ACCELERATOR MASS SPECTROMETRY; a powerful atom counting technique

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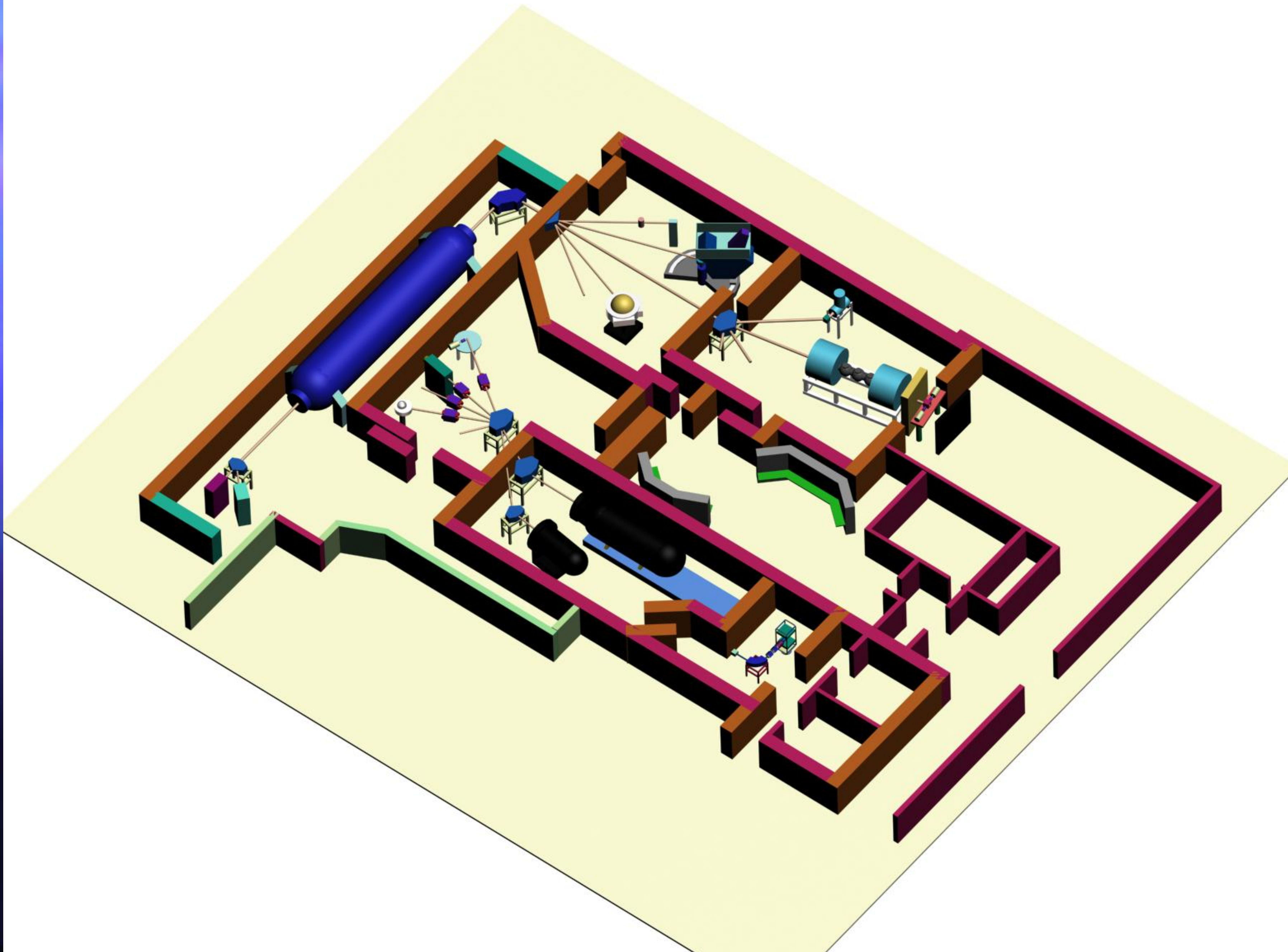


What is Accelerator Mass spectrometry (AMS)

The determination of the concentration of a given radionuclide in a sample can be done in 2 ways:

a) measure the radiation emitted during the decay

In many cases where concentrations and/or small or long $t_{1/2}$ this becomes impractical

1mg carbon = 6 x 10^7 at ^{14}C $\equiv$ ~1 decay/hour

b) count the number of atoms themselves

In a Mass Spectrometer a sample material is converted to an ion beam that is then magnetically (and electrostatically) analysed

→ MS separates ions by their mass only

Goal of AMS

However in many cases a high background (molecular, isobaric, …) makes it impossible to separate the ions of interest.

An unambiguous (A, Z) identification would solve this problem

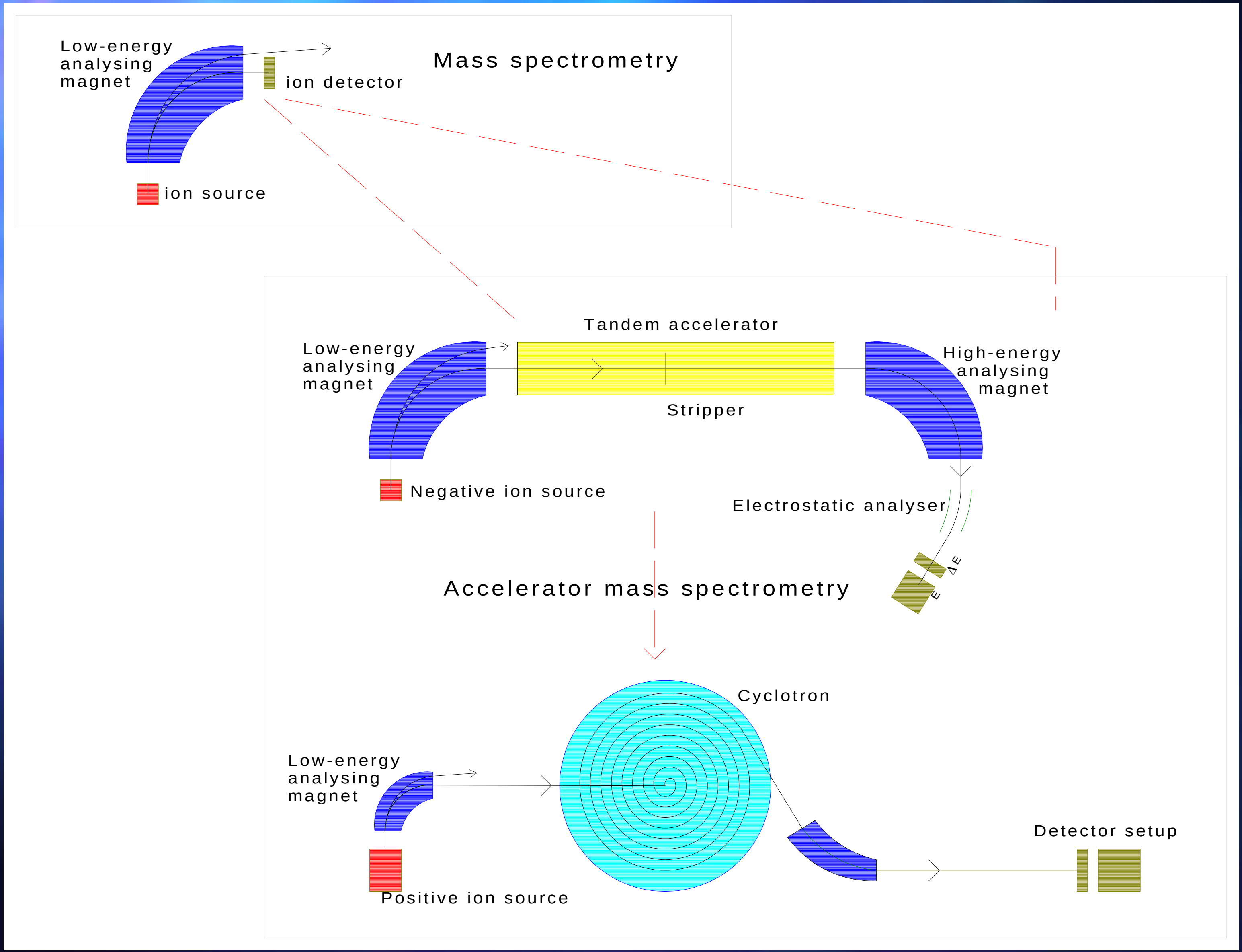
The use of an accelerator in AMS makes it possible to go to much higher energies (several MeV vs. keV) and the measurement of a range of properties that do not depend on ionic charge.

- Range
- Stopping power
- TOF

→ (A, Z)

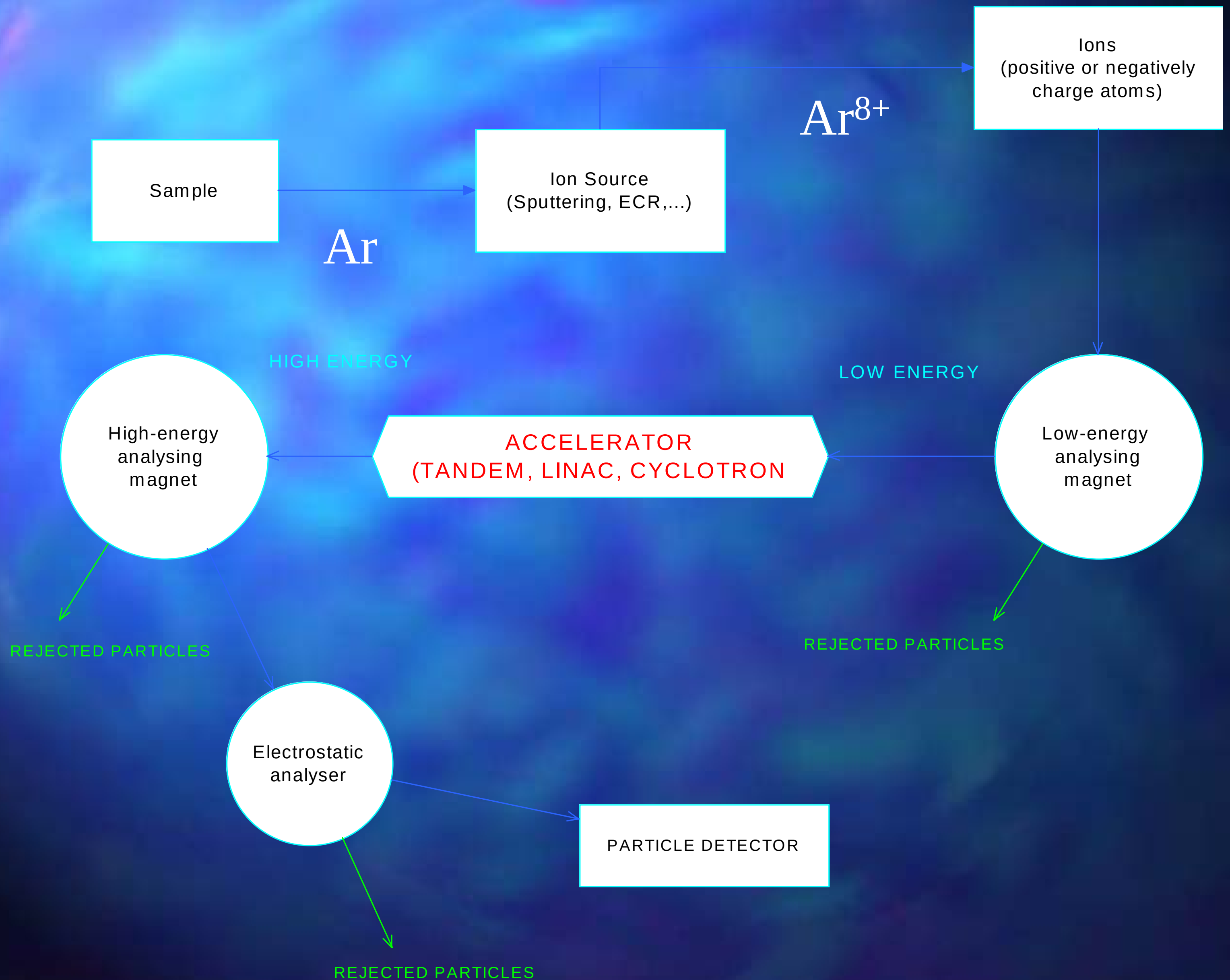
The high sensitivity of the method makes it possible to measure down to several counts per hour from a beam of the order of microamperes (1.6 μA = 1 x 10^{13} ions).

MS vs. AMS

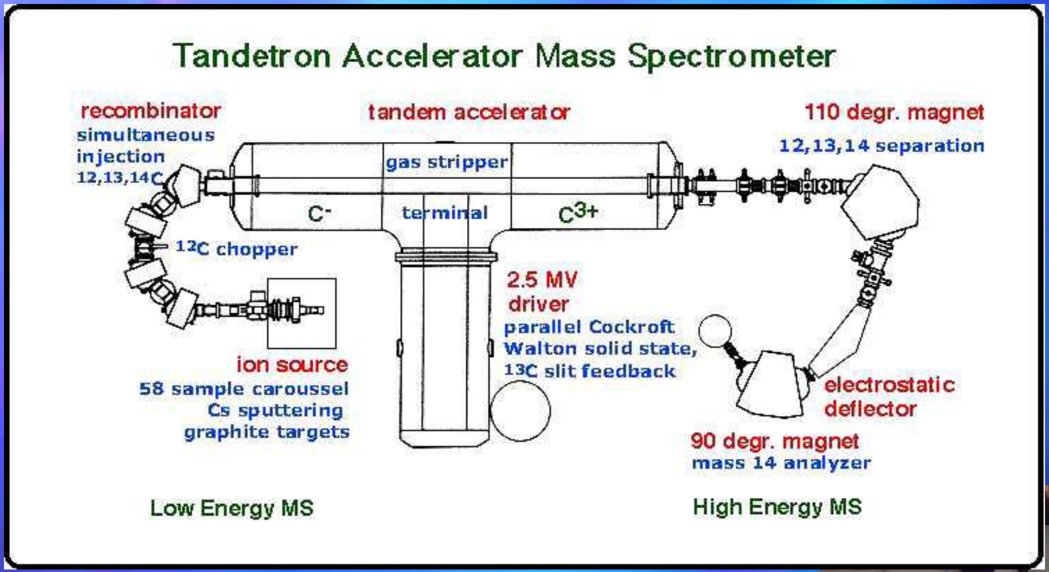


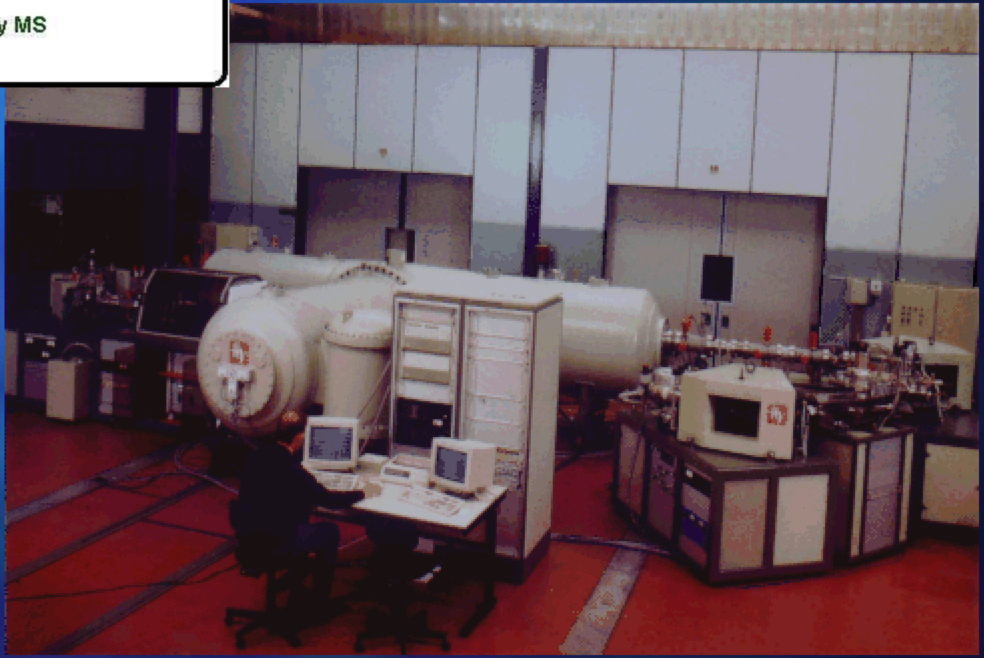
Low-energy analysing magnet
ion detector
Mass spectrometry
ion source
Tandem accelerator
Low-energy analysing magnet
High-energy analysing magnet
Stripper
Negative ion source
Electrostatic analyser
ΔE
E
Accelerator mass spectrometry
Cyclotron
Low-energy analysing magnet
Detector setup
Positive ion source

