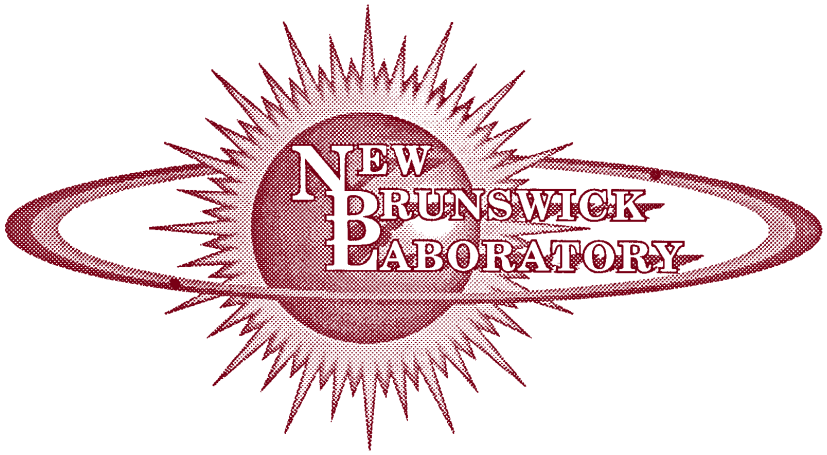
NBL-371
AUGUST 2002

MINUTES OF THE MEASUREMENT EVALUATION PROGRAM MEETING

URANIUM SAMPLE EXCHANGE
PLUTONIUM SAMPLE EXCHANGE
CALORIMETRY EXCHANGE



June 23, 2002

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 June 23, 2002

Safeguards Measurement Evaluation Program

8:30 AM	Sign in
9:00 AM	Welcome and Introductions (Jon Neuhoff, New Brunswick Laboratory)
9:15 AM	Programs and Initiatives of the Office of Plutonium, Uranium, and Special Materials Inventory (David Young, Department of Energy Headquarters, Office of Plutonium, Uranium, and Special Materials Inventory)
9:30 AM	Safeguards Measurement Evaluation Program: Overview (Jay Thompson, New Brunswick Laboratory)
9:45 AM	Interpretation of Measurement Evaluation Reports (Jay Thompson, New Brunswick Laboratory)
10:00 AM	Break
10:15 AM	Summary of 2001 Safeguards Measurement Evaluation Program Results (Jay Thompson, New Brunswick Laboratory)
10:30 AM	Overview of the Uranium by Davies-Gray Analysis at SRS (Kim Carter, Westinghouse Savannah River Company)
10:45 AM	IAEA, JSGO and JNC Interlaboratory Comparison Program (Mika Yoshida Sumi, Plutonium Fuel Center, Tokai Works, Japan Nuclear Cycle Development Institute)
11:00 AM	REIMEP and NUSIMEP: Present Status (Roger Wellum, IRMM Safeguards Co-ordinator, European Commission Joint Research Centre, Institute for Reference Materials and Measurements)
11:15 AM	Status of Reference Material Production (Jon Neuhoff, New Brunswick Laboratory)
11:30AM	Discussion and session wrap-up

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

U. S. Department of Energy
New Brunswick Laboratory
Measurement Evaluation Program
Agenda

Afternoon of June 23, 2002

Calorimetry Exchange Program

1:00 PM	Calorimetry Exchange Program: Overview (Jay Thompson, New Brunswick Laboratory)
1:15 PM	Annual Report Statistical Treatment (Dave Baran, New Brunswick Laboratory)
1:30 PM	Summary of 2001 Calorimetry Exchange Program Results (Dave Baran, New Brunswick Laboratory)
2:00 PM	Status of the Performance Demonstration Project (Bill Geist, Los Alamos National Laboratory/Larry Kayler, Rocky Flats Environmental Technology Site/Saleem Salaymeh, Westinghouse Savannah River Company)
2:30 PM	Break
2:45 PM	Transportable Calorimetry Laboratory (Cliff Rudy, Los Alamos National Laboratory)
3:00 PM	The Saga of the Hanford CalEx II Samples (Jay Thompson, New Brunswick Laboratory)
3:15 PM	Discussion
3:45 PM	Meeting wrap-up and closing remarks

NBL Measurement Evaluation Program Annual Meeting

June 23, 2002

Attendee List

Linda Baker	WSRC
Kimberly Carter	SRS
Linda Collins	Y-12
Susan Collins	WSRC/SRTC
Stephan Croft	Canberra
Norbert Ensslin	LANL
Bill Geist	LANL
Becky Guillen	LANL
Darryl Jackson	LANL
Larry Kayler	RFETS
Stewart Keeton	LLNL
Hideo Kobayashi	JNC
Ken Lewis	NBL
Steven Long	LANL
Olga Mafra	ABACC
Melanie May	DOE
Charles Petri	Hitech
Kenneth Raschke	LLNL
Wendy Rhodes	DOE
Stephan Richter	NBL
Cliff Rudy	LANL
Saleem Salaymeh	WSRC/SRTC
Jim Stewart	LANL
Mika Sumi	JNC
Naoki Surugaya	JNC
Jay Thompson	NBL
Stephan Vogt	IAEA
Roger Wellum	IRMM
Terri Welsh	PTH-Hanford
David Young	DOE

Since Last We Met...

- September 11th
- START-I Reductions Completed
- U.S. Withdrawal from the ABM Treaty
- Strategic Offensive Reductions Treaty
- Russia-NATO Link
- Russian Withdrawal from the START-II Treaty
- Administration Review of U.S. Nonproliferation Programs in Russia
- U.S. Nuclear Posture Review
- HEU Deal Renewal
- Portsmouth Gaseous Diffusion Plant Status
- Yucca Mountain Recommended as HLW Repository

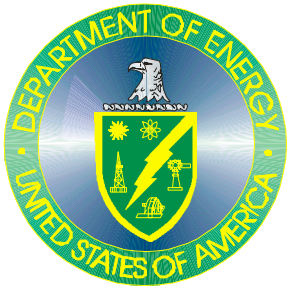
Since Last We Met...

- Pu Disposition
  - 34 metric tons of weapons-grade Pu in Russia (MOX)
  - 34 metric tons of weapons grade Pu in U.S. (+ 18 metric tons of non-weapons grade Pu) - MOX
  - RFETS - SRS Shipments
- Additional Protocol to U.S.-IAEA Nuclear Safeguards Agreement
- India-Pakistan Sanctions Waived; increased tensions
- "Axis of Evil"
  - Iraq
    - Revamped sanctions
    - Increased tensions
  - Iran
    - Bushehr/
    - Russian assistance
  - North Korea
- Nuclear Threat Initiative

In This “New World”...

Highly accurate and precise nuclear material measurements, made with traceable calibration standards and analytical methods demonstrated to be in control (through means such as interlaboratory comparison programs), are critical to national security and nuclear nonproliferation efforts.

Programs and Initiatives of the Office of Plutonium, Uranium, and Special Materials Inventory



David A. Young
(301) 903-0498

Office of Plutonium, Uranium, And Special Materials Inventory, SO-62

David W. Crawford
Director

Peter Dessaules
Deputy Director

Socorro Holmes
Secretary

New Brunswick Laboratory	Nuclear Information Development and Support	Data Validation And Assessment	Inventory Management And Operations
Margaret Tolbert, Director Wanda Mitchell, Deputy Dir. NBL Staff	Susanne Furr Timothy Beckham Alice Morris/SAIC	Edward Reynolds Rosemary Hazard Carol Raeder David Young	Peter Dessaules Bill Benton Ruth Watkins Len Myers/SAIC

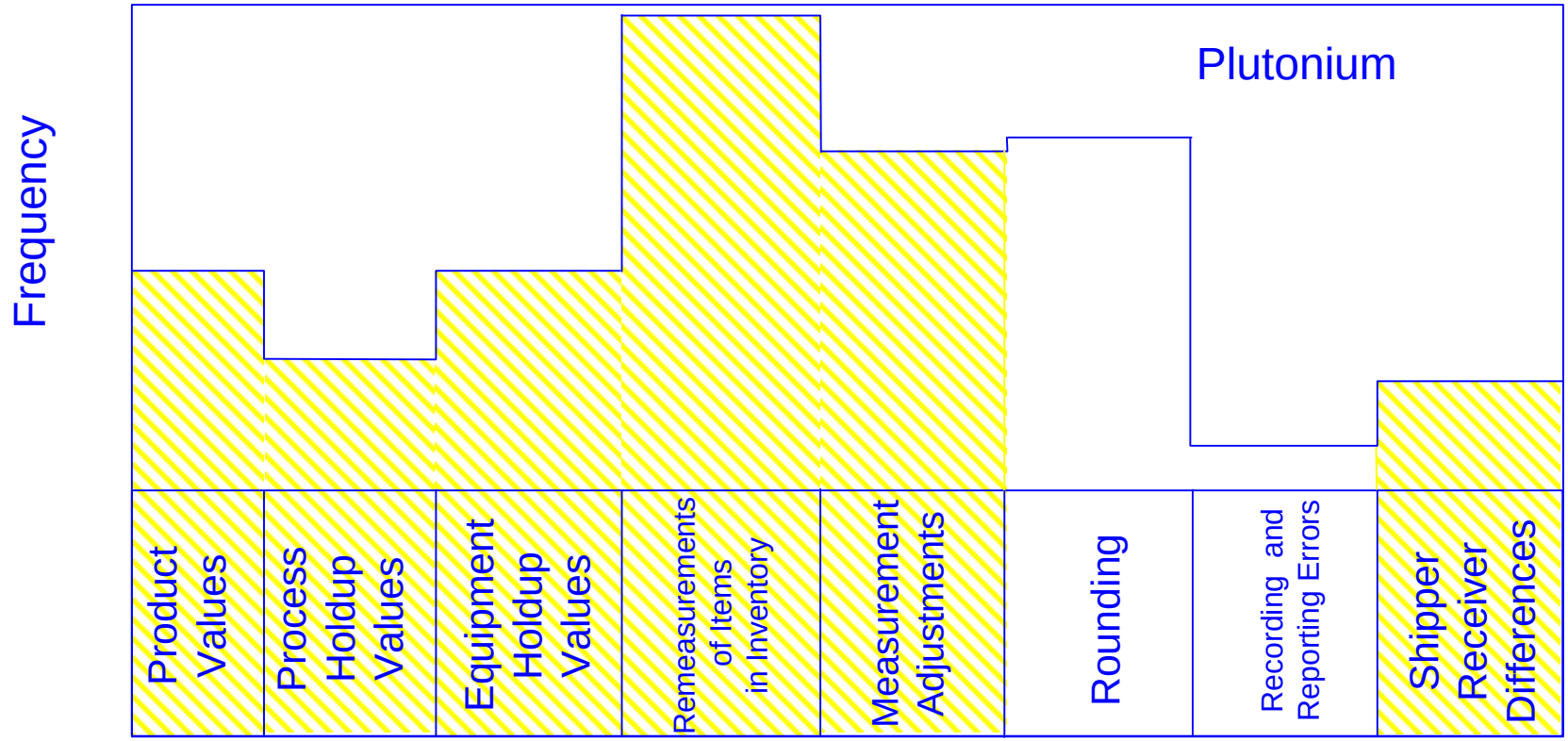
Cumulative Inventory Differences Trend Analysis

- Examine data for purpose of performing statistical trend analysis on cumulative inventory differences

- Objectives are to improve knowledge of accuracy of data in NMMSS and contributors to inventory uncertainty

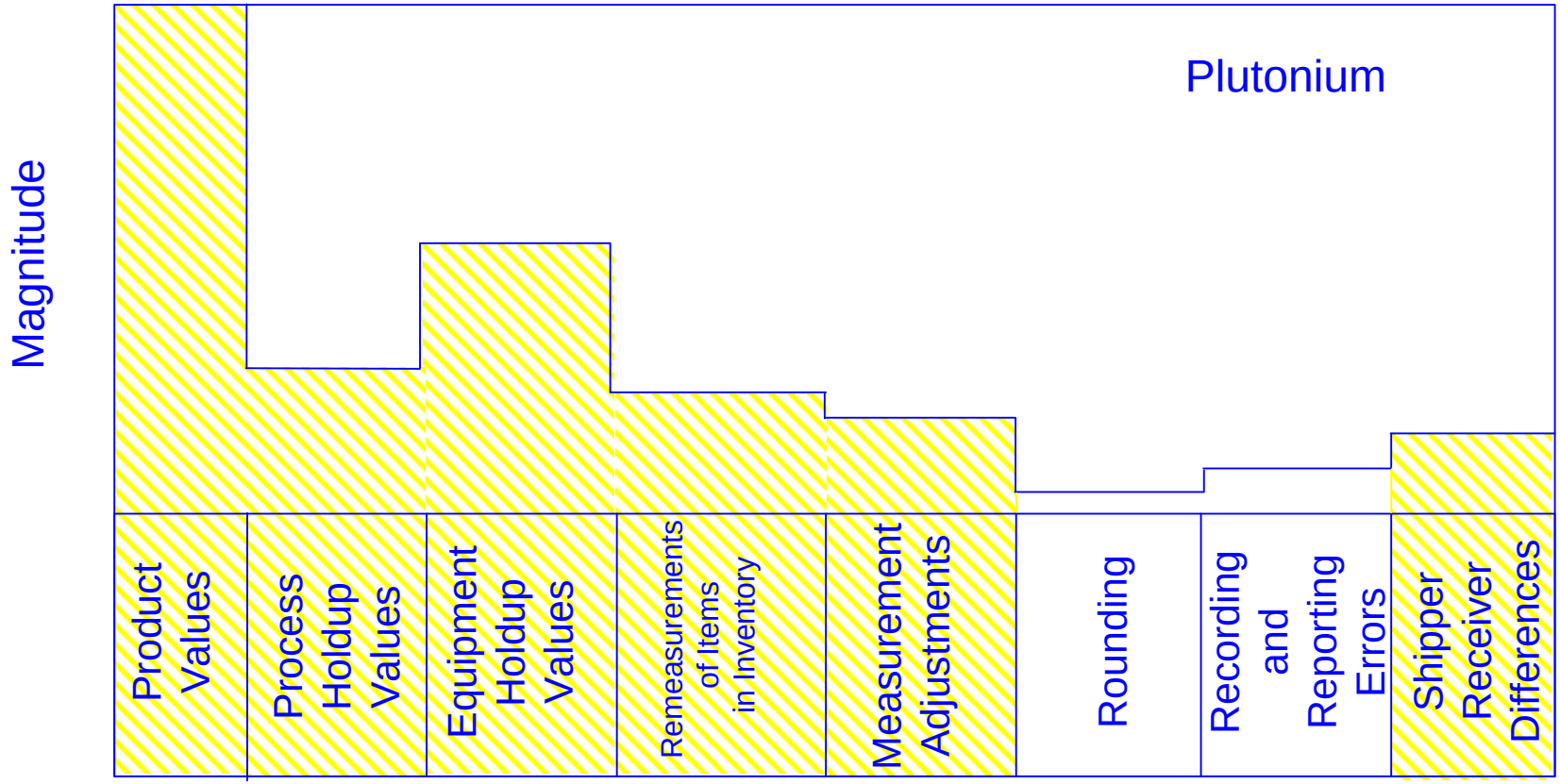
Distribution of Frequencies of Causes of IDs for Plutonium

(Shaded areas are measurements-driven)

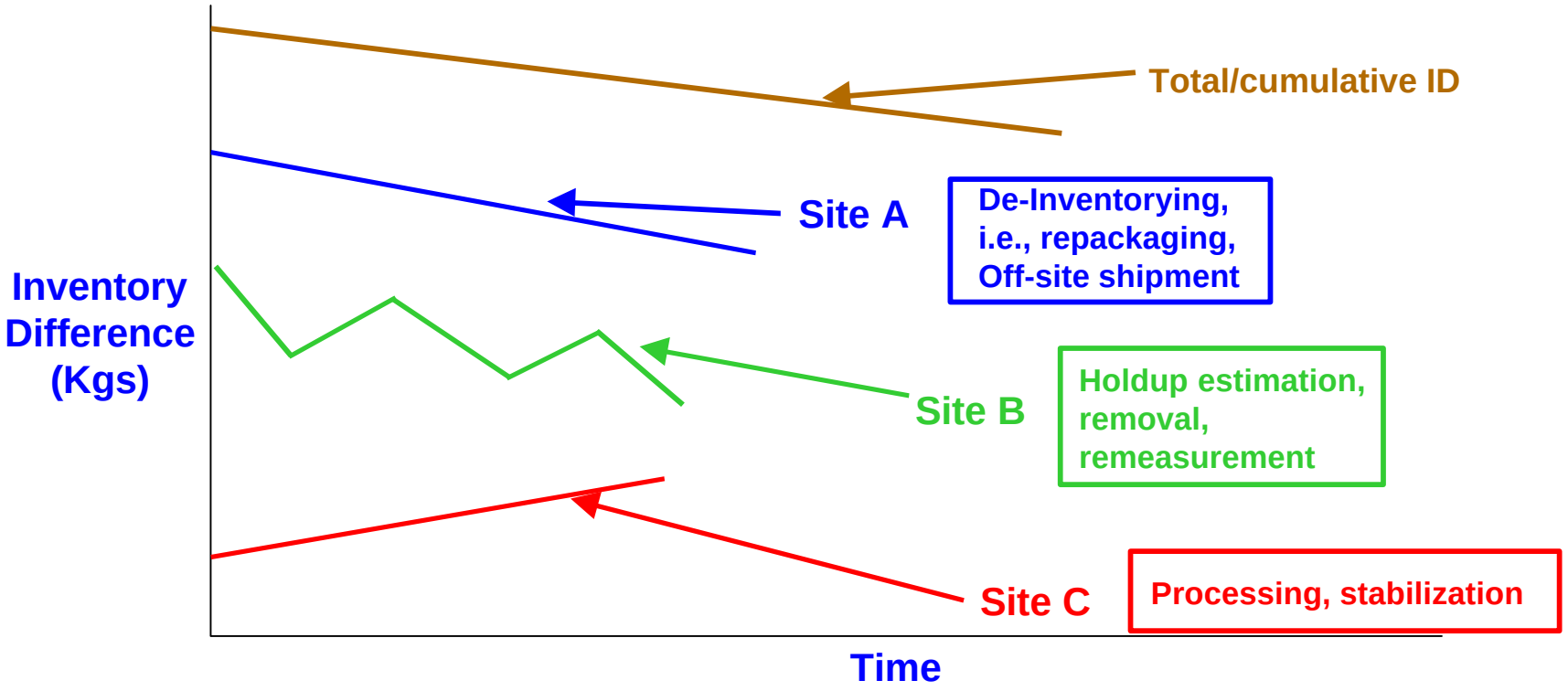


Distribution of Magnitudes of Inventory Differences for Plutonium

(Shaded areas are measurements-driven)



Projected Cumulative Inventory Difference Trends



Prioritization of Materials that Contributes Most to Inventory Uncertainty

(Hypothetical Response)

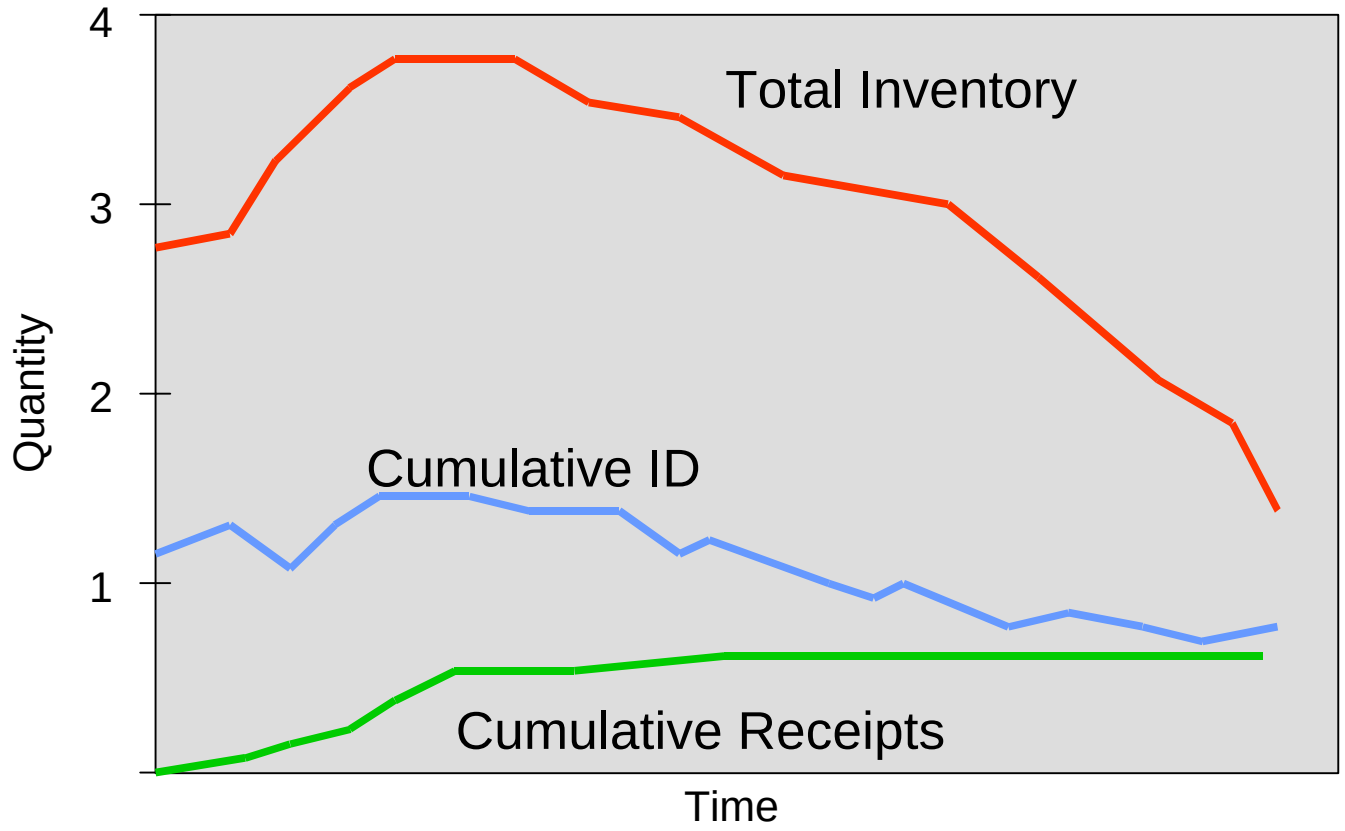
Priority	Material Form	Comments
1	Pu recyclable scrap with high Am Content	Contributes majority of uncertainty to MBA X according to POV.
2	Ash, Salts	May contribute as much as 5% to ID according to POV analysis of MBA Y when material is processed.
3	Pu sweepings	Contributes to uncertainty of MBA Z
4	In-Process oxide	Representative standards not available

Inventory Measurement Uncertainty Assessment

- Identify material strata and locations where greatest potential for measurement problems exists

- Objective is to prioritize “problem” materials so that resources can be better directed to address weaknesses in inventory quality

Hypothetical ID Trends as a Function of Total Inventory and Receipts



Routine inventory measurements quality assessments enable the DOE to better understand the quality and validity of nuclear material inventory information.

- characterize and quantify nuclear material inventories
- identify incomplete, inadequate, outdated and inaccessible data or data of questionable validity in a timely manner
- determine uncertainties in these inventories
- identify and prioritize major contributors to inventory uncertainties

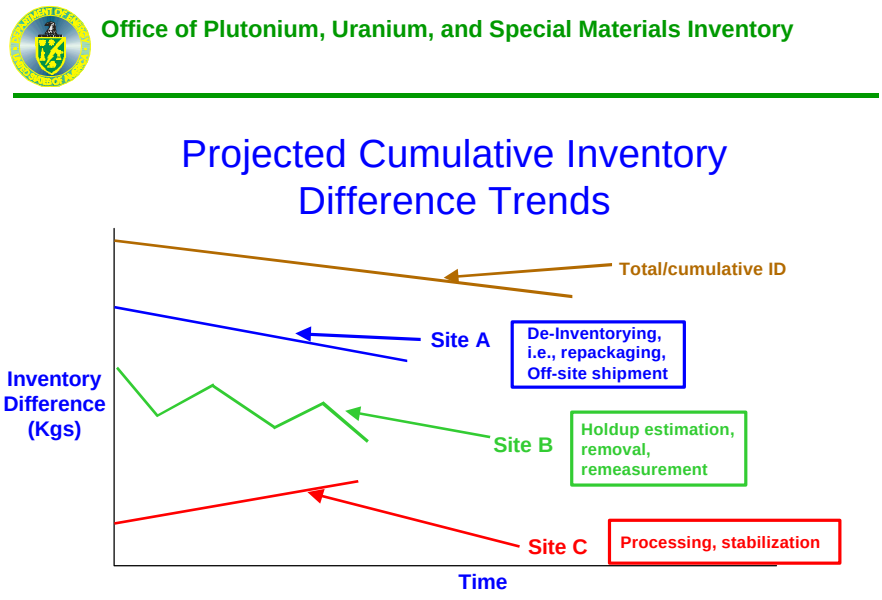
Nuclear Material Inventory Data Quality Initiatives

The Office of Plutonium, Uranium, and Special Materials Inventory (SO-62) is undertaking several initiatives for purposes of better understanding the quality of nuclear material inventory data and uncertainties around nuclear material inventories. Such inventory data issues have been the subject of various U.S. Government audits and are of media and Congressional interest. The intent is to examine various parameters such as inventory differences and measurement quality to determine where greater focus needs to be applied to improve confidence levels around inventories and address undesirable safeguards trends that can negatively impact on critical mission.

One inventory quality activity is the conduct of statistical trend analyses on cumulative inventory differences across the Department complex. These analyses are important in improving our knowledge of the quality of site inventory data in Nuclear Materials Management and Safeguards System or the NMMSS (the U.S. national nuclear materials database), as well as major contributors to total uncertainty around plutonium inventories.

The current cumulative inventory difference analyses are focused on three major plutonium sites: Rocky Flats, Hanford and Savannah River. Staff have found from recent visits to these sites that there is a great deal of interest in these analyses. In fact, some sites are already taking action to examine and routinely track cumulative inventory differences. What we hope to do at Headquarters is to work with sites in conducting their analyses and validating their data with that reported to the NMMSS.

Monitoring and analyzing the cumulative inventory difference provide a measure of assurance regarding the quality of inventory data. These analyses also serve as a forecasting tool for decision making regarding safeguards performance improvement. Through the analyses, inventory difference trends can be identified and corrected before they seriously impact on site missions. Our ultimate goal is not to validate site data against the NMMSS but integrate individual site data into one cross cutting trend analyses useful in validating site and NMMSS data and seeing how changes in the cumulative ID are related to specific site operations (Slide 1).



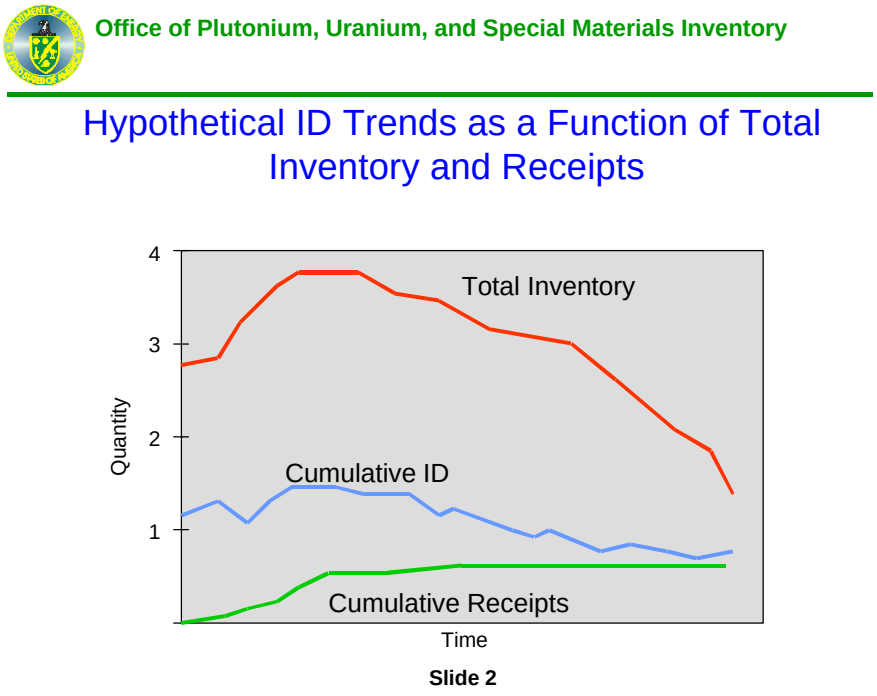
Slide 1

In reconstructing and mapping cumulative inventory difference data, my office is utilizing data in the 1996 Plutonium History Report as a baseline. We are compiling NMMSS data for performing these cumulative inventory data trend analyses. NMMSS data that has been acquired by our office from the three sites are (1) reported inventory difference, (2) total inventory, and (3) total shipments and receipts - all at the site level. These data are important in performing the trend analyses. We have some expectation and knowledge regarding historical inventory difference trends. We hope to predict future trends related to inventory differences. We also hope that any undesirable inventory difference trends are identified and corrected prior to impacting on major milestones. The product of these analyses will be more than just a report. We intend to produce a management tool for use in institutionalizing these analyses.

Slide 2 below shows how from examining cumulative inventory differences as a function of total inventory and receipts, several "flags" as to potential materials accountability issues are observed. Examples are:

1. Data disconnects or inconsistencies between site accountability systems and NMMSS, and

2. Abnormal trends related to increases or decreases in total inventories and materials received and the rate of change in the cumulative inventory differences.



Slide 2

A comprehensive inventory difference evaluation must include an understanding of inventory difference contributors. Once the inventory difference analysis work has been completed, we want to work backward in order to identify materials and isolate the major contributors to measurement problems to the inventory difference and the major contributors to the inventory differences**?** uncertainty. Without an estimate of the uncertainty, it is very difficult to lay a great deal of significance on the magnitude of the inventory difference. A lack of an estimate of the inventory difference is a problem results in the Department's inability to defend inventory differences.

We are working with EM sites in developing a list of problematic materials so that measurement resources can be applied effectively throughout the complex. From this we hope to obtain a prioritized list of nuclear material forms of which measurements have the greatest impact on uncertainties.

We know that major components or causes of inventory differences are measurements-related. In fact the largest contributor to inventory differences, as determined from past analyses, are actual accounting values or estimates ascribed various strata of the inventory, such as hold-up, input accountability values for plutonium in spent nuclear fuel and difficult to measure materials or discharges to waste. Identifying those materials and material streams where measurements are most challenging and drive the inventory differences is important not only to facilities and programs but also safeguards measurement technology developers.

Most nuclear materials accountability programs require that facilities summarize contributors to uncertainty broken down by material category and location. This is sometimes documented in MC&A plans. In fact, this materials categorization data is normally used by facilities to qualify measurement methods, perform variance propagation and analyze inventory and shipper/receiver differences. Our experience tells us that there are probably a half dozen or less material forms and strata that dominate the overall uncertainty. We want to know what they are (and so should sites!) in order enhance our understanding of inventory quality.

In summary, we recognize that by linking improved measurements and understanding of our nuclear materials inventory data with reductions in the cumulative inventory differences, we can hopefully raise the importance of improving the accountability and subsequently the quality of inventory data across the Department. The cumulative inventory difference and measurement assessments discussed above are intended to do this.

NEW BRUNSWICK LABORATORY

SAFEGUARDS MEASUREMENT EVALUATION PROGRAM

Jay M. Thompson

Measurement Evaluation Program Manager

(630) 252-2524

jay.thompson@ch.doe.gov

New Brunswick Laboratory

SAFEGUARDS MEASUREMENT EVALUATION PROGRAM - BASIC OPERATION

- Select, acquire, and characterize materials
- Distribute materials to participants
- Participants follow analysis plan and report results
- Perform statistical analysis
  - ⇒outlier tests
  - ⇒1-way ANOVA
- Report results
  - ⇒individual letter with statistical analysis and graph
  - ⇒comparative annual report

CHARACTERIZATION OF TEST MATERIALS

- Select and package material
- Design and follow statistical sampling plan
- Select analytical method
- Select quality control material(s)
- Design and follow analysis plan
- Statistically evaluate analytical data to assign reference value(s) and uncertainty

MATERIALS AVAILABLE

1. Uranyl nitrate solutions for U concentration
2. Uranyl nitrate solutions for ^{235}U enrichment
3. UO_2 pellets for U concentration and enrichment
4. UO_3 powder for U concentration
5. UF_6 (normal or low-enriched) solid for U concentration
6. UF_6 (low-enriched) solid for ^{235}U enrichment
7. Plutonium sulfate for isotopic abundances and IDMS

PARTICIPATING FACILITIES

- 4 DOE Contractor Laboratories
- 1 Federal Laboratory (NBL)
- 7 NRC Licensees*
- 9 International Laboratories
  - 1 Japanese
  - 2 Argentine
  - 6 Brazilian

PLUTONIUM ISOTOPIC EXCHANGE PARTICIPATING FACILITIES

- Los Alamos National Laboratory
- New Brunswick Laboratory
- Savannah River Site
- NMCC-TSC, Japan

BENEFITS OF PROGRAM

- Demonstrate comparability
- Evaluation of measurement systems
- Verification of achievement of performance criteria
- Validation of values used in propagation of variance
- Source of materials for performance tests
- Exchange of information

New Materials

- Concentrations
- New compounds
  - e.g., PuO_2

Requests

- Identify new materials
- Electronic submittal of analytical results
  - jay.thompson@ch.doe.gov
- Email NBL data evaluation reports
  - Volunteers?
- Updated distribution lists with full addresses

NEW BRUNSWICK LABORATORY

Interpretation of Measurement Evaluation Reports

Jay M. Thompson

SAFEGUARDS MEASUREMENT EVALUATION PROGRAM - BASIC OPERATION

- Select, acquire, and characterize materials
- Distribute materials to participants
- Participants follow analysis plan and report results
- **Perform statistical analysis**
  - ⇒**outlier tests**
  - ⇒**1-way ANOVA**
- **Report results**
  - ⇒**individual letter with statistical analysis and graph**
  - ⇒comparative annual report

Analysis Goals for SME Data

- Validate the data; evaluate for outliers
- Describe the data graphically
- Evaluate the data for day-to-day variations
- Estimate the accuracy and precision of the measurements
- Estimate the overall closeness of the data to the characterized values

General Analytical Scheme

- Each data set should consist of at least four measurements on each of at least two days

General Analytical Scheme

- This enables the estimation of:
  - variations in measurement uncertainties from day to day
  - measurement uncertainty within an analysis day
  - the overall measurement accuracy
- Comparison of the between day variation and the within day variation

Analysis Goals for SME Data

- **Validate the data**; evaluate for outliers
- Describe the data graphically
- Evaluate the data for day-to-day variations
- Estimate the accuracy and precision of the measurements
- Estimate the overall closeness of the data to the characterized values

DATA ENTRY AND REVIEW

- SME Program Manager receives program data and performs preliminary review
- Statistician enters the data into a FOXPRO database
- SME Program Manager compares the entered data with the submitted data
- Statistician runs outlier tests and one-way ANOVA
- SME Program Manager writes report to participating facility

Data Evaluation Report

Day to Day ANOVA analysis
Report for Laboratory (CODE)
UNH Solution – U Concentration
Davies-Gray Titration
Date of Report: May 13, 2002

Sample Number	Aliquant Number	Analysis Date	Reported %U	% Relative Difference	Analyst Code
98NU0075-067	1	04/18/02	1.00210	-0.0200	
98NU0074-092	1	04/18/02	1.00349	-0.0528	
98NU0075-067	2	04/18/02	1.00244	0.0140	
98NU0074-092	2	04/18/02	1.00376	-0.0259	
98NU0075-067	3	04/22/02	1.00245	0.0150	
98NU0074-092	3	04/22/02	1.00352	-0.0498	
98NU0075-067	4	04/22/02	1.00227	-0.0030	
98NU0074-092	4	04/22/02	1.00385	-0.0169	

Statistical Analysis Routine

- Calculate % Relative Difference (RD)

$$RD = \frac{R - C}{C} \times 100\%$$

  - *Where R = Value reported by laboratory*

    C = Characterized value of material

Analysis Goals for SME Data

- Validate the data; **evaluate for outliers**
- Describe the data graphically
- Evaluate the data for day-to-day variations
- Estimate the accuracy and precision of the measurements
- Estimate the overall closeness of the data to the characterized values

Reference for Outlier Detection

- Outliers in Statistical Data
  - Vic Barnett and Lewis, 1978
    - Sample kurtosis — p. 101
    - Grubbs T_1 and T_N statistics — p.93
    - Dixon's upper and lower R statistics — pp.97-99
    - Upper and lower outlier blocks, L_k — p. 96
    - Tietjen and Moore's E_k statistic — p. 102

Kurtosis Test

Test Statistic:

Let

$$\bar{x} = \sum_{i=1}^{n} \frac{x_i}{n} \quad \text{and} \quad s^2 = \sum_{i=1}^{n} \frac{(x_i - \bar{x})^2}{n-1}$$

Then the test statistic T is:

$$T = \frac{\sum_{i=1}^{n} (x_i - \bar{x})^4}{ns^4}$$

And is compared to tabulated values.

Kurtosis Test

KURTOSIS TEST FOR OUTLIERS
SCHEMATIC

RANK	ITEM	VALUE	OUTLIER?	STATISTICS
1	1	-0.052788	NO	|
2	3	-0.049800	NO	|
3	2	-0.025896	NO	+IQR+
4	5	-0.019954	NO	+-M-+
5	4	-0.016932	NO	+-M-+
6	8	-0.002993	NO	+IQR+
7	6	0.013968	NO	|
8	7	0.014966	NO	|

Grubbs and Dixon Tests

Grubbs Tests:

$$T1 = \frac{\bar{x} - x_{(1)}}{s}, \quad TN = \frac{x_{(n)} - \bar{x}}{s}$$

Dixon Tests:

$$Dixon(1) = \frac{x_{(2)} - x_{(1)}}{x_{(n)} - x_{(1)}}, \quad Dixon(N) = \frac{x_{(n)} - x_{(n-1)}}{x_{(n)} - x_{(1)}}$$

Grubbs and Dixon Tests

Grubbs:

T1 = 1.38 TN = 1.26

Dixon:

DIXON(1)= 0.0448 DIXON(N)= 0.0154

Other Outlier Tests

Upper and Lower Outlier Blocks

(for k highest or k lowest values)

$$\text{Let } \breve{x}_{n-k} = \sum_{i=k+1}^{n} \frac{x_{(i)}}{n-k} \text{ and } \hat{x}_{n-k} = \sum_{i=1}^{n-k} \frac{x_{(i)}}{n-k}$$

$$\text{then } L_k = \frac{\sum_{i=k+1}^{n} \left(x_{(i)} - \breve{x}_{n-k}\right)^2}{\sum_{i=1}^{n} \left(x_i - \bar{x}\right)^2} \textit{ and } U_k = \frac{\sum_{i=1}^{n-k} \left(x_{(i)} - \hat{x}_{n-k}\right)^2}{\sum_{i=1}^{n} \left(x_i - \bar{x}\right)^2}$$

Note: Small values indicate outliers.

Other Outlier Tests

Tietjen and Moore's E_k Statistics (for k outliers):

Let $r_j = \left|\bar{x} - x_j\right|$, i.e. the absolute deviation of the jth observation from the mean.

