



Pacific Northwest
NATIONAL LABORATORY

*Proudly Operated by **Battelle** Since 1965*

Low Radioactivity Argon for Rare Event Searches

HENNING O. BACK

Pacific Northwest National Laboratory

TAUP2017, Sudbury, ON, Canada

Terrestrial argon and ^{39}Ar sources

As I understand it ← it's more complicated



Pacific Northwest
NATIONAL LABORATORY

Proudly Operated by **Battelle** Since 1965

▶ ^{40}Ar comes from ^{40}K decay

▶ Atmosphere

- ^{39}Ar produced by cosmic rays
- ^{39}Ar concentration = $8 \times 10^{-16} \text{ }^{39}\text{Ar}/^{40}\text{Ar}$

▶ Crust

- No cosmic rays
- ^{39}Ar produced underground

▶ Mantle

- Very low U and Th
- Lowest ^{39}Ar levels

Atmospheric isotopic abundance

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

^{36}Ar – 0.334%

^{38}Ar – 0.063%

^{40}Ar – 99.604%

Solar system isotopic abundance

K. Lodders, Astrophys. J. 591 (2003) 1220-1247

^{36}Ar – 84.59%

^{38}Ar – 15.38%

^{40}Ar – 246 ppm

Terrestrial argon and ^{39}Ar sources

As I understand it ← it's more complicated



Pacific Northwest
NATIONAL LABORATORY

Proudly Operated by **Battelle** Since 1965

▶ ^{40}Ar comes from ^{40}K decay

▶ Atmosphere

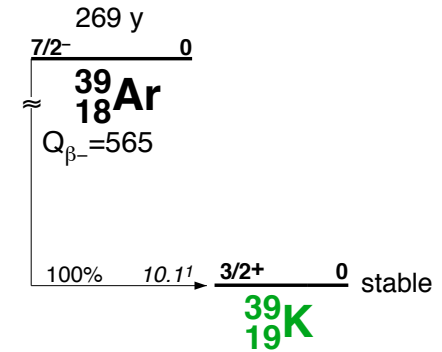
- ^{39}Ar produced by cosmic rays
- ^{39}Ar concentration = $8 \times 10^{-16} \text{ }^{39}\text{Ar}/^{40}\text{Ar}$

▶ Crust

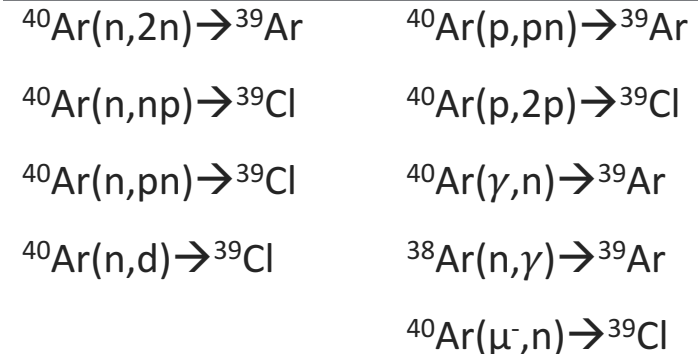
- No cosmic rays
- ^{39}Ar produced underground

▶ Mantle

- Very low U and Th
- Lowest ^{39}Ar levels



^{39}Ar production reactions



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

Terrestrial argon and ^{39}Ar sources

As I understand it ← it's more complicated



Pacific Northwest
NATIONAL LABORATORY

Proudly Operated by **Battelle** Since 1965

▶ ^{40}Ar comes from ^{40}K decay

▶ Atmosphere

■ ^{39}Ar produced by cosmic rays

■ ^{39}Ar concentration = $8 \times 10^{-16} \text{ }^{39}\text{Ar}/^{40}\text{Ar}$

