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# IER 121: Updates on the NeSO benchmark

Alexander R. Clark

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*Nuclear Criticality Safety Program Technical Program Review*

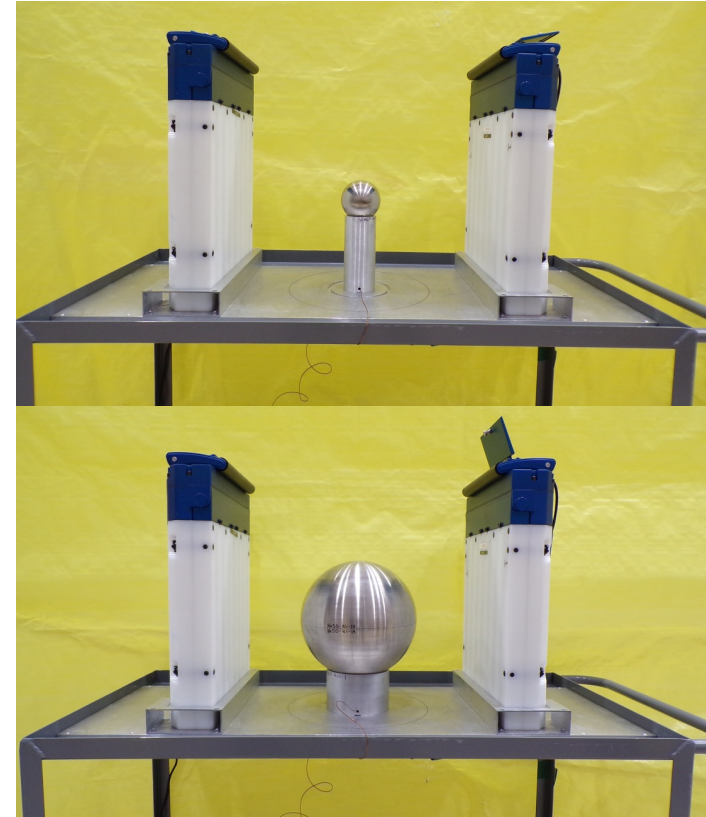
February 20-22, 2024

LA-UR 24-21132

# NeSO benchmark addresses need for improved neptunium nuclear data validation

- **Neptunium Subcritical Observation**
- Measurements performed during 2019 in collaboration with IRSN
- ~6 kg neptunium metal sphere clad with nickel and tungsten shells
- Measured configurations
  - Neptunium sphere bare and reflected by 0.6, 1.1, 1.6, 2.1, 2.6, 3.1, and 3.6 inches of nickel
  - Cf-252 source measurements for detector efficiency
- Purpose:
  - Improve characterization of the neptunium sphere
  - Validate neptunium nuclear data
  - Keff sensitivities to neptunium nuclear data are greater than any currently in the ICSBEP

ID	HEU mass (kg)	keff sensitivity for Np237 fission
SMF003	17.7 kg + 1000 kg NU	1.90E-03
SMF008	62.6	1.59E-01
SMF011	27.0	1.64E-01
SMF014	34.3	1.85E-01
NeSO bare	0	8.22E-01



Neptunium sphere measured by linked NOMADs in bare (top) and 3.6-in-nickel-reflected (bottom) configurations.

