Measurement of Fission Product Yields at the National Synchrotron Light Source II

M. Topsakal, A. Mattera, S. Ota, A. A. Sonzogni, E. A. Ricard, S. Gill, C. Morse, M. Jandel

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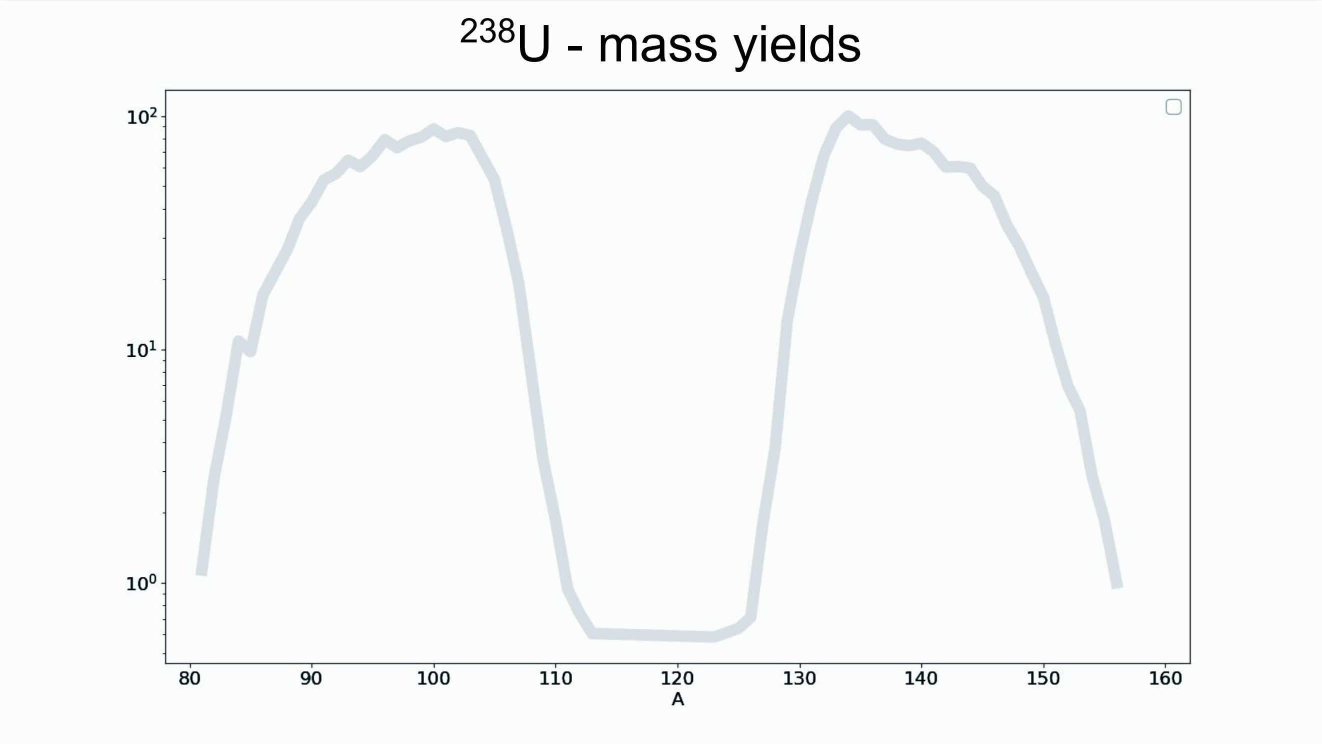
A brief history of Fission Yields evaluation in ENDF



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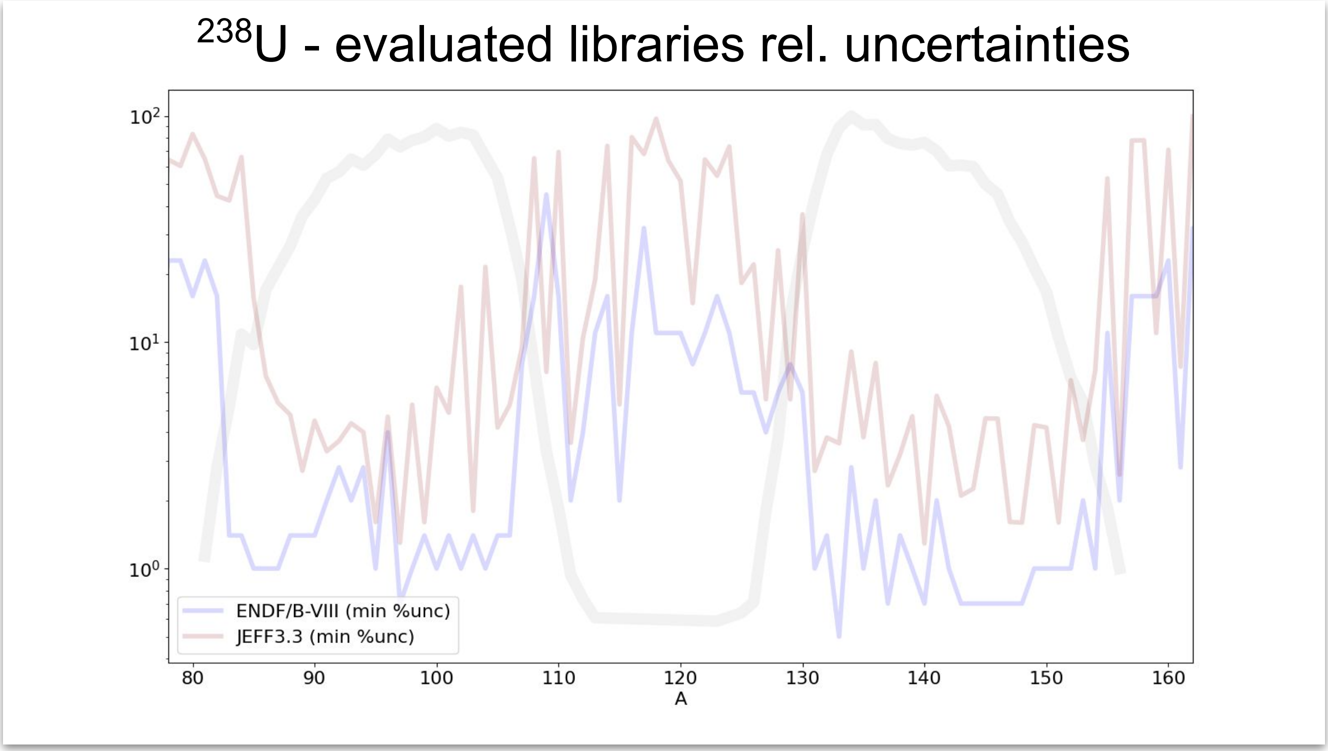

