LOW-RADIOACTIVITY UNDERGROUND ARGON

Pacific Northwest NATIONAL LABORATORY
Proudly Operated by Battelle Since 1965

Developing the First Supply of Underground Argon

THOMAS ALEXANDER

Pacific Northwest National Laboratory

Argon on Earth

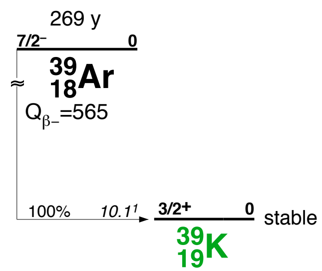
<u>Atmospheric isotopic abundance</u>

J.-Y. Lee, et al., Geochim. Cosmochim. Acta 70 (2006) 4507-4512

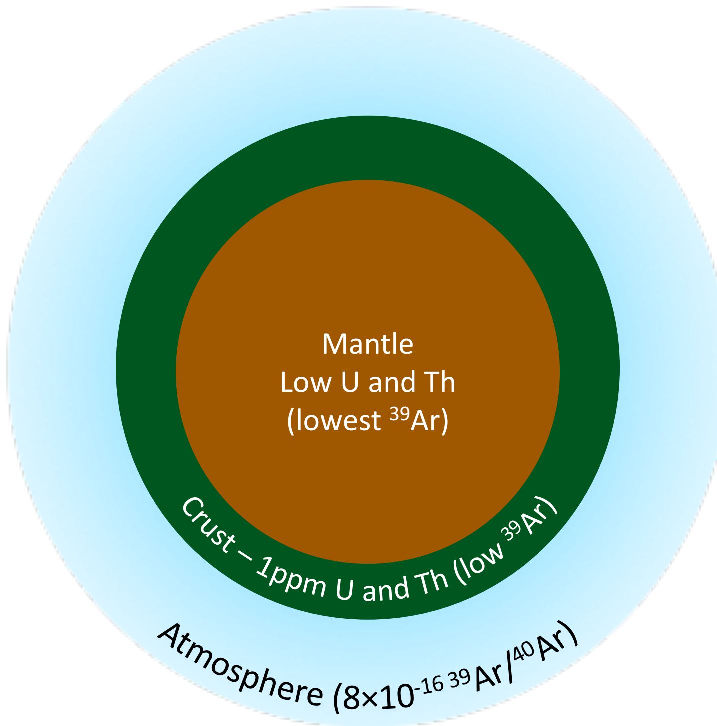
^{36}Ar – 0.334%

^{38}Ar – 0.063%

^{40}Ar – 99.604%



- ^{40}Ar comes from ^{40}K decay
- ^{39}Ar Origins
  - ^{40}Ar(n,2n)→^{39}Ar
  - ^{40}Ar(p,np)→^{39}Ar
- Underground via U + Th fission neutrons



The Hunt

2007: Helium Reserve ^{39}Ar content < 5% atmospheric levels

arXiv:0712.0381v1 [astro-ph] 3 Dec 2007

Discovery of underground argon with low level of radioactive ^{39}Ar and possible applications to WIMP dark matter detectors

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Abstract

We report on the first measurement of ^{39}Ar in argon from underground natural gas reservoirs. The gas stored in the US National Helium Reserve was found to contain a low level of ^{39}Ar. The ratio of ^{39}Ar to stable argon was found to be $\leq 4\times10^{-17}$ (84% C.L.), less than 5% the value in atmospheric argon ($^{39}Ar/Ar=8\times10^{-16}$). The total quantity of argon currently stored in the National Helium Reserve is estimated at 1000 tons. ^{39}Ar represents one of the most important backgrounds in argon detectors for WIMP

2007: Begin Searching CO_2 Wells

Notes on Trip to Bueyeros - May 23-26 2007 and Proposal for Small Scale Argon Production in the 2008 Campaign

F. Calaprice, E. de Haas, C. Galbiati, A. Goretti, A. Ianni, A. Nelson

Physics Department of Princeton University

February 10, 2008

	Gas from Liquid Trap	Gas from the PSA
Ar	290 ppm	2.5%
CH_4	11.5%	1200 ppm
CO_2	79.3%	6500 ppm
C_xH_y	0 ppb	0 ppb
H_2	467 ppm	750 ppm
H_2O	2860 ppm	5000 ppm
He	700 ppm	22.3%
N_2	8.7%	73.8%
O_2	550 ppm	120 ppm

Table 1: Composition of the stream from the liquid trap and of the stream from the PSA unit.

