An Analysis of Potential Sources of Non-fission Product Xenon Radionuclides (^{127}Xe, ^{129m}Xe, ^{125}Xe, ^{122}Xe)

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Quality Control Standards at Idaho National Laboratory

- INL Noble Gas Laboratory (NGL) produces Quality Control standards for laboratories that operate xenon radionuclide monitoring systems.
- Current standards provided to these labs consist of fission product xenon radionuclides (^{131m}Xe, ^{133}Xe, ^{133m}Xe, and ^{135}Xe)
- INL NGL's mission is to provide high quality standards that ensure high accuracy results being reported
  - INL NGL research can demonstrate potential interferences to quantification of fission product xenon radionuclides
  - Through research, INL NGL may demonstrate the need for calibration with other xenon radionuclides not currently in use
    - Potential needs for calibration standards containing non-fission products such as ^{127}Xe, or ^{129m}Xe may exist

Background: Detection of Non-fission Product Radioxenons

- Testing of a new continuous noble gas monitoring system (Xenon International, Teledyne Brown Engineering) showed some unusual radionuclide detections[1]
  - System was operated in proximity (~20 km) of Oak Ridge National Laboratory
    - Operators noted that a large number of samples showed all 4 radionuclides of interest[1] and more ^{133m}Xe than ^{133}Xe
    - Samples showing all 4 radionuclides are relatively uncommon when analyzing the atmosphere
    - ^{133m}Xe is almost never more abundant than ^{133}Xe

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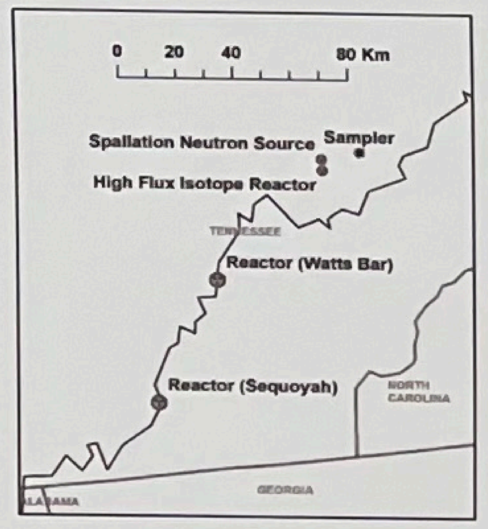
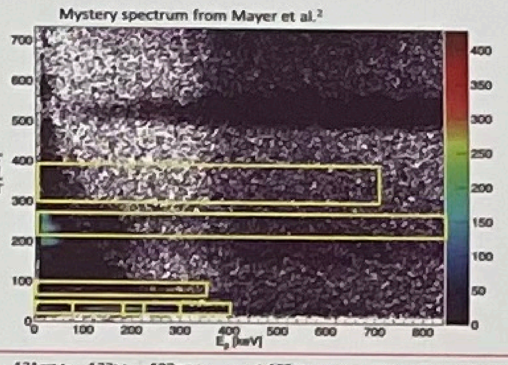


Unusual Spectra Observed

- Testing of a new continuous noble gas monitoring A spectra was published[2] of the unusual count pattern observed
- Although the automatic quantification algorithm showed hits for all 4 radionuclides of interest, it appears that no ^{135}Xe or ^{133}Xe is present

Mystery spectrum from Mayer et al.[2]



ROIs for ^{131m}Xe, ^{133}Xe, ^{133m}Xe, and ^{135}Xe are approximated in this plot for comparison. The pattern formed by the observed beta-gamma spectrum do not fit inside the defined ROIs.

