

# Study of cosmogenic activation above ground of Ar for DarkSide-20k

- GADMC and DarkSide-20k
- Methodology: flux, cross section
- Isotopes
- Production rates
- Activity and counting rates



**Susana Cebrián** [scebrian@unizar.es](mailto:scebrian@unizar.es)  
on behalf of the DarkSide-20k collaboration

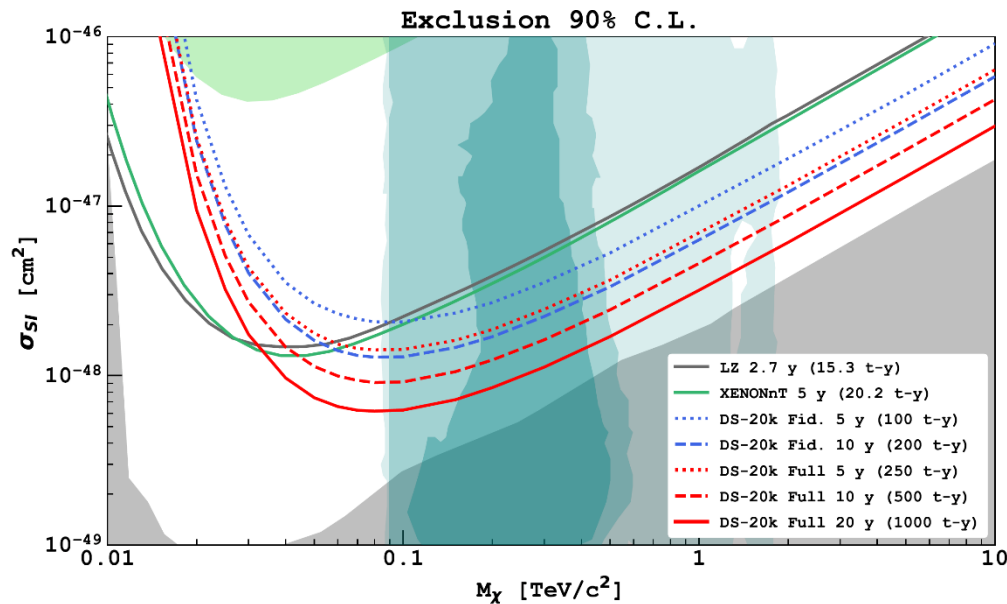
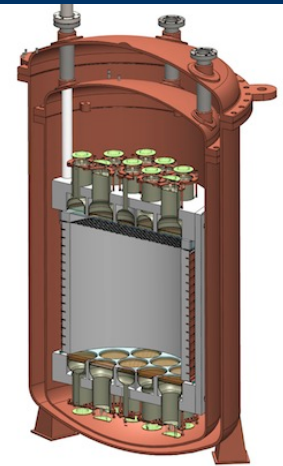
20<sup>th</sup> September 2023



# GADMC

## Global Argon Dark Matter Collaboration (GADMC):

joining ArDM, DarkSide-50, DEAP-3600, and MiniCLEAN to operate LAr TPCs reaching the scale of **multi-ton** detectors to push the sensitivity for WIMP detection down to the neutrino fog



**DarkSide-20k:** 20 t (fiducial) at Gran Sasso, starting in 2027

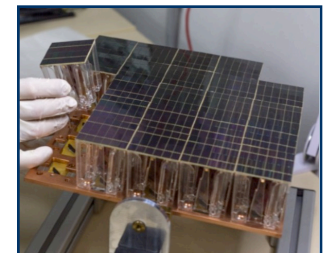
**DarkSide-LowMass:** 1 t (fiducial), S2 only to lower threshold → more ultrapure Ar needed

