Fission Product Yields for Neutrino Physics and Non-proliferation

The nuclear physics properties of fission products are important for many fields

- Neutrino Physics
- Nuclear Forensics
- Monitoring Reprocessing Facilities
- Weapons Physics
- Reactor Safeguards
- Stellar Astrophysics
- Nuclear Medicine

In many cases more detailed experimental data is needed
- Here are some examples from from the first two fields

Neutrino Physics

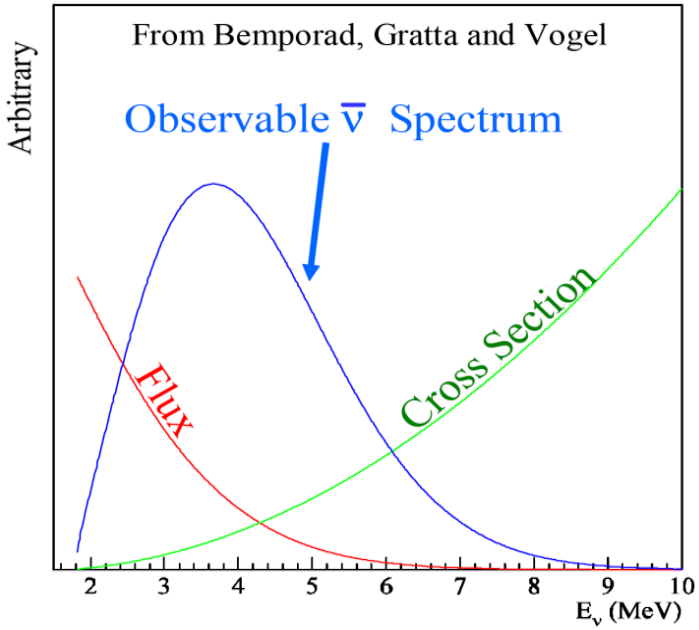
Reactors represent intense sources of anti-neutrinos and are used for neutrino oscillation studies

A 3 GW reactor emits 10^{21} antineutrinos per second
- All produced by the beta-decay of fission products

$$E_\nu \sim 0 - 10 \ \ MeV$$

Detected by Reines & Cowan via:

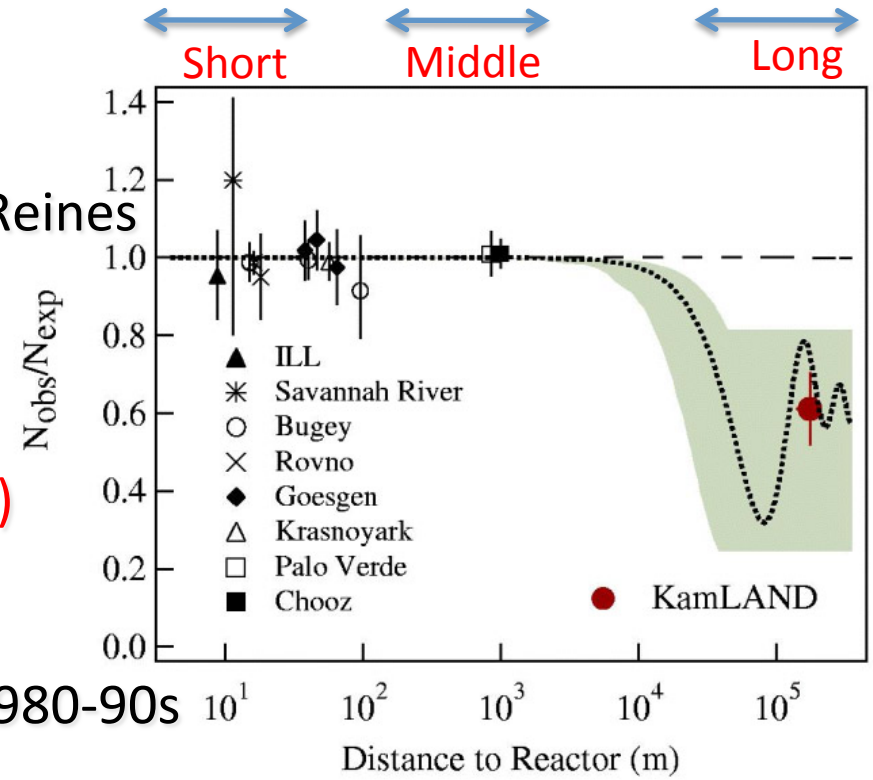
$$\bar{\nu}_e + p \rightarrow n + e^+$$



Reactor Neutrino Experiments

Discoveries since the 1950s:

- Neutrino discovered in 1953-56 by Reines and Cowan.
- Short baseline expts determined antineutrino spectrum/flux (maybe)
- Upper limit of mixing angle θ_{13} to $\sin^2 2\theta_{13}<0.17$ (Chooz, Palo Verde),1980-90s
- Anti-neutrino disappearance at KamLAND in 2003.
- Precision measurements of θ_{13} (Daya Bay, Double Chooz, RENO), 2014-present

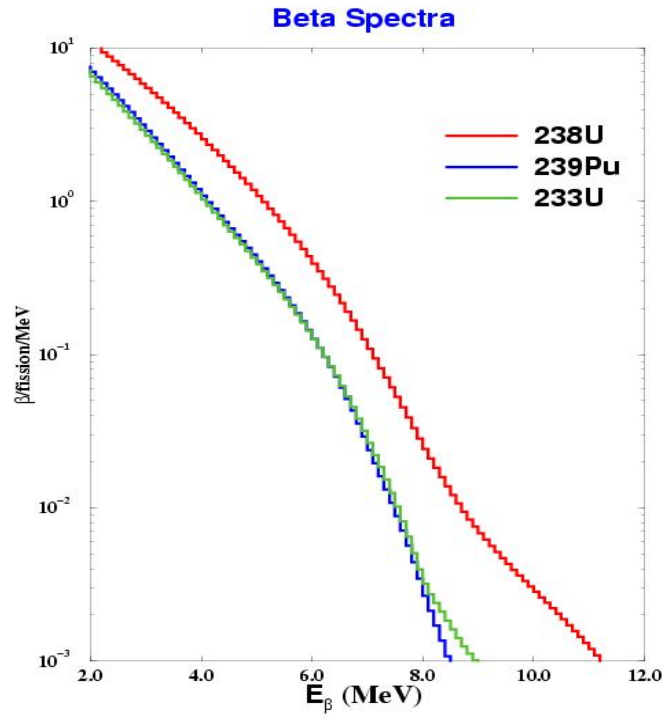


Anti-neutrino spectra are determined by the cumulative yields and the beta-decay spectra of the fission products

Equilibrium aggregate beta spectra:

