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Title: Report on proposed improvements to FRAM uncertainty

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Report on proposed improvements to FRAM uncertainty

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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

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Reason for proposed improvements

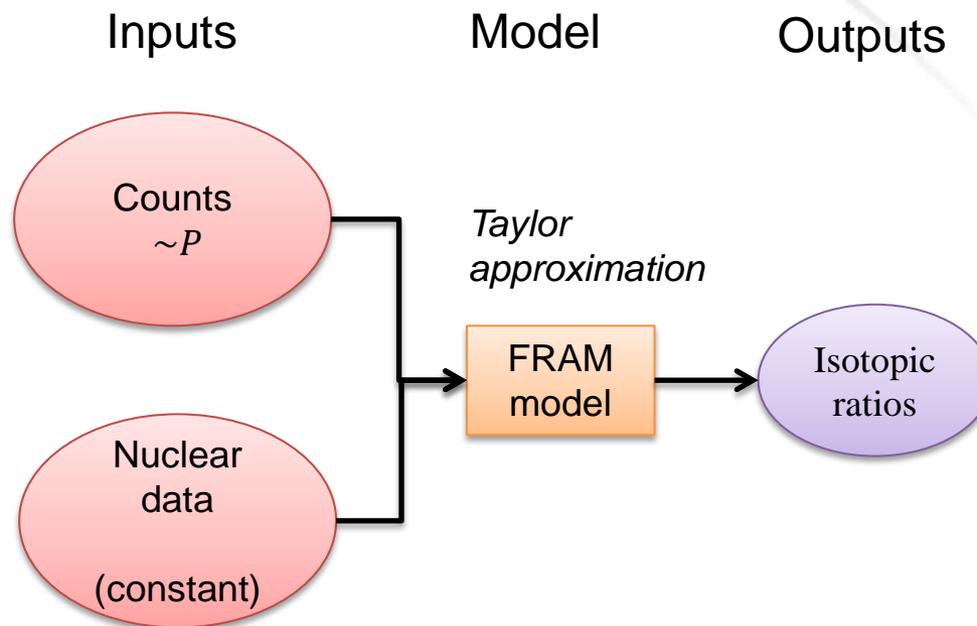
1. ISO 17025 requires accredited labs to include all relevant uncertainty contributors into calculations
2. C-AAC always strives to get the best data possible to provide results with overlapping uncertainties to clients (high sigma discrepancies observed between TIMS / NDA FRAM)

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

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Current FRAM uncertainty method



Current FRAM uncertainty method

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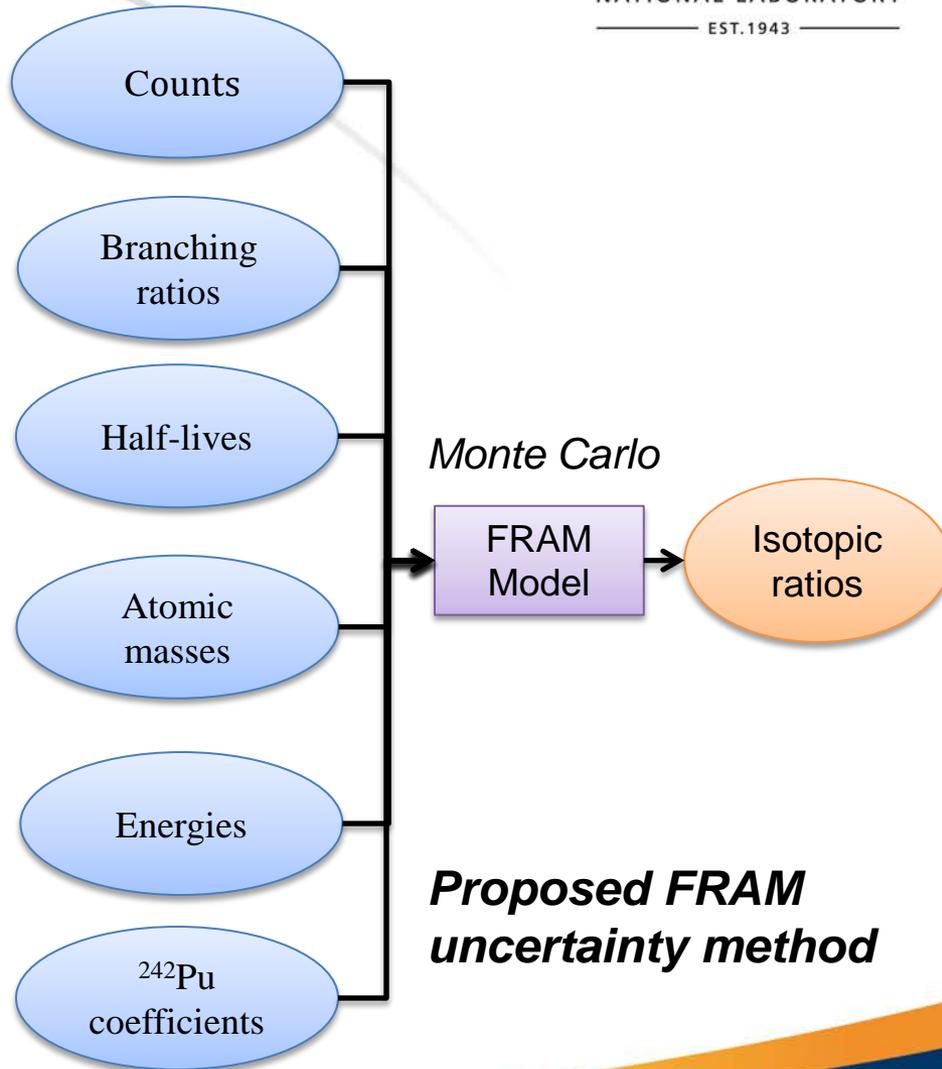
Introduce uncertainty from nuclear data:

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



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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



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File

Choose background .spc file: Z:\FRAM_MC\test\set2_background.spc

Choose directory of .spc files: Z:\FRAM_MC\Jung_6_25_2016

Choose .cmr file: Z:\FRAM_MC\test\pu_cx_120-460_np_267re.par

Background Subtract

Physical

Number of simulations: 10000

Convergence criteria M: 100

Num sims per budget component: 100

Convergence criteria n: 2

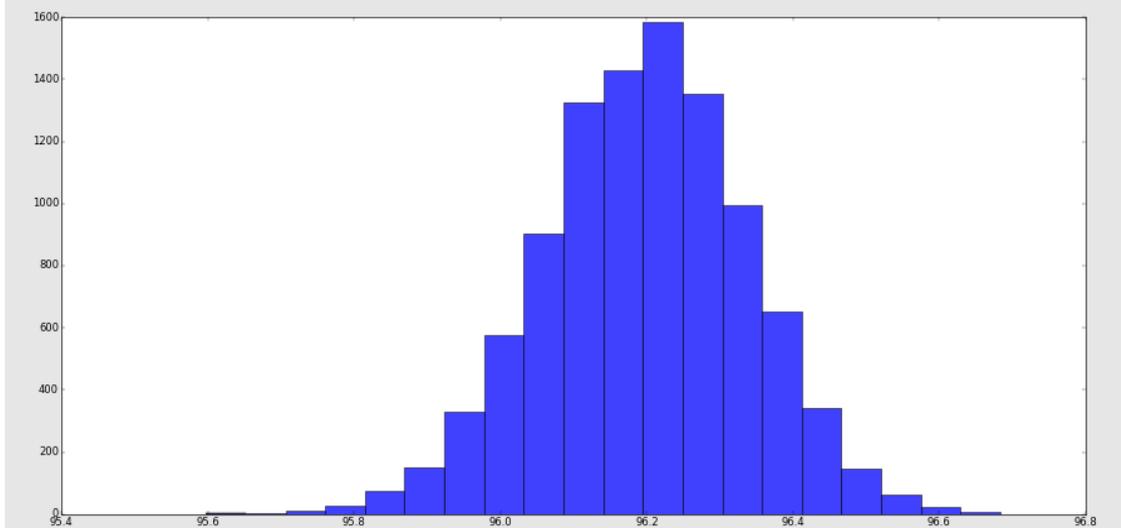
Uncertainty components

Determine uncertainty budget

Poisson statistics Half-lives Pu_242_coefficients

Branching ratios Atomic masses Photon energies

Execute ▶ 100%



x=96.3863 y=122.372 Pu239

Mass % results:

| | Pu238 | Pu239 | Pu240 | Pu241 | Pu242 | Am241 | age | U235 | Np237 | Am243 |
|-------------|------------|------------|------------|------------|------------|------------|------------|------------|------------|------------|
| MC_means | 3.9078e-03 | 9.6197e+01 | 3.7730e+00 | 1.5349e-02 | 1.0368e-02 | 2.2103e-01 | 2.1138e+04 | 5.3591e-02 | 1.4341e-02 | 1.0028e-06 |
| MC_std_unc | 1.9299e-04 | 1.3940e-01 | 1.3861e-01 | 7.4792e-04 | 3.3781e-03 | 1.0048e-02 | 6.4699e+02 | 1.3274e-02 | 3.6919e-04 | 3.9474e-07 |
| FRAM_values | 4.0000e-03 | 9.6208e+01 | 3.7625e+00 | 1.5300e-02 | 1.0000e-02 | 2.2070e-01 | 2.1128e+04 | 5.1070e-02 | 1.4310e-02 | 4.2830e-07 |
| FRAM_s_unc | 1.7040e-04 | 3.8483e-02 | 3.5368e-02 | 1.3464e-04 | 2.3500e-04 | 1.9642e-03 | 9.1200e+01 | 1.7190e-02 | 7.1550e-05 | 4.2826e-07 |
| FRAM_b_unc | 3.1000e-04 | 4.8104e-02 | 5.1923e-02 | 2.1420e-04 | 1.0830e-03 | 3.9285e-03 | 1.6500e+02 | 1.7190e-02 | 7.1550e-05 | 4.2826e-07 |

Uncertainty budget [%]

| | Pu238 | Pu239 | Pu240 | Pu241 | Pu242 | Am241 | age | U235 | Np237 | Am243 |
|---------------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|
| Poisson | 74.27 | 10.67 | 10.78 | 1.08 | 0.71 | 0.71 | 0.34 | 0.41 | 83.27 | 3.47 |
| 98.24 | | | | | | | | | | |
| BR | 21.57 | 73.72 | 73.55 | 95.26 | 1.88 | 93.21 | 95.20 | 0.50 | 90.36 | 0.08 |
| Energies | 4.15 | 15.50 | 15.61 | 3.17 | 0.24 | 6.35 | 4.28 | 16.23 | 4.64 | 0.50 |
| HL | 0.00 | 0.07 | 0.07 | 0.49 | 0.00 | 0.10 | 0.11 | 0.00 | 1.53 | 0.00 |
| masses | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 1.18 |
| PU_242_coeffs | 0.00 | 0.04 | 0.00 | 0.00 | 0.00 | 97.17 | 0.00 | 0.00 | 0.00 | 0.00 |

Monte Carlo Convergence Criteria (less than 1 implies convergence)

| | Pu238 | Pu239 | Pu240 | Pu241 | Pu242 | Am241 | Age | U235 | Np237 | Am243 |
|-------|-------|-------|-------|-------|-------|-------|-----|------|-------|-------|
| means | 0.8 | 0.5 | 0.5 | 3.3 | 1.4 | 0.4 | 2.8 | 0.5 | 1.5 | 1.4 |
| stds. | 0.5 | 0.4 | 0.4 | 2.1 | 1.4 | 0.3 | 1.7 | 0.4 | 1.0 | 1.1 |