Theorized Radionuclides Responsible for Unusual β/γ Coincidence Spectra

- Various xenon radionuclides are theorized to contribute to the spectrum recorded during testing of Xenon International
- ^{122}Xe, ^{125}Xe, ^{127}Xe , and ^{129m}Xe, are potentially radionuclides that could create this type of spectrum
- The spectrum of ^{127}Xe is shown on the right[3] and has counts in the spectra that overlaps with some of the 7 ROIs for determining the 4 radionuclides of interest

^{127}Xe counts overlap the 7-ROIs used for detection of fission product xenon radionuclides. Figure is from Klingberg et al.[3] (2015)

Theorized Radionuclides Responsible for Unusual β/γ Coincidence Spectra

- Coincidence counts observed during assay of ^{127}Xe by beta-gamma coincidence counting shows overlap with ^{135}Xe, ^{133}Xe, ^{133m}Xe and ^{131m}Xe ROIs
- Because ^{127}Xe was identified in an environmental sample, it is important to understand the potential for sources of ^{127}Xe
- Operators of systems benefit from understanding the potential of having ^{127}Xe present in samples
- Understanding types of facilities that have the potential to emit ^{127}Xe will put operators in mind to watch for ^{127}Xe interference if the potential exists

Potential Source#1: Spallation Neutron Source (SNS)

- Power Source = 1.4 MW pulsed (60 hz) beam (proton) accelerator
- Target Material = Liquid Mercury
- Neutron production Mechanism: High energy protons collide with mercury target atoms to knock out neutrons
- 19 instruments for neutron scattering experiments
- Mercury off-gas treatment system removes radioxenon by-products
  - In operation since 2006

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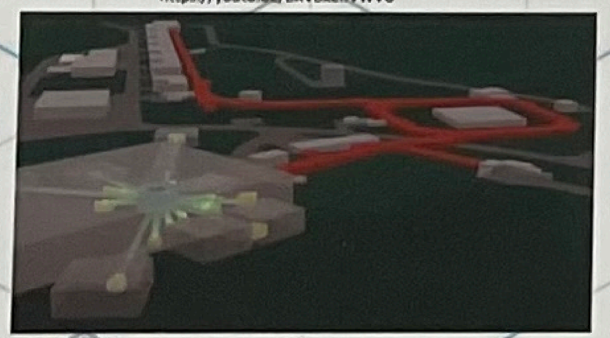
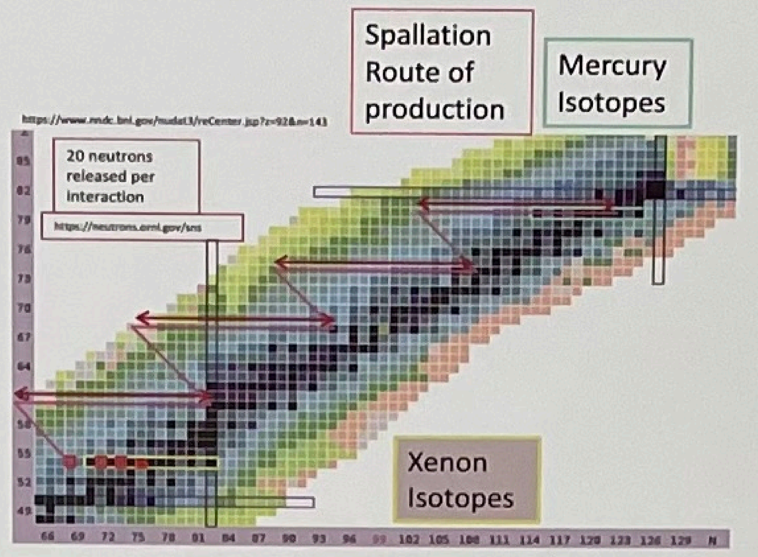


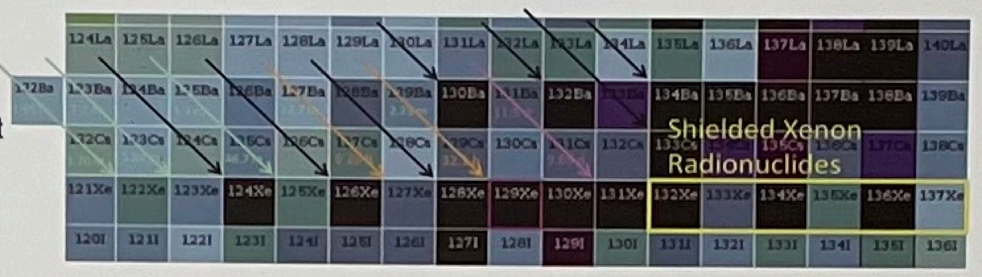
Production Routes for Non-fission production Radionuclides: SNS