▶ Crust

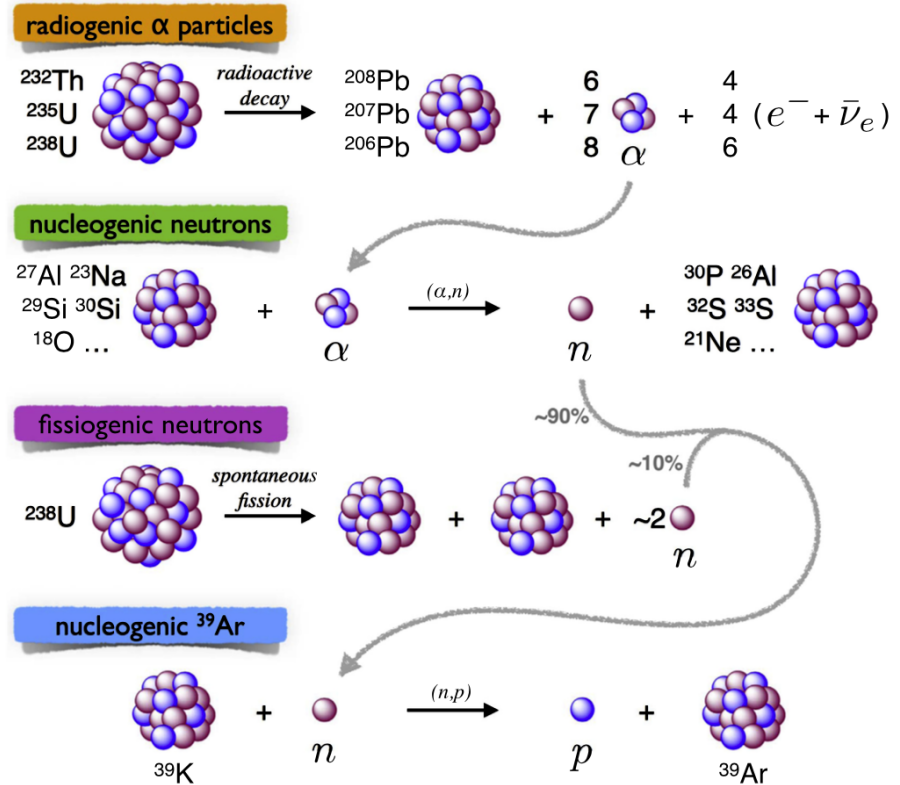
■ No cosmic rays

■ ^{39}Ar produced underground

▶ Mantle

■ Very low U and Th

■ Lowest ^{39}Ar levels



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

Terrestrial argon

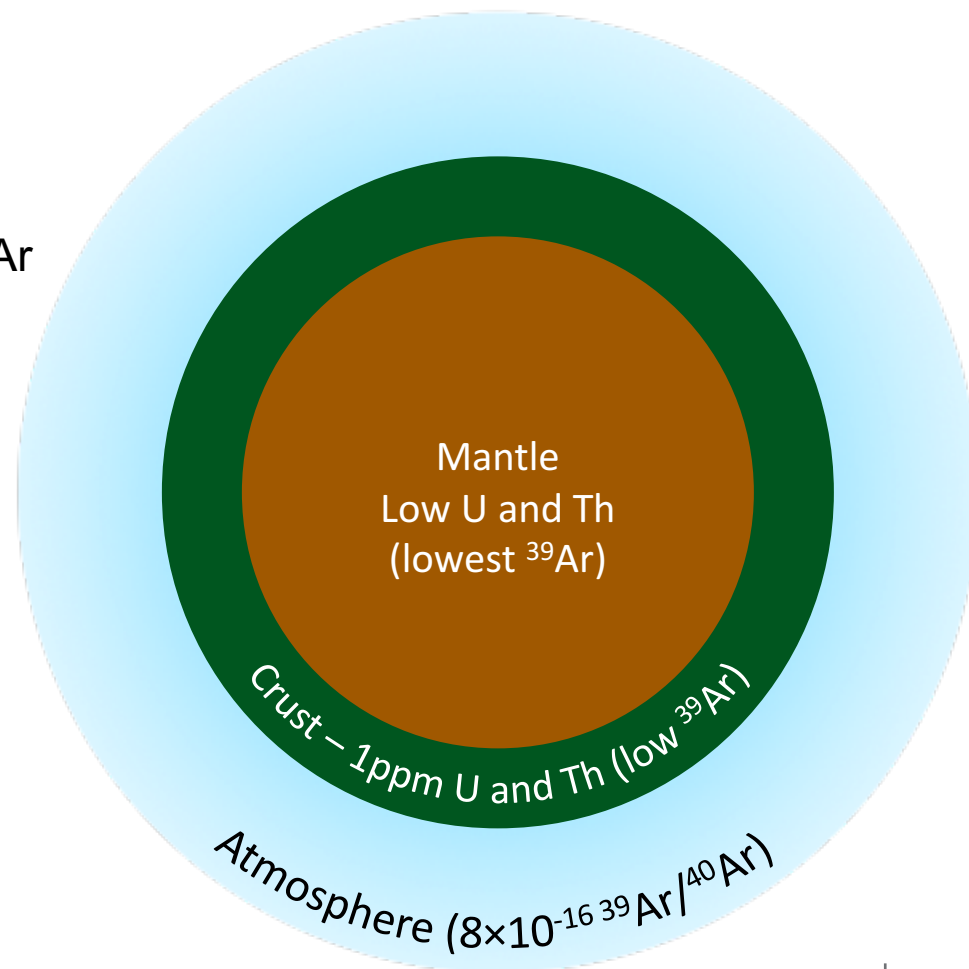
As I understand it ← it's more complicated



