generalized approaches that

- **apply to all deposit geometries.**
- **employ programmable algorithms for rapid plantwide measurements.**
- **are immune to user skill and subjectivity.**

III. Revised Models/GG Holdup

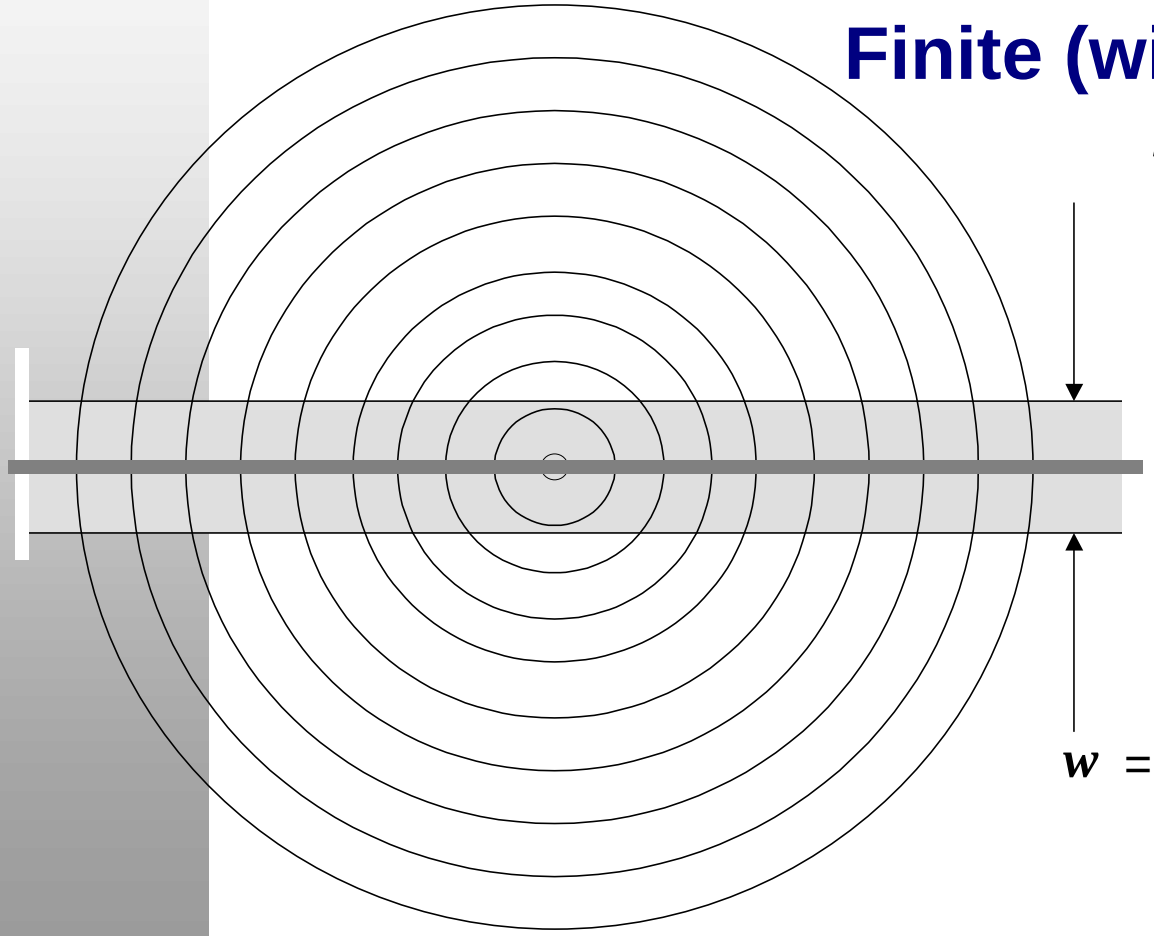


Finite (wide) point deposits

*Detector field of view with ideal point deposit superimposed on realistic point deposit with width **w**, area **a**.*

point deposit width = w ($a = p\, w^2/4$)

III. Revised Models/GG Holdup



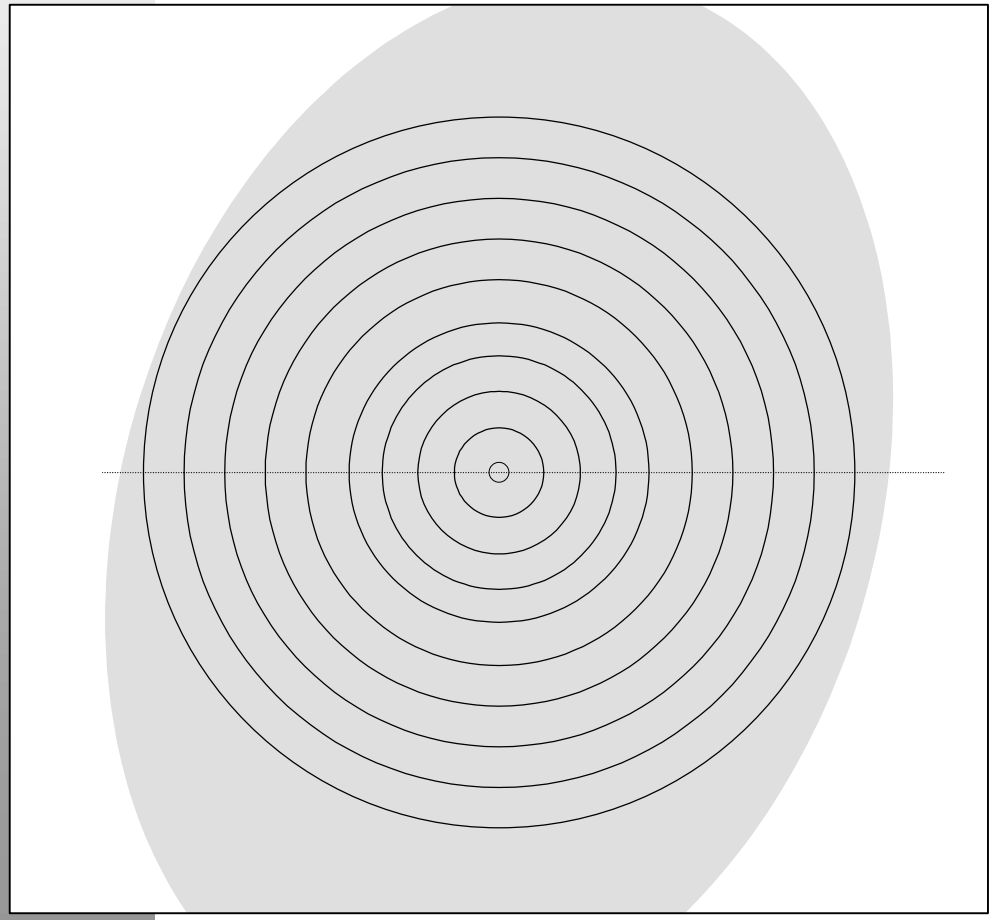
Finite (wide) line deposits

Detector field of view with ideal line deposit superimposed on realistic line deposit with width w.

w = line deposit width

III. Revised Models/GG Holdup

Area deposits are unaffected.



