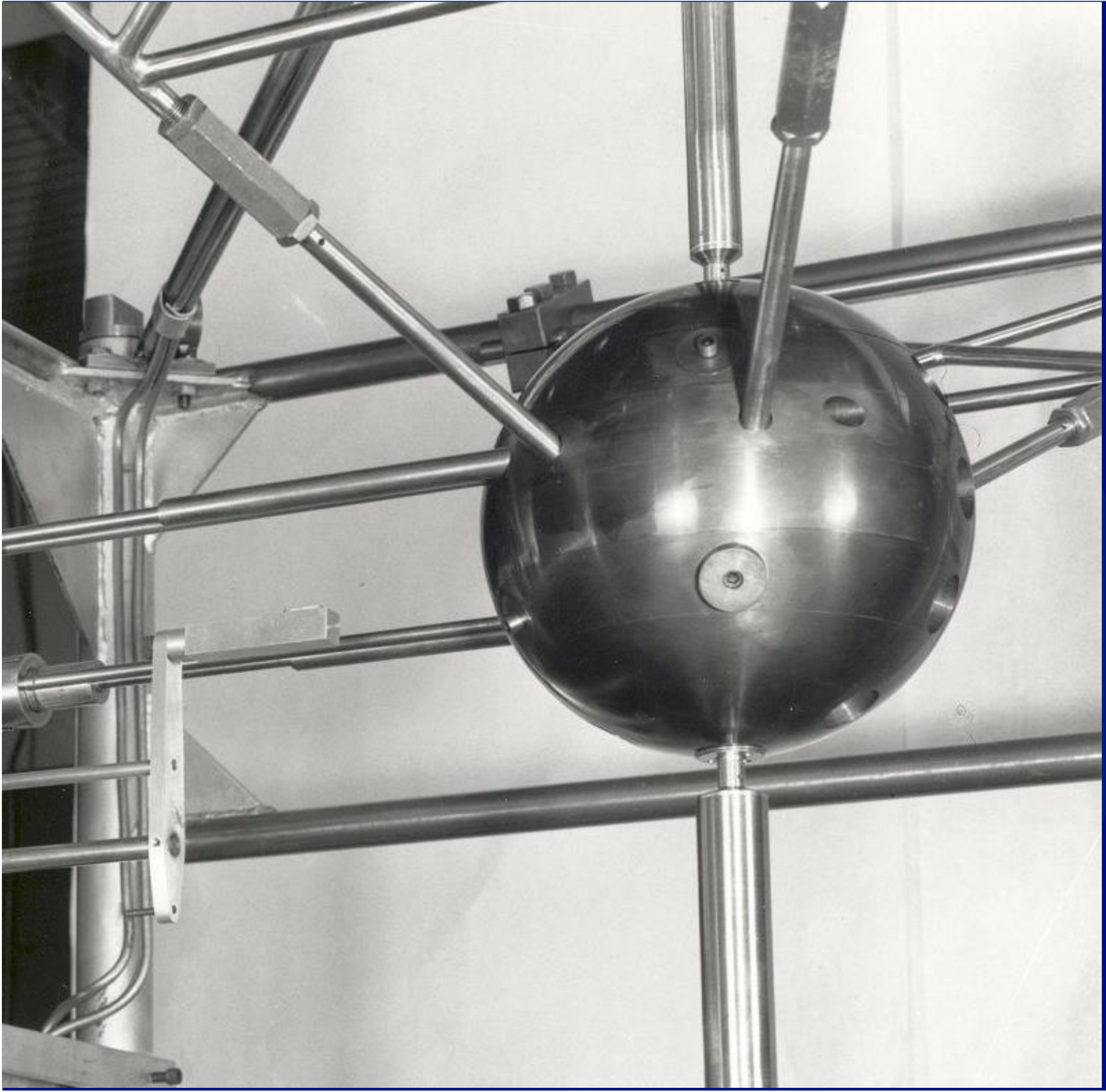

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LEAD ISOTOPE EVALUATIONS

Current progress and limitations

Peter Brain

INDEN Structural Meeting 2024
Vienna, Austria // Hybrid
December 16-22, 2024
LA-UR-24-33187

Outline

- Motivation
- RRR
- Fast Region
- Integral Experiments
- Recommendations for Future Work

Majority of work presented here is encompassed in Thesis (available upon request):
P. Brain, "NEUTRON EVALUATION AND VALIDATION OF NATURAL LEAD ISOTOPES FOR FAST SPECTRUM SYSTEMS", Ph.D. Thesis, MANE, RPI, Troy, NY 2023. Electronic

Motivation

Lead is everywhere

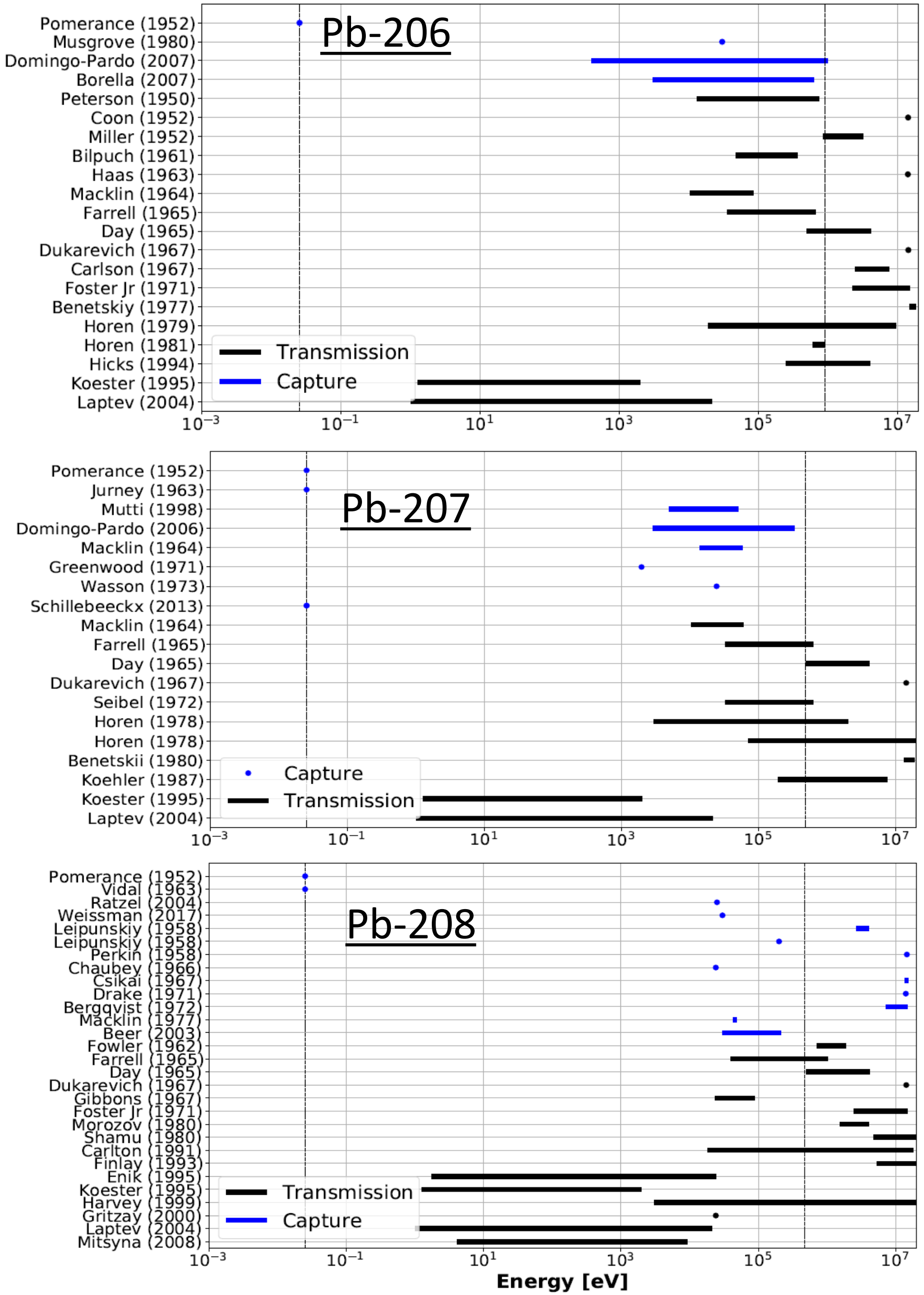
- Work here was sponsored by DOE-NEUP for Lead Fast Reactors
- JAEA – LANL collaboration for Accelerator Driven Systems
- Fusion considering Pb or PbLi in blanket
- Lead is used for shielding gamma in mixed source environments

NEUP Nuclear Energy University Program
U.S. Department of Energy

Pre-validation / Pre-Evaluation

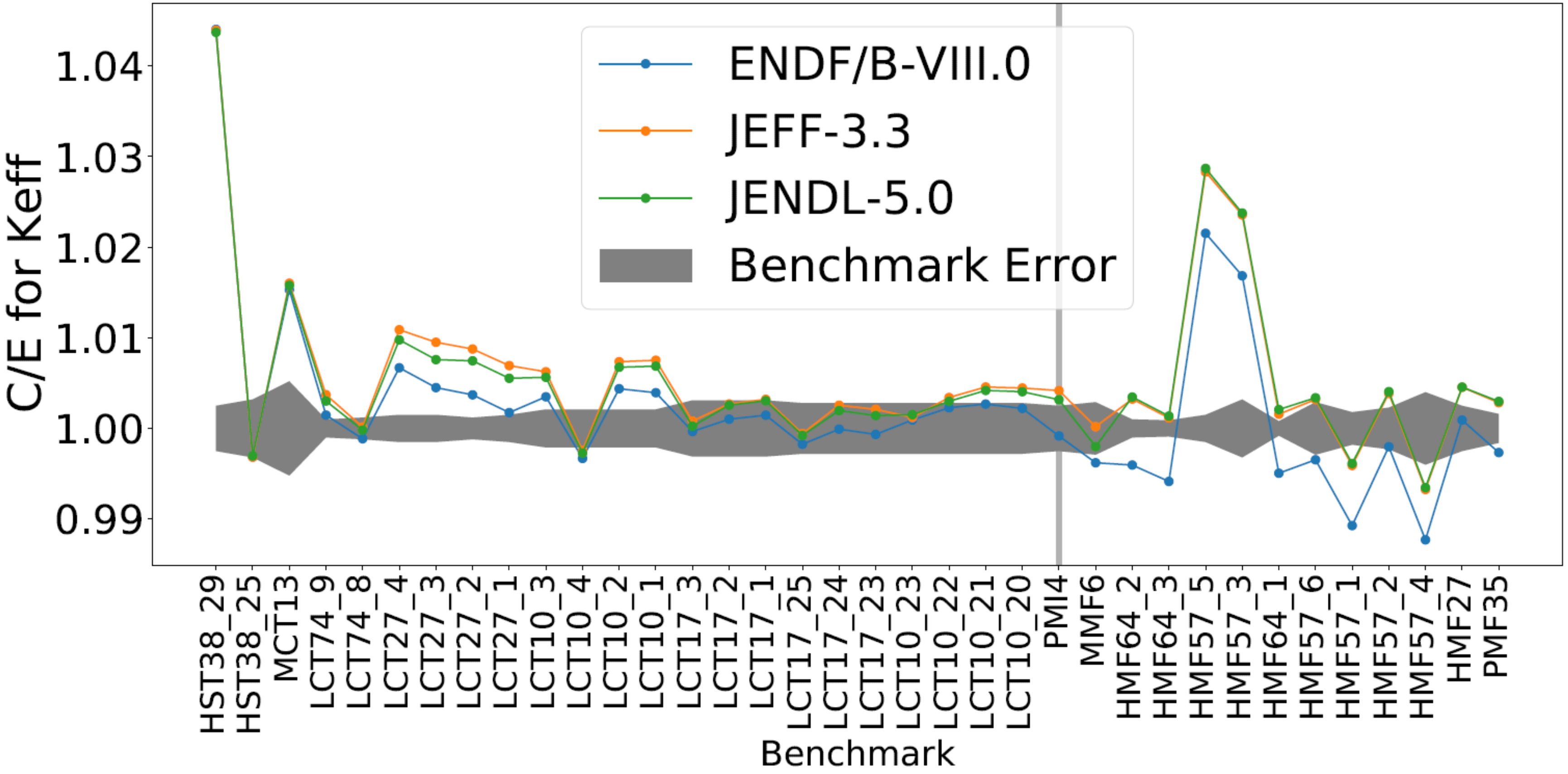
Summary of previous work

- ENDF/B-VIII.0 is a copy of JEFF-3.1
- Differential data exists throughout but the resolution of it varies greatly
  - Main differential data is ORELA (Horen, Harvey, Carlton) *Private Communication*
- No resonances below 3 keV except Pb-204
- Domingo-Pardo and Borella Pb-206/207 capture yields released post JEFF-3.1



