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Title: The Role of Monte Carlo Burnup Calculations in Quantifying Plutonium Mass in Spent Fuel Assemblies with Non-Destructive Assay

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The Role of Monte Carlo Burnup Calculations in Quantifying Plutonium Mass in Spent Fuel Assemblies with Non-Destructive Assay

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Abstract

MCNPX has been used to simulate the isotopic composition of spent nuclear fuel. This talk, prepared for the 33rd ESARDA Annual Meeting, first addresses the spatial variation of plutonium concentration across a representative 17x17 generic Westinghouse fuel assembly, and then examines how well plutonium mass can be predicted spatially across the assembly. In addition we provide an assessment of how well the quantity of plutonium in all internal fuel pins can be predicted from known plutonium quantities in the perimeter fuel pins. Furthermore, MCNPX has been used to simulate the response of a passive gamma system used in the assay of spent nuclear fuel. This talk also addresses source set-up and transport simulation challenges and methods, and discusses how these challenges were met and what methods were developed for use with SFL 2, including some preliminary results of gamma spectrum for various sides of a fuel assembly.

The Role of Monte Carlo Burnup Calculations in Quantifying Plutonium Mass in Spent Fuel Assemblies with Non-Destructive Assay

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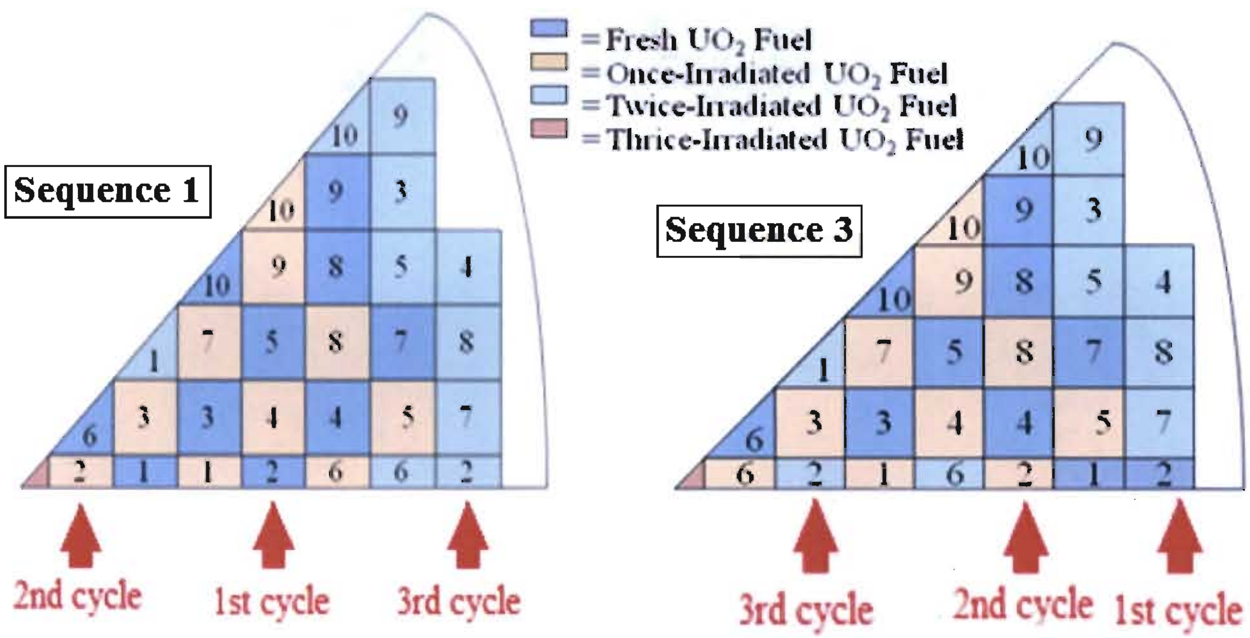
Outline

- **Spent fuel libraries (SFLs)**
- **Spatial Pu distribution**
  - ^{239}Pu accumulation
- **Assembly Pu prediction**
  - Spatial distribution prediction & total assembly prediction
- **Passive gamma simulations with SFL 2**
- **Future work**

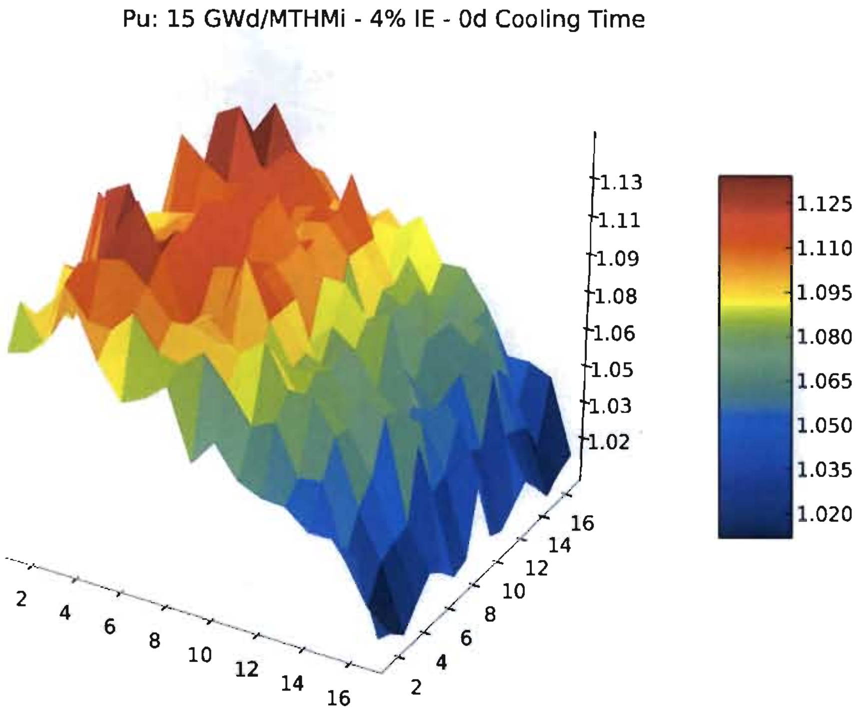
What Influences Plutonium Distribution?

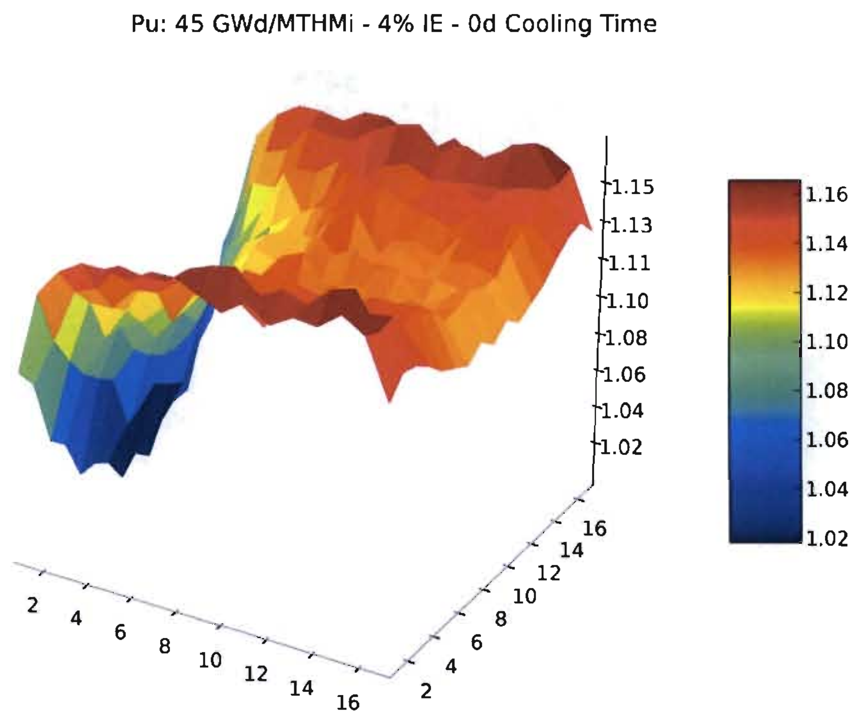
- **Among several factors, shuffling sequence drives plutonium distribution**
  - SFL #1: Infinitely reflected assembly
  - SFL #2: Multiple fuel shuffling sequences employed



