



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

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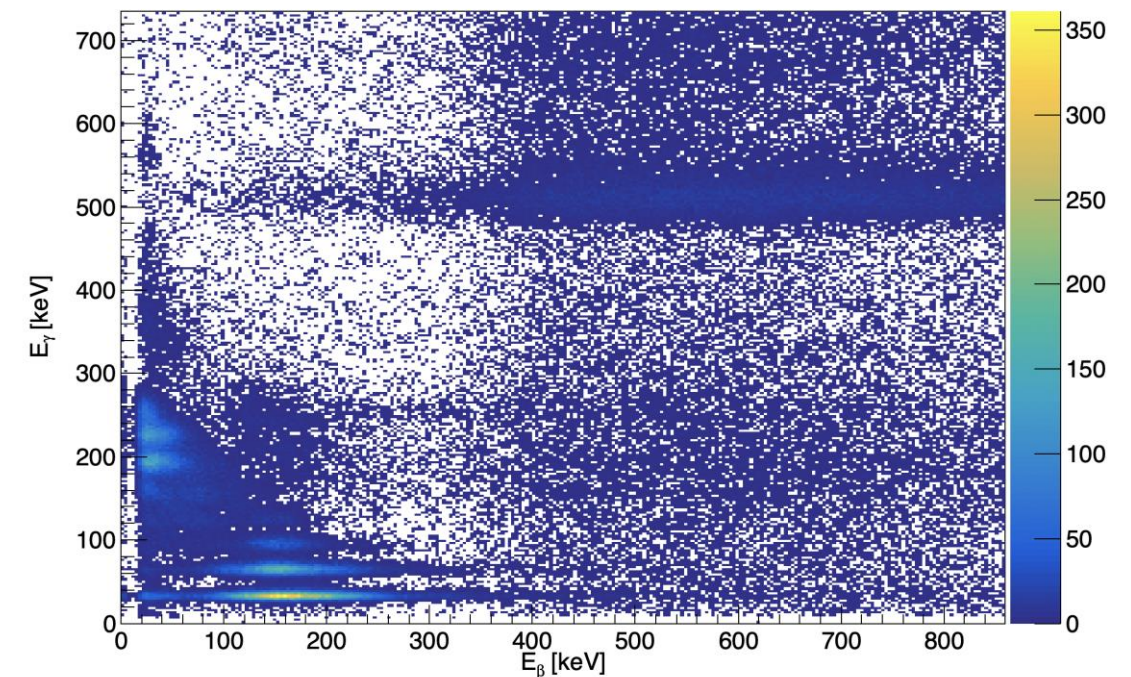


PNNL is operated by Battelle for the U.S. Department of Energy

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	4.52E+05	9.52E+00	9.55E+00
Xe	133*			
Xe	134*			
Xe	135			



Non-traditional Xenon Isotopes

- Many isotopes can be produced via neutron irradiation
 - ^{127}Xe
 - ^{125}Xe
 - $^{129\text{m}}\text{Xe}$
- 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 $(n,2n)$ 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.