Pre-validation / Pre-Evaluation

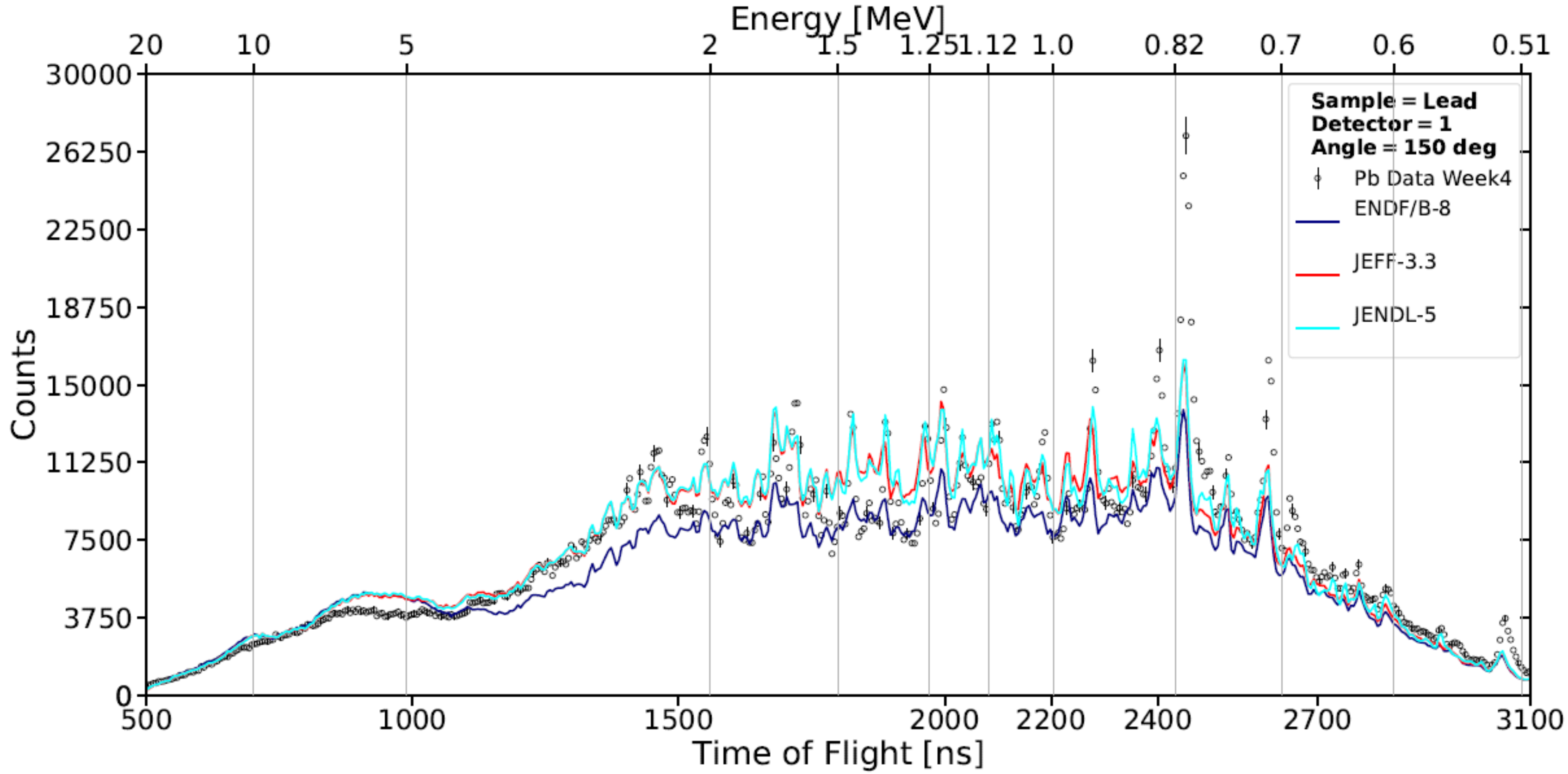
- Prediction of critical assemblies is all over the place, E/B-VIII.0 performing poorly in fast but best in thermal
- Missing intermediate spectra assemblies (this is common)

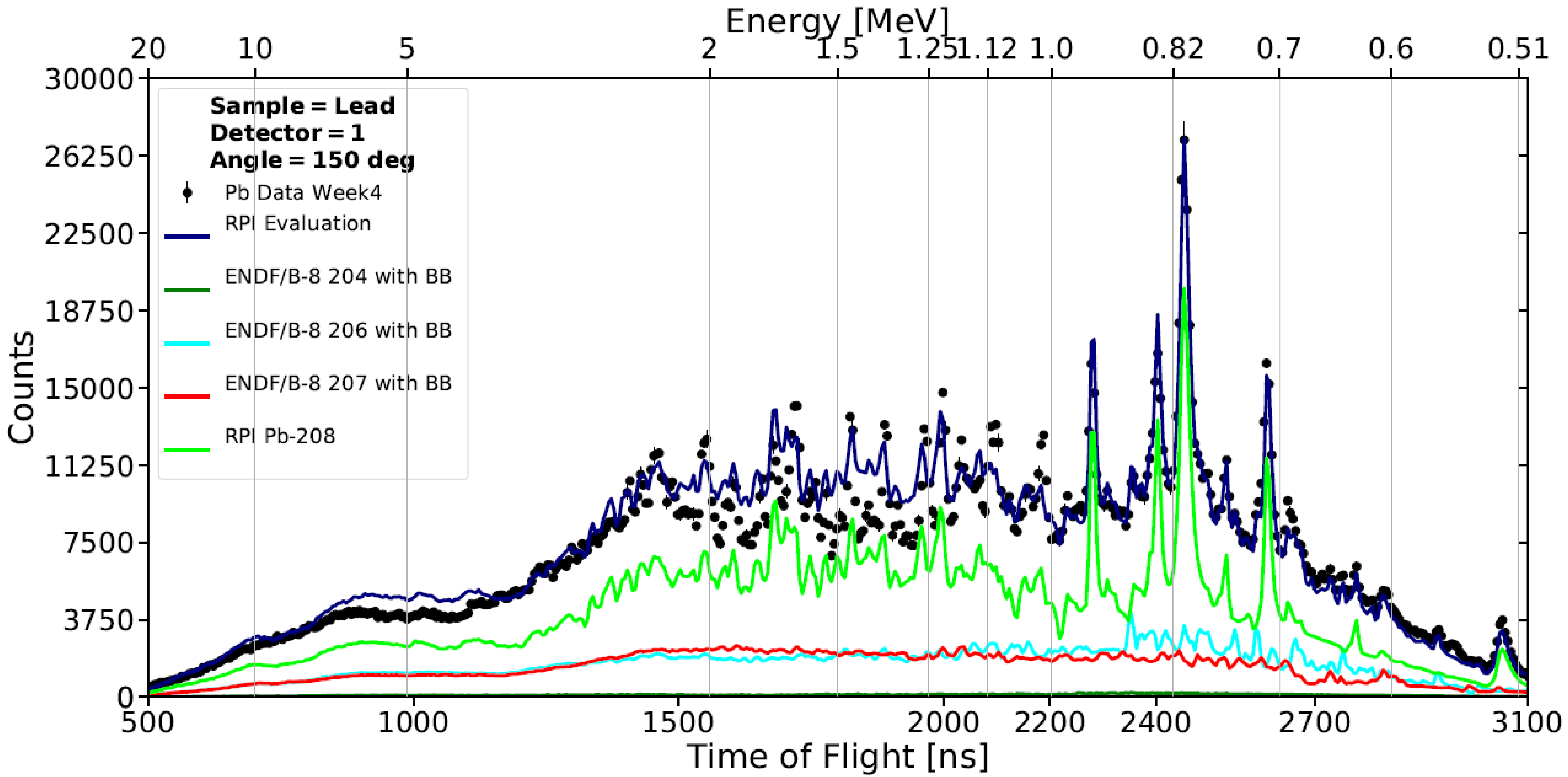


Pre-validation / Pre-Evaluation

Quasi-Differential Scattering

- Natural Pb quasi-differential scattering measurements show that below 2 MeV there is significant structure that is poorly recreated





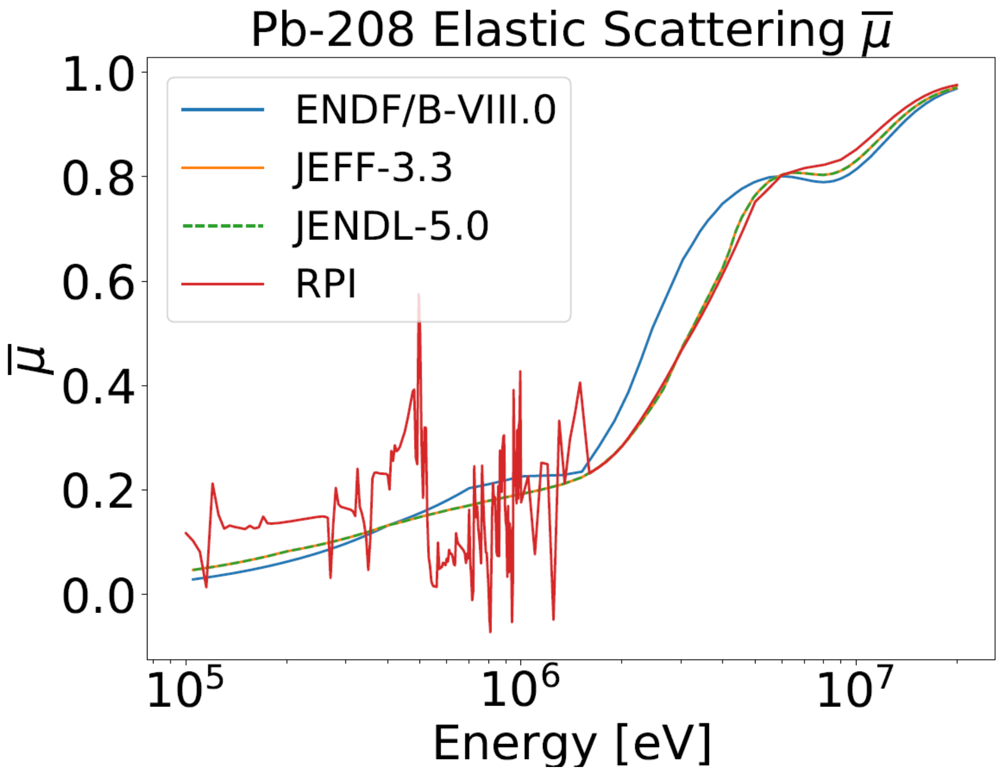
Energy [MeV]
20
10
5
2
1.5
1.25
1.12
1.0
0.82
0.7
0.6
0.51
Sample = Lead
Detector = 1
Angle = 150 deg
Pb Data Week4
RPI Evaluation
ENDF/B-8 204 with BB
ENDF/B-8 206 with BB
ENDF/B-8 207 with BB
RPI Pb-208
Counts
30000
26250
22500
18750
15000
11250
7500
3750
0
500
1000
1500
2000
2200
2400
2700
3100
Time of Flight [ns]

