Combining Reactor Fission Gas Data with Antineutrino Data

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Abstract

In previous work [1], we have shown that the average thermal neutron flux that reactor fuel has been exposed to can be determined from a measure of the $^{137}Cs/^{135}Cs$ ratio. However, this is only possible if the number of reactor shutdowns is know. Here we simply point out that, since antineutrino measurements can determine [2,3] the number of reactor shutdown and the total irradiation time, coupling fission gas measurements with antineutrino measurements would allow one to extract the $^{240}Pu/^{239}Pu$ ratio in reactor fuel.

1. A.C. Hayes and G. Jungman, Nucl. Insts. and Meth. A 690, 68 (2012).
2. N. Bowden, *et al.,* Nucl. Inst. Meth. A, 572 (2007) 985-998
3. Andriamirado *et al.,* PRD103, 03201 (2021).

Goal: To determine the grade of Pu of reactor spent fuel from fission gas and antineutrino measurements

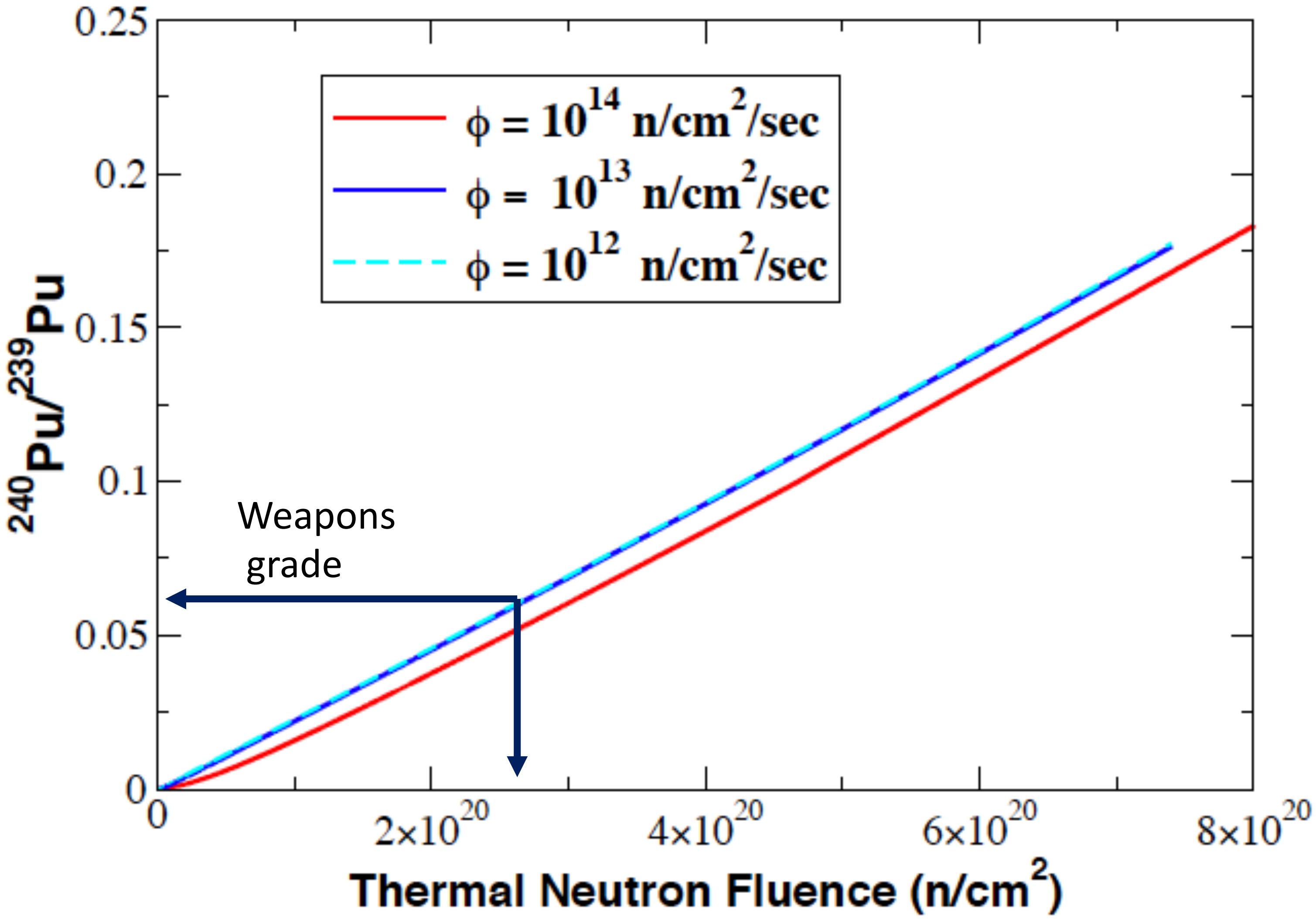
Assumptions:

1. The original unirradiated fuel is some form of uranium $^{238}U + ^{235}U$, but of unknown enrichment.

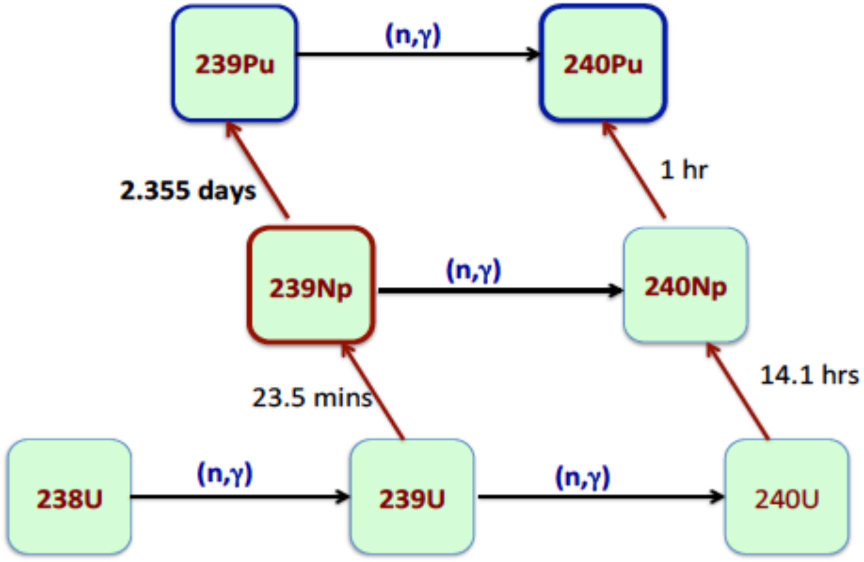
2. During the entire burn of the fuel antineutrino monitoring was sufficiently accurate to determine when the reactor was on or off.