Let $\bar{r} = \sum_{i=1}^{n} \frac{r_i}{n}$ and $\bar{r}_{n-k} = \sum_{i=1}^{n-k} \frac{r_{(i)}}{n-k}$

Then the test statistic E_k is given by

$$E_k = \frac{\sum_{i=1}^{n-k} \left(r_{(i)} - \bar{r}_{n-k}\right)^2}{\sum_{i=1}^{n} \left(r_i - \bar{r}\right)^2}$$

Other Outlier Tests

Upper and lower outlier blocks:

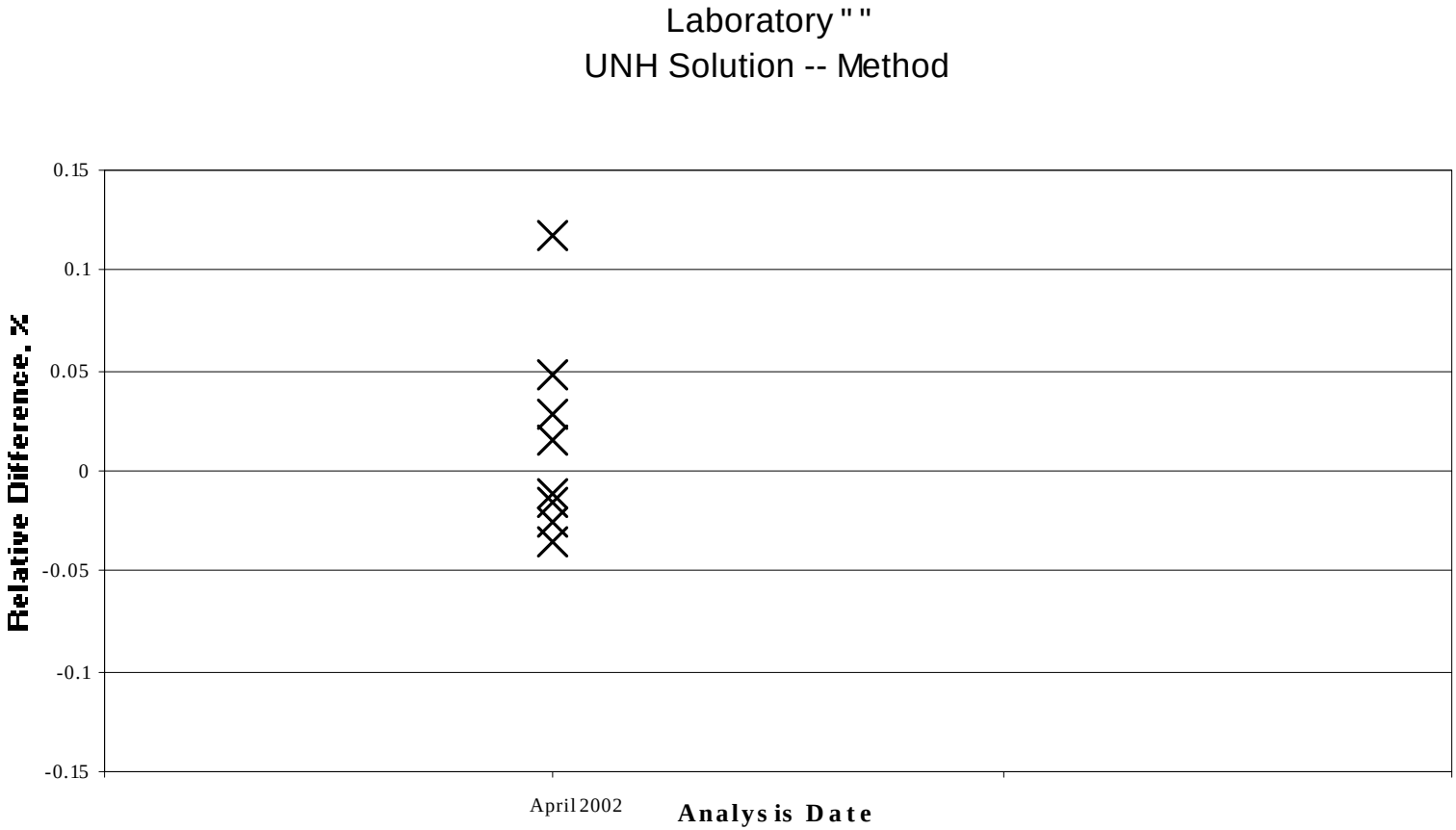
TEST OF 'K' LOWEST RESULTS, WHERE K=1, 4
L(K): 0.388 0.337 0.357 0.429
*
TEST OF 'K' HIGHEST RESULTS, WHERE K=1, 4
L(K): 0.460 0.413 0.445 0.519

Tietjen and Moore's E_k statistic

TEST OF 'K' MOST EXTREME RESULTS, WHERE K=1, 4
E(K): 0.691 0.502 0.221 0.061

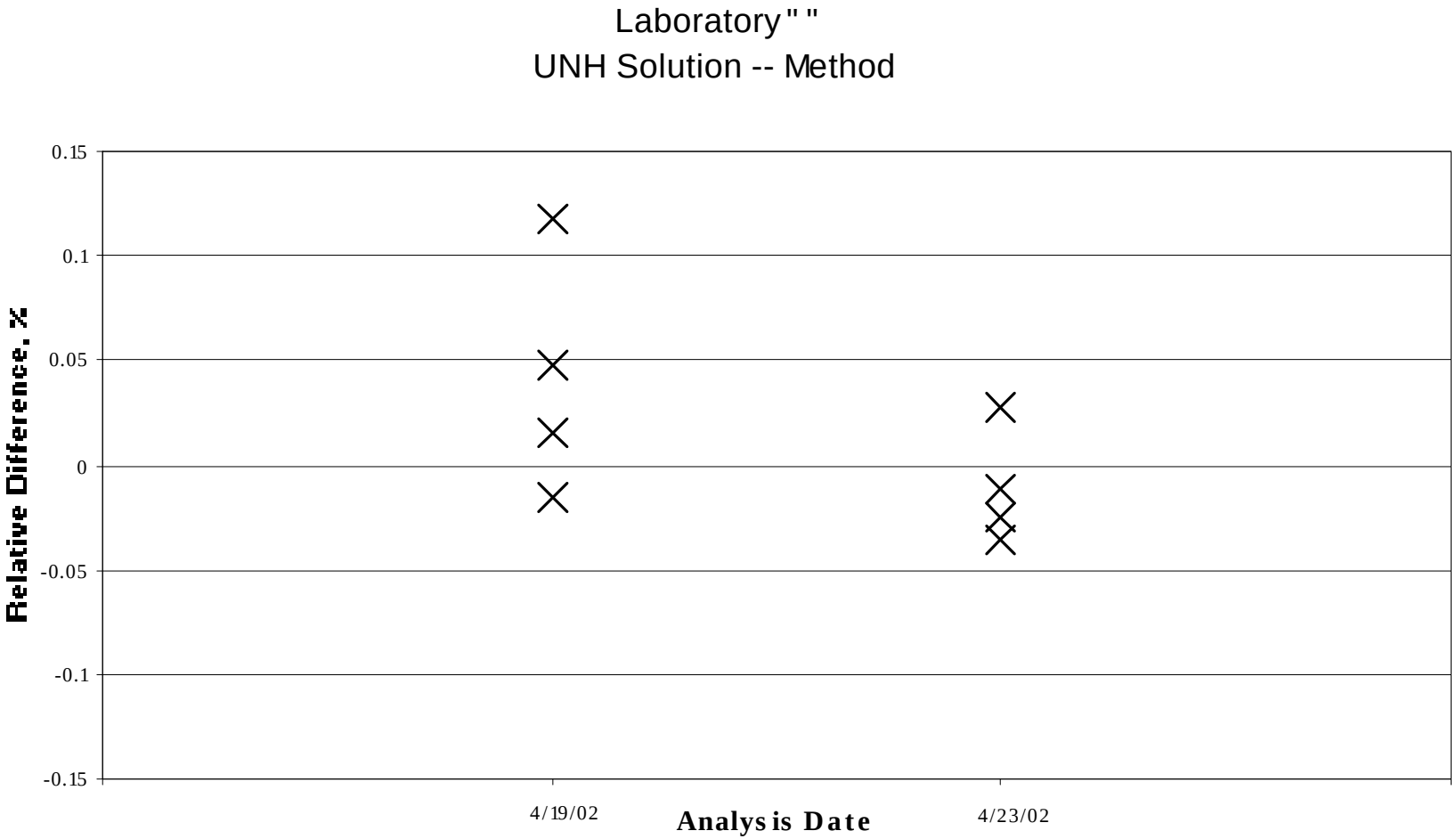
Declared Outlier

- For a point to be declared as an outlier, it should fail one test at the 99% level and another at the 95% level.
- Flagged outliers are reviewed prior to the final analysis



0000522.xls

New Brunswick Laboratory



0000522.xls

New Brunswick Laboratory

Analysis Goals for SME Data

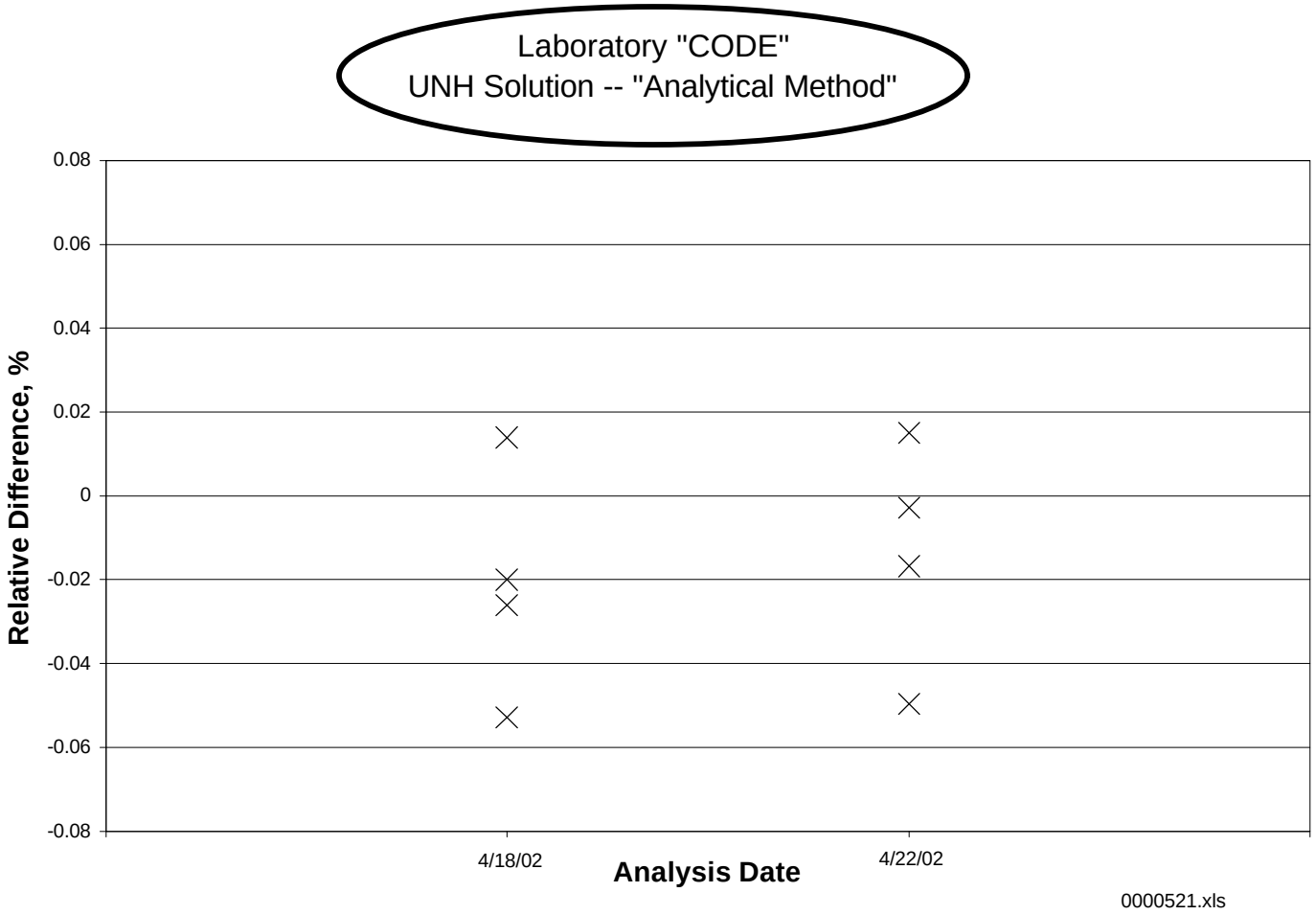
- Validate the data; evaluate for outliers
- **Describe the data graphically**
- Evaluate the data for day-to-day variations
- Estimate the accuracy and precision of the measurements
- Estimate the overall closeness of the data to the characterized values

Data Evaluation Report

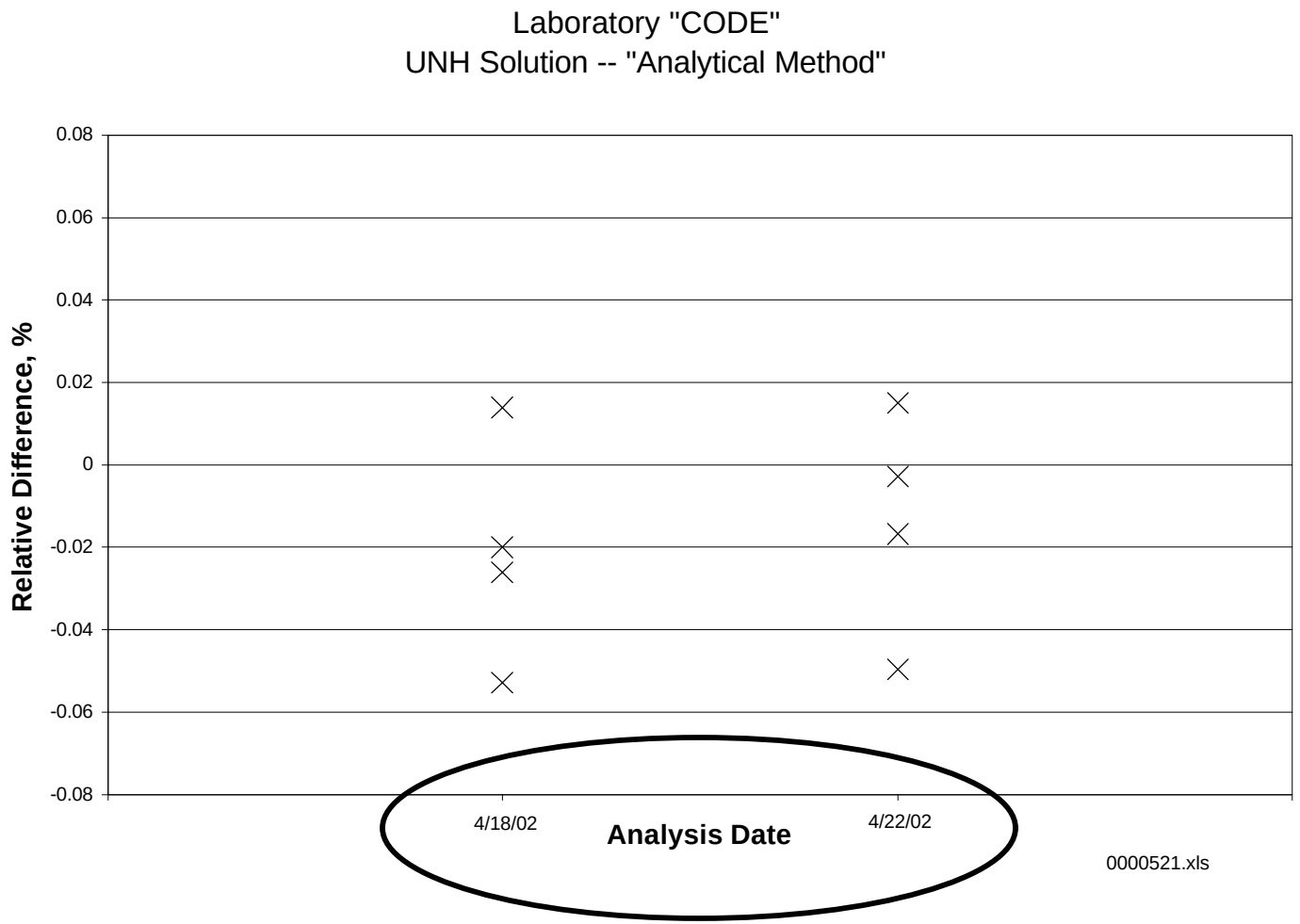
Day to Day ANOVA analysis
Report for Laboratory (CODE)
UNH Solution – U Concentration
Davies-Gray Titration
Date of Report: May 13, 2002

Sample Number	Aliquant Number	Analysis Date	Reported %U	% Relative Difference	Analyst Code
98NU0075-067	1	04/18/02	1.00210	-0.0200	
98NU0074-092	1	04/18/02	1.00349	-0.0528	
98NU0075-067	2	04/18/02	1.00244	0.0140	
98NU0074-092	2	04/18/02	1.00376	-0.0259	
98NU0075-067	3	04/22/02	1.00245	0.0150	
98NU0074-092	3	04/22/02	1.00352	-0.0498	
98NU0075-067	4	04/22/02	1.00227	-0.0030	
98NU0074-092	4	04/22/02	1.00385	-0.0169	

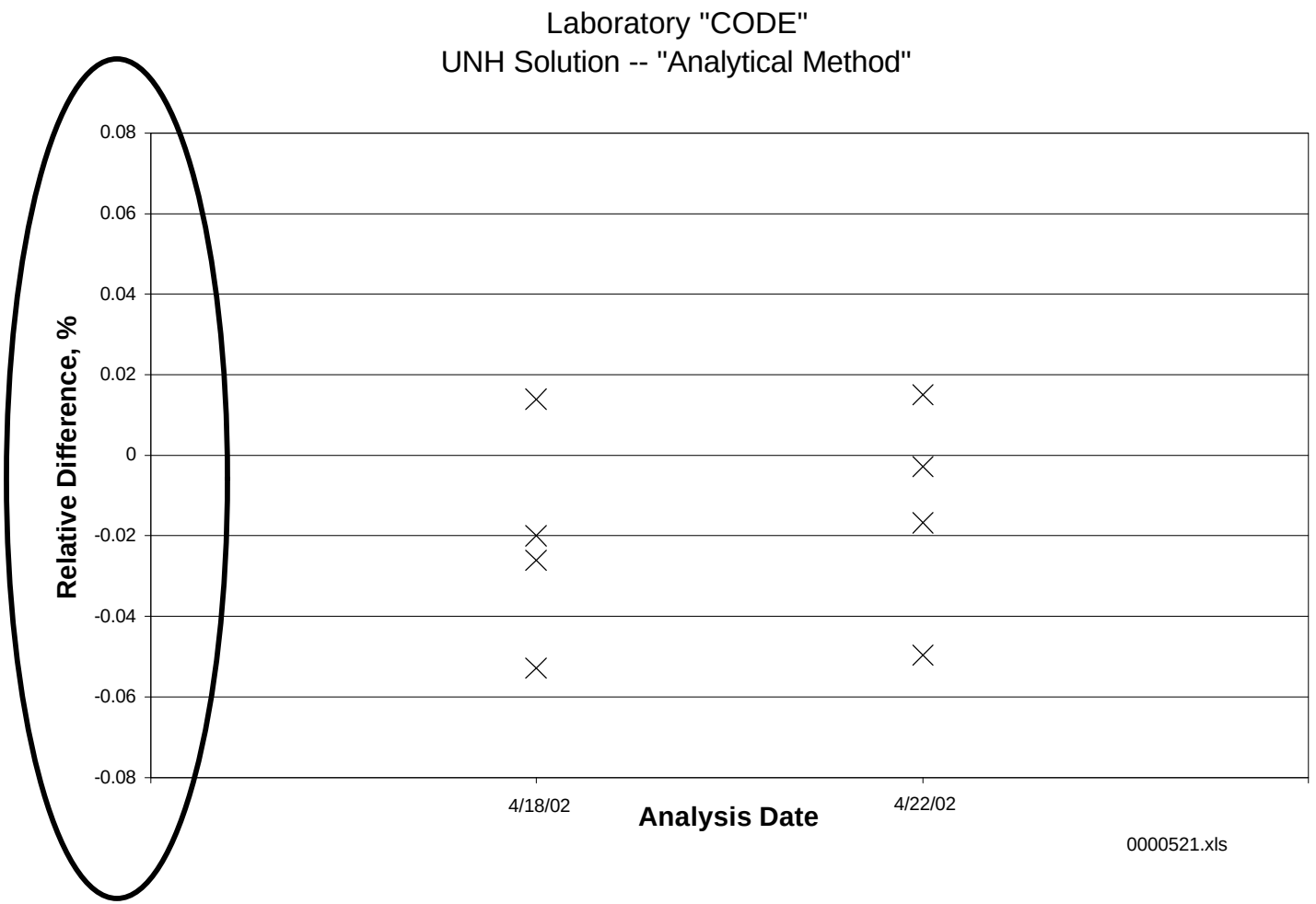
Data Plot


Laboratory "CODE"
UNH Solution -- "Analytical Method"
0.08
0.06
0.04
0.02
0
-0.02
-0.04
-0.06
-0.08
Relative Difference, %
4/18/02
4/22/02
Analysis Date
0000521.xls

Data Plot

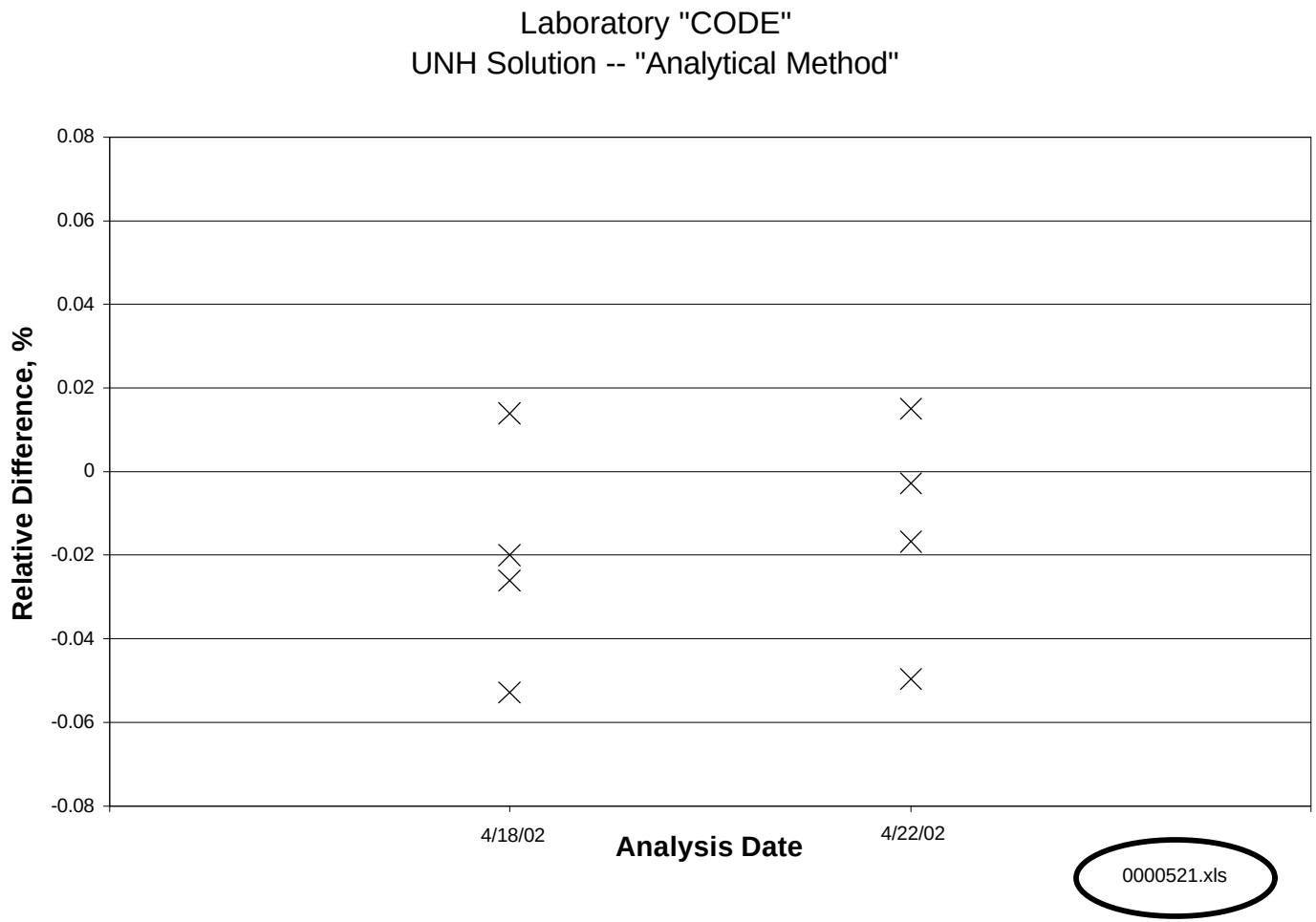

Laboratory "CODE"
UNH Solution -- "Analytical Method"
Relative Difference, %
0.08
0.06
0.04
0.02
0
-0.02
-0.04
-0.06
-0.08
4/18/02
4/22/02
Analysis Date
0000521.xls

Data Plot

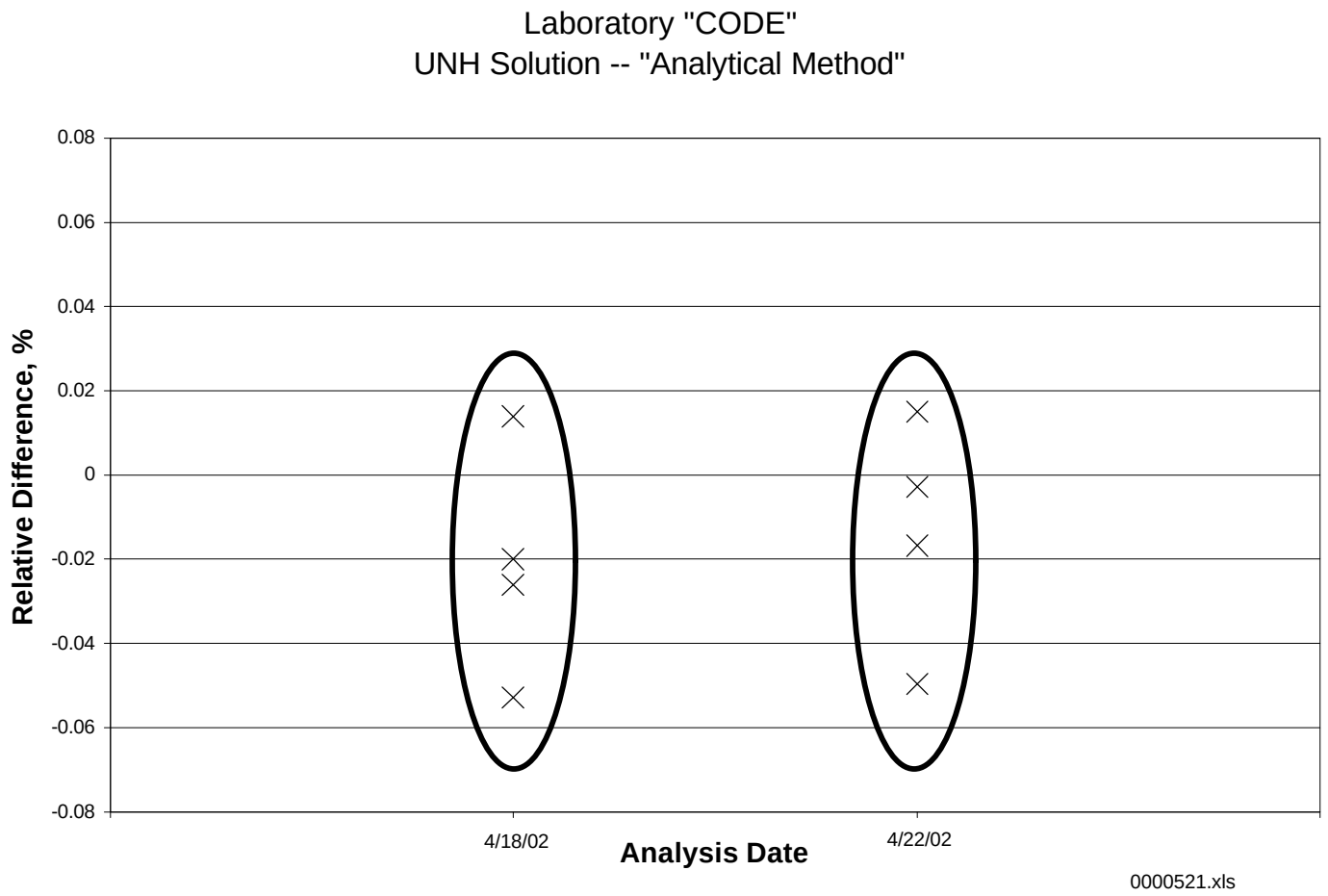


New Brunswick Laboratory

Data Plot


Laboratory "CODE"
UNH Solution -- "Analytical Method"
Relative Difference, %
0.08
0.06
0.04
0.02
0
-0.02
-0.04
-0.06
-0.08
4/18/02
4/22/02
Analysis Date
0000521.xls

Data Plot


Laboratory "CODE"
UNH Solution -- "Analytical Method"
0.08
0.06
0.04
0.02
0
-0.02
-0.04
-0.06
-0.08
Relative Difference, %
4/18/02
4/22/02
Analysis Date
0000521.xls

Analysis Goals for SME Data

- Validate the data; evaluate for outliers
- Describe the data graphically
- **Evaluate the data for day-to-day variations**
- Estimate the accuracy and precision of the measurements
- Estimate the overall closeness of the data to the characterized values

Statistical Analysis Routine

- Evaluate the day-to-day variation
  - If not significant, group all data together
  - If significant, use day-to-day variation to determine confidence limits (note that there will only be one degree of freedom in this case if analyses were run on two days)

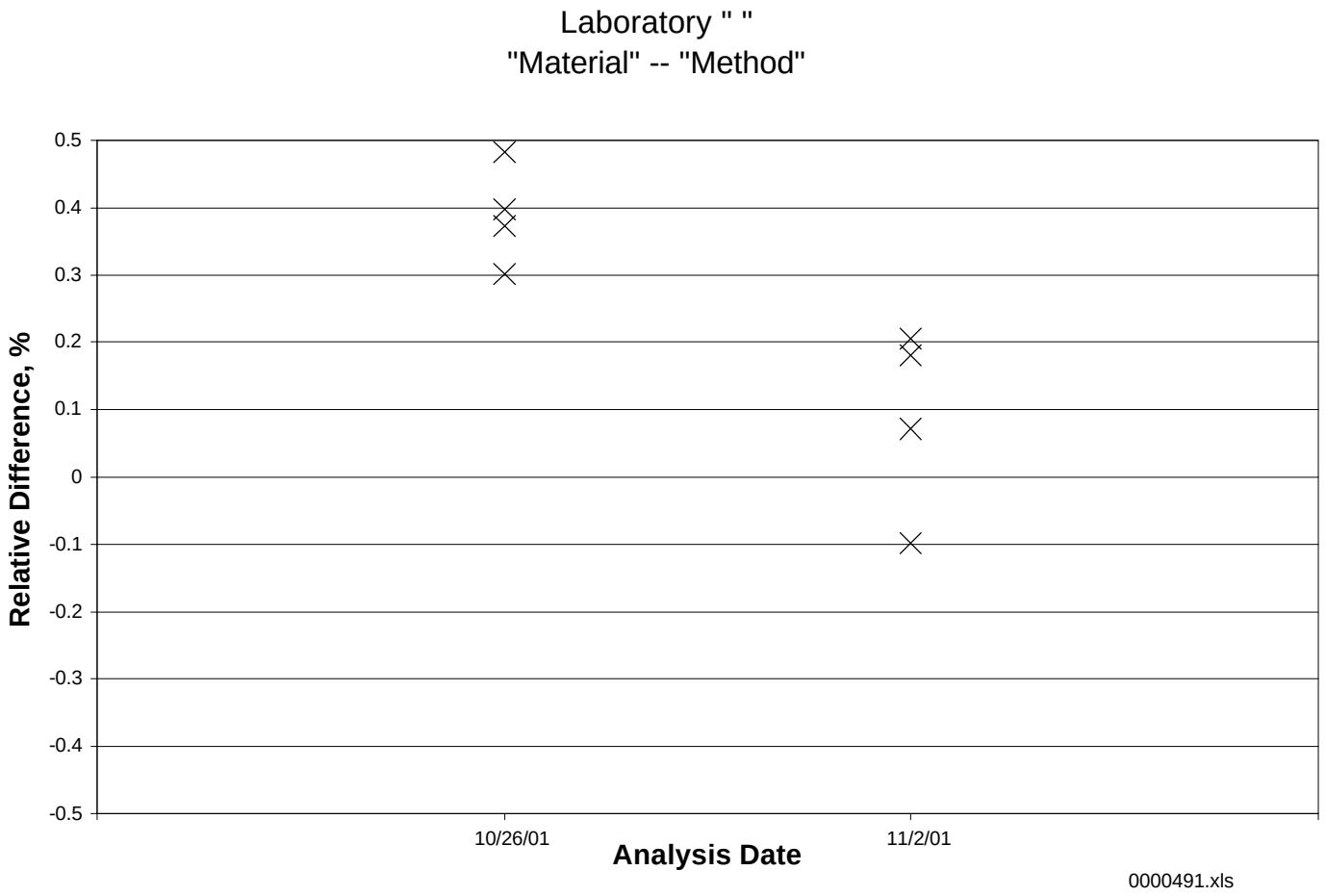
Day-to-Day Variation

- Reference: NUREG/CR-4604, Statistical Methods for Nuclear Material Management, Bowen and Bennett, 1988. See Chapter 5, Section 5.2, Table 5.3 and A6.
- If $\geq$ 95%, then day-to-day variation is significant

Day-to-Day Variation Not Significant

Number of Results Analyzed	8
Mean % Difference	-0.017
Mean Absolute % Difference	0.025
95% C. L. of Mean (df = 7)	0.021
Standard Deviation	0.026
Between-Day Standard Deviation (df = 1)	0.011
Within-Day Standard Deviation (df = 6)	0.027
Statistical Significance of Between-Day Standard Deviation	28.7%

Significant Day-to-Day Variation



Significant Day-to-Day Variation

Number of Results Analyzed	8
Mean % Difference	0.239
Mean Absolute % Difference	0.264
95% C. L. of Mean (df =1)	1.903
Standard Deviation	0.190
Between-Day Standard Deviation (df = 1)	0.423
Within-Day Standard Deviation (df = 6)	0.111
Statistical Significance of Between-Day Standard Deviation	99.1%

Significant Day-to-Day Variation

Sample Number	Aliquant Number	Analysis Date	Reported %U	% Relative Difference	Analyst Code
98EU0182-067	1	10/26/01	82.92000	0.0200	
98EU0184-092	1	10/26/01	83.00000	0.3980	
98EU0182-067	2	10/26/01	82.98000	0.3738	
98EU0184-092	2	10/26/01	83.07000	0.4826	
98EU0182-067	3	11/02/01	82.59000	-0.0980	
98EU0184-092	3	11/02/01	82.82000	0.1802	
98EU0182-067	4	11/02/01	82.73000	0.0714	
98EU0184-092	4	11/02/01	82.84000	0.2044	

Analysis Goals for SME Data

- Validate the data; evaluate for outliers
- Describe the data graphically
- Evaluate the data for day-to-day variations
- **Estimate the accuracy and precision of the measurements**
- Estimate the overall closeness of the data to the characterized values

Statistical Analysis Routine

- Relative Standard Deviation (RSD)
  - the standard deviation of the % Relative Differences of a set of measurements
  - is an indication of precision, independent of the characterized values

Magnitude of Random Error

Number of Results Analyzed	8
Mean % Difference	-0.017
Mean Absolute % Difference	0.025
95% C. L. of Mean (df = 7)	0.021
Standard Deviation	0.026
Between-Day Standard Deviation (df = 1)	0.011
Within-Day Standard Deviation (df = 6)	0.027
Statistical Significance of Between-Day Standard Deviation	28.7%

Compare to 2000 ITV u(r)

Magnitude of Bias

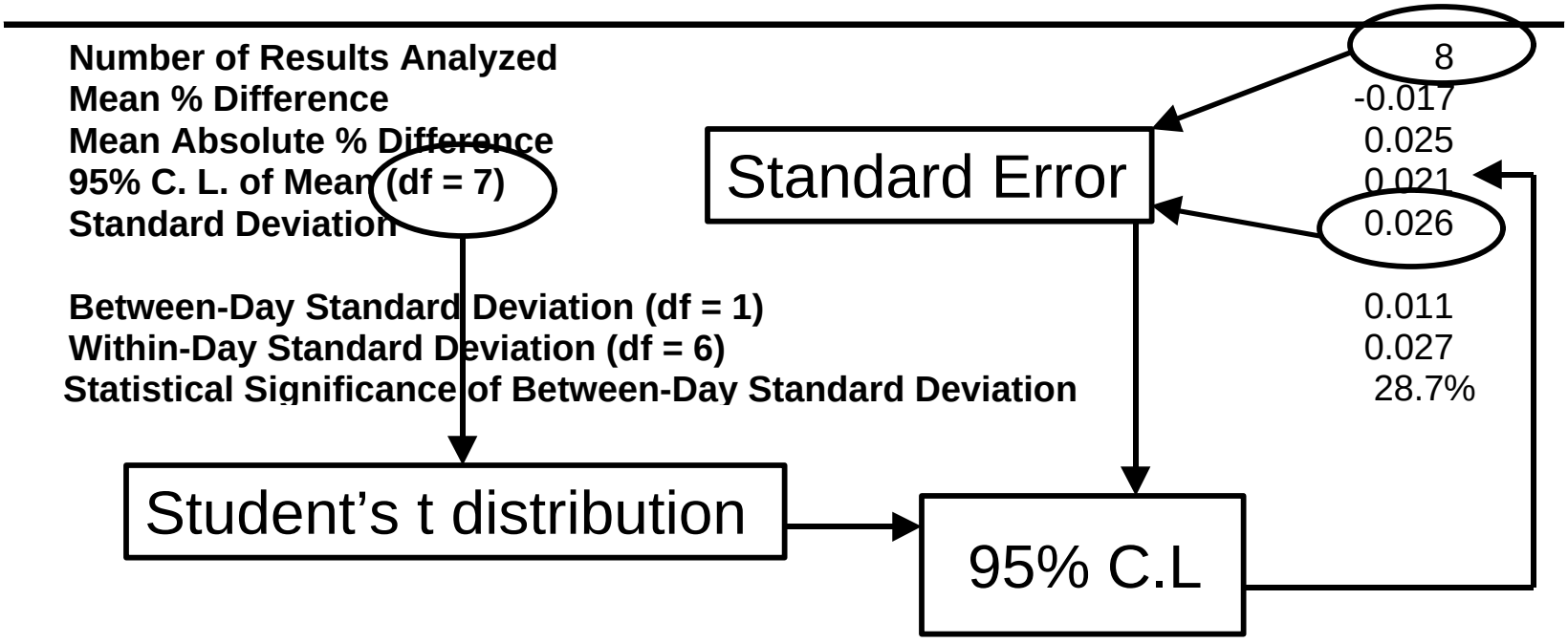
Number of Results Analyzed	8
Mean % Difference	-0.017
Mean Absolute % Difference	0.025
95% C. L. of Mean (df = 7)	0.021
Standard Deviation	0.026
Between-Day Standard Deviation (df = 1)	0.011
Within-Day Standard Deviation (df = 6)	0.027
Statistical Significance of Between-Day Standard Deviation	28.7%

Compare to 2000 ITV u(s)

Statistical Significance of the Bias

Number of Results Analyzed	8
Mean % Difference	-0.017
Mean Absolute % Difference	0.025
95% C. L. of Mean (df = 7)	0.021
Standard Deviation	0.026
Between-Day Standard Deviation (df = 1)	0.011
Within-Day Standard Deviation (df = 6)	0.027
Statistical Significance of Between-Day Standard Deviation	28.7%

95% Confidence Limit (no significant day-to-day variation)



Student's t Distribution

ν	$\alpha=0.05$
1	12.706
2	4.303
7	2.365
15	2.131
infinite	1.960

95% Confidence Limit (significant day-to-day variation)

Number of Results Analyzed	8
Mean % Difference	0.239
Mean Absolute % Difference	0.264
95% C. L. of Mean (df =1)	1.903
Standard Deviation	0.190
Between-Day Standard Deviation (df = 1)	0.423
Within-Day Standard Deviation (df = 6)	0.111
Statistical Significance of Between-Day Standard Deviation	99.1%

Note that the 95% C.L. is very large due to df=1

Significant Day-to-Day Variation

Analysis Goals for SME Data

- Validate the data; evaluate for outliers
- Describe the data graphically
- Evaluate the data for day-to-day variations
- Estimate the accuracy and precision of the measurements
- **Estimate the overall closeness of the data to the characterized values**

Mean Absolute % Difference

Number of Results Analyzed	8
Mean % Difference	-0.017
Mean Absolute % Difference	0.025
95% C. L. of Mean (df = 7)	0.021
Standard Deviation	0.026
Between-Day Standard Deviation (df = 1)	0.011
Within-Day Standard Deviation (df = 6)	0.027
Statistical Significance of Between-Day Standard Deviation	28.7%

Low if both accuracy and precision are small

Cover Letter

- Summarizes data evaluation
  - Results of outlier tests
  - Results of tests for day-to-day variation
  - Existence of bias
  - Comparison of uncertainties to 2000 International Target Values u(r) and u(s)

Caveats

- Statistically significant biases/variations may not be of practical significance
- Biases detected by SMEP analyses are short-term for typical reports; other time bases may give different results!
- 2000 ITVs are goals; required precision and accuracy are set by Operations Offices

Note on Use of International Target Values for Domestic Safeguards Measurements

Jim Crabtree and Melanie May,
SO-11, Safeguards and Security Policy

Mike Sparks, Lynne Preston, and Wendy Rhodes,
SO-13, Policy Integration and Technical Support

David Crawford, David Young, Ed Reynolds, Len Myers
SO-62, Office of Pu, U, and Special Materials Inventory

Jay Thompson, Wanda Mitchell, Kimberly Johnson-Miller,
New Brunswick Laboratory

Norbert Ensslin, Tom Burr, Morag Smith, Cliff Rudy, Brian Scott,
Los Alamos National Laboratory

Los Alamos
NATIONAL LABORATORY

What is our Interest in, and Concern with, International Target Values (ITVs)?

- The DOE Order on MC&A mentions measurement goals.

- But DOE facilities contain many material forms for which the ITVs are not appropriate.
  - These sites sometimes set their own Precision and Accuracy Goals (PAGs).

- We are preparing a “Modernization Plan” to guide domestic MC&A technology development:
  - Over the next year, LANL, LLNL, Y-12, SRS, and other MC&A technology development sites will work with SO to define technology development needs for existing and future facilities.
  - The Plan will contain a Measurement Matrix showing where improvements may be needed.

- David Crawford’s suggestions for developing the Measurement Matrix:
  - ITVs can provide a technical basis for quantitative “quality thresholds.”
  - The sites may also develop a list of other forms that are included in site-specific MC&A Plans.
  - Measurement Evaluation Program data from NBL should provide good input.

- So there is a lot of information already available that can be utilized in some hybrid way to provide performance criteria for the Measurement Matrix.

Measurement Matrix - current version only describes state of technology today

NM Type, Form, or Container	NM Inventory (kg)	Calorimetry w/Gamma Isotopics	Gamma-Ray Isotopics	Hybrid Densitometry XRF	Far-field Trans-Corr Gamma	Segmented Gamma Scanning	Tomographic Gamma Scanning	Passive Neutron Totals	Passive Neutron Coincidence	Passive Neutron Multiplicity	Active Neutron Coincidence	Active Neutron Multiplicity	Delayed Neutron Shuffler	Differential Die-Away Technique
Pu metal in cans		1						2	4	2				
Impure Pu metals & alloys		1								5 to 10				
Pu classified parts													3	
Pu oxide in cans		1							5	2				
Pu in shielded drums														
Pu fuel rods								5	2					
Pu assembly									2					
Pu solutions				1	1				5					
Pu scrap (low impurity)		2				10	6			3				
Pu residues (mid impurity)		2				15	6		10	5				
Pu residues (hi impurity)		2				15	8		40					
Pu waste (hi density)									20	5			10	
Pu waste (lo density)					5	4	2		20					30
Pu holdup					25			50						
Pu process monitoring					15			25						
Pu portal monitoring														
ID of shielded Pu														
HEU metal & oxide pieces											>10	5	4	
LEU metal								2	4		8		3	
U oxide in cans					2			10			3	5	2	
U in shielded drums														
UF6 solid								5			5			
U carbides													30	
U solutions				1	1								4	
U fuel pellets											5		3	
U fuel rods								10			3		3	
U waste (hi)											15		8	
U waste (lo)					10	5					15		20	30
U holdup					50									
U process monitoring					30									
U portal monitoring														
ID of shielded U														
U/Pu classified parts													3	
U/Pu oxide in cans										5	5		5	
U/Pu solutions				1	1									
U/Pu fuel pellets		1							2		5			
U/Pu fuel rods		2									3		1	
U/Pu waste (hi density)									20	5	30			
U/Pu waste (lo density)					15	10			15					30
ID of U in presence of Pu														
Irradiated nuclear fuel													25	25
Pu-238 oxide						5			5					
Pu-242 oxide														
Np-237 metal														
Np-237 oxide						10							8	
U-233 oxide														
U-235 with U-236														
Americium														
Alternate N.M. solutions				2										
Tritium waste														

Los Alamos NATIONAL LABORATORY

We Invite Further Discussions on this Topic

- Would anyone like to meet later at the INMM Meeting?
- There are two designated ad hoc meeting rooms,
  - Damselfish room,
  - Fantail room.
- These rooms are available on a first-come first-served basis. Should we request a time slot on Monday or later?
- What future steps should we undertake to continue and broaden this dialog? How can we involve the DOE facility sites in this process?

