

# Procuring 50 Tonnes of Underground Argon for DS-20k

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University of Houston

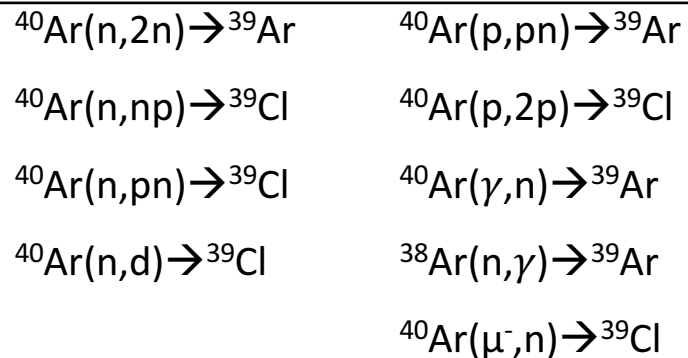
UCLA DM 2018  
February 23, 2018



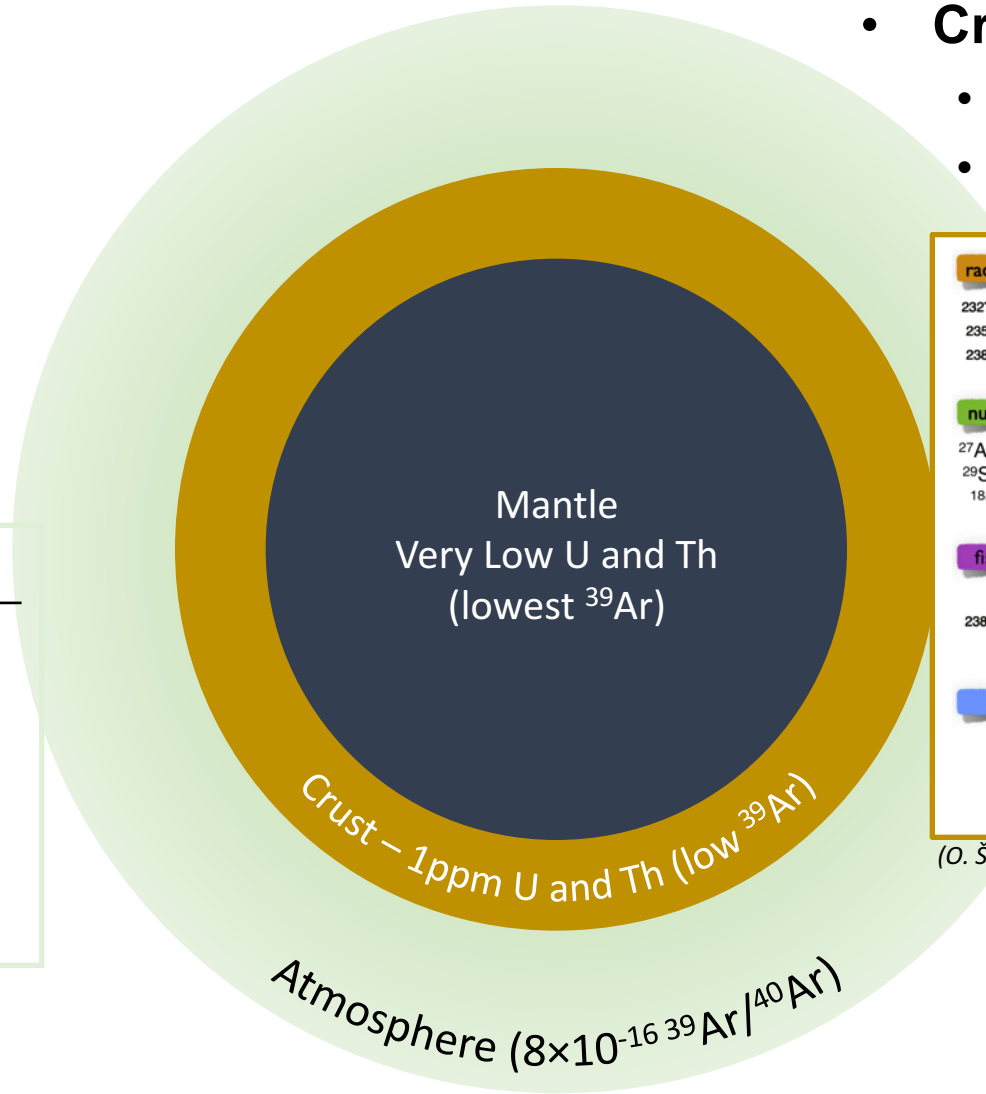
# Terrestrial Ar Isotope Production

- $^{36}\text{Ar}$  dominates in Universe
- $^{40}\text{Ar}$  comes from  $^{40}\text{K}$  decay
- **Atmosphere**
  - $^{39}\text{Ar}$  produced by cosmic rays

## $^{39}\text{Ar}$ production reactions

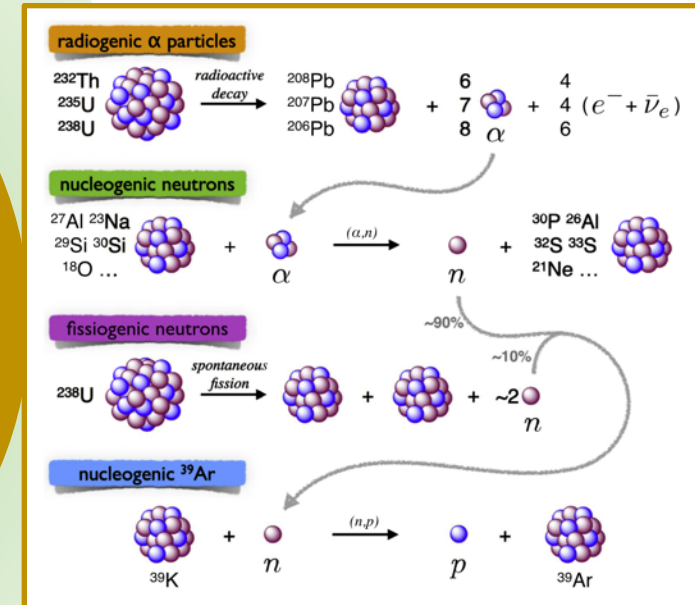


(Loosli & Oeschger, *Earth Planet. Sci. Lett.* 5 (1968) 191-198)



## • Crust

- No cosmic rays
- $^{39}\text{Ar}$  produced underground



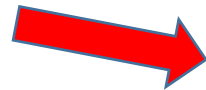
(O. Šrámek, et al., *Geochim. Cosmochim. Acta* 196 (2017) 370)



# Tacking Down a Source

2007

In USA Helium reservoir  $^{39}\text{Ar}$  content < 5% compared to atmospheric Argon



2007

The exploration was extended to  $\text{CO}_2$  wells



2008

@ Reliant plant  
Production rate  $\sim 0.5$  kg/day  
Reduction of  $^{39}\text{Ar}$  compared to atmospheric > factor 10

Available online at www.sciencedirect.com  
ScienceDirect  
Nuclear Instruments and Methods in Physics Research A 587 (2008) 46–51  
www.elsevier.com/locate/nucinst

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

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

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

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Physics Department of Princeton University

February 10, 2008

	Gas from Liquid Trap	Gas from the PSA
Ar	290 ppm	2.5%
CH <sub>4</sub>	11.5%	1200 ppm
CO <sub>2</sub>	79.3%	6500 ppm
C <sub>2</sub> H <sub>2</sub>	0 ppb	0 ppb
H <sub>2</sub>	467 ppm	750 ppm
H <sub>2</sub> O	2860 ppm	5000 ppm
He	700 ppm	22.3%
N <sub>2</sub>	8.7%	73.8%
O <sub>2</sub>	550 ppm	120 ppm

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

First Large Scale Production of Argon Depleted in  $^{39}\text{Ar}$  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  $\text{CO}_2$  liquifier of the Reliant Dry Ice Plant in Bueyeros, NM, with a special Vacuum Swing Adsorption plant. The  $\text{CO}_2$  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 (8–16%) in a single pass. The argon production rate is 0.5–0.6 kg/day. We determined that the underground argon is depleted in  $^{39}\text{Ar}$  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  $^{39}\text{Ar}$  activity is under way.

