

The role of fission yield correlations to obtain realistic uncertainty values in the summation method

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BROOKHAVEN
NATIONAL LABORATORY

a passion for discovery



U.S. DEPARTMENT OF
ENERGY

Office of
Science

Fission as a tool to study weak interactions

It was realized early on that the large antineutrino flux generated by a nuclear reactor, $> 10^{20}$ /second, could be used to detect antineutrinos

Detection of the Free Neutrino: a Confirmation

C.L. Cowan, F. Reines, F.B. Harrison,
H.W. Kruse, A.D. McGuire
Science 124, 3212 (1956).

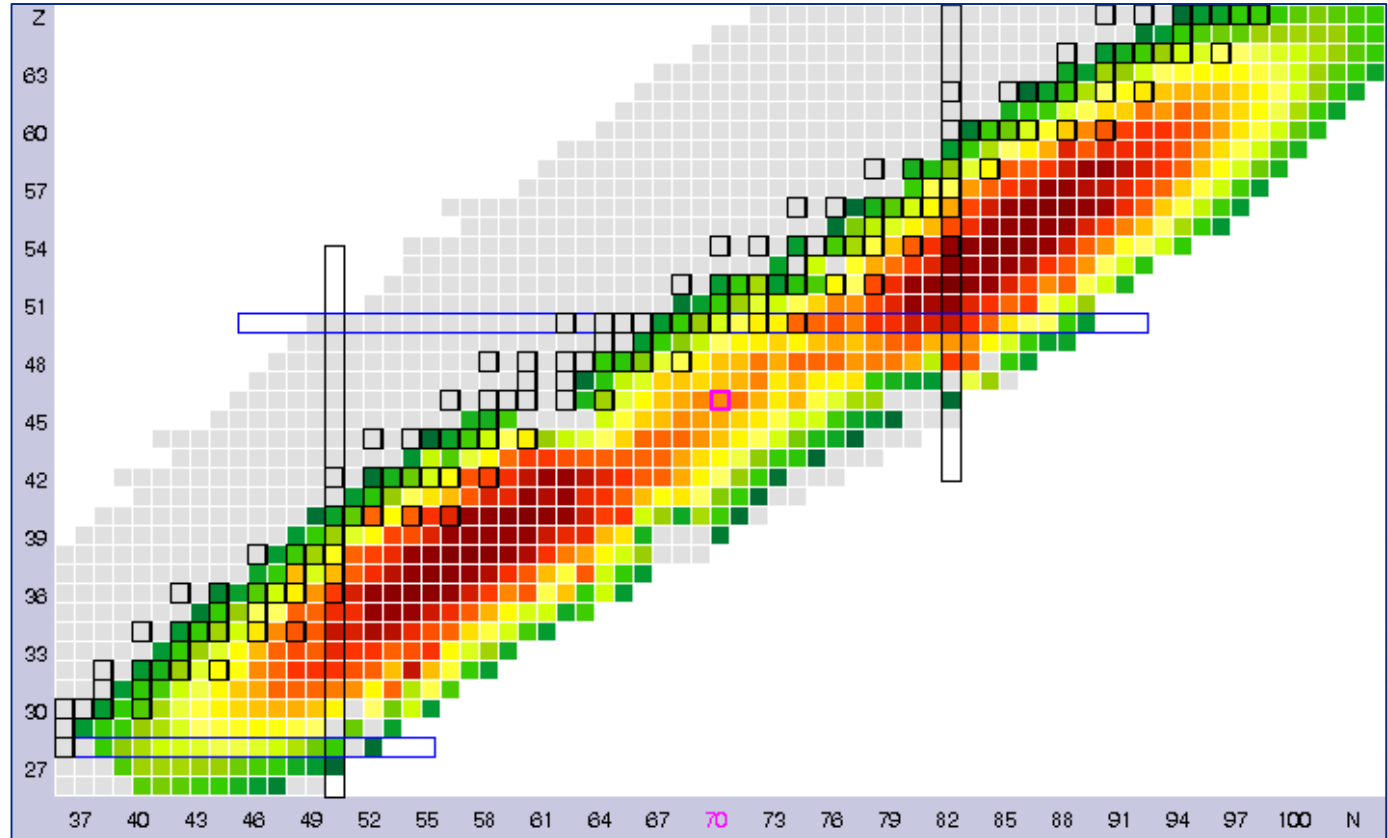
LANL team working at the Savannah
River Site

~25 years after first postulated by Pauli



How are antineutrinos produced in a nuclear reactor?

Electron antineutrinos are produced in the beta minus decay of the neutron rich fission products



In our technical parlance, reactor antineutrinos are delayed radiation, somewhat anticorrelated with beta delayed neutrons.

How are electron antineutrinos detected?

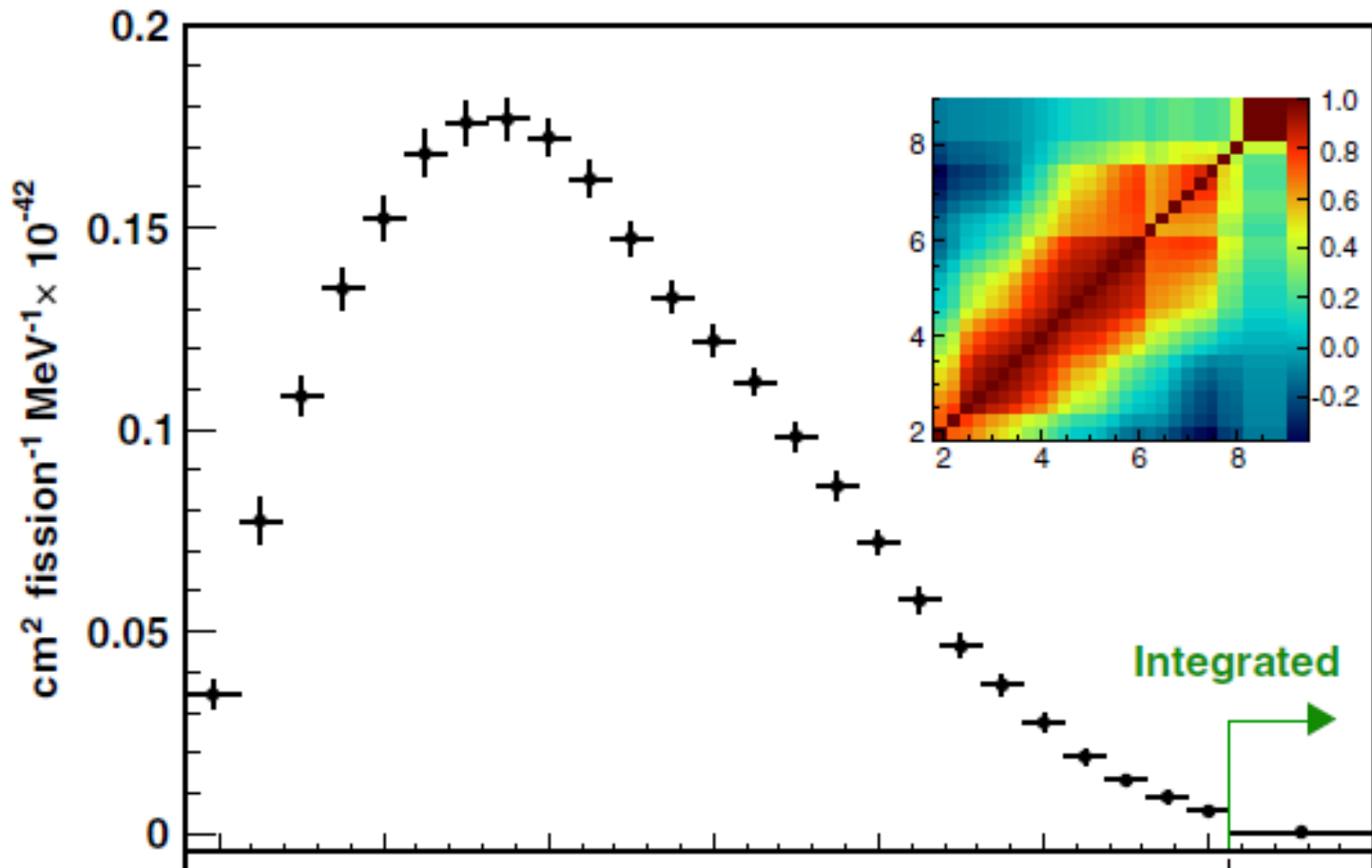
- Electron antineutrinos are captured by protons through an Inverse Beta Decay process:



- The positron annihilates and produces 2 511-keV gammas
- The neutron is captured some micro seconds later producing a distinct gamma event with ~ 8 MeV sum energy.
- Large detectors with Gd-loaded water and many scintillators.

What type of signal is measured?

The antineutrino spectrum will be the product of the spectrum generated by the fission products times the IBD cross section.



