

LA-UR-16-26709

Approved for public release; distribution is unlimited.

Title: Report on proposed improvements to FRAM uncertainty

Author(s): Yoho, Michael Duncan
Porterfield, Donovan R.
Rim, Jung Ho
Vo, Duc Ta

Intended for: Report

Issued: 2016-09-01

Disclaimer:

Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the Los Alamos National Security, LLC for the National Nuclear Security Administration of the U.S. Department of Energy under contract DE-AC52-06NA25396. By approving this article, the publisher recognizes that the U.S. Government retains nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.



Report on proposed improvements to FRAM uncertainty

M. Yoho
D. Porterfield
J. Rim
D. Vo

September 2016

UNCLASSIFIED



Operated by Los Alamos National Security, LLC for the U.S. Department of Energy's NNSA



My background

- 1st year PhD student UT Austin
- Department of Mechanical engineering
- NNIS fellowship
(Nuclear Nonproliferation International Safeguards Graduate Fellowship Program)
- UT advisor: S. Landsberger
(Nuclear and Radiation Engineering Program)
- LANL advisor: D. Porterfield
(C-AAC)



Clock tower



Austin, Texas

UNCLASSIFIED

Reason for proposed improvements

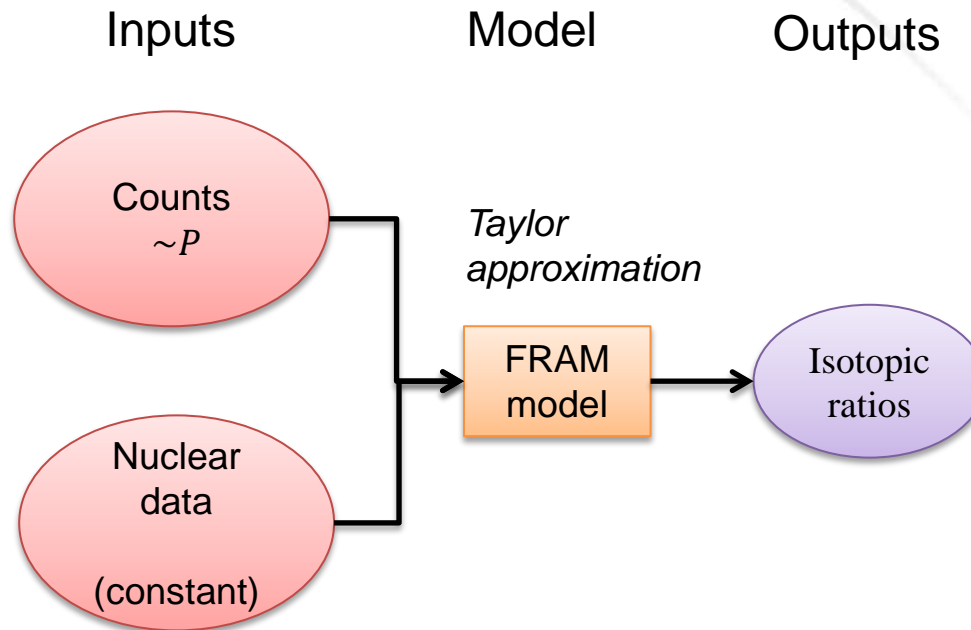
1. ISO 17025 requires accredited labs to include all relevant uncertainty contributors into calculations
2. C-AAC always strives to get the best data possible to provide results with overlapping uncertainties to clients (high sigma discrepancies observed between TIMS / NDA FRAM)

Plan of action to achieve these goals:

1. Develop new FRAM uncertainty model introducing more uncertainty contributors according to JCGM GUM (Joint Committee for Guides in Metrology, Guide to the Expression of Uncertainty in Measurement)
2. Critically analyze C-AAC NDA counting conditions