P. Agnes et al, Phys. Rev. D 107 (2023) 112006

**ARGO:** ~360 t, at SNOLAB

### Program:

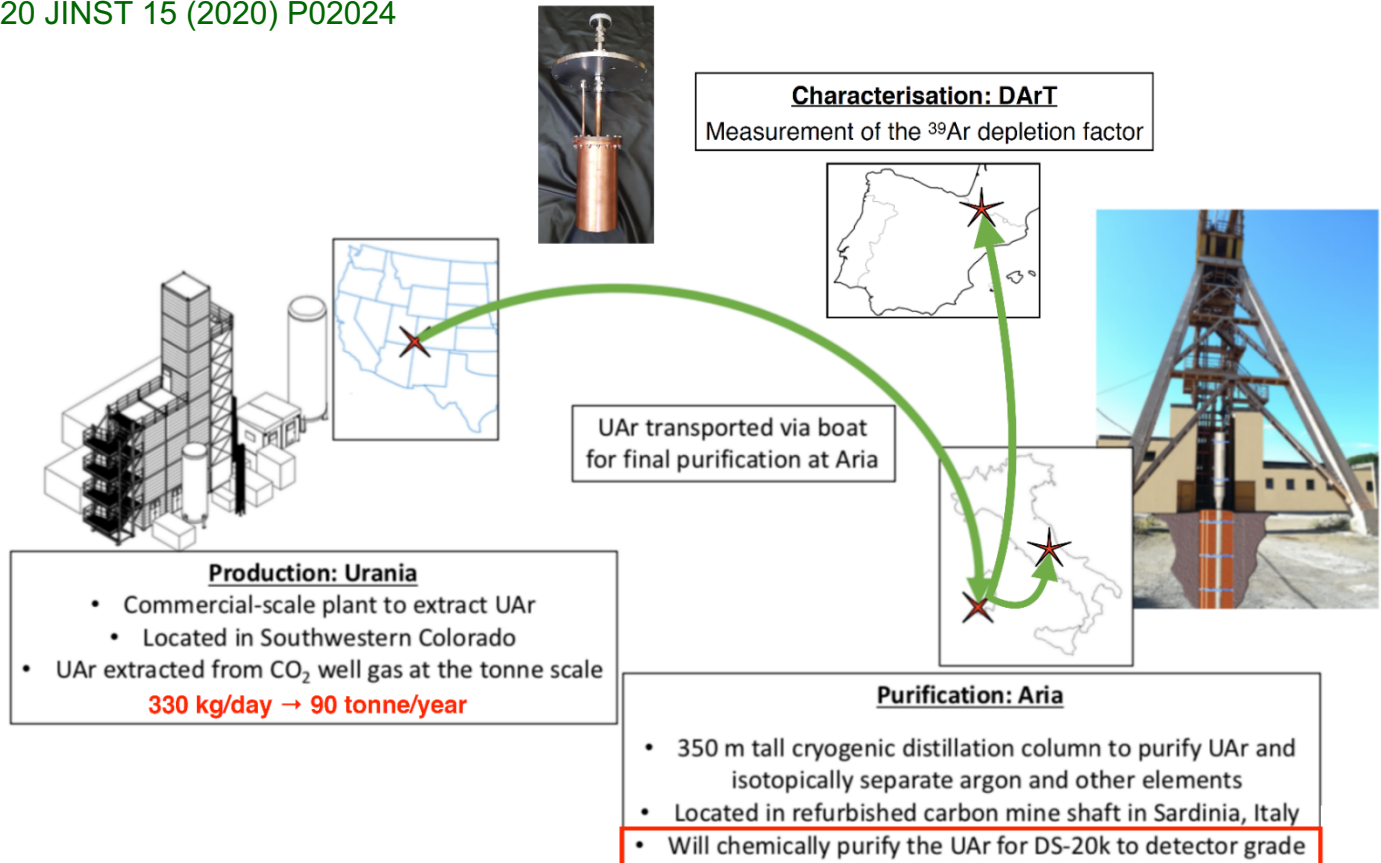
- Development of custom-designed **SiPMs**
- Design of **active vetos** to reject backgrounds
- Procurement of large amounts of radiopure **underground Ar (UAr)**



# GADMC: UAr journey

## Procurement of low-radioactivity UAr

- **Urania:** extraction from CO<sub>2</sub> wells in Colorado (as DarkSide-50)
- **Aria:** purification in a cryogenic distillation column in Sardinia  
P. Agnes et al, Eur. Phys. J. C 81 (2021) 359; Eur. Phys. J. C 83 (2023) 453
- **DArT:** quantification of <sup>39</sup>Ar in a chamber in the ArDM detector in Canfranc  
C.E. Aalseth et al, 2020 JINST 15 (2020) P02024

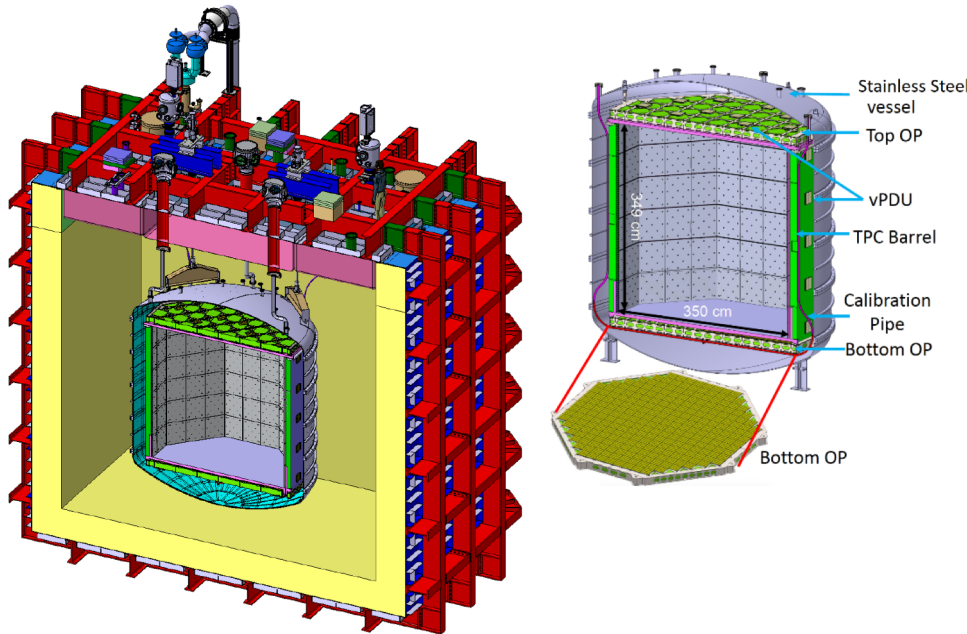


# DarkSide-20k

**Dual-phase TPC** read by **SiPMs** with 3D space reconstruction capability

Filled and surrounded by **UAr**: 99.2 t required, 51.1 t inside TPC, **20.2 t** fiducial mass

- **TPC**: Gd-PMMA vessel to moderate and capture neutrons
- **Inner veto**: stainless steel vessel
- **Outer veto**: 700 t of Atmospheric Argon, ProtoDUNE-like membrane cryostat



