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Selecting the Right Noble Gas Tracer: Balancing Experiment Needs and Practical Challenges

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Pacific Northwest National Laboratory

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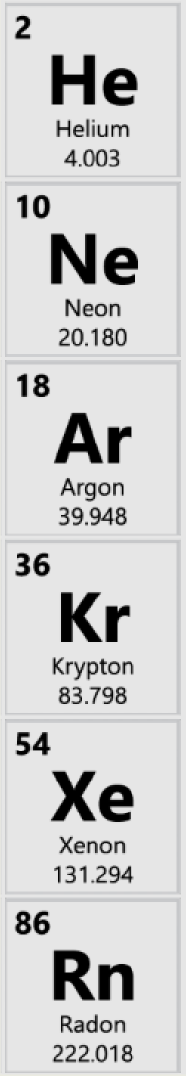
What makes a good noble gas tracer?

- Half-life considerations
  - Length of the sampling campaign
  - Operational constraints
- Relevance
  - Is this the species of interest or a surrogate?
- Measurement frequency
  - Is real-time, onsite, measurement needed?
  - Are periodic samples analyzed offsite sufficient?
- Acquisition
  - How hard is the tracer to produce?
  - Cost



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18
Ar
Argon
39.948

Ar-37 (Half-life 35.01 d)

- Ar-37 is an isotope of interest for nuclear explosion monitoring with work ongoing to develop and improve fieldable systems
- Potential for background challenges from ^{39}Ar at legacy test sites
- A 35-day half-life makes ^{37}Ar valuable for long-term (year+) studies of argon in the environment
- Least soluble of the tracers considered here
  (lowest impact from water in the testbed)

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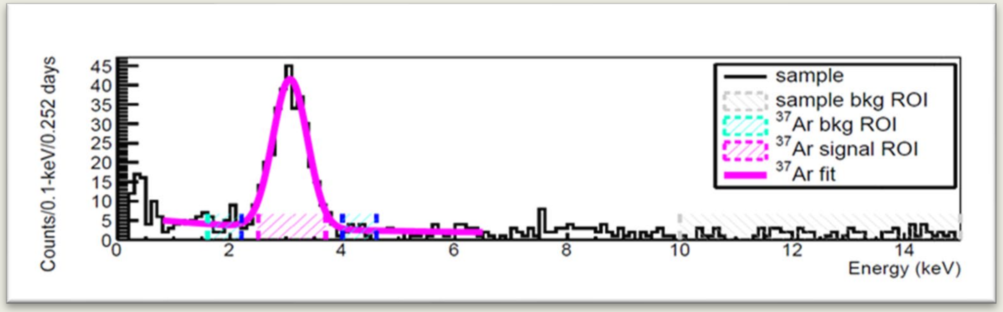
18 Ar Argon 39.948

Ar-37 (Half-life 35.01 d)

- Acquisition
  - Production of pure ^{37}Ar requires irradiation of enriched ^{36}Ar (0.334%)
    - Higher complexity, high cost
- Detection
  - Requires collection of discrete samples for analysis (no real-time capability)
  - Potentially challenging measurement outside of established laboratories



Ar-37 spectrum from a field collected sample.

C. Johnson, "*UNESE Phase 2: Gas migration studies in a tunnel test location*", SnT 2019.

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54
Xe
Xenon
131.294

Xenon-133 (Half-life 5.25 d)

- Xe-133 is an isotope of critical interest in nuclear explosion monitoring
  - Can leverage and exercise existing techniques for sampling and detection
- A 5-day half-life is workable for short (< 1 month) transport studies
  - Presents operational timing challenges
- Xenon sorption on geologic zeolites can be impactful in certain geologies