Sequence 1 Pu spatial distribution

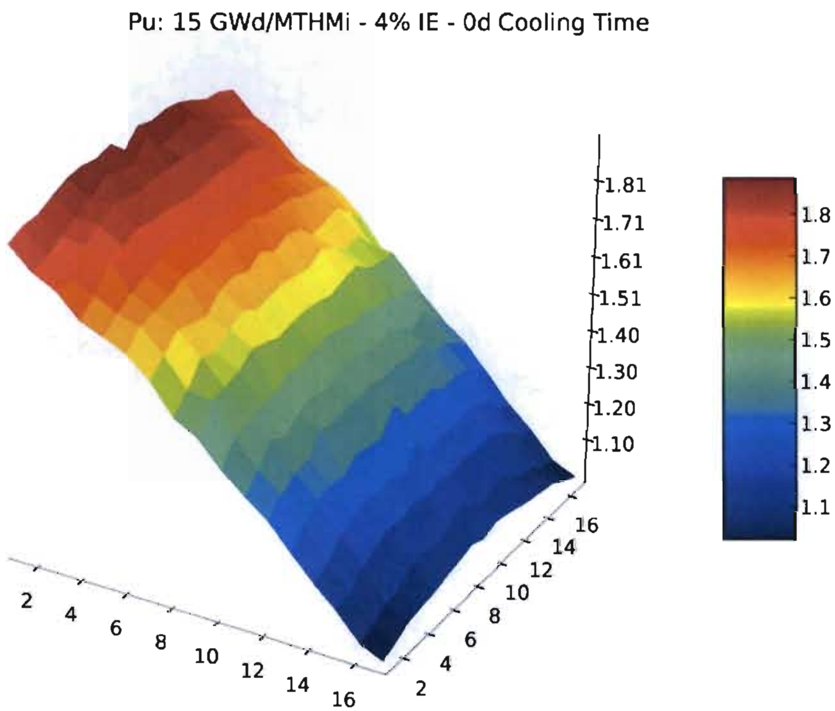
- **Z-axis normalized to minimum pin Pu mass**
  - Shuffling sequence #1

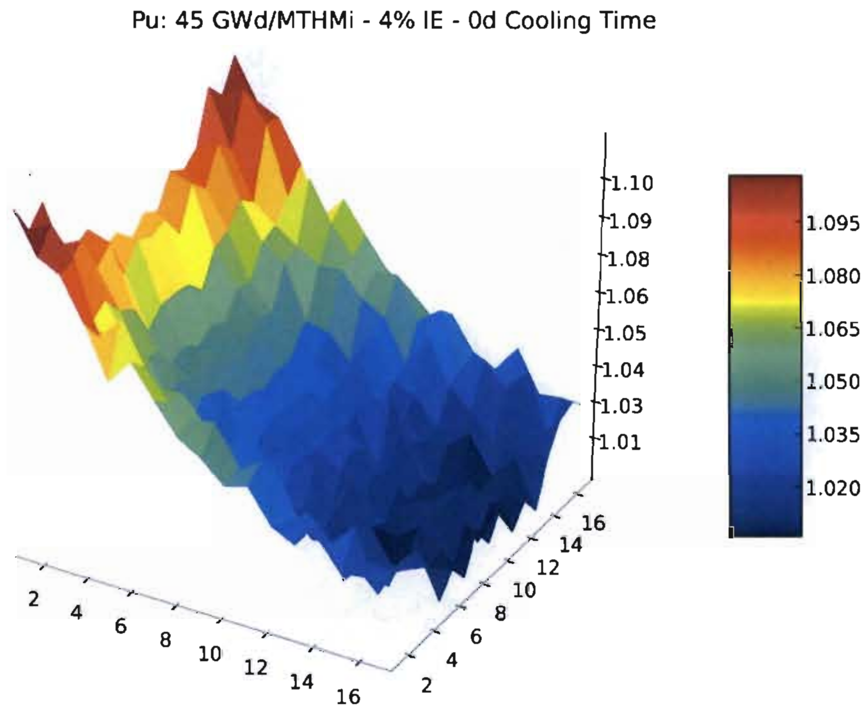




Sequence 3 Pu spatial distribution

- **Z-axis normalized to minimum pin Pu mass**
  - Shuffling sequence #3

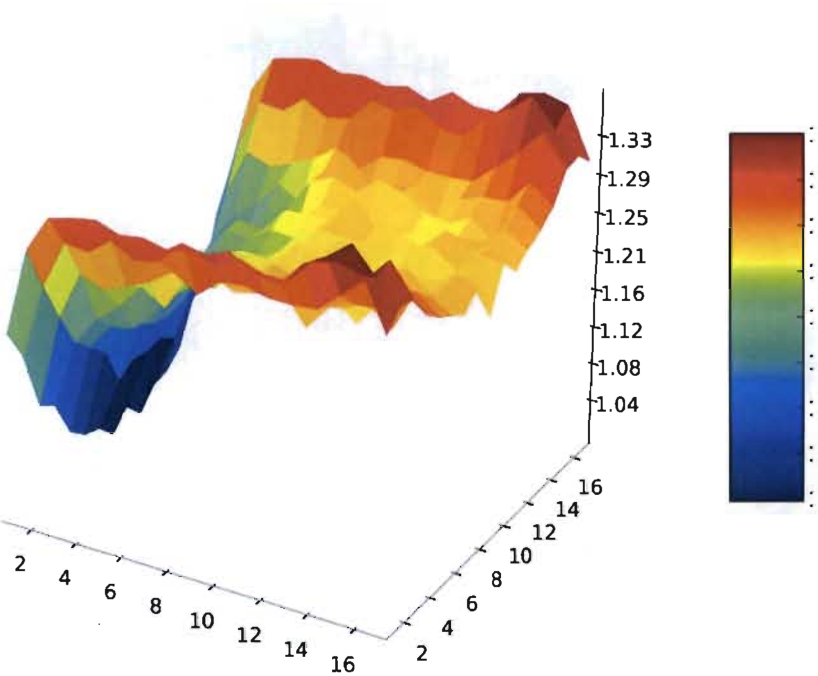




^{239}Pu spatial distribution – sequence 1 and 3

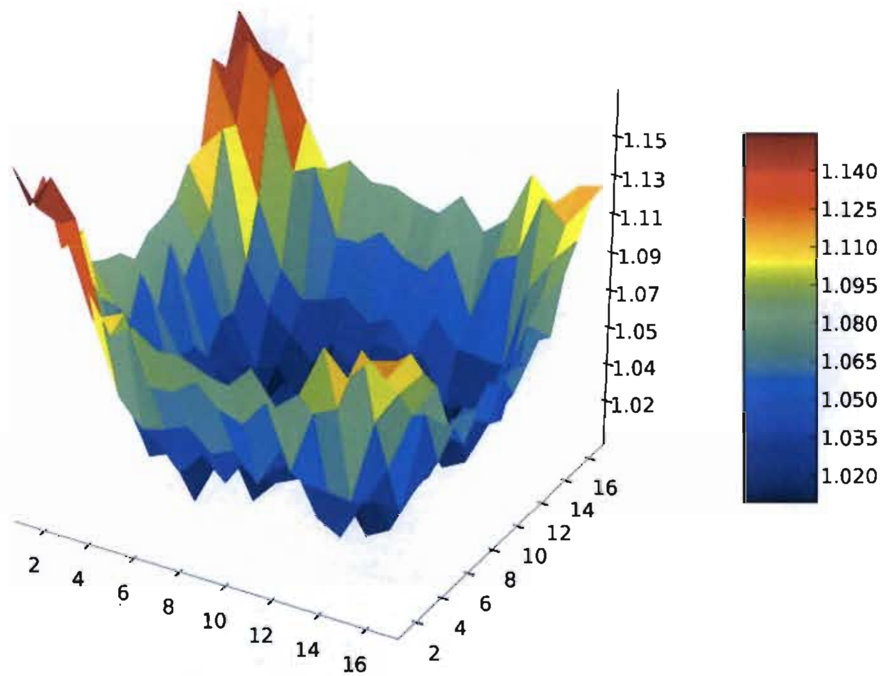
- **At Higher burnups, Pu becomes more peripherally weighted**
  - ^{239}Pu drives elemental distribution
  - Highest concentrations is at corners

Pu239: 45 GWd/MTHMi - 4% IE - 0d Cooling Time



Sequence 1

Pu239: 45 GWd/MTHMi - 4% IE - 0d Cooling Time



Sequence 3

Pu spatial distribution

- **^{239}Pu clearly accumulates preferentially on periphery**
  - Also is dominant contributor to overall Pu inventory
  - % Contribution to Pu inventory:

45 GWd/MTU	Sequence 1	Sequence 2	Sequence 3
Pu (g)	5024.82	5081.89	4983.85
Pu239 (g)	2575.09	2682.90	2663.92
Pu239 % Contribution	***51.25%***	***52.79%***	***53.45%***
% Pu Change from Sequence 1		1.14%	-1.93%
% Pu239 Change from Sequence 1		4.19%	-0.71%

  - In all cases, contributes greater than 50%
  - Accumulation distribution varies for different isotopes, ex: ^{238}Pu, ^{242}Pu weighted toward internal