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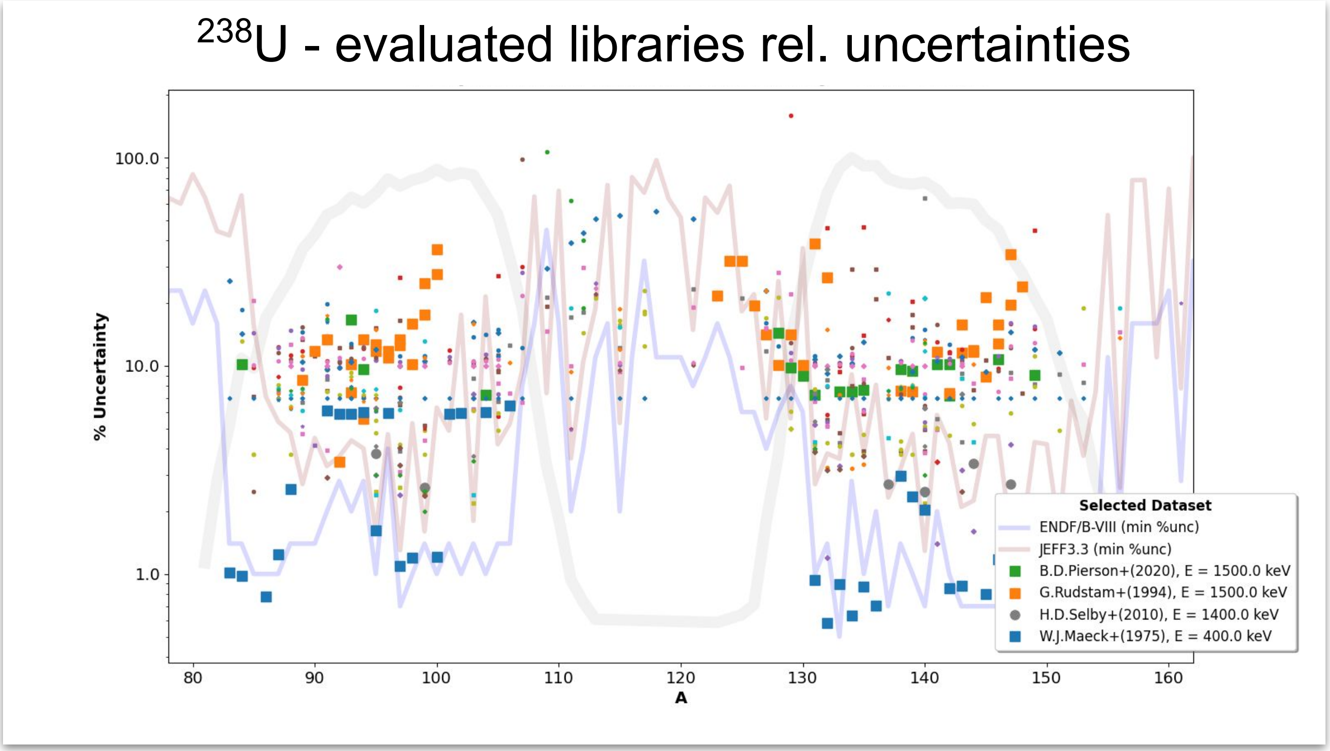

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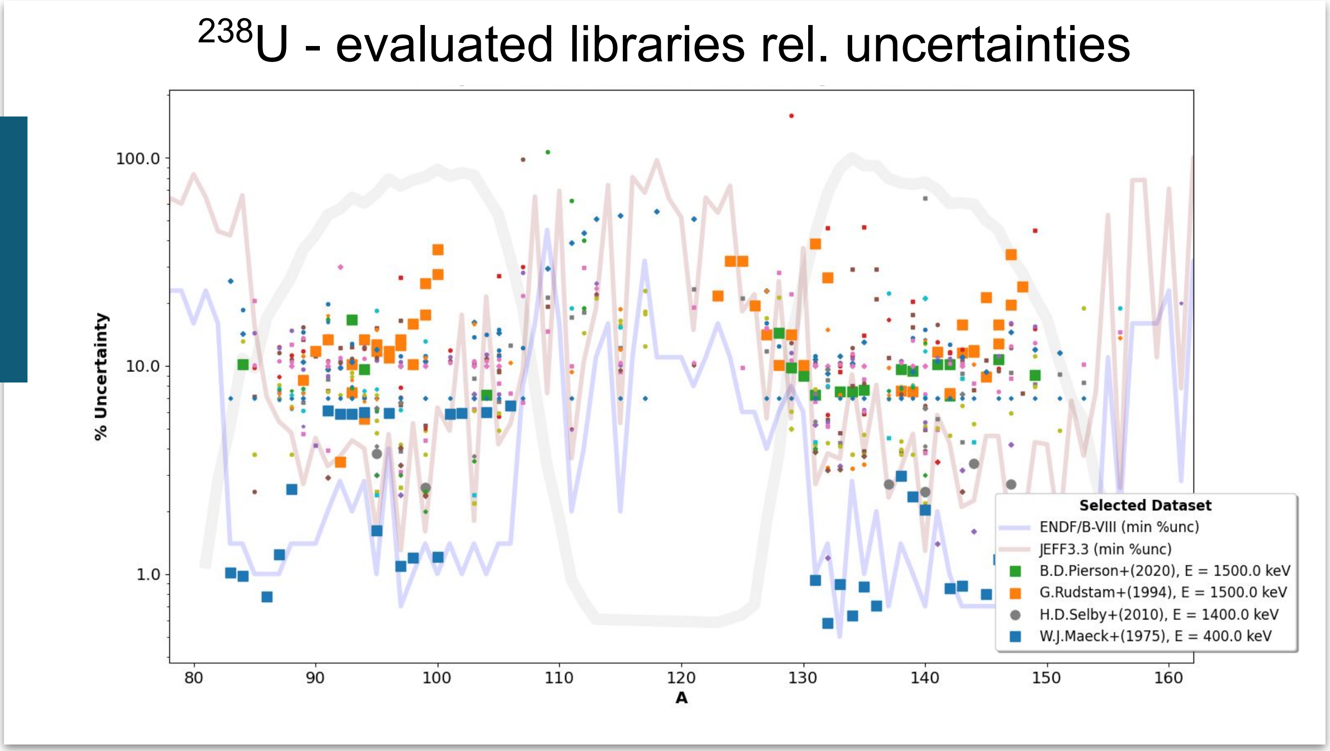
A brief history of Fission Yields evaluation in ENDF