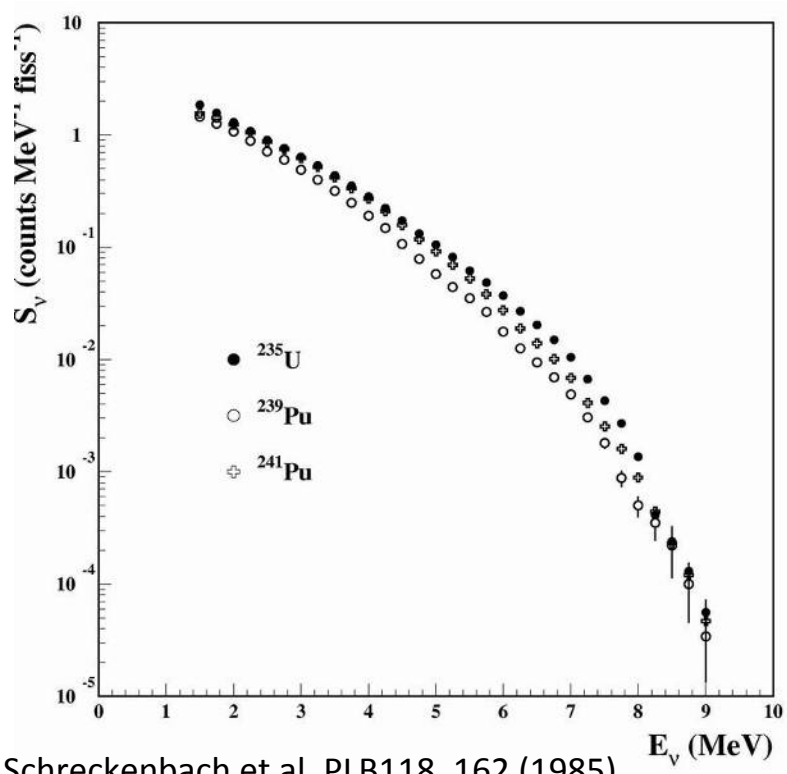
$$S_k(E) = \sum_{FF} Y_{FF}(Z,A,m) S(E,Z,A,m)$$

$$S(E,A,Z,m) = \sum_i B^i S(E,Z,A,m,E_0^i)$$



- Spectra are quite actinide dependent
- β-decay branching ratios and end-point energies only known for ~95% of the decays
- Many of the cumulative fission yields are uncertain

Neutrino community determined the antineutrino spectra from measurements of aggregate beta-decay spectra



K. Schreckenbach et al. PLB118, 162 (1985)
A.A. Hahn et al. PLB160, 325 (1989)

- Measurements at ILL of thermal fission beta spectra for ^{235}U, ^{239}Pu, ^{241}Pu
- Converted to antineutrino spectra by fitting to 30 end-point energies
- Use Vogel *et al*. and Mueller *et al*. database estimates for ^{238}U
  ^{238}U ~ 7-8% of fissions =>small error

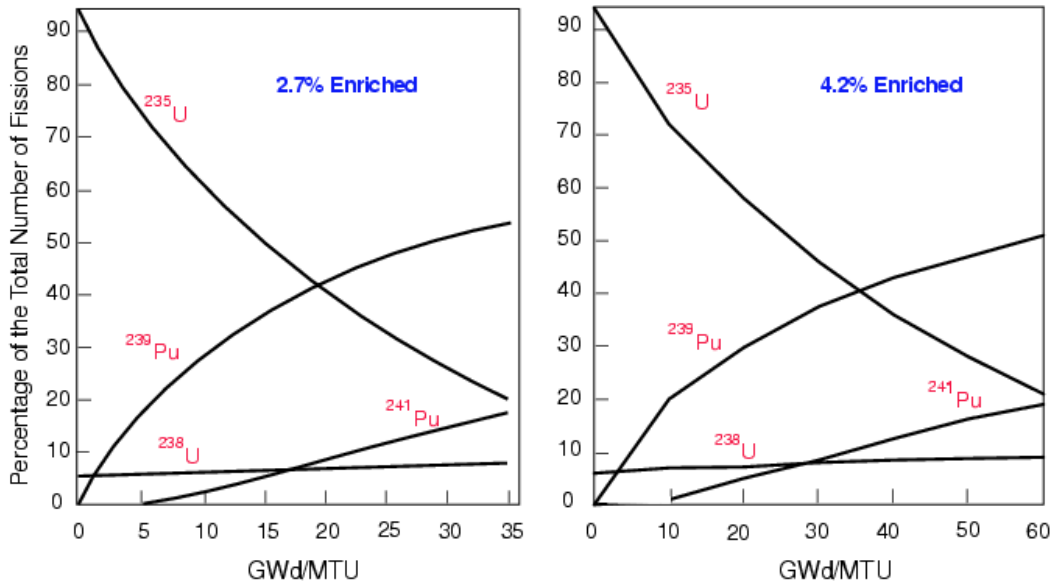
FIT

$$S_\beta(E) = \sum_{i=1,30} A_i S^i(E, E_o^i)$$

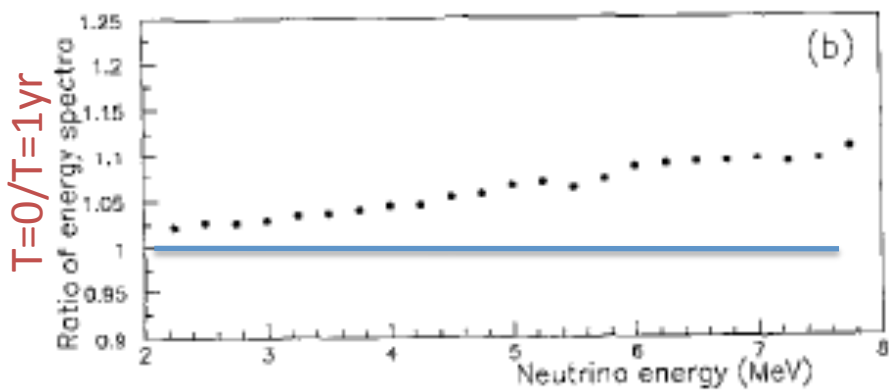
$$S^i(E, E_0^i) = E_\beta p_\beta (E_0^i - E_\beta)^2 F(E, Z)$$

As Burn Proceeds:
Different Combination of Isotopes Fissioning

^{239}Pu steadily grows in via:
^{238}U+n→^{239}U→^{239}Np→^{239}Pu
Followed by higher mass Pu



This change translates into a change in the antineutrino spectrum emitted from the beginning to the end of a burn cycle



⇒ 5% fewer detected antineutrinos after 1 yr
⇒ Must be taken into account in osc expts.
⇒ Basis of non-proliferation schemes

Bugey-3 energy-dependent decrease after 1 year

Calculating the antineutrino flux at a given reactor

1. Measure **thermal power** in primary & secondary loop
W_{th} uncertainty ~<2%, but KamLAND quote 0.6-0.7%
Uncertainty usually dominated by water flow measurement.

