Nuclear Data Activities on Fission Yields at LBNL

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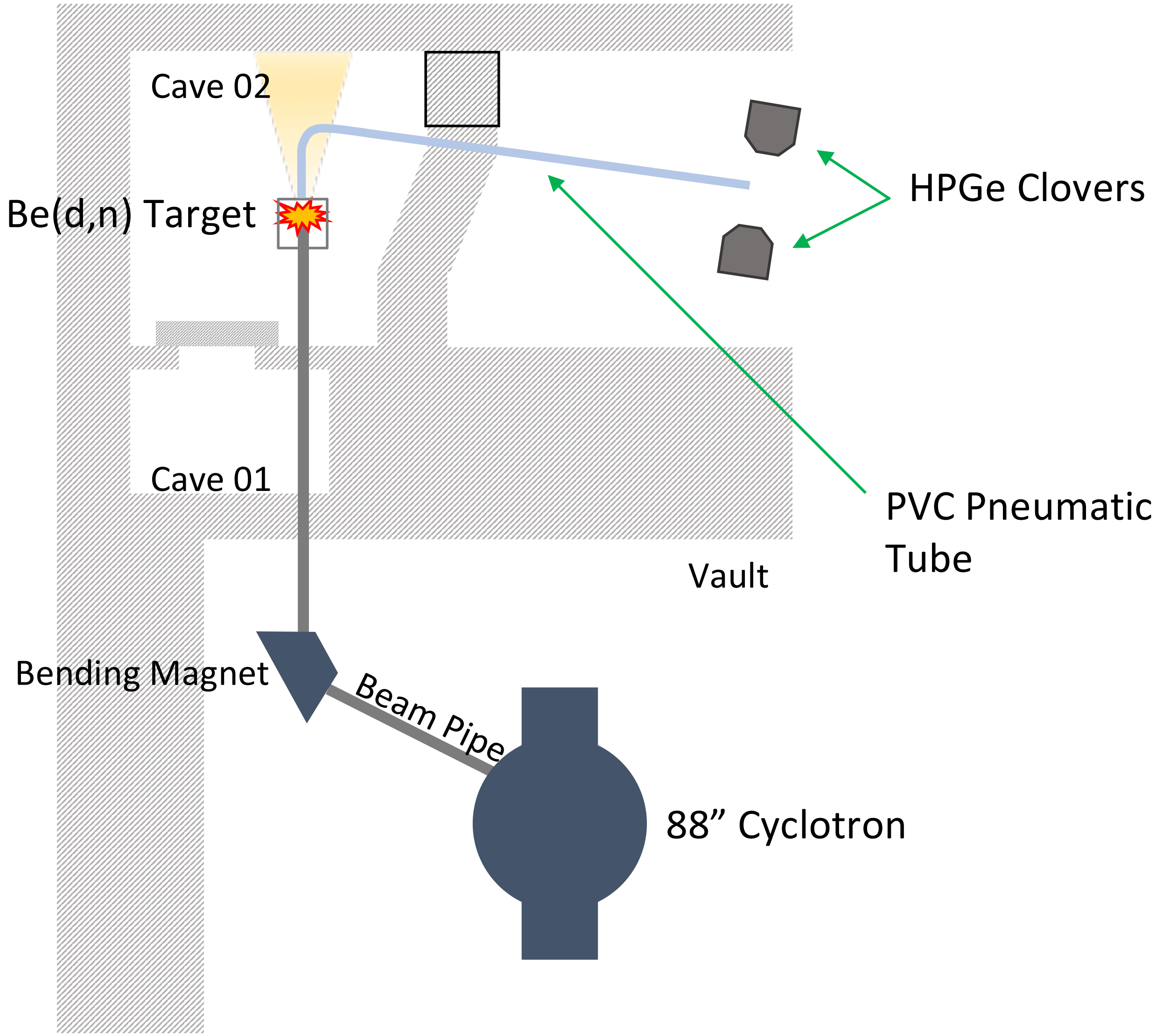
October 19, 2021

Fission Yield Measurements at the Fast Loading User Facility for Fission Yields (FLUFFY)

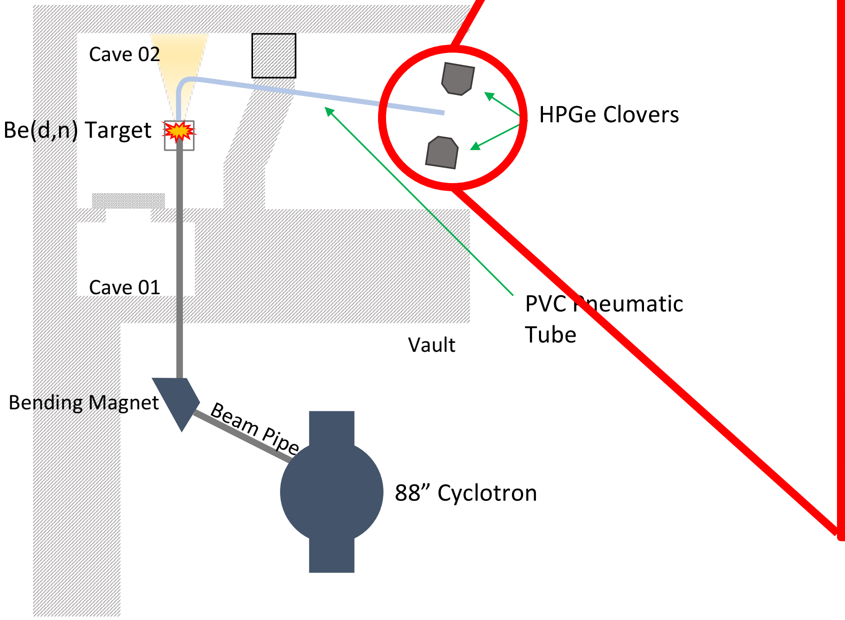
FLUFFY

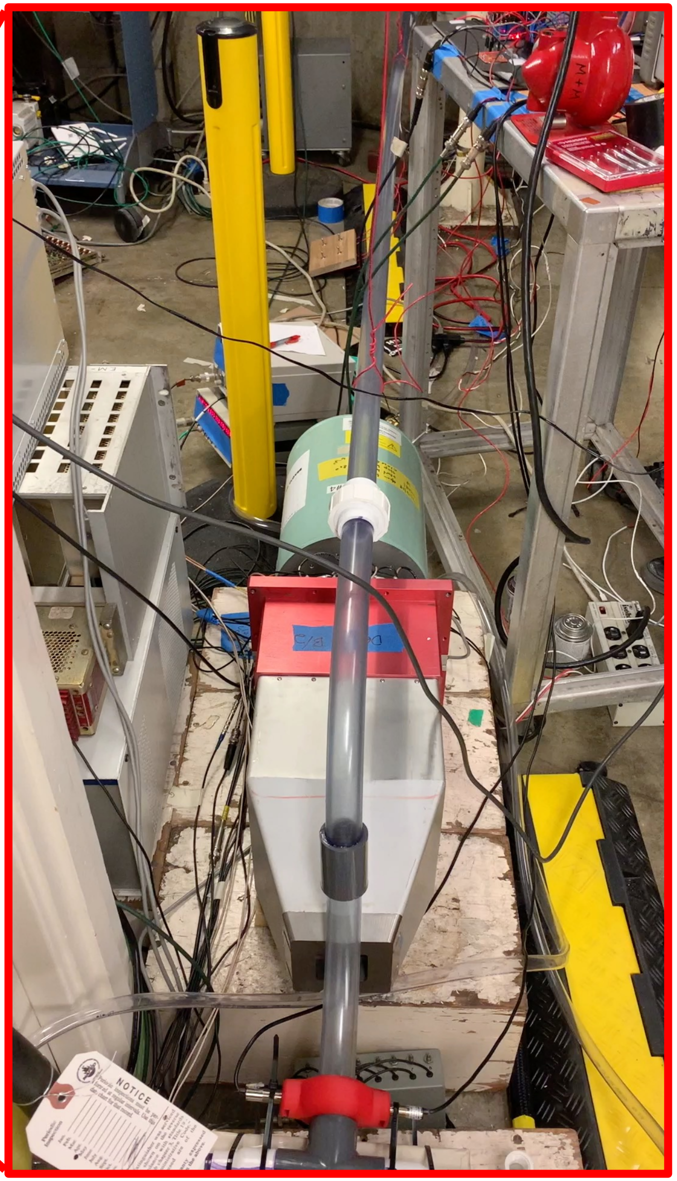
- The Fast Loading User Facility for Fission Yields (FLUFFY) was developed at LBNL to rapidly shuttle actinide samples between a neutron source and counting array.
- Transport times of <1 second allow observation of short-lived fission products.



FLUFFY

- Flux: **7.2 x 10^8 n/cm^2/s**
- This high flux along with the rapid transport time allows for the observation of 80+% of the yield in peak mass chains.

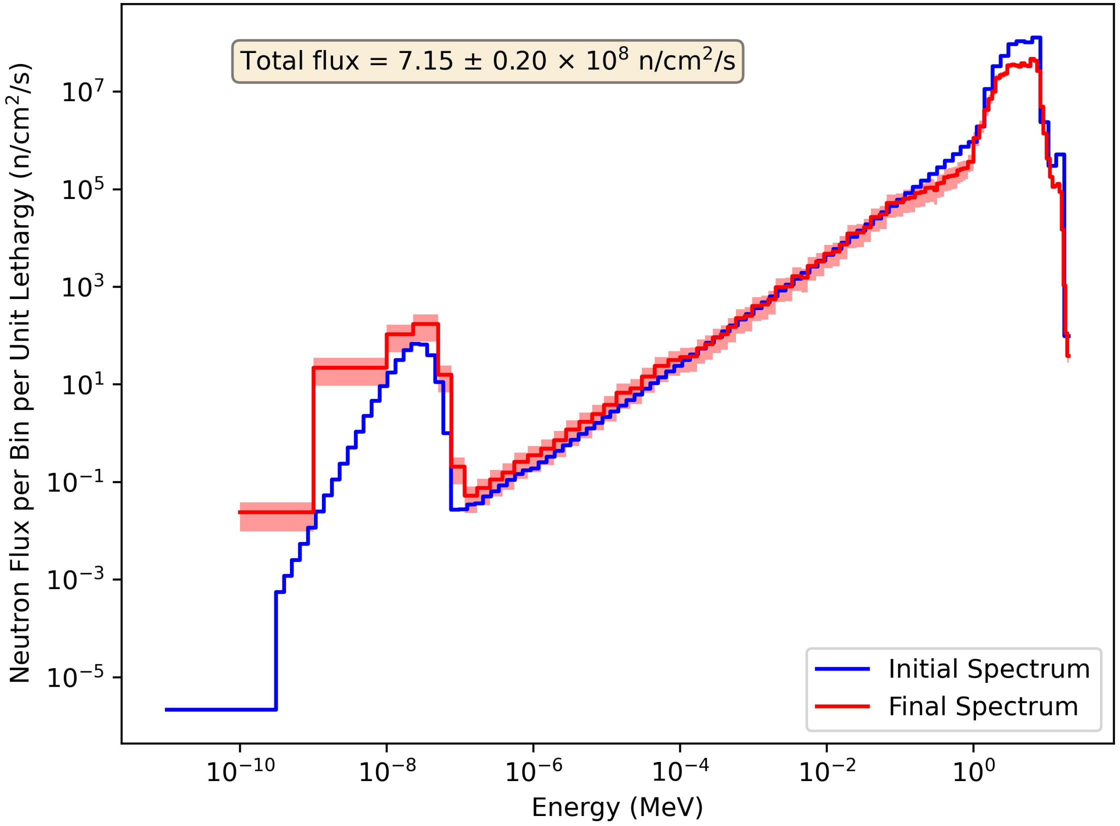




July 2020 Experiment

- On July 21-26, 2020, ^{238}U and ^{235}U samples were irradiated at the Fast Loading User Facility for Fission Yields (FLUFFY) at LBNL's 88-inch cyclotron.
  - 24 hours of 1 s-25 s ^{238}U data (455.3 mg)
  - 24 hours of 5 s-125 s ^{238}U data (455.3 mg)
  - ~16 hours of 1 s-25 s ^{235}U data (~20 mg)
  - Neutron energy spectrum data for

  14 MeV deuteron breakup on graphite
    - 8 hour foil pack irradiation
    - Single nTOF irradiation

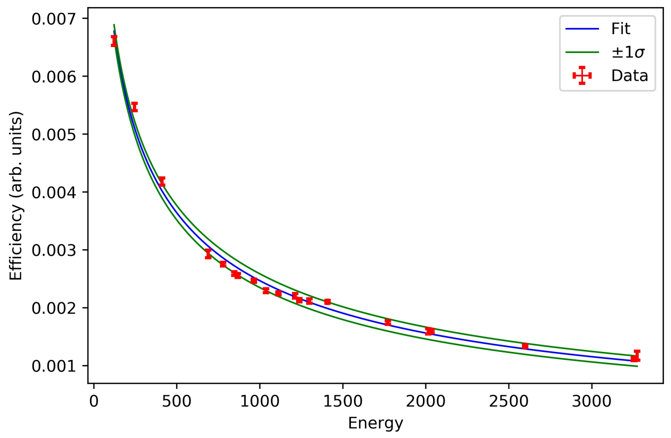


Measured energy spectrum for 14 MeV deuteron breakup on graphite.