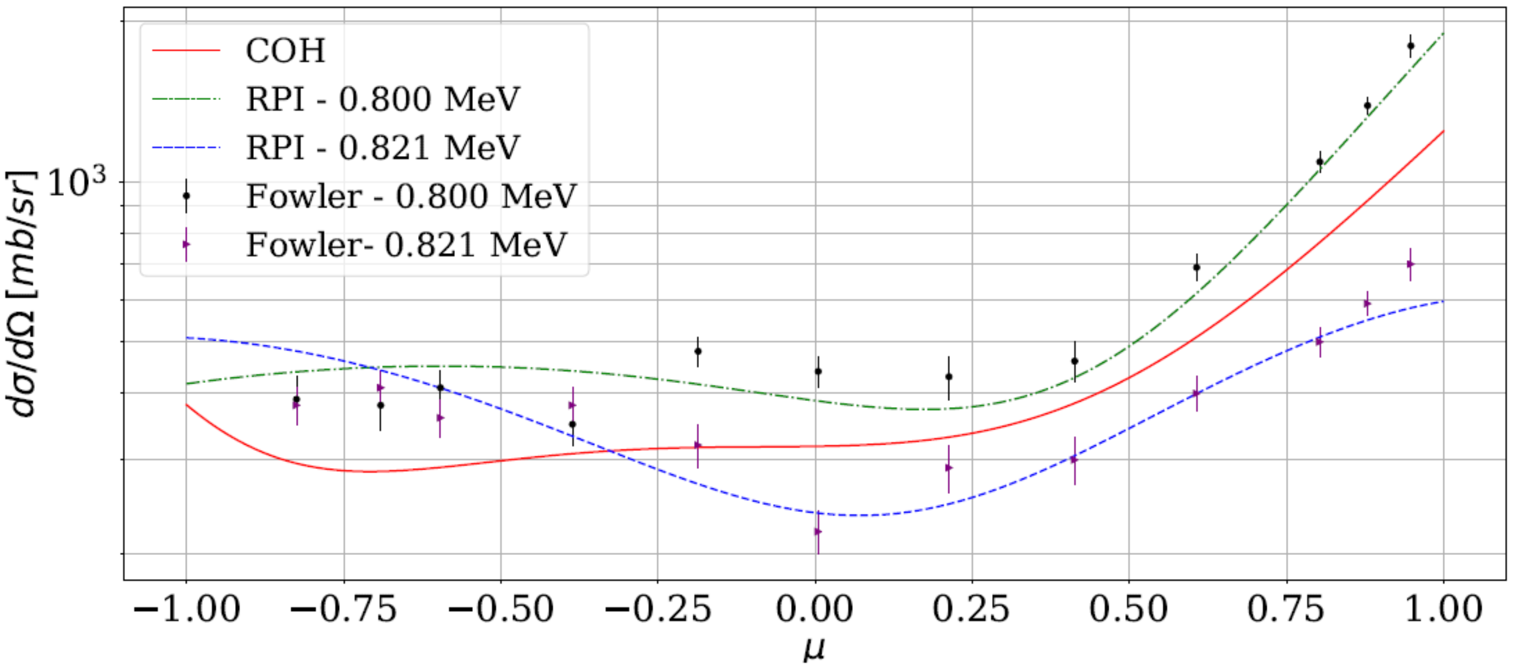
Resolved Resonance Region Evaluation

- Targeted Pb-206, Pb-207, Pb-208
- Adding capture and tweaking inelastic contribution for Pb-206/207
- **Resolving an additional 500 keV of resonances for Pb-208 using novel methodology with Quasi-Diff Data and transmission**
- Reconstructing the whole RRR with Blatt-Biedenharn

Impact of using BB compared to OMP

Resonance derived scattering distributions is more physically accurate from theory and experiments

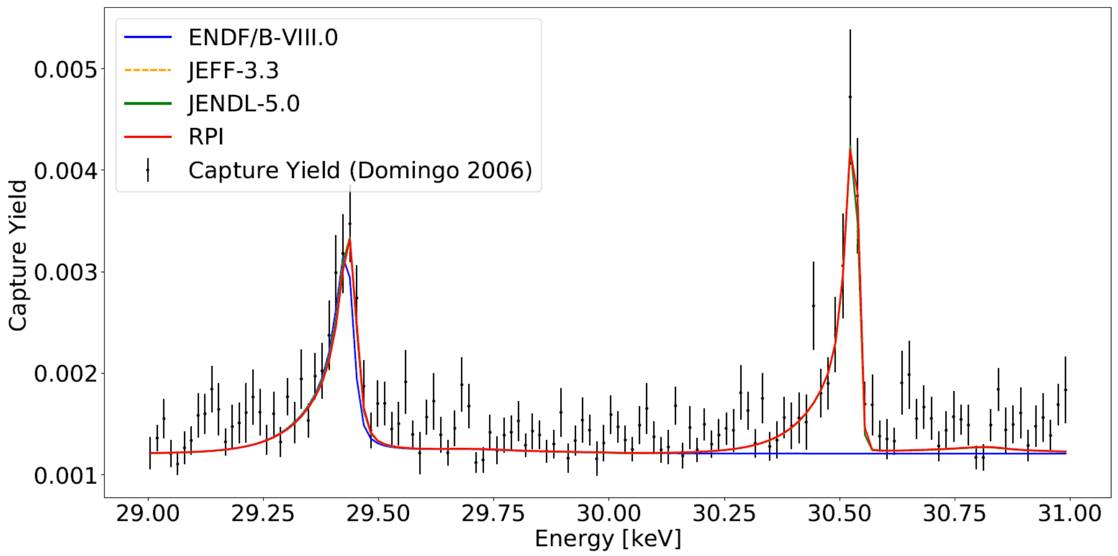


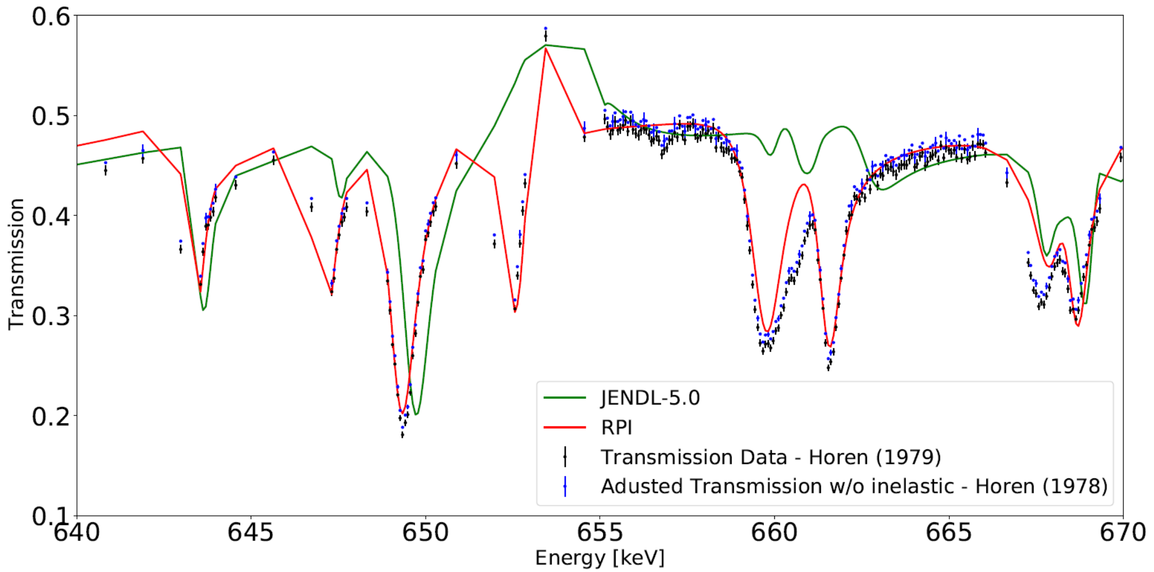


Pb-207 RRR

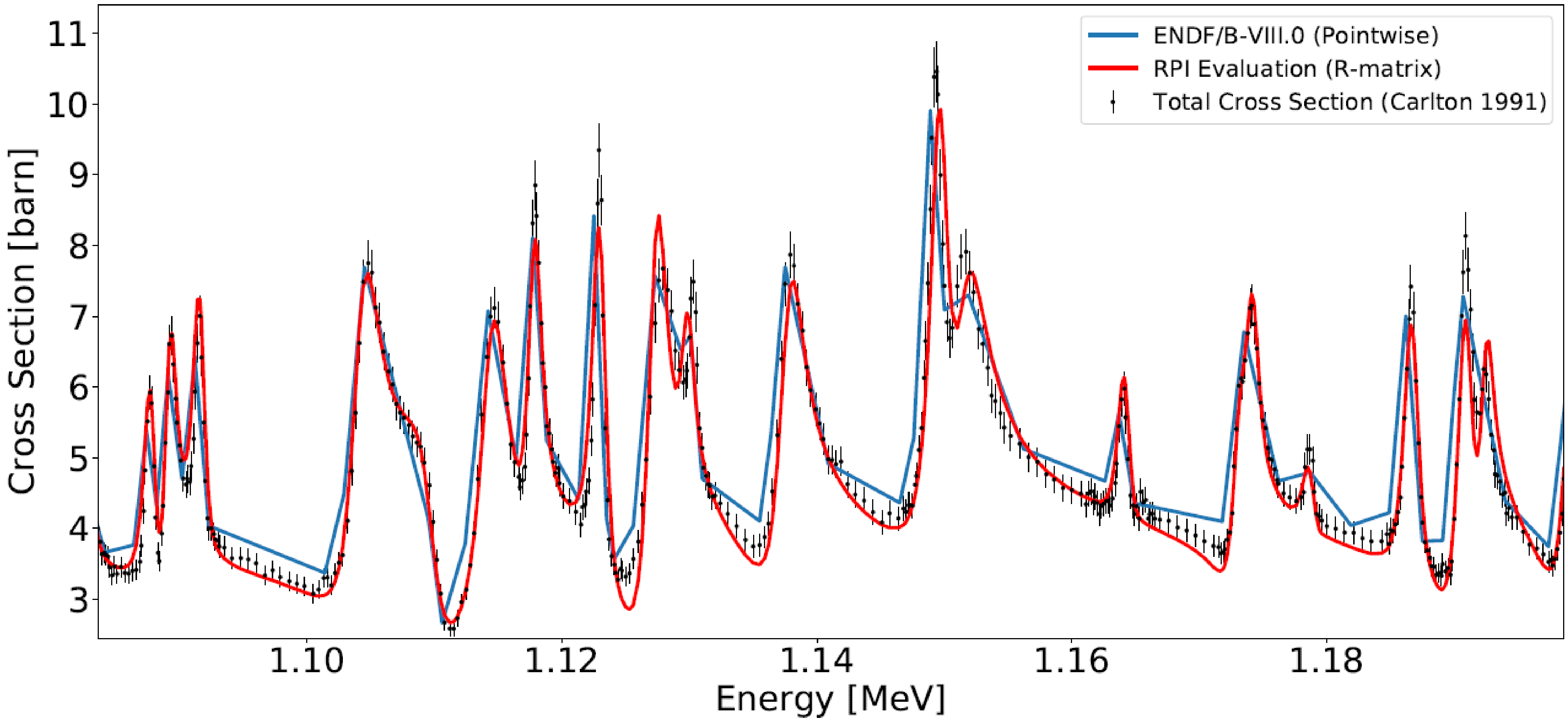
Similar work for Pb-206

- Adopted JEFF-3.3 RRR parameters ($E_{lim} = 675\text{keV}$) and adjusted them based on CoH-3 inelastic calculations
- Reworked bound s-wave resonance to be consistent with thermal scattering lengths and decay gammas