2. Calculate **number of fissions** from W_{th}
E_f good to ~0.5-1% (?)

Kopeiken et al. Phys. Atom Nucl. 67 (2004) 1892

$$N_f = W_{th} / E_f$$

$$E_f = E_{tot} - < E_\nu > - \Delta E_{\beta\gamma} + E_{(n,\gamma)}$$

3.Power company reactor **burn simulation** to determine which isotopes are fissioning

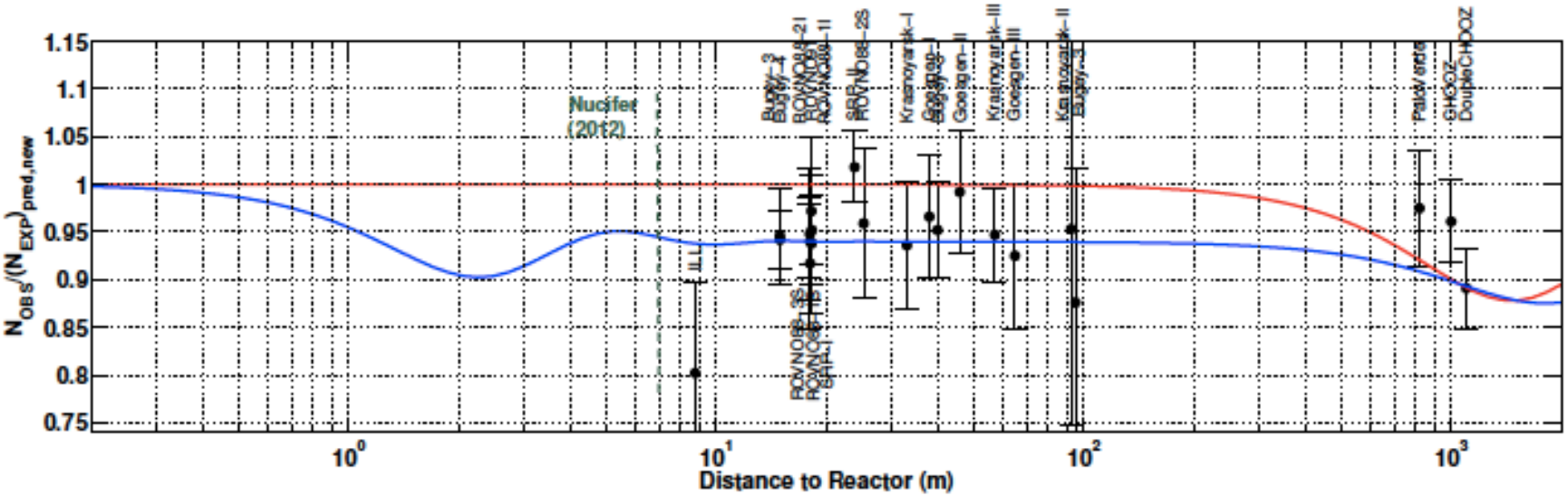
$$S(E_\nu) = \sum_i^{isotopes} f_i S_i(E_\nu)$$

4. Knowledge of **individual antineutrino spectra** for each fissioning isotope

4. has recently become a serious problem – observing an anomaly and a `bump'

Mention *et al.* PRD 073006 2011.

Short Baseline Reactor Anti-Neutrino Anomaly



Improved Antineutrino Spectra: Th. A. Mueller, et *al. PRC, C83 (2011*) 054615

A reanalysis of the conversion of the aggregate beta-spectrum to an anti-neutrino spectrum

⇒ All short baseline experiments saw ~6% too few antineutrinos

⇒ If oscillations would require 1 eV sterile neutrino

Known corrections to β-decay are the main source of the anomaly

$$S(E_e,Z,A) = \frac{G_F^2}{2\pi^3} p_e E_e (E_0 - E_e)^2 C(E) F(E_e,Z,A)(1+\delta(E_e,Z,A))$$

Fractional corrections to the individual beta decay spectra:

$$\delta(E_e,Z,A) = \delta_{rad} + \delta_{FS} + \delta_{WM}$$

δ_{rad} = Radiative correction (used formalism of Sirlin)

δ_{FS} = Finite size correction to Fermi function

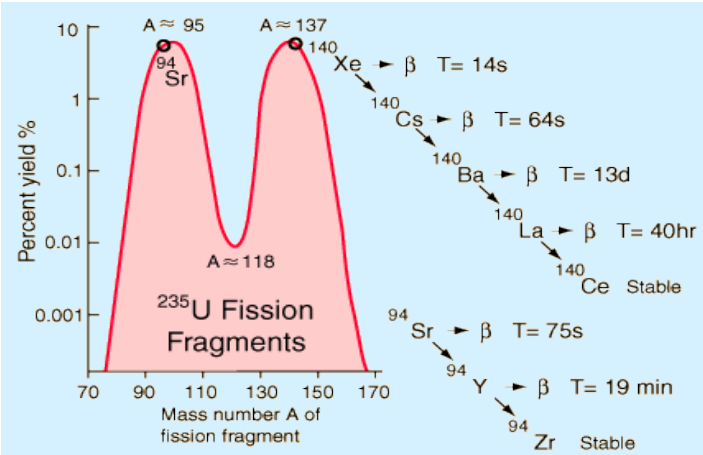
δ_{WM} = Weak magnetism

Originally approximated as:

$$\delta_{FS} + \delta_{WM} = 0.0065(E_\nu - 4\,MeV))$$

The difference between this original treatment and an improved treatment of these corrections is the main source of the anomaly

However, 30% of the transitions are forbidden => the uncertaunty in the anomaly large