Principle of AMS



Typical AMS setup





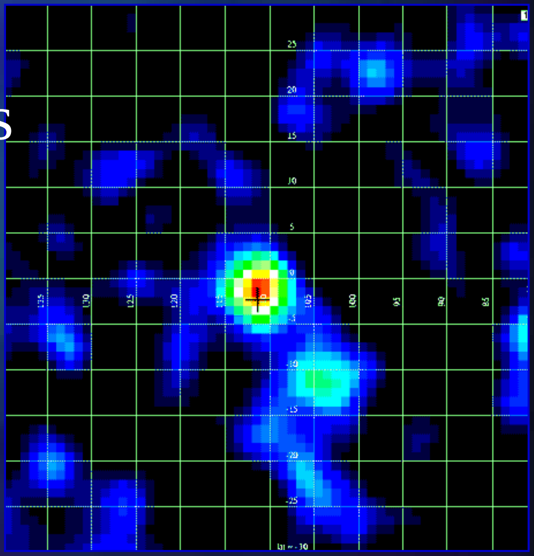
From carbon dating the Ice Man:

^{14}C age = 5300 years

To nuclear Astrophysics:

The detection of the decay of ^{44}Ti by Compton gamma-ray obs.

A clear indicator for ongoing ^{44}Ti nucleosynthesis



The measurement of the cross-section of the suspected main production channel of ^{44}Ti:
$^{40}Ca(\alpha, \gamma)^{44}Ti$

Some applications of AMS

AMS can be used in many different fields and adapted to different isotopes.

The following slides will illustrate:

- the application to environmental studies
- AMS technique developed for 2 different isotopes

AMS is however also applied to:

- Nuclear physics
  ($t_{1/2}$ measurements, cross-section,…)
- Nuclear astrophysics
- Archeology
  (^{14}C, ^{10}B,…)

Why is there a need for isotope tracers in sciences?

Natural resources on Earth are limited

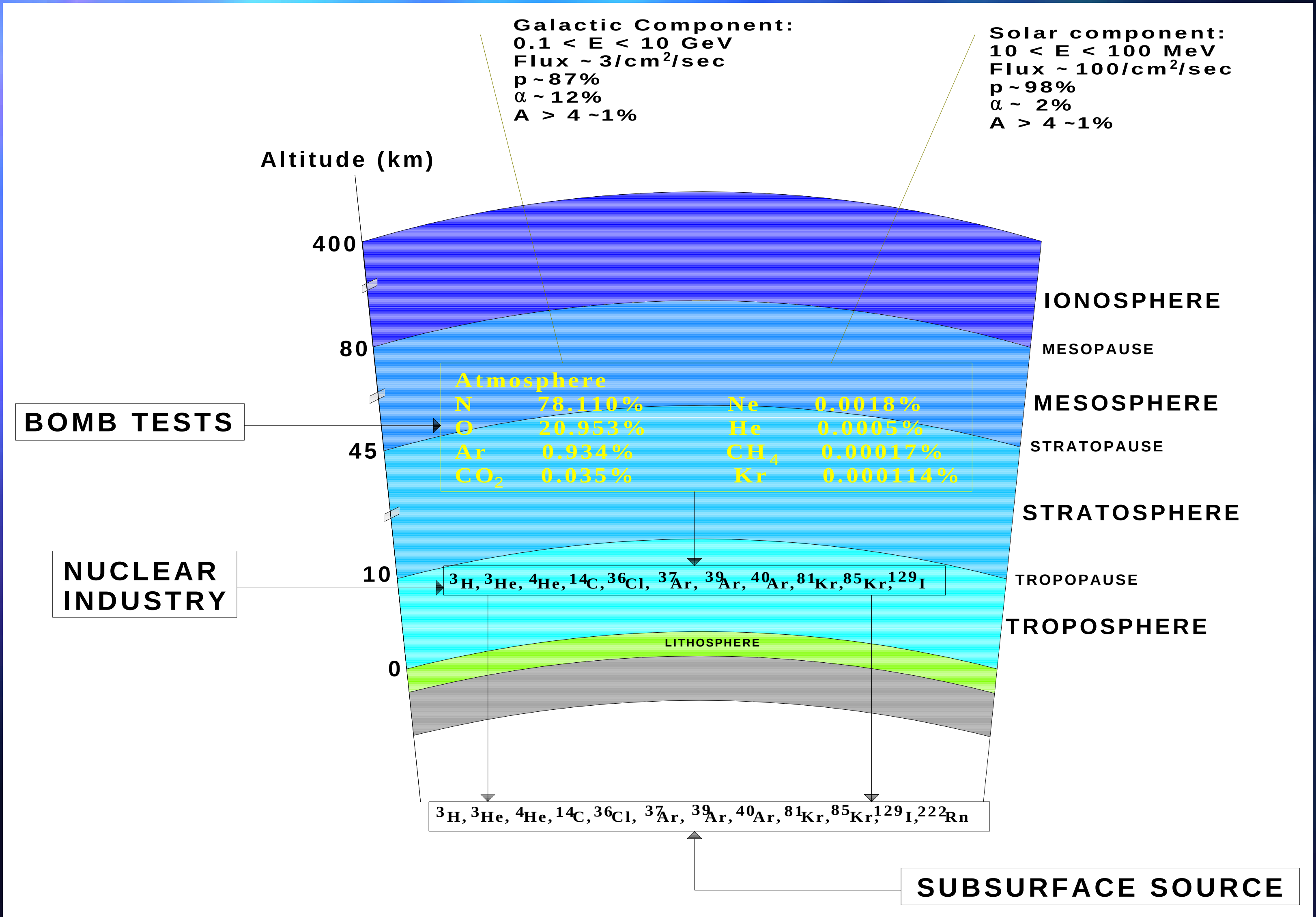
Human activities are no longer negligible

Therefore a better understanding of environmental systems is necessary

The environment is not a controlled laboratory but an extremely complex dynamical system

Special tools are needed to trace the main environmental transport processes and to determine their dynamics: **ISOTOPES**

Cosmogenic radionuclides as tracers



What potential application of these tracers makes the development of a detection technique so important - I

^{81}Kr with $t_{1/2}$ = 229,000 years, is possibly the only cosmo-genic radionuclide that has the potential to become an absolute chronometer for dating polar ice caps and old groundwater.

Properties of ^{81}Kr

$t_{1/2}$ = 230,000 years
$^{81}Kr/Kr = 5.2 \times 10^{-13}$

- Produced in the atmosphere by cosmic ray induced spallation and neutron activation of stable krypton
- The atmosphere is the only major reservoir of ^{81}Kr on earth
- The contribution from fission products are negligible
  - ^{81}Kr is shielded by stable ^{81}Br from b-decay feeding through mass-81 fission products.
  - The direct ^{81}Kr fission yield has been estimated to be as low as 7×10^{-11}

Why use AMS to measure ^{81}Kr?

The solubility of Kr in ocean water is $9.5x10^{-5}$ cm^3 STP/liter

→ 1 l of ocean water contains ~1,200 ^{81}Kr atoms

This results in 1 radioactive decay in 300 years

Low level counting on very large samples is not possible anymore due to the heavy background of the β-decay of anthropogenic ^{85}Kr

~ 50 years ago — modern

($^{85}Kr/Kr$: $3x10^{-18}$ → $1.3x10^{-11}$)

AMS for ^{81}Kr, three main difficulties

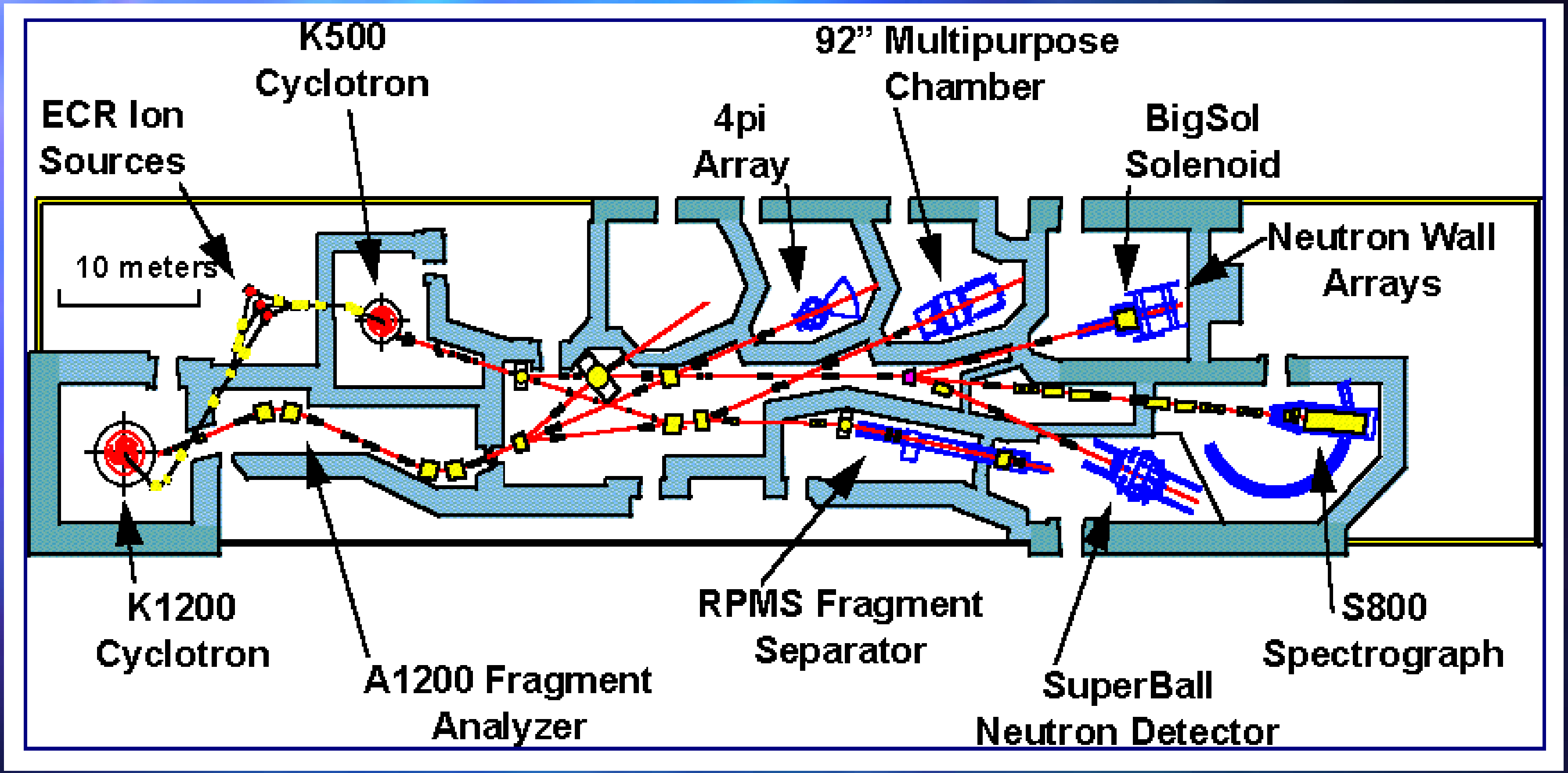
- ^{81}Kr – ^{81}Br isobar separation in the cyclotron

($\Delta_{cycl.}(M/Q)=3 \times 10^{-4}$ *vs.* $\Delta_{Isob.}(M/Q)=4 \times 10^{-6}$).

- Small gas samples to be transferred to the ion source ---→ SKIPI
- Overall transmission

(1kg of modern water or ice contains ~1500 atoms of ^{81}Kr)

MSU EXPERIMENTAL FACILITY



For the purposes of our experiment we used the Superconducting Electron Cyclotron Resonnance ion source (SCECR) coupled to the K1200 Cyclotron and the A1200 fragment analyser. Our detection system was mounted at the end of the A1200.