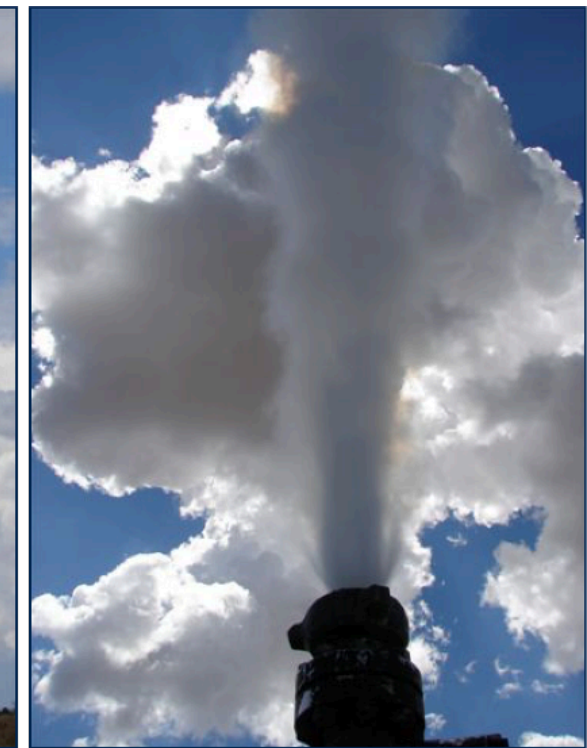
Depleted argon is of interest for the construction of large scale WIMP dark matter searches and of detectors of reactor neutrinos for non-proliferation efforts. WIMP dark matter searches of high sensitivity may require depleted argon targets of 10 tons or more. Underground argon offers an affordable 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.

Key words: Dark Matter; Low Background Detectors; Cryogenic Noble Gases.

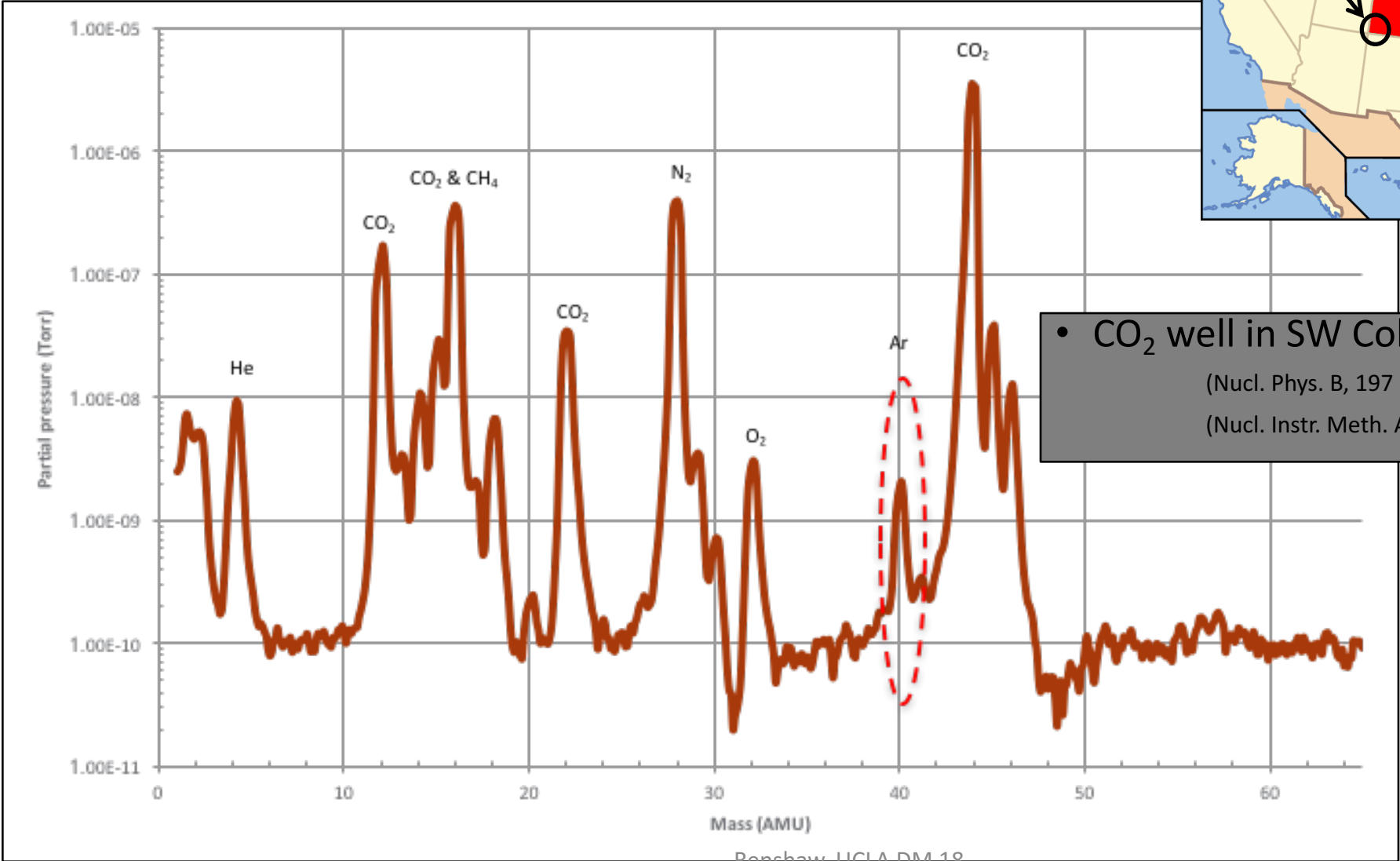


# Source Found

New exploration at Doe Canyon in 2008 ...



# DarkSide UAr Source

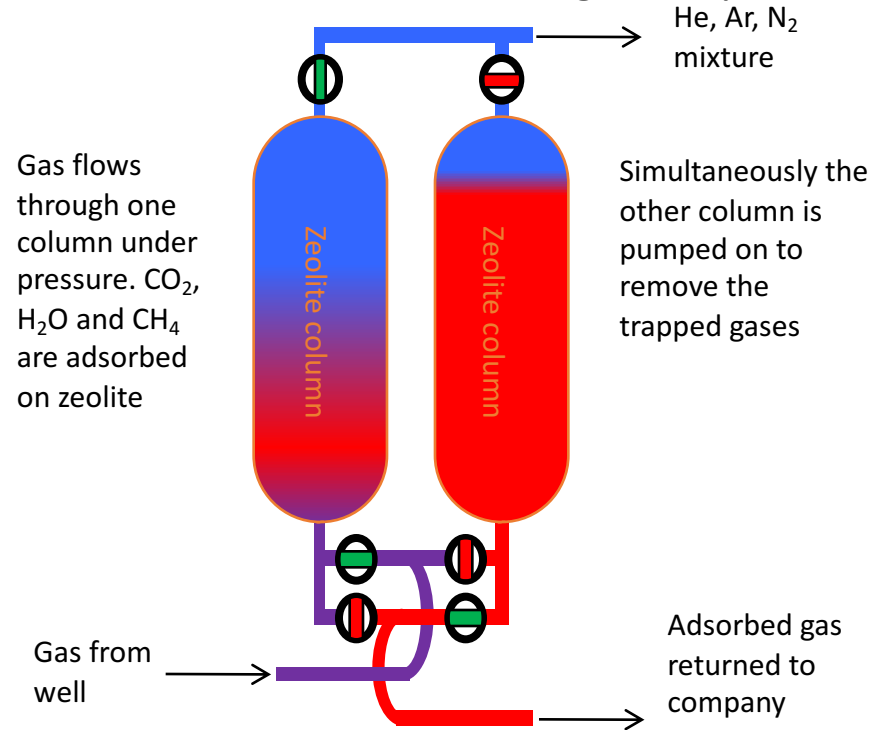


- CO<sub>2</sub> well in SW Colorado with 400 ppm Ar  
(Nucl. Phys. B, 197 (2009) 70-73)  
(Nucl. Instr. Meth. A 587 (2008) 46-51)