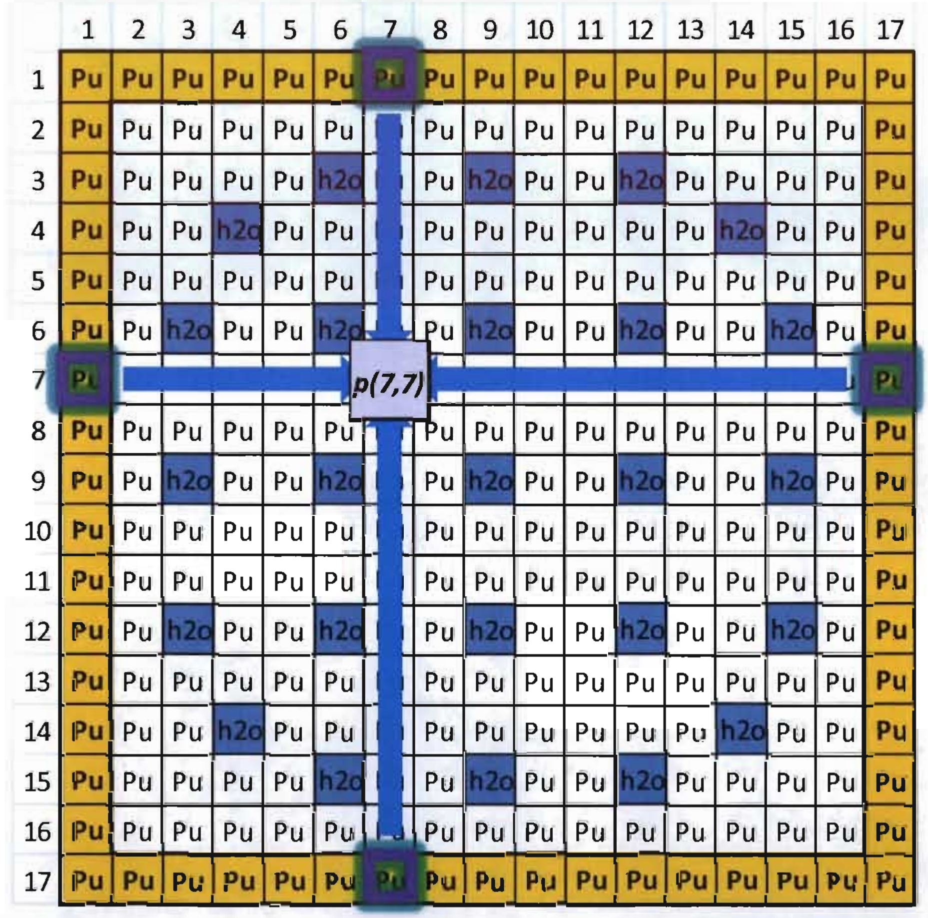
Spatial Pu prediction approach

- **How accurately can pin-by-pin isotopic distribution be predicted from knowledge of edge content?**

Approach:
Use SFL 1 (infinite reflection) edge pin, and internal pin Pu values to generate internal fuel pin predictors.

Use predictors and edge pin knowledge to predict internal Pu content for the three shuffling cases.

Ex: p(7,7) = predictor for pin 7,7



Spatial Pu prediction approach

- **Using edge pin knowledge and internal predictors**
  - Predict Pu mass for pin 7,7

$\widetilde{Pu}_{7,7}$ = predicted Pu mass at 7,7

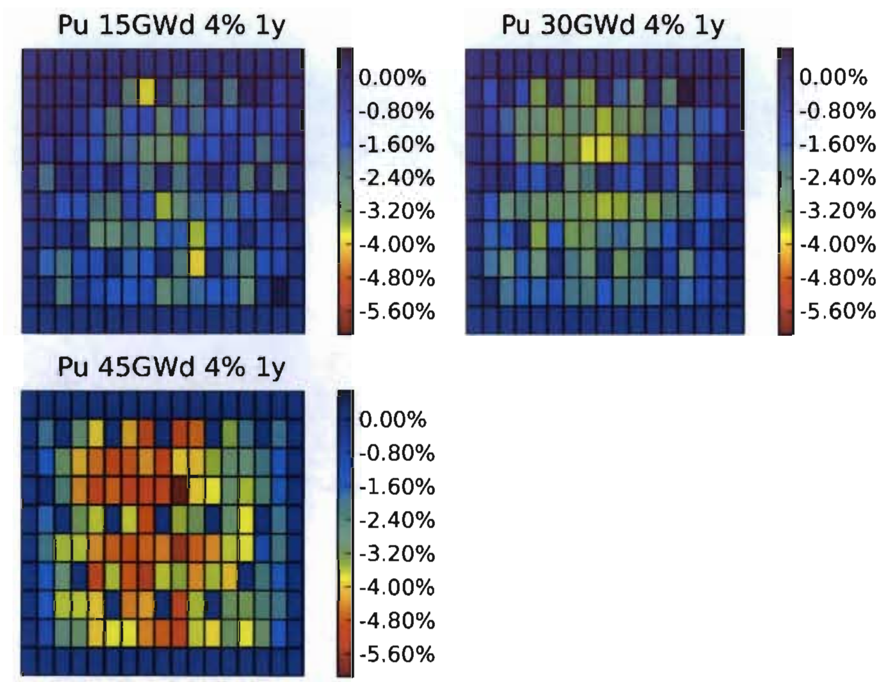
Same process repeated for all internal fuel pins

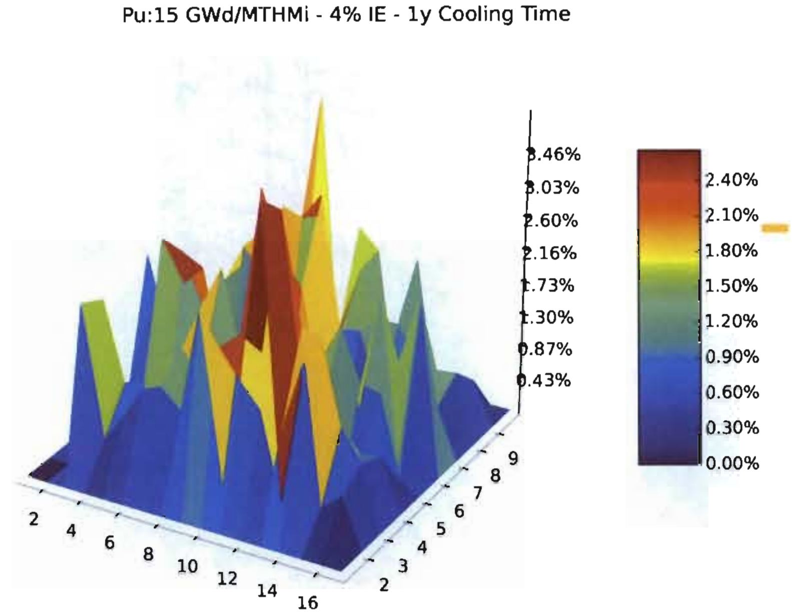
Compare Pu prediction to true value from shuffling sequence simulation