Experimental setup (diagram)

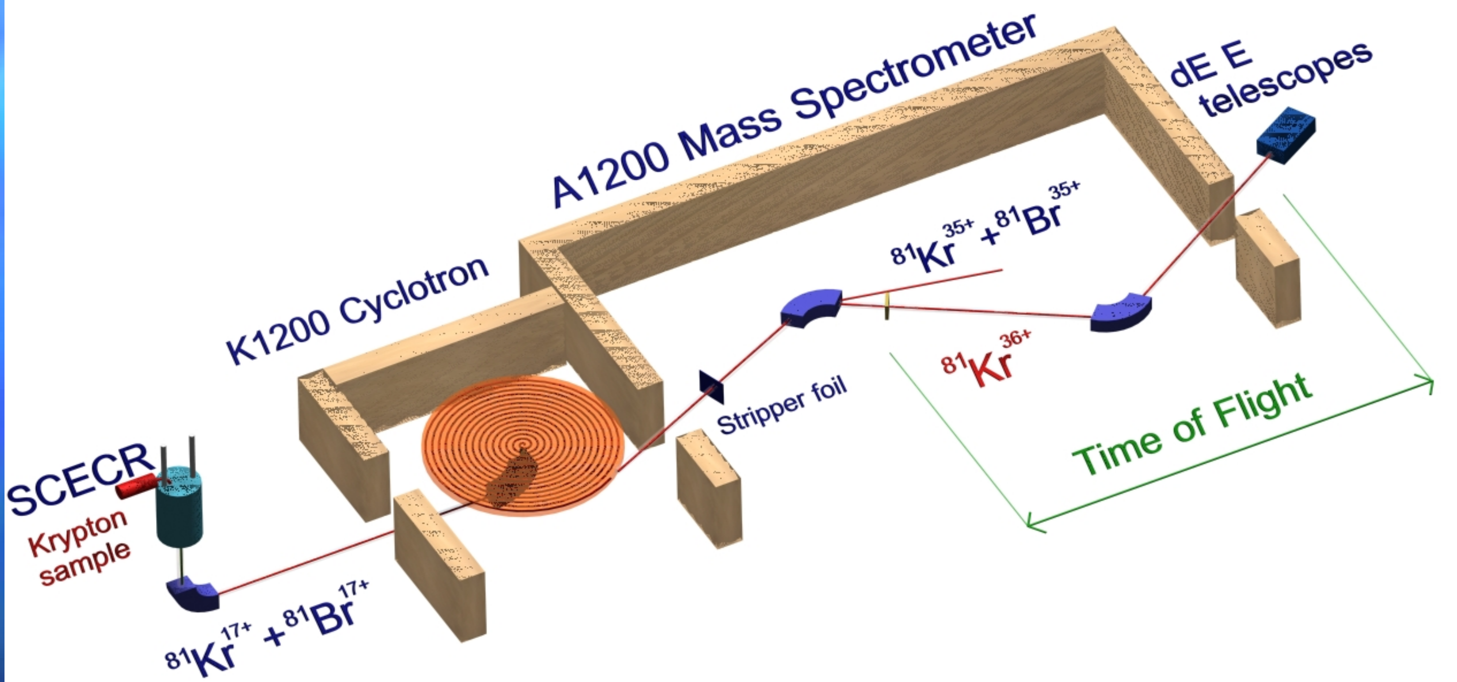


Table of the nuclides

40 2 8 18 10 2	Zr 1855° 4409° +4 91.224 3.72×10^{-8}%	Zr79	Zr80 0+	Zr81 15 s ECp	Zr82 32 s 0+ EC	Zr83 44 s (1/2-) ECp	Zr84 25.9 m 0+ EC	Zr85 7.86 m 7/2+ * EC	Zr86 16.5 h 0+ EC	Zr87 1.68 h (9/2)+ * EC	Zr88 83.4 d 0+ EC	Zr89 78.41 h 9/2+ * EC	Zr90 0+ * 51.45	Zr91 5/2+ 11.22	Zr92 0+ 17.15	Zr93 1.53E+6 y 5/2+ β^-	Zr94 0+ 17.38
Y 1522° 3345° +3 88.90585 1.51×10^{-8}%	Y77	Y78	Y79 14.8 s (5/2+) ECp	Y80 35 s (3,4,5) EC	Y81 70.4 s (5/2+) EC	Y82 9.5 s 1+ EC	Y83 7.08 m (9/2+) * EC	Y84 4.6 s 1+ * EC	Y85 2.68 h (1/2)- * EC	Y86 14.74 h 4- * EC	Y87 79.8 h 1/2- * EC	Y88 106.65 d 4- * EC	Y89 1/2- * 100	Y90 64.10 h 2- * β^-	Y91 58.51 d 1/2- * β^-	Y92 3.54 h 2- β^-	Y93 10.18 h 1/2- * β^-
Sr75 71 ms ECp	Sr76 8.9 s 0+ EC	Sr77 9.0 s (5/2+,7/2+) ECp	Sr78 2.5 m 0+ EC	Sr79 2.25 m 3/2(-) EC	Sr80 106.3 m 0+ EC	Sr81 22.3 m 1/2- EC	Sr82 25.55 d 0+ EC	Sr83 32.41 h 7/2+ * EC	Sr84 0+ 0.56	Sr85 64.84 d 9/2+ * EC	Sr86 0+ 9.86	Sr87 9/2+ * 7.00	Sr88 0+ 82.58	Sr89 50.53 d 5/2+ β^-	Sr90 28.78 y 0+ β^-	Sr91 9.63 h 5/2+ β^-	Sr92 2.71 h 0+ β^-
Rb74 64.9 ms (0+) EC	Rb75 19.0 s (3/2-,5/2-) EC	Rb76 36.5 s 1(-) EC	Rb77 3.75 m 3/2- EC	Rb78 17.66 m 0(+) * EC	Rb79 22.9 m 5/2+ EC	Rb80 34 s 1+ EC	Rb81 4.576 h 3/2- * EC	Rb82 1.273 m 1+ * EC	Rb83 86.2 d 5/2- EC	Rb84 32.77 d 2- * EC,β^-	Rb85 5/2- 72.165	Rb86 18.631 d 2- * EC,β^-	Rb87 4.75E10 y 3/2- β^- 27.835	Rb88 17.78 m 2- β^-	Rb89 15.15 m 3/2- β^-	Rb90 158 s 0- * β^-	Rb91 58.4 s 3/2(-) β^-
Kr73 27.0 s 5/2- ECp	Kr74 11.50 m 0+ EC	Kr75 4.3 m (5/2)+ EC	Kr76 14.8 h 0+ EC	Kr77 74.4 m 5/2+ EC	Kr78 0+ 0.35	Kr79 35.04 h 1/2- * EC	Kr80 0+ 2.25	Kr81 2.29E+5 y 7/2+ * EC	Kr82 0+ 11.6	Kr83 9/2+ * 11.5	Kr84 0+ 57.0	Kr85 10.756 y 9/2+ * β^-	Kr86 0+ 17.3	Kr87 76.3 m 5/2+ β^-	Kr88 2.84 h 0+ β^-	Kr89 3.15 m (3/2+,5/2+) β^-	Kr90 32.32 s 0+ β^-
Br72 78.6 s 3+ * EC	Br73 3.4 m 1/2- EC	Br74 25.4 m (0-) * EC	Br75 96.7 m 3/2- EC	Br76 16.2 h 1- * EC	Br77 57.036 h 3/2- * EC	Br78 6.46 m 1+ EC,β^-	Br79 3/2- * 50.69	Br80 17.68 m 1+ * EC,β^-	Br81 3/2- 49.31	Br82 35.30 h 5- * β^-	Br83 2.40 h 3/2- β^-	Br84 31.80 m 2- * β^-	Br85 2.90 m 3/2- β^-	Br86 55.1 s (2-) β^-	Br87 55.60 s 3/2- β^-n	Br88 16.34 s (1,2-) β^-n	Br89 4.348 s (3/2-,5/2-) β^-n
Se71 4.74 m 3/2-,5/2- EC	Se72 8.40 d 0+ EC	Se73 7.15 h 9/2+ * EC	Se74 0+ 0.89	Se75 119.779 d 5/2+ EC	Se76 0+ 9.36	Se77 1/2- * 7.63	Se78 0+ 23.78	Se79 1.13E6 y 7/2+ * β^-	Se80 0+ 49.61	Se81 18.45 m 1/2- * β^-	Se82 1.08E+20 y 0+ $\beta^-\beta^-$ 8.73	Se83 22.3 m 9/2+ * β^-	Se84 3.1 m 0+ β^-	Se85 31.7 s (5/2+) β^-	Se86 15.3 s 0+ β^-	Se87 5.29 s (5/2+) β^-n	Se88 1.53 s 0+ β^-n
As70 52.6 m 4(+) EC	As71 65.28 h 5/2- EC	As72 26.0 h 2- EC	As73 80.30 d 3/2- EC	As74 17.77 d 2- EC,β^-	As75 3/2- * 100	As76 1.0778 d 2- β^-	As77 38.83 h 3/2- β^-	As78 90.7 m 2- β^-	As79 9.01 m 3/2- β^-	As80 15.2 s 1+ β^-	As81 33.3 s 3/2- β^-	As82 19.1 s (1+) * β^-	As83 13.4 s (5/2-,3/2-) β^-	As84 4.02 s * β^-n	As85 2.021 s (3/2-) β^-n	As86 0.945 s β^-n	As87 0.48 s (3/2-) β^-n
Ge69 39.05 h 5/2- EC	Ge70 0+ 21.23	Ge71 11.43 d 1/2- * EC	Ge72 0+ 27.66	Ge73 9/2+ * 7.73	Ge74 0+ 35.94	Ge75 82.78 m 1/2- * β^-	Ge76 0+ 7.44	Ge77 11.30 h 7/2+ * β^-	Ge78 88.0 m 0+ β^-	Ge79 18.98 s (1/2)- * β^-	Ge80 29.5 s 0+ β^-	Ge81 7.6 s (9/2+) * β^-	Ge82 4.60 s 0+ β^-	Ge83 1.85 s (5/2+) β^-	Ge84 966 ms 0+ β^-n	Ge85 535 ms β^-n	Ge86 0+
Ga68 67.629 m 1+ EC	Ga69 3/2- 60.108	Ga70 21.14 m 1+ EC,β^-	Ga71 3/2- 39.892	Ga72 14.10 h 3- * β^-	Ga73 4.86 h 3/2- β^-	Ga74 8.12 m (3-) * β^-	Ga75 126 s 3/2- β^-	Ga76 32.6 s (2+,3+) β^-	Ga77 13.2 s (3/2-) β^-	Ga78 5.09 s (3+) β^-	Ga79 2.847 s (3/2-) β^-n	Ga80 1.697 s (3) β^-n	Ga81 1.217 s (5/2-) β^-n	Ga82 0.599 s (1,2,3) β^-n	Ga83 0.31 s β^-n	Ga84 85 ms β^-n	54
Zn67 5/2- 4.1	Zn68 0+ 18.8	Zn69 56.4 m 1/2- * β^-	Zn70 5E+14 y 0+ 0.6	Zn71 2.45 m 1/2- * β^-	Zn72 46.5 h 0+ β^-	Zn73 23.5 s (1/2)- * β^-	Zn74 95.6 s 0+ β^-	Zn75 10.2 s (7/2+) β^-	Zn76 5.7 s 0+ β^-	Zn77 2.08 s (7/2+) * β^-	Zn78 1.47 s 0+ β^-	Zn79 995 ms (9/2+) β^-n	Zn80 0.545 s 0+ β^-n	Zn81 0.29 s β^-n	Zn82 0+		

Main problem: reduce the ^{81}Br intensity

$^{81}Kr^{17+} + {}^{81}Br^{17+}$ → Stripper foil (Be 18.8 mg/cm^2)

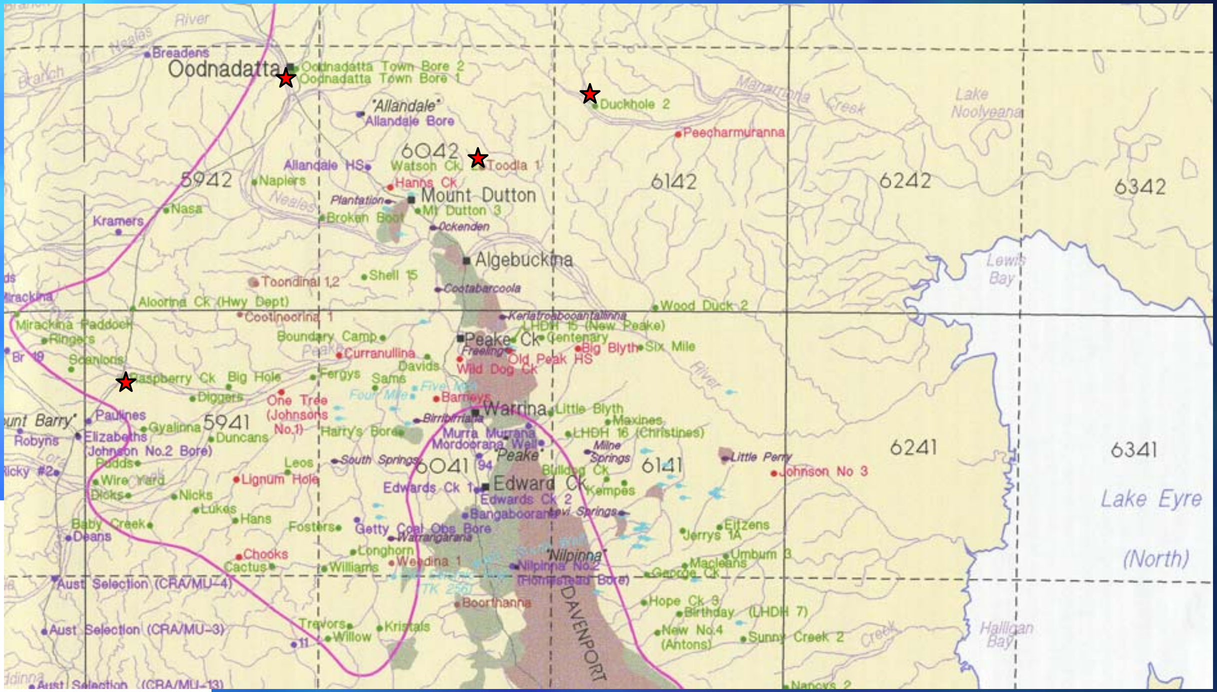
A1200

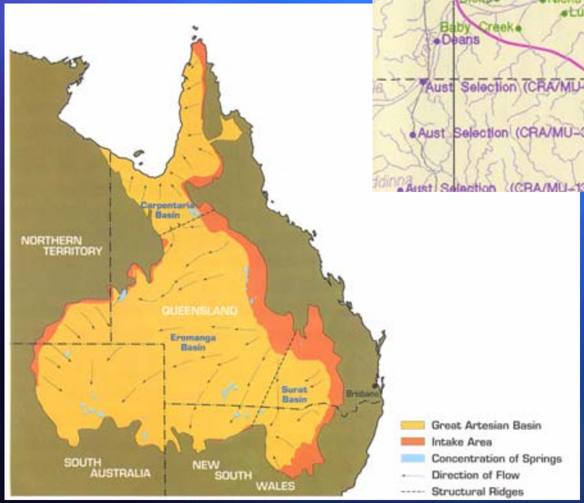
$^{81}Br^{35+}$

$\Sigma\ {}^{81}Kr\ ({}^{81}Kr_{primary} + {}^{81}Kr_{react.\ Prod})$

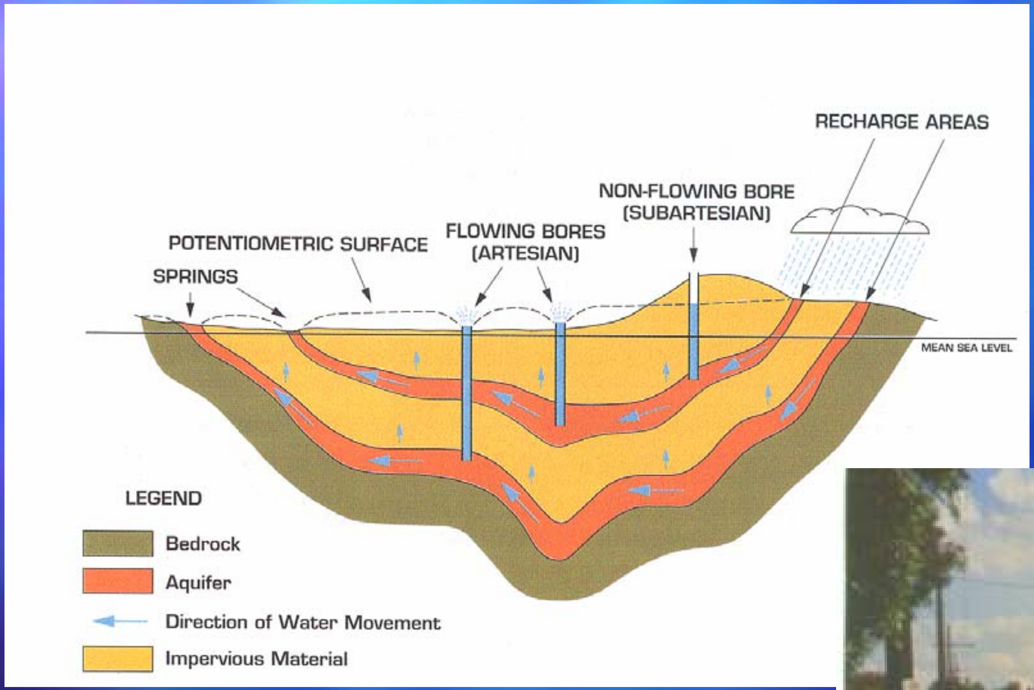
In order to reduce this factor, a substantial reduction in the Br Background intensity in the beam must be made

