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Measurements of Ar-39 in Shallow Subsurface Soil

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CTBTO PREPARATORY COMMISSION

INTRODUCTION

We previously reported measuring Ar-39, which has a 269 year half-life, in gas samples from shallow subsurface soil above UNEs. In this work we report on measurements of the Ar-39 background in shallow subsurface soil not associated with UNEs.

METHODS/DATA

Samples were taken at several locations across the western U.S. Air was processed in PNNL's Ar-37 Field System with the concentrated argon then measured in internal proportional counters.

START

RESULTS

Atmospheric measurements were consistent with published results (16.6 mBq/m^3). Shallow subsurface measurements ranged from atmospheric to 3.5x atmospheric levels.

CONCLUSION

We present the first measurements of Ar-39 background in shallow subsurface soil. The results were orders of magnitude less than those observed above UNEs. Ar-39 appears to be a UNE signature that can be detected at the surface for many decades.

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Measurement of Ar-39 in Shallow Surface Soil

It has previously been reported at SnT that PNNL has measured Argon-39 at historic underground nuclear explosion (UNE) sites at the Nevada National Security Site (NNSS) in gas samples from shallow (few meters or less) subsurface soil. Considerable Argon-39 was observed at UNE sites sampled. Thus, the detection of Argon-39 in such samples at strengths sufficiently above background can help identify possible UNEs, though Argon-39's long half-life precludes constraining when the UNE occurred. While a published Argon-39 background value for atmospheric air exists (16.6 mBq/m^3 whole-air equivalent), there were no published Argon-39 background values for shallow subsurface air samples. We report on such measurements at a number of locations across the western United States of America, in an attempt to characterize the range of backgrounds that might exist.

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Motivation

PNNL has previously reported measuring Argon-39 in shallow soil gas at a few NNSS UNE locations (all that we sampled)

Sample Description	Ar-39 Conc. (Bq/m3 whole air)
U20az borehole near chimney	1036
U20az surface borehole (2.4 – 8.1 m)	63 - 102
U19c borehole to chimney	1868
U2ez surface borehole (1.6 – 8.7 m)	5 - 34
U12p tunnel	0.1 - 23
U12p tunnel just outside entrance	5.9
U12p chimney	1460 - 7182
U12p surface atmosphere near SGZ	0.5-0.8

U20az: 20-150 kT UNE
U19c: 20-200 kT UNE
U2ez is in alluvium and approximately 300 m from a couple UNEs (20-200 kT)
U12p: tunnel site of four < 20 kT UNEs

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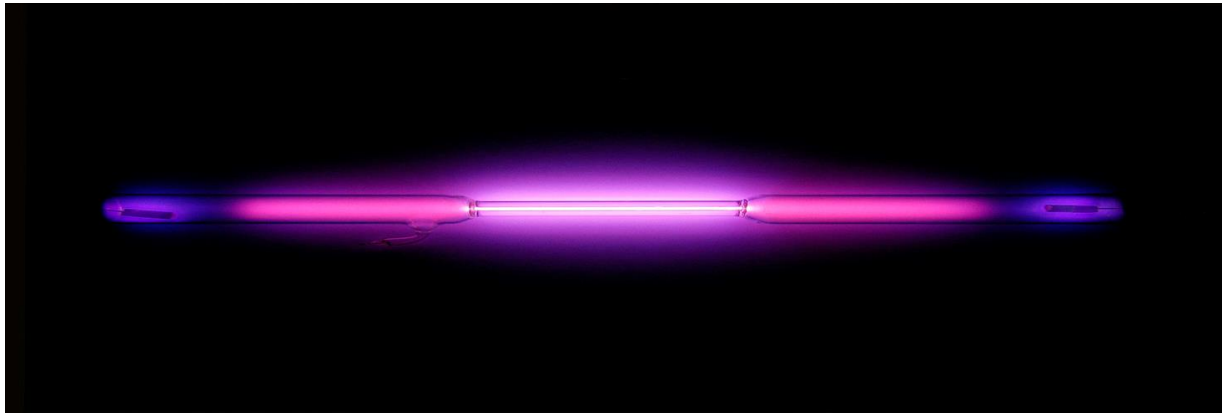
Motivation

-Ar-39 UNE results were reported in:
Johnson et al., J. Env. Rad. 237 (2021) 106715
-Natural Ar-39 background in atmospheric air: 0.017 Bq/m^3 of whole air
Benetti et al., NIM A 574 (2007) 83
-No reported results on Ar-39 backgrounds in shallow soil gas
-A decade ago, there was an effort to understand the range of Ar-37 backgrounds in shallow soil gas in order to understand the limits of its use in detecting UNEs
-We decided to investigate the range of Ar-39 backgrounds in shallow soil gas.
This was not a dedicated project, but pieced together from measurements made for other projects.