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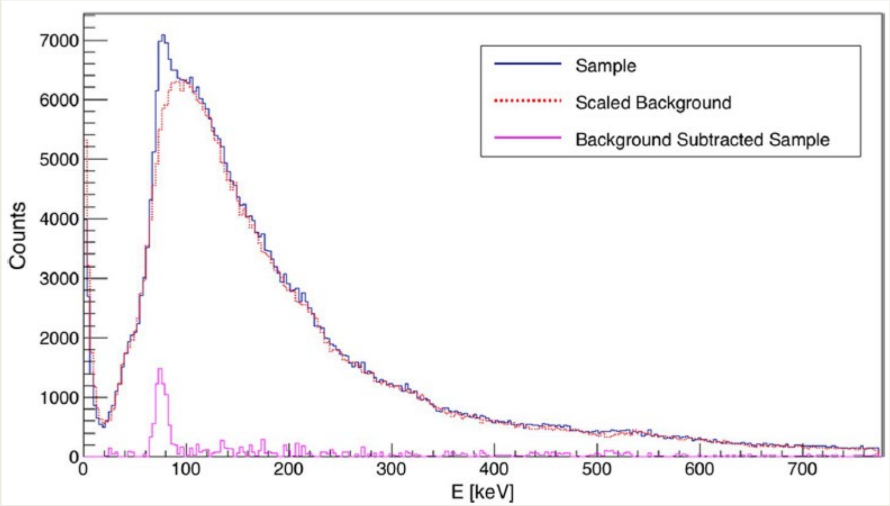
54 Xe Xenon 131.294

Xenon-133 (Half-life 5.25 d)

- Acquisition
  - Can be produced via irradiation of ^{132}Xe
  - Available from medical isotope suppliers
- Detection
  - Straightforward real-time detection
  - Counting of discrete samples can be conducted via γ-singles (on location) or via β-γ detection (offsite)



Field collected ^{133}Xe γ spectrum measured onsite.

C Johnson *et al.*, *"Injection and sampling of* ^{133}*Xe in shallow boreholes in alluvium,"* JRNC, 2022.

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Xe
Xenon
131.294

Xenon-127 (Half-life 36.34 d)

- Xenon-127 is not a current isotope of interest for nuclear explosion monitoring

However…

Other xenon isotopes are of critical interest

- A 36-day half-life makes ^{127}Xe uniquely valuable for long-term (year+) studies of xenon in the environment
  - The assumption becomes that all xenon isotopes behave similarly

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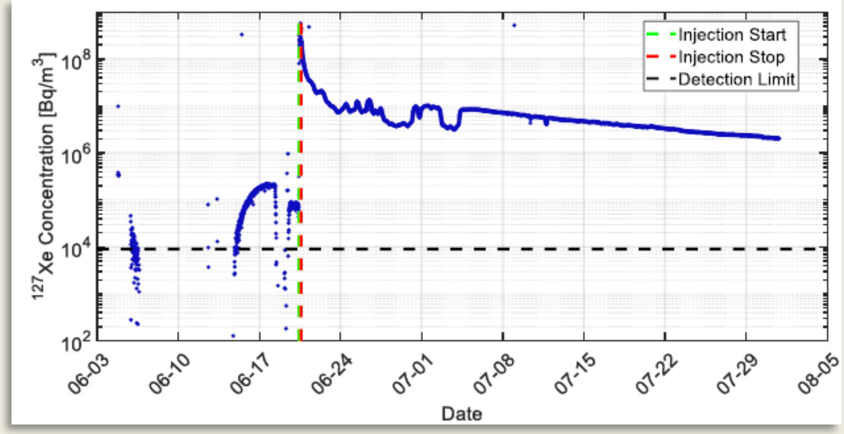
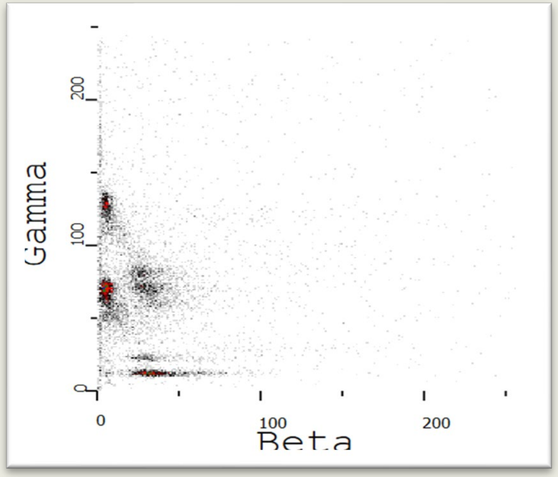
Xe
Xenon
131.294