Dating water from the Great Artesian Basin of Australia





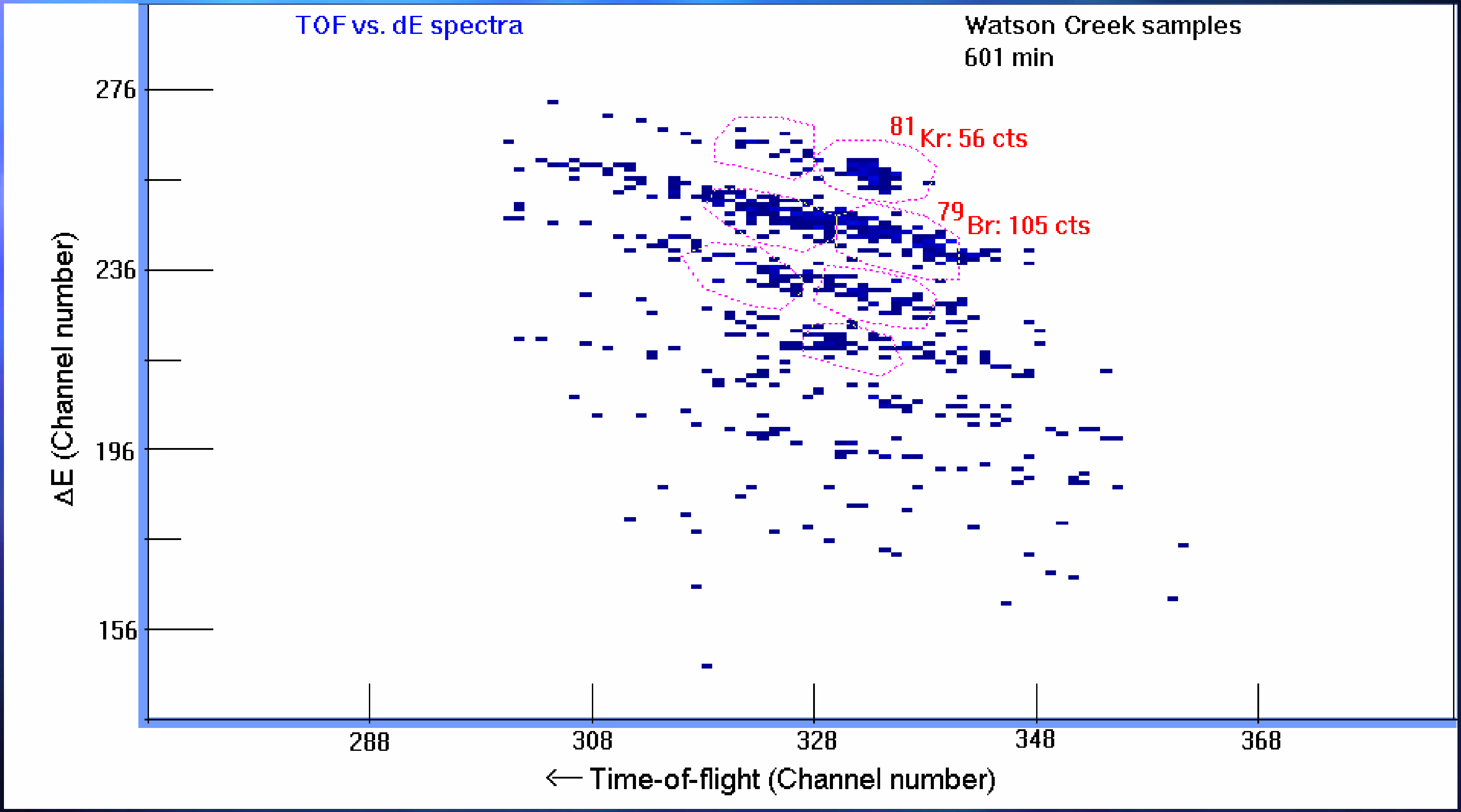
The GAB sampling trip



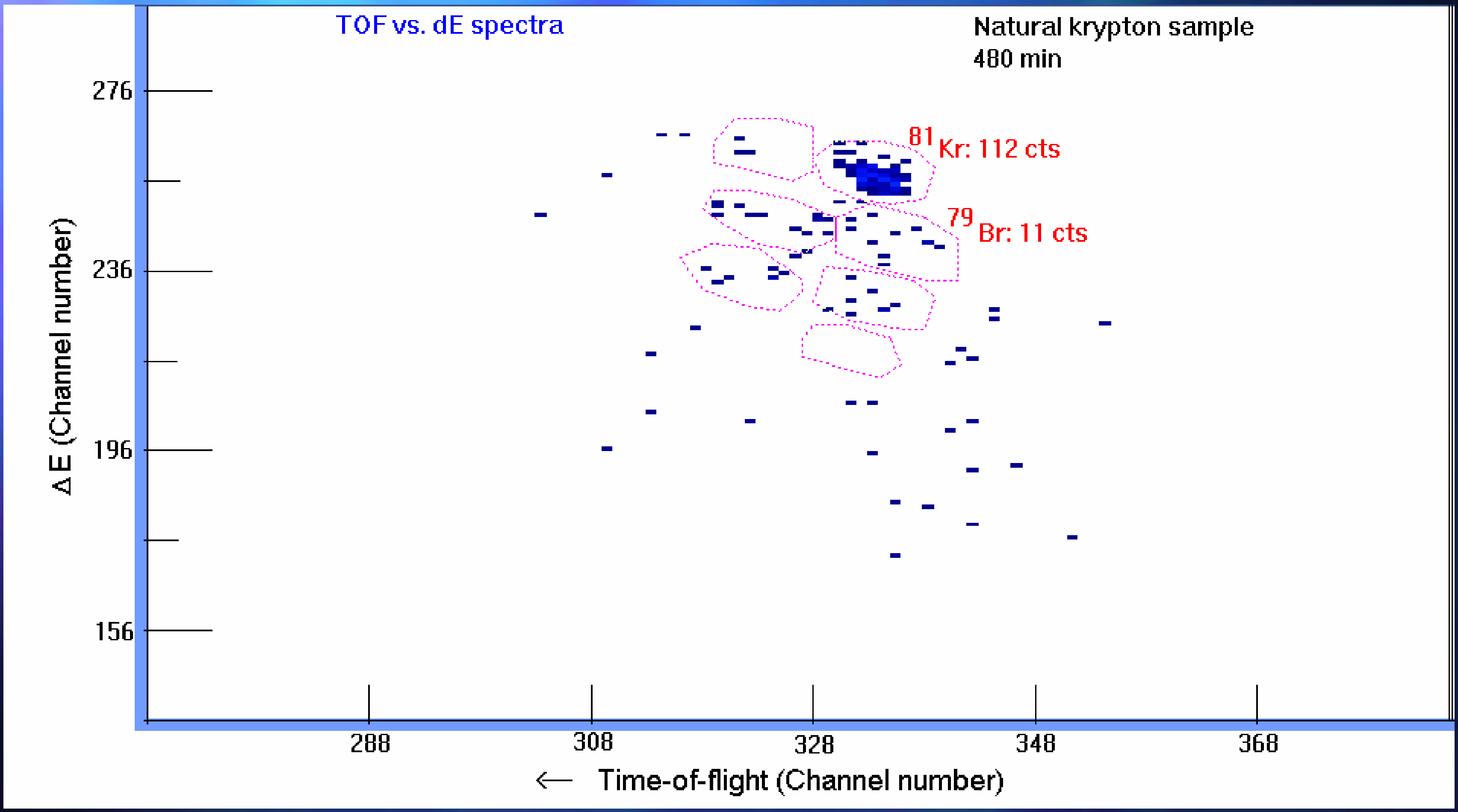
Water sampling

Oh the hard life of the scientist

Groundwater sample (Watson Creek)



Natural Krypton sample



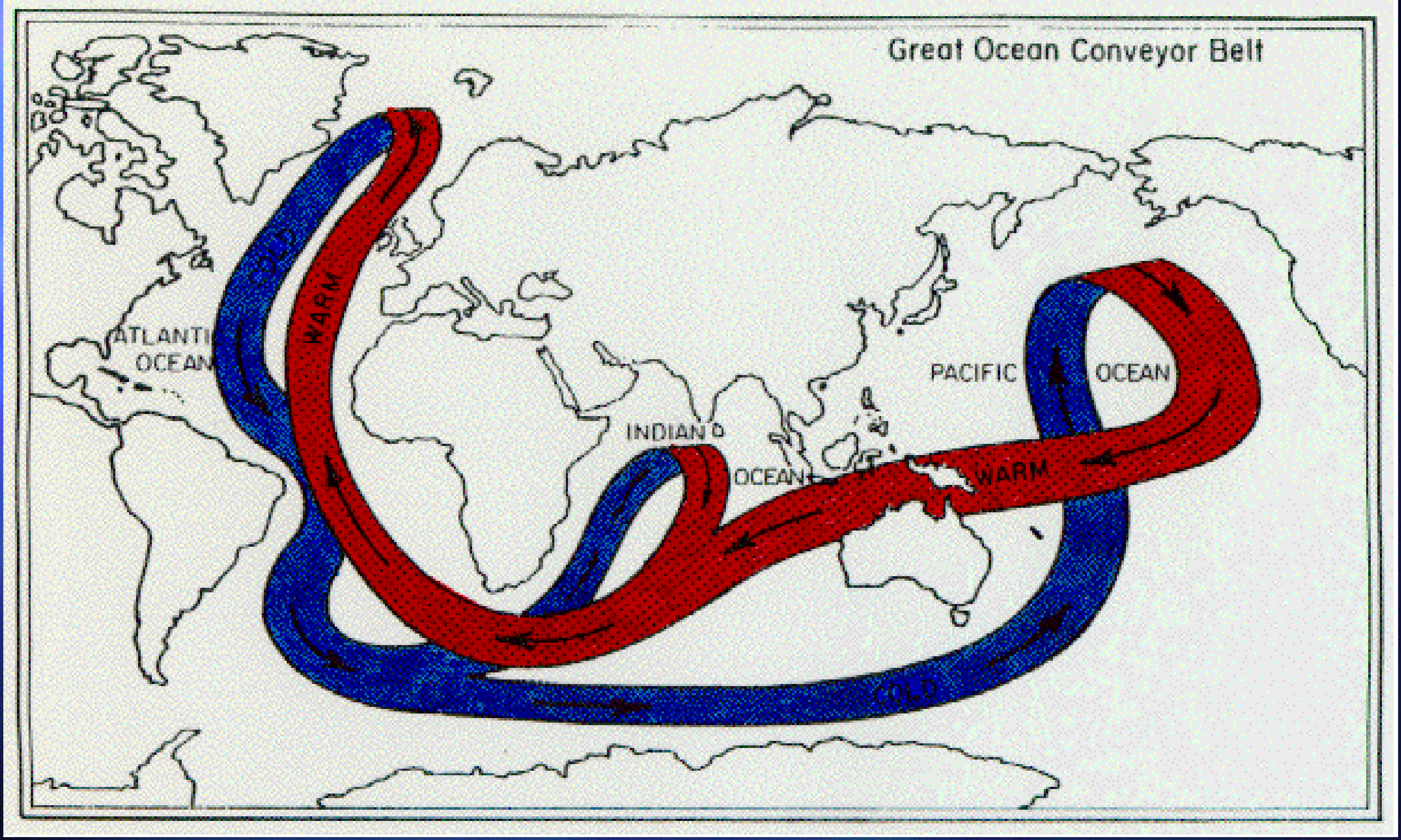
Experimental results

	Age (years)	Uncertainty	
Atmosphere	0		
Raspberry Creek	225,000	±42,000	±(12.7% * τ)
Oodnadatta	354,000	±50,500	±(15.3% * τ)
Duck Hole	287,000	±44,200	±(13.4% * τ)
Watson Creek	402,000	±51,000	±(15.4% * τ)

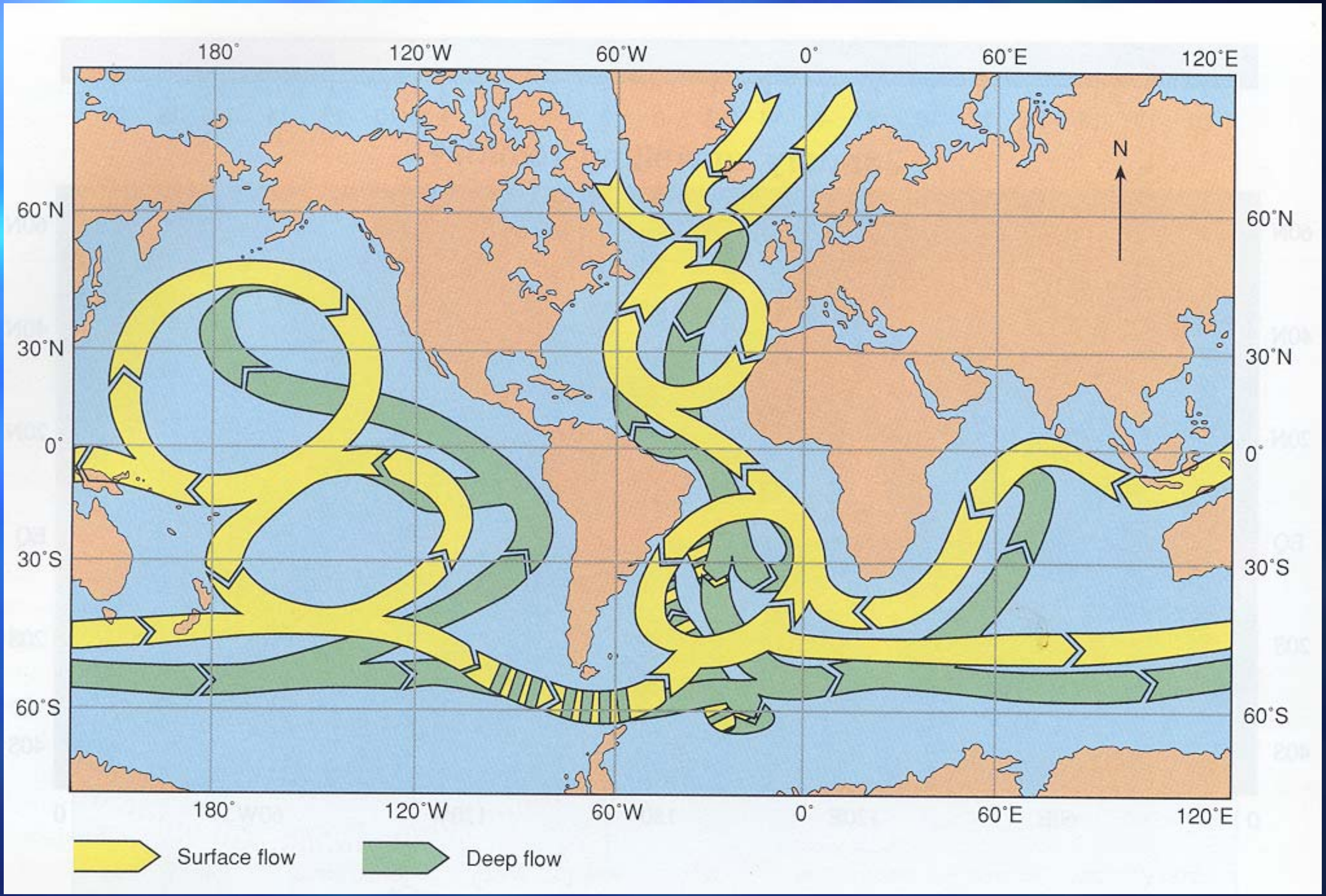
What potential application of these tracers makes the development of a detection technique so important - II

^{39}Ar with $t_{1/2}$ = 269 years, it is particularly well suited to study the "Great Atlantic conveyor belt" with its cycle of 1000 years

Atlantic conveyor belt circulation



Concept of the Conveyor belt



The application of ^{39}Ar dating to groundwater is limited by the fact that underground production in granitic rock $^{39}K(n,p)^{39}Ar$ can be substantial.

