LOW-RADIOACTIVITY UNDERGROUND ARGON

Pacific Northwest NATIONAL LABORATORY

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Background Sources of ^{37}Ar

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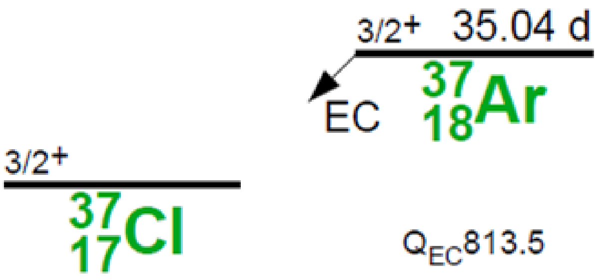
Low Radioactivity Underground Argon Workshop | March 2018

PNNL-SA-133058

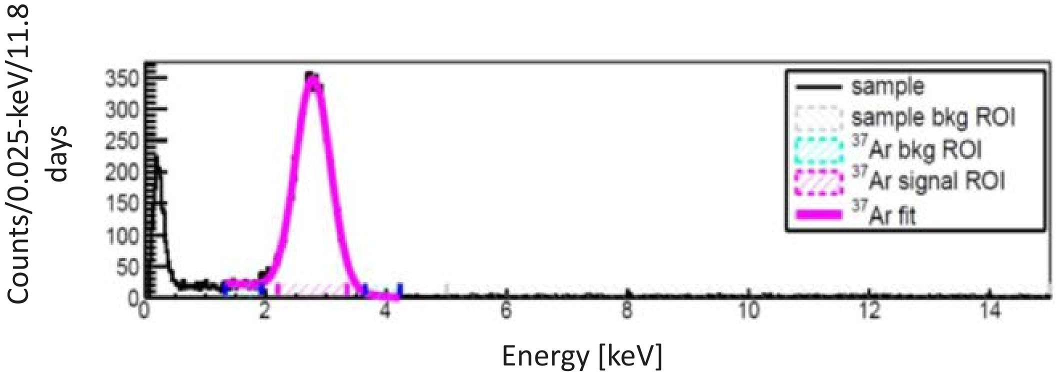
The Underground Nuclear Explosion Signatures Experiment (UNESE) was created to apply a broad range of research and development (R&D) techniques and technologies to nuclear explosion monitoring and nuclear nonproliferation. It is a multi-year research and development project sponsored by NNSA DNN R&D, and is collaboratively executed by Lawrence Livermore National Laboratory, Los Alamos National Laboratory, National Security Technologies, Pacific Northwest National Laboratory, and Sandia National Laboratories.

The basics of ^{37}Ar

- ^{37}Ar decays via electron capture
- Half-life of 35.04 days
- Auger electrons and x-rays sum to binding energy of inner shell vacancy in ^{37}Cl
- ~90.2% of decays in channel centered around 2.82-keV for ^{37}Ar



Firestone, 1996.



Williams et al. *Applied Radiation and Isotopes*, 2016.

A note about units

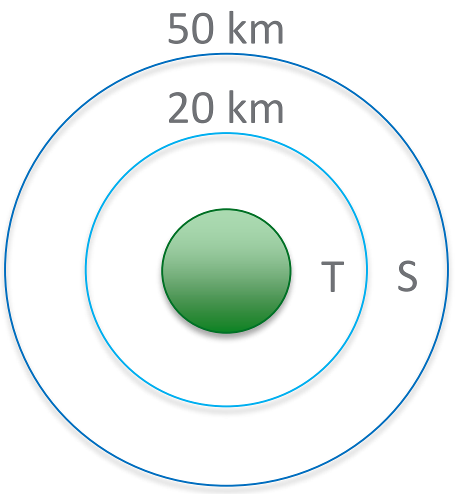
- Within the references used to create these slides there are two primary unit conventions:
  - Concentration per unit air (typically mBq/m^3 air)
  - Concentration per unit argon (typically dpm/L argon)
- For consistency values will be presented as mBq/m^3 air
  - In cases where the original units from the reference is different, will be noted by (orig. dpm/L argon) after the value
- Conversion method used:

$$\frac{dpm}{L\ Ar} \times \frac{16.667\ mBq}{dpm} \times \frac{0.00934\ L\ Ar}{L\ air} \times \frac{1000\ L\ air}{m^3\ air}$$

$$1\frac{dpm}{L\ Ar} = 155.7\frac{mBq}{m^3\ air}$$

Atmospheric background

- Atmospheric ^{37}Ar produced primarily by two mechanisms:
  - Spallation on ^{40}Ar: >90%
  - ^{36}Ar(n,γ)^{37}Ar reaction: <10%
- Atmospheric neutrons produced primarily by interactions between primary galactic cosmic rays and isotopes of O and N
- Production of ^{37}Ar per gram of air peaks at 20 km altitude
- Estimated stratospheric activities:
  - Mean solar activity: 9.3 mBq/m^3 air (orig. dpm/L argon)
  - High solar activity: 0.5 mBq/m^3 air (orig. dpm/L argon)
- Typical tropospheric activity:
  - 1 – 6 mBq/m^3 air (orig. dpm/L argon)
  - Includes mixing from stratosphere
  - Corresponds to ^{37}Ar/Ar ratio of 5×10^{-20}



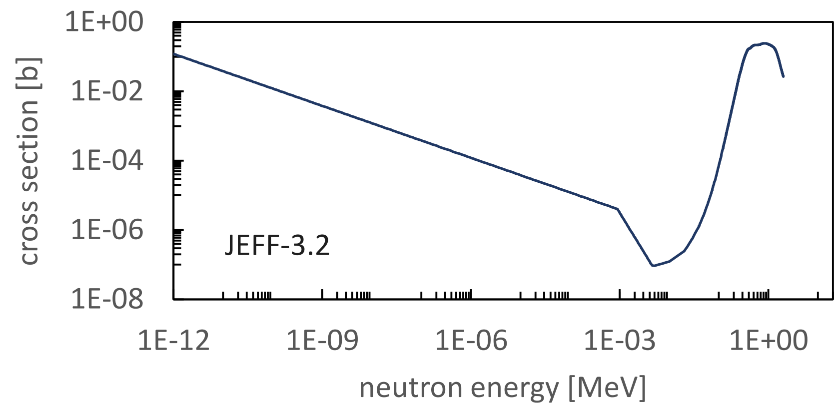
Reactor produced atmospheric source

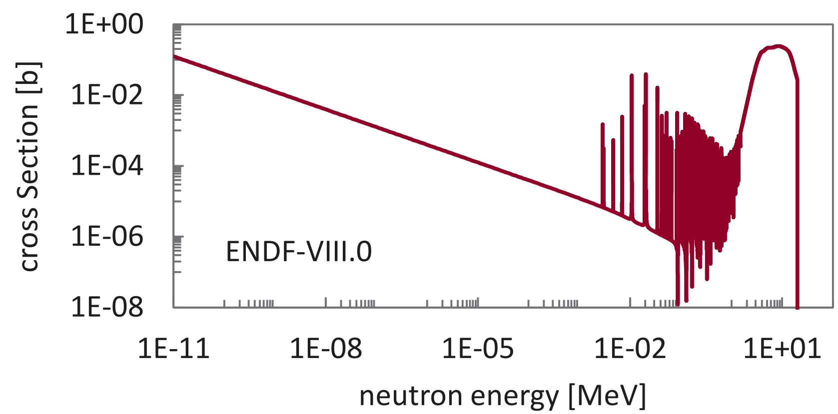
- ^{37}Ar also produced in larger quantities by air activation around nuclear reactors
- Potentially large source of atmospheric ^{37}Ar, but limited in range
- Measurements/Estimates
  - Research reactor, 1 MW (in stack): 80 MBq/year (Johnson 2017)
  - Power reactor, BWR: 111 GBq/year (Matuszek 1975)
  - Power reactor, PWR: 574 GBq/year (Matuszek 1975)
  - Power reactor, HWPWR: 70 GBq/year (Matuszek 1975)