2008: First Production

First Large Scale Production of Argon Depleted in ^{39}Ar from Underground Wells

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Abstract

We report on the first large-scale production of depleted argon from underground gas wells. We processed the exhaust stream of the CO_2 liquifier of the Reliant Dry Ice Plant in Bueyeros, NM, with a special Vacuum Swing Adsorption plant. The CO_2 gas fed directly from the well into the liquifier contains argon at the concentration of 40-70 ppm, and the argon concentration in the exhaust stream of the liquifier is in the range 200-450 ppm. The Vacuum Swing Adsorption plant produces crude argon, concentrating the argon to the level of 80,000–100,000 ppm (8–10%) in a single pass. The argon production rate is 0.5–0.6 kg/day. We determined that the underground argon is depleted in ^{39}Ar by a factor 10 or more relative to atmospheric argon activated by cosmic rays, by analysis in a low-background proportional counter. More accurate analysis of the ^{39}Ar activity is under way.

Depleted argon is of interest for the construction of large scale WIMP dark matter searches and of detectors of reactor neutrinos for non-proliferation efforts. WIMP dark matter searches of high sensitivity may require depleted argon targets of 10 tons or more. Underground argon offers an affordable solution for the production of depleted argon targets. Prior to this work, only a few grams of depleted argon from underground wells were separated and purified from natural gas. To the best of our knowledge, the work reported in this paper is the first production of depleted argon from underground sources at the kg-scale.

2009: Move to Colorado

First Large Scale Production of Low Radioactivity Argon From Underground Sources

H. O. Back[a,*], F. Calaprice[a], C. Condon[a], E. de Haas[a], R. Ford[b], C. Galbiati[a], A. Goretti[a], T. Hohmann[a], An. Ianni[a], B. Loer[a,1], D. Montanari[a,1], A. Nelson[a], A. Pocar[c]

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-ph.IM] 1 May 2012

Abstract

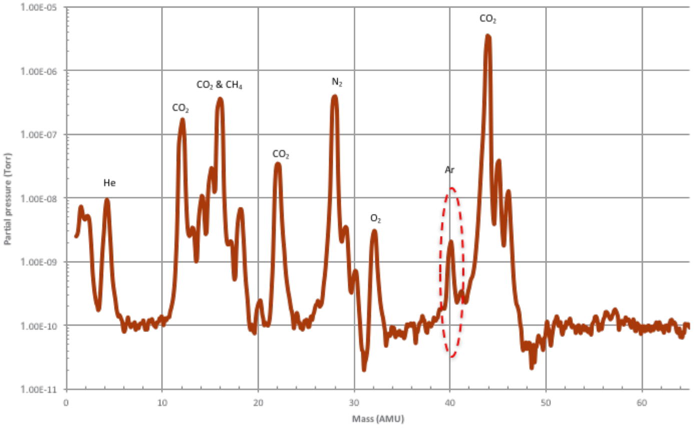
We report on the first large-scale production of low radioactivity argon from underground gas wells. Low radioactivity argon is of general interest, in particular for the construction of large scale WIMP dark matter searches and detectors of reactor neutrinos for non-proliferation efforts. Atmospheric argon has an activity of about 1 Bq/kg from the decays of ^{39}Ar; the concentration of ^{39}Ar in the underground argon we are collecting is at least a factor of 100 lower than this value.

The argon is collected from a stream of gas from a CO_2 well in southwestern Colorado with a Vacuum Pressure Swing Adsorption (VPSA) plant. The gas from the well contains argon at a concentration of 400-600 ppm, and the VPSA plant produces an output stream with an argon concentration at the level of 30,000-50,000 ppm (3-5%) in a single pass. This gas is sent for further processing to Fermilab where it is purified by cryogenic distillation. The argon production rate is presently 0.5 kg/day.