3. Fission gases produced in the the fuel during the burn can be measured.

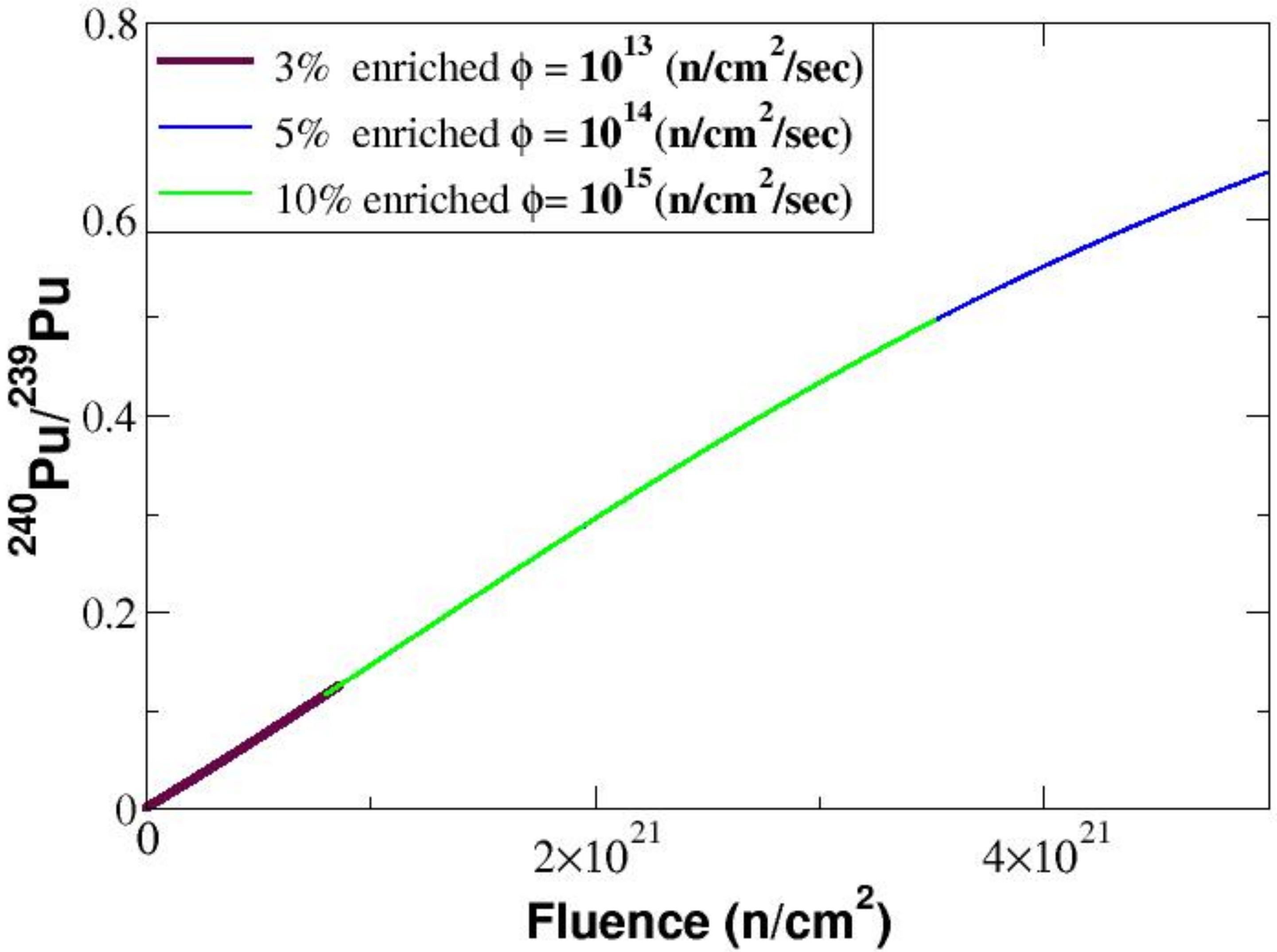
The grade of the plutonium ($^{240}Pu/^{239}Pu$) in any uranium fuel depends on the total neutron fluence (n/cm^2) of the irradiation



- For very high flux ~ 10^{14} n/cm^2/sec the curve depends on the flux because of the time for ^{239}Np to equilibrate.
- But the gases determine the flux, so we always know which curve we're on.