**Goal:  $<0.1$  events in 200 t·y**

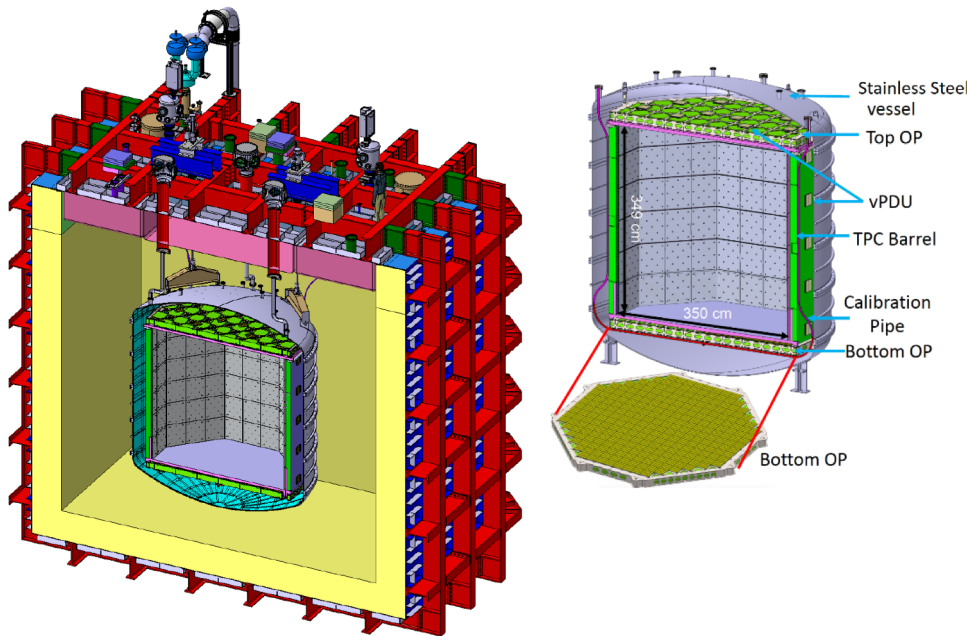
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Paolo Franchini talk, Friday morning

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➔ But **beta/gamma** background from **primordial or cosmogenic radioactivity** must be analyzed due to acceptance losses for ER+NR pile-up in TPC or accidental coincidences between TPC and Veto

# Methodology

**Goal:** to determine if **activity induced by exposure to cosmic rays** during fabrication, transport and storage of components could be an issue in order to identify limits on the surface residency time and assess the necessity of taking some steps:

- Storing materials underground
- Avoiding flights
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1. To know the **production rates**  $R$  of relevant isotopes in the targets, from
  - Scarce experimental data from irradiation/controlled exposure experiments
  - Calculations from **production cross sections** and **cosmic ray spectrum**:

$$R = N_t \int \sigma(E)\phi(E)dE$$

$N_t$  = number of target nuclei  
 $\phi$  = flux of cosmic rays  
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3. To compute the **background rate** generated by Monte Carlo simulation: G4DS

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At the Earth's surface nuclide production is dominated by **neutrons** because of the absorption of charged particles in the atmosphere

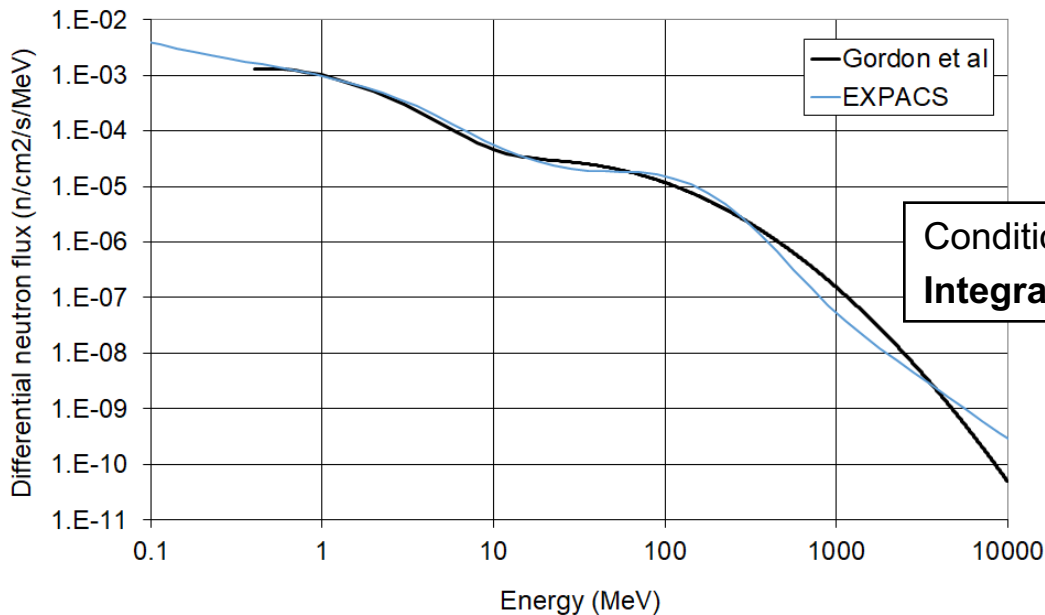
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M.S. Gordon et al, IEEE Trans. Nucl. Sci. 51 (2004) 3427



Conditions of New York City at sea level  
**Integral flux (10 MeV-10 GeV):  $3.6 \times 10^{-3} \text{ cm}^{-2} \text{ s}^{-1}$**

