Addressing Nuclear Data Needs with FIER

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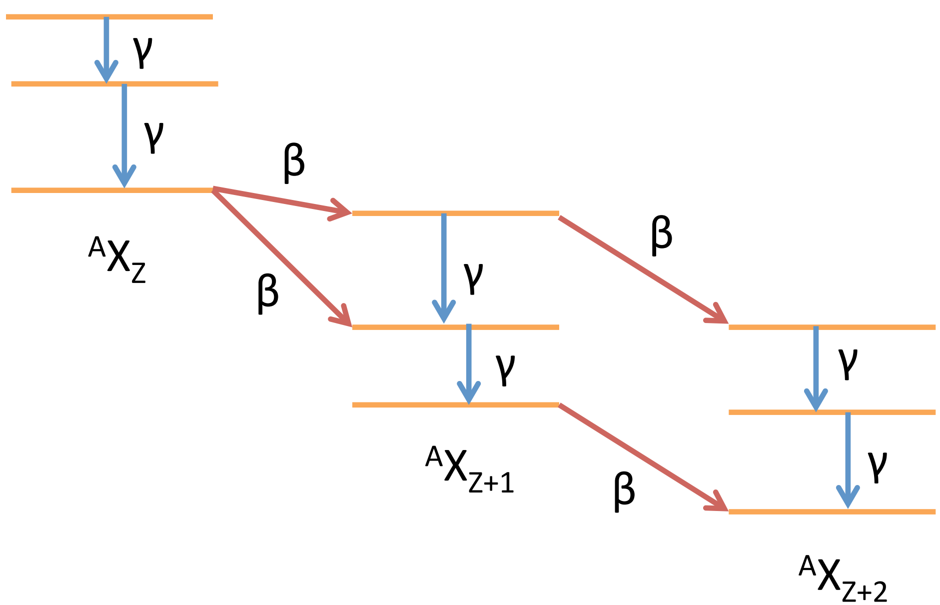
What is FIER?

- FIER (Fission Induced Electromagnetic Response) is an analytical tool that calculates delayed gamma-ray spectra that result from any arbitrary irradiation scheme.



$$\frac{\partial N_1(t)}{\partial t} = Y_1 R - \lambda_1 N_1(t)$$

$$\frac{\partial N_2(t)}{\partial t} = \beta_{1,2}\lambda_1 N_1(t) - \lambda_2 N_2(t)$$

$$\frac{\partial N_3(t)}{\partial t} = \beta_{2,3}\lambda_2 N_2(t) - \lambda_3 N_3(t)$$

$$\vdots$$

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FIER Inputs

- FIER uses evaluated nuclear data to perform its calculations:
  - Independent fission yields Y_i
  - Decay modes and branching ratios $\beta_{i-1,i}$
  - Half-lives/decay constants λ_i
  - Gamma energies and intensities $I_i(E_\gamma)$

- Currently fission yields are taken from England and Rider [1] and decay data are taken from the ENDF Decay Sub-Library [2]

[1] T. England, B. Rider, Evaluation and compilation of fission product yields, Tech. Rep. LA-UR-94-3016, Los Alamos National Lab, Oct. 1994
[2] D. Brown, Release of the ENDF/B-VII.1 Evaluated Nuclear Data File, Tech. Rep. BNL-98708-2012-IR, Brookhaven National Lab, Nov. 2012

FIER Workflow

- FIER uses the listed decay modes of each nuclear species to produce decay chains
- From each decay chain, decay stems are extracted

decay chains

$$
\begin{tikzcd}[column sep=small]
(x,f) \arrow[r] & \dots \arrow[r] \arrow[d] & {}^{134}Sn \arrow[r] \arrow[d] & {}^{134}Te \arrow[r] & {}^{134m}I \arrow[r] & {}^{134}I \arrow[r] & \dots \\
(x,f) \arrow[r] & \dots \arrow[r] & {}^{133}Sb \arrow[r] & {}^{133}Te \arrow[r] & {}^{133m}Te \arrow[r] & {}^{133}I \arrow[r] & \dots
\end{tikzcd}
$$

decay stems for ^{133}Te

$$(x,f) \rightarrow \dots \rightarrow {}^{134}Sn \rightarrow {}^{133}Sb \rightarrow {}^{133}Te$$

$$(x,f) \rightarrow \dots \rightarrow {}^{133}Sb \rightarrow {}^{133}Te$$

$$(x,f) \rightarrow \dots \rightarrow {}^{133}Sb \rightarrow {}^{133}Te$$

- For each decay stem, the population of its last species is calculated. The population of a species is the sum of its populations calculated from each decay stem.
- Decay stems are how the complex system of decay chains is linearized.

Production and Decay from Irradiation

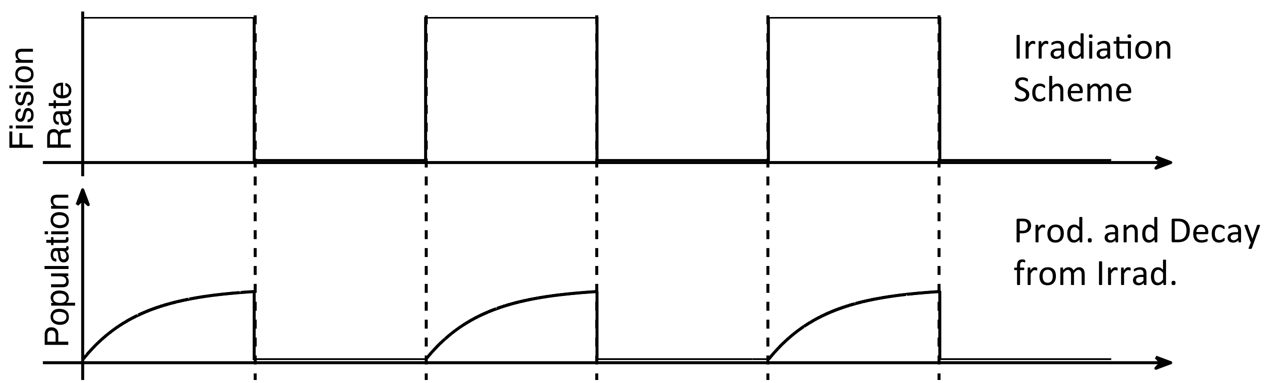
- The population calculation is the sum of two non-physical solutions to the production and decay series of differential equations.

$$\frac{\partial N_1(t)}{\partial t} = Y_1 R - \lambda_1 N_1(t)$$
$$\frac{\partial N_2(t)}{\partial t} = \beta_{1,2}\lambda_1 N_1(t) - \lambda_2 N_2(t)$$
$$\frac{\partial N_3(t)}{\partial t} = \beta_{2,3}\lambda_2 N_2(t) - \lambda_3 N_3(t)$$

$$\rightarrow \quad Y_1 R \neq 0, \quad N_1(0) = 0 \quad \rightarrow$$

$$\boxed{N_i(t) = \left[Y_1 R \prod_{l=1}^{i-1} \beta_{l-1,l}\lambda_l \right] \sum_{j=1}^{i} \frac{1 - e^{-\lambda_j t}}{\lambda_j \prod_{\substack{k=1 \\ k\neq j}}^{i} (\lambda_k - \lambda_j)}}$$

- FIER calculates the populations of the fission products at the end of each time step in the irradiation using the continuous production solution to the Bateman equations



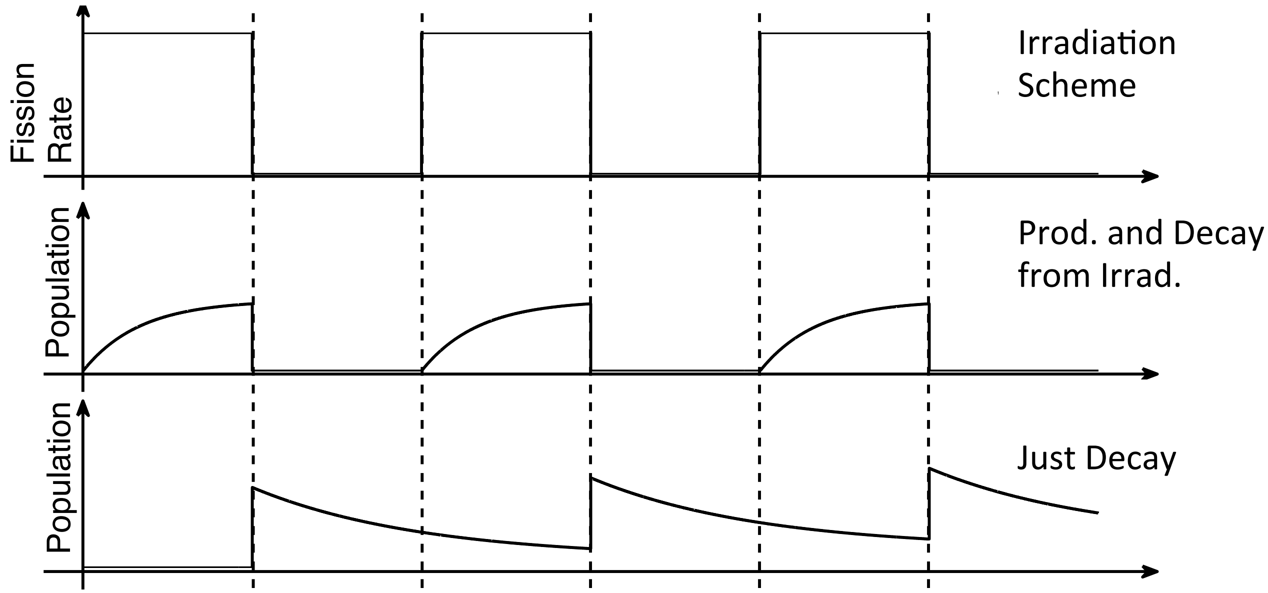
Just Decay

$$\frac{\partial N_1(t)}{\partial t} = Y_1 R - \lambda_1 N_1(t)$$
$$\frac{\partial N_2(t)}{\partial t} = \beta_{1,2}\lambda_1 N_1(t) - \lambda_2 N_2(t)$$
$$\frac{\partial N_3(t)}{\partial t} = \beta_{2,3}\lambda_2 N_2(t) - \lambda_3 N_3(t)$$