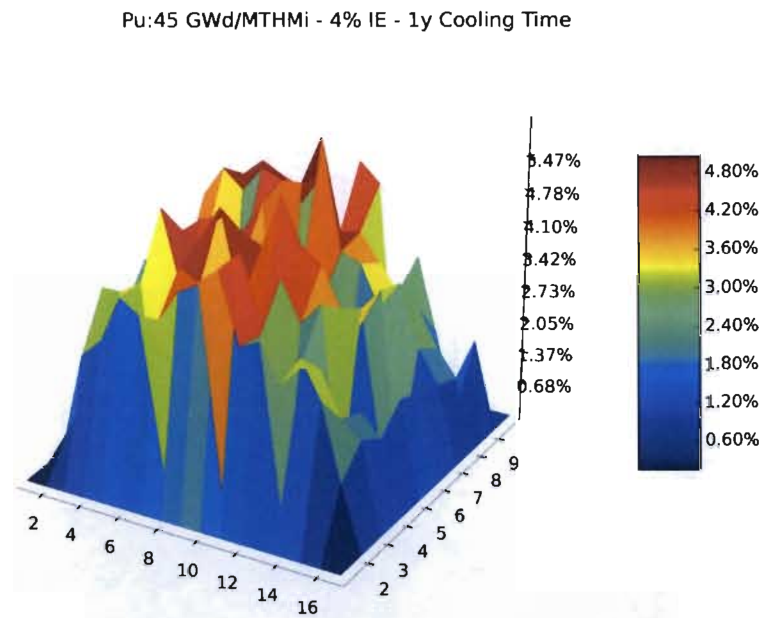
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
1	Pu	Pu	Pu	Pu	Pu	Pu	Pu	Pu	Pu	Pu	Pu	Pu	Pu	Pu	Pu	Pu	Pu
2	Pu	?	?	?	?	?		?	?	?	?	?	?	?	?	?	Pu
3	Pu	?	?	?	?	h2o		?	h2o	?	?	h2o	?	?	?	?	Pu
4	Pu	?	?	h2o	?	?		?	?	?	?	?	?	h2o	?	?	Pu
5	Pu	?	?	?	?	?		?	?	?	?	?	?	?	?	?	Pu
6	Pu	?	h2o	?	?	h2o	?	?	h2o	?	?	h2o	?	?	h2o	?	Pu
7	Pu						$\widetilde{Pu}_{7,7}$	?									Pu
8	Pu	?	?	?	?	?	?	?	?	?	?	?	?	?	?	?	Pu
9	Pu	?	h2o	?	?	h2o		?	h2o	?	?	h2o	?	?	h2o	?	Pu
10	Pu	?	?	?	?	?		?	?	?	?	?	?	?	?	?	Pu
11	Pu	?	?	?	?	?		?	?	?	?	?	?	?	?	?	Pu
12	Pu	?	h2o	?	?	h2o		?	h2o	?	?	h2o	?	?	h2o	?	Pu
13	Pu	?	?	?	?	?		?	?	?	?	?	?	?	?	?	Pu
14	Pu	?	?	h2o	?	?		?	?	?	?	?	?	h2o	?	?	Pu
15	Pu	?	?	?	?	h2o		?	h2o	?	?	h2o	?	?	?	?	Pu
16	Pu	?	?	?	?	?		?	?	?	?	?	?	?	?	?	Pu
17	Pu	Pu	Pu	Pu	Pu	Pu	Pu	Pu	Pu	Pu	Pu	Pu	Pu	Pu	Pu	Pu	Pu

Spatial Pu prediction results

- **Shuffling Sequence #1, relative errors**

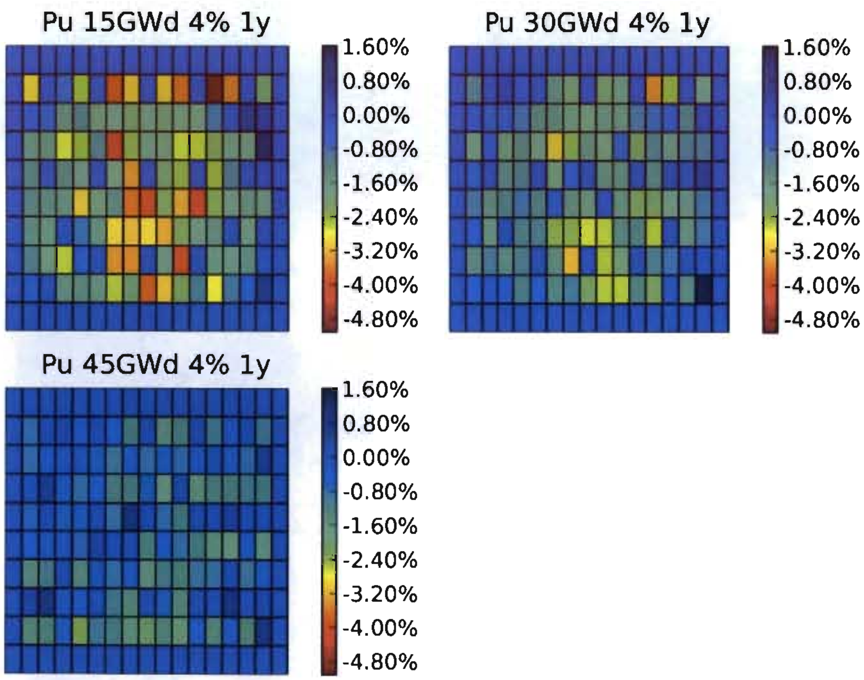


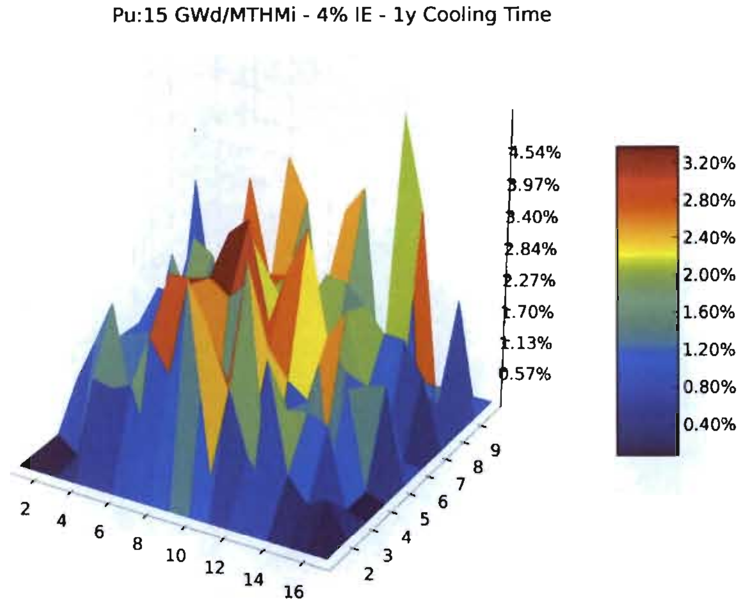


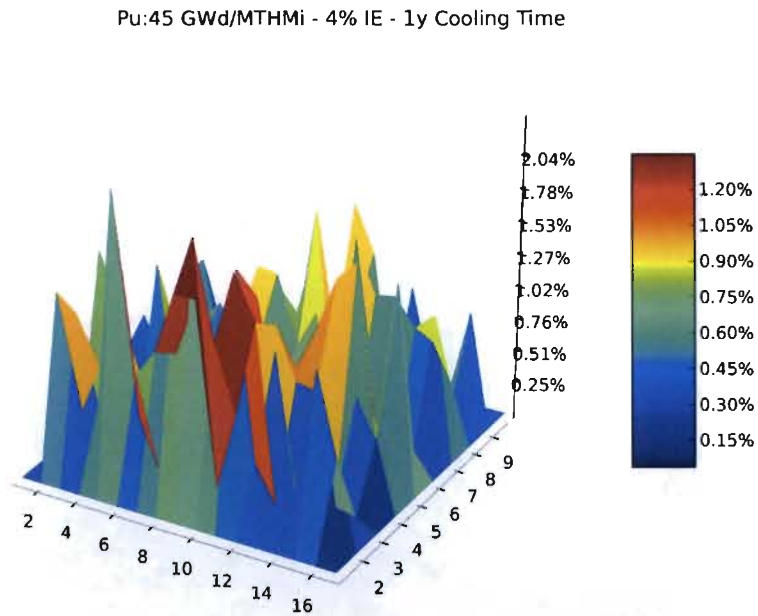


Spatial Pu prediction results

- **Shuffling Sequence #3, relative errors**







Spatial Pu prediction conclusions

- **Strong sensitivity to fuel rotation scheme**
  - Shuffling Sequence 1 errors increase as BU increases
    - Max pin error of 2% at 15 GWd/MTU, max pin error of 6% at 45 GWd/MTU
  - Shuffling Sequence 3 errors do not increase as BU increases
    - Max pin error of 5% at 15 GWd/MTU, max pin error of 1.5% at 45 GWd/MTU
  - The highest errors corresponded with the most asymmetric power distributions

- **A smarter prediction is needed and expected to yield more accurate results**
  - Using the infinite assembly to predict asymmetric burns, shuffling sequence 3 at 15GWd/MTU yields the largest error swing
  - Train predictors for various gradients or use smarter prediction that incorporates asymmetric knowledge for better prediction