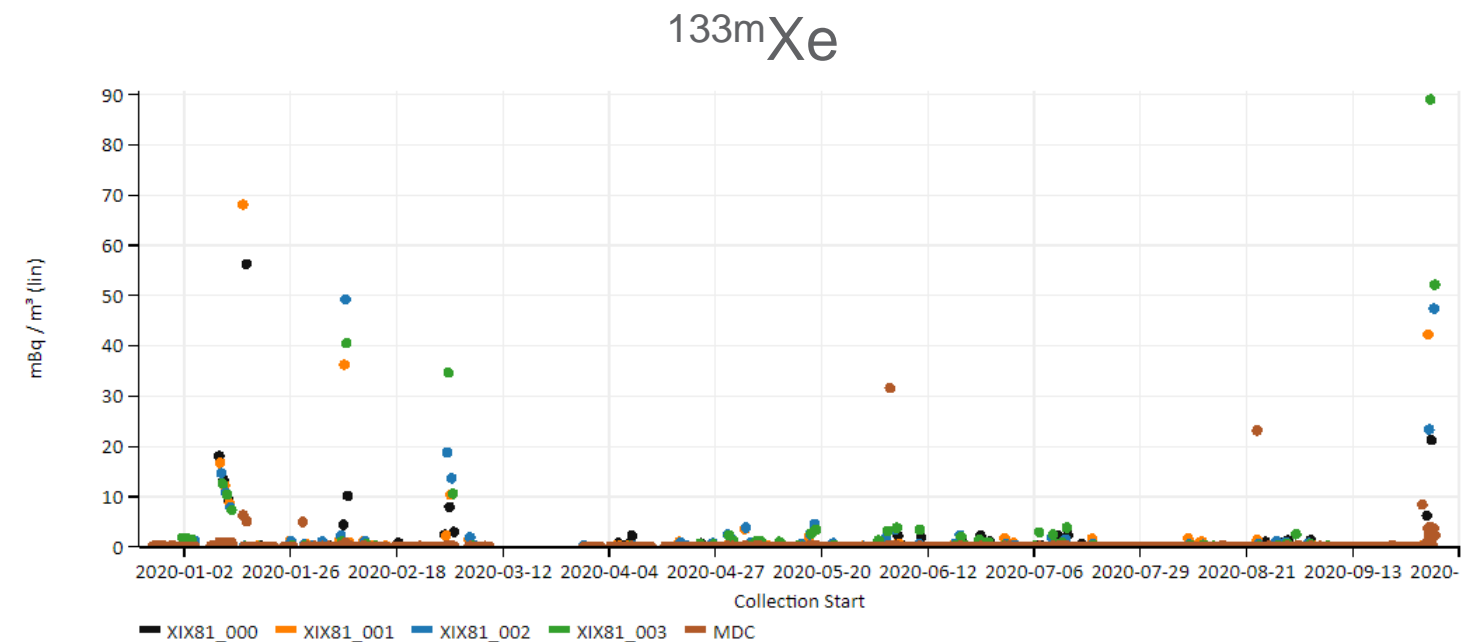
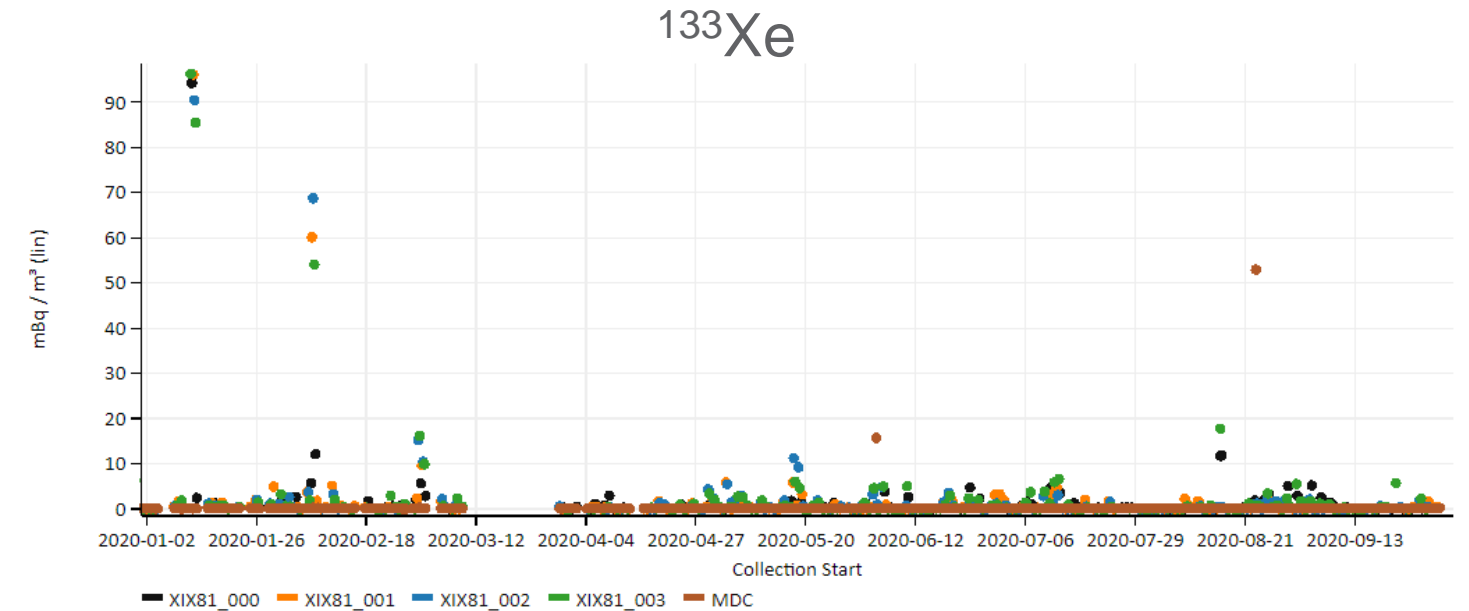
Xenon International

- Next generation atmospheric radioxenon system
- Faster and more sensitive than current certified 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 , $^{131\text{m}}\text{Xe}$, $^{133\text{m}}\text{Xe}$
 - <0.5 mBq/SCM for ^{135}Xe
- Developed at PNNL
 - Transitioned to Teledyne Brown Engineering for production (Knoxville, TN)