$$\rightarrow \quad Y_1 R = 0, \quad N_1(0) = N_1^0 \neq 0 \quad \rightarrow$$

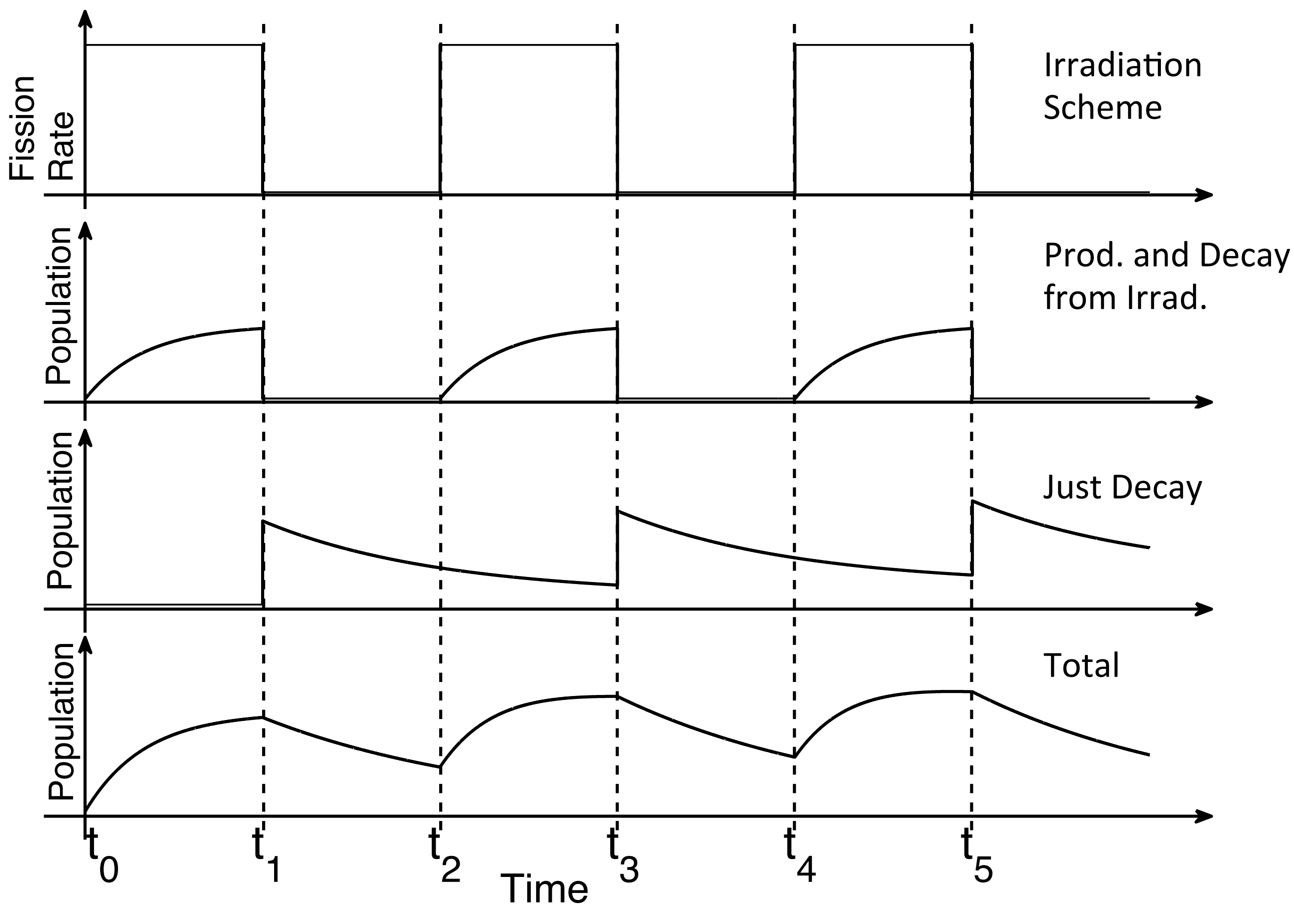
$$\boxed{N_i(t) = \left[N_1^0 \prod_{l=1}^{i-1} \beta_{l+1}\lambda_l \right] \sum_{j=1}^{i} \frac{e^{-\lambda_j t}}{\prod_{\substack{k=1 \\ k\neq j}}^{i} (\lambda_k - \lambda_j)}}$$

- The decay of the existing population at the beginning of each time step is calculated using the batch decay solution of the Bateman equations



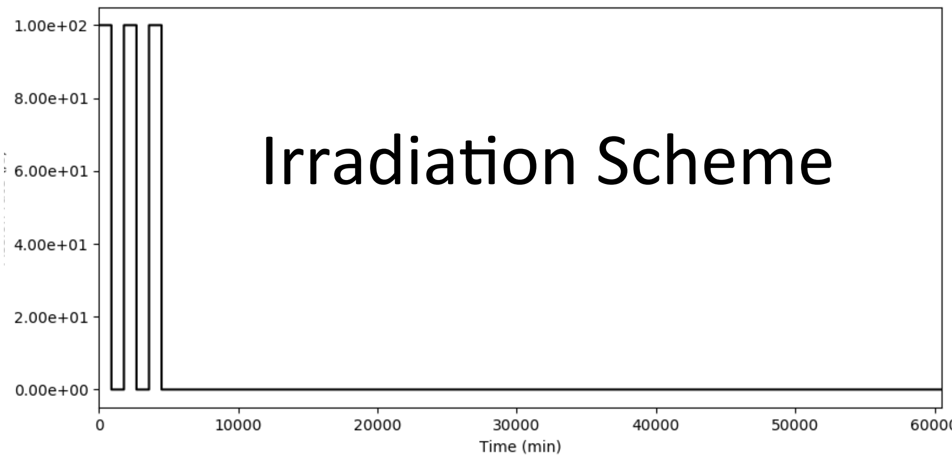
Total

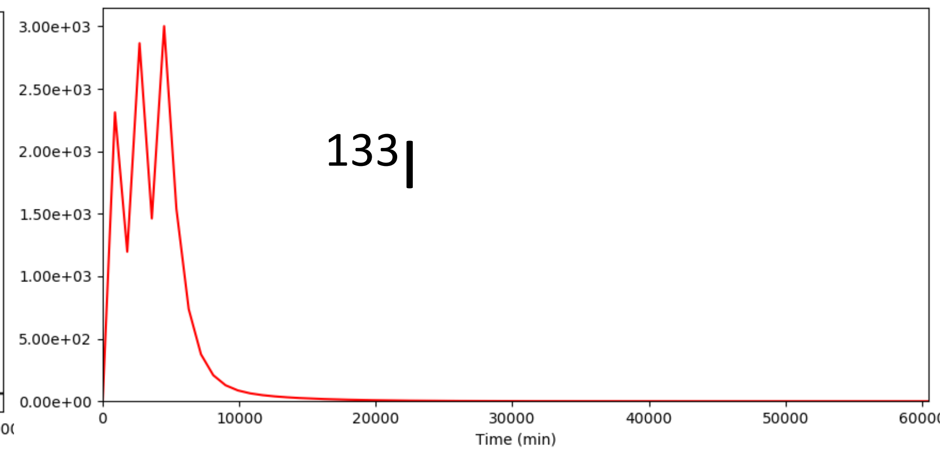
- The sum of the continuous production and batch decay solutions yield the total population of each species:

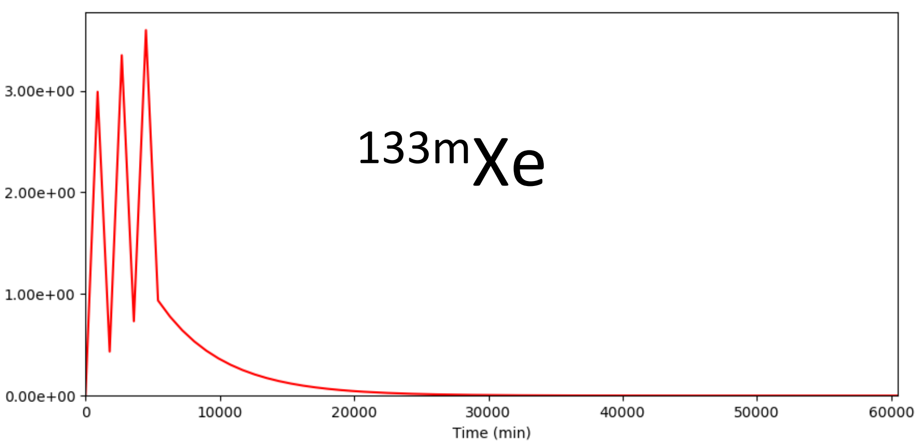


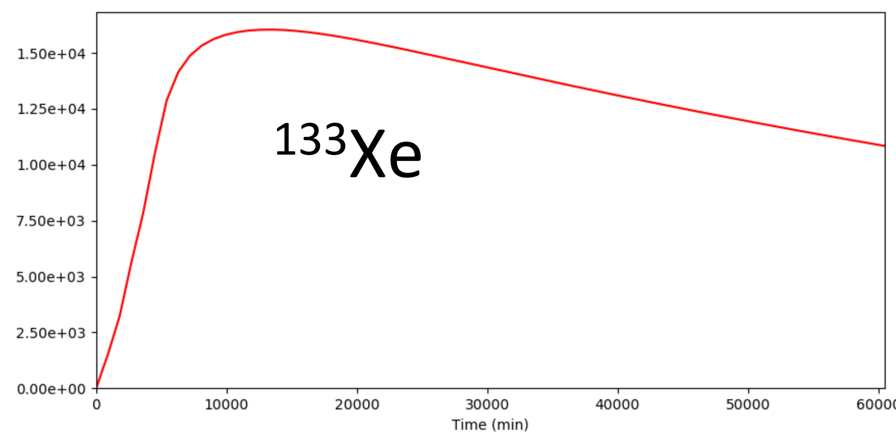
A More Complex Example

$$(x,f) \rightarrow \ldots \rightarrow {}^{133}I \rightarrow {}^{133m}Xe \rightarrow {}^{133}Xe$$









Spectra Calculation

- Once the populations at the end of the irradiation are calculated, the delayed gamma-ray spectra can be calculated
- The number of gamma emissions in a time window is the integral of the activity of its emitter:

$$C_i(E_m) = \int_{t_1}^{t_2} I_m \lambda_i N_i(t) dt$$

$$C_i(E_m) = \left[I_m \lambda_i N_1^0 \prod_{l=1}^{i-1} \beta_{l+1} \lambda_l \right] \sum_{j=1}^{i} \frac{-e^{-\lambda_j t}}{\lambda_j \prod_{\substack{k=1 \\ k \neq j}}^{i} (\lambda_k - \lambda_j)} \Bigg|_{t_1}^{t_2}$$

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Monte Carlo Uncertainty

- Uncertainty evaluation is a new capability for a delayed gamma ray model
- Users can obtain more accurate estimates of the model uncertainty using Monte Carlo analysis
- Numerous FIER trials are calculated, each with a statistically varied input data library
- The standard deviation of these trials provides the uncertainty in the model output
- This method accounts for correlation in the model output
- While this method is more accurate, it is computation time and memory intensive

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FIER Applications

- Delayed gamma rays provide a unique signature for fission isotopes:
  - Post-detonation forensics
  - Non-destructive assay
- Gamma decay heat could be calculated with FIER
  - Nuclear power
- Fission product build-up can be calculated with FIER
  - Medical isotope production
- Integral testing of nuclear data efficacy

