SMR.1744 - 15

SCHOOL ON ION BEAM ANALYSIS AND ACCELERATOR APPLICATIONS

13 - 24 March 2006

Accelerator mass spectrometry of heavy radioisotopes

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Accelerator Mass Spectrometry of Heavy Radioisotopes

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University of Vienna

School on Ion Beam Analysis and Accelerator Applications
Miramare – Trieste, 17 March 2006

Radioisotopes measured with AMS

Isotope	**Half-life (year)**
^{3}H	12
^{44}Ti	60
^{63}Ni	100
^{32}Si	140
^{39}Ar	269
^{14}C	**5 730**
^{59}Ni	75 000
^{41}Ca	104 000
^{81}Kr	230 000
^{36}Cl	301 000
^{26}Al	720 000
^{10}Be	1 520 000
^{53}Mn	3 600 000
^{182}Hf	8 900 000
^{129}I	17 000 000
^{236}U	23 000 000
^{244}Pu	81 000 000

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^{53}Mn	3 600 000
^{182}Hf	**8 900 000**
^{129}I	**17 000 000**
^{236}U	**23 000 000**
^{244}Pu ($^{239, 240, 242}$Pu)	**81 000 000**

Radioisotopes are measured with AMS through isotope ratios

Typical range:

radioisotope/stable isotope = 10^{-12} to 10^{-16}

The main challenge in measuring such minute isotope ratios is the separation of the radioisotope from interfering background of stable isobars.

There are three good cases if negative ions are used

$^{14}\mathrm{C}^{-}$ – $\cancel{^{14}\mathrm{N}}^{-}$

$^{26}\mathrm{Al}^{-}$ – $\cancel{^{26}\mathrm{Mg}}^{-}$

$^{129}\mathrm{I}^{-}$ – $\cancel{^{129}\mathrm{Xe}}^{-}$

All other radioisotopes are more difficult and must be cleaned up from stable isobar interference.

Sometimes special negative molecules help

$^{41}\mathrm{CaH}_3^{-}$ – $^{41}\xcancel{\mathrm{K}}\mathrm{H}_3^{-}$

$^{10}\mathrm{BeF}^{-}$ – $^{10}\xcancel{\mathrm{B}}\mathrm{F}^{-}$

$^{182}\mathrm{HfF}_5^{-}$ – $^{182}\xcancel{\mathrm{W}}\mathrm{F}_5^{-}$

Measurement of the ^{129}I/^{131}I Ratio in Chernobyl Fallout

Paper presented at the Adriatico Conference on „Environmental Physics - Atmospheric Aerosol“, Trieste 22-25 July 1986

Both ^{129}I and ^{131}I are fission products, but with vastly different half-lives:

^{129}I: 1.6 million years
^{131}I : 8.0 days

W. Kutschera, D. Fink, M. Paul, G. Hollos, A. Kaufmann
Physics Scripta 37 (1988) 310-313

Abstract

Rainwater collected in the Munich area approximately one week after the Chernobyl reactor accident was investigated for its content of the radioisotopes ^{129}I ($T_{1/2}$ = 1.6 × 10^7 yr) and ^{131}I ($T_{1/2}$ = 8.04 d). For the time of release, an isotopic ratio of $^{129}I/^{131}I = 19 \pm 5$ was found. This value was obtained from a gamma-ray activity measurement of ^{131}I with a Ge detector and a concentration measurement of ^{129}I with accelerator mass spectrometry. From the measured ratio an operating time of the reactor prior to the accident in the vicinity of two years can be estimated, which is in fair agreement with estimates from other long-lived to short-lived radioisotope ratios in the Chernobyl fallout. Some measurements of ^{131}I activity in thyroids of persons living in the Munich area are also reported.

Table I. *Activity of* ^{131}*I in human thyroids (Munich area).*

Date of measurement	Person #	Activity[a] (Bq)	Main living site	Dose[b] (mrem)
6 May 1986	1	227	country	45
	2	98	city	20
	3	91	city	18
	4	108	city	22
11 May 1986	2	60	city	12
	5	120	country	24
	6	117	country	23
27 May 1986	2	15	city	3
	5	36	country	7

[a] Bq = 1 decay/sec. Uncertainty = ±20%.

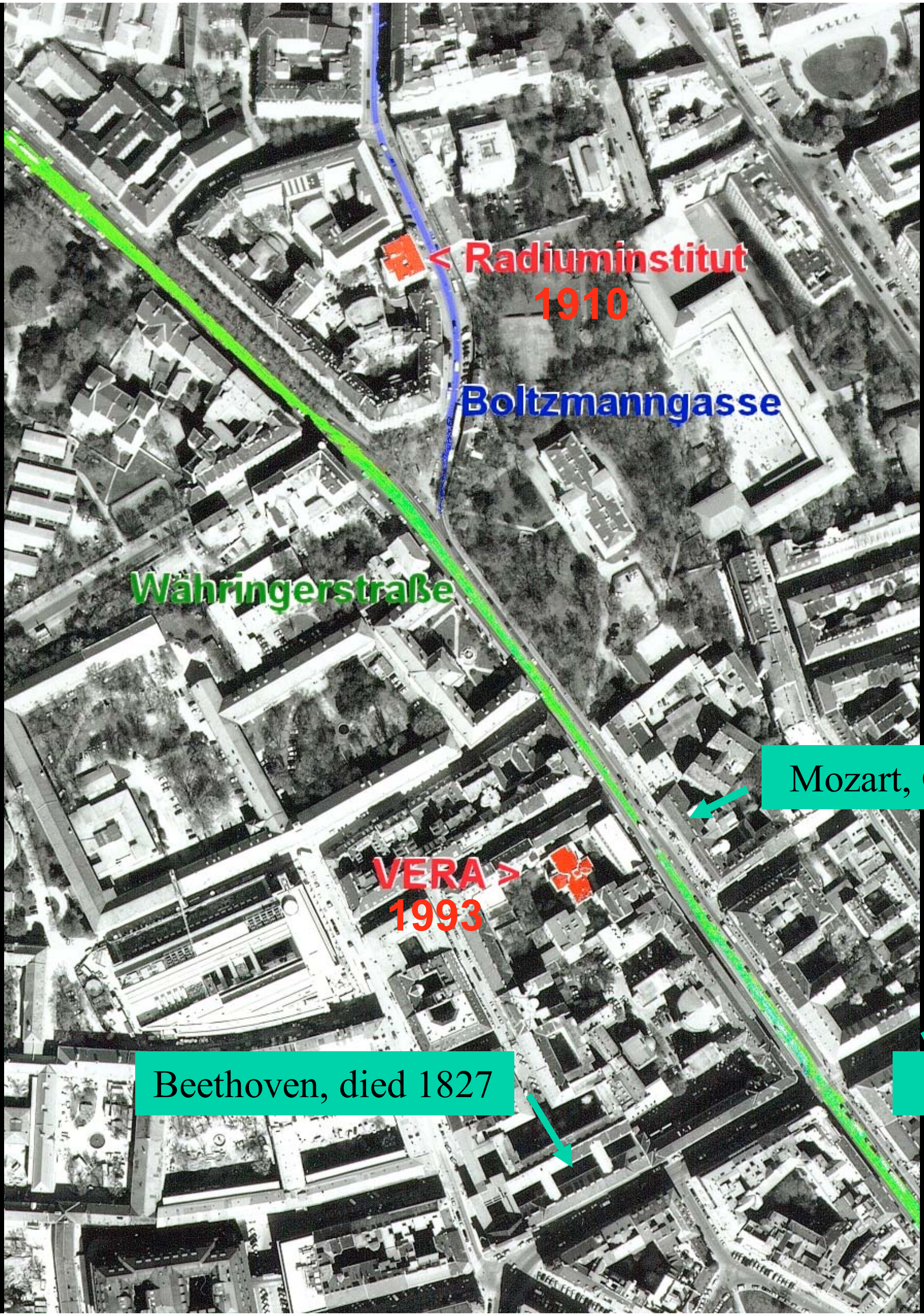
[b] Conversion = 0.2 mrem/Bq (= 2 μSv/Bq).

Vienna Environmental Research Accelerator
VERA

Institute für Isotope Research and Nuclear Physics
University of Vienna
Währinger Str. 17, A-1090 Vienna, Austria

VERA is an AMS facility for “all“ isotopes based on a 3-MV Pelletron tandem accelerator

P.Steier et al., Nucl. Instrum. & Methods 223-224 (2004) 67-71
W. Kutschera, Int. J. Mass Spectrometry 142 (2005) 145-160

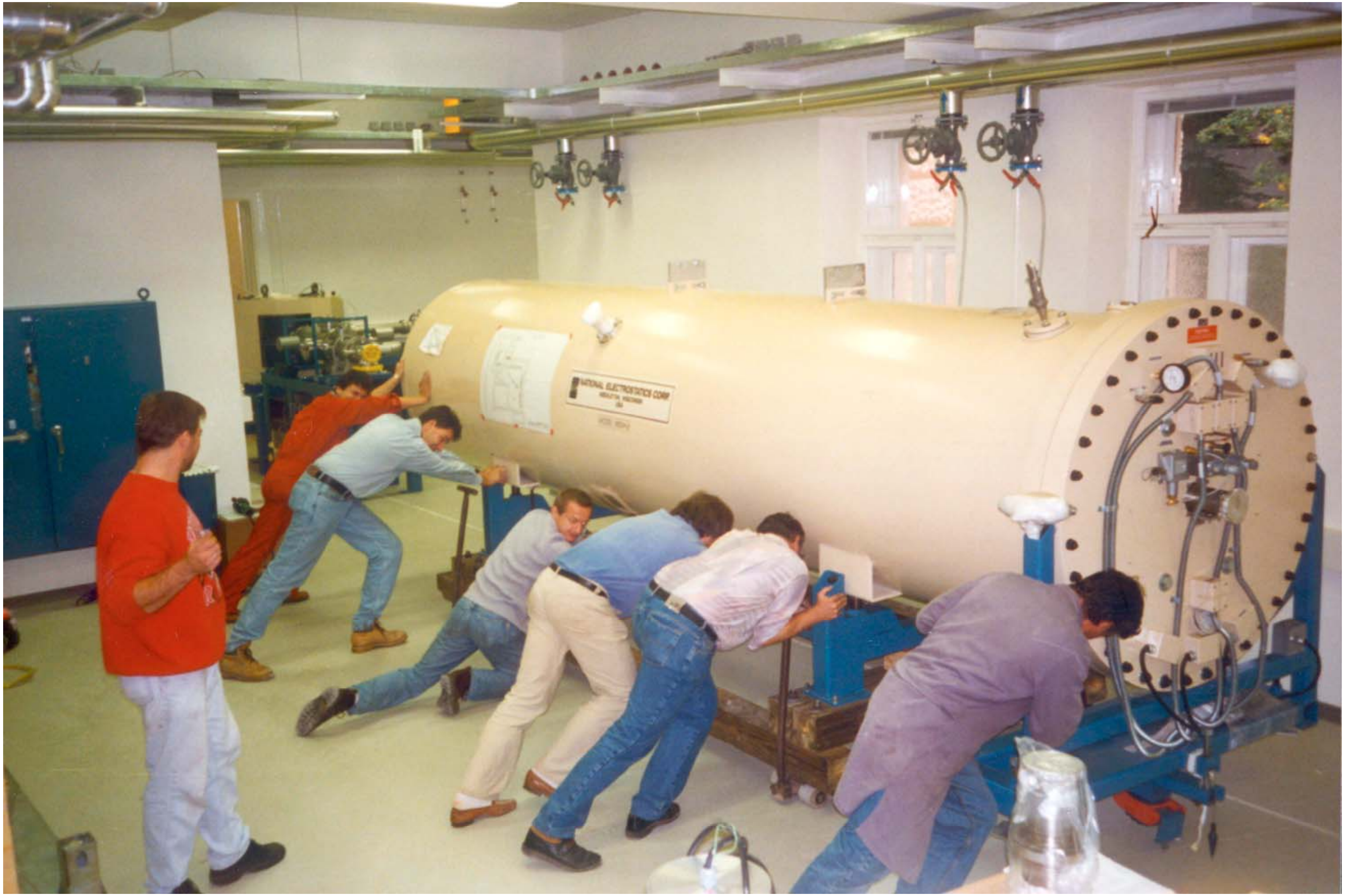


Mozart, Cosi fan tutte

Beethoven, died 1827

Sigmund Freud appartm.