- The path to create ^{122}Xe from the mercury target (shown in pink zig zag above) requires 4 spallation interactions to reduce mass of mercury to xenon
  - ^{202}Hg (Rel. Abund. = 29.86%) – 4 interactions x (20 neutrons / interaction) = ^{122}Xe



Radionuclides Shielded from Production Route at SNS

- Spallation Sources produce radionuclides through decay cascades from above
- All fission product xenon radionuclides are shielded except ^{131m}Xe
- ^{131m}Xe formation is impeded by relatively long-lived ^{131}Ba ($T_{1/2}$ = 11.5 d) and ^{131}Cs ($T_{1/2}$ = 9.69 d)



Potential source#2: High Flux Isotope Reactor (HFIR)

- HFIR: "Beryllium-reflected, light-water cooled and –moderated, flux-trap type reactor" (https://neutrons.ornl.gov/hfir/parameters)
- Isotope Production (^{252}Cf, others)[4]
- ^{252}Cf Starting Materials = ^{242}Pu, ^{243}Am, ^{244}Cm
- Neutron Scattering experiments
- [Too many types to list here]

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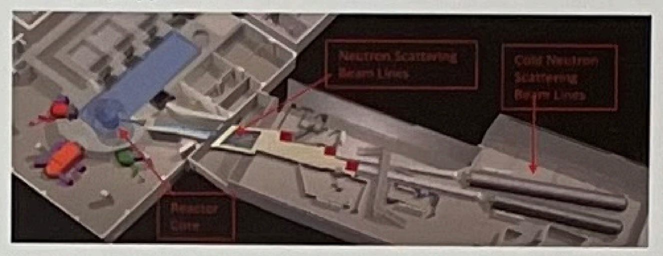
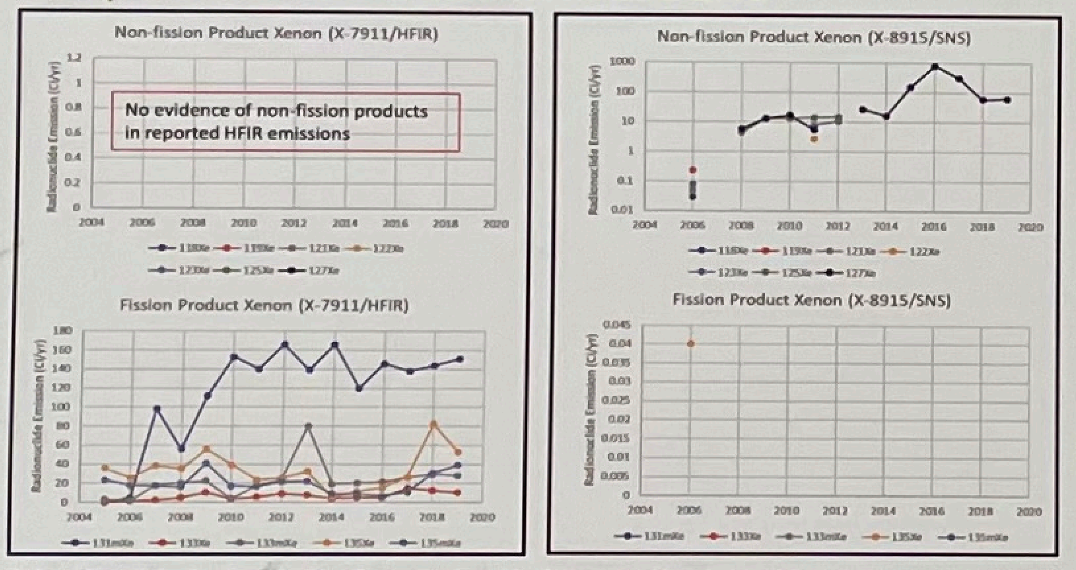