Keywords: depleted argon, dark matter, pressure swing adsorption

Exploration at Doe Canyon

CO_2 well in SW Colorado with 400ppm Ar
(Nucl. Phys. B, 197 (2009) 70-73)
(Nucl. Instr. Meth. A 587 (2008) 46-51)

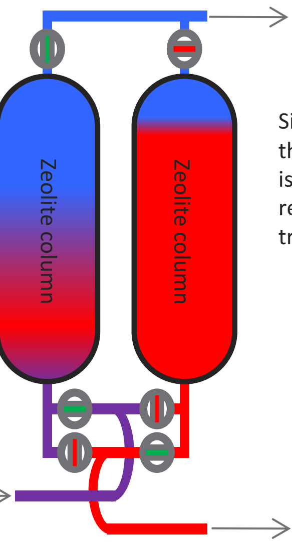


Argon Extraction-Cortez, Colorado

Approximate Input:

Gas Type	Concentration from well
Carbon Dioxide	96%
Nitrogen	2.4%
Methane	0.57%
Helium	0.43%
Other hydrocarbons	0.21%
Argon	440 ppm

Gas flows through one column under pressure. CO_2, H_2O and CH_4 are adsorbed on zeolite



He, Ar, N_2 mixture

Simultaneously the other column is pumped on to remove the trapped gases

Gas from well

Adsorbed gas returned to company

Gas Type	Concentration in Output
Helium	85-95%
Argon	3-6%
Nitrogen	1-10%
Methane, Oxygen	Trace
Carbon Dioxide	Trace
Other Hydrocarbons	Trace

Average Production: 140g/day

First Measurement of Underground Argon

2011-2012

A Study of the Residual ^{39}Ar Content in Argon from Underground Sources

J. Xu[a], F. Calaprice[a,*], C. Galbiati[a], A. Goretti[b], G. Guray[a], T. Hohman[a,1], D. Holtz[a,2], A. Ianni[b], M. Laubenstein[b], B. Loer[a,3], C. Love[c,4], C.J. Martoff[c], D. Montanari[a,3], S. Mukhopadhyay[d], A. Nelson[a], S.D. Rountree[e], R.B. Vogelaar[e], A. Wright[a]

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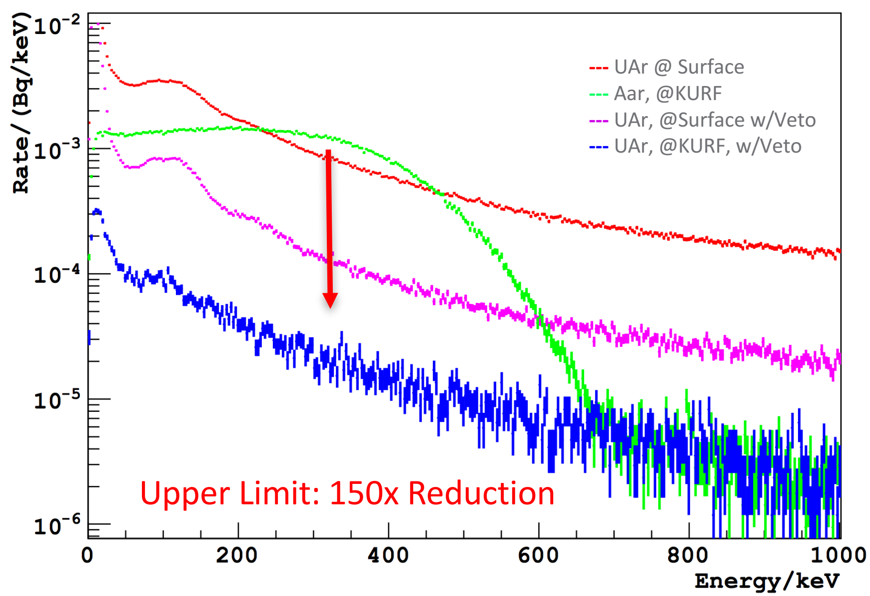
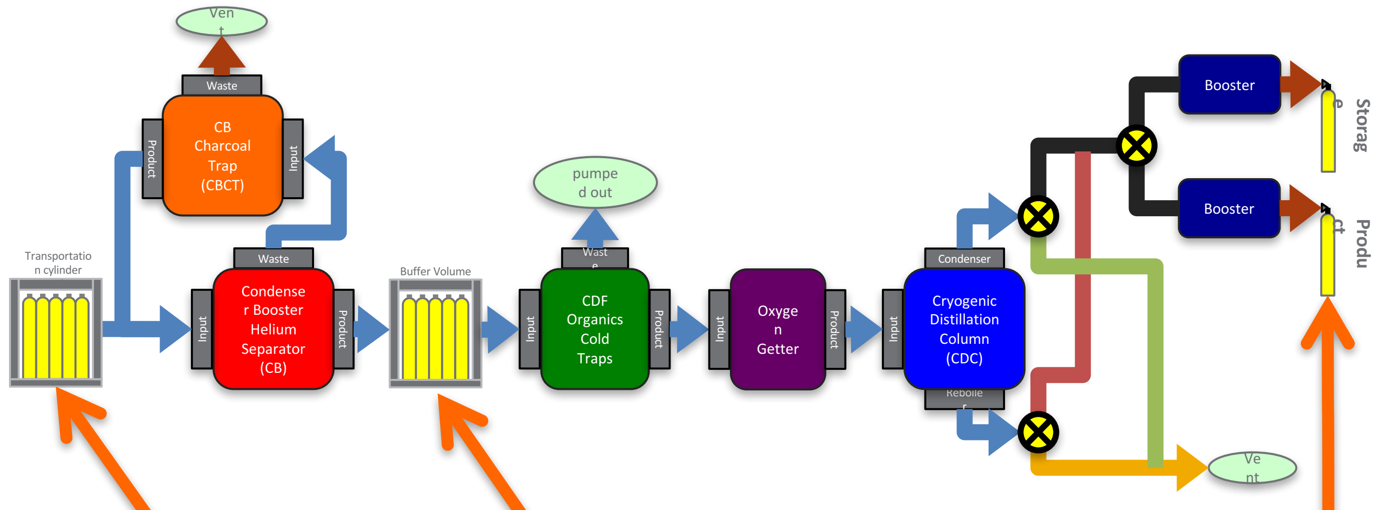
Abstract