Detector field of view shown with ideal area deposit. (Area deposits are not subject to finite-source effects.)

III. Revised Models/GG Holdup

Example of a nearly ideal line geometry

At this measurement distance (40 cm), the vertical pipe appears as a narrow line in a relatively wide field of view.

III. Revised Models/GG Holdup

Example of a “finite” line source

At the same measurement distance (40 cm), the larger diameter horizontal pipe is a significant fraction of the width of the field of view.

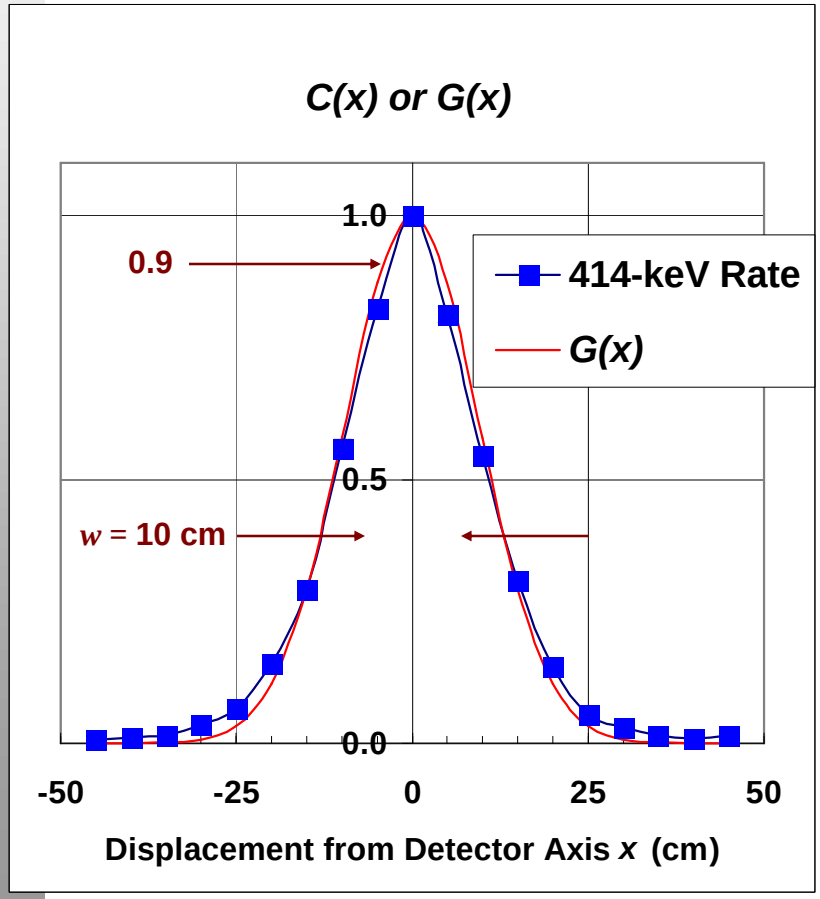
III. Revised Models/GG Holdup

Example of a wider “finite” line source

Often, measurement distance is limited by the equipment height. Duct width is always a significant fraction of the width of the field of view for many common ventilation ducts.

III. Revised Models/GG Holdup

Origin of the *finite-source effect*



*The normalized radial response of a collimated detector at r = 40 cm. The GG model requires the full peak response to a point or line. The average response to the illustrated point or line (**w** =10 cm) is 90% of peak. A negative bias results.*

III. Revised Models/GG Holdup

Choosing finite width w for point or line deposits

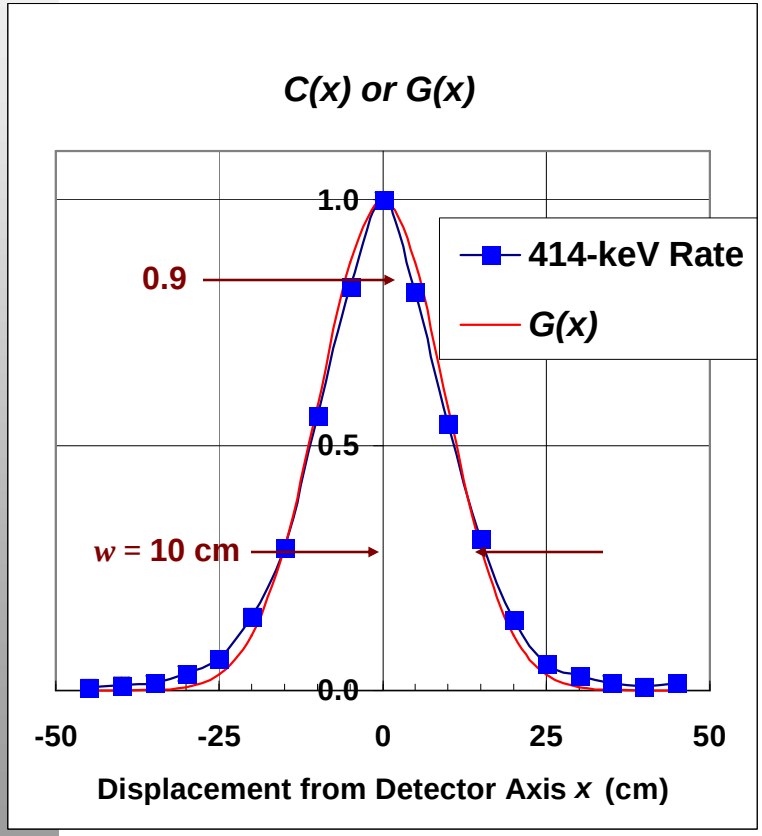
User chooses the width parameter w.

- Basis of choice:
  - i. Knowledge of equipment
  - ii. Knowledge of process
  - iii. Radiation measurements
- w is also used to correct for *self-attenuation*.
- Effects of uncertainty in w diminish self-consistently in the corrected* holdup measurement.

* for *finite-source* and *self-attenuation effects*

III. Revised Models/GG Holdup

Correcting for the revised-model *finite-source effect*



Correction Steps

1.) Fit radial response data. Gaussian fit, G(x), is shown here in red.

