Pacific Northwest
NATIONAL LABORATORY

Production and Abatement of Non-Traditional Xenon Isotopes at a Spallation Neutron Source

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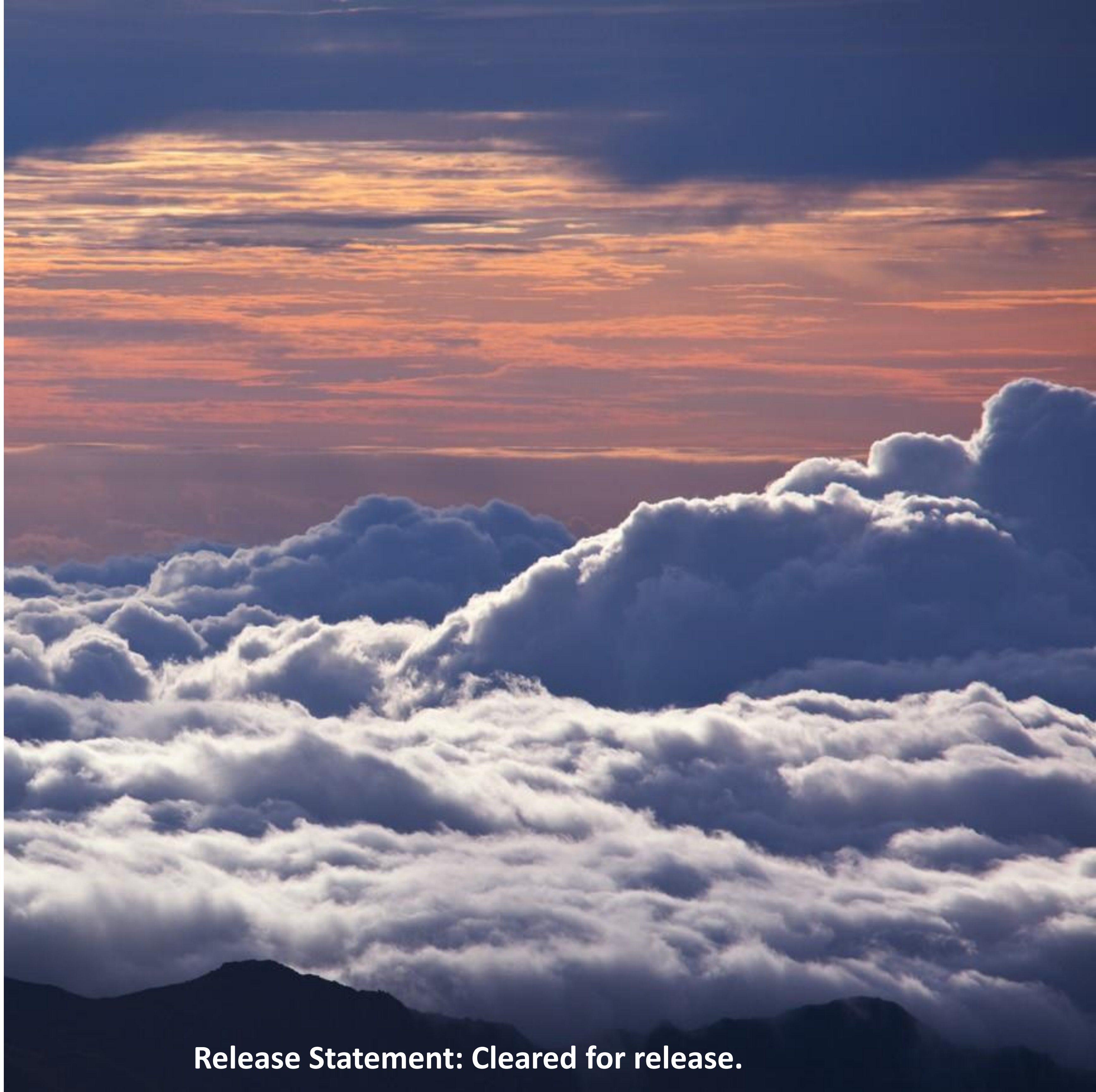


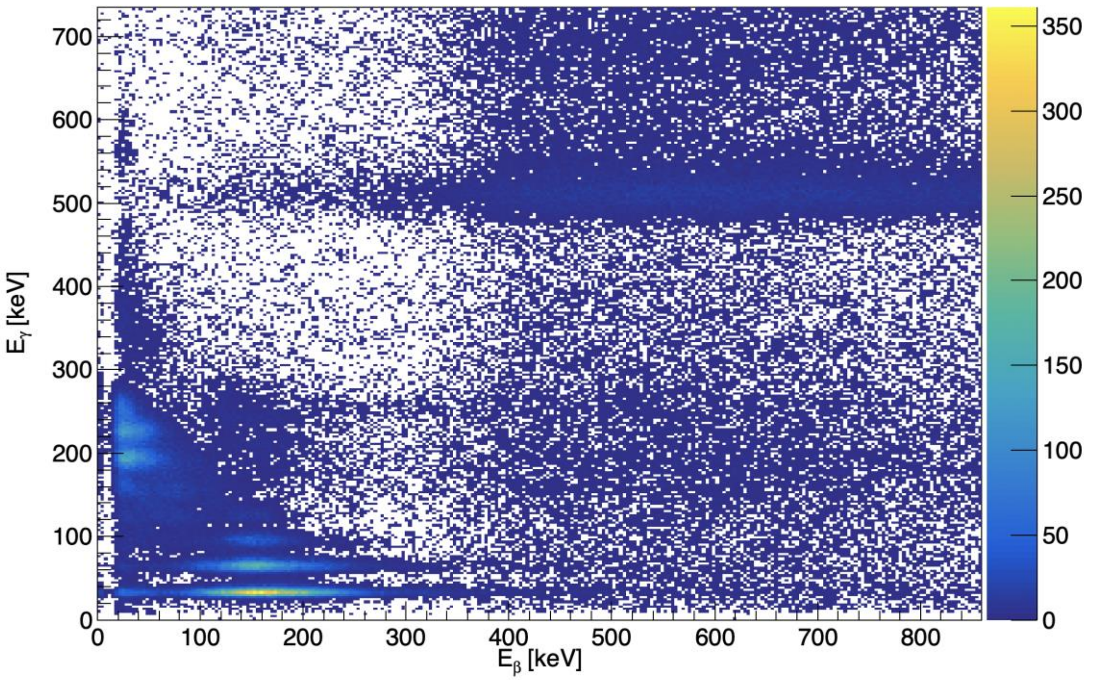
Release Statement: Cleared for release.

New Sources of Radioxenon

- Anthropogenic sources of radioxenon are expanding
  - Current Nuclear Reactors
  - Medical Isotope Production
  - Accelerator Facilities
    - ✓ E.g., Spallation Neutron Source
  - Advanced Nuclear Reactors
    - ✓ E.g., Molten Salt Reactors

- These new sources of radioxenon are also expanding the isotopes potentially detected at IMS stations