Shallow subsurface production

- Produced in the subsurface primarily by the ^{40}Ca(n,α)^{37}Ar reaction
- Other reactions:
  - $^{37+X}$K(μ,X·n)^{37}Ar
  - ^{39}K(n,2n+p)^{37}Ar
- Cosmic ray secondary particles are primary neutron source
  - Creates depth dependence

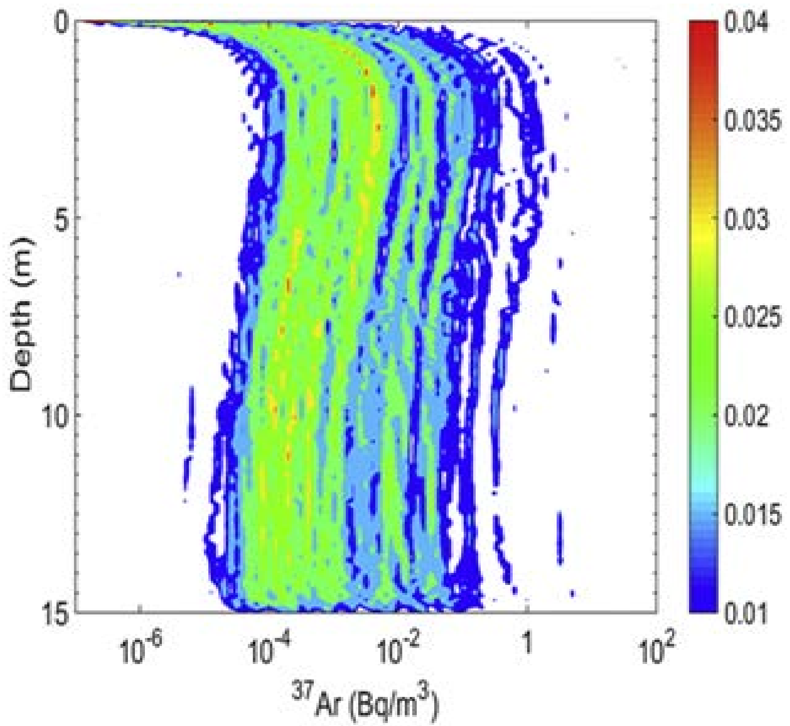
^{40}Ca(n,α)^{37}Ar cross section





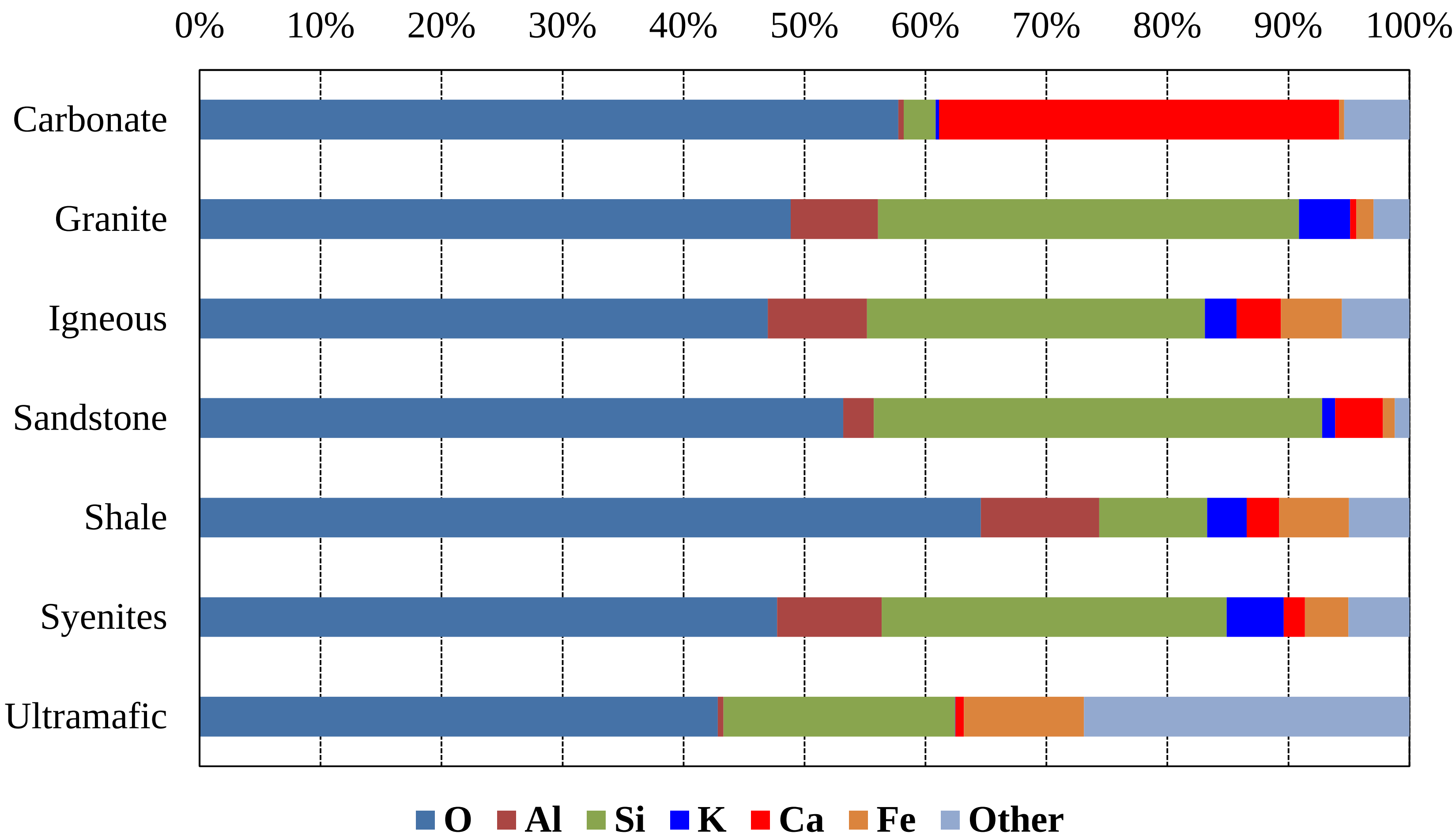
Variations in a simple geology

- Maximum concentration between 2-5 meters