Los Alamos
NATIONAL LABORATORY

NEW BRUNSWICK LABORATORY

SAFEGUARDS MEASUREMENT EVALUATION PROGRAM

RESULTS for 2001

Jay Thompson

New Brunswick Laboratory

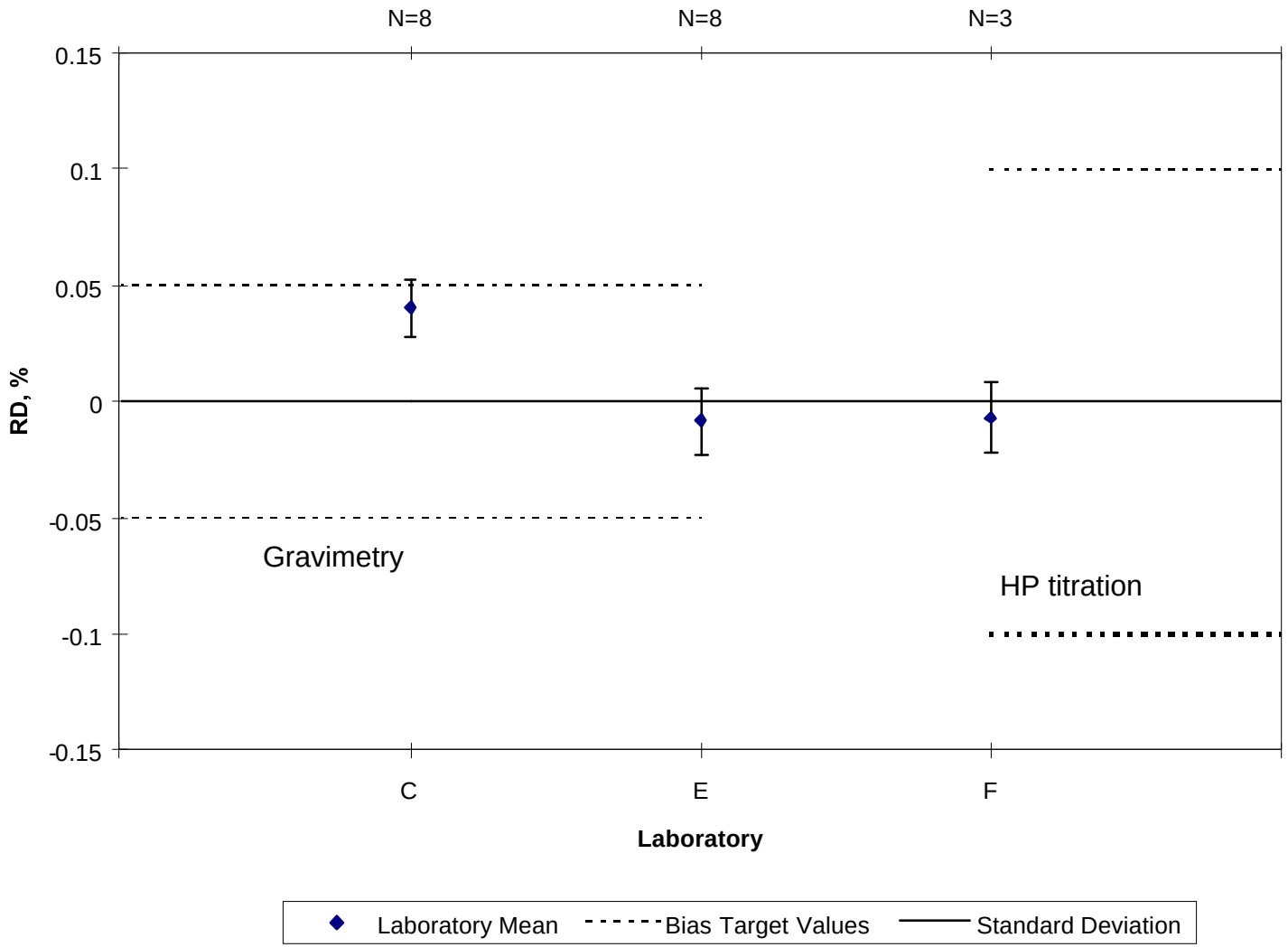
MATERIALS AVAILABLE

1. Uranyl nitrate solutions for U concentration
2. Uranyl nitrate solutions for ^{235}U enrichment
3. UO_2 pellets for U concentration and enrichment
4. UO_3 powder for U concentration
5. UF_6 (normal or low-enriched) solid for U concentration
6. UF_6 (low-enriched) solid for ^{235}U enrichment
7. Plutonium sulfate for isotopic abundances and IDMS

PARTICIPATING FACILITIES

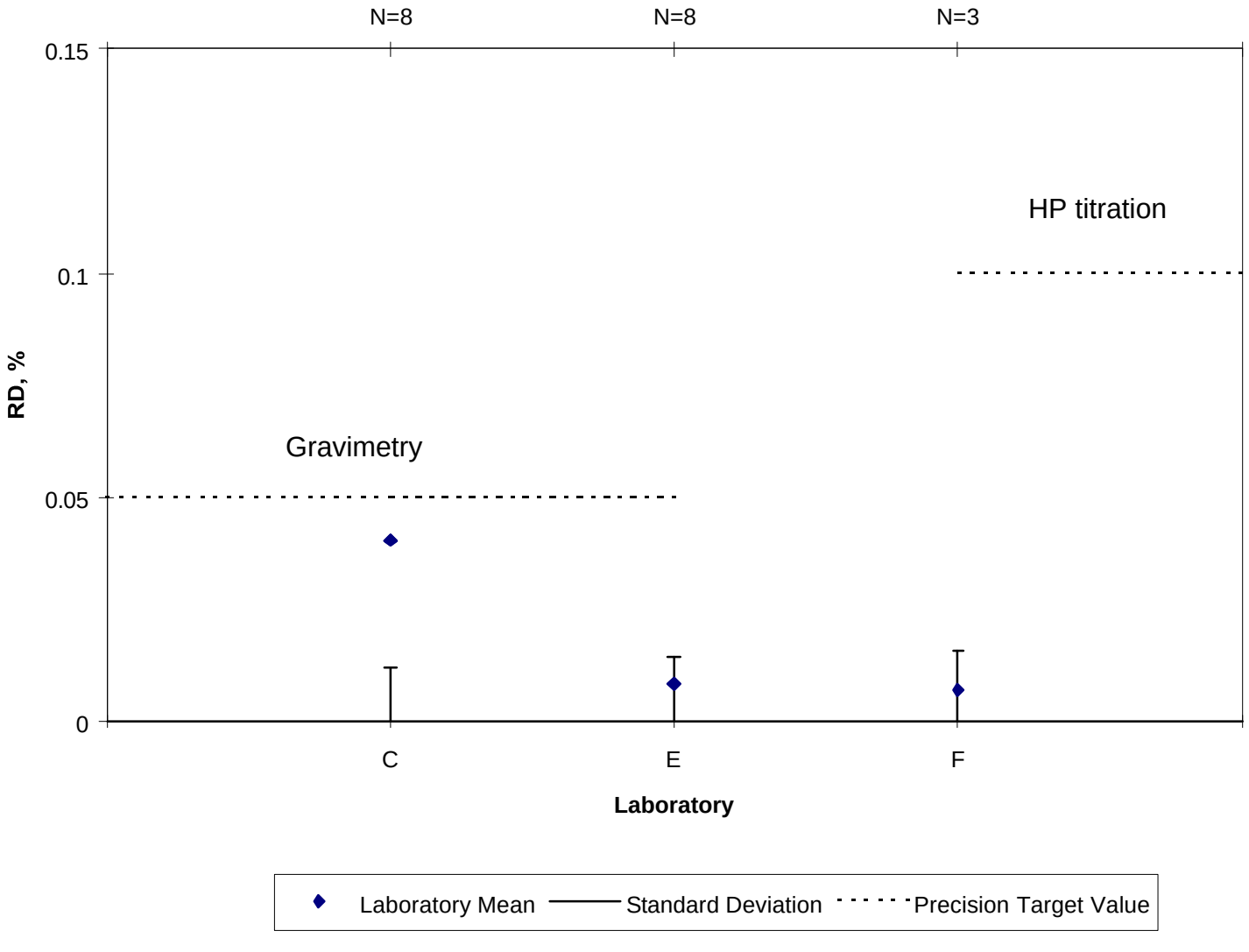
- 4 DOE Contractor Laboratories
- 1 Federal Laboratory (NBL)
- 7 NRC Licensees
- 9 International Laboratories (1 Japanese, 2 Argentine, 6 Brazilian)

New Brunswick Laboratory Safeguards Measurement Evaluation Program
UF6 - Percent U

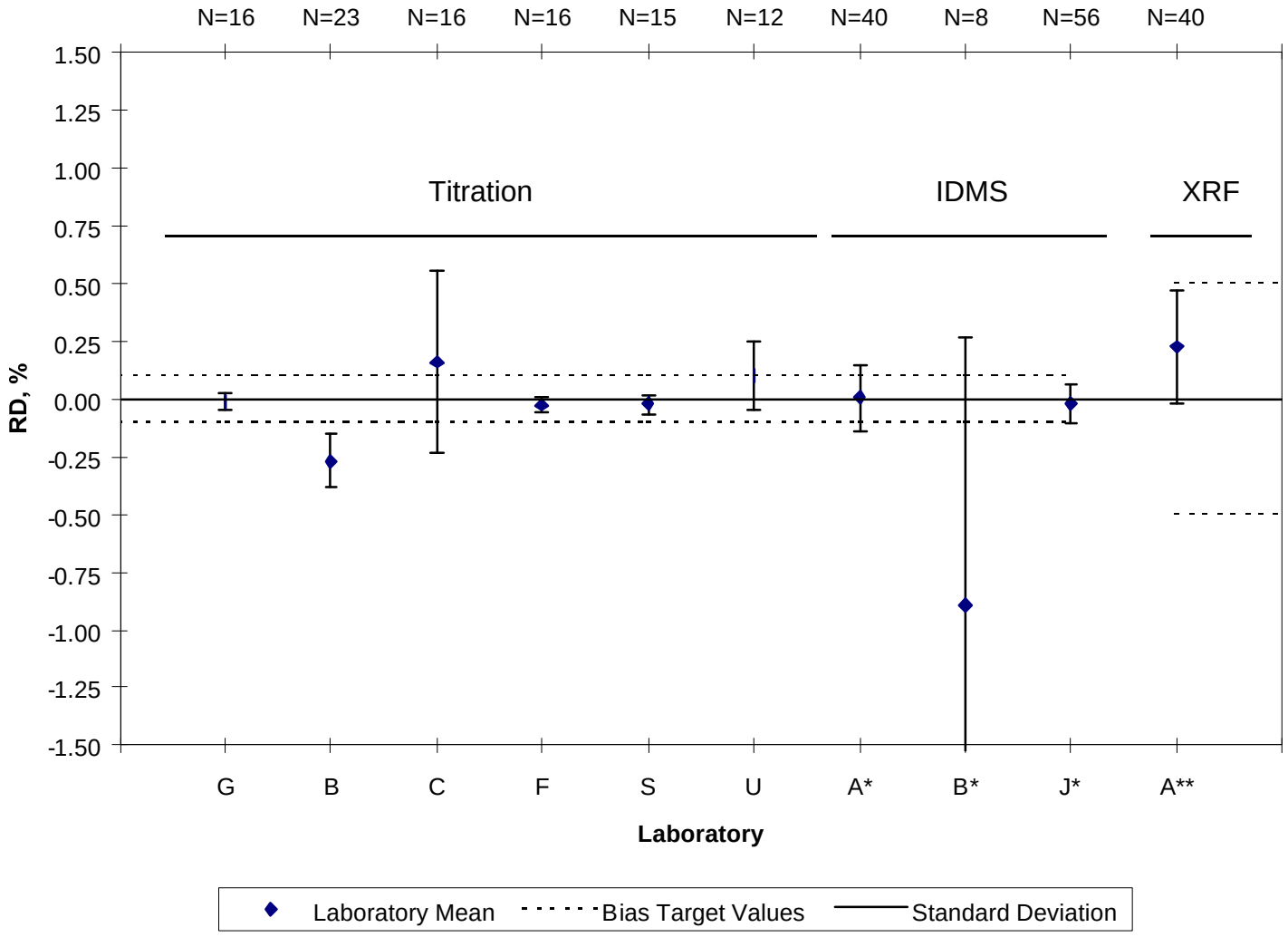


New Brunswick Laboratory

New Brunswick Laboratory Safeguards Measurement Evaluation Program
UF6 - Percent U

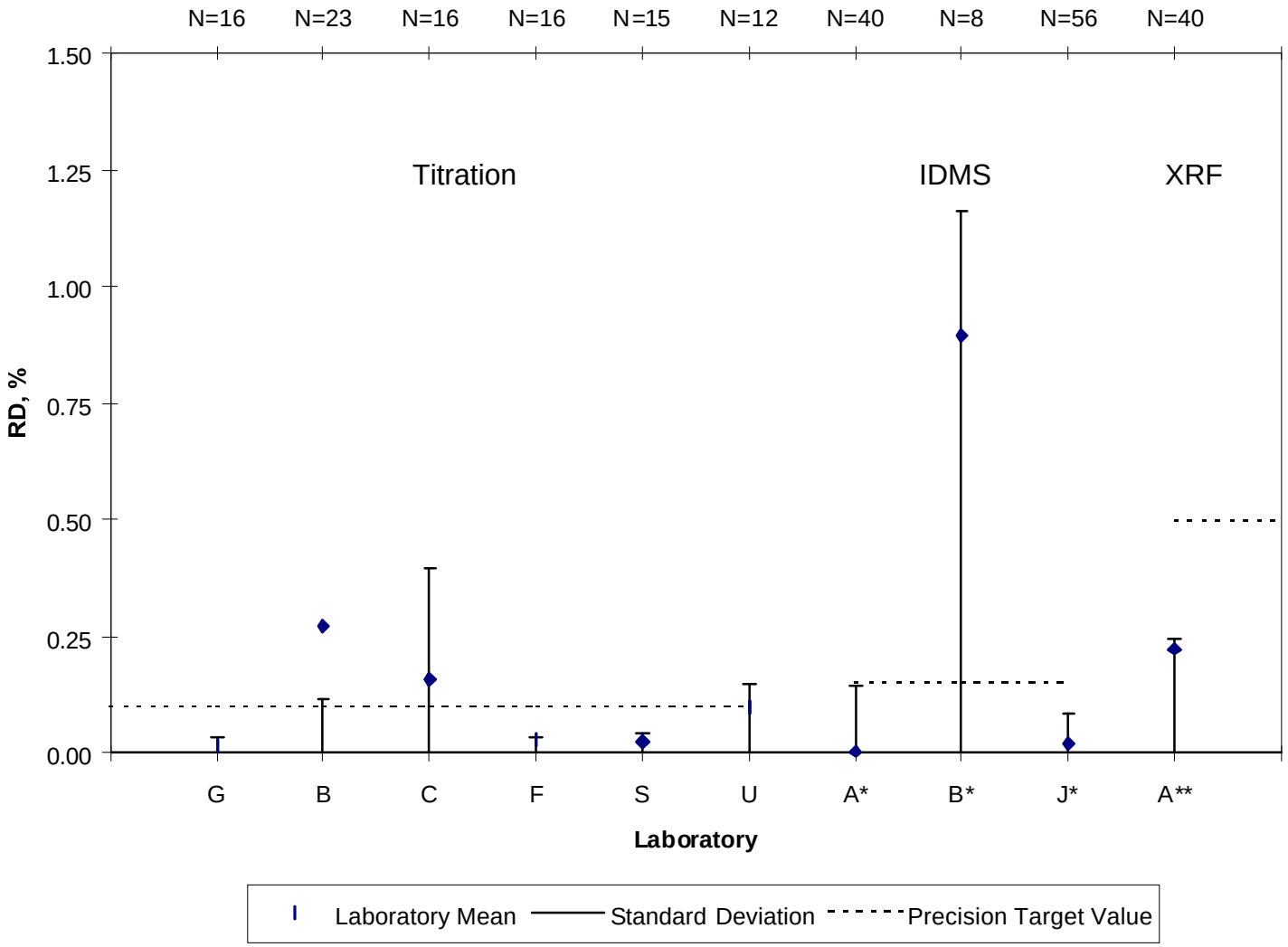


New Brunswick Laboratory



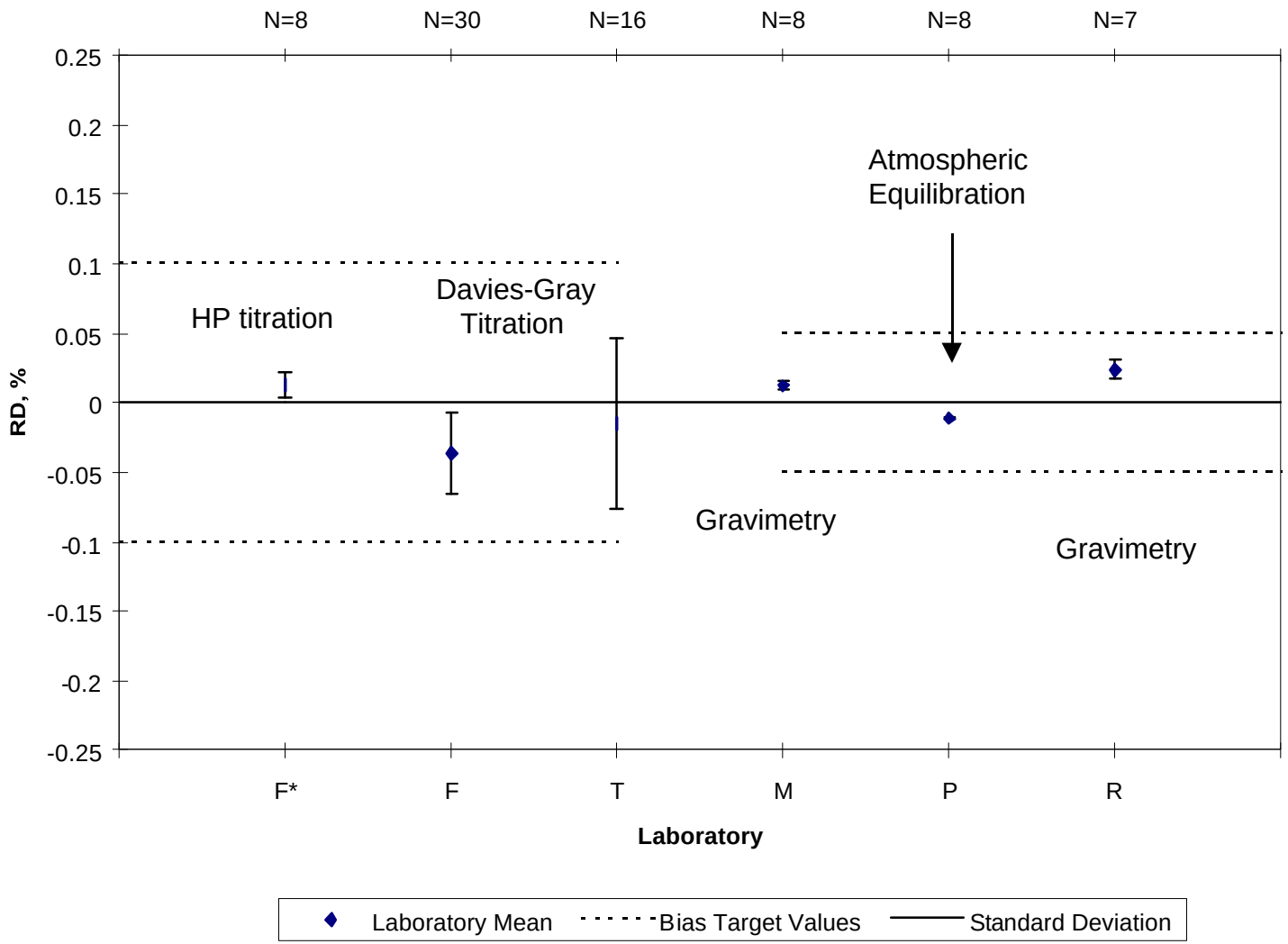
New Brunswick Laboratory

New Brunswick Laboratory Safeguards Measurement Evaluation Program
UNH - Percent U

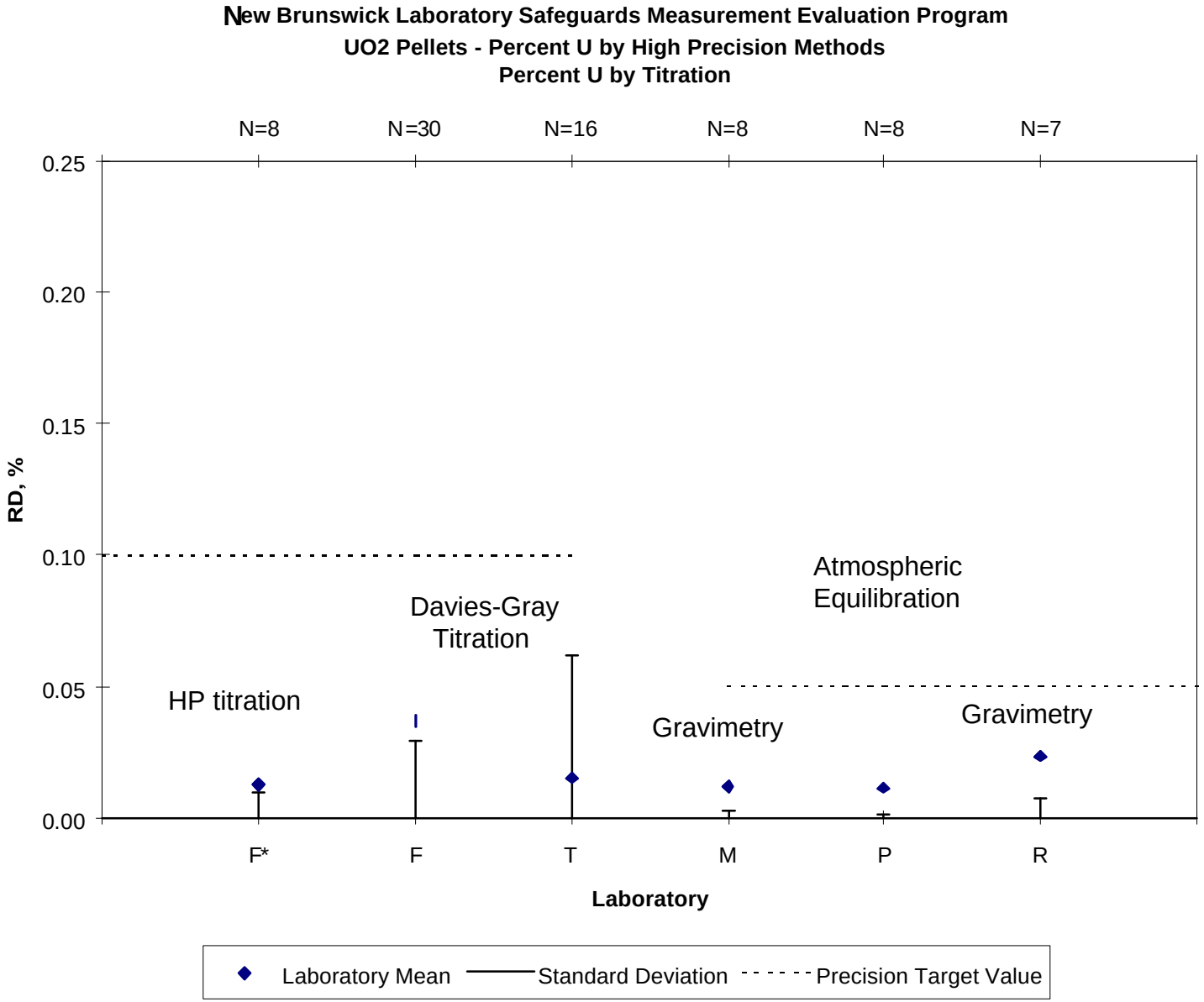


New Brunswick Laboratory

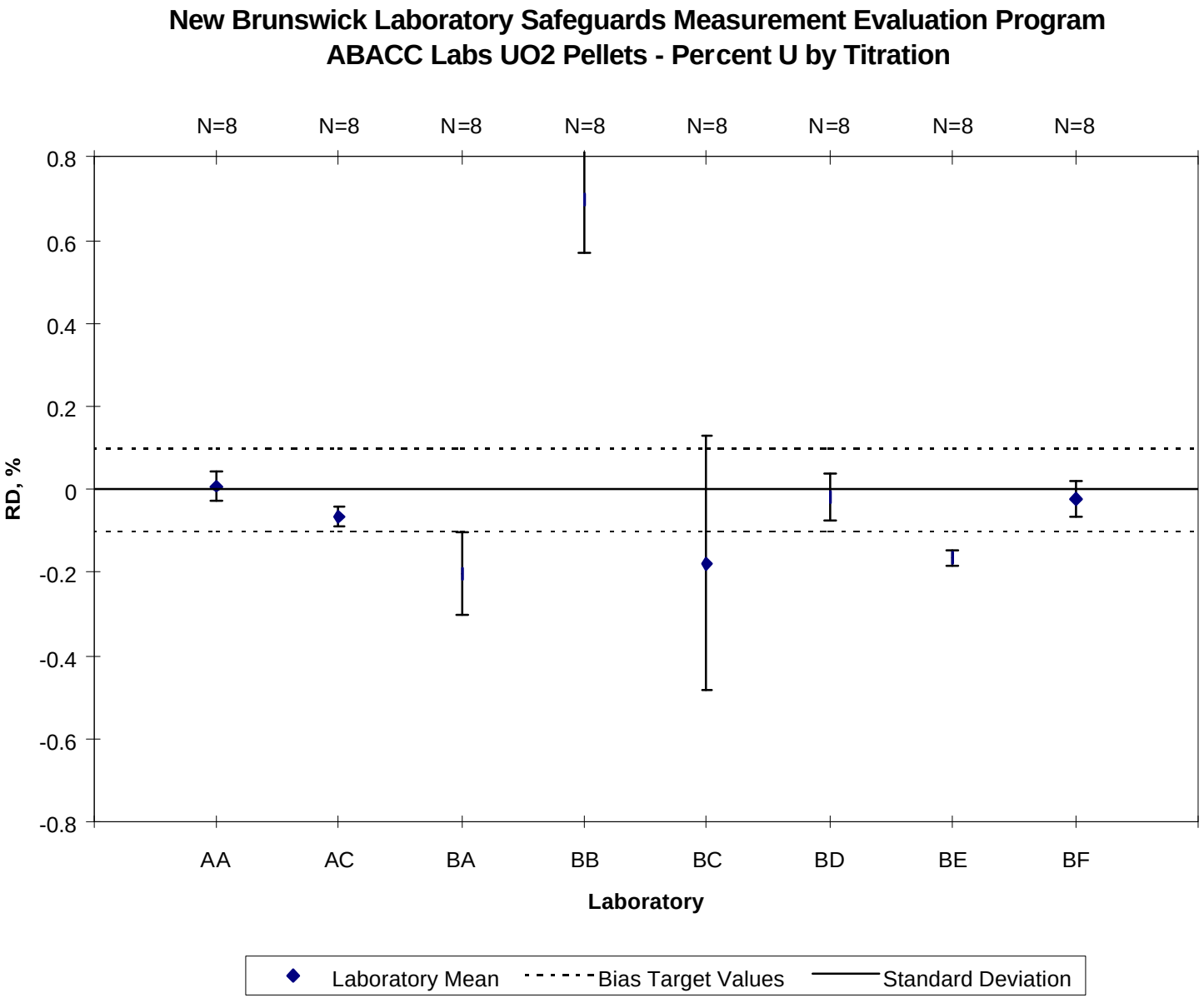
New Brunswick Laboratory Safeguards Measurement Evaluation Program
UO2 Pellets - Percent U by High Precision Methods
Percent U by Titration



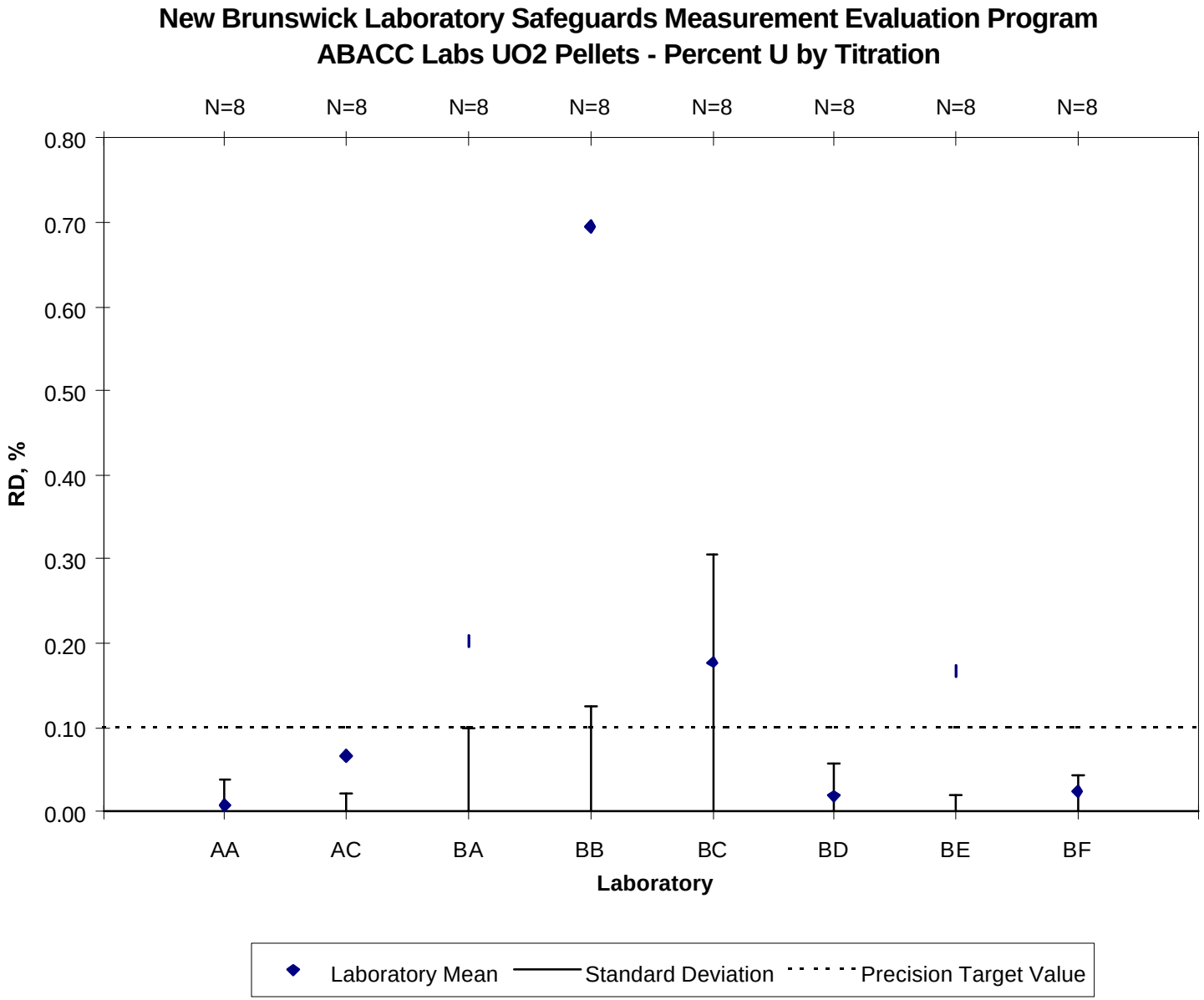
New Brunswick Laboratory



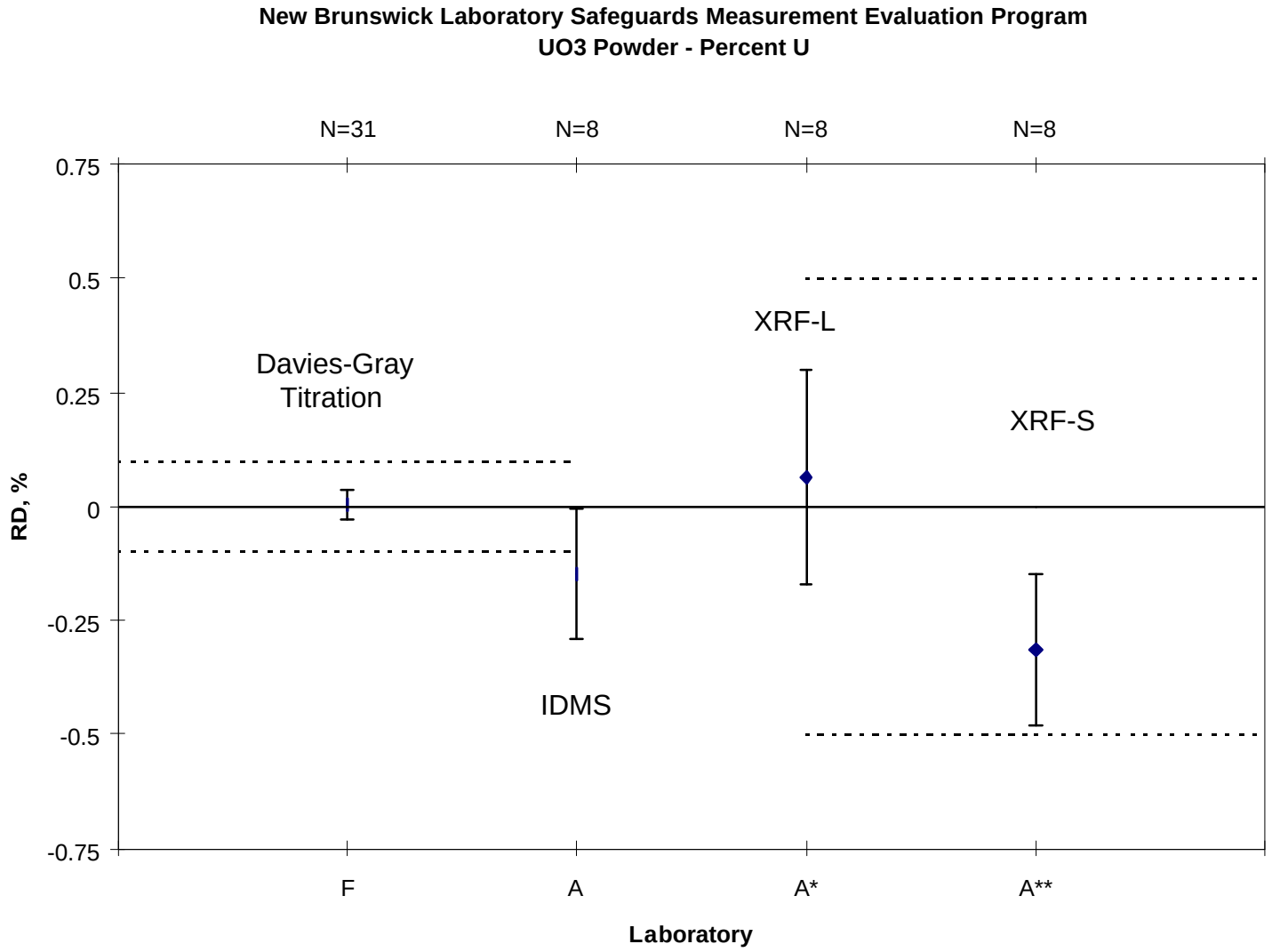
New Brunswick Laboratory



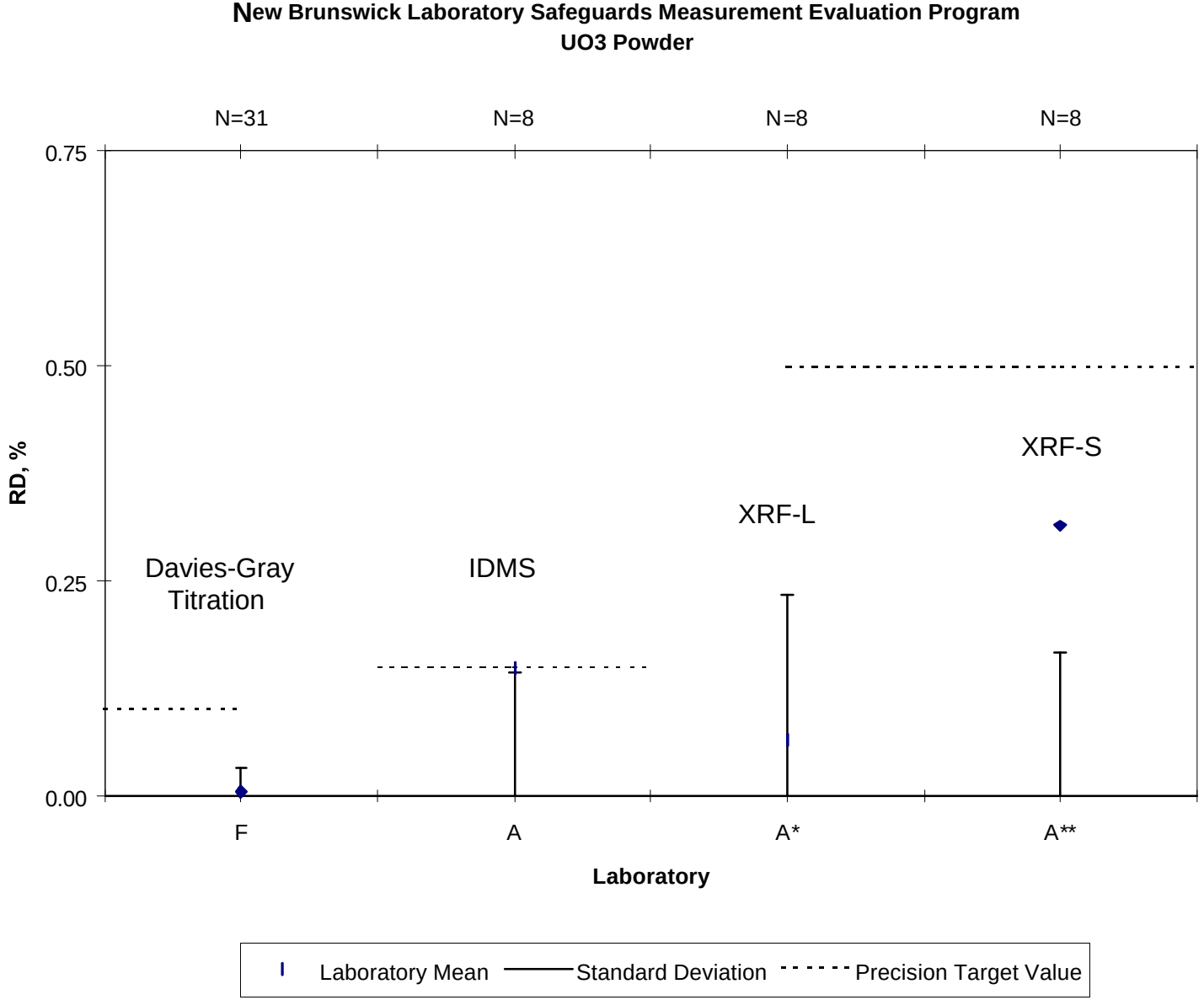
New Brunswick Laboratory



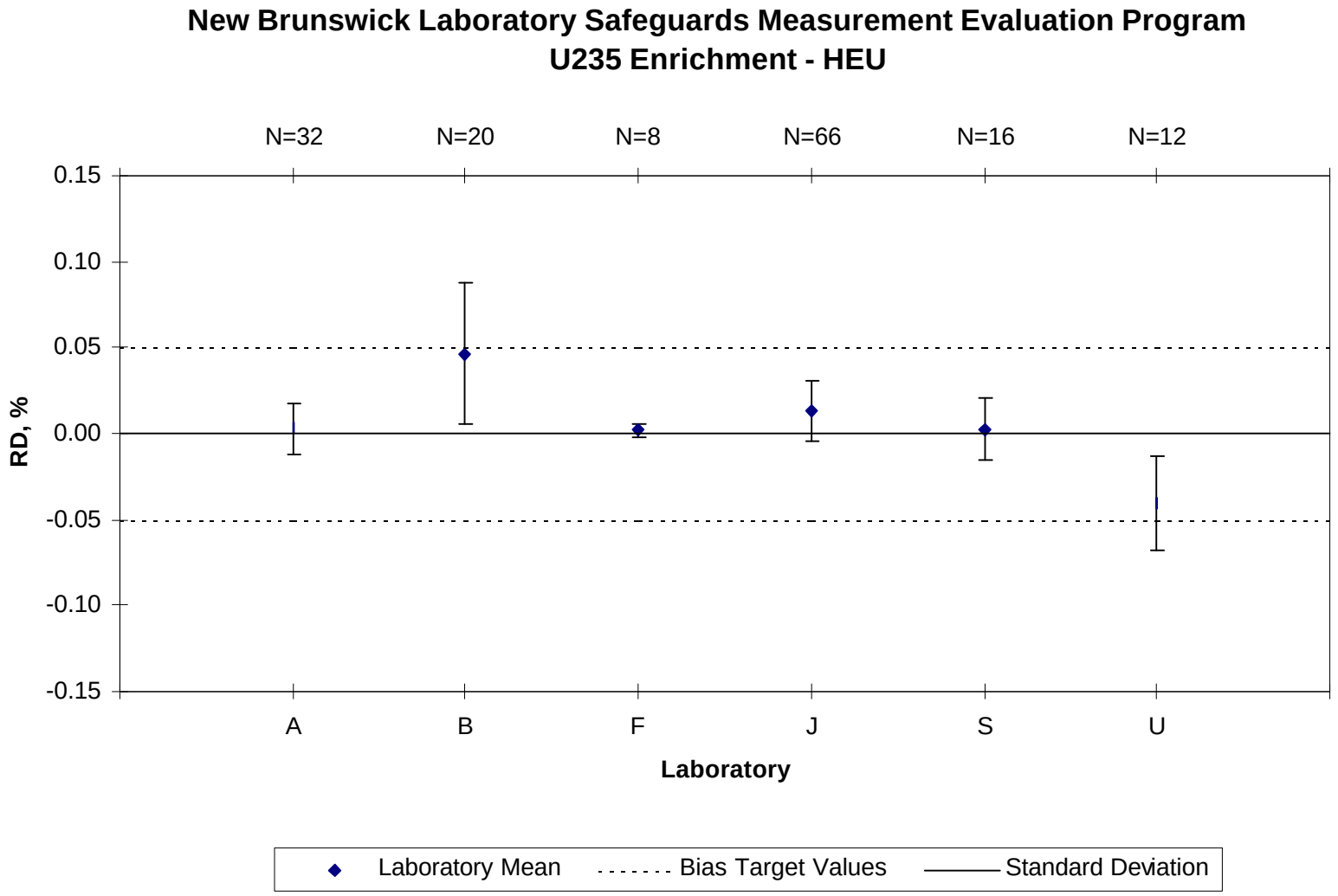
New Brunswick Laboratory



New Brunswick Laboratory

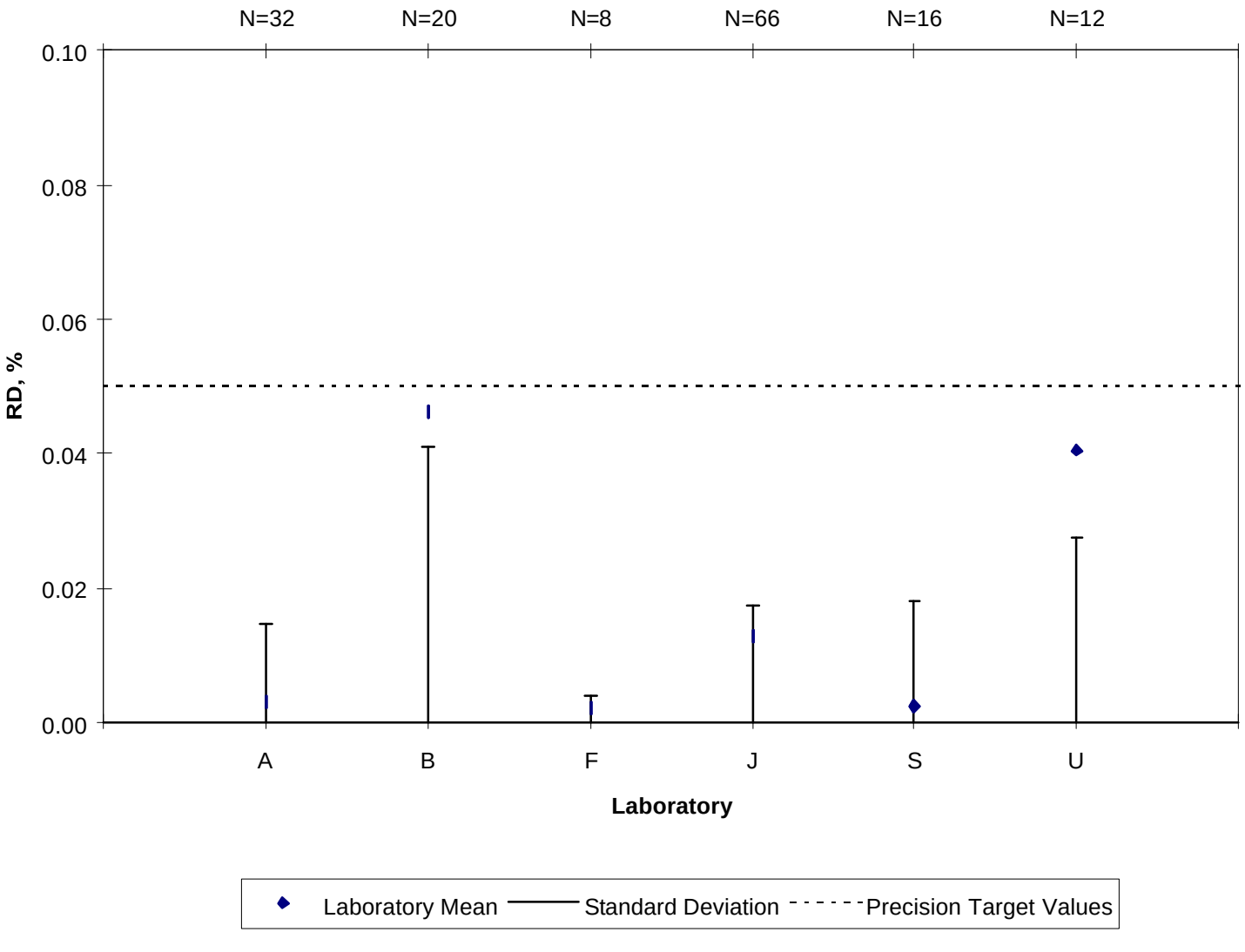


New Brunswick Laboratory

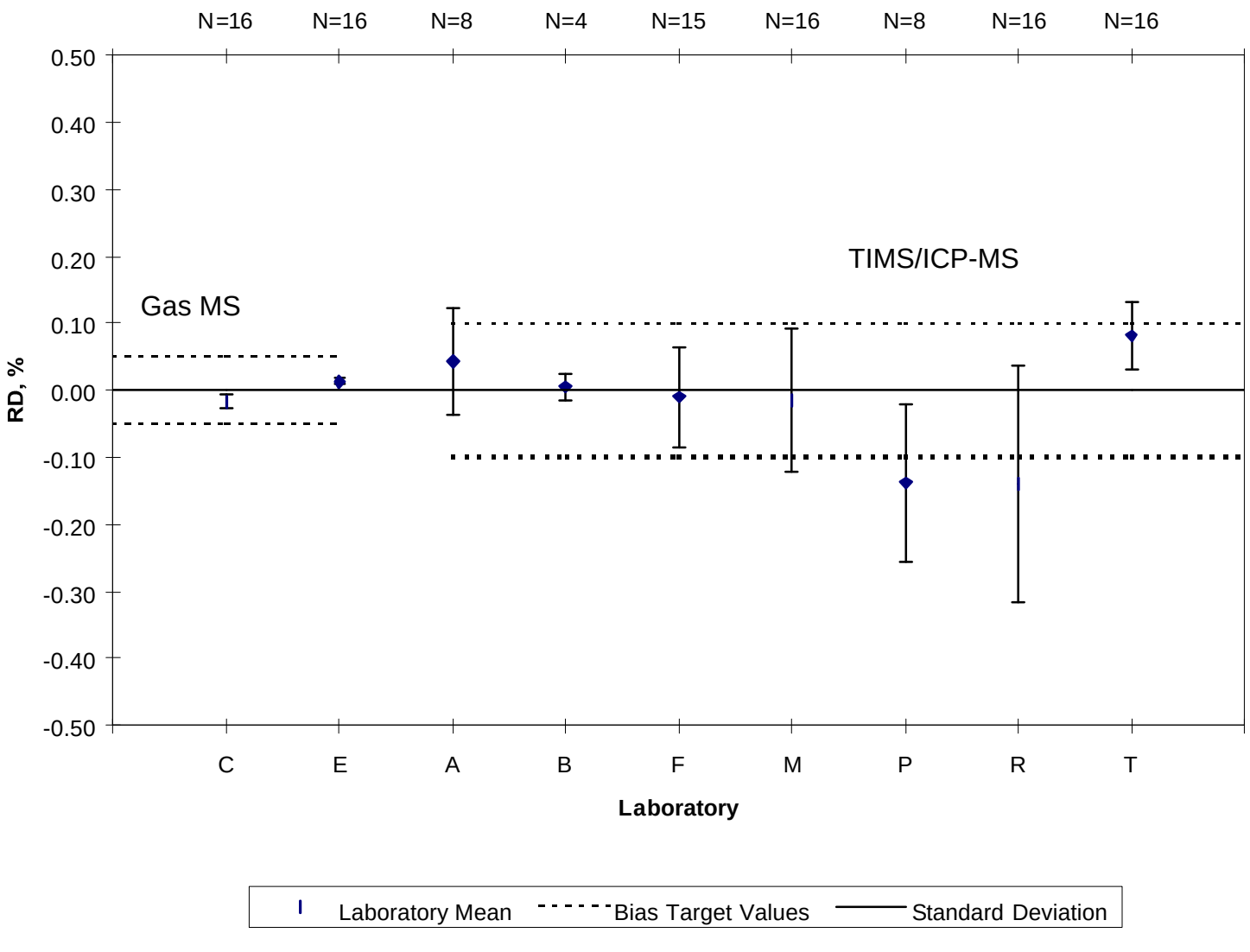


New Brunswick Laboratory

New Brunswick Laboratory Safeguards Measurement Evaluation Program
U235 Enrichment - HEU