2.) Determine CF_{FINITE}*:

$$CF_{FINITE} = 2n \bullet [1 + G(w/2)]^{-n},$$

where

$n_{POINT} = 2$

and

$n_{LINE} = 1$.

* Multiplies uncorrected specific mass of point or line deposit

III. Revised Models/GG Holdup

Summary of the *finite-source correction*

- Only *one new* (empirical) parameter, ***w***.
- No additional measurements required.
  (Radial response data measured during the GG calibration.)
- Fitting and evaluation of CF_{FINITE} is simple.
- Applies to *all* generalized point and line deposits.
- Process is ***straightforward*** to automate.
- Removes negative bias from uncorrected results.
- Facility measurements of ^{239}Pu glove box holdup required values of CF_{FINITE} up to 1.25. These GG holdup results agreed with on-line neutron coincidence measurements.

III. Revised Models/GG Holdup

Ideal holdup deposit has no *self-attenuation*

Generalized-geometry model of holdup ….

…. assumes non-attenuating point, line and area deposits.

All holdup deposits attenuate their own gamma rays.

- Self-attenuation is greater for thicker deposits.
- Self-attenuation is greater for lower-energy gamma-rays.
- **Ignoring *self-attenuation* causes negative bias in holdup result.**

III. Revised Models/GG Holdup

***Self-attenuation* correction also uses width *w*.**

- The measured **GG specific holdup mass** is the ***isotope***
  i. **mass** – for a point deposit.
  ii. **mass/length** – for a line deposit.
  iii. **mass/area** or $(\rho x)_{MEAS}$ – for an area deposit.

- Correcting for *self attenuation* requires knowing the measured *areal density* of the **element (E)**, $(\rho x)_{MEAS,E}$.

- All GG results for point and line holdup deposits can be converted to $(\rho x)_{MEAS,E}$ using the **isotope enrichment** e **and parameter *w***.

III. Revised Models/GG Holdup

Self-attenuation correction **algorithm uses** $(\rho x)_{MEAS,E}$

The true areal density (ρx) of the holdup deposit

– *i.e.*, corrected for self attenuation –

is a simple function of $(\rho x)_{MEAS,E}$.

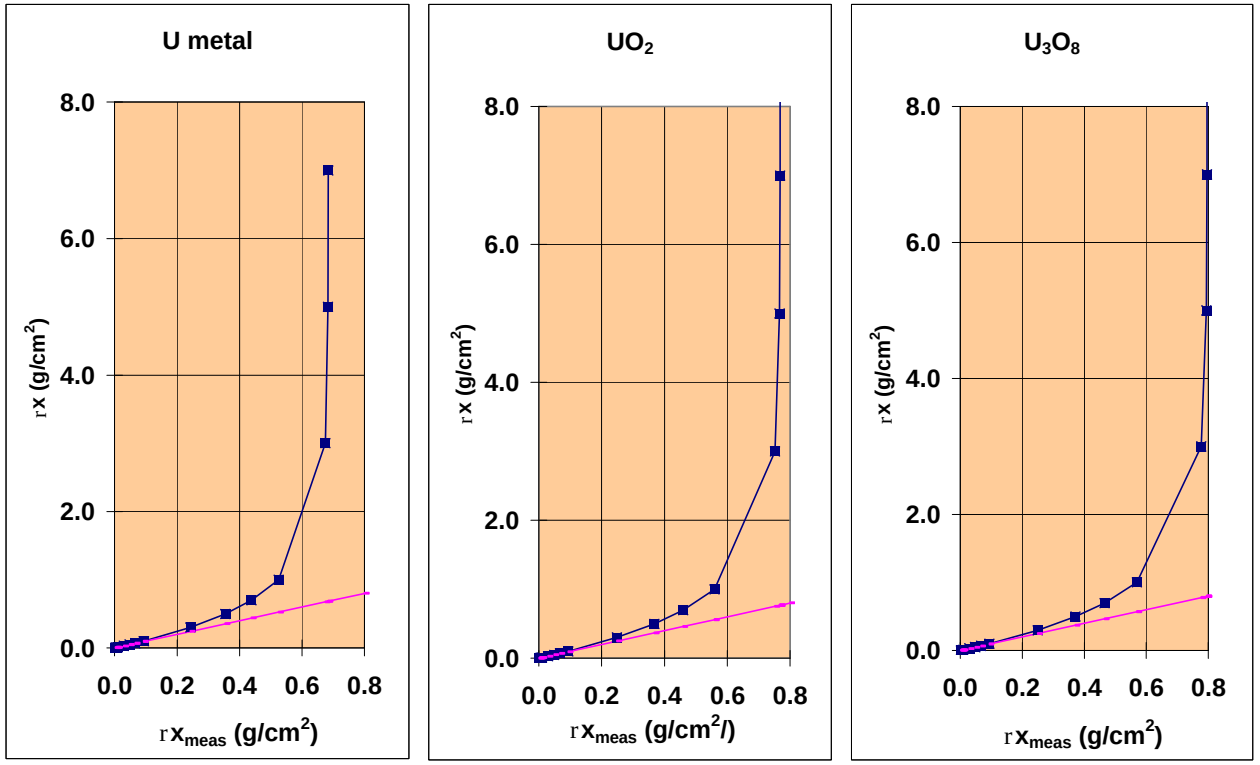
$$(\rho x) = - \varepsilon(\ln[1 - \mu(\rho x)_{MEAS,E}]) / \mu \qquad \text{[Eq.1]}$$

where

μ is the deposit mass attenuation coefficient.

III. Revised Models/GG Holdup

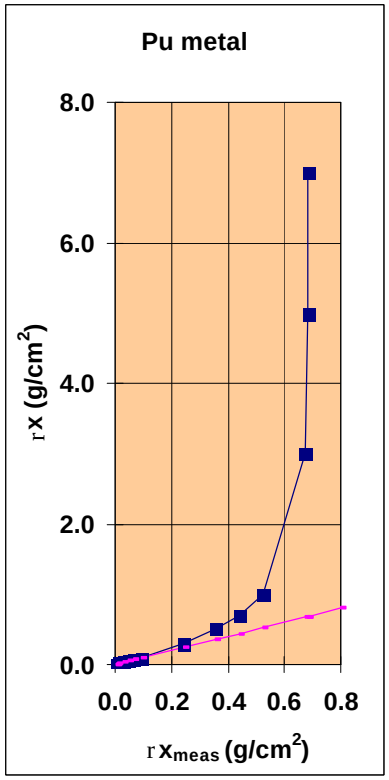
Plot Eq. 1 for uranium *self-attenuation correction.*



True vs measured areal density (g/cm^2) of uranium for 186-keV gamma rays from uranium metal, UO_2 & U_3O_8.