Pacific Northwest
NATIONAL LABORATORY

Proudly Operated by **Battelle** Since 1965

- ▶ ^{40}Ar comes from ^{40}K decay
- ▶ Atmosphere
 - ^{39}Ar produced by cosmic rays
 - ^{39}Ar concentration = $8 \times 10^{-16} \text{ }^{39}\text{Ar}/^{40}\text{Ar}$
- ▶ Crust
 - No cosmic rays
 - ^{39}Ar produced underground
- ▶ Mantle
 - Very low U and Th
 - Lowest ^{39}Ar levels

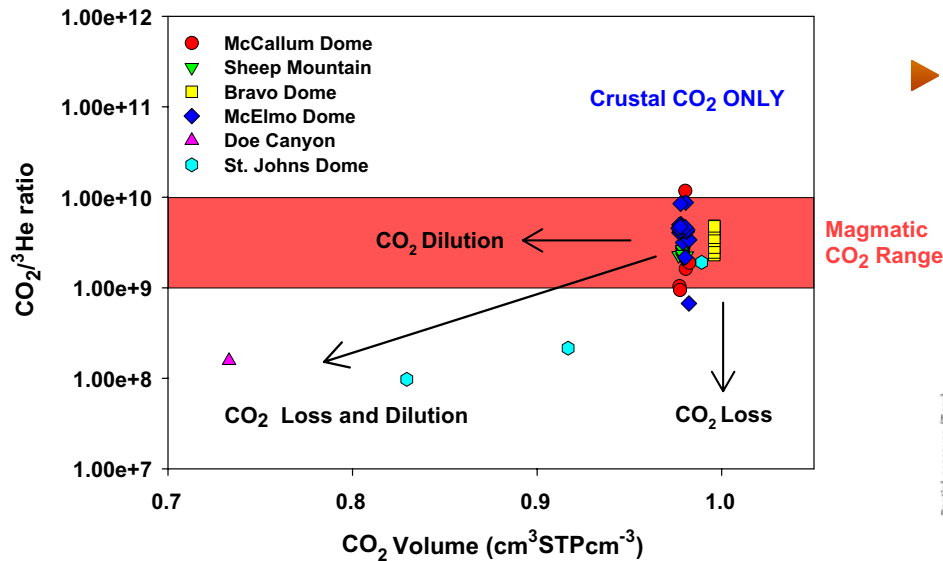


A low-radioactivity UAr source CO₂ well in SW Colorado

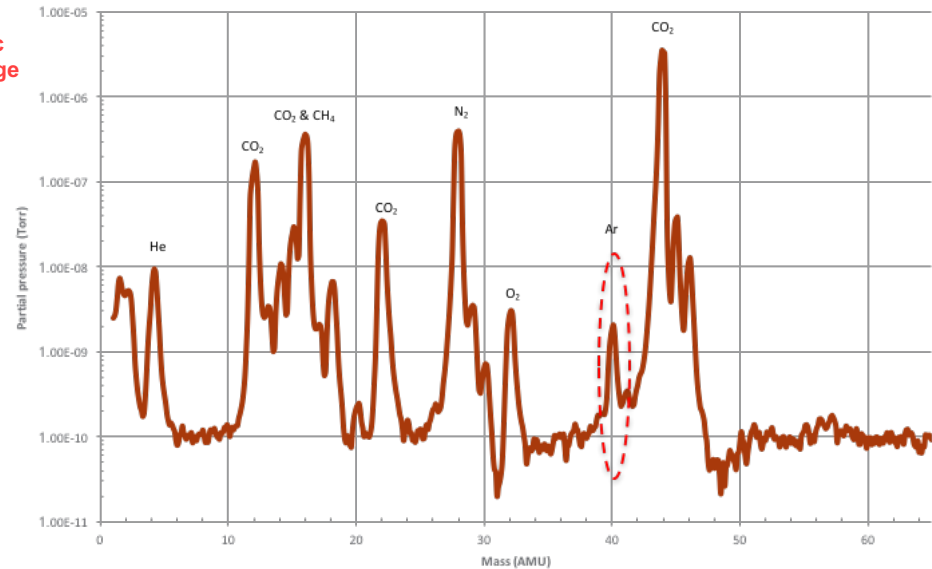
- ▶ Geological formations trap gases underground
- ▶ CO₂ in US Southwest is magmatic-like