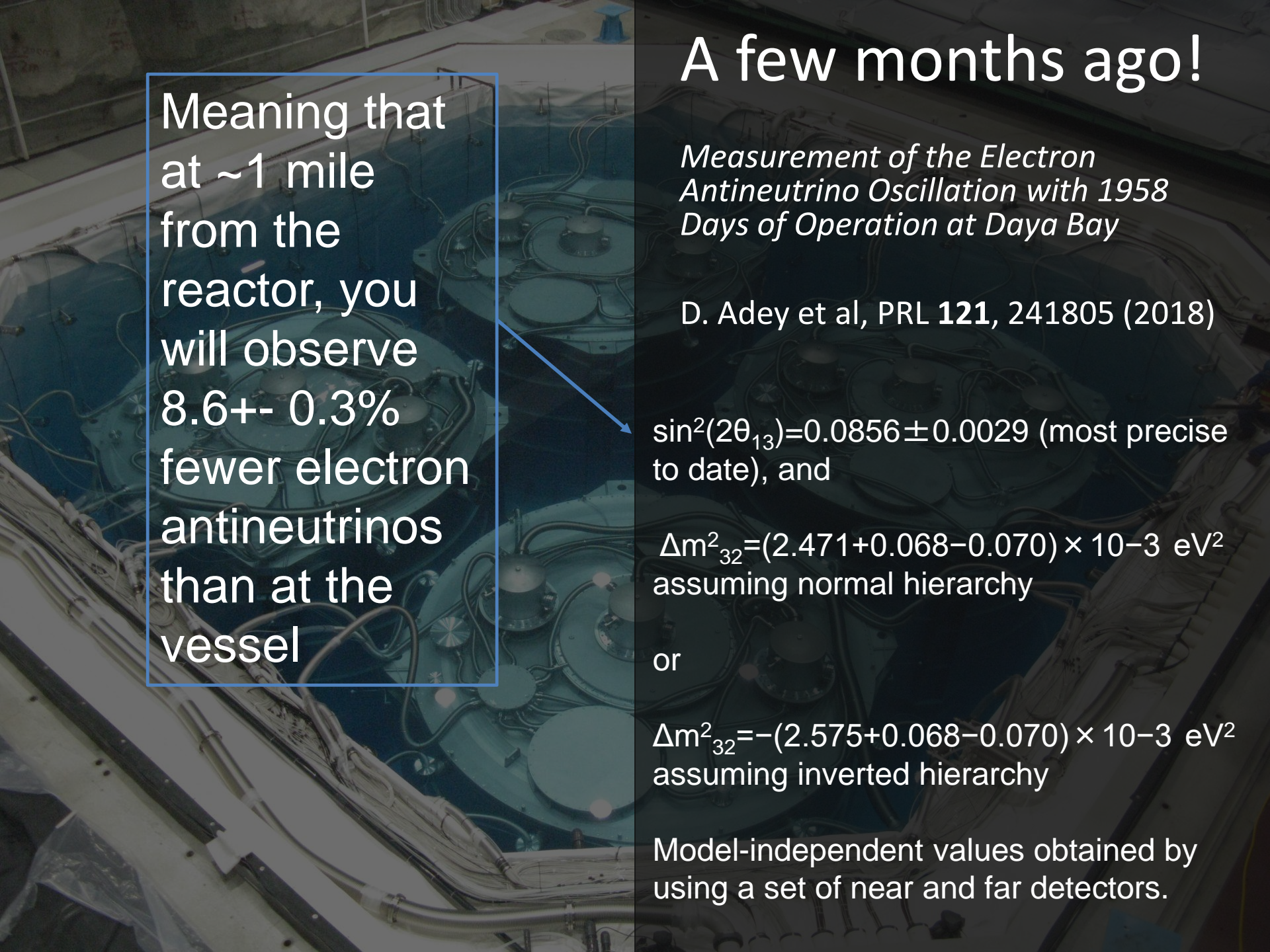
Neutrino Oscillations

About 10 years ago, data taking started at three large nuclear reactor antineutrino experiments:

- Daya Bay (China),
- Double Chooz (France),
- RENO (Korea)

The goal was to measure θ_{13} and Δm_{32}^2 by placing detector at around 1 mile and 400 yards from the reactors.





Meaning that
at ~1 mile
from the
reactor, you
will observe
8.6± 0.3%
fewer electron
antineutrinos
than at the
vessel

A few months ago!

*Measurement of the Electron
Antineutrino Oscillation with 1958
Days of Operation at Daya Bay*

D. Adey et al, PRL **121**, 241805 (2018)

$\sin^2(2\theta_{13})=0.0856\pm 0.0029$ (most precise
to date), and

$\Delta m^2_{32}=(2.471+0.068-0.070)\times 10^{-3}$ eV²
assuming normal hierarchy

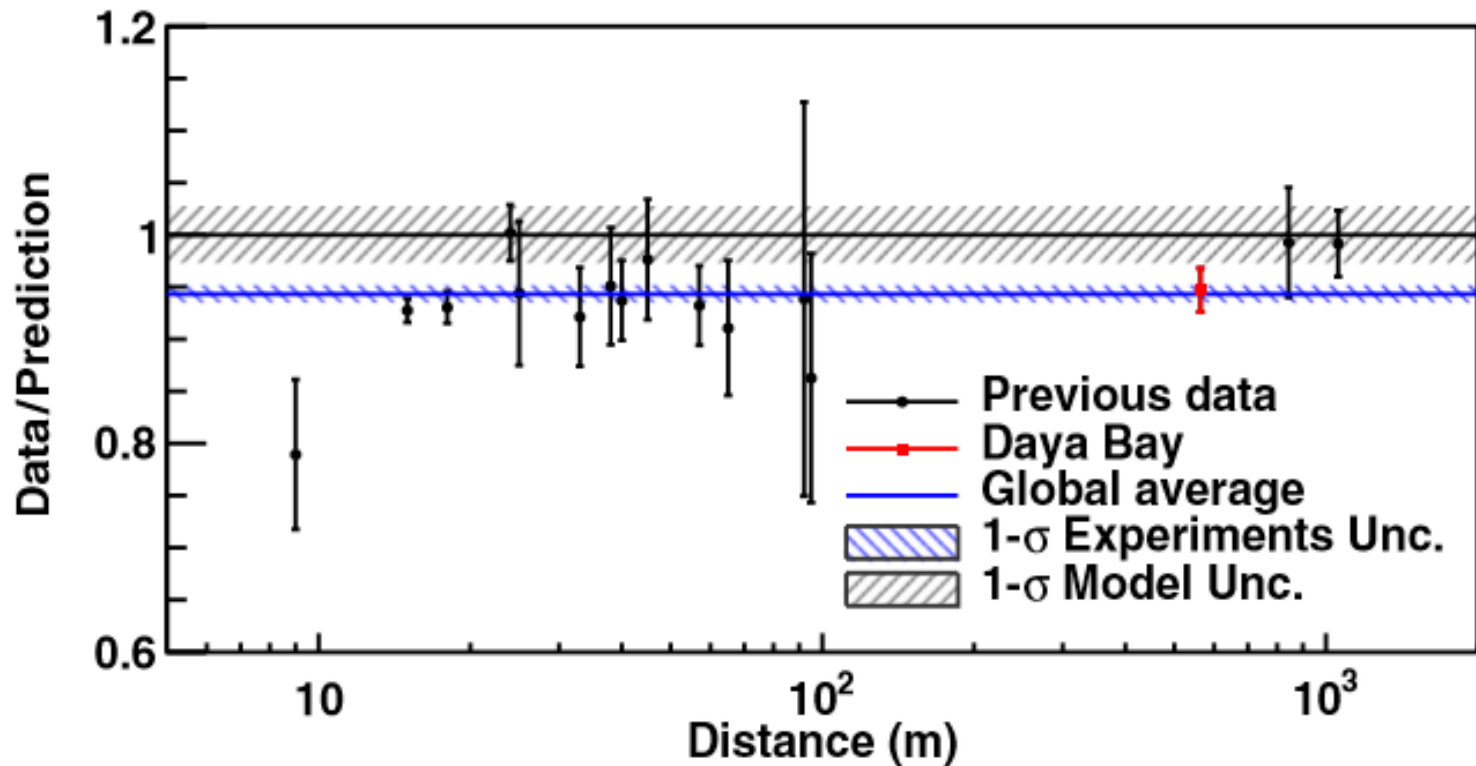
or

$\Delta m^2_{32}=- (2.575+0.068-0.070)\times 10^{-3}$ eV²
assuming inverted hierarchy

Model-independent values obtained by
using a set of near and far detectors.

Additionally!