Positioning of the 3-MV tandem accelerator of VERA in the "Kavalierstrakt", Währingerstr. 17, A-1090 Wien (1995)



+3 MV Tandem Accelerator

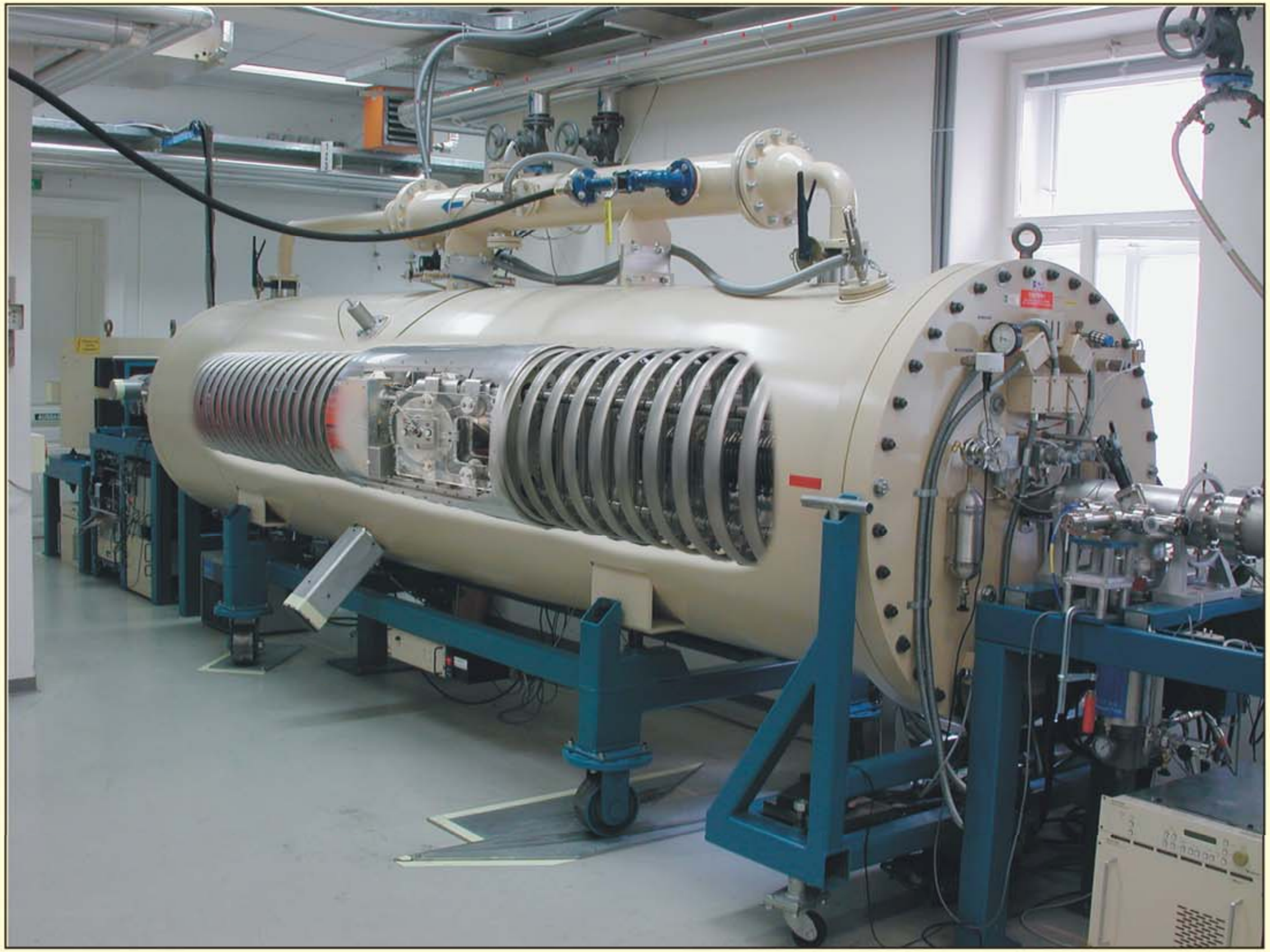
Pelletron type

2 charging
chains

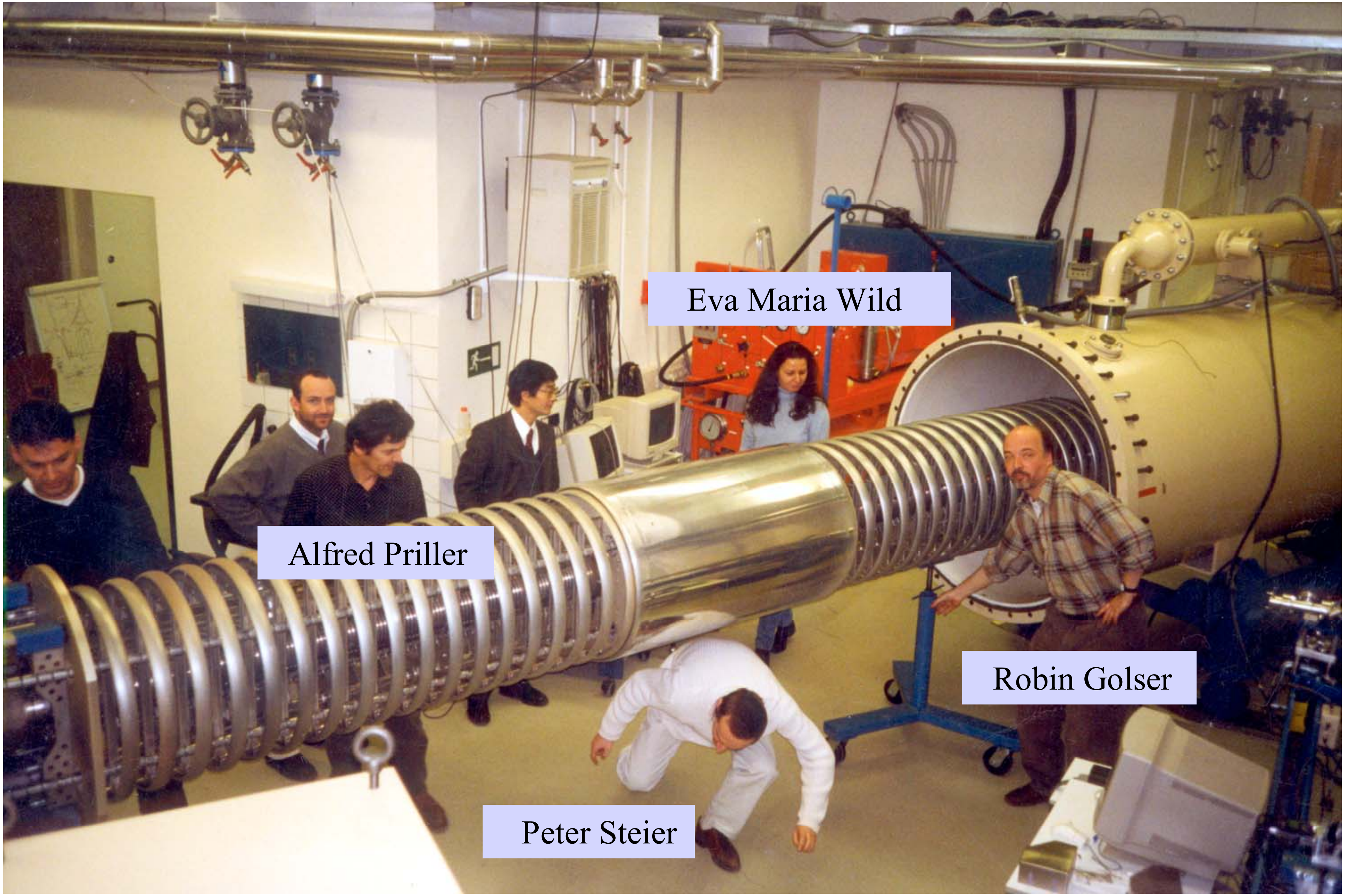
maximal charging
current: 230 µA

Ar gas stripper/
foil stipper

insulating gas:
SF_6



Column structure of the 3-MV tandem accelerator of VERA



Staff of VERA

Alfred Priller
Technical head of VERA
(paleoclimate, loess)

Peter Steier
Operations manager
(glacier dating, DNA dating, heavy isotopes)

Eva Maria Wild
Sample preparation
(archaeology, paleoclimate)

Robin Golser
Atomic physics
(exotic atoms, PIXE)

Anton Wallner
Astrophysics
(supernova remnant, stellar nucleosynthesis)