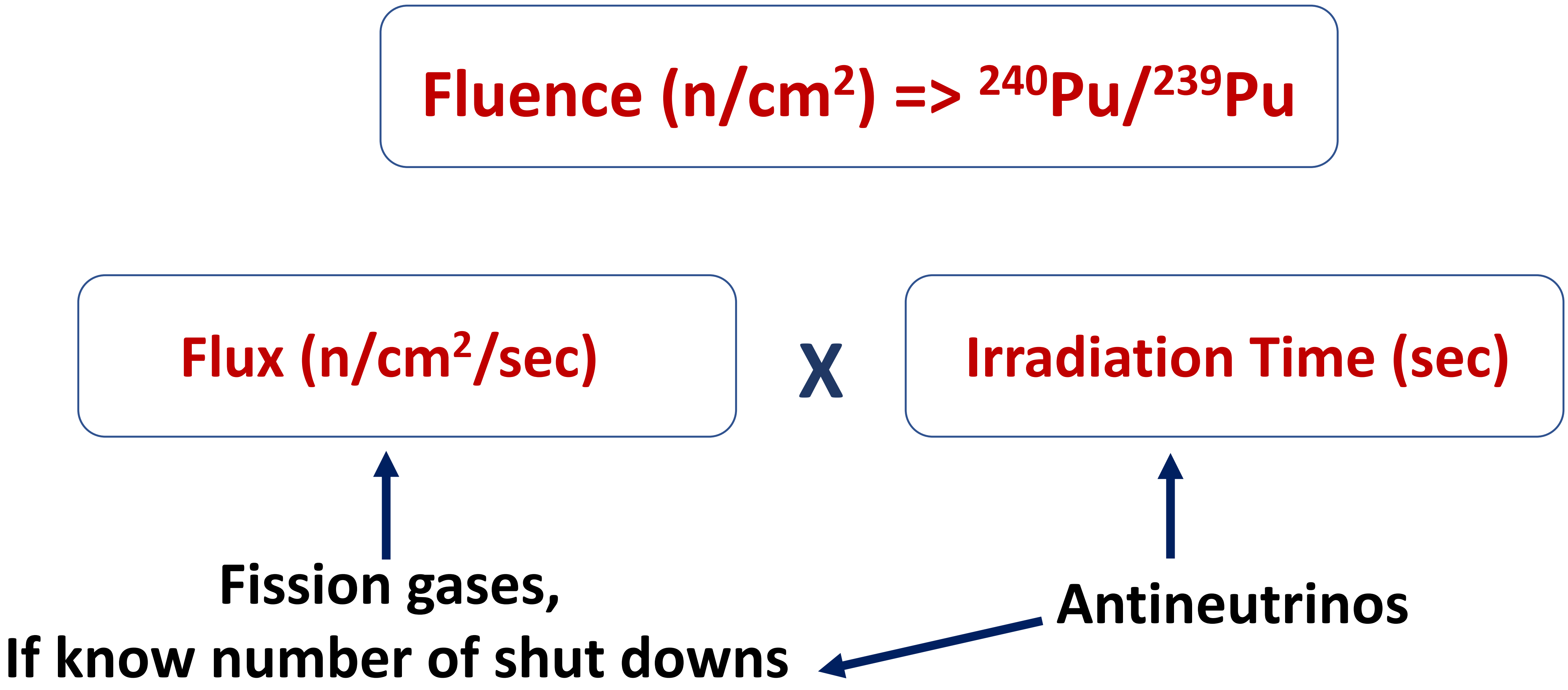


The enrichment of the fuel does not effect the Pu grade - only the fluence (and flux if very high) matter

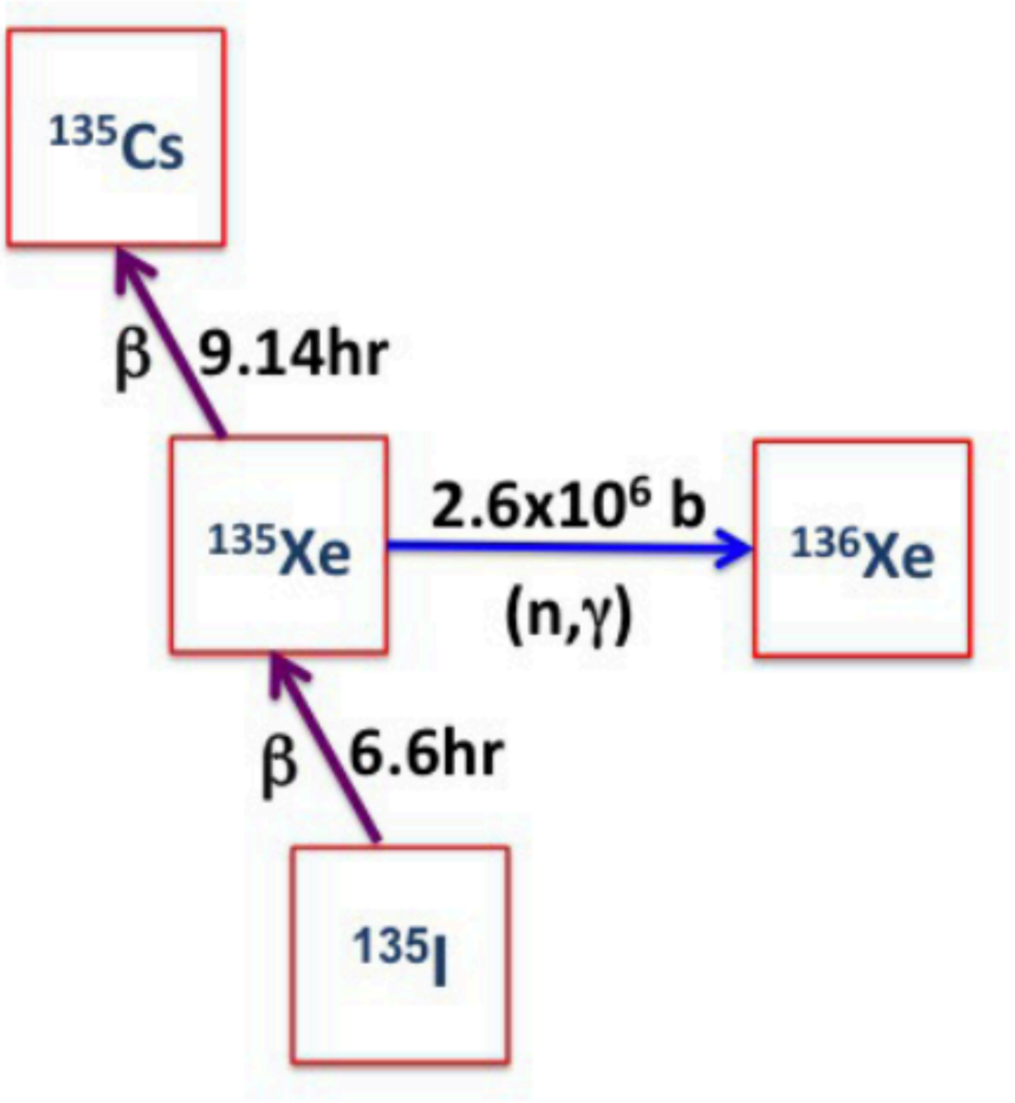


In these simulations we artificially set the half-life of ^{239}Np to 1 hour (instead of 2.3 days) to suppresses the dependence on flux.

