

Development of a Field Portable Ar-37 Monitoring Capability



Pacific Northwest
NATIONAL LABORATORY

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The Argon-37 project was initiated in October 2013 and continued through March 2017 with funding from the U.S. Department of State. The outcome of this first phase of the two-phase project was development and fabrication of a laboratory bench argon separation and measurement system, and a field prototype separation and measurement system demonstrating all needed science and engineering properties.

Goals and Objectives

The goal of the High Throughput Argon-37 Collection and Measurement project is to produce two prototype Argon-37 gas collection and analysis measurement systems capable of demonstrating all needed science and engineering properties to provide increased confidence that a nuclear explosion took place at a particular location in support of the suite of detection and analysis technology available for nuclear explosion monitoring.

Argon-37 in the Atmosphere

Argon-37 is produced from neutron interactions with calcium in soil, $^{40}\text{Ca}(n,\gamma)^{37}\text{Ar}$. For field inspections following a nuclear explosion, the 35-day half-life of ^{37}Ar provides both high specific activity and sufficient time for completion of an inspection before radioactive decay limits sensitivity.

Because argon is abundant in air (0.93%), a relatively large amount of argon can be collected from a modest volume of air as compared to other noble gases. Thus in optimizing an ^{37}Ar detector design it is practical to maximize detection sensitivity by maximizing the amount of argon able to be measured as a single sample.

Argon-37 Key Performance Parameters

Characteristics	Threshold	Objective
	(Min. Requirement)	(Desired Goal)
Isotope Measured	^{37}Ar	^{37}Ar
Measurement mode	Auger Electron / X-Ray	Auger Electron / X-Ray
Count time	12 hours/Variable	12 hours/Variable
Minimum Detectable Concentration (MDC)	15 mBq/m ³ per 12 hour count	10 mBq/m ³ per 12 hour count
Operational Mode	With/without xenon system pre-processing	With/without xenon system pre-processing
Data Collection and Analysis	Automated control with operator present	Automated control
Communication	Local/remote control	Local/remote control
Power	National and International power	National and International power

Argon-37 Field Prototype Gas Processing



The system is designed to accept several different sample input configurations: Tedlar bag filled with soil gas, SCUBA tanks, or input from a xenon processing system. The system removes nitrogen, removes oxygen, purifies the argon, then counts the argon gas in one of seven nuclear detectors.