Cs-Beam Sputter Source for Negative Ions

40 Samples

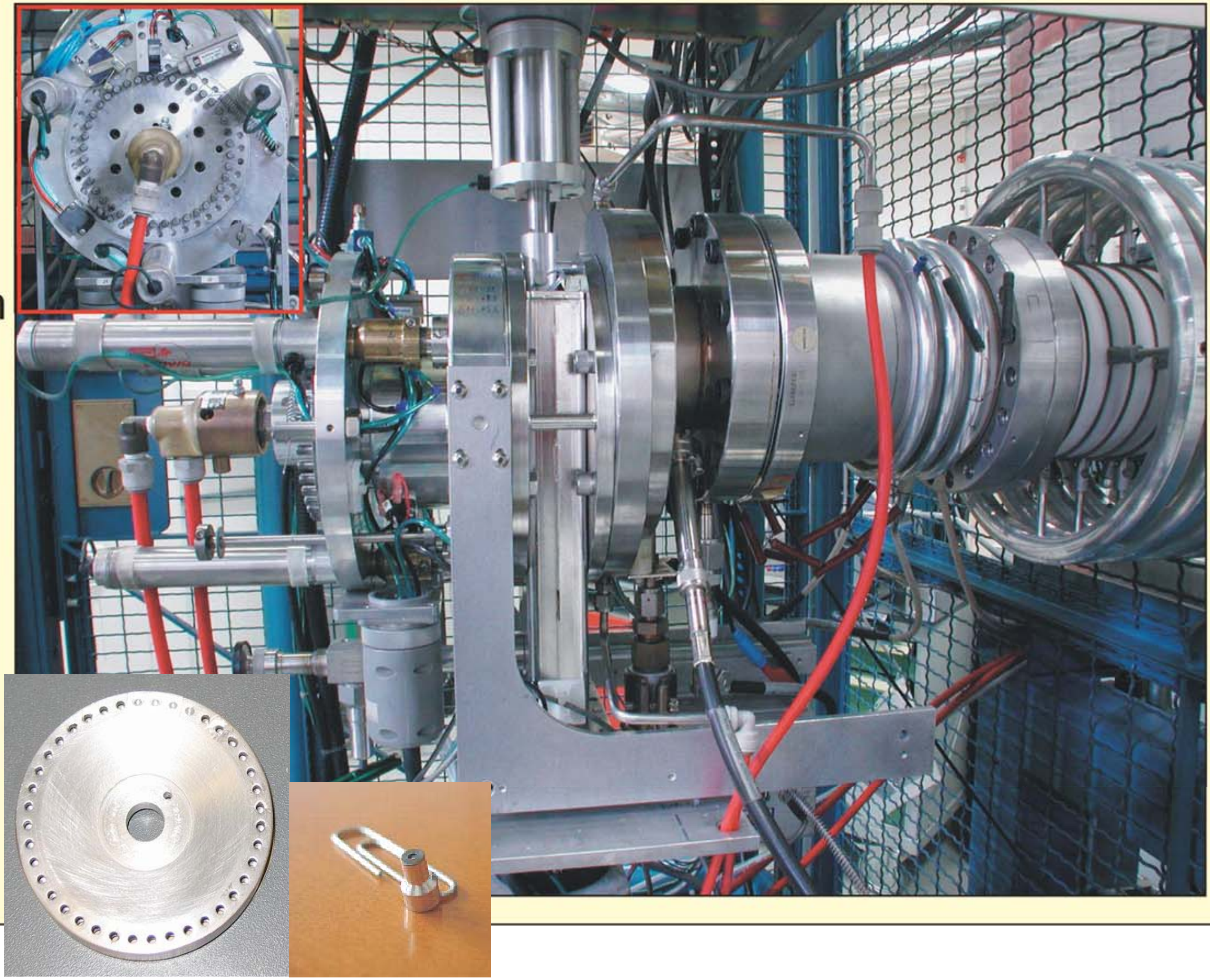
max. 75 keV
Preacceleration

Ion Currents:

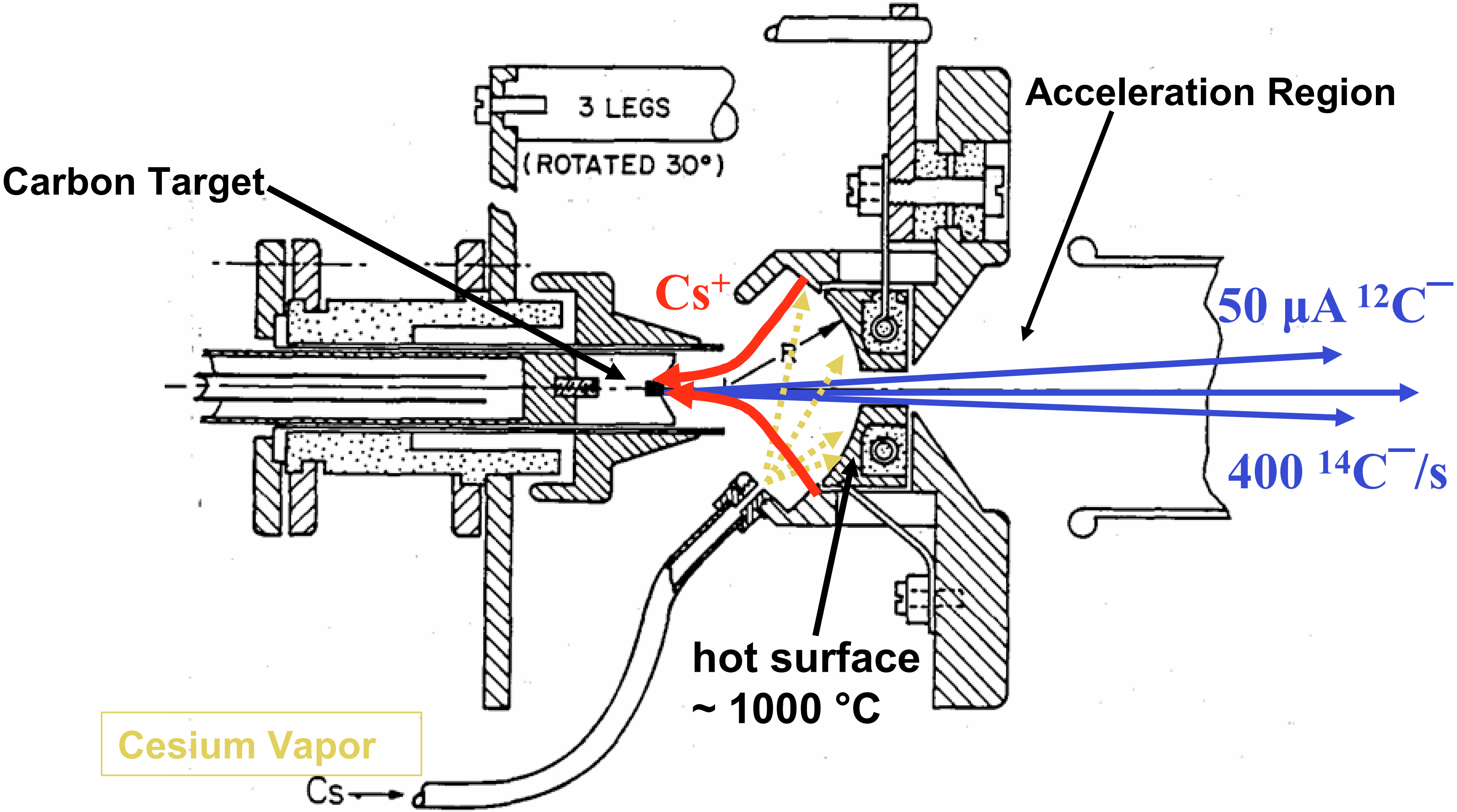
C^-: 60 μA

BeO^-: 3 μA

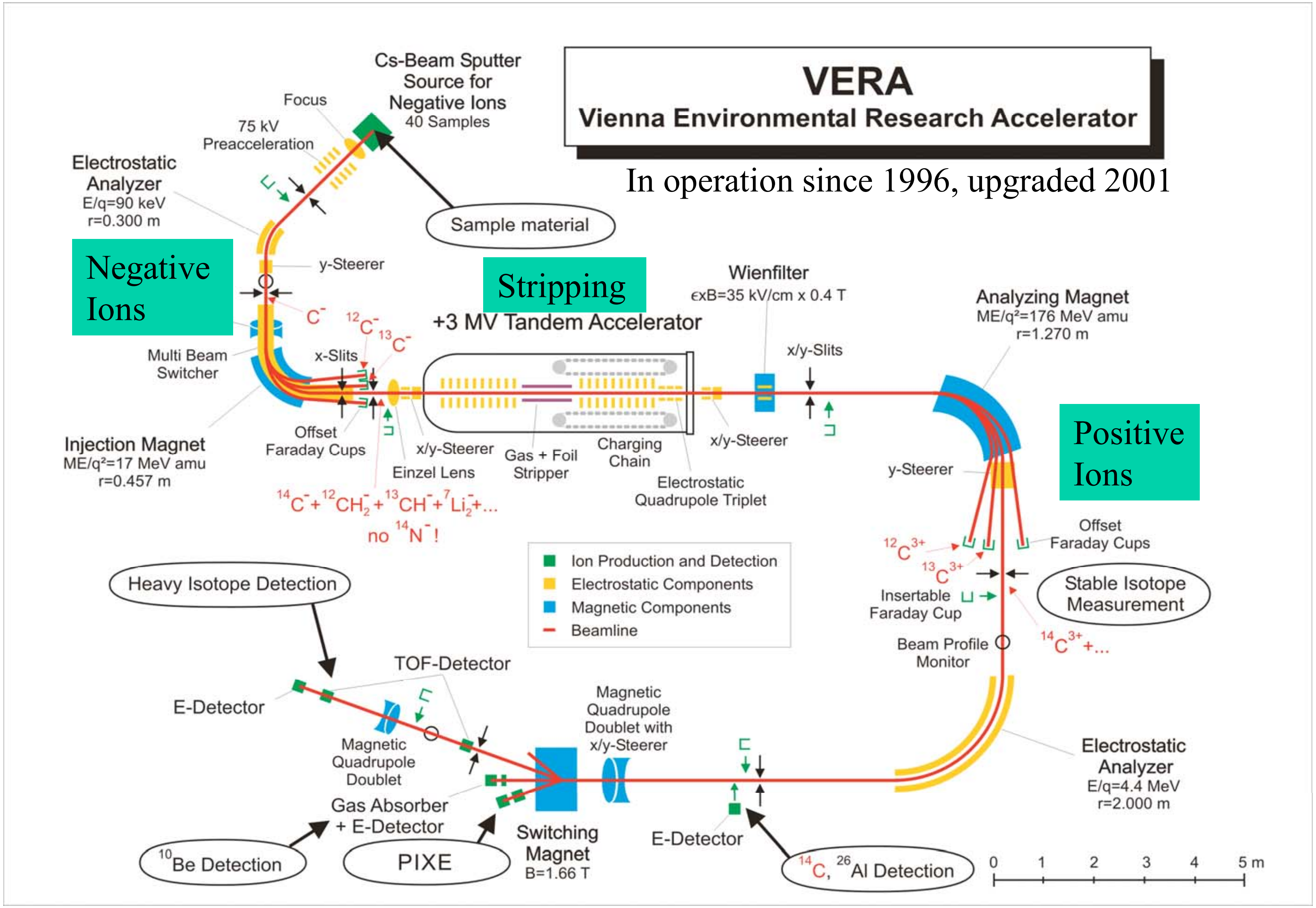
UO^-: 100 nA



The Cesium-Beam Sputter Source for Negative Ions



^{1}H, ^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl, ^{41}Ca, ^{55}Fe, ^{129}I, ^{182}Hf, ^{210}Pb, ^{236}U, $^{239\text{-}244}$Pu, $(^{43}Ca^{19}F_4)^{--}$, $(H_2)^{-}$