A~95 Peak

Br, Kr, Rb, Y, Sr, Zr mostly

Nb, Mo, Tc often allowed

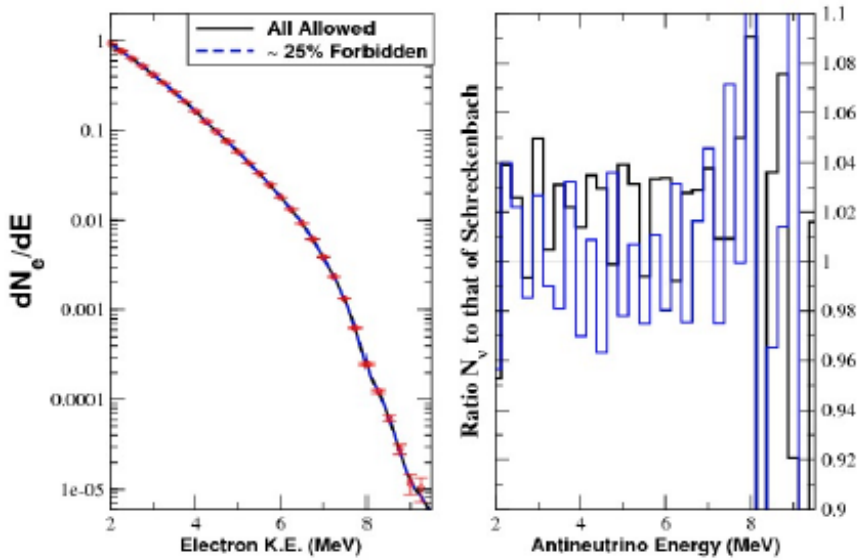
A~ 137 Peak

Sb, I, Te, Xe, Cs, Ba, Pr, La

- mostly forbidden

Forbidden:
Not Fermi (0+) or GT (1+)
i.e, $\Delta L>0$, $\Delta\pi=+/-1$

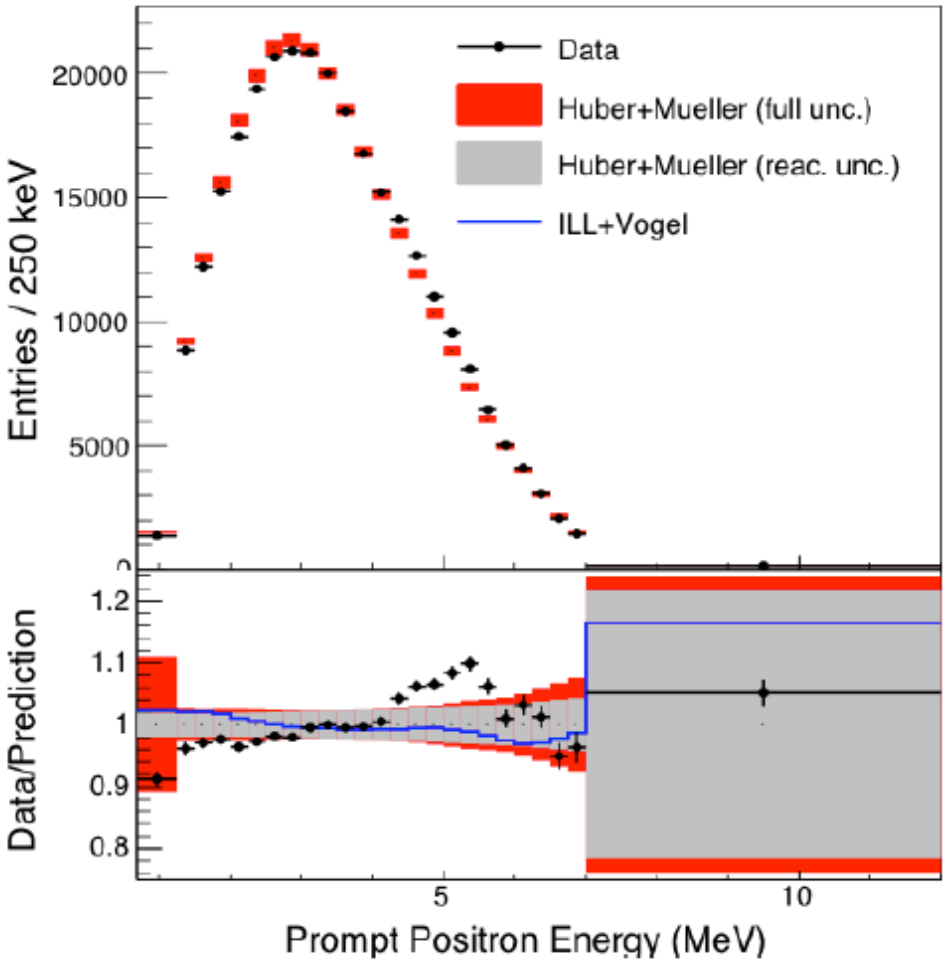
The forbidden transitions tend to dominate the high energy component of spectrum
This makes the weak magnetism and finite size correction quite uncertain

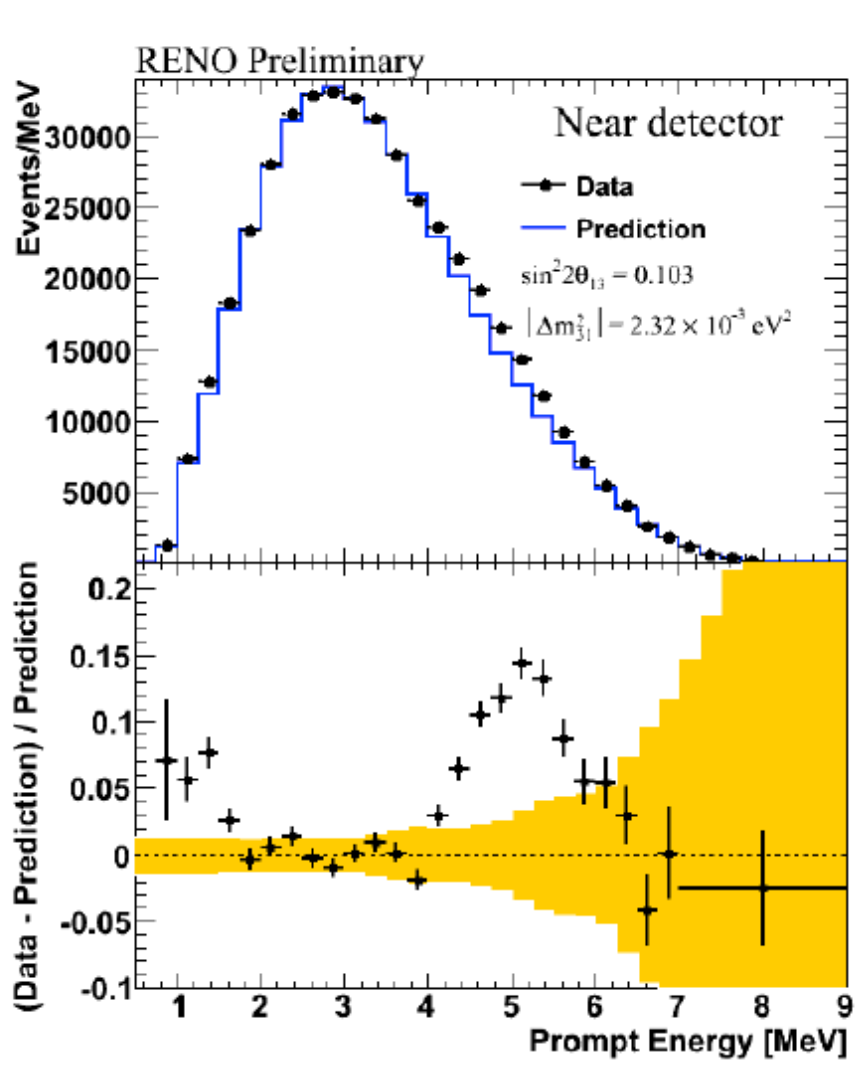


If all allowed the fit to Schreckenbach
=> Antineutrino spectrum +3%

If 25% forbidden
⇒ Find an equally good fit with no increase in the antineutrino spectrum