  - Shallower – transport losses
  - Deeper – decreased neutron flux
- Various factors impact subsurface gas concentrations
  - Figure – homogenous sandy soil, varied parameters
    - Ca
    - Neutron flux
    - Transport parameters
    - Water infiltration



Guillon et al. *Journal of Environmental Radioactivity*, 2016.

Geology

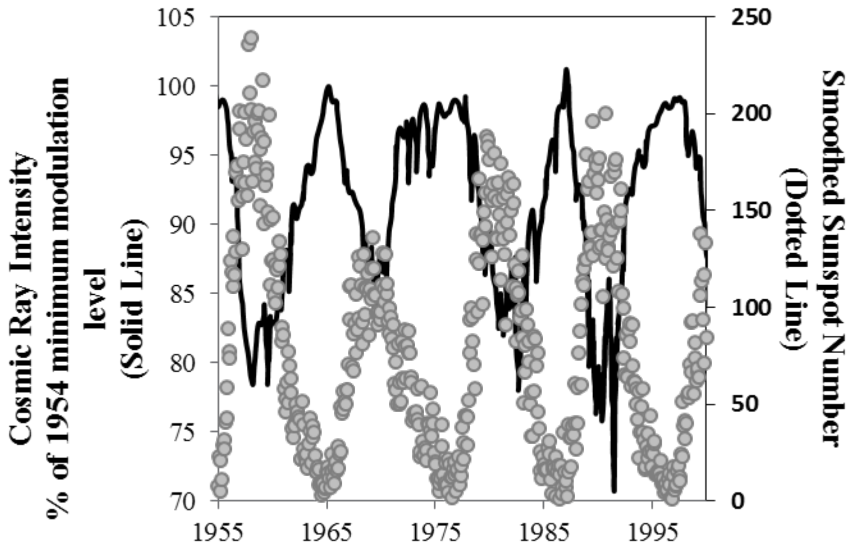


Johnson et al. *Journal of Environmental Radioactivity*, 2015.