VERA

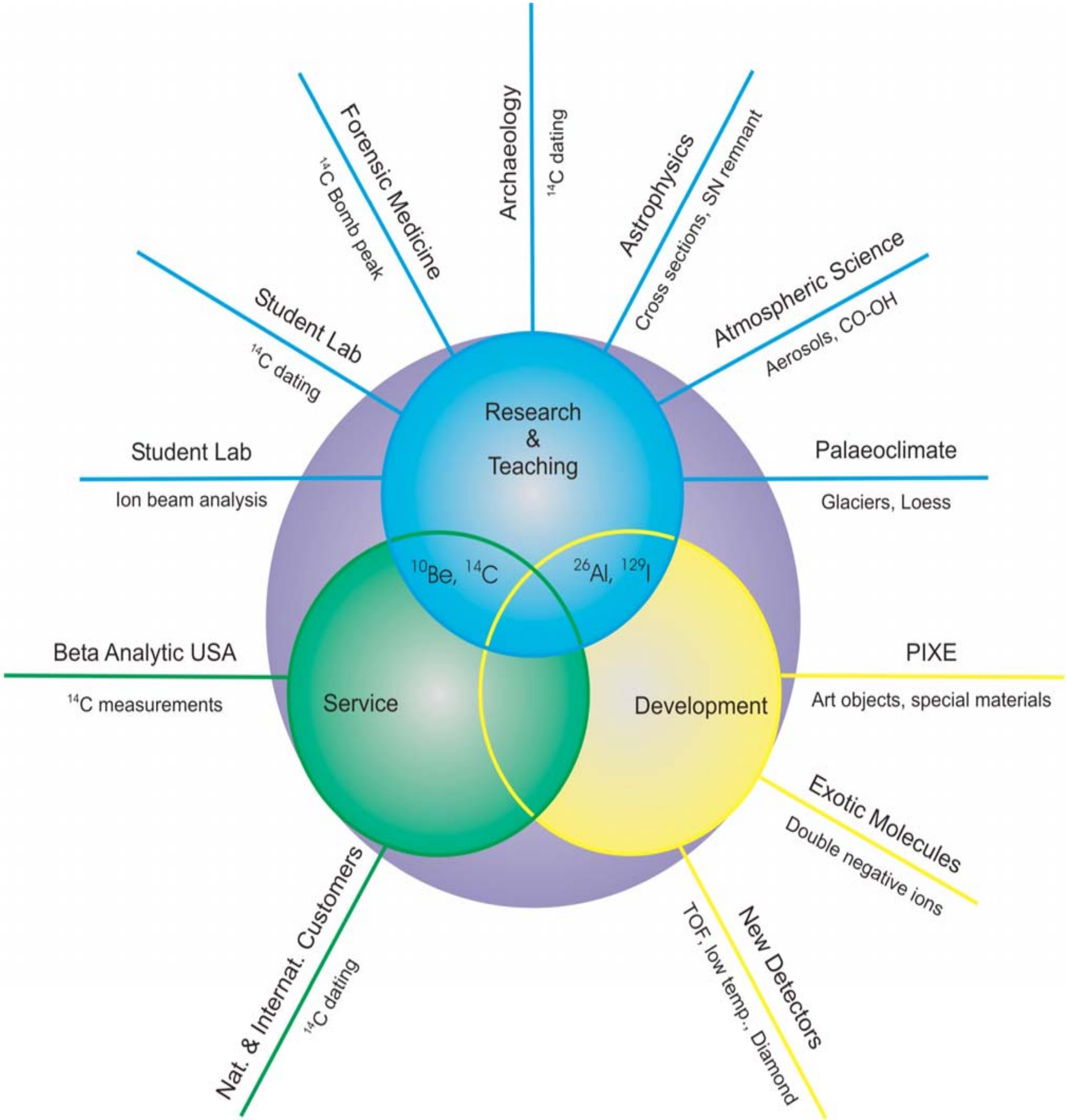
Vienna Environmental Research Accelerator

In operation since 1996, upgraded 2001

Negative Ions

Stripping

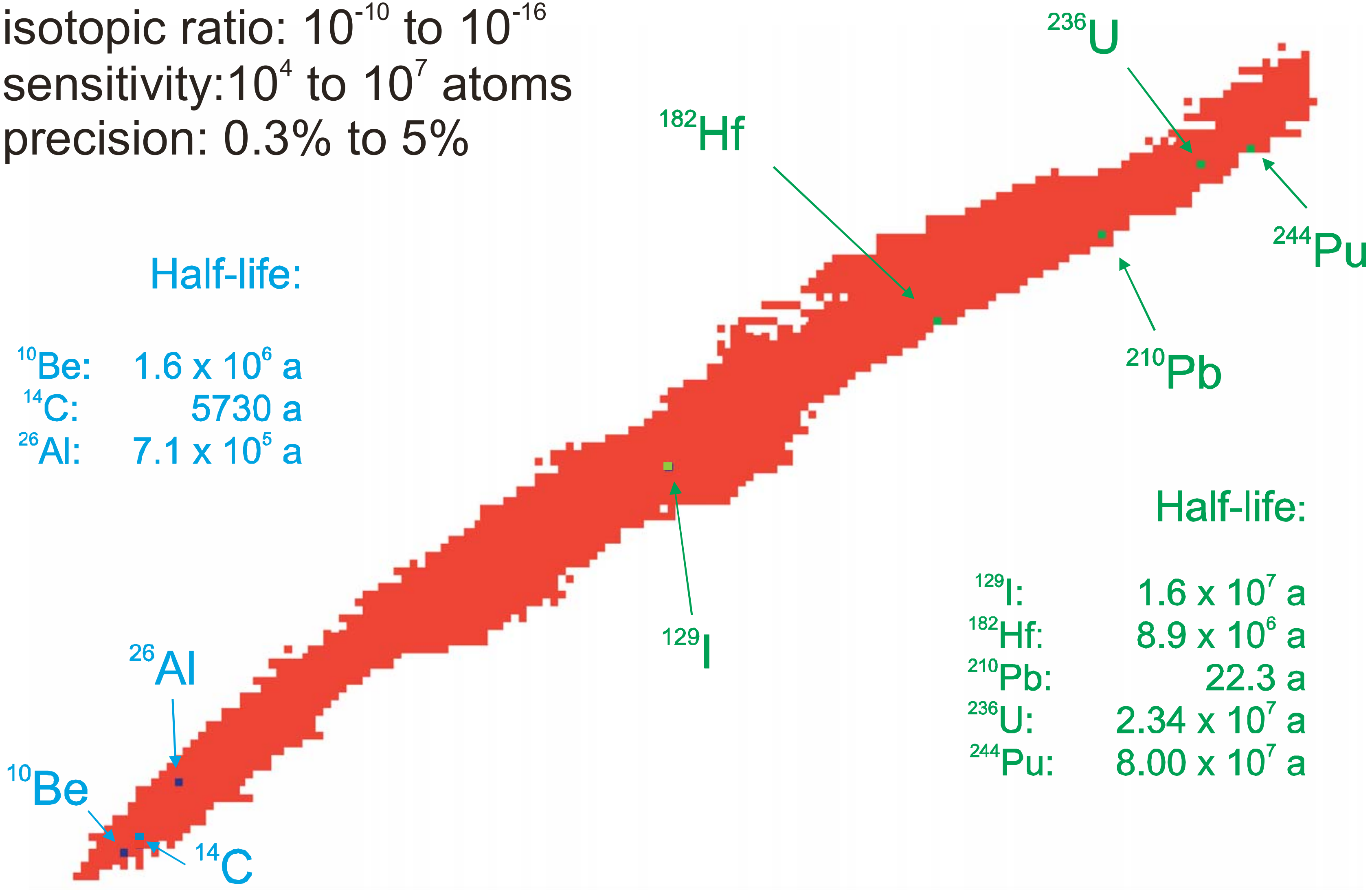
Positive Ions



The World of VERA

Long-lived radionuclides measured at VERA

isotopic ratio: 10^{-10} to 10^{-16}
sensitivity: 10^{4} to 10^{7} atoms
precision: 0.3% to 5%



Half-life:

^{10}Be:	1.6×10^{6} a
^{14}C:	5730 a
^{26}Al:	7.1×10^{5} a

Half-life:

^{129}I:	1.6×10^{7} a
^{182}Hf:	8.9×10^{6} a
^{210}Pb:	22.3 a
^{236}U:	2.34×10^{7} a
^{244}Pu:	8.00×10^{7} a

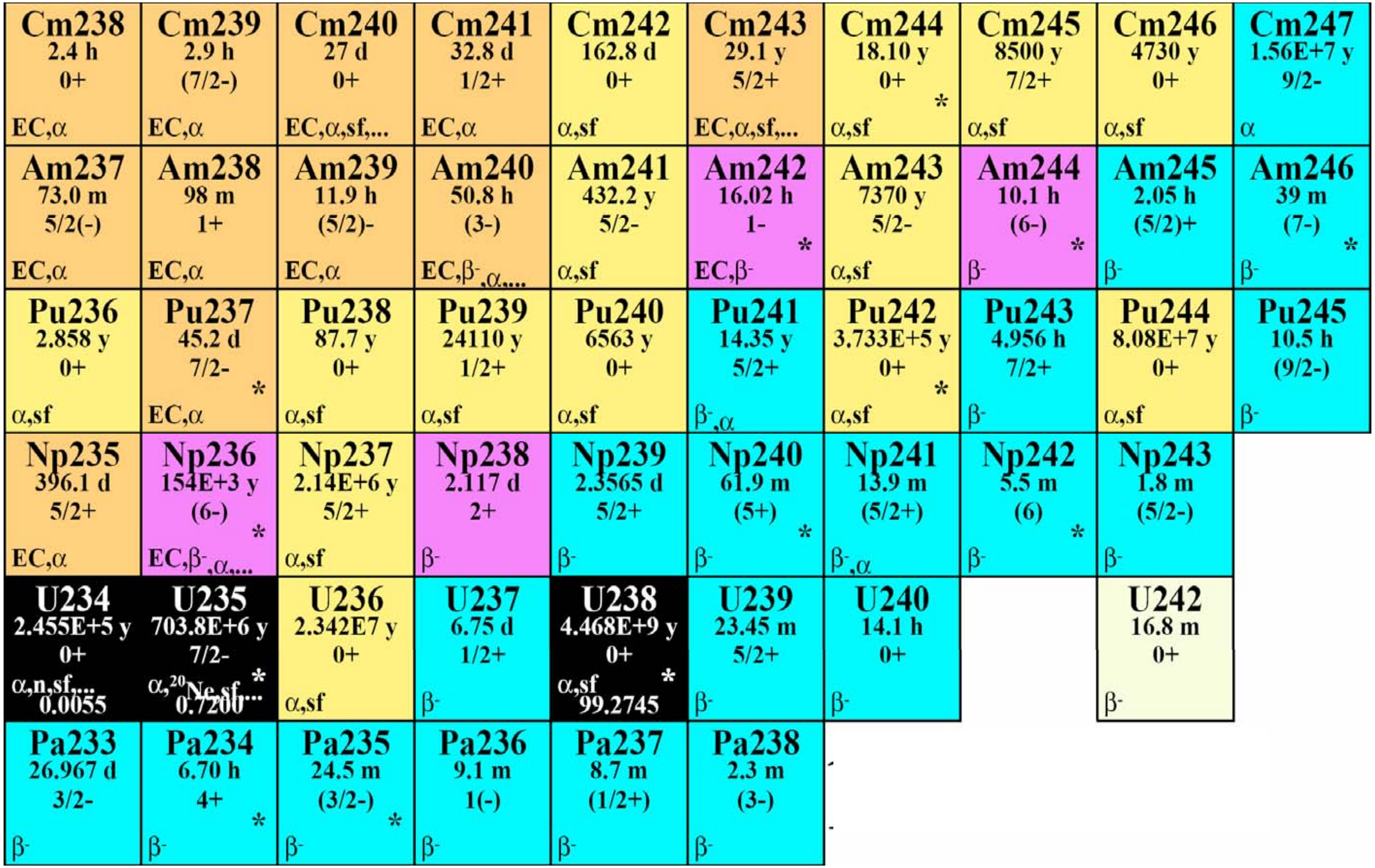
Some AMS-nuclides at VERA (3-MV)