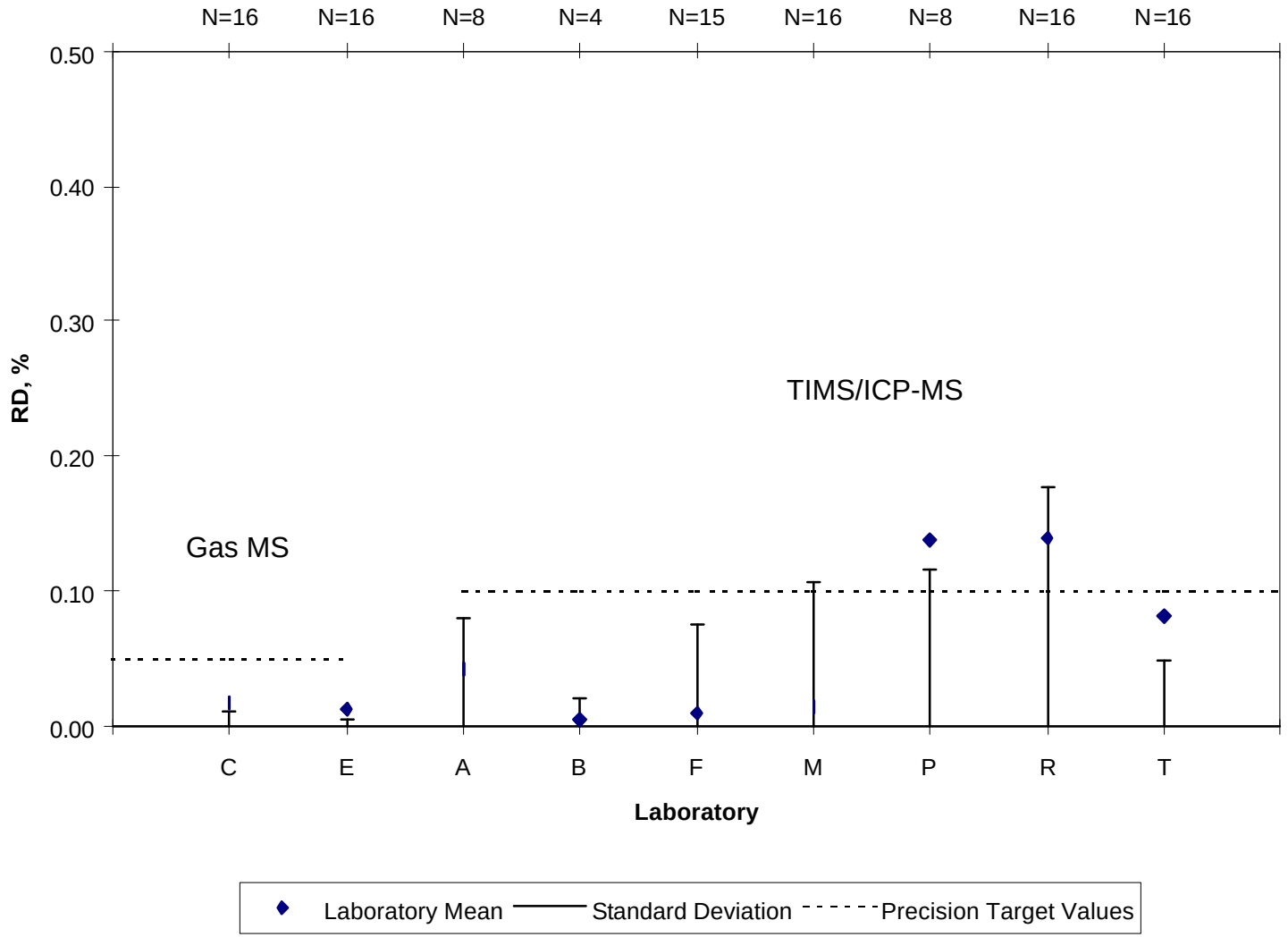


New Brunswick Laboratory



New Brunswick Laboratory

New Brunswick Laboratory Safeguards Measurement Evaluation Program
U235 Enrichment - LEU

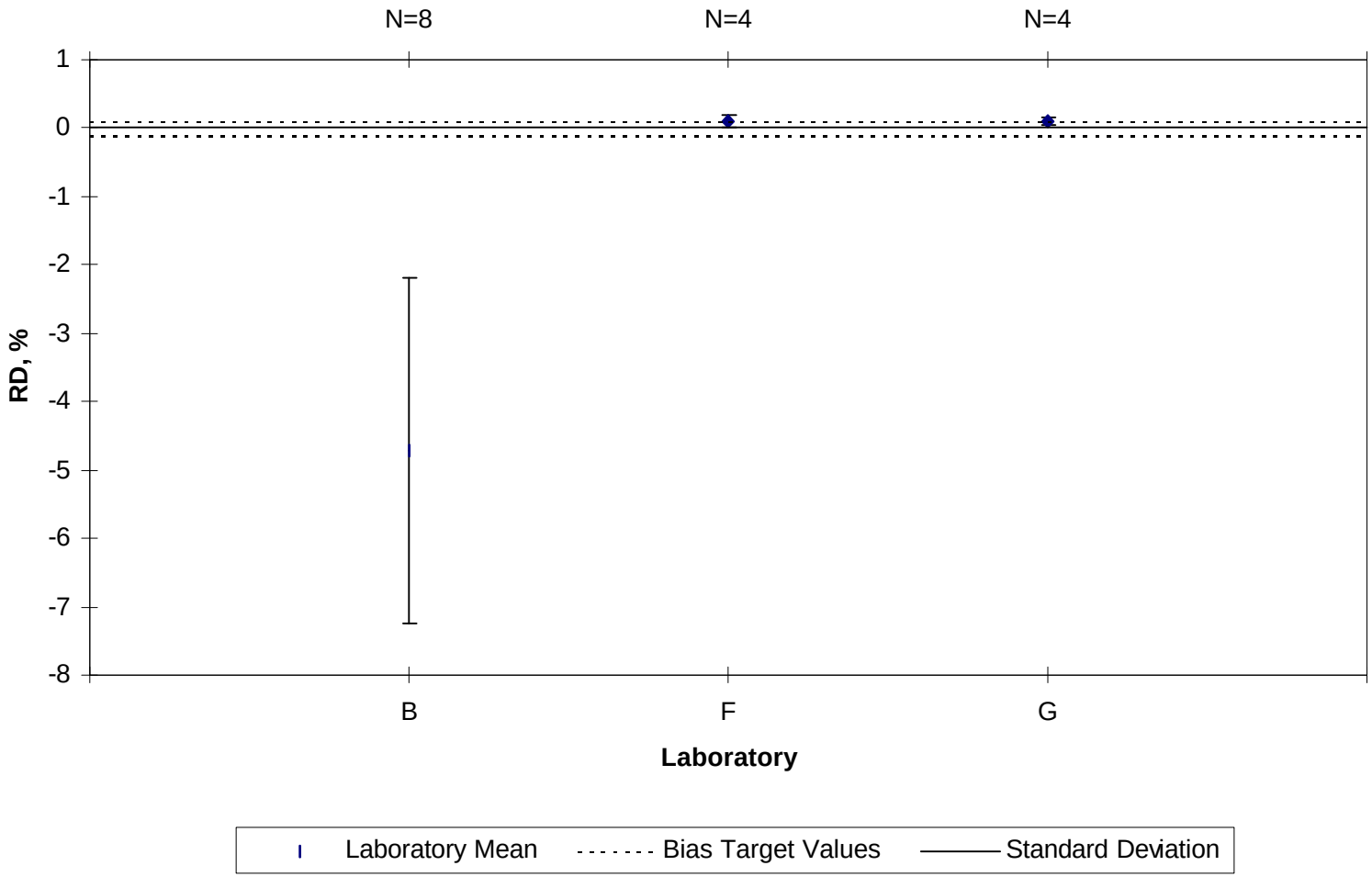


New Brunswick Laboratory

PLUTONIUM ISOTOPIC EXCHANGE PARTICIPATING FACILITIES

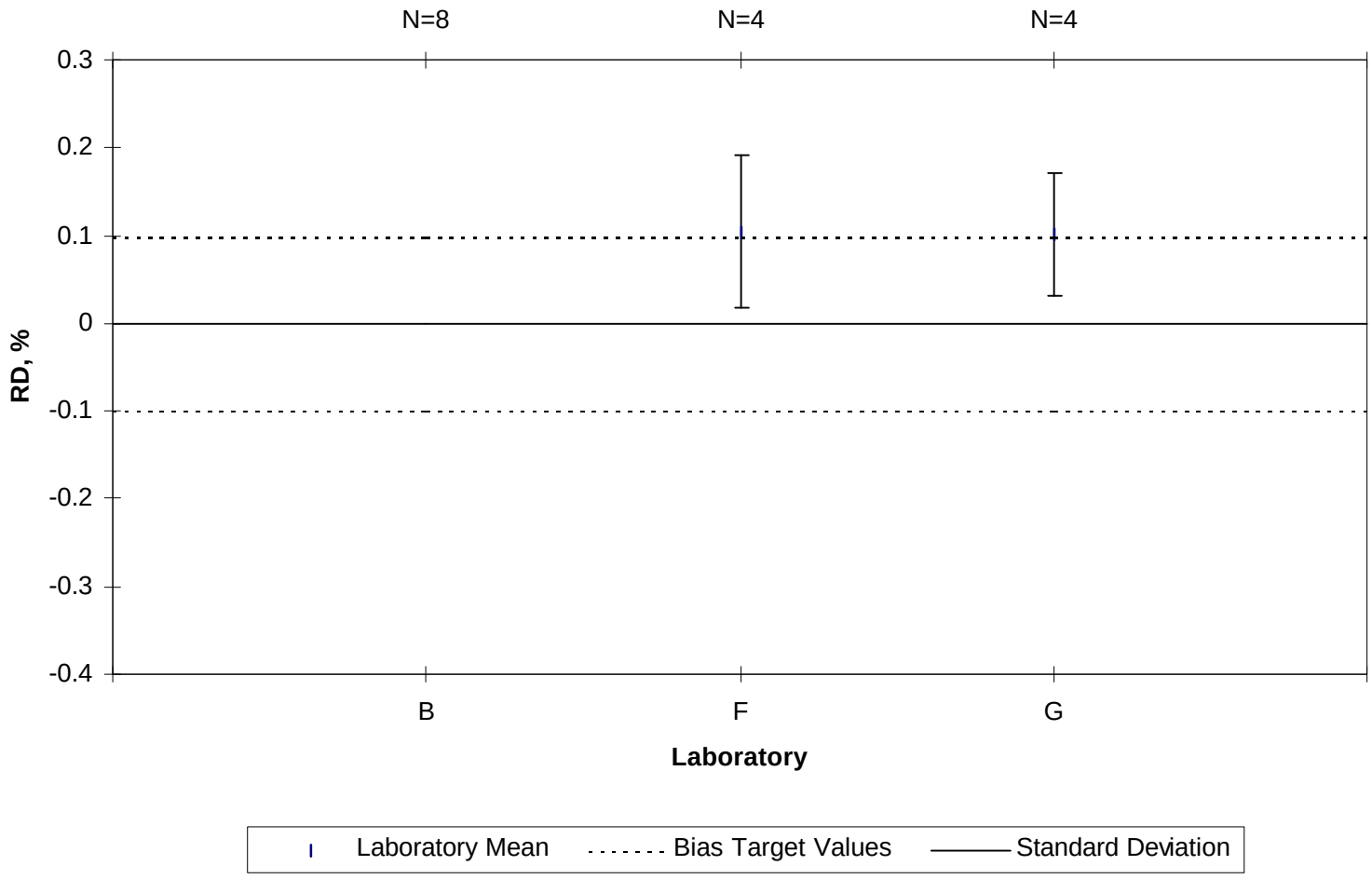
- Los Alamos National Laboratory
- New Brunswick Laboratory
- Savannah River Site
- NMCC, Japan

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



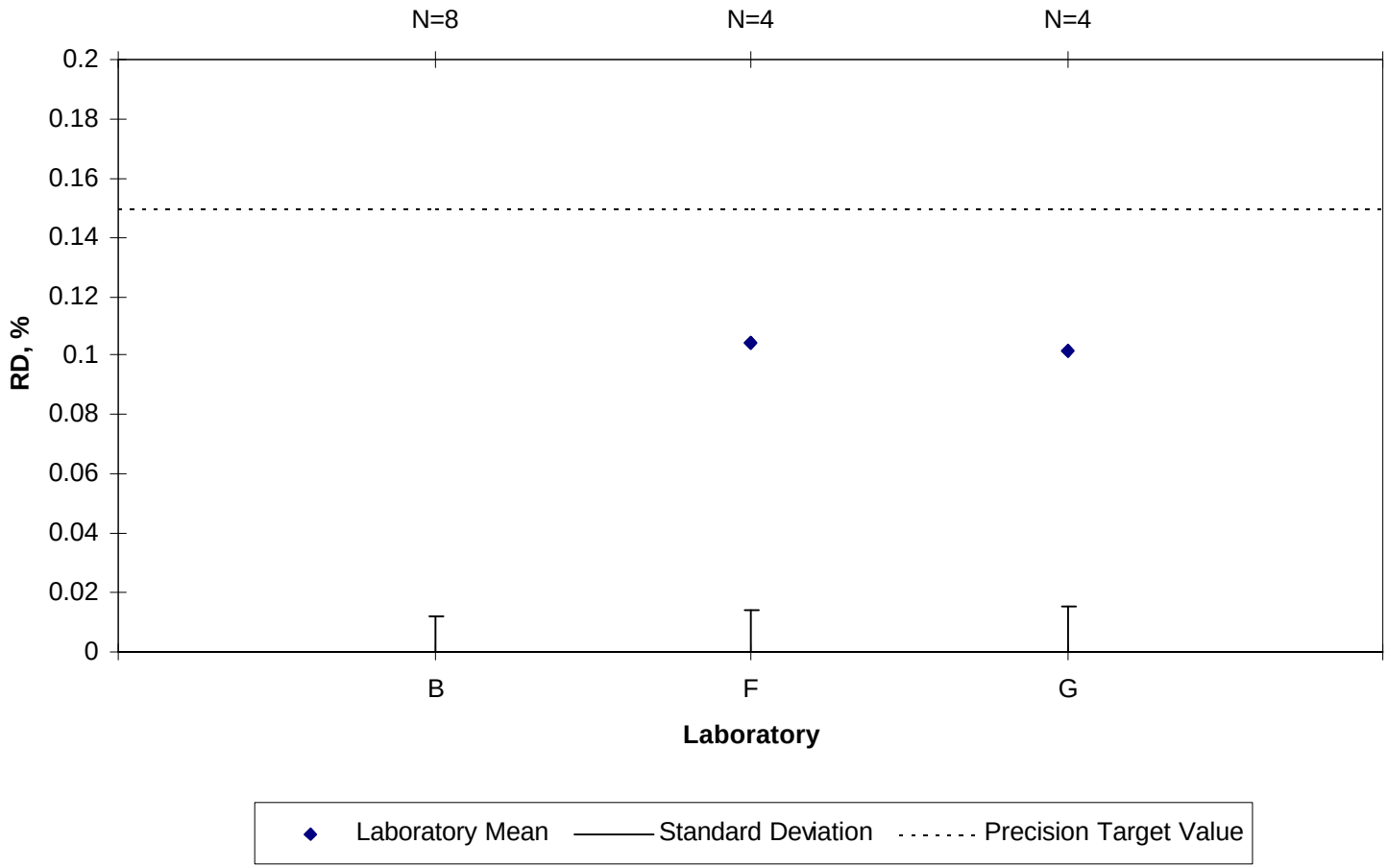
New Brunswick Laboratory

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

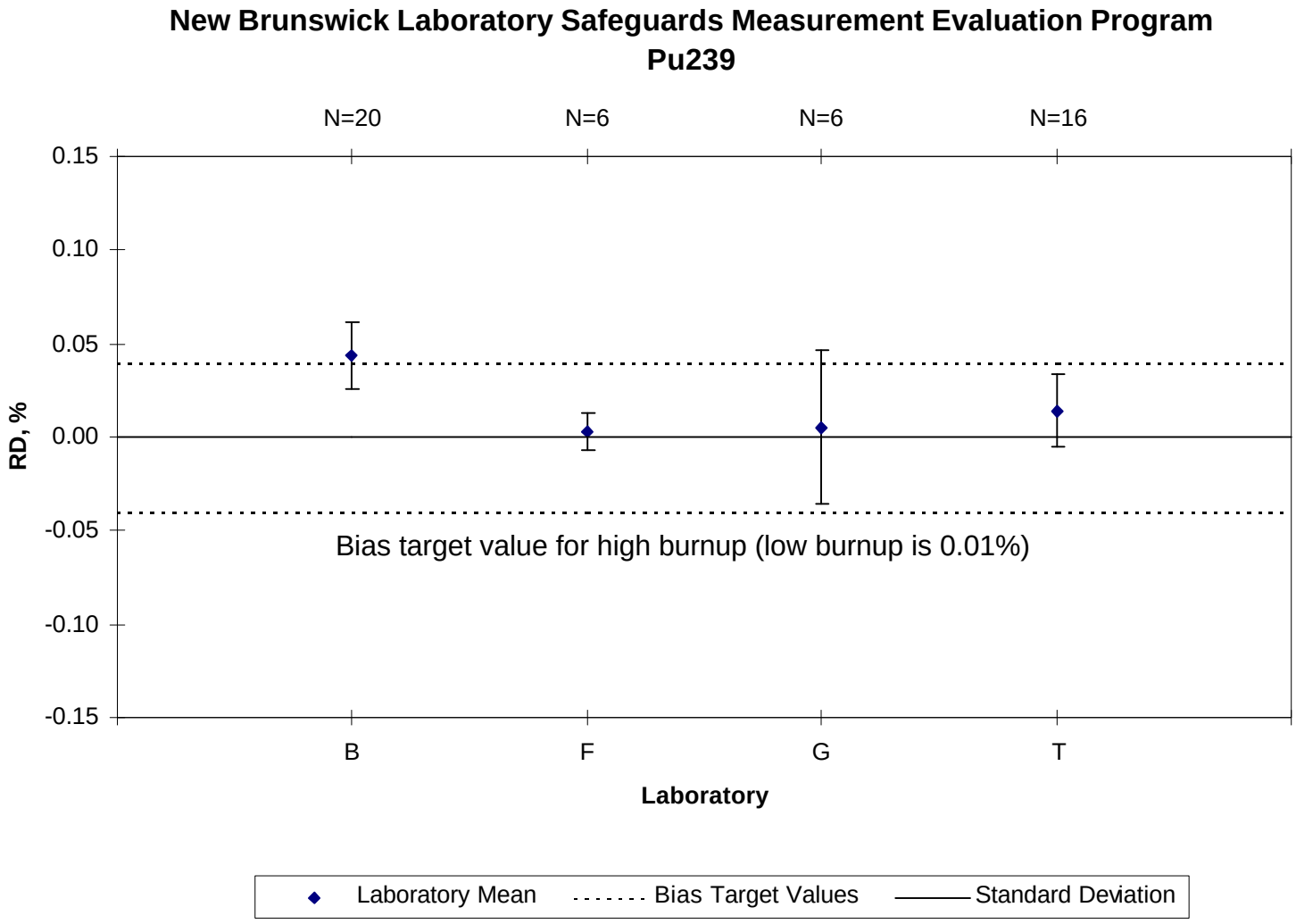


New Brunswick Laboratory

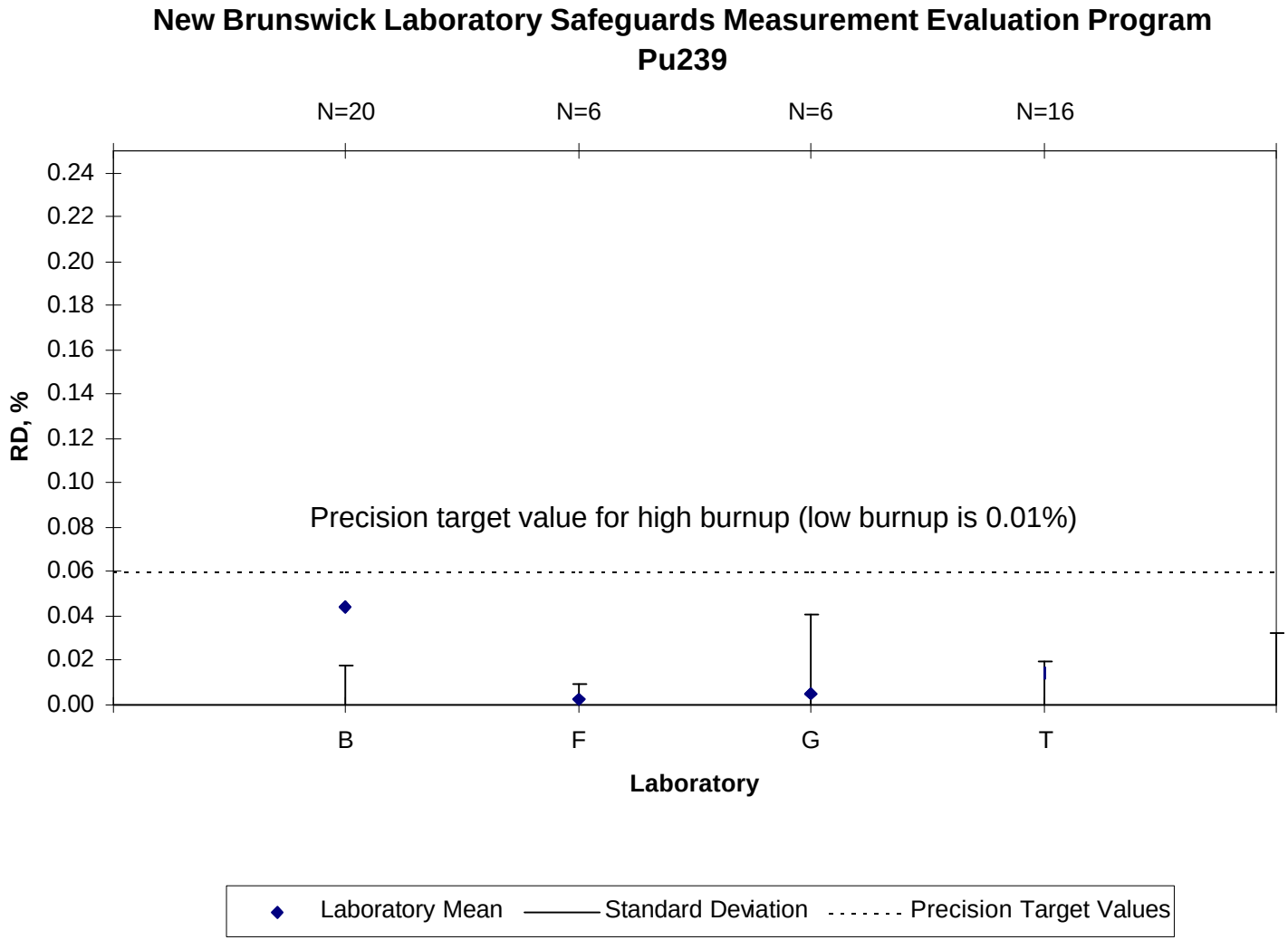
New Brunswick Laboratory Safeguards Measurement Evaluation Program
Pu Sulfate - Percent Pu



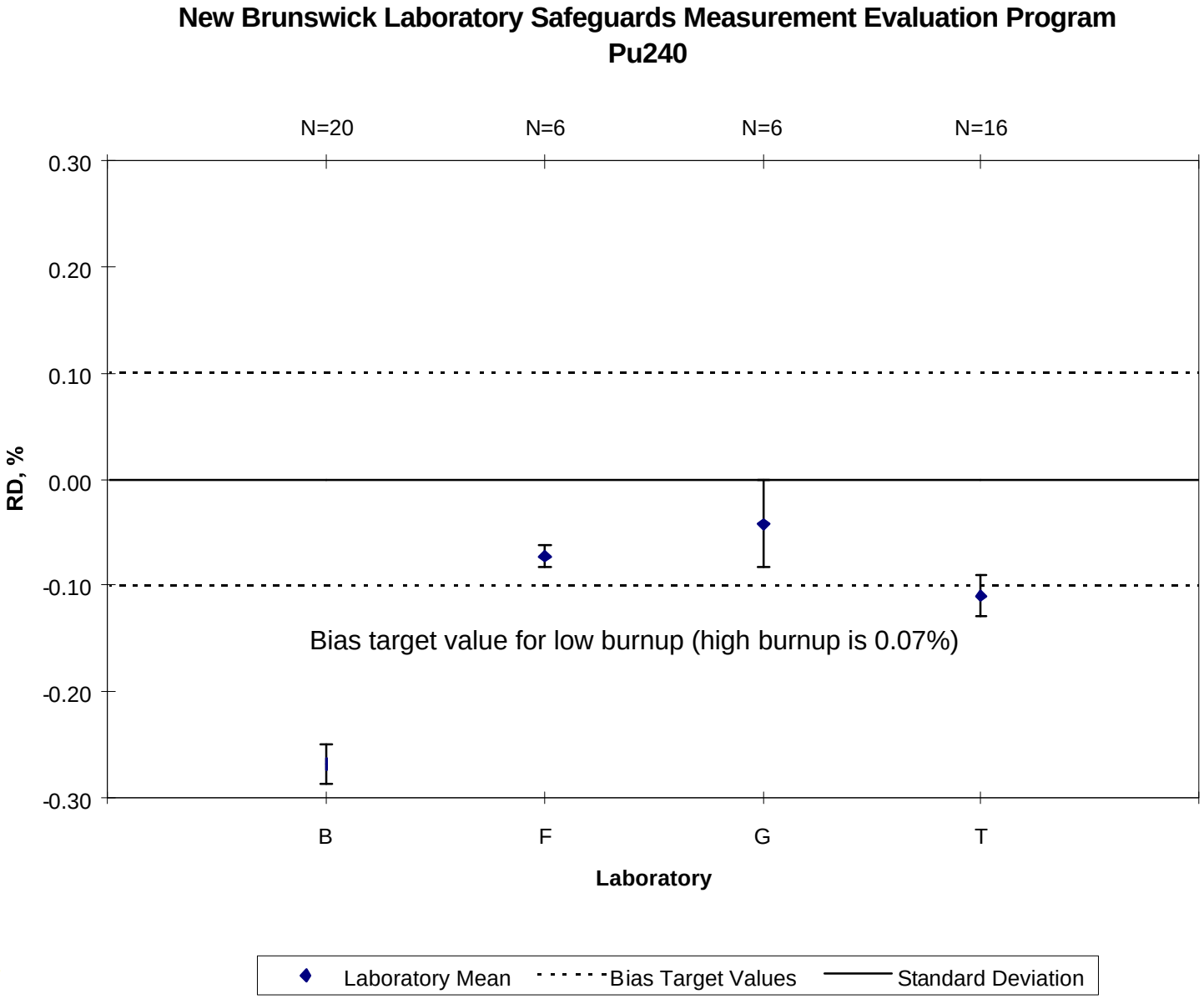
New Brunswick Laboratory



New Brunswick Laboratory

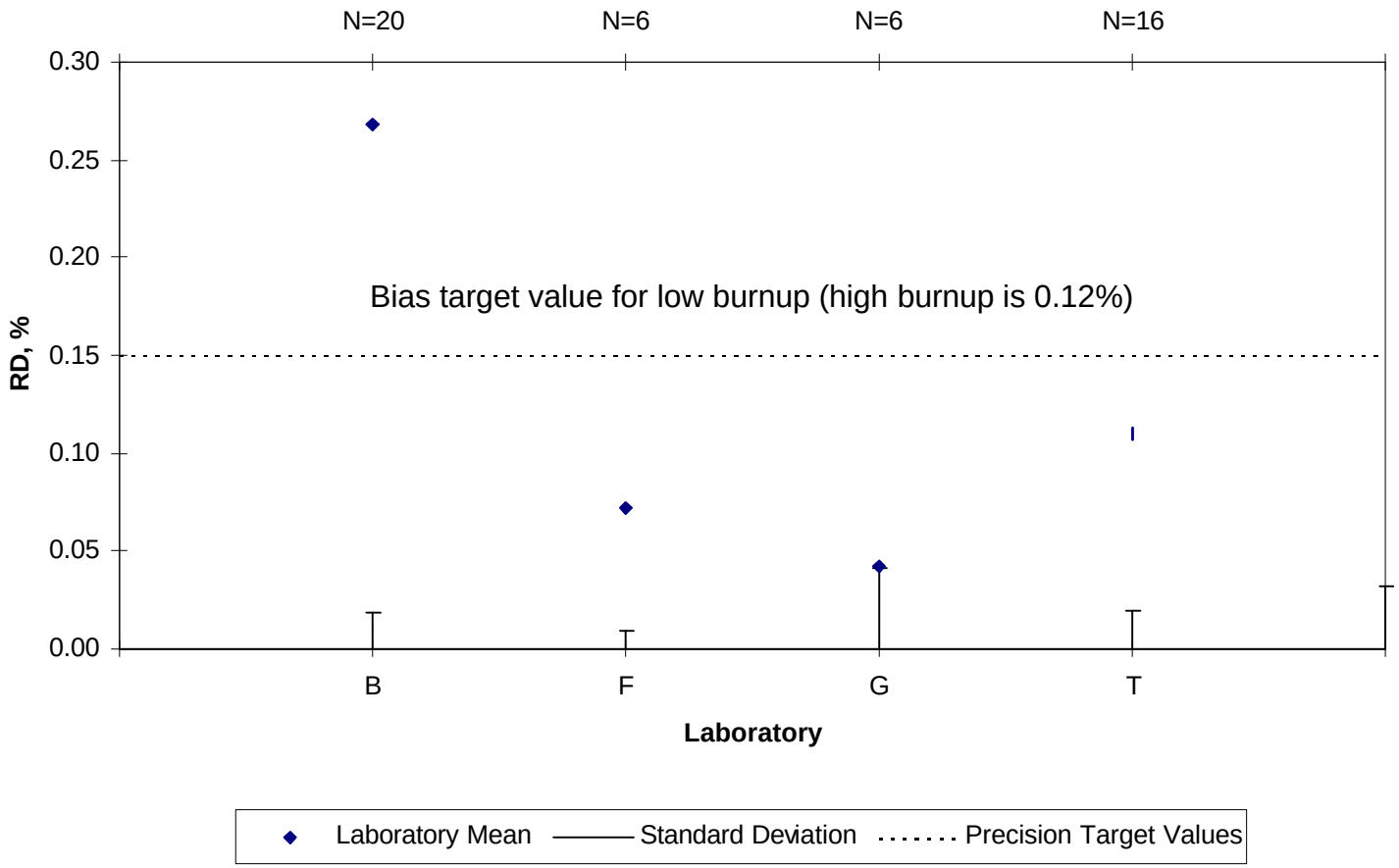


New Brunswick Laboratory



New Brunswick Laboratory

New Brunswick Laboratory Safeguards Measurement Evaluation Program
Pu240



New Brunswick Laboratory

Overview of the Davies-Gray Analysis at SRS

Kimberly Carter - Chemist

Savannah River Site

Aiken, South Carolina

June 23, 2002

Outline

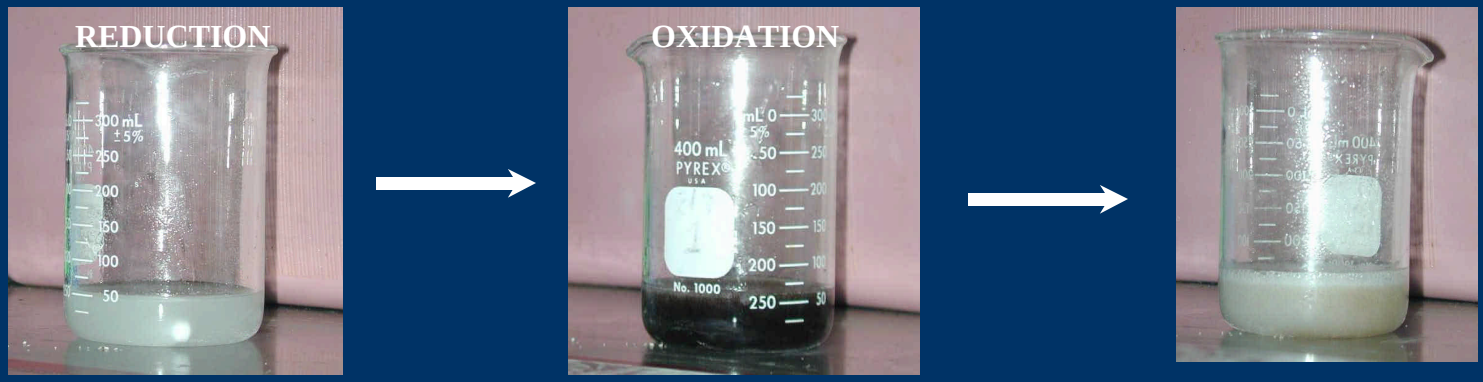
- Method Description
- Calculations
- Limitations
- Quality of Data
- Method Measurement Control
- Samples
- Method Problems
- Future Testing

Method Description

- Gravimetric redox titration
- Method use:
  - Uranium (enriched) nuclear material accountability measurements
  - Characterization of uranium standards for the Quality Control & Standards group
  - Validation of other uranium measurement methods
  - Participation in the NBL-SMEP

Method Description

- Fumed to near dryness in sulfuric acid
- Aliquot re-dissolved in a sulfamic acid-strong phosphoric acid mixture
- Ferrous sulfate reduces U^{+6} to U^{+4}
- Nitrate ion with the Mo $^{+6}$ catalysts oxidizes the excess ferrous ion



Method Description

- Uranium remains in the reduced state
- $K_2Cr_2O_7$ titrant with a vanadium catalyst is used to oxidize U^{+4} to U^{+6}

- Endpoint occurs between 590 - 610mV
- Isotopic weights to calculate equivalent weight of uranium & density results are needed before we submit results to the customer

Calculations

$$\mathbf{U,\ ^{mg}/_{g} =}$$

$$\frac{(K_2Cr_2O_{7wt\ before} - K_2Cr_2O_{7wt\ after}) \times K_2Cr_2O_7\ \text{Conc.} \times \text{U g eq. wt.} \times 0.9993}{S_{wt} \times 49.0302}$$

$$\mathbf{U,\ ^{g}/_{l} =}$$

$$\text{U, } ^{mg}/_{g} \times \text{Sample density}$$

Where,

49.0302 = gram wt. eq. of the $K_2Cr_2O_7$
0.9993 = air buoyancy correction for sample densities
119.02 = U gram wt. eq. for normal or depleted U
Isotopics used to determine the U gram wt. eq. for enriched U

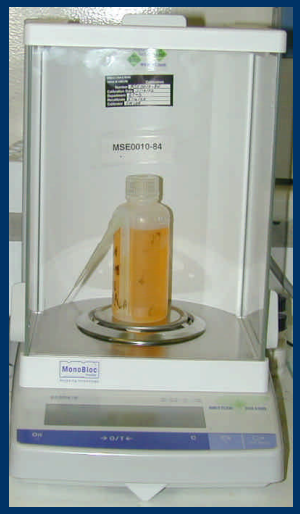
*Note: Calculations performed via LIMS

Method Limitations

- Interferences - silver, mercury, palladium, & platinum
- Lab temperature must be between 21°C to 26°C

Quality of Data

- **Method used for uranium solutions > 0.5 g/L U**
- **Precision: +/- 0.25% @ 95% C.L.**



Method Measurement Control

- Controlled by Laboratory Information Management System (LIMS)
- Only qualified technicians can run blind standards
- An “in control” blind synthetic result is required on each shift by analyst to report sample analyses by the method

Measurement Control

- Out of control “LOCKS” the method from reporting results.
  - A blind greater than 3 s.d. from known value
  - Two consecutive blinds between 2 and 3 sd
- A blind within 2 s.d. will “unlock” the method.
- Method control charts are also reviewed by chemists weekly for other adverse trends.

Measurement Control

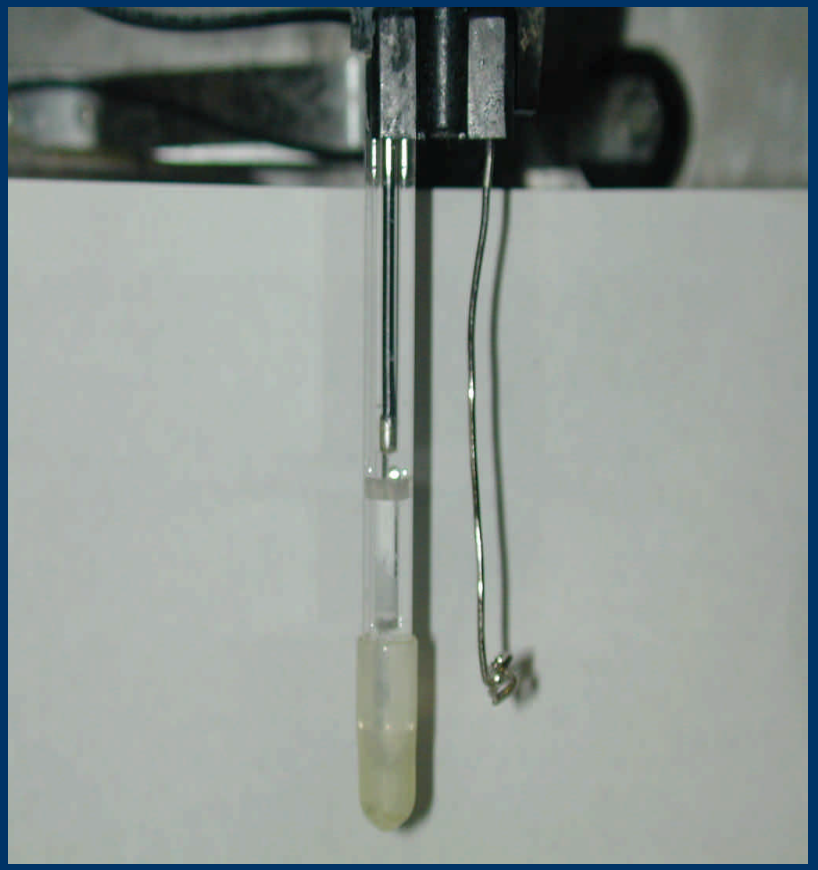
- Accountability measurements are bracketed with blind standards.

Advantages of Davies-Gray

- Extremely precise method for determining uranium concentration
- Inexpensive
- Quicker turn around time than Isotopics by Mass Spec

Method Issues

- Shift operation
- Operator technique (i.e. consistency, over-titration)

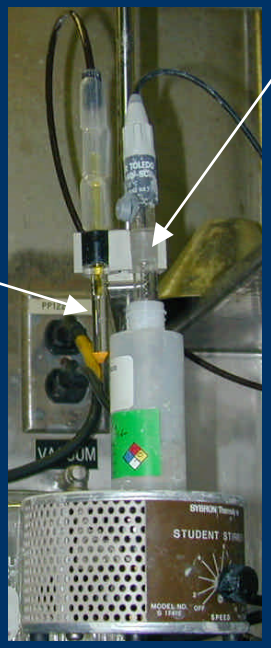


Future Plans

- **Autotitration**

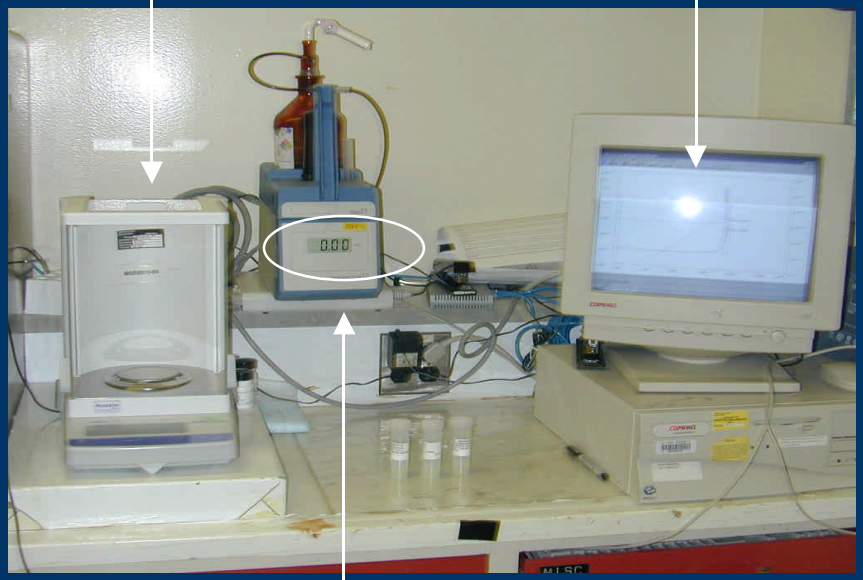
Combination Pt/
calomel electrode

Titration
Tip



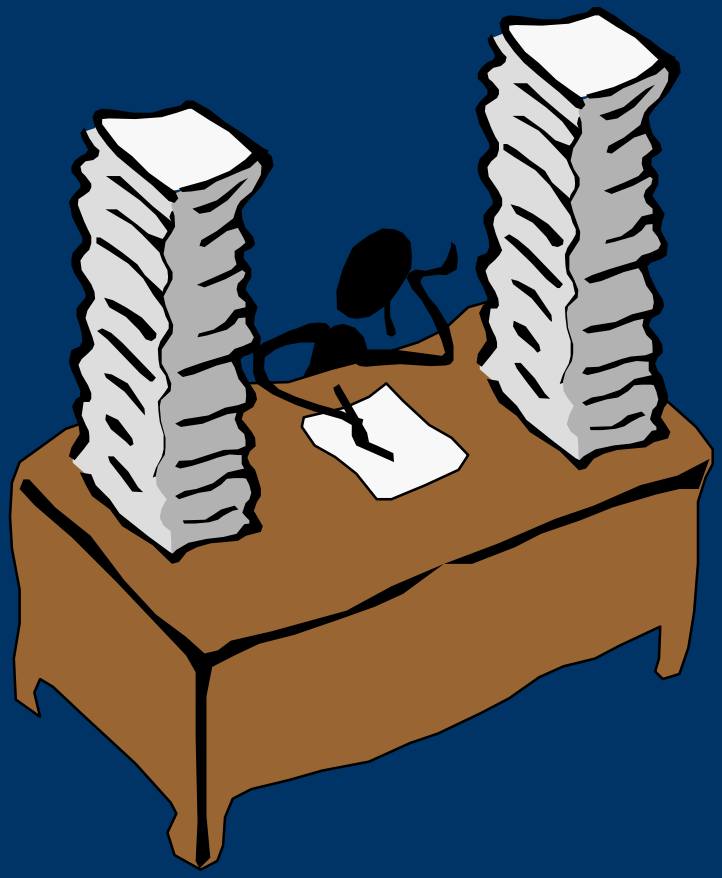
Balance

Computer w/
Multi-Tasking Software



Piston Burette TITRONIC T110 w/
Interchangeable Unit

Any Questions?