Total assembly Pu prediction

- **Predict total assembly Pu mass from perimeter measurement**
  - Ratio sum of bundle perimeter Pu masses to internal Pu masses

$$Pu_{ratio} = \frac{\sum x}{\sum y}$$

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
1	x	x	x	x	x	x	x	x	x	x	x	x	x	x	x	x	x
2	x	y	y	y	y	y	y	y	y	y	y	y	y	y	y	y	x
3	x	y	y	y	y	h2o	y	y	h2o	y	y	h2o	y	y	y	y	x
4	x	y	y	h2o	y	y	y	y	y	y	y	y	y	h2o	y	y	x
5	x	y	y	y	y	y	y	y	y	y	y	y	y	y	y	y	x
6	x	y	h2o	y	y	h2o	y	y	h2o	y	y	h2o	y	y	h2o	y	x
7	x	y	y	y	y	y	y	y	y	y	y	y	y	y	y	y	x
8	x	y	y	y	y	y	y	y	y	y	y	y	y	y	y	y	x
9	x	y	h2o	y	y	h2o	y	y	h2o	y	y	h2o	y	y	h2o	y	x
10	x	y	y	y	y	y	y	y	y	y	y	y	y	y	y	y	x
11	x	y	y	y	y	y	y	y	y	y	y	y	y	y	y	y	x
12	x	y	h2o	y	y	h2o	y	y	h2o	y	y	h2o	y	y	h2o	y	x
13	x	y	y	y	y	y	y	y	y	y	y	y	y	y	y	y	x
14	x	y	y	h2o	y	y	y	y	y	y	y	y	y	h2o	y	y	x
15	x	y	y	y	y	h2o	y	y	h2o	y	y	h2o	y	y	y	y	x
16	x	y	y	y	y	y	y	y	y	y	y	y	y	y	y	y	x
17	x	x	x	x	x	x	x	x	x	x	x	x	x	x	x	x	x

Assembly Pu prediction

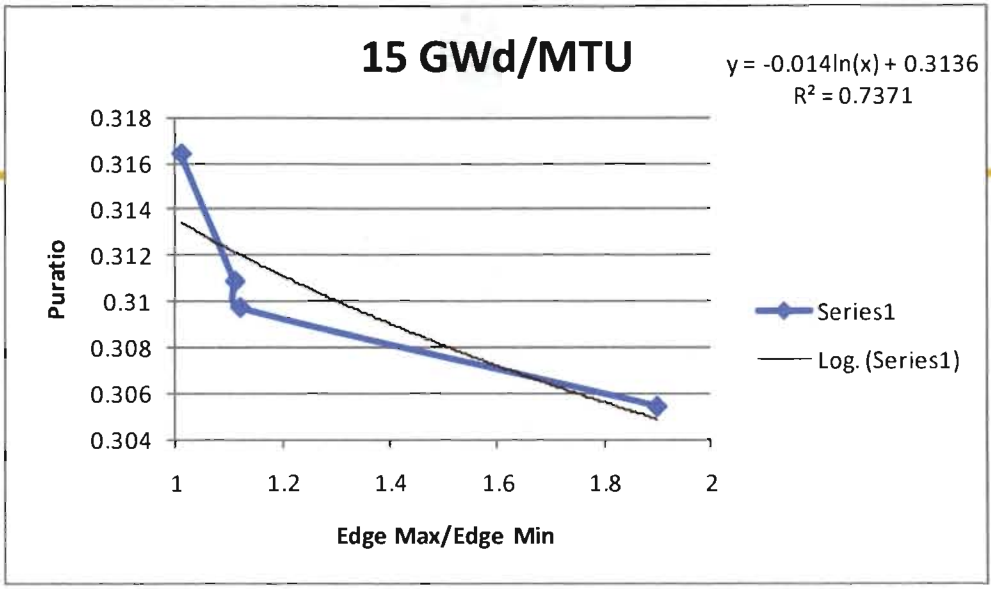
- **Simplistic approach:**
  - Take Pu_{ratio} from infinitely reflected assembly at (15,30,45 GWd/MTU)
  - Use ratio as predictor for the 3 different shuffling sequences

		Infinite ratio					
		Shuffling Sequence 1		Shuffling Sequence 2		Shuffling Sequence 3	
		Prediction (g)	% Error	Prediction (g)	% Error	Prediction (g)	% Error
15 GWd/MTU	0d	2029.87	**-1.74%**	1990.21	**-2.10%**	1848.49	**-3.45%**
	5y	2043.02	**-1.69%**	2004.67	**-2.04%**	1855.69	**-3.45%**
	80y	1925.01	**-1.34%**	1891.61	**-1.72%**	1742.65	**-3.63%**
30 GWd/MTU	0d	3079.52	**-2.29%**	3118.38	**-1.70%**	2991.93	**-1.89%**
	5y	3032.16	**-2.22%**	3065.12	**-1.68%**	2952.17	**-1.89%**
	80y	2715.03	**-2.20%**	2740.98	**-1.73%**	2647.73	**-1.91%**
45 GWd/MTU	0d	3677.34	**-3.68%**	3785.31	**-1.41%**	3722.93	**-1.04%**
	5y	3577.47	**-3.61%**	3689.09	**-1.35%**	3646.23	**-0.96%**
	80y	3142.06	**-3.71%**	3236.61	**-1.43%**	3203.43	**-0.88%**

  - Maximum error with "dumb" guess is <4%
  - Largest errors predictably correspond to locations with strong gradients

Assembly Pu prediction

- **More advanced approach**
  - Tally maximum & minimum values on perimeter for 4 assemblies (characterize gradient)
  - Fit function to plot of: max/min ratio vs. edge/internal ratio – at each burnup (15,30,45)
  - Maximum error reduces to ~1.2%



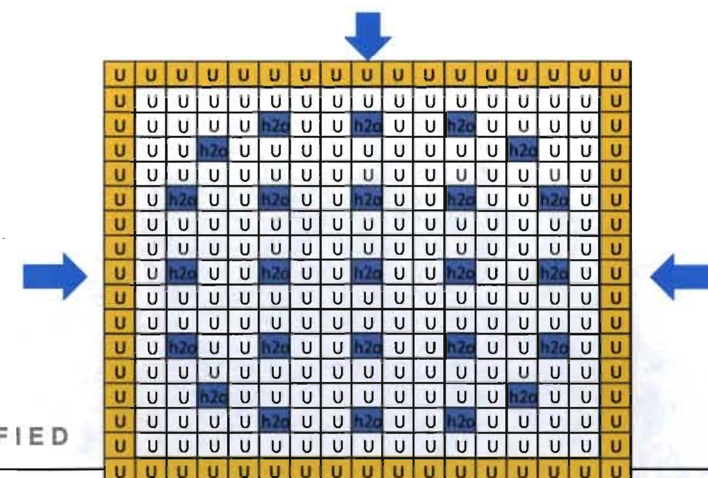
		Fitted prediction					
		Shuffling Sequence 1		Shuffling Sequence 2		Shuffling Sequence 3	
		Prediction (g)	% Error	Prediction (g)	% Error	Prediction (g)	% Error
15 GWd/MTU	0d	2057.64	**-0.40%**	2018.20	**-0.72%**	1919.86	**0.28%**
	5y	2072.20	**-0.28%**	2034.05	**-0.61%**	1926.60	**0.24%**
	80y	1960.96	**0.50%**	1927.77	**0.16%**	1814.60	**0.35%**
30 GWd/MTU	0d	3119.41	**-1.02%**	3166.21	**-0.19%**	3062.52	**0.43%**
	5y	3073.34	**-0.89%**	3114.20	**-0.10%**	3021.55	**0.42%**
	80y	2765.54	**-0.38%**	2796.61	**0.27%**	2719.02	**0.73%**
45 GWd/MTU	0d	3776.43	**-1.08%**	3874.21	**0.91%**	3778.17	**0.43%**
	5y	3668.65	**-1.15%**	3773.79	**0.91%**	3696.08	**0.39%**
	80y	3225.35	**-1.16%**	3303.94	**0.62%**	3247.23	**0.48%**

Passive gamma source generation

- **How large of an effect do the asymmetries introduced in SFL 2 impact gamma signals?**
  - BAMF tool developed for SFL 1 modified and enhanced
    - Determine photon source in each fuel pin, create source file, transport photons and tally flux individually across four assembly boundaries
    - Simulate detector response by "pushing" flux to detector and performing pulse-height tally
    - Each step performed within one Perl script, serves as "go" button that takes spent fuel composition, creates MCNP input deck with source definitions and performs transport