Since 2011 we learned that we are missing about 6% of electron antineutrinos near the reactor (model dependent)



Antineutrinos from fission products

How to Spot a Nuclear Bomb Program? Look for Ghostly Particles

The New York Times

By **Kenneth Chang**

March 27, 2018

**Watchman
Project
LLNL**



The Boulby mine in northeast England will be home to the Watchman experiment, which aims to detect antineutrinos from a nearby nuclear power plant. Science and Technology Facilities Council Boulby Underground Laboratory

How to Calculate a Reactor Antineutrino Spectrum

Know the composition of the core in terms of fission fractions

^{235}U : 0.586 ^{238}U : 0.076 ^{239}Pu : 0.288 ^{241}Pu : 0.050

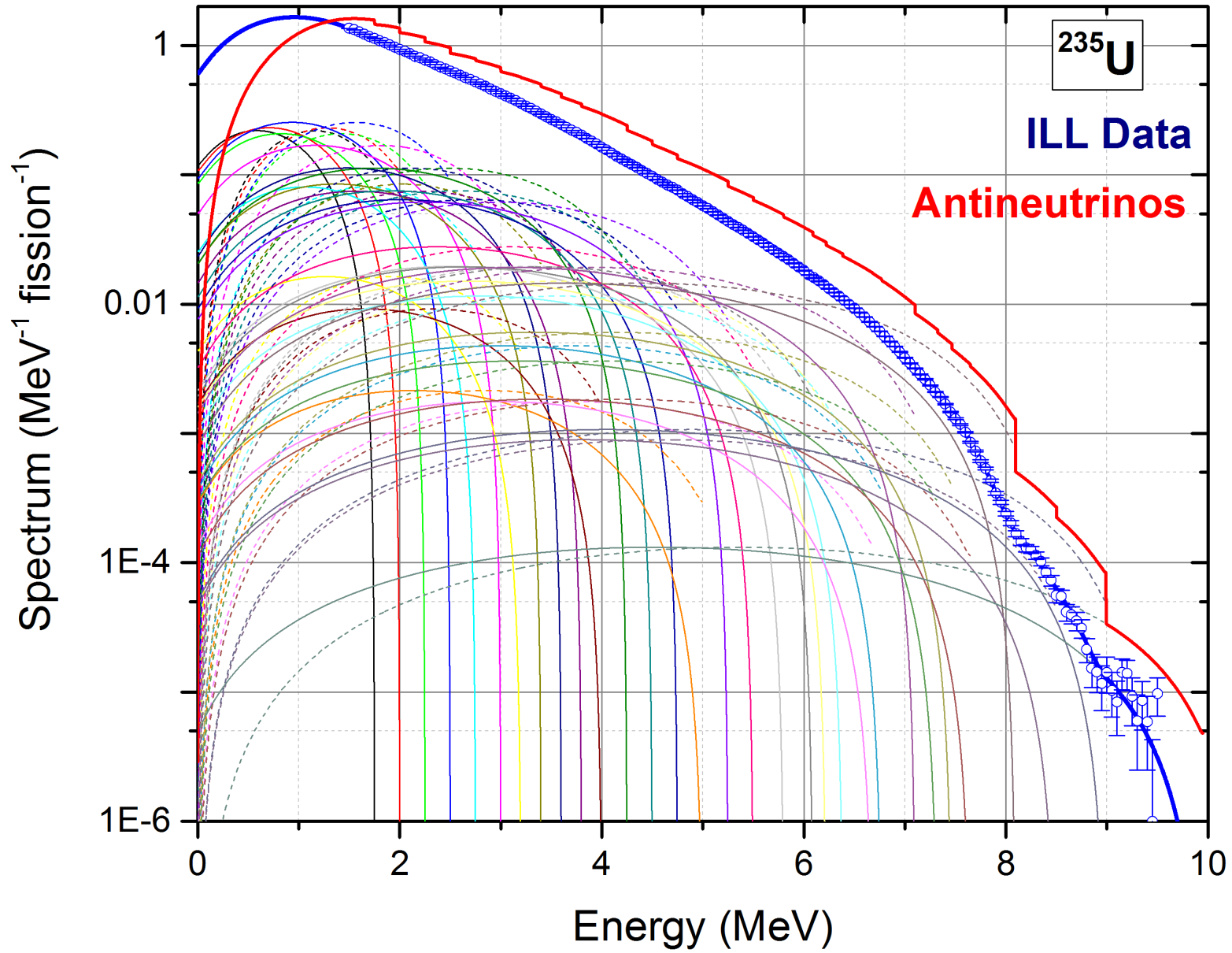
$$\begin{aligned} \text{Spectrum (reactor)} = & c_5 \times \text{Spectrum}(^{235}\text{U}) + \\ & c_8 \times \text{Spectrum}(^{238}\text{U}) + \\ & c_9 \times \text{Spectrum}(^{239}\text{Pu}) + \\ & c_1 \times \text{Spectrum}(^{241}\text{Pu}) \end{aligned}$$

The ^{235}U , $^{239,241}\text{Pu}$ spectra are obtained from the ILL electron spectra measurements (Conversion).

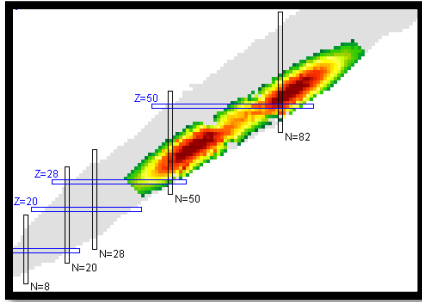
For ^{238}U , using fission yield and decay databases (Summation).

Signal = Spectrum x Inverse beta decay cross section,

Conversion Method



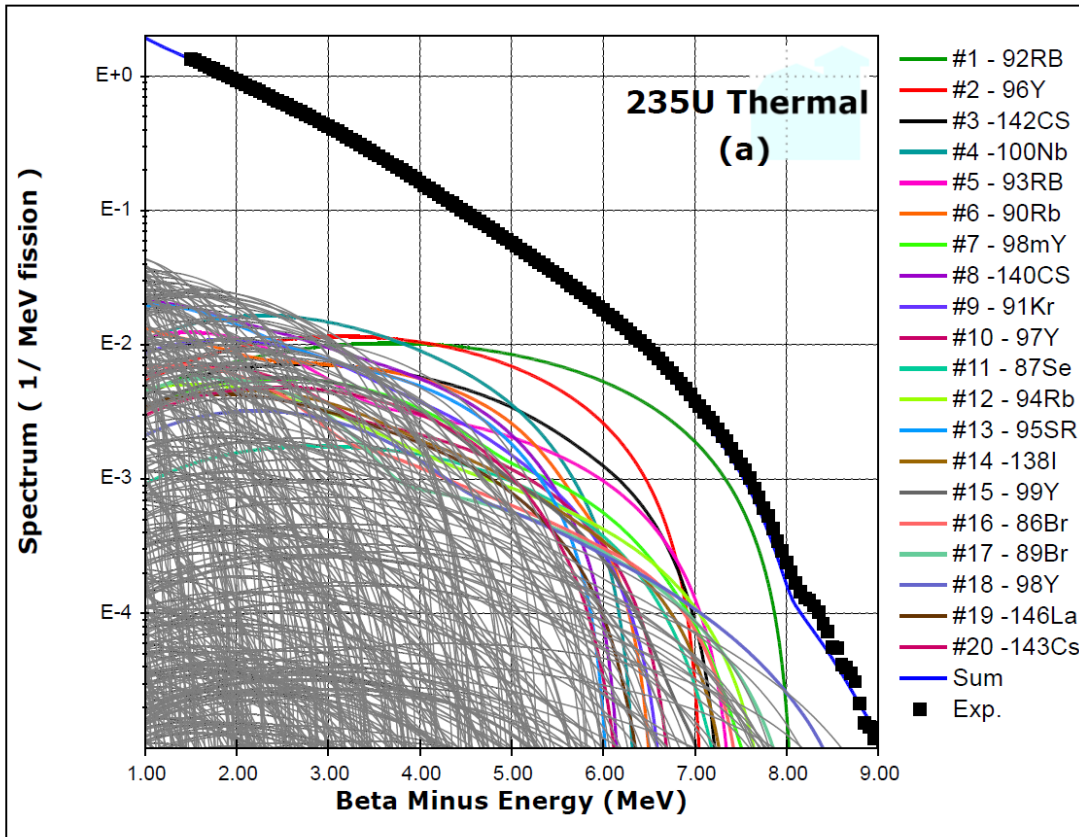
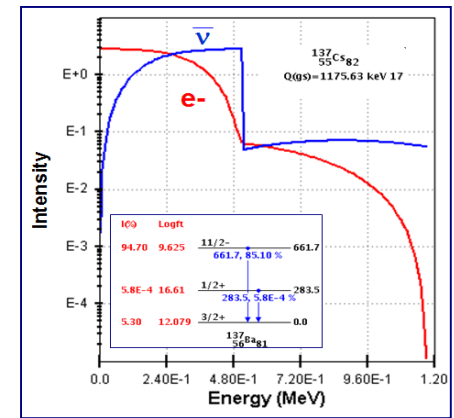
Using Our Databases