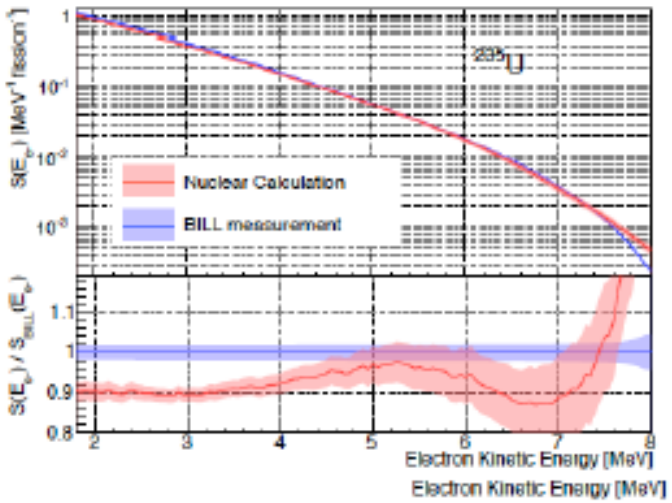
A significant shoulder (the bump) is seen in the near and far detectors at all current reactor neutrino experiments at E_{prompt} =4-6 MeV

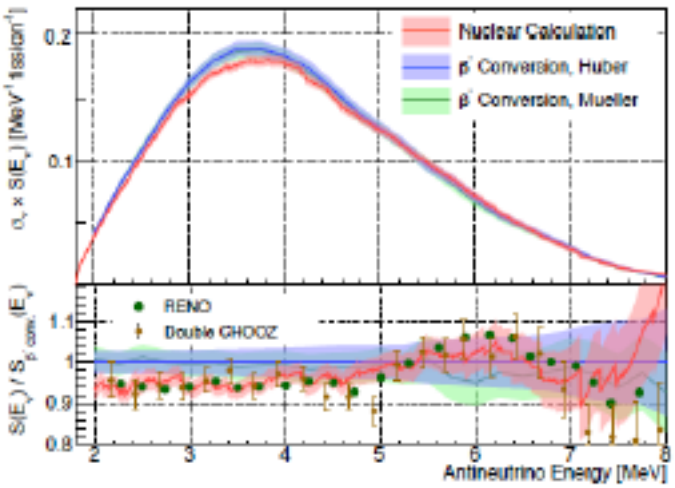




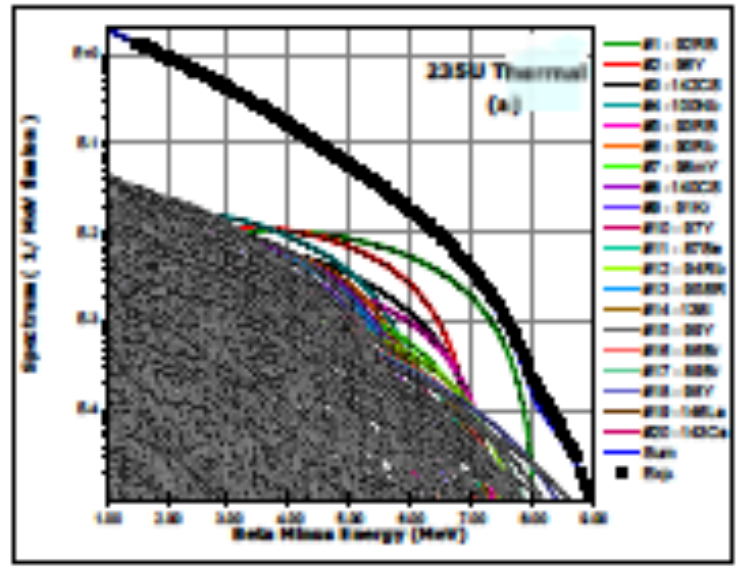
Eν=Eprompt + 0.78 MeV

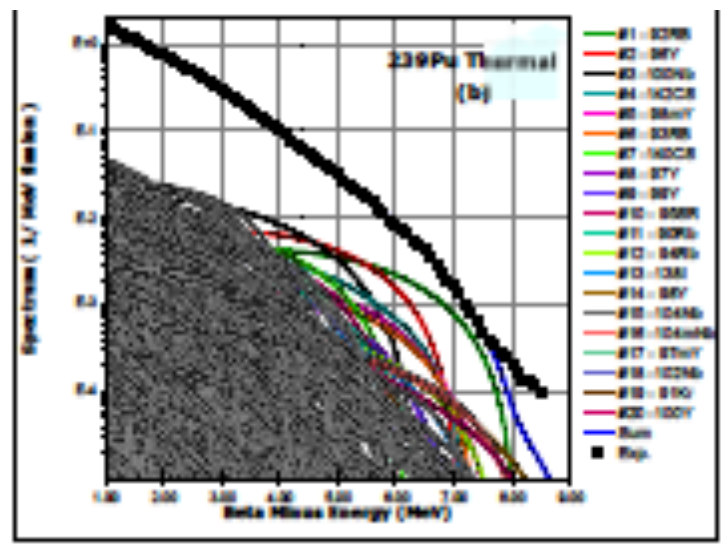
Analyses of the Bump using ENDF versus JEFF cumulative yields differ





ENDF, Dwyer + Langford,
Phys. Rev. Lett. 114 012502 (2015)