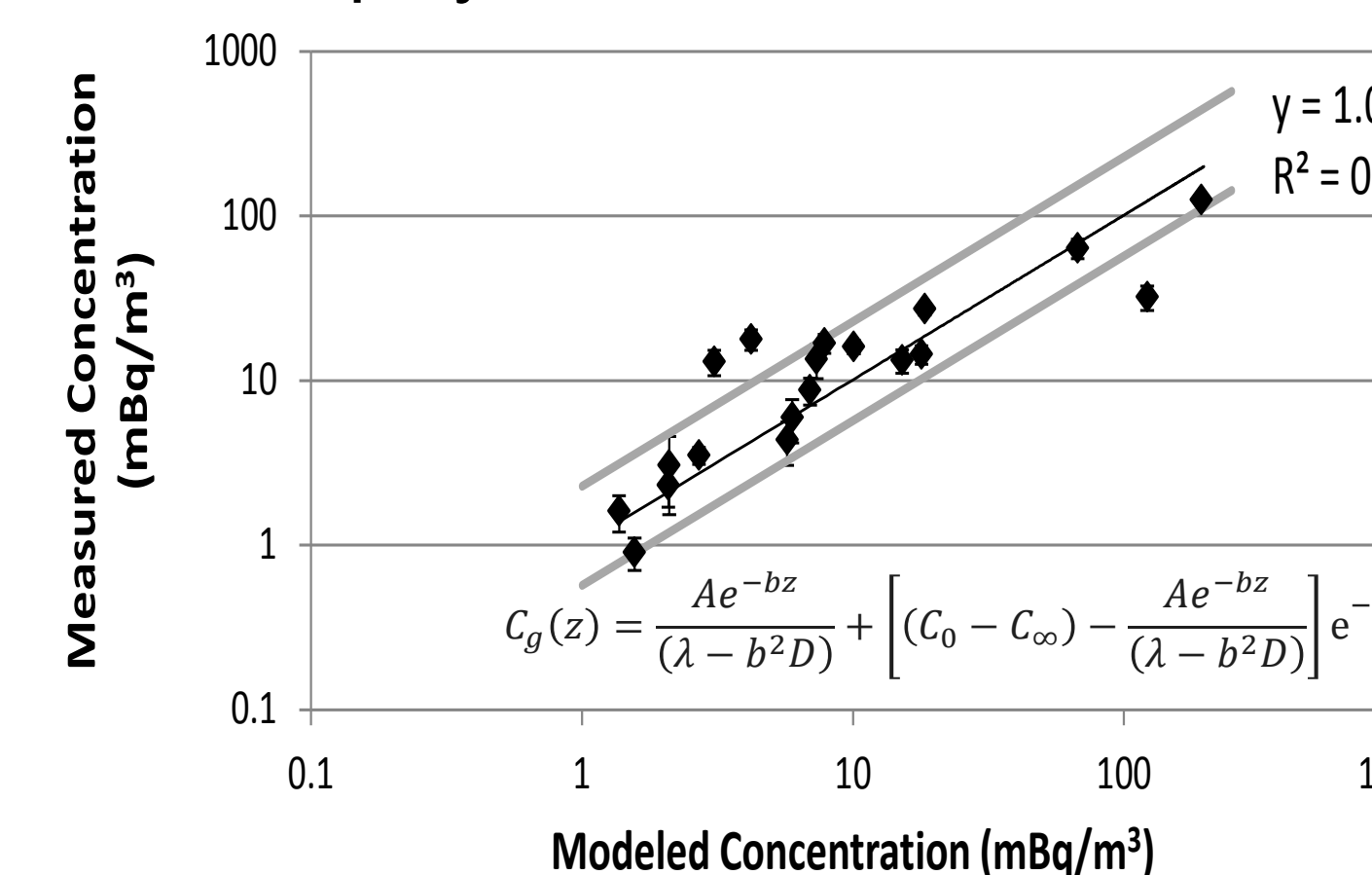
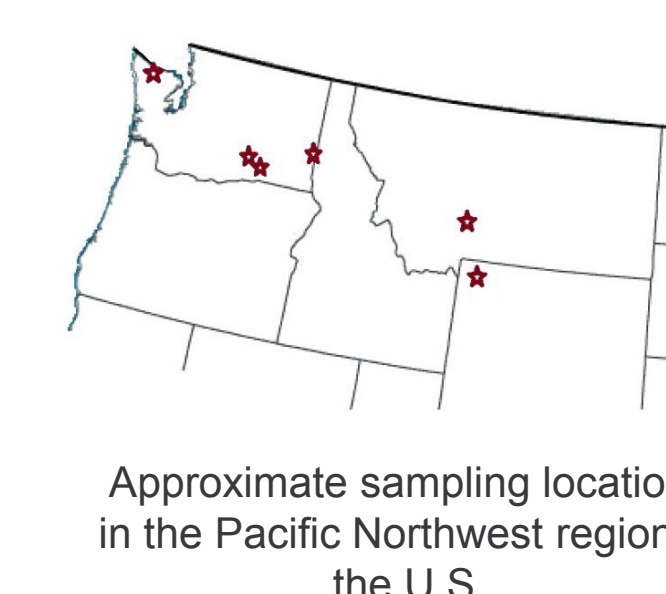


The system can automatically process samples every two hours and put purified argon into a nuclear detector which counts the gas for 12 hours. Six samples per 24 hours can be processed and counted.

Sample Measurements on the Argon-37 System

Sampling in Pacific Northwest

Soil gas samples were obtained from areas around the Pacific Northwest region of the U.S. that were predicted to have high calcium, and hence, high ^{37}Ar concentrations. Samples were processed and measured on the Argon-37 bench-top system at PNNL.