Acknowledgments

- Alma Stiffin - NBL
- Kimberly Johnson-Miller - NBL
- Iris Frank - NBL
- Jaime Murphy - SRS

Interlaboratory Comparison for the Analysis of Plutonium and Uranium

Mika Yoshida Sumi, Toru Suzuki, Hideo Kobayashi
Japan Nuclear Cycle Development Institute

1. Abstract

The analytical laboratory of the Plutonium Fuel Center (PFC) of Japan Nuclear Cycle Development Institute (JNC) is participating in a regular interlaboratory comparison program for the analysis of plutonium and uranium. This program has been carried out four rounds a year since 1994 based on the framework of international safeguards implementation for direct use plutonium handling laboratories in Japan. Destructive analysis (DA) samples for safeguards purpose with various material types are utilized for the program. After preparation of the verification samples for transportation to the safeguards laboratories, additional aliquots are taken for delivery to the accountancy analysis laboratories of JNC for parallel analysis. The results of the individual laboratories are compared to the grand mean of all participating laboratories.

The program allows for a fast turn-around of results and any follow-up actions can be taken place immediately. This interlaboratory comparison program has been successful in its contribution to monitor, improve and assure the quality of the analytical measurements.

2. Introduction

Until 2000 fiscal year, 24 rounds of interlaboratory comparison analysis have been carried out by a collaborative effort of the IAEA, Safeguards Analytical Laboratory (IAEA-SAL) and Section for Statistical Analysis at department of SG, the domestic safeguards laboratory (NMCC-SAL) and the JNC laboratories involved in measurements for nuclear material accountancy. IAEA recommended the interlaboratory comparison analysis program to PFC to improve the performance of analysis in 1994 and has organized the program till now. The primary purpose of this program is to promptly detect any measurement biases as well as to provide those laboratories an external quality assurance (QA) tool for maintaining and improving a high level of quality in nuclear material accountancy analysis.

The analytical results of plutonium and uranium concentrations and their isotope compositions of randomly selected DA samples are commonly utilized for the interlaboratory comparison evaluation and for the inspection evaluation. All the analytical results from the participants are collected at IAEA for evaluation, and then its report is sent to each laboratory for its own further evaluation. PFC conducts a survey on analytical procedures and calculations whenever the program detects irregular data in the evaluation. Also long term biases are an important subject to be checked based on re-evaluating the accumulated data.

This paper discusses the entire scheme of the interlaboratory comparison program and specifically PFC's experience gained through the past six years of the program.

3. Scheme of the program

The entire scheme of the interlaboratory comparison program and overviews of the evaluation at the PFC laboratory is described in this section.

3.1 Materials used

Various types of materials are taken as DA samples during routine inspections for safeguards purposes from plutonium handling facilities of JNC such as Plutonium Fuel Production Facility (PFPF), Plutonium Fuel Fabrication Facility (PFFF), Plutonium Conversion Development Facility (PCDF) and Tokai Reprocessing Plant (TRP), and some of those samples are utilized for this program. The samples have wide variety of Pu/U content and uranium enrichment and they have no reference value, because the samples originate from process materials. Therefore, the evaluation should be relative due to regarding the grand mean of all the participants for each sample as reference value. On the contrary the program brings advantage for each participant that it enables wide range evaluation of plutonium concentration.

The materials used in the program are shown in Table 1.

Table 1. Materials used in the program

Sampling Facility	TRP	PCDF	PFPF			PFFF
Sample	Pu nitrate solution	1:1MOX powder	FBR "MONJU" pellet	FBR "JOYO" pellet	PuO_2 powder	ATR "FUGEN" pellet
Pu/U ratio	1/0	1/1	1/3	1/3	1/0	1/20
U-235 [wt%]		0.2 - 4.3	0.3	18		0.7

3.2 Participating laboratories

When the program was launched in 1994, four JNC laboratories, as mentioned above, and two safeguards laboratories (IAEA-SAL and NMCC-SAL) were the participants of the program. However, PFFF has left the program since the end of 1996, when the PFPF laboratory expanded its analysis capability to take over PFFF material accountancy samples. Also PCDF has left participation in this program since 1997 because TRP has taken over the PCDF material accountancy samples into its laboratory.

At present, two safeguards laboratories and two laboratories in JNC (PFPF and TRP) are members of the program.

3.3 Sample preparation and distribution

IAEA inspectors randomly select sample for the interlaboratory comparison analysis from DA samples taken during safeguards inspections at the JNC plutonium handling facilities before sample treatment.

The Pu containing samples have to undergo following special sample treatments before they can be shipped to the safeguards laboratories as so called Type-A quantities.

a. Pu nitrate solutions are diluted on a weight basis

b. Samples of PuO_2, 1:1MOX and FBR pellet are dissolved in nitric acid with traces of fluoric acid.

c. Pellets of ATR, because of their low Pu concentration, are crushed and small sub-samples are weighed into so-called BC-4 vial.

In general, replicated aliquots containing about 3mg of Pu each are sent for analysis. In the case of samples from a. and b., these are taken into penicillin vials and evaporated for analysis before shipment.

This procedure of sample preparation is digested in Fig.1.

Safeguards laboratories analyze these DA samples as usual verification samples but the results are regarded not only for verification but also for the interlaboratory comparison.

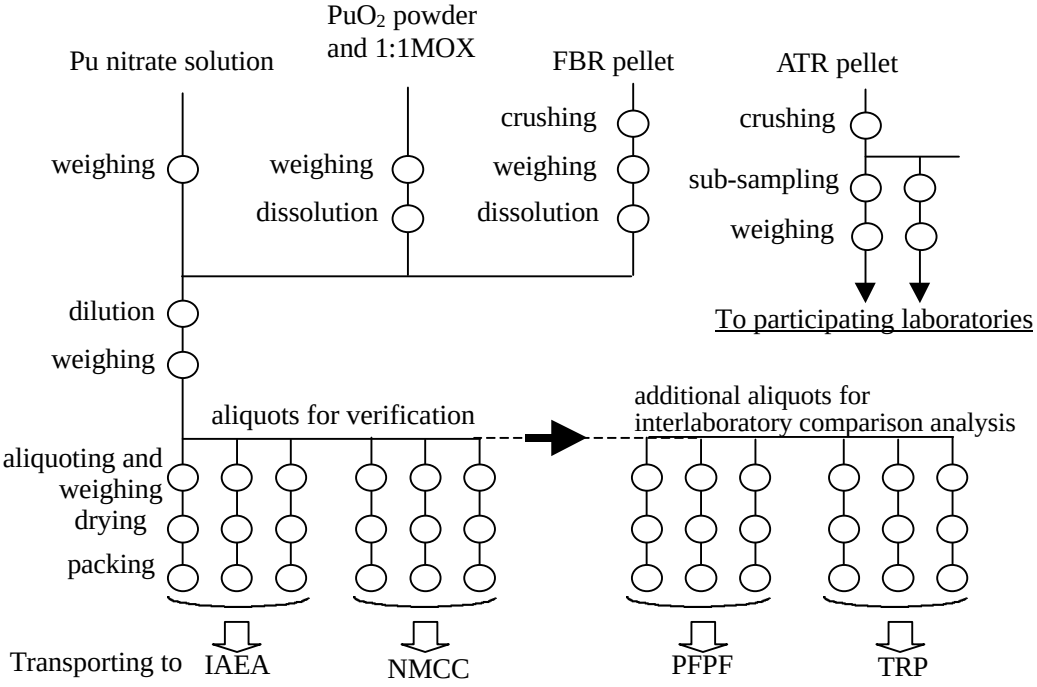


Fig. 1 Sample preparation and distribution procedure

3.4 Analytical method in PFC laboratories

PFFF used to apply titration procedure for plutonium concentration analysis, while PFPF used to apply controlled potential coulometry at the beginning of interlaboratory comparison program. In order to improve measurement accuracy, the PFPF laboratory has introduced IDMS in 1997, with sufficient capacity to also accommodate all accountancy samples from PFFF. Currently IDMS is the only analytical method to cover all the accountancy purpose analysis in PFC.

Large Size Dried (LSD) spikes for IDMS prepared from plutonium metal reference material in the laboratories of PFC and prepared by AEA Technology in UK have been utilized [1].

After anion ion exchange, purification and separation, plutonium and uranium isotope composition are measured with thermal ionization mass spectrometry, followed by plutonium-238 correction with measurement results of alpha-spectrometry. The total evaporation method for IDMS has been applied since 1999.

3.5 Flow of the information of analysis results

The analysis results obtained in the JNC laboratories and the NMCC-SAL are gathered at the Japan Safeguards Office (JSGO), and then forwarded to the IAEA.

IAEA Section for Statistical Analysis evaluates the whole analysis results gathered from all participant laboratories including the IAEA-SAL. The results of IAEA's evaluation are reported to the participant laboratories once a year.

The following analysis results associating with relevant information are summarized in every interlaboratory comparison analysis report.

- plutonium concentration (mgPu / vial) and isotopic composition
- uranium concentration (mgU/vial) and enrichment
- analytical techniques
- analysis date

PFC carries out the statistical evaluation for its own purposes additionally, based on the accumulated information reported from IAEA till then.

3.6 IAEA's evaluation

The data from all participants are provided to the IAEA as plutonium and uranium concentration. After decay correction of the plutonium concentration and isotopic composition to the sampling date, these analytical results for each laboratory are compared with the grand mean of all the corresponding data of participants. The comparison result is reported to the participants once a year.

3.7 PFC's evaluation

In addition to the IAEA evaluation from the viewpoint of safeguards implementation, statistical evaluation is carried out by PFC for its own purposes that involve detecting irregular data and possible long term bias of PFC.

Detecting irregular data has been performed by checking the difference between each of the PFC laboratory and the average value of two safeguards laboratories against the control limit calculated based on the "1993 International Target Value (ITV) for Uncertainty Components in Fissile Isotope and Element Accountancy for the Effective Safeguarding of Nuclear Materials"[2].

The latter, estimation of long-term bias, has been performed by checking the interlaboratory analysis results obtained in a year whether the corresponding annual average of the difference against the grand mean has statistically significance compared with 2 confidence level, calculated from the ITV.

4. PFC's experience in the interlaboratory comparison

PFC's evaluation of 24 rounds interlaboratory comparison analysis results is summarized in this section. The contribution of the interlaboratory comparison analysis at PFC quality control is also described.

4.1 Evaluation on irregular data of PFC analysis results

Irregular data can be checked by plotting the relative difference of the PFC analysis results against

the average value of the two safeguards laboratories and comparing the relative difference with the control limit calculated from the ITV. The control chart of Pu concentration analysis is shown in Fig. 2, as an example.

As shown in this figure, a few data being out of or close to control limit have been found. In such cases, the procedure, calibration of equipments, calculation process and repeatability have been checked out. Although no clear reason which causes data being out of control has been identified so far, this kind of action plays the important role to maintain and improve the quality control of analytical measurement.

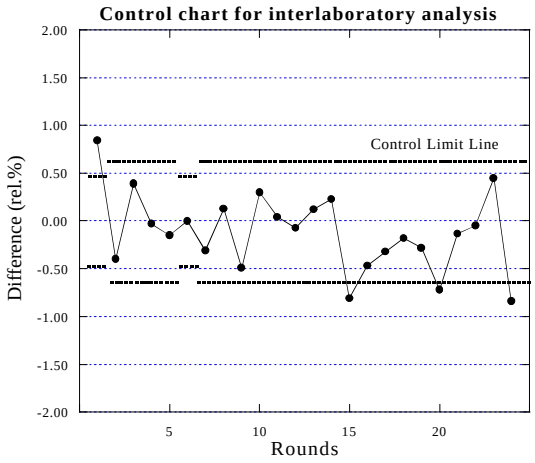


Fig.2 The example of the control chart for interlaboratory analysis result of PFC laboratory

4.2 Evaluation on possible bias of PFC analysis results

Fig. 3 to 5 show annual average differences from the grand mean with 95% confidence limit, while the control limit lines indicate 2 sigma limits calculated from the ITV.

No significant biases have been observed so far in plutonium concentration, uranium concentration nor isotopic ratio measurements. Therefore, it can be concluded that the material accountancy analysis in PFC laboratory is maintained in good condition. This result also shows that the LSD spikes prepared in PFC and AEA are of sufficient quality and accuracy. However, small negative biases on the Pu238/239, Pu242/239 ratios and uranium concentration might be existed.

In 1999 the total evaporation method has been introduced for the mass spectrometric measurements. The comparison of the Pu240/239 ratio in the year 2000, for instance, could suggest improvement in measurement precision. The intercomparison program will be used for further confirmation on this improvement.

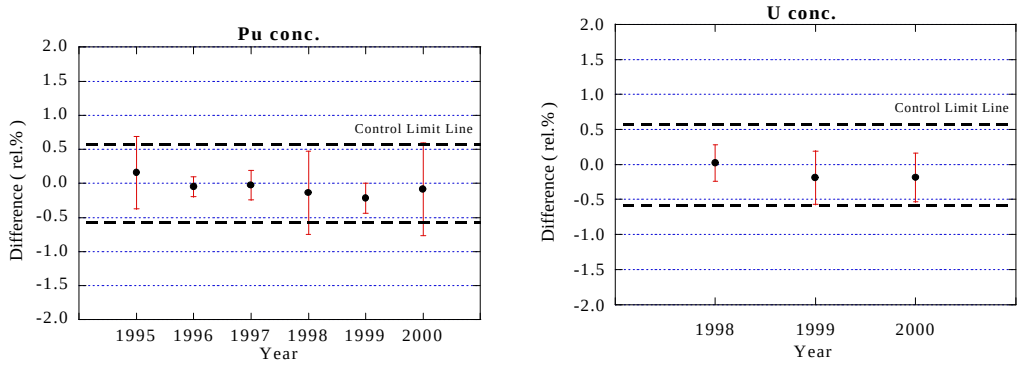


Fig.3. Difference against the grand mean on plutonium and uranium concentration results

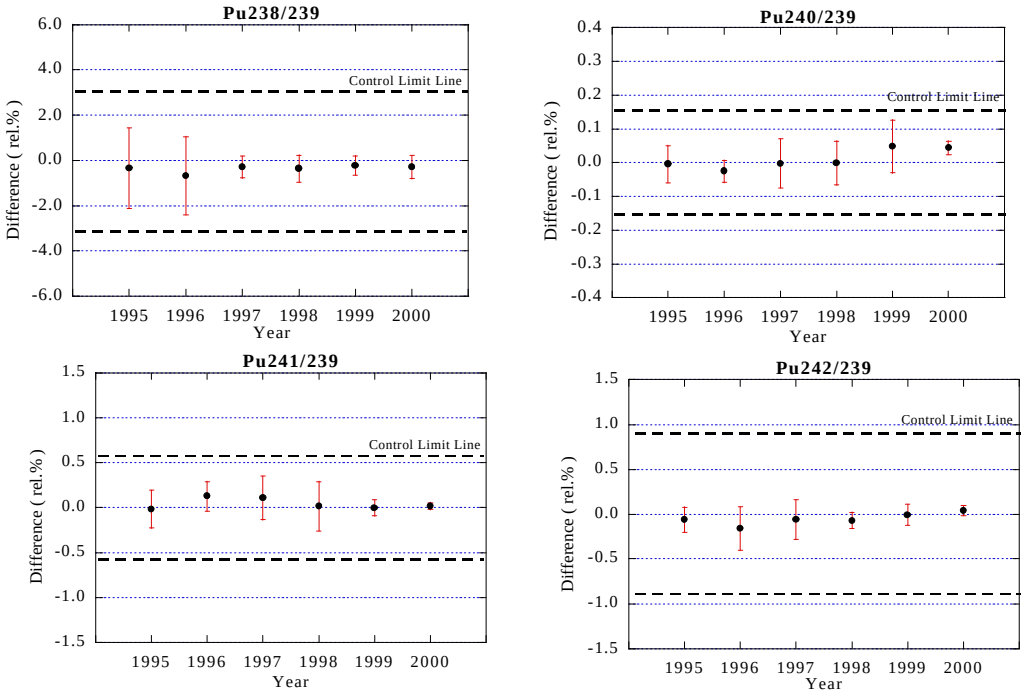


Fig.4. Difference against the grand mean on plutonium isotope ratios

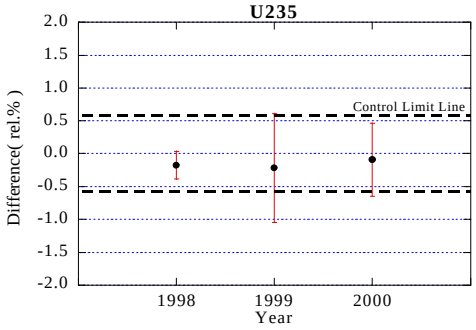


Fig.5. Difference against the grand mean on uranium enrichment results

4.3 Contribution to quality control of material accountancy analysis

It is not easy for Japanese laboratories to participate in interlaboratory comparison programs on plutonium analysis hosted by foreign laboratories due to the difficulty for overseas transportation of

nuclear material samples. This program provides a precious opportunity for quality control of material accountancy analysis to the Japanese laboratories. Only a little additional effort during sample treatment and distribution is required, while samples represent actual process materials. Those features are beneficial for PFC as well as to the other participants. PFC can evaluate its analysis performance efficiently by participating in the program. PFC makes use of the program as an important tool for confirming the reliability of nuclear material accountancy analysis.

Because of insufficient experience on IDMS in the past, procurement of reliable LSD spikes and establishment of IDMS operation were the crucial issues in the initial stage of IDMS introduction into PFC. This program also played an important role to confirm the reliability of LSD spikes and contributed to improve the analyst's skill.

A continuation of the program certainly has the merit for the participants in view assuring that long-term biases do not exist.

4.4 Examination on reliability of the interlaboratory comparison method

The program has disadvantage to utilize samples without reference values. Therefore maintaining the reliability of grand mean is essential to ensure the performance of the program. In order to evaluate the reliability, dispersions of results of four laboratories for each sample type of each round are calculated. As an example, results on plutonium concentration are summarized in Table 2. Each dispersion is indicating stable condition except two cases. The first case is plutonium nitrate solution in 1997 and the second case is ATR pellet from 1998 through 2000. In the first case there was a relatively large fluctuation among the average values of the laboratories, which could indicate a sample treatment problem. In the latter cases differences might be caused by sample oxidation and/or change of moisture content because the sample is powder of a crushed pellet. In this case, it is important to check the validity of the sample treatment and shipment procedure.

Table 2 Dispersions of the grand mean for plutonium concentration for each sample type in an interlaboratory comparison year

(CV%)

Sample type	1995	1996	1997	1998	1999	2000
Pu nit.	0.34	0.09	0.57	0.08	0.18	0.08
1:1MOX	0.38	0.09	0.21	0.14	0.14	0.16
FBR pellet	0.21		0.22	0.27	0.14	0.43
ATR pellet	0.09	0.19	0.06	0.51	0.48	0.48
PuO_2		0.11				

5. Conclusion

Experiences gained so far through the interlaboratory comparison program are summarized as follows.

- This interlaboratory comparison program, in which the DA samples prepared for safeguards are utilized, can provide a precious opportunity for quality control of material accountancy analysis to the Japanese plutonium handling laboratories.

- In the sample treatment and distribution procedure, this program involves little additional effort and costs but ensures that the samples represent actual process materials and that their measurement is conducted by routine analytical procedures. It provides a mean to monitor the accuracy of routine analysis.
- The program also allows for checking the quality and accuracy of the LSD spikes which are a potential source of systematic errors.
- Monitoring long term biases is effective in maintaining the accuracy of material accountancy.

6. Acknowledgement

The work of interlaboratory comparison is the results of the effort of many people of the IAEA, NMCC and JNC.
The authors would like to thank E. Kuhn of IAEA Tokyo office, Y. Kuno of IAEA-SAL and S. Deron of former IAEA-SAL for there active support for interlaboratory comparison analysis and evaluation and valuable discussion. We also wish to thank T. Ohtani of JNC for his helpful comments and review of this paper.

7. References

[1] M. YOSHIDA, T. SUZUKI, H. KOBAYASHI, T. OHTANI, Experience of Determination of Plutonium and Uranium Contents in MOX Fuel by IDMS. 42nd INMM Annual Meeting, California, USA, July 2001
[2] E. KUHN, et. al., 1993 International Target Value for Uncertainty Components in Fissile Isotope and Element Accountancy for the Effective Safeguarding of Nuclear Materials. IAEA STR-294

joint research centre
EUROPEAN COMMISSION

irmm
Institute for Reference Materials and Measurements

External QC programmes at IRMM

REIMEP / NUSIMEP

EUROPEAN COMMISSION
JOINT RESEARCH CENTRE

joint research centre
EUROPEAN COMMISSION

REIMEP

- REIMEP 15 completed (and reported in 2001)
  - Uranium isotopics in UF_6
  - 9 laboratories provided results
  - Excellent results generally for $^{235}U/^{238}U$ ratio
  - Good results for $^{236}U/^{238}U$ ratio; some discrepancy for $^{234}U/^{238}U$ (still to be resolved)

joint research centre
EUROPEAN COMMISSION

Next REIMEP campaigns

1 Pu isotopic abundances

2 Simulated dissolver tank solution

Both being planned (perhaps later 2002)

joint research centre
EUROPEAN COMMISSION

Pu isotopic abundances

- Planned to prepare 4 solutions, each with microgram amounts of Pu. Transport as solutions or dried nitrates
- ^{239}Pu enrichments from 60 - 90%
- Either by certification of existing material
- Or mixing certified starting materials (probably this option)

irmm
nstitute for Reference
Materials and Measurements

joint research centre
EUROPEAN COMMISSION

Simulated dissolver tank solution

- U/Pu ca 100:1
- U concentration ~ 150 mg/g
- Uranium non-natural
- Pu equivalent to high or medium burnup
- Measure U/Pu ratio, U and Pu isotopic abundances

joint research centre
EUROPEAN COMMISSION

QC campaigns: general thoughts

Internal QC: repeated measurements on a well-known sample

Informs us if the measurement is consistent over time

joint research centre
EUROPEAN COMMISSION

Round Robin

⇒ Uses non-certified material

⇒ Easy to set up

⇒ Shows relation between labs

⇒ No information regarding correctness of measured values nor their uncertainties

joint research centre
EUROPEAN COMMISSION

Certification by selected labs

⇒ Attempts to address last point

⇒ Much extra work required

⇒ Runs into problem of combining replicates in a certification

⇒ Works best with a clear hierarchy of labs

Use of certified materials

⇒ Time consuming

⇒ Restricted choice of materials

⇒ Gives ‘absolute’ value for comparisons

⇒ A lot of weight for accreditation!

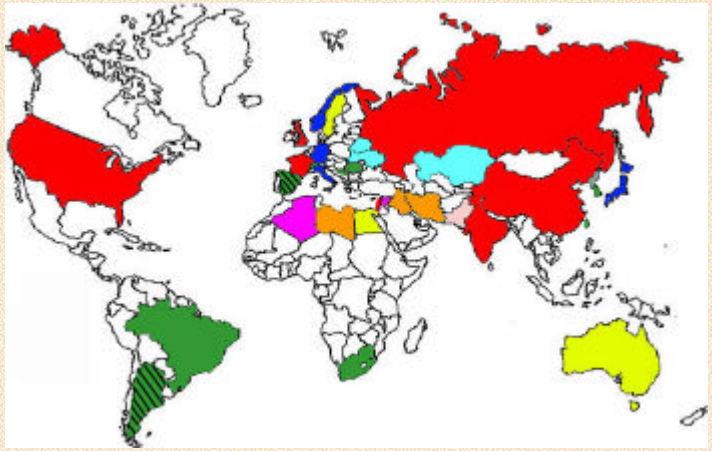
NBL Reference Material Production Status

Jon W. Neuhoff
New Brunswick Laboratory
U.S. Department of Energy
June 23, 2002

NBL Has the Responsibility

- For the preparation, certification, and distribution of Certified Reference Materials (CRMs) for nuclear material safeguards measurements
- To provide CRMs which ensure traceability of DOE nuclear materials measurements to a national and international measurements database
- To collaborate with other laboratories in the preparation and characterization of Working Reference Materials (WRM) for the purpose of ensuring national and international comparability of nuclear material safeguards measurements

NBL Domestic Versus International Focus



- Primary focus is on meeting the needs of U.S. DOE and NRC-licensed facilities
- However, this is balanced with needs stemming from the increased importance of nuclear nonproliferation, international safeguards, MPC&A, and IAEA support activities

NBL CRMs - Current Availability

- 53 NBL CRMs are available for purchase within the following categories:
  - Plutonium & Uranium Assay CRMs (42-A (1-4), 112-A, 113-B, 116, 122, 125-A, 126, 129, 145)
  - Thorium & Uranium Impurity CRMs (66(1-7), 123(1-7), 124(1-7)
  - Plutonium & Uranium Isotopic CRMs (111-A, 113-B, 116, 122, 125-A, 126, 128, 130, 135, 136, 137, 138, U0002, U005-A, U010, U015, U020-A, U030-A, U100, U150, U200, U350, U500, U750, U800, U850, U900, U930-D, U970)
  - Uranium NDA CRMs (146, 149, 969)
  - Thorium & Uranium Ore CRMs (1-A, 3-B, 4, 5, 101-A, 102-A, 103-A, 104-A, 105-A, 106-A, 107-A, 108-A, 109-A, 110-A)

NBL CRMs - Archived

- Certain NBL CRMs are archived (many are CRMs that were replaced/repackaged):
  - Plutonium & Uranium Assay CRMs (15, 17, 17-A, 18, 42, 97, 112, 113, 113-A, 114, 115, 120, 125, 127)
  - Uranium Impurity CRMs (98(1-7), 121)
  - Thorium & Uranium-Thorium Assay CRMs (19, 20, 71, 118, 119)
  - Plutonium & Uranium Isotopic CRMs (111, 113, 113-A, 117, 127, 131, U005, U020, U030, U050, U930, U930a-c, SRM 944, SRM 949, SRM 949a-f)
  - Thorium & Uranium Ore CRMs (3, 3-A, 3-C, 6, 6-A, 7, 7-A, 101, 102, 103, 104, 105, 106, 107, 108, 109, 110)
  - Potassium Dichromate CRM (99)

NBL CRMs - Restricted Sales Policy

- NBL policy is to restrict sale of CRMs due to the following reasons:
  - Low CRM inventory (“endangered species”)
  - Material is a “national resource” that requires strict control to ensure availability for critical applications (e.g., Pu-244)
  - CRM pulled for re-verification of specific attributes

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

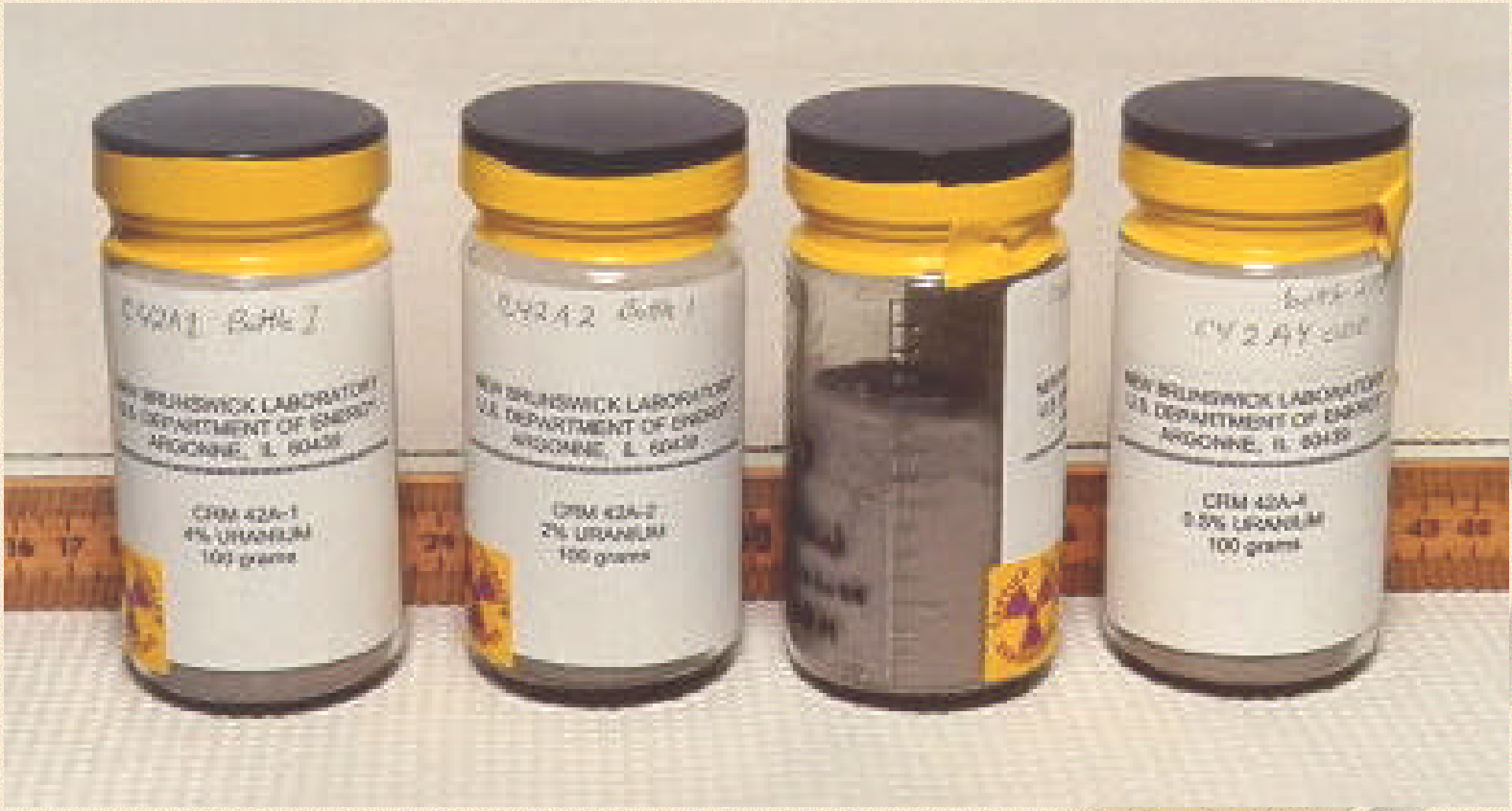
CRM 146 - Enriched Uranium Gamma Spectrometry Standard

New Brunswick Laboratory

CRM 149 - Uranium NDA Standard for Active Well Coincidence Counters

New Brunswick Laboratory

CRM 42-A(1-4) - Normal Uranium Counting Standard



New Brunswick Laboratory

NBL CRMs - Active Projects

- CRM 115 (Depleted Uranium Metal Assay Standard) **(3rd quarter FY 2002)**
- Pu-242 WRMs **(3rd quarter FY 2002)**
- CRM 113-B (Enriched Uranium Hexafluoride Assay and Isotopic Standard) (re-verification of isotopic abundance) **(4th quarter FY 2002)**
- CRM U045 (Uranium Isotopic Standard, 4.5% Enriched) **(4th quarter FY 2002)**
- CRM U630 (Uranium Isotopic Standard, 63% Enriched) **(4th quarter FY 2002)**
- CRM 116 (Enriched Uranium Metal) **(4th quarter FY 2002)**

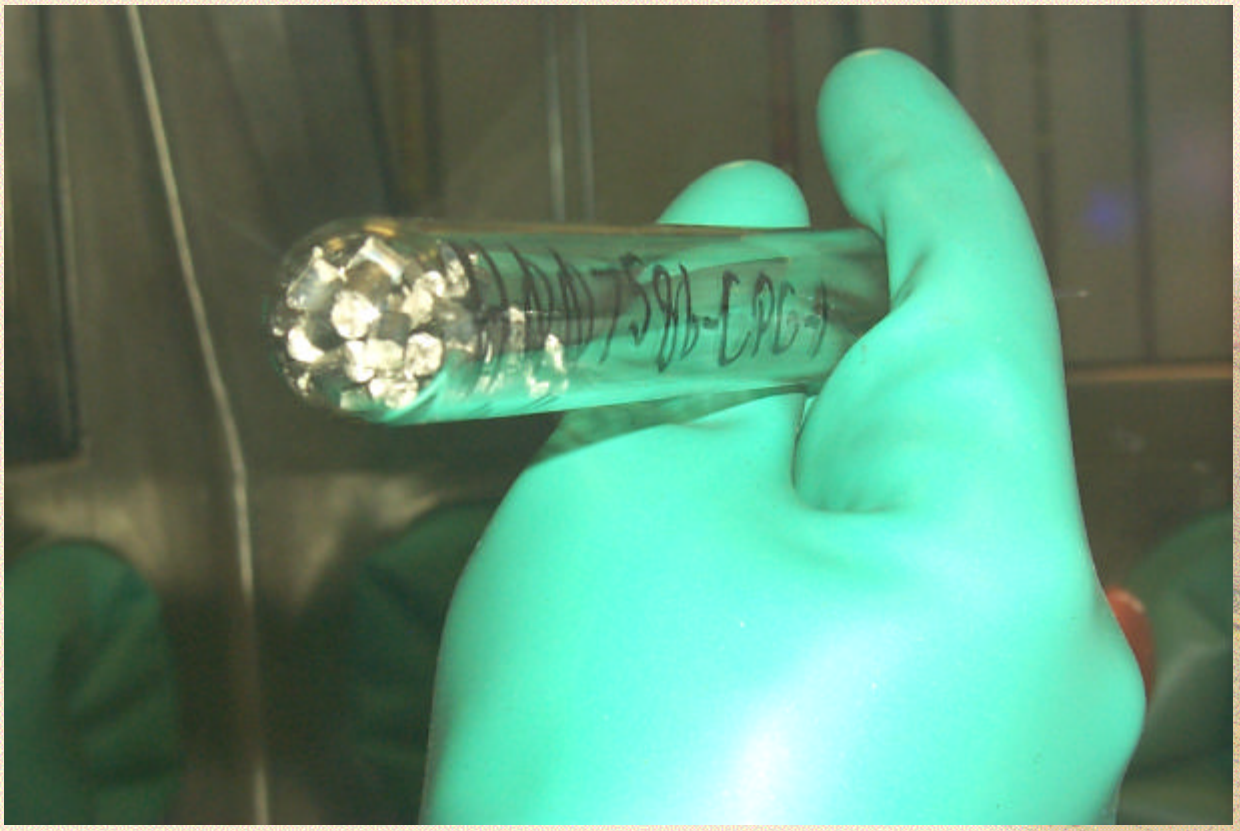
NBL CRMs - Active Projects

- CRM 126-A (Plutonium Metal Assay and Isotopic Standard) **(1st quarter FY 2003)**
- CRM U005-A (Uranium Isotopic Standard, 0.5% Enriched) **(1st quarter FY 2003)**
- CRM 129-A (Normal Uranium Assay and Isotopic Standard) **(2nd quarter FY 2003)**
- NDA Calibration and Standards Support Project **(ongoing)**
- International nonproliferation reference materials **(ongoing)**

CRM 115 - Depleted Uranium Metal Assay Standard

New Brunswick Laboratory

CRM 126-A - Plutonium Metal Assay and Isotopic Standard



New Brunswick Laboratory

CRM 129-A - Normal Uranium Assay and Isotopic Standard

New Brunswick Laboratory

NBL CRMs - Future Plans for Uranium CRMs

- CRM 17-B (Normal Uranium Tetrafluoride Assay Standard)
- CRM 148 (U-233/U-236 Double Atom Spike Standard)
- CRM 135-A (U-235 Spike in UNH Standard)
- CRM 112-B (Uranium Assay and Isotopic Standard)
- CRM U030-B (Uranium Isotopic Standard, 30% Enriched)
- CRM U010-A (Uranium Isotopic Standard, 1% Enriched)
- CRM U007 (Uranium Isotopic Standard, 7% Enriched)
- CRM U0002-A (Uranium Isotopic Standard, 0.2% Enriched)

NBL CRMs - Future Plans for Uranium CRMs

- CRM U015-A (Uranium Isotopic Standard, 1.5% Enriched)
- CRM U500 (Uranium Isotopic Standard, 50% Enriched)
- CRM 124-A (Uranium Oxide Impurity Standard)
- Uranium Isotope Calibration Mixes
- Californium Shuffler NDA Standard
- Enriched UF_6 Mixes
- Uranium NDA WRMs
- Uranium Environmental WRMs (isotopic, particle)
- Pitchblende Ore-Dunite Mix Assay Standard

NBL CRMs - Future Plans for Plutonium CRMs

- CRM 138-A (Plutonium Isotopic Standard)
- CRM 147 (Plutonium NDA Standard)
- CRM 122-A (Plutonium Oxide Assay and Isotopic Standard)
- CRM 144 (Plutonium Triple Atom Spike)
- CRMs 140-143 (Plutonium Isotopic Standards)
- Plutonium Double Atom Spike
- Plutonium Impurity Standard
- Plutonium NDA Standards
- Plutonium Environmental WRMs (isotopic, particle)

NBL CRMs - Future Plans for Other CRMs

- Mixed Oxide (MOX) Standard
- CRM 66 (Thorium Oxide Impurity Standard)
- Neptunium Oxide Assay Standard
- Am-241/Am-243 Spike Standard
- Am-241 in Pu Standard
- Irradiated material standards
- HTGR standards

Identified CRM Needs

- Plutonium Metal
- Uranium and Plutonium LSD Spikes
- Pu-244, Pu-242, Pu-240, U-233, Cm-244, Cm-248, Am-243 Spikes
- Uranium/Plutonium Double Spikes
- “High burn-up” PuO_2
- Impurities in Plutonium
- 18% Enriched Uranium Metal
- Uranium/Plutonium Particle Reference Materials
- Np-236 and U-236 for Accelerator Mass Spectrometry

THE NEW BRUNSWICK LABORATORY

CALORIMETRY EXCHANGE PROGRAM

Jay Thompson and David T. Baran

Phone: 630-252-2524

jay.thompson@ch.doe.gov

david.baran@ch.doe.gov

CALORIMETRY EXCHANGE PARTICIPATING FACILITIES

- Hanford
- Lawrence Livermore National Laboratory
- Los Alamos National Laboratory
- Rocky Flats Analytical Laboratory
- Savannah River Site

1979 Standard

- 455 g PuO_2
- 400 g Pu
- ^{240}Pu content about 6% of total Pu
- 1 watt heat output
- Characterized but not traceable

CALORIMETRY EXCHANGE
NBL Enhancements

- Calculation and inclusion of Pu mass
- NBL destructive measurements of 6-watt std
- “Official” decay spreadsheet
- Outlier identification
- Instrument-to-instrument ANOVA
- 2000 ITVs
- Interim values for 6 watt standard

BENEFITS OF PROGRAM

- Demonstrate comparability
- Evaluation of measurement systems
- Verification of achievement of performance criteria
- Validation of values used in propagation of variance
- Exchange of information

A Request!

Measure the 6 Watt Standard!