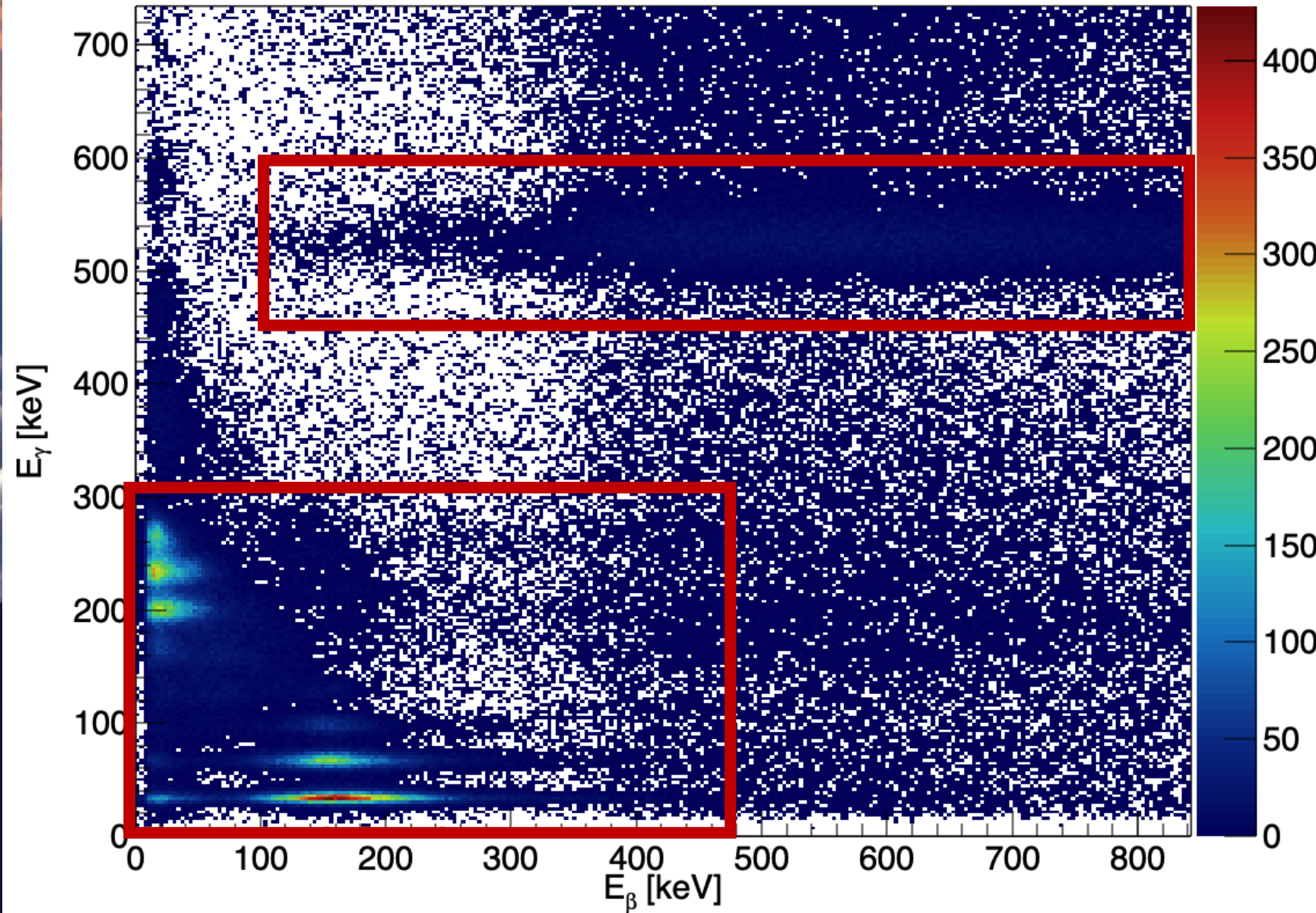
Puzzling Observations and Analyses

- 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
 - $^{133\text{m}}\text{Xe}$ – unusual to have higher concentration than ^{133}Xe
 - $^{131\text{m}}\text{Xe}$ – similar concentration to $^{133\text{m}}\text{Xe}$
- Further investigation showed that the gamma-ray spectrum has unusual features.