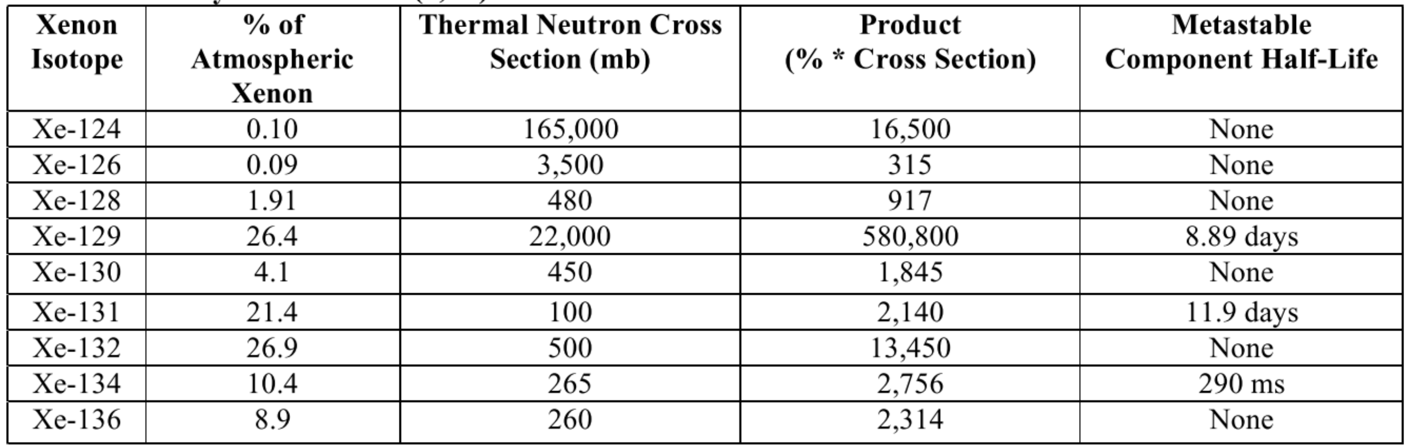
Xe	119	3.48E+02	8.43E-01	2.34E-02
Xe	121	2.41E+03	1.27E+01	7.57E+00
Xe	122	7.24E+04	8.44E+00	8.29E+00
Xe	123	7.49E+03	2.46E+01	2.08E+01
Xe	125	6.08E+04	9.80E+01	9.65E+01
Xe	125*	5.70E+01	1.20E+01	3.75E-09
Xe	127	3.15E+06	1.08E+02	1.08E+02
Xe	127*	6.92E+01	2.12E+00	3.13E-08
Xe	129*	7.68E+05	6.17E+00	6.16E+00
Xe	131*	1.03E+06	4.43E+00	4.42E+00
Xe	133			
Xe	133*			
Xe	134*			
Xe	135			



Non-traditional Xenon Isotopes

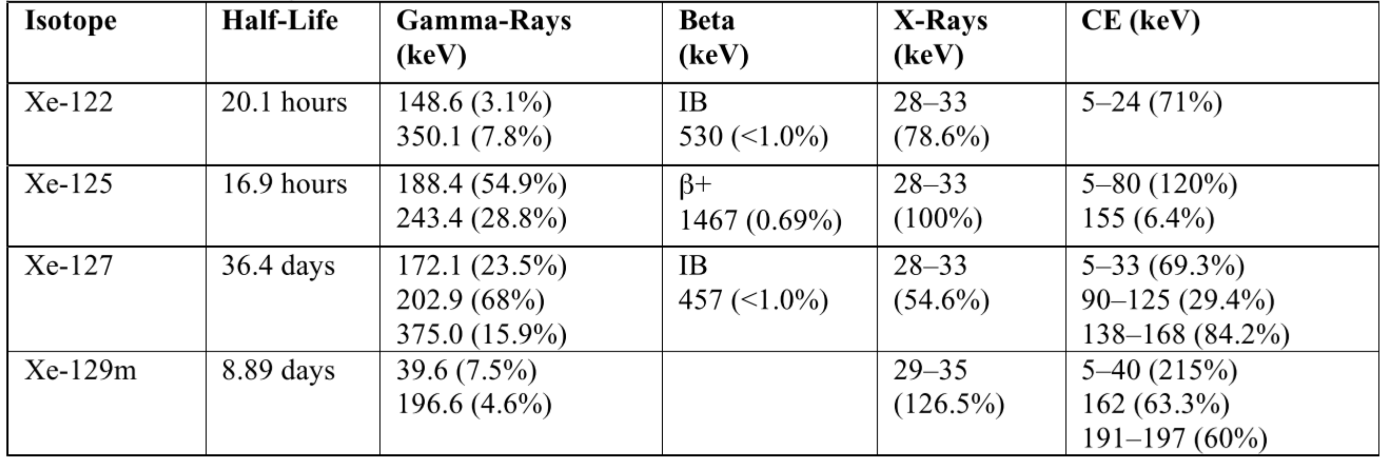
- Many isotopes can be produced via neutron irradiation
    - ^{127}Xe
    - ^{125}Xe
    - ^{129m}Xe
- Neutron irradiation isotopes have been previously investigated and observed
- ^{122}Xe produced as medical isotope via proton or alpha bombardment
    - Not studied previously

Table 1. A listing of the all the stable xenon isotopes along with pertinent information for each. Clearly Xe-124 has the highest thermal neutron cross-section followed by Xe-129. The production of Xe-129m actually comes from a (*n*,2*n*) reaction on Xe-130.



Xenon Isotope	% of Atmospheric Xenon	Thermal Neutron Cross Section (mb)	Product (% * Cross Section)	Metastable Component Half-Life
Xe-124	0.10	165,000	16,500	None
Xe-126	0.09	3,500	315	None
Xe-128	1.91	480	917	None
Xe-129	26.4	22,000	580,800	8.89 days
Xe-130	4.1	450	1,845	None
Xe-131	21.4	100	2,140	11.9 days
Xe-132	26.9	500	13,450	None
Xe-134	10.4	265	2,756	290 ms
Xe-136	8.9	260	2,314	None

Table 2. Data taken from *Table of Radioactive Isotopes*, edited by E. Browne, R. B. Firestone, and V. S. Shirley, 1986.