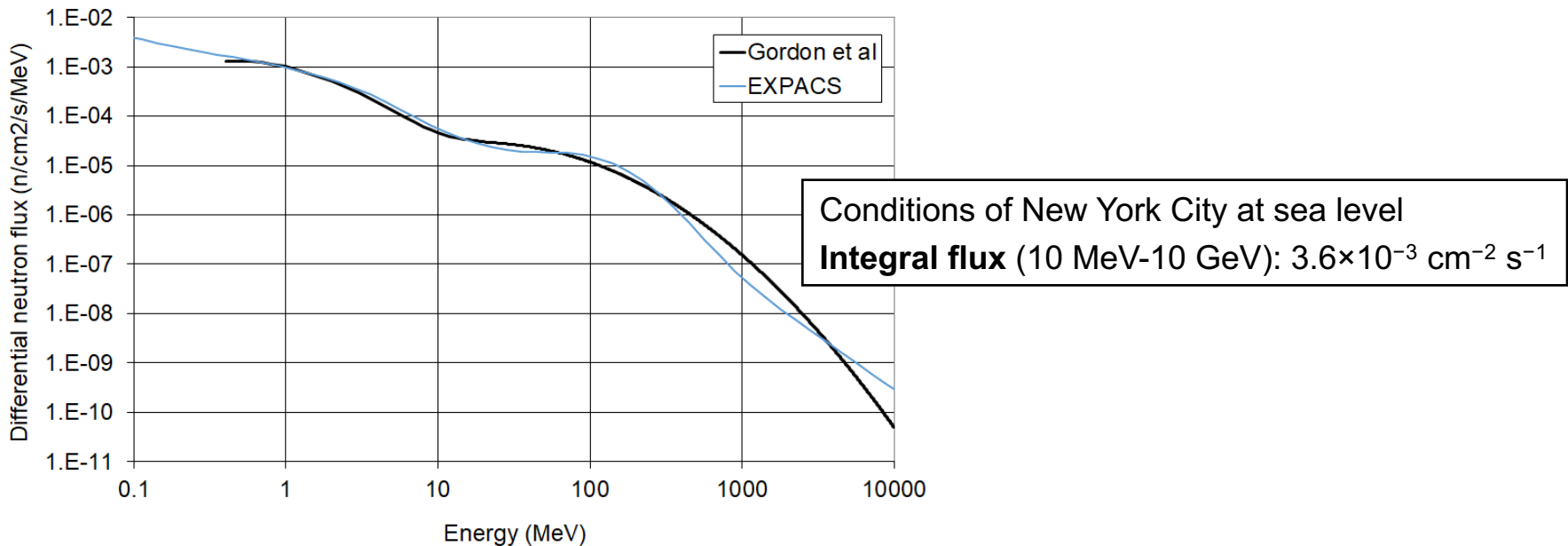
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As it depends on altitude and geomagnetic rigidity, **correction factors** must be applied to the parametrization

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**JENDL** (Japanese Evaluated Nuclear Data Library)

- Using GNASH code
- For **neutrons and protons up to 200 MeV, from 20 MeV to 3 GeV** in High Energy File

# Isotopes

- **Relevant cosmogenic products**

**$^{39}\text{Ar}$** :  $\beta^-$  emitter with  $Q=565$  keV,  $T_{1/2}=\mathbf{269}$  y mainly produced by  $^{40}\text{Ar}(n,2n)^{39}\text{Ar}$

Measured activity in

- **Atmospheric Ar**:  $\sim 1$  Bq/kg (WARP, ArDM, DEAP)
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**$^{42}\text{Ar}$** :  $\beta^-$  emitter,  $Q=599$  keV,  $T_{1/2}=\mathbf{32.9}$  y producing  $^{42}\text{K}$  ( $\beta^-$  emitter,  $Q=3525$  keV,  $T_{1/2}=12.36$  h)  $\rightarrow$  potential background for neutrinoless double beta decay

In Atm Ar: DBA:  $92^{+22}_{-46}$   $\mu\text{Bq/kg}$ , GERDA: 50-100  $\mu\text{Bq/kg}$ , DEAP  $(40.4 \pm 0.5.9)$   $\mu\text{Bq/kg}$

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**$^3\text{H}$** :  $\beta^-$  emitter with  $Q=18.6$  keV,  $T_{1/2}=\mathbf{12.3}$  y

- Quantified yields in **Ge** detectors (EDELWEISS, CDMSlite) and observed hints in **NaI(Tl)** crystals (ANAIS, COSINE)
- Purification systems for LAr should remove all non-noble radionuclides, as also assumed in **LXe**, but tritium proposed as a possible explanation for the XENON1T excess

# Production rates

- **Production rates R** (sea level): available information
  - First measurement for  $^{39}\text{Ar}$  and  $^{37}\text{Ar}$  in an **irradiation experiment** at Los Alamos (LANL) with a **neutron beam**, quantifying products with a proportional counter at PNNL + calculations at sea level from alternate mechanisms

R. Saldanha et al, Phys. Rev. C 100 (2019) 024608

Reaction	Estimated $^{39}\text{Ar}$ production rate [atoms/(kg <sub>Ar</sub> day)]	Fraction of total AAr (%)
$^{40}\text{Ar} (n, 2n)^{39}\text{Ar} + ^{40}\text{Ar}(n, d)^{39}\text{Cl}$	$759 \pm 128$	72.3
$^{40}\text{Ar} (\mu, n)^{39}\text{Cl}$	$172 \pm 26$	16.4
$^{40}\text{Ar} (\gamma, n)^{39}\text{Ar}$	$89 \pm 19$	8.5
$^{40}\text{Ar} (\gamma, p)^{39}\text{Cl}$	$23.8 \pm 8.7$	2.3
$^{40}\text{Ar} (p, 2p)^{39}\text{Cl}$	<0.1	<0.01
$^{40}\text{Ar} (p, pn)^{39}\text{Ar}$	$3.6 \pm 2.2$	0.3
$^{38}\text{Ar}(n, \gamma)^{39}\text{Ar}$	$\ll 0.1$ (UAr) $1.1 \pm 0.3$ (AAr)	- 0.1
Total	$1048 \pm 133$	100