# DS-50 UAr Extraction: Doe Canyon

## Vacuum-Pressure Swing Adsorption



Contaminants trapped in VPSA zeolite		
C <sub>3</sub> H <sub>8</sub>	C <sub>7</sub> H <sub>14</sub>	C <sub>7</sub> H <sub>16</sub>
C <sub>5</sub> H <sub>10</sub> O	C <sub>6</sub> H <sub>13</sub> I	C <sub>6</sub> H <sub>12</sub> O
C <sub>5</sub> H <sub>12</sub>	C <sub>6</sub> H <sub>13</sub> I	C <sub>5</sub> H <sub>8</sub> O <sub>2</sub>
C <sub>6</sub> H <sub>14</sub>	C <sub>7</sub> H <sub>16</sub>	C <sub>8</sub> H <sub>16</sub>
C <sub>5</sub> H <sub>10</sub>	C <sub>7</sub> H <sub>16</sub>	C <sub>8</sub> H <sub>16</sub>
C <sub>5</sub> H <sub>10</sub> O	C <sub>6</sub> H <sub>12</sub> O	C <sub>8</sub> H <sub>18</sub>
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C <sub>6</sub> H <sub>14</sub>	C <sub>7</sub> H <sub>16</sub>	C <sub>6</sub> H <sub>10</sub> O <sub>2</sub>
C <sub>6</sub> H <sub>12</sub> O	C <sub>6</sub> H <sub>6</sub>	C <sub>8</sub> H <sub>18</sub>
C <sub>6</sub> H <sub>12</sub>	C <sub>6</sub> H <sub>6</sub>	C <sub>9</sub> H <sub>20</sub>

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

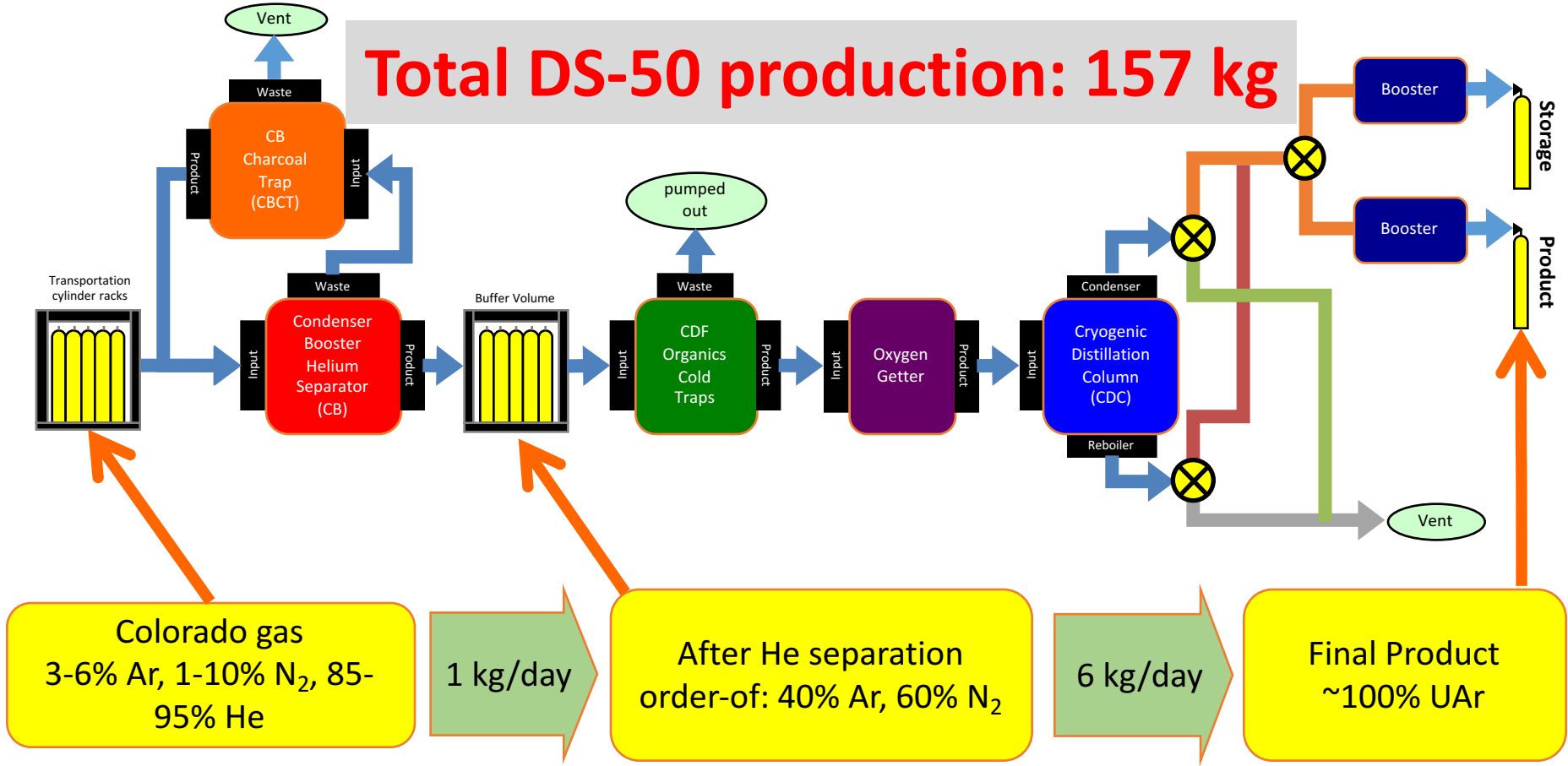


- Approximate product composition:
  - He – 85-95%
  - Ar – 3-6%
  - N<sub>2</sub> – 1-10%
- Average production rate:
  - 140 g/day



# DS-50 UAr Purification: Fermilab

**Total DS-50 production: 157 kg**



Colorado gas  
3-6% Ar, 1-10% N<sub>2</sub>, 85-95% He

1 kg/day

After He separation  
order-of: 40% Ar, 60% N<sub>2</sub>

6 kg/day

Final Product  
~100% UAr



Contaminants frozen in cryogenic systems					
C <sub>3</sub> H <sub>8</sub>	C <sub>5</sub> H <sub>10</sub> O	C <sub>7</sub> H <sub>14</sub>	C <sub>6</sub> H <sub>12</sub> O	C <sub>7</sub> H <sub>16</sub>	C <sub>8</sub> H <sub>18</sub>
C <sub>5</sub> H <sub>10</sub> O	C <sub>5</sub> H <sub>10</sub> O	C <sub>6</sub> H <sub>13</sub> I	C <sub>6</sub> H <sub>12</sub> O	C <sub>6</sub> H <sub>12</sub> O	C <sub>8</sub> H <sub>18</sub>
C <sub>5</sub> H <sub>12</sub>	C <sub>6</sub> H <sub>14</sub>	C <sub>6</sub> H <sub>13</sub> I	C <sub>7</sub> H <sub>16</sub>	C <sub>5</sub> H <sub>8</sub> O <sub>2</sub>	C <sub>6</sub> H <sub>10</sub> O <sub>2</sub>
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C <sub>5</sub> H <sub>10</sub>	C <sub>6</sub> H <sub>12</sub>	C <sub>7</sub> H <sub>16</sub>	C <sub>6</sub> H <sub>6</sub>	C <sub>8</sub> H <sub>16</sub>	C <sub>9</sub> H <sub>20</sub>