$$S(E) = \sum CFY_i S_i(E)$$

Cumulative Fission Yields

Individual Spectra



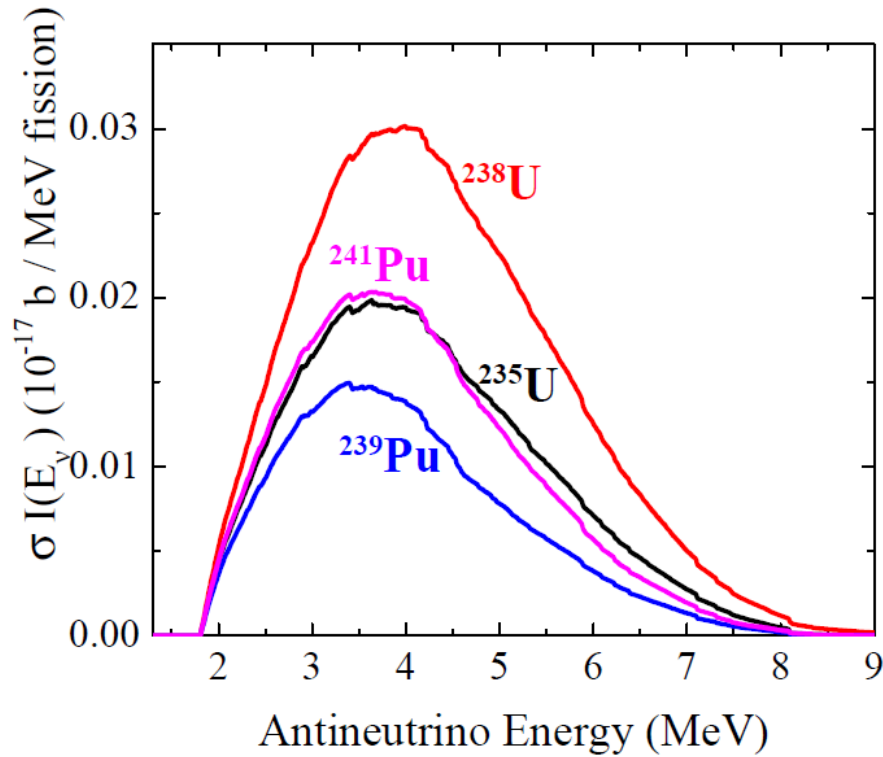
Comparison with the measured electron spectra.

Surprisingly, fewer contributors at high energy.

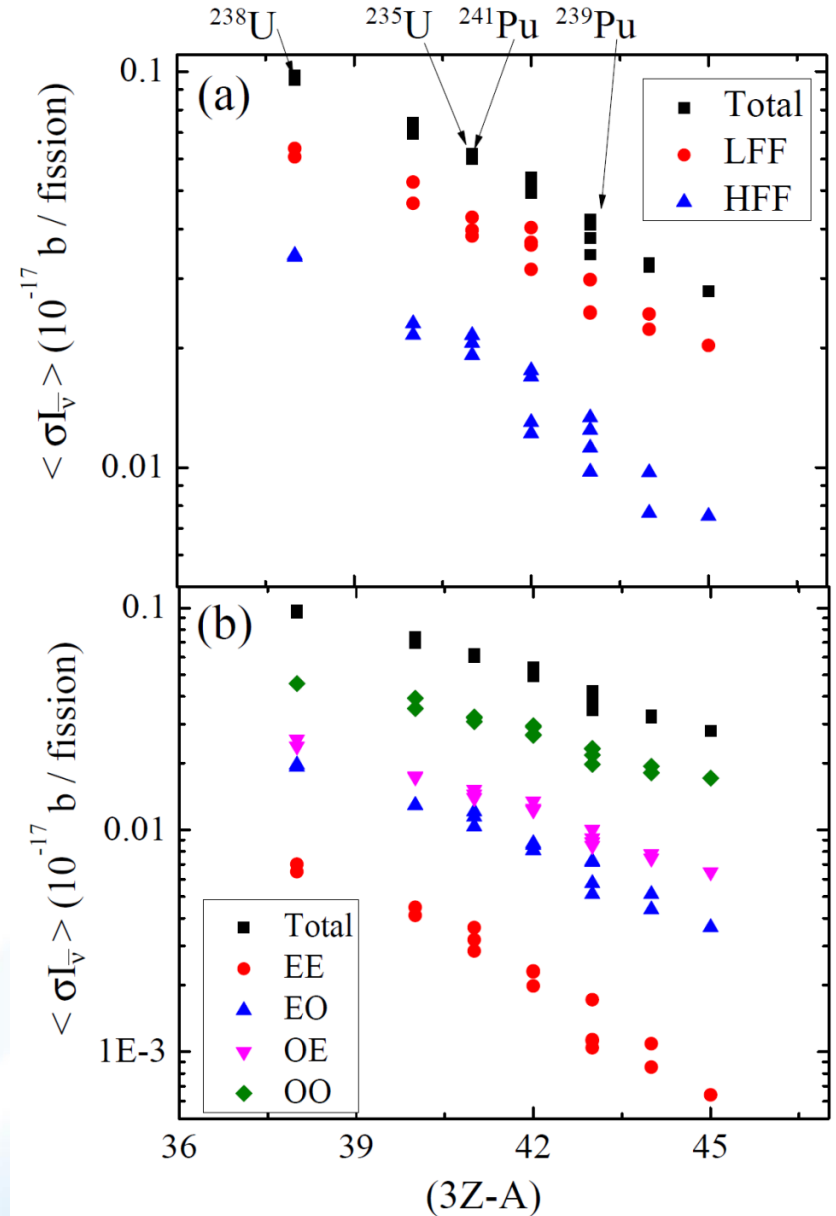
⁹²Rb and ⁹⁶Y are the two top contributors.

First calculation of this type performed by P. Vogel et al in 1981 using ENDF/B-V.

Systematics for all fissioning systems



3Z-A



Summation Method

Using nuclear databases, the **IBD antineutrino yield** under equilibrium conditions can be calculated as

$$N_{\bar{\nu}} = \sum C_i \int \sigma_{IBD}(E) S_i(E) dE = \sum C_i N_i,$$

where **C_i** is the cumulative fission yield and **$S_i(E)$** the antineutrino spectrum for the fission products. The uncertainty can be calculated as

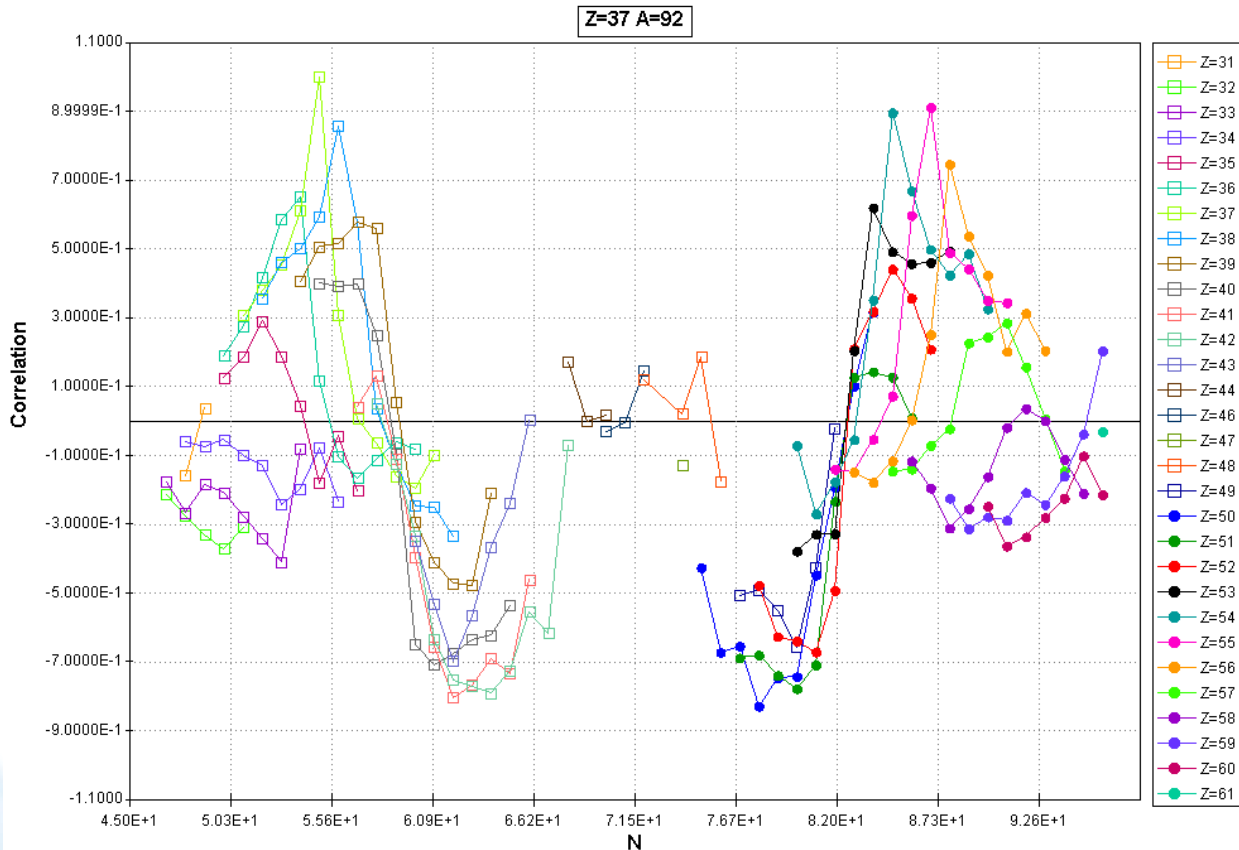
$$\Delta^2 N_{\bar{\nu}} = \sum_i (\Delta C_i N_i)^2 + (C_i \Delta N_i)^2 + \sum_i \sum_{j \neq i} N_i N_j \Delta C_i \Delta C_j \text{Cor}(C_i, C_j).$$

where **$\text{Cor}(C_i, C_j)$** is the correlation among cumulative fission yields.