III. Revised Models/GG Holdup

Plot Eq. 1 for plutonium *self-attenuation correction.*



NOTE: The true areal density ρx cannot be determined if the measured value is so large that $\mu(\rho x)_{MEAS}$ approaches 1. (TEST EACH MEASUREMENT FOR "INFINITE THICKNESS"!) The ρx for holdup deposits <u>rarely</u> exceeds 0.3 g/cm^2. Typically (for 186-414 keV):

$$\mu(\rho x)_{MEAS} < 0.5$$

True vs measured areal density (g/cm^2) of plutonium for 414-keV gamma rays from plutonium metal.

III. Revised Models/GG Holdup

***Self-attenuation correction* for point deposit requires determining $(\rho x)_{MEAS,E}$**

For a point holdup deposit, get $(\rho x)_{MEAS,E}$ from the **GG specific point mass**, ε and w:

- **isotope mass** $\div \varepsilon$ = element mass
- element mass $\div$ point area = element areal density
- point deposit area = $\pi w^2/4$

Therefore, for a point holdup deposit:

$$(\rho x)_{MEAS,E} = \textbf{GGH specific mass} \div (\varepsilon \cdot \rho w^2/4)$$

III. Revised Models/GG Holdup

***Self-attenuation correction* for line deposit requires determining $(\mathrm{rx})_{MEAS,E}$**

For a line holdup deposit, get $(\rho x)_{MEAS,E}$ from the **GG specific line mass**, ε and w:

- **isotope mass/length** $\div \varepsilon$ = element mass/length
- element mass/length $\div$ line width = element areal density
- line deposit width = w

Therefore, for a line holdup deposit:

$$(\rho x)_{MEAS,E} = \textbf{GGH specific mass} \div (\varepsilon \cdot w)$$

III. Revised Models/GG Holdup

***Self-attenuation correction* for area deposit requires determining $(\rho x)_{MEAS,E}$**

For an area holdup deposit, get $(\rho x)_{MEAS,E}$ from the **GG specific area mass** and ε (***w** is not needed)*:

- **isotope mass/area** $\div \varepsilon$ = element mass/area
- element mass/area = element areal density

Therefore, for an area holdup deposit:

$$(\rho x)_{MEAS,E} = \textbf{GGH specific mass} \div \varepsilon$$

III. Revised Models/GG Holdup

Correcting $(\rho x)_{MEAS,E}$ for *self-attenuation*

Correction Steps

1.) Obtain $(\rho x)_{MEAS,E}$ using ε and w as in 3 previous slides.

2.) Use [Eq. 1] to get $(\rho x)_E$ from $(\rho x)_{MEAS,E}$.*

3.) Convert back to the **(true) isotope** areal density.

$$(\rho x) = \varepsilon \bullet (\rho x)_E$$

4.) Convert back to **corrected (true) GGH specific mass**

Point:	***true isotope mass***	$= (\rho w^2/4) \bullet (\rho x)$
Line:	***true isotope mass/length***	$= w \bullet (\rho x)$
Area:	***true isotope mass/area***	$= (\rho x)$

* Step 2 is actually the self-attenuation correction step.

III. Revised Models/GG Holdup

Summary (like slide 32) of *self-attenuation correction*

- Uses the *same* (empirical) width parameter, w.
- No additional measurements are required.
- All algorithms are analytical and simple.
- Applies to *all* generalized point, line and area deposits.
- Process is **straightforward** to automate.
- Removes negative bias from uncorrected results.
- Screens for “infinitely thick” deposits.
- Facility measurements of ^{239}Pu glove box holdup required self-attenuation corrections up to 1.11. These GG holdup results agreed with on-line neutron coincidence measurements.

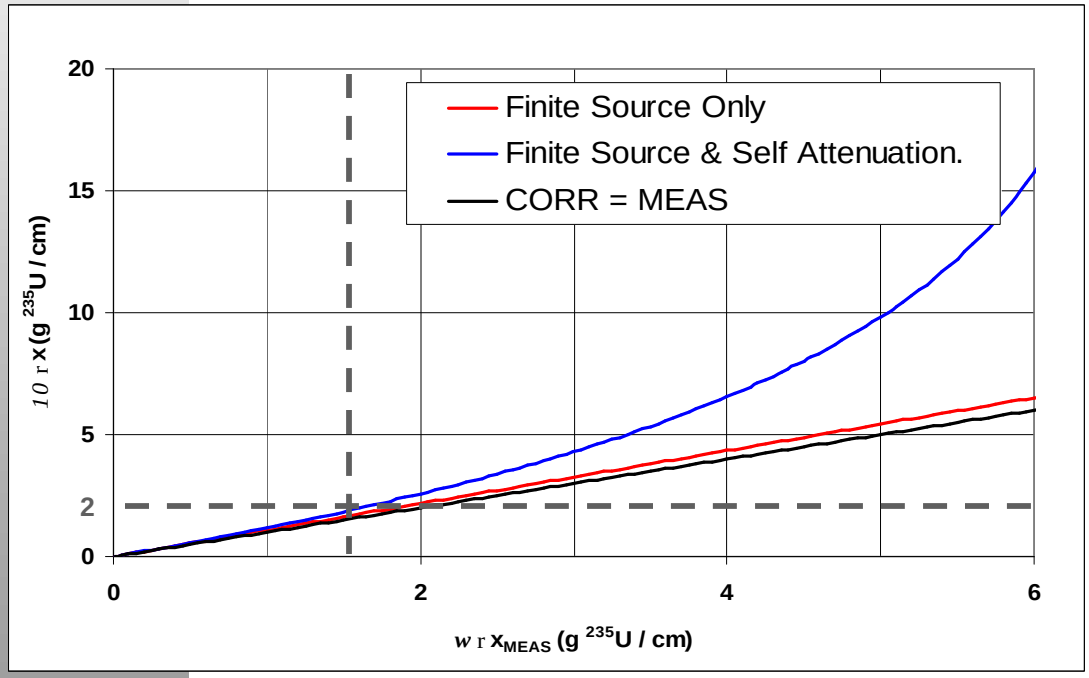
IV. Results/GG Holdup

Effect of uncertainty in w is self consistently minimized in using revised models.