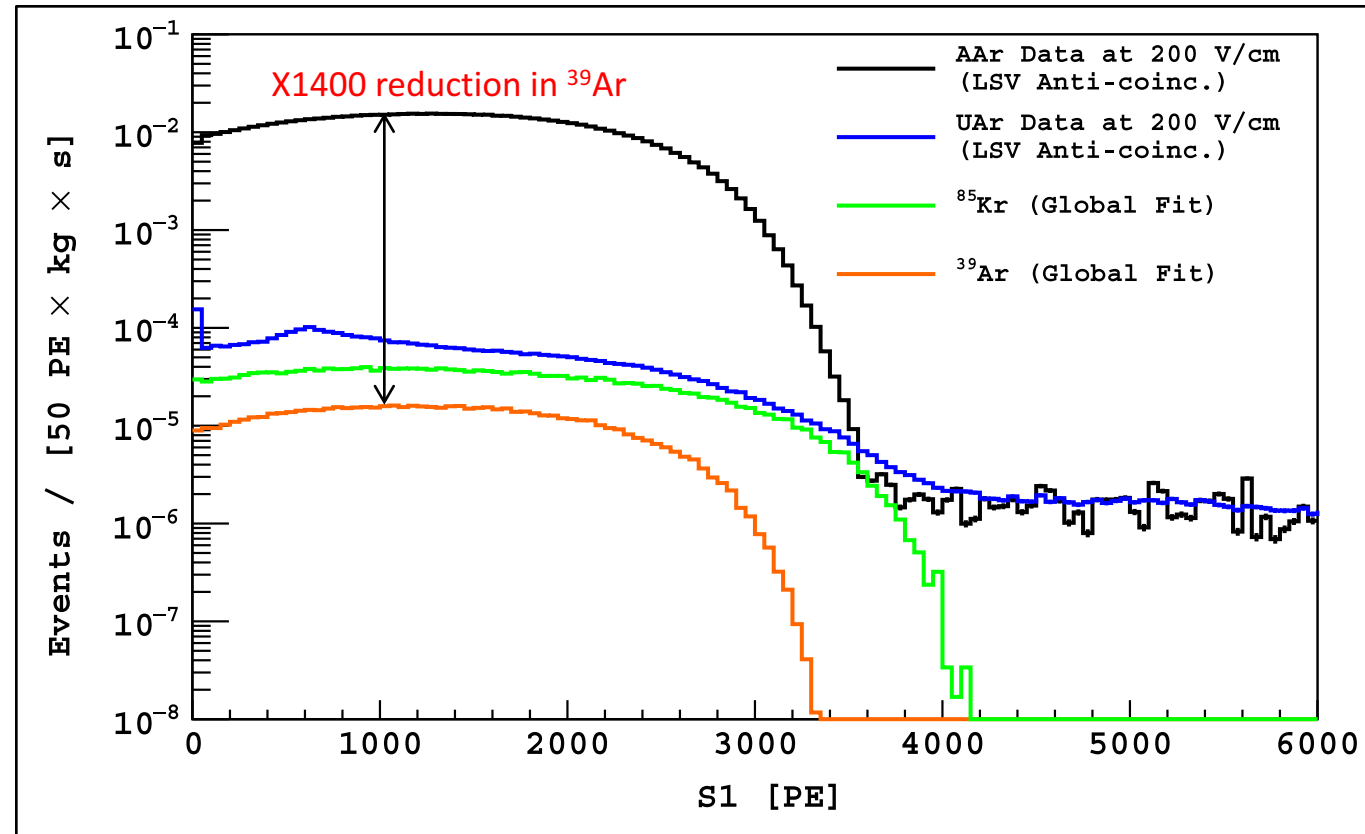
# Successful Target Production

- $^{39}\text{Ar}$  –  $0.73 \pm 0.11$  mBq/kg
- $^{85}\text{Kr}$  –  $2.05 \pm 0.13$  mBq/kg

**TOTAL UAr MASS ~ 157.5 kg**  
**Filled into Darkside-50**  
**on April 3, 2015**

- Residual contamination after all processing (measured by PNNL):

	Concentration	moles	mass (g)
Nitrogen	279 ppm	1.120	31.37
Oxygen	192 ppm	0.773	24.74
Methane	95 ppm	0.380	6.08
Helium	3 ppm	0.014	0.054
Carbon Dioxide	14 ppm	0.055	2.42



*P. Agnes et al. (DarkSide Collaboration), Phys. Rev. D 93, 081101(R)*

**$^{39}\text{Ar} < 0.07\%$  of atmospheric argon**

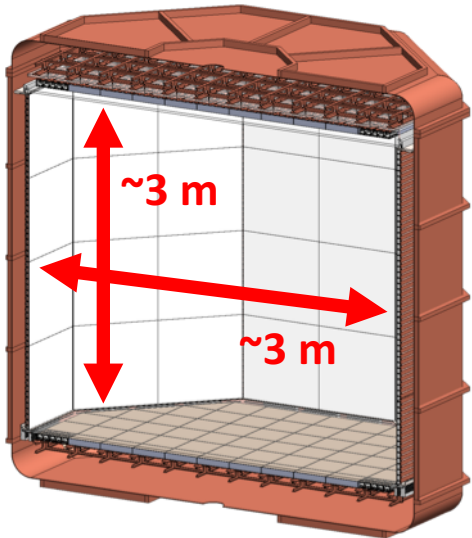




# Scaling-Up UAr Production

ArDM  
DarkSide  
DEAP  
MiniCLEAN

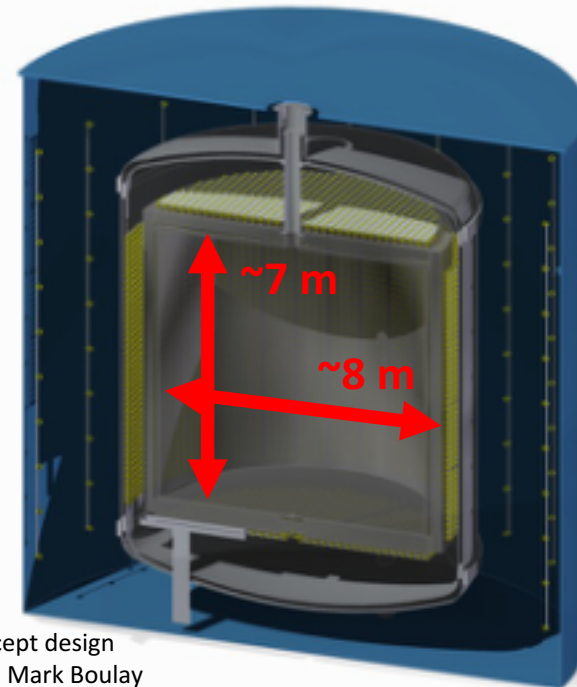
A Single Global Program for Direct Dark Matter Searches  
Currently taking data: ArDM, DarkSide-50, **DEAP-3600**  
**Next step: DarkSide-20k at LNGS (2021-)**  
Last Step: **300 tonnes detector**, location t.b.d (2027-)



## DarkSide-20k

- 30 tonnes UAr total
- Target needed by 2021
- With construction, need to produce target on timescale of 1 year

→ Target ~250 kg/day extraction rate

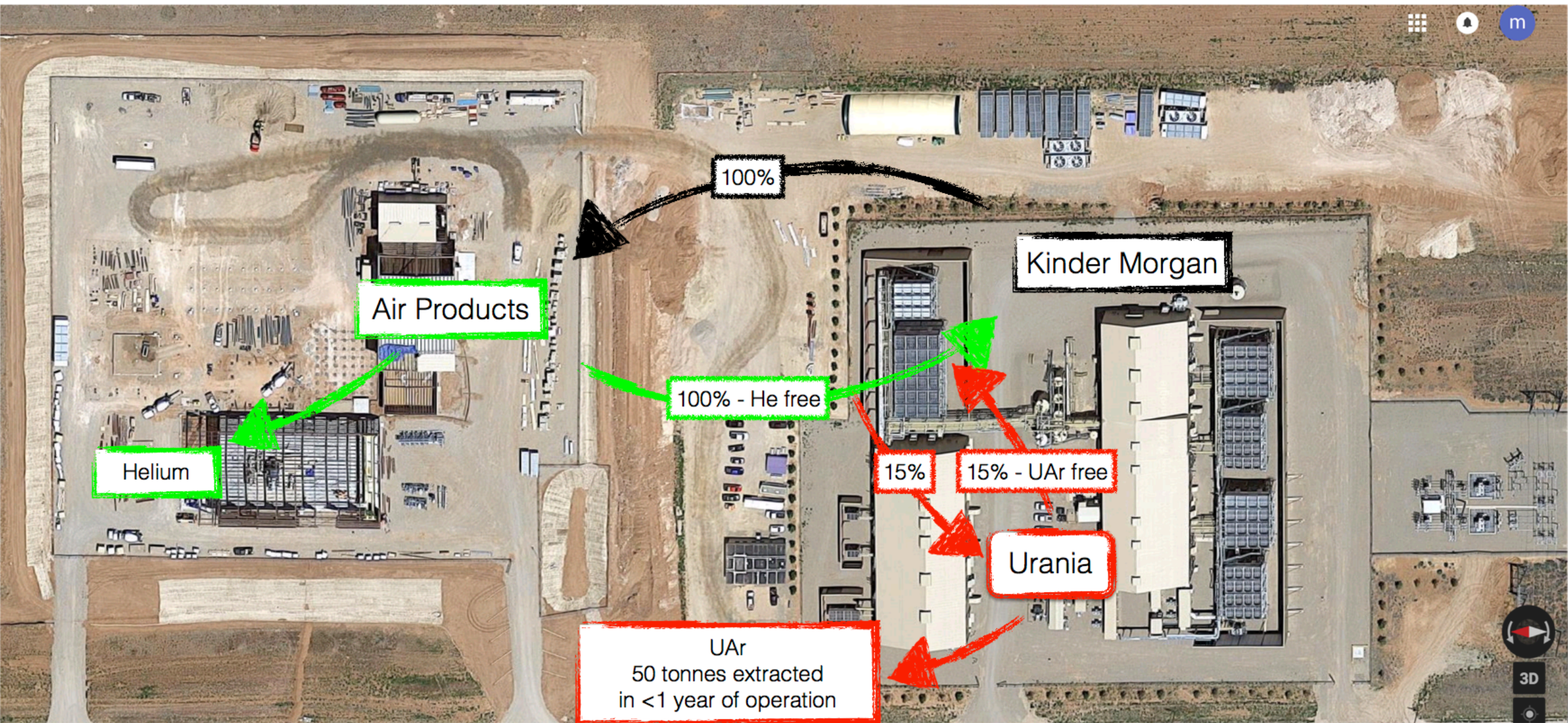
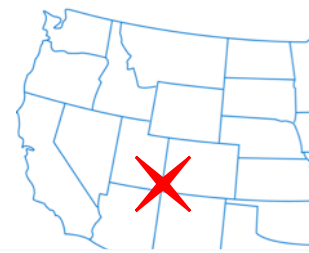