Summation Method

Unfortunately, the fission yield databases **don't contain any correlation data**.

We used GEF¹ to obtain Independent fission yield correlations



Independent fission yield correlations for ^{92}Rb in $^{235}\text{U}(n,f)$.

The largest correlations will be for nearby nuclides as well as for nuclides with $Z=92-37$ and $A=236-92-2$.

Positive correlations among nuclides in the same fission mode: symmetric, asymmetric (S1), very asymmetric (S2).

Negative correlations among different fission modes.

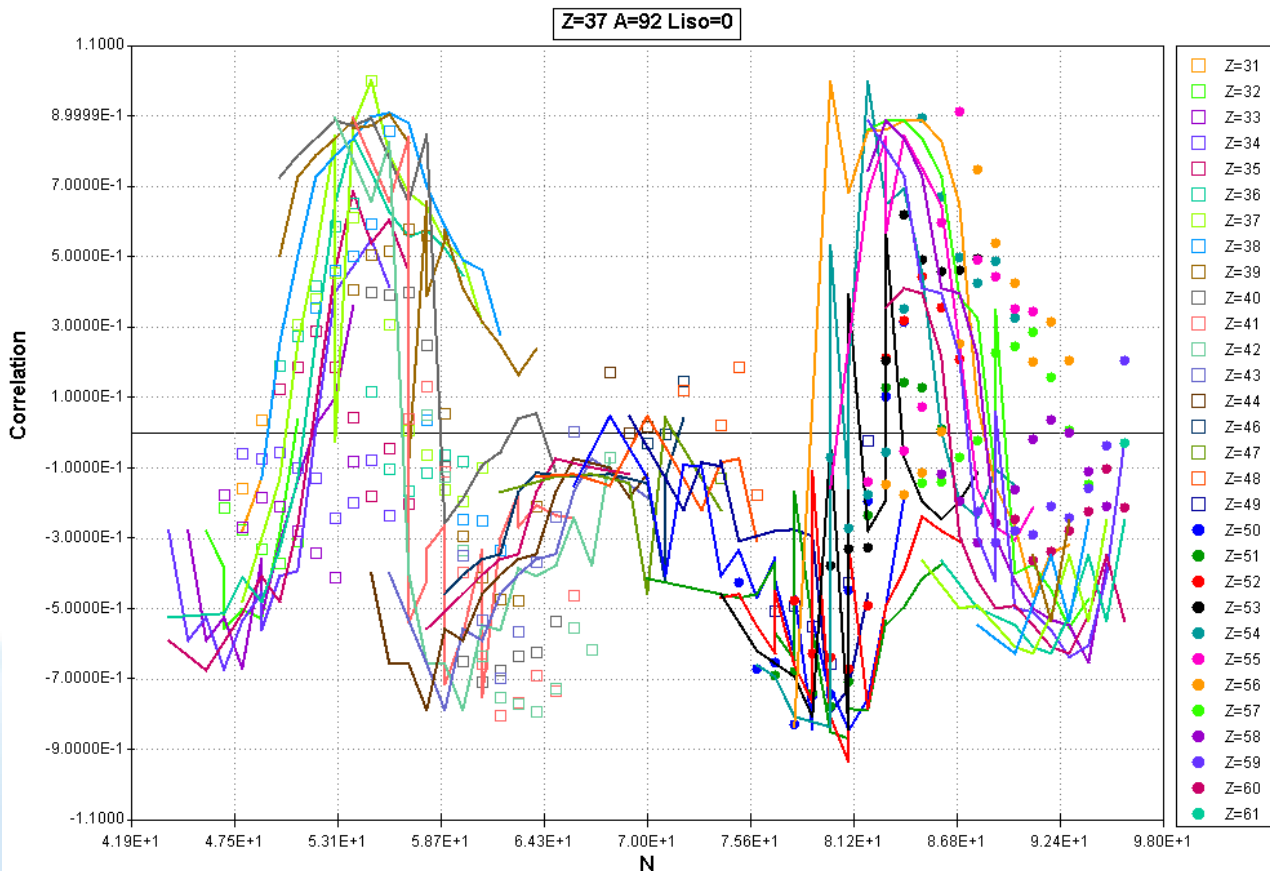
¹ "General Description of Fission Observables: GEF Model Code", K.-H. Schmidt, B. Jurado, C. Amouroux, C. Schmitt, Nucl. Data Sheets **131**, 107 (2016).

Summation Method

Using ENDF/B-VIII.0 decay data, we can calculate cumulative yield correlations:

$$C_i = I_i + \sum b_{ik} I_k, \text{ where } b_{ik} \text{ are decay probabilities and } I_k \text{ are independent yields.}$$

$$\text{For instance, } C(^{92}\text{Rb}) = I(^{92}\text{Rb}) + I(^{92}\text{Kr}) + 0.0195 \times I(^{93}\text{Kr}) + 0.67 \times I(^{92}\text{Br}) + 0.68 \times I(^{93}\text{Br})$$



^{92}Rb Cumulative (lines) and independent (symbols) fission yield correlations for $^{235}\text{U}(n,f)$.

The CFY correlations get broader and shifted to lower Z values

Some issues combining GEF (Monte Carlo) with a JEFF-3.3 yields (deterministic)

Summation Method

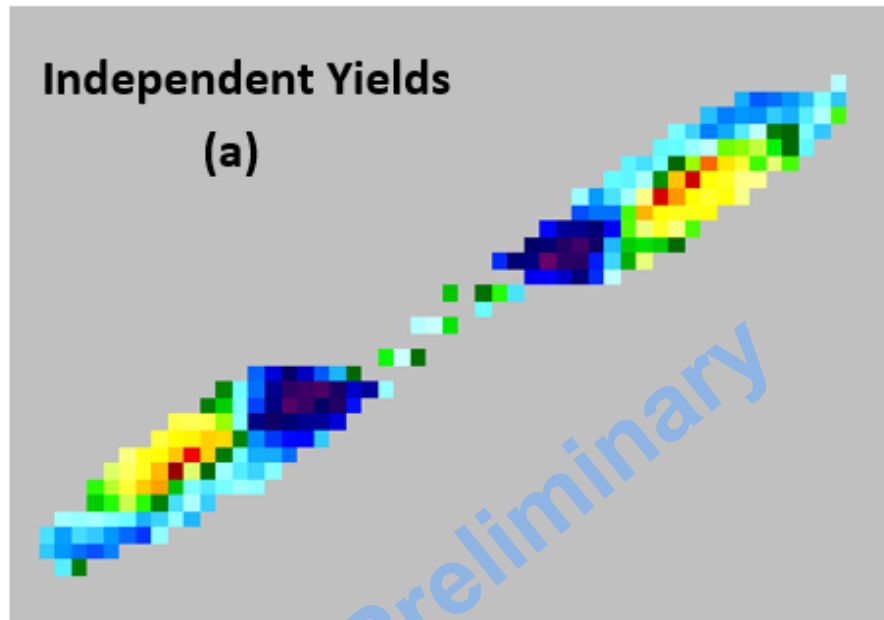
Independent and cumulative fission yield correlations for ^{92}Rb in $^{235}\text{U}(n,f)$.

Cumulative fission yield correlations are considerable wider than the independent ones due to the link among different fission products provided by beta and IT decay.

Z

Independent Yields

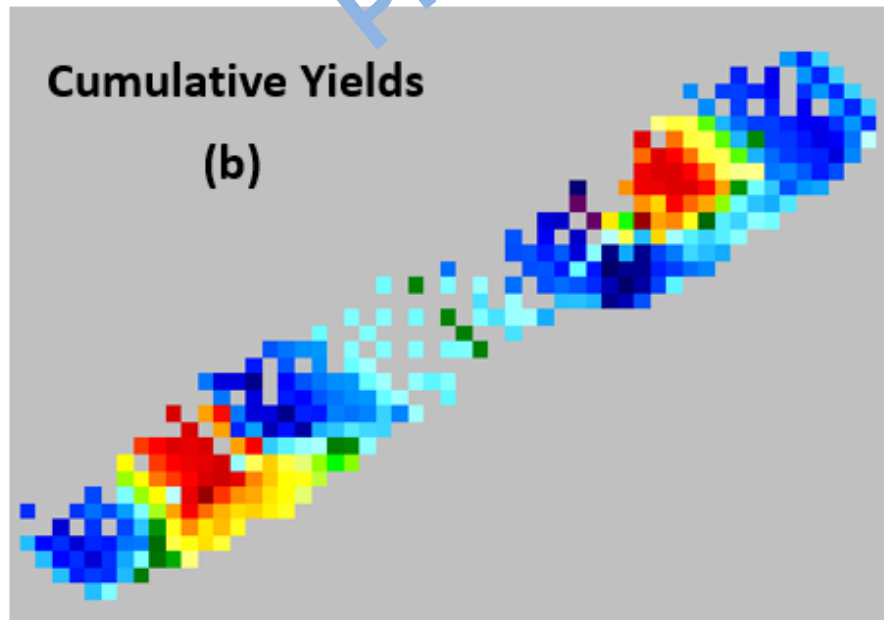
(a)