The discovery of argon from underground sources with significantly less ^{39}Ar than atmospheric argon was an important step in the development of direct-detection dark matter experiments using argon as the active target. We report on the design and operation of a low background detector with a single phase liquid argon target that was built to study the ^{39}Ar content of the underground argon. Underground argon from the Kinder Morgan CO_2 plant in Cortez, Colorado was determined to have less than 0.65% of the ^{39}Ar activity in atmospheric argon.

Keywords: underground argon, dark matter search technique,



Purification– Fermilab, Illinois, USA



Colorado gas
3-6% Ar, 1-10% N_2, 85-95% He

1 kg/day

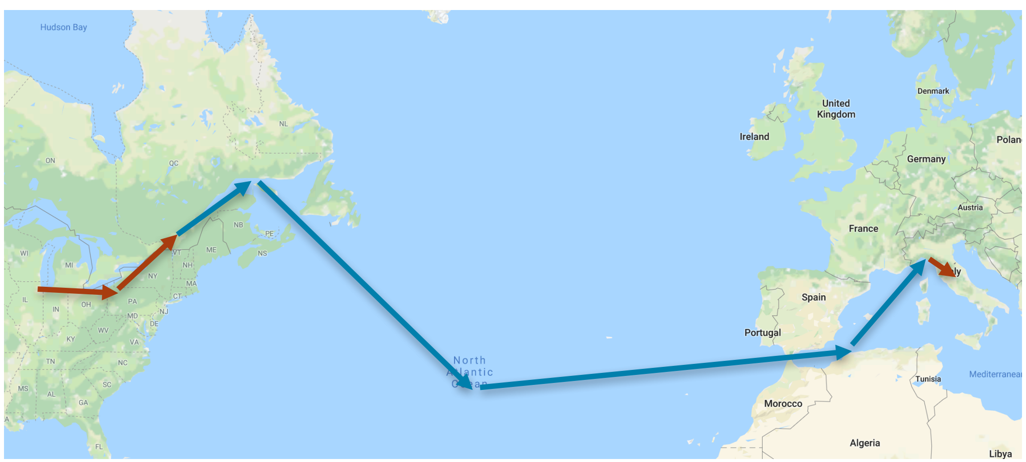
After He separation order-of: 40% Ar, 60% N_2

Total DS-50 production: 157 kg

Final contamination concentration	
Nitrogen	279 ppm
Oxygen	192 ppm
Methane	95 ppm
Helium	3 ppm
Carbon Dioxide	14 ppm