# NeSO benchmark is difficult to model correctly due to poorly-characterized impurities

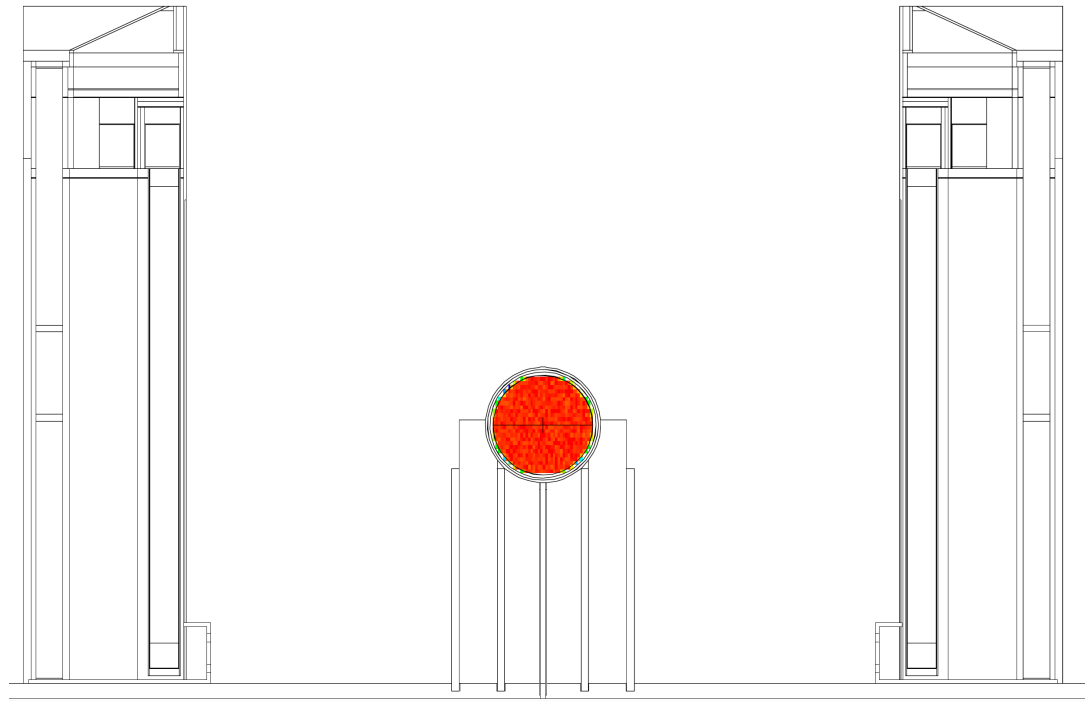
- Originally 8 kg of  $^{237}\text{NpO}_2$  from Savannah River, Purex reprocessing of DOE reactor fuels.
- Cast for neptunium sphere was too large, resulting in a concentration of impurities distributed non-uniformly
- Composition analysis done by C-NR in 2022 found “significant quantities and unusual isotope profiles of americium and curium contaminants”<sup>1</sup>
- Subcritical neutron measures (i.e. leakage multiplication and count rate) simulated with both original (SPEC-MET-FAST-008) and C-NR neptunium compositions disagree with measurement

isotope	measured atoms/g A solution	% unc.	Neutrons/s
$^{237}\text{Np}$	1.87E+18	1.35	0.68
$^{238}\text{Pu}$	2.37E+13	0.98	196.61
$^{239}\text{Pu}$	5.33E+14	0.90	0.04
$^{240}\text{Pu}$	3.84E+13	0.95	125.45
$^{241}\text{Pu}$	3.78E+11	4.90	0.00
$^{242}\text{Pu}$	5.48E+12	1.20	30.19
$^{244}\text{Pu}$	$L_D = 9.1\text{E}+09$		0.00
$^{241}\text{Am}$	7.90E+12	0.91	0.03
$^{242\text{m}}\text{Am}$	$L_D = 9.0\text{E}+09$		0.00
$^{243}\text{Am}$	2.00E+15	0.90	0.00
$^{244}\text{Cm}$	3.05E+11	1.42	20515.15
$^{233}\text{U}$	7.79E+13	0.37	0.00
$^{234}\text{U}$	1.42E+13	0.41	0.00
$^{235}\text{U}$	4.95E+14	0.38	0.00
$^{236}\text{U}$	2.80E+12	0.41	0.00
$^{238}\text{U}$	5.05E+13	1.94	0.00

1. Sean D. Reilly et. al, “Trace Actinide Signatures of a Bulk Neptunium Sample”, *Analytical Chemistry* 2023 95 (23), 9123-9129