UNCLASSIFIED

Current FRAM uncertainty method



Current FRAM uncertainty method

UNCLASSIFIED

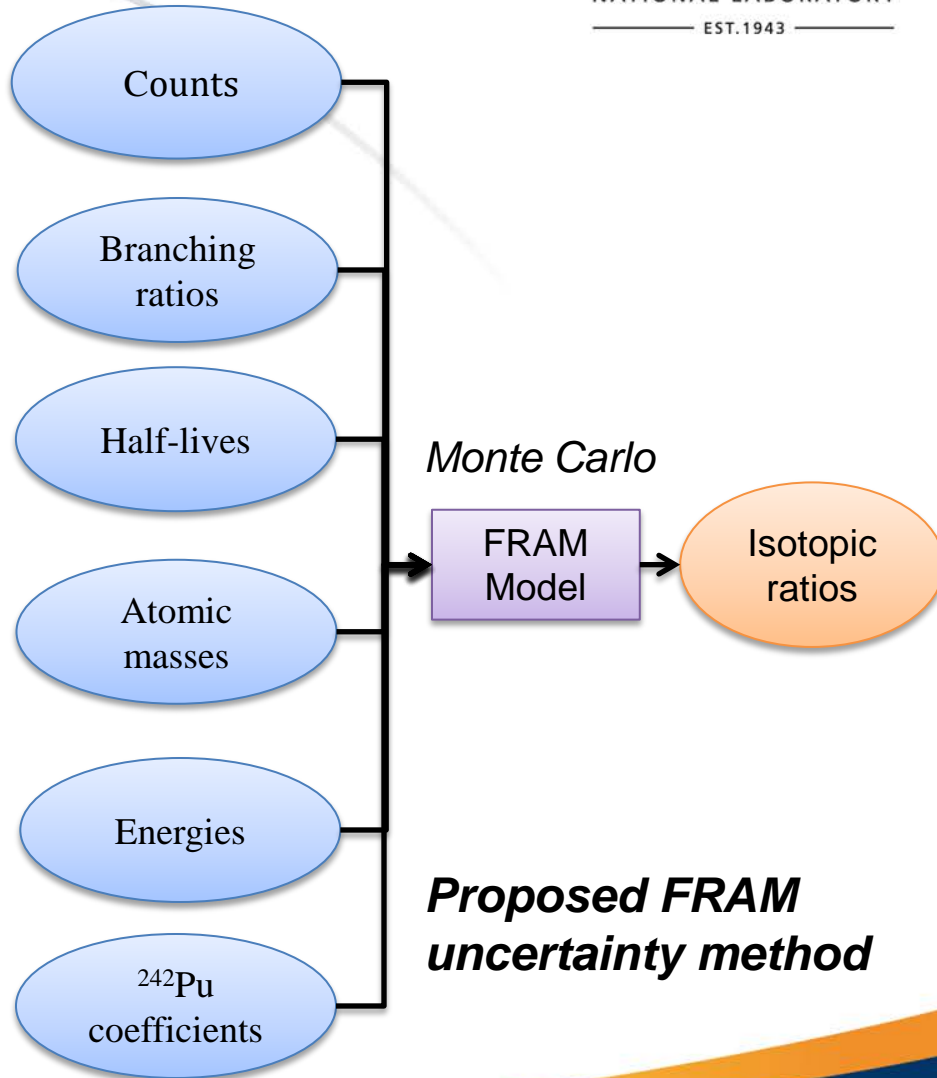
Introduce uncertainty from nuclear data:

Inputs:

1. Counts in each channel $\sim P$
2. Nuclear data $\sim N$
3. ^{242}Pu coefficients $\sim N$

Model: Keep FRAM model

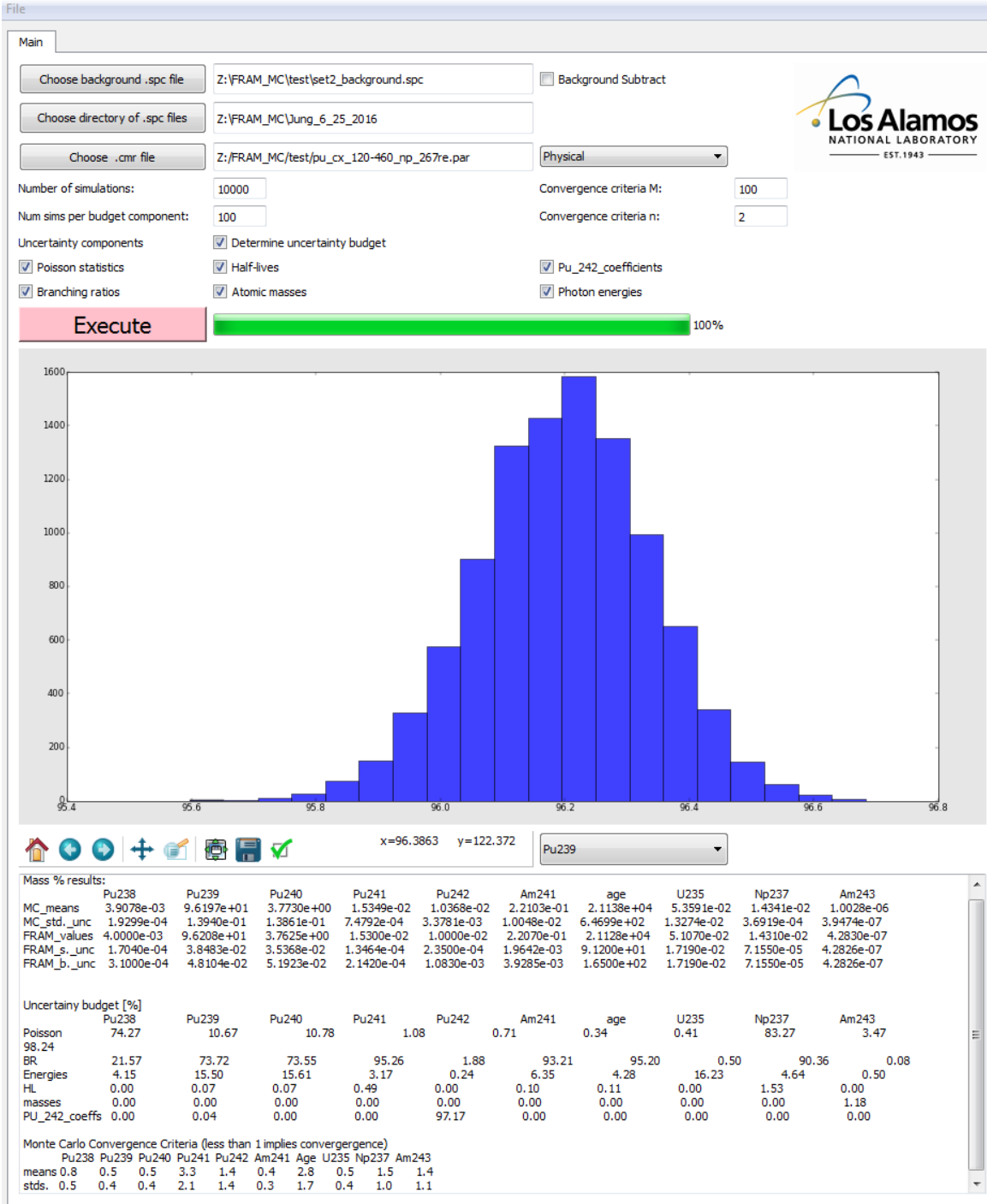
Outputs: Distributions found via Monte Carlo modelling



UNCLASSIFIED

Pre-alpha program features:

- Background subtraction
- Uncertainty budget generation
- Turn on/off uncertainty contributors
- Monte Carlo convergence criteria reporting
- Real-time visual feedback for mass % distributions
- Queueing



Problem: branching ratio correlation

- Studies measure multiple branching ratios with the same standard, geometry, etc.
- May introduce correlation
- Plan of action:
 - Determine covariance matrix from examining original studies
 - Model branching ratios as multivariate Normal