Radio-nuclide	Half-life (Myr)	Overall Efficiency	Detection Limit		Precision
^{10}Be	1.5	$5\text{x}10^{-5}$	^{10}Be/^{9}Be	$< 2\text{x}10^{-14}$	< 3%
^{14}C	5730 yr	$2\text{x}10^{-2}$	^{14}C/^{12}C	$< 3\text{x}10^{-16}$	< 0.5 %
^{26}Al	0.7	$5\text{x}10^{-4}$	^{26}Al/^{27}Al	$< 6\text{x}10^{-16}$	< 1.0 %
^{129}I	15.7	$1\text{x}10^{-2}$	^{129}I/^{127}I	$= 2\text{x}10^{-14}$	2 %
^{182}Hf	8.9	$1\text{x}10^{-4}$	^{182}Hf/^{180}Hf	$= 1\text{x}10^{-11}$	5 %
^{236}U	23.4	--	^{236}U/^{238}U	$= 6\text{x}10^{-12}$	5 %
^{244}Pu	81.0	$> 4\text{x}10^{-5}$	--		5 %

Determination of Plutonium in environmental samples by AMS and Alpha Spectrometry

Proceedings of the 8th International Conference in Application of Nuclear Techniques, Crete, Greece, 12-18 September 2004

E. Hrenecek, P. Steier, A. Wallner
Applied Radiation and Isotopes 63 (2005) 633-638

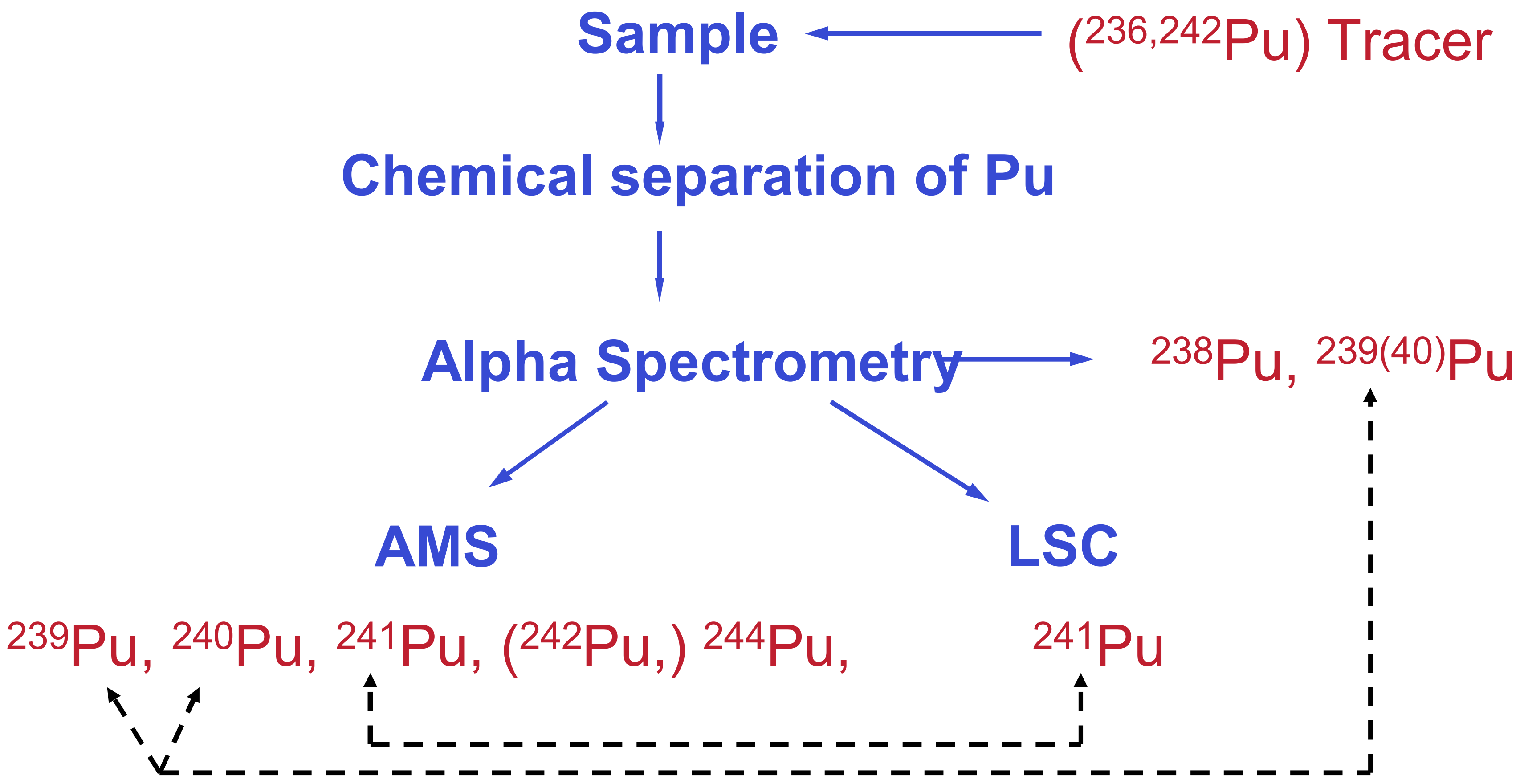


Cm	Cm238 2.4 h 0+ EC,α	Cm239 2.9 h (7/2-) EC,α	Cm240 27 d 0+ EC,α,sf,...	Cm241 32.8 d 1/2+ EC,α	Cm242 162.8 d 0+ α,sf	Cm243 29.1 y 5/2+ EC,α,sf,...	Cm244 18.10 y 0+ * α,sf	Cm245 8500 y 7/2+ α,sf	Cm246 4730 y 0+ α,sf	Cm247 1.56E+7 y 9/2- α
Am	Am237 73.0 m 5/2(-) EC,α	Am238 98 m 1+ EC,α	Am239 11.9 h (5/2)- EC,α	Am240 50.8 h (3-) EC,β-,α,...	Am241 432.2 y 5/2- α,sf	Am242 16.02 h 1- * EC,β-	Am243 7370 y 5/2- α,sf	Am244 10.1 h (6-) * β-	Am245 2.05 h (5/2)+ β-	Am246 39 m (7-) * β-
Pu	Pu236 2.858 y 0+ α,sf	Pu237 45.2 d 7/2- * EC,α	Pu238 87.7 y 0+ α,sf	Pu239 24110 y 1/2+ α,sf	Pu240 6563 y 0+ α,sf	Pu241 14.35 y 5/2+ β-,α	Pu242 3.733E+5 y 0+ * α,sf	Pu243 4.956 h 7/2+ β-	Pu244 8.08E+7 y 0+ α,sf	Pu245 10.5 h (9/2-) β-
Np	Np235 396.1 d 5/2+ EC,α	Np236 154E+3 y (6-) * EC,β-,α,...	Np237 2.14E+6 y 5/2+ α,sf	Np238 2.117 d 2+ β-	Np239 2.3565 d 5/2+ β-	Np240 61.9 m (5+) * β-	Np241 13.9 m (5/2+) β-,α	Np242 5.5 m (6) * β-	Np243 1.8 m (5/2-) β-	
U	U234 2.455E+5 y 0+ α,n,sf,... 0.0055	U235 703.8E+6 y 7/2- * α,^{20}Ne,sf,... 0.7200	U236 2.342E7 y 0+ α,sf	U237 6.75 d 1/2+ β-	U238 4.468E+9 y 0+ * α,sf 99.2745	U239 23.45 m 5/2+ β-	U240 14.1 h 0+ β-		U242 16.8 m 0+ β-	
Pa	Pa233 26.967 d 3/2- β-	Pa234 6.70 h 4+ * β-	Pa235 24.5 m (3/2-) * β-	Pa236 9.1 m 1(-) β-	Pa237 8.7 m (1/2+) β-	Pa238 2.3 m (3-) β-				

Pu isotopes: ^{242}Pu as spike material (reference)

AMS measurement: $^{239,240,241(244)}$Pu relative to ^{242}Pu

Analytical Strategy



Sample

(^{236,242}Pu) Tracer

Chemical separation of Pu

Alpha Spectrometry

^{238}Pu, $^{239(40)}$Pu

AMS

LSC

^{239}Pu, ^{240}Pu, ^{241}Pu, (^{242}Pu,) ^{244}Pu,

^{241}Pu

Isotope ratios

source	^{238}Pu/$^{239(40)}$Pu	reference
Global weapons test fallout	0.03	Bunzl et al. 1987
Chernobyl fallout	0.33 – 0.44	IAEA 1986
Irish Sea sediment	0.05 – 0.3	Kershaw et al. 1995
Thule sediment	0.019	Aarkog 1971
Mururoa test site	0.0044	Danesi et al. 2002
Fangataufa test site	0.38	Mulsow et al. 1999

Isotope ratios

source	$^{240}Pu/^{239}Pu$	reference
Global fallout	0.18	Buessler et al. 1987
Chernobyl fallout	0.39	MacKenzie 2000
Irish Sea sediment	0.05 – 0.25	Kershaw et al. 1995
Thule sediment	0.058	Komura et al. 1984
Mururoa average	~ 0.035	Chiappini et al. 1999
Fangataufa	0.05	Chiappini et al. 1999