One of many parameters that define magmatic
(*Geochim. et Cosmochim. Acta* 72 (2008) 1174–1198)



- ▶ CO₂ well in SW Colorado with 400ppm Ar
(*Nucl. Phys. B*, 197 (2009) 70-73)
(*Nucl. Instr. Meth. A* 587 (2008) 46-51)



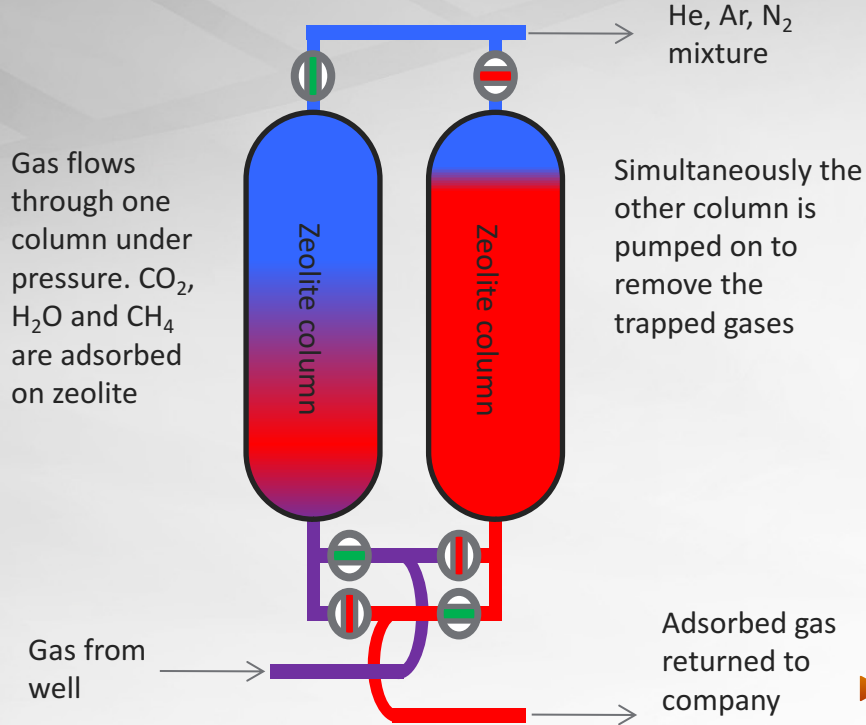
DarkSide-50 target production UAr extraction - Colorado