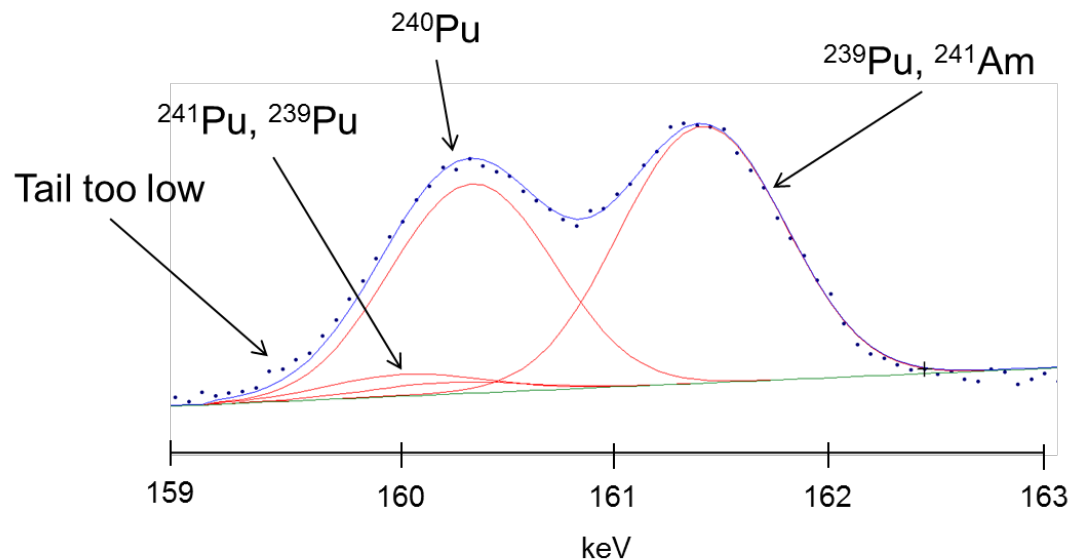
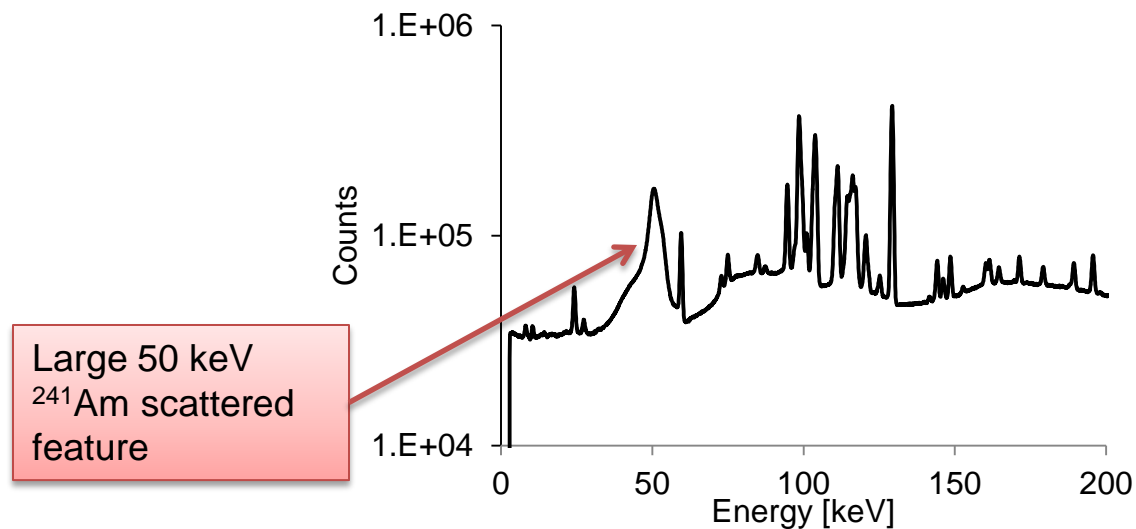
ENSDF file data for ^{239}Pu

Study	345 keV branching ratio	375 keV branching ratio	Relationship
1966 Ah02	8.7 (9)	25 (3)	
1976 GuZN	8.93 (18)	25.1 (5)	Both go up
1980 Despres	8.75 (30)	24.9 (8)	Both go down
1982 He02	8.67 (13)	24.2 (3)	Both go down
1984 lw02	8.61 (11)	24.2 (3)	Similar

UNCLASSIFIED

Additional uncertainty source: tail fitting

- No model is perfect
- For long counting times, tail model error will dominate Poisson statistics
- Exacerbated for spectra with high amount of scattered ^{241}Am photons
- 59 keV photons scatter off lead shield and coincidentally sum to around 158 and 162 keV, around tails of important ^{240}Pu 160 keV ROI