## Future 300 tonnes detector

- ~500 tonnes UAr total
- Target needed by 2027
- With 250 kg/day and continued production after DS-20k target production would have target procured in time

→ **No additional scale up of UAr plant required!!!**

# Enter the Age of Urania



Air Products

Helium

Kinder Morgan

100%

100% - He free

15%

15% - UAr free

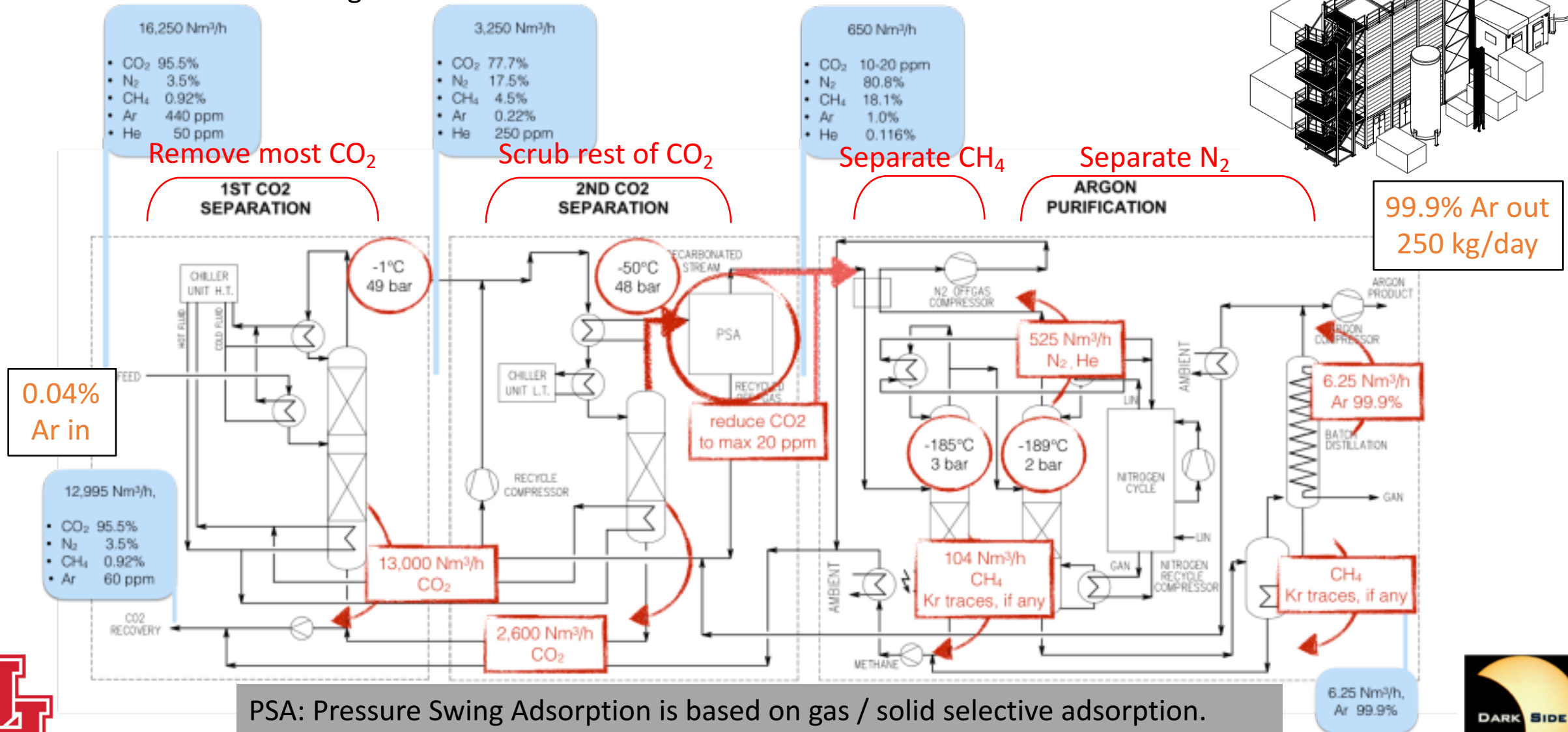
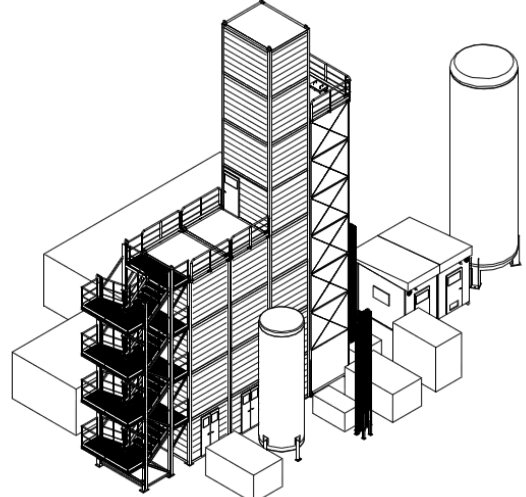
Urania