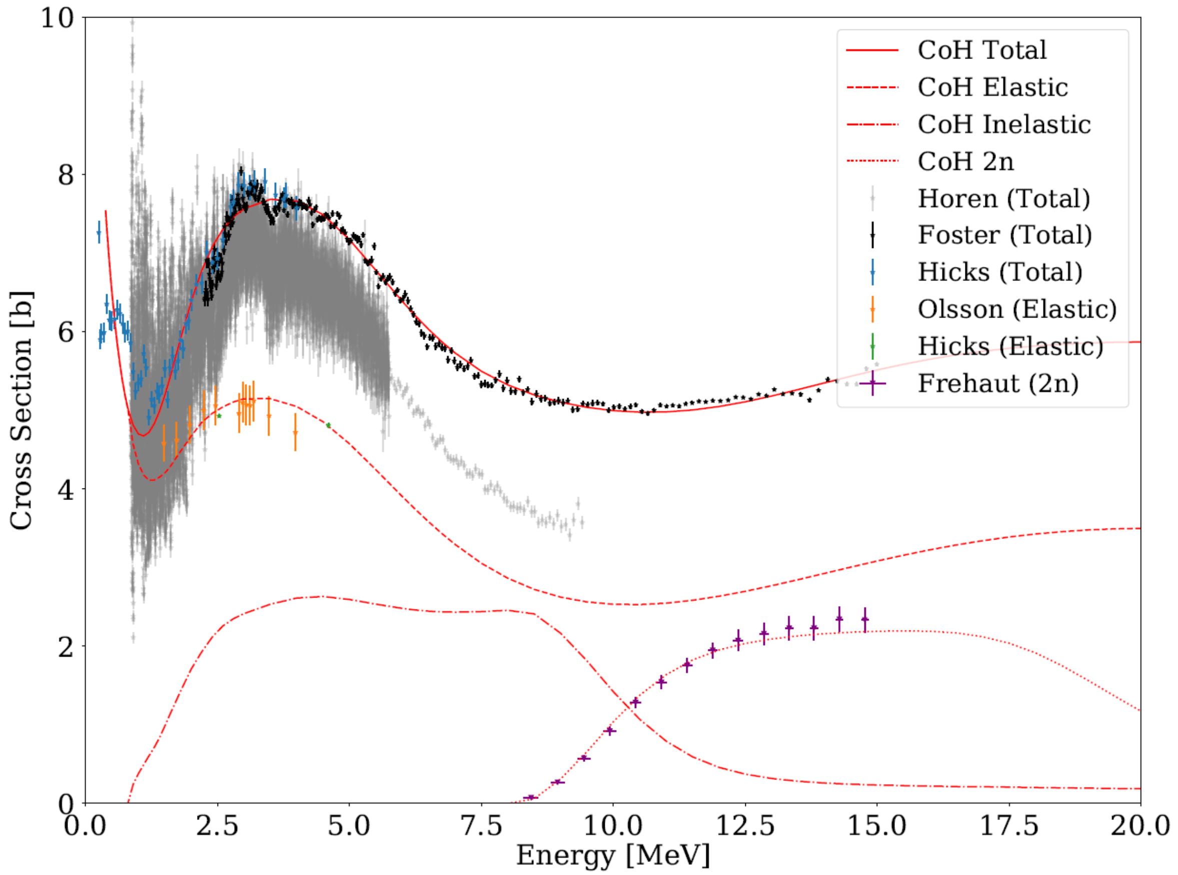
Pb-208 total cross section with new RRR



Fast Region Evaluation

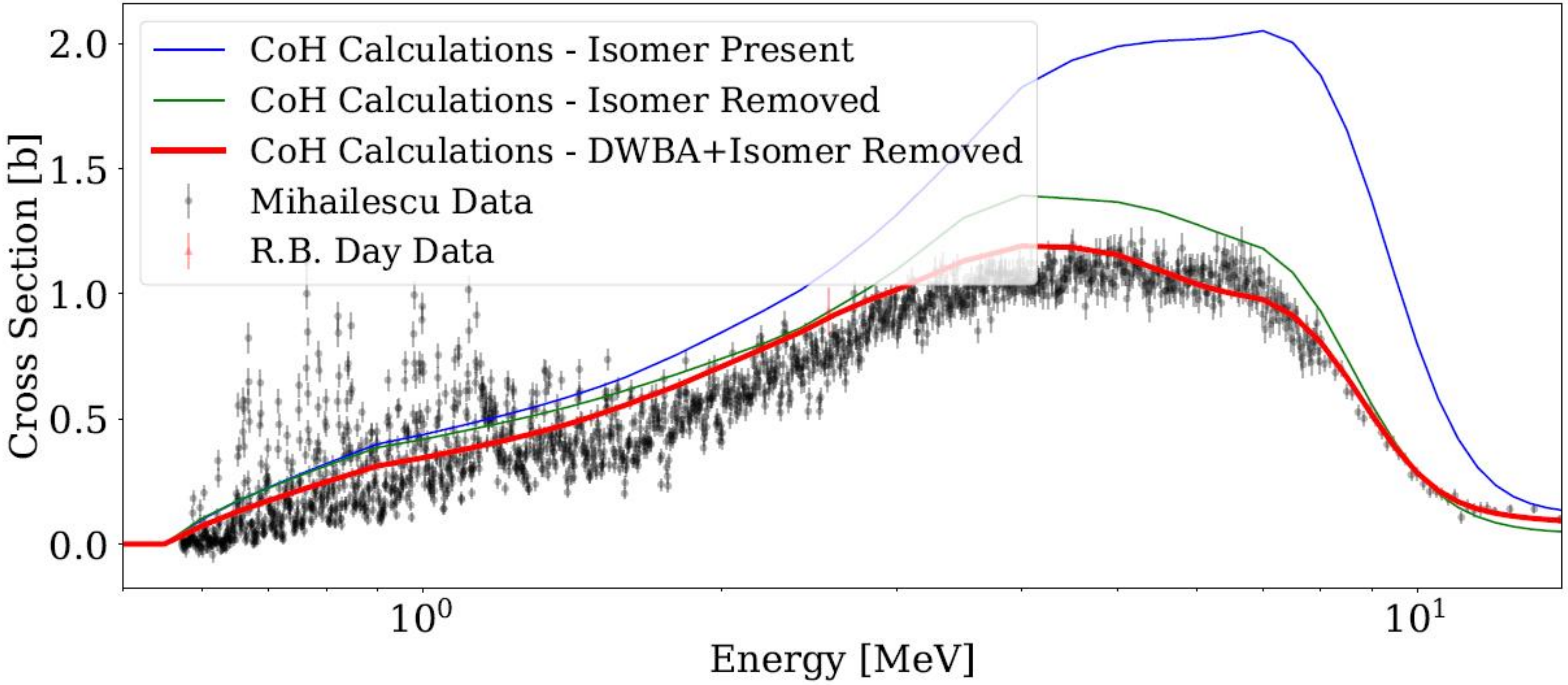
Limited cross section data after 4 MeV

- Elastic, inelastic, 2n, and total of Pb-206 and Pb-207 are major channels
- CoH used for Hauser-Feshbach calculations with vibrational deformation and coupled-channels
- Good reproduction of averaged cross sections



Fast Region Evaluation

Pb-207 inelastic cross section can be accounted for



Summary of Evaluation

New cross sections and covariances

- 208Pb
  - Full RRR evaluation and RRR extension up to 1.5 MeV using SAMMY.
  - Adoption of Kawano evaluation for En ≥ 2.5 MeV, between 1.5 and 2.5 MeV total cross section interpolated again.
  - Full covariance below 20 MeV.
- 207Pb
  - Full RRR evaluation up to 0.675 MeV using SAMMY.
  - Full fast region evaluation up to 20 MeV using CoH-3. ESAD (MF=4, MT=2) taken from ENDF/B-VIII.0 from 0.7 to 2 MeV.
  - Full covariance below 20 MeV.
- 206Pb
  - Full RRR evaluation up to 0.900 MeV using SAMMY.
  - Full fast region evaluation up to 20 MeV in CoH-3. ESAD (MF=4, MT=2) taken from BROND-3.1 from 0.5 to 3.0 MeV.
  - Full covariance below 20 MeV.

Status of Lead Containing Integral Experiments

"Benchmark" does not mean "Trust Blindly"!

- The purpose of integral experiments changed overtime from criticality limits to nuclear data validation → Evaluation methodology changed!
- These two are not equivalent and leads to trouble!

Benchmark	*Page Count*	*Year*	*# Configs*	*# Pb Configs*	*Notes*
PMF-35	21	1965	1	1	Extrapolation to k_{eff}
HMF-27	22	1962	1	1	Extrapolation to k_{eff}
HMF-57	37	1958	6	6	Extrapolation to k_{eff}
HMF-64	34	1991	3	3	Extrapolation to k_{eff}
MMF-06	100	1991	3	3	
PMI-04	60	1969	1	1	
LCT-10	67	1970-80	30	5	$U(4.31)O_2$
LCT-17	82	1970-80	30	5	$U(2.53)O_2$
LCT-27	101	1983	4	4	
LCT-74	444	2008	30	2	
MCT-13	85	1976	31	5	MOX/Plexiglass fuel blocks
HST-38	91	1988	30	2	Cases 29/30 calculate poorly

HMF-57

Livermore Critical Mass Lab done in early 1960s

- 2 spheres and 4 cylinders with varying amounts of Pb reflection
  - Highest multiplications were <= 100
- The two cylinder cases which calculate way-off have more lead reflection
- ICSBEP write-up says that a trend can be observed with Keff vs. SA of U/Pb