- *Finite-source & self-attenuation corrections both* rely on w.
- If w is overestimated, the algorithms:
  1. over-correct for the finite-source effect.
  2. under-correct for the self-attenuation effect.
  3. in combination tend to mutually compensate for error.
- If w is underestimated, the algorithms:
  1. under-correct for the finite-source effect.
  2. over-correct for the self-attenuation effect.
  3. in combination mutually compensate for error.
- **It is most important to make both corrections.**

IV. Results/GG Holdup

Corrected vs. measured specific mass
93%-^{235}U line source, w_{true} = 10 cm



1. Self-attenuation correction dominates for thick deposits.

2. ρx (g ^{235}U/cm^2)

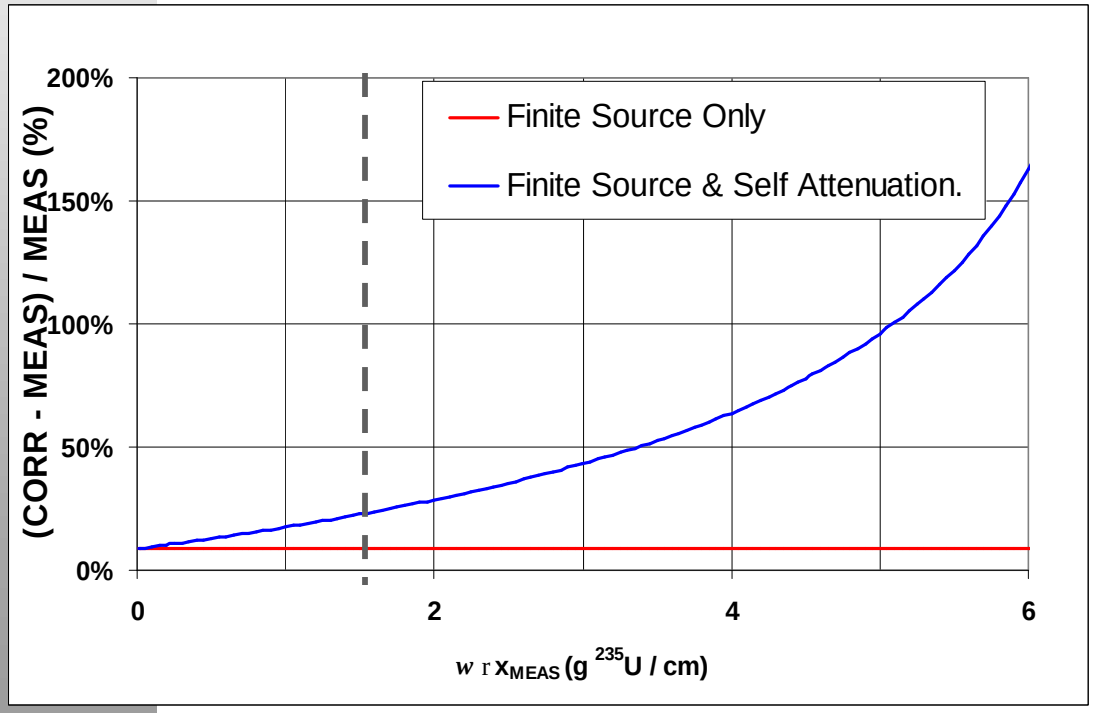
Measured	True
0.10	0.12
0.15	0.19
0.20	0.26

3. Most holdup is below the dashed lines (– – – – -).

4. Uncorrected (black) result is always less than corrected (biased negative).

IV. Results/GG Holdup

$(Corr–Meas)_{rel}$ vs. measured specific mass 93%-^{235}U line source, w_{true} = 10 cm.



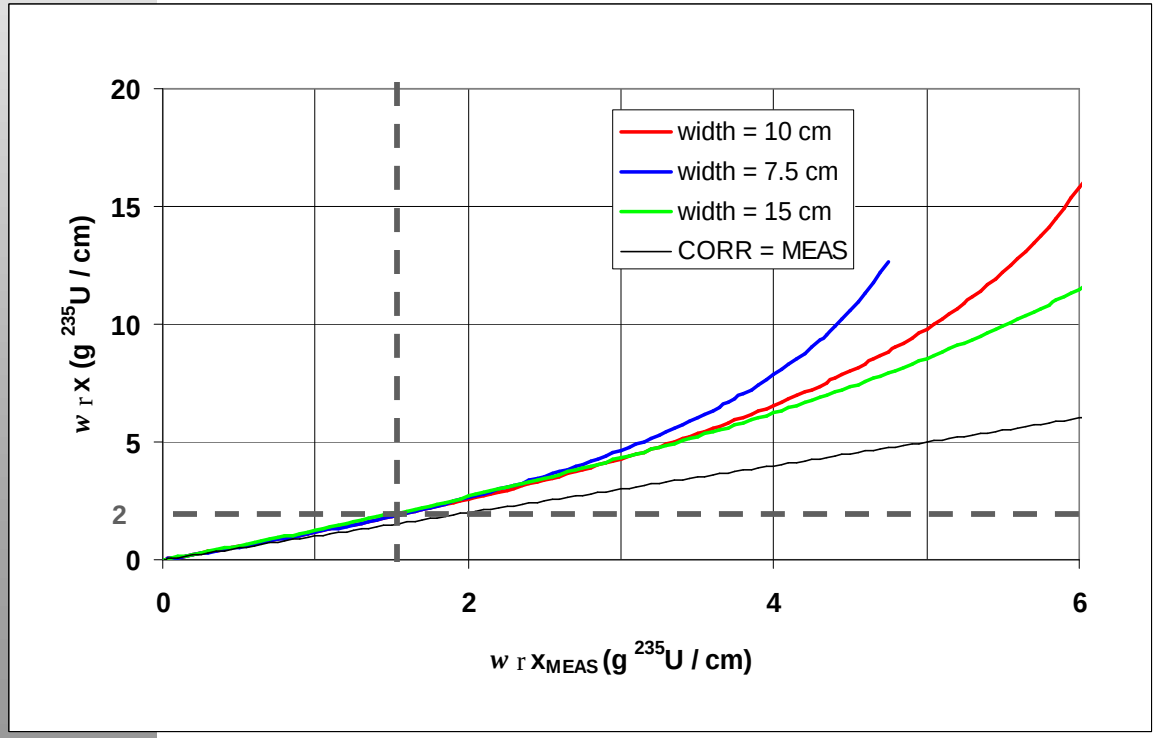
1. Corrections to measured ρx can exceed 20% in the thickness range of most holdup:
   ρx (g ^{235}U/cm^2) < 0.2

2. Note that magnitudes of corrections for the finite-source and self-attenuation effects are comparable in the range of most holdup.

IV. Results/GG Holdup

Corrected vs. measured specific mass
93%-^{235}U line source, w_{true} = 10 cm.