The Experimental Data

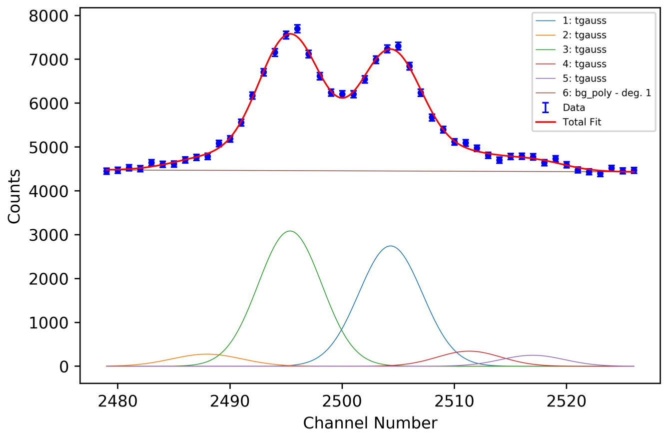
- The experimental data produced by FLUFFY is γ emissions as a function of time since irradiation start, time since capsule arrival at counting station, and as a function of the emitting product isotope:

$$\boldsymbol{N_{\gamma}(Z, A, I, t_0, t_1, n_{cycles})}$$



γ Efficiency

+

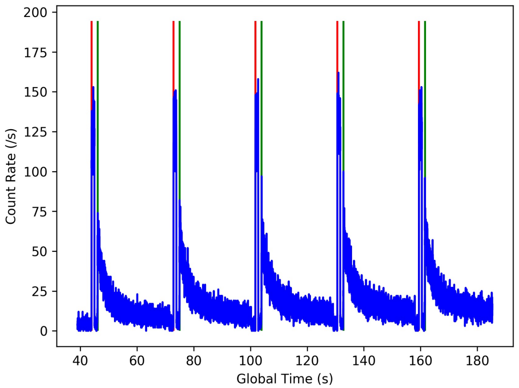


γ Counts

The FIER Model

- The Fission Induced Electromagnetic Response (FIER) code offers a model that produces analogous FPY γ emission data.

Irradiation Scheme



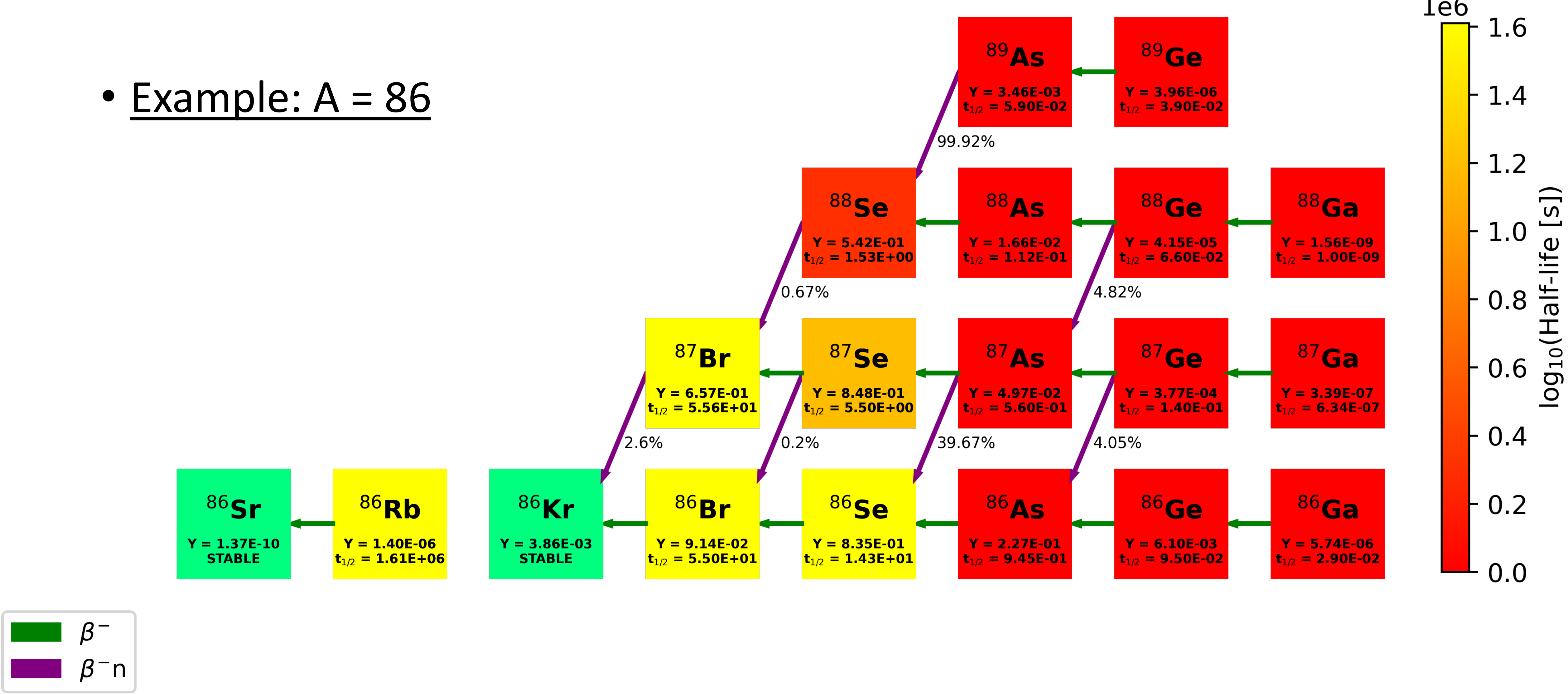
→ FIER → $\boldsymbol{N_\gamma(Z, A, I, t_0, t_1, n_{cycles})}$

- Chi-squared minimization between FIER and experimental data is used to determine fission yields and correct decay data:

$$\chi^2 = \frac{\left[N_\gamma(Z, A, I, t_0, t_1, n_{cycles}) - FIER(Z, A, I, t_0, t_1, n_{cycles})\right]^2}{\sigma_{N_\gamma}^2}$$

Mass Chain Analysis

- <u>Example: A = 86</u>



Mass Chain Analysis

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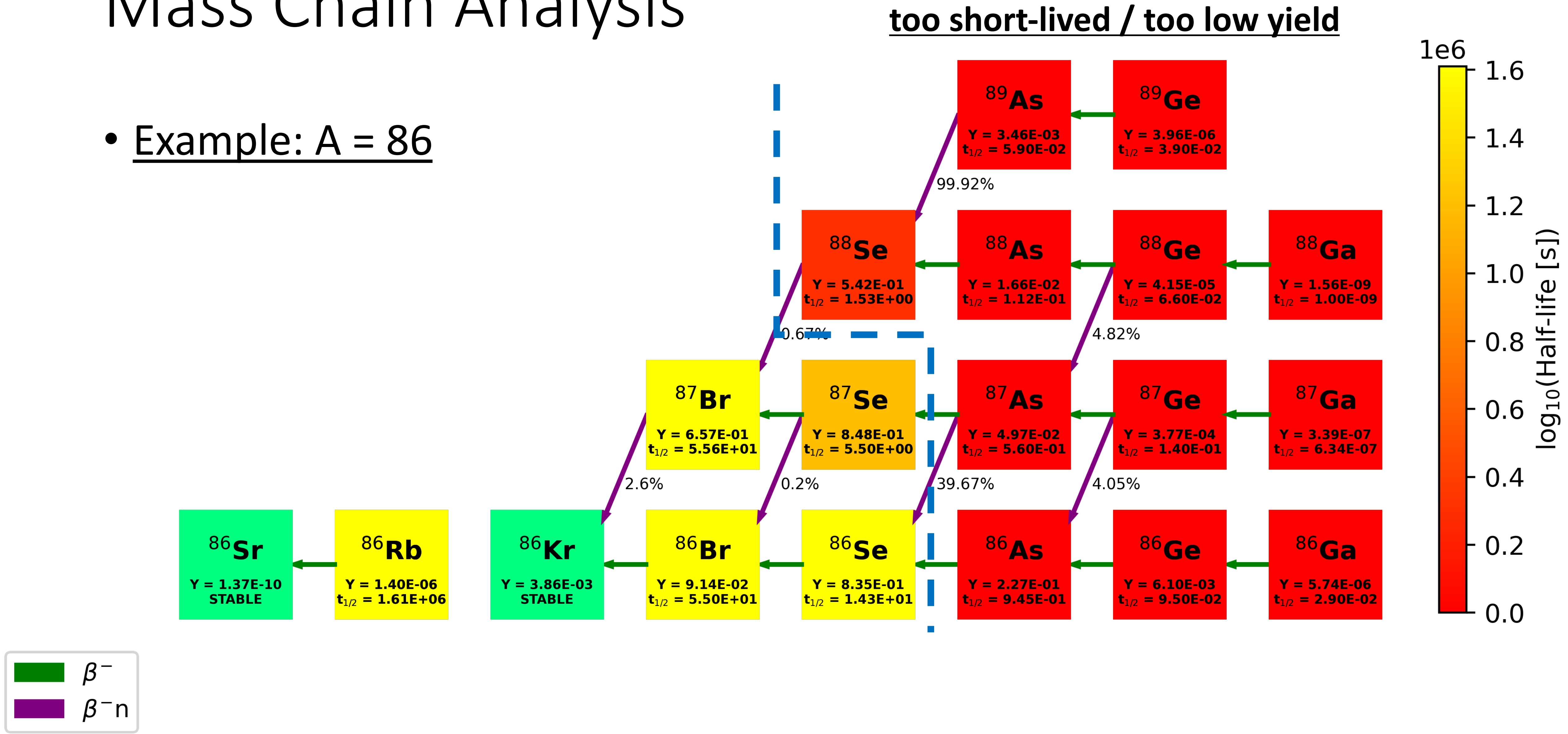