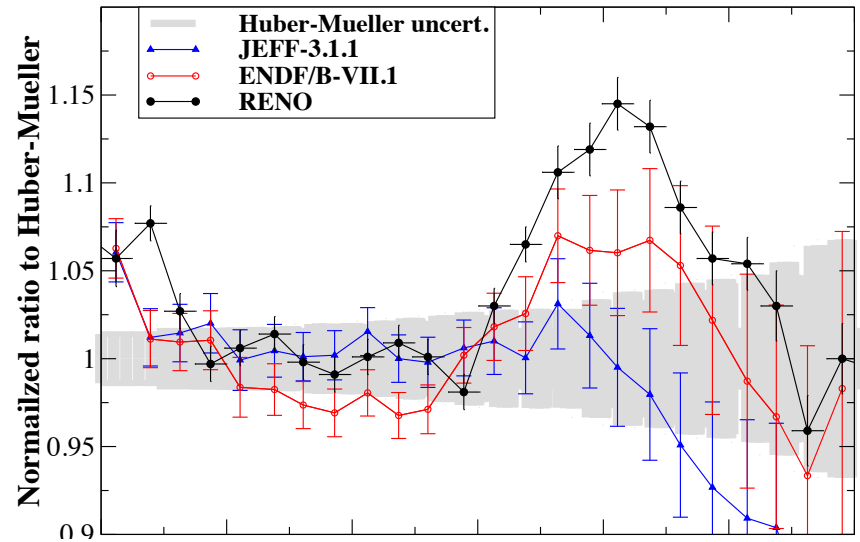


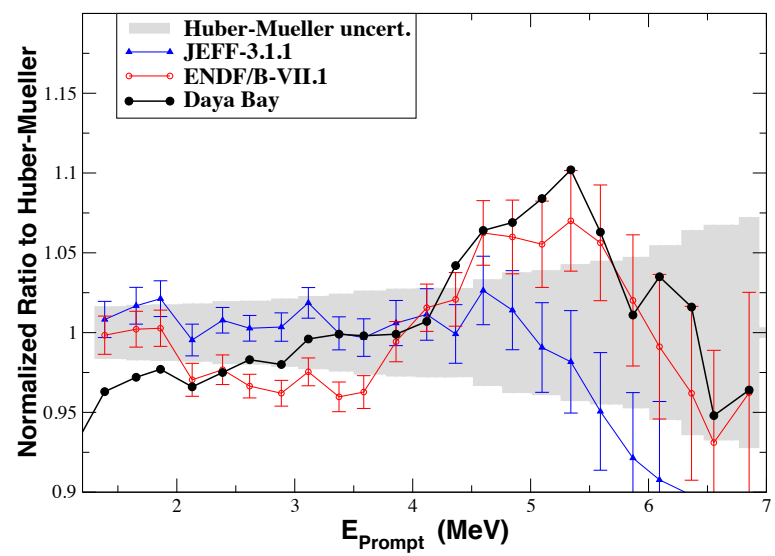


A. A. Sonzogni, T. D. Johnson, and E. A. McCutchan, Phys. Rev. C 91, 011301(R) (2015)

ENDF versus JEFF predictions for the BUMP

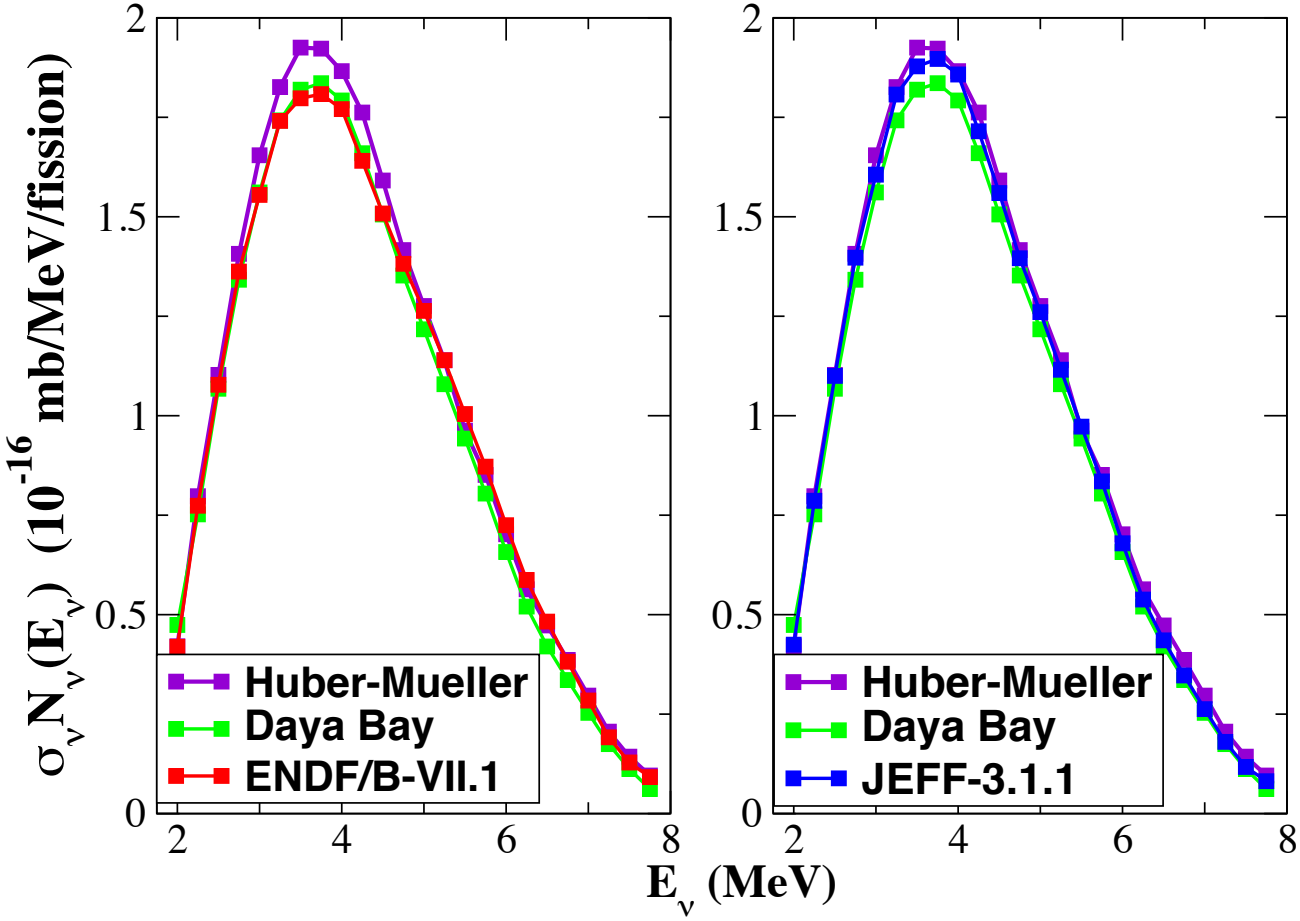
Nucleus	JEFF Y_{F_i} (%)		ENDF Y_{F_i} (%)	
	^{235}U	^{239}Pu	^{235}U	^{239}Pu
^{89}Br	1.36	0.50	1.08	0.35
^{90}Br	0.49	0.10	0.56	0.25
^{95}Rb	0.66	0.26	0.77	0.44
^{96}Y	4.72	2.88	6.0	4.35
^{97}Y	2.08	1.22	4.89	3.75
^{98}Y	1.07	0.68	1.92	1.52
^{98m}Y	1.97	1.87	1.11	1.19
^{100}Y	0.30	0.21	0.61	0.35
^{134m}Sb	0.52	0.19	0.36	0.20