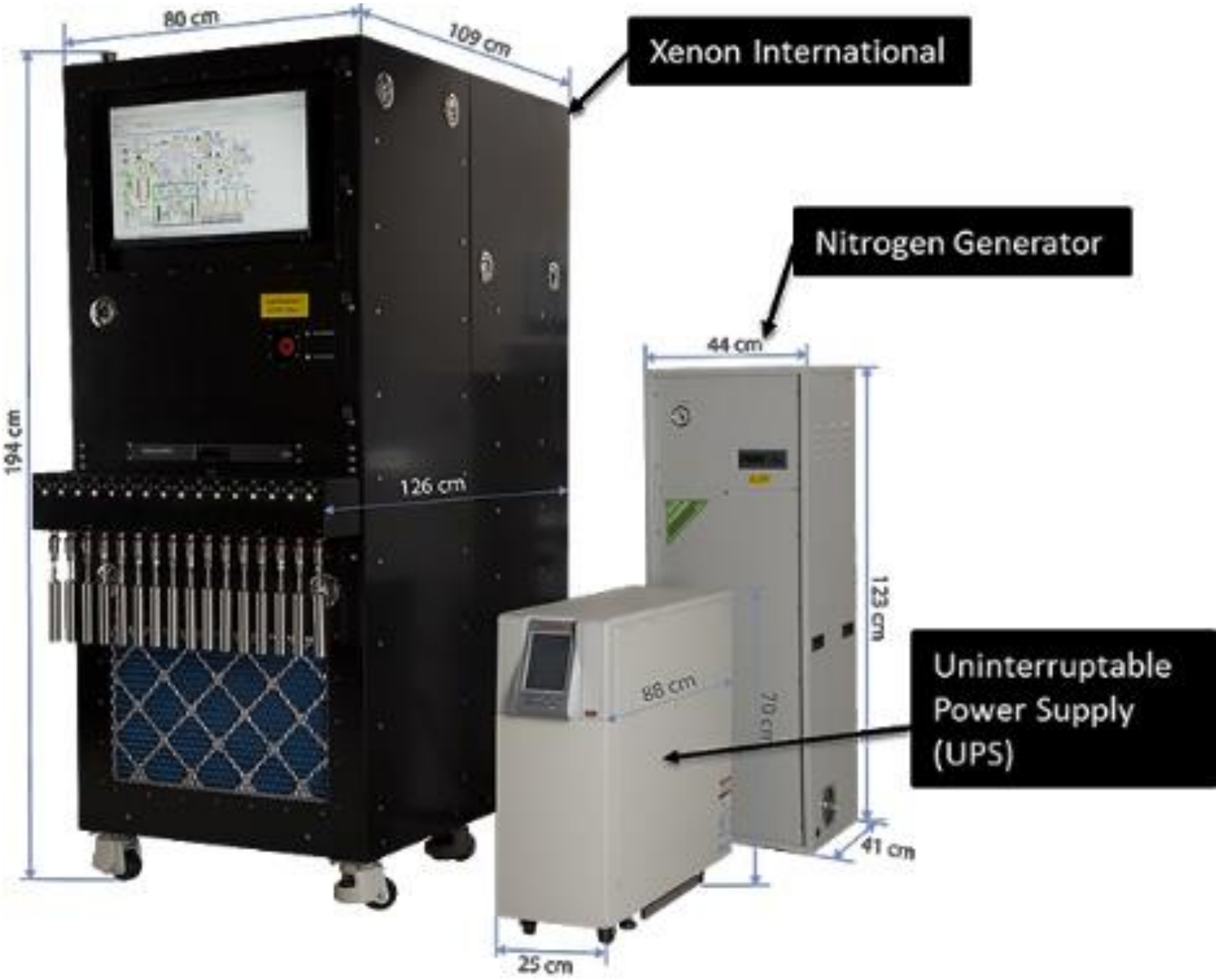


Isotope	Half-Life	Gamma-Rays (keV)	Beta (keV)	X-Rays (keV)	CE (keV)
Xe-122	20.1 hours	148.6 (3.1%) 350.1 (7.8%)	IB 530 (<1.0%)	28–33 (78.6%)	5–24 (71%)
Xe-125	16.9 hours	188.4 (54.9%) 243.4 (28.8%)	β+ 1467 (0.69%)	28–33 (100%)	5–80 (120%) 155 (6.4%)
Xe-127	36.4 days	172.1 (23.5%) 202.9 (68%) 375.0 (15.9%)	IB 457 (<1.0%)	28–33 (54.6%)	5–33 (69.3%) 90–125 (29.4%) 138–168 (84.2%)
Xe-129m	8.89 days	39.6 (7.5%) 196.6 (4.6%)		29–35 (126.5%)	5–40 (215%) 162 (63.3%) 191–197 (60%)

McIntyre, Justin I et al.. 2008. "Generation of Radioxenon Isotopes." In *Proceedings of the 30th Seismic Research Review: Ground Based Nuclear Explosion Monitoring Technologies*, 793–801.

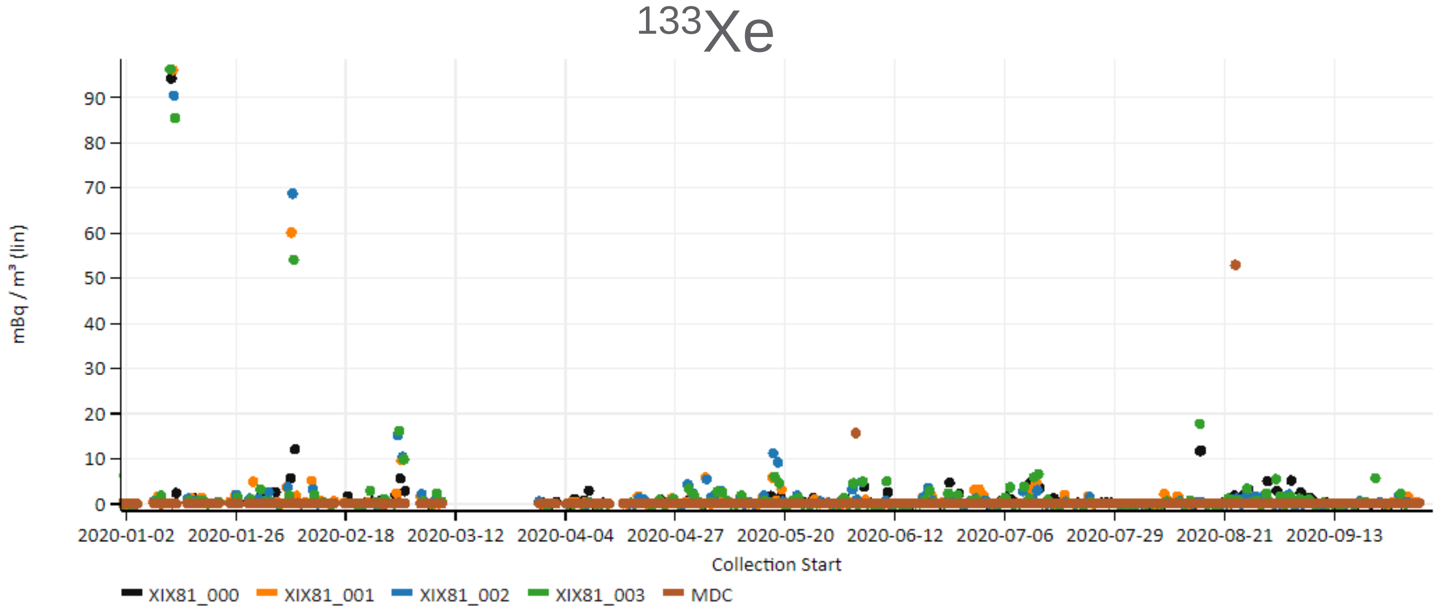
Xenon International

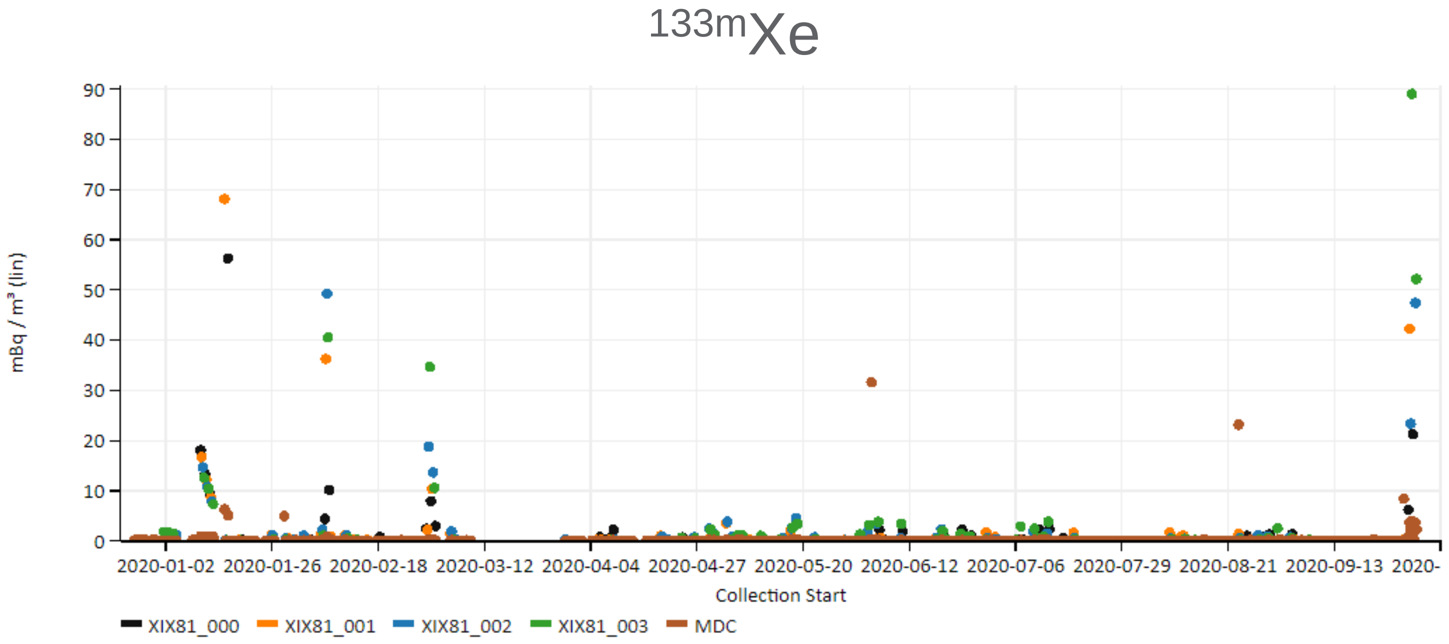
- Next generation atmospheric radioxenon system
- Faster and more sensitive than current generation systems
  - ~2.5 cc of xenon in 6 hours
  - Compared to ~1 cc for SAUNA II in 12 hours
- MDCs
  - <0.15 mBq/SCM for ^{133}Xe, ^{131m}Xe, ^{133m}Xe
  - <0.5 mBq/SCM for ^{135}Xe
- Developed at PNNL
  - Transitioned to Teledyne Brown Engineering for production (Knoxville, TN)



