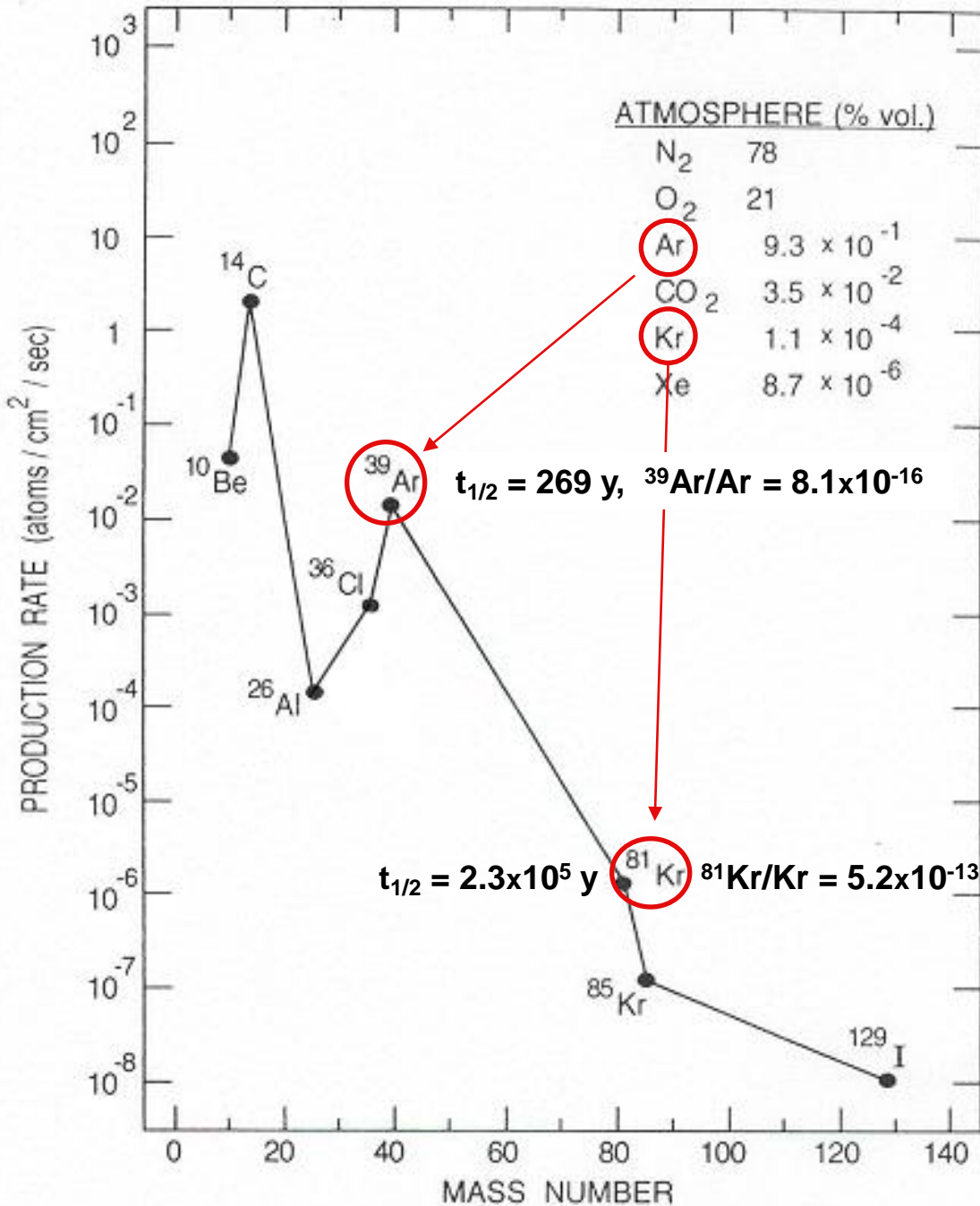


Atom Counting of Noble Gas Radioisotopes with Accelerators – Successes and Limitations

Walter Kutschera

*Vienna Environmental Research Accelerator (VERA)
Faculty of Physics, University of Vienna*



Production rate of long-lived radionuclides in the atmosphere

W.K. et al, NIMB 92 (1994) 241

Successes

- Dating old groundwater with ^{81}Kr in the Great Artesian Basin by measuring $^{81}\text{Kr}/\text{Kr}$ atom ratios in the 10^{-13} range
- Dating ocean water with ^{39}Ar in the Southern Atlantic by measuring $^{39}\text{Ar}/\text{Ar}$ atom ratios in the 10^{-16} range

Limitations

- Attempts to measure $^{39}\text{Ar}/\text{Ar}$ ratios down to the 10^{-18} range to test underground argon for its suitability for Dark Matter Searches

USA

Physics Department, University of Notre Dame

P. Collon, M. Bowers, D. Robertson, C. Schmitt

Physics Division, Argonne National Laboratory

J. Caggiano, J.L. Jiang, A. Heinz, D. Henderson, H.Y. Lee, Richard Pardo, K.E. Rehm, R.H. Scott, R. Vondrasek

National Superconducting Cyclotron Laboratory, Michigan State University

T. Antaya, D. Anthony, D. Cole, B. Davids, M. Fauerbach, R. Harkewicz, M. Hellstrom, D.L. Morrissey, B.M. Sherrill, M. Steiner

Lamont-Doherty Earth Observatory of Columbia University

P. Schlosser, W.M. Smethie Jr.

Department of Physics, Princeton University

F. Calaprice, C. Galbiati, T. Hohman, B. Loer

Lawrence Livermore National Laboratory

J. Moran

US Geological Survey, Idaho

L. De Wane Cecil

CANADA

Institute for Groundwater Research, University of Waterloo

S.K. Frape

EUROPE

VERA Laboratory, University of Vienna

W. Kutschera, R. Golser

Atom Institute, Technical University of Vienna

M. Bichler

Isotope Hydrology Section, IAEA Vienna

M. Gröning

Institute of Physics, University of Bern

B.E. Lehmann, H.H. Loosli, R. Purtschert

Swiss Federal Institute of Environmental Science and Technology (EAWAG) Dübendorf

W. Aschbach-Hertig, R. Kipfer

Institute of Nuclear Physics, Catholic University of Louvain

Y. El Masri, P. Leleux

Geological Institute, Kola Scientific Centre, Russian Academy of Sciences, Apaty

I.N. Tolstikhin

Racah Institute of Physics, Hebrew University of Jerusalem

Michael Paul

AUSTRALIA

Department of Water, Land and Biodiversity Conservation (DWLBC), South Australia, Adelaide

A. Love

CSIRO Land and Water, South Australia, Adelaide

A. Herczeg



Bernhard
Lehmann

H₂O degassing
apparatus
Univ. of Bern, CH
16 000 L H₂O/sample
→ 320,000 cm³ gas → 0.5 cm³ Kr



Bernhard Lehmann (1946-2005) taking a groundwater sample in the Great Artesian Basin in Australia in 1998.