The two evaluations disagree on many of the fission yields for nuclei dominating the bump

ENDF versus JEFF predictions for the normalization of the antineutrino spectra

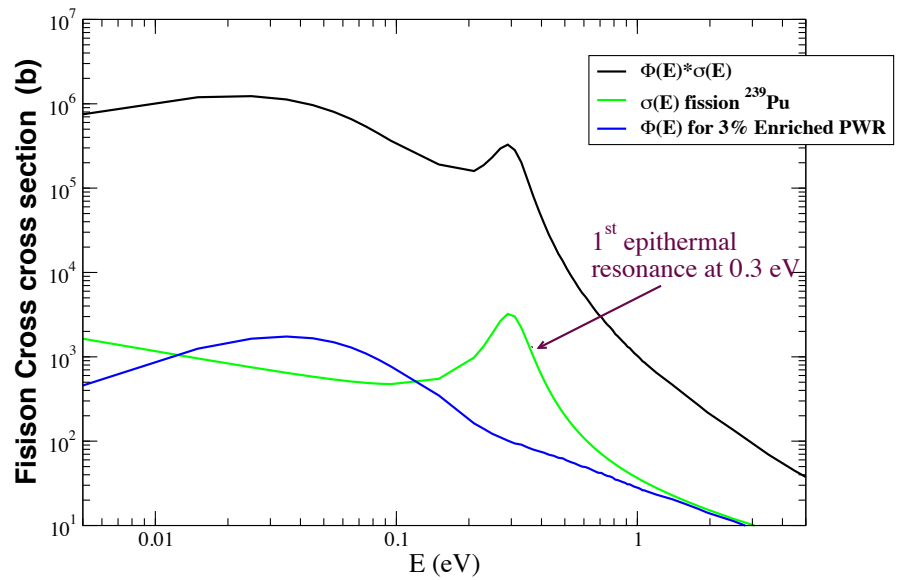


The ENDF/B-VII.1 predictions are closer to Daya Bay , while the JEFF-3.1.1 predictions are closer to the Huber-Mueller model that suggests an anomaly

The hardness of the reactor spectrum could be an issue

The original fission aggregate beta spectra were measured at ILL, France
- Heavy water reactor
⇒ Extremely thermal

The Daya Bay, RENO and Double Chooz measurements are all at PWR reactors
⇒ Significant epithermal neutron component to the flux



Could the JEFF-3.1.1 yields be closer to thermal
And the ENDF/B-VII.1 yields be thermal/epithermal?

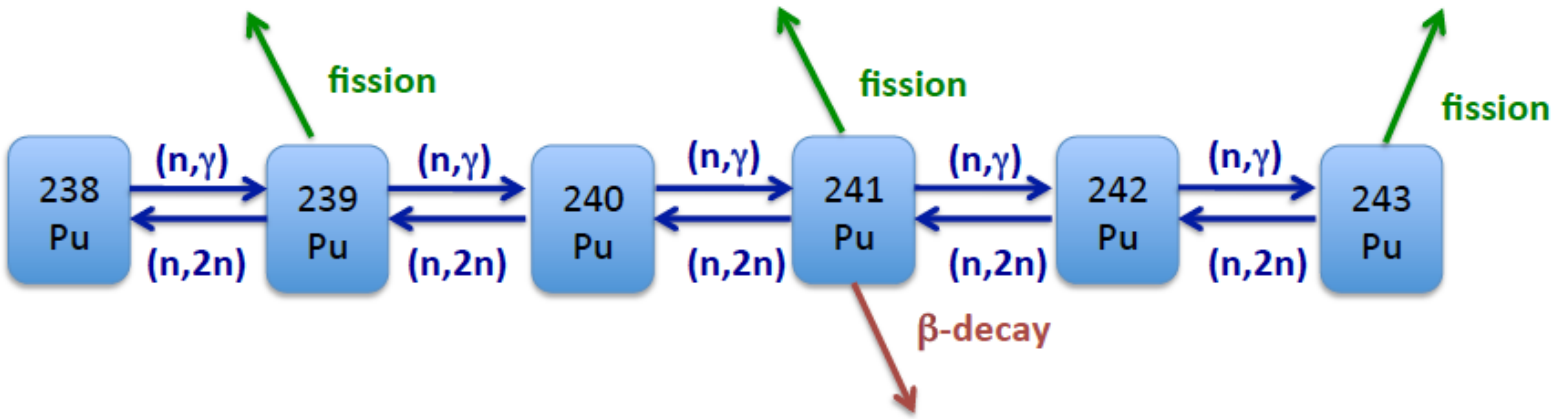
Experimental measurements of the dominant fission yields would be very valuable in addressing the these neutrino issues

Nuclear Forensics

Nuclear Forensics relies on measuring the actinide ratios in the debris

Timescale is controlled by the difficult U & Pu chemistry:

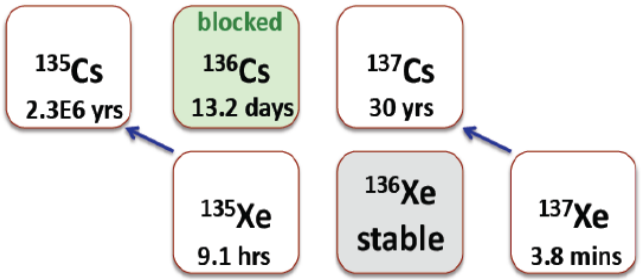
- Separate key actinides and measure their isotopic ratios by mass spectroscopy.

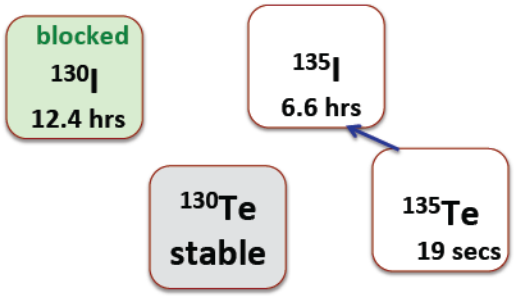


- Actinides provide the most detailed information
- But there is a pressing need to get key information on a faster timescale

The volatile fission products could be collected and measured rapidly by flying into the plume

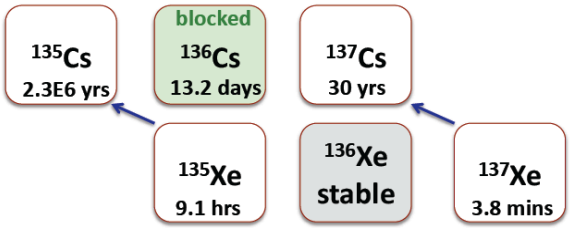
1. Collect the volatile fission products on special filters
2. Assay key fission products on a rapid timescale using γ-ray spectroscopy
3. Deduce the fuel and whether the device was boosted from blocked Cs and I isotopes