Puzzling Observations and Analyses - Measured at TBE in Knoxville, Tennessee

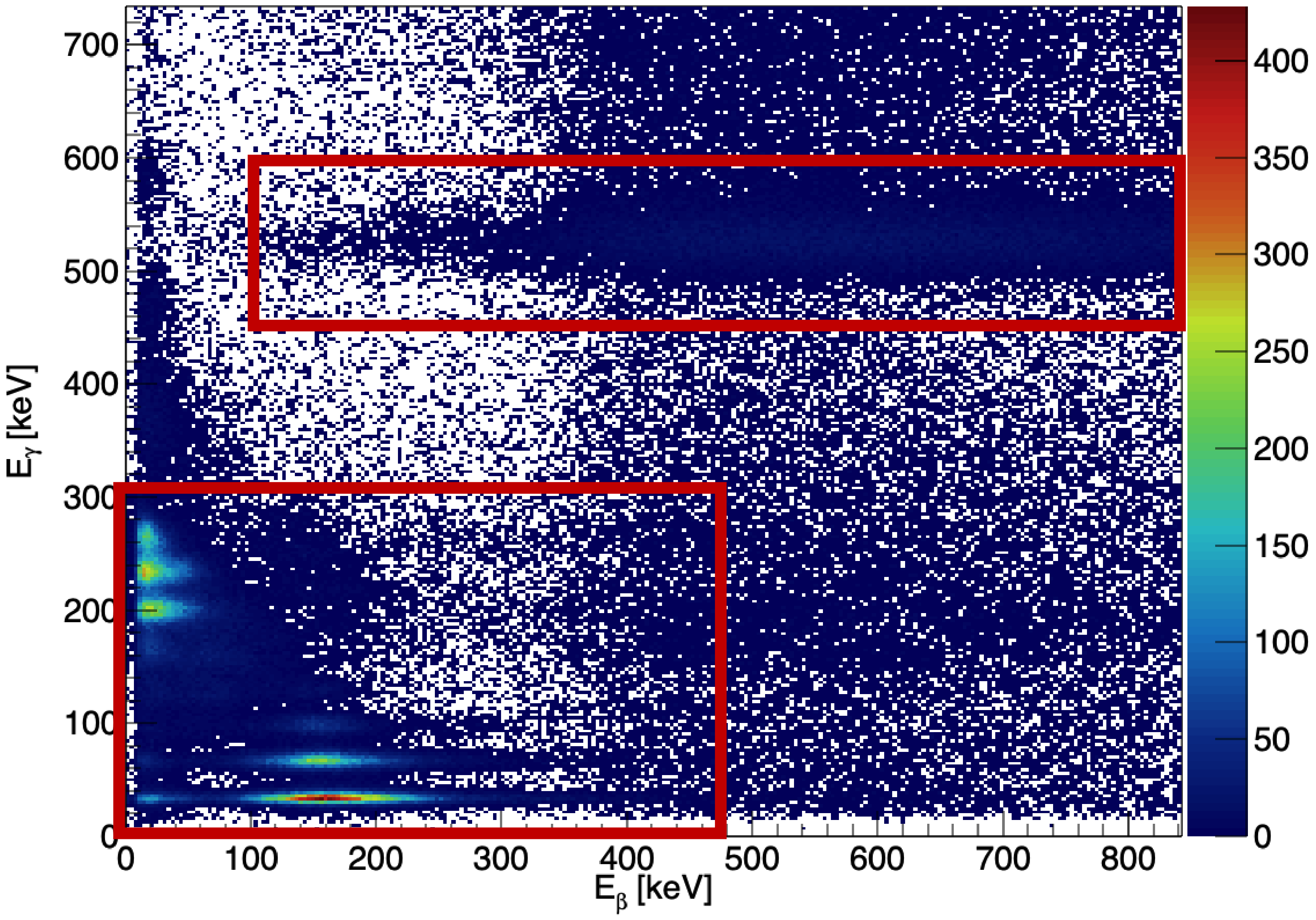
- At initial glance, everything seems to be going well.
  - ^{133}Xe – no unusual levels observed for the area
  - ^{135}Xe – high but also not too unusual for the area
  - ^{133m}Xe – unusual to have higher concentration than ^{133}Xe
  - ^{131m}Xe – similar concentration to ^{133m}Xe
- Further investigation showed that the gamma-ray spectrum has unusual features.