To determine the total fluence we divide the problem into one of determining the flux and irradiation time separately



Determining the flux

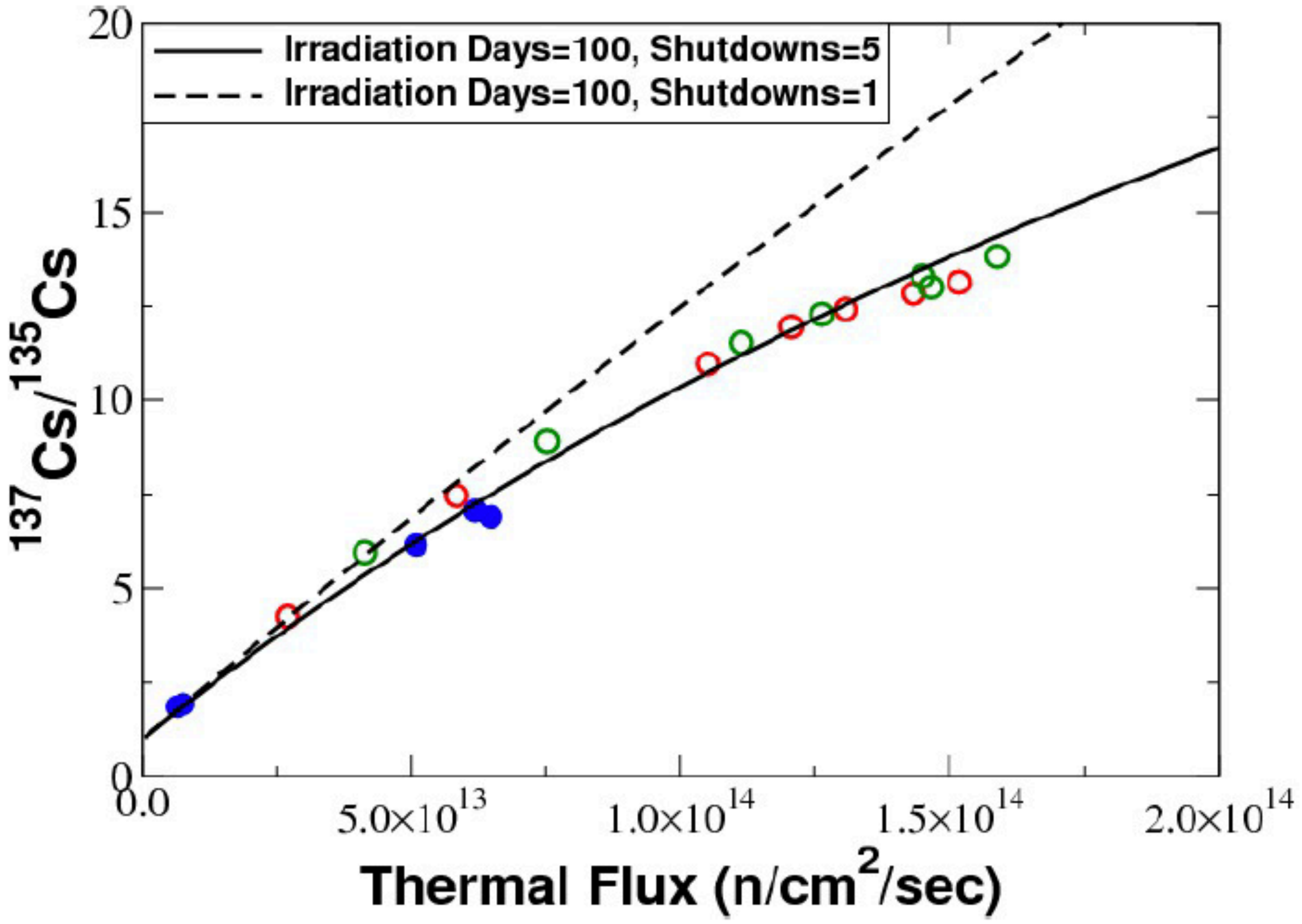


The competition between:

$^{135}Xe \rightarrow {}^{135}Cs$ and $^{135}Xe(n,\gamma)\,{}^{136}Xe$

causes the ^{136}Xe and ^{135}Cs to be sensitive functions of the thermal neutron flux.