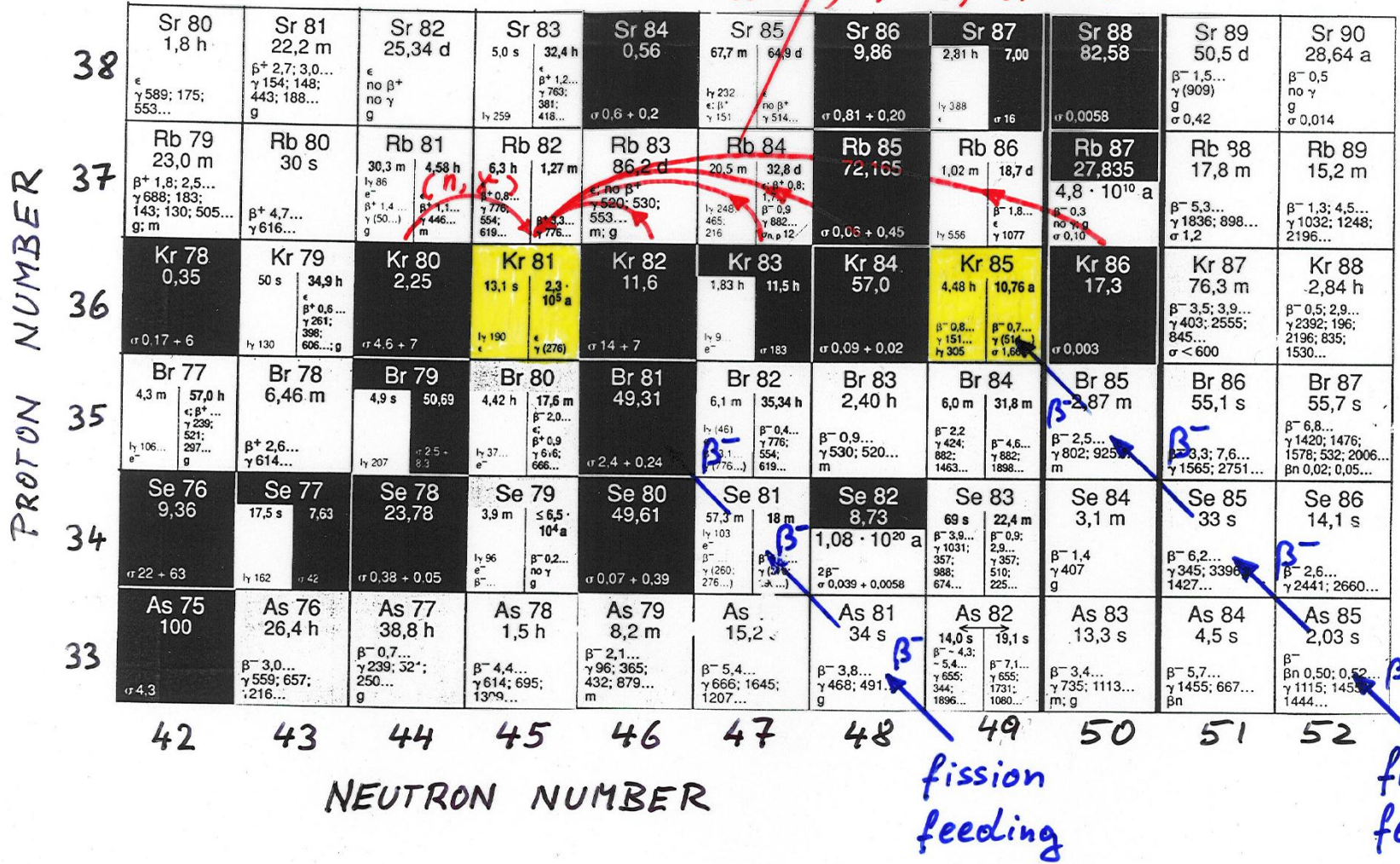
Water fountain from a well in the Great Artesian Basin of Australia

The production of ^{81}Kr (230,000 y) and ^{85}Kr (10.8 y) from cosmic rays in the atmosphere and from fission