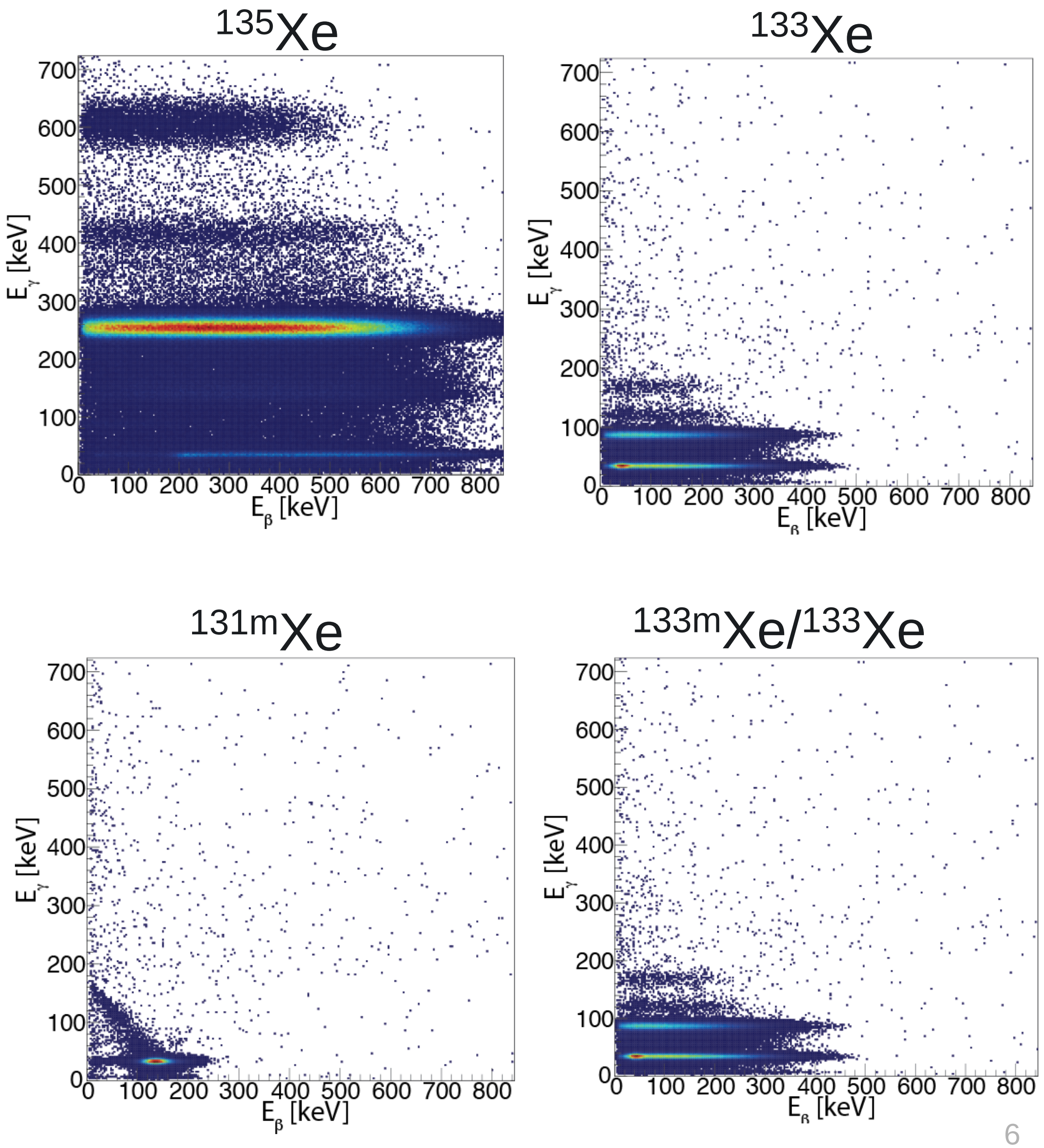


Atypical Coincidence Signatures - Measured at TBE in Knoxville, Tennessee

Spectrum observed on Xenon International



Typical Xenon Signatures



^{135}Xe

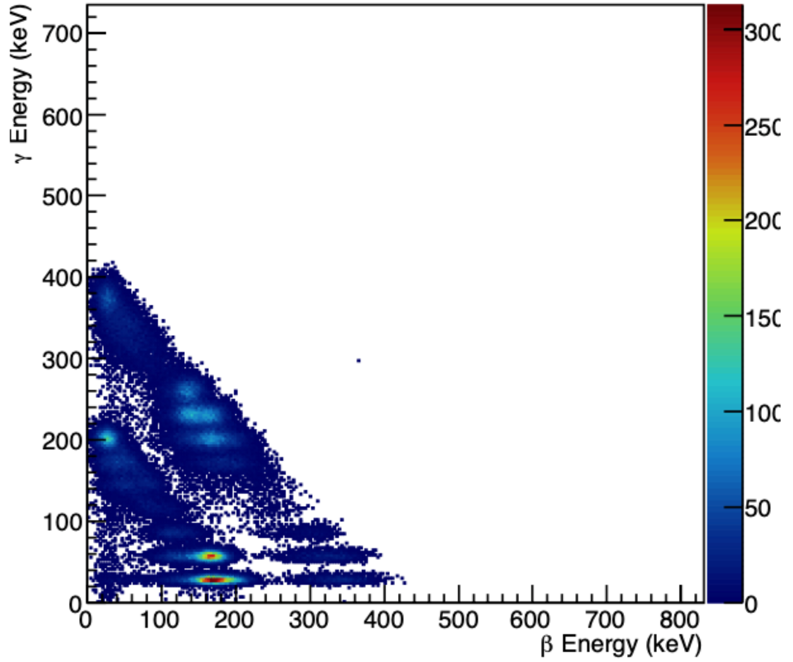
^{133}Xe

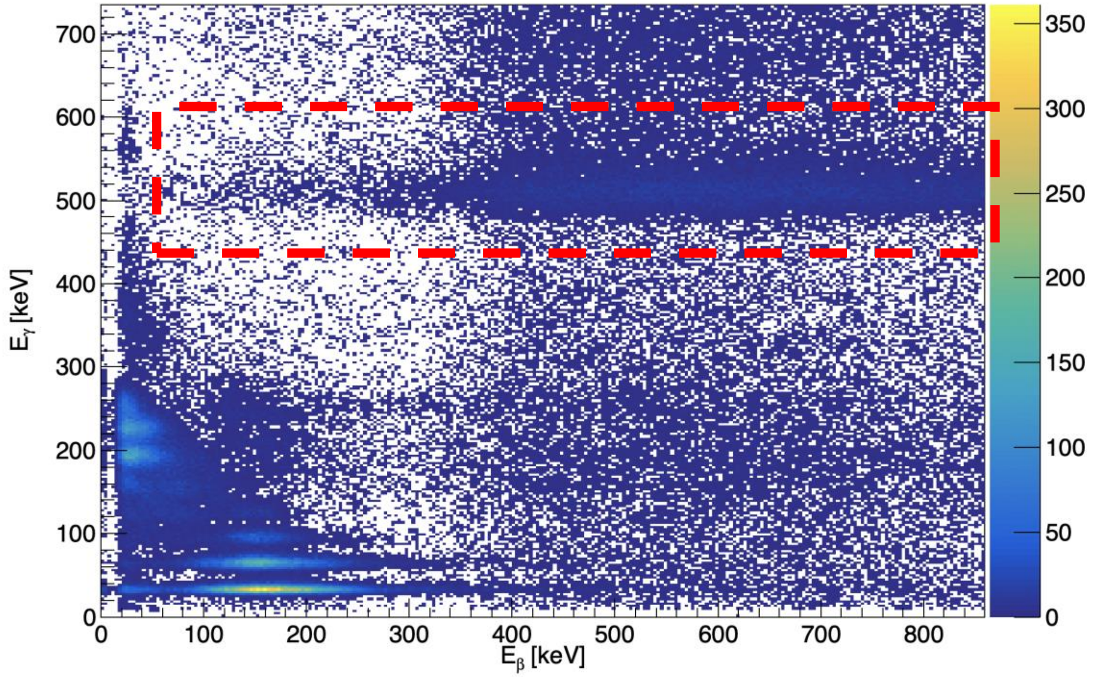
^{131m}Xe

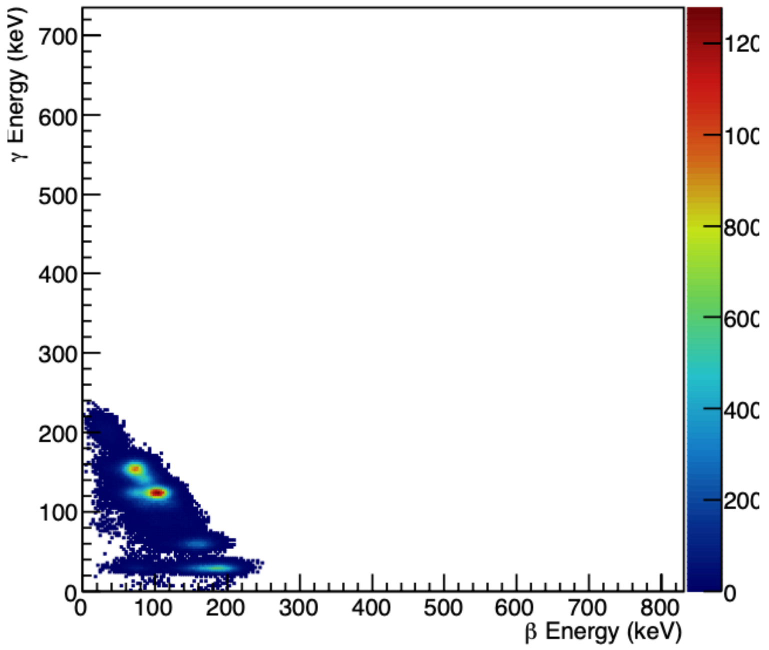
^{133m}Xe/^{133}Xe

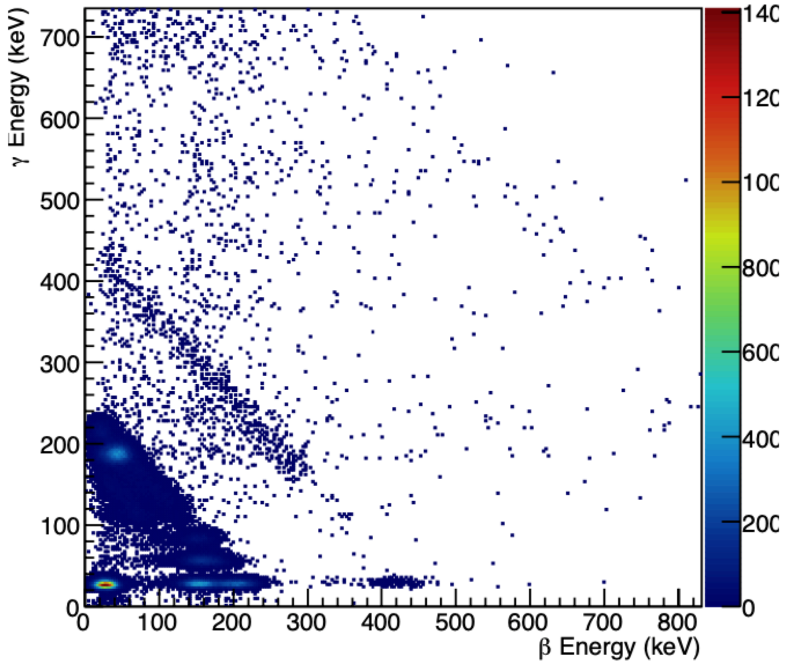
Monte Carlo Simulations

- Monte Carlo simulations performed to observe clean signatures and extract efficiencies
- Region at 511 keV in gamma energy not explained with these simulations