UNCLASSIFIED

^{240}Pu 160.3 keV ROI

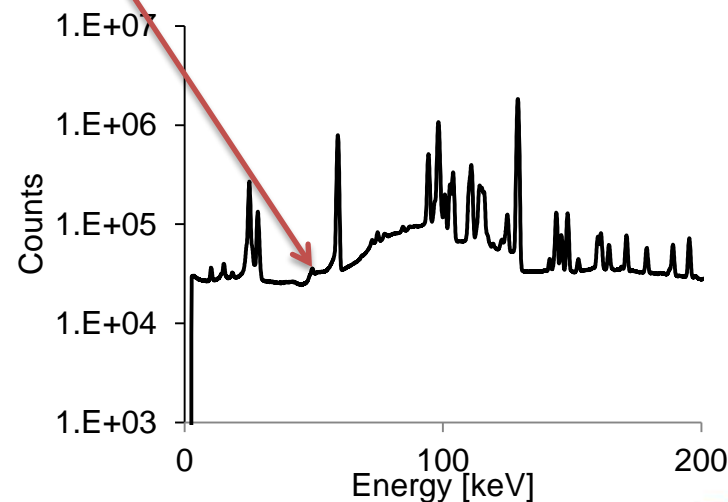
Plan to minimize ^{241}Am scattered photons: encase sample in pewter container

- Already in use
- Substantially reduces 50 keV spectral feature
- Cadmium cup alternative deemed overly toxic