Reported Emissions from HFIR and SNS

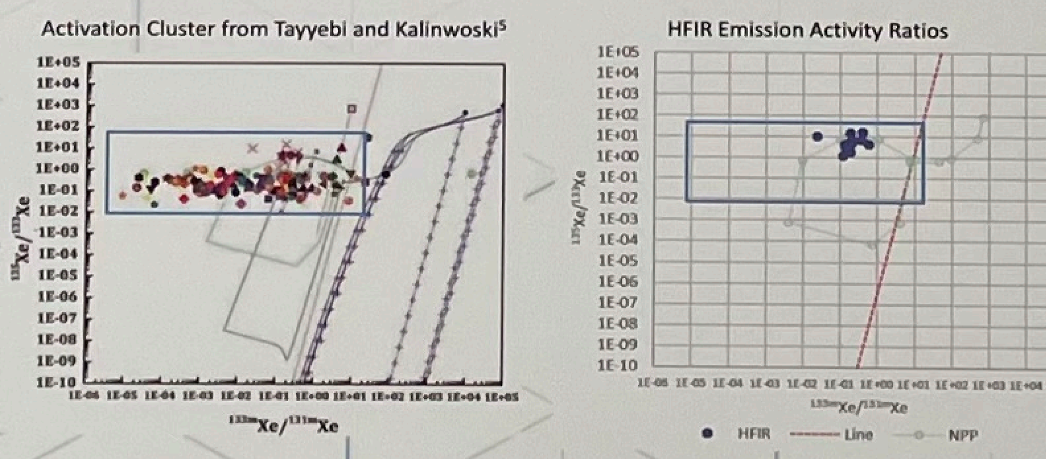


Radionuclide emission data from: Oak Ridge Reservation Annual Site Environmental Reports: years 2005 to 2019

- HFIR emissions are purely fission/activation product radionuclides
- No observed non-fission products reported
- SNS shows a single report of a fission/activation product, but the reported emissions are dominated by non-fission products
- ^{127}Xe emissions are easily detectable in reported SNS emissions

HFIR emission Activity Ratios align with Activation Cluster

- Isotope Ratio region identified as Activation Cluster by Tayyebi and Kalinowski[5]
- HFIR reported emissions lie within Activation Cluster
- Also lies within the aged fission gas from nuclear power plant region



Analysis of Another High Flux Reactor: INL Advanced Test Reactor

- Four stack emission grab samples (5 min assay) from 8/10/2023 were averaged to obtain observed daily emission activities
- ^{135}Xe to ^{41}Ar ratio = 0.010 ± 0.002

Radionuclide	Classification	Ci/day
^{41}Ar	Pure Activation	10.0 ± 0.4
^{133}Xe	Activation or Fission	0.08 ± 0.04
^{135}Xe	Activation or Fission	0.10 ± 0.02
^{135m}Xe	Activation or Fission	0.10 ± 0.02
^{138}Xe	Activation or Fission	0.26 ± 0.04
^{88}Kr	Activation or Fission	0.04 ± 0.01
^{87}Kr	Activation or Fission	0.04 ± 0.01

Recreation of ^{135}Xe:^{41}Ar ratio by Air Activation Modeling (ATR Neutron Flux)

- Using measured neutron fluxes from ATR, the activation of air was modeled
  - ^{135}Xe to ^{41}Ar ratio (Modeled, no decay) = 1.30E-07
  - ^{135}Xe to ^{41}Ar ratio (Stack Emission, Observed) = 0.010 ± 0.002
- The ratios can be reconciled by assuming decay (residence time in ATR building before exiting the stack)
  - ^{135}Xe to ^{41}Ar ratio (Modeled, 36.7 ± 1.5 hr decay) = 0.010
  - Other Activation Product emitted in this scenario →

Radionuclide	Classification	Stack Emission (No decay) Ci/day	Stack Emission (36.7 hr Decay) Ci/day
^{41}Ar	Pure Activation	10	10
^{125}Xe	Pure Activation	1.4E-05	5 ± 2
^{127}Xe	Pure Activation	4.6E-09	0.008 ± 0.003
^{131m}Xe	Activation or Fission	1.4e-07	0.14 ± 0.16
^{133}Xe	Activation or Fission	6.9e-07	0.6 ± 0.1
^{133m}Xe	Activation or Fission	1.6e-07	0.11 ± 0.01
^{135}Xe	Activation or Fission	1.3e-06	0.09 ± 0.02

If stack emission ^{135}Xe is from purely activation of air, the quantity of ^{125}Xe would be easy to detect, but it is not observed/ reported in stack emissions assay.