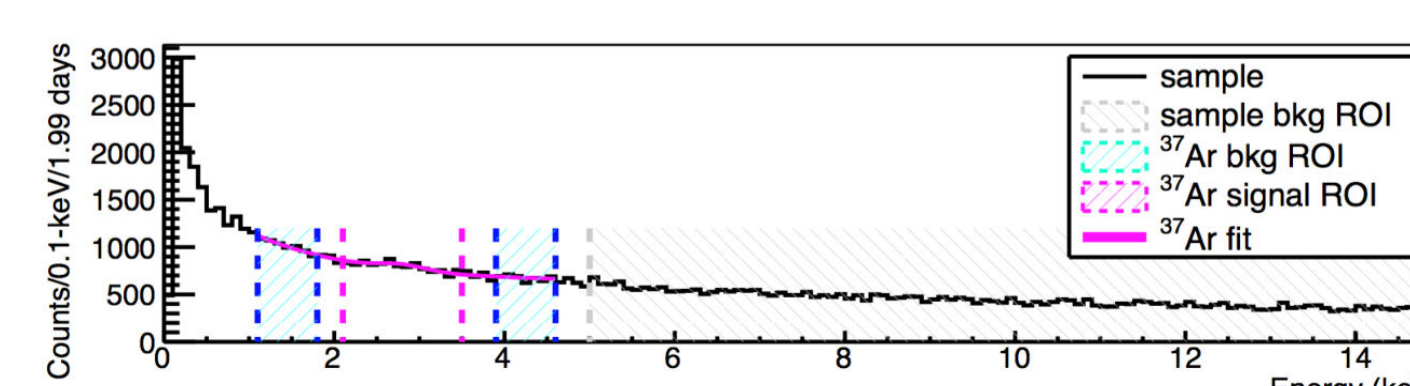
Constants	Value
λ (1/day)	0.0198
T (day)	3
Λ (g/cm ²)	200
ϵ	0.07
ρ_s (g/cm ³)	2.65
$D_{g,0}$ (cm ² /day)	15552
$D_{g,eff}$ (cm ² /day)	2100
C_0	1
C_∞	0.5
$\Psi'_1(0)$	0.00141

Comparison of the measured ^{37}Ar concentrations with calculated production rate (A) and the modeled concentration (B). Error bars represent the error of the measurement. Grey lines represent 2x and 1/2 of the best fit equation.

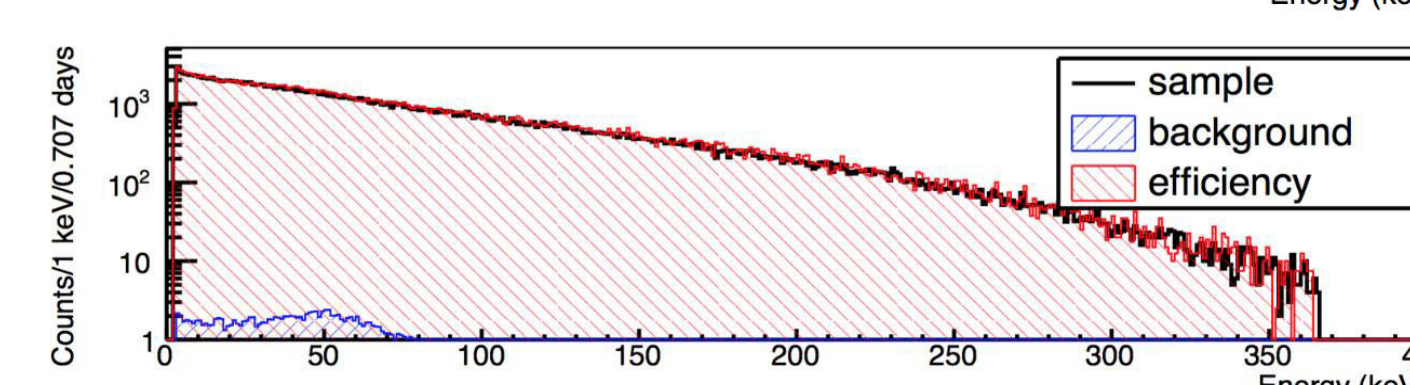
Sampling in Nevada

In experiments conducted at the Nevada National Security Site (NNSS), formerly known as the Nevada Test Site, Pacific Northwest National Laboratory (PNNL) and National Security Technologies (NSTec) simulated the radioactive noble gases that would be created during a nuclear explosion and measured surface concentrations over a period of months.

Pacific Northwest National Laboratory detected ^{39}Ar at the location of an underground nuclear explosion on the NNSS. The presence of ^{39}Ar was not anticipated at the outset of the experimental campaign but results from this work demonstrated that it is present, along with ^{37}Ar and ^{85}Kr in the subsurface at the site of an underground nuclear explosion.



Argon sample collected from the shallow borehole and analyzed. A background is shown in magenta where the ^{37}Ar peak would be located. The background activity in this sample was greatly elevated (860x "normal air") so the sensitivity in the ^{37}Ar ROI was diminished.