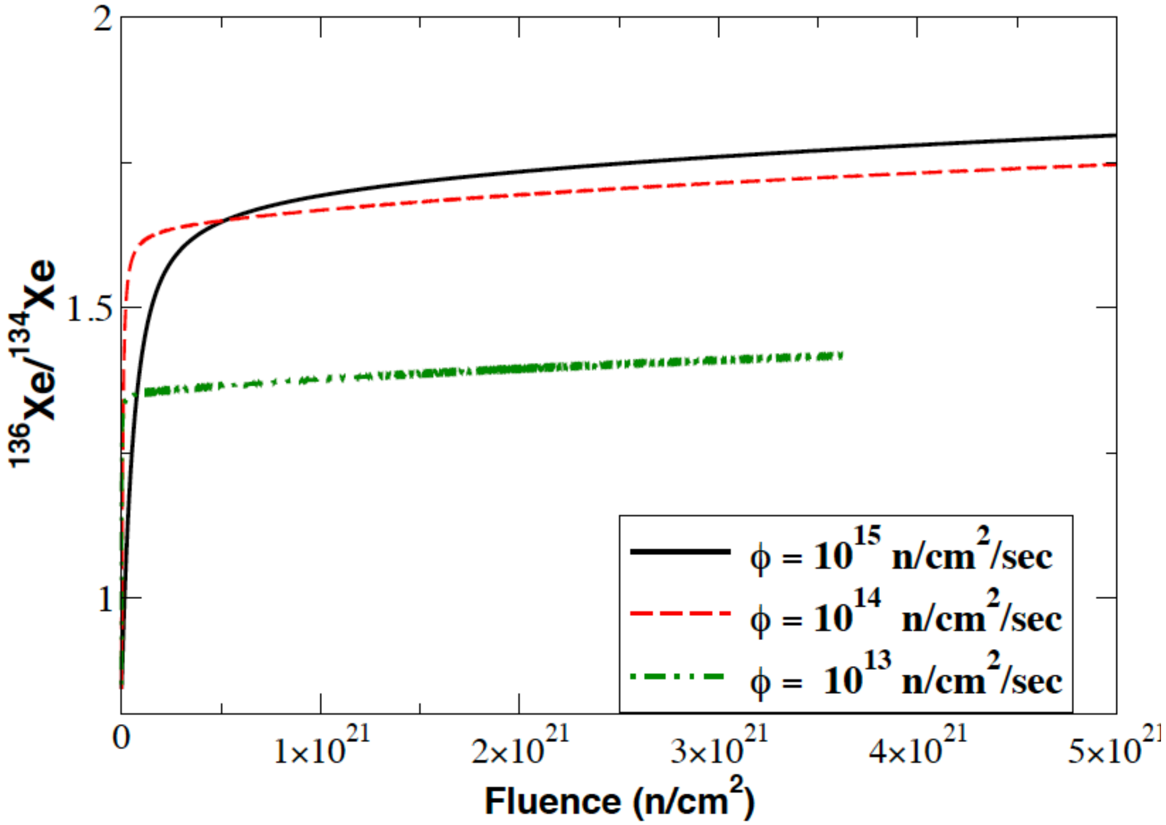
$^{137}Cs/^{135}Cs$ scales linearly with the flux, but reactor shutdowns affect the ratio significantly at high flux

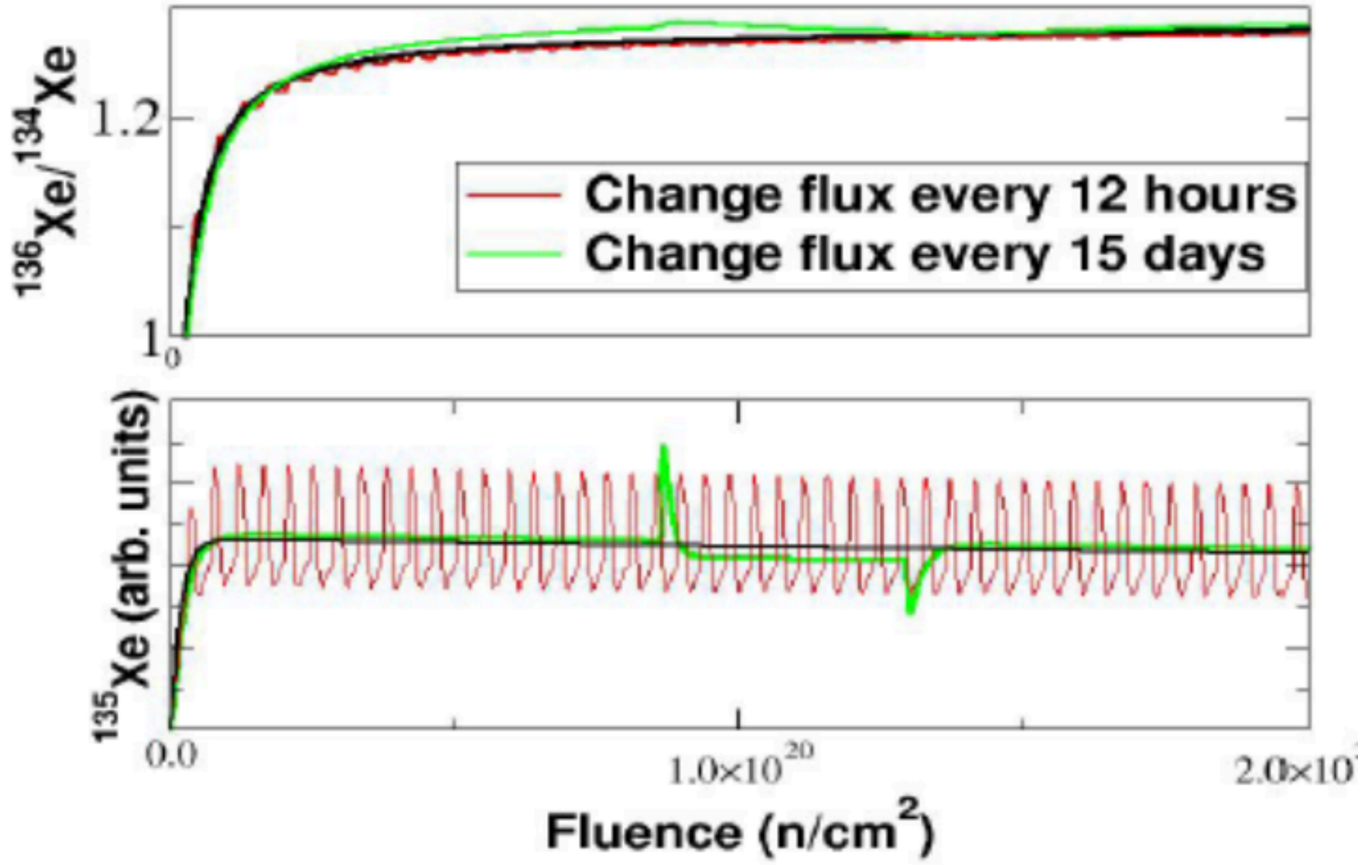


99% ^{235}U enriched samples
Irradiated 185 days over 320 days

94% ^{235}U enriched samples
Irradiated 100days over 35 days

Reactor neutron flux variations are a classic way to spoof many antineutrino schemes, but ^{135}Cs and ^{136}Xe always measure the the average thermal neutron flux, which is what determines the Pu grade.