-Ar-39 has a 269-year half-life and beta decays (565 keV end-point) to K-39



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Sampling for shallow subsurface background Ar-39

- Samples taken from 0.8 to 3.8 m depth, usually from several holes within a given area
- Also took air samples at Richland and Death Valley

Ch – Chinook Pass (near Mt. Rainier NP)
DV – Death Valley NP
NNSS – Nevada National Security Site (for reference)
R – Richland (PNNL)
TS – Tule Springs National Monument

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Sampling for shallow subsurface background Ar-39

Manual Installation
- Post drive
- Jackhammer
- Handyman Jack

Completion
- SS screens
- Poly tubing
- Sand
- Bentonite

Sample volumes of ~ 500 L monitored for CO_2 and Rn concentrations

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Analysis of shallow subsurface background Ar-39

- Earlier samples (Chinook Pass) were collected as part of the development of PNNL's Ar-37 Field System (See T3.2 oral presentation by Hayes.). Samples were only measured up to 15 keV since Ar-37's signal peaks at 2.8 keV.

- Argon is processed from whole air, and the concentrated argon is mixed with methane to make P10 gas for the internal proportional counters used to make the radioargon measurements

- Using the 5-15 keV (narrow) range from the earlier samples, we could perform a measurement of Ar-39, though with large error bars due to the ~10% detector efficiency.

Argon-37 Analysis Field System test at NNSS during UNESE Experiment

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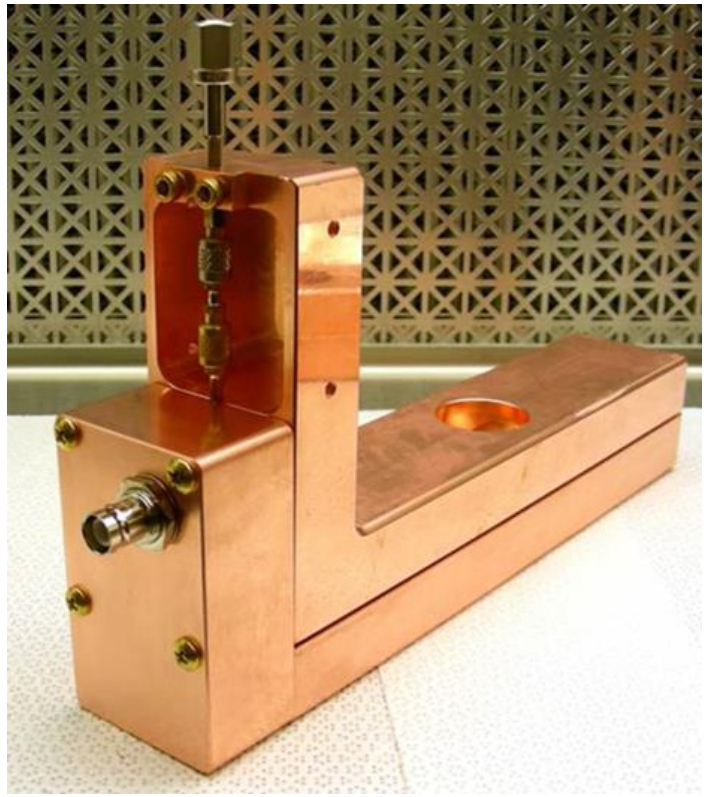
Analysis of shallow subsurface background Ar-39

- Later Ar-39 specific samples were measured with ultra-low-background proportional counters in PNNL's Shallow Underground Laboratory.
- These samples were measured over the 5 – 400 keV (full) range, where the detector efficiency is ~82%.
- The background of these detectors has been characterized using P10 gas made with geologic argon (i.e., no ^{39}Ar).