# Consider several impurity bounding cases to determine plausible range of simulated subcritical measures

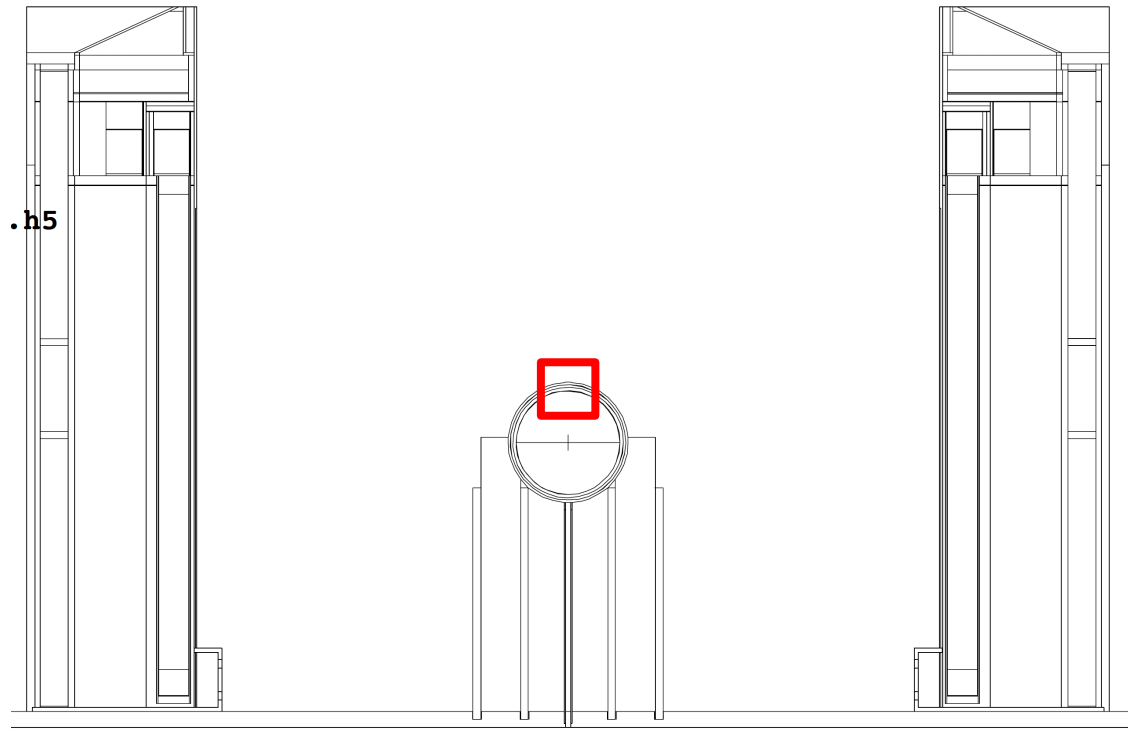
- Vary the neutron source volume distribution
  - Uniform
  - point\_top
  - point\_detector
  - point\_center
  - zplane\_pos\_x
    - x=1 is closest to uniform
    - x=6 is closest to point
    - x=3a-d are subdivisions between x=3 and x=4



MCNP6.3 FMESH plot of uniformly sampled source sites.

# Consider several impurity bounding cases to determine plausible range of simulated subcritical measures

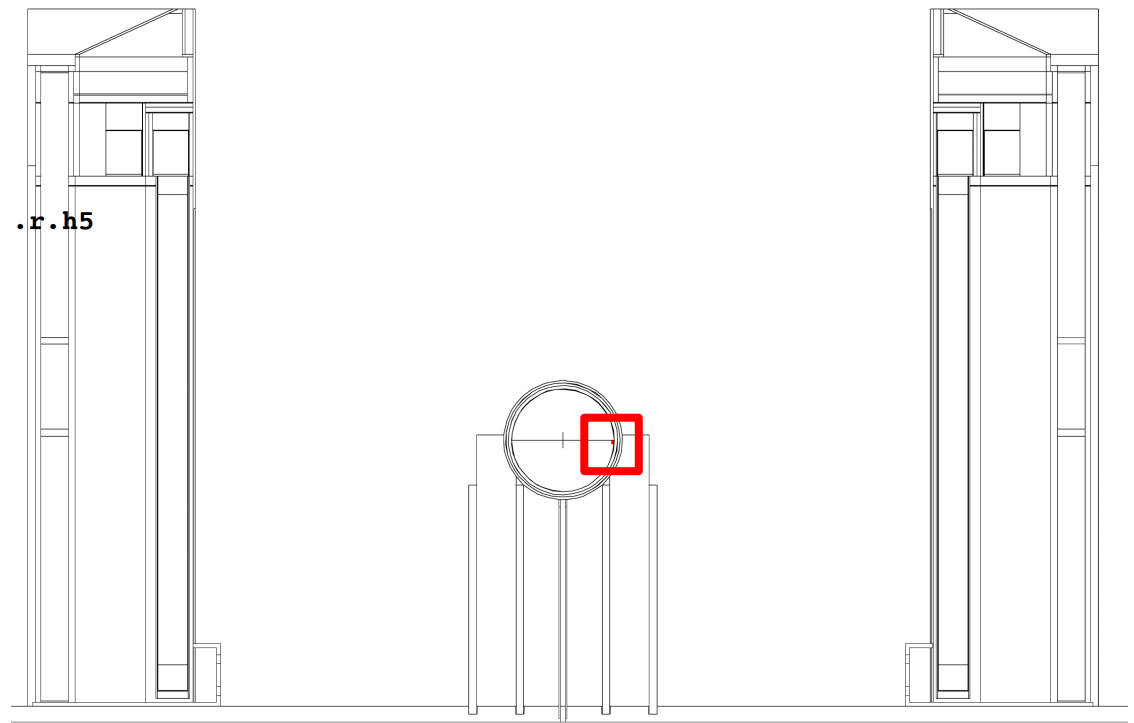
- Vary the neutron source volume distribution
  - Uniform
  - point\_top
  - point\_detector
  - point\_center
  - zplane\_pos\_x
    - x=1 is closest to uniform
    - x=6 is closest to point
    - x=3a-d are subdivisions between x=3 and x=4