- NBL has received Hanford samples to resolve NBL/LANL coulometry results
- Traceable Working Standard Certificate to follow

More Requests

- Identify new materials
- Electronic submittal of results
  - jay.thompson@ch.doe.gov
- Updated distribution list for the annual report with full addresses

THE NEW BRUNSWICK LABORATORY

CALORIMETRY EXCHANGE PROGRAM

Report/Graph Calculations

Jay Thompson and Dave Baran

Phone: 630-252-2524

jay.thompson@ch.doe.gov

david.baran@ch.doe.gov

Explanation of Calculations

- Same for all parameters
  - 238,239,240,241,Am241, peff, power, mass
- Same format/formula as used by Mound
  - Exception is outlier tests
- Reports deal with “error” (M-A) and “% error” (M-A)/A
- Graphs deal with ratio (M/A)

Report

	XXXX
Mean error	1
Standard deviation	2
Uncertainty in the mean	3
Mean error (%)	4
Standard deviation (%)	5
Uncertainty in the mean (%)	6
Number of points	7
Outliers	8

Lab name

All data, excluding lab declared outliers

Lab declared outliers and NBL determined outliers

New Brunswick Laboratory

Mean Error - item #1

$$error = \text{measured value} - \text{standard value}$$

Standard value is decay corrected to the date of the measurement

$$\text{mean error} = \sum_{i}^{N} \frac{error_i}{N}$$

Just the simple average

Standard Deviation - item #2

$$\text{standard deviation} = \sqrt{\frac{\text{var}(error) * N}{N - 1}}$$

FoxPro calculates population variance, we want the sample variance

Uncertainty in the mean - item #3

Two methods to calculate this

1. Using facility declared σ
2. Statistical treatment, σ_m

$$\text{unc} = \frac{1}{N} * \sqrt{\sum_i^N s_i^2} \qquad \text{or} \qquad \text{unc} = \sqrt{\frac{\text{var}(error)}{N-1}}$$

Error (%) - items #4, #5, #6

$$\text{pcterr} = \frac{\text{measured value} - \text{standard value}}{\text{standard value}}$$

$$\text{mean error (\%)} = \sum_{i}^{N} \frac{pcterr_i}{N}$$

Similar calculations for Standard Deviation (%) and Uncertainty in the mean (%)
Use pcterr instead of error

Outlier determination

- C++ code, OUTLIER
  - performs multiple outlier test (Kurtosis, Dixon, Grubbs, Range, One tail test of each extreme, two tail test of extreme pairs)
  - forces review of data
- Graphical representation of the data
- Consultation with NBL statistician

Graphs - by lab

- Using Mound format
- graphing ratio (M/A)
  - decay corrected to measurement date
- mean and 2 σ limits (quarterly and annual)
- error bars are facility declared uncertainties

Comparison graphs

- NBL format
- mean of ratio (M/A)
- error bars are error in the mean

THE NEW BRUNSWICK LABORATORY

CALORIMETRY EXCHANGE PROGRAM - CY2001

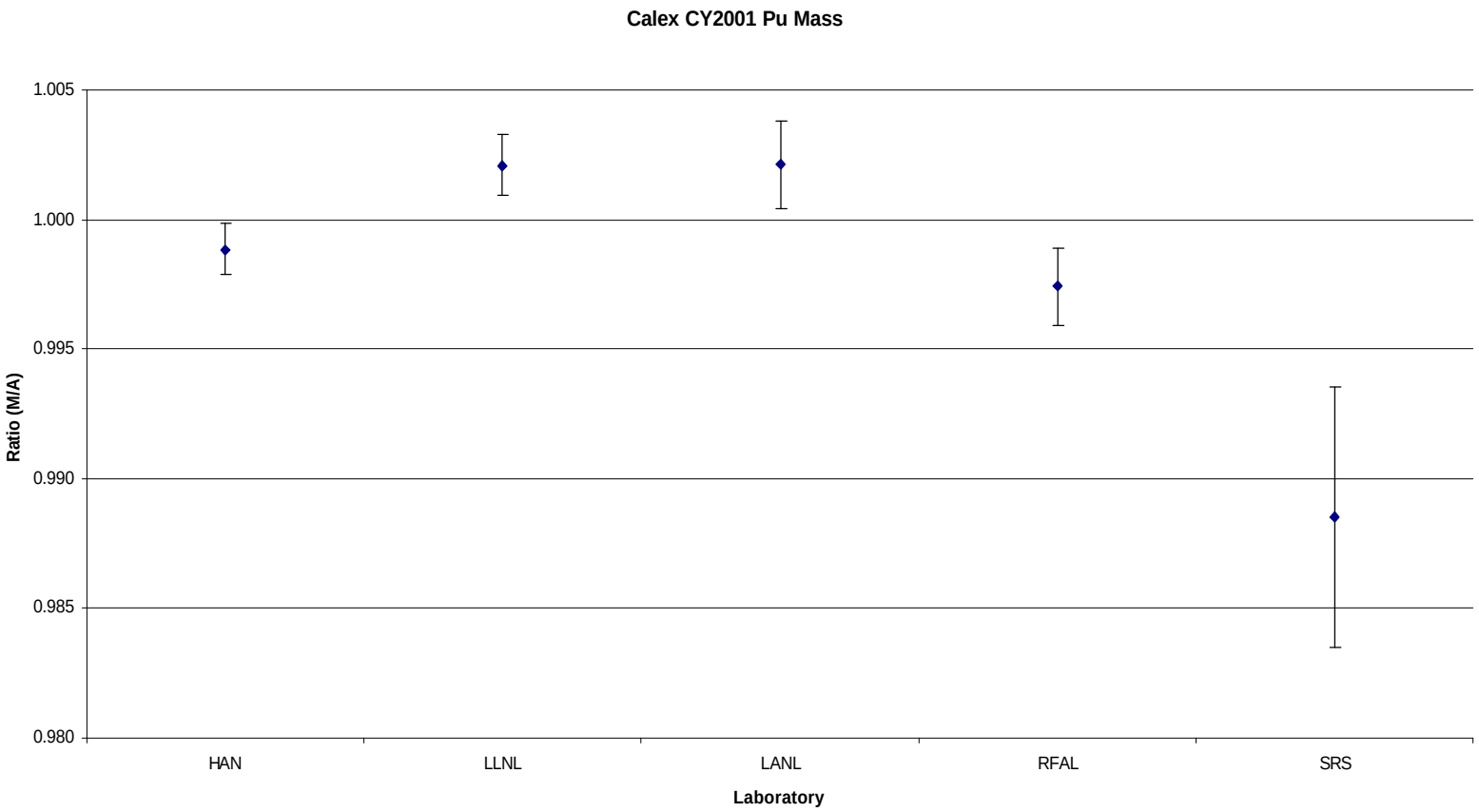
Jay Thompson and Dave Baran

Phone: 630-252-2524

jay.thompson@ch.doe.gov

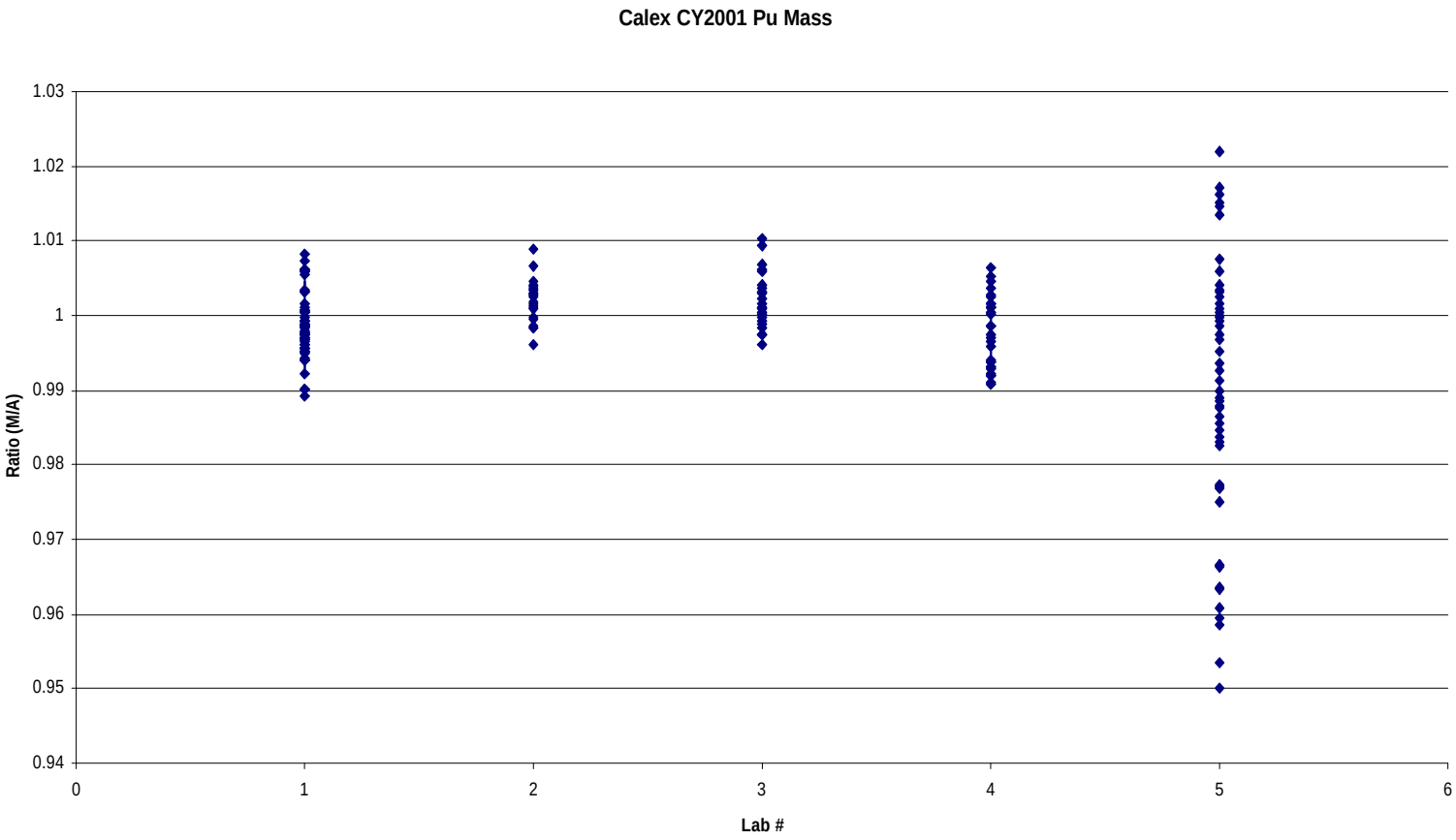
david.baran@ch.doe.gov

Results - Pu mass



New Brunswick Laboratory

Results - Pu mass

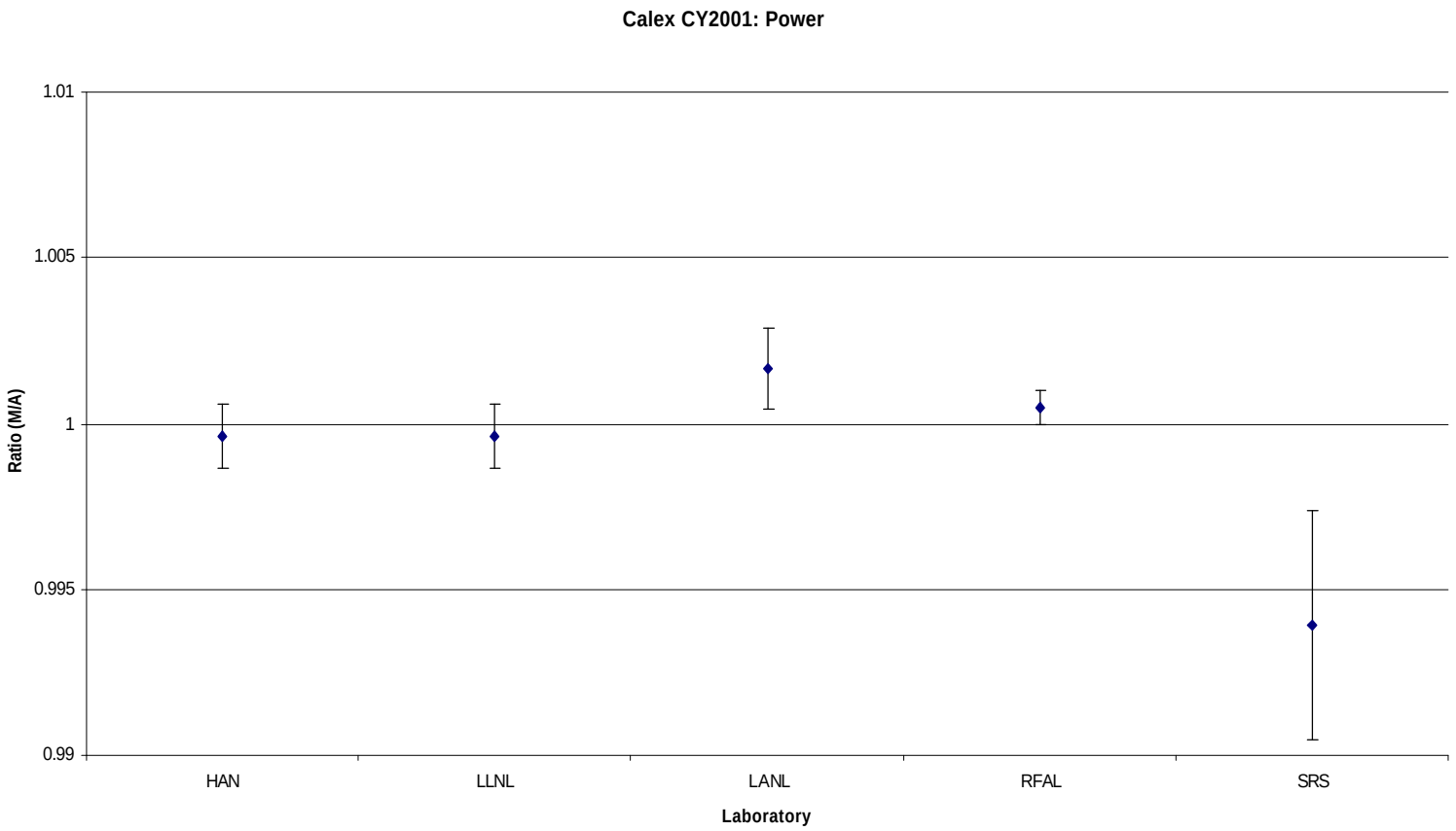


New Brunswick Laboratory

Results - Pu mass

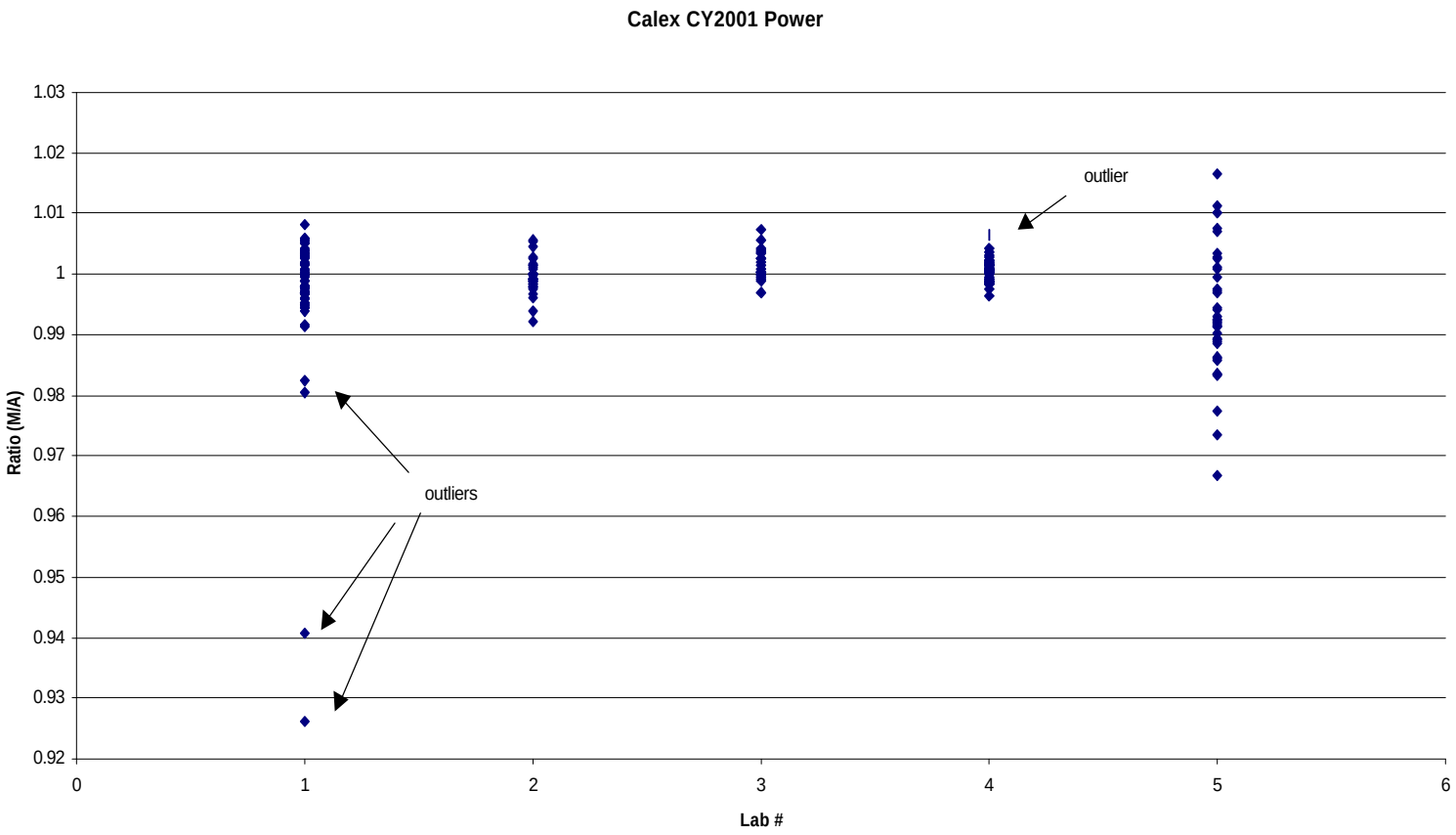
- Lab to lab statistically significant variation (ANOVA)
- 4 labs had multiple systems
  - 1 had system to system variation
- Difficult to do statistics
  - most systems have few data points

Results - Power



New Brunswick Laboratory

Results - Power

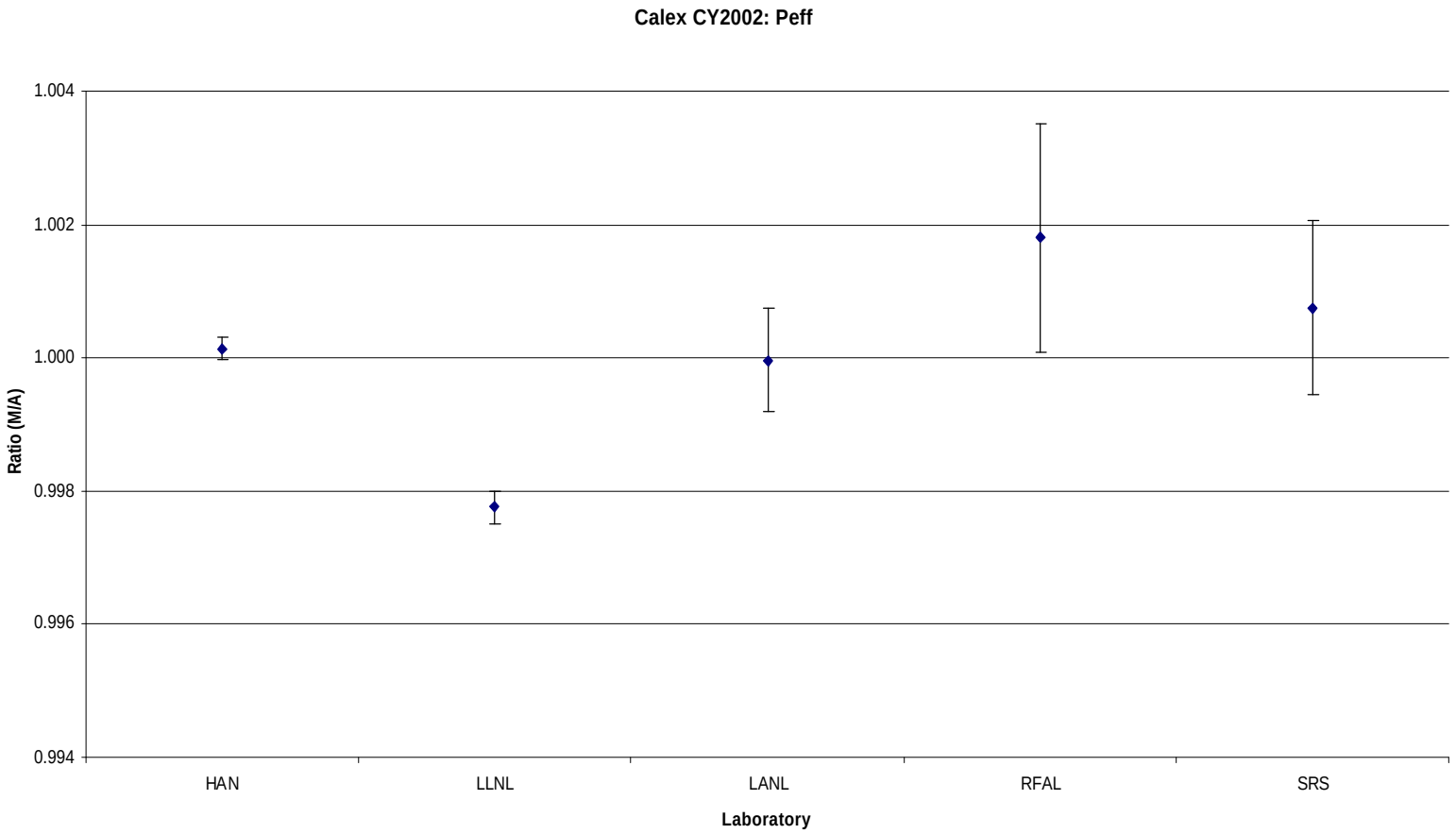


New Brunswick Laboratory

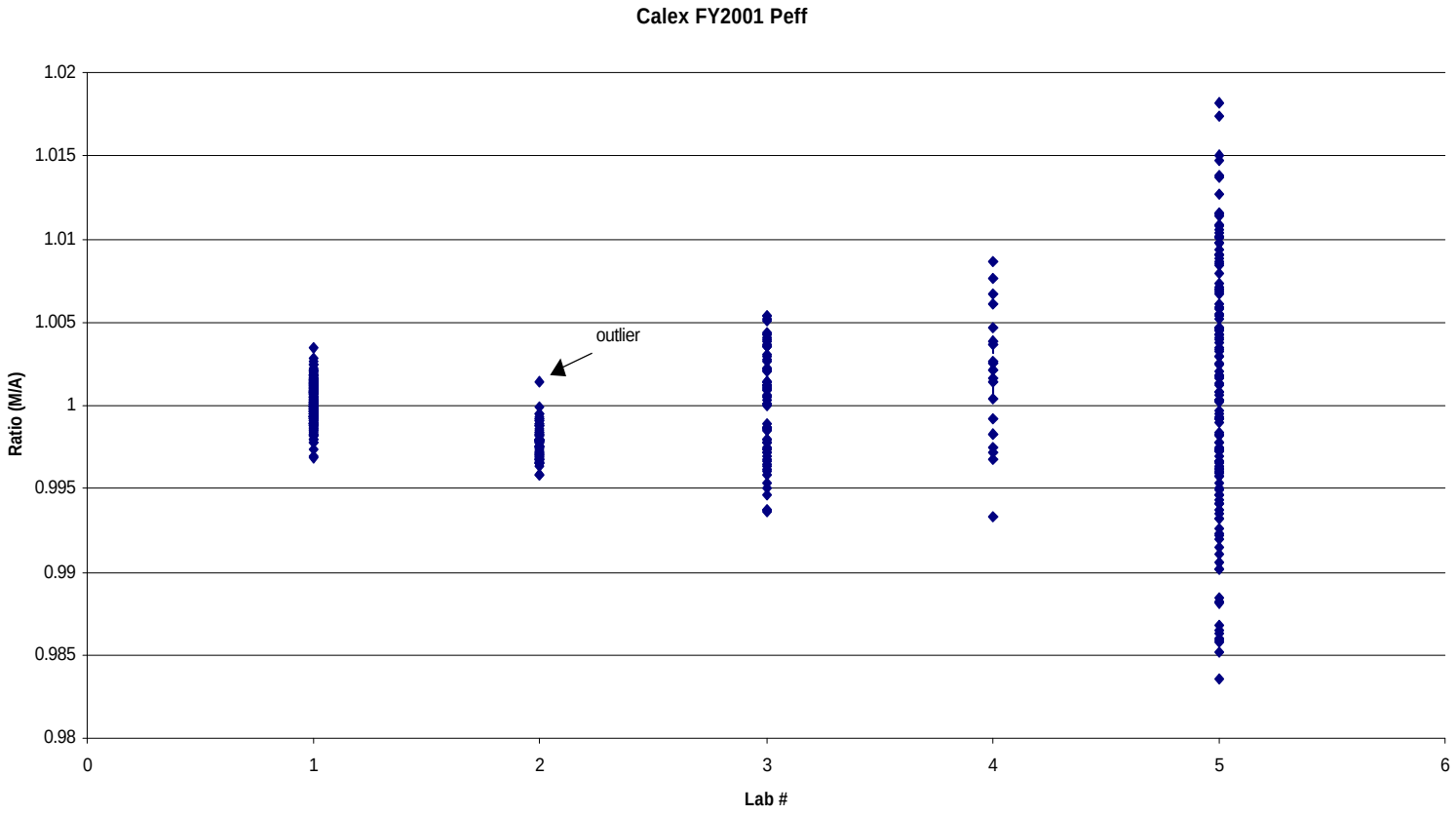
Results - Power

- Lab to lab statistically significant variation
- 40 calorimeters
  - 9 had single data point
  - 11 statistically significantly biased
  - bias range from 0.05% to 0.61%
- 1 lab had statistically significant variation among its systems

Results - P_{eff}



Results - P_{eff}

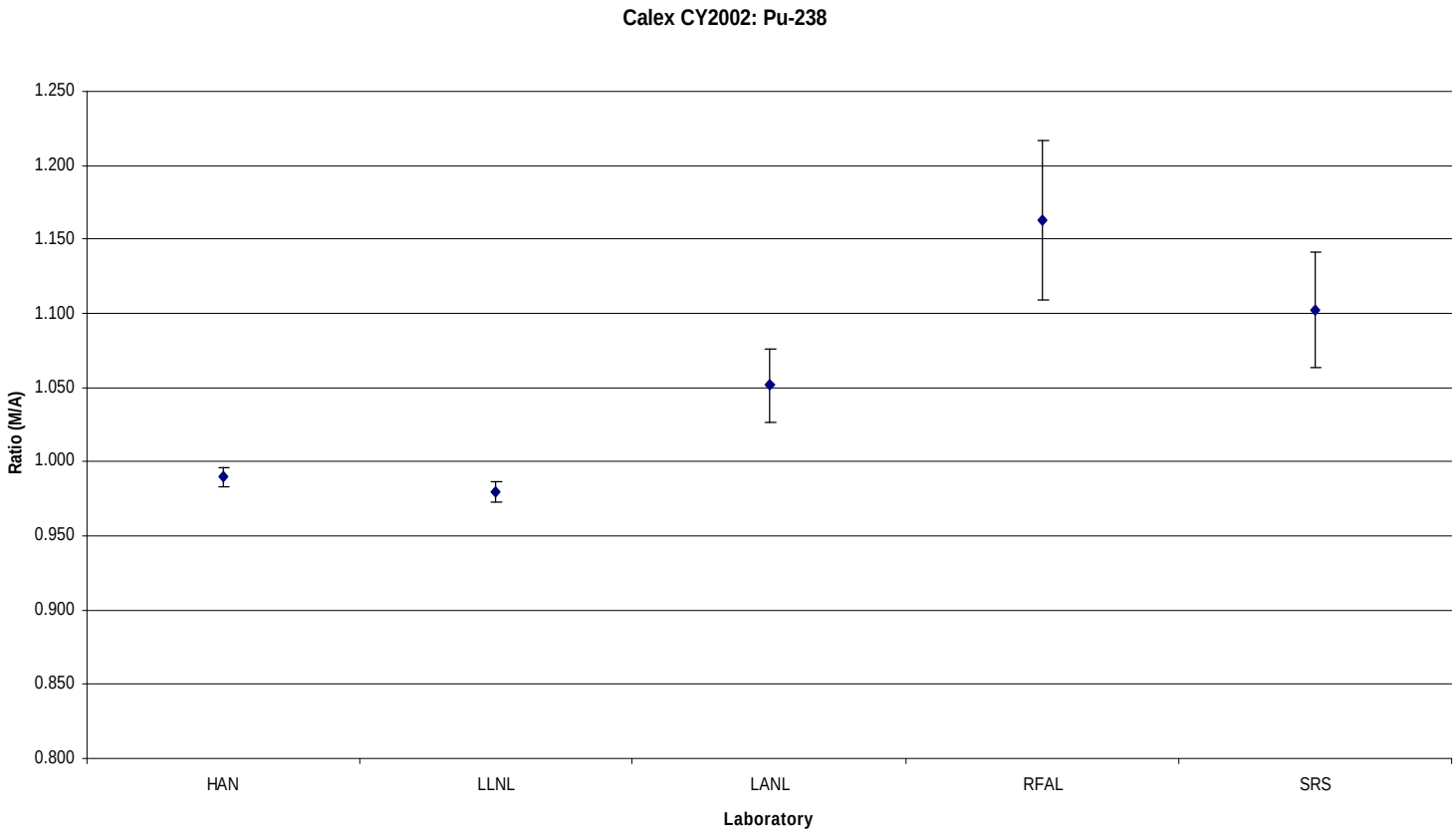


New Brunswick Laboratory

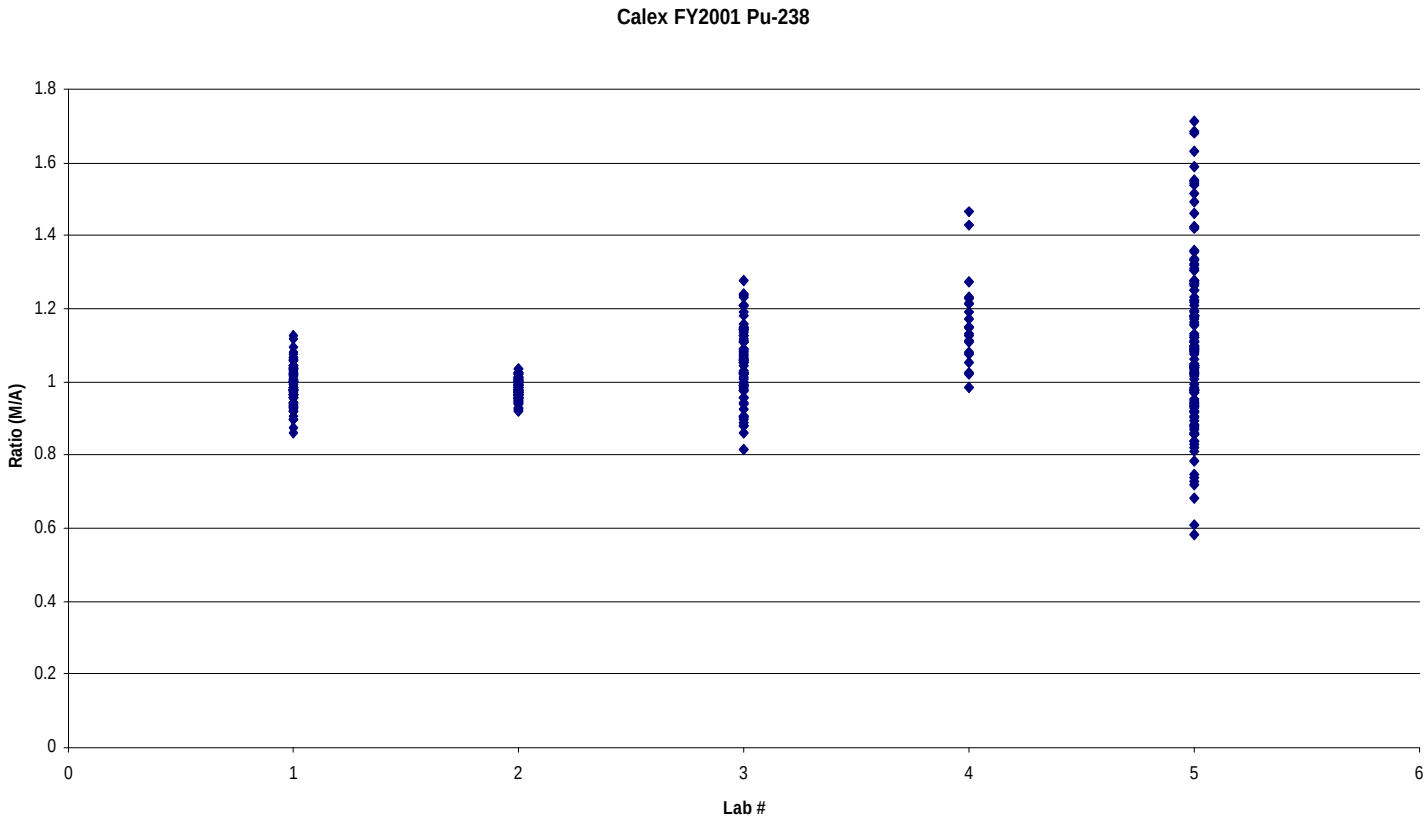
Results - P_{eff}

- Lab to lab statisically significant variation
- 13 isotopic systems
  - 7 statistically significantly bias
  - bias ranges from 0.04% to 0.32%
- 3 labs show statistically significant variation among its systems