$^{136}Xe/^{134}Xe$ also a flux diagnostic, but less sensitive than ^{135}Cs

Analytic expression can be derived for the Cs and Xe flux relations and their dependence of shutdowns

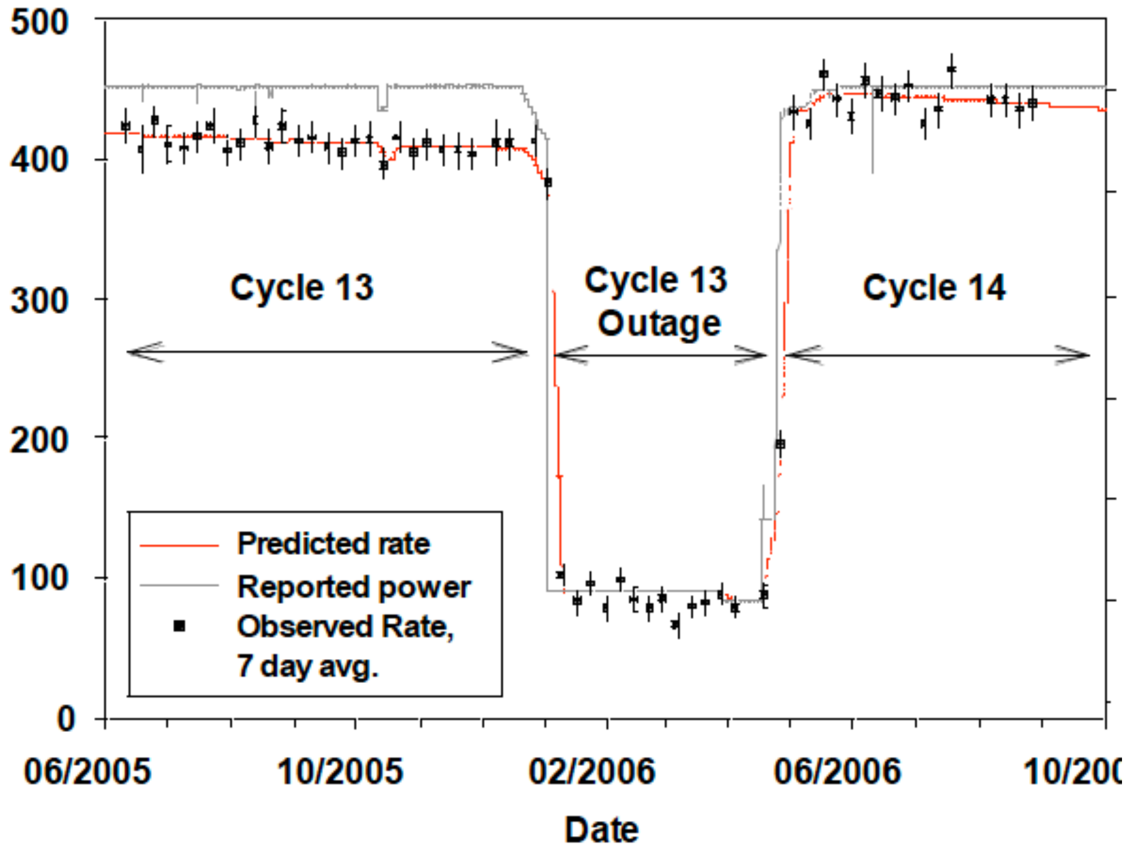
$$N_{135Cs}/N_{137Cs} = \frac{\bar{f}_{135I}}{\bar{f}_{137Cs}} \left[\frac{\lambda_{135Xe}}{\lambda_{135Xe} + \phi_T \sigma_a} + \frac{\phi_T \sigma_a}{\lambda_{135Xe} + \phi_T \sigma_a} \times \left(\frac{P}{\lambda_{135I} T_{irrad}^{total}} \right) \left(1 + \frac{\lambda_{135I}}{\lambda_{135Xe} + \phi_T \sigma_a} \right) \right].$$

Comes from the fact that you can't restart a reactor until all the ^{135}Xe decays to ^{135}Cs