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

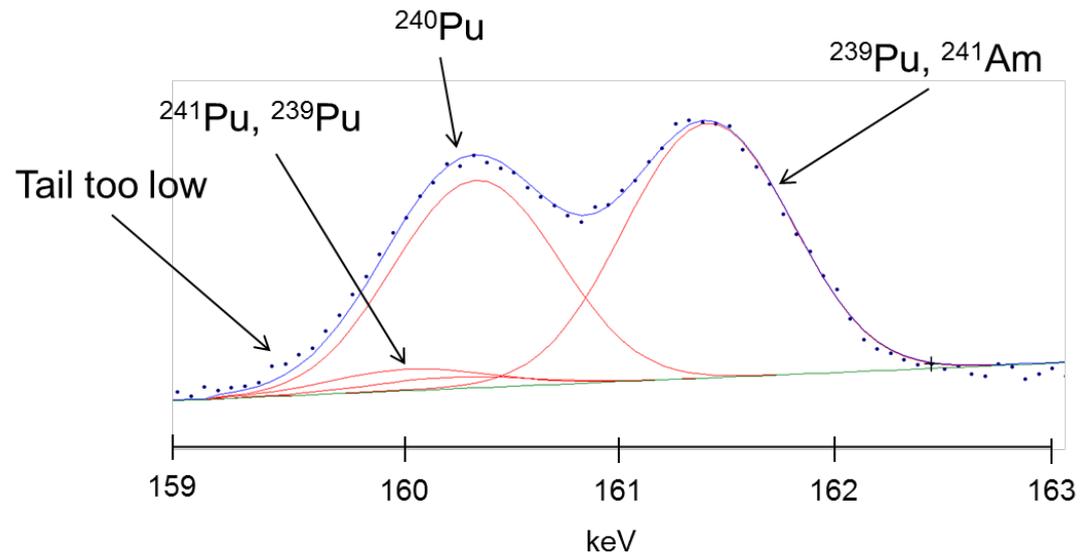
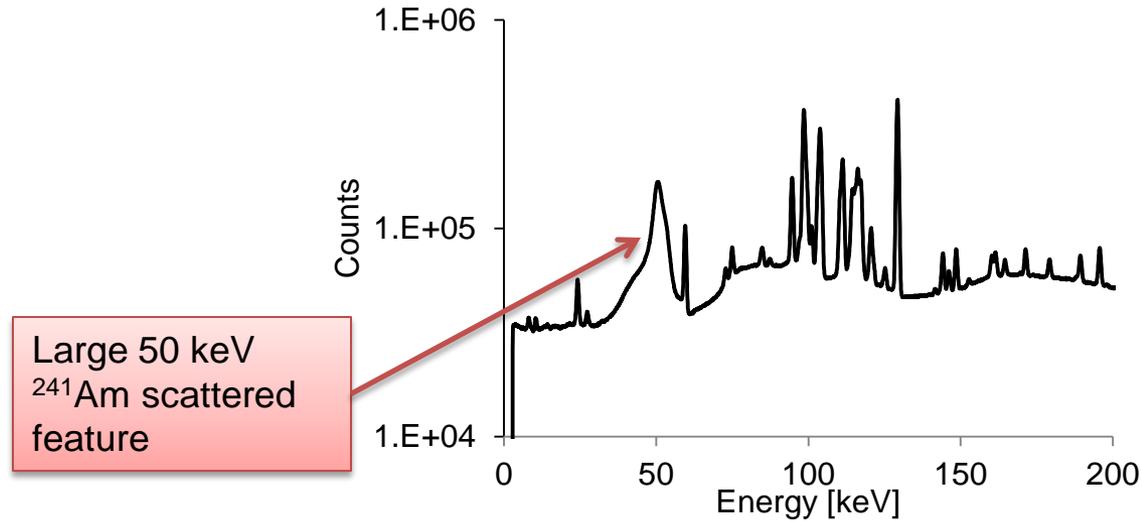
ENSDF file data for ^{239}Pu

| 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 |

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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

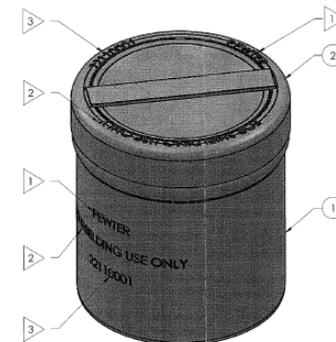


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^{240}Pu 160.3 keV ROI

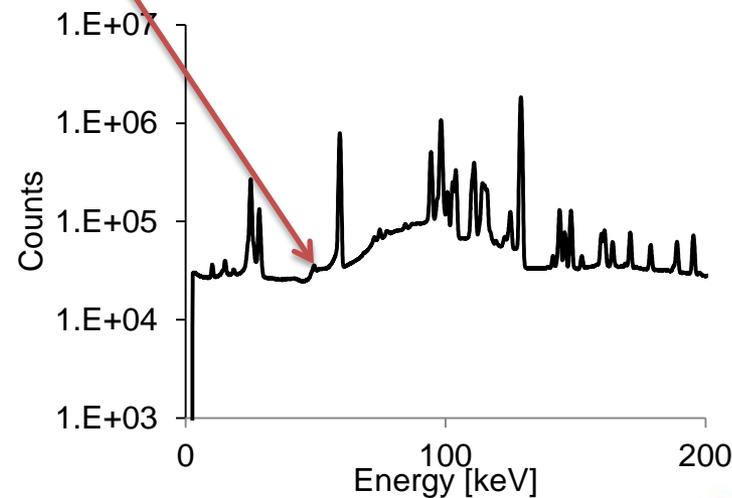
Plan to minimize ^{241}Am scattered photons: encase sample in pewter container