Xenon-127 (Half-life 36.34 d)

- Acquisition
  - Production of pure ^{127}Xe requires irradiation of enriched ^{126}Xe (0.089%)
    - Higher complexity, very high cost
- Detection
  - Straightforward real-time detection
  - Multiple high intensity (>15%) gamma emission peaks (172 keV, 203 keV, 375 keV)
  - Options also exist for laboratory beta-gamma detection





Field collected real-time ^{127}Xe γ singles concentrations (top) and a ^{127}Xe β-γ spectrum (bottom).

C. Johnson, "*UNESE Phase 2: Injection and measurement of gaseous tracers at U-12p Tunnel*", 2020.

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86
Rn
Radon
222.018

Radon-222 (Half-life 3.82 d)

- Radon-222 is not an isotope of monitoring interest, but can be significant as a background in detector systems
- Potentially provides natural tracer for study of subsurface changes
  - Naturally produced by decay of ^{238}U
  - Can be present in very high subsurface concentrations (>100,000 Bq/m^3)
- Useful for understanding of sampling borehole quality
- Most soluble of the tracers considered here

  (highest impact from water in the testbed)

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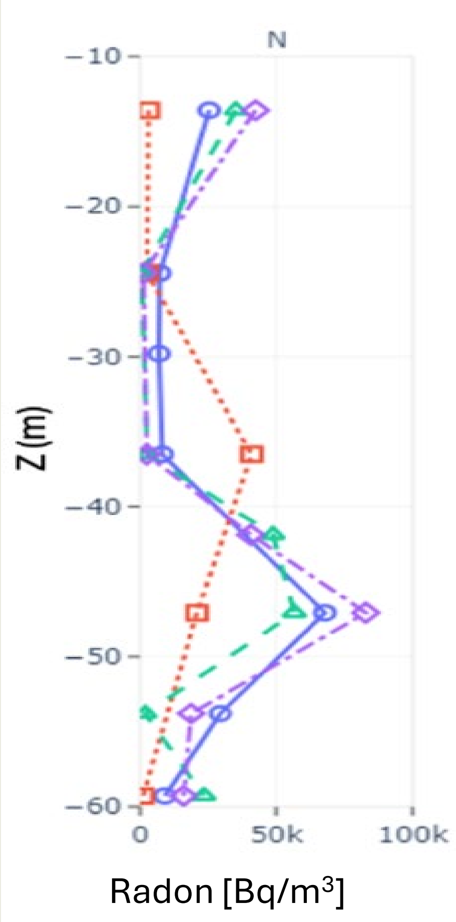
86
Rn
Radon
222.018

Radon-222 (Half-life 3.82 d)

- Acquisition
  - Straightforward production via radon generator
  - Can also be leveraged as a naturally present tracer in certain geologies
- Detection
  - Simple real-time alpha detection with commercial radon detectors



Radon concentration as a function of depth across 4 sampling campaigns.

C. Johnson *et al.*, "Field measurement of subsurface radon content from a series of underground chemical explosions," submitted to JRNC, 2025.

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What about stable gases?

- Stable gases can also be used as tracers
- Aim is to perturb natural abundance (isotopic or elemental)
- Acquisition
  - Can be directly purchased from gas companies
  - Required quantity highly dependent on tracer choice
    - Elemental perturbation requires significant tracer volume (100’s of L or more)
    - Rare isotopes require less tracer, but are costly and harder to acquire
- Detection
  - Potential for real-time detection via in-field mass spectrometry
  - Sensitivity may require off-site laboratory measurements
    - Small required sample size

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Conclusions

- **There is no one perfect noble gas tracer**
- Tracers should be chosen based on experiment needs
  - Relevance, cost, detectability, attainability, duration
- Noble gases aren't perfectly unreactive – considerations such as solubility and sorption should be kept in mind
- Radioactive tracers provide significantly improved real-time detection and direct measurement of isotopes of interest
- Stable tracers are generally cheaper and can be handled more easily

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Questions?

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