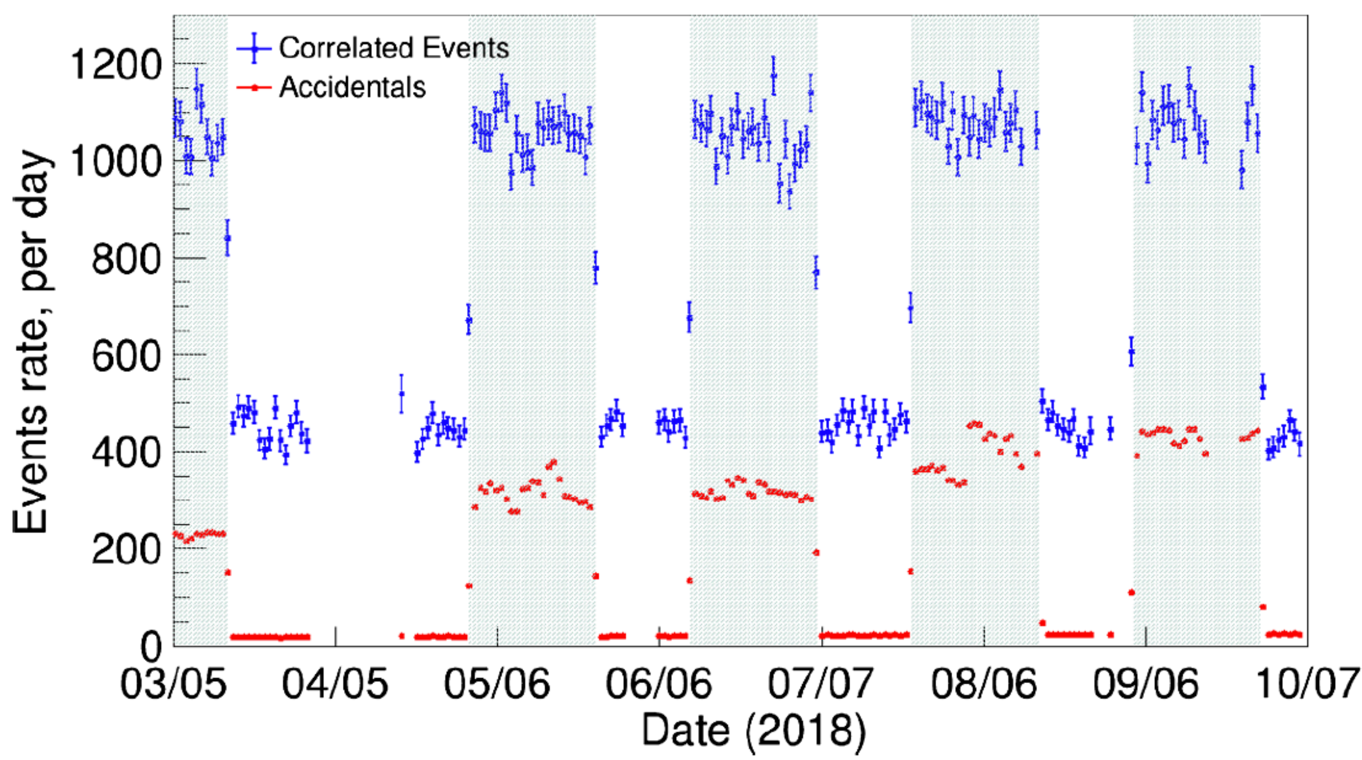
Dependence on shutdowns

P=number of shutdowns

Need a second diagnostic for T_{irrid} and P (# of shutdowns) - This is exactly what Antineutrinos are good at!

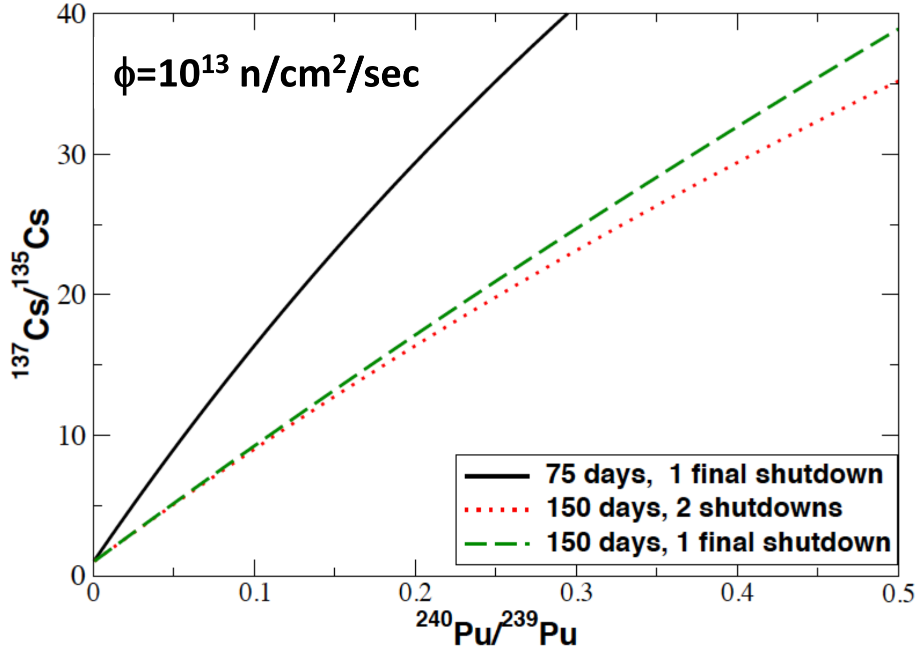


SONGS detector at San Onofre Reactor, N. Bowden, *et al.,* Nucl. Inst. Meth. A, 572 (2007) 985-998

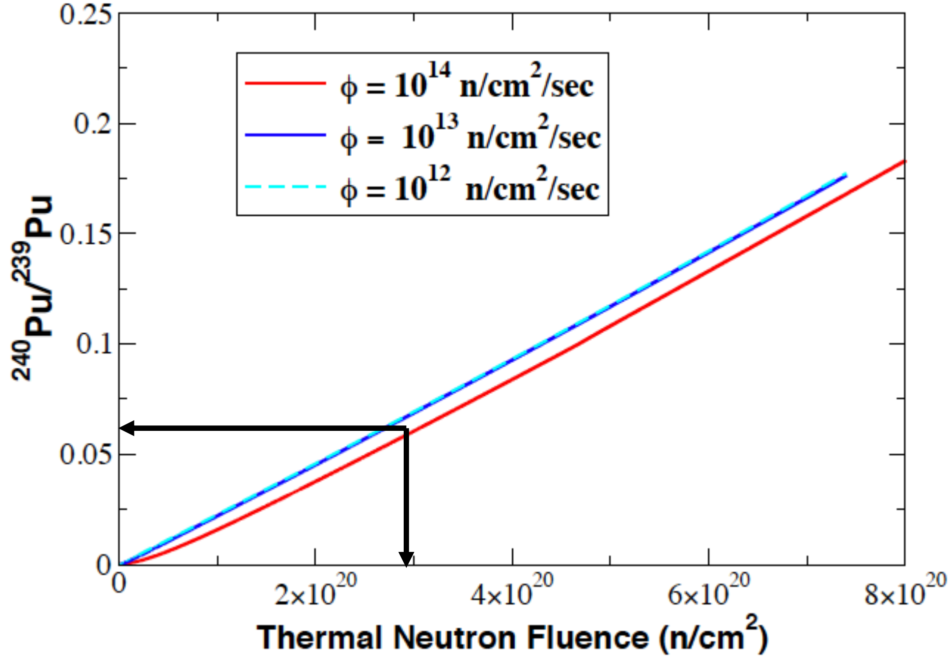


PROSPECT at 85 MW HFIR, Andriamirado *et al.,* PRD103, 032001 (2021).