Assumed w = 7.5 & 15 cm. (The w_{true} is 1/3 larger & smaller than these.)



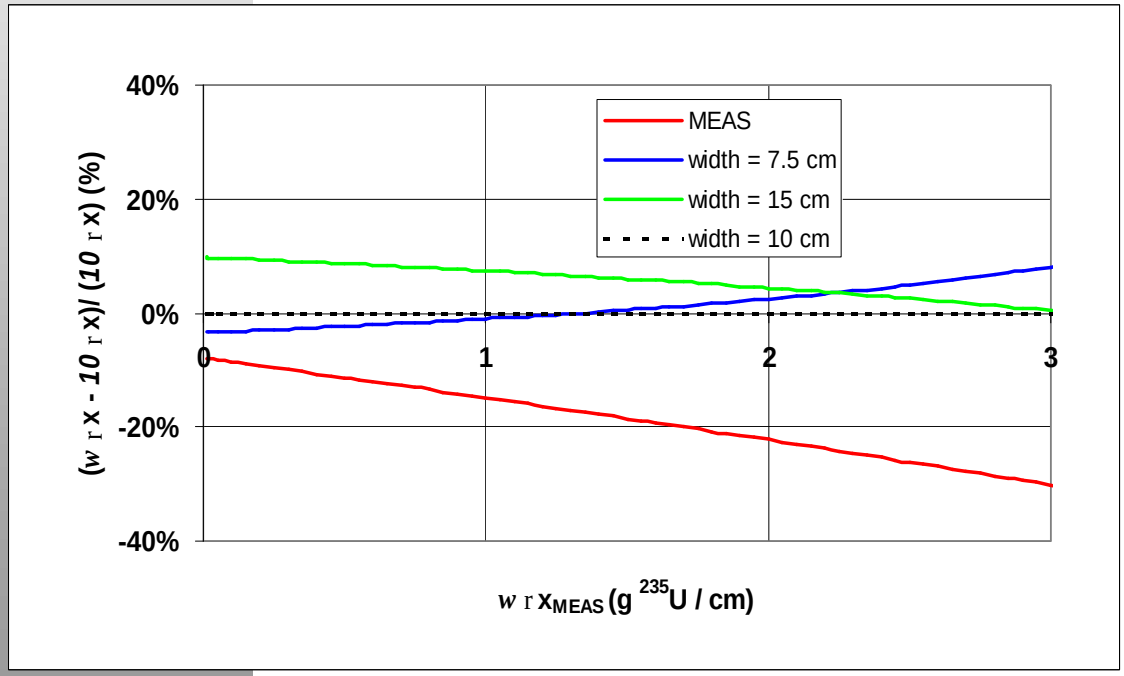
1. Above ρx = 0.5 g ^{235}U/cm^2 where self-attenuation governs corrections, the holdup result is affected greatly by an incorrect choice of ***w***.

2. See next slide for effects (of incorrect choice of ***w***) when ρx is in the range of most holdup.

IV. Results/GG Holdup

Corrected vs. measured specific mass
93%-^{235}U line source, w_{true} = 10 cm.

Assumed w = 7.5 & 15 cm. (The w_{true} is 1/3 larger & smaller than these.)



1. When ρx is in the range of holdup deposits (self-attenuation & finite-source effects are comparable) the effects tend to cancel with incorrect choice of ***w***.
2. Incorrect choice of ***w*** causes a + or - effect (which precludes bias) that is less than the negative bias incurred without the corrections.

IV. Results/GG Holdup

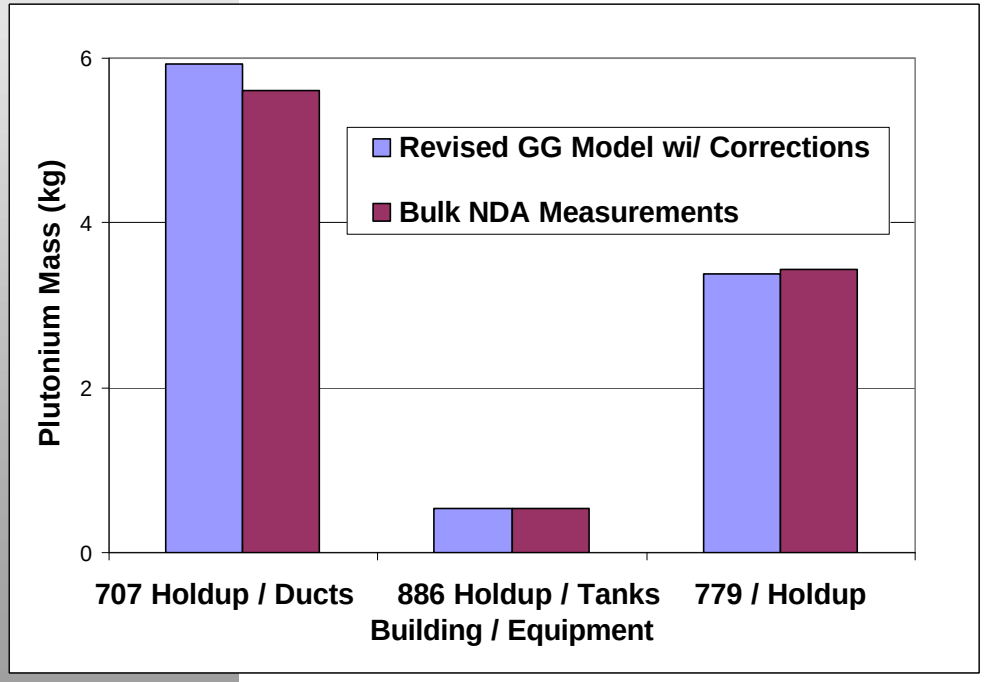
This approach for unbiased plantwide accountability of holdup is in use*.

- Assume revised GG holdup models: self-attenuating P, L or A deposits with finite dimensions.
- Always estimate w and perform both the finite-source and self-attenuation corrections.
- Always screen for infinite thickness and enforce cleanout for those occurrences.

*** Los Alamos, Oak Ridge, Rocky Flats**

IV. Results/GG Holdup

Verifying recent plantwide accountability measurements of holdup at Rocky Flats*



- GG holdup measurements performed facility-wide with the new corrections for finite-source effects and γ-ray self-attenuation are in progress at Rocky Flats ETS.
- The GG holdup results agree with reference values obtained by NDA measurements of materials from controlled cleanout of the facilities.
- Verifications of the GG holdup results at Rocky Flats are ongoing.