MCNP6.3 FMESH plot of point source sites at the top.

# Consider several impurity bounding cases to determine plausible range of simulated subcritical measures

- Vary the neutron source volume distribution
  - Uniform
  - point\_top
  - point\_detector
  - point\_center
  - zplane\_pos\_x
    - x=1 is closest to uniform
    - x=6 is closest to point
    - x=3a-d are subdivisions between x=3 and x=4

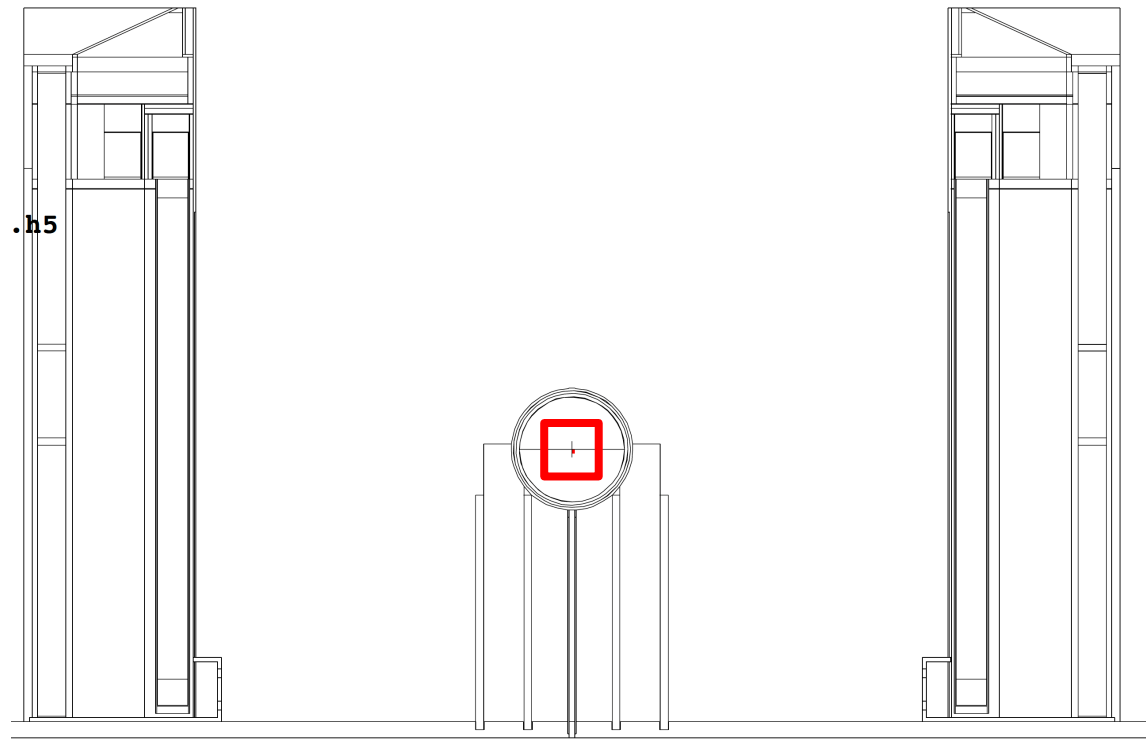


MCNP6.3 FMESH plot of point source sites closer to one NOMAD than another.