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$^{40}\text{Ar} (n, 4n)^{37}\text{Ar}$	$51.0 \pm 7.4$
$^{40}\text{Ar} (\gamma, 3n)^{37}\text{Ar}$	$3.5 \pm 0.7$
$^{40}\text{Ar} (p, p3n)^{37}\text{Ar}$	$1.3 \pm 0.4$
$^{36}\text{Ar}(n, \gamma)^{37}\text{Ar}$	$0.9 \pm 0.3$ (UAr) $36 \pm 11$ (AAr)
$^{38}\text{Ar}(n, 2n)^{37}\text{Ar} + ^{38}\text{Ar}(\gamma, n)^{37}\text{Ar} + ^{38}\text{Ar}(p, pn)^{37}\text{Ar}$	<0.05 (UAr) $0.43 \pm 0.05$ (AAr)
Total	$56.7 \pm 7.5$ (UAr) $92 \pm 13$ (AAr)

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- Rates for  $^{37}\text{Ar}$ ,  $^{39}\text{Ar}$  and  $^{42}\text{Ar}$  from neutrons, protons and muons estimated by **GEANT4** simulation too

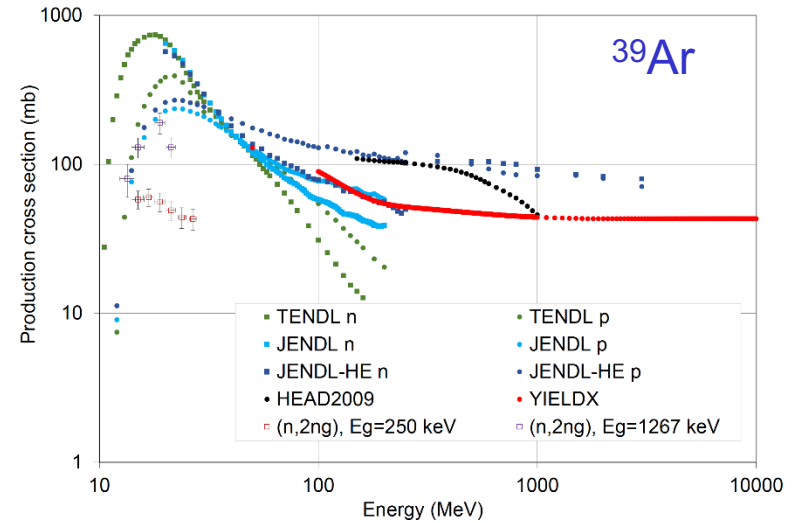
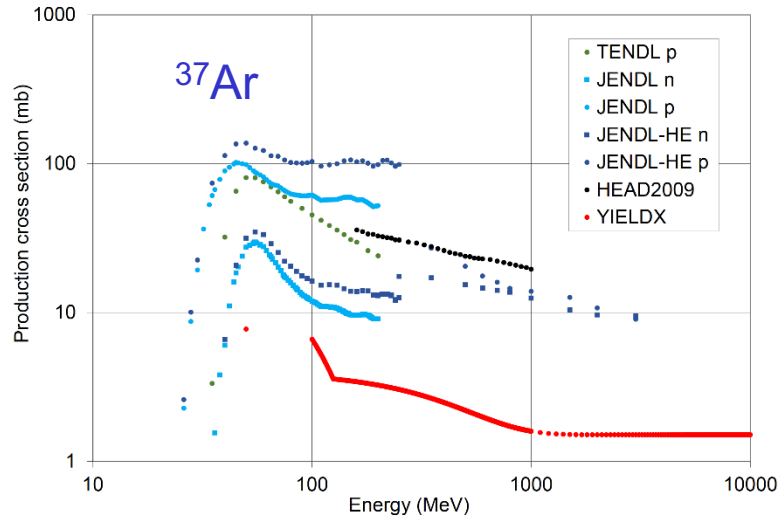
C. Zhang, D.M. Mei, Astropart. Phys. 142 (2022) 102733



# Production rates

- Production rates **R** (sea level): new calculations

$$R = N_t \int \sigma(E)\phi(E)dE$$

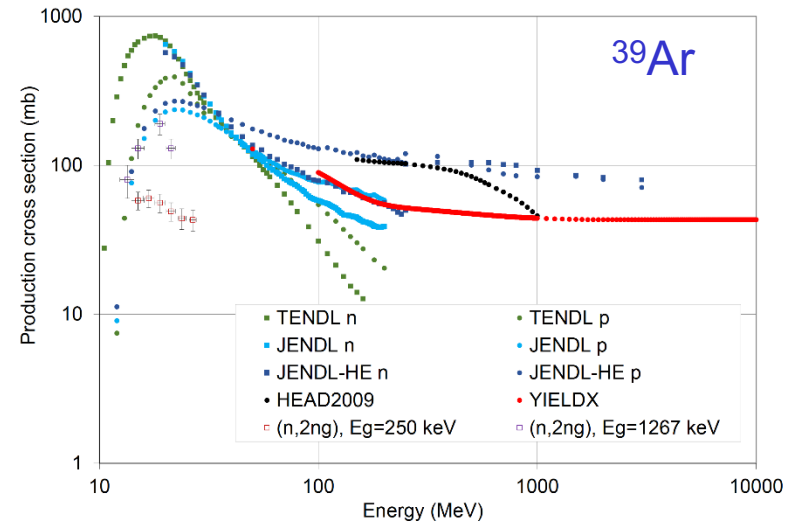
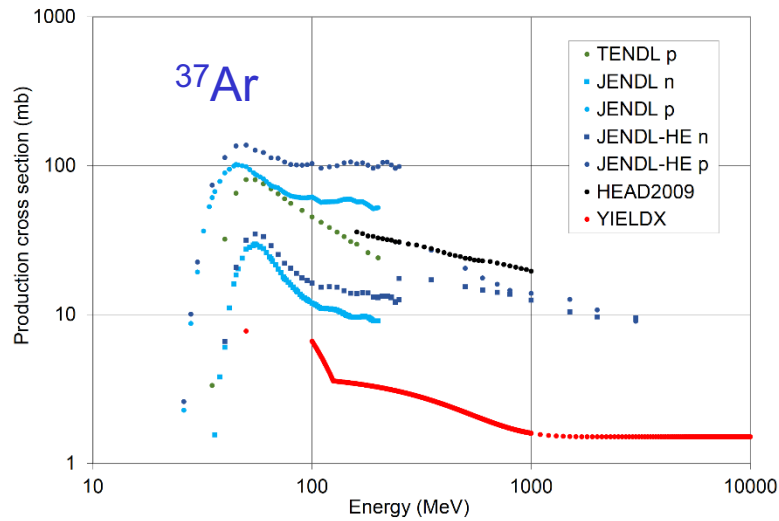