HMF-57

Livermore Critical Mass Lab done in early 1960s

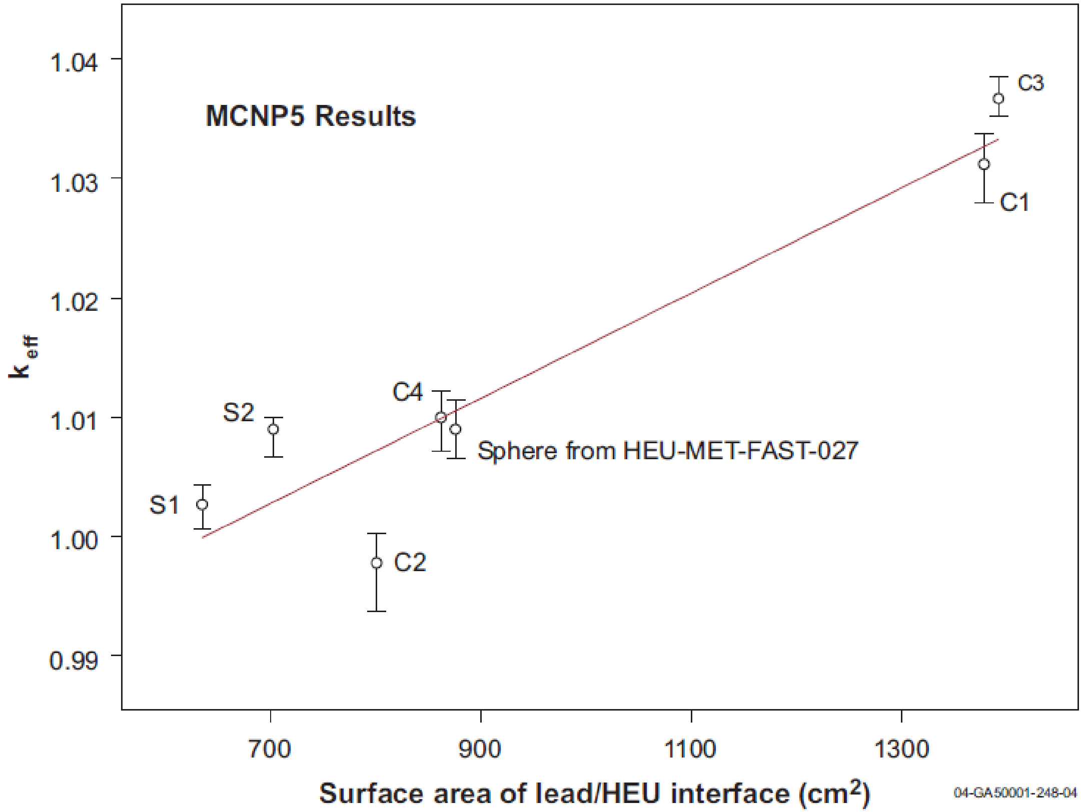
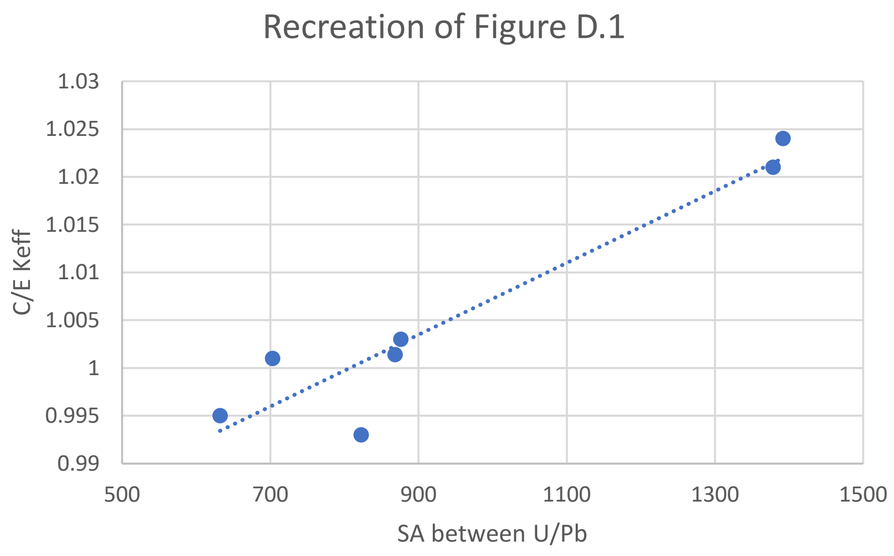


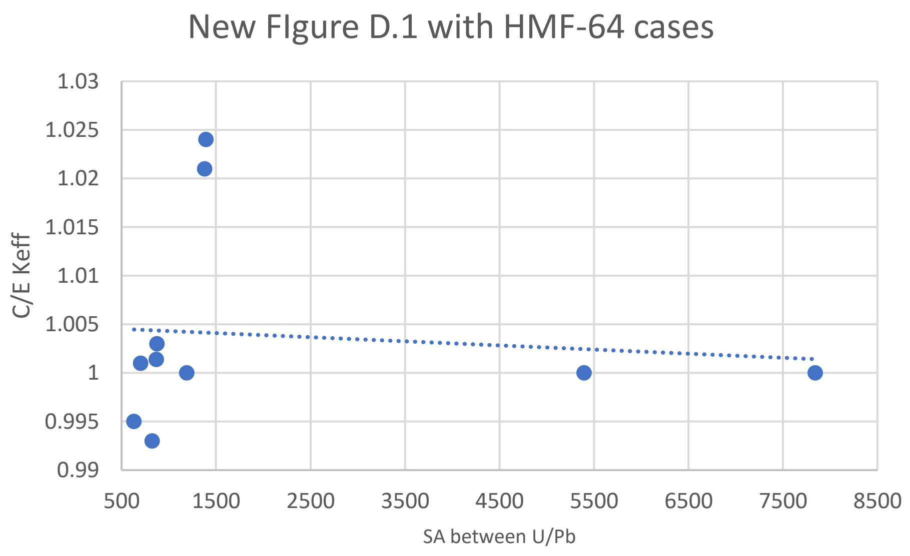
Figure D.1. A Comparison of Calculated k_{eff} as a Function of HEU/Lead Surface Area.

HMF-57

Recreating Figure D.1

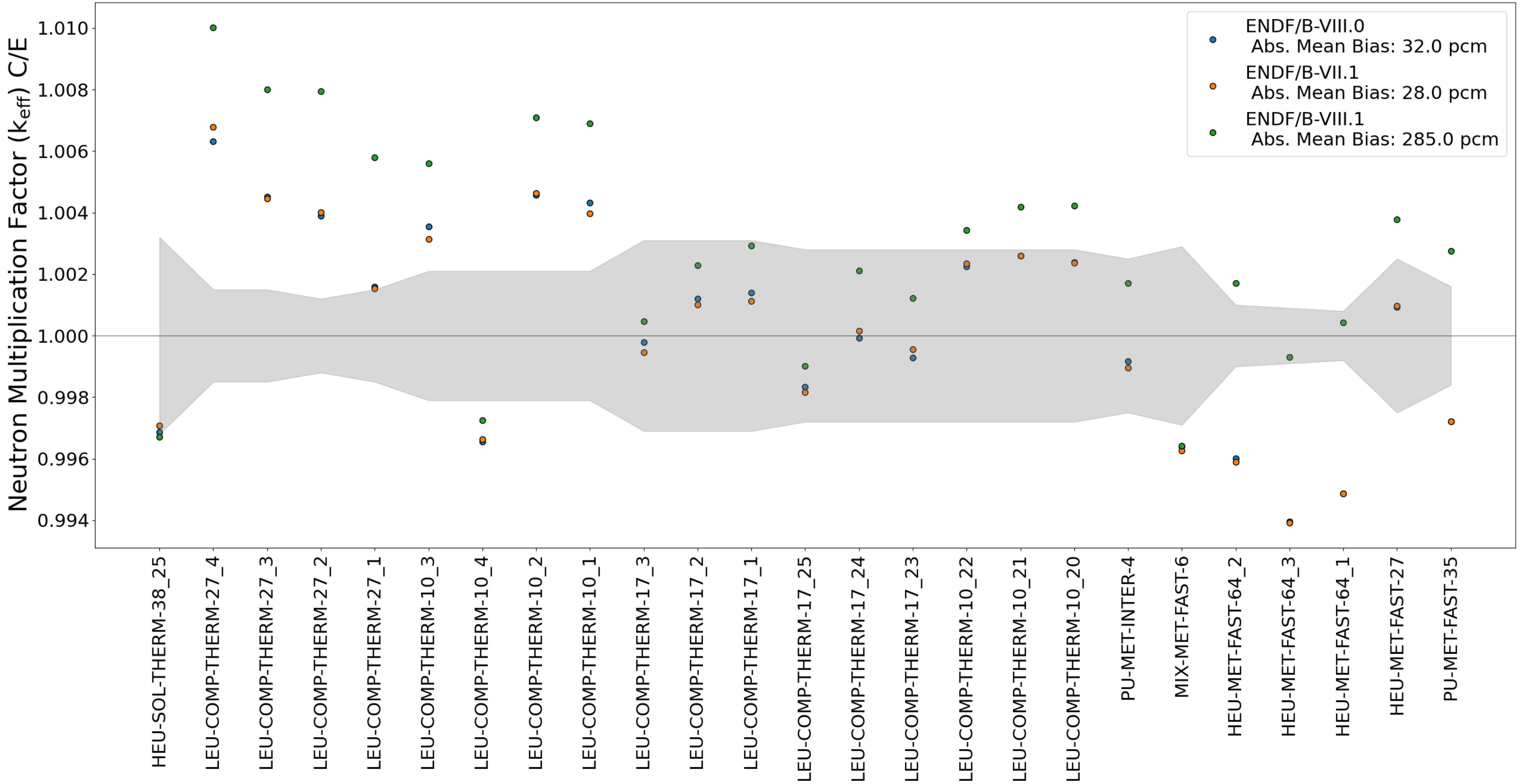
- After inclusion of HMF-64, the relationship breaks down





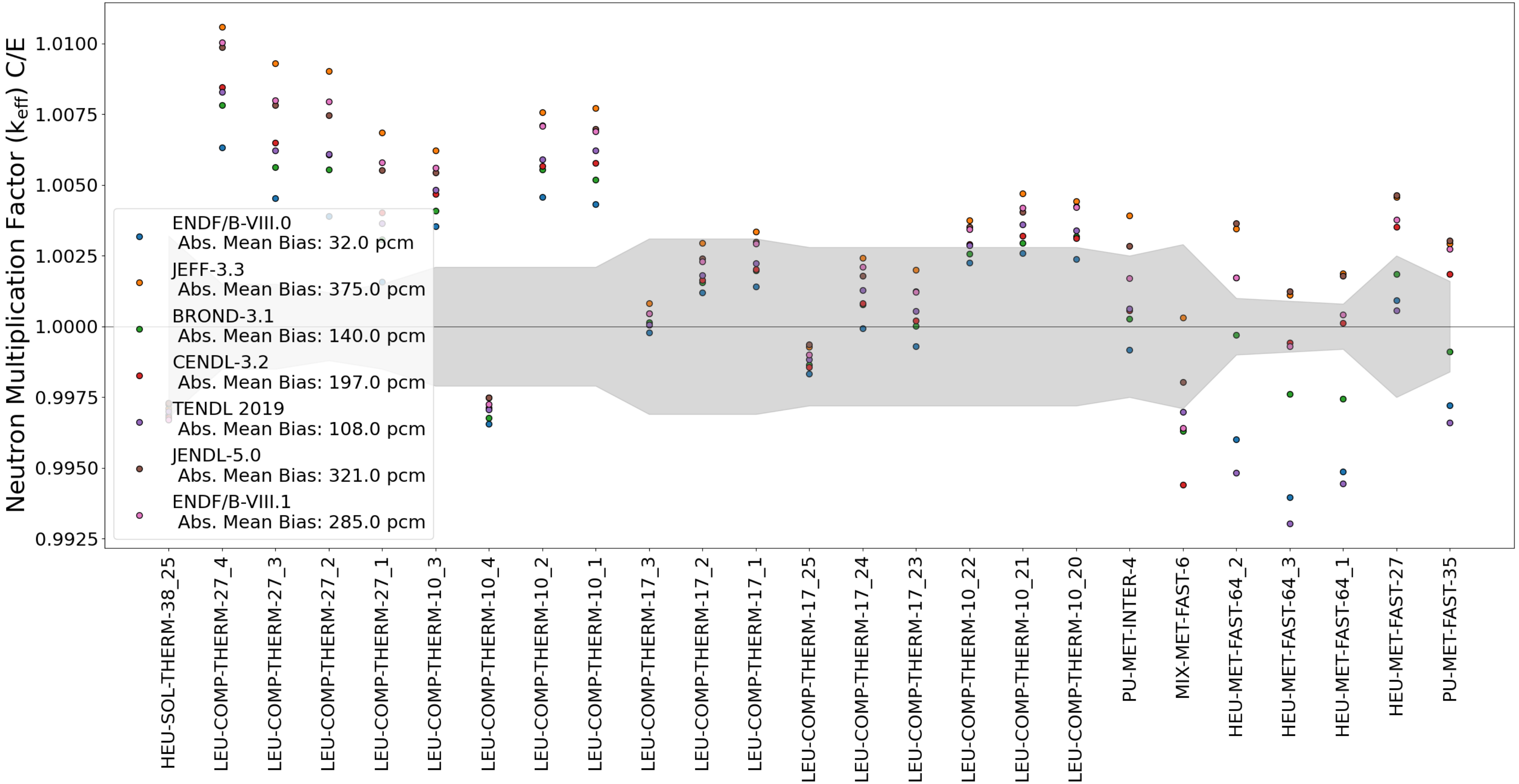
Simulating Pb Benchmarks with Various Evaluations