Thus, by combining reactor fission gas measurements with antineutrino measurements the grade of Pu in the spent fuel can be determined



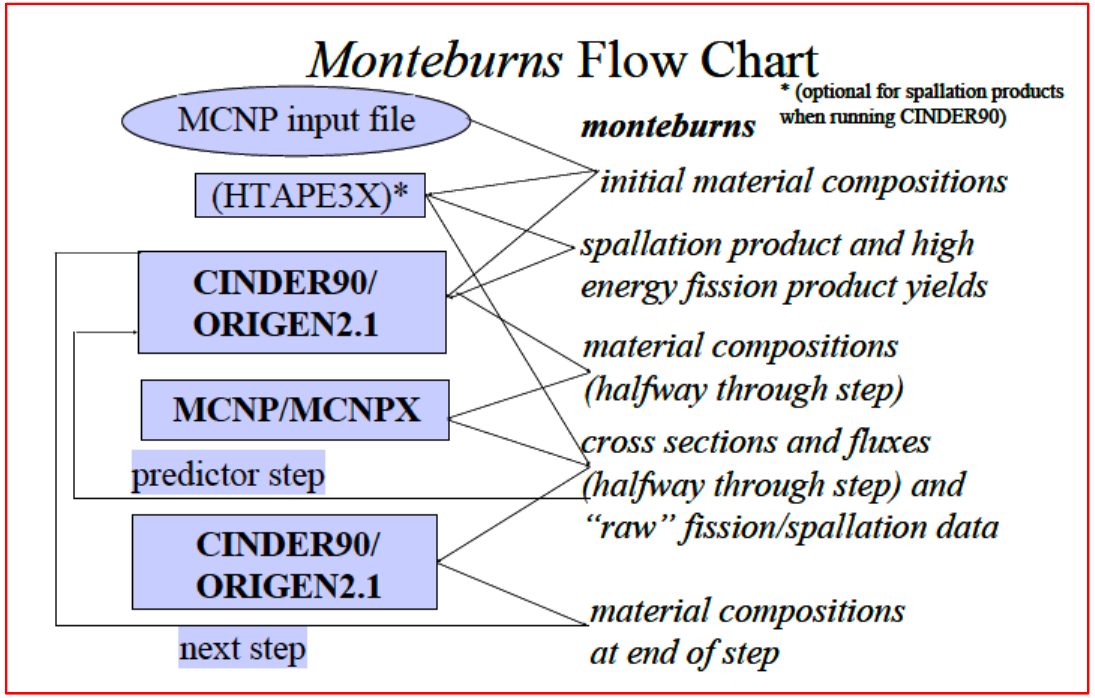
Antineutrino measurements of T_{irr} and P break any degeneracy in $T_{ir}r/P$ from Cs measurements alone.



Antineutrino measurements also eliminate the uncertainty in the Pu grade that arises if there is a very high flux $\phi \sim 10^{14}$ n/cm^2/sec

Uncertainties arising from nuclear data

Carried out benchmark reactor simulations using Monteburns with Origin 2.1 and CINDER90



Operating History for H. B. Robinson Unit 2 PWR

Cycle number	Interval time, days	Downtime following interval (days)	Average Power (MW/MTU)	Burnup (GWd/MTU)	Average soluble boron, ppm (wt)
1	243.5	40	32.23	7.85	652.5
2	243.5	64	30.10	15.18	247.5
3	156.0	39	28.35	19.60	652.5
4	156.0	3631	26.99	23.81	247.5

Comparisons for central rod positions

Isotope	Measured (g/gUO2)	*Monteburns* with ORIGEN2.2	*Monteburns* with CINDER90	% Relative Error
U235	7.21E-03	7.31E-03	7.33E-03	1.7
U236	2.74E-03	2.64E-03	2.59E-03	-5.40
U238	0.847	8.41E-01	8.41E-01	-0.70
Pu238	6.95E-05	6.55E-05	6.71E-05	-3.42
Pu239	4.02E-03	3.92E-03	4.04E-03	0.59
Pu240	1.67E-03	1.67E-03	1.70E-03	1.94
Pu241	5.04E-04	5.30E-04	5.34E-04	5.90
Np237	2.60E-04	2.36E-04	2.45E-04	-5.91
	(Ci/gUO2)			
Tc99	8.09E-06	8.76E-06	7.21E-06	-10.86
Cs137	5.39E-02	5.27E-02	4.95E-02	-8.07

- ^{238}U , 235U ^{239}Pu comparisons reasonably good
- Minor actinides off by up to 6%
- Only have comparison to long-live fission products and ^{137}Cs and ^{99}Tc off by ~ 8-10%.

One rod was measured in detail at LANL at 112 inches above the bottom of the fuel, providing more stringent tests of simulation predictions of fission products.

isotope ratio	% difference
$^{235}U/U$	-0.5
$^{238}U/U$	+0.01
$^{239}Pu/Pu$	-0.5
$^{148}Nd/^{238}U$	+0.3

Isotope ratio	% difference
$^{234}U/U$	-7.8
$^{236}U/U$	-0.1
$^{240}Pu/Pu$	+5.0
$^{241}Pu/Pu$	-7.1
$^{242}Pu/Pu$	6.2
$^{241}Am/Am$	-15
$^{242}Am/Am$	7.5
$^{242}Cm/Cm$	0.4
$^{243+244}Cm/Cm$	+1.6

Atoms per gm of UO_2	% diff
^{90}Sr	-13.2
^{125}Sb	+12.6
^{134}Cs	-9.0
^{137}Cs	-2.9
^{144}Ce	-1.9
^{154}Eu	-68
^{155}Eu	+43

Major actinides are well reproduced
Minor actinides are off by up to 10%
Most fission products are off up to 10%
Eu fission products are poorly reproduced

Summary

- The $^{240}Pu/^{239}Pu$ ratio depends on the total neutron fluence, and can be determined with a measure of the flux, the irradiation time, and the number of reactor shutdowns.
- The $^{137}Cs/^{135}Cs$ ratio is a sensitive measure of the the reactor thermal neutron flux, but requires knowledge of the number of reactor shutdowns.
- Antineutrino monitoring determines irradiation times and the number of reactor shutdowns. Coupling fission gas and antineutrino measurements could result in a powerful diagnostic.
- Benchmark simulations suggest that the major actinides can be reproduced accurately.
- However, fission product yields can be off by more than 10%, . There is a need for more accurate fission product yields, beyond those important for antineutrino spectra alone.