Side 2

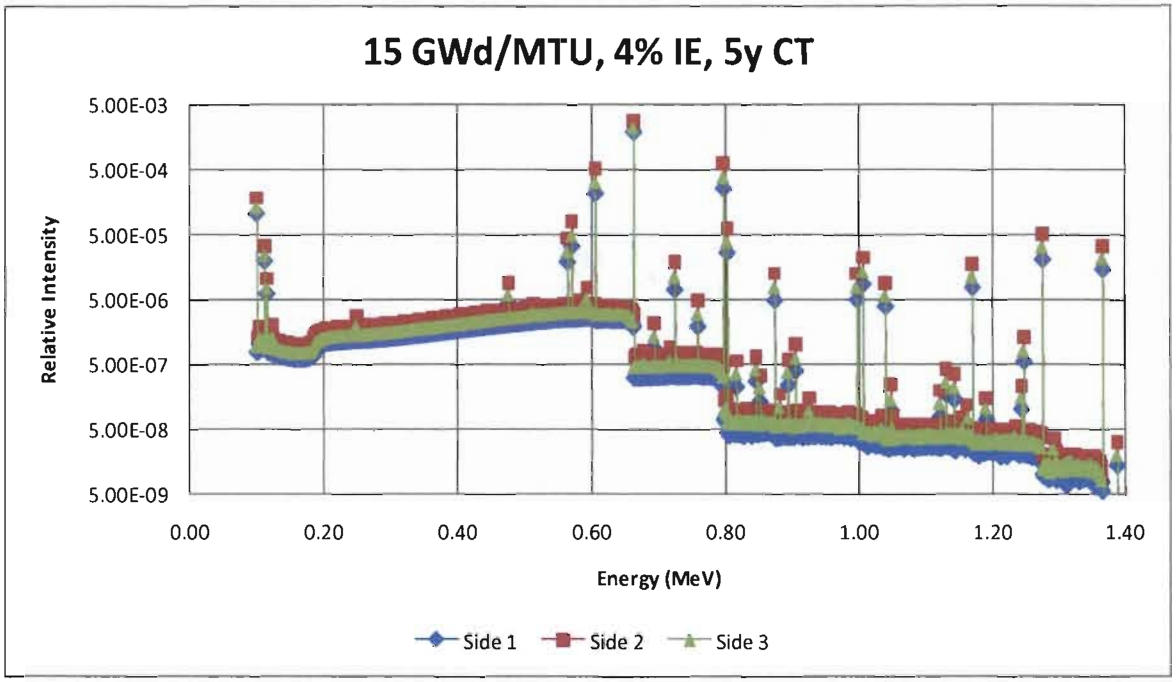
Side 1



Side 3

Passive gamma results

- **Photon signal for shuffling sequence 3 at 15 GWd/MTU**
  - Most extreme gradient experienced in all three shuffling schemes
  - Side 2, highest burned side

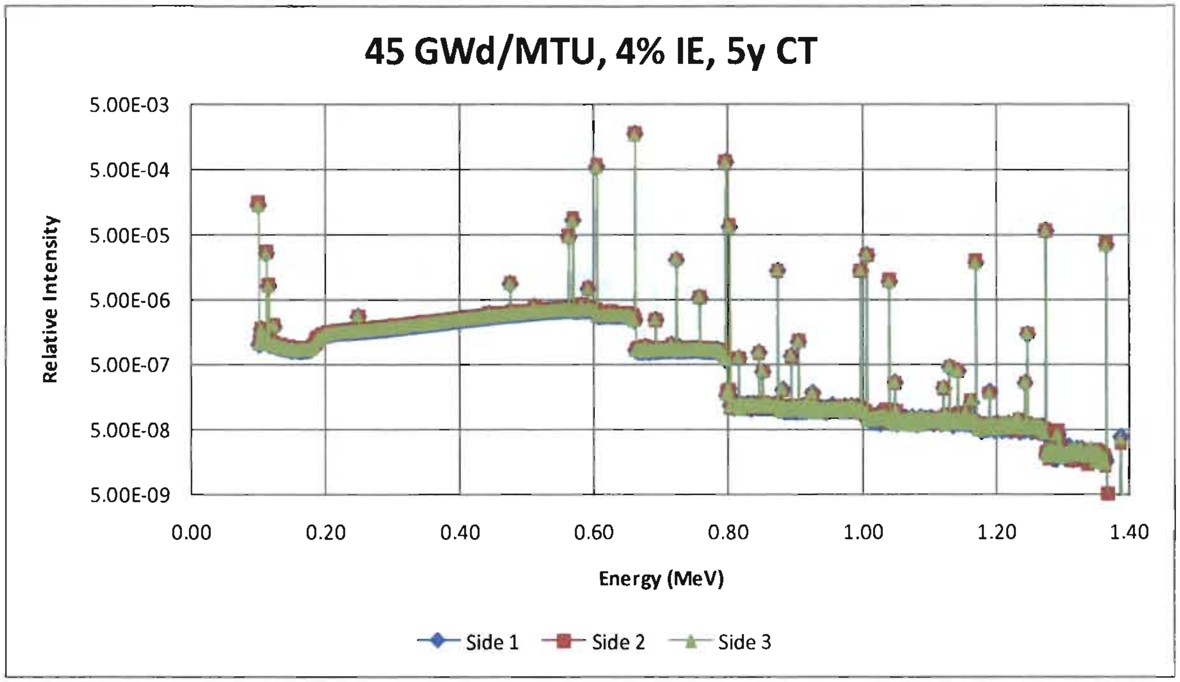


Relative difference from side 2 activity (5y cooling time)		
	Side 1	Side 3
Cs-134 (.6047 MeV)	-58.84%	-40.86%
Cs-137 (0.6617 MeV)	-32.49%	-23.86%
Cs-134 (0.7959 MeV)	-58.39%	-39.55%

- ❑ ^{137}Cs signal ~24% - 33% signal intensity variation
- ❑ ^{134}Cs signal ~40% - 60% signal intensity variation
  - ❑ ^{134}Cs depends on square of flux through (n,γ) capture reaction

Passive gamma results

- **Photon signal for shuffling sequence 1 at 45 GWd/MTU**
  - Spatial burnup distribution is quite flat
  - Side 2 is still highest burned side



Relative difference from side 2 activity (5y cooling time)		
	Side 1	Side 3
Cs-134 (.6047 MeV)	-6.51%	-3.88%
Cs-137 (0.6617 MeV)	-5.21%	-4.51%
Cs-134 (0.7959 MeV)	-5.86%	-3.40%

- ❑ ^{137}Cs signal ~4% - 5% signal intensity variation
- ❑ ^{134}Cs signal ~4% - 6% signal intensity variation

Passive gamma ratios

- **Ratios often used for evaluation of assembly average parameters**
- **How much variation is introduced, due to shuffling scheme?**
  - $^{134}Cs/^{137}Cs$ ratio calculated, all gammas from 134Cs included in signal
  - Sequence 3,15 GWd/MTU (strong gradient), Sequence 1, 45 GWd/MTU

Sequence 3 - 15 GWd/MTU - 5y			
	cs134	cs137	**ratio**
Side 1	5.43E-04	1.84E-03	**0.2955**
Side 2	1.31E-03	2.72E-03	**0.4801**
Side 3	7.84E-04	2.07E-03	**0.3784**

Sequence 1 - 45 GWd/MTU - 5y			
	cs134	cs137	**ratio**
Side 1	1.37E-03	1.69E-03	**0.8100**
Side 2	1.46E-03	1.79E-03	**0.8178**
Side 3	1.41E-03	1.71E-03	**0.8258**

  - Strong variation seen at 15 GWd/MTU, 20 & 40% signal difference from peak ratio
  - Weak variation seen at 45 GWd/MTU, 2% signal difference from peak ratio
    - High burnup bundles will typically have relatively even distribution

Future Work

- **Develop better spatial Pu predictor algorithm, using perimeter knowledge**
- **Characterize assembly prediction fidelity by applying to greater number of assemblies**
- **Use tally-tagging, characterize significantly contributing pins to HPGE/collimator geometry**
  - Retool source generation to only generate in significantly contributing pins
- **Characterize HPGE sensitivity to detector location, centered on fuel pin, +/- various offsets (0.1, 0.5, 1, 2 cm)**
- **How well does Passive Gamma signal scale with Pu content?**

Questions?

- Jackg@lanl.gov

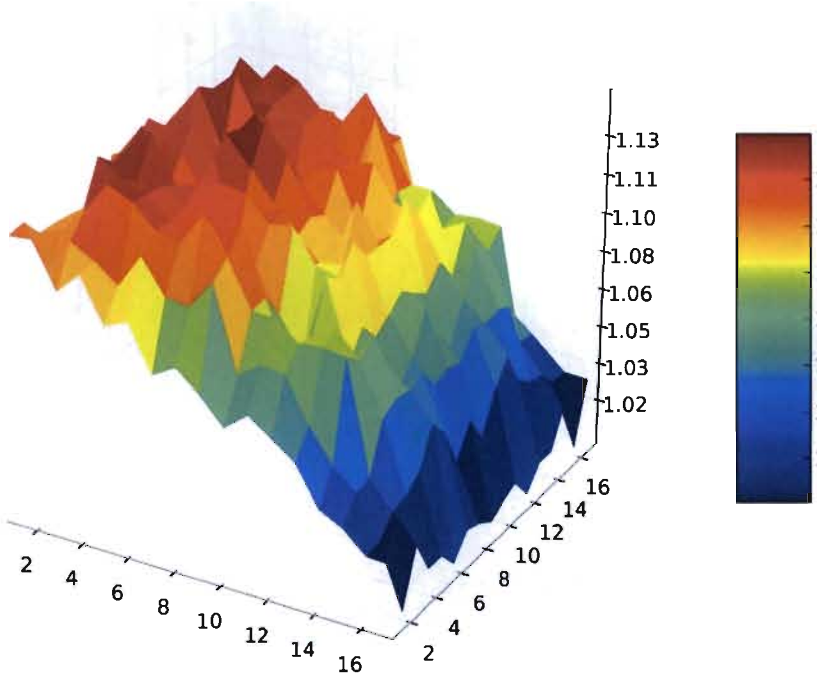
Additional Slides

- **Additional slides follow if time allows, and info provides useful supplemental information.**