ENDF/B uncertainties follow closely these experimental data



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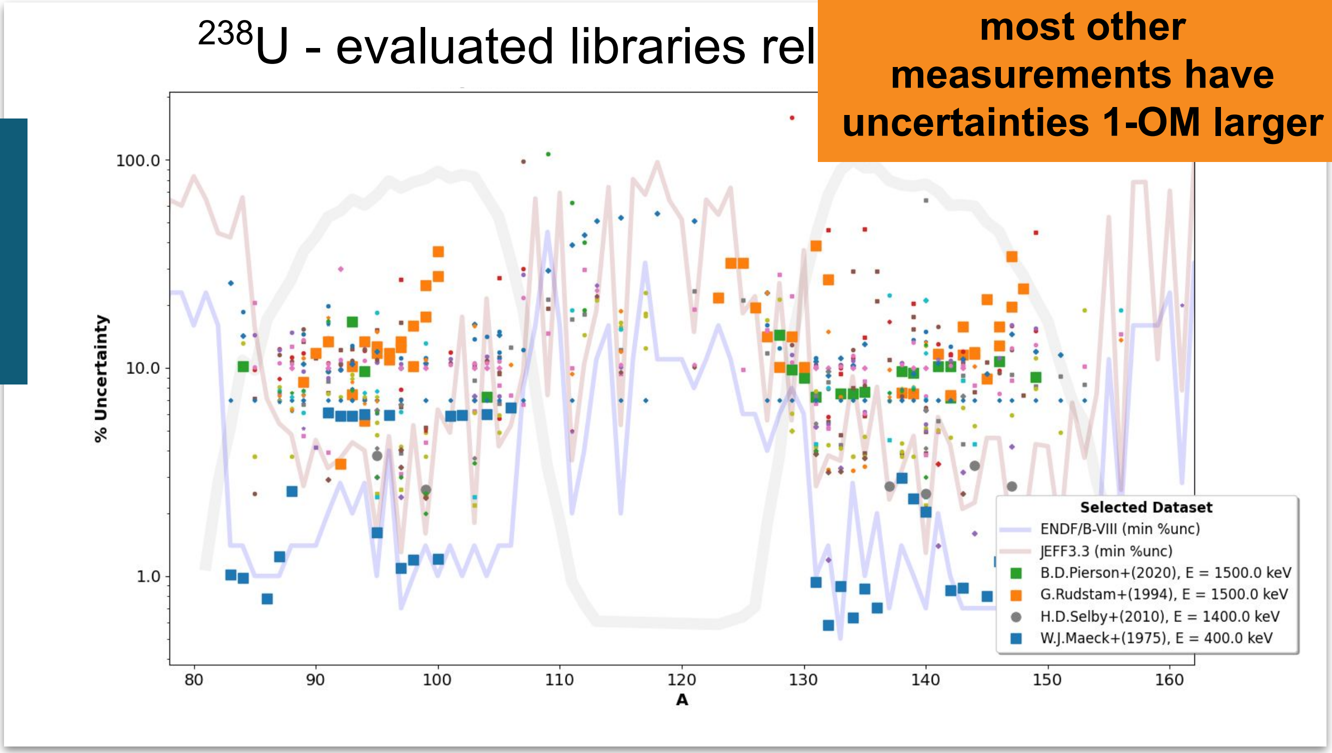

most other measurements have uncertainties 1-OM larger

ENDF/B uncertainties follow closely these experimental data

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Why are they so precise?

Uncertainty Budget

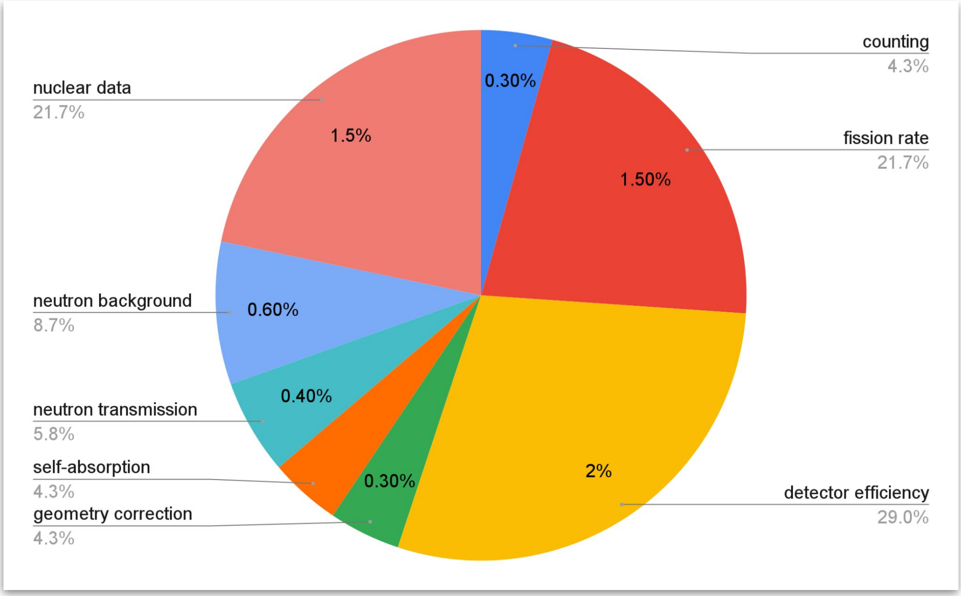

TABLE 6.1.1. UNCERTAINTY SOURCES OF ABSOLUTE FISSION YIELD MEASUREMENT WITH THE γ spectroMETRIC METHOD

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f. neutron transmission correction	0.4	(for thermal neutrons)
g. neutron background correction	0.6-1.0	(for high energy neutrons)
total systematic error	2.6-2.7	$\Delta y_{sys} = \sqrt{\sum_i \Delta y^2_{sys_i}}$
total error	2.6-3.4	$\Delta y_{tot} = \sqrt{\Delta y^2_{sys} + \Delta y^2_{sta}}$

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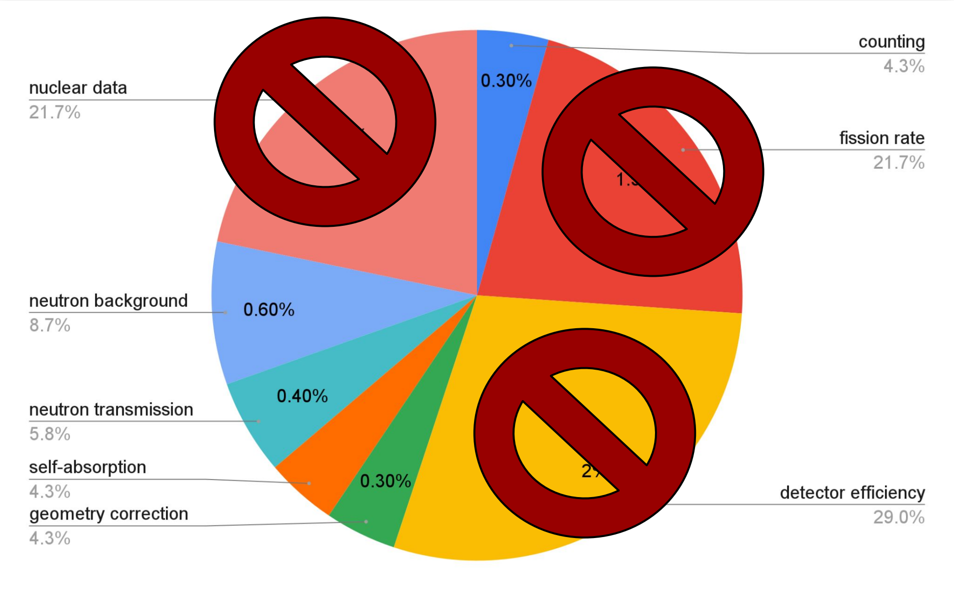
Why are they so precise?