Experimental Comparison

- Delayed gamma-ray spectra were measured using two HPGe's (high purity Germanium detectors)
- The fission source was the Godiva critical assembly at the Nevada Nuclear Test Site
  - Godiva is a pulsed device with a pulse width of ~35 μs [3]

Courtesy of Wikimedia

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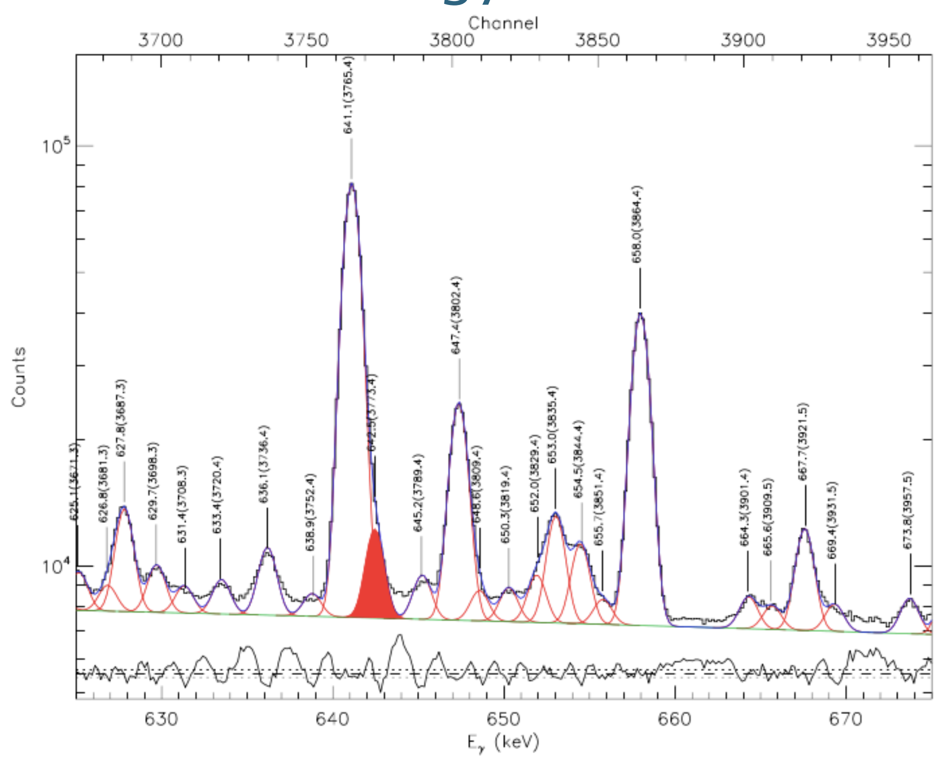
[3] R. Mosteller, J. Goda, Analysis of Godiva-IV Delayed-Critical and Static-Super-Prompt-Critical Conditions, Tech. Rep. LA-UR-09-01007, Los Alamos National Lab, May 2009

Experimental Comparison - XGAM

- The delayed gamma-ray spectra that were collected from the experiment were then analyzed using the XGAM spectral analysis code [4]
- XGAM is well suited for analyzing complicated gamma-ray spectra such as those resulting from fission across the full energy window of the detector



[4] W. Younes, An Overview of the XGAM Code and Related Software for Gamma-ray Analysis, Tech. Rep. LLNL-TR-665689, Lawrence Livermore National Lab, Dec. 2014

Experimental Comparison

- These counts are then efficiency corrected to get the number of source emissions
- FIER was run using an irradiation and counting scheme matching that of the experiment
- The experimentally measured gamma emissions extracted using XGAM and the FIER output were compared using the following percent difference figure-of-merit:

$$\varepsilon = \frac{C_{XGAM} - C_{FIER}}{C_{XGAM}}$$

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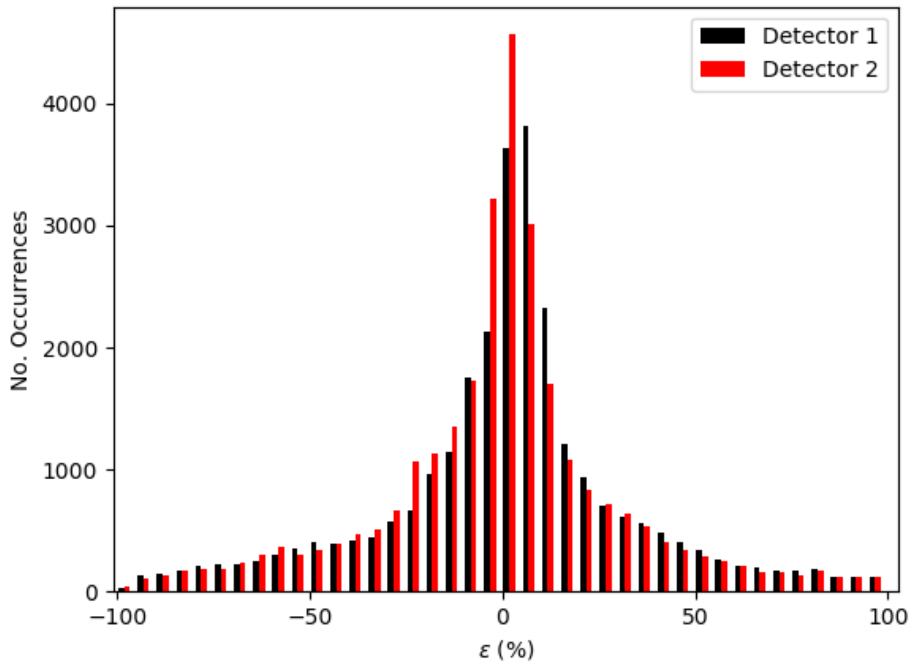
Experimental Comparison

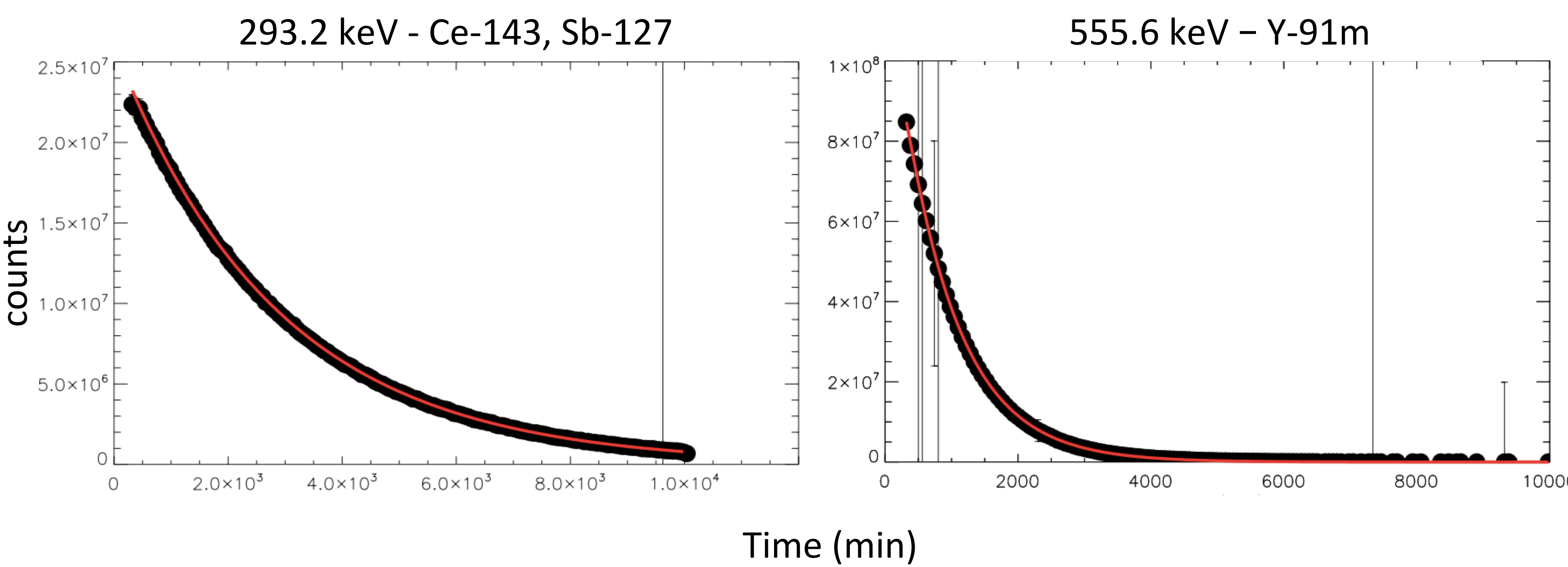
- The results are in generally good agreement, however, there is a roughly symmetric disagreement.
- The source of this disagreement could arise from uncertainty in the data analysis or from nuclear data inaccuracies/deficiencies/discrepancies



35 gamma rays,
163 time steps

Case 1 – Good Agreement

- There are several examples where FIER and the experimental data agree very well:



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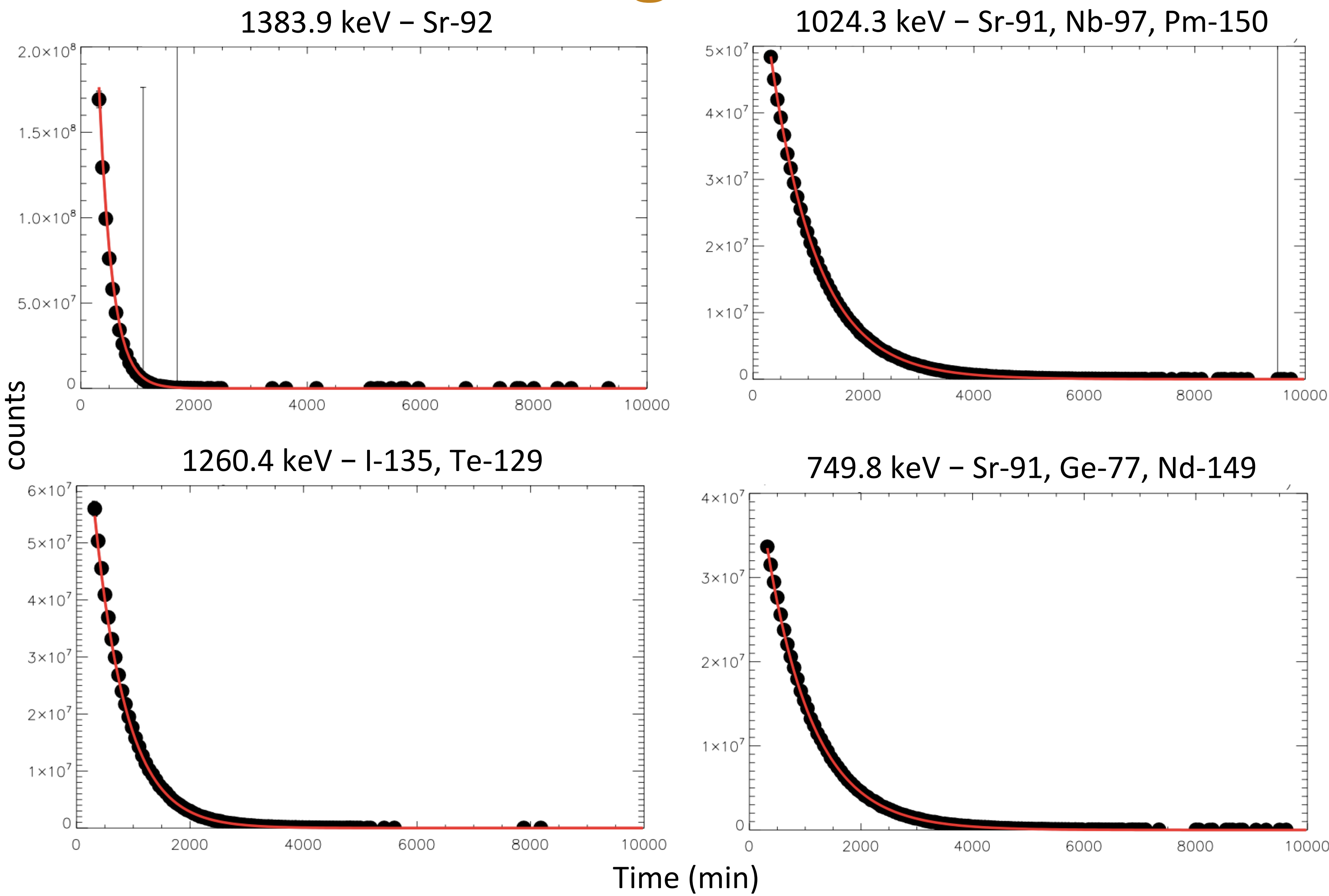
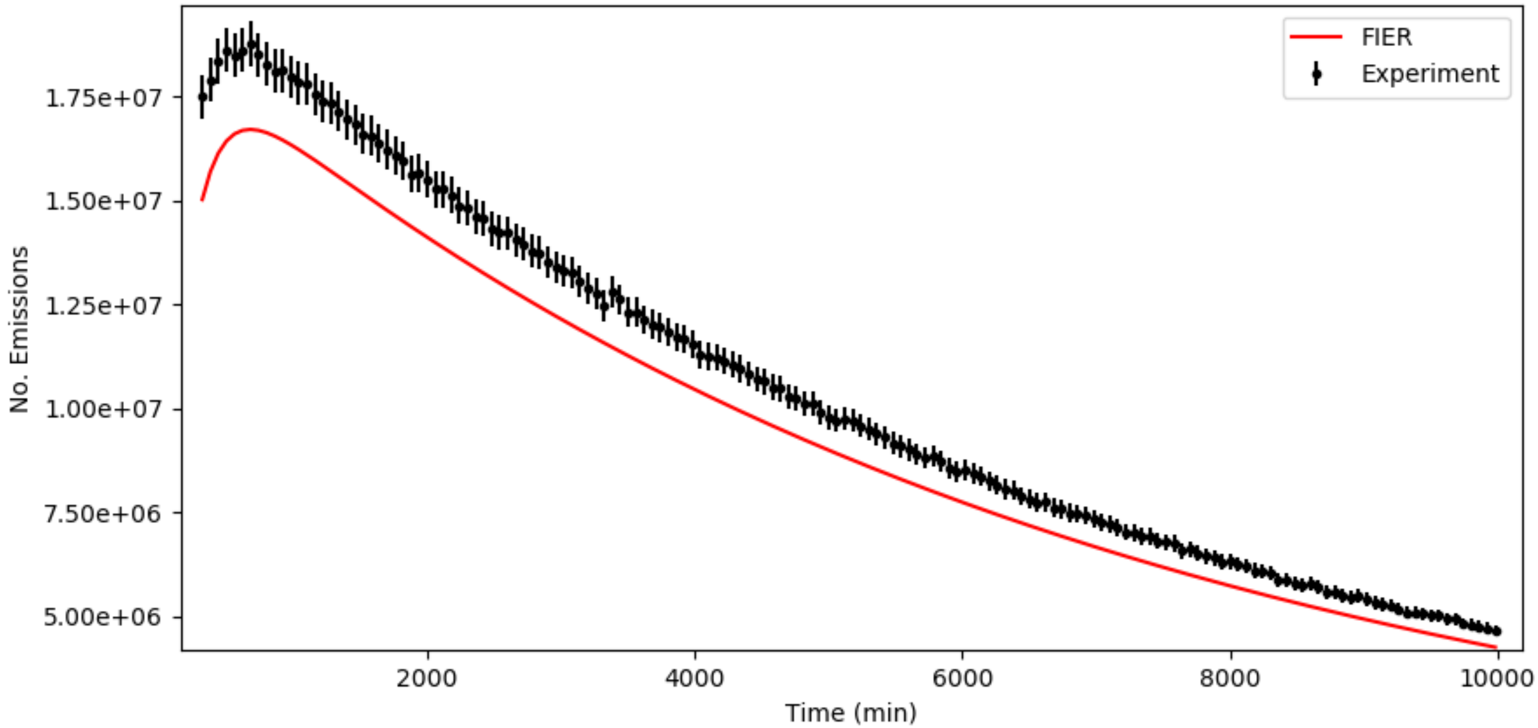


Case 2: 667.7 keV γ from I-132, Cs-132, Sb-127

- It can be seen there is a discrepancy between the experimental data and the FIER model. It appears to be a magnitude offset.



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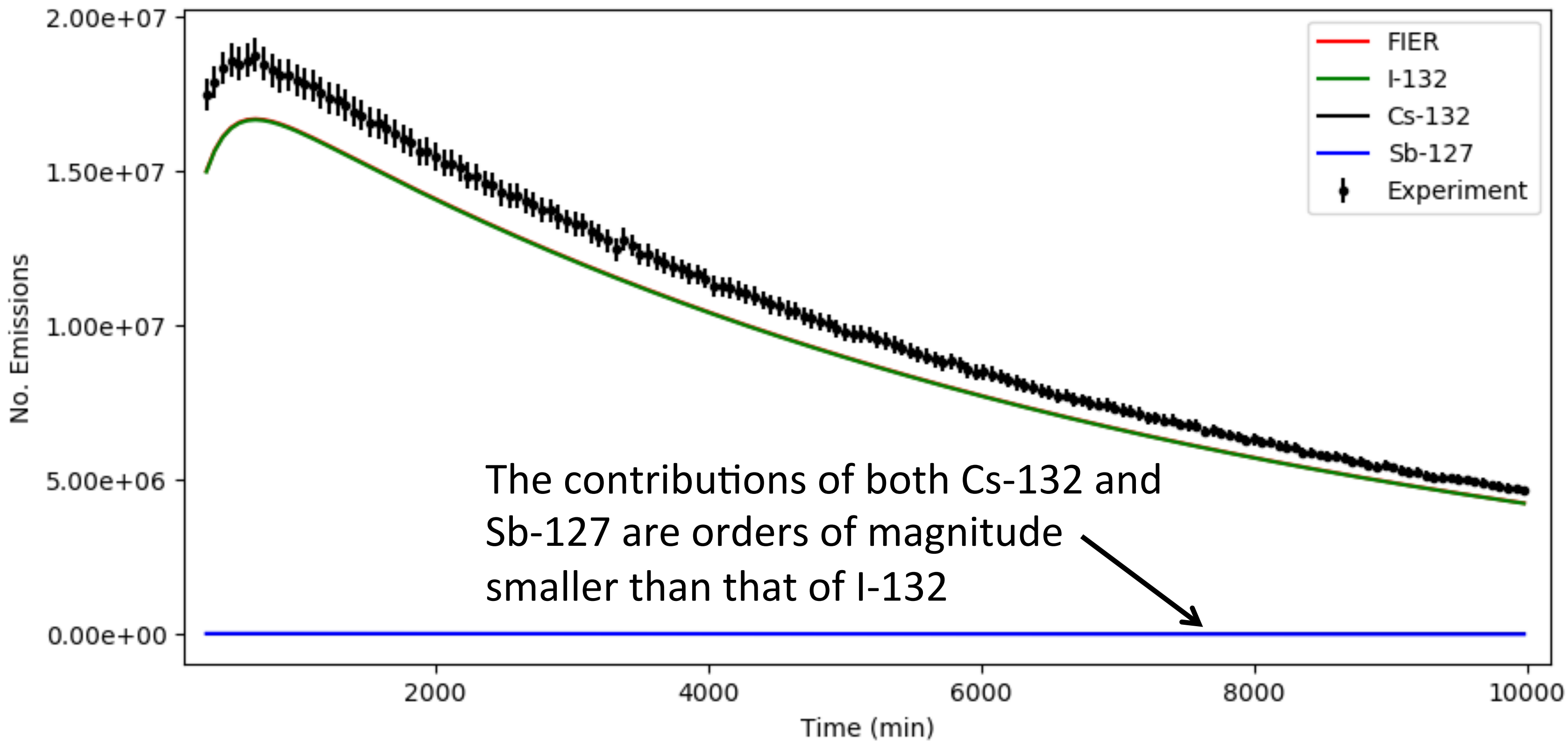
Case 2: 667.7 keV γ from I-132, Cs-132, Sb-127

- FIER allows us to break this line up into its various components:



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Case 2: 667.7 keV γ from I-132, Cs-132, Sb-127

- Because the discrepancy is a magnitude offset that is constant in time, the discrepancy could be caused by:
  - the gamma intensity could be too small
  - a fission yield of one of the long-lived species in the decay chain leading to I-132 could be too small