# Consider several impurity bounding cases to determine plausible range of simulated subcritical measures

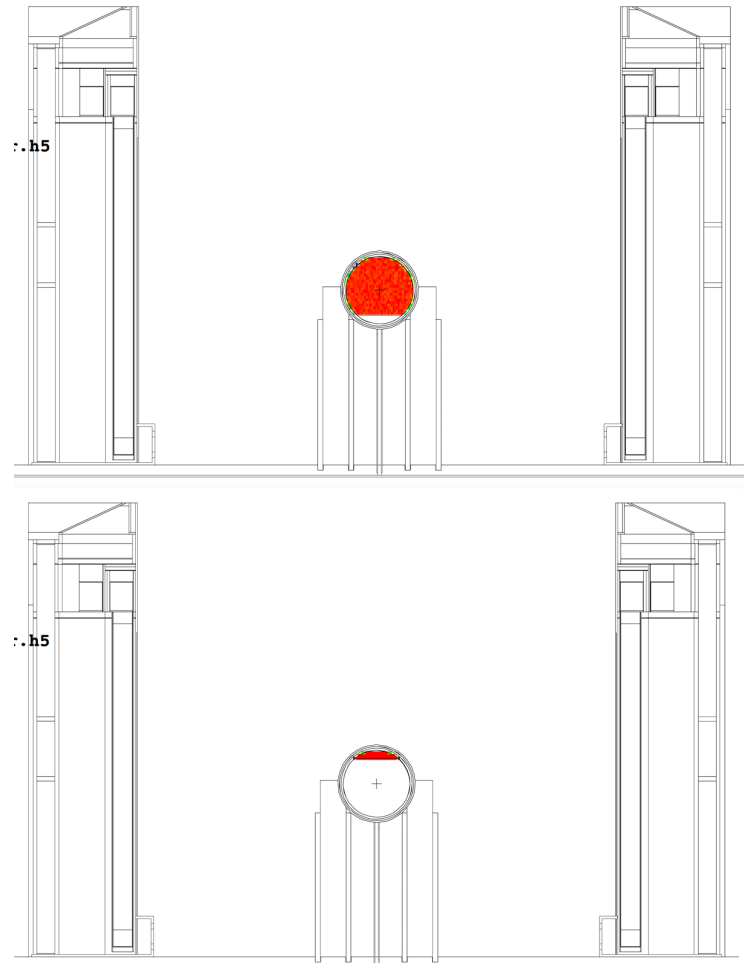
- Vary the neutron source volume distribution
  - Uniform
  - point\_top
  - point\_detector
  - point\_center
  - zplane\_pos\_x
    - x=1 is closest to uniform
    - x=6 is closest to point
    - x=3a-d are subdivisions between x=3 and x=4



MCNP6.3 FMESH plot of point source sites at the center.

# Consider several impurity bounding cases to determine plausible range of simulated subcritical measures

- Vary the neutron source volume distribution
  - Uniform
  - point\_top
  - point\_detector
  - point\_center
  - zplane\_pos\_x
    - x=1 is closest to uniform
    - x=6 is closest to point
    - x=3a-d are subdivisions between x=3 and x=4



MCNP6.3 FMESH plots of uniformly sampled source sites above zplane\_pos\_1 (top) and zplane\_pos\_6 (bottom).