Astropart. Phys. 152 (2023) 102878

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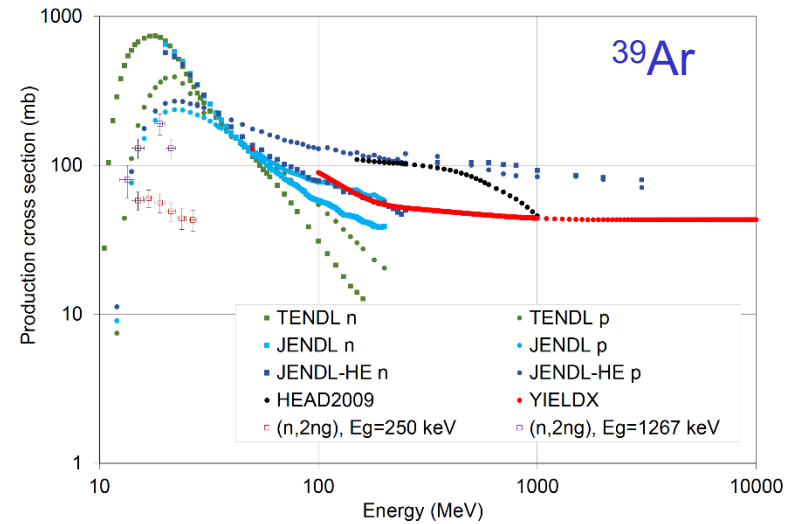
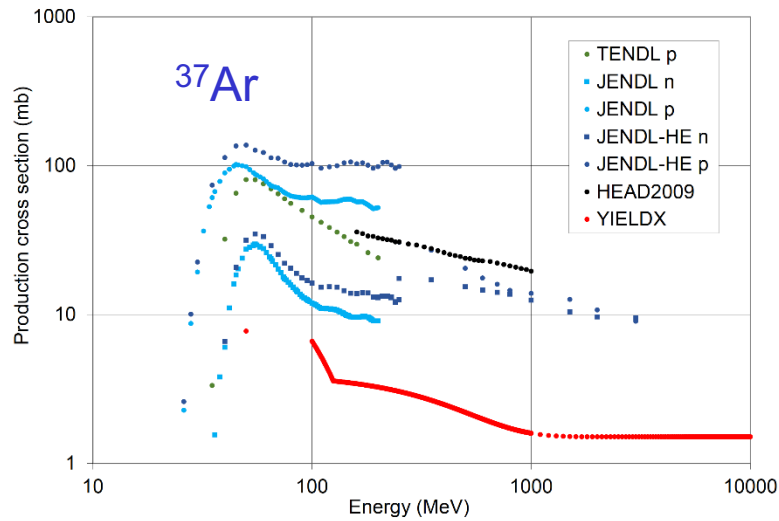
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This work:	Cut (MeV)	R (atoms/kg/day)	This work	Cut (MeV)	R (atoms/kg/day)
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TENDL(p)+YIELDX	100	93.5	TENDL+YIELDX	100	697.1
TENDL(p)+YIELDX	200	122.7	TENDL+YIELDX	200	646.0
JENDL-HE(n)	30	63.9	TENDL+JENDL-HE(n)	20	804.3
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Measurement [35]		51.0 ± 7.4			759 ± 128
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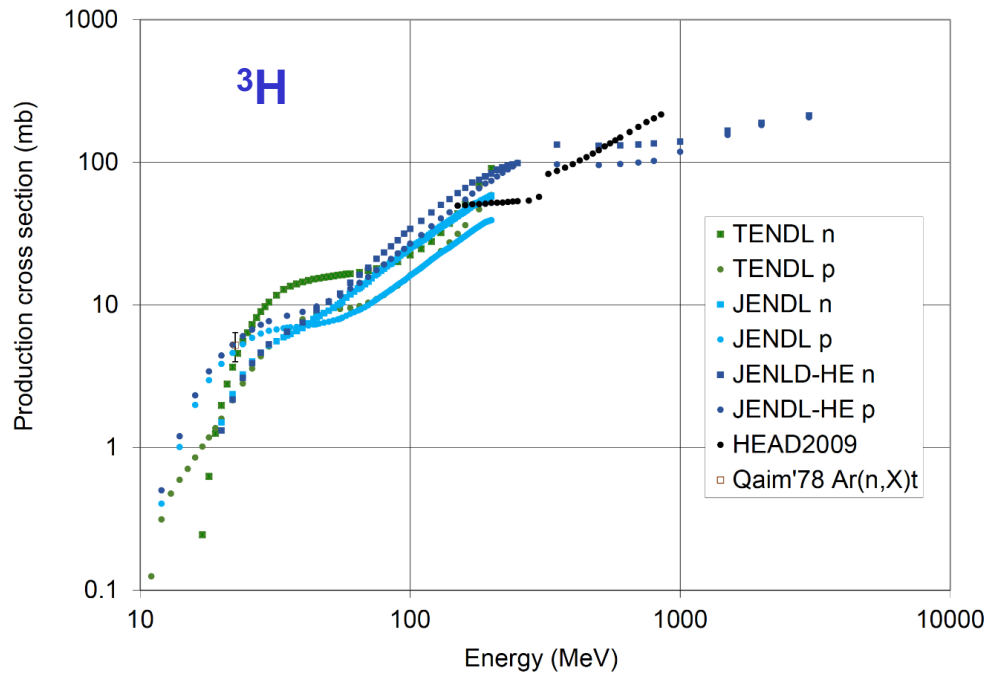
<sup>39</sup>Ar: fully compatible with measured value and several calculations