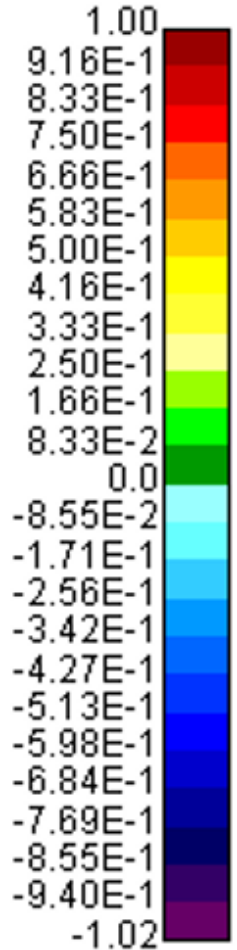
Z

Cumulative Yields

(b)



N



Summation Method

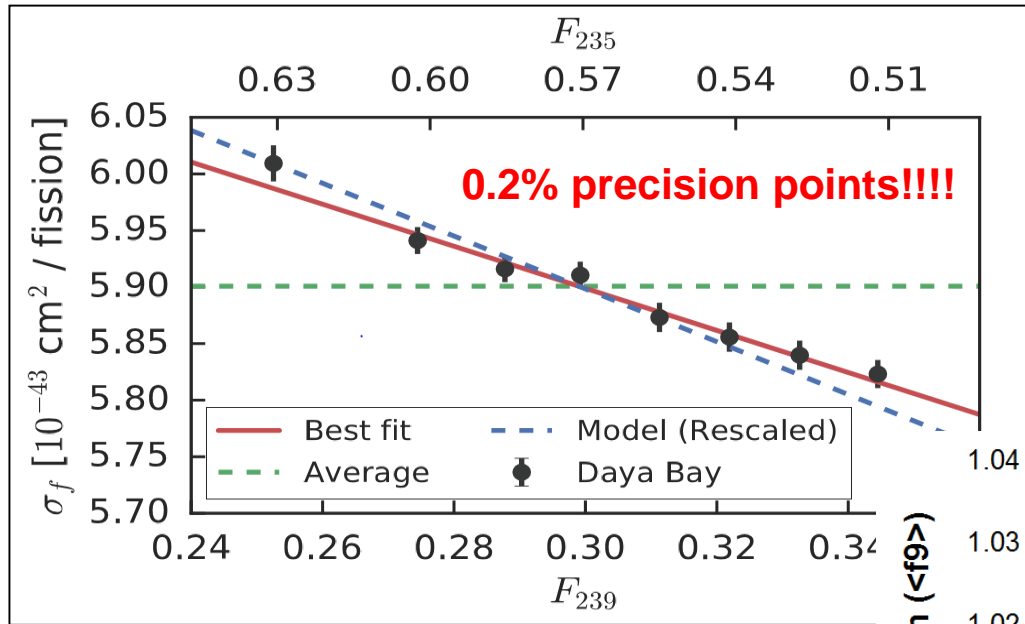
Yield results (10^{-43} cm²/fission) using ENDF/B-VIII.0 decay data, JEFF-3.3 yields and GEF IFY correlations, compared to Huber-Mueller and Daya Bay 2017 evolution data.

Nucleus	Summation Yield	Sum. Δ Yield	Sum. Δ yield No Corrs.	Yield from theory	H-M Yield	H-M Δ Yield	DB17 Yield	DB17 Δ Yield
²³⁵ U	6.37	4.59%	1.76%	4.5%	6.69	2.2%	6.17	2.8%
²³⁸ U	9.69	3.43%	2.04%	14%	10.1	10%		
²³⁹ Pu	4.39	5.18%	2.08%	8.4%	4.36	4.4%	4.27	6.1%
²⁴¹ Pu	6.25	5.11%	2.36%	15%	6.04	10%		

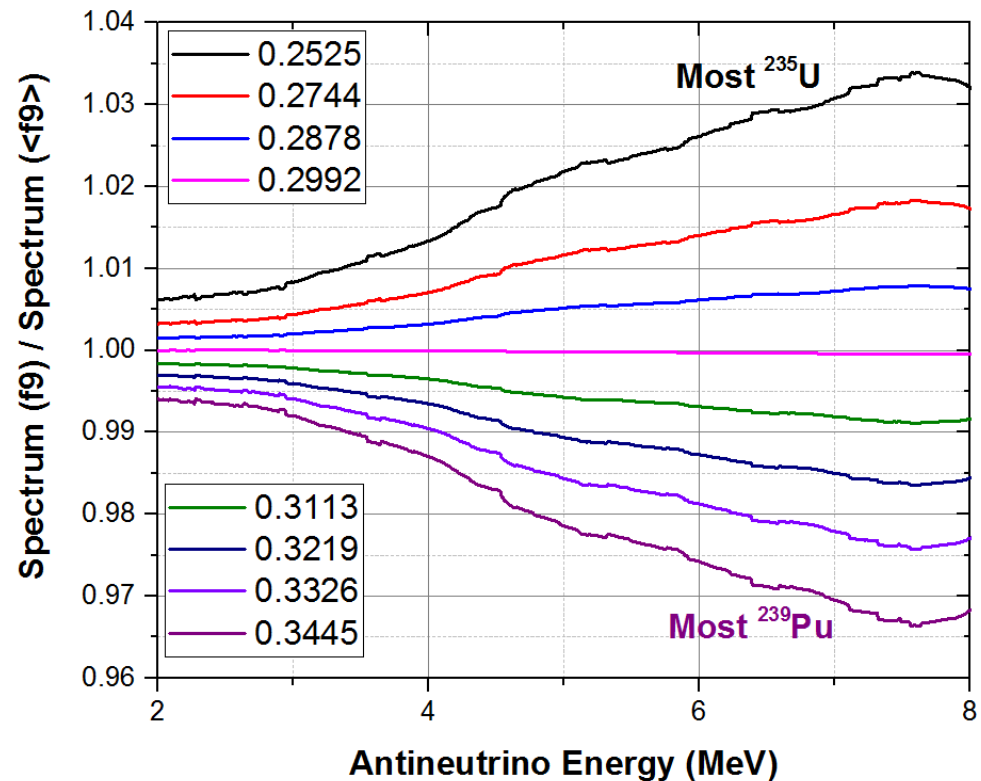
Conclusion: All summation calculations **must include the correlations** among Cumulative Fission Yields!

DB17: F.P. An *et al.*, PRL118, 251801 (2017).