Neutron flux

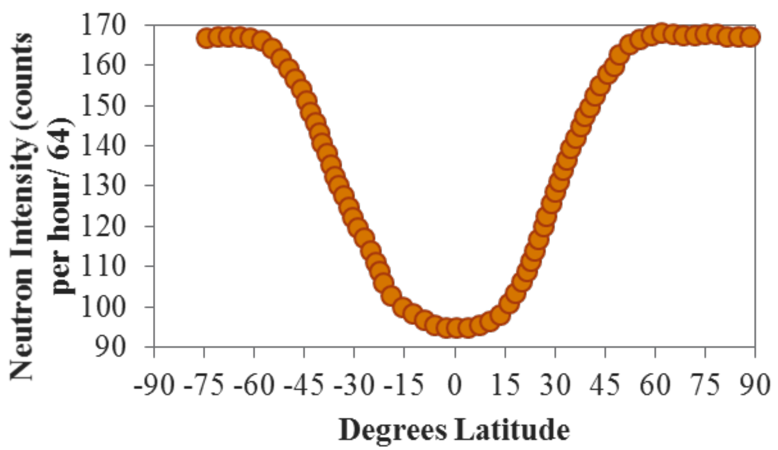
Solar Cycle

- Neutron intensity varies as an inverse of solar activity
- Production rate of ^{37}Ar varies by about a factor of 1.15 between solar minimum and maximum



Latitude

- Neutron intensity varies as a function of latitude
- Production rate of ^{37}Ar varies by more than a factor of 2 between 15° and 60°



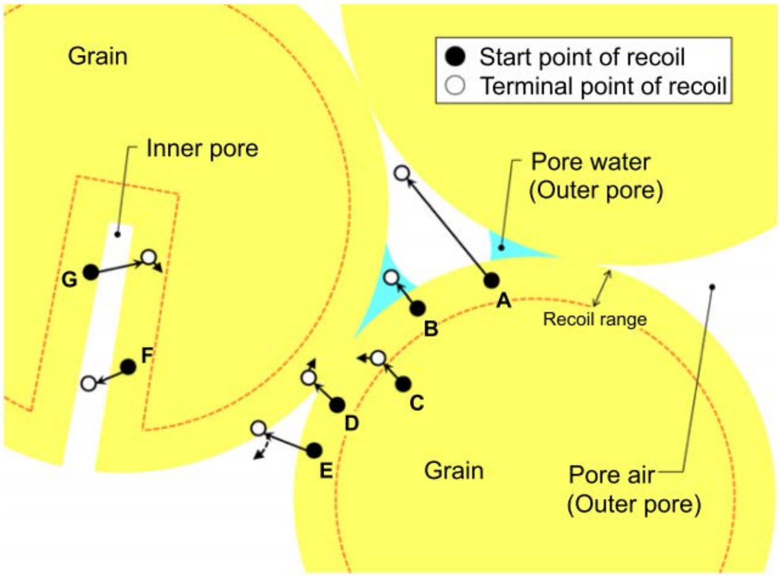
Johnson et al. *Journal of Environmental Radioactivity*, 2015.

Shallow subsurface measurements

Location	Depth [m]	Ca [%]	^{37}Ar Conc [mBq/m^3 air]	Reference
Richland, WA	4.3	0.75	1.6 ± 0.4	Fritz 2018
Yellowstone NP	1.45	20	125 ± 9.3	Fritz 2018
Olympic NP	0.79	0.6	3.0 ± 1.5	Fritz 2018
Olympic NP	1.52	0.4	18 ± 2.5	Fritz 2018
München, DE	9.0	90.0	3.0 ± 1.0	Riedmann 2011
Baltenswil, CH	2.1	65	119.3 ± 29.0	Riedmann 2011
Baltenswil, CH	9.6	65	9.9 ± 3.0	Riedmann 2011