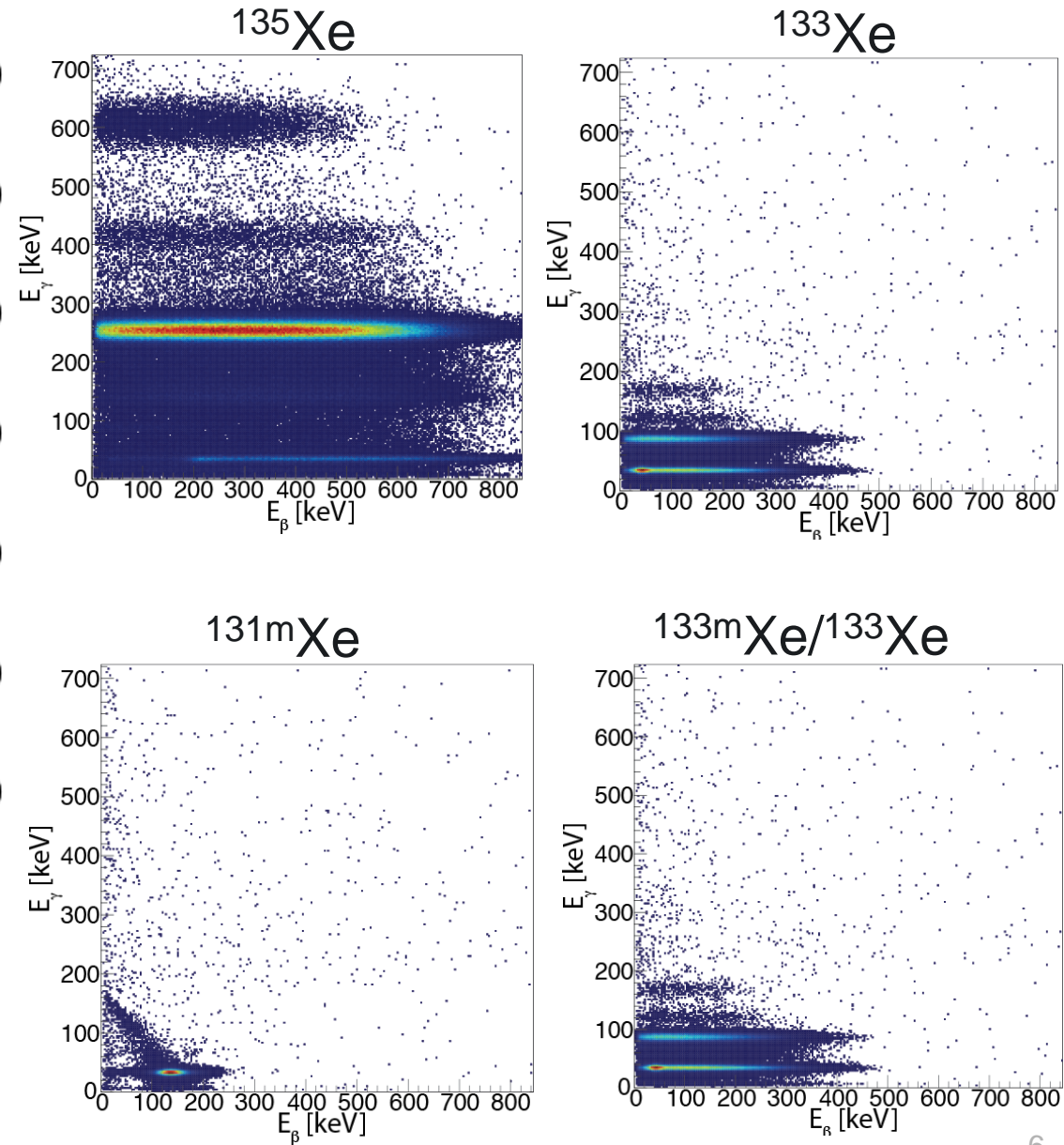


Atypical Coincidence Signatures

Spectrum observed on Xenon International



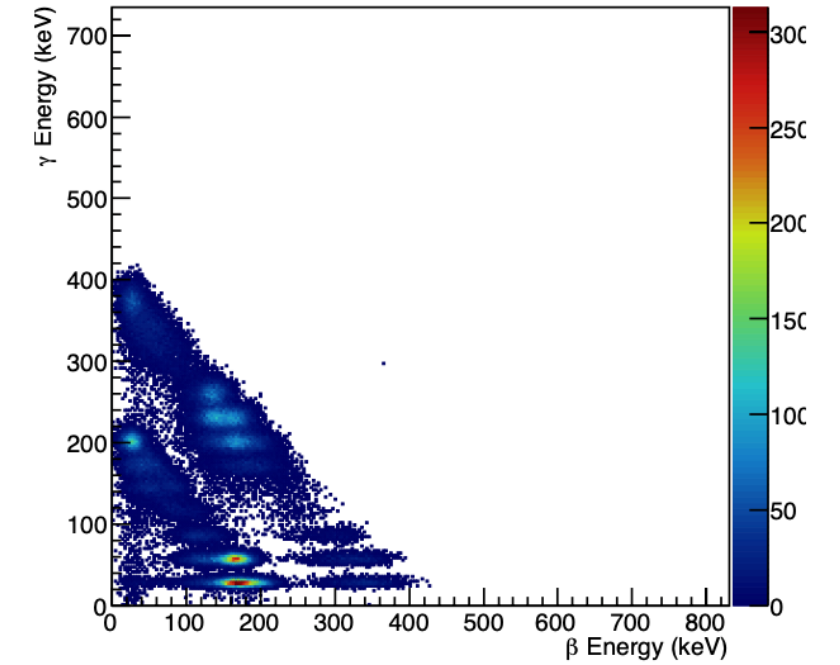
Typical Xenon Signatures