^{127}Xe Simulation

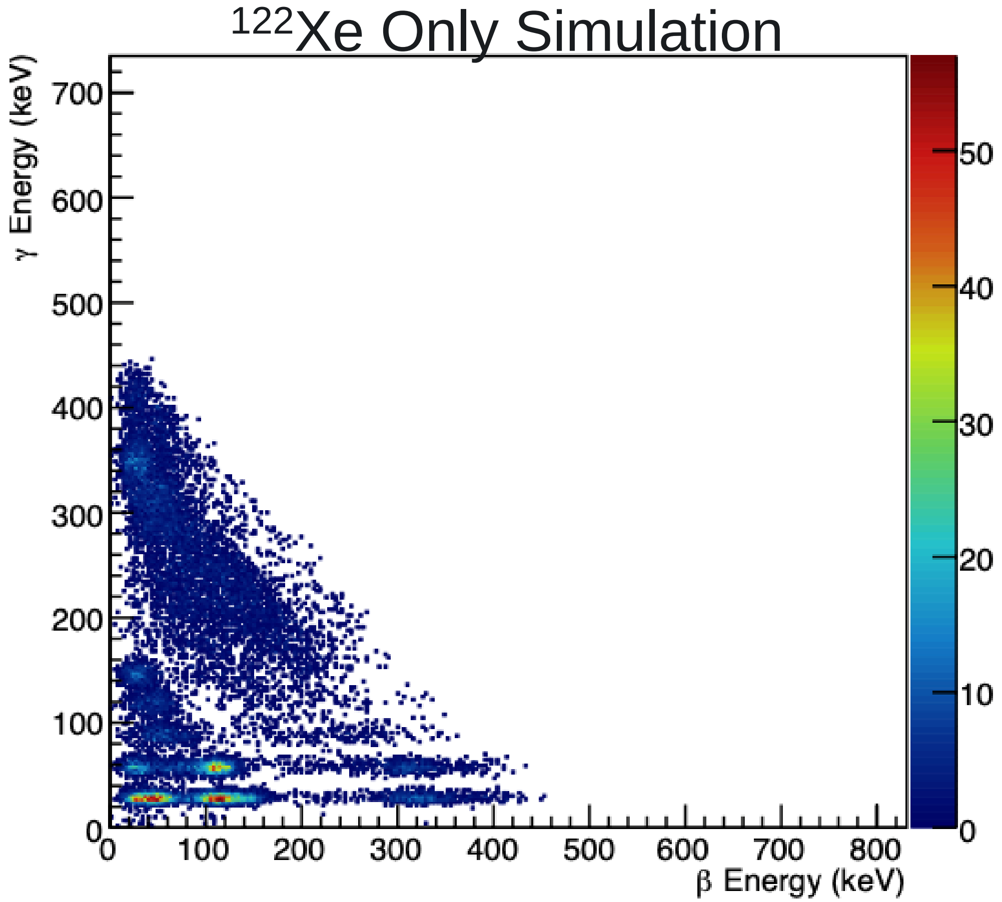

Xenon International Measurement

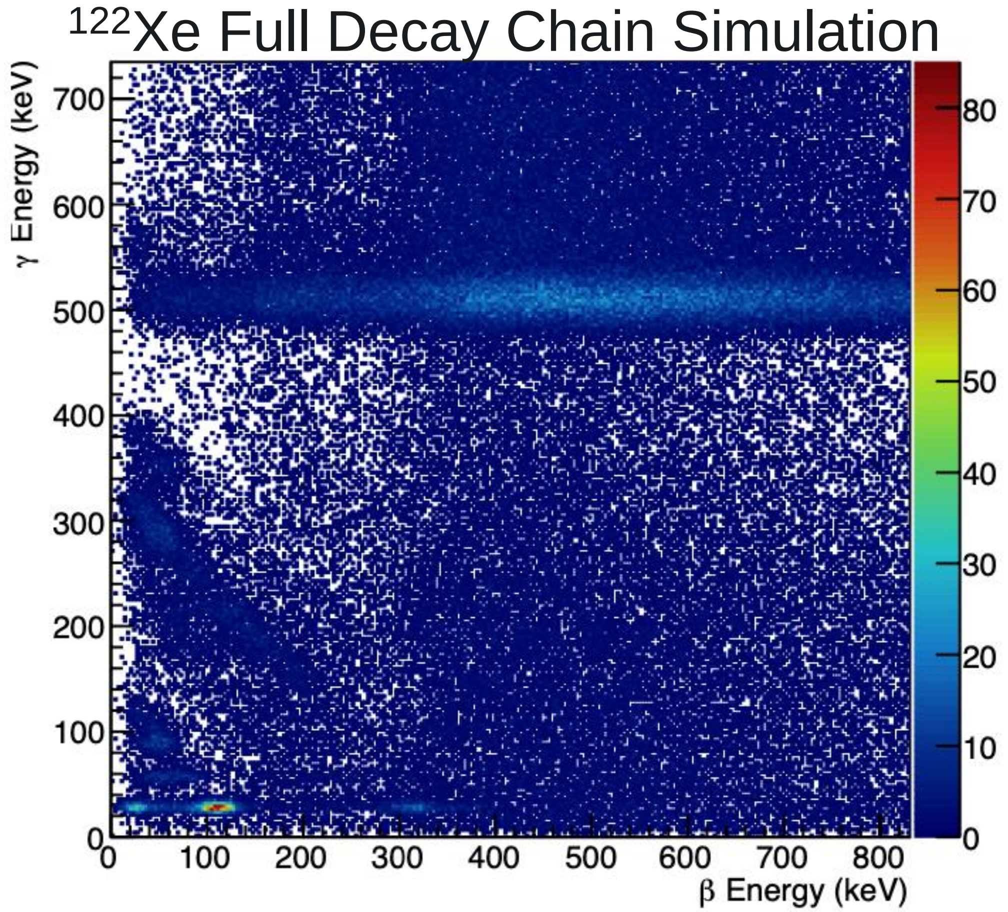

^{129m}Xe Simulation


^{125}Xe Simulation

Missing Piece

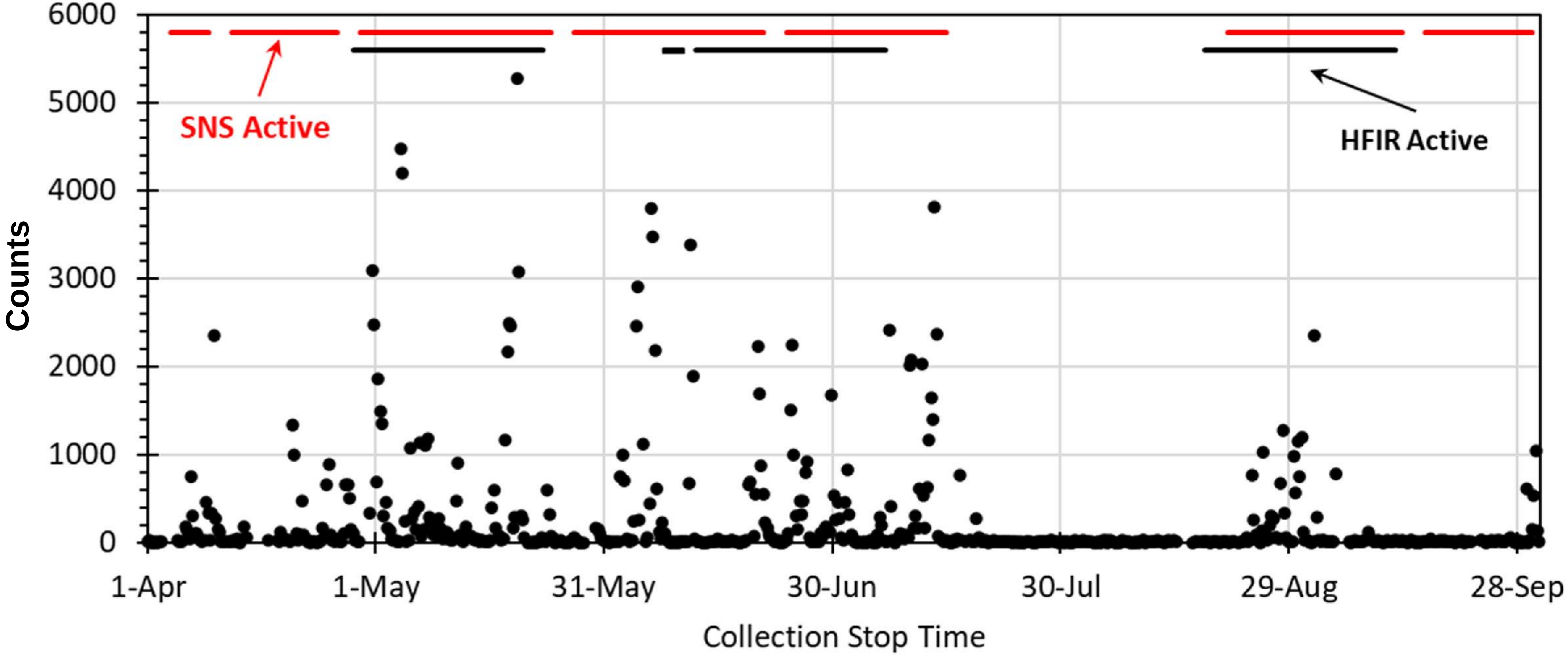
- ^{122}Xe simulation alone did not seem to generate missing signature
    - Including daughter ^{122}I ($T_{1/2}$=3.63 minutes) produces missing signature
- Only possible source of production of ^{122}Xe is via spallation in the mercury target at Spallation Neutron Source (SNS)