Results - Pu 238

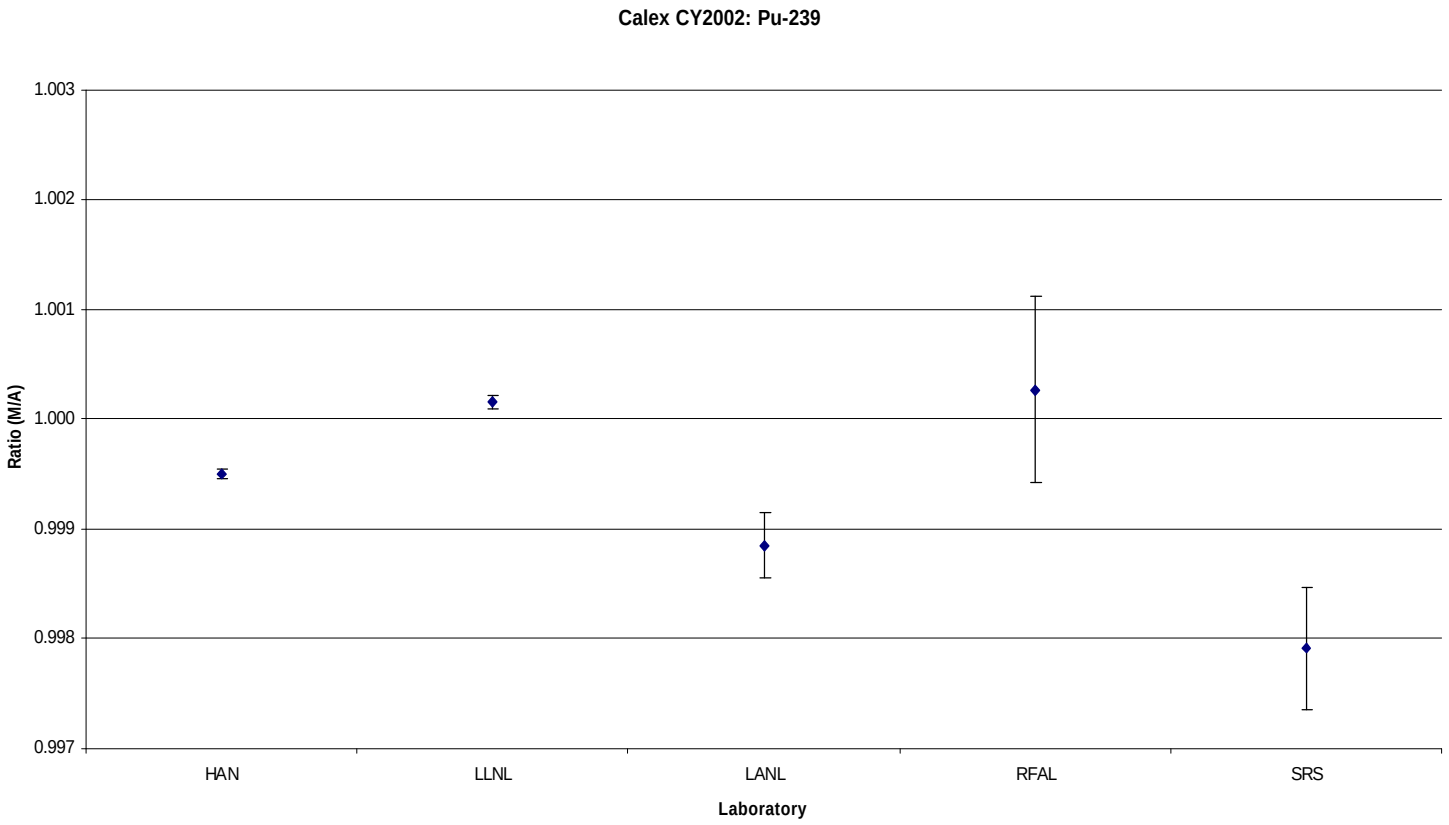


Results - Pu 238



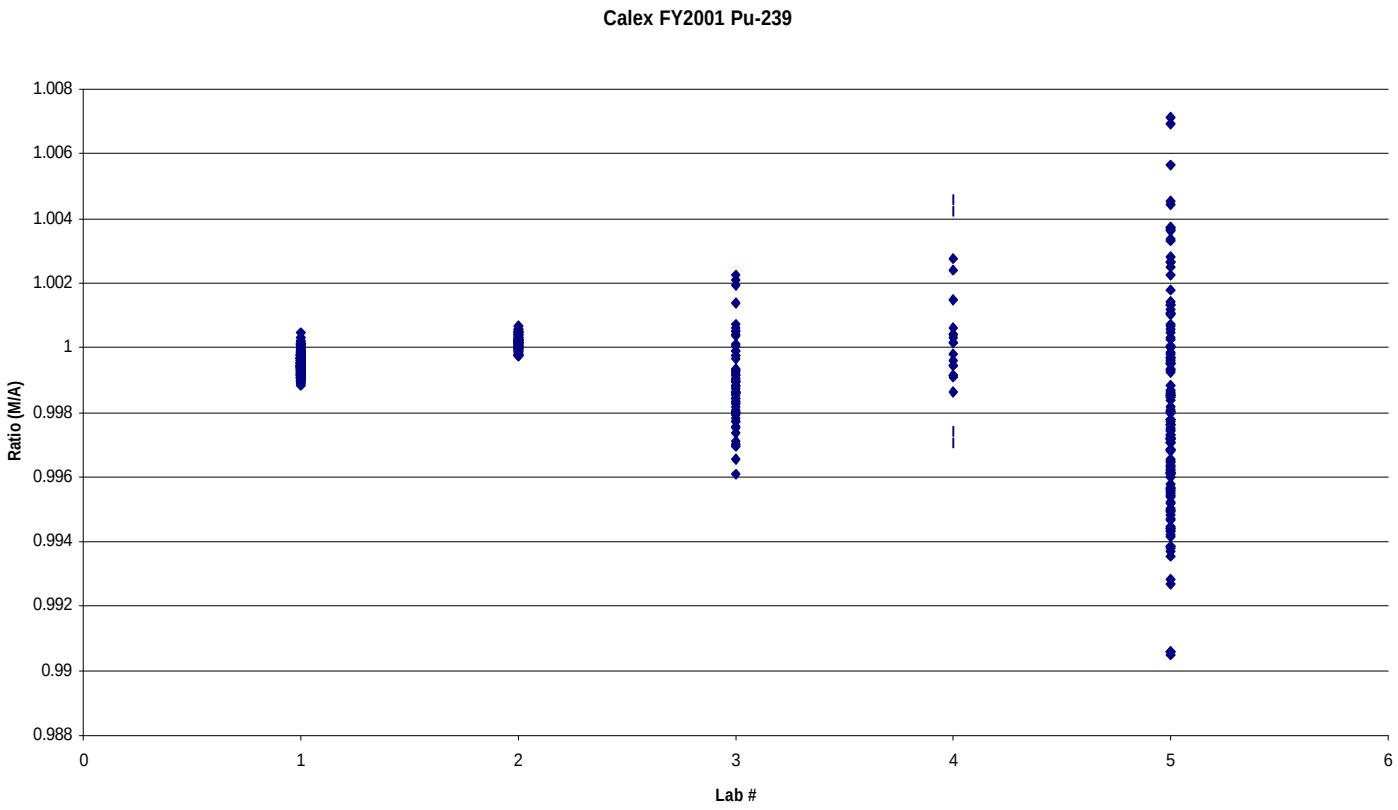
New Brunswick Laboratory

Results - Pu 239



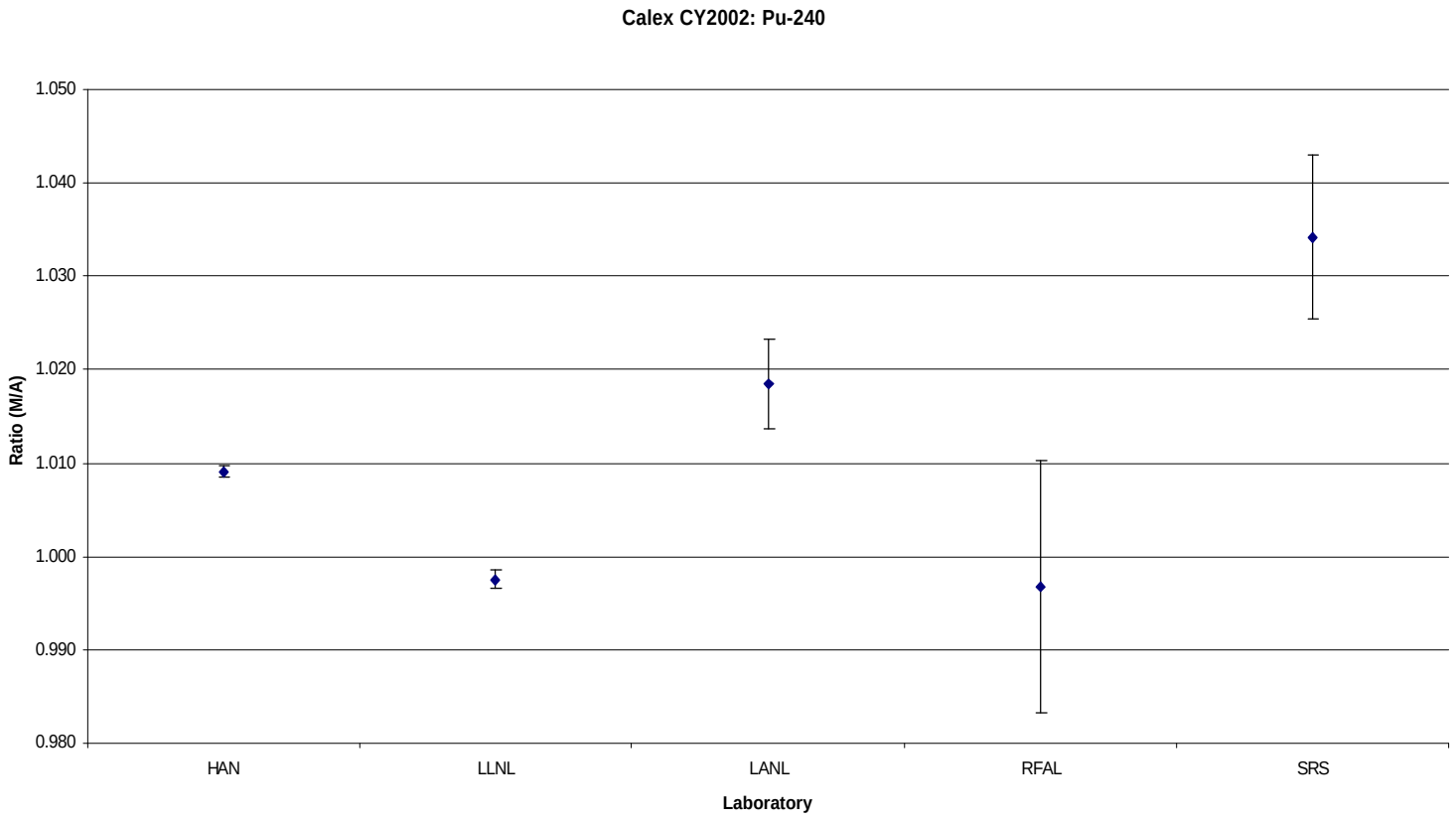
New Brunswick Laboratory

Results - Pu 239



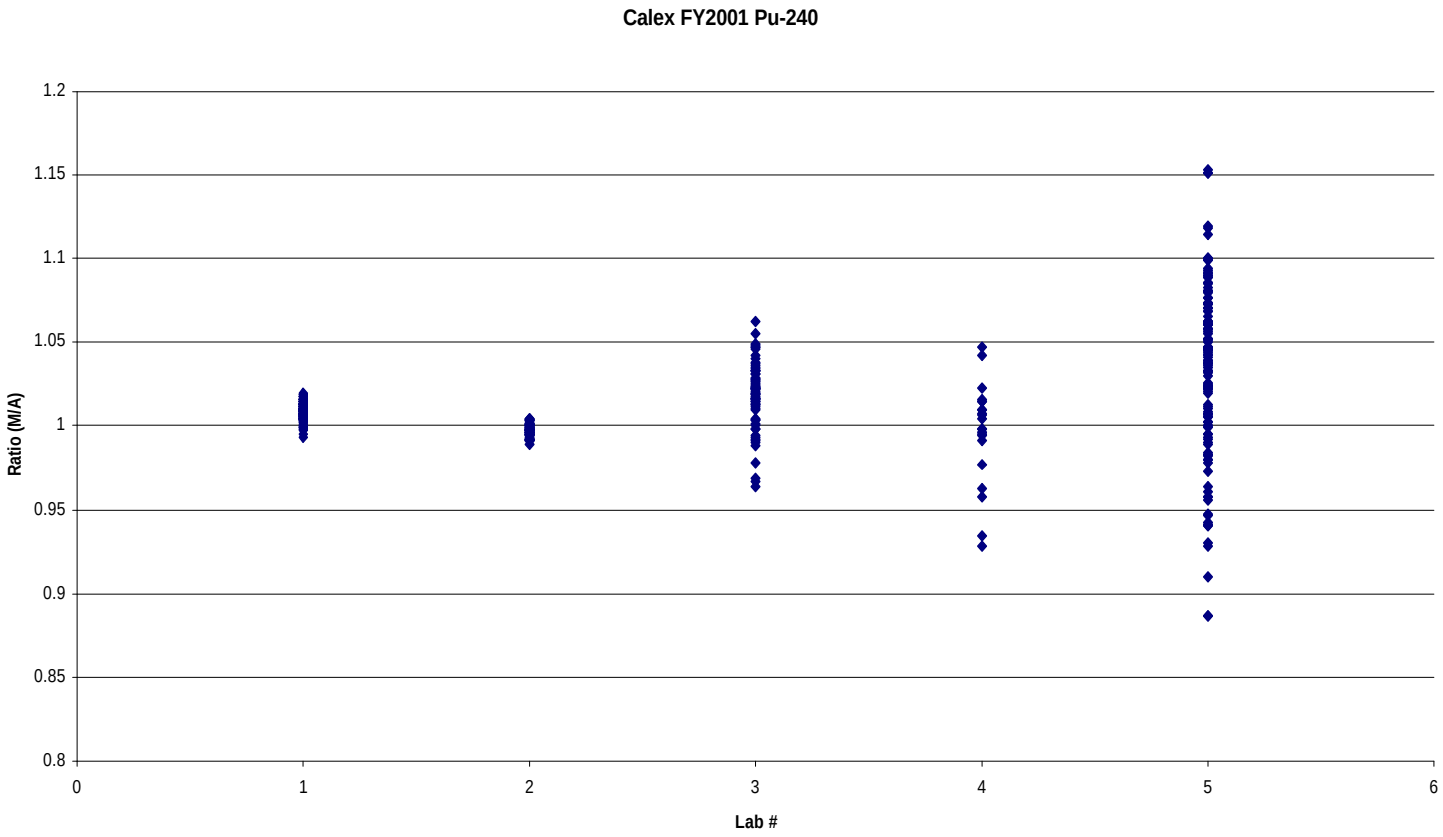
New Brunswick Laboratory

Results - Pu 240



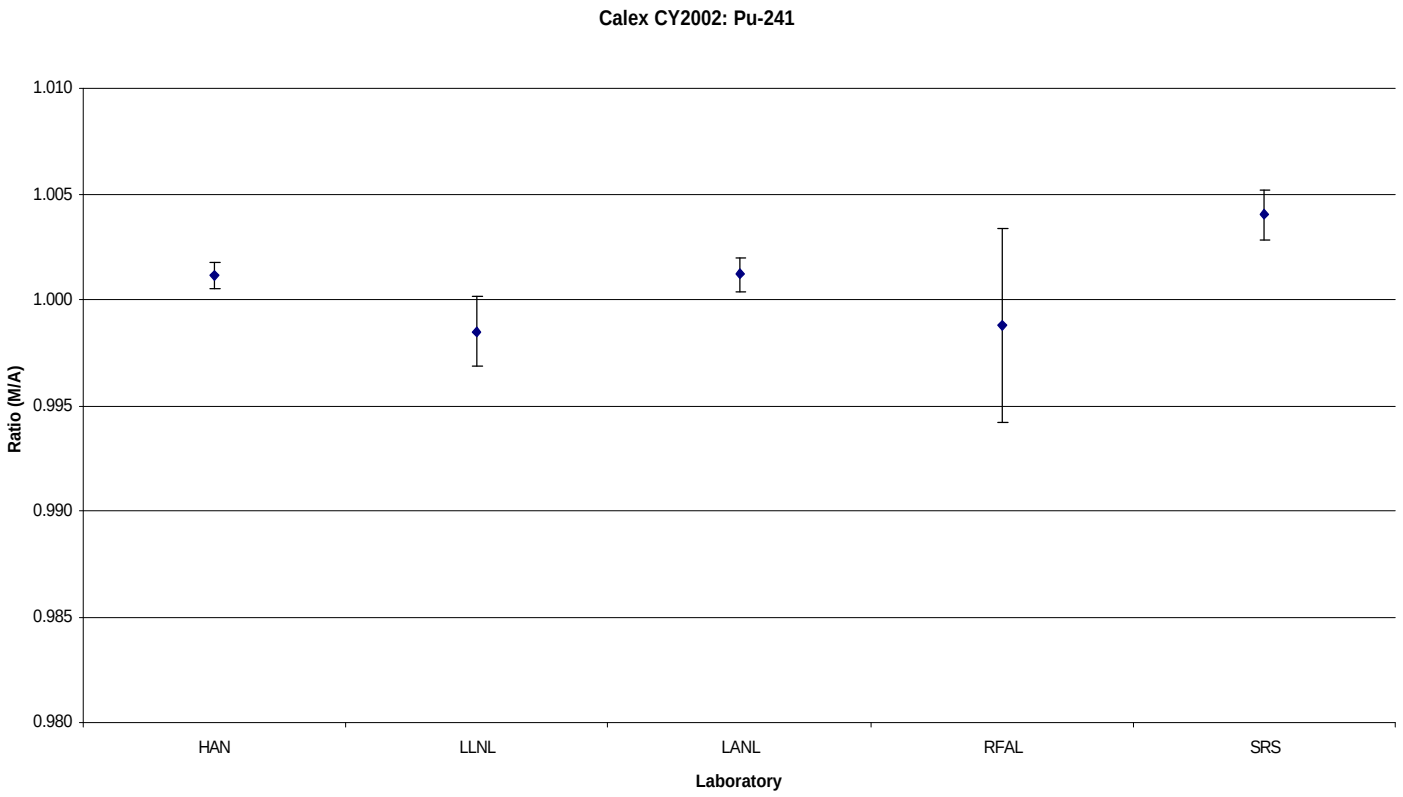
New Brunswick Laboratory

Results - Pu 240



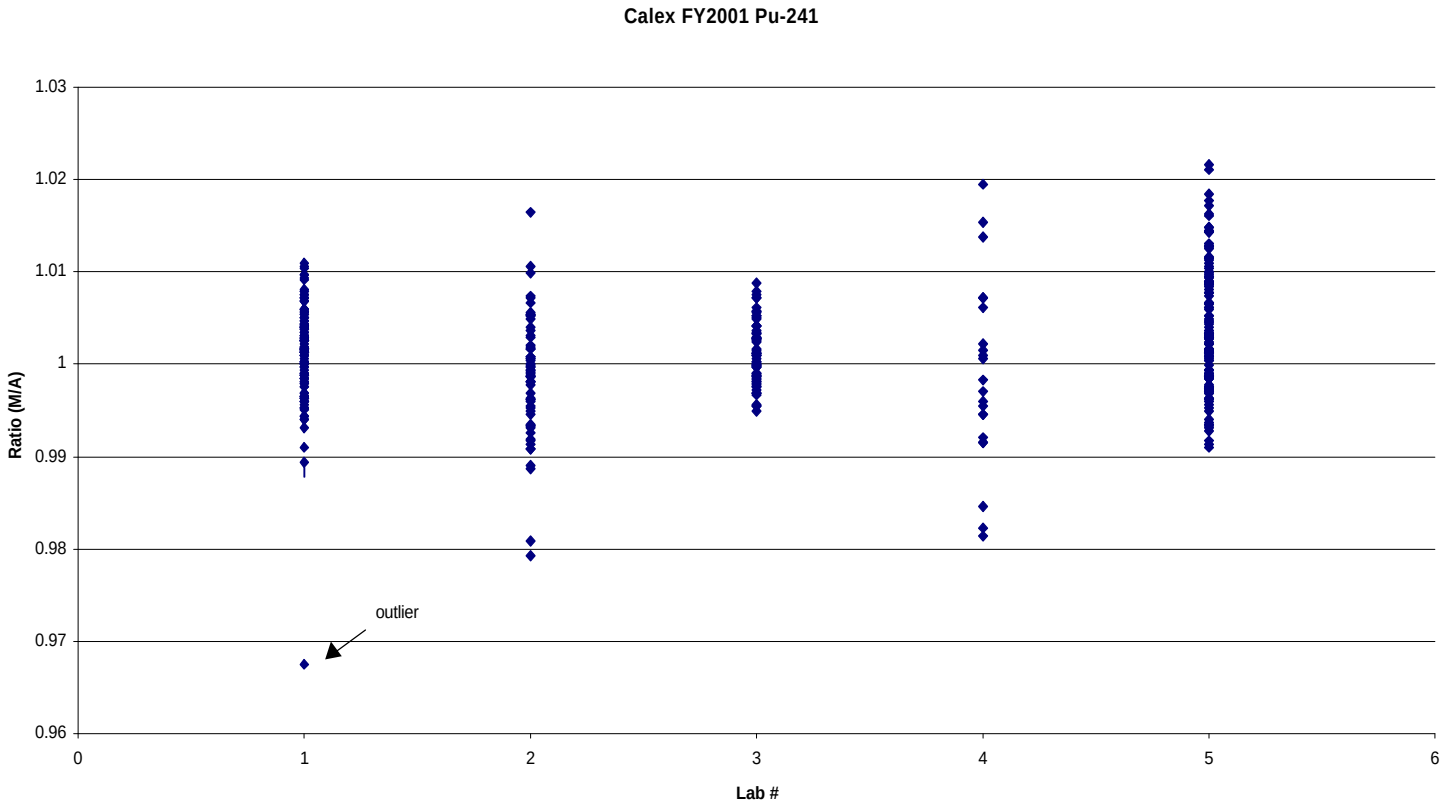
New Brunswick Laboratory

Results - Pu 241



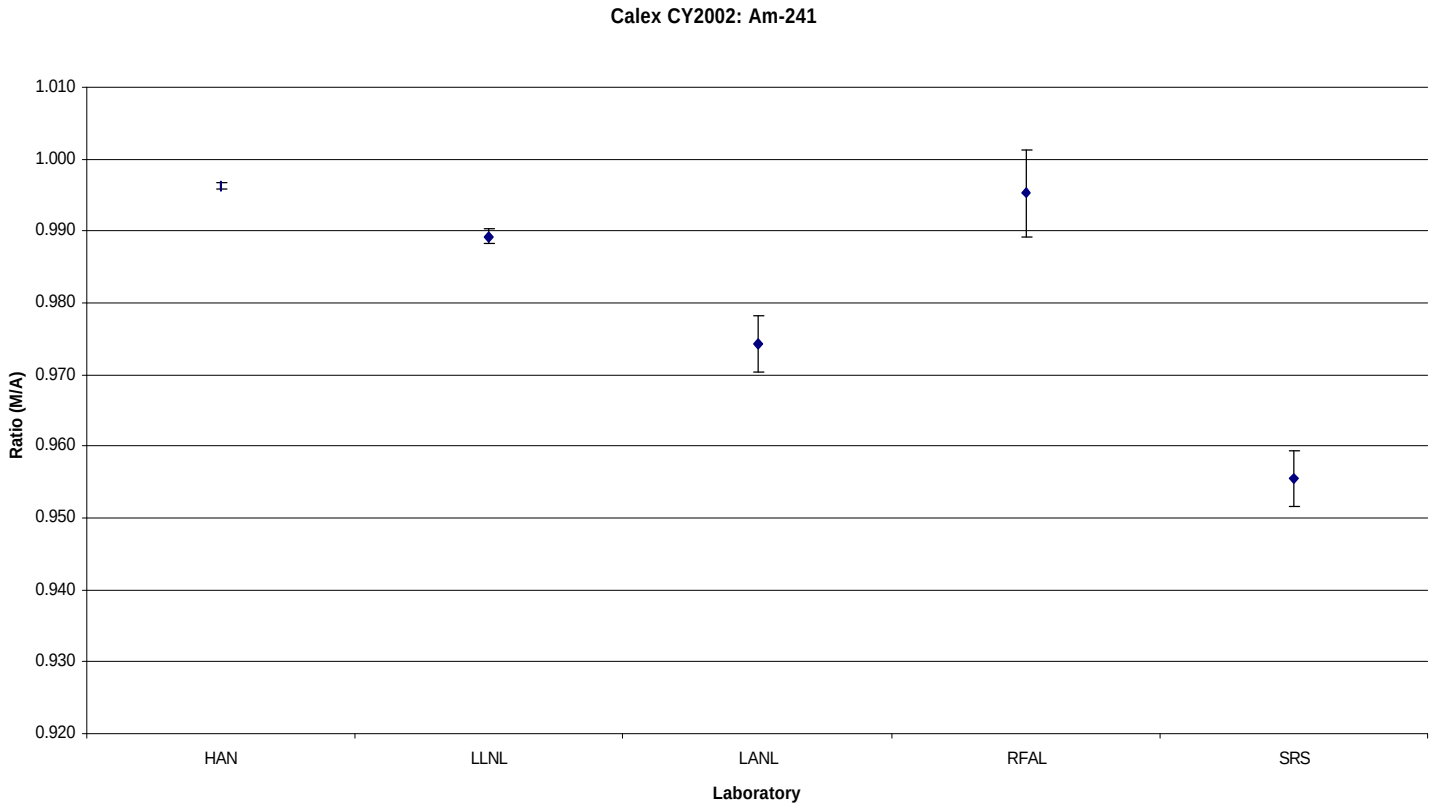
New Brunswick Laboratory

Results - Pu 241



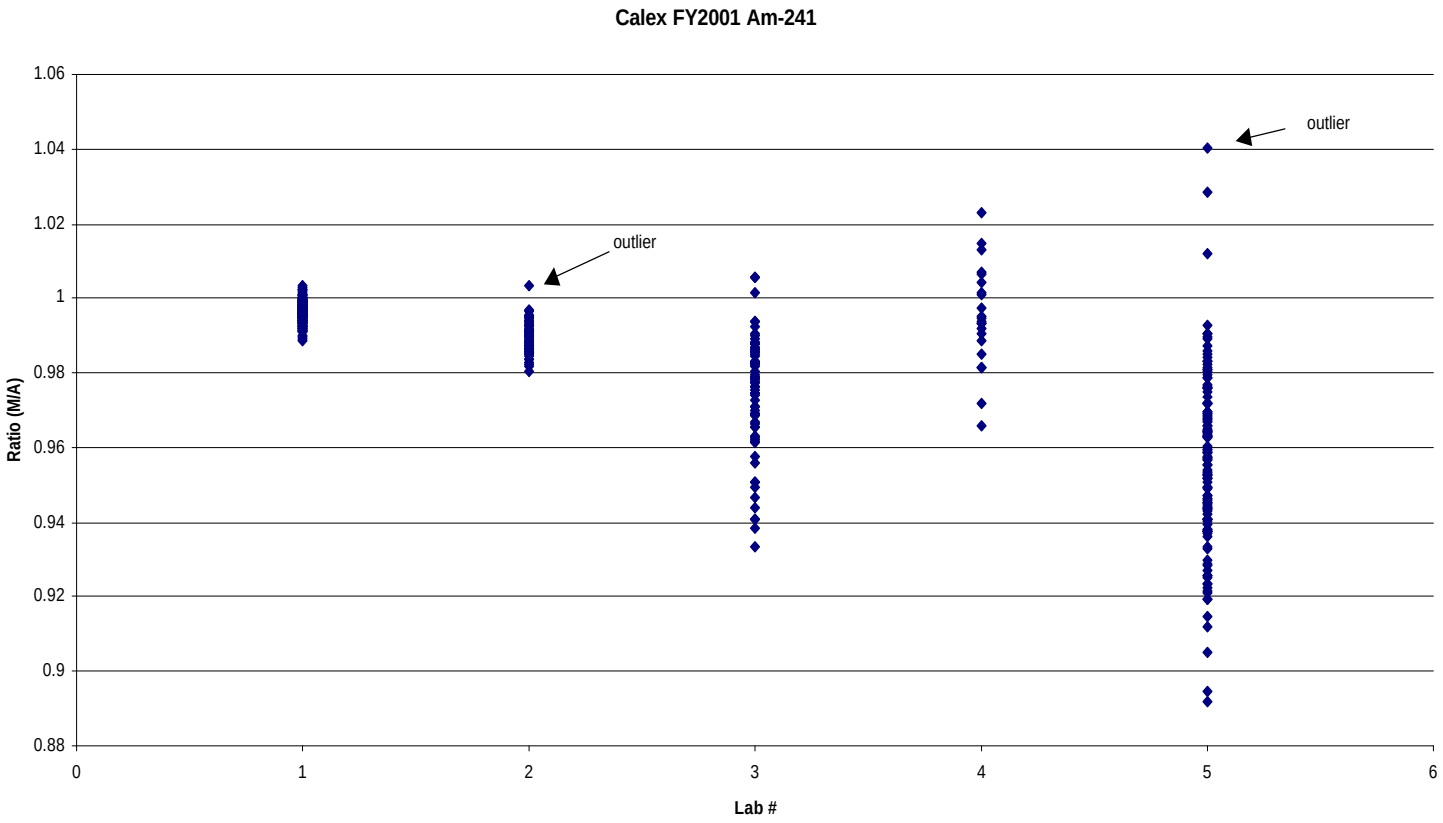
New Brunswick Laboratory

Results - Am 241



New Brunswick Laboratory

Results - Am 241



New Brunswick Laboratory

Status of the Performance Demonstration Project

Bill Geist - LANL

Larry Kayler - RFETS

Saleem Salaymeh - SRS

LA-UR-02-3740

This work is sponsored by the DOE Office of Security Policy, Policy Integration and Technical Support branch

Los Alamos
NATIONAL LABORATORY

Performance Demonstration Project (PDP)

Goals

- 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.

Data Evaluation

- Evaluate calorimeter, isotopic, and neutron data in different facilities.
- Involves RFETS, SRS, LLNL, NBL, and LANL.
- Two reference sets, Calex standards and the 6 working reference standards from RFETS.
- Calex 1 standard is a Pu heat standard of roughly 400 grams (1 watt).
- Calex 2 standard is a Pu heat standard of roughly 1750 grams (6 watt).
- 6 working standards were made at RFETS in support of shipment of materials to KAMS at SRS. These include 3 metal and 3 oxide items.

PDP - Measurements

Phase 1:

- Measure the 6 working reference standards at both RFETS and SRS and compare results.
- 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.
- ^{252}Cf measurements to provide traceability to the KAMS NMC.

Phase 2:

- Measure and collect Calex 1 and 2 standards data from LLNL, LANL, and SRS.

PDP - What has been done?

Developed the measurement protocol

- Input from RFETS, SRS, LANL, NBL, DOE EM, and DOE SO-13.
- Identified the key activities, time schedules, and deliverables.
- Identified the measurement details: number of measurements, statistical precision....

Activity at Rocky Flats (Larry Kayler)

- Completed measurements of 6 standards in both 3013 and 9975 containers.
- Review of data by key players at RF in April 2002.

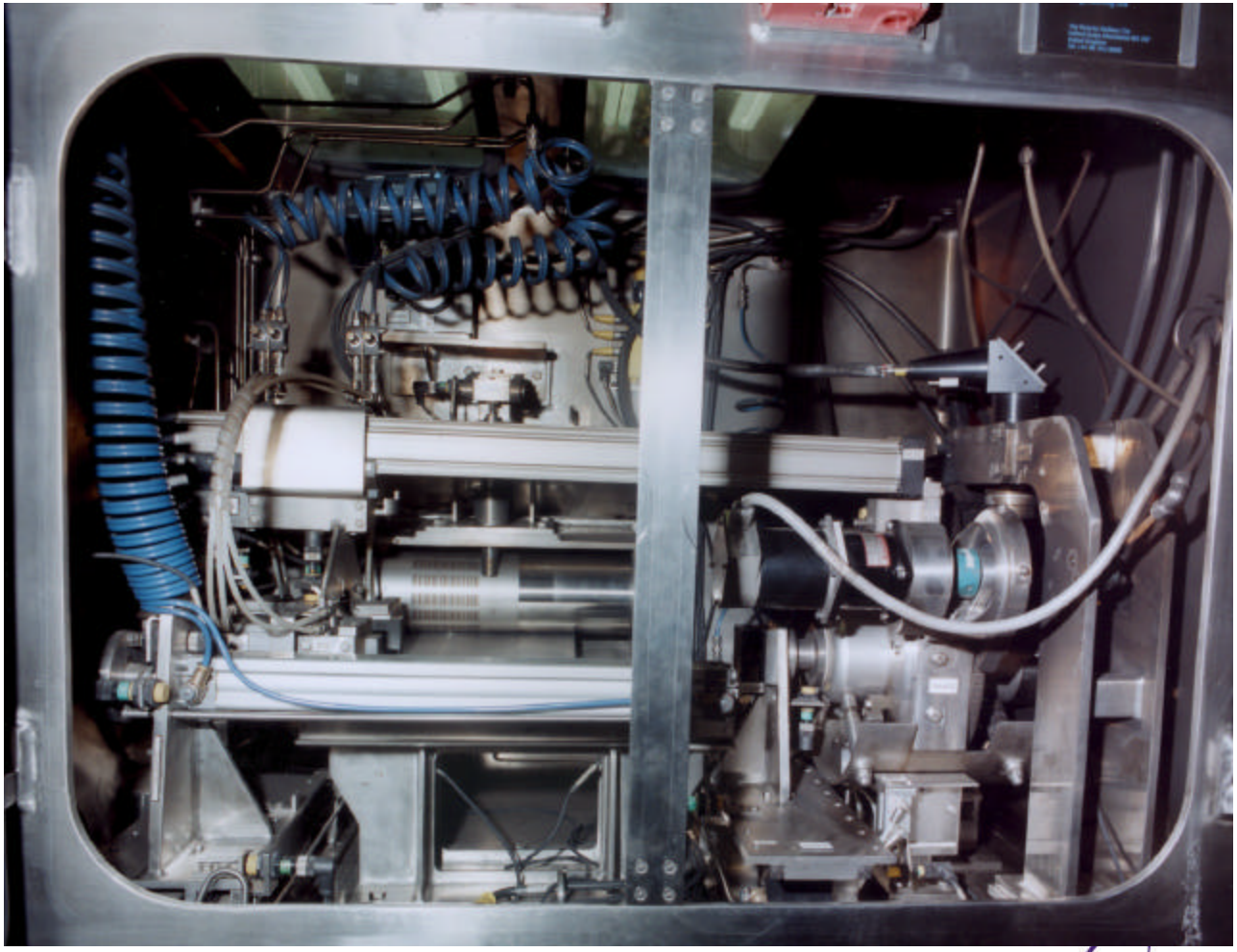
Activity of SRS (Saleem Salaymeh)

- Cf measurements in the KAMS counter.
- Cf and Calex measurements at FB-Line.
- After shipments, measurements of standards at KAMS.

3013 Container Neutron Isotopic and Calorimeter Measurements

Los Alamos NATIONAL LABORATORY

3013 Container
Automated Production Process



Los Alamos
NATIONAL LABORATORY

3013 Initial Characterization Measurements

In support of the shipper receiver agreement between SRS and RFETS, 3 Oxide and 3 Metal 3013 samples were prepared and characterized by three or more replicate calorimetry and gamma spec measurements. These samples provided the basis for the PDP study.

Subsequently, these same 3013 samples were measured using the LNMC. Again, each sample was measured a minimum of three times.

The results of this characterization study were provided to SRS and LANL for further analysis.

3013 Primary RFETS Measurement Method
LANL Water Bath Calorimeters

Los Alamos
NATIONAL LABORATORY

3013 Supplemental Measurement Method
ANTECH Air Bath Calorimeters

Los Alamos
NATIONAL LABORATORY

3013 Gamma Spec System w/Prompt Gamma

Los Alamos
NATIONAL LABORATORY

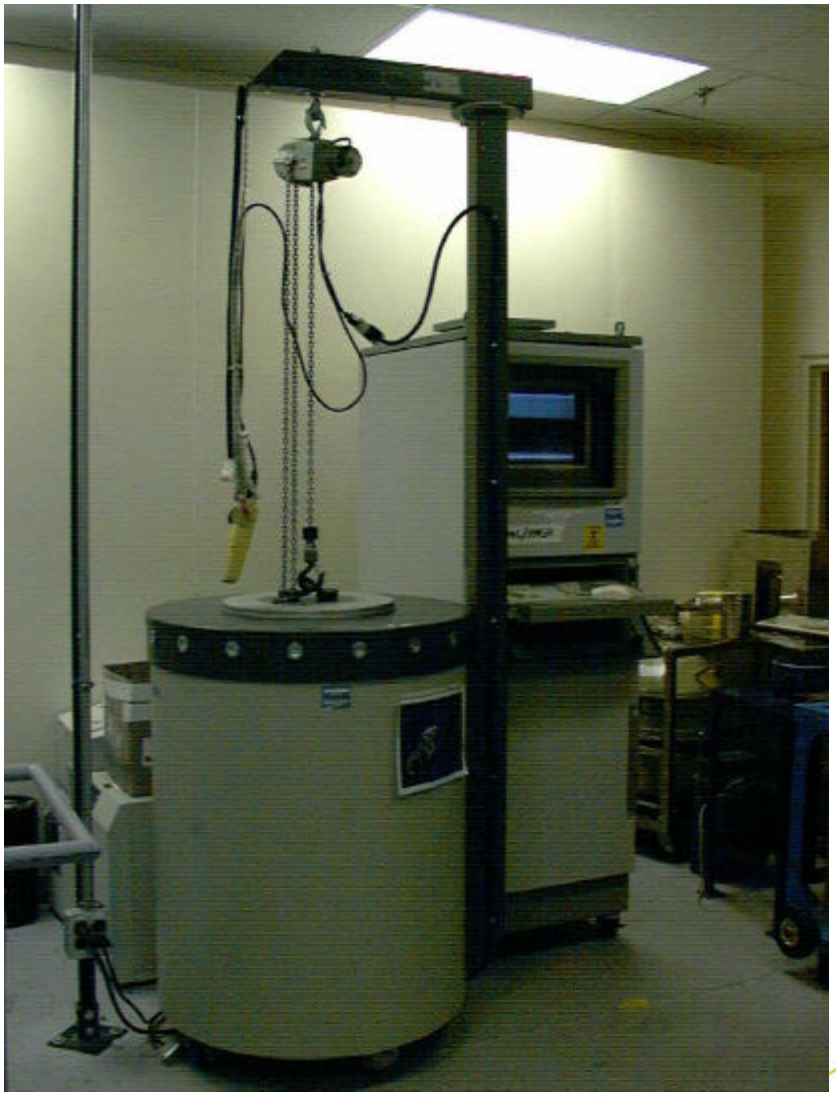
3013 Neutron Measurements

B-371 LNMC

3013 LNMC Measurements

B-371 LNMC

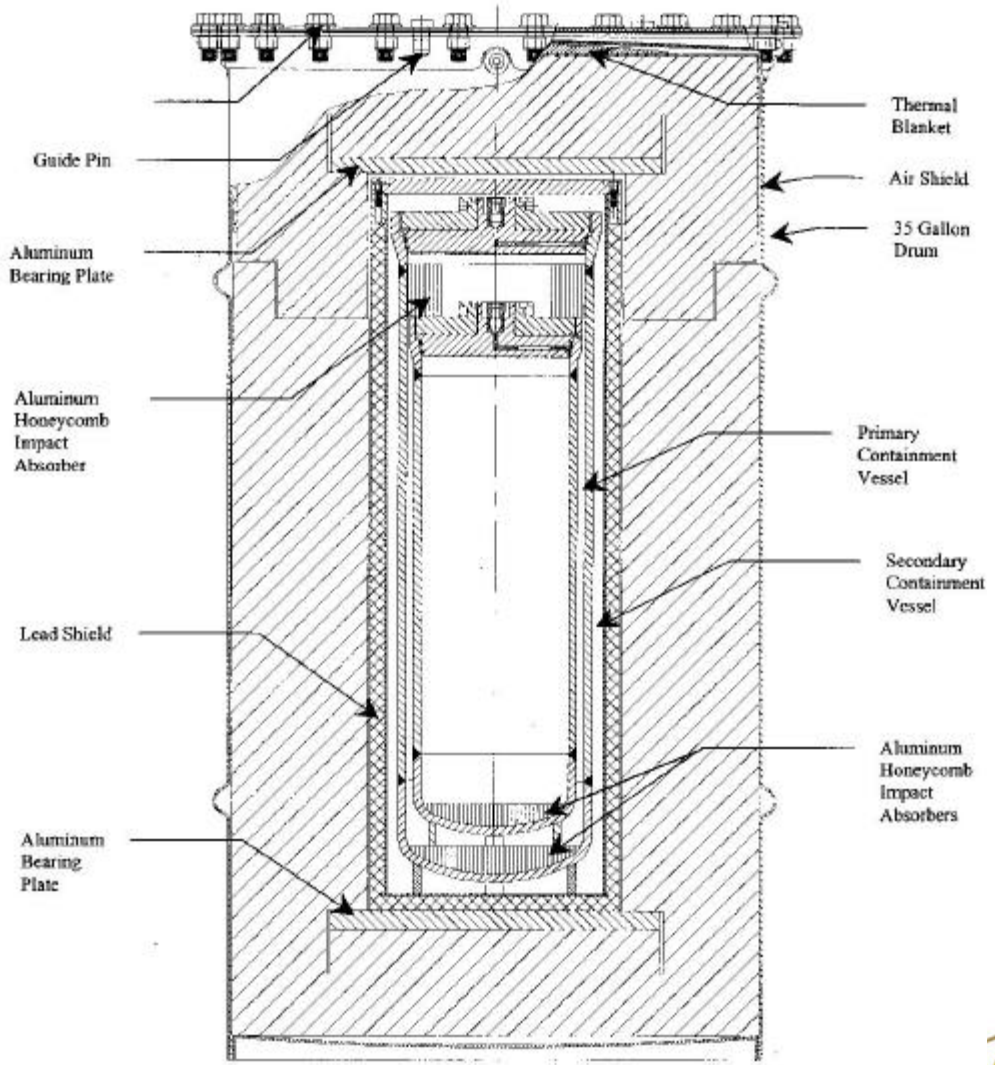
3013 Proposed Measurement Method
Small NMC with AMSR and INCC



Los Alamos
NATIONAL LABORATORY

9975 Neutron Measurements

9975 Specification Drawing



Los Alamos NATIONAL LABORATORY

9975 LNMC Performance Demonstration

B-371 LNMC

Los Alamos
NATIONAL LABORATORY

9975 LNMC Configuration Changes

- To allow measurement of the 9975 in the LANL LNMC, the top plug on the instrument had to be raised 4-inches to allow the container to fit.

- Measurements were made on a ^{252}Cf check source installed in a fixture that places the source in the center of the volume of the chamber.

- After reconfiguring the moveable top plug to allow sufficient height for the 9975, the ^{252}Cf source was measured again in the same location (no longer the center) and the results recorded.

- Additional measurements were made placing the ^{252}Cf source in various positions within the chamber to investigate variations in response due to position.

9975 LNMC Measurements

- The previously characterized 3013 oxide and metal samples were packaged in standard 9975 shipping containers.
- Each container was measured four times.
- Standard daily performance checks were completed each measurement day prior to performing measurements for record
- In between replicate measurements, each 9975 was rotated randomly to simulate the normal randomness of loading and unloading between measurements.
- All data collected were provided to SRS and LANL

B-371 NDA Tech Support Team

Not pictured - Dr. Michelle Cameron, NDA PI
Dr. John Conway, Senior Chemist.

LA-UR-02-3740

Los Alamos
NATIONAL LABORATORY

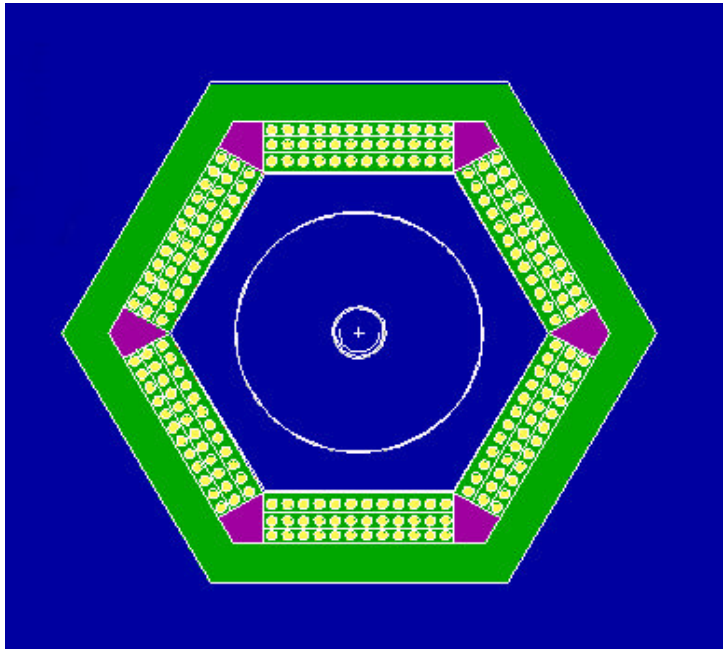
Performance Demonstration Project

Performance Demonstration Project
SRS Activities:

- Cf measurements in the KAMS counter
- Cf measurements in the FB-Line counter
- Calex I & II measurements in the FB-Line counter
- After shipments from RFETS, measurements of working standards at KAMS

Performance Demonstration Project

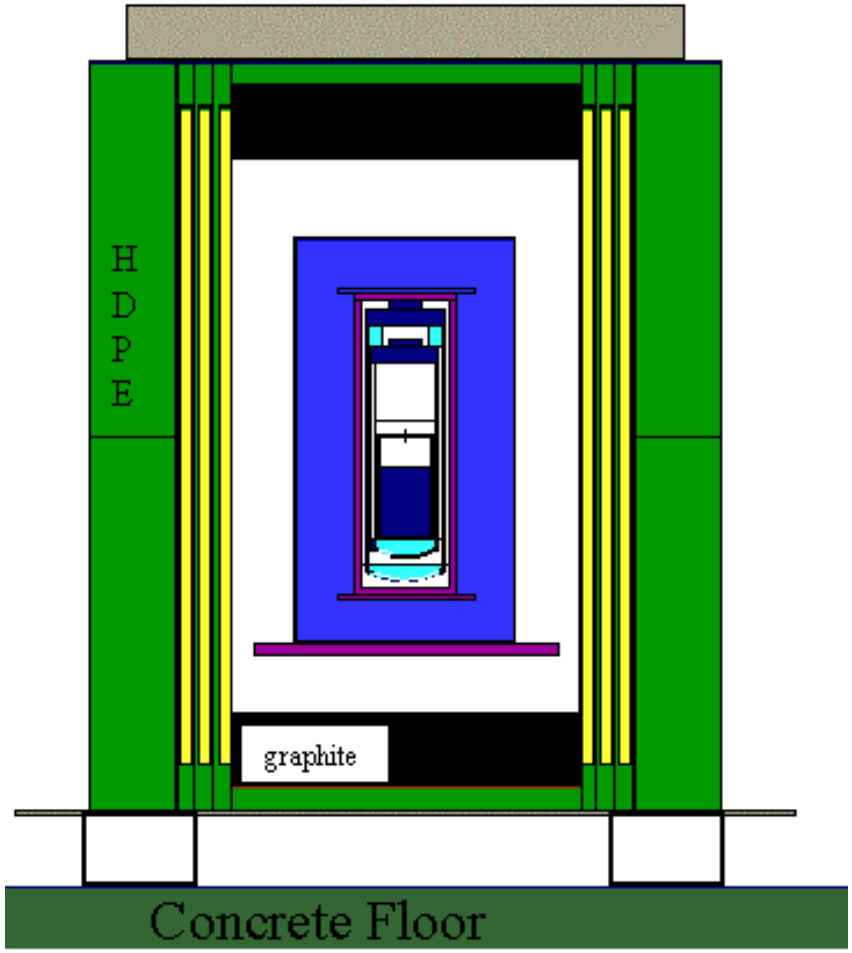
Drum Neutron Multiplicity Counter



- Basic Counter
  - Hexagonal Shape
  - 3 Rings of Tubes
  - Graphite Endplugs
  - Unpowered rollers for load/unload of drums

Performance Demonstration Project

Drum Neutron Multiplicity Counter

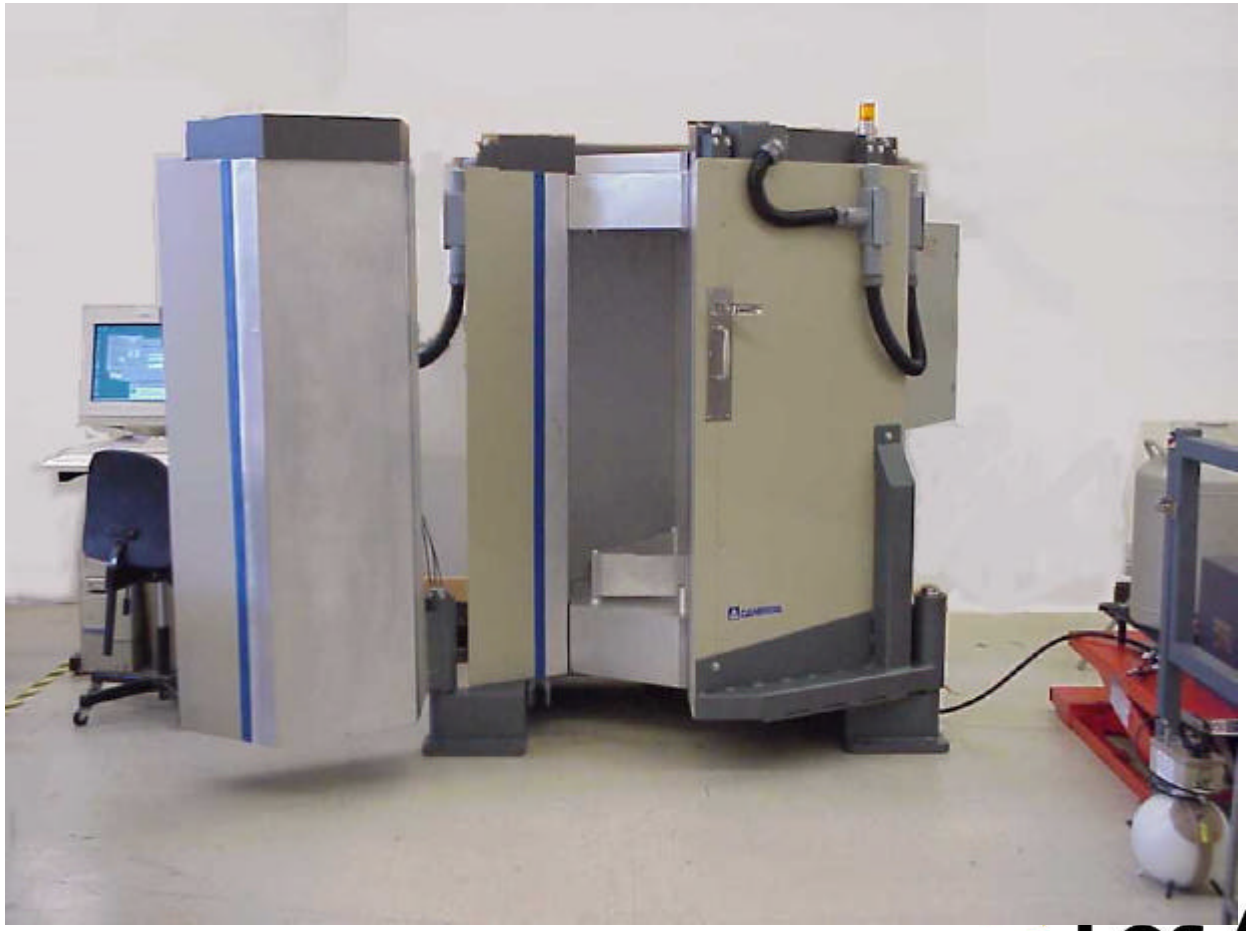


- Final Counter
  - 198 tubes
  - 3 rings
  - 10 atm

Los Alamos
NATIONAL LABORATORY

Performance Demonstration Project

Drum Neutron Multiplicity Counter



Performance Demonstration Project

KAMS Drum Neutron Multiplicity Counter

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

Performance Demonstration Project

FB-Line Neutron Multiplicity Counter

High Voltage	1680 V
Pre-delay time	3 μsec
Die-Away time	50.4 μsec
Gate Width	32 μsec
Dead time Parameters (NCC)	$a = 21.02 \times 10^{-9}$
	$b = 0.0020\ \mu sec^2$
Dead time parameters (multiplicity)	$c = 50.0 \times 10^{-9}$ sec
Dead time parameter (τ)	19.15 ± 0.45 nsec
Doubles Gate Fraction	0.4426
Triples Gate Fraction	0.1919
Efficiency (Cf-252 point source)	0.578
Efficiency (Pu-240 estimated)	0.567

Performance Demonstration Project

Californium Source Measurements in the KAMS NMC:

Item ID	Singles	Doubles	Triples
BKG	16.94	0.07	0
Cfone	52051.4	24009.94	6397.2
Cftwo	51981	23867.77	6410.2
Cftree	51973.7	23950.11	6482.9
Cffour	52005.1	24043.83	6649.7

Performance Demonstration Project

Californium Source Measurements in the FB-Line NMC:

Item ID	Singles	Doubles	Triples
BKG	35.6	1.05	c
Cfone	56335.3	23407.9	5444.1
Cftwo	56309.3	23432.4	5489.3
Cftree	56334.5	23411.9	5555.2
Cffour	56326.2	23397.7	5373.9

Performance Demonstration Project

Ratios of KAMS to FB-Line:

Item ID	KAMS to FB Singles Ratio	KAMS to FB Doubles Ratio	KAMS to FB Triples Ratio
BKG	0.476	0.067	
Cfone	0.924	1.026	1.175
Cftwo	0.923	1.019	1.168
Cftree	0.923	1.023	1.167
Cffour	0.923	1.028	1.237

Performance Demonstration Project

Status of the Performance Demonstration Project at SRS

Saleem Salaymeh and Raymond Dewberry

Savannah River Technology Center

Linda Baker and Don Faison

Central Laboratory Facility

David Eisele and Don McCurry

KAMS Facility

Savannah River Site

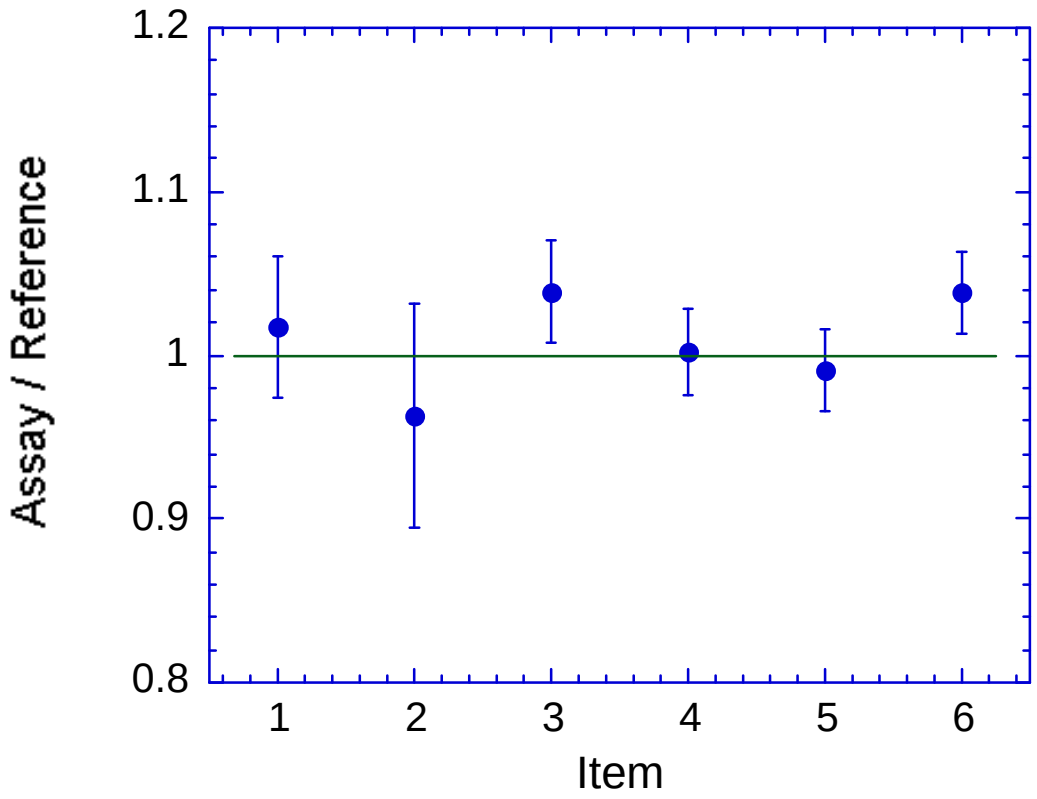
Aiken, SC 29808

LA-UR-02-3740

Los Alamos NATIONAL LABORATORY

Preliminary Results for 3013 Containers

- Reference values were determined from calorimetry.
- The mass values determined from the neutron data agree well with the reference values.