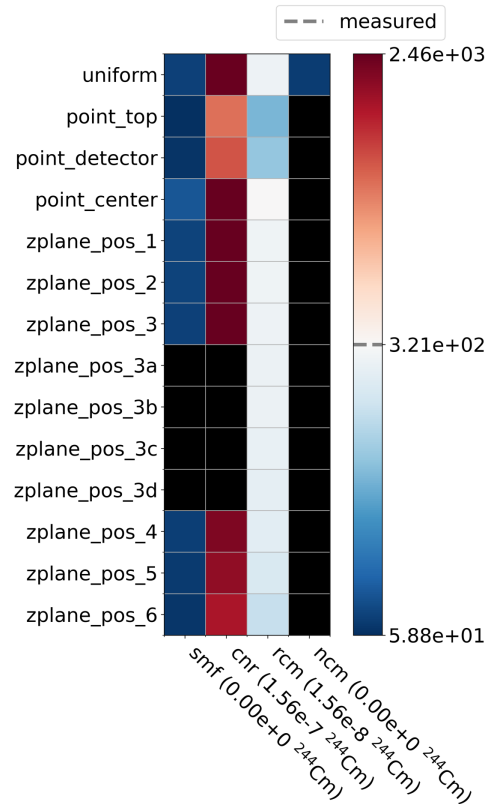
# Consider several impurity bounding cases to determine plausible range of simulated subcritical measures

- Vary the neutron source material composition
  - SMF: the benchmark SPEC-MET-FAST-008
  - CNR: the 2022 C-NR analysis
  - RCM: the 2022 C-NR analysis with reduced Cm-244 content
  - NCM: the 2022 C-NR analysis no Cm-244 content
- Use the **MCNP® Intrinsic Source Constructor (MISC)** to compute the source energy distribution for each material composition

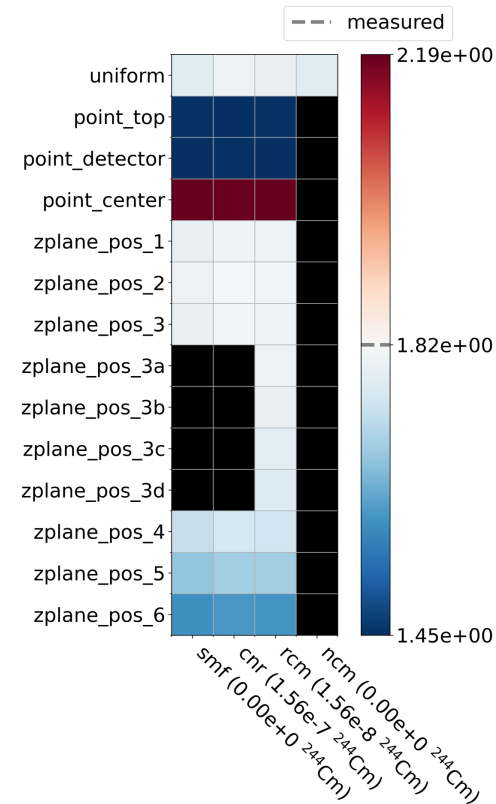
ID	Cm-244 atom fraction
SMF	0
CNR	1.55847E-07
RCM	1.55847E-08
NCM	0

# Subcritical measures vary strongly with respect to source volume distribution and material composition

NeSO\_Exp\_1 mean neutron count rates



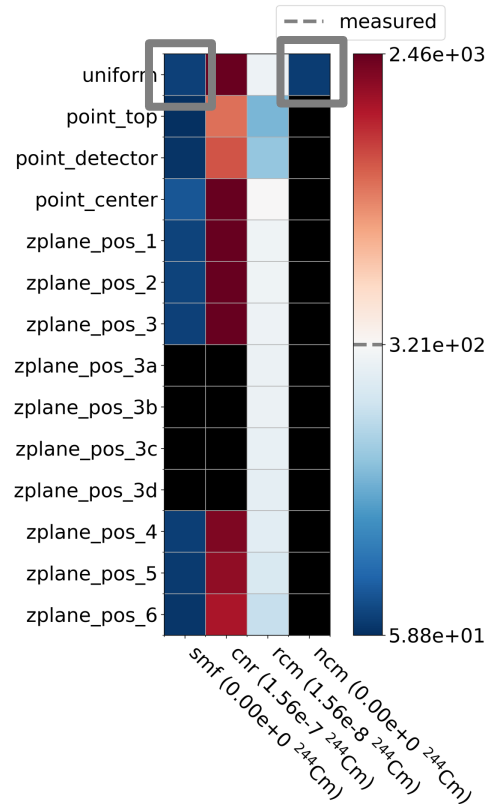
NeSO\_Exp\_1 neutron leakage multiplication