Mass Chain Analysis

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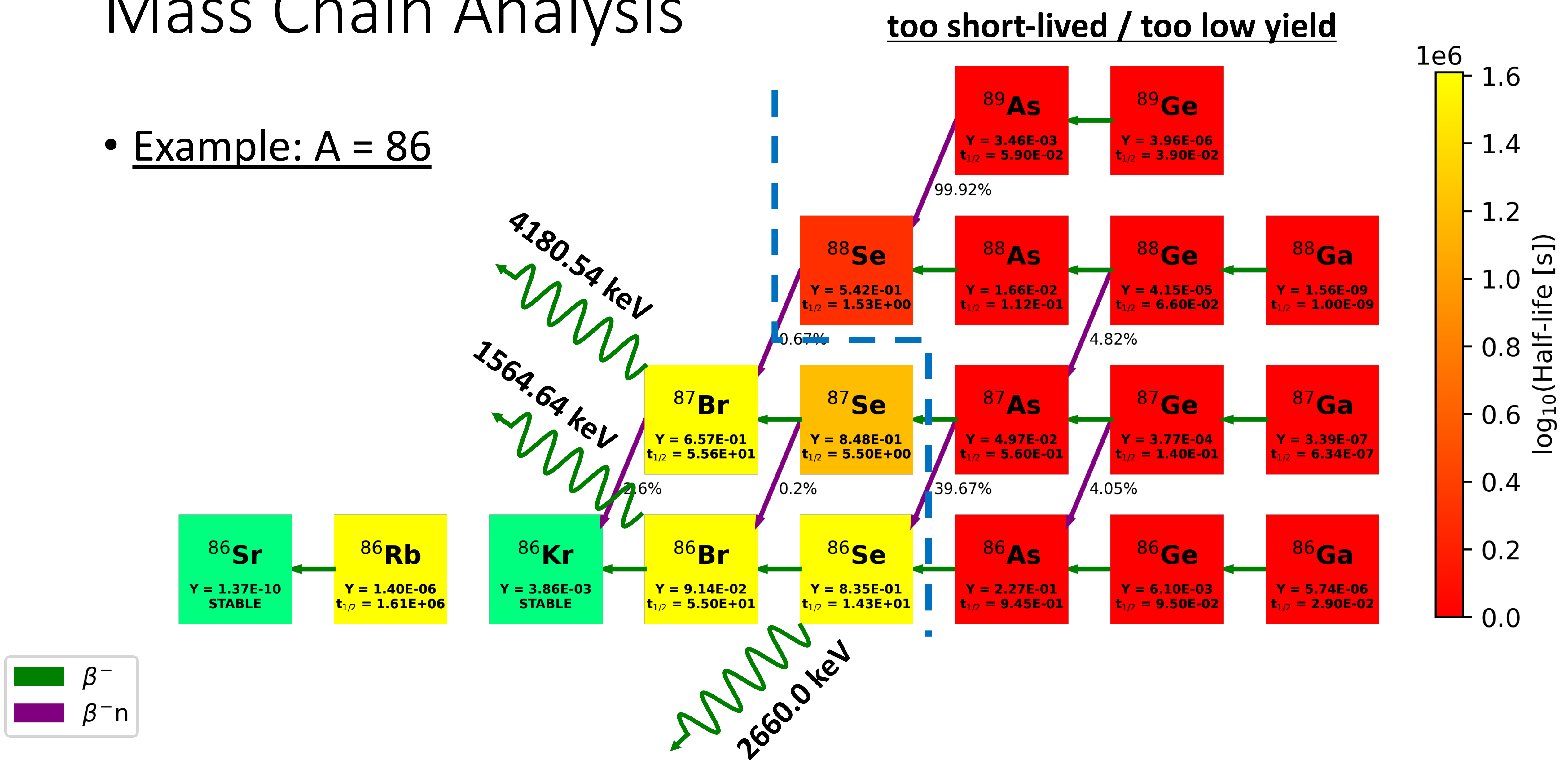
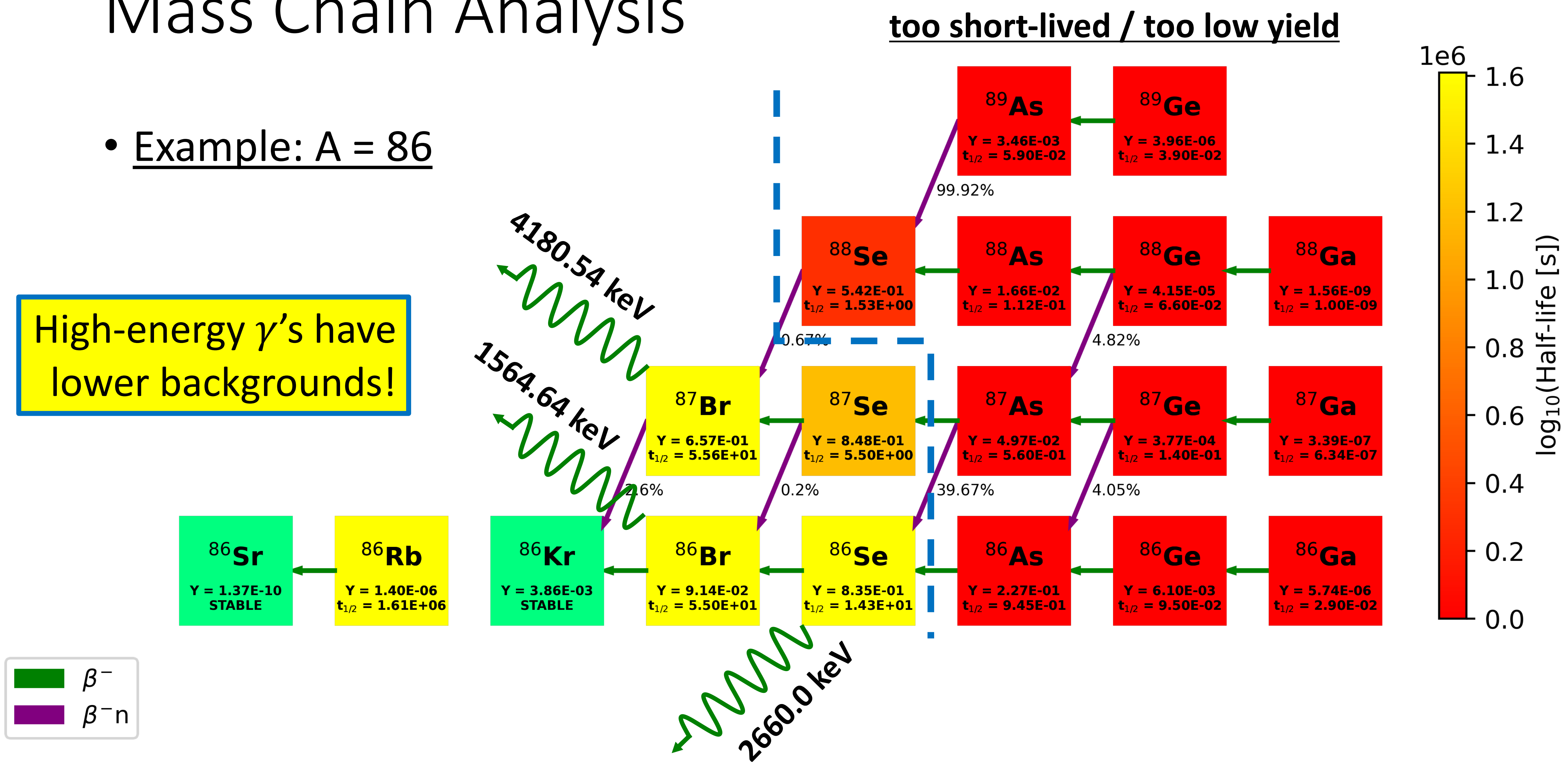


Mass Chain Analysis

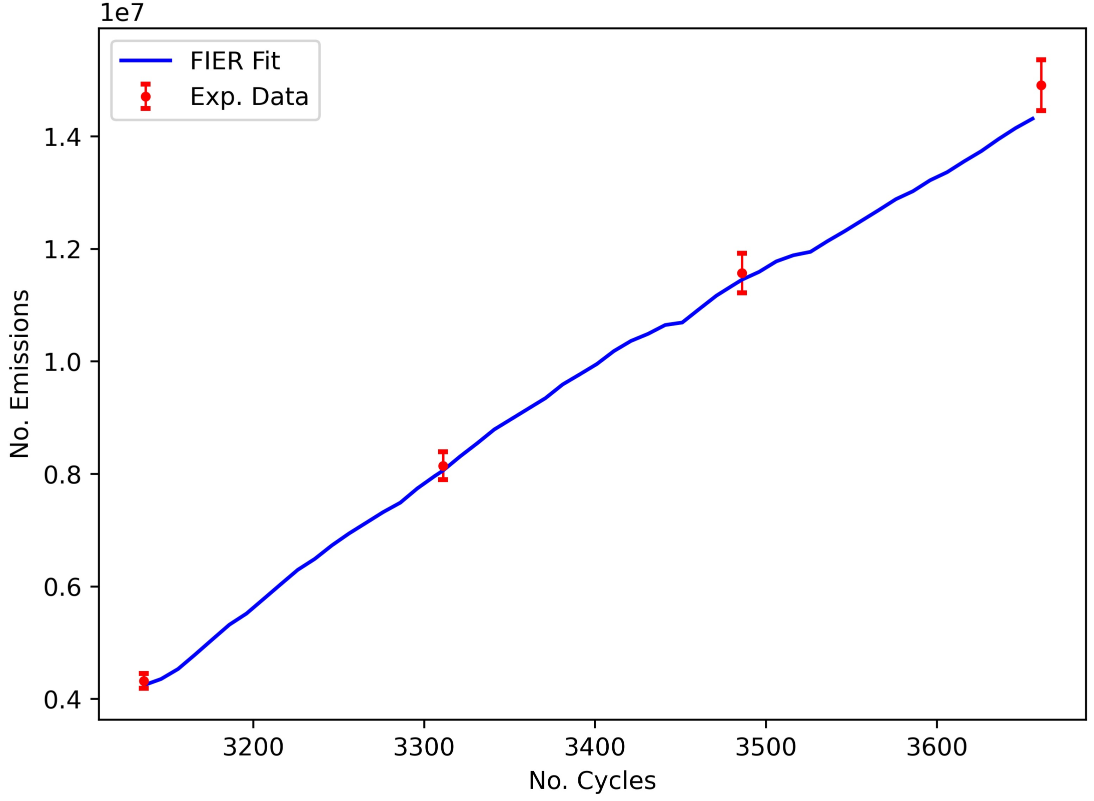
- Example: A = 86

High-energy γ's have lower backgrounds!