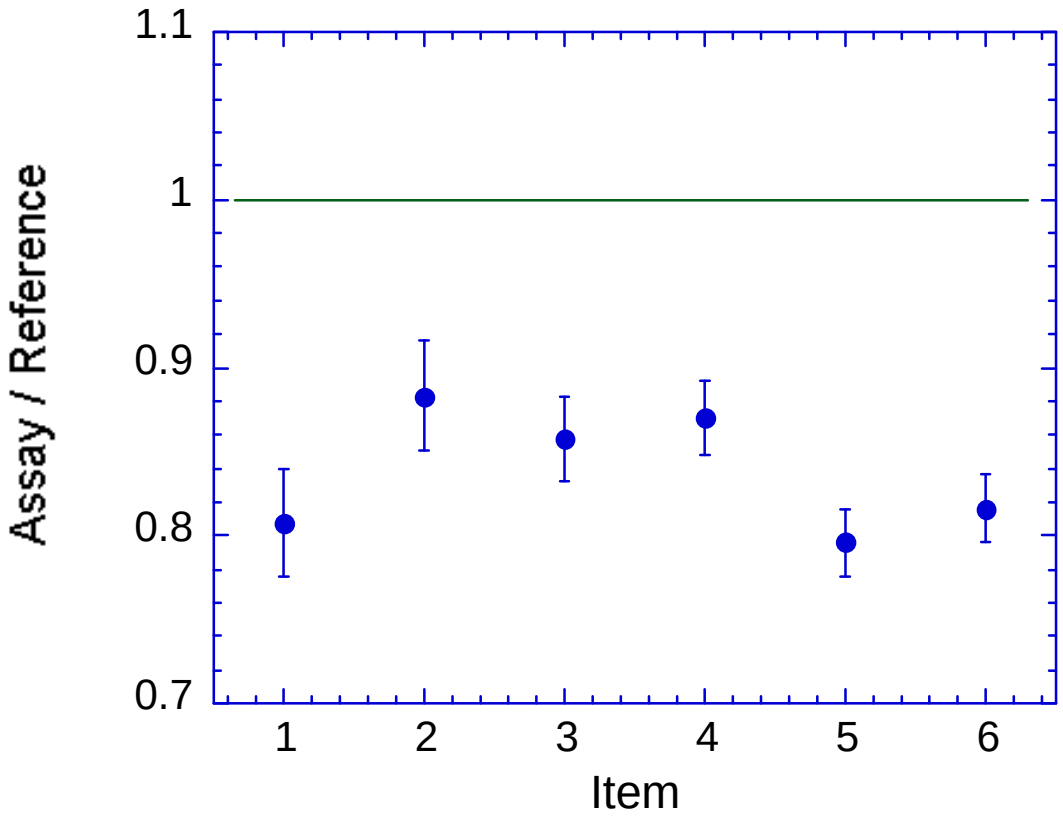


Average A/R = 1.009
1 σ rsd = 0.030

Los Alamos NATIONAL LABORATORY

Preliminary Results for 9975 Containers

The neutron data have a 10% to 20% discrepancy from the reference data.



Cause of the bias:

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

Los Alamos NATIONAL LABORATORY

Conclusions

- Most of the first phase measurements have been completed.
- Working on the data analysis of the 3013 and 9975 containers.
- Good results for NMC assay of items in 3013 containers.
- Biased results for NMC assay of items in 9975 containers.
- Future work:
  - Complete analysis of the data.
  - Send results to NBL for a statistical analysis.
  - Determine the cause of the bias in the 9975 data.
  - Collect and analysis measurement control data.

Transportable Calorimetry Laboratory

Clifford Rudy, Phillip Hypes

Los Alamos National Laboratory, NIS-5

NBL Measurement Evaluation Program Meeting

June 23, 2002

Orlando, Florida

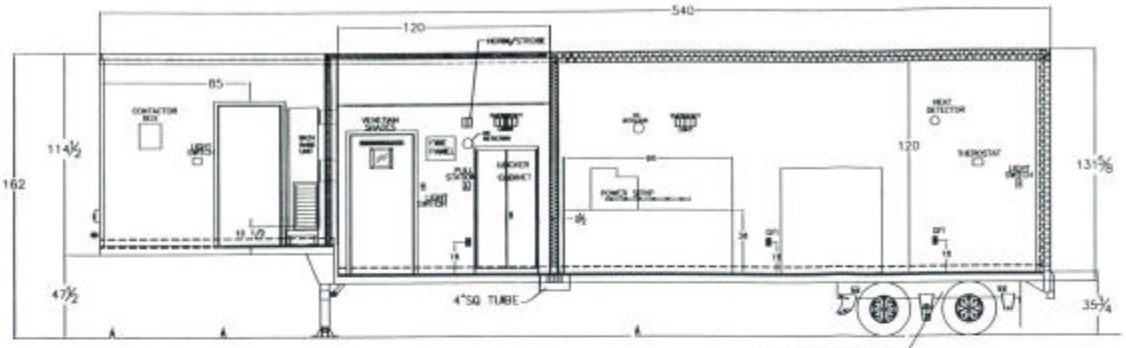
This work supported by the US Department of Energy, Office of Nonproliferation and National Security, Office of Safeguards and Security

Transportable Calorimetry Laboratory

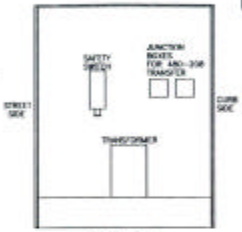
- Develop transportable cal lab capable of being moved to different DOE facilities
- Produce working standards using Cal/Iso
  - HEU,Pu
- DOE Verification of Inventories
- Staffed by facility personnel or NBL personnel

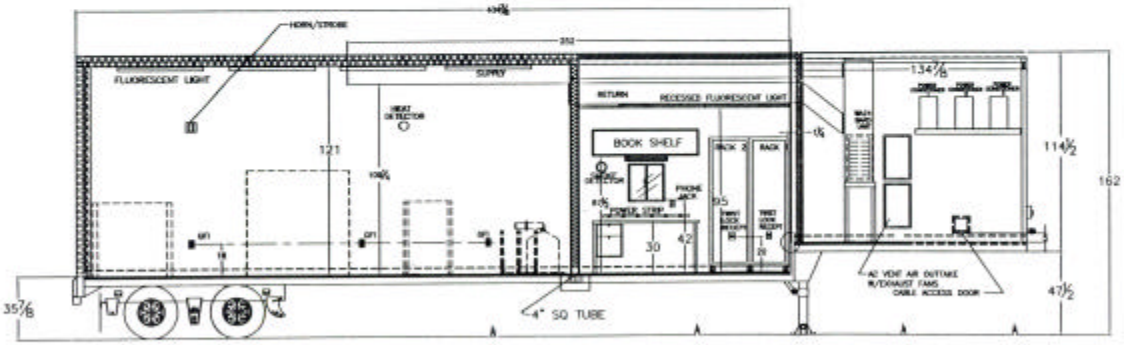
Transportable Calorimetry Laboratory

- Holds 2”, 5” and 13.5” diameter calorimeters
- Calorimeters sensitive enough to measure HEU
- Lab large enough to hold large volume calorimeter that will measure 55 gallon drum

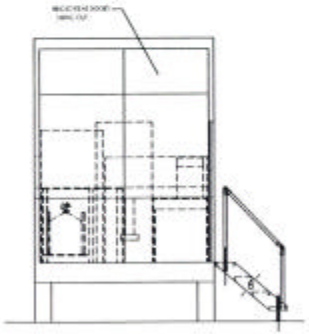


INTERIOR CURB SIDE ELEVATIONS





INTERIOR STREET SIDE ELEVATIONS



PARTS LIST

EMI TECHNOLOGIES, INC.
3200 N. TELSHOR BLVD. LAS CRUCES, NM 88011
(505) 532-9190 FAX (505) 532-0884

LOS ALAMOS/ CALORIMETRY TRAILER
INTERIOR SIDE ELEVATIONS

D | 77004 | 8262-1003

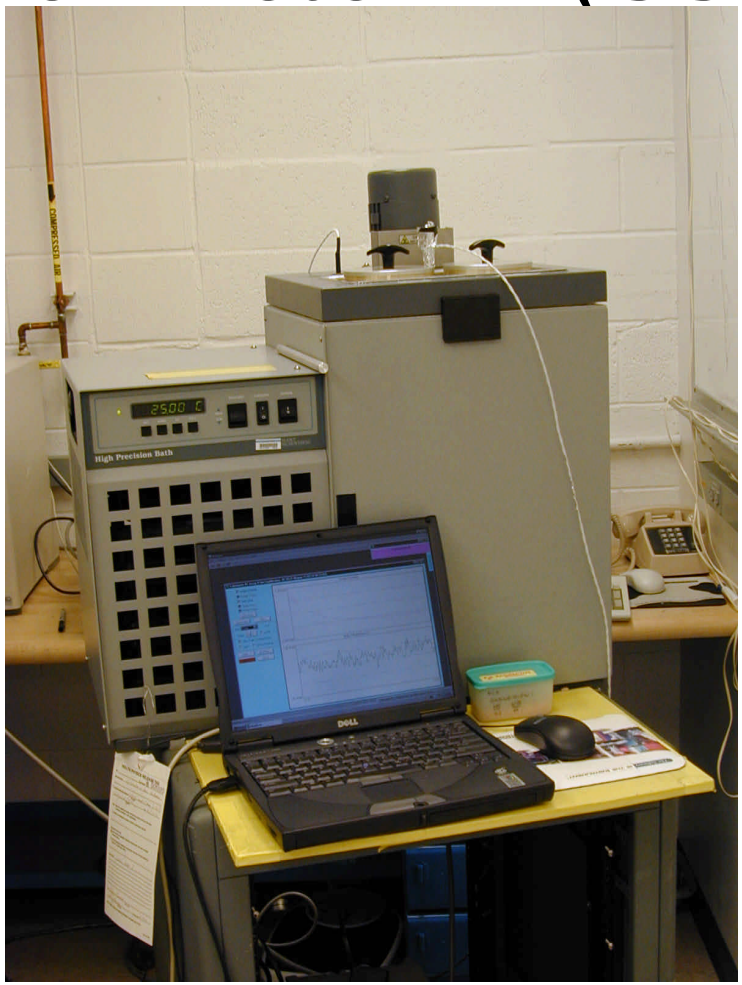
L. APODACA 2/20/02

Safeguards Science and Technology
Los Alamos National Laboratory

Transportable Calorimeter Laboratory

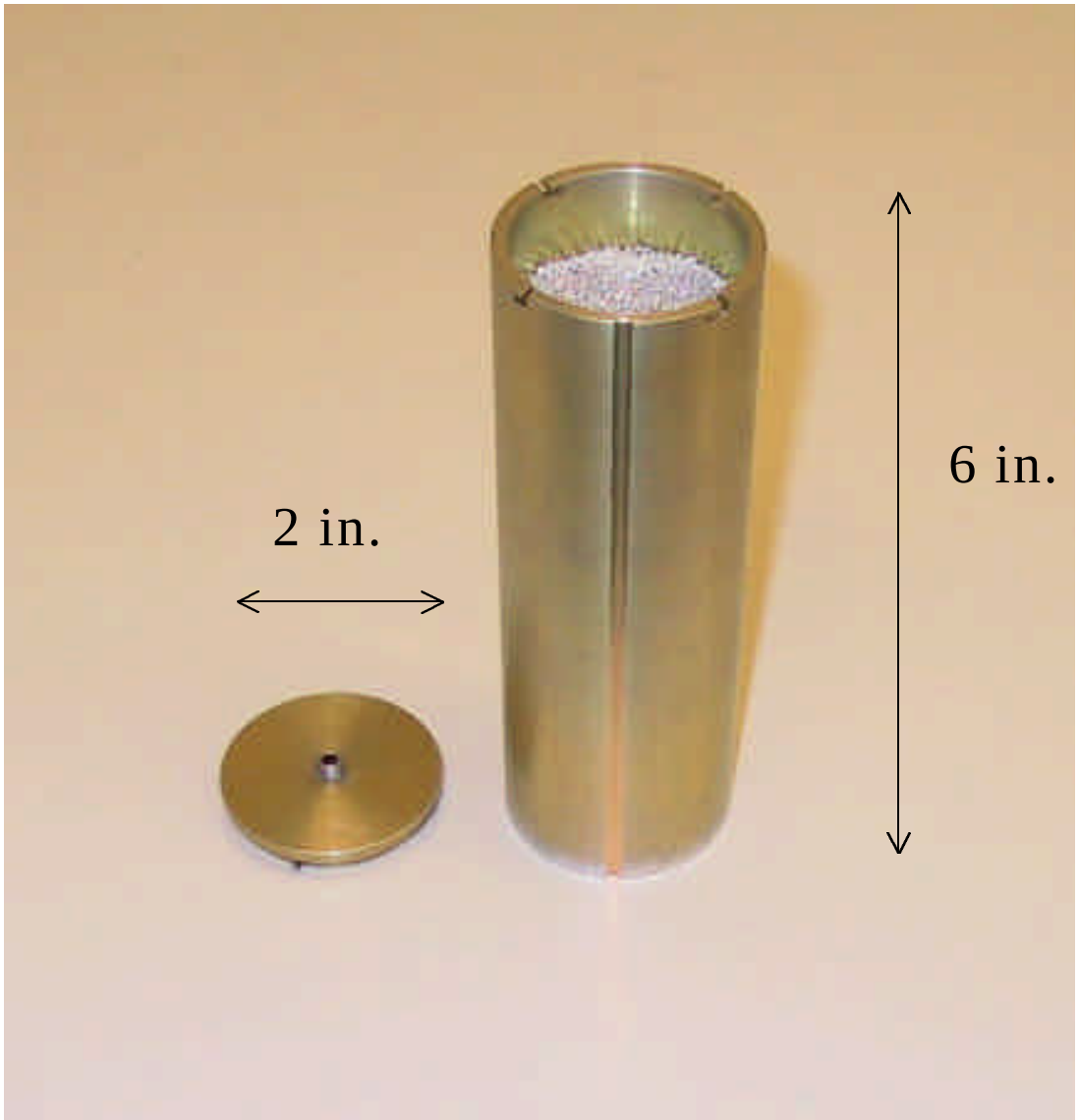
Transportable Cal Lab

Transportable Cal Lab Calorimeter #1(SSC2)

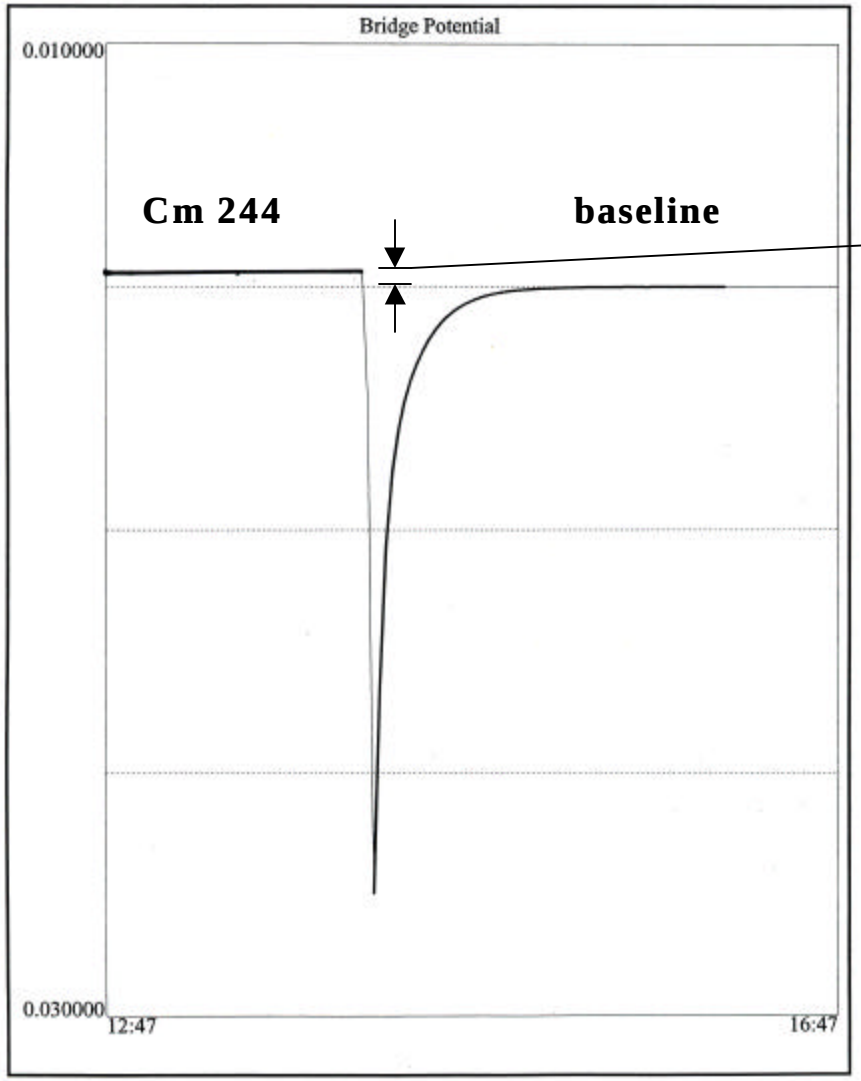


Calorimeter #1(SSC2) Water Jackets

Cal #1(SSC2) Cal can



Transportable Lab Calorimeter #1
Measurement of 0.0019 g Cm-244 sample



Replicate Measurement Results

625 μV

623 μV

627 μV

625 μV

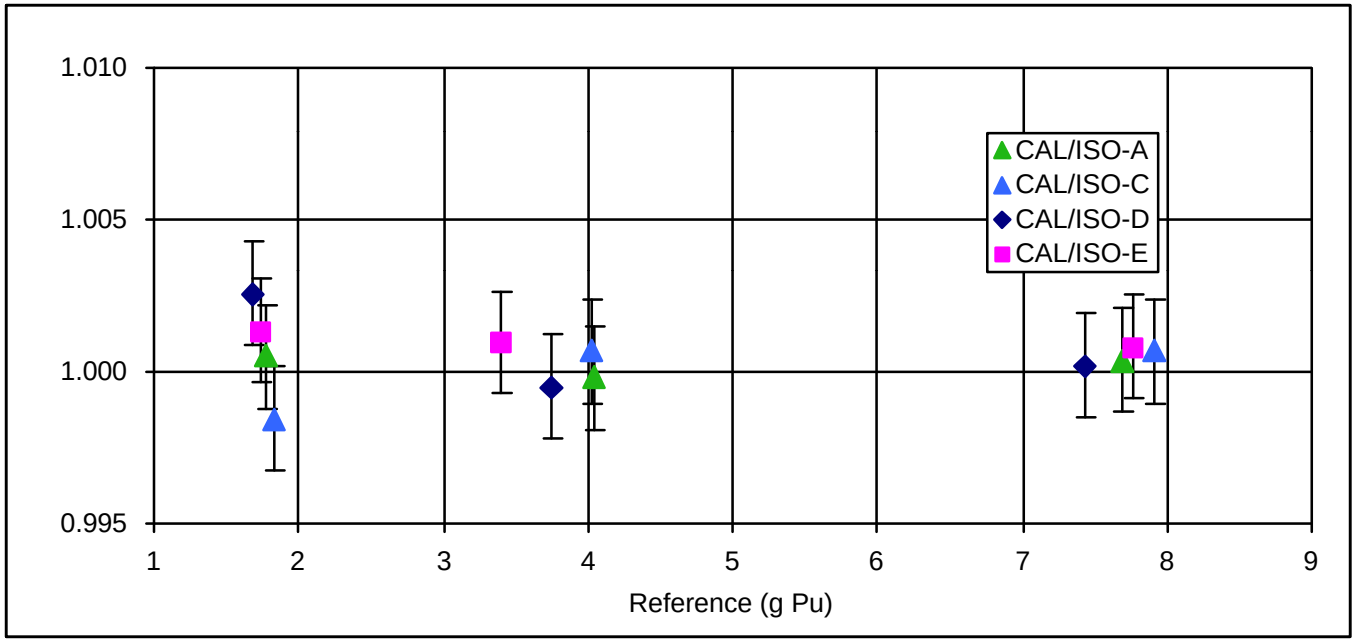
627 μV

626 μV

Precision=0.2% RSD

Power:Book - cal average => 0.8%

Mass vs. SSC2 Cal/Iso Assay Comparison on Low-Power Pure Pu Metal Standards



2 grams of Pu thermal power equivalent to about 2 kg HEU

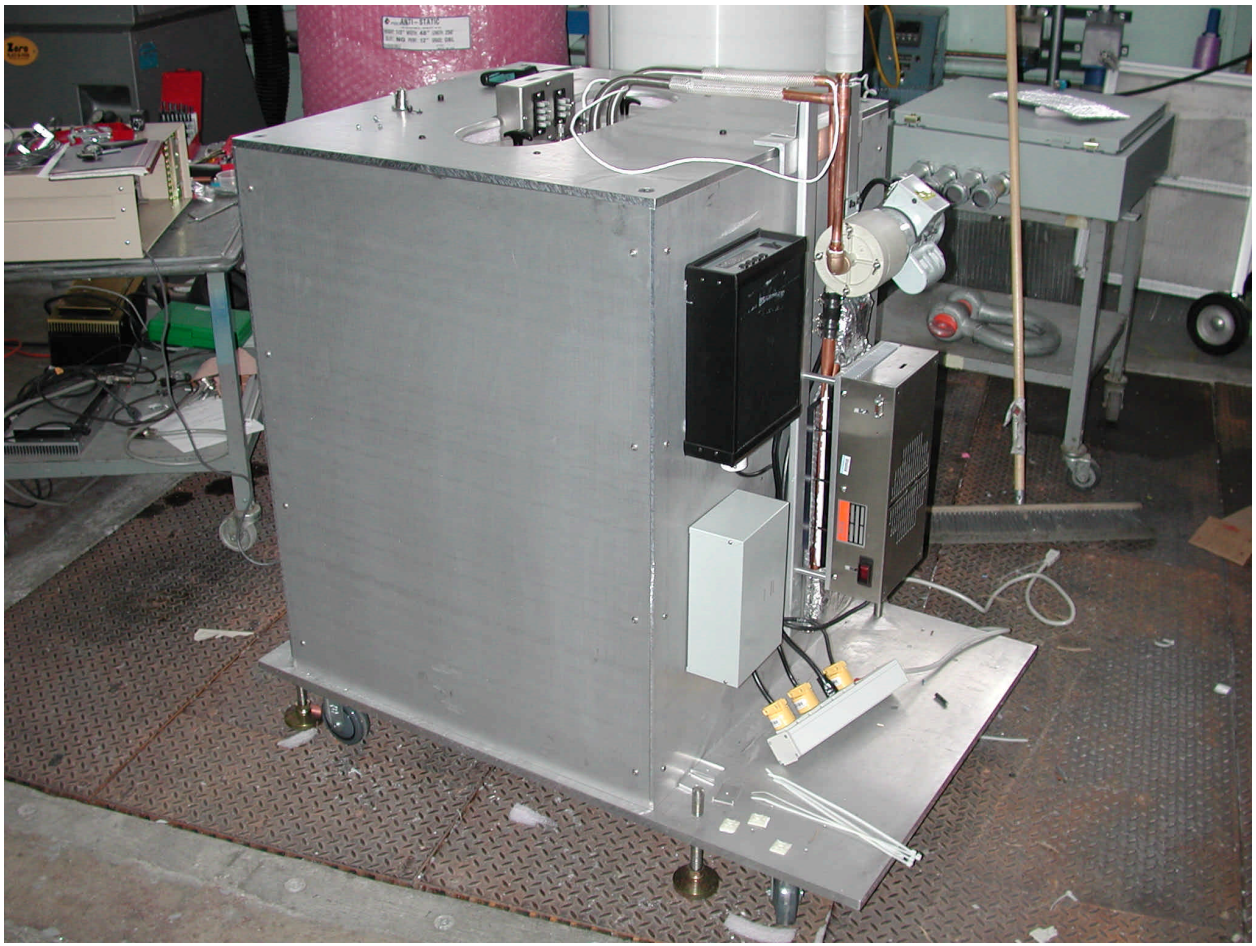
Uncertainties in the graph are standard deviation of the average

Transportable Cal Lab

Calorimeter #2 will be similar to this one:

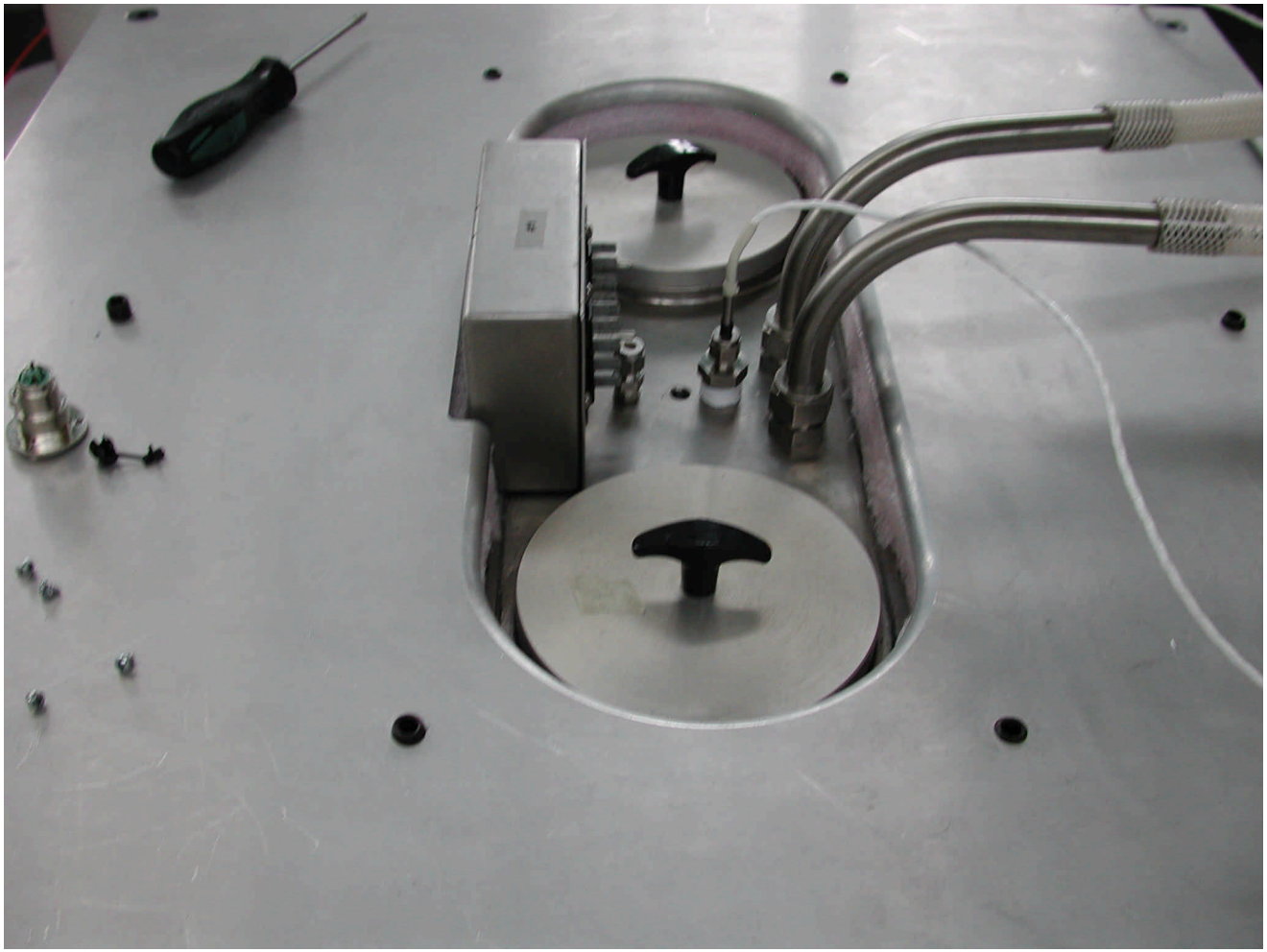
precision about 80 microWatts

Transportable Calorimeter #2

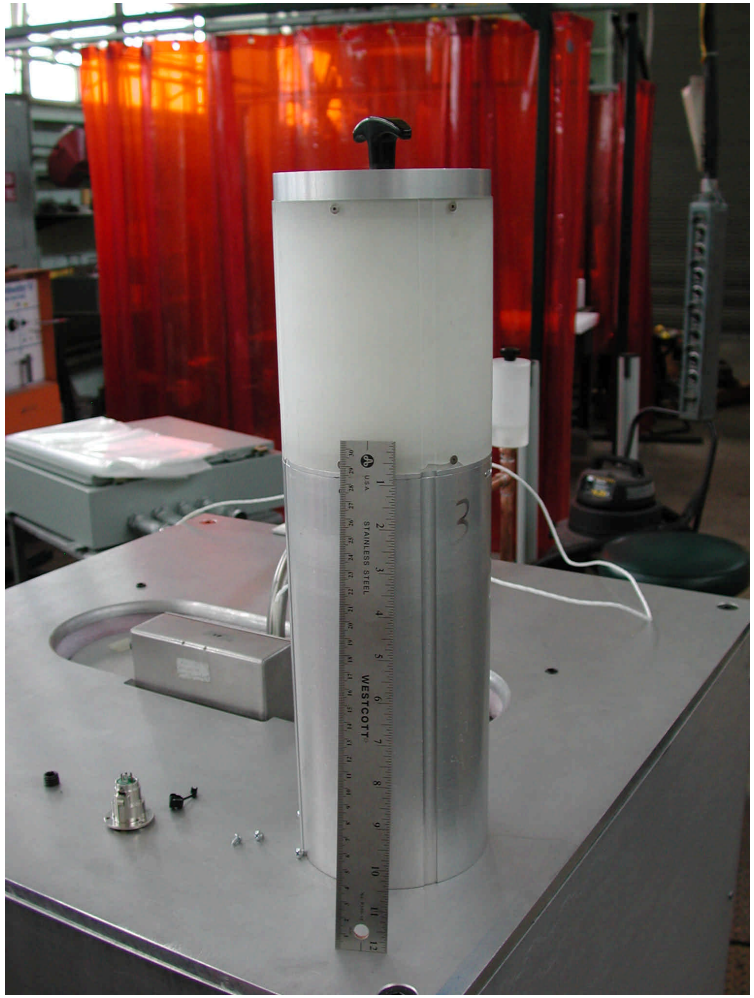


Transportable Calorimeter #2 Water Bath Temperature Control System

5” Calorimeter Top



Cal Can (with inner baffle)
5” Calorimeter



Calorimeter #3, 13.5 “ cal

- Holds a 5 gallon bucket
- Target precision = 100 microWatts
- Solid State sensors

Thermels for 13. 5” Diameter Calorimeter

Transportable Cal lab plans

- Ship lab to another DOE site to be used by lab or facility.
- LANL will train facility personnel in calorimeter operations
- 55 gallon drum calorimeter now under construction to be installed at later date

The Saga of the CALEX II Samples

Jay M. Thompson

New Brunswick Laboratory

Outline

- Creation of the CalEx II standards
- Results of analyses to date
- The samples from Hanford
- New adventures

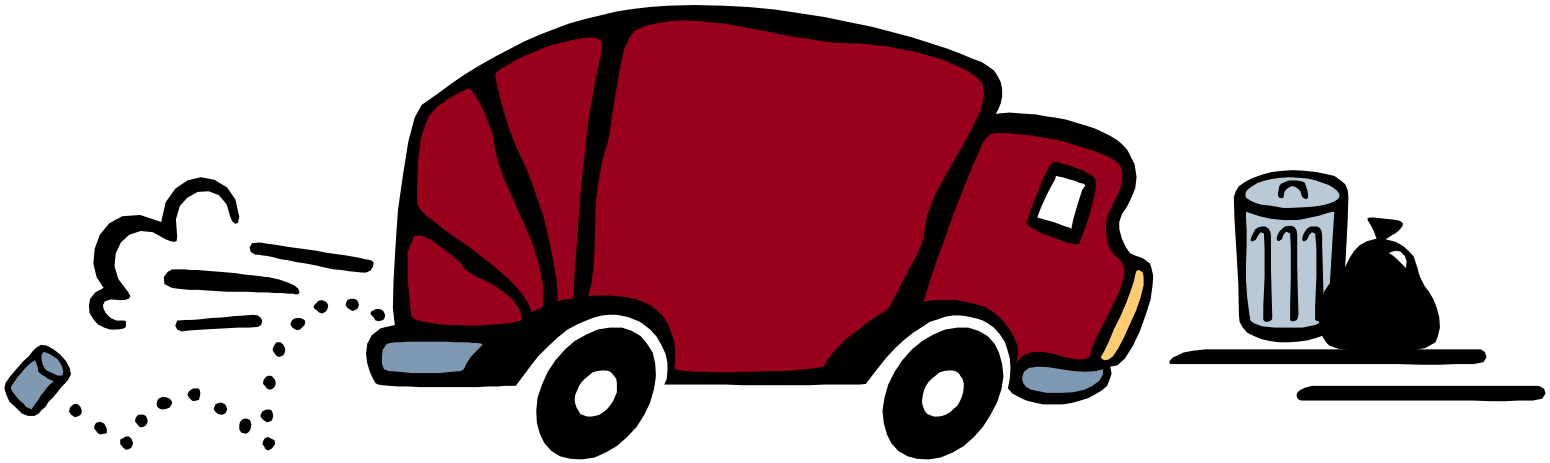
Creation of the CalEx II Standards

- Ten standards created at LANL in 1995
  - 2000.0 g of PuO_2 each
  - 12% ^{240}Pu
  - Approximately 6.2 watts each
  - Two at RF*
  - One each at LLNL, ANL-W, and SRS
  - Five at LANL

CalEx II Standards

- ~~10~~ 9 CalEx II standards

CalEx II Standards



The remaining CalEx II standards need to be preserved.

CalEx II Samples

- Three sample sets
  - LANL
  - NBL
  - Hanford
- LANL and NBL samples were analyzed

Plutonium Isotopics

- LANL and NBL isotopics agreed
- Interim values (decay date 10/26/1999)
  - Pu-238: 0.0780 Atom %
  - Pu-239: 86.7528
  - Pu-240: 12.1466
  - Pu-241: 0.8173
  - Pu-242: 0.2054

Americium-241

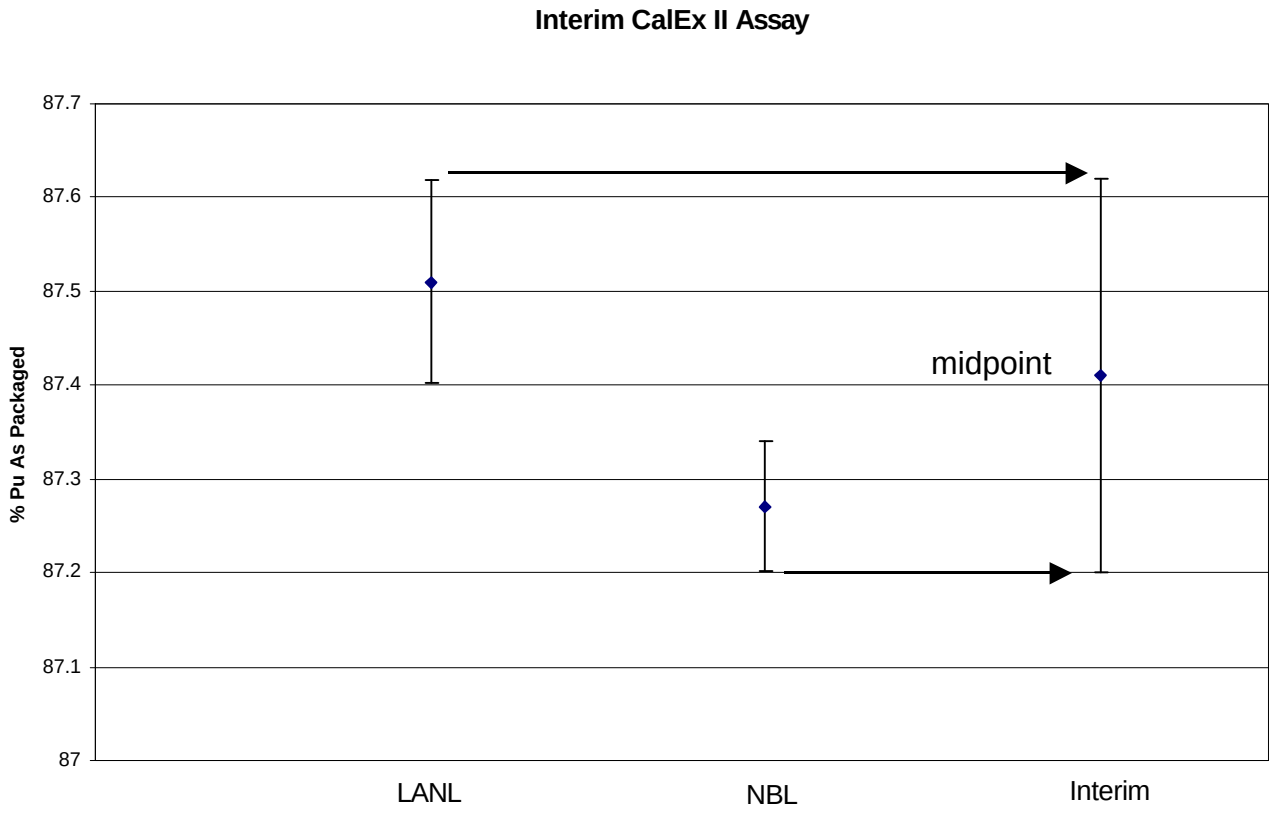
- Am-241: 5853 ug/g

Plutonium Assay (As Packaged)

- LANL: 87.510 wt% $1\sigma = 0.056$
- NBL: 87.271 $1\sigma = 0.036$

- Interim value: 87.41%Pu ± 0.21 (95% C.L.)

Interim Assay Value



New Brunswick Laboratory

Resolution of the Assay Difference

- Additional analysis is needed
- Sources of material
  - Hanford samples
  - Standards

The Acquisition Proceeded at a Remarkable Pace

The Saga of the CalEx II Samples

- The samples were shipped

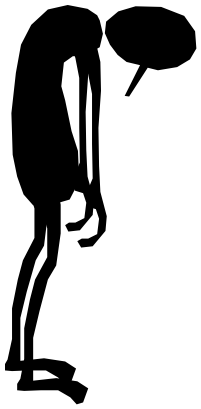
The Saga of the CalEx II Samples

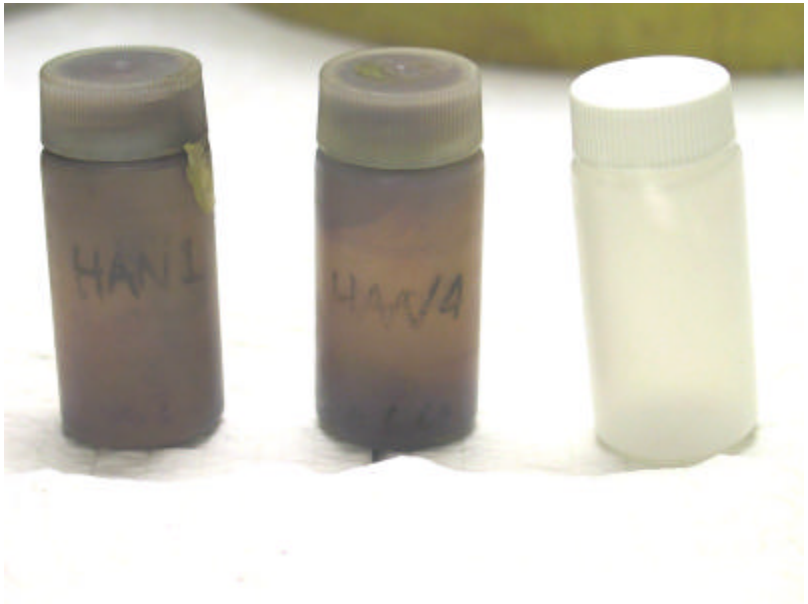
The samples arrived at NBL on May 14, 2002

A Celebration Followed

Sample Vials

- 18 ml HDPE vials
- Screw caps
- Non-inert atmosphere
- Moisture pickup
- External contamination





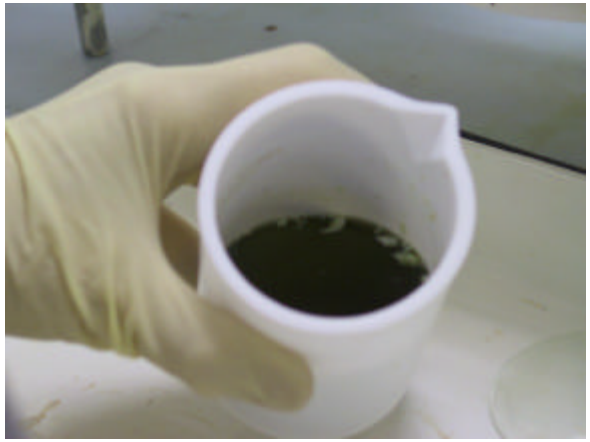
What Do We Do Next?

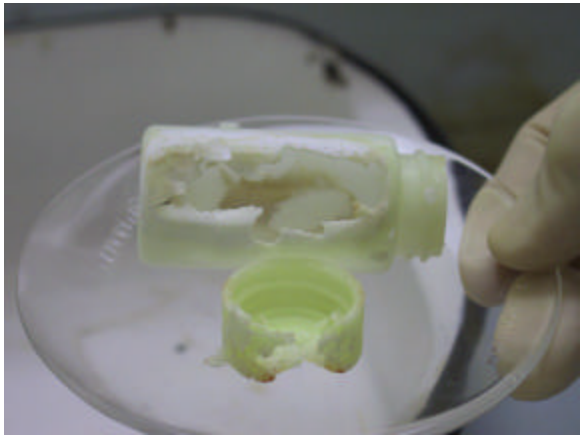
Analysis Plan

Analysis Plan

- Calorimetry
- Verification of isotopics
- Assay
  - total dissolution of contents
    - Measure residue in vial
  - use original packaged weight
  - Verify ^{241}Am

The ~~Vile~~ Vial Experiment





Conclusions

- Total dissolution not an option
- Anticipate more conventional analysis
  - Take subsamples
  - Calcine to constant weight
  - Coulometry
- Welcome suggestions for a new analysis plan

See You In Phoenix, Arizona in 2003!!

DOE AND NBL BACKGROUND AND MISSION

OWNERSHIP

The New Brunswick Laboratory (NBL) is owned and operated by the U.S. Department of Energy (DOE). Although it is part of the DOE Chicago Operations Office system, its primary sponsor is the Office of Plutonium, Uranium, and Special Materials Inventory (SO-62) in the DOE Office of Security.

DOE MISSION

DOE is entrusted to contribute to the welfare of the nation by providing the scientific foundation, technology, policy, and institutional leadership necessary to achieve efficiency in energy use, diversity in energy sources, a more productive and competitive economy, improved environmental quality, and a secure national defense.

NBL MISSION

NBL serves as the U.S. government central authority for nuclear materials measurements and measurement evaluation. It is also the U.S. government certifying authority for nuclear reference materials. These functions assure that the United States maintains an accurate and reliable nuclear safeguards program, particularly in the area of nuclear materials accountability. NBL program and technical capabilities not only enhance domestic nuclear security but also support international nonproliferation efforts. Its nuclear material measurements and measurement evaluation roles allow the federal government to perform independent technical audits and validate nuclear material measurements made by contractors. NBL also has the technical capability for the independent resolution of measurement and safeguards anomalies that may arise from nuclear operations and the transfer of materials between sites.

NBL HISTORY

NBL was established by the Atomic Energy Commission in New Brunswick, NJ in 1949. It was initially staffed by scientists from the National Bureau of Standards who had contributed to the science of measuring nuclear materials for the Manhattan Project. At first, the NBL mission was to provide the federal government with the capability to assay uranium-containing materials for the nation's developing atomic energy program. Over the years, NBL expanded its capabilities, improving methods and procedures, developing new ones, and certifying additional reference materials for use around the world. It incorporated the capability to make plutonium measurements in 1959. During the period from 1975 to 1977, NBL was relocated from New Jersey to the current site at Argonne, Illinois.

Since its beginning, NBL has been a center of excellence in the analytical chemistry and the science of measuring nuclear materials. In this role, NBL continues to make state-of-the-art measurements of elemental and isotopic composition for a wide range of nuclear materials.