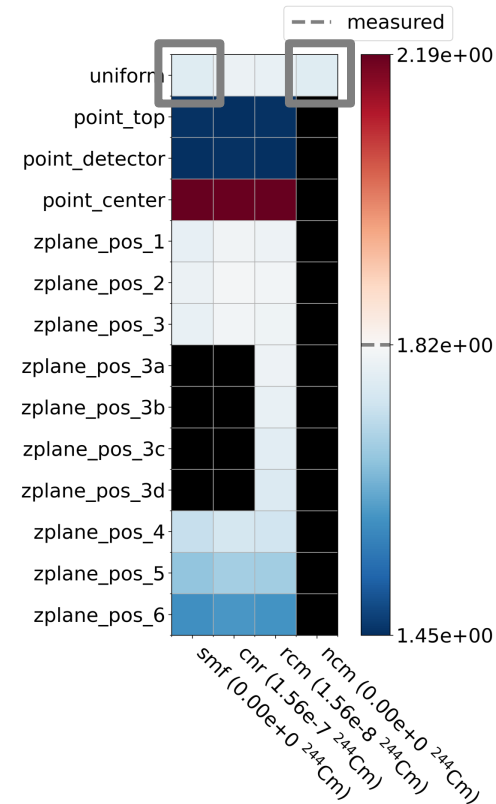
Plots of mean neutron count rate (left) and neutron leakage multiplication (right) as a function of volume distribution and material composition IDs and annotated with the measured quantity. The mean neutron count rate varies strongly w.r.t. material composition while neutron leakage multiplication varies strongly w.r.t. volume distribution. Black elements indicate non-existent combinations.

# Subcritical measures vary strongly with respect to source volume distribution and material composition

NeSO\_Exp\_1 mean neutron count rates



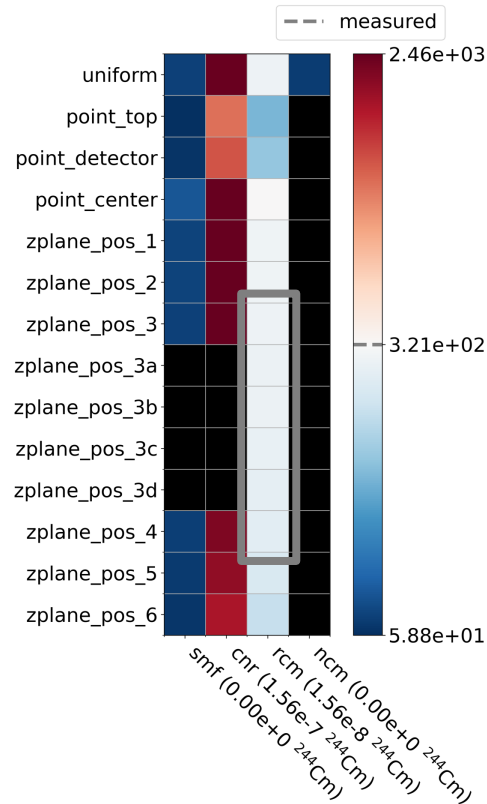
NeSO\_Exp\_1 neutron leakage multiplication



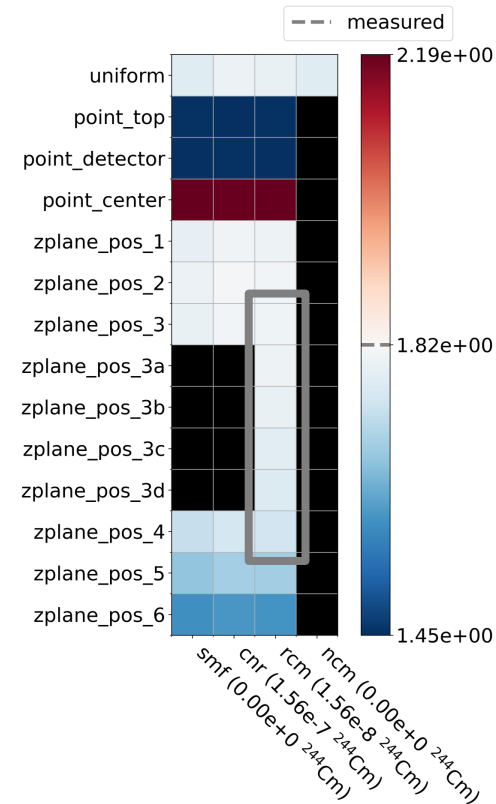
Plots of mean neutron count rate (left) and neutron leakage multiplication (right) as a function of volume distribution and material composition IDs and annotated with the measured quantity. The mean neutron count rate varies strongly w.r.t. material composition while neutron leakage multiplication varies strongly w.r.t. volume distribution. Black elements indicate non-existent combinations.