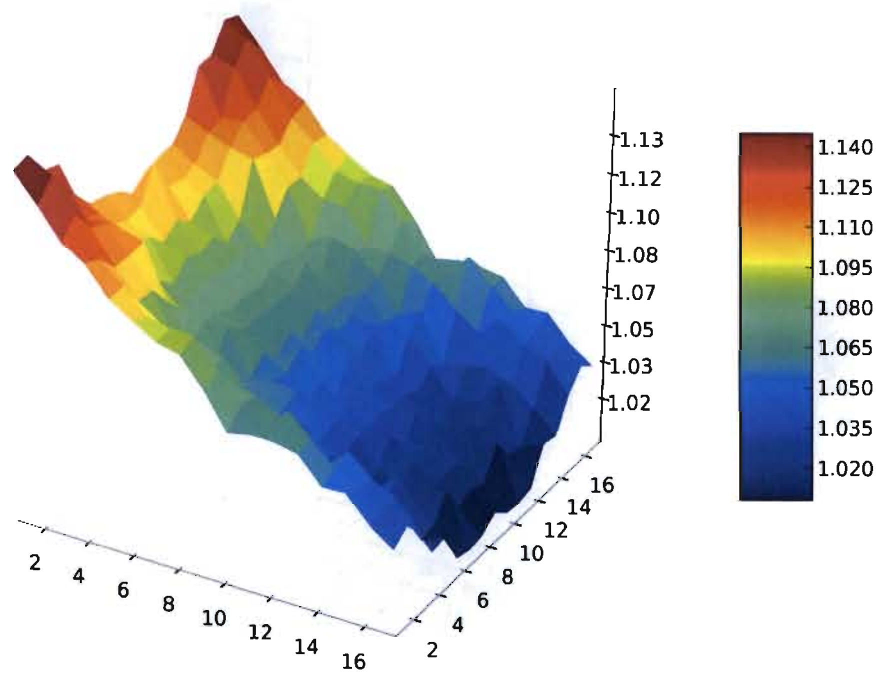
Pu spatial distribution

- **How does Pu vary spatially?**
  - Shuffling sequence #2

Pu: 15 GWd/MTHMi - 4% IE - 0d Cooling Time

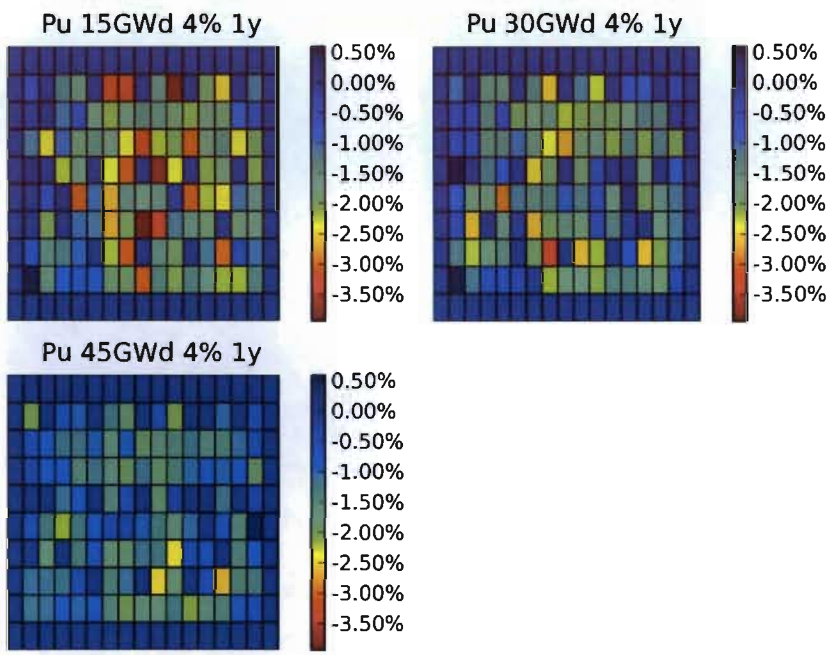


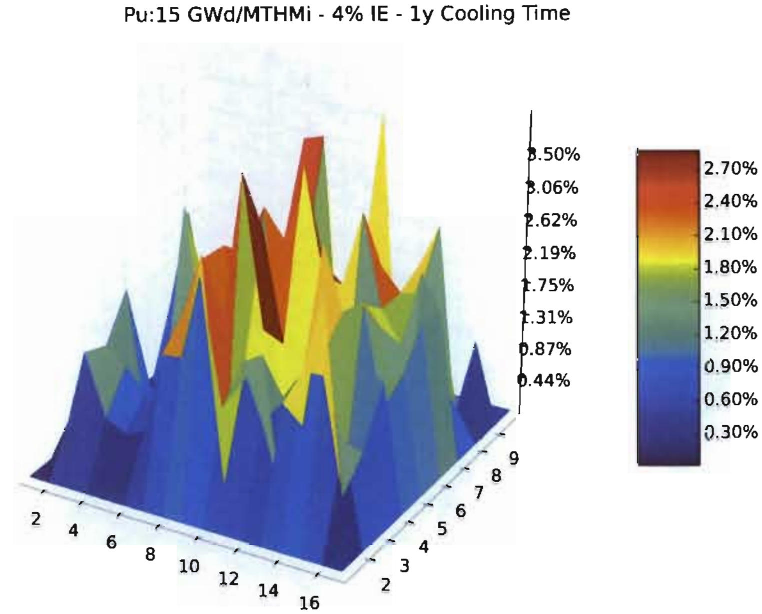
Pu: 45 GWd/MTHMi - 4% IE - 0d Cooling Time