- The discrepancy is less likely to be the result of the gamma intensity as it appears to be well characterized with an uncertainty of less than 1%

$$I_\gamma = 98.7 \pm 0.66\%$$

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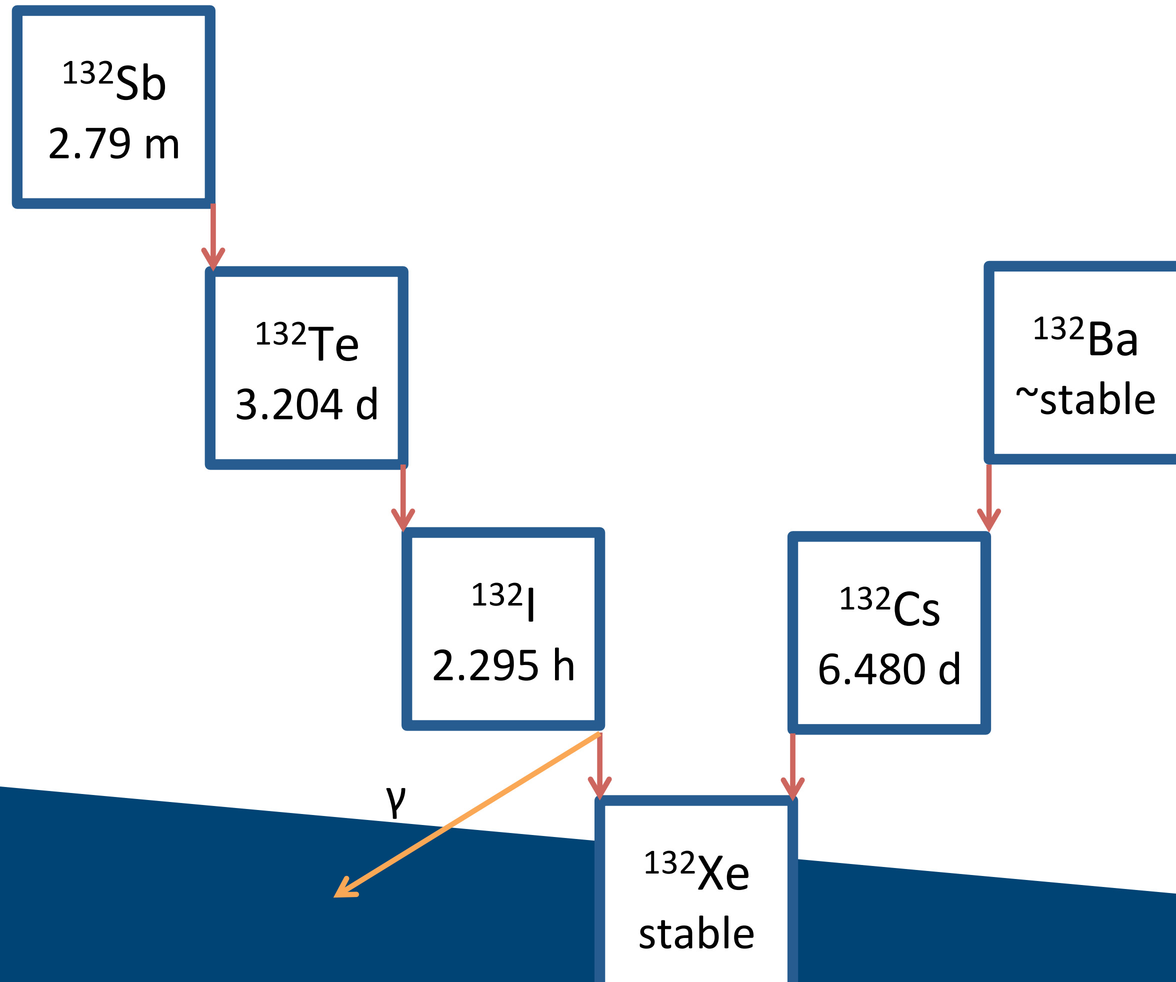
Case 2: 667.7 keV γ from I-132, Cs-132, Sb-127

- Use the mass chain evaluation to determine which species are relevant on this time scale.

- This gamma was observed over one week in one hour bins, therefore look at the independent or cumulative yields of long-lived species in the chain



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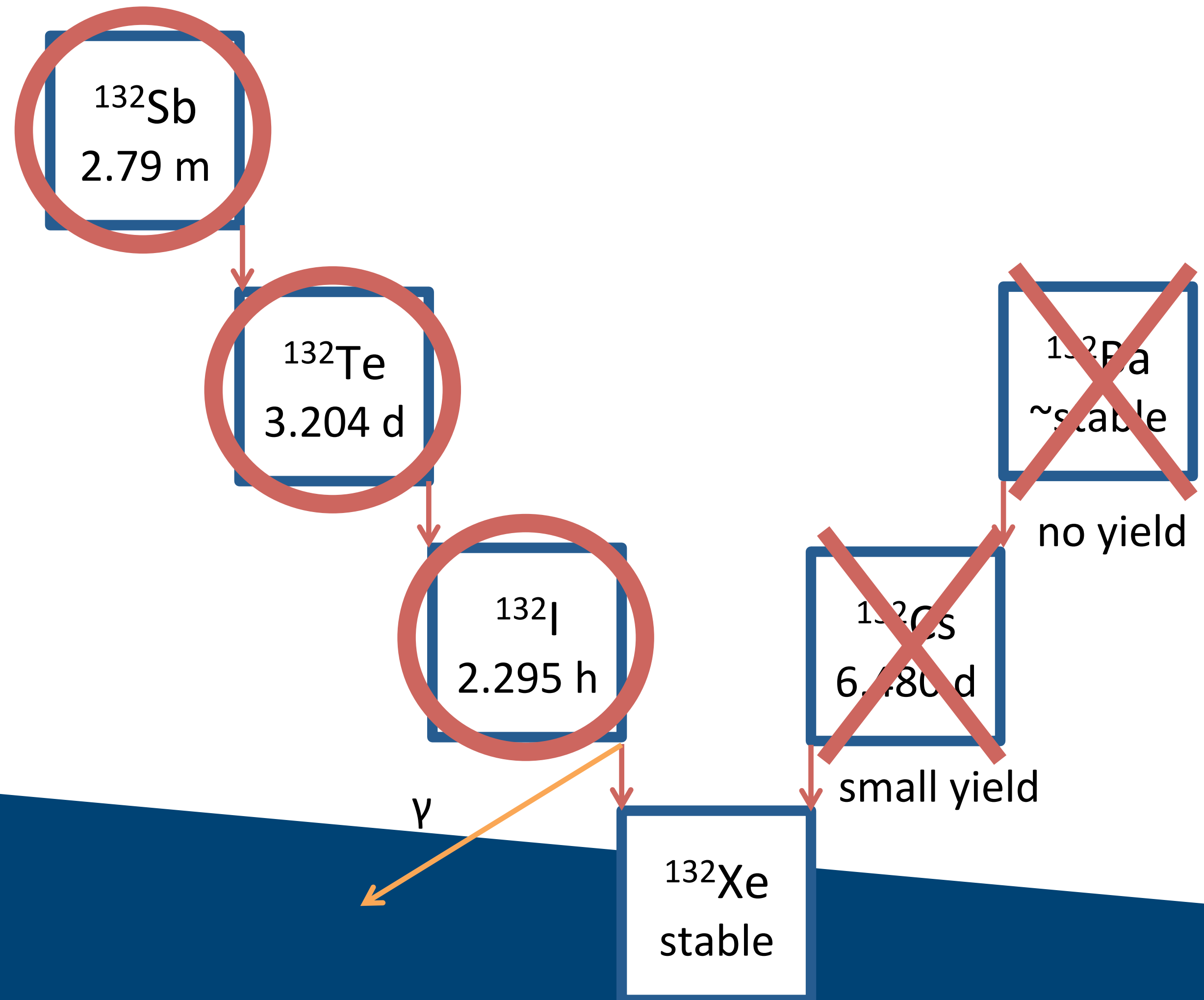

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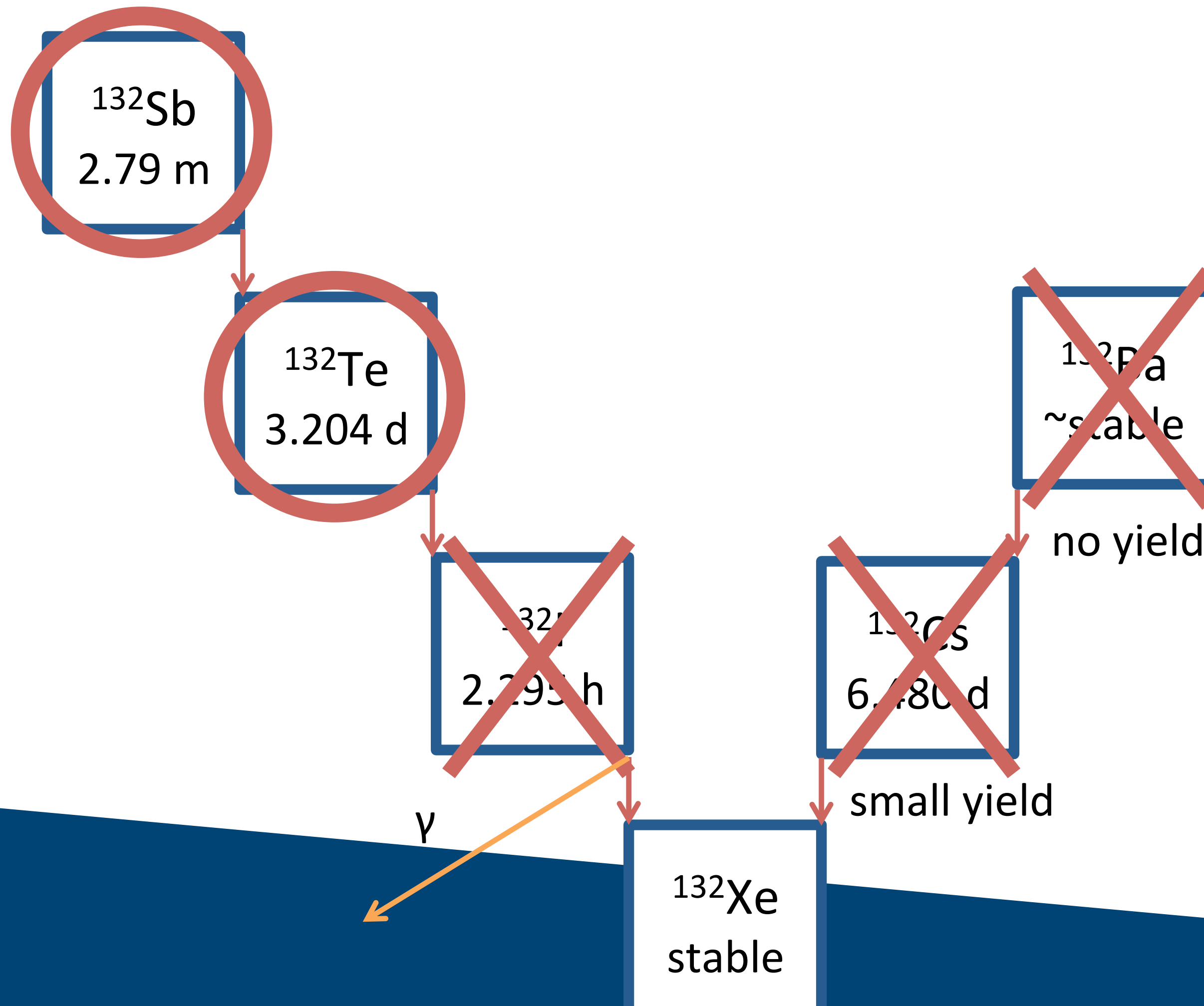
Case 2: 667.7 keV γ from I-132, Cs-132, Sb-127

- Use the mass chain evaluation to determine which species are relevant on this time scale.