Uncertainty Budget



MS measurements make **no use of Nuclear Data** to extract the absolute yields

The **fission rate** does not need to be determined as long as the majority of the yields can be measured (**normalized to 100%** / mass peak)

The uncertainties were estimated as the standard deviation of repeated measurements (possibly not fully considering systematic effects)

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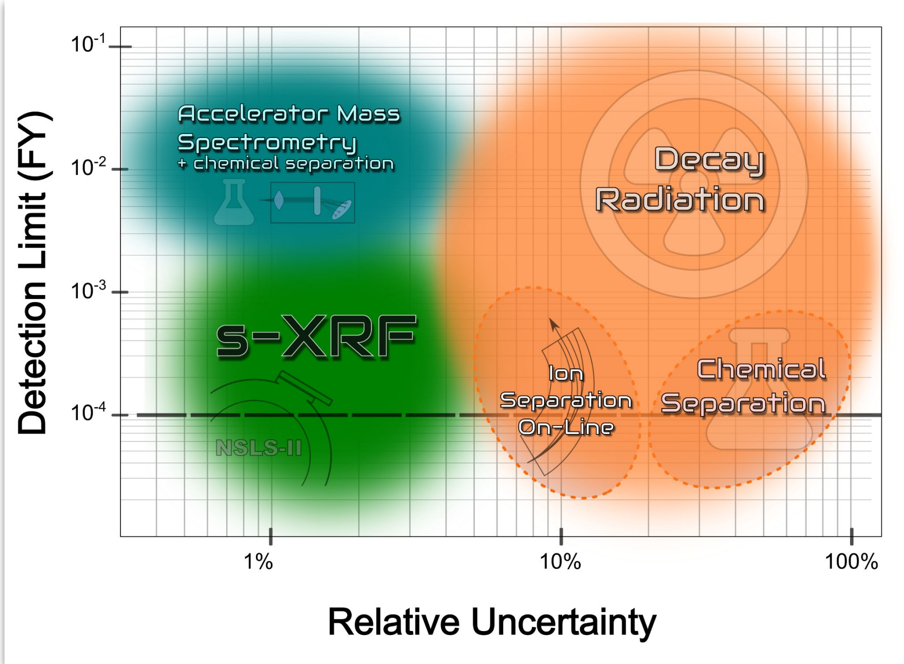
Fission Product Yields at NSLS-II

detection limit → XRF can reach **ppm-level concentrations** of elements in a substrate: this would allow us to get to **low-yield fission products** and a **full normalization** (no fission rate), with no limitations on the measured elements

A calibration of the setup with standard samples would let us not rely on decay data for determination of the absolute yields



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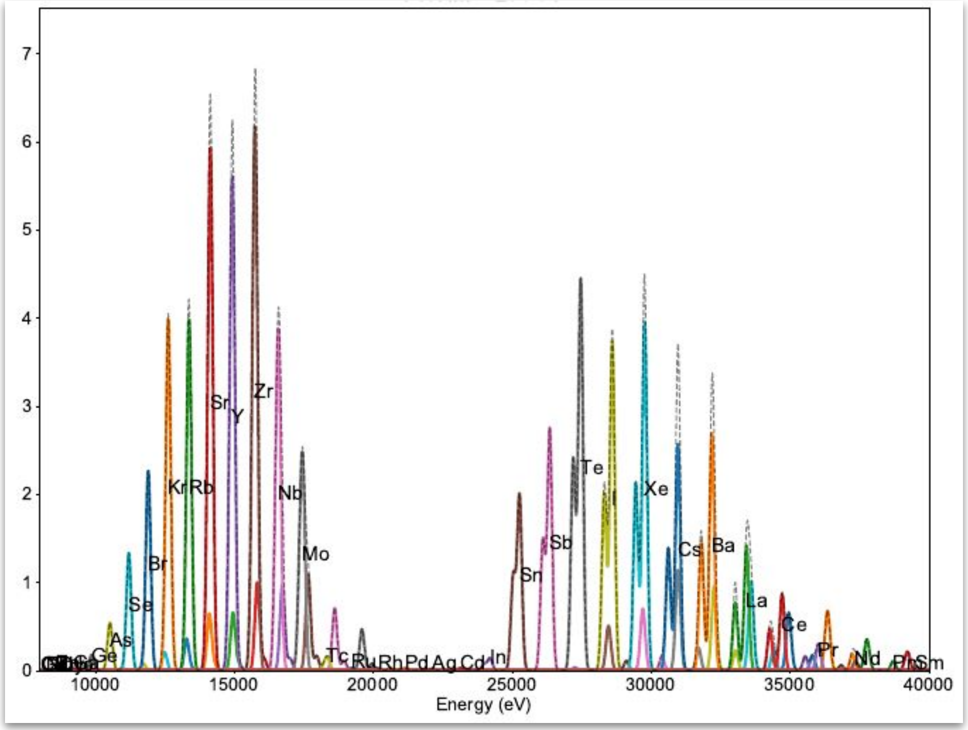
Fission Product Yields at NSLS-II

Long-term goal: use the Light Source to **quantify** the amount of fission products in a sample

NSLS-II combined with the new Ge-XRF detector at NE can allow us to reach low-yield fission products

Measure Z-distribution (XRF) of long-lived or stable fission products *à-la* Maeck (but looking at elements instead of masses) - it provides different information, but still valuable for an evaluation as an 'anchor point'



calculated X-ray peaks from the fission products in a ^{238}U sample irradiated in a fast neutron spectrum

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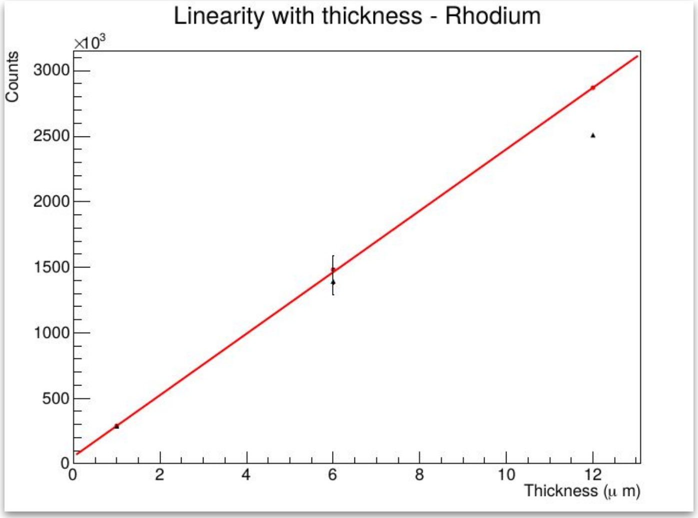
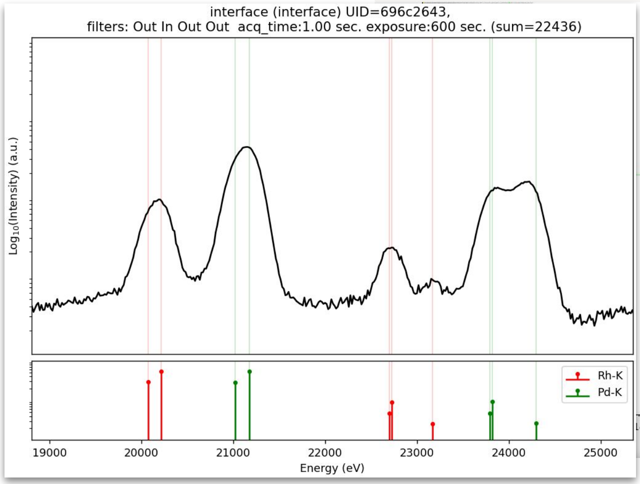
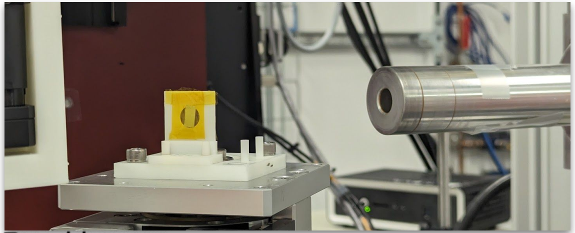
First tests