Properties of ^{39}Ar

$t_{1/2}$ = 269 years
$^{39}Ar/Ar = 8.1 \times 10^{-16}$

- Mainly produced through cosmic ray induced spallation on argon in the atmosphere $^{40}Ar(n, 2n)^{39}Ar$ Q= -9.87 MeV
- Anthropogenic production is estimated to be below 5% [Loosli 1983]
- Subsurface production can be significant in rocks with high uranium content $^{39}K(n,p)^{39}Ar$

Activity of 1 l water

- 1 l ocean sea water contains ~ 6500 ^{39}Ar atoms (In ocean water: Ar solubility ≡ 0.4 cm^3 STP/l)
- $Activity_{(t=0)}$ $=5.3 \times 10^{-7}$ Bq or ~17 decays per year.

This tends to make statistics rather poor

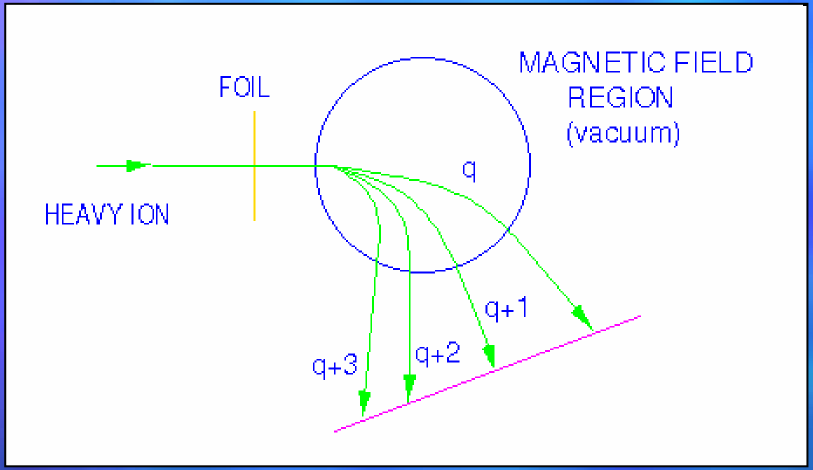
How can ^{39}Ar be counted?

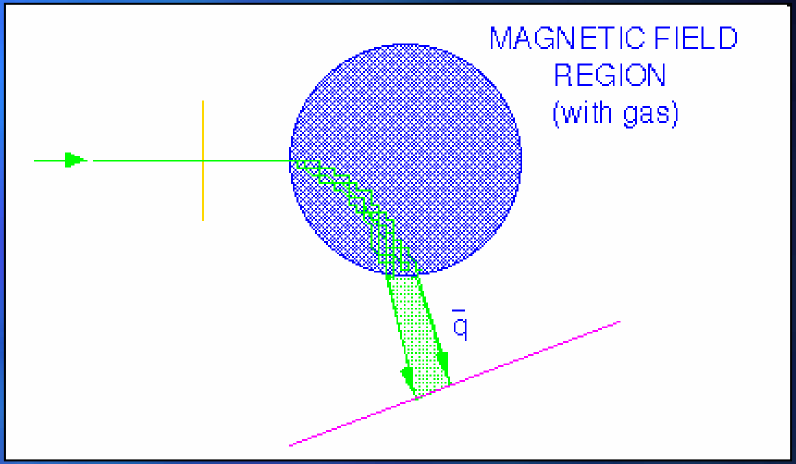
- Low Level Counting
  - Possible on large samples (~1000 l), done by H.H. Loosli in Bern
- Laser
  - The extremely low concentration makes this a very difficult isotope for laser techniques
- AMS (with small vol. samples)
  - several difficulties ($\Delta M/M$, low concentration, ...)

AMS for ^{39}Ar

- 5 Main difficulties
  - The $^{39}Ar/Ar = 8.1 \times 10^{-16}$ ratio
  - Isobar separation between ^{39}K and ^{39}Ar ($\Delta M/M = 1.55 \times 10^{-5}$)
  - A tandem (as used in traditional AMS lab) cannot be used for noble gasses
  - Source efficiency
  - Overall transmission

Principle of the gas filled magnet

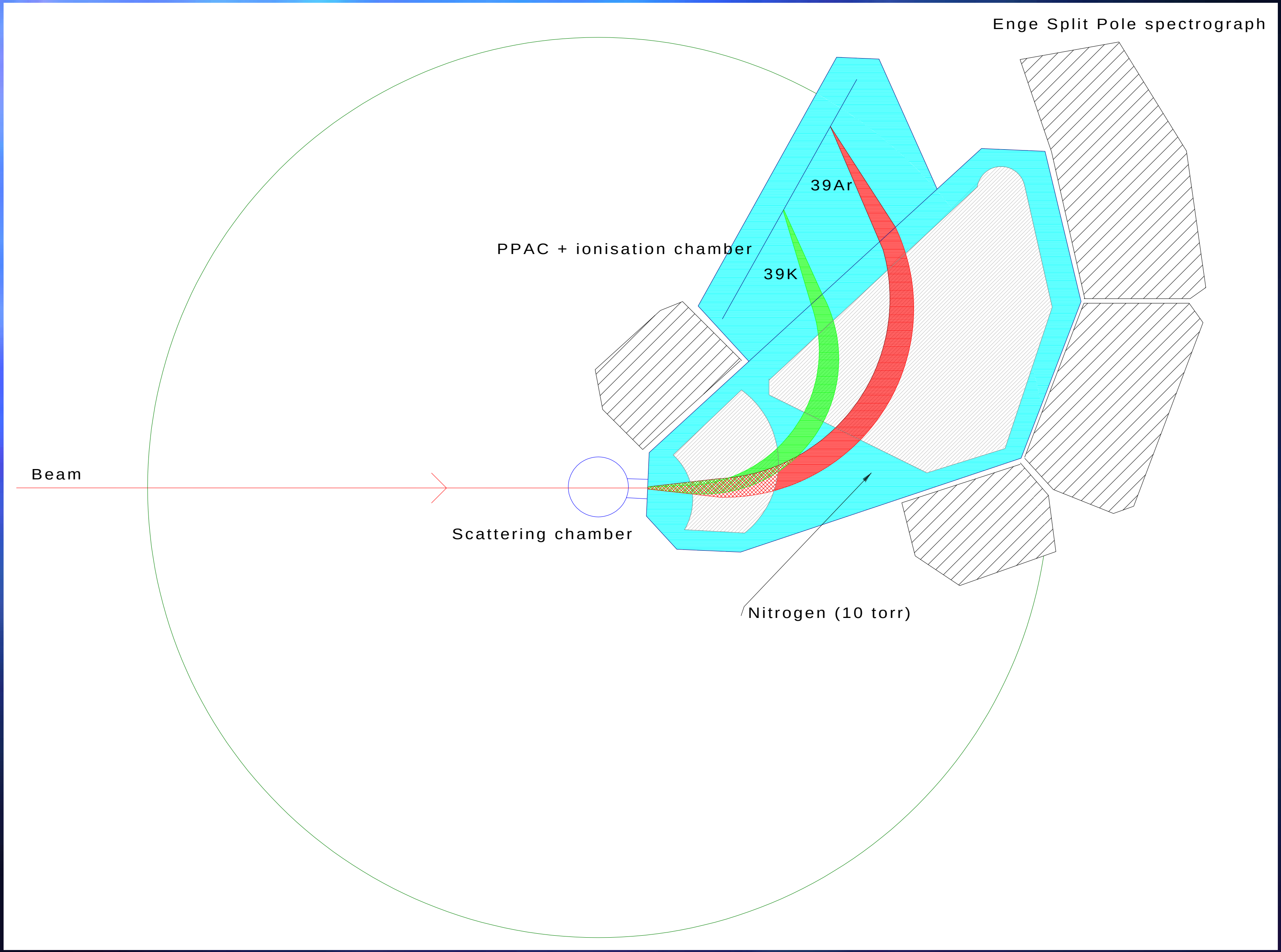




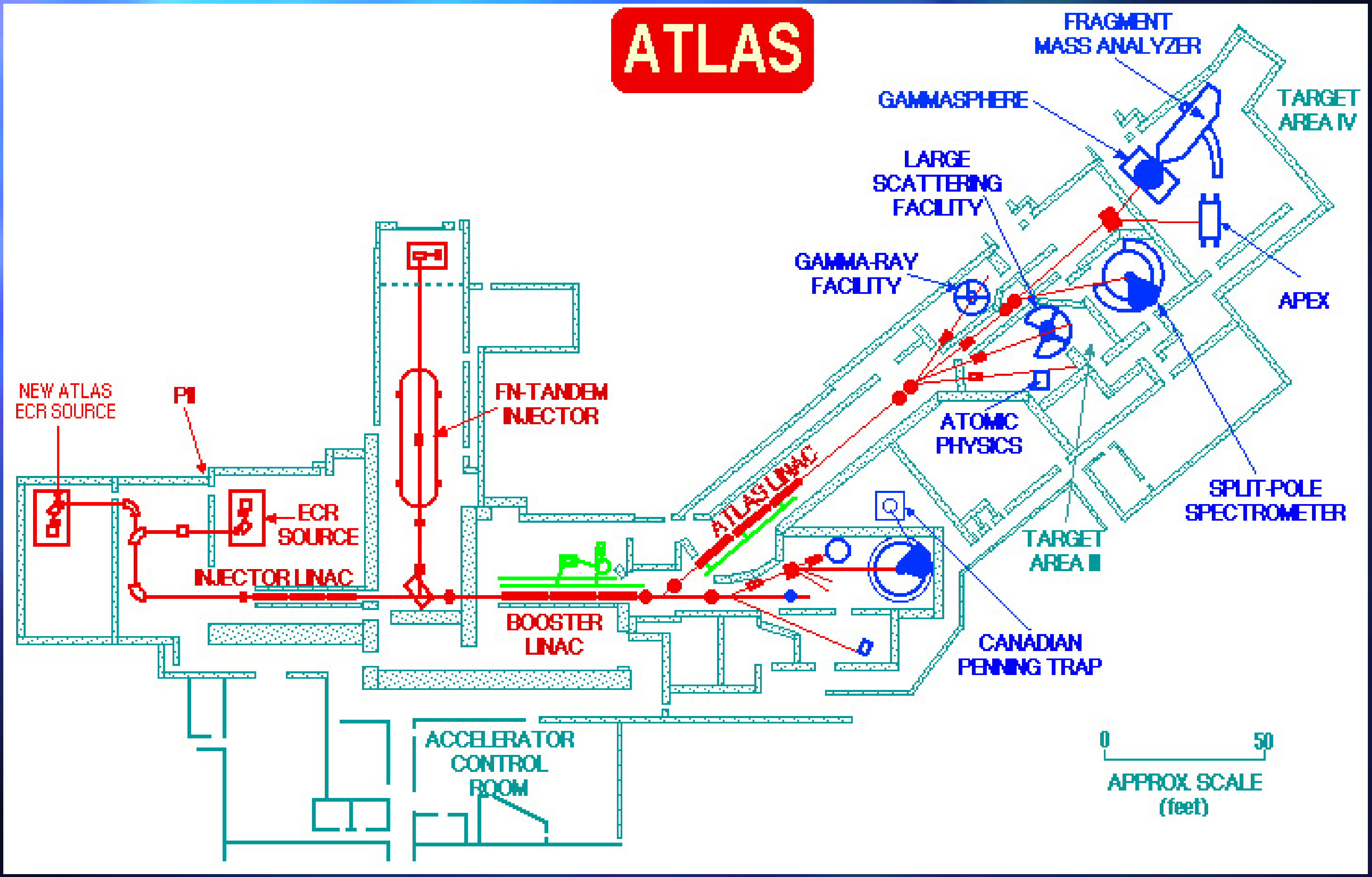
In the gas filled magnetic region, the discreet charge states coalesce around a trajectory defined by the mean charge state of the ion in the gas