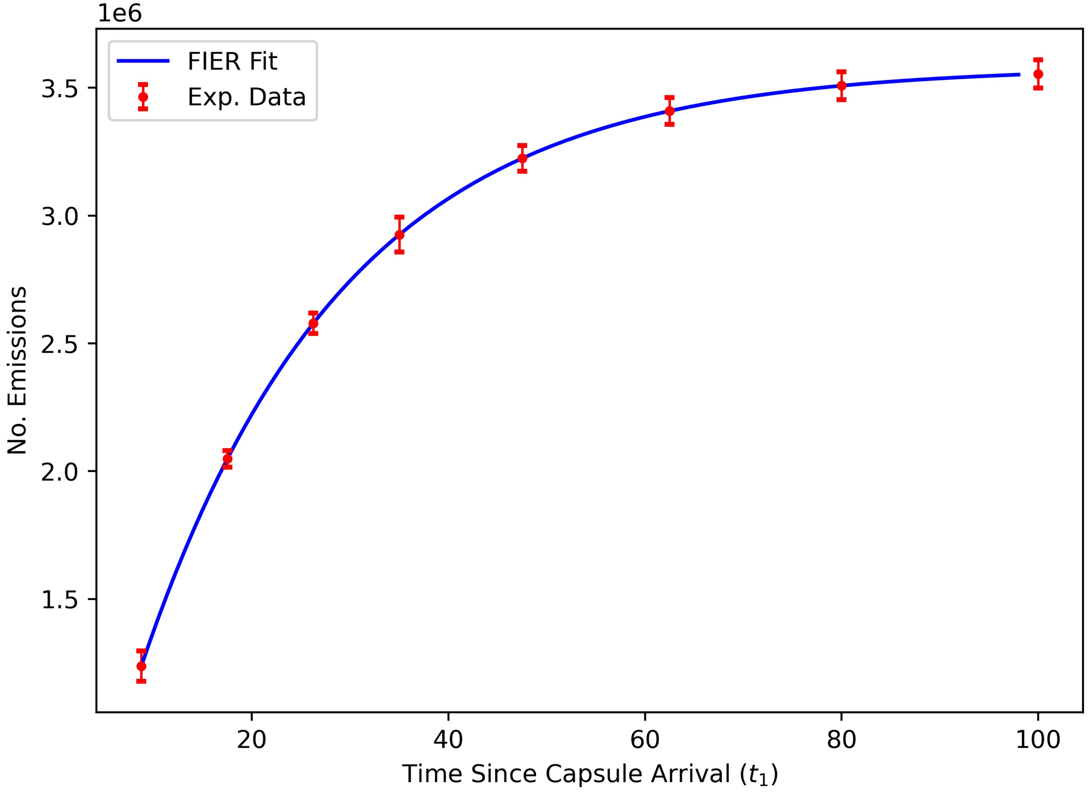


Results

- An example of results from the A = 86 mass chain



Emissions of 1564.0 keV γ's from ^{86}Br as a function of time since capsule arrival fit with FIER. ^{238}U(n,f) data.



Emissions of 2660.0 keV γ's from ^{86}Se as a function of time since capsule arrival fit with FIER. ^{238}U(n,f) data.

Results

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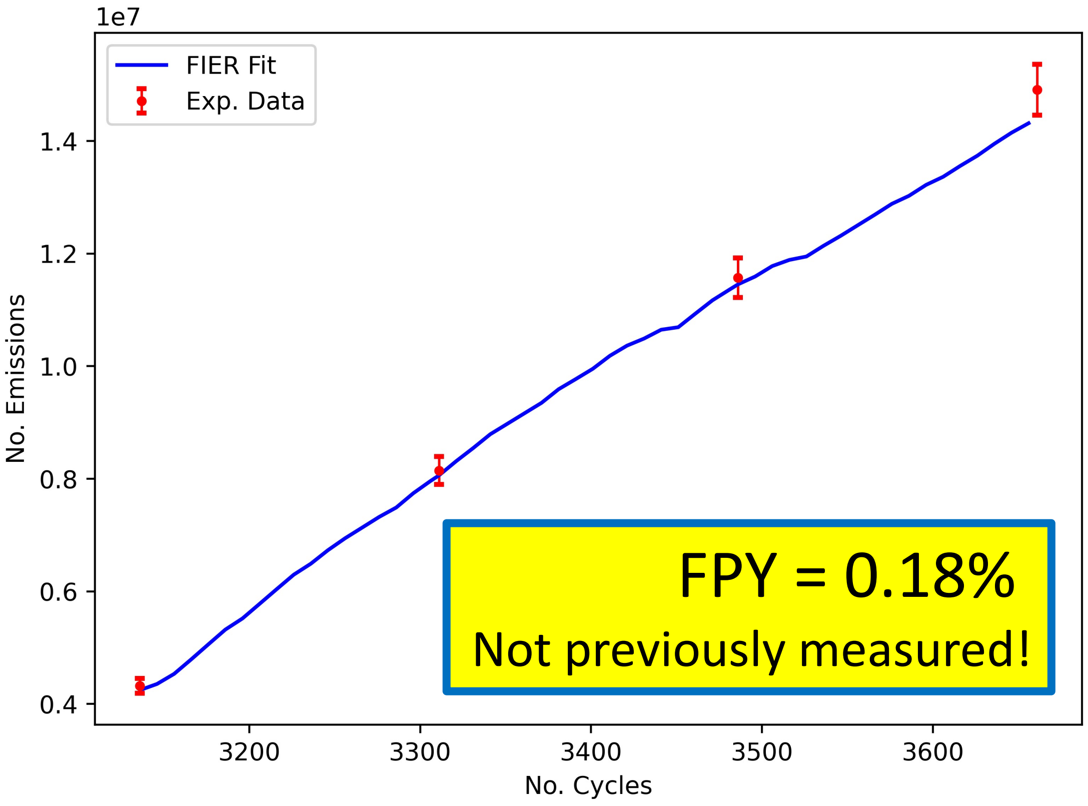

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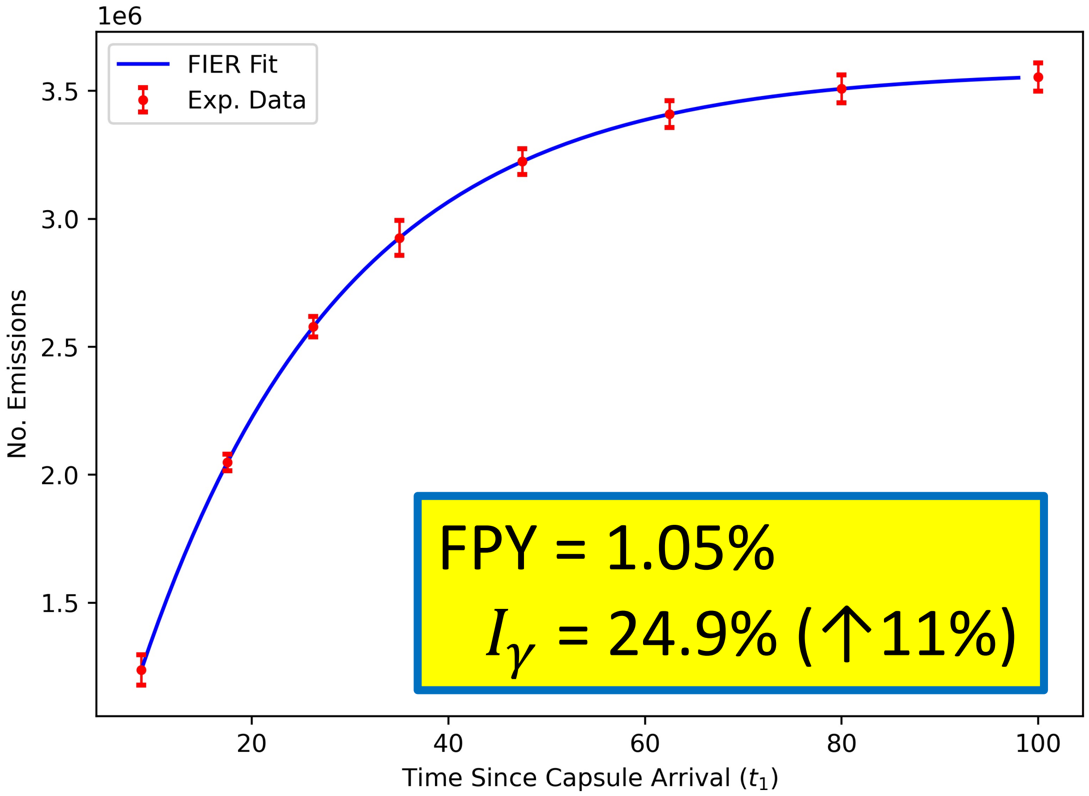
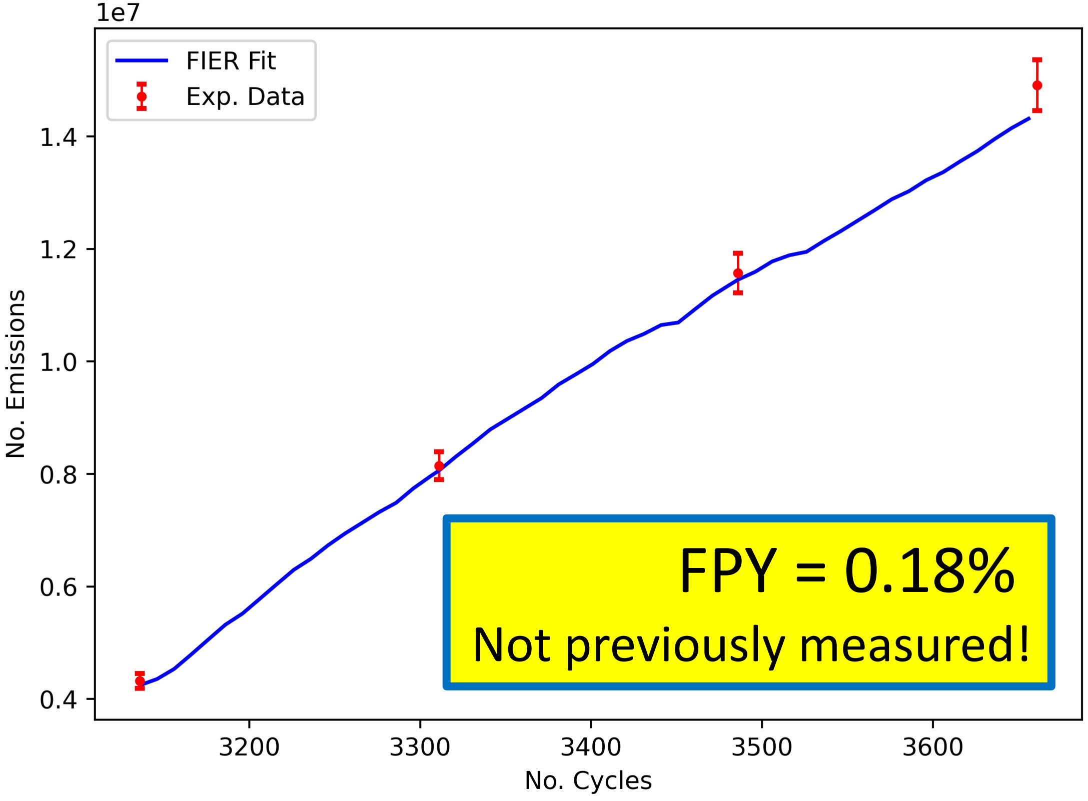


Emissions of 2660.0 keV γ's from ^{86}Se as a function of time since capsule arrival fit with FIER. ^{238}U(n,f) data.

Results

γ emission rates from the daughter FP simultaneously constrain the FPY and I_γ of the parent.