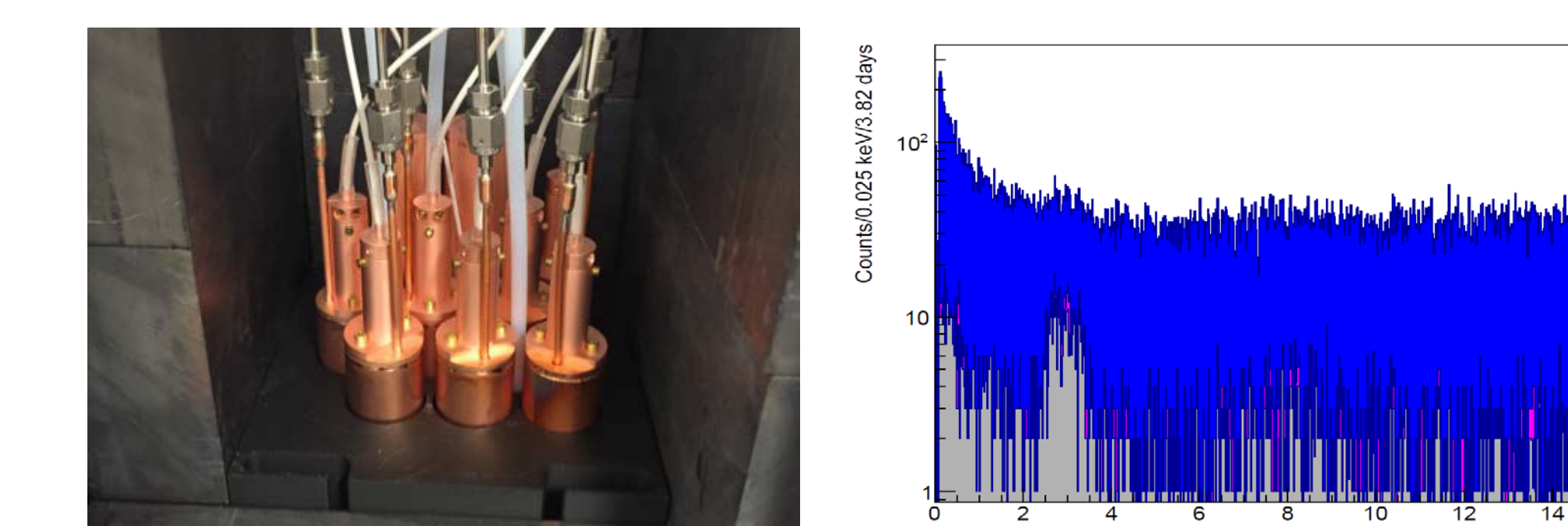


Argon sample recalibrated from 0-400 keV to look at higher energy events and compared to an ^{39}Ar efficiency spectrum (red).