check of linearity & attenuation corrections





28-ID-2 XPD beamline

70 keV X-ray beam

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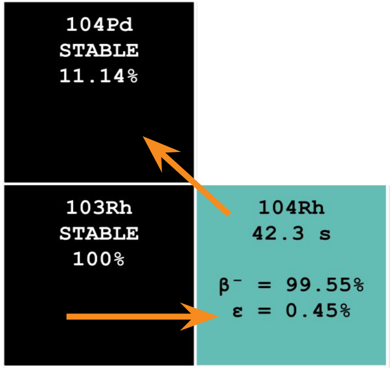
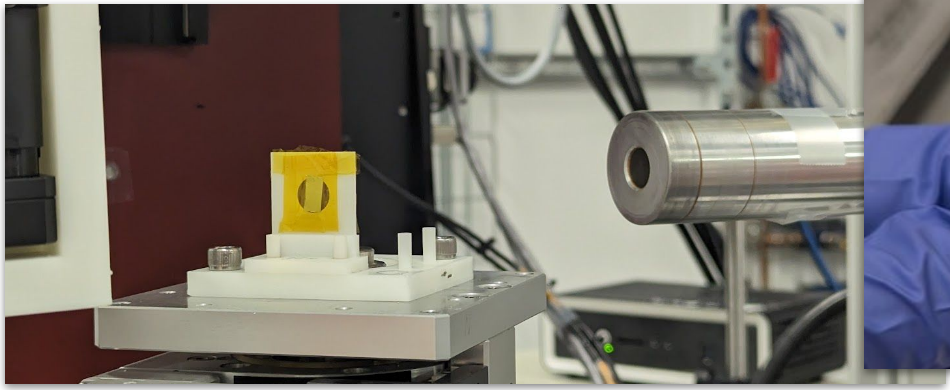
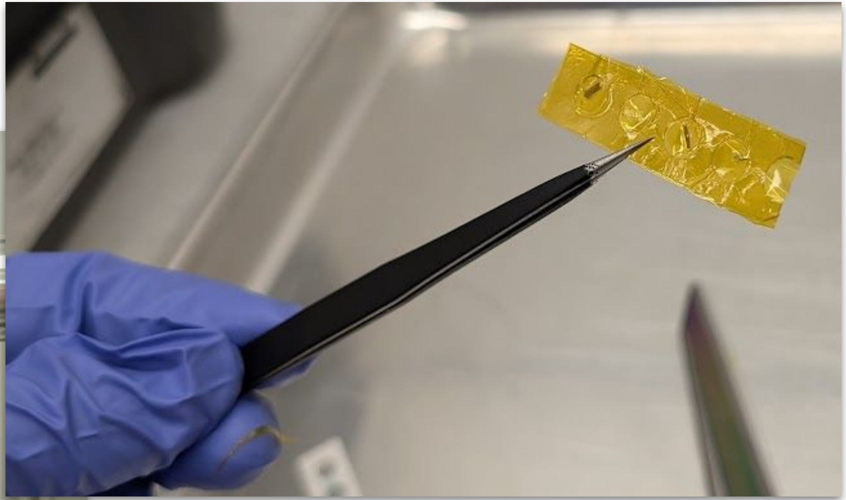
Step 1: Feasibility Study



test the method in a more simple experiment: neutron capture on ^{103}Rh

Rh samples irradiated at Lowell Research Reactor (thermal capture)

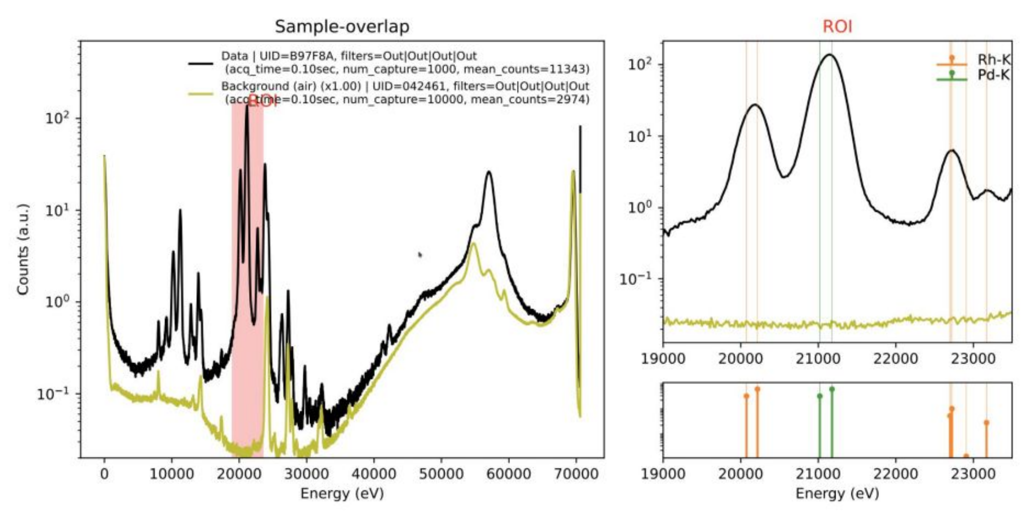


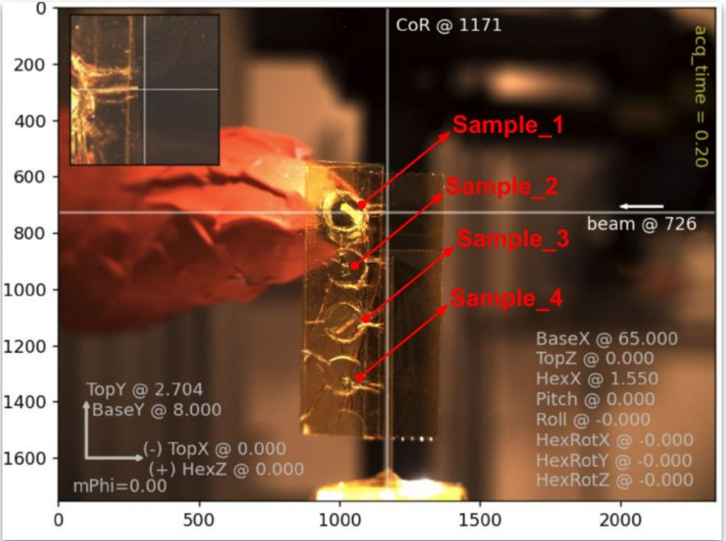


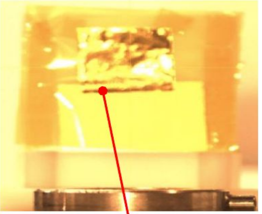
M. Jandel

Step 1: Feasibility Study

Oct 2024 run







Sample_overlap

we confirmed that we can easily quantify elements when concentrations are similar