<sup>37</sup>Ar: larger discrepancies

# Production rates

- Production rates R** (sea level): new calculations

$$R = N_t \int \sigma(E)\phi(E)dE$$



Astropart. Phys.152 (2023) 102878

Same approach but including more data

J. Amaré et al, Astropart. Phys.97 (2018) 96

	R (atoms/kg/day)
TENDL	115.1
HEAD2009	177.2
JENDL-HE	221.6
Estimated rate in this work	<b>168 ± 53</b>
Not used for estimation:	
TALYS [16]	44.4
GEANT4 [33]	84.9
ACTIVIA [33]	82.9

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- **Activity A** from measured production rates for  $^{37}\text{Ar}$ ,  $^{39}\text{Ar}$  and estimated rate for  $^3\text{H}$  for realistic exposure conditions

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Fix tentative **exposure times** and **places (altitude)** for shipping:

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$^{39}\text{Ar}$	Neutrons	Muons	Protons	$\gamma$ rays	Total
<i>R</i> (atoms/kg/day) [35]	$759 \pm 128$	$172 \pm 26$	$3.6 \pm 2.2$	$112.8 \pm 20.9$	
Urania	$0.551 \pm 0.093$	$0.0483 \pm 0.0073$	$0.0035 \pm 0.0022$	$0.0127 \pm 0.0024$	$0.616 \pm 0.093$
US	$0.139 \pm 0.024$	$0.0148 \pm 0.0022$	$0.0009 \pm 0.0005$	$0.0056 \pm 0.0010$	$0.161 \pm 0.024$
Overseas	$0.359 \pm 0.061$	$0.081 \pm 0.012$	$0.0017 \pm 0.0010$	$0.053 \pm 0.010$	$0.495 \pm 0.063$
Aria	$0.321 \pm 0.054$	$0.073 \pm 0.011$	$0.0015 \pm 0.0009$	$0.048 \pm 0.0088$	$0.444 \pm 0.056$
Italy	$0.0536 \pm 0.0090$	$0.0121 \pm 0.0018$	$0.0003 \pm 0.0002$	$0.0080 \pm 0.0015$	$0.0739 \pm 0.0093$
Final	$1.42 \pm 0.24$	$0.229 \pm 0.035$	$0.0078 \pm 0.0048$	$0.127 \pm 0.024$	$1.79 \pm 0.24$
(%)	79.6	12.8	0.4	7.1	
$^{37}\text{Ar}$	Neutrons	Thermal neutrons	Protons	$\gamma$ rays	Total
<i>R</i> (atoms/kg/day) [35]	$51 \pm 7.4$	$0.9 \pm 0.3$	$1.3 \pm 0.4$	$3.5 \pm 0.7$	
Urania	$87 \pm 13$	$2.99 \pm 0.92$	$0.93 \pm 0.19$	$0.239 \pm 0.080$	$91 \pm 13$
US	$24.5 \pm 3.6$	$0.81 \pm 0.25$	$0.453 \pm 0.091$	$0.116 \pm 0.039$	$25.9 \pm 3.6$
Overseas	$37.5 \pm 5.4$	$0.95 \pm 0.29$	$2.57 \pm 0.51$	$0.66 \pm 0.22$	$41.7 \pm 5.5$
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Italy	$9.2 \pm 1.3$	$0.234 \pm 0.072$	$0.63 \pm 0.13$	$0.162 \pm 0.054$	$10.2 \pm 1.3$
Final	$8.0 \pm 1.2$	$0.209 \pm 0.064$	$0.52 \pm 0.10$	$0.135 \pm 0.045$	$8.9 \pm 1.2$
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If no purification  
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$^3\text{H}$	
R (atoms/kg/day)	$168 \pm 53$
Urania	$2.66 \pm 0.84$
US	$0.67 \pm 0.21$
Overseas	$1.73 \pm 0.54$
Aria	$1.55 \pm 0.49$
Italy	$0.259 \pm 0.082$
Final	$6.5 \pm 2.1$

# Activity and counting rates

**Activity A** when all UAr is in Gran Sasso and **counting rate** in DarkSide-20k

Isotope	Activity ( $\mu\text{Bq/kg}$ )	TPC rate (Hz)	Veto rate (Hz)
$^{39}\text{Ar}$	$20.7 \pm 2.8$	$1.03 \pm 0.14$	$0.662 \pm 0.090$
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G4DS simulation of  $\gamma/\beta$  emissions from the full set of detector components to estimate rates in the TPC and in the Veto from measured activities

$\gamma$ :  $\sim 50$  Hz in TPC,  $\sim 100$  Hz in Veto

$\beta$ : 36 Hz in TPC, 26 Hz in Veto

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Study of cosmogenic activation above ground for the DarkSide-20k experiment, *Astropart. Phys.*152 (2023) 102878

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- Study of cosmogenic activation above ground for the DarkSide-20k experiment, *Astropart. Phys.*152 (2023) 102878
- Results useful to set exposure limitations for the large amounts of LAr necessary in future projects for dark matter (**DarkSide-Low Mass, ARGO**) and even for neutrino experiments (COHERENT, LEGEND-1000, DUNE)

EXTRA SLIDES

# Correction factors

- Production rates R** correction factors **f** for exposure at Colorado

Urania facilities: 2164 m

$$I_2 = I_1 \exp\left(\frac{A_1 - A_2}{L}\right), \quad (1)$$

where  $I_1$  is the cascade flux at some altitude (pressure)  $A_1$ , and  $I_2$  is the flux at altitude  $A_2$ , both altitudes being expressed in  $\text{g}/\text{cm}^2$ . To convert terrestrial altitudes to

J.F. Ziegler, IBM J. Res. Develop. 42 (1998) 117.