UAr  
50 tonnes extracted  
in <1 year of operation



# Urania Process Overview

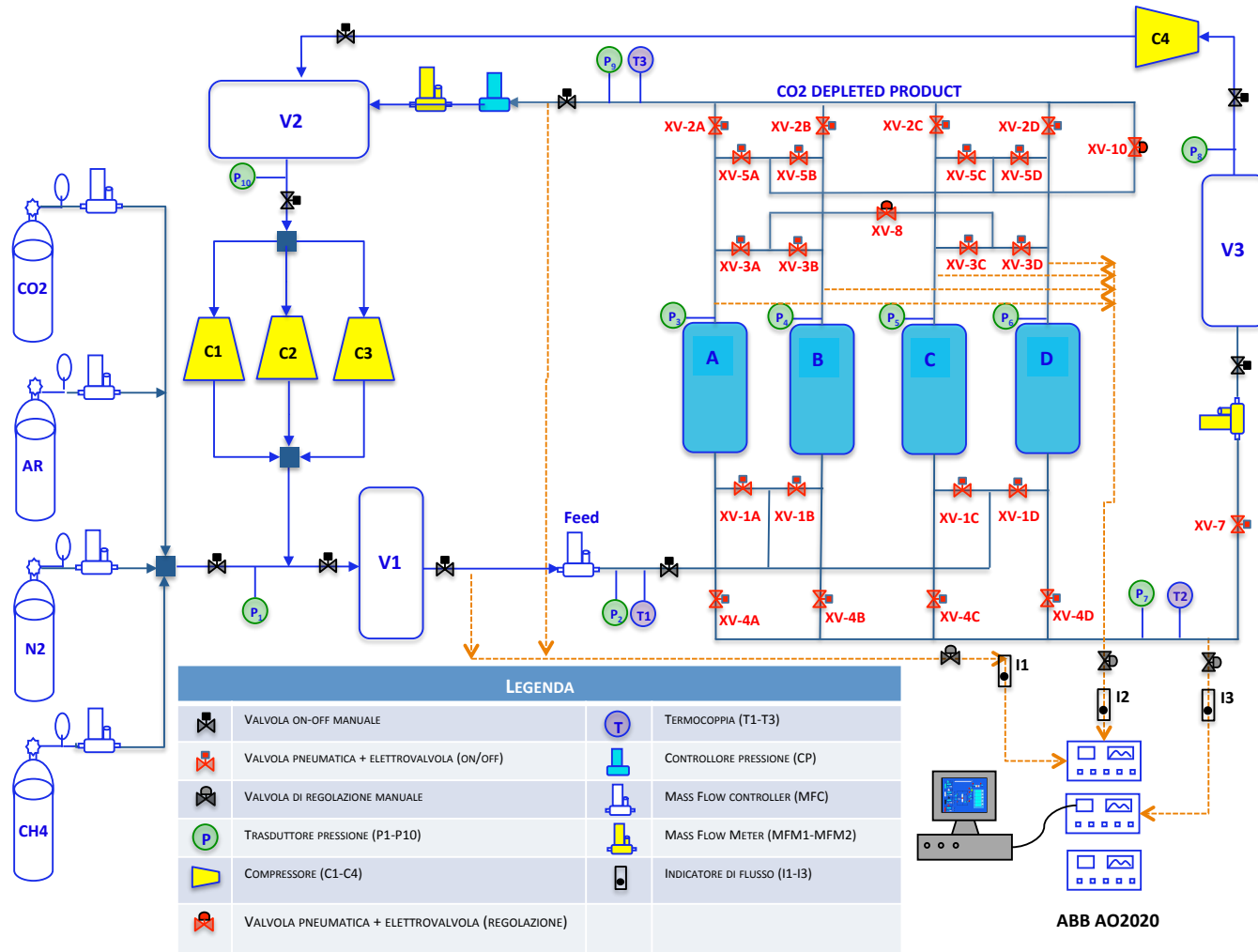
Same source as DarkSide-50 target



PSA: Pressure Swing Adsorption is based on gas / solid selective adsorption. Design cannot be properly performed without experimental validation.



# Urania PSA R&D



Pilot Plant is under test in Naples

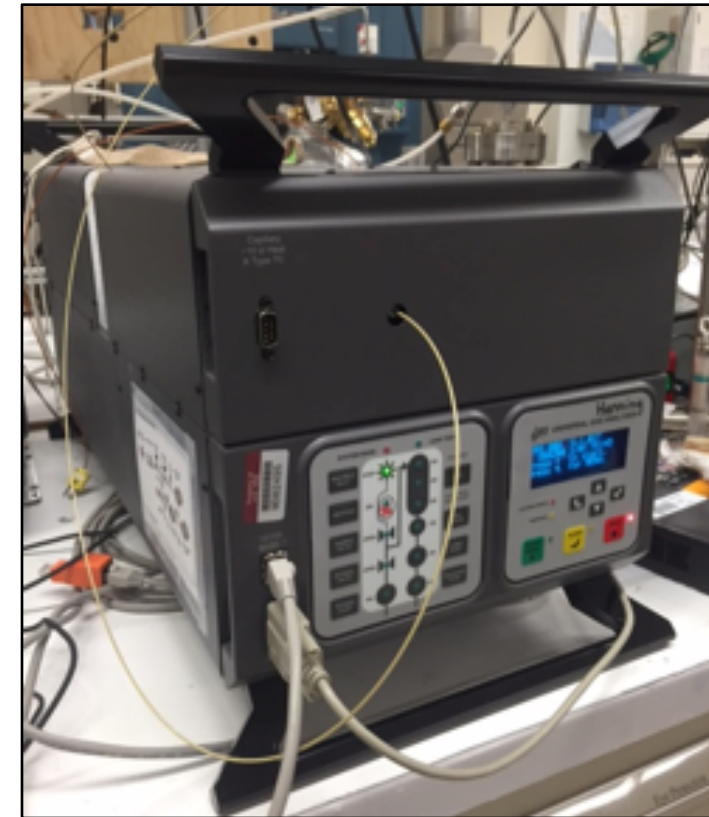
# Gas Input Stability Monitoring

- Monitor the long term stability of the input gas composition at Doe Canyon
  - Does the helium composition fluctuate?
  - Is there something we are missing?
- Critical to the long term operational stability
- PNNL developed method to measure gas composition over long periods of time (weeks to months)
  - Autonomous and remotely accessible UGA

## Long term run taken last year

- **Residual oxygen in DS-50 UAr is from air infiltration**
- Precision gas analysis of CO<sub>2</sub>
  - O<sub>2</sub> = 6.7 ppm
  - Ar = 427 ppm
- Ar:O<sub>2</sub> ratio in air 0.045
- AAr concentration in the UAr:
  - $(6.7\text{ppm} * 0.045)/427\text{ppm} = 7.0\text{e-}4$
- <sup>39</sup>Ar rate in UAr due to AAr:
  - $1 \text{ Bq/kg} * 7.0\text{e-}4 = \mathbf{0.70 \text{ mBq/kg}}$
- Potential <sup>39</sup>Ar reduction without air infiltration:
  - DS-50 <sup>39</sup>Ar rate = 0.73 mBq/kg
  - **Residual after air infiltration correction = 30 μBq/kg**
  - **33,000 time lower than AAr!**

LOGAN (LONg-term Gas ANalyzer)



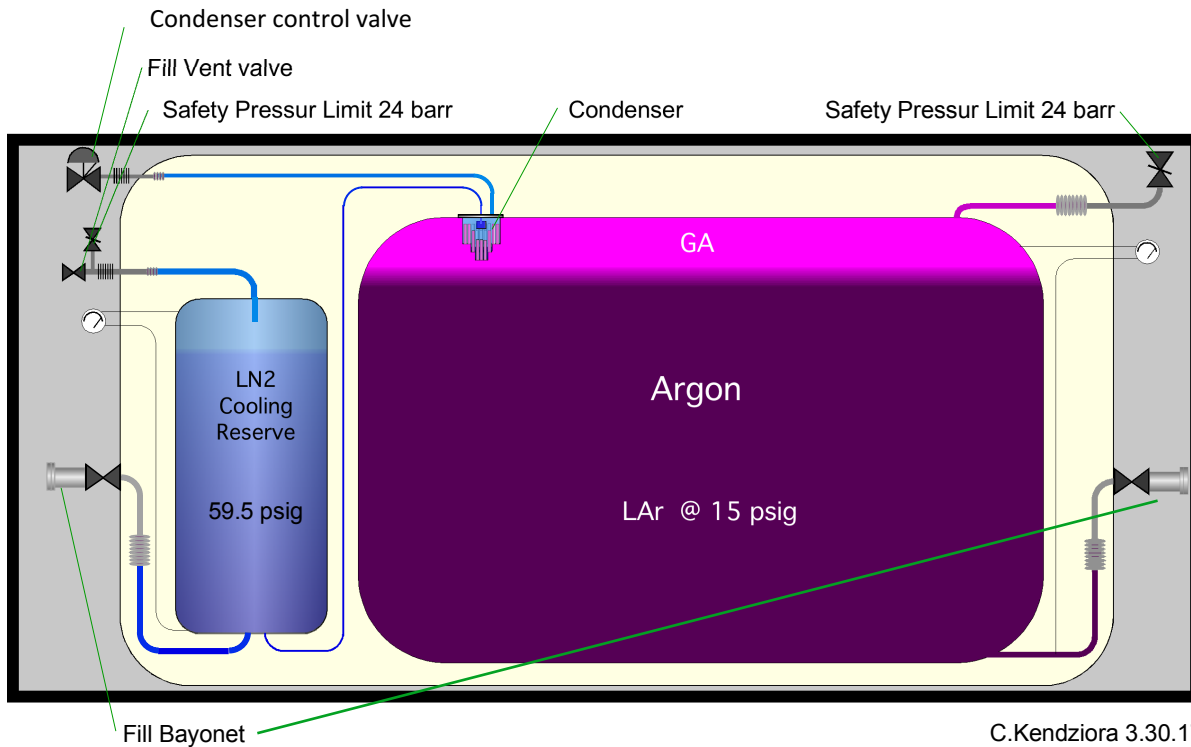
Funded by: PNNL Lab Directed R&D



# UAr Shipping and Storage

Custom designed cryogenic shipping vessel (Wessington Cryogenics)

- LN<sub>2</sub> fed UAr condenser (UCLA)
- UAr-pressure dependent valve controls cooling (UCLA)