ENDF/B-VIII.1 only has major change from changing elastic scattering



Simulating Pb Benchmarks with Various Evaluations

All International – No one performs good in thermal and fast



Future Work

New data is needed for Pb evaluation to make progress

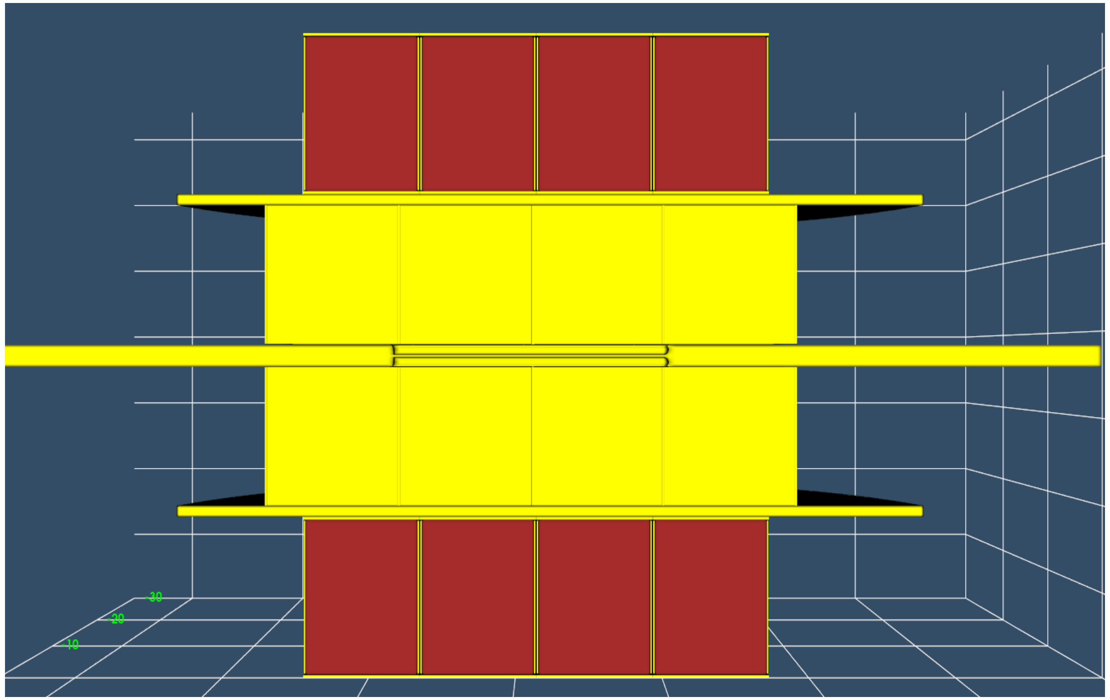
- Areas for refining cross sections for fission and fusion applications are below

Resolution	**Measurements**	**Sample**	**Energy**
Differential	Transmission	^{206,208}Pb	1 - 4 MeV
	Capture Yield	^{206,207,208,nat}Pb	$E_n \geq 1$ MeV
Quasi-Differential			
	High Energy Scattering	^{206rad,208rad}Pb[2]	0.5 - 20 MeV
	Mid Energy Scattering	^{206rad,208rad,nat}Pb	3 keV - 1 MeV
	Pulsed Leakage Experiment	^{nat}Pb	DT or D-D 14 MeV / 2.2 MeV
Integral Critical			
	Fast reflected ^{235}U/^{239}Pu	^{nat}Pb	0.1 - 5 MeV
	Thermal reflected LEU	^{nat}Pb	$E_n \leq 5$ MeV
	Sub-critical Measurements with BeRP [167]	^{nat}Pb	0.1 - 5 MeV

Example of New Integral Experiments

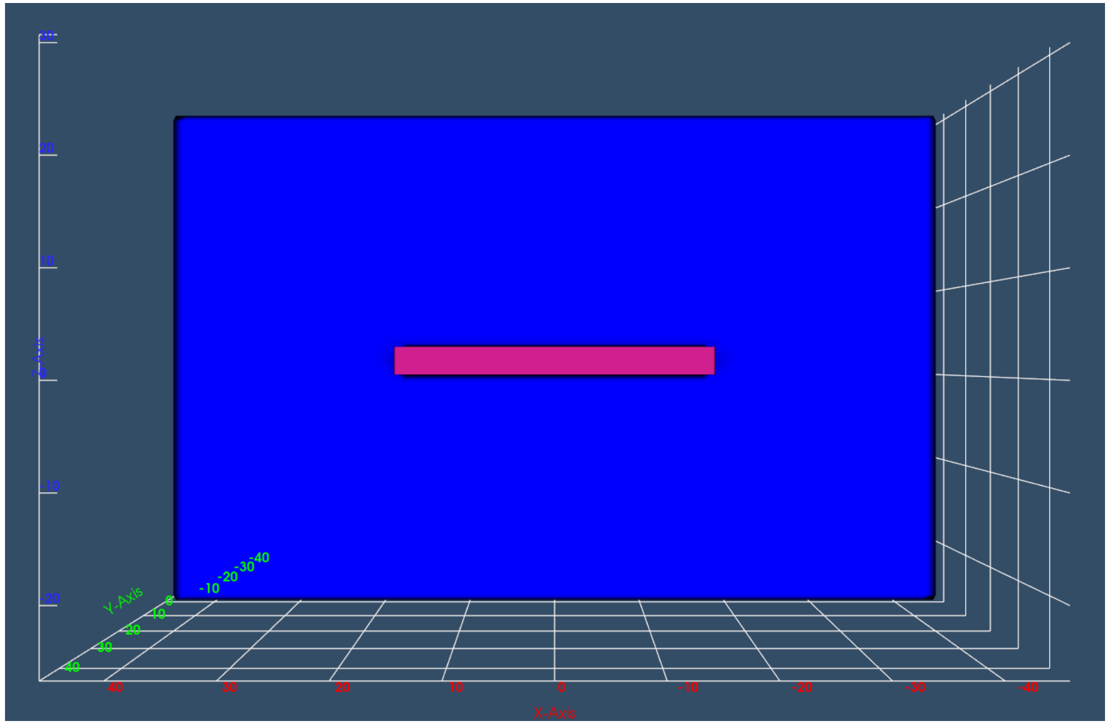
Thales with Pb

- ΔKeff from E/B-8.0 → 8.1 = 750 pcm



Remaking HMF-57

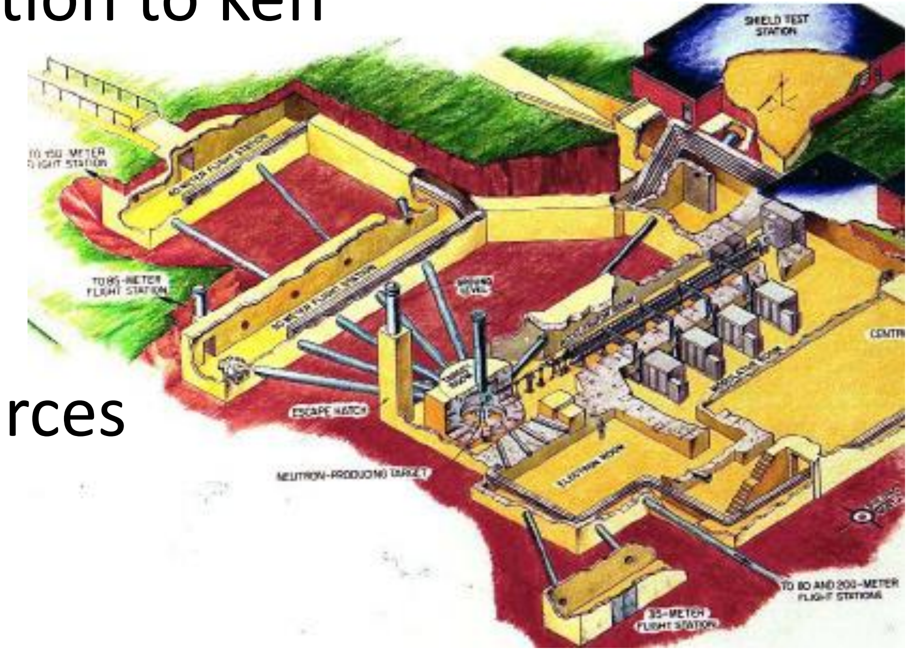
- ΔKeff from E/B-8.0 → 8.1 = 400 pcm



Where are We Going?

Structural materials need to escape fission interactions

- Leverage secondary integral responses in addition to keff
    - Subcritical Noise, Leakage, Activation Foils
- Thick sample differential measurements
- Quasi-differential scattering/emission
- Leakage from Cf-252, D-D, or D-T shielded sources