$$\frac{^{81}\text{Kr}}{\text{Kr}} = (5.2 \pm 0.4) \times 10^{-13}$$

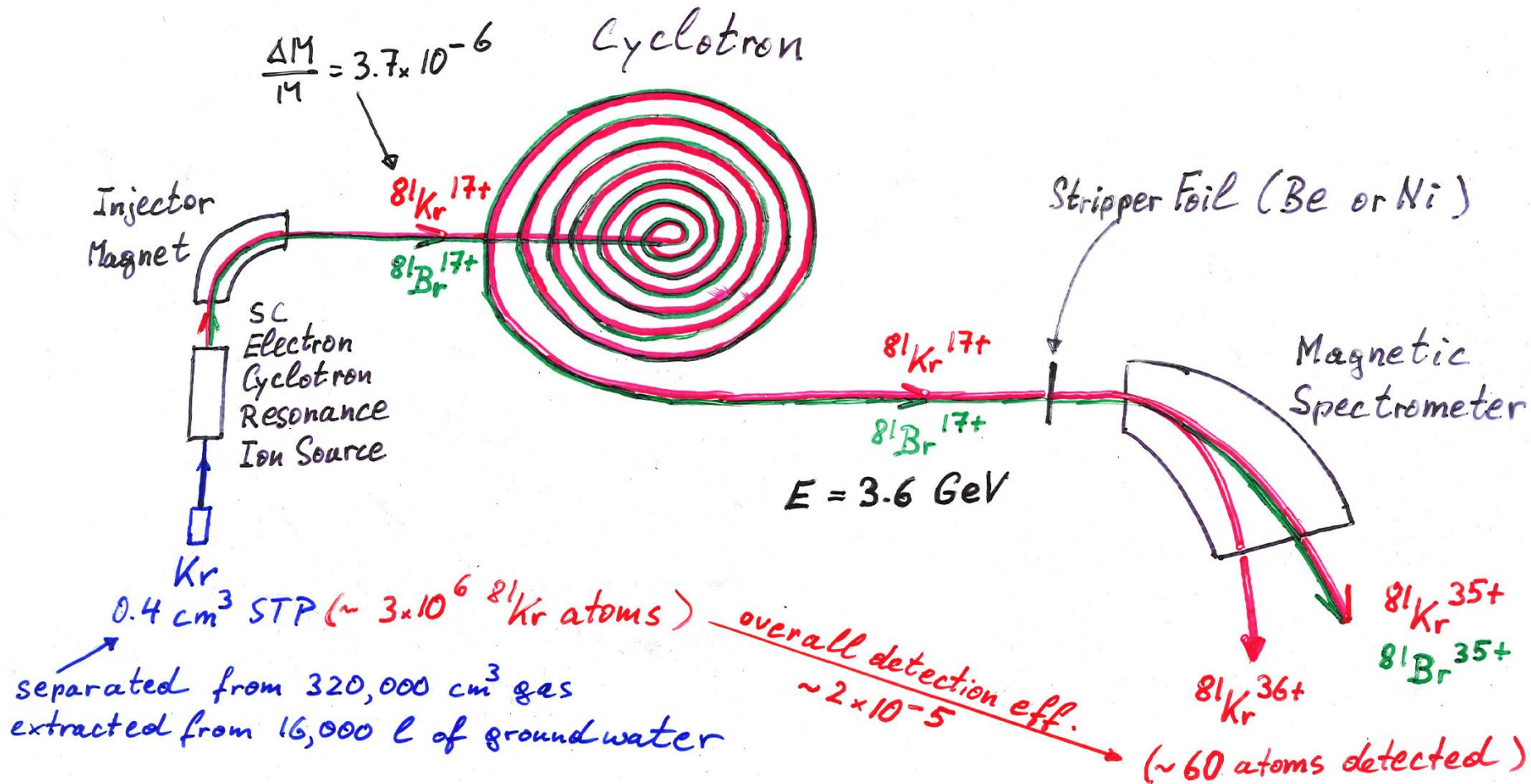
$$\frac{^{85}\text{Kr}}{\text{Kr}} = 3 \times 10^{-18} \text{ (cosmogenic)} \rightarrow 2 \times 10^{-11} \text{ (anthropogenic)}$$