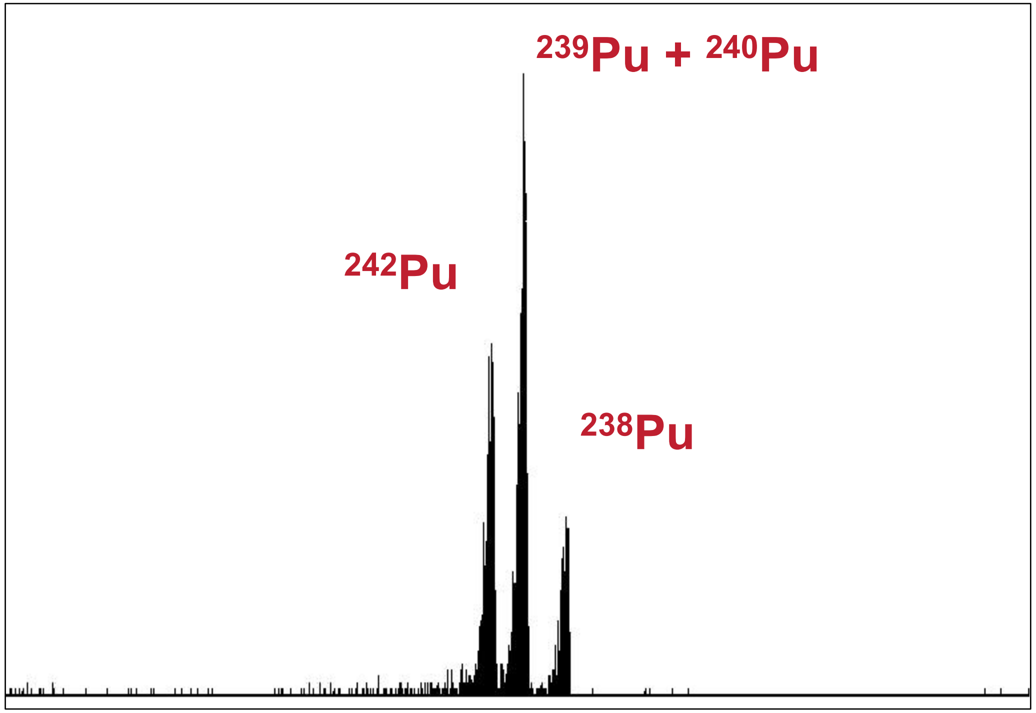
Sample preparation for alpha counting

- Leaching 8 M HNO_3
- Anion Exchange AG 1x8
- 8 M HNO_3, $NaNO_2$ for Pu(IV)
- 10 M HCl for Th(IV)
- 0,1 M NH_4I / 9 M HCl for Pu
- Alpha Spectrometry: Mikroprecipitation with $NdF_{3,}$
- Cellulosenitrate filter

  0.1 µm pore size

Alpha Spectrometry

- ^{242}Pu Tracer
- 2.5 to 6 g Sample size
- Detection limit:
  0.1 to 0.3 Bq / sample
- ^{238}Pu / $^{239(240)}$Pu



Energy of alpha particles

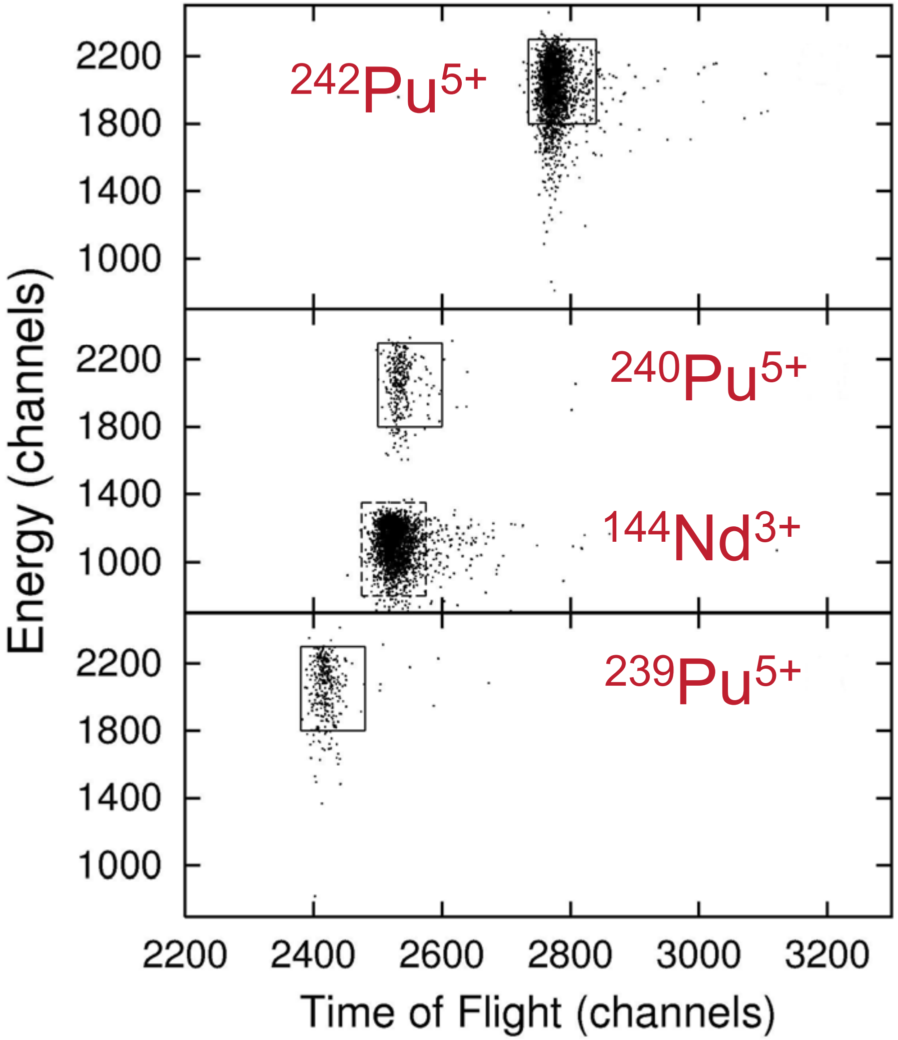
Sample Preparation for AMS

- Sample preparation:
  - – combustion of filter
  - – dissolution in HCl
  - – 10 mg Fe carrier, $Fe(OH)_3$
  - – 800°C Fe_2O_3

AMS measurement

$^{238}UO^{-} \rightarrow {}^{238}U^{5+}$, used for tuning (current)

$^{242}PuO^{-} \rightarrow {}^{242}Pu^{5+}$, scaling and ion counting



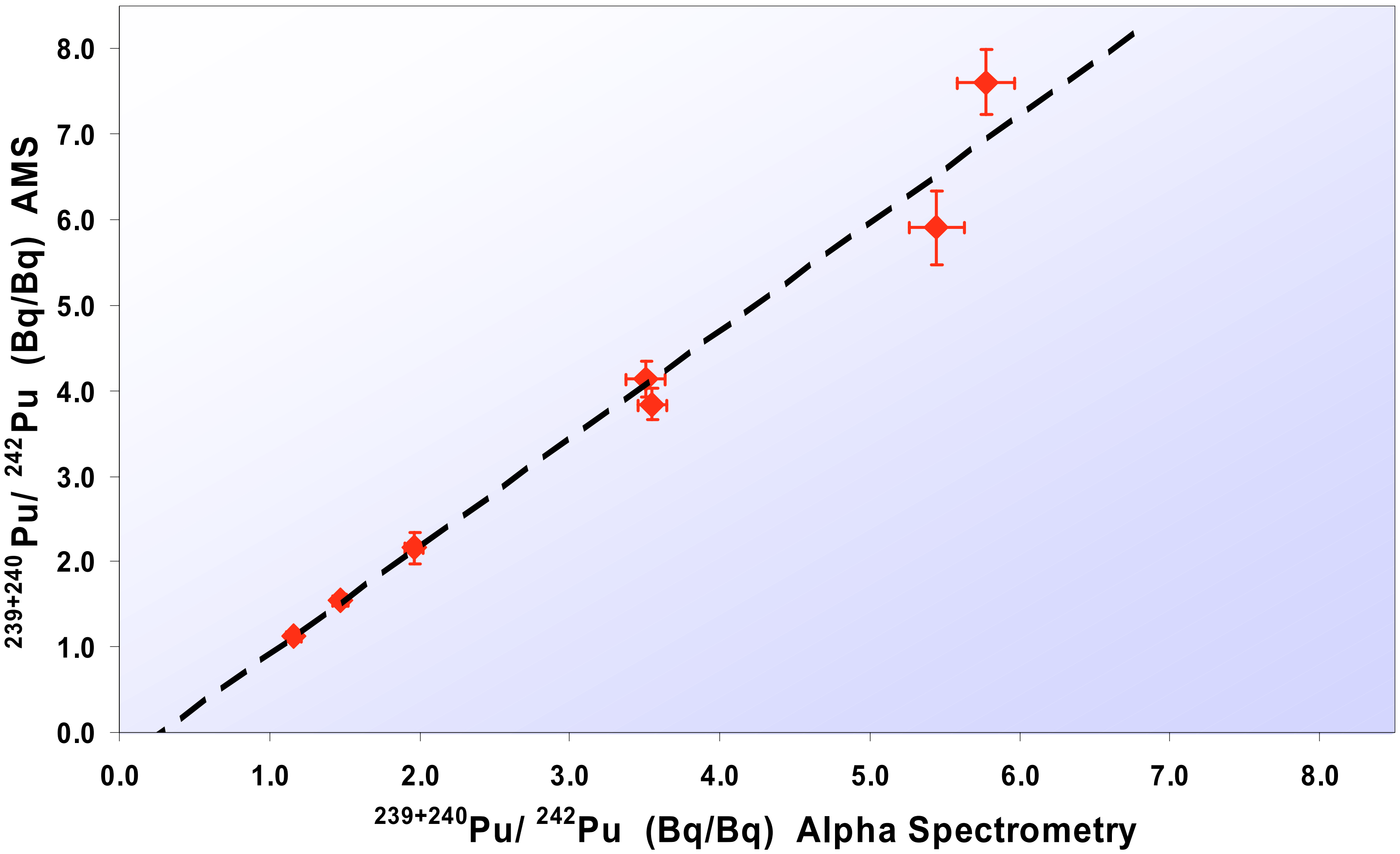
M/Q !