# Subcritical measures vary strongly with respect to source volume distribution and material composition

NeSO\_Exp\_1 mean neutron count rates



NeSO\_Exp\_1 neutron leakage multiplication



Plots of mean neutron count rate (left) and neutron leakage multiplication (right) as a function of volume distribution and material composition IDs and annotated with the measured quantity. The mean neutron count rate varies strongly w.r.t. material composition while neutron leakage multiplication varies strongly w.r.t. volume distribution. Black elements indicate non-existent combinations.

# Conclusions

- Difficulties in casting the neptunium sphere resulted in difficulties in characterizing the intrinsic source term due to impurities
- Considered a variety of bounding cases for source term volume distribution and impurity content to explore plausible subcritical measure values
- Identified a source characterization that provides good agreement for both mean neutron count rate and neutron leakage multiplication
- Only leakage multiplication will be used as a benchmark parameter
  - Count rate varies too strongly with Cm-244 content
  - Credible volume distribution leads to acceptable leakage multiplication uncertainty

# Future work

- Complete update and review of Section 1 for the CED-3b milestone
- Document our impurity characterization in Sections 1 and 2
- Perform uncertainty quantification of other benchmark factors for Section 2
- Receive feedback on our approach to characterize the impurity



# Acknowledgements

- This work was supported by the DOE Nuclear Criticality Safety Program, funded and managed by the National Nuclear Security Administration for the Department of Energy. This measurement was done in collaboration with IRSN.



# Backup slides

# MCNP material cards

```
c Np sphere region with impurities defined by the 2022 CNR analysis
c Info given:
c Mass: 6070.4 g
c Outer radius: 1.6335 in
c Computed:
c Outer radius: 4.14909 cm
c Volume: 299.1901 cm^3
c Density: 20.28944 g/cm^3
c -----
m11 91233 3.39722000e-08
    92233 4.20042000e-05
    92234 7.69203000e-06
    92235 2.63314000e-04
    92236 1.49758000e-06
    92238 2.70173000e-05
    93237 9.98267000e-01
    94238 1.25480000e-05
    94239 2.84015000e-04
    94240 2.05193000e-05
    94241 1.91089000e-07
    94242 2.93069000e-06
    94244 4.21287000e-06
    95241 1.08435000e-08
    95243 1.06684000e-03
    96244 1.55847000e-07

c Np sphere region without impurities, defined by SMF-008
c Info given:
c Mass: 6070.4 g
c Outer radius: 1.6335 in
c Computed:
c Outer radius: 4.14909 cm
c Volume: 299.1901 cm^3
c Density: 20.28944 g/cm^3
c -----
m1 91233 3.39457000e-08
    92233 3.67200000e-05
    92234 5.94873000e-06
    92235 2.75678000e-04
    92236 1.53736000e-06
    92238 3.06067000e-05
    93237 9.97488000e-01
    93239 1.56963000e-09
    94238 1.59834000e-05
    94239 3.18880000e-04
    94240 2.27554000e-05
    94241 5.77639000e-07
    94242 3.14018000e-06
    95241 6.55803000e-06
    95243 1.79349000e-03
```

MCNP material cards from the SMF-008 benchmark specification<sup>1</sup> (left) and the 2022 C-NR analysis<sup>2</sup> (right).

1. Nuclear Energy Agency, "NEPTUNIUM-237 SPHERE SURROUNDED BY HEMISPHERICAL SHELLS OF HIGHLY ENRICHED URANIUM", *International Handbook of Evaluated Criticality Safety Benchmark Experiments*, NEA/NSC/DOC/(95)03/VII (SPEC-MET-FAST-008), Table 6, 2020
2. Sean D. Reilly et. al, "Trace Actinide Signatures of a Bulk Neptunium Sample", *Analytical Chemistry* 2023 95 (23), 9123-9129