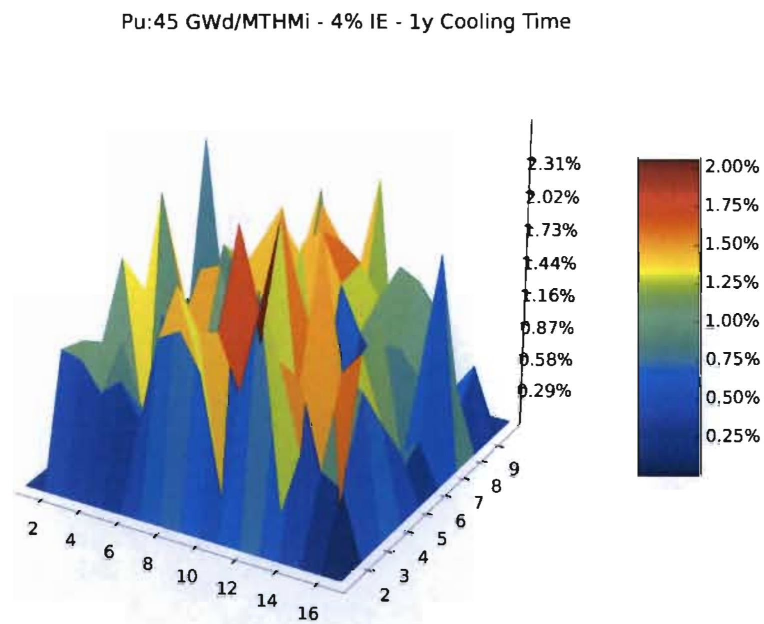


Spatial Pu prediction

- **Shuffling Sequence 2**







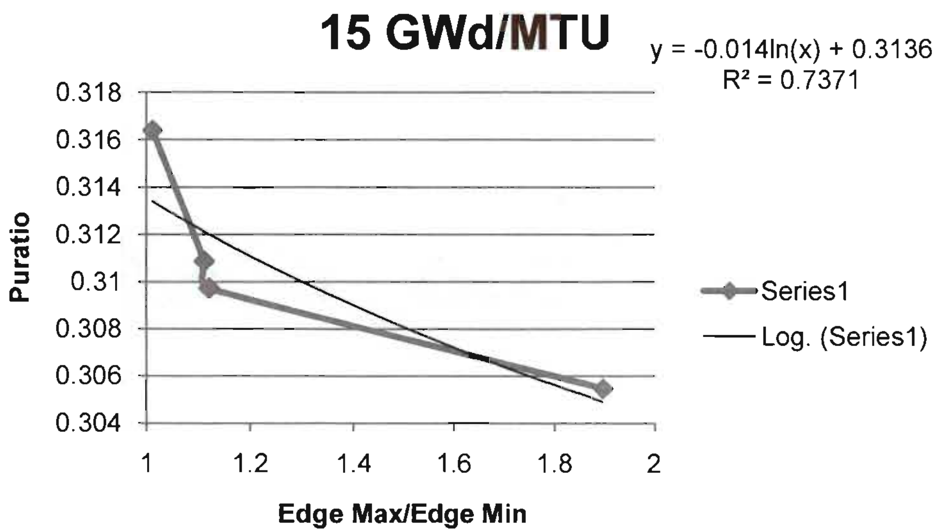
Spatial Pu prediction equation

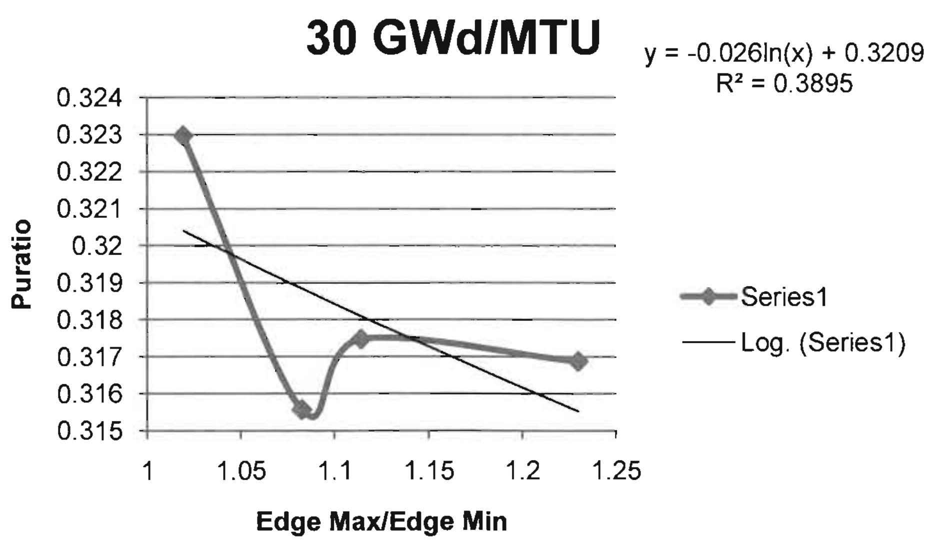
- **Pin-by-pin Pu prediction equation:**
  - Use SFL 1 (infinite reflection) distribution to generate internal predictions, then

$$Predictor_{i,j} = \frac{Pu_{i,j}}{\left[Pu_{j,0} * \frac{(xl - i)}{xl} + Pu_{j,xl} * \frac{i}{xl} + Pu_{0,i} * \frac{(yl - j)}{yl} + Pu_{yl,i} * \frac{j}{yl}\right]}$$

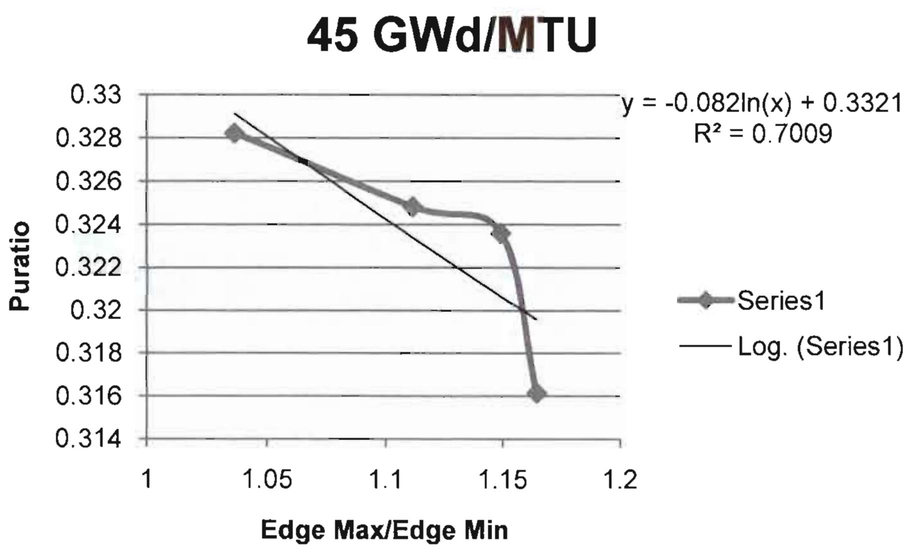
$$xl = number\ of\ pins\ x\ direction;\ yl = number\ of\ pins\ in\ y\ direction$$

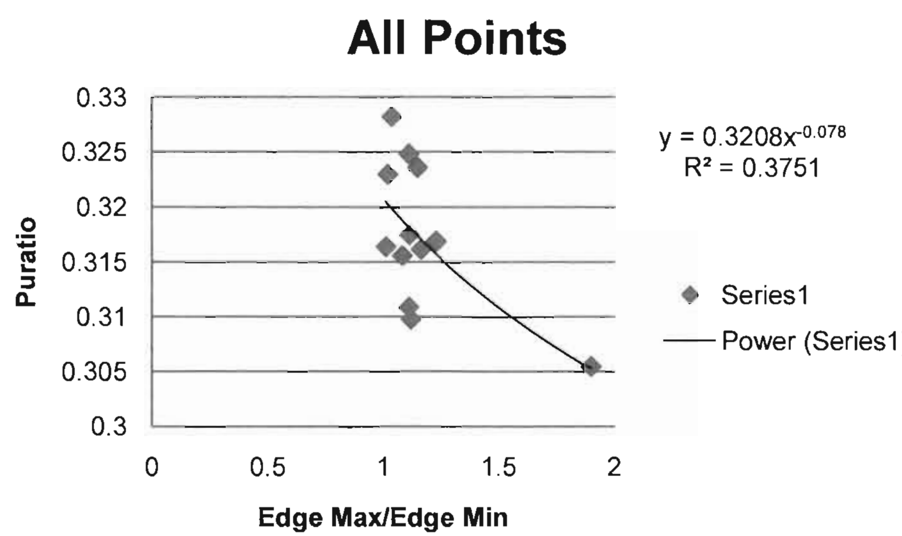
Fits


15 GWd/MTU
y = -0.014ln(x) + 0.3136
R² = 0.7371
0.318
0.316
0.314
0.312
0.31
0.308
0.306
0.304
Puratio
1
1.2
1.4
1.6
1.8
2
Edge Max/Edge Min
Series1
Log. (Series1)


30 GWd/MTU
y = -0.026ln(x) + 0.3209
R² = 0.3895
0.324
0.323
0.322
0.321
0.32
0.319
0.318
0.317
0.316
0.315
Puratio
1
1.05
1.1
1.15
1.2
1.25
Edge Max/Edge Min
Series1
Log. (Series1)

Fits


45 GWd/MTU
0.33
0.328
0.326
0.324
0.322
0.32
0.318
0.316
0.314
Puratio
1
1.05
1.1
1.15
1.2
Edge Max/Edge Min
y = -0.082ln(x) + 0.3321
R² = 0.7009
Series1
Log. (Series1)


All Points
0.33
0.325
0.32
0.315
0.31
0.305
0.3
Puratio
0
0.5
1
1.5
2
Edge Max/Edge Min
y = 0.3208x-0.078
R² = 0.3751
Series1
Power (Series1)

Assembly Pu Prediction

- **Using "All Points" fitted function (if you didn't know burnup)**

		All Points Fitted prediction					
		Shuffling Sequence 1		Shuffling Sequence 2		Shuffling Sequence 3	
		Prediction (g)	% Error	Prediction (g)	% Error	Prediction (g)	% Error
15 GWd/MTU	0d	2018.56	**-2.29%**	1980.42	**-2.58%**	1916.53	**0.10%**
	5y	2032.76	**-2.18%**	1995.88	**-2.47%**	1921.87	**0.00%**
	80y	1923.25	**-1.43%**	1891.30	**-1.74%**	1808.09	**-0.01%**
30 GWd/MTU	0d	3119.57	**-1.02%**	3166.04	**-0.20%**	3061.13	**0.38%**
	5y	3073.49	**-0.89%**	3114.02	**-0.11%**	3020.28	**0.38%**
	80y	2765.69	**-0.38%**	2796.56	**0.27%**	2718.13	**0.70%**
45 GWd/MTU	0d	3807.44	**-0.27%**	3915.20	**1.97%**	3840.68	**2.09%**
	5y	3700.76	**-0.29%**	3813.36	**1.97%**	3758.54	**2.09%**
	80y	3254.28	**-0.27%**	3346.51	**1.91%**	3305.09	**2.27%**