*** The verification data were provided by Frank Lamb of Rocky Flats ETS.**

V. Solution Measurements

Applications to *in-situ* measurements of solutions

- The point calibration standards for holdup are also used to calibrate *in-situ* measurements of solutions in tanks and columns of various dimensions.
- The experimental method for absolute calibration for solution measurements is identical to that for holdup measurements.
- *In-situ* solution measurements also rely heavily on models in combination with the measured calibration.
- Because of differences (see slides 5 & 10) between holdup and solutions, algorithms relating the calibrated response to the SNM concentration are much more complex for solutions vs. holdup.
- Methods developed at Los Alamos for measurements of solutions *in-situ* are used routinely at Y-12 for accountability of solution inventory. Results to ~10% (typ., 100 s) are unbiased.

V. Solution Measurements

Self-attenuation algorithm for *in-situ* solutions

- It is not possible to solve for ρy analytically using $(\rho y)_M$ because the **solvent** (S) also contributes. Compare the relationship:

$$(\rho y)_{MEAS} = \rho y \left\{ 1 - [\, \exp(-\mu_U \rho y) \,] [\, \exp(-m\rho y)_S \,] \right\}$$

$$\frac{}{(\mu_U \rho y) + (m\rho y)_S}$$

to Eq. 1 on slide 35. Therefore, the analysis for solution measurements is numerical.

- Variable acid molarity and partially full horizontal tanks increase the complexity of *in-situ* solution measurements.

V. Solution Measurements

Examples of *in-situ* solution inventory measurements

In-situ measurements of solutions in progress at Y-12 (4/00). Note:

- contact geometry.
- telescoping poles.
- backshields.
- Partially-full horizontal tanks require special treatment because ***y*** (slide 51) ≠ diameter.

V. Solution Measurements

Examples of *in-situ* solution inventory measurements

Measuring columns is more difficult than vertical tanks.

Solvent extraction (SE) columns, including the normal steel portions, may or may not contain *stator rods* and *sieve plates*.

V. Solution Measurements

Examples of *in-situ* solution inventory measurements

The SE aqueous/organic *interphase* has a discontinuous concentration that is difficult to locate by count rate. Visually: access is limited, solution and glass are murky, most equipment is opaque, *etc.* Note the use of flashlights.

V. Solution Measurements

Results of *in-situ* solution inventory measurements

Verification of Quantitative NDA Measurements with Reference Solutions in 10-cm Diameter Cylinders

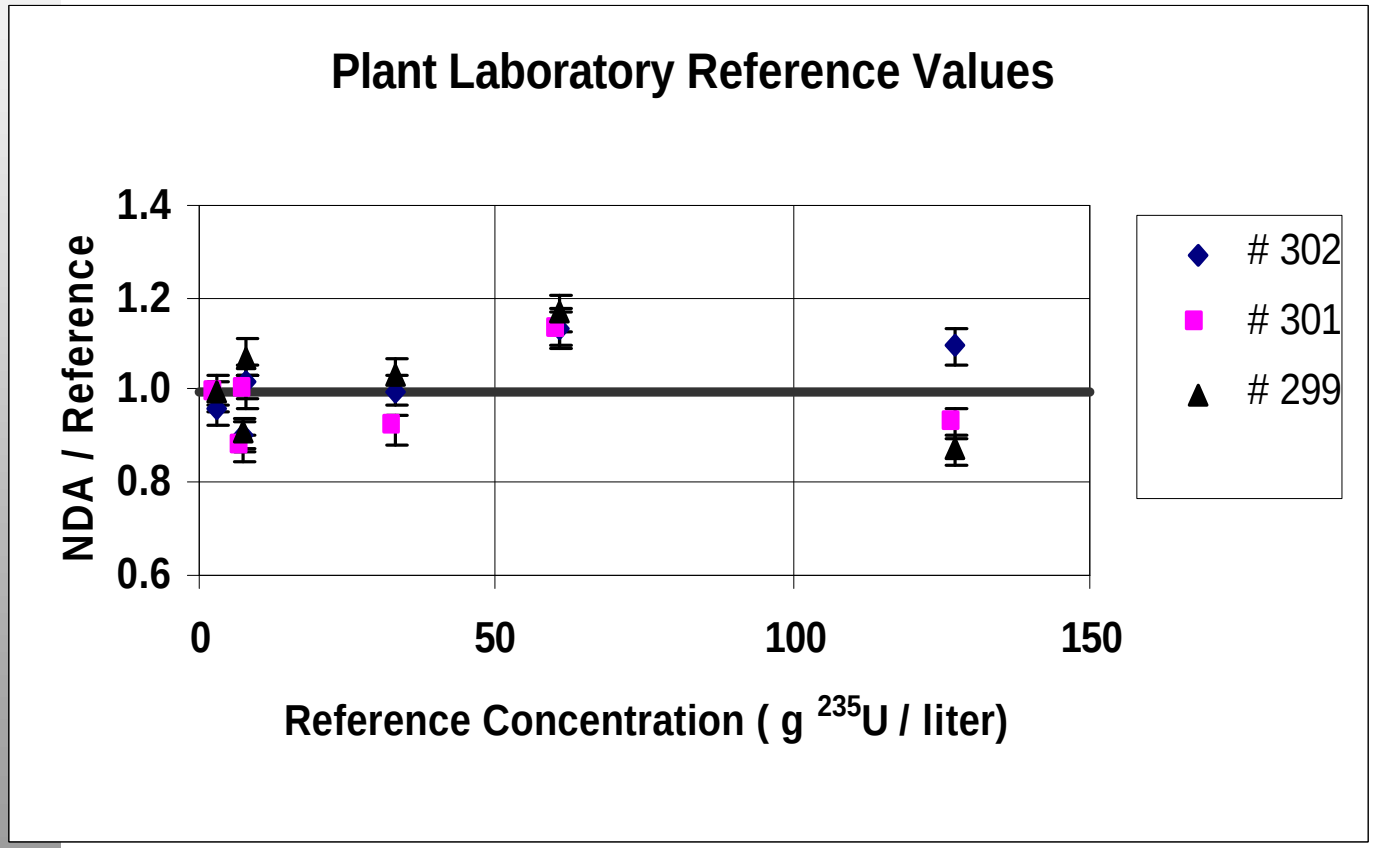
Solution ID	Type	Reference Values (g ^{235}U / liter)		Portable NDA Results (g ^{235}U / liter), % 1_S				NDA / Reference					
				MCA / Detector Serial Numbers				Plant Laboratory			9212 Laboratory		
		Plant Lab.[1]	9212 Lab.[2]	N302/HY599	N301/100063	N299/100059	% RSD	1	2	3	1	2	3
716-0084	OR	127.41	125.00	139	118	111	12.0%	1.09	0.93	0.87	1.12	0.94	0.89
				3.6%	7.2%	1.3%							
716-0083	AQ	60.55	60.03	68.8	68.3	70.7	1.8%	1.14	1.13	1.17	1.15	1.14	1.18
				1.8%	3.5%	2.5%							
715-8178	OR	32.83	32.92	32.8	30.0	33.8	6.1%	1.00	0.91	1.03	1.00	0.91	1.03
				3.0%	1.9%	4.2%							
715-9998	AQ	7.82	7.75	7.97	7.78	8.38	3.8%	1.02	0.99	1.07	1.03	1.00	1.08
				2.9%	5.5%	1.6%							
715-9974	OR	7.50	7.18	6.76	6.54	6.80	2.1%	0.90	0.87	0.91	0.94	0.91	0.95
				2.1%	3.7%	4.2%							
716-0248	AQ	3.09	2.93	2.97	3.05	3.09	2.0%	0.96	0.99	1.00	1.01	1.04	1.05
				2.9%	1.2%	2.3%							
59-7020	AQ	0.004	0.03	0.019	0.024	0.013	29.5%	4.75	6.00	3.25	0.63	0.80	0.43
				33.5%	29.5%	10.9%							