^{125}Xe as an Indicator of Air-Activation non-Fission Products

- Modeling of air activation products (this work) shows that ^{125}Xe is the largest radioxenon activity when air is activated, next followed by fission product radioxenon (^{131m}Xe, ^{133}Xe, ^{133m}Xe, and ^{135}Xe)
- ^{127}Xe is produced 3 orders of magnitude lower activity than ^{125}Xe (see plots)
  - ^{125}Xe should be relatively easy to detect in stack emission gamma quantification systems
  - If ^{125}Xe is found to be present, ^{127}Xe will likely not be detectable, but is likely present at 3 orders of magnitude lower activity

ATR emissions dominated by Fission Products

- The existence of extremely short-lived (<1 hr) radionuclides in the stack emissions indicates that 36.7 hour decay time for activation products is not feasible
- Significant fraction of fission ^{135}Xe explains the ^{135}Xe:^{41}Ar ratio better than activation and decay

^{127}Xe emission is negligible compared to fission product radioxenon emitted from high flux reactors (both ATR and HFIR)

Radionuclide	Half-life	Classification	Ci/day
^{41}Ar	109.61 min	Activation	10.0 ± 0.4
^{133}Xe	5.2475 day	Activation or Fission	0.08 ± 0.04
^{135}Xe	9.14 hr	Activation or Fission	0.10 ± 0.02
^{135m}Xe	15.29 min	Activation or Fission	0.10 ± 0.02
^{138}Xe	14.14 min	Activation or Fission	0.26 ± 0.04
^{87}Kr	76.3 min	Activation or Fission	0.04 ± 0.01
^{88}Kr	2.825 hr	Activation or Fission	0.04 ± 0.01

Analysis of Neutron Spallation Sources World-wide

- ORNL SNS is in Knoxville, TN, USA
- Japan operates a similar spallation source to the ORNL SNS (liquid mercury target)
- Solid target (W, Pb) spallation sources are sealed and emit no significant spallation products

Country	Facility	Category	Incident Particle	Target
Canada	TRIUMF	Spallation	H-	W
China	CSNS	Spallation	H-	W
Japan	J-PARC	Spallation	H-	Hg (Liquid)
Russia	INR-RAS	Spallation	H+/H-	
Russia	JINR	Spallation		Pb
Sweden	ESS	Spallation	H-	W
Switzerland	CERN:nTOF Fa...	Spallation		Pb
Switzerland	SINQ	Spallation	P	Pb
UK	ISIS Facility	Spallation		W
USA	LANSCE	Spallation		W
USA	SNS	Spallation	H-	Hg (Liquid)
Korea[6]	KOMAC	Spallation	H+	W

Table Data from:
-https://nucleus.iaea.org/sites/accelerators/Pages/default.aspx
-Lee P et al.[6] (Korean SNS planned, not in operation currently)

Solid vs. Liquid Spallation Targets

- Liquid targets are continuously purged of neutron spallation products
  - Both Japanese and American treatment systems are similar and developed by shared experience[7]
  - Mercury off-gas treatment systems prioritize separation and emission of spallation products while preventing mercury emissions
- Solid targets (W, Pb, Ta) appear to be operated without any attempts to remove accumulated neutron spallation products
  - Because no attempts are made to remove contaminants while targets are in service, containments are designed with the goal of retaining all spallation products

Conclusions

- Liquid mercury target spallation neutron sources are the most likely source of detectable ^{127}Xe
  - Proton rich neutron spallation products are produced in the absence of fission product xenon radionuclides
  - Facilities using liquid mercury targets are constantly removing neutron spallation products to prolong the life of mercury target containers
- Solid target spallation neutron sources also produce proton rich neutron spallation products but are more likely to retain produced radionuclides
- Neutron activation of air in nuclear power plants and research reactors produces significantly more ^{125}Xe than ^{127}Xe
  - If gamma spectrum of stack-emissions doesn't show signs of ^{125}Xe, the emissions are likely dominated by fission gas, and won't be a strong source of ^{127}Xe

References

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