$$B\rho \propto mv / \bar{q}$$

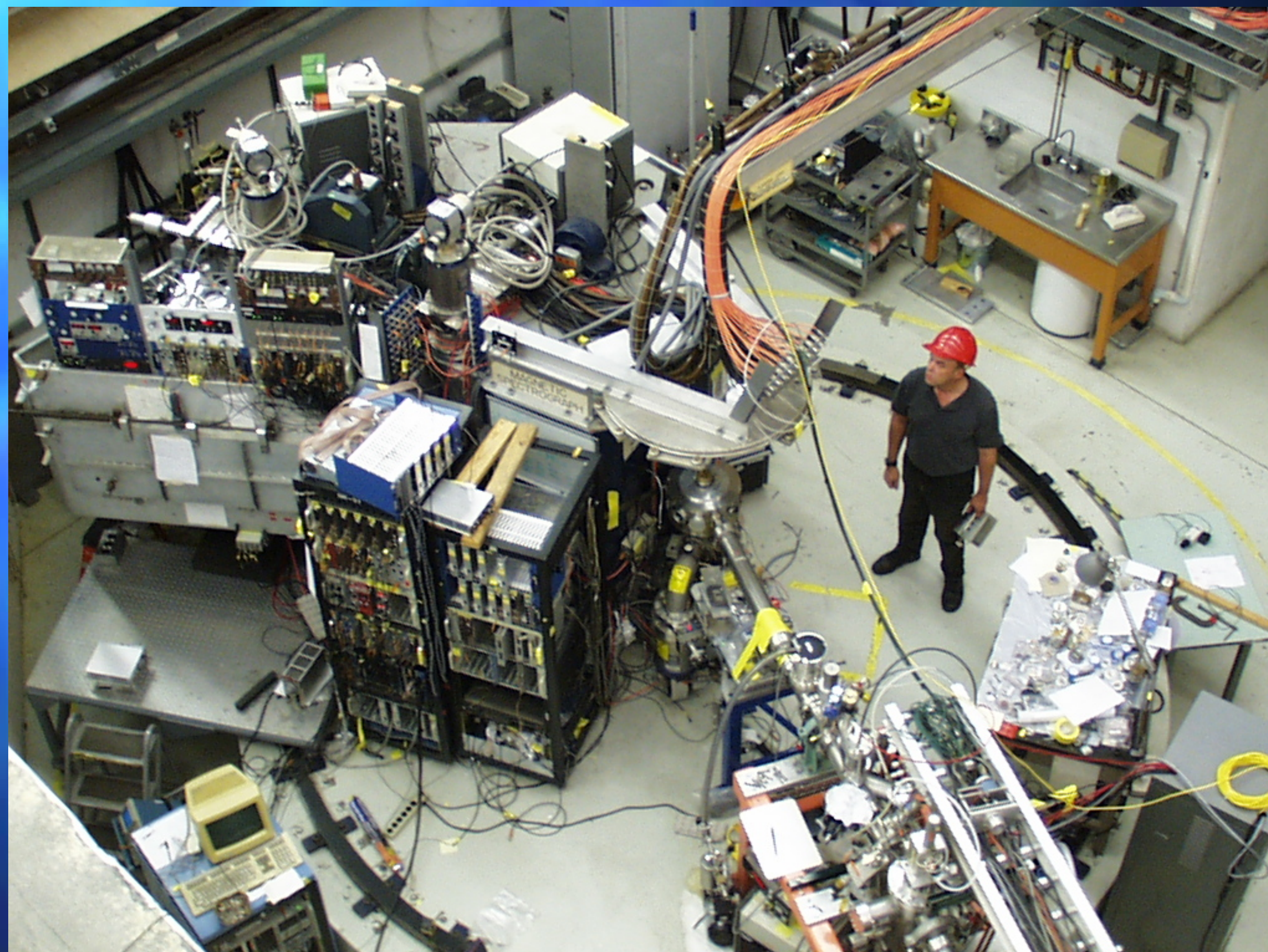
Gas filled magnet setup



ATLAS layout



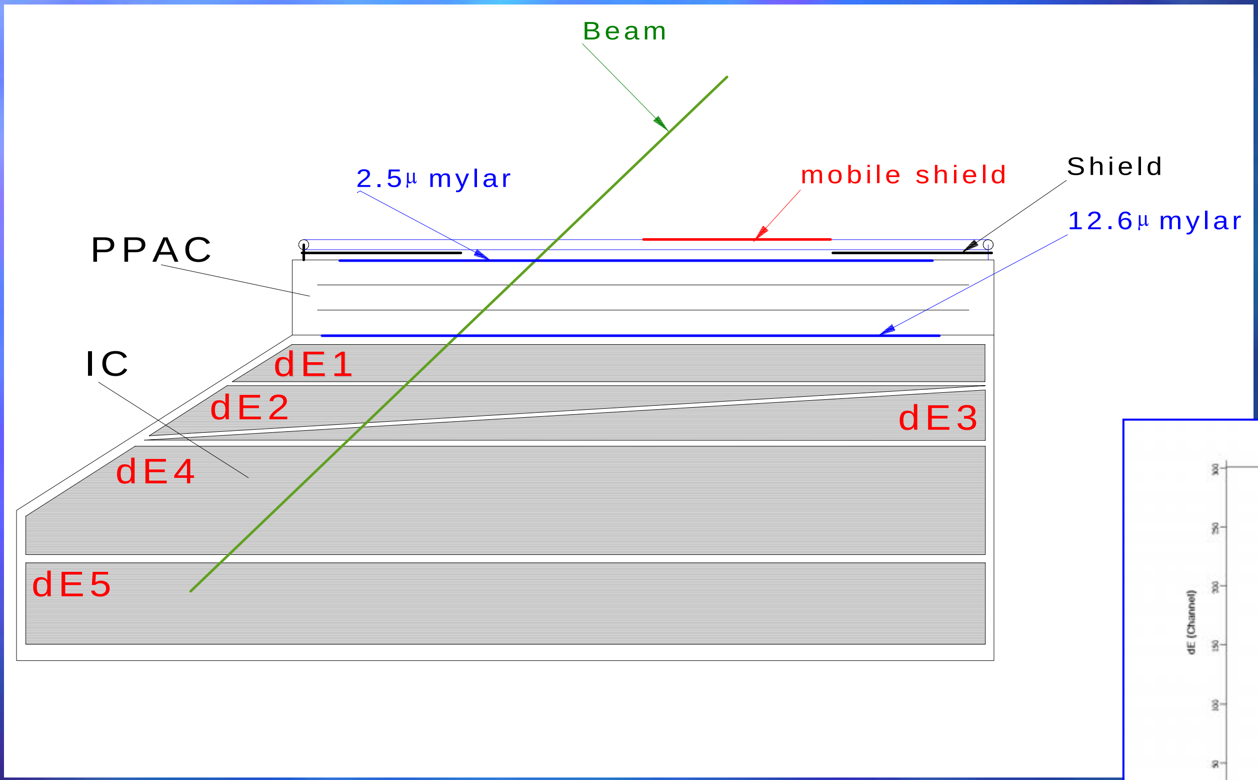
Split-Pole Enge Spectrograph

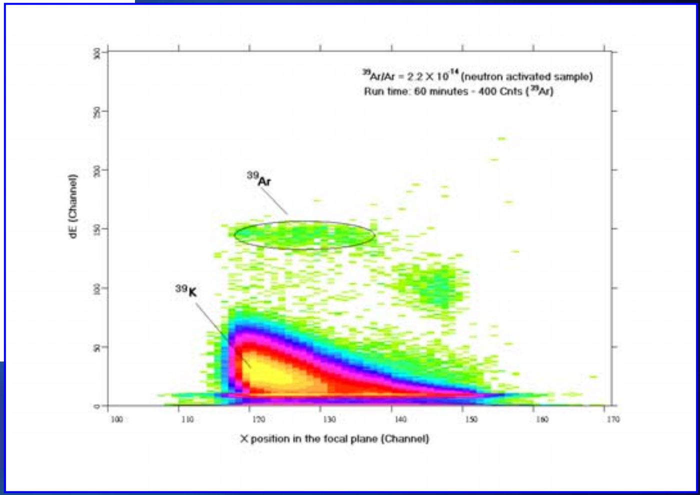


Experimental setup I

- Initial beam tuning
  - As it is not possible to tune on $^{39}Ar^{8+}$ it was decided to use as pilot beam : $^{78}Kr^{16+}$ from the ECR source
- Beam energy
  - $^{78}Kr^{16+}$ Energy: 464 MeV resulting in a $^{39}Ar^{8+}$ beam with 232 MeV
- Total transmission ~20% (without stripping)

Later detector set-up





Beam:

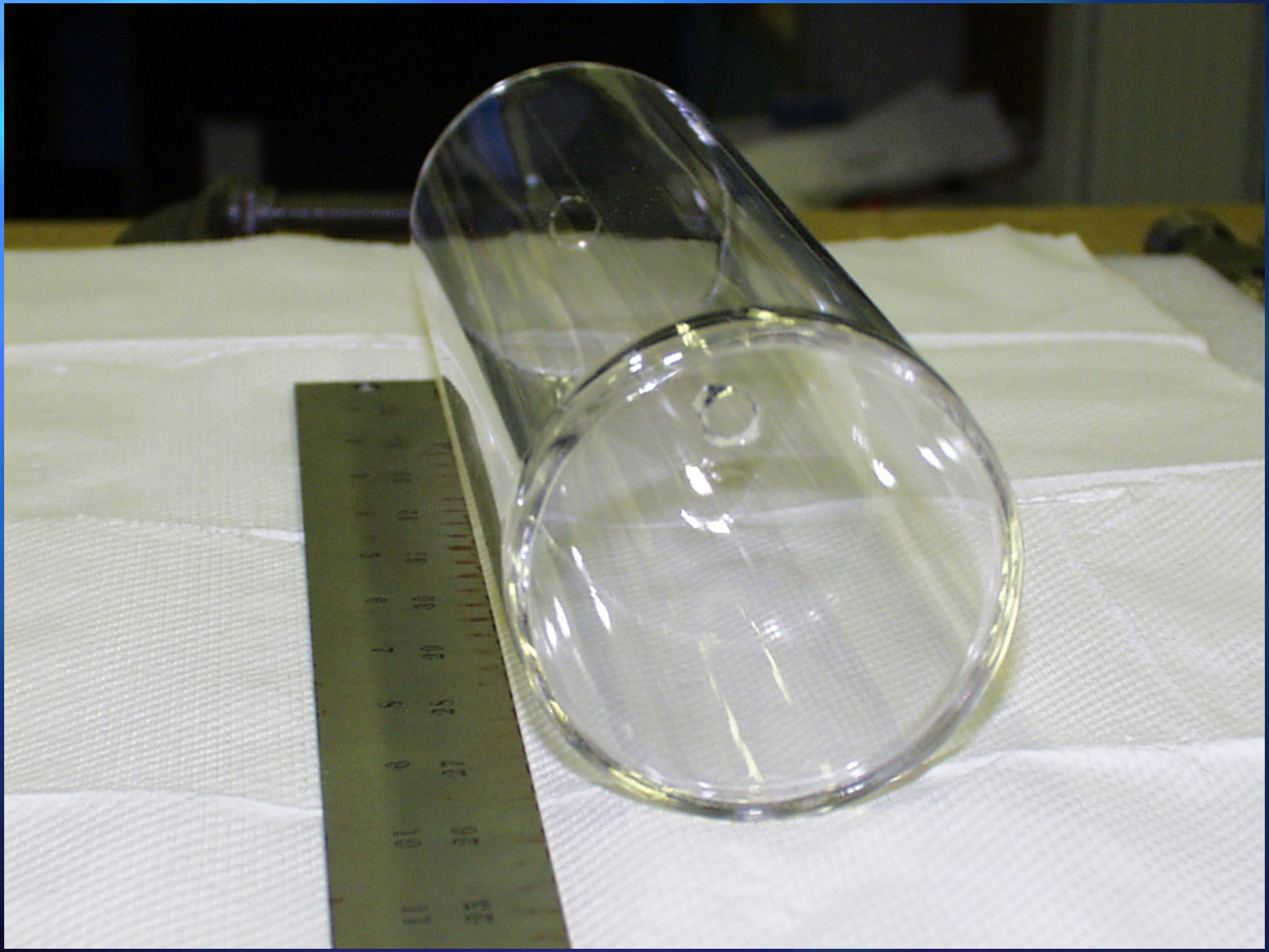
Pπ = 113.6 MeV
Booster = 348.8 MeV
ATLAS = 464 MeV

Detect:

N2 = 12.1 Torr
PPAC = 3 torr (Isob)
IC = 21 torr (Isob)

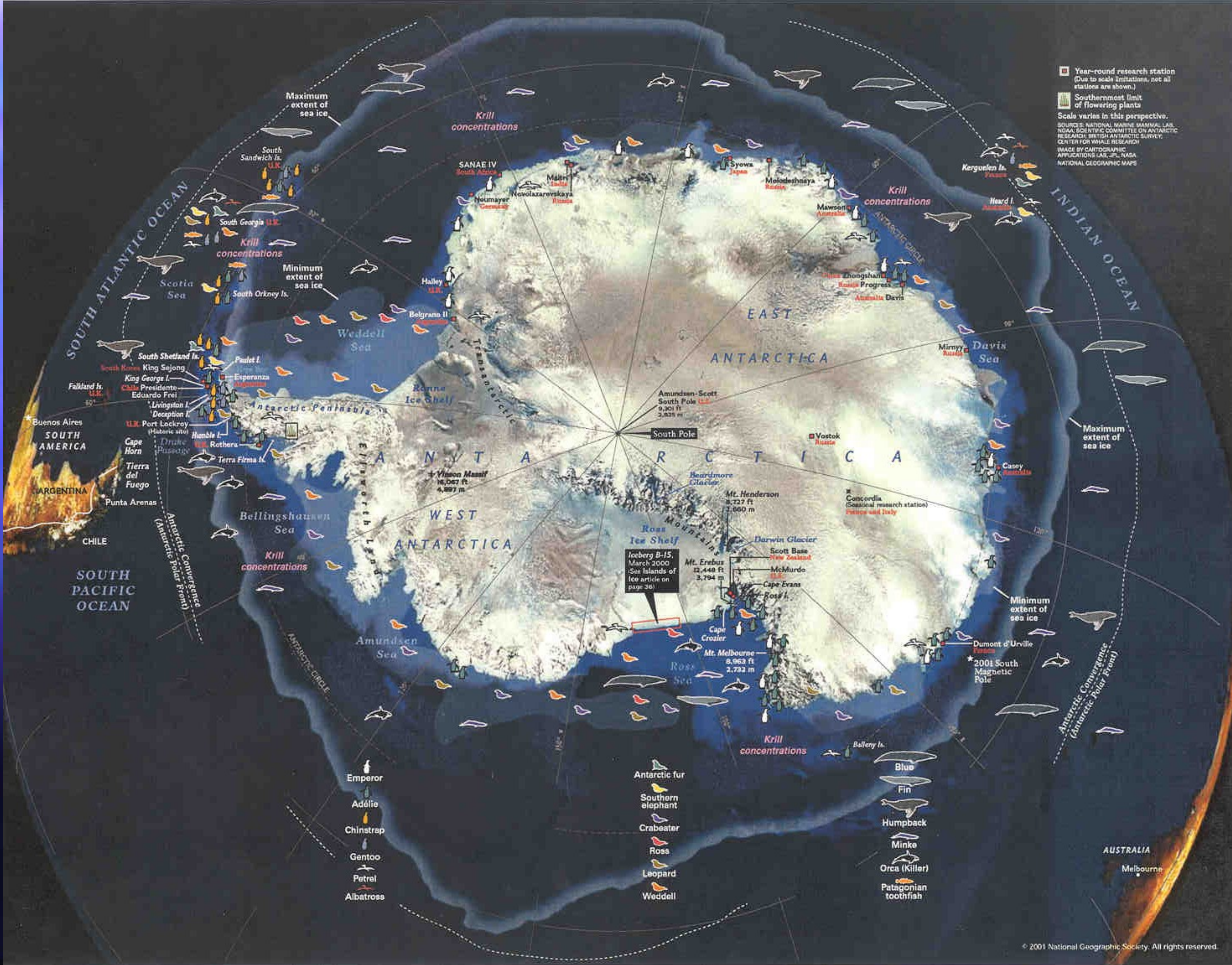
Cath: - 430 V
Anode: + 575 V
Grid: + 300
Div: +240V / -365V