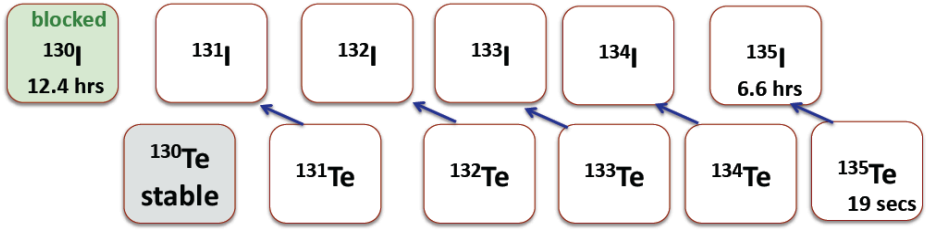


Timescale possible because no major chemical separation is needed for Cs and I

Sensitivity of Cesium and Iodine to the type of explosion - blocked fission products retain fission information



The required cesium R-values are measured
But the iodine isotopes of interest are not



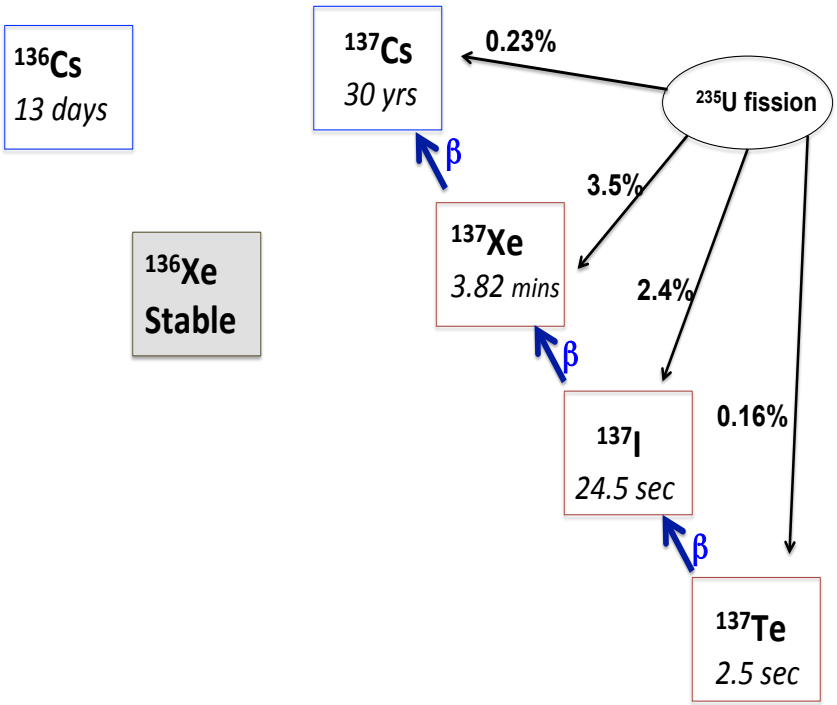
Radiochemistry always evaluated in terms of R-values:

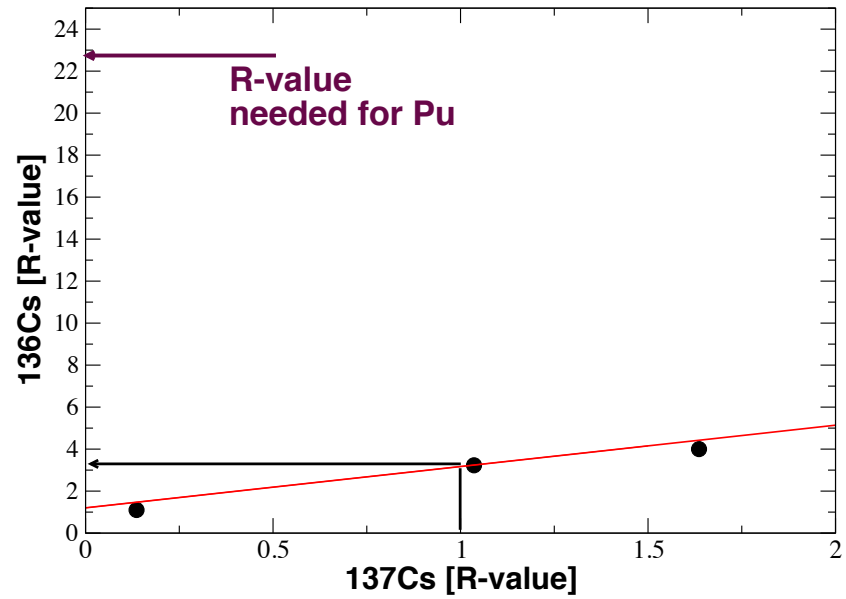
$$R = \left({}^{Y_x}\!/_{99Mo} \right) \Big/ \left({}^{Y_x}\!/_{99Mo} \right)_{thermal}$$

Fission type	136Cs	137Cs	130I	135I
235U 1 MeV	2.05	1.003	0.08	1.0
235U 14 MeV	45.4	1.015	138	0.78
239Pu 1 MeV	22.7	1.03	20.8	1.02
239Pu 14 MeV	164	0.905	655	0.68

Varies by orders of magnitude — **Always ~1** — **Varies by orders of magnitude** — **Close to ~1**

Debris Fractionation
- always occurs but can be corrected for





The transport of ^{137}Cs depends ^{137}I and ^{137}Xe.

^{136}Cs has no precursors.

=> ^{136}Cs and ^{137}Cs from one another.

Fractionation is corrected for by using the fact that the 137Cs/99Mo ratio is close to unity for all fissions

Uncertainties in the method are small compared to the sensitivity of ^{136}Cs/^{99}Mo to the nature of the fission

Summary

- **Reactor neutrino physics is currently facing two major puzzles**
  - The anomaly , which is a 6% deficit in the antineutrino flux at all short baseline experiments
  - The shoulder (bump) at $E\nu$=4.5-6.5 MeV
  - ENDF and JEFF give very different predictions for these because their yields for the important fission product are different
  - handful of fission products, that dominate the high-energy spectrum, need to be measured

- **Nuclear Forensics could involve a new early phase**
  - The blocked cesium and iodine fission products can be used to determine whether the fuel is uranium and plutonium and whether 14 MeV fission is involved
  - the ^{130}I and ^{135}I thermal, fast and 14 MeV fission yields need to be measured

Summary cont.

- **Nuclear Forensics could involve a new early phase**
  - The blocked cesium and iodine fission products can be used to determine whether the fuel is uranium and plutonium and whether 14 MeV fission is involved
  - the ^{130}I and ^{135}I thermal, fast and 14 MeV fission yields need to be measured