- As the discrepancy is a magnitude offset and the discrepancy persists for a week after fission has ceased, focus on the longer lived Te–132 and its parent Sb–132



Case 2: 667.7 keV γ from I-132, Cs-132, Sb-127

- Inspect the independent fission yields of Te-132 and Sb-132:

$$Y_{Te132} = 1.074 \pm 0.172\% \qquad Y_{Sb132} = 1.720 \pm 0.550\%$$

- Indeed either fission yield could be the culprit, with an uncertainty of over 15% and 30%, respectively.
- Sb-132 has a 2.79 minute half-life. Given the time scale of the experiment, it will have all decayed to Te-132 before counting began. Thus it is more useful to discuss the cumulative yield of Te-132:

$$\xi_{Te132} = 2.794 \pm 0.576\%$$

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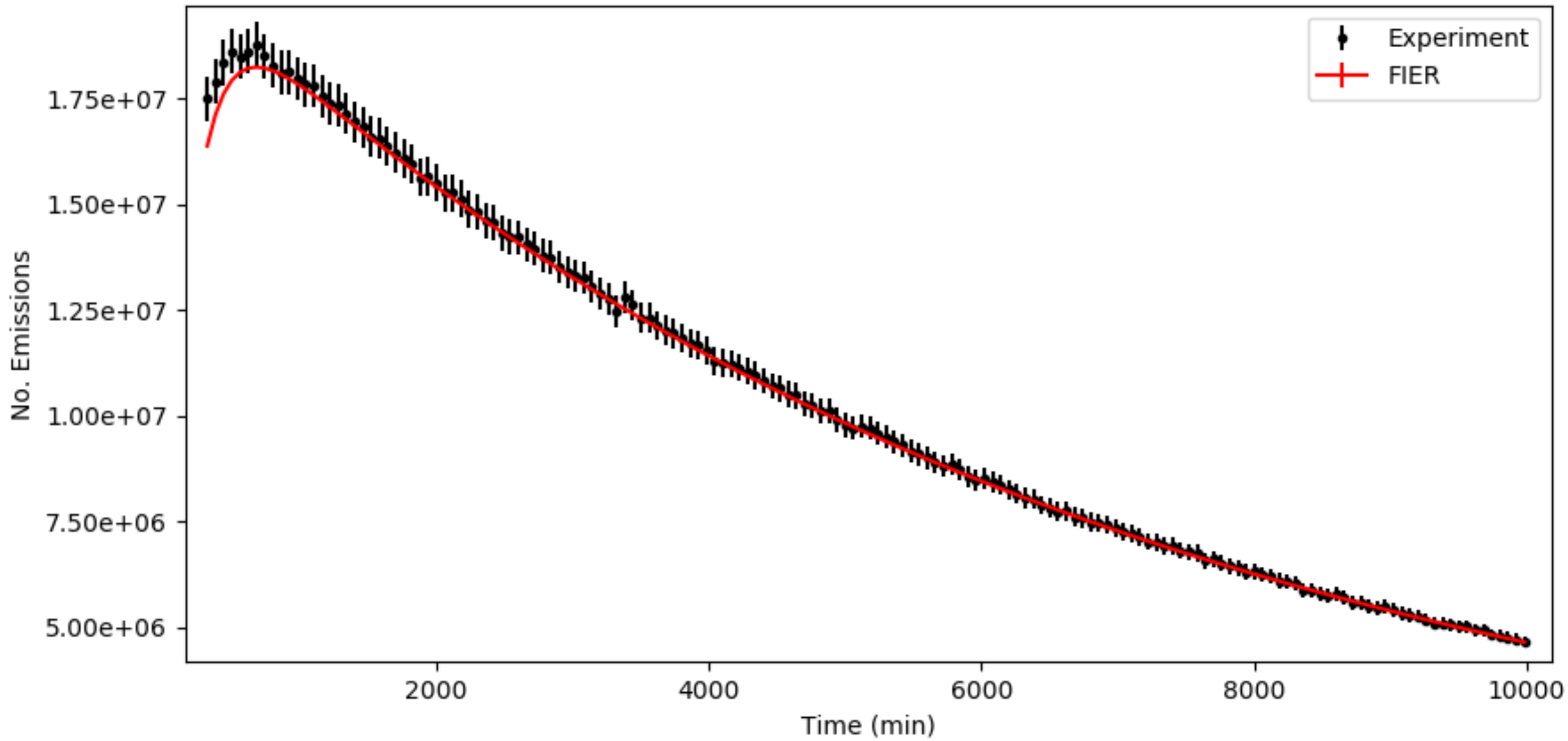
Case 2: 667.7 keV γ from I-132, Cs-132, Sb-127

- When the cumulative yield of Te-132 is increased by 0.75σ to 3.20%, the discrepancy is largely resolved.



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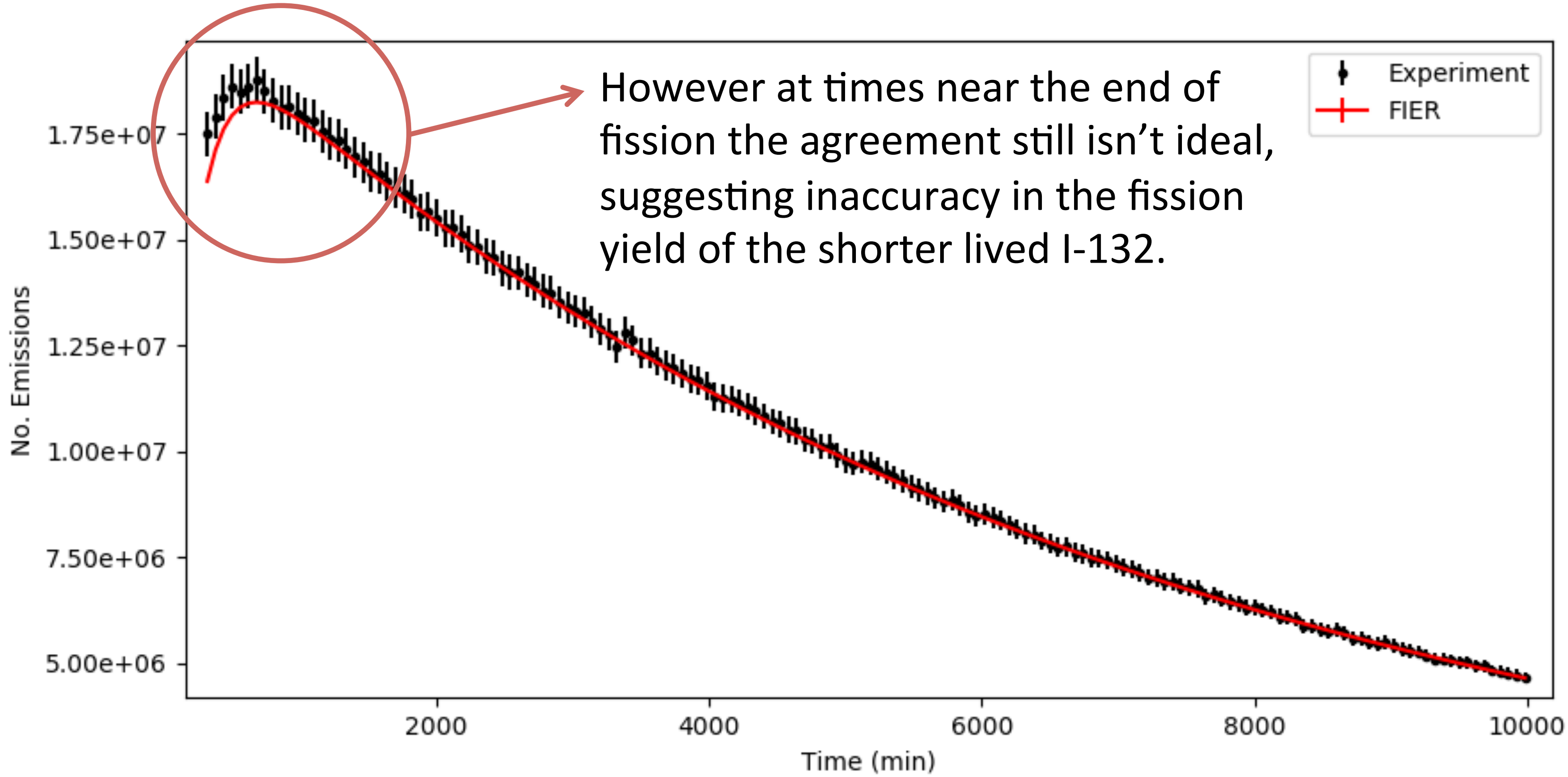
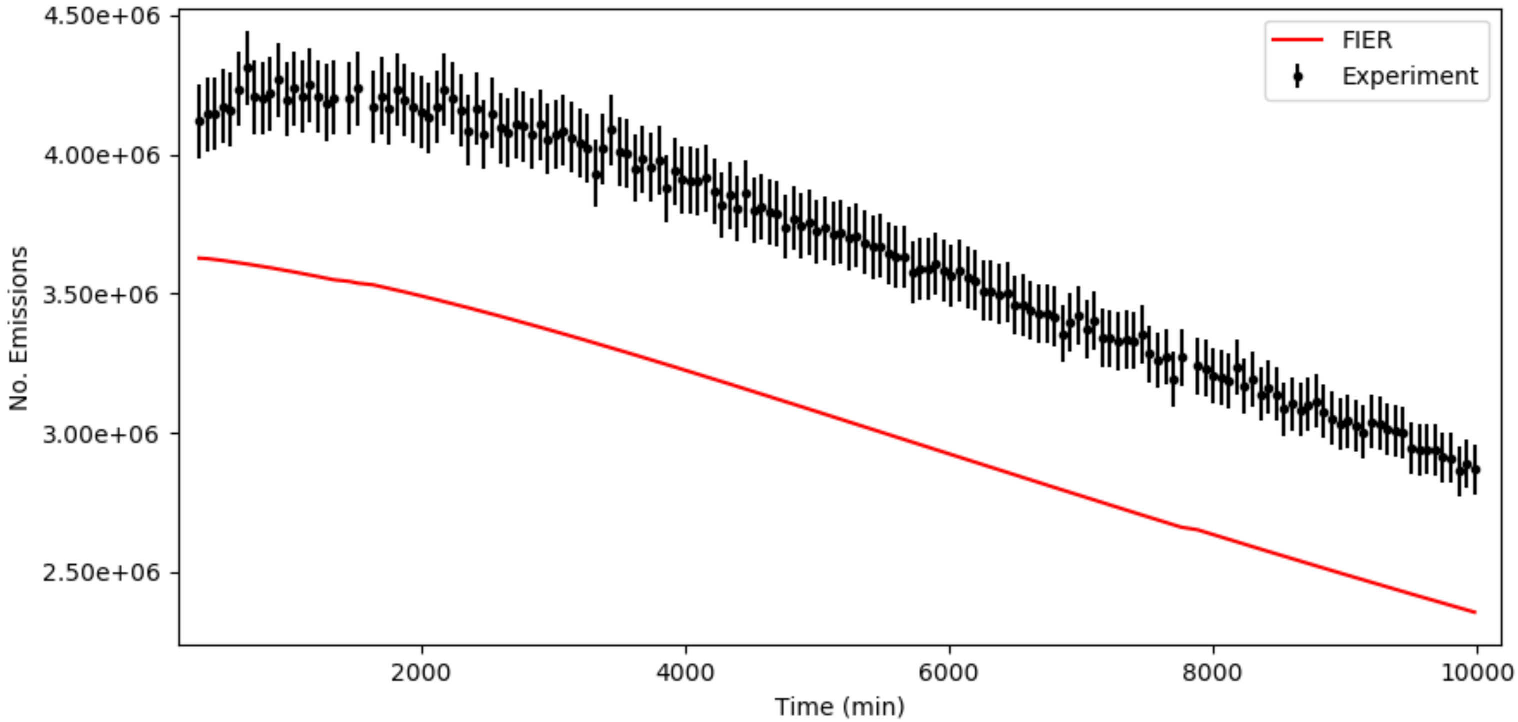