A brief note about emanation

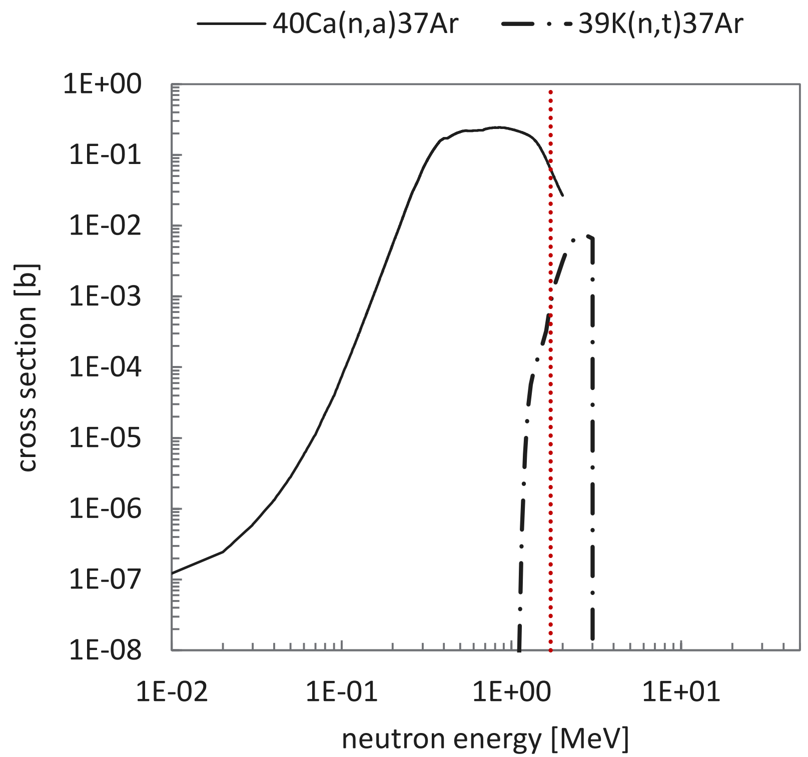
- Production rates do not provide the full picture on how much argon may be measured in soil gas
- Most gas remains trapped in material
  - May be trapped in the material
  - May recoil into adjoining grains
- Some gas escapes
  - This is the gas measured during subsurface gas sampling
- Percentage of gas that escapes:
  - **Emanation fraction/coefficient**



Sakoda et al., Appl. Radiat. Isot., 2011

Deep underground

- Primary reactions:
  - $^{40}Ca(n,\alpha)^{37}Ar$
  - $^{39}K(n,2n+p)^{37}Ar$
- Primary neutron sources:
  - Spontaneous fission
  - (α,n) reactions on light elements
    - α particles produced by α decay of natural U, Th
- Average neutron energy from both (α,n) reactions and spontaneous fission:
  - 1.7 MeV



Current measurements/predictions

- Measured in groundwater at depths below cosmic muons (and neutrons)
  - Range of values between 1.9-1900 mBq/m^3 air (orig. dpm/L argon) (Lehmann 1993)
- Estimated production rates in specific geologies (Lehmann 1997):
  (Sorted by neutron flux from highest to lowest)
  - Stripa Granite: 3.9×10^{-2} atoms/cm^3 rock per year
  - Typical Granite: 1.1×10^{-2} atoms/cm^3 rock per year
  - Milk River Sandstone: 1.3×10^{-2} atoms/cm^3 rock per year

Expanded predictions

- In order to predict ^{37}Ar production deep underground, need
  - Estimated neutron flux
  - $^{40}Ca(n,\alpha)^{37}Ar$ cross sections
  - Ca concentration of surrounding geology
- Estimation of emanation (ε) needed to infer air concentrations from production rates in rock
  - Recent measurements estimate ε to be 1-15% for ^{37}Ar

Calculations in brief

- Neutron production rate (P_n) in continental crust, oceanic crust, and depleted upper mantle drawn from Šrámek, 2017
  - Production rate in neutrons/kg rock/year
  - Assume average neutron energy of 1.7 MeV
- $^{40}Ca(n, \alpha)^{37}Ar$ cross section (σ) at 1.7 MeV (ENDF VIII)
  - 1.93 mb
- Calcium content known for each geology
- Assumes spherical kilogram of rock of known density to convert neutron production rate, P_n, to neutron flux, Φ
- Production rate