- An example of results from the A = 86 mass chain



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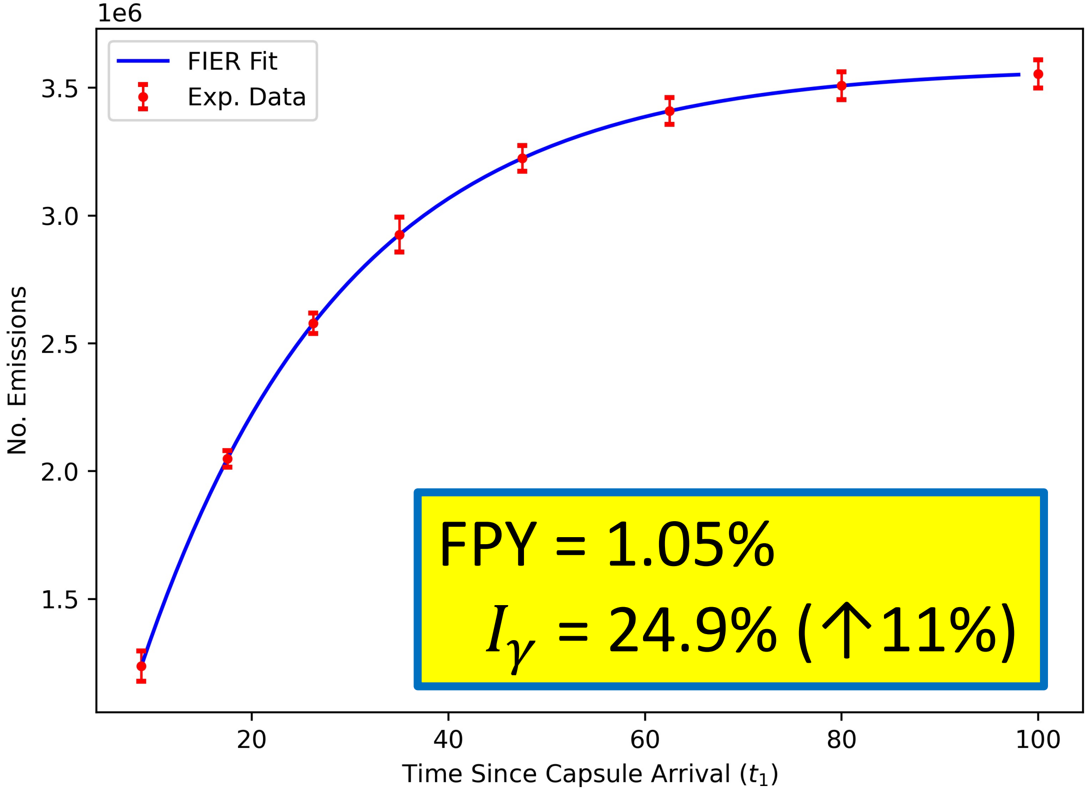

Emissions of 2660.0 keV γ's from ^{86}Se as a function of time since capsule arrival fit with FIER. ^{238}U(n,f) data.

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Fission Yield Correlation/Covariance Matrices

Motivation

- Neither the ENDF/B-VIII.0 or JEFF-3.1 fission yield evaluations include information on covariances between fission yields. [1,2]
- Covariances between fission yields affect several important applications:
  - Forensics and safeguards calculations
  - Reactor antineutrino rates
  - Reactor inventory, decay heat, and poisoning

[1] – Evaluation and Compilation of Fission Product Yields – T.R. England and B.F. Rider (1994)   [2] – R. W. Mills, Fission product yield evaluation, Ph.D. thesis, University of Birmingham (1995)

Previous Work

- Pigni et al. – 2013
  - Variance estimation with Wahl systematics
- Schmidt – 2013
  - Parameter perturbation in the GEF code
- Leray et al. – 2017
  - Parameter perturbation in the GEF code
- Kawano and Chadwick – 2013
  - Bayesian method for ^{239}Pu FPY

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- Work by Pigni, Schmidt, and Leray relies on an underlying model of fission and parameter uncertainties.
- Results of these work are not readily accessible due in part to ENDF format limitations.

Motivation

- The goal of this work is to generate a set of covariance matrices for the fissioning systems of a given fission yield evaluation **with as little fission model bias/uncertainty as possible.**
- This method seeks to use simple conservation rules in order to constrain a sample space for Monte-Carlo estimation.
- The resulting covariance matrix will predominantly reflect the evaluated uncertainties in the independent fission yields.
- **Public availability of the covariance matrices is a high priority.**

Conserved Relationships

- In order to obtain correlation, conserved quantities can be enforced upon a set of resampled fission yields [1]:

Total Yield:

$$\sum_i Y_i = 2$$

[1] - Generation of Fission Yield Covariances to Correct Discrepancies in the JEFF Fission Yield Library – L. Fiorito et al. (2015) - https://www.oecd-nea.org/science/wpec/sg37/Meetings/2015_May/SG37_8_LF.pdf

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Charge Parity:

$$\sum_i Y_i(Z_1, A_i) = \sum_i Y_i(Z_{CN} - Z_1, A_i)$$

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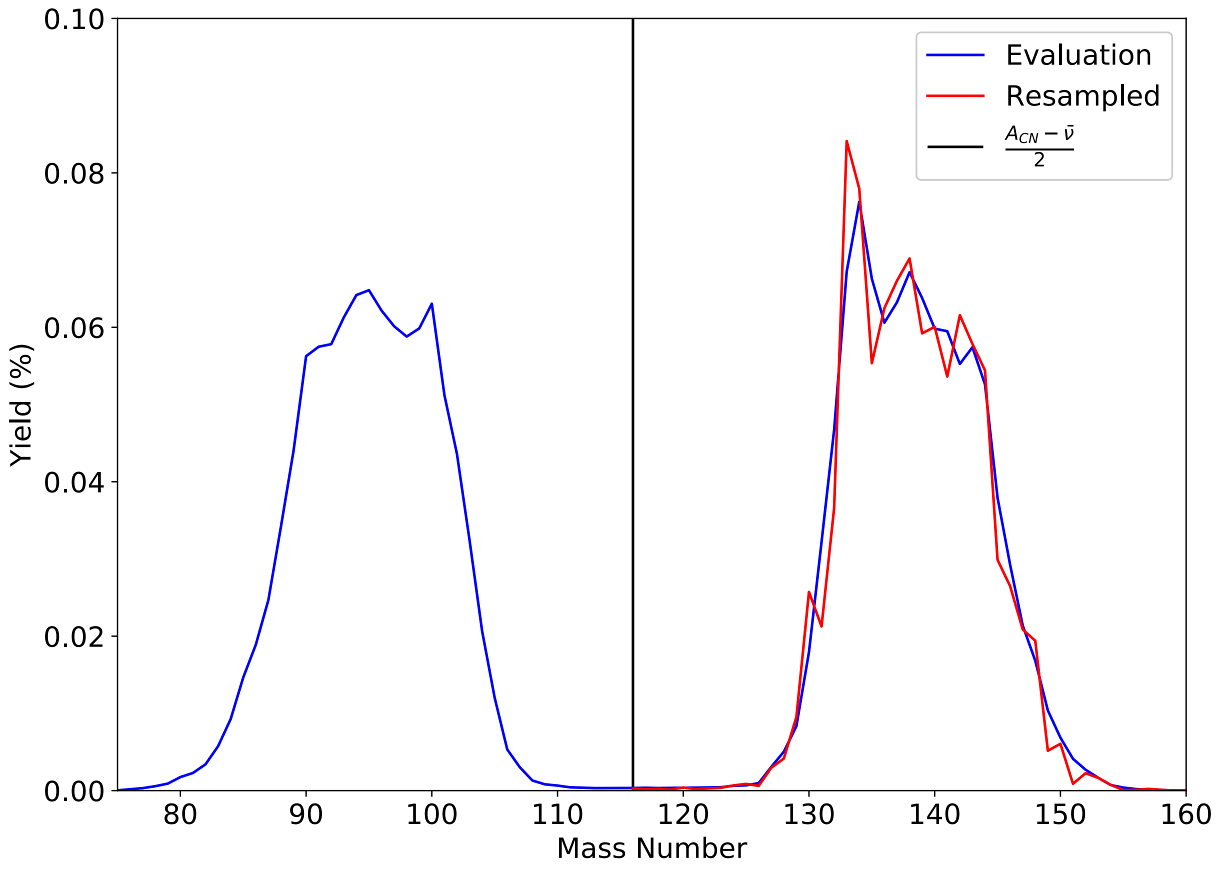
Mass Symmetry:

$$\sum_{A_i > \frac{A_{CN} - \bar{\nu}}{2}} Y_i(A_i) = 1$$

[1] - Generation of Fission Yield Covariances to Correct Discrepancies in the JEFF Fission Yield Library – L. Fiorito et al. (2015) - https://www.oecd-nea.org/science/wpec/sg37/Meetings/2015_May/SG37_8_LF.pdf

FY Covariance Matrix Generation

- The way in which a set of fission yields are resampled can be structured to conserve these relationships:
- **1)** Randomly selected the “light” or “heavy” side of the fission product spectrum to resample.
- **2)** Randomly select (weighted by uncertainty) a product in each A chain, resample its yield about its evaluated uncertainty.
- **3)** Scale all other yields in that A chain by the same percent change.



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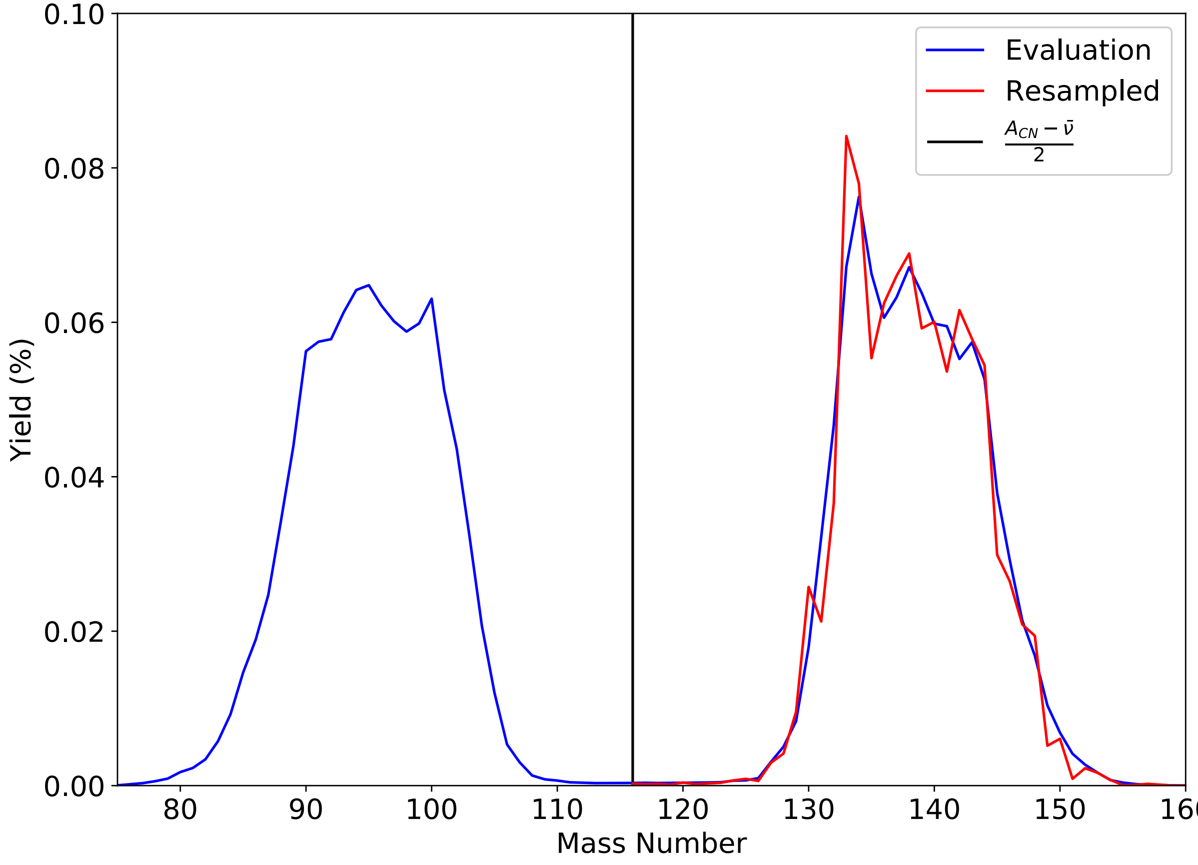


Step 3 is allowed if the Z distribution for a given A is Gaussian, which empirical data and the ENDF/B-VII.0 evaluation supports [1].