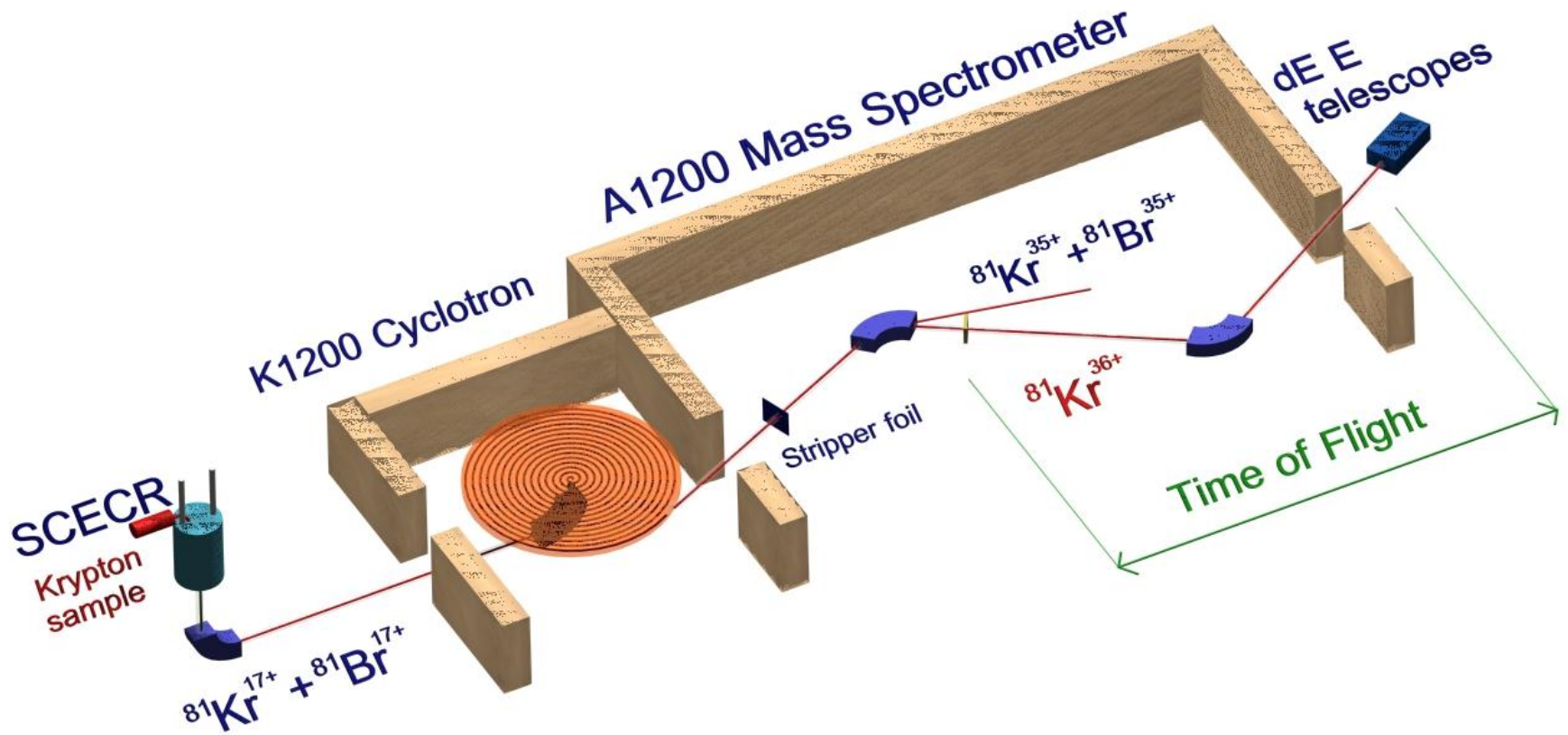
$(p, pxn); (n, xn); (\gamma, xn)$



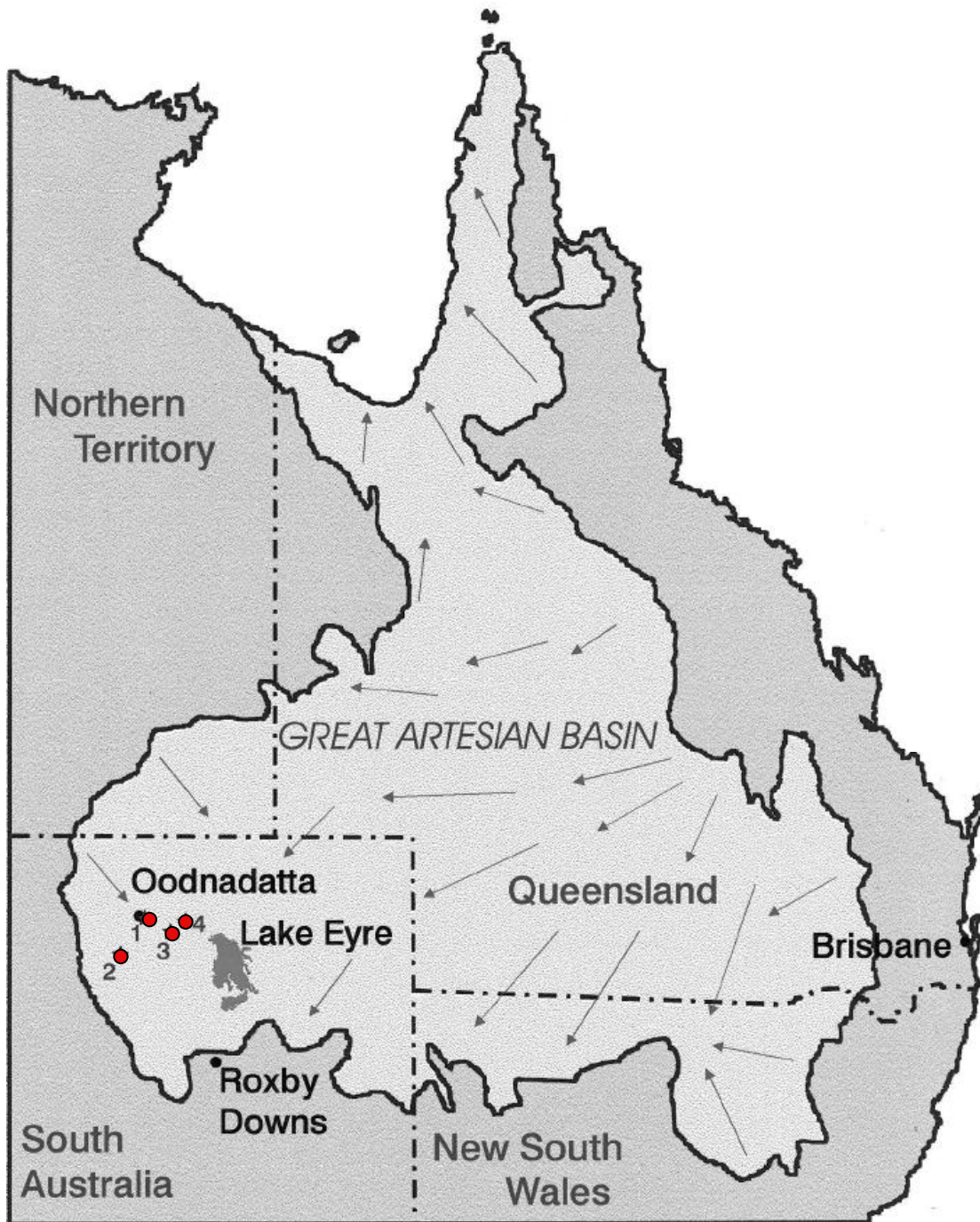
^{81}Kr AMS detection in groundwater samples from the Great Artesian Basin in Australia through full-stripping at the K1200 Superconducting Cyclotron of Michigan State University

P. Collon et al., Earth Planet. Sci. Lett. 182 (2000) 103





Schematic layout of the experimental setup at the superconducting cyclotron at Michigan State University



Area of the Great Artesian Basin in Australia

~ 1.7 million km²

~ 1/5 of Australia

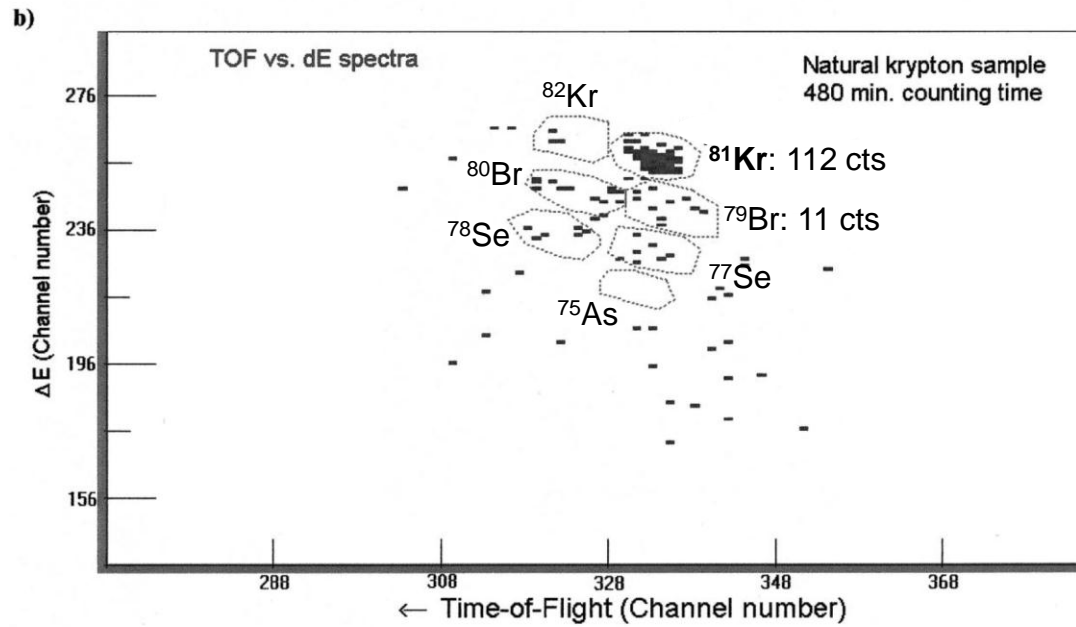
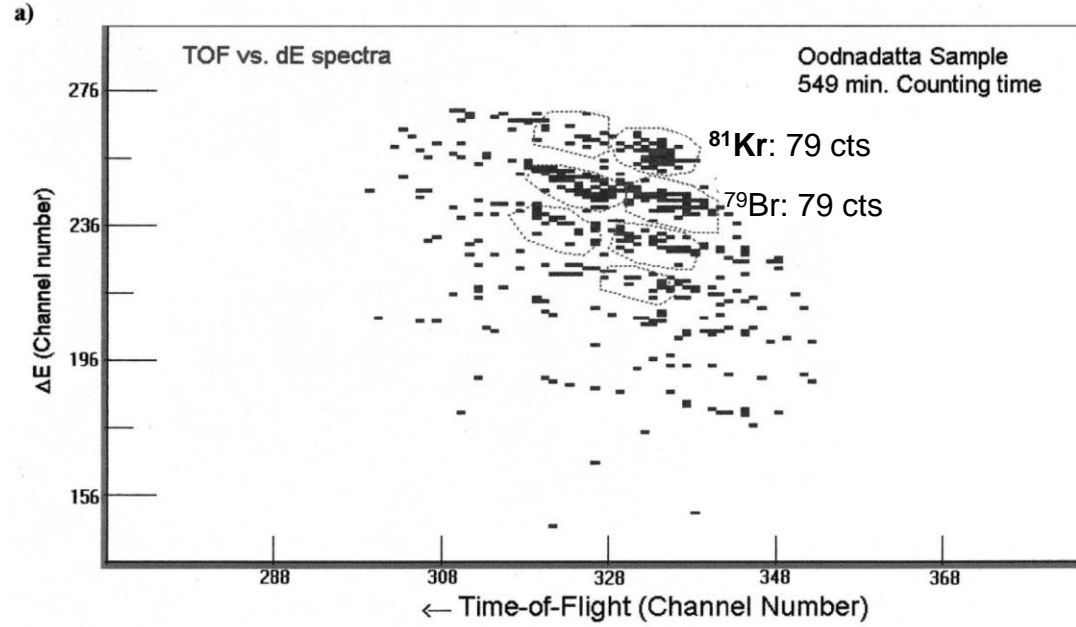
~ size of Alaska