Pewter container

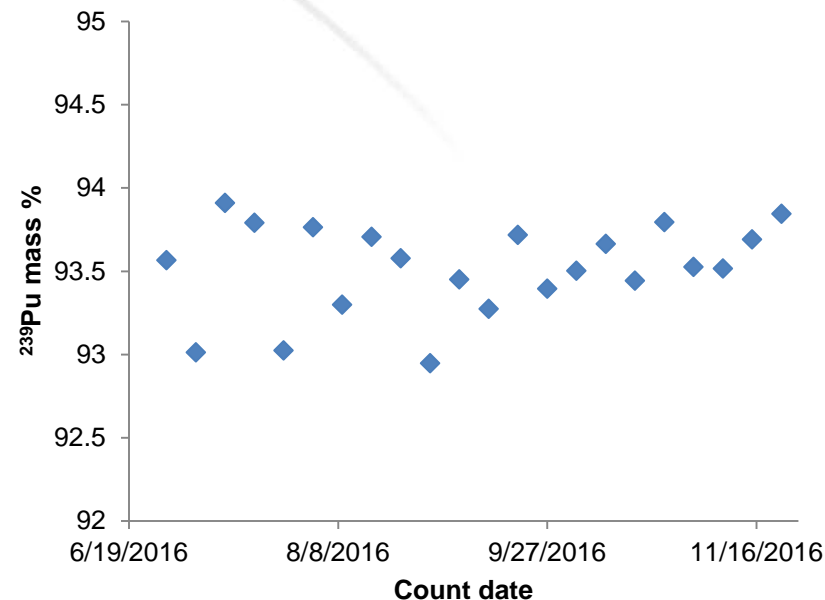
Reduced ^{50}keV feature



UNCLASSIFIED

Suggestion to deal with model inaccuracies

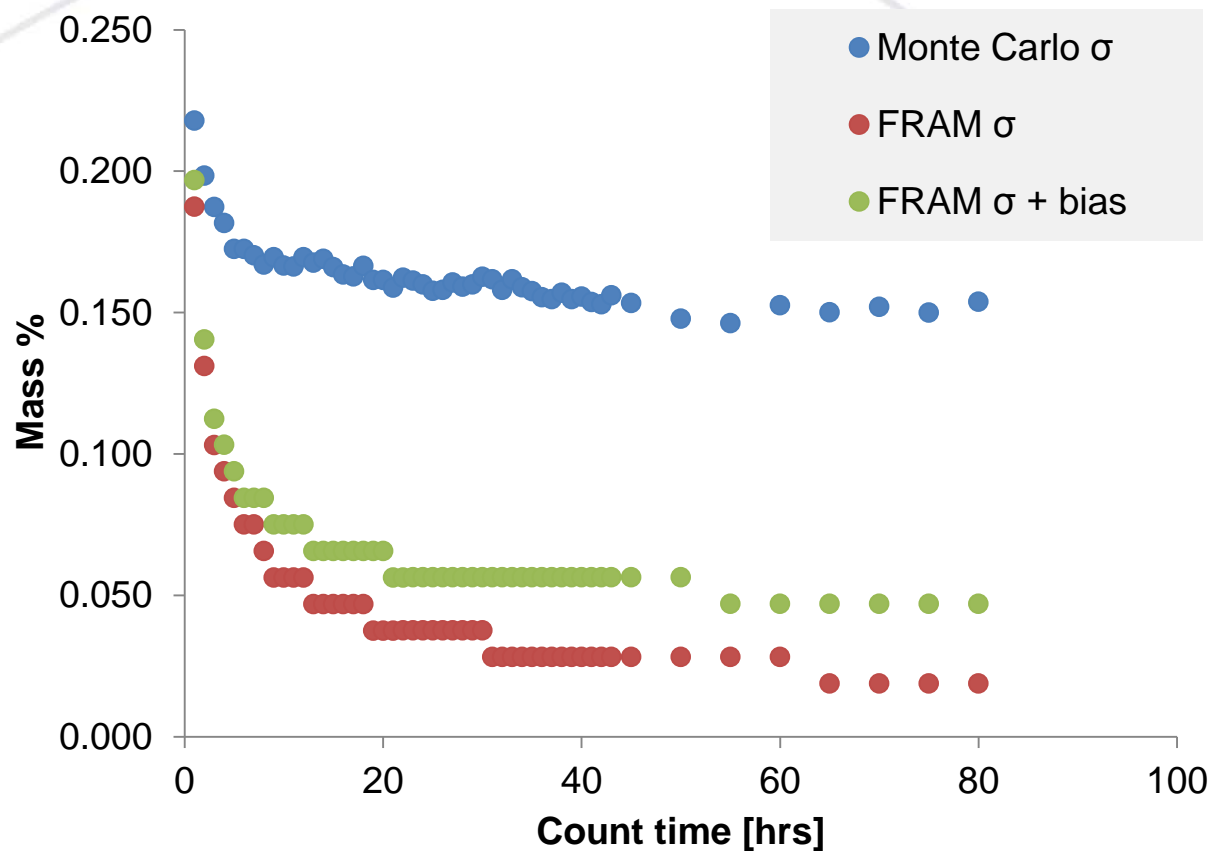
- Utilize control chart data with CRMs at specified:
 - Sample geometry
 - Sample height
 - Same detector
 - DT, etc...
- From this historic data, determine bias via NIST SOP No. 29 (2014)
- It is suggested to not alter the reported value, but rather increase the uncertainty
- Bias correction will be for specific C-AAC detectors with its specific counting configuration



Example control chart

UNCLASSIFIED

Comparison of MC uncertainty to FRAM uncertainty



Uncertainty for CRM 126A ^{239}Pu mass % as a function of counting time

UNCLASSIFIED

Discussion of comparison

- For counting times < 3 hours, Monte Carlo uncertainty approximates FRAM uncertainty (with reported bias)
- For longer counting times (3 – 100 hours) nuclear data uncertainty plays a larger role
- The next two slides depict biases between FRAM and DA/TIMS CRM mass % results $(\text{FRAM} - \text{CRM})/\sigma$
 - Chart 1: conventional FRAM σ
 - Chart 2: Monte Carlo σ

UNCLASSIFIED

Bias chart using conventional FRAM uncertainty

■ 2+ σ events: **44**
(33%)

■ 3+ σ events: **19**

■ 7+ σ events: **3**

■ Spectra with *
have significant
 ^{241}Am scattering
features at 50
keV (no pewter)

CRM	DT [%]	LT [h]	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am	^{237}Np
136	11.3	16	-1.6	0.2	0.3	-2.4		
126-A	14.7	16	2.8	-0.4	0.5	-3.8	0.1	-0.7
*126-A	14.5	16	1.1	-1.8	1.9	0.7	-0.6	-0.6
*126-A	11.5	17	0.1	-1.2	1.2	0.1	-0.1	-0.1
126-A	24.8	19	-2.0	1.5	-1.4	-2.6	0.9	-0.8
138	11.1	19	-0.5	0.6	-0.6	-2.8		
*126-A	12.6	23	0.9	-2.6	2.7	0.1	-2.3	-1.6
*126-A	11	24	0.8	-1.5	1.5	-0.9	-1.4	-0.4
137	11.9	27	-1.3	2.4	-0.7	-0.3		
*126-A	14.1	28	0.0	-2.8	2.7	-1.6	0.4	-1.4
*126-A	13.3	41	1.7	-2.5	2.6	-2.6	-1.1	-1.7
*126-A	13.9	61	2.5	-2.3	2.4	-1.0	0.0	-1.4
*126-A	13.4	64	1.4	-6.0	5.9	-3.1	-1.5	-1.8
*126-A	11.1	66	0.2	-1.3	1.4	-1.4	-1.0	-0.6
138	6.3	71	-0.2	-2.7	2.8	-2.7		
*126-A	6.8	80	3.7	-0.3	0.8	-0.8	2.3	-1.1
126-A	12.5	83	6.2	0.1	0.7	-0.4	2.5	-0.9
136	8.8	84	-0.7	4.9	-3.1	-1.2		
126-A	8.5	86	4.1	-0.9	1.4	-1.0	2.4	-0.8
DHS13LANL1A	8.4	86	-1.0	-4.7	4.6	-3.8	1.2	2.0
*126-A	13	102	1.1	-6.8	7.2	-4.1	-1.6	-1.3
*126-A	13.3	126	2.7	-7.7	7.6	-3.3	-1.1	-1.4
137	8.5	127	-1.1	5.1	-0.8	-1.7		
		Ave. bias:	0.9	-1.3	1.8	-1.8	-0.1	-0.9