Using a quartz liner in the plasma chamber



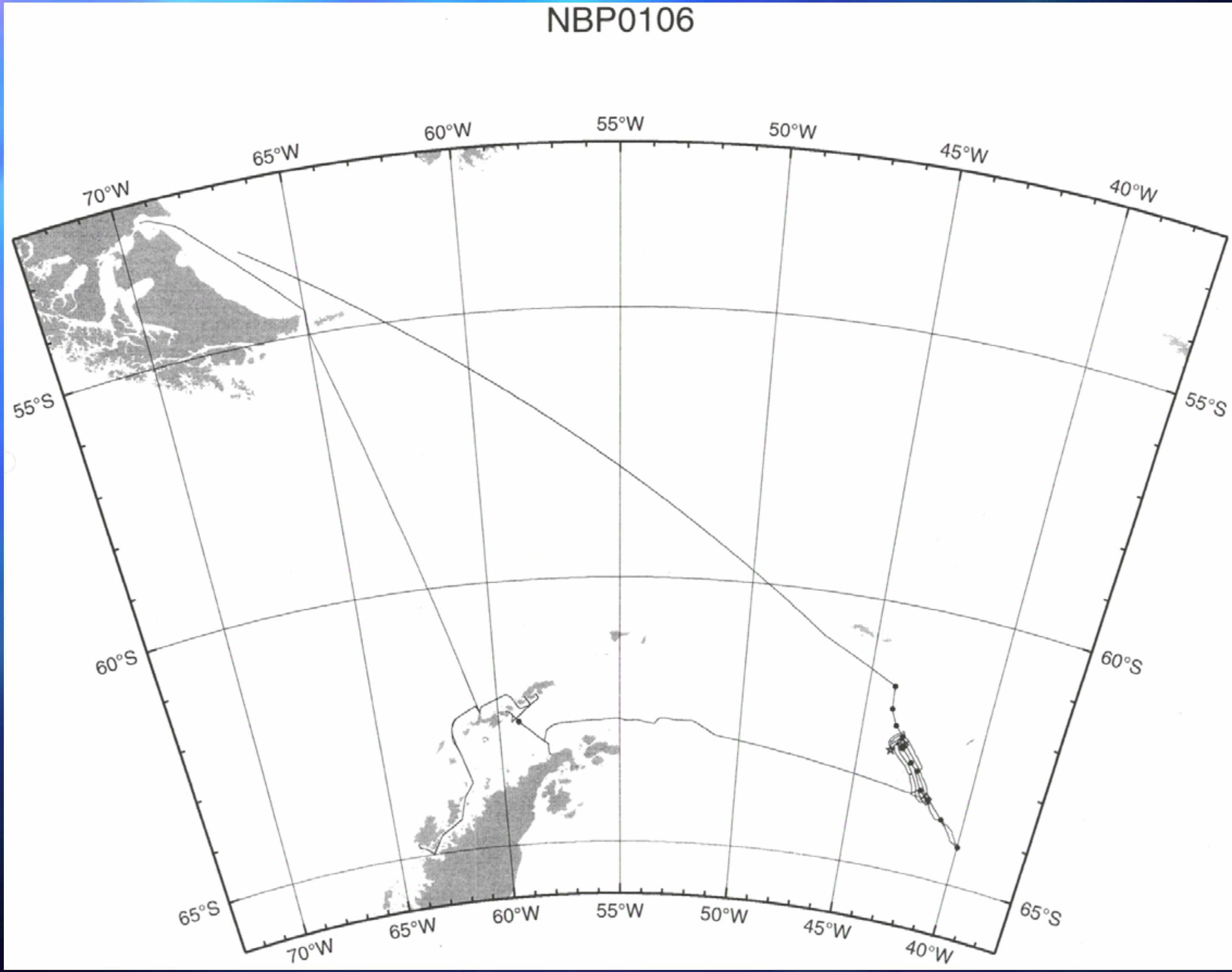
How to sample ocean water?

R/V Nathaniel B. Palmer



Year-round research station (Due to scale limitations, not all stations are shown.)
Southernmost limit of flowering plants
Scale varies in this perspective.
SOUTH ATLANTIC OCEAN
INDIAN OCEAN
SOUTH PACIFIC OCEAN
Maximum extent of sea ice
Minimum extent of sea ice
Krill concentrations
South Sandwich Is. U.K.
South Georgia U.K.
Scotia Sea
South Orkney Is.
South Shetland Is.
Weddell Sea
Halley U.K.
Belgrano II Argentina
SANAE IV South Africa
Neumayer Germany
Novolazarevskaya Russia
Maitri India
Syowa Japan
Molodezhnaya Russia
Mawson Australia
ANTARCTIC CIRCLE
Zhongshan
Progress
Davis
Kerguelen Is. France
Heard I. Australia
EAST ANTARCTICA
Mirnyy Russia
Davis Sea
Amundsen-Scott South Pole U.S.
South Pole
Vostok Russia
ANTARCTICA
Casey Australia
Concordia (Seasonal research station) France and Italy
Beardmore Glacier
Mt. Henderson
Darwin Glacier
Scott Base New Zealand
McMurdo U.S.
Cape Evans
Ross I.
Mt. Erebus
Cape Crozier
Mt. Melbourne
Ross Ice Shelf
Ross Sea
Iceberg B-15, March 2000 (See Islands of Ice article on page 36)
Transantarctic Mountains
Dumont d'Urville France
2001 South Magnetic Pole
Antarctic Convergence (Antarctic Polar Front)
Balleny Is.
Ronne Ice Shelf
Antarctic Peninsula
Ellsworth Land
Vinson Massif
WEST ANTARCTICA
Bellingshausen Sea
Amundsen Sea
Falkland Is. U.K.
Buenos Aires
SOUTH AMERICA
ARGENTINA
CHILE
Cape Horn
Tierra del Fuego
Punta Arenas
Drake Passage
King George I.
South Korea King Sejong
Chile Presidente Eduardo Frei
Esperanza Argentina
Paulet I.
Livingston I.
Deception I.
U.K. Port Lockroy (Historic site)
Humble I.
U.K. Rothera
Terra Firma Is.
AUSTRALIA
Melbourne
Emperor
Adélie
Chinstrap
Gentoo
Petrel
Albatross
Antarctic fur
Southern elephant
Crabeater
Ross
Leopard
Weddell
Blue
Fin
Humpback
Minke
Orca (Killer)
Patagonian toothfish