$$P_{Ar37} = \Phi \sigma N_{Ca}$$

Expanding to underground ^{37}Ar activities

- Calculate activity from production rate, ^{37}Ar decay constant, and emanation fraction
  - $T_{1/2}(^{37}Ar) = 35.04$ days
  - ε assumed to be 10%
- Activity will be in Bq/m^3 rock/year
  - In order to convert to m^3 air must assume some porosity (ϕ)

$$A_{Ar37} = \frac{P_{Ar37}\lambda\varepsilon}{\phi}$$

Estimated ^{37}Ar production deep underground

Geology	Neutron Production Rate [n/year/kg rock] (Šrámek 2017)	^{37}Ar Production Rate [atoms/year/kg rock]	^{37}Ar Production Rate [mBq/m^3 air/year]
Upper CC	10 680	1.79×10^{-4}	4.30×10^{-5}
Middle CC	6114	1.53×10^{-4}	3.92×10^{-5}
Lower CC	1129	5.25×10^{-5}	1.43×10^{-5}
Oceanic Crust	260	1.45×10^{-5}	3.66×10^{-6}
Depleted Upper Mantle	22.4	3.95×10^{-7}	1.20×10^{-7}

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Conclusions

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- Atmospheric production dominated by spallation reactions on ^{40}Ar by cosmic ray neutrons
  - Average tropospheric concentrations: 1 – 6 mBq/m^3 air
  - Average stratospheric concentrations: 9 mBq/m^3 air (orig. dpm/L argon)
- Shallow underground production dominated by $^{40}Ca(n,\alpha)^{37}Ar$ reaction from cosmic ray neutrons
  - Concentrations between 1-125 mBq/m^3 air measured
- Deep underground production primarily from $^{40}Ca(n,\alpha)^{37}Ar$ reaction
  - Neutrons produced by:
    - (α,n) reactions on light elements
    - spontaneous fission
  - Production rates between 1×10^{-7}- 4×10^{-5} mBq/m^3/year estimated

Selected References

Atmospheric :

- Machta, L. Argon-37 as a measure of atmospheric vertical mixing. in *Noble Gases* 58–68 (1973).
- Loosli, H. H., Oeschger, H. & Wiest, W. Argon 37, Argon 39, and Krypton 81 in the Atmosphere and Tracer Studies Based on These Isotopes. *J. Geophys. Res.* **75,** 2895–2900 (1970).
- Johnson, C. *et al.* Production and release rate of 37Ar from the UT TRIGA Mark-II research reactor. *J. Environ. Radioact.* **167**, 249-253 (2017).
- Matuszek, J, Paperiello, C, Kunz, C. Reactor Contributions to Atmospheric Noble Gas Radioactivity Levels. in *Noble Gases* 360-364 (1973).

Shallow Subsurface:

- Johnson, C., Armstrong, H., Wilson, W. H. & Biegalski, S. R. Examination of radioargon production by cosmic neutron interactions. *J. Environ. Radioact.* **140,** 123–129 (2015).
- Guillon, S. *et al.* Alteration of natural (37)Ar activity concentration in the subsurface by gas transport and water infiltration. *J. Environ. Radioact.* **155–156,** 89–96 (2016).
- Riedmann, R. a & Purtschert, R. Natural 37Ar concentrations in soil air: implications for monitoring underground nuclear explosions. *Environ. Sci. Technol.* **45,** 8656–64 (2011).
- Fritz, B. G. *et al.* Prediction of sub-surface 37 Ar concentrations at locations in the Northwestern United States. *J. Environ. Radioact.* **181,** 1–7 (2018).

Deep Underground

- Lehmann, B. E., Davis, S. N. & Fabryka-Martin, J. T. Atmospheric and subsurface sources of stable and radioactive nuclides used for groundwater dating. *Water Resour. Res.* **29,** 2027–2040 (1993).
- Lehmann, B. E. & Purtschert, R. Radioisotope dynamics - The origin and fate of nuclides in groundwater. *Appl. Geochemistry* **12,** 727–738 (1997).
- Šrámek, O., Stevens, L., McDonough, W. F., Mukhopadhyay, S. & Peterson, R. J. Subterranean production of neutrons, 39Ar and 21Ne: Rates and uncertainties. *Geochim. Cosmochim. Acta* **196,** 370–387 (2017).