UNCLASSIFIED



Bias chart using Monte Carlo uncertainty

■ 2+ σ events: **11**
(9%)

■ 3+ σ events: **0**

■ All average
biases below
1 σ

CRM	DT [%]	LT [h]	²³⁸ Pu	²³⁹ Pu	²⁴⁰ Pu	²⁴¹ Pu	²⁴¹ Am	²³⁷ Np
136	11.3	16	-1.1	0.1	0.2	-0.3		
126-A	14.7	16	2.7	-0.2	0.3	-0.4	0.1	-0.4
126-A	14.5	16	1.1	-1.1	1.2	0.1	-0.2	-0.4
126-A	11.5	17	0.2	-1.0	1.1	0.0	-0.1	-0.1
126-A	24.8	19	-2.0	1.3	-1.2	-0.8	0.7	-0.6
138	11.1	19	-0.5	0.3	-0.3	-0.5		
126-A	12.6	23	0.8	-1.6	1.6	0.0	-0.6	-1.0
126-A	11.0	24	0.8	-1.4	1.4	-0.3	-0.5	-0.3
137	11.9	27	-0.8	0.7	-0.3	0.0		
126-A	14.1	28	0.0	-1.3	1.4	-0.2	0.2	-0.9
126-A	13.3	41	1.7	-1.2	1.3	-0.2	-0.6	-1.0
126-A	13.9	61	2.1	-0.8	0.8	-0.1	0.0	-0.9
126-A	13.4	64	1.3	-2.3	2.4	-0.4	-0.5	-1.1
126-A	11.1	66	0.2	-1.0	1.0	-0.3	-0.4	-0.4
138	6.3	71	-0.2	-0.5	0.5	-0.3		
126-A	6.8	80	1.4	0.0	0.1	-0.1	0.7	-0.7
126-A	12.5	83	1.4	0.0	0.1	0.0	0.7	-0.6
136	8.8	84	-0.4	0.7	-0.5	-0.1		
126-A	8.5	86	1.4	-0.1	0.2	-0.1	0.6	-0.5
DHS13LANL1A	8.4	86	-0.9	-1.1	1.1	-0.7	0.5	1.1
126-A	13.0	102	0.9	-2.3	2.4	-0.4	-0.6	-0.8
126-A	13.3	126	2.1	-2.2	2.3	-0.3	-0.4	-0.9
137	8.5	127	-0.6	0.6	-0.1	-0.2		
		Ave. bias:	0.5	-0.6	0.7	-0.2	0.0	-0.6

UNCLASSIFIED



Discussion

- It is unclear if the discrepancy between CRM sheets and FRAM is due to
 1. High ^{241}Am scattering
 2. Not taking into account uncertainty from nuclear data
- For these long duration spectra, introducing nuclear data uncertainty improved statistical agreement between CRM sheets and FRAM

UNCLASSIFIED

Discussion / future work

- Note that the nuclear data uncertainty model does not improve FRAM accuracy:
 - Rather, it is an attempt to get a more accurate picture of uncertainty

- Future work:
 1. Introduce branching ratio correlations into model
 2. Determine if pewter improves FRAM accuracy
 3. Analyze FRAM 200 spectra dataset used in previous ANOVA study
 4. Consider implementation of control chart bias correction
 5. Count for shorter durations

UNCLASSIFIED

Thank you

UNCLASSIFIED