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



Pewter container

Reduced ^{50}keV feature



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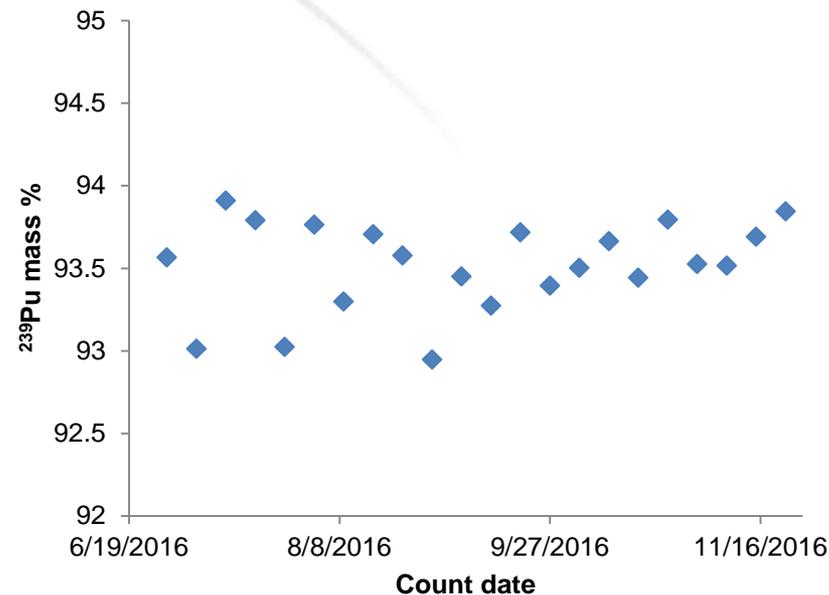
Suggestion to deal with model inaccuracies

- 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

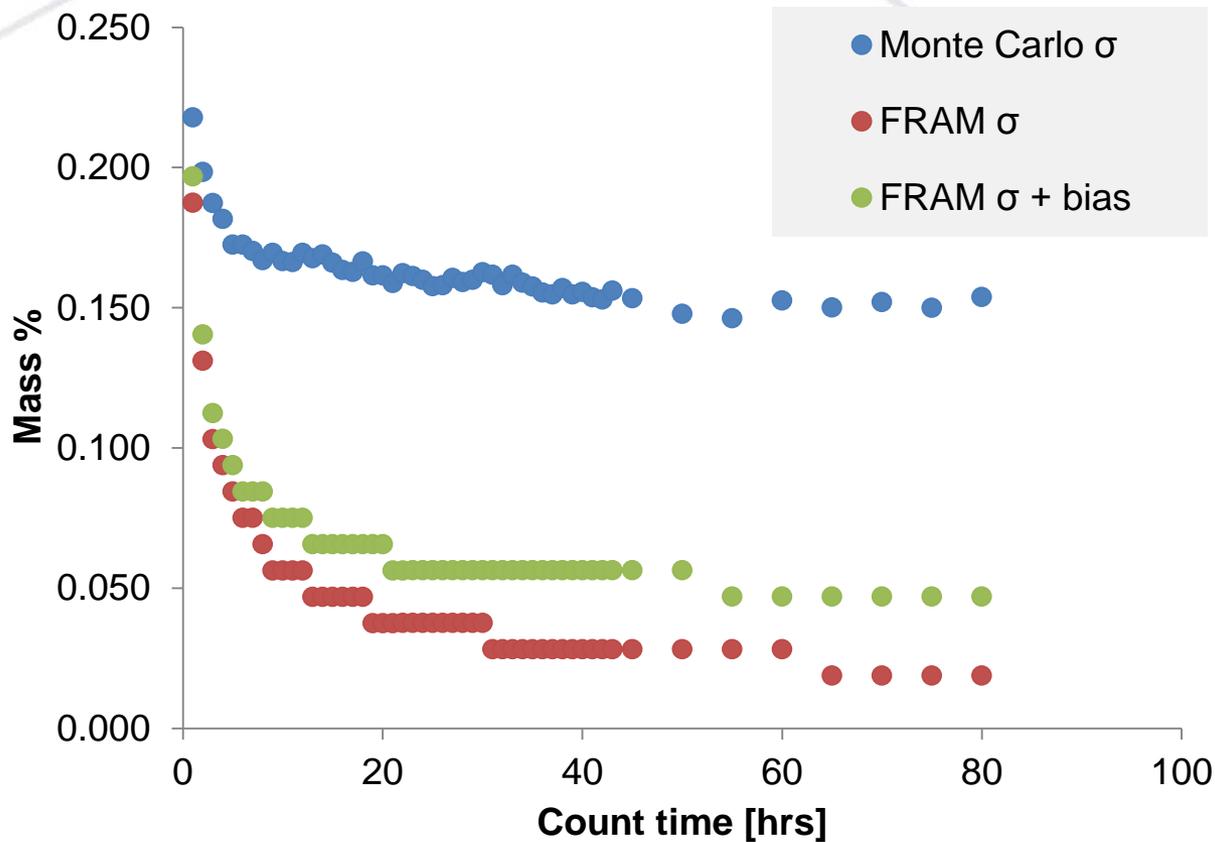
- Bias correction will be for specific C-AAC detectors with its specific counting configuration



Example control chart

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Comparison of MC uncertainty to FRAM uncertainty



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

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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

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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 |

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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

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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

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Thank you

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