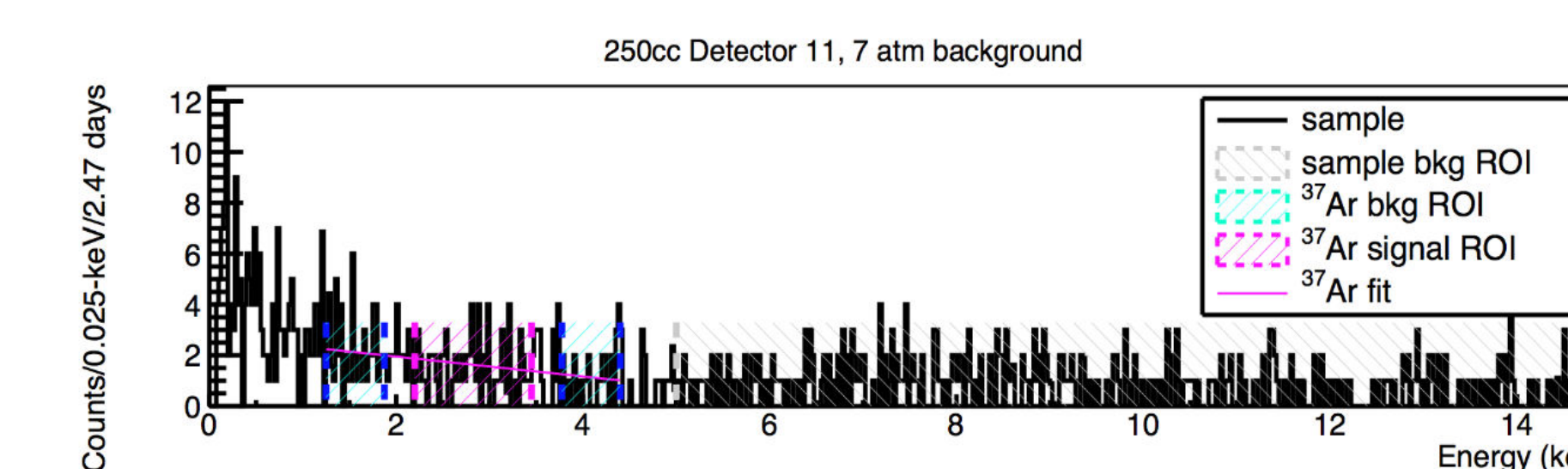
Nuclear Detection

Argon-37 Signature

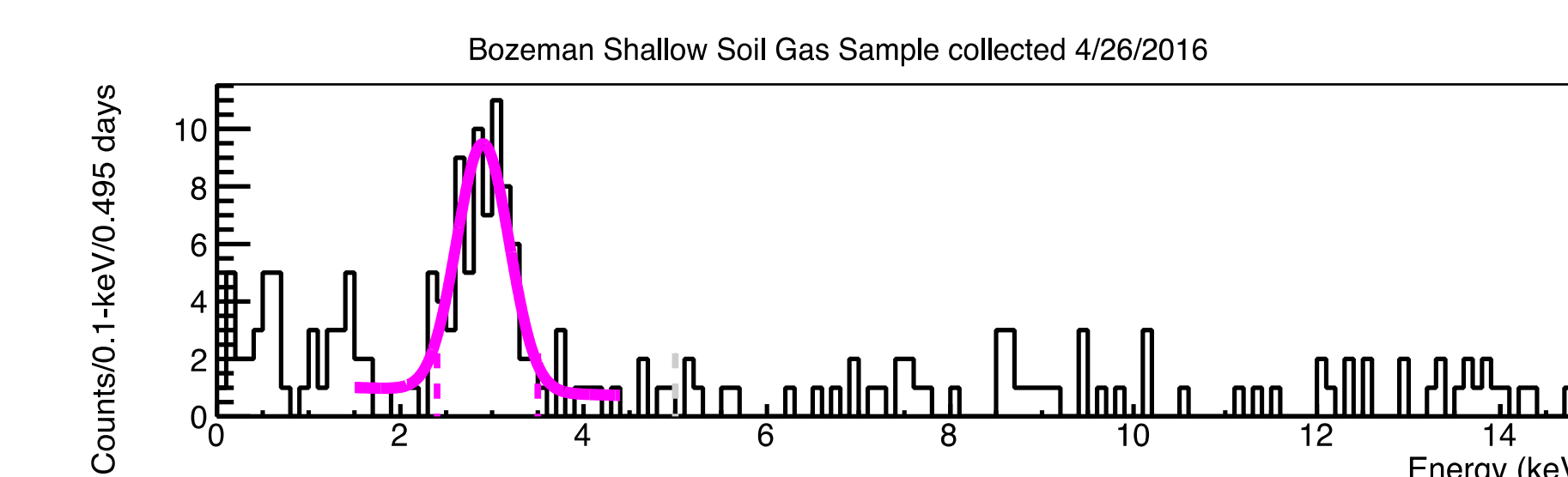
Argon-37 decay is rather challenging to detect, since it decays via electron capture, and emits only low-energy Auger electrons, neutrinos, and x-rays. During analysis, a series of analysis cuts are applied to the digitized events before a net background is calculated. First, events in coincidence with the anti-cosmic veto shield are rejected; this eliminates about 93% of the gross counts. Next, pulse-shape discrimination (PSD) is applied to identify events not arising from gas gain.



Stacked Histogram of the Events Being Rejected by way of Cosmic Veto (blue), Events Failing the PSD Cut (magenta), and Good Events (grey)



Laboratory Bench System 2.5-day measurement: an Upper Limit for this Sample of ~1 mBq/m³ (95% Confidence Level) was Obtained



Twelve-hour Measurement of ^{37}Ar Activity Near the KPP Detection limit (12.1 mBq/m³)

Conclusion

PNNL has successfully designed and tested an end-to-end ^{37}Ar processing capability for above ground measurements in a field environment. A fully functional ^{37}Ar field prototype system has been built, and over 100 samples have been processed on the bench-top system. The ^{37}Ar Field Prototype design will be incorporated into two TRL-6 ^{37}Ar field measurement systems during the next two years.