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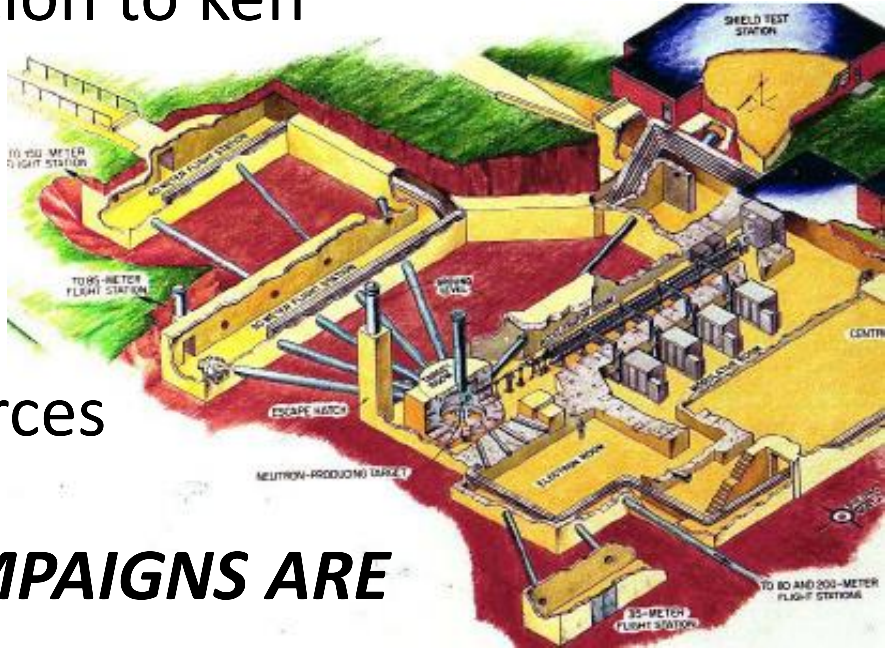
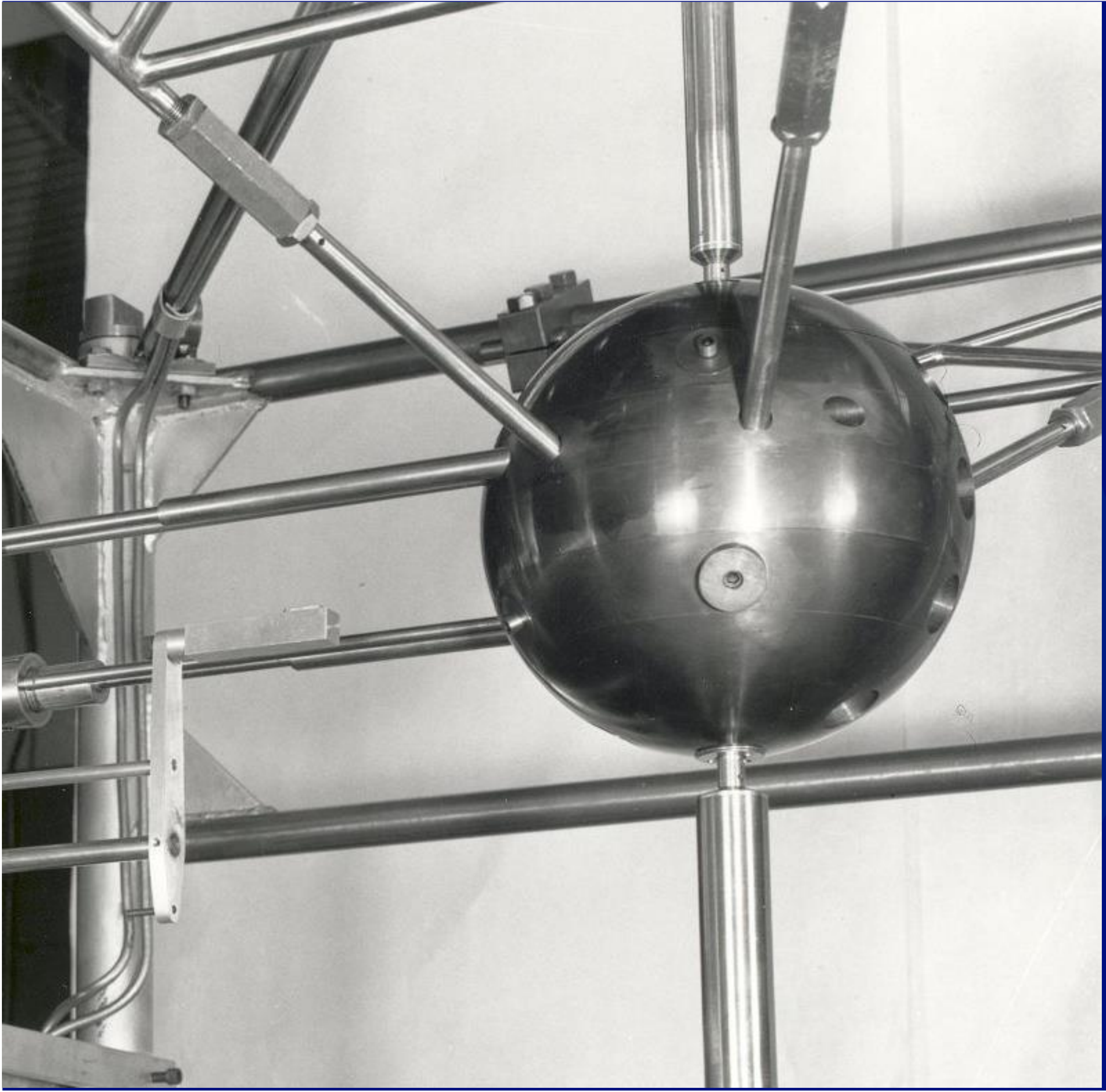


PERIODIC TABLE WORTH OF CAMPAIGNS ARE NEEDED



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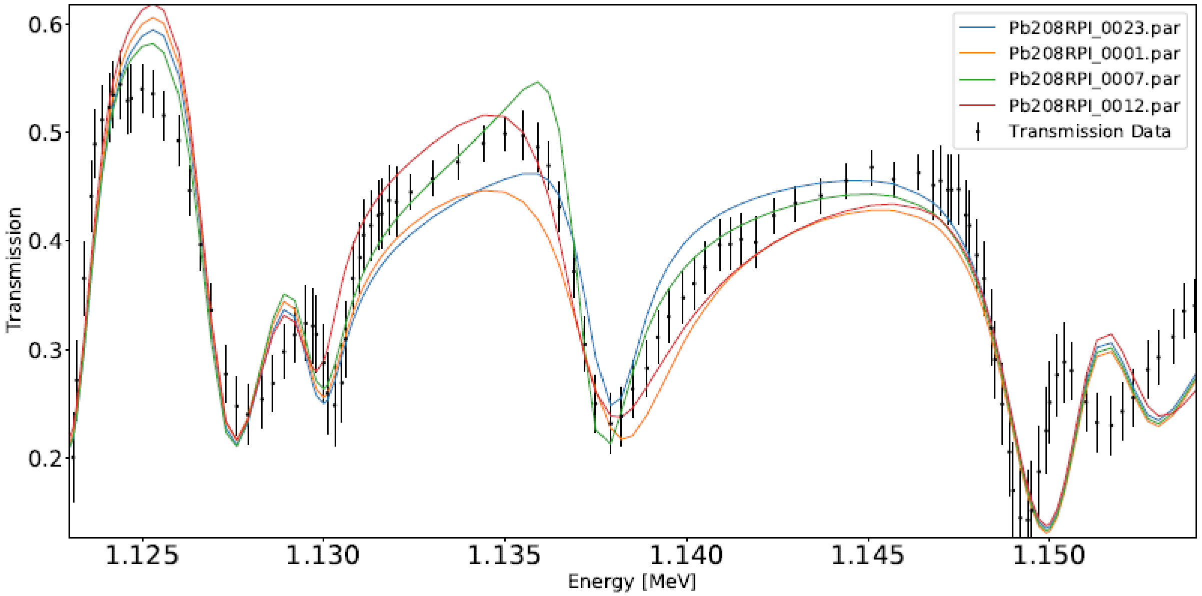
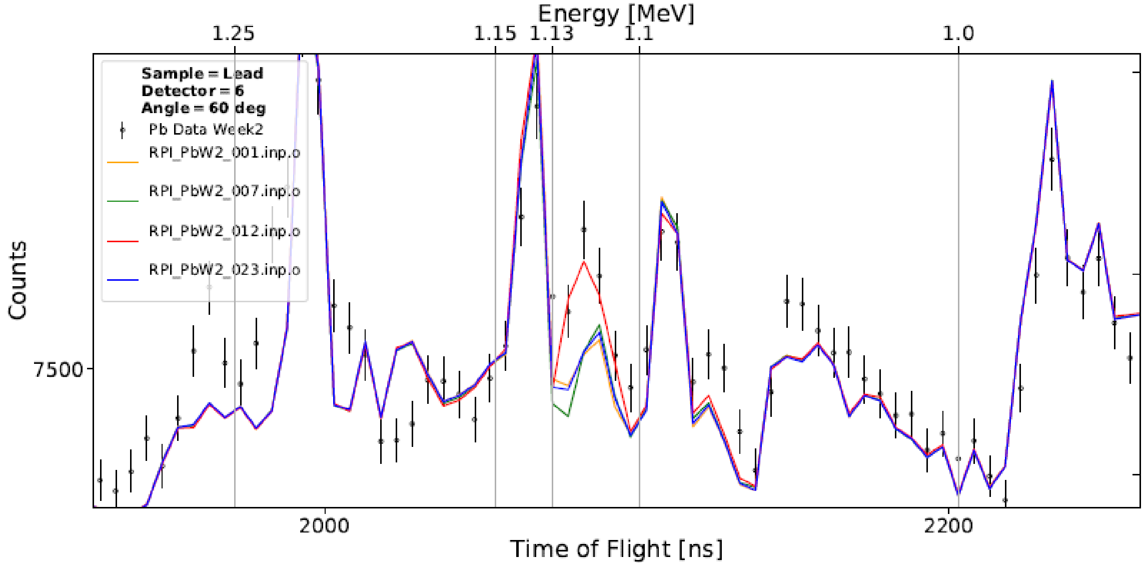
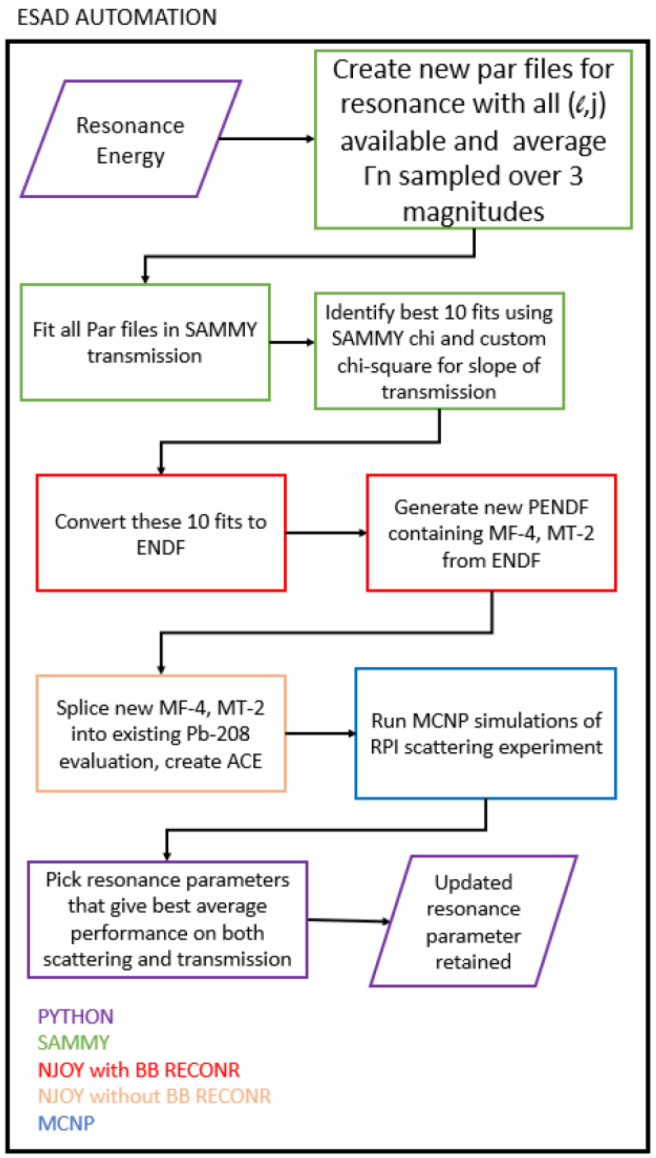
ESAD Automation Method







HST-38

WINCO Tank experiment from 1980s

- Two cases containing Pb, one calculates fine while other is 4000pcm off
- Upon further inspection the last two cases in the experiment campaign calculate uncharacteristically high
- Looked into similar configurations of material and logbooks to see what is the cause

Table 45. Sample Calculation Results (United States).