Correlation with SNS and HFIR Operation

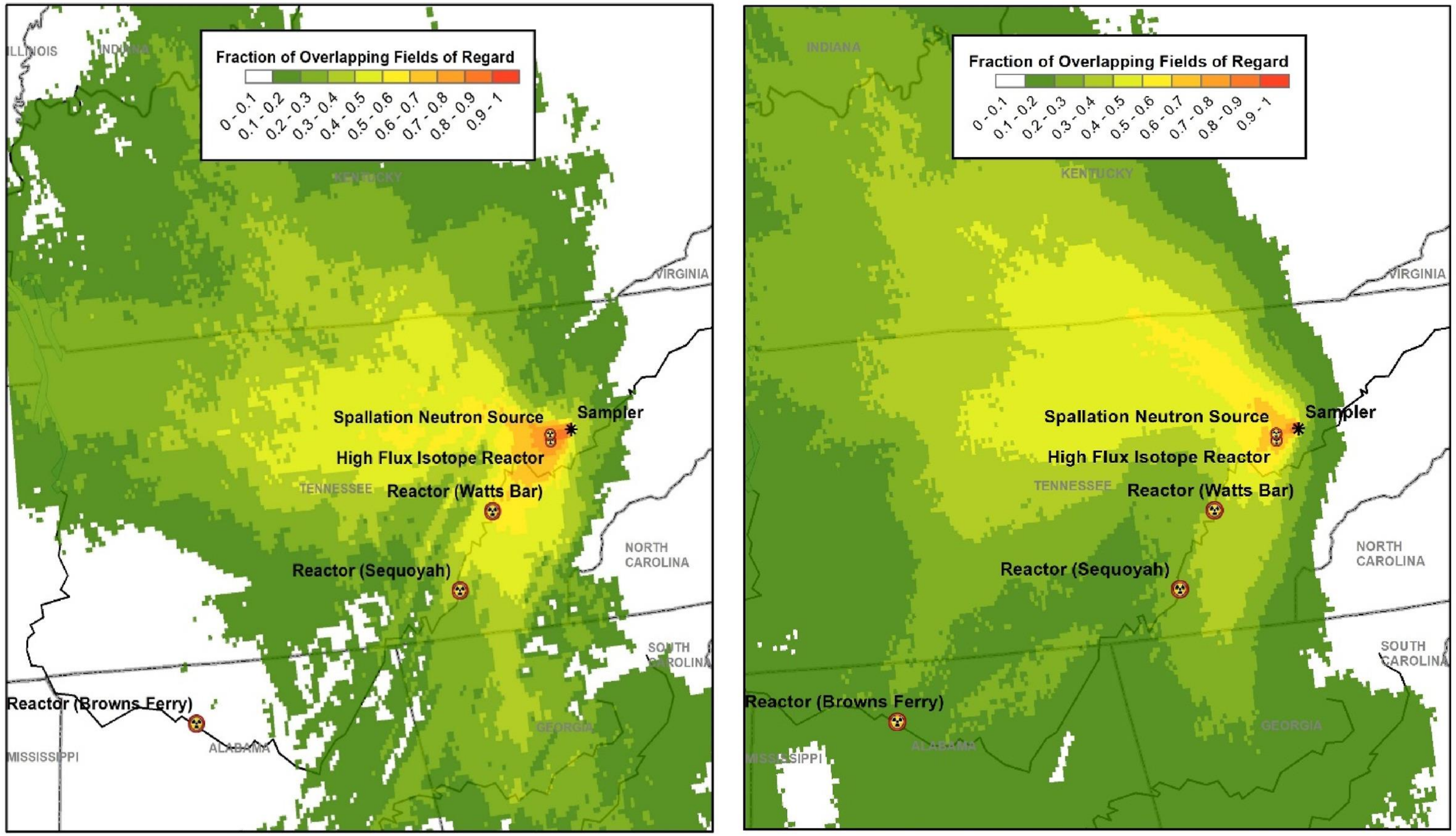
Radioxenon Data from 2020 not Associated with Traditional Radioxenon Isotopes



Eslinger, Paul W. et al. 2022. “Determining the Source of Unusual Xenon Isotopes in Samples.” *Journal of Environmental Radioactivity* 247 (June): 106853. https://doi.org/10.1016/J.JENVRAD.2022.106853.

Atmospheric Transport Modeling (ATM)

- ATM suggests that likely release is from HFIR or SNS
- Wind patterns are not consistent with release from the nuclear power plants
- Production mechanism suggests
  - SNS only
  - SNS and HFIR
- Publication on ATM



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How is the Non-Traditional Radioxenon Produced?

- Proton bombardment of a liquid mercury target producing neutrons
  - Trace amounts of uranium within the target
  - Irradiation of natural xenon from air
- Proton bombardment producing ^{122}Xe