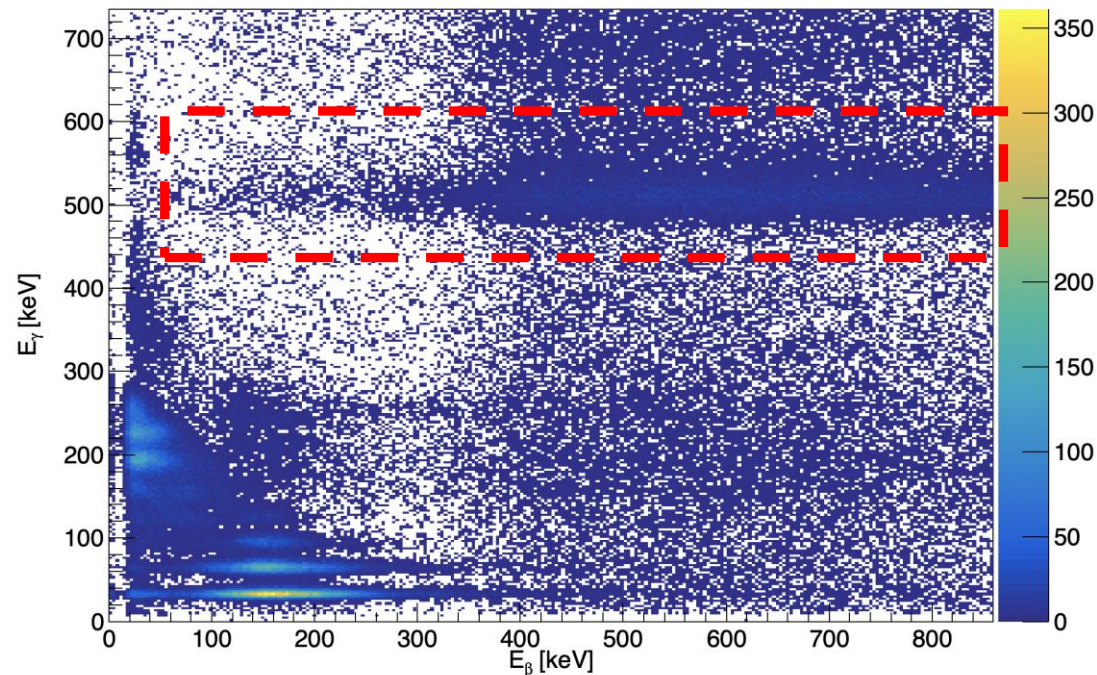
Monte Carlo Simulations

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

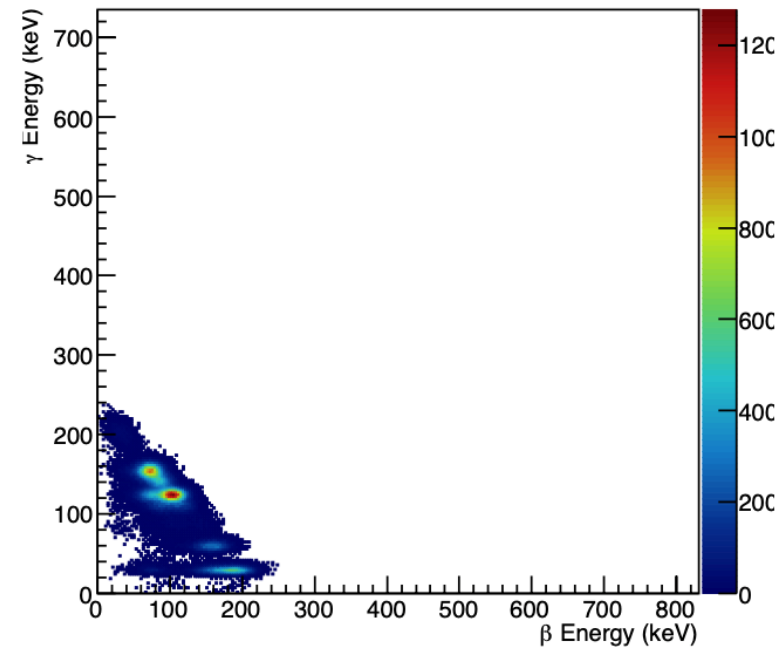
^{127}Xe Simulation