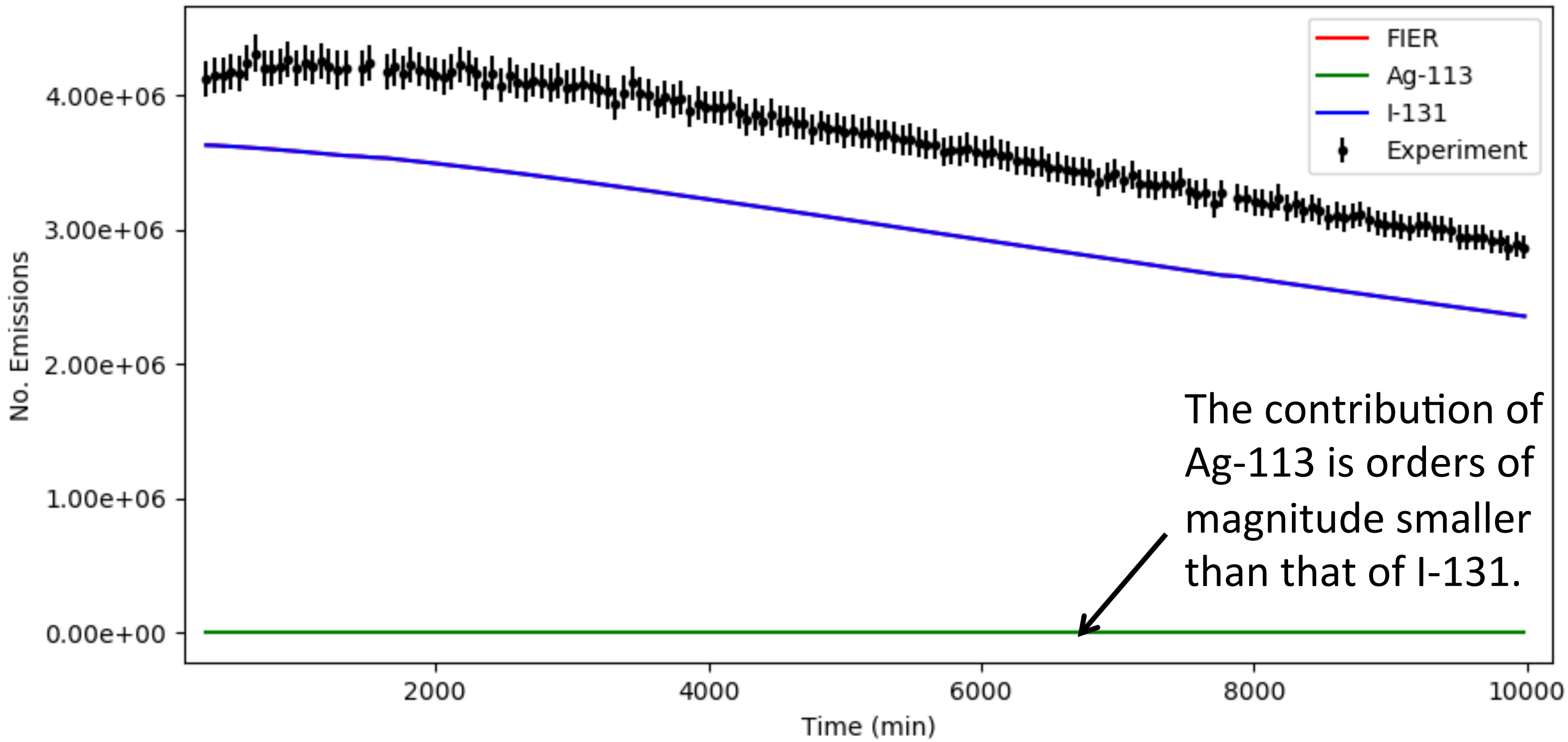
Case 3: 364.5 keV γ from I-131 and Ag-113

- It can be seen there is a discrepancy between the experimental data and the FIER model. It appears to be a magnitude offset and perhaps a half-life discrepancy.



Case 3: 364.5 keV γ from I-131 and Ag-113

- Inspect the nuclide contributions:



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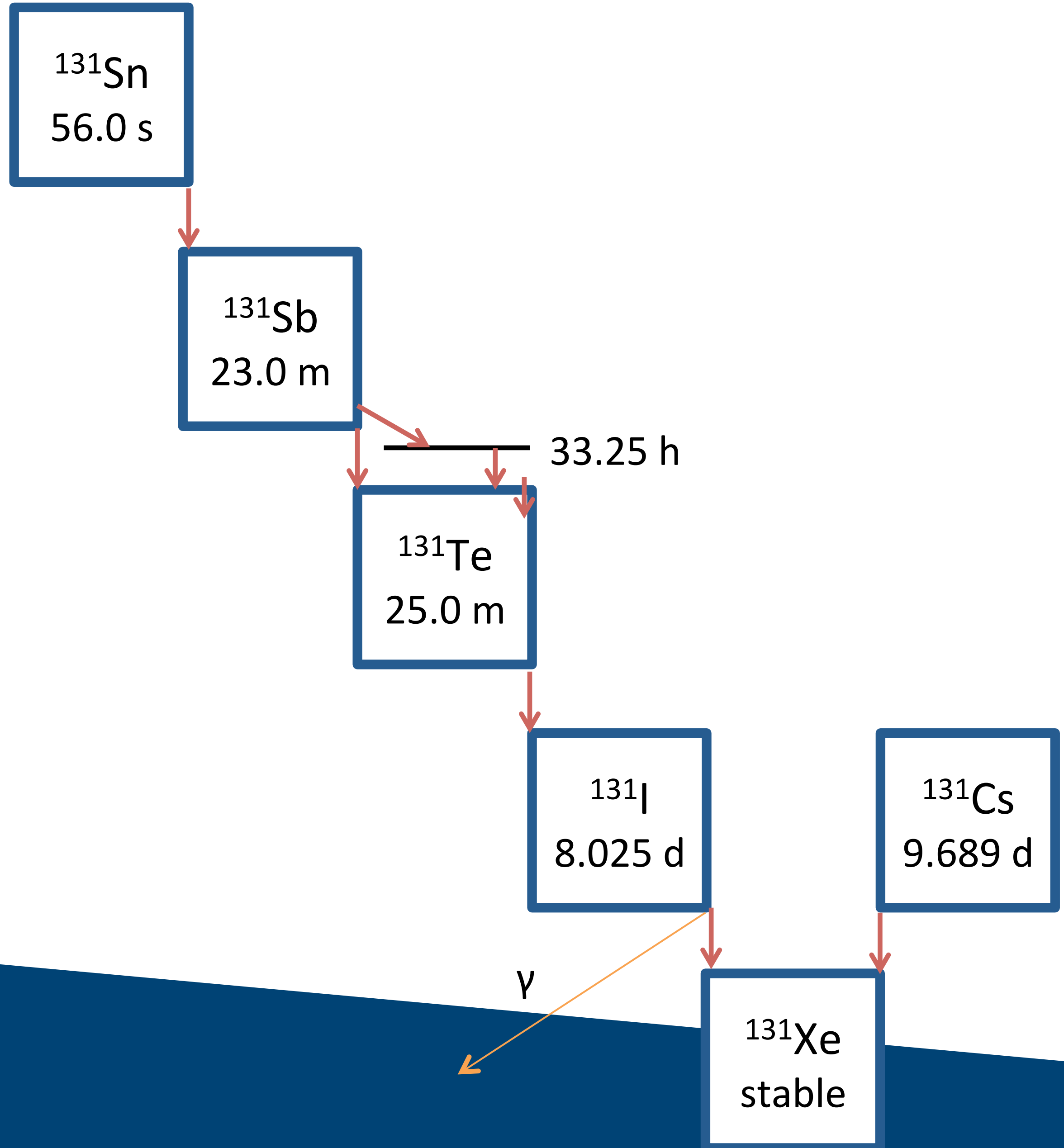
Case 3: 364.5 keV γ from I-131 and Ag-113

- Inspect the mass chain:



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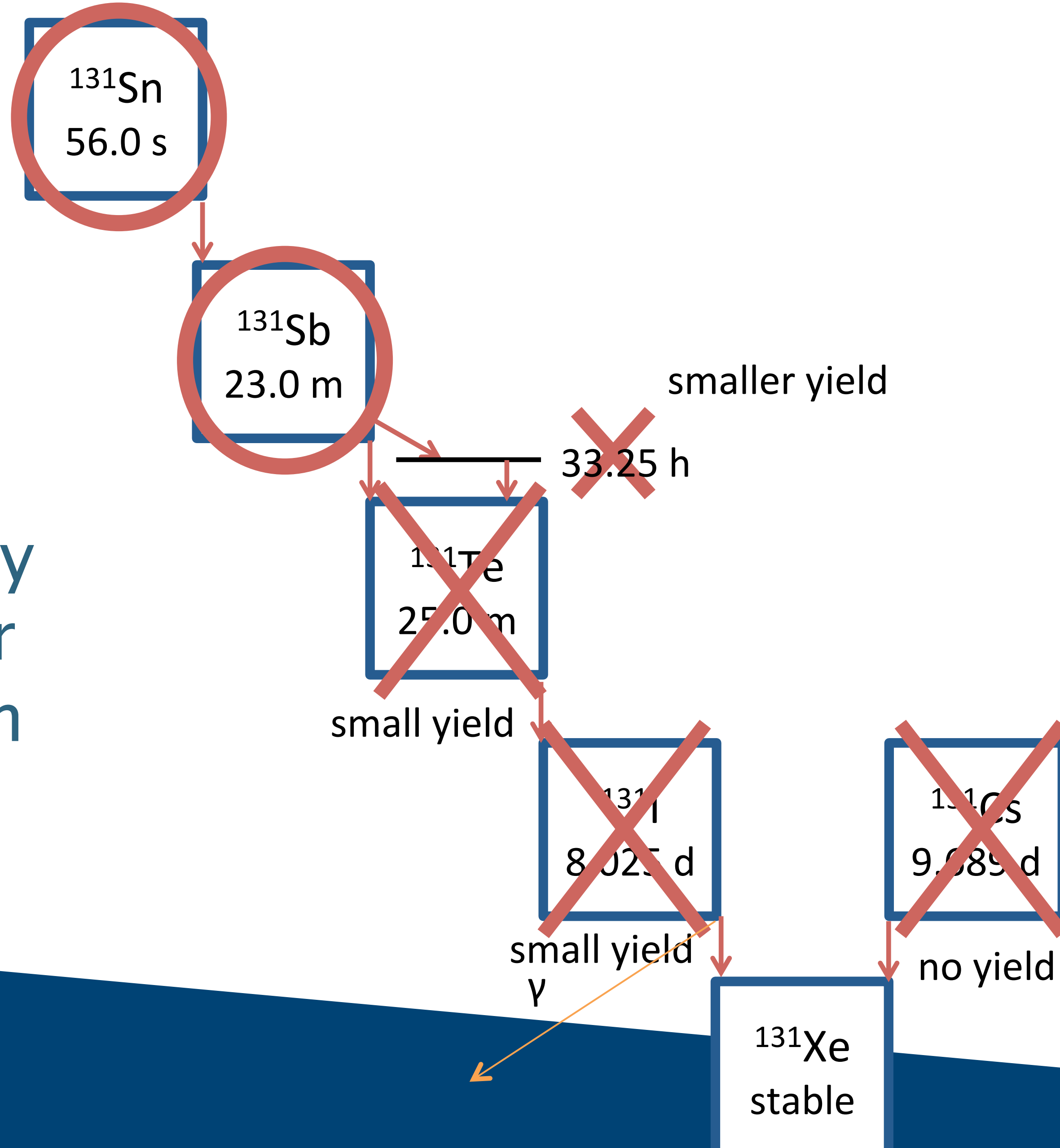
Case 3: 364.5 keV γ from I-131 and Ag-113

- Sn-131 and Sb-131 make up the majority of the yield for the mass chain



Case 3: 364.5 keV γ from I-131 and Ag-113

- Inspect the independent fission yields of Sb-131 and Sn-131:

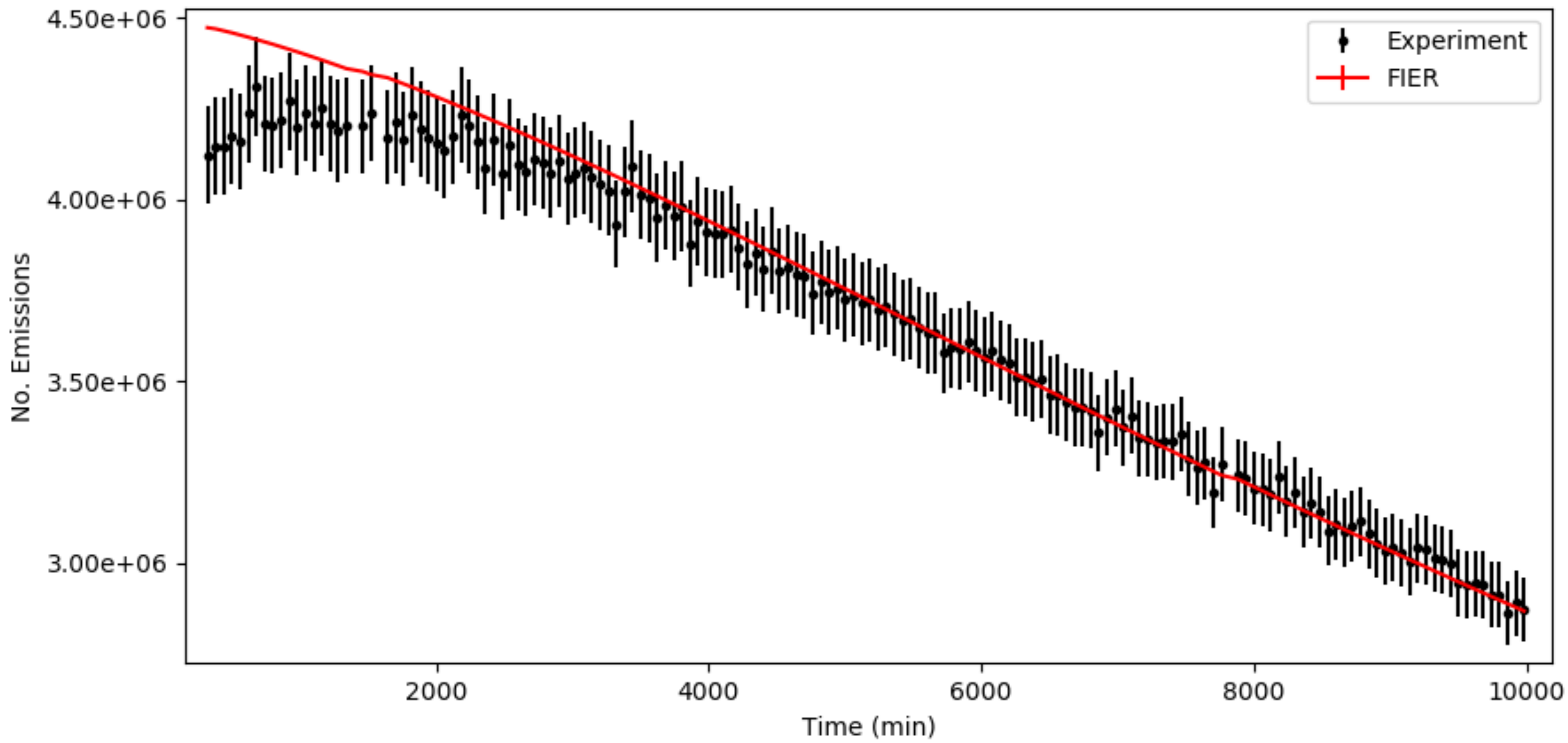
$$Y_{Sb131} = 1.496 \pm 0.344\% \quad Y_{Sn131} = 0.693 \pm 0.443\%$$

- Sn-131 has a 56 second half-life. Given the time scale of the experiment, it will have all decayed to Sb-131 before counting began. Thus it is more useful to discuss the cumulative yield of Sb-131:

$$\xi_{Sb131} = 2.189 \pm 0.561\%$$

Case 3: 364.5 keV γ from I-131 and Ag-113

- Increasing the cumulative yield of Sb-131 by 1.25σ to 2.9% resolves the discrepancy at later times:



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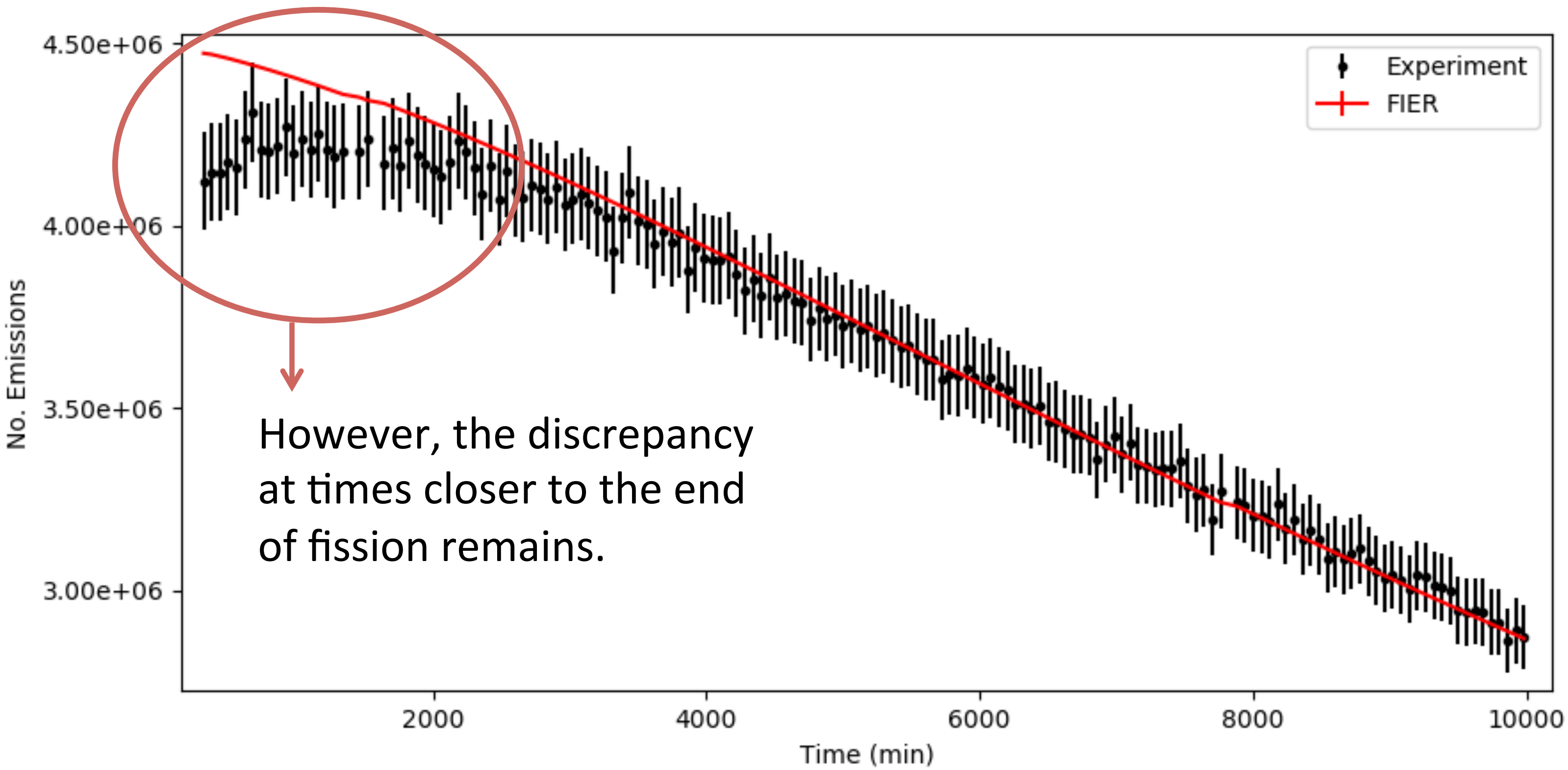

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Conclusion and Future Work

- With the ability to calculate fission production populations and delayed gamma-ray spectra, FIER is useful to numerous applications
- We've demonstrated that model output from FIER can be used as an integral benchmark the efficacy of nuclear data
- A publication is written and the code is being prepared for public release
- Extend FIER's delayed particle capabilities beyond gammas to betas, neutrinos, beta-neutrons, alphas, etc.

Thank you!

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Bibliography

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