## Open Questions

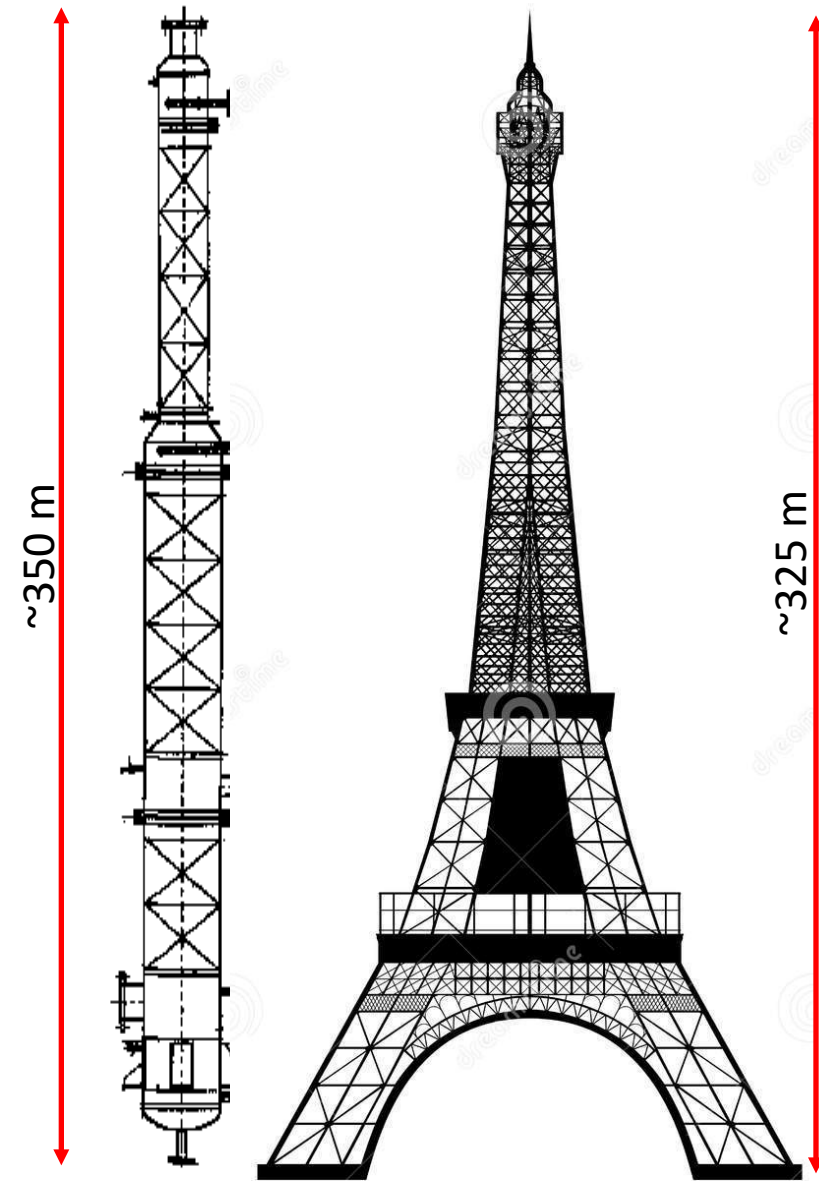
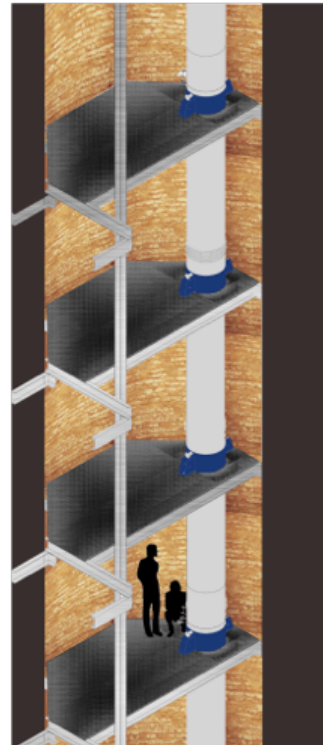
- What is the cosmogenic <sup>39</sup>Ar production rates at various altitudes, and how does this affect shipping and storage of UAr?
- What is the maximum time allowed at Doe Canyon elevation?
- Is underground storage needed?
- How do we store the argon for long terms, and what are the associated costs?

In progress:

- Comparison of cosmogenic activation codes and our own analytical estimate for <sup>39</sup>Ar production
- Measurement of <sup>39</sup>Ar activation rate by beam measurement

# DS-20k UAr Purification: Aria

- DarkSide project
- Final argon purification for DS-20k
- Capable of isotope separation through cryogenic distillation
- Can further deplete UAr of  $^{39}\text{Ar}$
- Located in coal mine shaft in Sardinia, Italy
- Funded by INFN, NSF, RAS, Carbosulcis and Princeton University
- Contributions by CERN for leak testing



Renshaw, UCLA DM 18

# Aria: Path from Fabrication to Production

**CERN (Geneva):** Global leak check test of the fully assembled modules



**Polaris (Misinto):** Production site, individual parts leak check test and SI installation



**CarboSulcis (Sardinia):** Final destination: Seruci 0 & 1 installation, testing & operation





# Status: Full Tower – 30 Modules

- First 3 modules

- Column module #1 built 2016
- Top module (condenser) built 2016
- Bottom module (re-boiler) built 2016

- Column completion

- Of remaining 27 column modules:
  - 4 modules (#28-25) built end-2016
  - 4 modules (#24-21) built 2017

- All 11 modules

- Successfully leak-tested at CERN

## Seruci-0

Top + Column-1 + Bottom

- On surface
- No module inter-connection via flanges
- Interconnections with welding (need adapting mechanical pieces)

Additional advantage:

- *Will serve to validate and adjust Seruci-1 welding scheme*



# Summary

- DarkSide-50 successfully produced 157 kg argon target with 1400x less  $^{39}\text{Ar}$  than atmospheric argon
  - Challenges to DarkSide-50 target production are understood (minor contaminations)
- Residual  $^{39}\text{Ar}$  in DarkSide-50 target likely from an air infiltration
  - intrinsic  $^{39}\text{Ar}$  in UAr < DarkSide-50 target ←
- Plans for producing and purifying 50 tons of UAr for DarkSide-20k are firmly in place
- Further reduction of  $^{39}\text{Ar}$  possible through cryogenic distillation with Aria



Thank you!



Photo by Y.Suvorov



# Agreement with Kinder Morgan (KM)

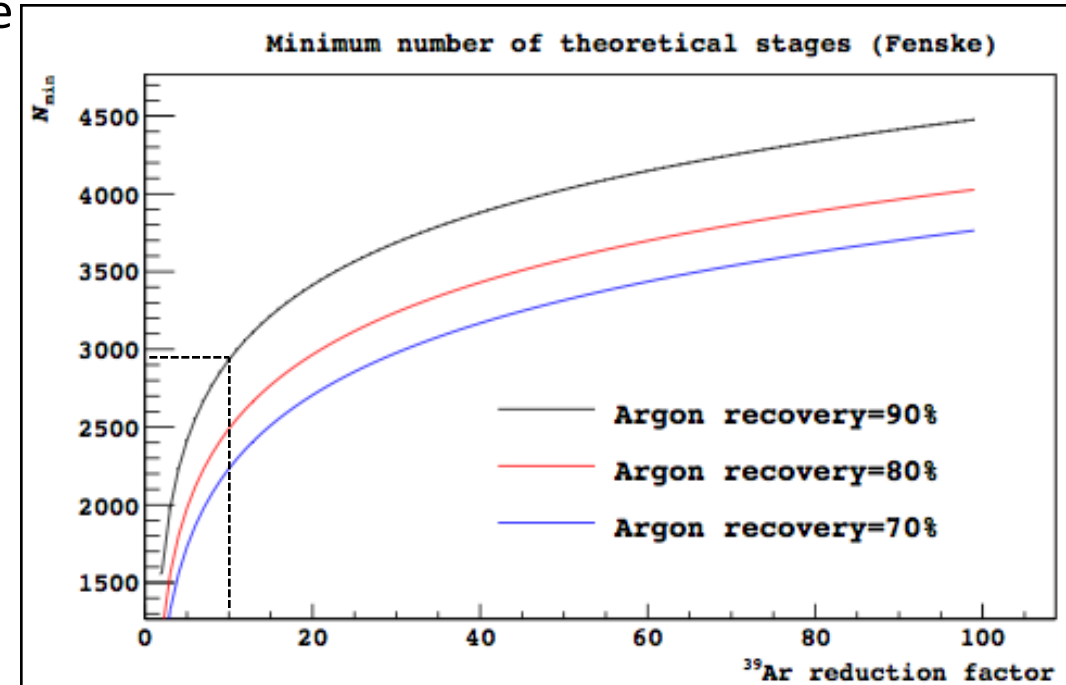
- Currently spans from January 1, 2017 through December 31, 2019, with option for renewal
- Agreement grants access to KM side stream of up to 5,500 Mscf to procure up to 50 tonnes of underground argon.
- Agreement grants permission to install the “Argon Extraction Plant” on an area of 16.0 x 19.2 square meters.
- Gas flow at outlet must stay within  $\pm 3\%$  of inlet gas flow.