Pacific Northwest
NATIONAL LABORATORY

Proudly Operated by **Battelle** Since 1965

Vacuum-Pressure Swing Adsorption



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



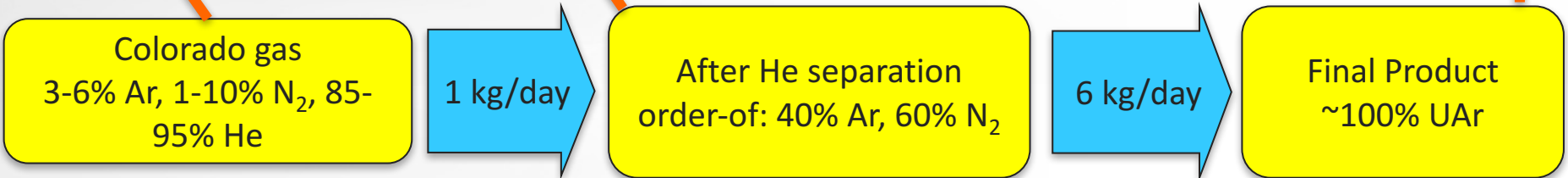
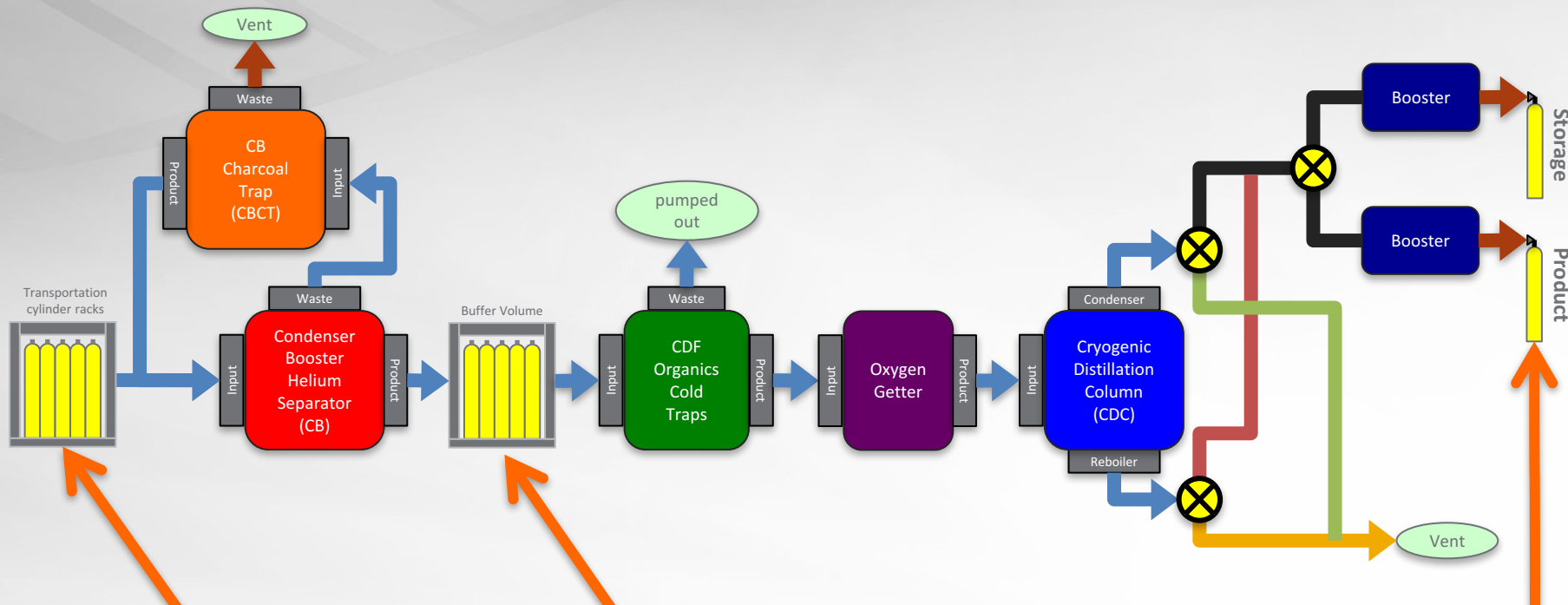
- ▶ **Approximate product composition:**
 - He – 85-95%
 - Ar – 3-6%
 - N₂ – 1-10%
- ▶ **Average production rate:**
 - 140 g/day

Contaminants trapped in VPSA zeolite

C ₃ H ₈	C ₇ H ₁₄	C ₇ H ₁₆
C ₅ H ₁₀ O	C ₆ H ₁₃ I	C ₆ H ₁₂ O
C ₅ H ₁₂	C ₆ H ₁₃ I	C ₅ H ₈ O ₂
C ₆ H ₁₄	C ₇ H ₁₆	C ₈ H ₁₆
C ₅ H ₁₀	C ₇ H ₁₆	C ₈ H ₁₆
C ₅ H ₁₀ O	C ₆ H ₁₂ O	C ₈ H ₁₈
C ₅ H ₁₀ O	C ₆ H ₁₂ O	C ₈ H ₁₈
C ₆ H ₁₄	C ₇ H ₁₆	C ₆ H ₁₀ O ₂
C ₆ H ₁₂ O	C ₆ H ₆	C ₈ H ₁₈
C ₆ H ₁₂	C ₆ H ₆	C ₉ H ₂₀