IBD antineutrino yield as function of F_{239}

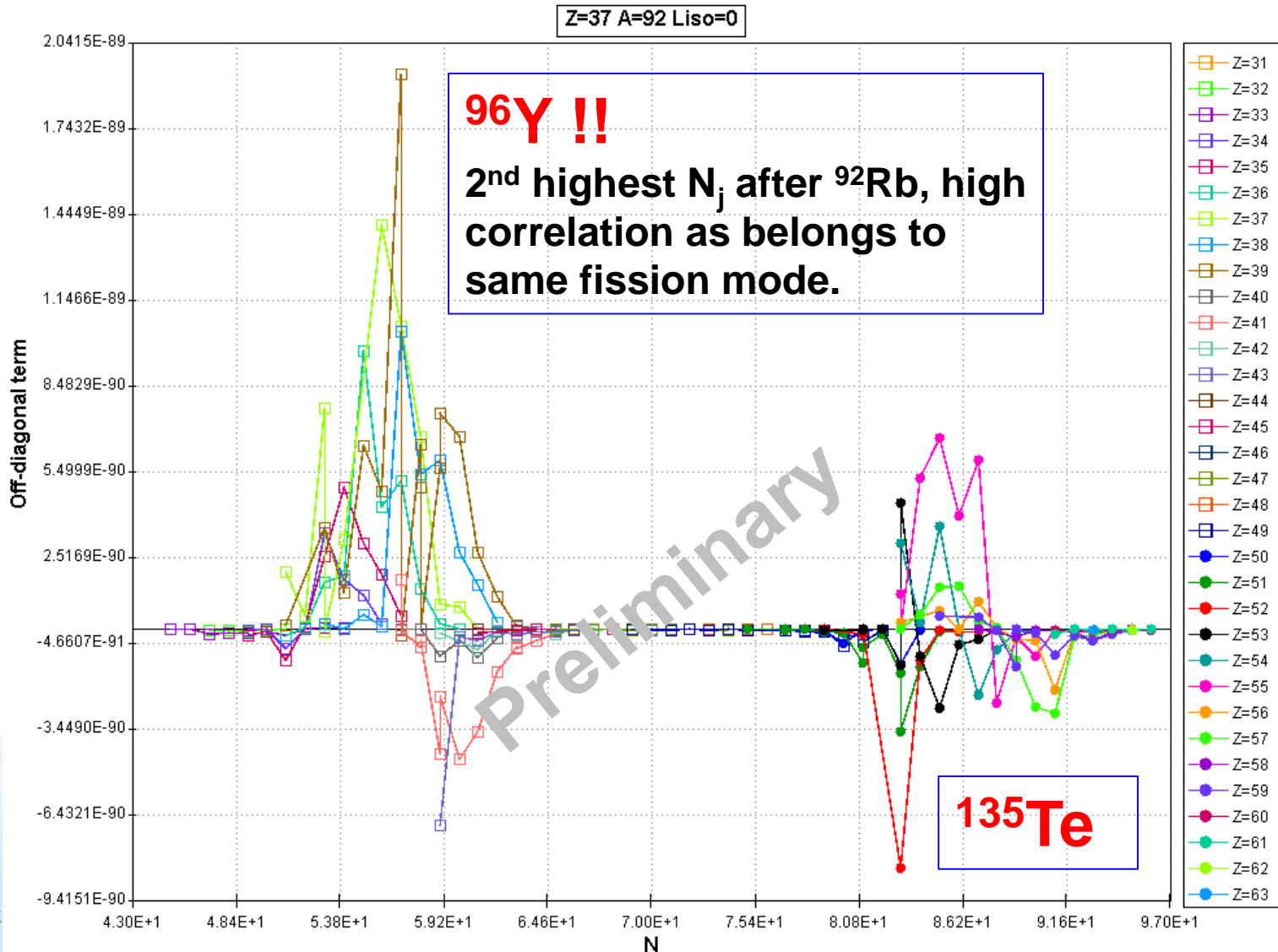


F.P. An *et al*, PRL **118**, 251801 (2017).



Summation Method

Let's plot the off-diagonal uncertainty terms,
 $N_i N_j \Delta C_i \Delta C_j \text{Cor}(C_i, C_j)$, involving ^{92}Rb



Summation Method

¹³⁵Te

High beta-minus Q-value (6 MeV) and fission yield (0.033) due to shell effects.

Z	¹³³ Xe 5.2475 D β ⁻ : 100.00% 427.4	¹³⁴ Xe >5.8E+22 Y 10.4357% 2β ⁻ -1234.667	¹³⁵ Xe 9.14 H β ⁻ : 100.00% 1168	¹³⁶ Xe >2.4E+21 Y 8.8573% 2β ⁻ -90.5	¹³⁷ Xe 3.818 M β ⁻ : 100.00% 4182.2	¹³⁸ Xe 14.08 M β ⁻ : 100.00% 2015	¹³⁹ Xe 39.68 S β ⁻ : 100.00% 5056	¹⁴⁰ Xe 13.60 S β ⁻ : 100.00% 4064	¹⁴¹ Xe 1.73 S β ⁻ : 100.00% β-n: 0.04% 6280
53	¹³² I 2.295 H β ⁻ : 100.00% 3575	¹³³ I 20.83 H β ⁻ : 100.00% 1785	¹³⁴ I 52.5 M β ⁻ : 100.00% 4082	¹³⁵ I 6.58 H β ⁻ : 100.00% 2634	¹³⁶ I 83.4 S β ⁻ : 100.00% 6884	¹³⁷ I 24.5 S β ⁻ : 100.00% β-n: 7.14% 6027	¹³⁸ I 6.23 S β ⁻ : 100.00% β-n: 5.56% 7992	¹³⁹ I 2.280 S β ⁻ : 100.00% β-n: 10.00% 7174	¹⁴⁰ I 0.86 S β ⁻ : 100.00% β-n: 9.30% 9380
52	¹³¹ Te 25.0 M β ⁻ : 100.00% 2231.7	¹³² Te 3.204 D β ⁻ : 100.00% 515	¹³³ Te 12.5 M β ⁻ : 100.00% 2921	¹³⁴ Te 41.8 M β ⁻ : 100.00% 1510	¹³⁵ Te 19.0 S β ⁻ : 100.00% 6050	¹³⁶ Te 17.63 S β ⁻ : 100.00% β-n: 1.31% 5120	¹³⁷ Te 2.49 S β ⁻ : 100.00% β-n: 2.99% 7053	¹³⁸ Te 1.4 S β ⁻ : 100.00% β-n: 6.30% 6284	¹³⁹ Te >150 NS β ⁻ : 100.00% β-n 8266
51	¹³⁰ Sb 39.5 M β ⁻ : 100.00% 5067	¹³¹ Sb 23.03 M β ⁻ : 100.00% 3229.6	¹³² Sb 2.79 M β ⁻ : 100.00% 5553	¹³³ Sb 2.34 M β ⁻ : 100.00% 4014	¹³⁴ Sb 0.78 S β ⁻ : 100.00% 8513	¹³⁵ Sb 1.679 S β ⁻ : 100.00% β-n: 22.00% 8038	¹³⁶ Sb 0.923 S β ⁻ : 100.00% β-n: 18.30% 9918	¹³⁷ Sb 492 MS β ⁻ : 100.00% β-n: 49.00% 9.24E+3	¹³⁸ Sb 348 MS β ⁻ : 100.00% β-n: 72.00% 1.15E+4
50	¹²⁹ Sn 2.23 M β ⁻ : 100.00% 4.04E+3	¹³⁰ Sn 3.72 M β ⁻ : 100.00% 2153	¹³¹ Sn 56.0 S β ⁻ : 100.00% 4717	¹³² Sn 39.7 S β ⁻ : 100.00% 3089	¹³³ Sn	¹³⁴ Sn	¹³⁵ Sn	¹³⁶ Sn	¹³⁷ Sn

Z	¹³¹ Ba	¹³² Ba	¹³³ Ba	¹³⁴ Ba	¹³⁵ Ba	¹³⁶ Ba	¹³⁷ Ba	¹³⁸ Ba	¹³⁹ Ba	¹⁴⁰ Ba	¹⁴¹ Ba	¹⁴² Ba	¹⁴³ Ba	¹⁴⁴ Ba	¹⁴⁵ Ba	¹⁴⁶ Ba	¹⁴⁷ Ba
	¹³⁰ Cs	¹³¹ Cs	¹³² Cs	¹³³ Cs	¹³⁴ Cs	¹³⁵ Cs	¹³⁶ Cs	¹³⁷ Cs	¹³⁸ Cs	¹³⁹ Cs	¹⁴⁰ Cs	¹⁴¹ Cs	¹⁴² Cs	¹⁴³ Cs	¹⁴⁴ Cs	¹⁴⁵ Cs	¹⁴⁶ Cs
54	¹²⁹ Xe	¹³⁰ Xe	¹³¹ Xe	¹³² Xe	¹³³ Xe	¹³⁴ Xe	¹³⁵ Xe	¹³⁶ Xe	¹³⁷ Xe	¹³⁸ Xe	¹³⁹ Xe	¹⁴⁰ Xe	¹⁴¹ Xe	¹⁴² Xe	¹⁴³ Xe	¹⁴⁴ Xe	¹⁴⁵ Xe
	¹²⁸ I	¹²⁹ I	¹³⁰ I	¹³¹ I	¹³² I	¹³³ I	¹³⁴ I	¹³⁵ I	¹³⁶ I	¹³⁷ I	¹³⁸ I	¹³⁹ I	¹⁴⁰ I	¹⁴¹ I	¹⁴² I	¹⁴³ I	¹⁴⁴ I
52	¹²⁷ Te	¹²⁸ Te	¹²⁹ Te	¹³⁰ Te	¹³¹ Te	¹³² Te	¹³³ Te	¹³⁴ Te	¹³⁵ Te	¹³⁶ Te	¹³⁷ Te	¹³⁸ Te	¹³⁹ Te	¹⁴⁰ Te	¹⁴¹ Te	¹⁴² Te	¹⁴³ Te
	¹²⁶ Sb	¹²⁷ Sb	¹²⁸ Sb	¹²⁹ Sb	¹³⁰ Sb	¹³¹ Sb	¹³² Sb	¹³³ Sb	¹³⁴ Sb	¹³⁵ Sb	¹³⁶ Sb	¹³⁷ Sb	¹³⁸ Sb	¹³⁹ Sb	¹⁴⁰ Sb		
50	¹²⁵ Sn	¹²⁶ Sn	¹²⁷ Sn	¹²⁸ Sn	¹²⁹ Sn	¹³⁰ Sn	¹³¹ Sn	¹³² Sn	¹³³ Sn	¹³⁴ Sn	¹³⁵ Sn	¹³⁶ Sn	¹³⁷ Sn	¹³⁸ Sn	¹³⁹ Sn		
	¹²⁴ In	¹²⁵ In	¹²⁶ In	¹²⁷ In	¹²⁸ In	¹²⁹ In	¹³⁰ In	¹³¹ In	¹³² In	¹³³ In	¹³⁴ In	¹³⁵ In	¹³⁶ In	¹³⁷ In			
48	¹²³ Cd	¹²⁴ Cd	¹²⁵ Cd	¹²⁶ Cd	¹²⁷ Cd	¹²⁸ Cd	¹²⁹ Cd	¹³⁰ Cd	¹³¹ Cd	¹³² Cd	¹³³ Cd	¹³⁴ Cd					
	75		77		79		81	83		85		87		89			N

Another Summation Calculation

E_i : Energy converted into heat in the reactor following fission, used to calculate a nuclear reactor's antineutrino spectrum:

$$\frac{d^2\phi(E, t)}{dEdt} = \frac{W_{th}(t)}{\sum_i f_i(t) e_i} \sum_i f_i(t) S_i(E) c_i^{ne}(E, t) + S_{SNF}(E, t),$$

Target	E_i (Kopeikin 2004)	% Δ	E_i (Ma2013)	% Δ
235U	201.92+-0.46	0.22%	202.36+-0.26	0.12%
238U	205.52+-0.96	0.47%	205.99+-0.52	0.25%
239Pu	209.99+-0.60	0.29%	211.12+-0.34	0.16%
241Pu	213.60+-0.65	0.30%	214.26+-0.33	0.15%

E_i s are derived using nuclear data.
Uncertainties seem unreasonably low.