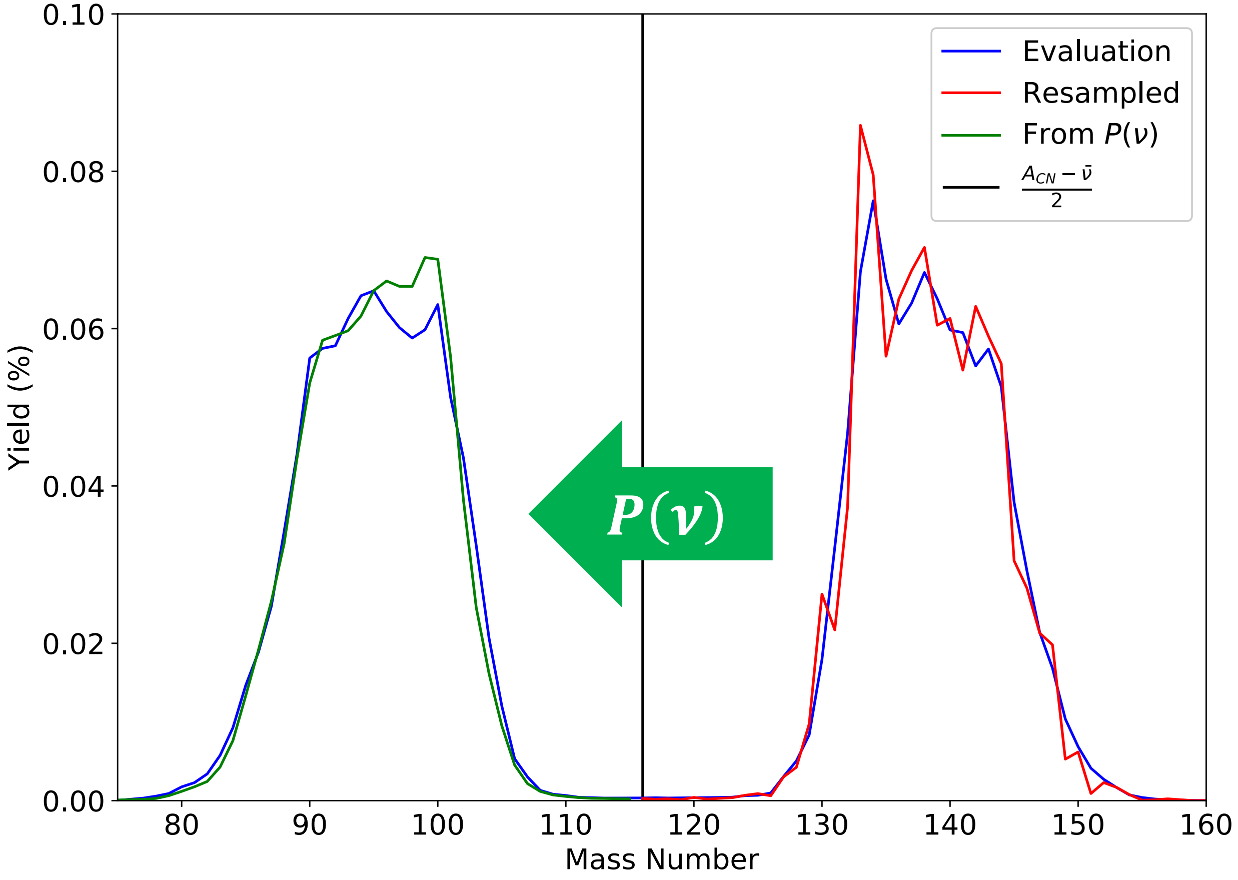
[1] – Evaluation and Compilation of Fission Product Yields – T.R. England and B.F. Rider (1994)

FY Covariance Matrix Generation

- **4)** Normalize the resampled yields such that they sum to 1.
- **5)** Generate the fission yields on the complementary side of the fission product spectrum using the neutron multiplicity of the compound system.

$$Y_{frac}(Z_{CN} - Z, A_{CN} - A - \nu\ ) = P(\nu)\, Y(Z, A)$$

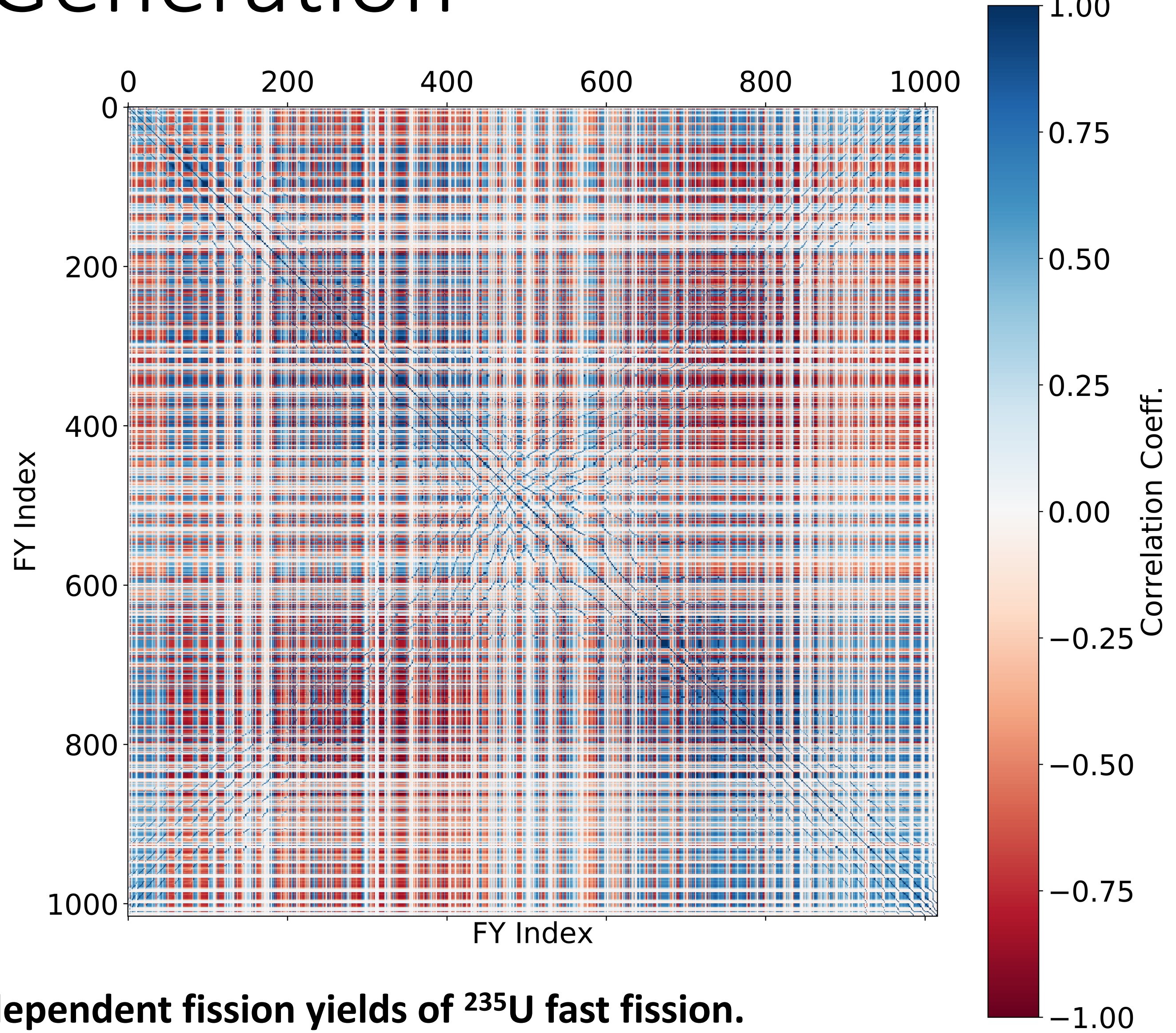
$$Y(Z_{CN} - Z, A_i) = \sum_{\nu} Y_{frac}(Z_{CN} - Z, A_i)$$



By Step 5 we've ensured all of the conservation rules are met.

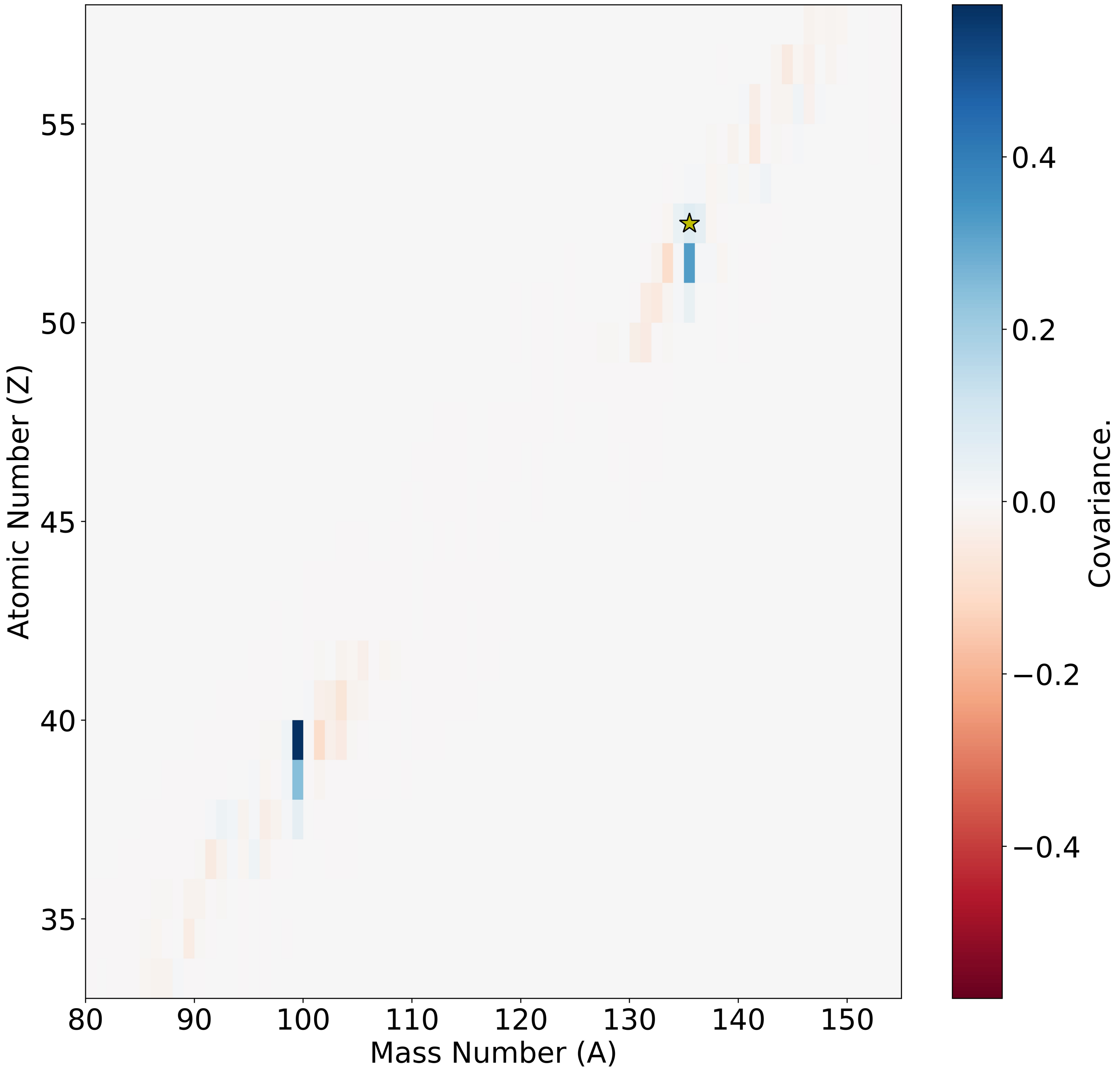
FY Covariance Matrix Generation

- **6)** Repeat steps 1-5) N times. Select N such that statistical noise is minimized.
- **7)** Calculate the resulting correlation matrix from the N trials.