DarkSide-50 target production

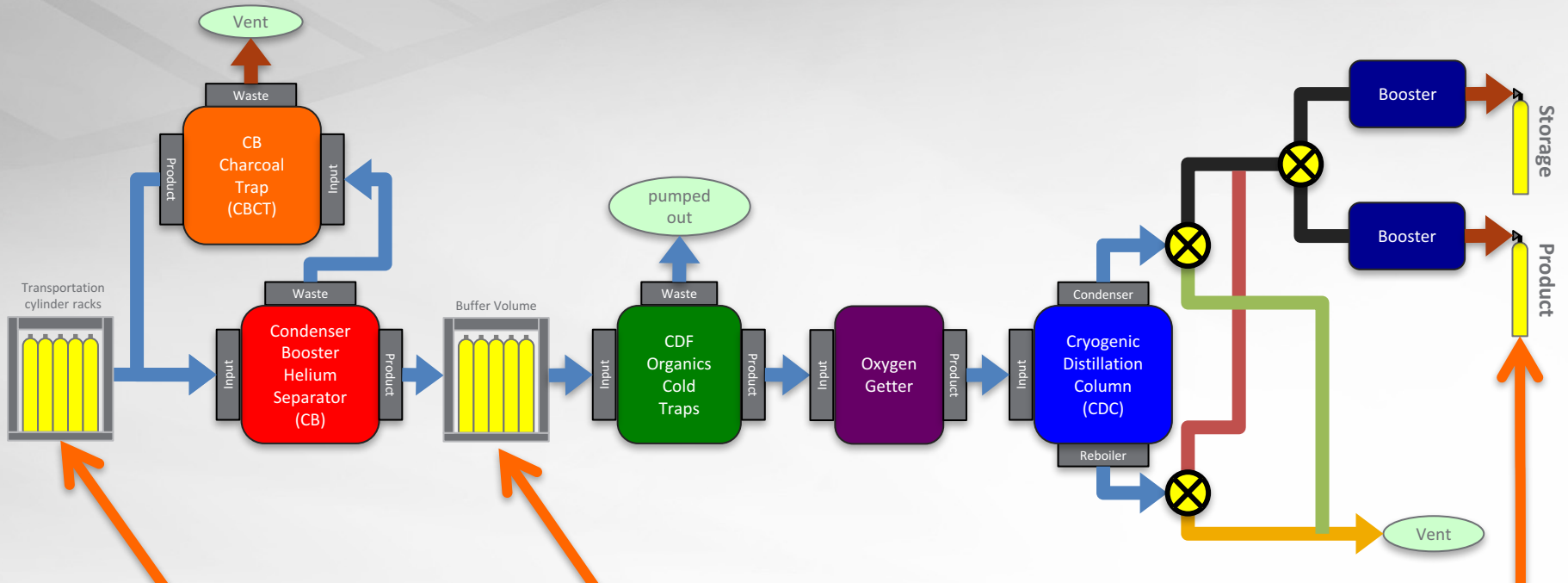
UAr purification – Fermilab, Illinois, USA



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

DarkSide-50 target production

UAr purification – Fermilab, Illinois, USA



Colorado gas
3-6% Ar, 1-10% N₂, 85-95% He

1 kg/day

After He separation
order-of: 40% Ar, 60% N₂

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

Cryogenic distillation column



Fall 2010

The end DarkSide-50 UAr production



Pacific Northwest
NATIONAL LABORATORY

Proudly Operated by **Battelle** Since 1965

Vacuum-pressure swing adsorption



07/29/2011 AM 10:31

March 2016



Jan. 2016

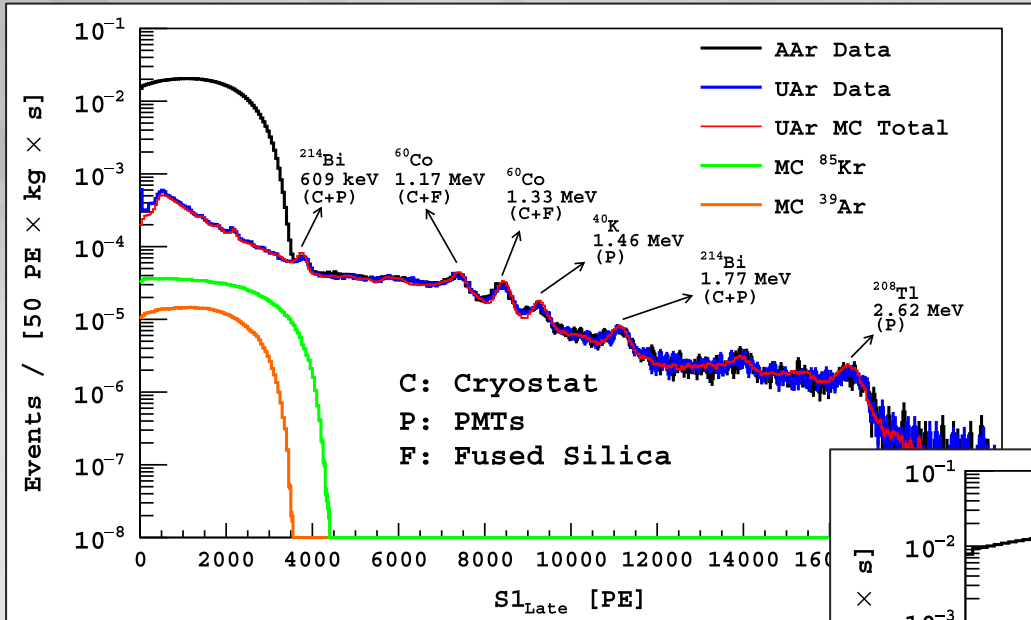
Underground argon in DarkSide-50

Phys. Rev. D 93, 081101(R) (2016)