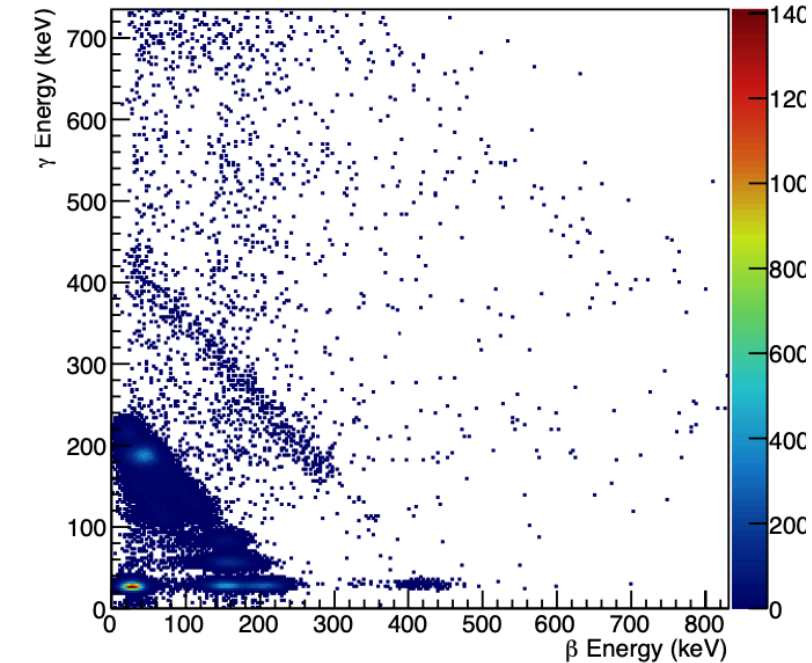
Xenon International Measurement



$^{129\text{m}}\text{Xe}$ Simulation

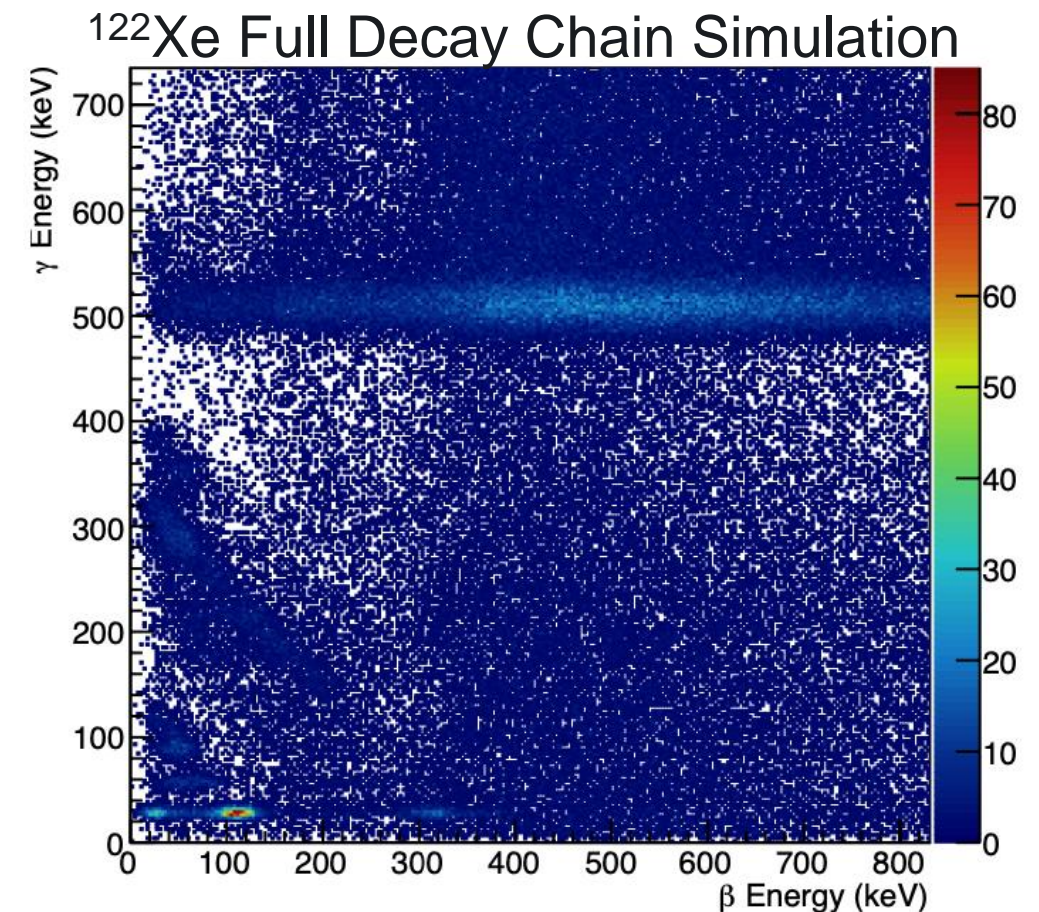
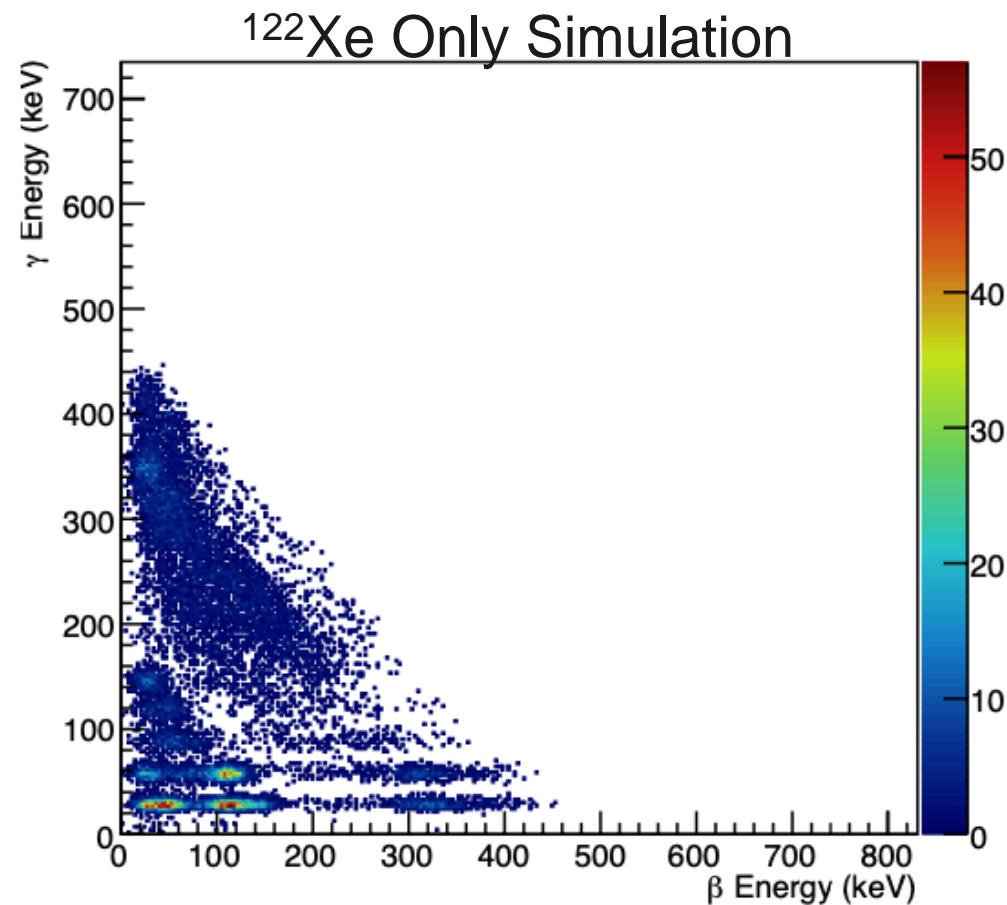