[1] Davies-Gray (g U/ g) plus density (g / cm^3) plus IDMS (g ^{235}U / g U)

[2] Transmission-corrected high-resolution gamma-ray analysis

V. Solution Measurements

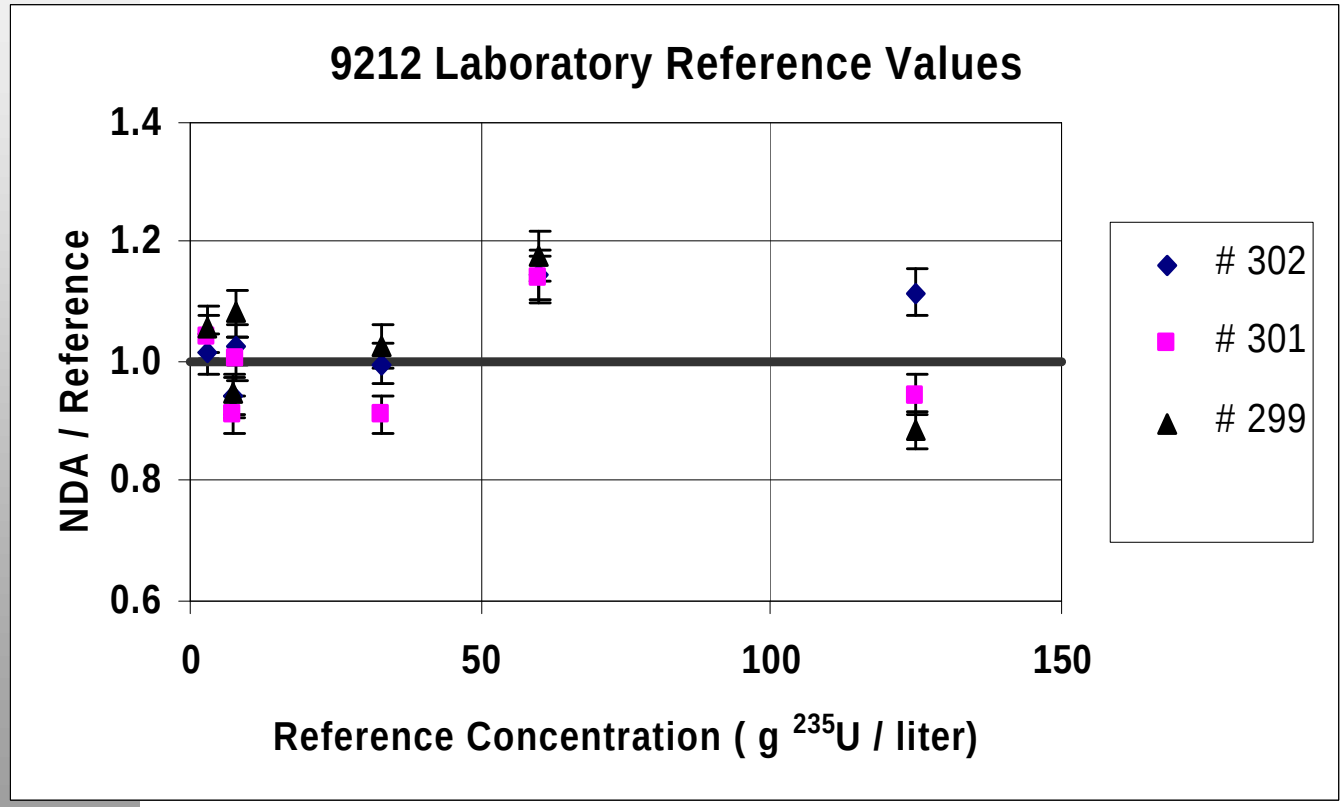
Results of *in-situ* solution inventory measurements



The ratio of measured-to-reference concentration of ^{235}U vs. reference value determined by destructive analysis.

V. Solution Measurements

Results of *in-situ* solution inventory measurements



The ratio (measured-to-reference) of ^{235}U concentrations vs. reference value determined by HR TC gamma-ray NDA.

VI. Discussion, Conclusions

References on the *In-Situ* Models

Revised GG Holdup Models for *In-Situ* Measurements

"Achieving Higher Accuracy in the Gamma-Ray Spectroscopic Assay of Holdup." P. A. Russo, T. R. Wenz, S. E. Smith and J. F. Harris. Los Alamos National Laboratory report LA-13699-MS, September 2000, 50 pp.

Solution Models for *In-Situ* Inventory Measurements

"*In-Situ* Measurement of Process Solution Inventory." P. A. Russo, T. R. Wenz, and K. A. Veal. Los Alamos National Laboratory report LA-UR-00-2470, June 2000, 60 pp.

VI. Discussion, Conclusions

Conclusions from *in-situ* results to date

- Although small (usually <10%) for most holdup, ignoring finite-source (FS) & self-attenuation (SA) effects introduces negative bias in every measurement. A 10% bias in the plant-wide holdup is a very large absolute quantity.
- Revised models (with FS & SA corrections) comply with needs: 1) Approach is generalized (easily automated). 2) Applies to very short measurements (5-15 s).
- Self-consistent implementation of revised GG holdup model minimizes bias in holdup measured at individual locations and plant-wide.
- Application to solutions: 1) Requires more complex models. 2) Indicates RSD of 10% with no apparent bias for 100-s counts.