Pacific Northwest
NATIONAL LABORATORY

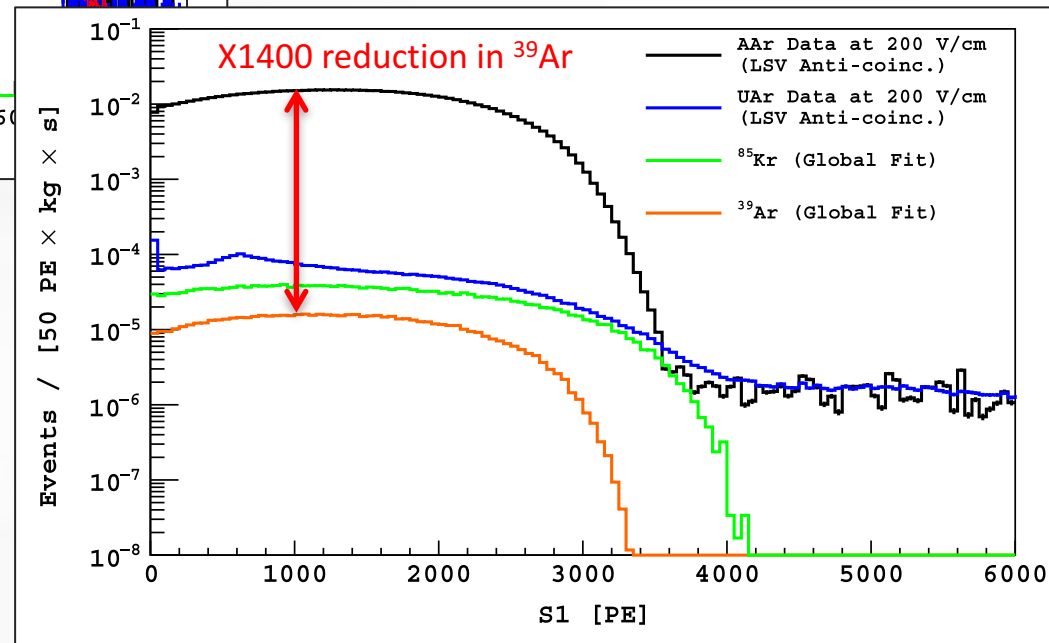
Proudly Operated by **Battelle** Since 1965



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

^{39}Ar specific activity = 0.73 mBq/Kg

Residual ^{39}Ar is likely from air infiltration during production



DarkSide-20k target

Obtaining 50 tons of UAr

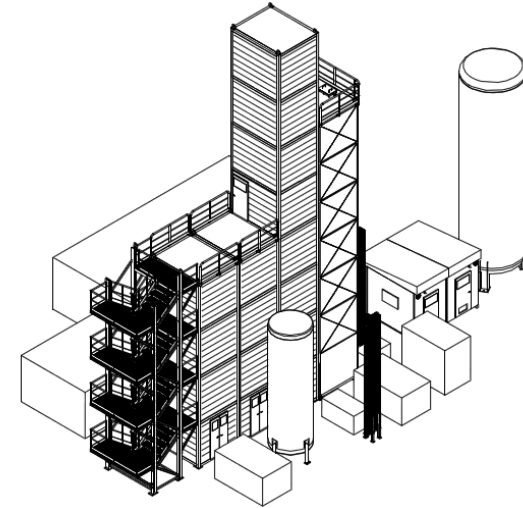


Pacific Northwest
NATIONAL LABORATORY

Proudly Operated by **Battelle** Since 1965

► Urania (planned)

- Extracts argon from CO₂
- Same source as DarkSide-50 target
- Production:
 - 100 kg/day
 - 99.9% pure