Internal-source proportional counter

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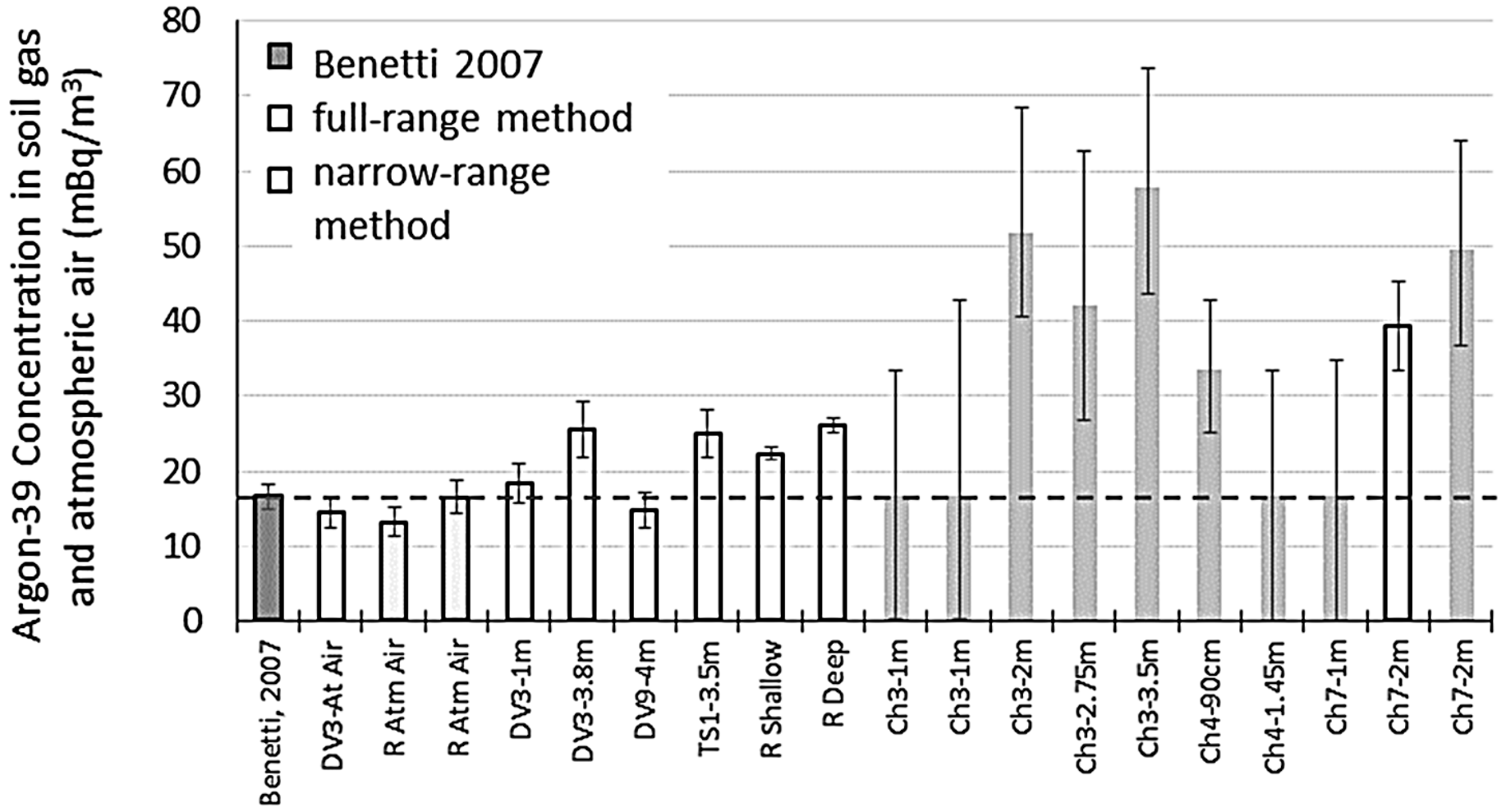
Results



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Results

- Atmospheric air ^{39}Ar measurements consistent with Benetti et al.
- Observed ^{39}Ar concentrations in shallow soil gas varied from atmospheric levels to about 3.5x atmospheric levels. All measured values were much below those observed for UNEs (table on slide 3 where concentrations are in Bq/m^3).
- Sample analyzed both by narrow (5-15 keV) and full (5-400 keV) range method (and different detectors) showed consistent results
- Deeper samples from same location tend to show higher concentrations
- Higher latitudes and elevations tend to show higher concentrations, as expected for cosmic ray activation. (DV-9 is 205 m in elevation while DV-3 is at 1042 m.)
- See Fritz et al., J. Env. Rad. 228 (2021) 106513 for more details

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Conclusions & Future Work

- The work presented was a small effort to make the first measurements of the range of background Ar-39 levels that might be present in shallow soil gas, with some emphasis on geologies similar to the NNSS.
- Ar-39 should be present in shallow soil gas samples taken from UNE sites for a very long time, so it is important to understand the background levels that might exist.
- More samples covering a range of geologies would be useful.
- A more concerted effort to study background concentrations with depth at some locations would be useful, as would modeling of shallow concentrations.
- PNNL and NNSS are beginning a new project to collect shallow soil gas samples at additional UNE sites, and may collect background samples at NNSS as well.

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