The Target Leaves FNAL for LNGS

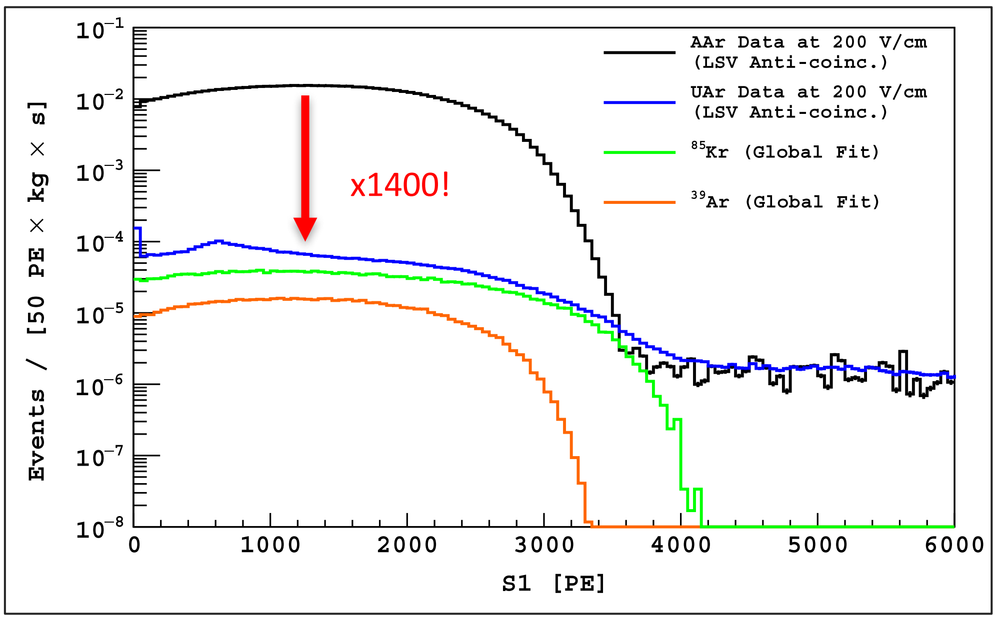
- Underground Argon tested in SCENE cryostat
  - Test final polishing technique (Zirconium Getter)
  - Recaptured in Condenser Booster, "Lossless test"
- Moved to coast by truck, transported by sea
  - Reduce possible cosmogenic activation
- Final ~10 kg flown
- Detector filled April 2015

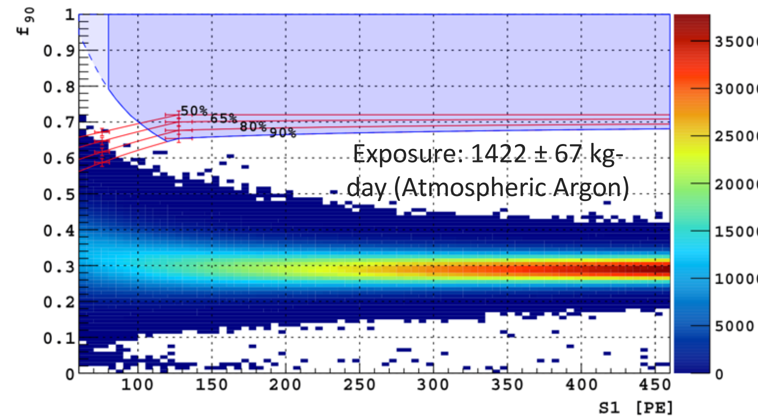


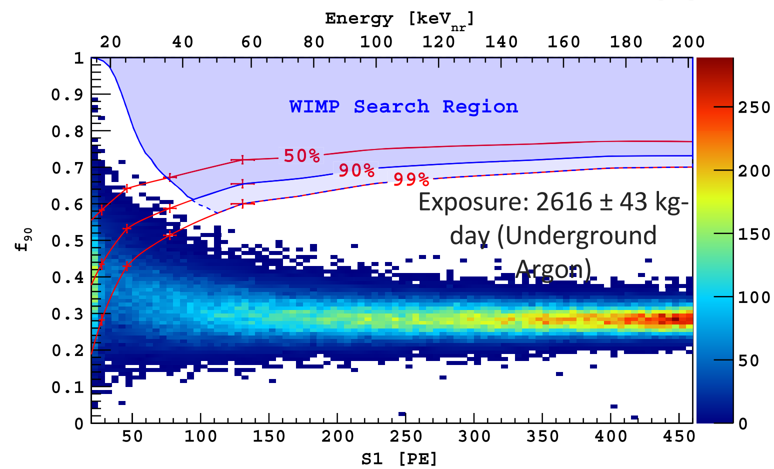
DarkSide-50: The First Underground Argon-based Dark Matter Detector.