Water storage

~ 8.7 x 10¹⁵ litre

~ ½ Great Lakes

- Sampling sites



Identification of ^{81}Kr events from a GAB groundwater sample (a) and Atmospheric krypton (b). From Collon et al., EPSL 182 (2000) 103.

Summary of $^{81}\text{Kr}/\text{Kr}$ isotopic ratios for Krypton gas extracted from groundwater samples of the Great Artesian Basin in Australia

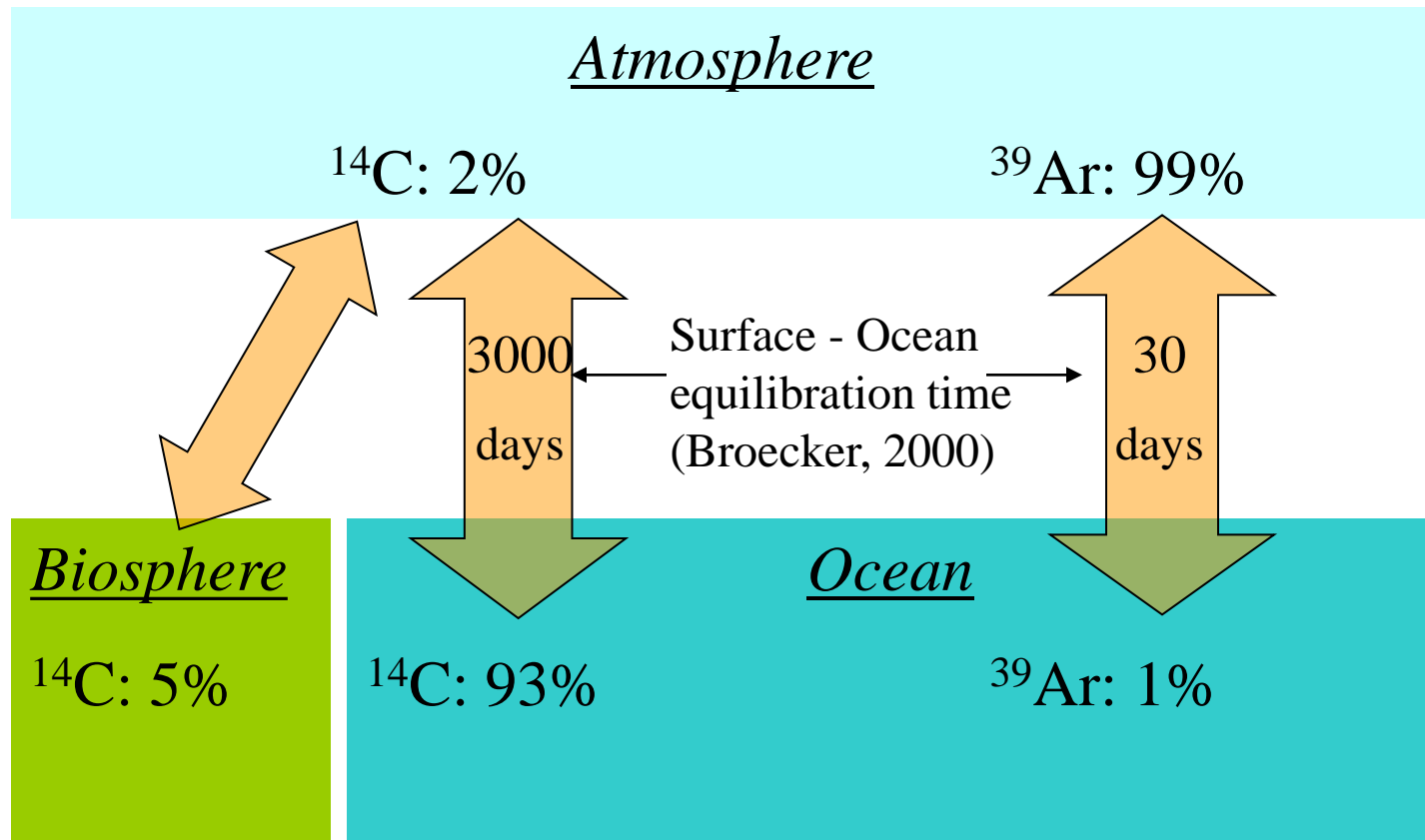
Sample name	Well location (well depth)	$^{81}\text{Kr}/\text{Kr}$ (10^{-13})	Mean residence time (10^3 yr)	
			^{81}Kr ^a	^{36}Cl ^b
Raspberry Creek	2 (127 m)	2.63 ± 0.32	230 ± 40	
Oodnatta	1 (479 m)	1.78 ± 0.26	350 ± 50	340 - 540
Duck Hole	4 (251 m)	2.19 ± 0.28	290 ± 40	470 - 760
Watson Creek	3 (289 m)	1.54 ± 0.22	400 ± 50	480 – 890
Atmospheric Kr		5.20 ± 0.40	~ 0	