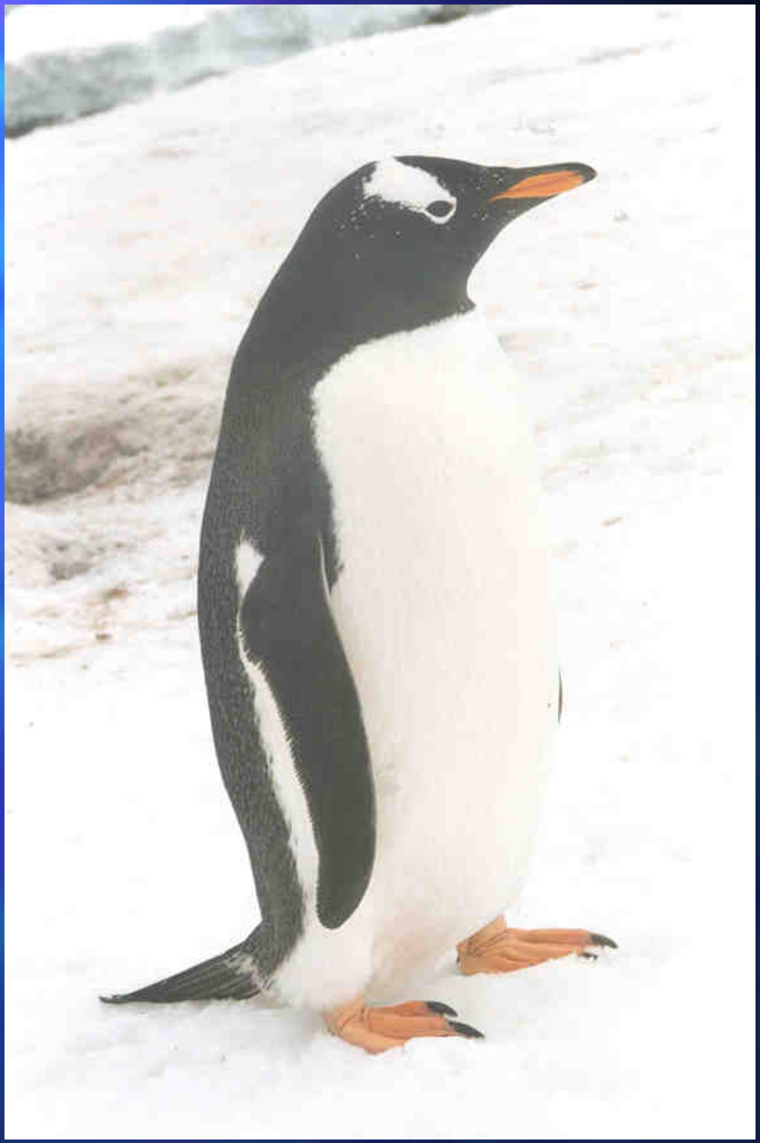
Nathaniel B.Palmer cruise 0106



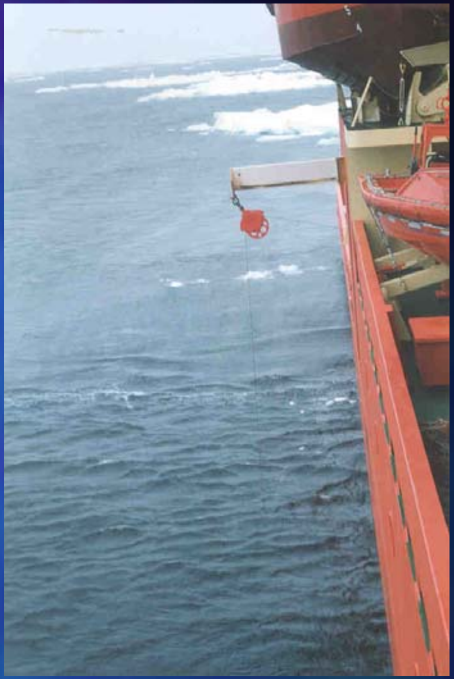

CRUNCHER

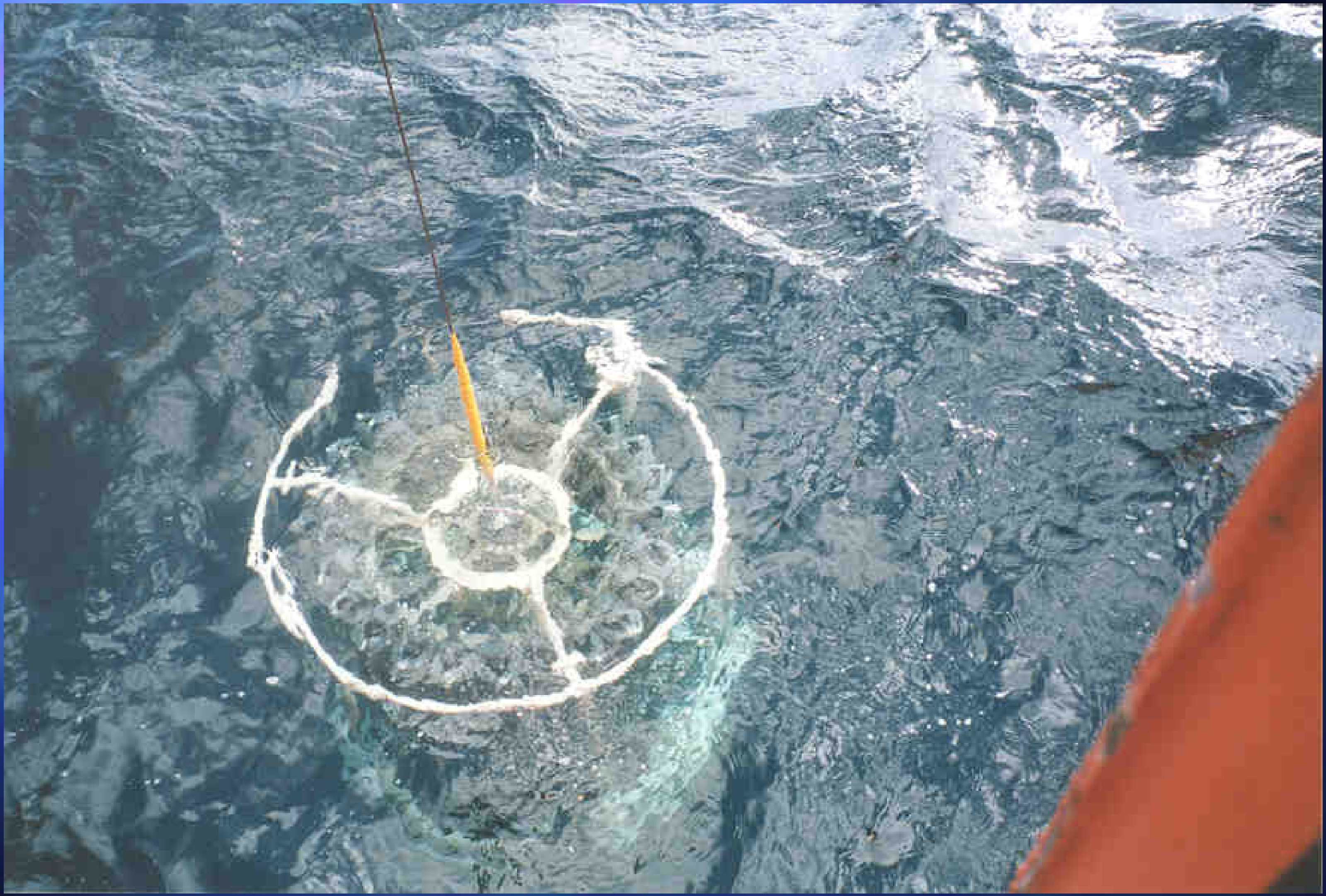

SANTIAGO
3755
ARICA
PTA. ARENAS
1325
POLO NORTE
POLO SUR
MAC MURDO
CAMBERRA


93

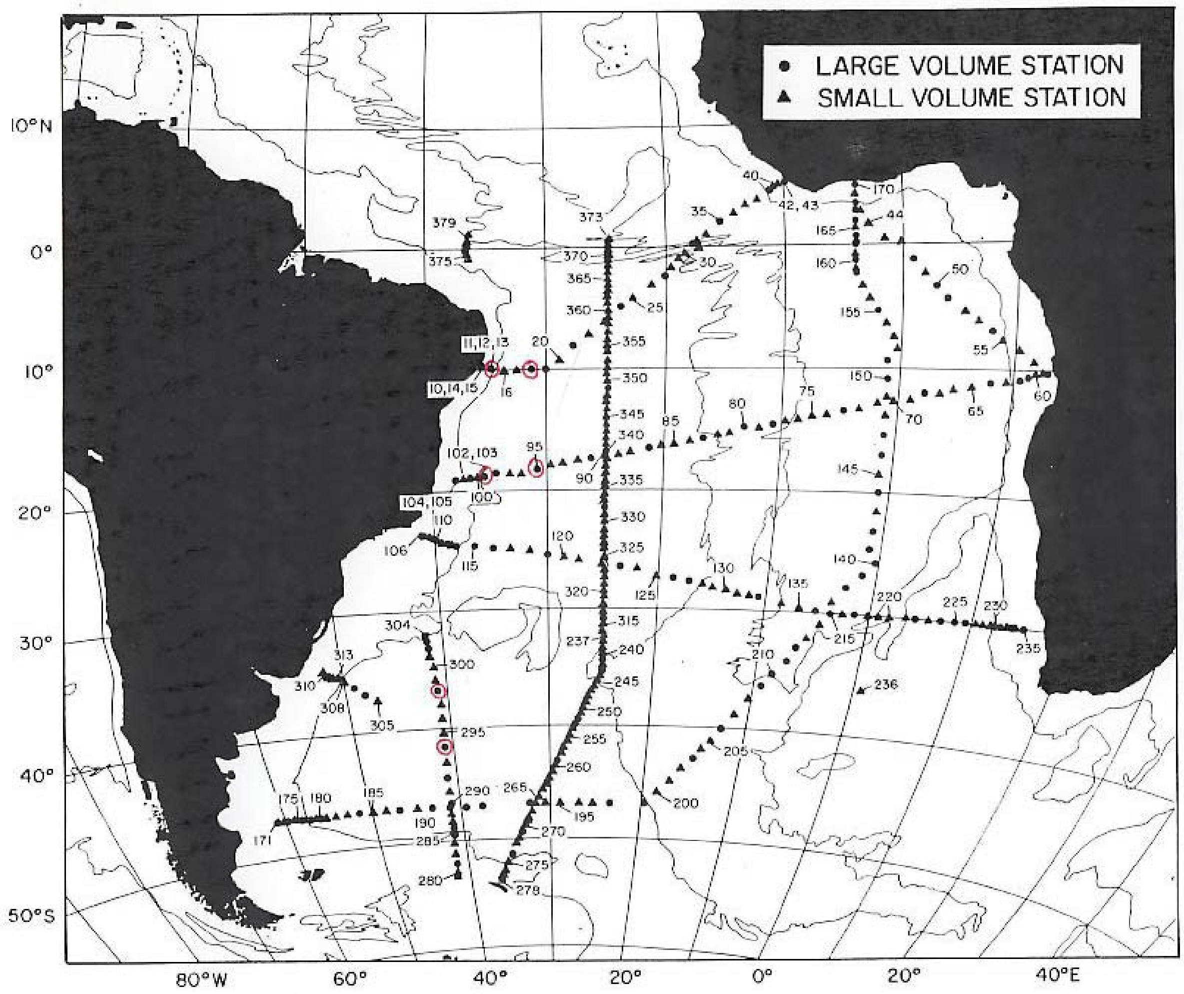


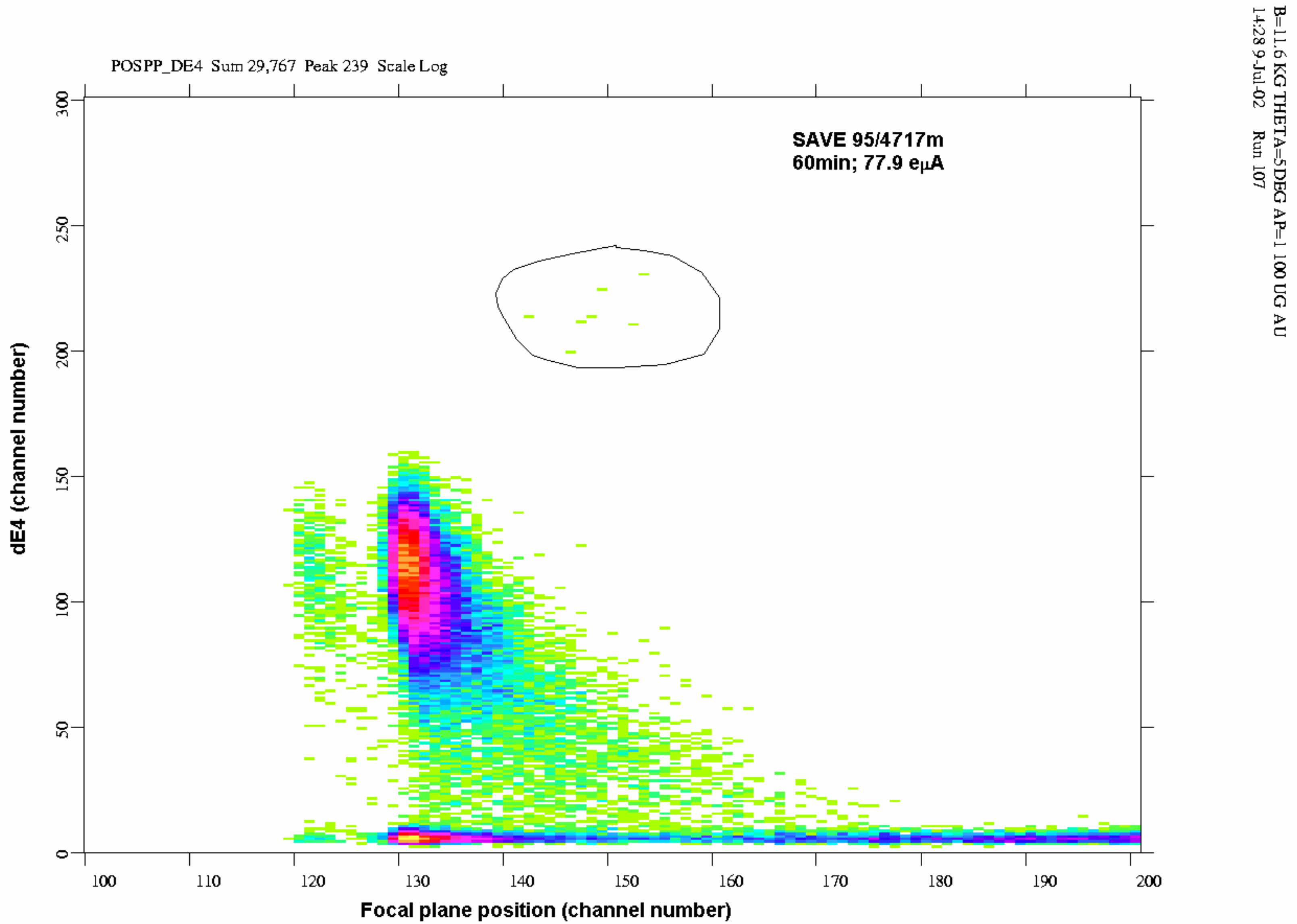
Water sampling rosette



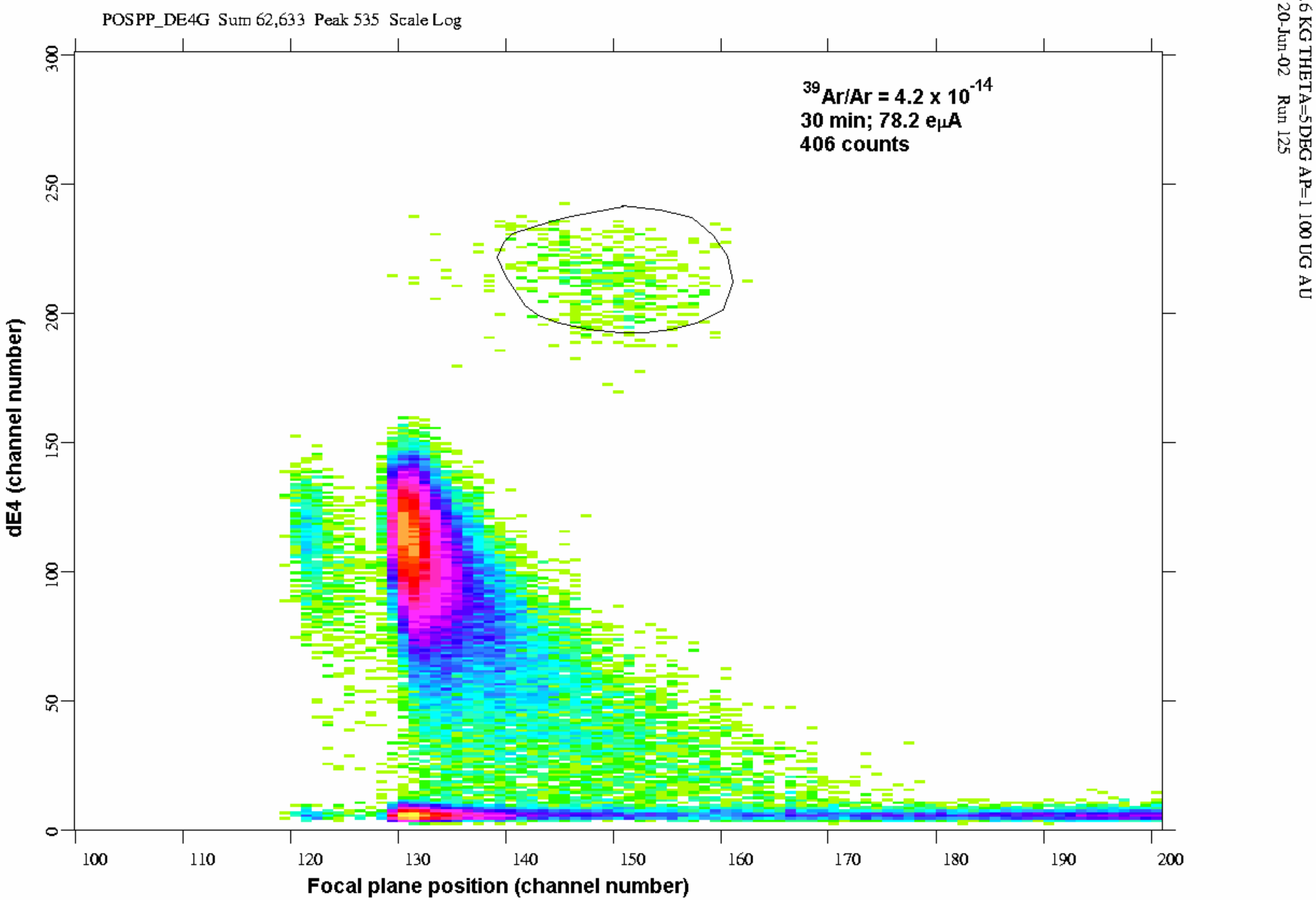


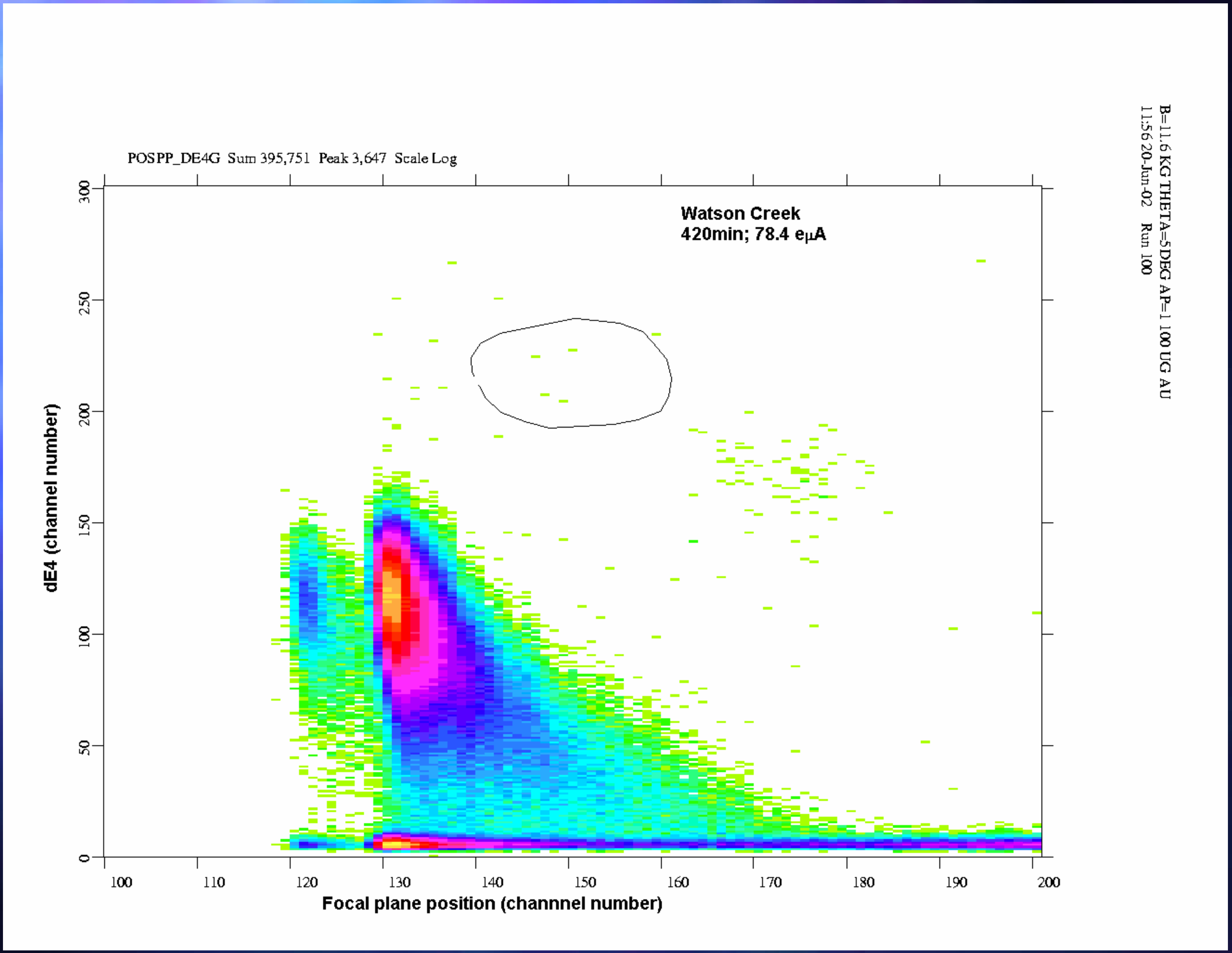
Ocean water samples





POSPP_DE4 Sum 29,767 Peak 239 Scale Log
SAVE 95/4717m
60min; 77.9 eμA
B=11.6 KG THETA=5DEG AP=1 100 UG AU
14:28 9-Jul-02 Run 107
dE4 (channel number)
Focal plane position (channel number)
0
50
100
150
200
250
300
100
110
120
130
140
150
160
170
180
190
200


POSPP_DE4G Sum 62,633 Peak 535 Scale Log
^{39}Ar/Ar = 4.2 x 10^{-14}
30 min; 78.2 eμA
406 counts
dE4 (channel number)
Focal plane position (channel number)
B=11.6 KG THETA=5DEG AP=1 100 UG AU
14:18 20-Jun-02 Run 125


POSPP_DE4G Sum 395,751 Peak 3,647 Scale Log
Watson Creek
420min; 78.4 eμA
B=11.6 KG THETA=5DEG AP=1 100 UG AU
11:56 20-Jun-02 Run 100
dE4 (channel number)
Focal plane position (channnel number)
0
50
100
150
200
250
300
100
110
120
130
140
150
160
170
180
190
200

Results from May 2002 AMS run

Sample	CPS / eμA	$^{39}Ar / Ar$		"age"
n-act	2.69×10^{-3}	5.80×10^{-14}		
^{nat}Ar	3.57×10^{-5}	7.70×10^{-16}		
SAVE 294 / 5000	1.67×10^{-5}	3.59×10^{-16}		44% mod.
SAVE 294 / 850	2.43×10^{-5}	5.23×10^{-16}		65% mod.
Watson creek	2.02×10^{-6}	4.35×10^{-17}		5.4%
SAVE 95 / 4717	1.21×10^{-5}	2.61×10^{-16}		32% mod.
^{nat}Ar	3.96×10^{-5}	8.53×10^{-16}		
n-act	2.79×10^{-3}	6.01×10^{-14}		

$3.76 \times 10^{-5} \equiv 8.1 \times 10^{-16}$