Physics Letters B, 743 (2015) & Phys. Rev. D 93, 081101(R) (2016)

- 157 kg produced in 5 years
- ^{39}Ar – 0.73 mBq/kg
- ^{85}Kr – 2.05 mBq/kg







Tackling Future Challenges

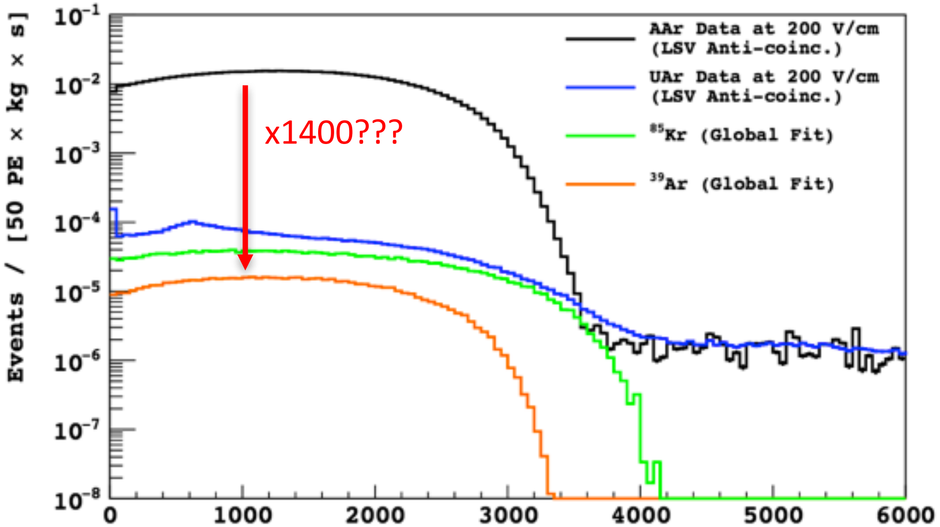
- **VPSA Challenges:**
  - Operating R&D plant as production facility
  - Lower production rate than expected
  - Zeolite poisoning
    - Reduced adsorption efficacy over time
    - 900 lbs of zeolite was replaced 3 times

- **Distillation/Purification Challenges**
  - Unexpected helium and oxygen levels
  - Minor contaminations in VPSA output clogged distillation column
  - Low production rate from VPSA (recapture of UAr from waste streams)

Contaminants found in Zeolite and Purification Systems					
C_3H_8	$C_5H_{10}O$	C_7H_{14}	$C_6H_{12}O$	C_7H_{16}	C_8H_{18}
$C_5H_{10}O$	$C_5H_{10}O$	$C_6H_{13}I$	$C_6H_{12}O$	$C_6H_{12}O$	C_8H_{18}
C_5H_{12}	C_6H_{14}	$C_6H_{13}I$	C_7H_{16}	$C_5H_8O_2$	$C_6H_{10}O_2$
C_6H_{14}	$C_6H_{12}O$	C_7H_{16}	C_6H_6	C_8H_{16}	C_8H_{18}
C_5H_{10}	C_6H_{12}	C_7H_{16}	C_6H_6	C_8H_{16}	C_9H_{20}

Why Did We See Oxygen? One Possibility.

- Could all the ^{39}Ar have come from an air infiltration?
  - ^{39}Ar concentration air equivalent:
    1.95 $mols_{Air}/kg_{UAr}$
- Air equivalent for O_2 contamination in CO_2
  - Assume all O_2 is from air
  - Precision gas analysis of CO_2 [one snapshot]:
    - O_2 = 6.7 ppm
    - Ar = 427 ppm
  - DS-50 target production history:
    - Always measured O_2/Ar ~1.5-2%



Phys. Rev. D. **93** (2016)