^a P.Collon et al., Earth Planet. Sci. Lett. 182 (2000) 103-113

^b A. J. Love et al., Water Resour. Res. 36/6 (2000) 1561-1574

**Dating ocean water of the Southern Atlantic with ^{39}Ar
by measuring $^{39}\text{Ar}/\text{Ar}$ atom ratios
with AMS in the 10^{-16} range**

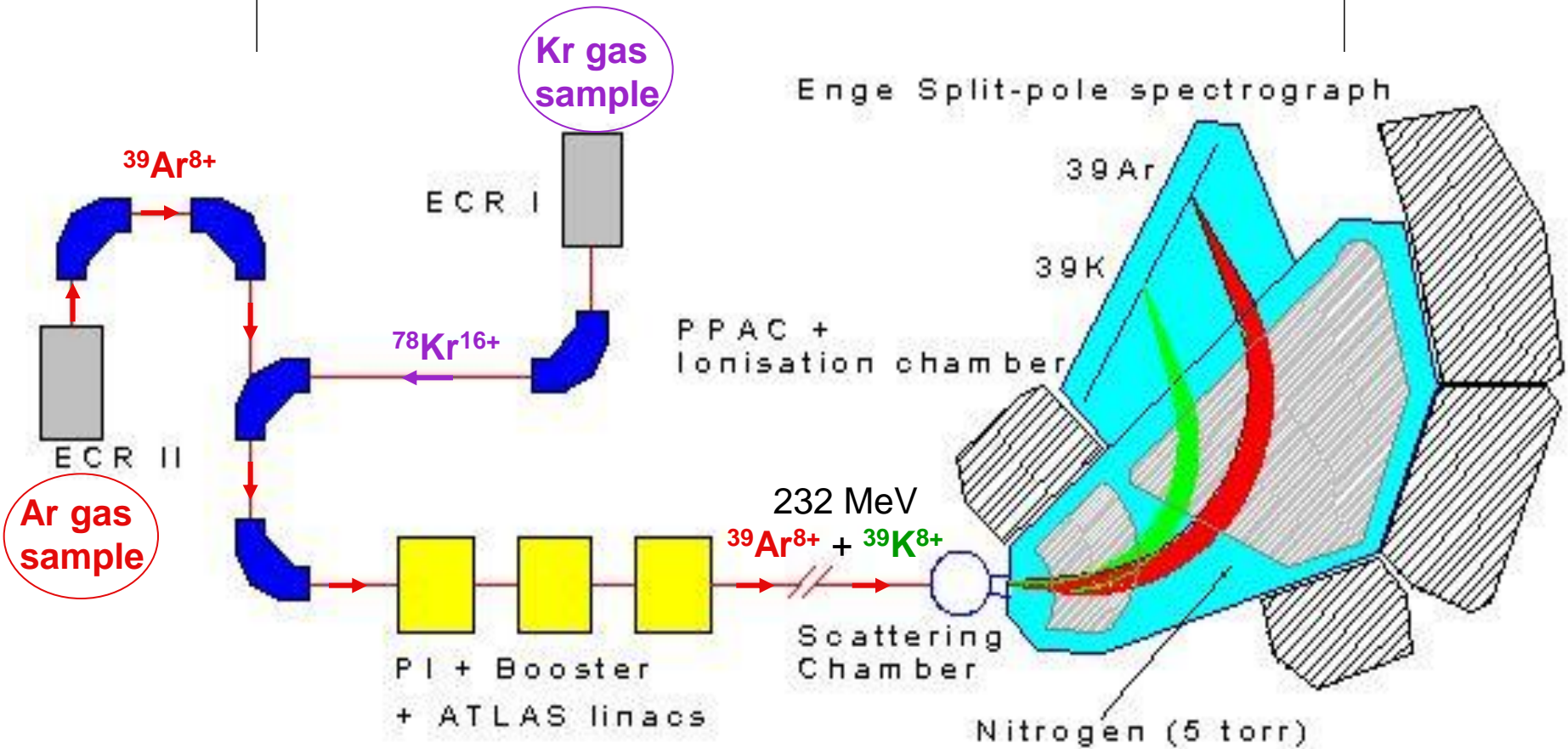
Dynamics and inventory of ^{14}C and ^{39}Ar in the ocean

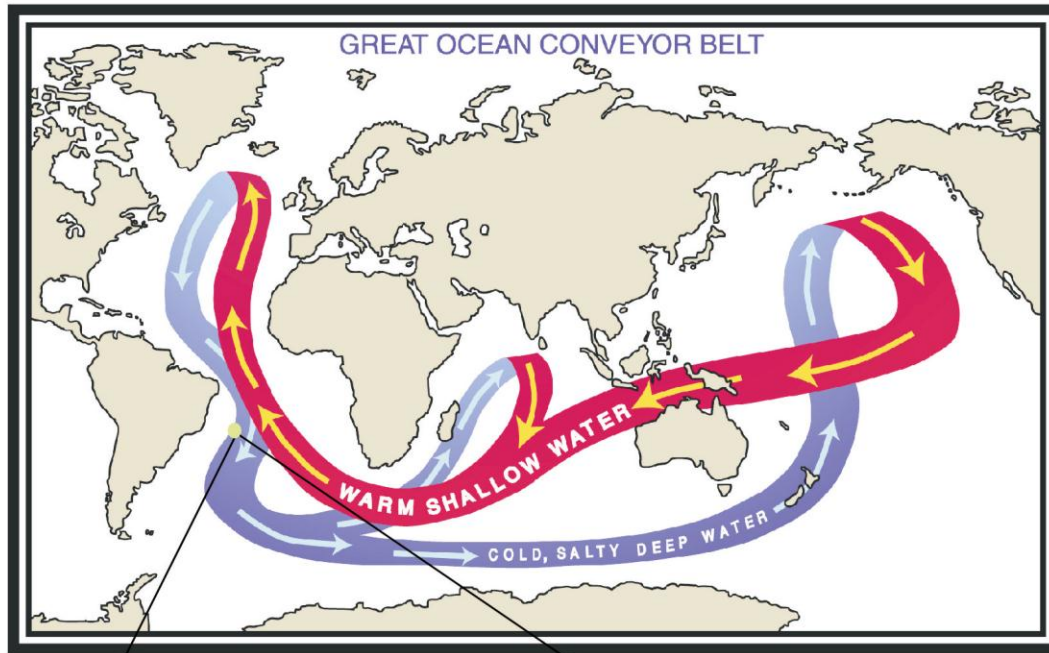


	^{14}C	^{39}Ar
Half-life	5730 y	269 y
Isotope ratio (atmosphere)	$^{14}\text{C}/^{12}\text{C} = 1.2 \times 10^{-12}$	$^{39}\text{Ar}/^{40}\text{Ar} = 8.1 \times 10^{-16}$
Atom concentration (ocean)	1.8×10^9 ^{14}C at/litre	6.5×10^3 ^{39}Ar at/litre