Table 3. Sea-level particle absorption lengths.

Particle	Length L ( $\text{g}/\text{cm}^2$ )
Electrons	100
Protons	110
Pions	113
Neutrons	136
Muons and muon capture	261

- Protons:** f=8.67
- Muons:** f=2.48
- Neutrons:** extrapolation for Urania location of deduced factors  $f$  due to altitude and geomagnetic rigidity at Denver and Leadville.

Location	$H$ (ft)	$A$ ( $\text{g}/\text{cm}^2$ )	$f$ from [47]	Relative $I$ to Urania	Deduced $f$ for Urania
Denver	5280	852.3	4.11	0.659	6.24
Leadville	10200	705.2	12.86	1.942	6.62
Urania	7100	795.5			<span style="border: 1px solid red; padding: 2px;">6.43</span>

Table 1

Cosmogenic activation rates of three argon isotopes:  $^{37}\text{Ar}$ ,  $^{39}\text{Ar}$  and  $^{42}\text{Ar}$ . The simulation results are also compared with the ones from measurements and estimations[56].

	$^{37}\text{Ar}$	$^{39}\text{Ar}$	$^{42}\text{Ar}$
	atoms/kg <sub>Ar</sub> /day		
Neutrons (this work)	176.01	857.73	$4.60 \times 10^{-3}$
Neutrons (measurement[56])	$51.0 \pm 7.4$	$759 \pm 128$	-
Muons (this work)	2.40	52.27	$1.57 \times 10^{-4}$
Muons (calculation[56])	-	$172 \pm 26$	-
Protons (this work)	6.20	28.53	$1.05 \times 10^{-3}$
Protons (calculation[56])	$1.73 \pm 0.35$	$3.6 \pm 2.2$	-
Total (this work)	184.61	938.53	$5.81 \times 10^{-3}$
Total (Ref.[56])	$52.73 \pm 7.75$	$934.6 \pm 156.2$	-

Table 2

Cosmogenic activation rates of other long-lived isotopes.

Isotope, Half Life	Neutron	Muon	Proton
	atoms/kg <sub>Ar</sub> /day		
$^3\text{H}$ , 12.32y	$3.00 \times 10^{-4}$	$6.56 \times 10^{-6}$	$1.05 \times 10^{-4}$
$^7\text{Be}$ , $1.387 \times 10^6\text{y}$	$3.43 \times 10^{-3}$	$6.47 \times 10^{-3}$	$9.62 \times 10^{-3}$
$^{10}\text{Be}$ , 53.22d	$7.05 \times 10^{-3}$	$5.22 \times 10^{-3}$	$1.10 \times 10^{-2}$
$^{14}\text{C}$ , $5.703 \times 10^3\text{y}$	0.10	$9.85 \times 10^{-3}$	$4.56 \times 10^{-2}$
$^{22}\text{Na}$ , 2.602y	0.37	$2.35 \times 10^{-2}$	$9.92 \times 10^{-2}$
$^{26}\text{Al}$ , $7.17 \times 10^5\text{y}$	0.63	$4.24 \times 10^{-2}$	0.12
$^{32}\text{Si}$ , 153y	7.00	0.14	0.47
$^{32}\text{P}$ , 14.268d	15.7	0.38	1.05
$^{33}\text{P}$ , 25.3d	22.5	0.42	1.32
$^{35}\text{S}$ , 87.37d	74.5	1.66	3.18
$^{36}\text{Cl}$ , $3.01 \times 10^5\text{y}$	75.5	1.23	3.32
$^{40}\text{K}$ , $1.248 \times 10^9\text{y}$	1.80	$5.86 \times 10^{-2}$	0.56
$^{41}\text{Ca}$ , $9.94 \times 10^4\text{y}$	$1.85 \times 10^{-3}$	$1.02 \times 10^{-4}$	$5.68 \times 10^{-4}$

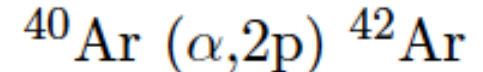
C. Zhang, D.M. Mei, *Astropart. Phys.*  
142 (2022) 102733

# $^{42}\text{Ar}$

## Production of $^{42}\text{Ar}$ in underground argon

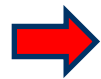
Production mechanisms:

- Two-step neutron capture: high neutron flux required due to short half-life of  $^{41}\text{Ar}$  (1.8 h) → not possible underground
- $(\alpha,2p)$  reaction: zero  $\sigma$  values  $<15$  MeV (TALYS2017) → not possible for  $\alpha$  from radioactivity



- Neutron reactions on Ca:

Isotope	Natural abundance (%)	Reaction	
$^{43}\text{Ca}$	2.086	$^{43}\text{Ca}(n,2p)^{42}\text{Ar}$	Zero $\sigma$ values $<17$ MeV (TALYS2017) → not possible for fission or $(\alpha,n)$ neutrons
$^{44}\text{Ca}$	0.135	$^{44}\text{Ca}(n,n2p)^{42}\text{Ar}$	Zero $\sigma$ values $<23$ MeV (TALYS2017) → not possible for fission or $(\alpha,n)$ neutrons



Precise estimate requires: neutron spectrum underground from other sources, Ca concentration in rock, emanation factor