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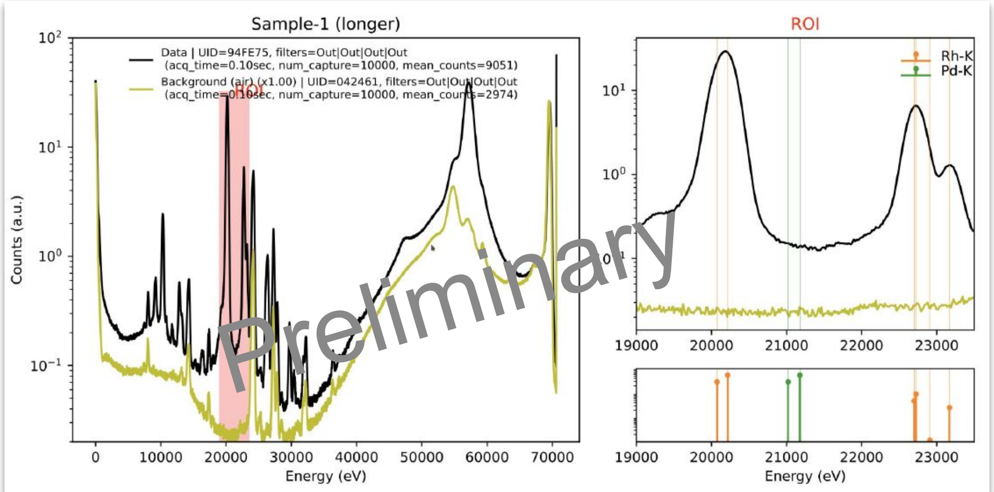
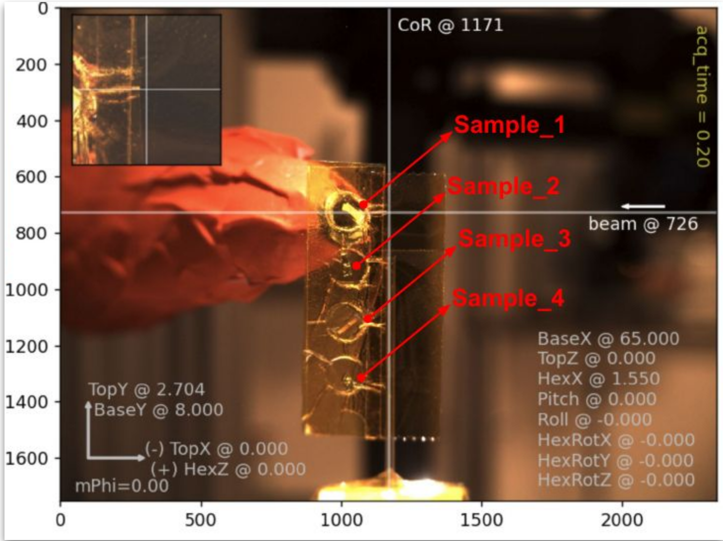
Step 1: Feasibility Study

Oct 2024 run





the Rh peak seems to drown the Pd at product concentrations of ~100 ppm

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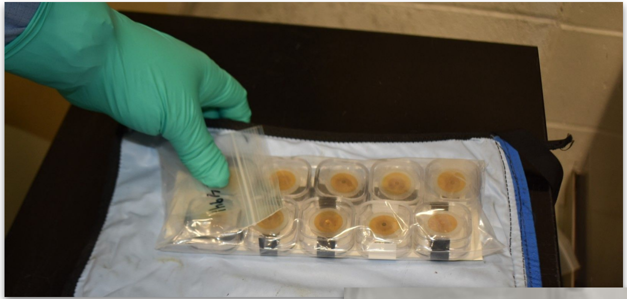
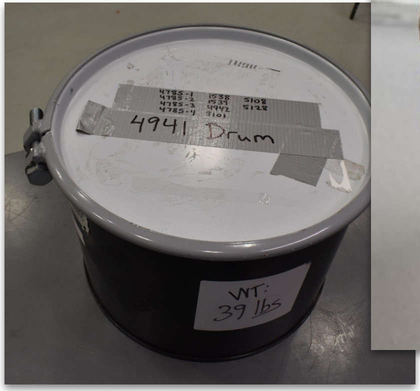
Beamtime with high-dose targets from NSUF

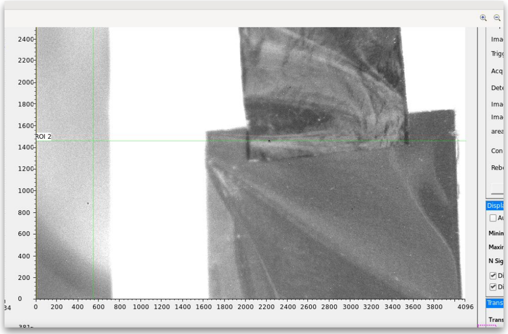
NSUF
Nuclear Science
User Facilities

Sample	Elem.	Integrated Fluence (n/cm²)	approx. product concentration	Applic.
034-08-331 149-08-331	Ag	1.47×10^{22} 2.9×10^{22}	2-4%	FY
052-08-331 109-08-331 148-08-331	W	1.05×10^{22} 1.58×10^{22} 2.94×10^{22}	5-100‰✠	IF
09-157-033 09-157-034 09-157-035	Hf, Al	1.29×10^{22} 1.3×10^{22} 1.3×10^{22}	100-600 ppm✠	IF
10-242-0011	Mo	1.05×10^{21}	20-30 ppm	FY

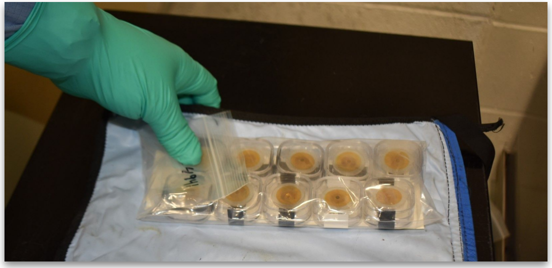




Next steps



- manufacture ad-hoc non-irradiated targets to study the effect of a strong substrate signal on the detection limit?
- simulation of signal and full X-ray measurement setup
- using AMS to complement the Z-distributions with A-distributions for specific fission products?



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Summary

- Exploratory study to test measurement of reaction rates (and Fission Yields) using s-XRF
- Samples irradiated at UMass Lowell measured at NSLS-2 at the end of October -- no apparent signal from 100 ppm of Pd on the strong Rh background
- Two additional beamtimes scheduled for early 2025 to assay high-dose samples from NSUF and ad-hoc samples fabricated to replicate

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JEFF did not adopt them in the evaluation