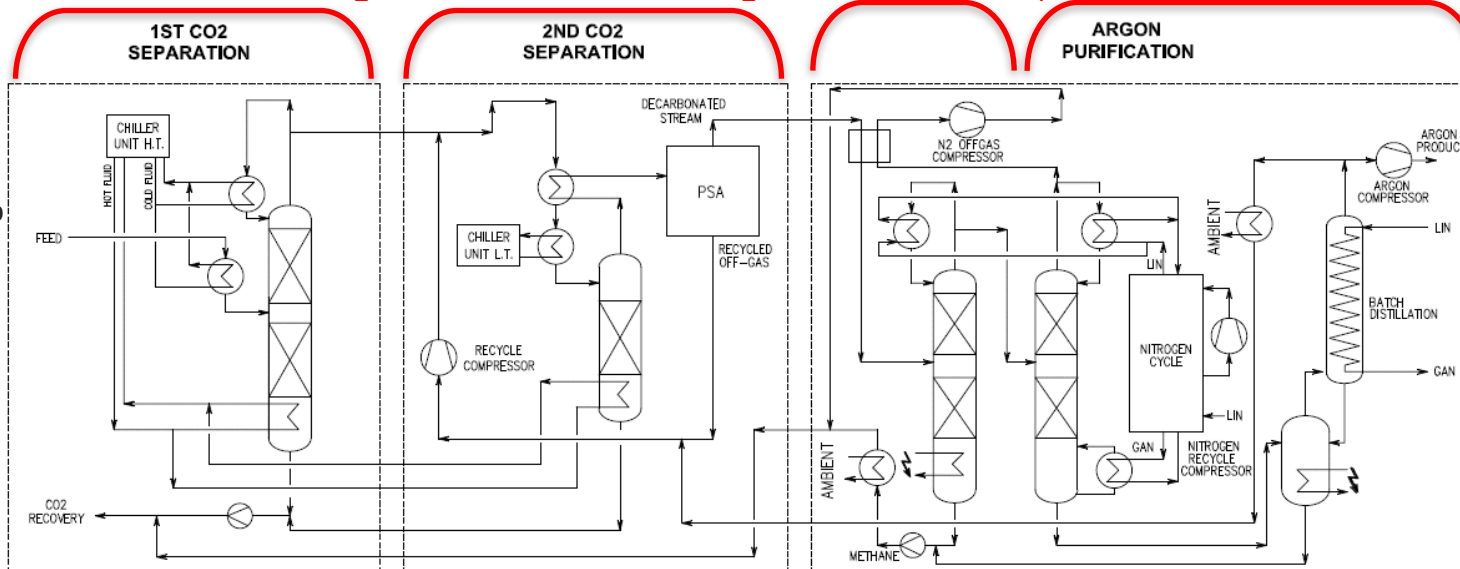


Remove most CO₂

Scrub rest of CO₂

Separate CH₄

Separate N₂



99.9%
Ar out

DarkSide-20k target

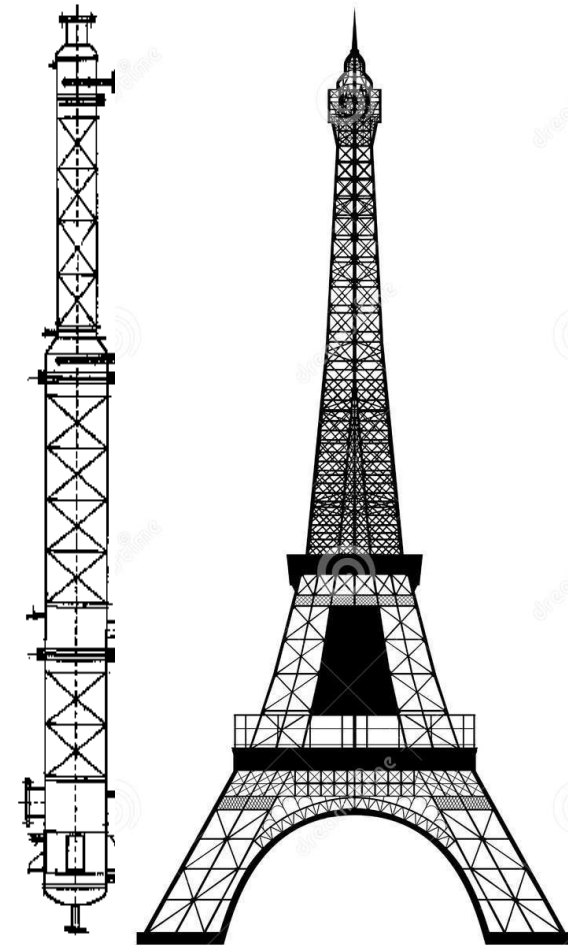
Obtaining 50 tons of UAr

► **Aria** (*under construction*)

- 350m distillation column
- Capable of isotope separation through distillation
 - Noble gas isotopes have different vapor pressure (i.e., non-zero relative volatilities)
- Final argon purification for DS-20k
- Can further deplete UAr of ^{39}Ar
- Located in coal mine shaft in Sardinia, Italy



TAUP2017





Conclusions / Highlights

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