Code (Cross Section Set) → Case Number ↓	KENO (44-Group ENDF/B-V)	MCNP (Continuous Energy ENDF/B-V)	COG[a] (Continuous Energy ENDF/B-VI.7)
Case 1	1.0009 ± 0.0004	1.0010 ± 0.0004	1.0018 ± 0.0006
Case 2	0.9998 ± 0.0004	0.9999 ± 0.0004	0.9960 ± 0.0006
Case 3	1.0015 ± 0.0004	1.0019 ± 0.0004	0.9999 ± 0.0006
Case 4	1.0014 ± 0.0004	1.0022 ± 0.0004	1.0020 ± 0.0006
Case 5	1.0015 ± 0.0004	1.0022 ± 0.0004	1.0016 ± 0.0006
Case 6	1.0006 ± 0.0004	0.9999 ± 0.0004	1.0016 ± 0.0006
Case 7	1.0024 ± 0.0004	1.0019 ± 0.0004	1.0021 ± 0.0006
Case 8	1.0023 ± 0.0004	1.0023 ± 0.0004	1.0003 ± 0.0006
Case 9	1.0020 ± 0.0004	1.0010 ± 0.0004	1.0001 ± 0.0006
Case 10	1.0016 ± 0.0004	1.0015 ± 0.0004	1.0016 ± 0.0006
Case 11	1.0019 ± 0.0004	1.0026 ± 0.0004	1.0034 ± 0.0006
Case 12	1.0013± 0.0004	1.0020 ± 0.0004	1.0018 ± 0.0006
Case 13	1.0069 ± 0.0004	1.0077 ± 0.0004	1.0071 ± 0.0006
Case 14	1.0067 ± 0.0004	1.0070 ± 0.0004	1.0087 ± 0.0006
Case 15	1.0063 ± 0.0004	1.0072 ± 0.0004	1.0075 ± 0.0006
Case 16	1.0046 ± 0.0004	1.0066 ± 0.0004	1.0070 ± 0.0007
Case 17	1.0019 ± 0.0004	1.0022 ± 0.0004	1.0021 ± 0.0006
Case 18	1.0012 ± 0.0004	1.0028 ± 0.0004	1.0020 ± 0.0006
Case 19	1.0020 ± 0.0004	1.0032 ± 0.0004	1.0035 ± 0.0006
Case 20	1.0039 ± 0.0004	1.0039 ± 0.0004	1.0041 ± 0.0006
Case 21	1.0001 ± 0.0004	1.0016 ± 0.0004	0.9986 ± 0.0006
Case 22	0.9994 ± 0.0004	1.0018 ± 0.0004	0.9981 ± 0.0006
Case 23	1.0007 ± 0.0004	1.0013 ± 0.0004	0.9991 ± 0.0006
Case 24	1.0026 ± 0.0004	1.0032 ± 0.0004	1.0037 ± 0.0006
Case 25	1.0029 ± 0.0004	1.0032 ± 0.0004	1.0027 ± 0.0006
Case 26	1.0026 ± 0.0004	1.0030 ± 0.0004	1.0038 ± 0.0006
Case 27	1.0021 ± 0.0004	1.0030 ± 0.0004	0.9970 ± 0.0006
Case 28	1.0001 ± 0.0004	1.0017 ± 0.0004	1.0038 ± 0.0006
Case 29[b]	1.0476 ± 0.0004	1.0481 ± 0.0004	1.0477 ± 0.0006
Case 30[b]	1.0127 ± 0.0004	1.0094 ± 0.0004	1.0175 ± 0.0006

(a) Results provided by Dave Heinrichs of Lawrence Livermore National Laboratory.
(b) Anomalous results

- Look into cases 1, 21, 28, 29, 30

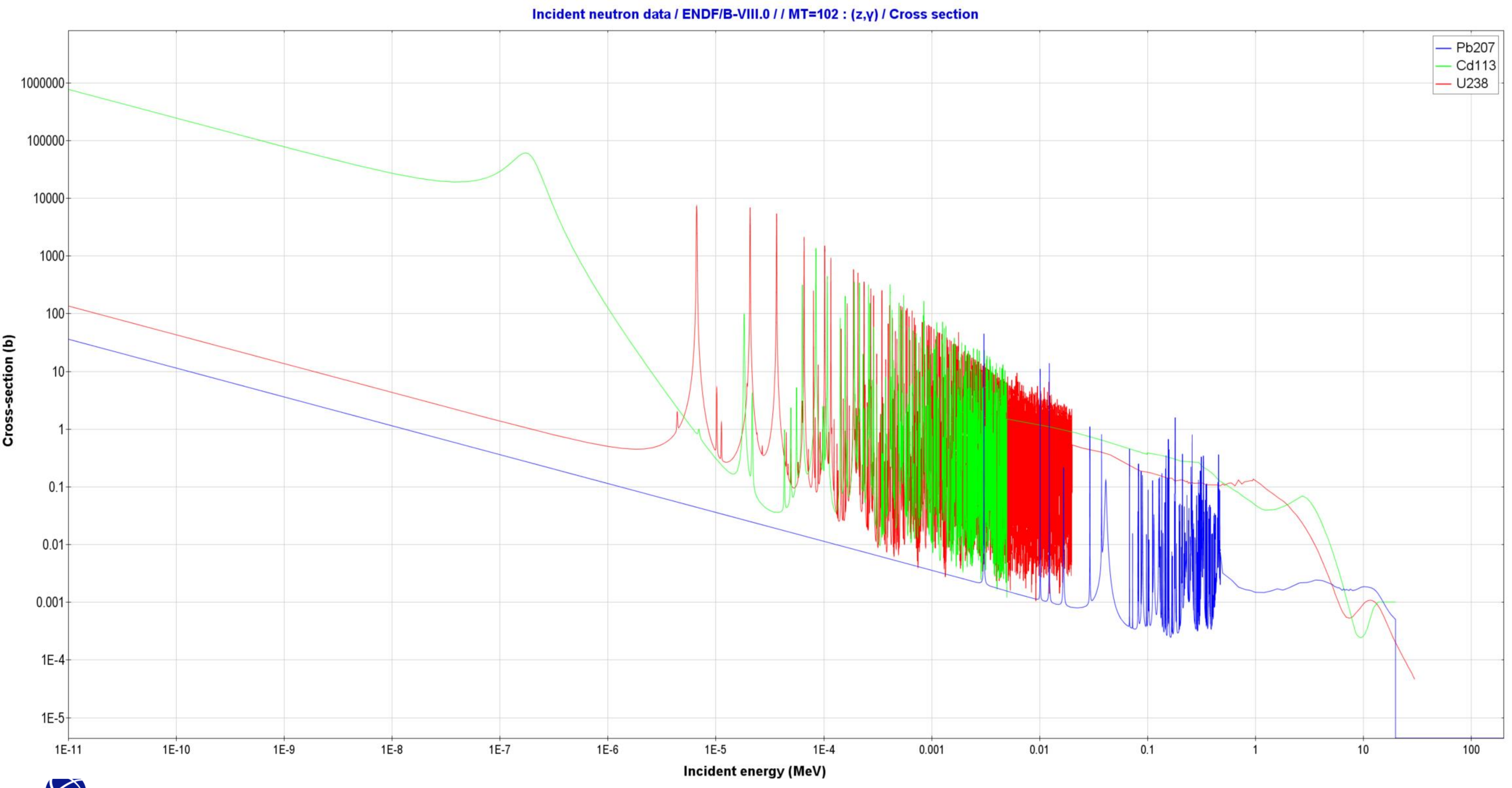
Table 32. Critical Configurations.

Case	Absorber-reflector plates on top of Tank #1 (Top Tank)	Absorber-reflector plates on top of Tank #2 (Bottom Tank)	Separation gap (cm) (a)
1	-	-	9.37768
2	-	POLY-2	1.55702
3	POLY-2	-	16.4719
4	SS	-	10.7823
5	-	SS	9.26592
6	SS	POLY-2	3.08102
7	POLY-2	BP-2	12.23772
8	POLY-2	BP-6	5.84454
9	POLY-2	BP-2 and BP-6	0.38608
10	POLY-2	BP-7	7.78256
11	BP-7	-	11.05408
12	BP-2	-	11.22934
13	-	1 Boraflex	5.50926
14	-	2 Boraflex's	3.87604
15	-	3 Boraflex's	2.6797
16	-	4 Boraflex's	1.6129
17	POLY-3	BP-7	2.84988
18	-	BP-2	4.61518
19	HRS	BP-2	5.9309
20	SS	BP-2	6.1214
21	-	POLY-3, HRS and POLY-1	5.22224
22	-	POLY-6, HRS, and POLY-1	3.24866
23	-	POLY-6, SS, and POLY-1	3.01244
24	-	Pyrex	3.48742
25	Lead	BP-2	5.08254
26	Beryllium	BP-2	6.42366
27	DU	BP-2	5.40766
28	-	Cadmium and POLY-1	5.68096
29	-	Lead and POLY-1	4.96316
30	-	POLY-6, DU and POLY-1	3.2258

(a) This is the distance between the bottom of Tank #1 and the top of any plates stacked on Tank #2. If no plates are on top of Tank #2, then the separation gap is the distance between the two tanks.

Without Poly and Interstitial material, the WINCO tanks are supercritical at any separation less than 9.4 cm

Somehow the 1/8'' of Pb is beating 1/16'' of Cd for thermal neutron absorber. Case 30 is an additional 0.7'' of poly and 1/8'' of DU.



Incident neutron data / ENDF/B-VIII.0 / / MT=102 : (z,γ) / Cross section
Pb207
Cd113
U238
1000000
100000
10000
1000
100
10
1
0.1
0.01
0.001
1E-4
1E-5
Cross-section (b)
1E-11
1E-10
1E-9
1E-8
1E-7
1E-6
1E-5
1E-4
0.001
0.01
0.1
1
10
100
Incident energy (MeV)

Poly Definitions Wrong?

Logbooks report changing descriptions

- Logbook and configuration table in ICSBEP entry are different and show different thicknesses
- Further review revealed that between ‘86 and ‘88 the polyethylene was relabeled but no information exists between which naming is which

Table 5. Dimensions of the Absorber-Reflector Plates.

Plate ID		**Diameter (in.)**	**Thickness (in.)**
POLY-1		27.704 ± 0.001	0.512 ± 0.001
POLY-2		27.004 ± 0.007	1.854 ± 0.002
POLY-3		27.696 ± 0.001	0.511 ± 0.001
POLY-6		27.010 ± 0.001	0.776 ± 0.011
BP-2		28.230 ± 0.001	0.482 ± 0.003
BP-6		28.232 ± 0.001	1.011 ± 0.004
BP-7		28.243 ± 0.001	0.478 ± 0.005
Boraflex		27.668 ± 0.067	0.082 ± 0.001
Pyrex		29.073 ± 0.003	1.113 ± 0.001
SS		29.997 ± 0.001	0.251 ± 0.001
HRS		27.730 ± 0.001	0.234 ± 0.001
Lead		28.634 ± 0.002	0.125 ± 0.002
Beryllium		Fourteen 2.865 × 16.008 × 0.255-inch Plates (see Figure 7) [a]	
Depleted Uranium		2 Rectangular Depleted-Uranium Plates (see Figure 8) [a]	
Cadmium[b]	CD-1	28.62 ± 0.02	0.0687 ± 0.001
	CD-2	28.60 ± 0.02	0.0681 ± 0.001

(a) obtained from lab notebook
(b) two half disks (see Figure 9)

Polyethelene				
POLY-1	0.2558 +/- .0019	2.573	28.661	0.000
POLY-2	0.5132 +/- .0013	4.930	28.504	0.000
POLY-3	0.5128 +/- .0010	4.973	28.610	0.000
POLY-4	0.9980 +/- .0014	9.637	28.524	0.000
POLY-5	1.0238 +/- .0048	9.957	28.602	0.000
POLY-6	2.0493 +/- .0060	19.226	28.602	0.000
POLY-7	2.0103 +/- .0081	19.093	28.591	0.000
Borated Polyethelene				
BP-1 (5 wt.% B)	0.4725 +/- .0029	4.539	28.472	0.000
BP-2 (5 wt.% B)	0.4840 +/- .0071	4.770	28.504	0.000
BP-3 (5 wt.% B)	0.4853 +/- .0096	4.712	28.563	0.000
BP-4 (5 wt.% B)	0.4890 +/- .0115	4.729	28.543	0.000
BP-5 (5 wt.% B)	1.0303 +/- .0207	9.556	28.496	0.000
BP-6 (5 wt.% B)	1.0193 +/- .0093	10.268	28.614	0.000
BP-7 (32 wt.% B)	0.4808 +/- .0024	6.092	28.465	0.000
BP-8 (32 wt.% B)	0.4873 +/- .0040	6.091	28.622	0.000

Updating polyethylene description

Adding more poly to increase capture

- Adding 1.3 inches of polyethylene to stack which could be a combination of several poly slabs
- Result is a reduction in keff from 1.040 to 0.996 (-4400 pcm change)