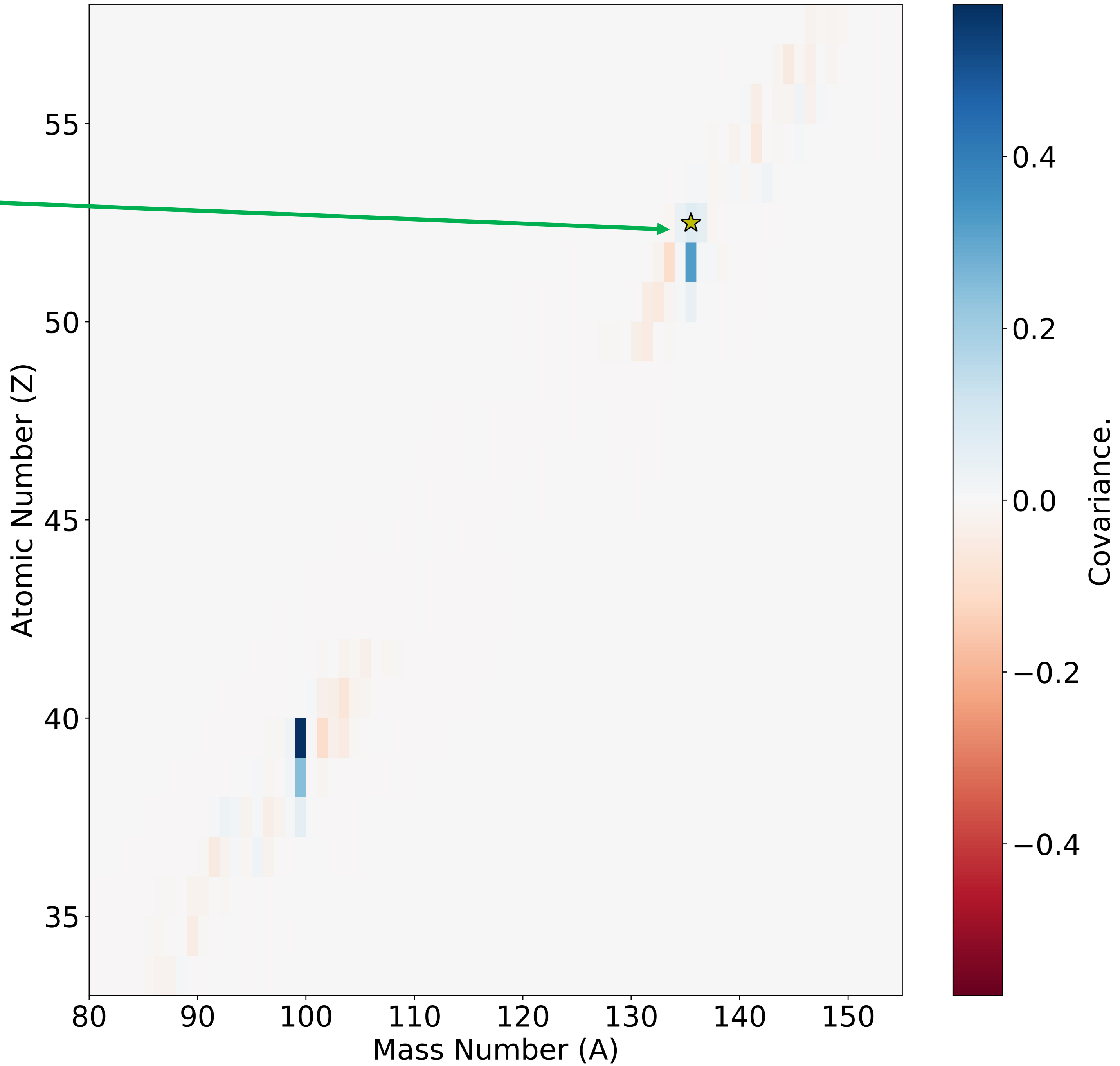


Correlation matrix for independent fission yields of ^{235}U fast fission.

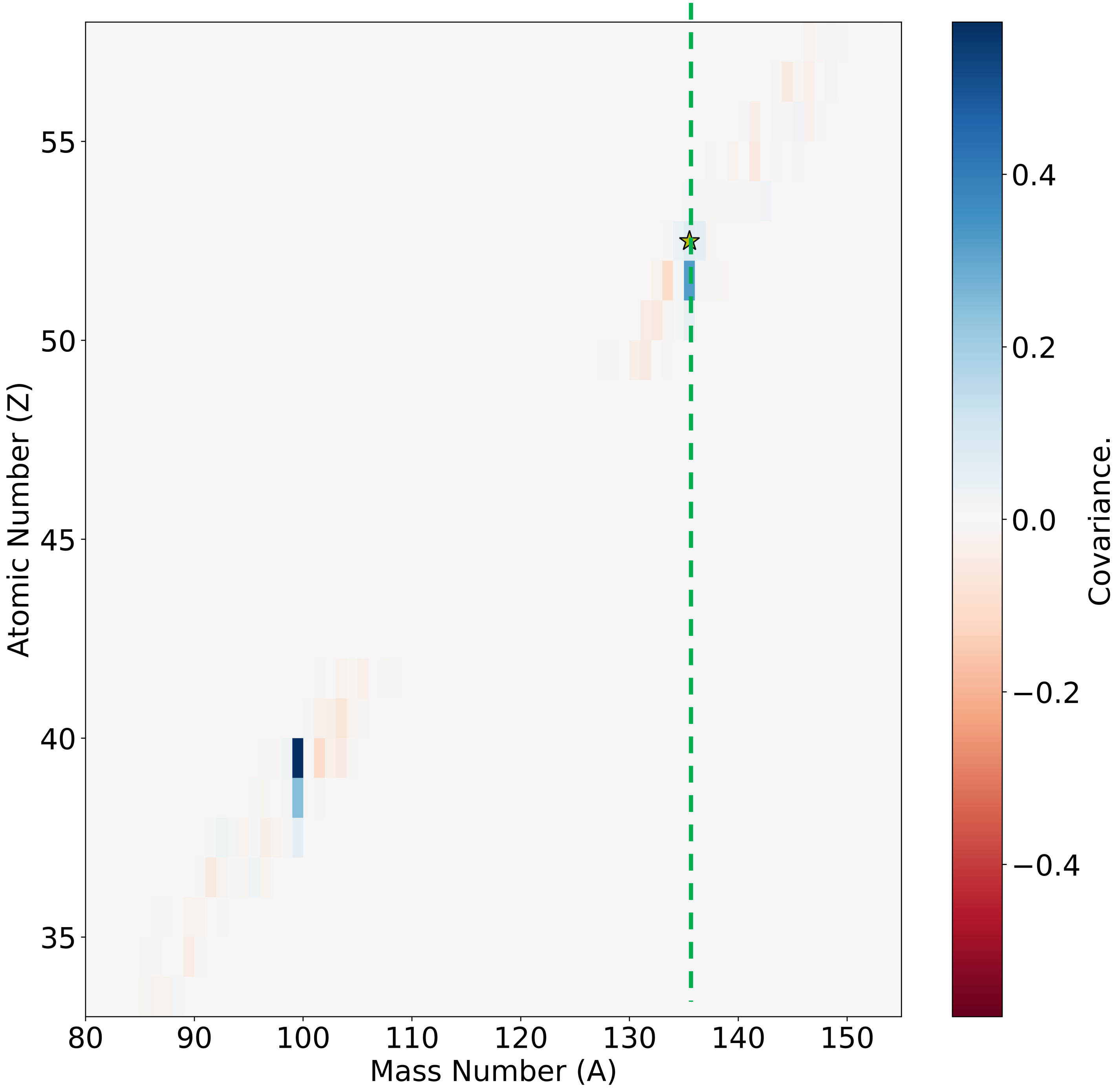
- **Example:** ^{135}Te
- Presented is the covariance between independent yields as function of Z and A and that of ^{135}Te.
- The evaluated yield for ^{135}Te is $2.47 \pm 0.57\%$



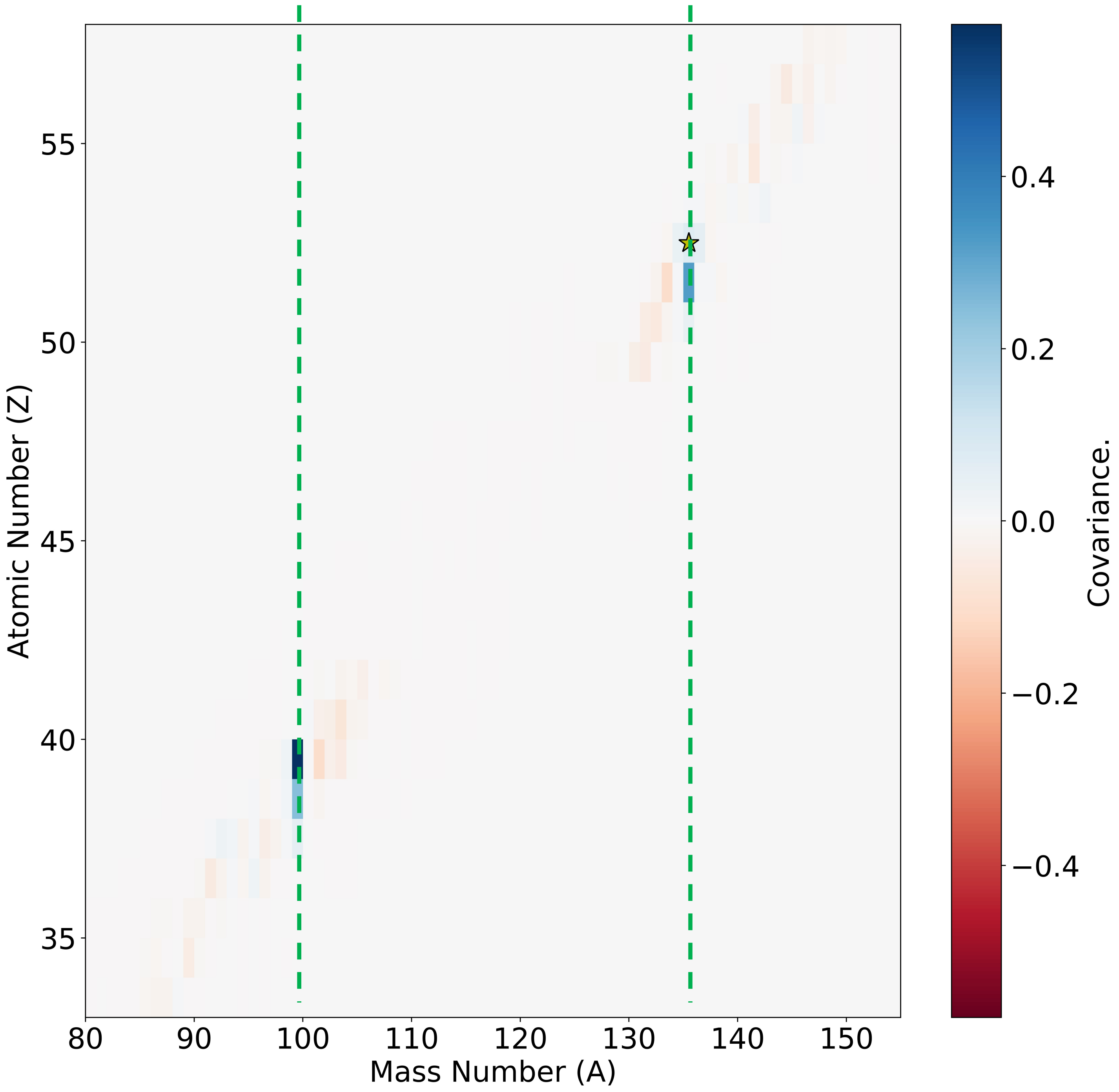
- **Features:**
- ^{135}Te is positively correlated with itself.