\*\*\*\*None of this is possible without the incredible support from Kinder Morgan and all of their staff at the Doe Canyon Facility\*\*\*\*



# Aria Principle of Separation

- Based on difference in volatility between molecules of the same compound containing different isotopes of the same element
- Fieschi-Terzi model gives the ratio between the vapor pressure of different isotopes
- Model tested on  $p(^{36}\text{Ar})/p(^{40}\text{Ar})$ , compared with experimental data from Boato-Scoles
- Model then extended to  $p(^{39}\text{Ar})/p(^{40}\text{Ar})$
- Estimated the number of theoretical equilibrium stages with Fenske equation



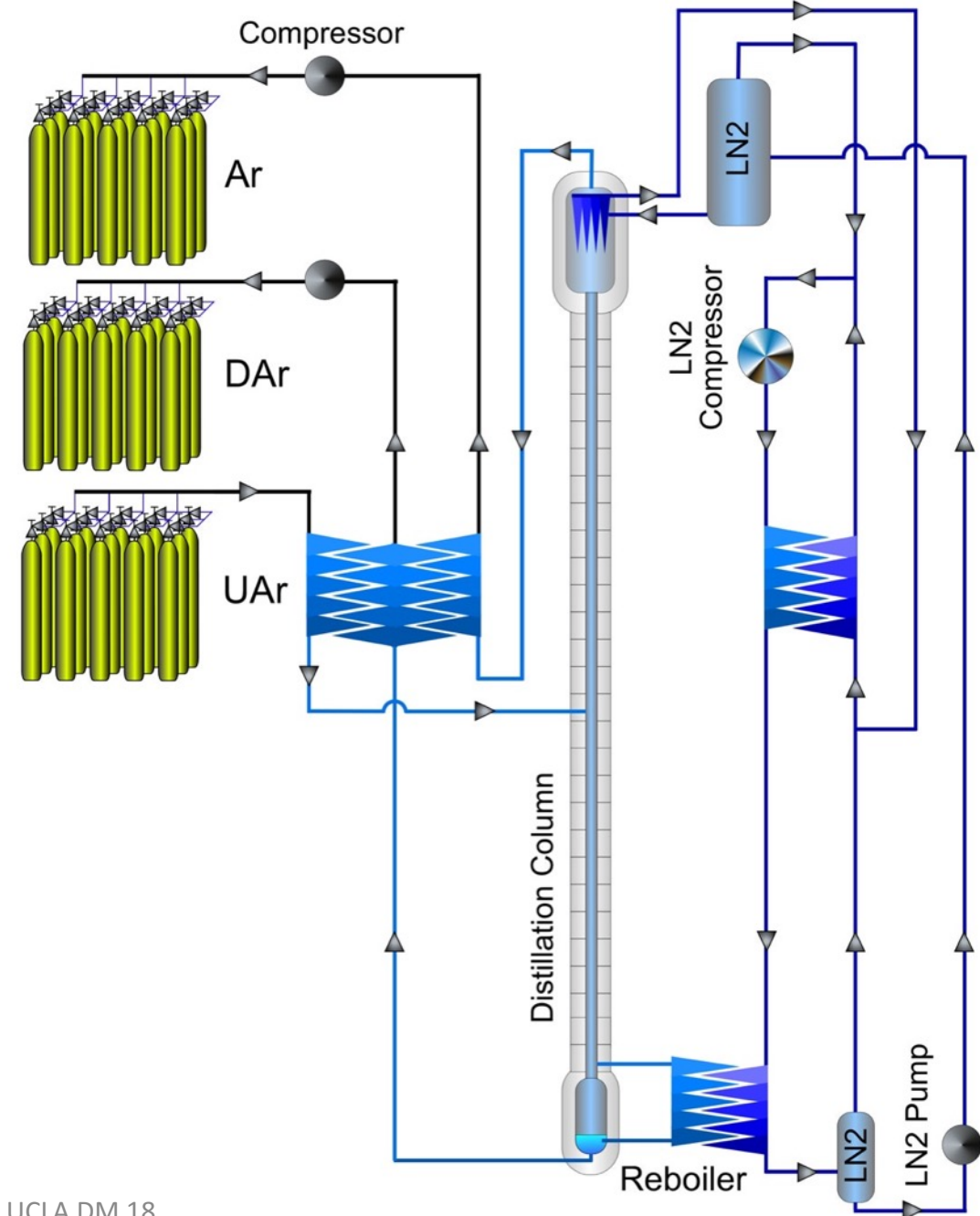
Thousands of equilibrium stages are needed

Renshaw, UCLA DM 18



# Aria Column Overview

- 28 Modules: 12 m each
- 1 Condenser module: 7 m
- 1 Reboiler module: 5m
- Total height: 348 m
- Outer Diameter of the column: 323.8 mm
- Inner Diameter of the column: 317.8 mm
- Outer Diameter of cold box: 711.2 mm
- Packing: CY from Sulzer
- Number of theoretical stages: 2870





# Aria: Beyond Argon

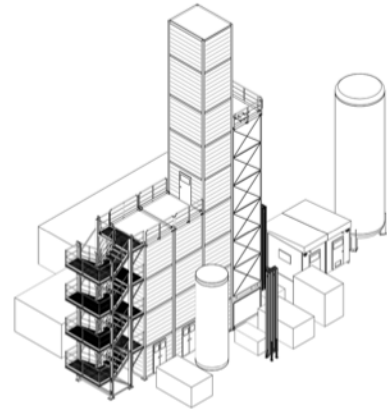
## Isotope Market Study Summary

Isotopo	2010	2011	2012	2017	CAGR 2012-17
C-13	46.963	50.316	54.241	95.791	12.05
D2	34.052	36.416	39.091	72.765	13.23
O-18	19.531	21.197	23.013	38.593	10.89
N-15	7.325	7.869	8.323	13.66	10.42
Altri	1.893	2.029	2.165	3.228	8.32
Totale	109.764	117.827	126.833	224.037	12.05



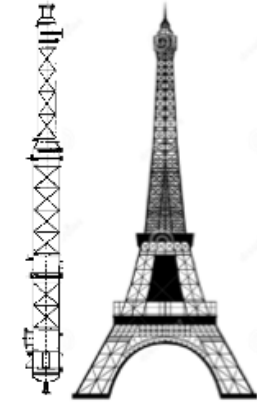
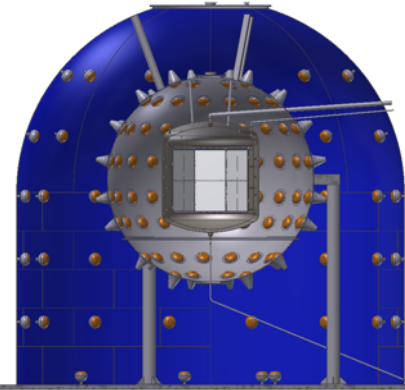


# Boarder Impact and Transfer Technology

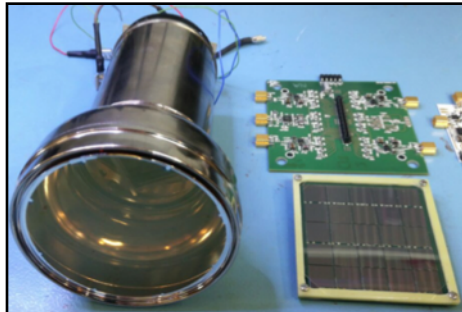


Urania  
Cortez, Colorado

DarkSide-20k  
LNGS



Aria  
Seruci,  
Sardegna



NOA  
LNGS

3D $\pi$



EB Welding  
Ortona - Italy



# Applications

- SiPM - **NOA + 3D $\pi$** 
  - PET
  - LiDAR
- Electron-Beam Welding (EB) - **NOC**
  - Reactor
  - SubMarine
- Urania:
  - TOF-PET - **3D $\pi$**
- Aria:
  - Tracer PET - **3D $\pi$**
  - Breath Test
  - <sup>15</sup>N New Generation Nuclear Power Plants