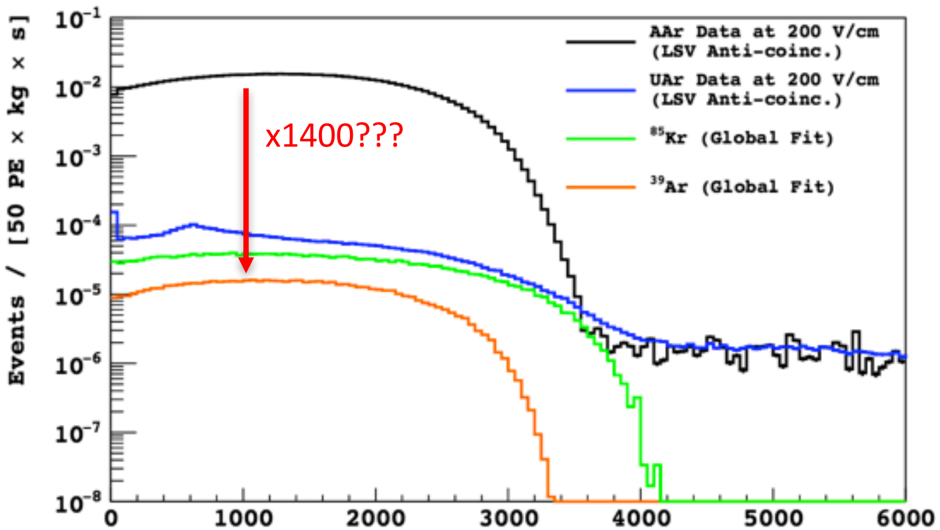
6.7 ppm_{O2} / 427 ppm_{UAr} ➔ 0.392 $mols_{O2}/kg_{UAr}$ ➔ **1.87 $mols_{Air}/kg_{UAr}$**

Why Did We See Oxygen? One Possibility.

- ^{39}Ar concentration air equivalent: **1.95 mols$_{Air}$/kg$_{UAr}$**
- O2 Contamination in Argon: **1.87 mols$_{Air}$/kg$_{UAr}$**

Air equivalent for ^{85}Kr contamination

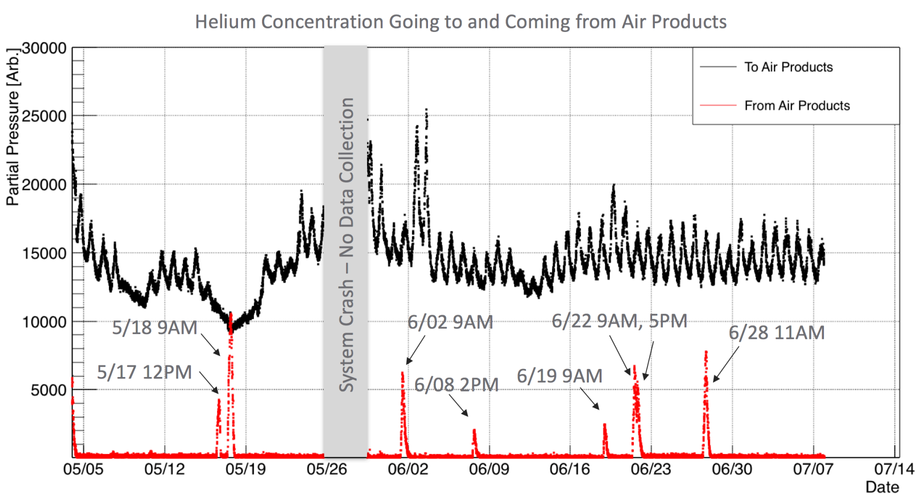
- Assume all ^{85}Kr is from air
  - ^{85}Kr concentration in air = 1.3 Bq/m$^3_{air}$
  - **2.05 mBq/kg$_{UAr}$ ➔ 0.070 mols$_{Air}$/kg$_{UAr}$**
- Assume ^{85}Kr can absorb on zeolite ~ like N_2
  - N_2 reduction factor through VPSA = 35
- **0.070 mols$_{Air}$/kg$_{UAr\text{-}DS50}$ × 35 = 2.45 mols$_{Air}$/kg$_{UAr\text{-}CO2}$**
  - Very rough, NOT proof of contamination



Phys. Rev. D. 93 (2016)

LOGAN: The LOng-term Gas ANalyzer

- Based on an SRS UGA
  - Great for qualitative, or if well calibrated, quantitative, measurements
- Ruggedized
- Custom control software
- Autonomous



UAr production – Onwards and Upwards