most other measurements have uncertainties 1-OM larger

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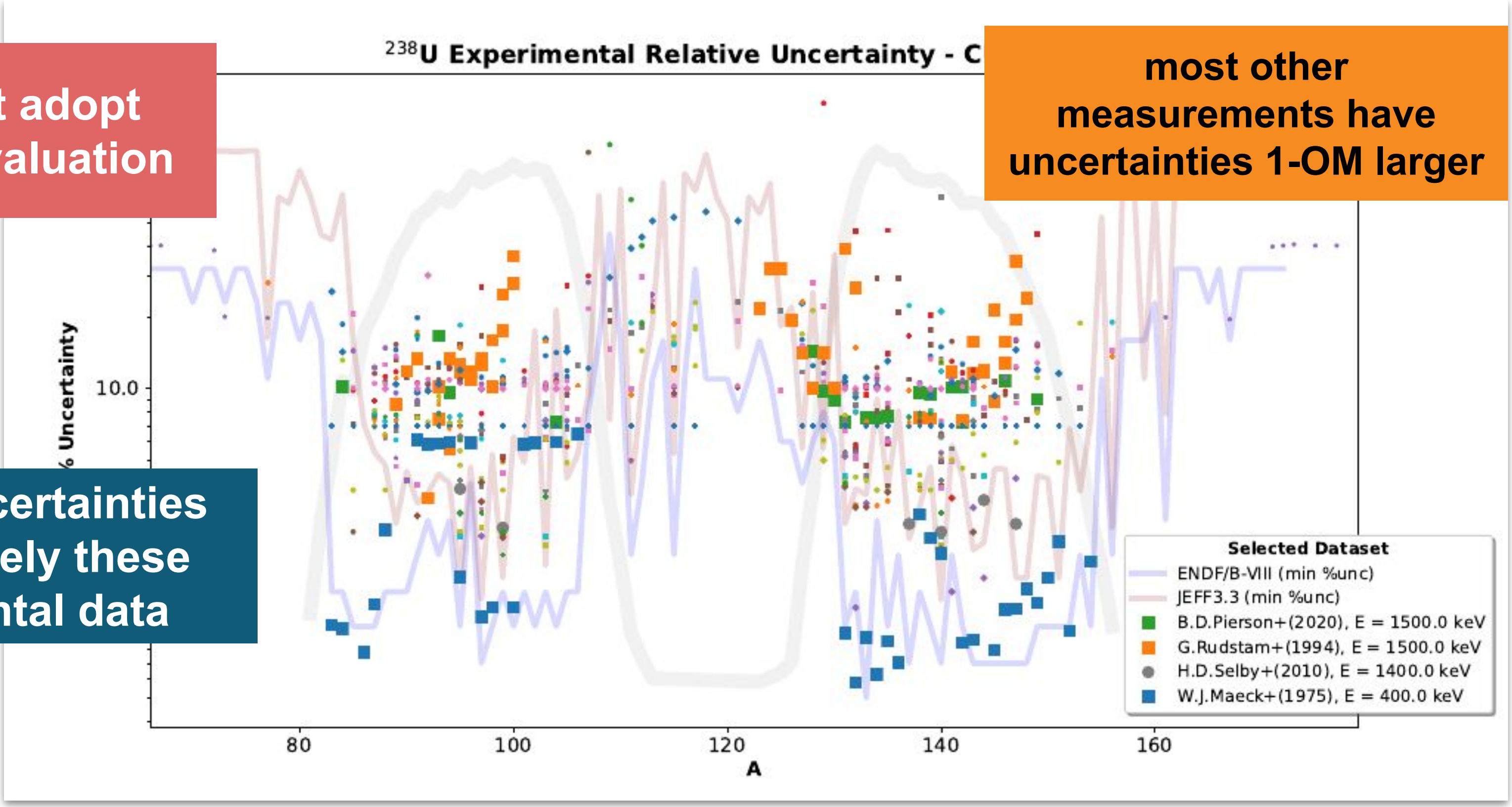

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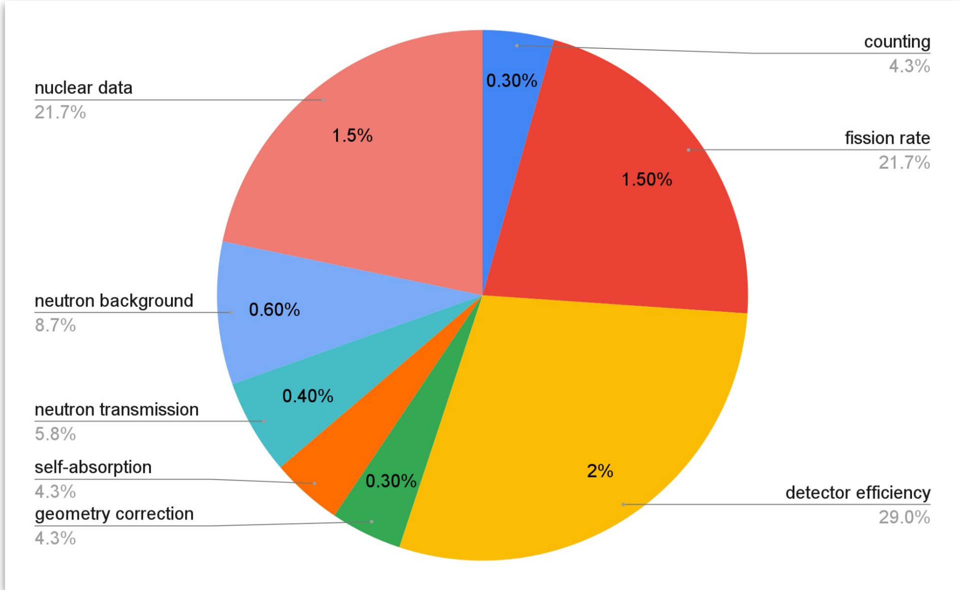
Uncertainty budget

for a typical FY measurement



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and… Nuclear Data uncertainties (can go anywhere from 1 to 10-20%)

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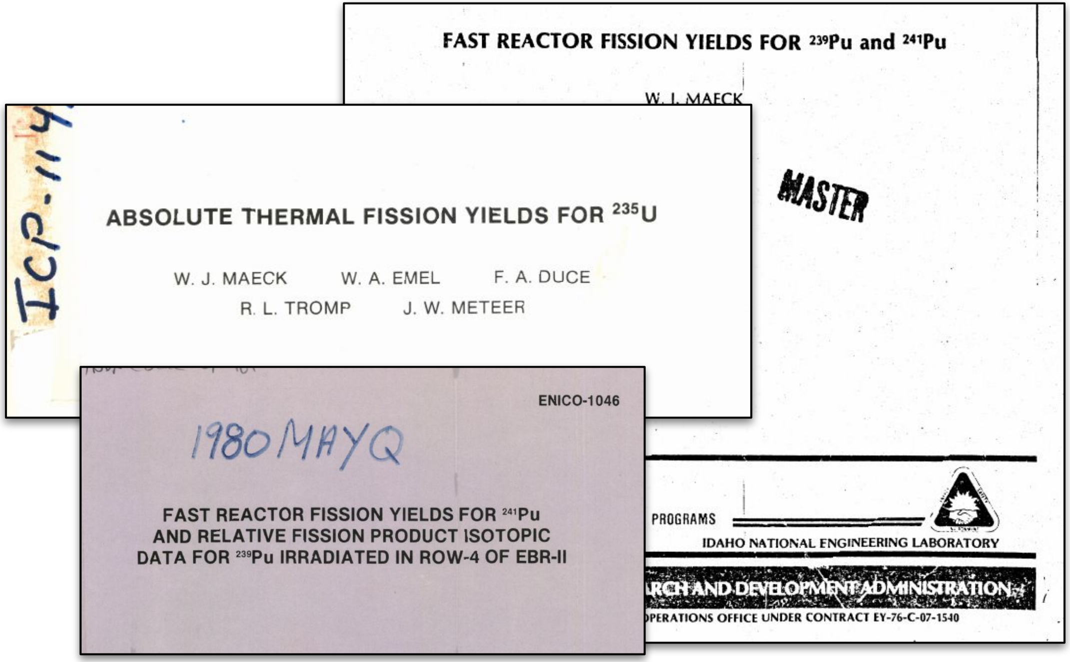
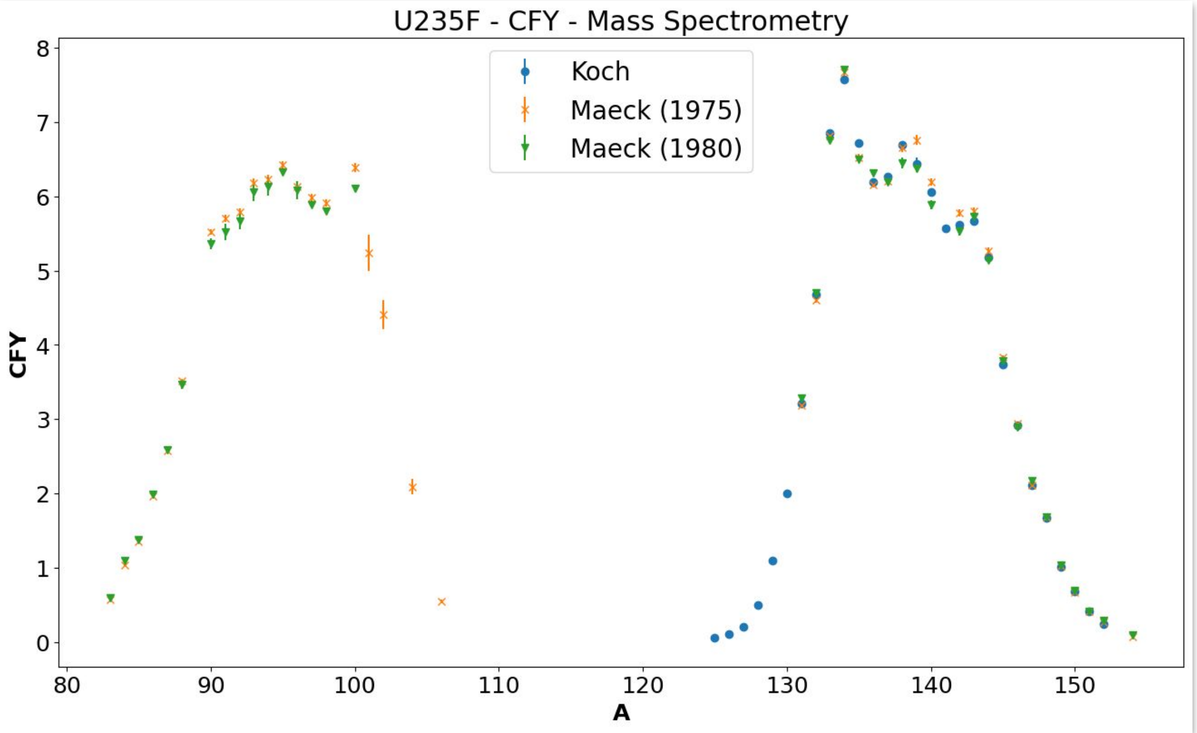
High-precision End-of-chain yields measurement