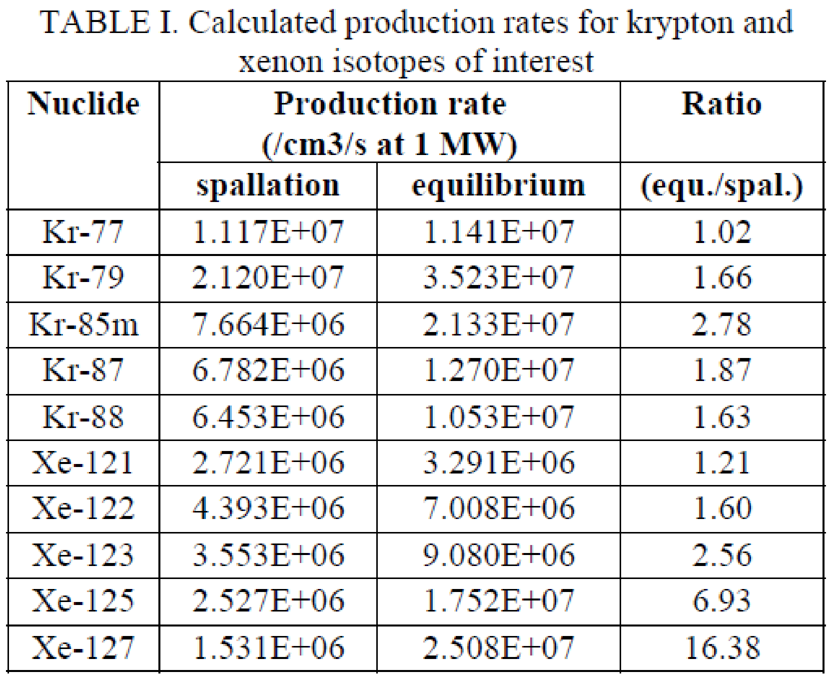
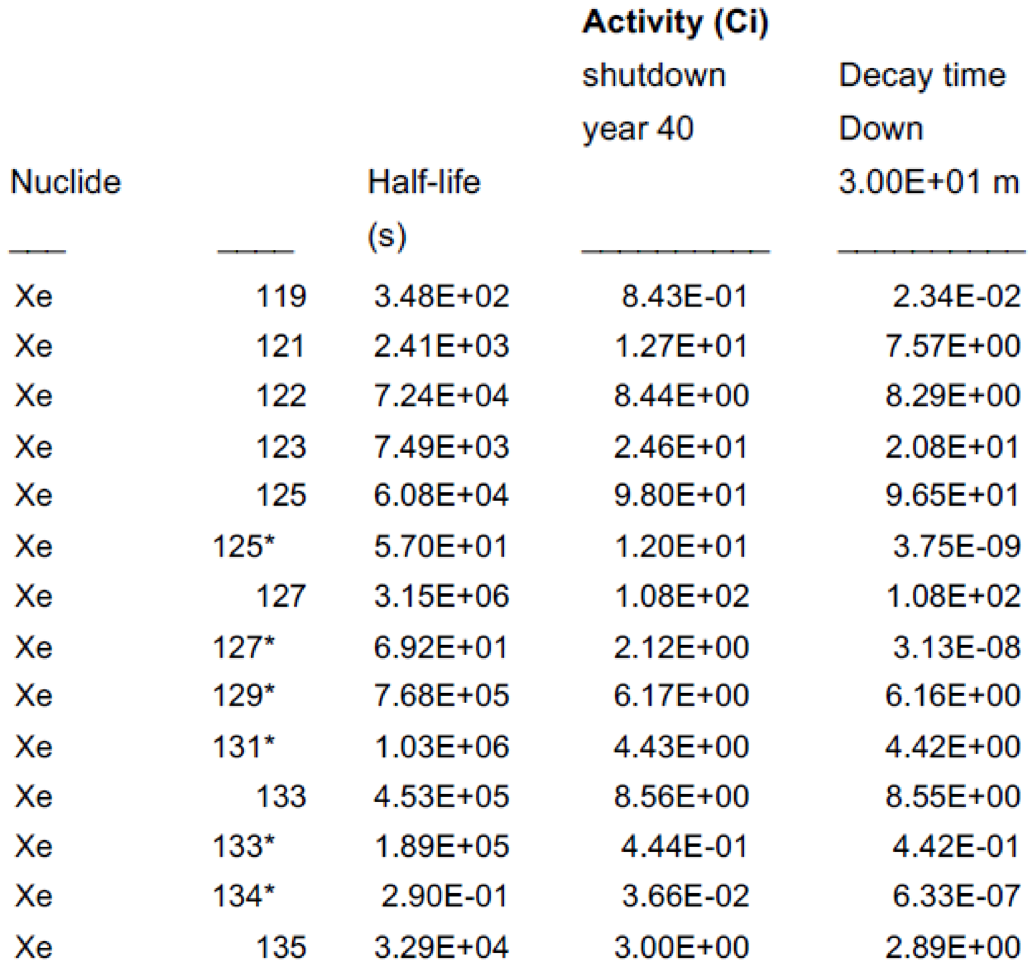


TABLE I. Calculated production rates for krypton and xenon isotopes of interest

Nuclide	Production rate (/cm3/s at 1 MW)		Ratio
	spallation	equilibrium	(equ./spal.)
Kr-77	1.117E+07	1.141E+07	1.02
Kr-79	2.120E+07	3.523E+07	1.66
Kr-85m	7.664E+06	2.133E+07	2.78
Kr-87	6.782E+06	1.270E+07	1.87
Kr-88	6.453E+06	1.053E+07	1.63
Xe-121	2.721E+06	3.291E+06	1.21
Xe-122	4.393E+06	7.008E+06	1.60
Xe-123	3.553E+06	9.080E+06	2.56
Xe-125	2.527E+06	1.752E+07	6.93
Xe-127	1.531E+06	2.508E+07	16.38



Nuclide		Half-life (s)	Activity (Ci) shutdown year 40	Decay time Down 3.00E+01 m
Xe	119	3.48E+02	8.43E-01	2.34E-02
Xe	121	2.41E+03	1.27E+01	7.57E+00
Xe	122	7.24E+04	8.44E+00	8.29E+00
Xe	123	7.49E+03	2.46E+01	2.08E+01
Xe	125	6.08E+04	9.80E+01	9.65E+01
Xe	125*	5.70E+01	1.20E+01	3.75E-09
Xe	127	3.15E+06	1.08E+02	1.08E+02
Xe	127*	6.92E+01	2.12E+00	3.13E-08
Xe	129*	7.68E+05	6.17E+00	6.16E+00
Xe	131*	1.03E+06	4.43E+00	4.42E+00
Xe	133	4.53E+05	8.56E+00	8.55E+00
Xe	133*	1.89E+05	4.44E-01	4.42E-01
Xe	134*	2.90E-01	3.66E-02	6.33E-07
Xe	135	3.29E+04	3.00E+00	2.89E+00

DeVore, Joe R, Lu, Wei, and Schwahn, Scott O. 2013. "NOBLE GAS PRODUCTION FROM MERCURY SPALLATION AT SNS". United States

How is the radioxenon abated?

- How much gets out of the target?
  - Target material: liquid mercury
  - Other sources use different target materials

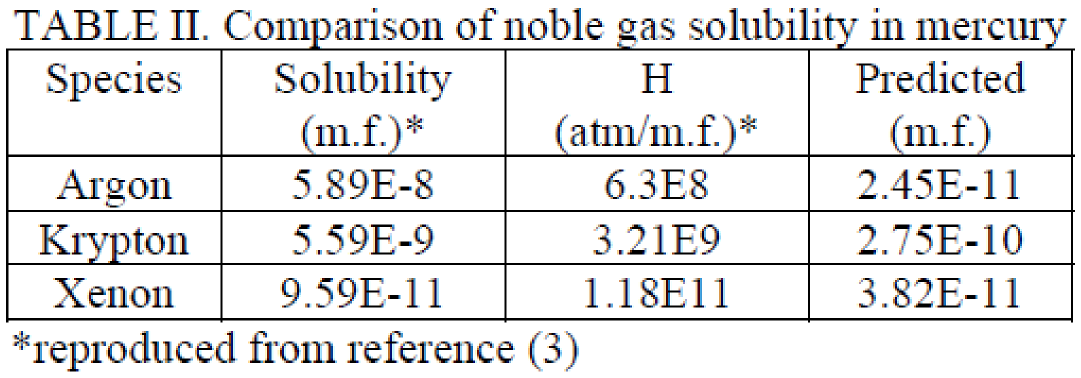


TABLE II. Comparison of noble gas solubility in mercury

Species	Solubility (m.f.)*	H (atm/m.f.)*	Predicted (m.f.)
Argon	5.89E-8	6.3E8	2.45E-11
Krypton	5.59E-9	3.21E9	2.75E-10
Xenon	9.59E-11	1.18E11	3.82E-11

*reproduced from reference (3)

- What traps do the gases go through before being released?
  - Helium off gases and purge for studying the production rates
  - HEPA filters for particulate
  - Cryogenic sulfur-impregnated charcoal absorber

- How long is the xenon contained before being released?
  - Initial retention time of approximately 350 minutes (1/3 ^{122}Xe half-life)

DeVore, Joe R, Lu, Wei, and Schwahn, Scott O. 2013. "NOBLE GAS PRODUCTION FROM MERCURY SPALLATION AT SNS". United States

Other possible sources around the world

- Neutron Activation Sources
  - Accelerators
  - Reactor cover gas
    - ✓ Cover gas hold-up times impact the short-lived isotope interferences
- Spallation sources
  - SNS (this presentation)
  - Los Alamos Neutron Science Center
  - ISIS neutron source in the UK
  - Japan Proton Accelerator Research Complex (J-PARC) in Japan
  - European Spallation Source in Sweden
  - China Spallation Neutron Source
- Are there other commercial sources of these isotopes?

Conclusion and Outlook

- Newly observed isotopes seen near HFIR and SNS suggest new source of interfering xenon background (non-traditional production mechanisms)
- New isotopes interfere with all traditional ROIs used for activity calculations
  - Algorithms will continue to calculate normally
  - Concentrations and ratios will not make sense
- Non-traditional xenon isotopes are produced through a series of mechanisms
  - Spallation on mercury targets
  - Air activation
  - Fission isotopes
- Even with wind predominantly in the other direction, they can still be detected at near-by stations
- These non-traditional isotopes may be produced at other facilities around the world

Thank you