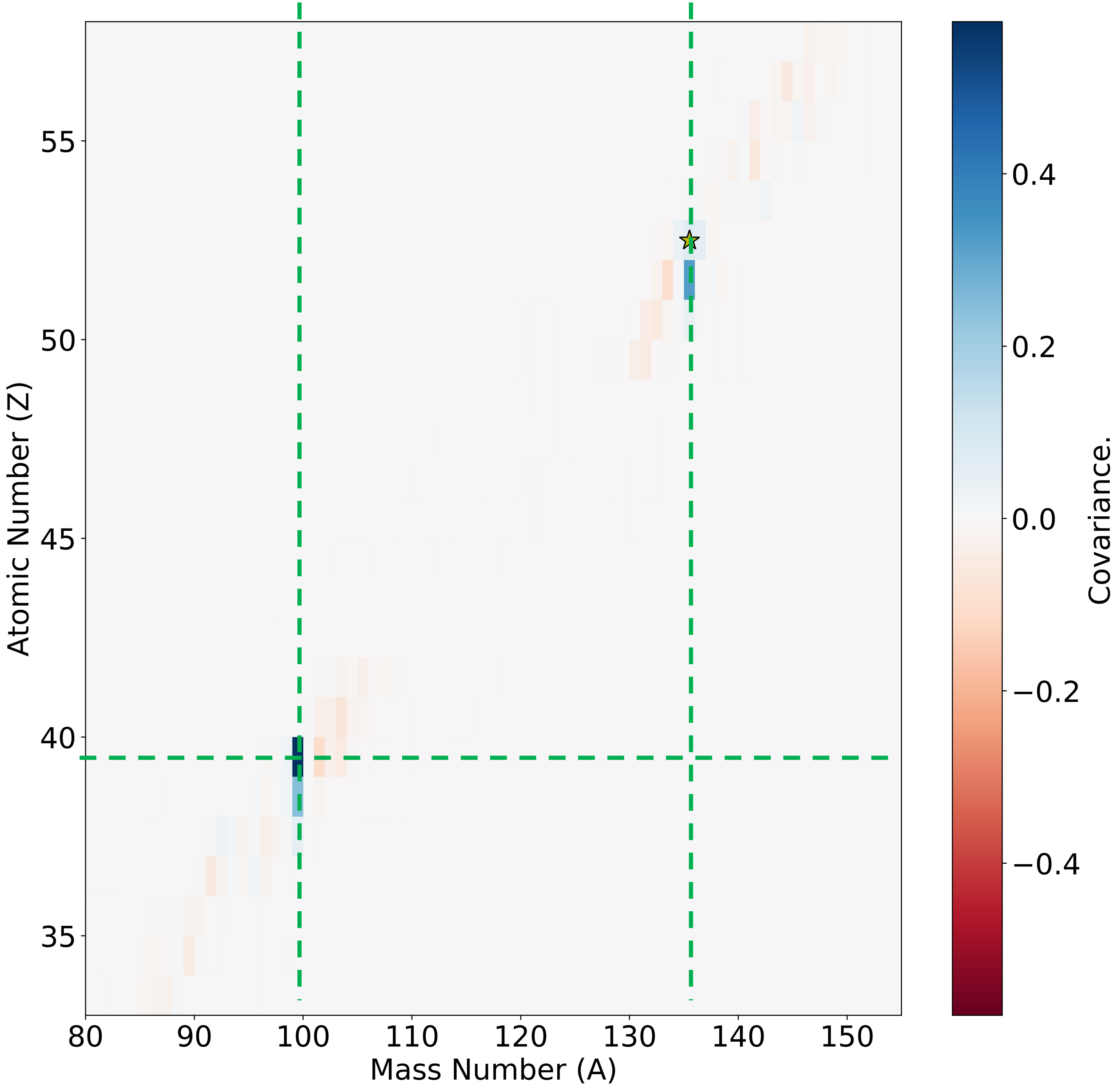
- **Features:**
- ^{135}Te is positively correlated with itself.
- Products along the A chain have positive correlation.



- **Features:**
- ^{135}Te is positively correlated with itself.
- Products along the A chain have positive correlation.
    - This positive correlation is reflected along a complementary $A = 99$ chain.



- **Features:**
- ^{135}Te is positively correlated with itself.
- Products along the A chain have positive correlation.
  - This positive correlation is reflected along a complementary $A = 99$ chain.
- Products along A chains that do not have complementary Z have negative correlation.



Conclusions

- A model-agnostic method for independent fission yield covariance matrix generation is being developed.
- **This method has been successfully applied to all compound systems in the ENDF/B-VIII.0 and JEFF-3.1 evaluations.**
- The results demonstrate expected behavior and trends.
- Final results serve as an interim solution for independent fission yield covariance matrices until a new evaluation is completed.
  - The results are publicly available at nucleardata.berkeley.edu/FYCOM
  - **Publication accepted to Atomic Data and Nuclear Data Tables on April 20, 2021.**

Template of Expected Experimental Uncertainties in Fission Yield Measurements

The Templates Project

- Dr. Denise Neudecker of Los Alamos National Laboratory is a leading an effort to assemble “templates” of expected experimental uncertainties in various nuclear properties measurements.
- Each “template” is a peer-reviewed guide to common experimental techniques for a given nuclear property. Tables enumerate expected averages/bounds for standard sources of uncertainty.
- I have written the “Fission Yields” chapter of this manuscript.
    - This covers neutron-induced fission product yields only.

The Templates Project

- The templates that will be published seek to address two primary goals:

- **<u>Guide for Experimentalists:</u>** the templates will help experimentalists plan their experiments and data analysis to be a thorough and precise as possible.

- **<u>Guide for Evaluators:</u>** the templates will help evaluators assess the quality of different experiments and filling in missing uncertainty data in legacy measurements.

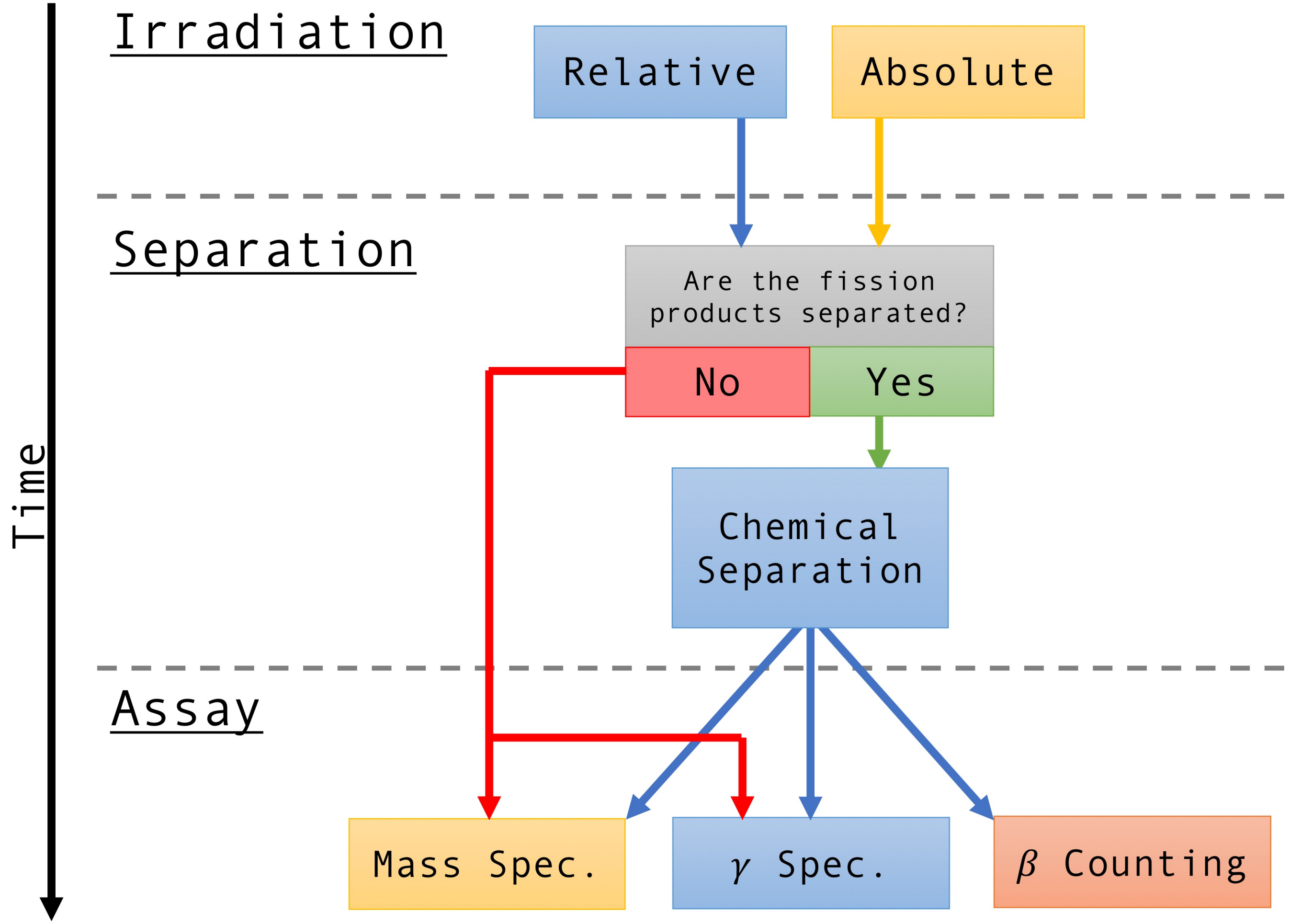
My Contribution: Fission Yields Template

- Three sources of data were used to produce the fission yields template:
  - 40+ peer-reviewed publications
  - Consultation with experts
    - Fredrik Tovesson
    - Dana Duke, Kristina Montoya
    - Bruce Pierson
  - 18214 EXFOR entries spanning 1943-2019
    - Semi-automated Python script for processing these entries
- Longest part of the template paper
- ~6 month effort

Activation-type Experiments

- Three major steps:
  - Irradiation
  - Separation (optional)
  - Assay

- Each step has its own template.



Activation-type Experiments

- Here's an example of the tables produced for the template:

- Expected minimum values for uncertainties associated with the irradiation of an actinide target (i.e., determination of number of fissions).

Symbol	Correlation
Relative and Absolute:	
$P(E_{inc})$	–
Absolute Only:	
N_f	Fully Correlated
ϕ	Fully Correlated
σ_f	Fully Correlated
N	Fully Correlated
	(strongly if different targets)
t_i	Fully Correlated
B_u	Strongly Correlated
c_T	Strongly Correlated
F_C	Fully Correlated
$\oslash$	–
Ω	Uncorrelated
c	Uncorrelated

Symbol	Minimum σ
Relative and Absolute:	
$\delta P(E_{inc})$	
nTOF	
-ΔL	1 mm
-Δt	1 ns
-δc	Poisson dist.
Foil Act.	
-δA_{mon}	Poisson dist.
-δc	Poisson dist.
-$\delta \sigma_{mon}$	given by evaluation
Absolute Only:	
δN_f	1
-$\delta\phi$	1
-$\delta\sigma_f$	given by evaluation
-δN	0.001
- δw	abundance: See databases
	or given by enrichment
- δt_i	~0
-δB_u	1
-δc_T	1.5
-δF_C	1.5
$\delta\oslash$	<0.1
δN	0.001
- δw	enrichment: given by manufacturer
	abundance: See databases
$\delta\Omega$	<0.1
δc	Poisson dist.

Pipeline to Publication

- Broad community input on the FY template was gathered from March 13 to April 16, 2021.
- Final sets of feedback from 11 individuals were received.
- The entire template manuscript was submitted to *Nuclear Data Sheets* in June 2021.

Templates of Expected Measurement Uncertainties

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Thank You!

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