Results of measurements at Idaho National Lab reactor (ETR and EBR), and only published in a series of INL technical reports

Isotope Dilution Mass Spectrometry on $^{239,241}Pu$ and ^{235}U targets irradiated with thermal & fast neutrons



What can be improved?



Ba and Ce elements could not be measured (by Koch) because of **heavy contamination of natural-occurring isotopes** - and Maeck's results were found to be up to 14% too low in a later measurement by the same group (several times the quoted error).

Maeck also had difficulties measuring **low-yield fission products (< 0.1 %)** and isotopes of Iodine, Tellurium and Ruthenium ~70% of the fission products in the heavy peak could measured by Koch; ~90% by Maeck

Maeck **never published** final results in a peer-reviewed publication

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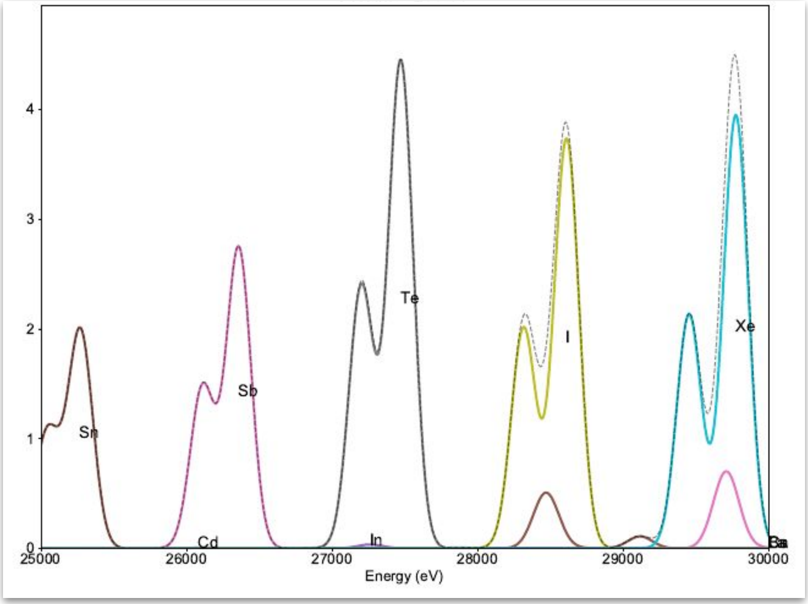
Challenges

activation level / contamination of samples (NSLS-II can measure only low-activity radioactive samples) - it may take too long to irradiate and let samples decay

ability of the X-ray detector to resolve contributions in a complex spectrum (fission yields)

long irradiations → correction for n-capture in fission products



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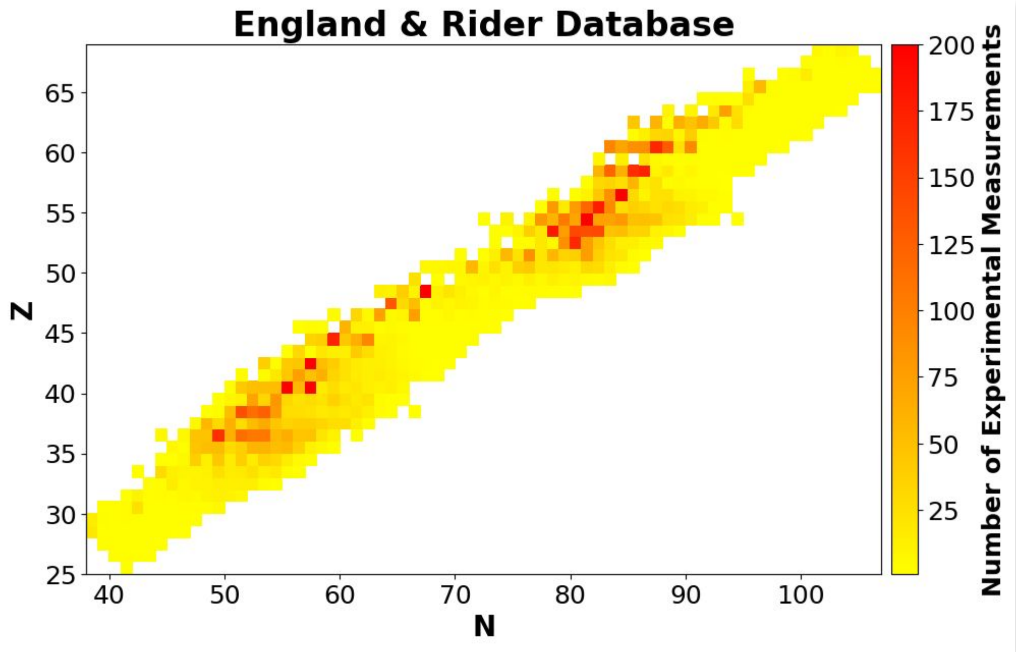
A brief history of Fission Yields evaluation in ENDF

start from **experimental data**, when available…

Very uneven distribution of measurements: some fission products have been measured 250x, others only once or twice and only for some fissioning systems



Number of experimental data points available for: $^{233,235,238}U$, $^{239,241}Pu$, ^{232}Th (thermal, fast, high-energy)

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