^{39}Ar detection at the ATLAS linear accelerator at Argonne accelerating $^{39}\text{Ar}^{8+}$ ions to 232 MeV and separating it from the $^{39}\text{K}^{8+}$ isobaric background in the gas-filled magnet

P. Collon et al. NIMB 223-224 (2004) 428





Dating of deep ocean water with ^{39}Ar ($t_{1/2} = 269 \text{ a}$) using AMS

Sample: SAVE #95

Water depth = 4717 m

$^{39}\text{Ar}/^{40}\text{Ar} = (2.6 \pm 0.6) \times 10^{-16}$

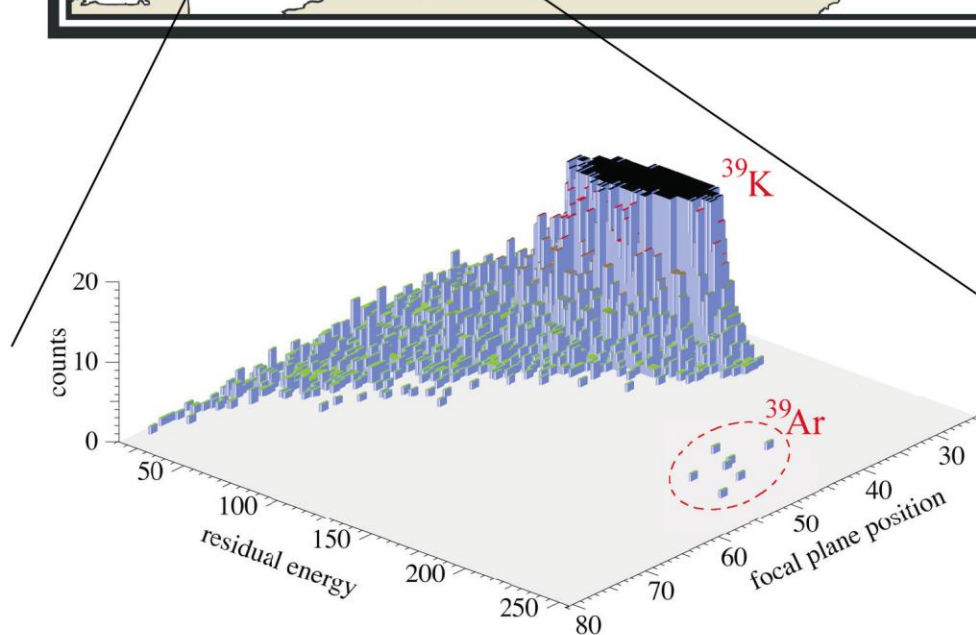
= 32% atmospheric Ar

“Age“ (decay) = 440 years

P. Collon et al.

Ann.Rev. Nucl.Part. Sci.

54 (2004) 39-67



Results of $^{39}\text{Ar}/^{40}\text{Ar}$ measurements at Argonne

Sample	$^{39}\text{Ar}/^{40}\text{Ar}$ (10^{-16})	Fraction of atm. argon*	“Decay age“ (years)
Neutron activated argon	580 ± 40		
Atmospheric argon	7.7 ± 0.9	95%	
South Atlantic Ventilation Experiment			
#294, water depth = 850 m	5.2 ± 0.7	65%	170
#294, water depth = 5000 m	3.5 ± 0.6	44%	320
#95, water depth = 4717 m	2.6 ± 0.6	32%	440
Great Artesian Basin, Australia, Watson			
creek well, ground water (0.4 Myr)	0.4 ± 0.2	5%	1200
Atmospheric argon*	8.5 ± 1.2	105%	
Neutron activated argon	600 ± 40		

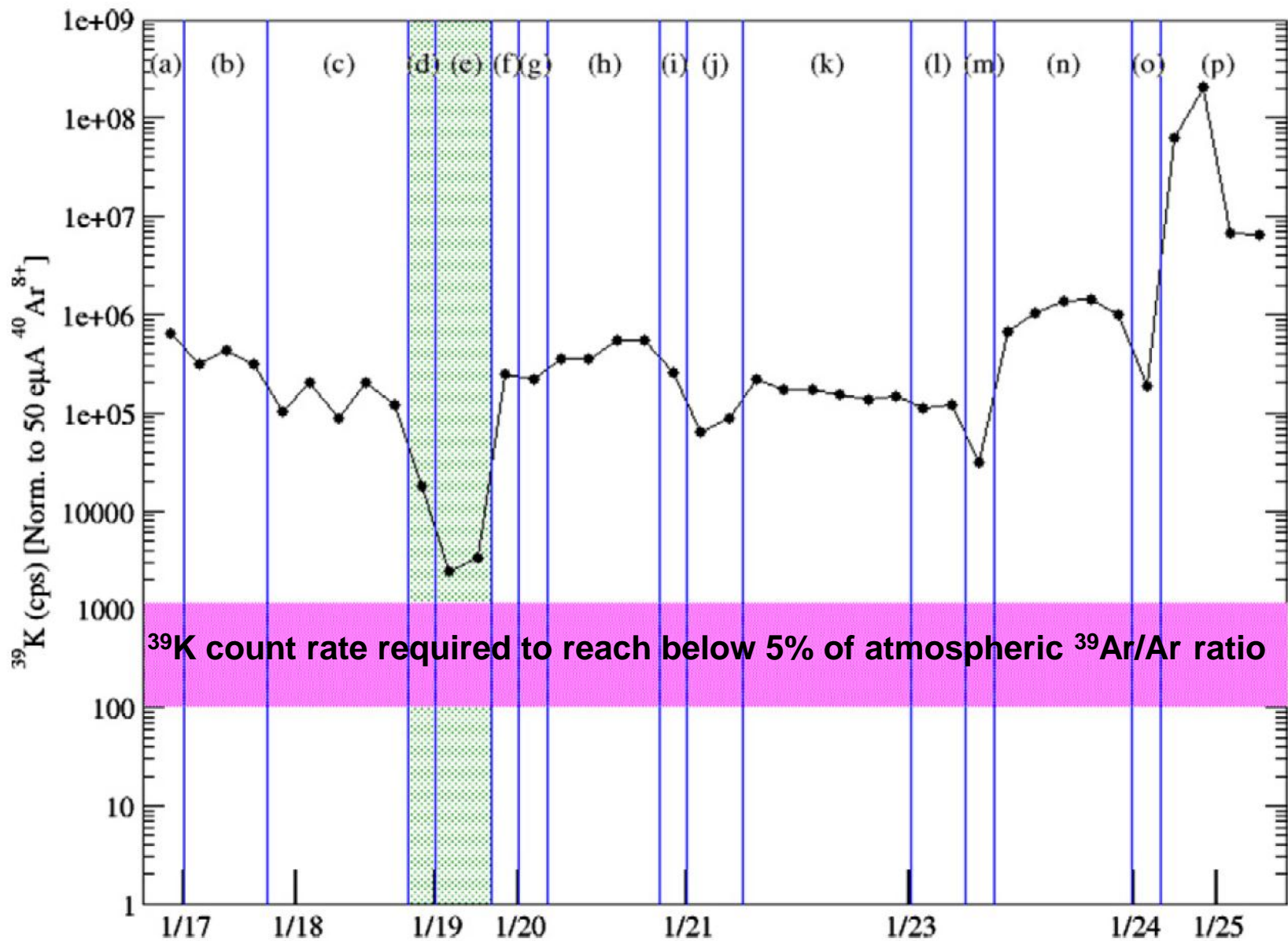
*Normalized to $^{39}\text{Ar}/^{40}\text{Ar} = 8.1 \times 10^{-16} = 100\%$, measured by Low Level Counting (Loosli)