^{125}Xe Simulation

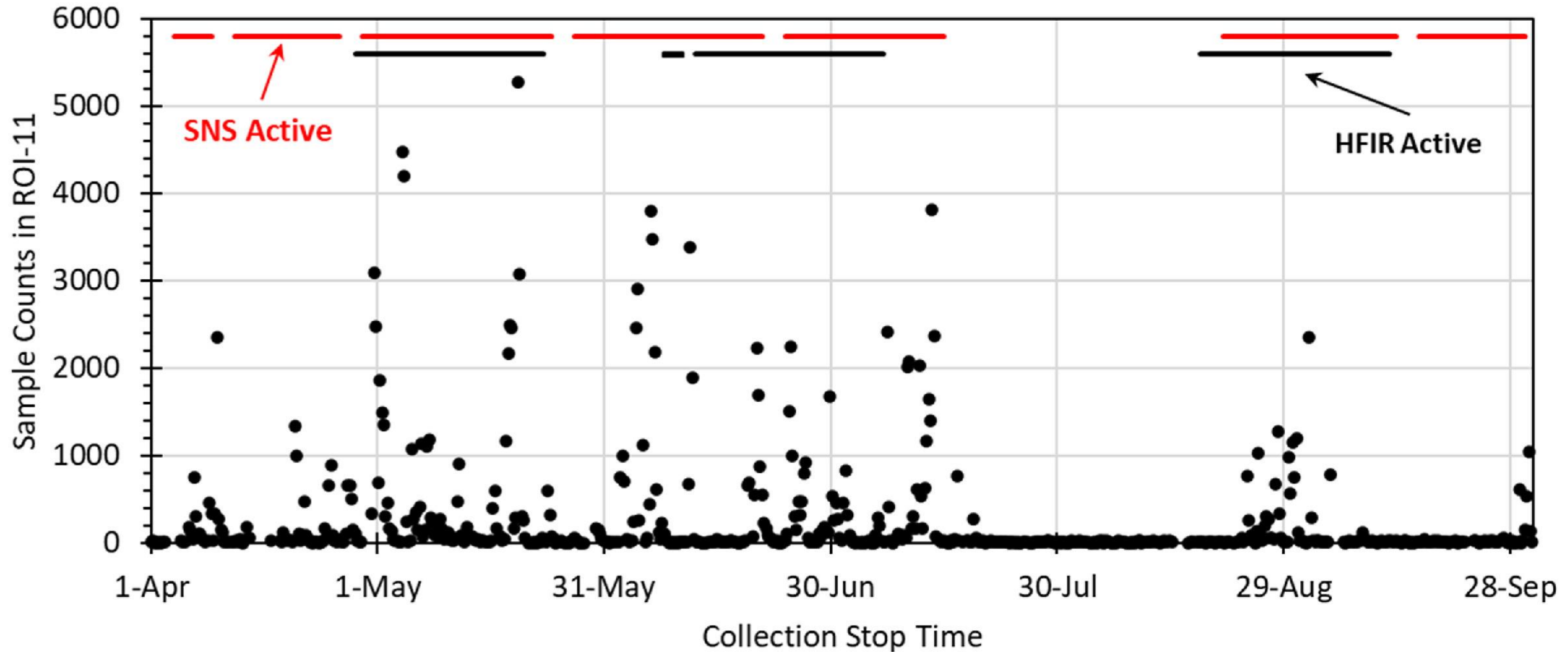


Missing Piece

- ^{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)