Another Summation Calculation

E_i is calculated as

$$E_i = M_{ti} - \sum C_{ik} M_k - (N_{pi} - 1) M_n - E_{\bar{\nu}i} - \Delta E_{\beta\gamma i} + E_{nci},$$

Summation!

M: mass excess

C: cumulative fission yield for the most stable fission product

N_p : prompt neutron multiplicity

$E_{\bar{\nu}i}$: energy carried away by the antineutrinos

Will briefly discuss $\sum CM$ and $E_{\bar{\nu}i}$ next

Another Summation Calculation

$$Y = \sum C_i M_i,$$

$$\Delta^2 Y = \sum_i (\Delta C_i M_i)^2 + (C_i \Delta M_i)^2 +$$

$$\sum_i \sum_{j \neq i} M_i M_j \Delta C_i \Delta C_j \text{Cor}(C_i, C_j) +$$

$$\sum_i \sum_{j \neq i} C_i C_j \Delta M_i \Delta M_j \text{Cor}(M_i, M_j),$$

Very preliminary results

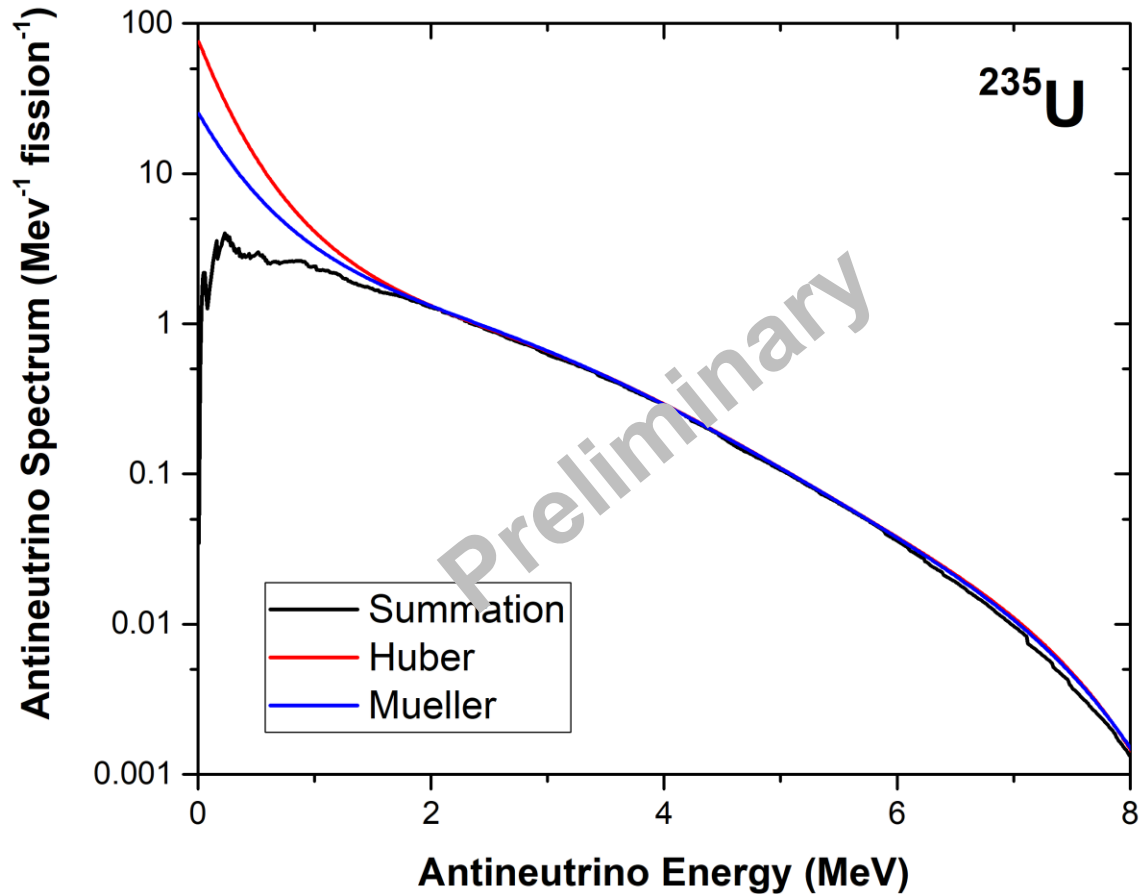
Target	Y (MeV)	ΔY (MeV) no CFY cors.	ΔY (MeV) with CFY cors
^{235}U	-173.27	0.73	0.91
^{238}U	-173.06	2.11	3.24
^{239}Pu	-173.56	2.41	3.06
^{241}Pu	-173.57	3.29	4.30

Energy carried away by antineutrinos

For energies lower than 2 MeV, one can only use Summation

Vogel is rendered obsolete by Mueller/BNL

Mueller and Huber are not independent as they are based on the ILL data. Note nice agreement with Summation.



For ^{235}U we obtain:

$$3.89 \pm 0.13 \text{ (0-2 MeV)} + 5.28 \pm 0.13 \text{ (>2 MeV)} = 9.17 \pm 0.18 \text{ MeV}$$

$$\text{For } ^{239}\text{Pu}: 3.54 \pm 0.08 + 5.47 \pm 0.17 = 9.01 \pm 0.27 \text{ MeV}$$

$$\text{For } ^{241}\text{Pu}: 3.98 \pm 0.20 + 5.01 \pm 0.17 = 9.01 \pm 0.27 \text{ MeV}$$

Comparison with ENDF/B energies

MT=1 MF=458

	235U		238U E7.1		239Pu E7.1		239Pu E8.0		241Pu	
Fragments KE	1.69E+08	4.90E+05	1.70E+08	4.90E+05	1.76E+08	4.00E+05	1.76E+08		1.75E+08	6.80E+05
Prompt neutrons	4.84E+06	7.00E+04	4.56E+06	1.00E+05	6.13E+06	1.00E+05	6.07E+06		5.99E+06	1.30E+05
Delayed neutrons	7.40E+03	1.11E+03	1.80E+04	2.70E+03	2.99E+03	4.40E+02	3.14E+03		5.00E+03	1.00E+03
Prompt gammas	6.60E+06	5.00E+05	6.68E+06	5.30E+05	6.74E+06	4.70E+05	6.37E+06		7.64E+06	6.90E+05
Delayed gammas	6.33E+06	5.00E+04	8.25E+06	7.00E+04	5.17E+06	6.00E+04	5.17E+06	6.00E+04	6.40E+06	9.00E+04
Delayed betas	6.50E+06	5.00E+04	8.48E+06	8.00E+04	5.31E+06	6.00E+04	5.31E+06	6.00E+04	6.58E+06	9.00E+04
Delayed neutrinos	8.75E+06	7.00E+04	1.14E+07	1.10E+05	7.14E+06	9.00E+04	7.14E+06	9.00E+04	8.85E+06	1.20E+05
Total - neutrinos	1.93E+08	1.50E+05	1.98E+08	3.16E+05	1.99E+08	1.09E+06	1.99E+08		2.02E+08	2.77E+05
Total	2.02E+08	1.30E+05	2.09E+08	3.00E+05	2.06E+08	1.18E+06	2.06E+08		2.11E+08	2.50E+05
Ma 13	1.93E+8		1.95E+08		1.99E+8		1.99E+8		2.02E+08	
ENDF/B-Ma13	5.54E+04		3.13E+06		-8.38E+05		-1.16E+06		1.01E+04	

ENDF/B energy values and uncertainties are from systematics. Values Could be improved.

Noticed some deviations of **3 MeV** for ^{238}U and **1 MeV** for ^{239}Pu .

Conclusions

- ❑ We introduced for the first time fission yield correlation matrices in summation calculations.
- ❑ We observe sensible changes in the uncertainties if these matrices are included. A method to develop high-fidelity correlation matrices should be developed later.
- ❑ We need to reach consensus about summation uncertainties in yields and energies.
- ❑ Formats, we need to extend ENDF-6 to accommodate correlation matrices for proper documentation and storage of the work done.