P. Collon et al., Nucl. Instr. Meth. B 223-224 (2004) 428-434

Attempts to measure $^{39}\text{Ar}/\text{Ar}$ ratios down to the 10^{-18} range to test argon sufficiently depleted in ^{39}Ar , which would make it suitable for large liquid-argon detectors for Dark Matter Searches

Collaboration with F. Calaprice et al. from Princeton

P. Collon et al, NIMB 283 (2012) 77



Summary of ^{39}K count rate (counts per sec, cps) at the detector (normalized to $50 \mu\text{A}$ of $^{40}\text{Ar}^{8+}$ at the ion source) as a function of different ion source support gasses and conditions in January 2009



The quartz liner of the ECR source



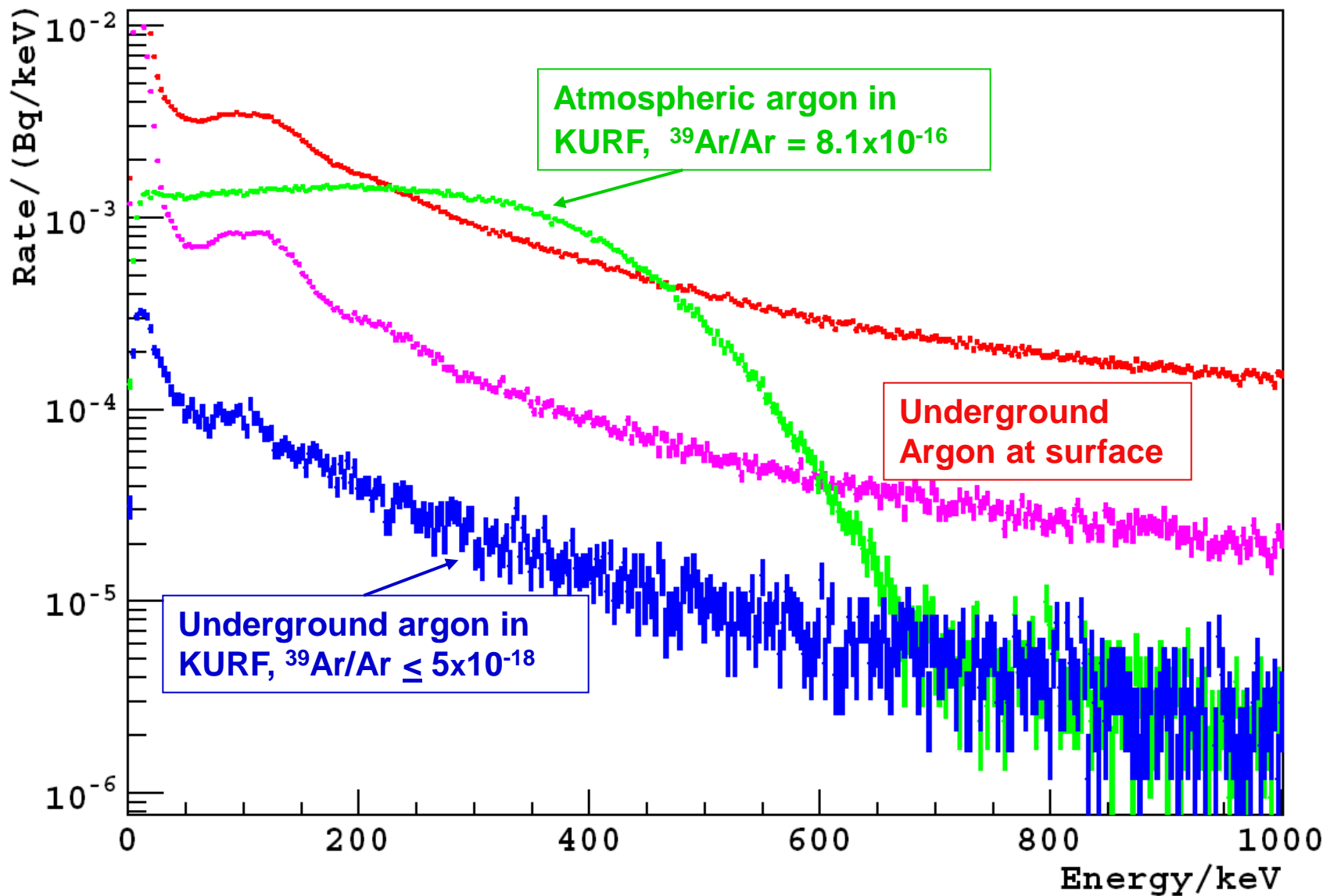
Trying to extract an Al liner from the ECR source



Not so happy with the result

Testing argon with the lowest ^{39}Ar content

- Decay counting with liquid argon detector
- Atom counting with ATTA?



Energy spectra of the ^{39}Ar beta decay (endpoint energy = 565 keV) recorded in a 0.56 kg liquid argon detector in the Kimballton Underground Research Facility (KURF) in Virginia.

J. Xu, F. Calaprice et al. (Princeton), arXiv:1204.6011v1_26 April 2012

Conclusion

AMS is a wonderful tool – but not for noble gas radionuclides