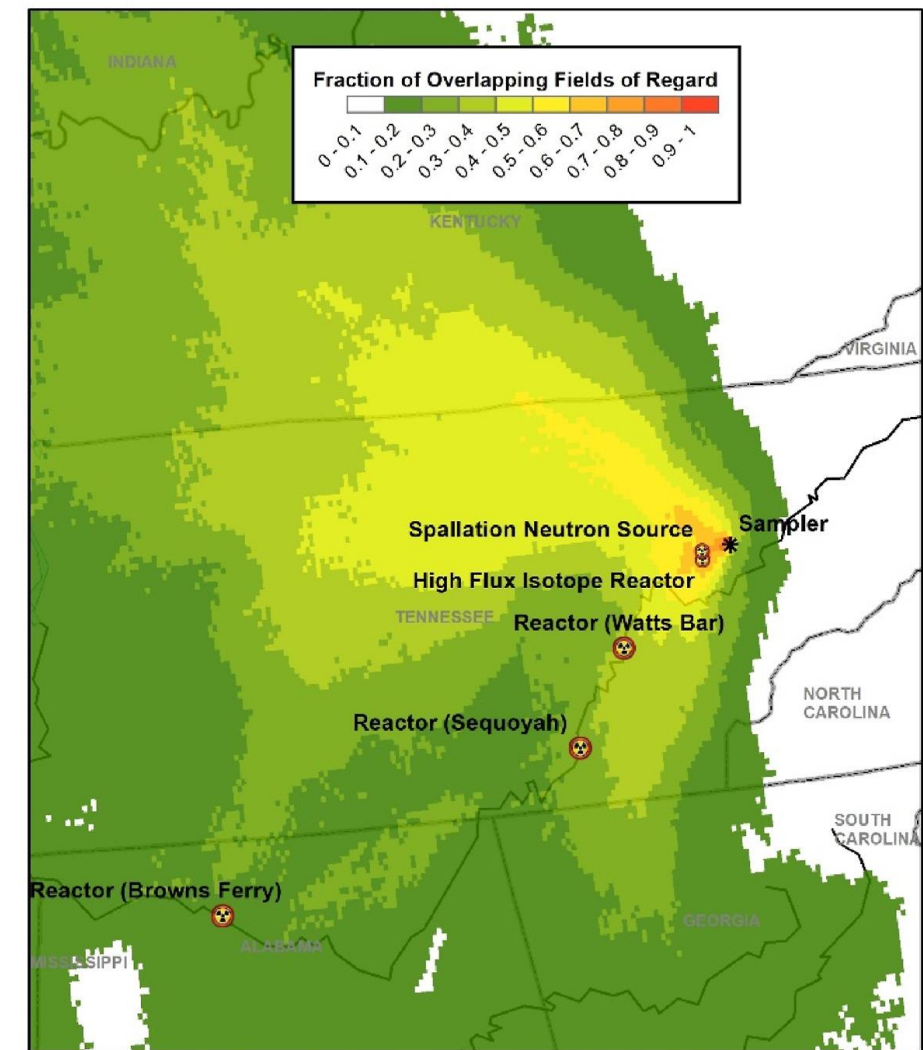
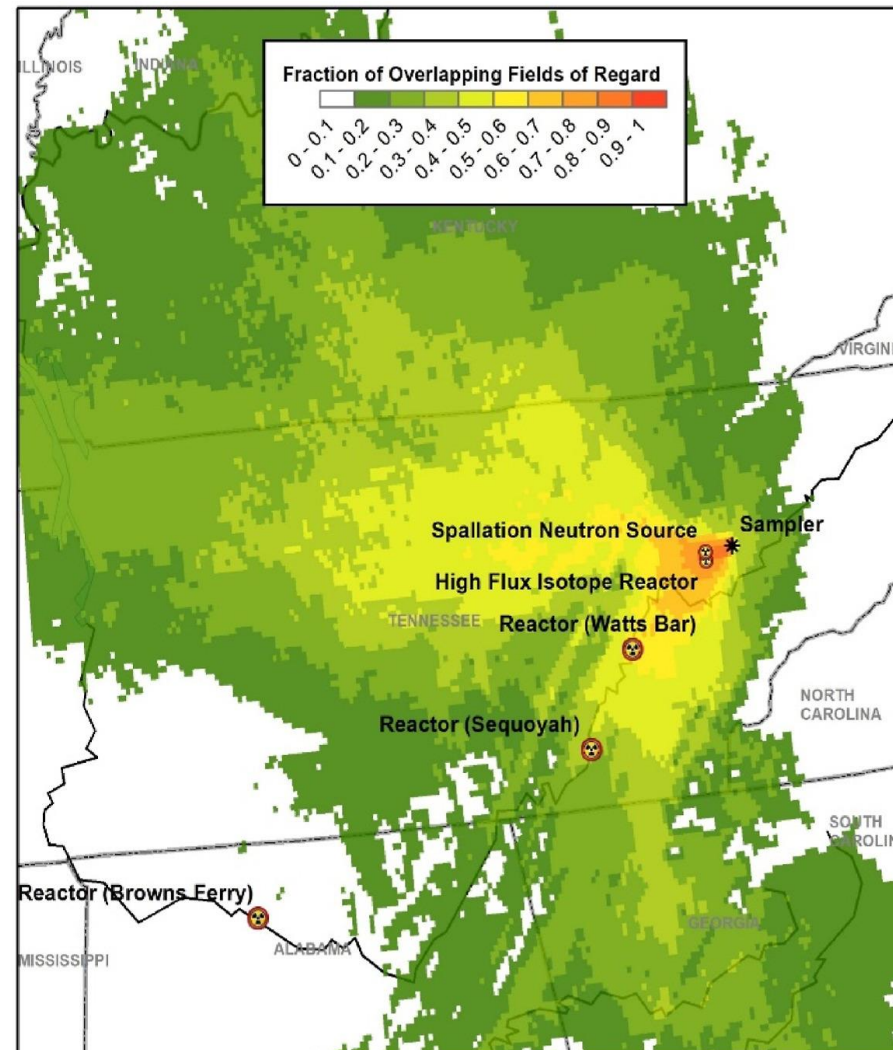
Correlation of ^{122}Xe with SNS and HFIR Operation



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



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>.

How is the Non-Traditional Radioxenon Produced?

- Neutron bombardment of a liquid mercury target
 - 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 (/cm ³ /s at 1 MW)		Ratio (equ./spal.)
	spallation	equilibrium	
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

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)

Other possible sources around the world

- Neutron Activation Sources
 - Accelerators
 - Reactor cover gas
 - ✓ Cover gas hold-up times impact the short-lives 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



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Thank you