240/5 = 144/3

Results

	AMS		Alpha Spectrometry	
	^{240}Pu/^{239}Pu [at/at]	$^{239(40)}$Pu [Bq/kg]	$^{239(40)}$Pu [Bq/kg]	^{238}Pu [Bq/kg]
11.4.1 Fangataufa (Kilo) loose coral rocks	0.049 ± 0.002	51.3 ± 2.6	43.5 ± 1.6	15.0 ± 0.7
11.4.3	0.049 ± 0.003	19.3 ± 0.8	19.8 ± 0.9	6.8 ± 0.4
11.4.4	0.050 ± 0.007	93.6 ± 4.7	71.8 ± 2.4	26.1 ± 1.0
9.3.6 Mururoa (Faucon) top soil	0.018 ± 0.002	175 ± 15	159.1 ± 4.9	0.77 ± 0.18
9.3.18	0.017 ± 0.002	115 ± 5	109.9 ± 3.7	0.59 ± 0.16
9.3.25	0.018 ± 0.003	221 ± 11	204.3 ± 5.4	1.21 ± 0.19
7.2.4.2 Mururoa (Colette)	0.019 ± 0.002	539 ± 39	497 ± 17	2.24 ± 0.46

Comparison AMS - α-spectrometry

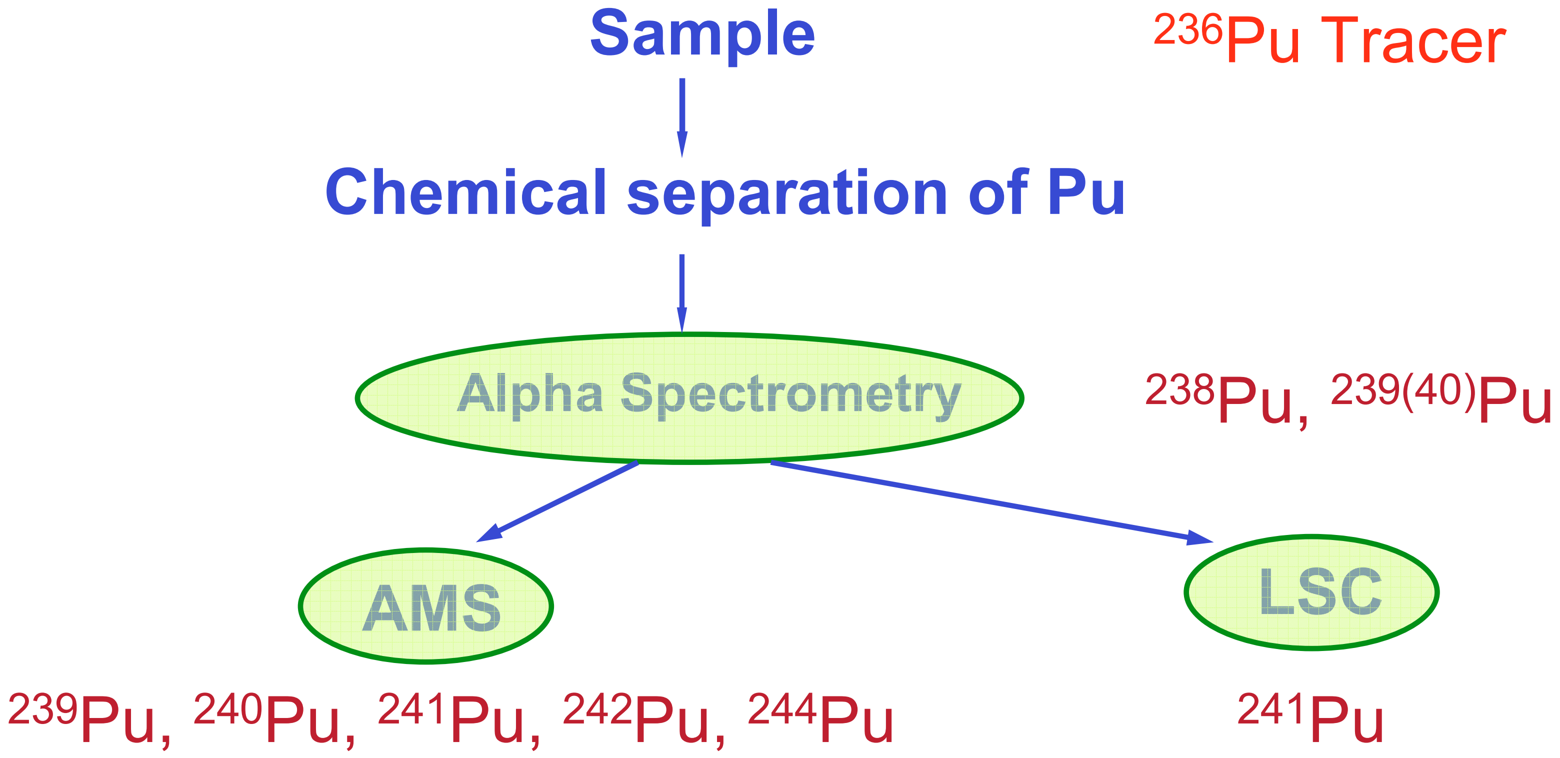


Results

Test site	$^{238}Pu/^{239(40)}Pu$ (Alpha Spec)	
Mururoa	0.0051 ± 0.0006	0.0044 Danesi et al. (2002)
Fangataufa	0.35 ± 0.01	0.38 Mulsow et al. (1999)

Test site	$^{240}Pu/^{239}Pu$ (AMS)	
Mururoa	0.018 ± 0.001	< 0.03 safety tests Chiappini et al. (1999)
Fangataufa	0.049 ± 0.001	0.05 Chiappini et al. (1999)

Outlook: Analytical Strategy



Sample

^{236}Pu Tracer

Chemical separation of Pu

Alpha Spectrometry

^{238}Pu, $^{239(40)}$Pu

AMS

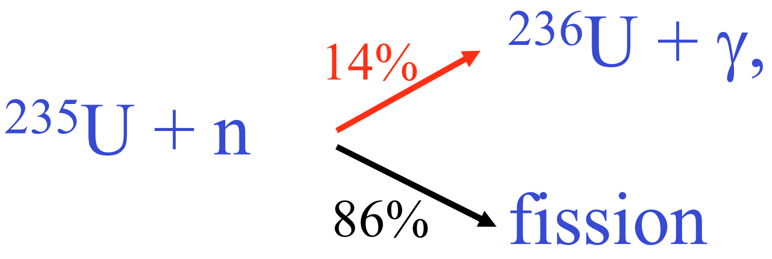
^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{244}Pu

LSC

^{241}Pu

⇨ ➢Complete set of „Pu-isotope signature“

^{236}U, a natural and anthropogenic neutron monitor



half-life = 23 million years

Expected ^{236}U/^{238}U isotope ratios:

Uranium mineral: ~ 10^{-11}
1 ppm U in rock: ~10^{-14}
Power reactor: ~ 10^{-3} (after burning off 1% ^{235}U)

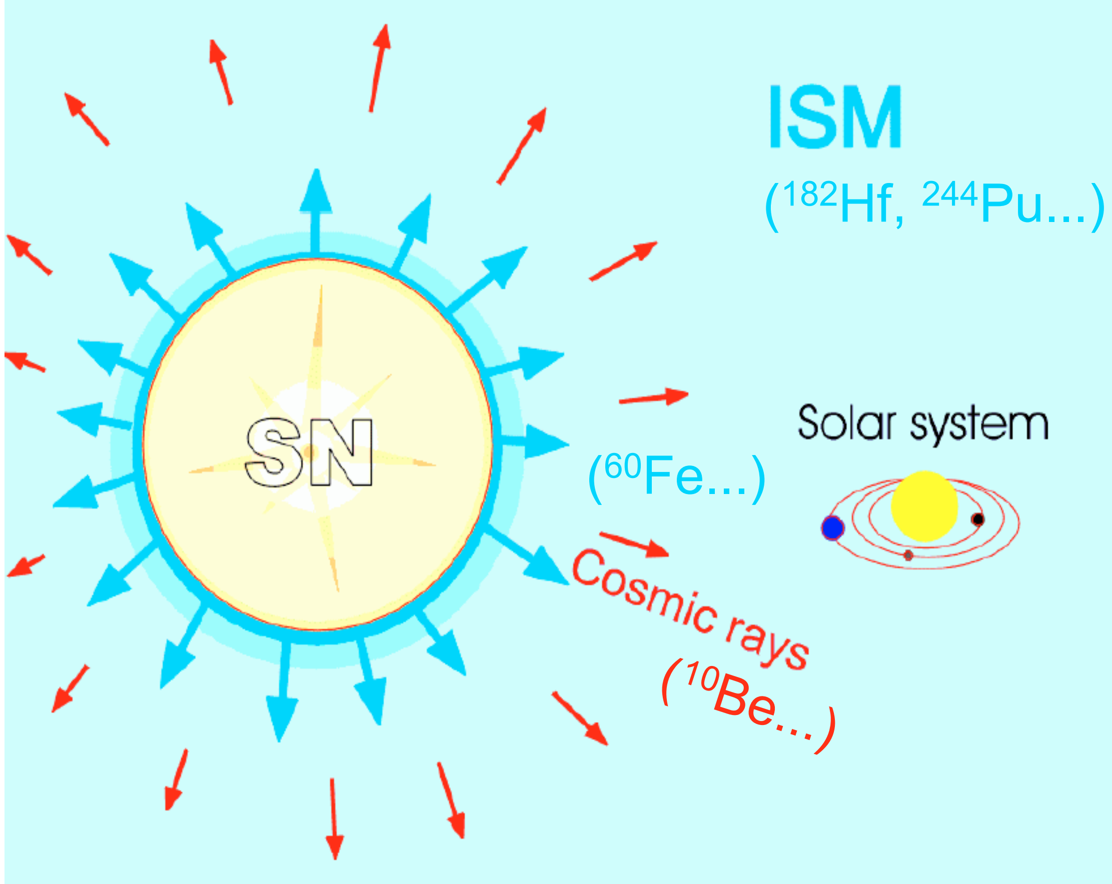
Lowest ^{236}U/^{238}U ratio measured at VERA: 6.1×10^{-12}

Probably the most interesting frontier for heavy-isotope AMS:

Searching for live supernova remnants on Earth

Live long-lived radionuclides

nearby supernova: < 100 pc, rate ~ 0.3 -10 $(Ma)^{-1}$

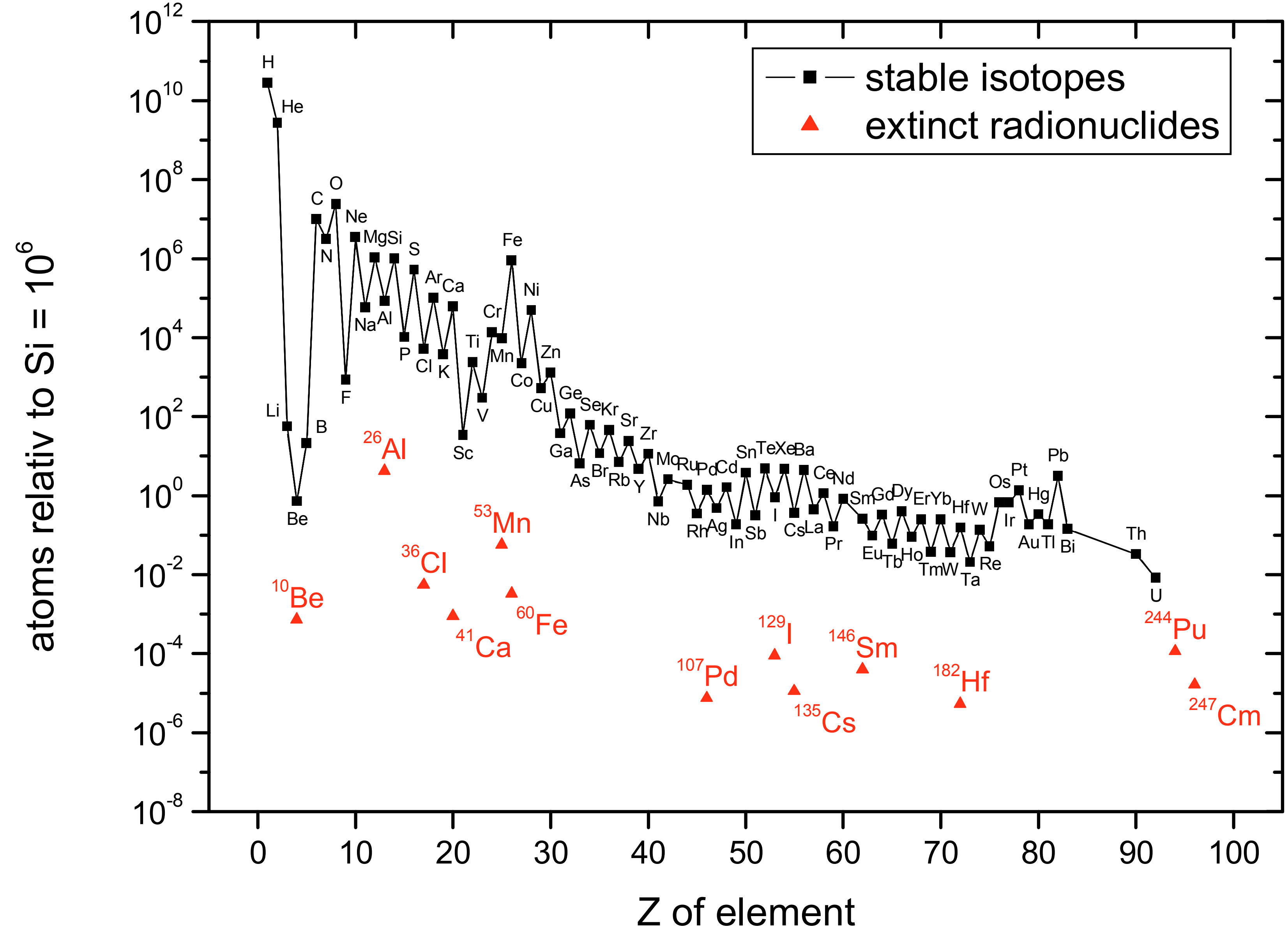


Crab-nebula (SN from 1054 AD)

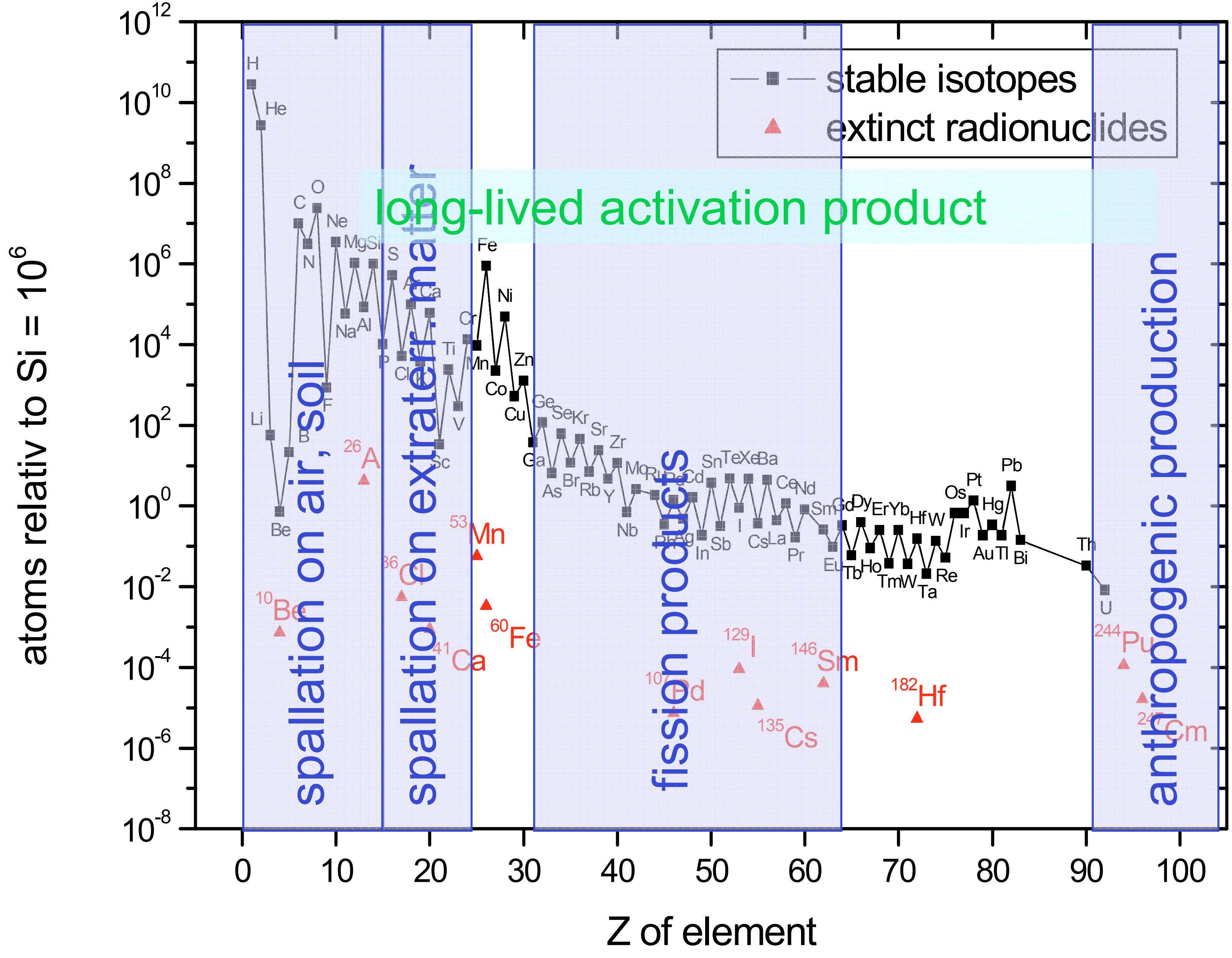
Supernova Remnant at 2000 pc distance

http://antwrp.gsfc.nasa.gov/apod/image/9911/crab_vlt_big.jpg

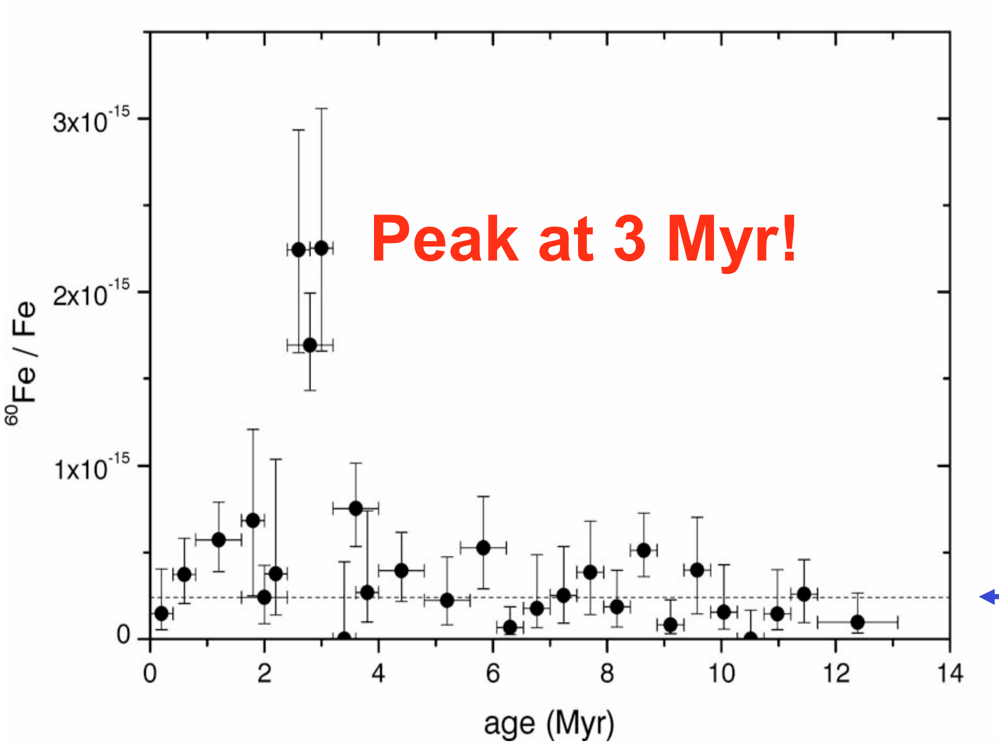
Solar system abundance



Production of long-lived solar system radionuclides



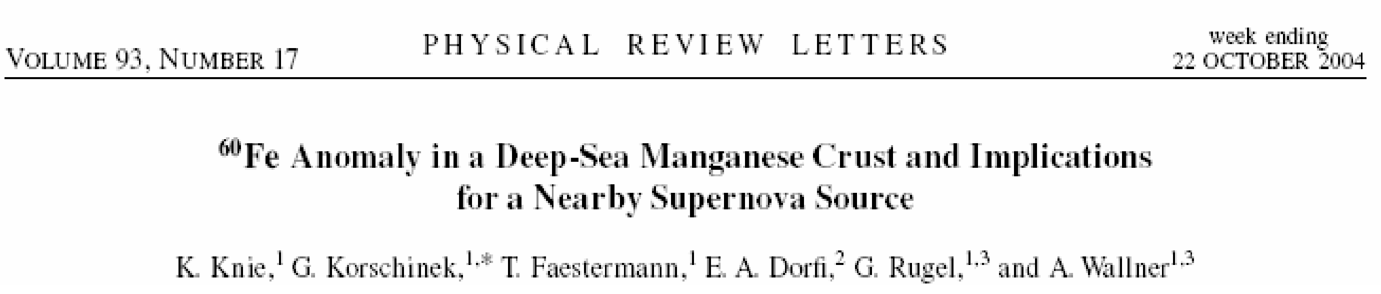
^{60}*Fe-signal in deep-sea crust*



Peak at 3 Myr!

AMS measurement of
^{60}Fe content of crust

← background level



VOLUME 93, NUMBER 17 PHYSICAL REVIEW LETTERS week ending 22 OCTOBER 2004

^{60}Fe Anomaly in a Deep-Sea Manganese Crust and Implications for a Nearby Supernova Source

K. Knie,[1] G. Korschinek,[1,*] T. Faestermann,[1] E. A. Dorfi,[2] G. Rugel,[1,3] and A. Wallner[1,3]

^{182}Hf would be a good candidate for a supernova remnant:

But interference from stable isobar ^{182}W limits ^{182}Hf/^{180}Hf currently to ~ 10^{-11}.

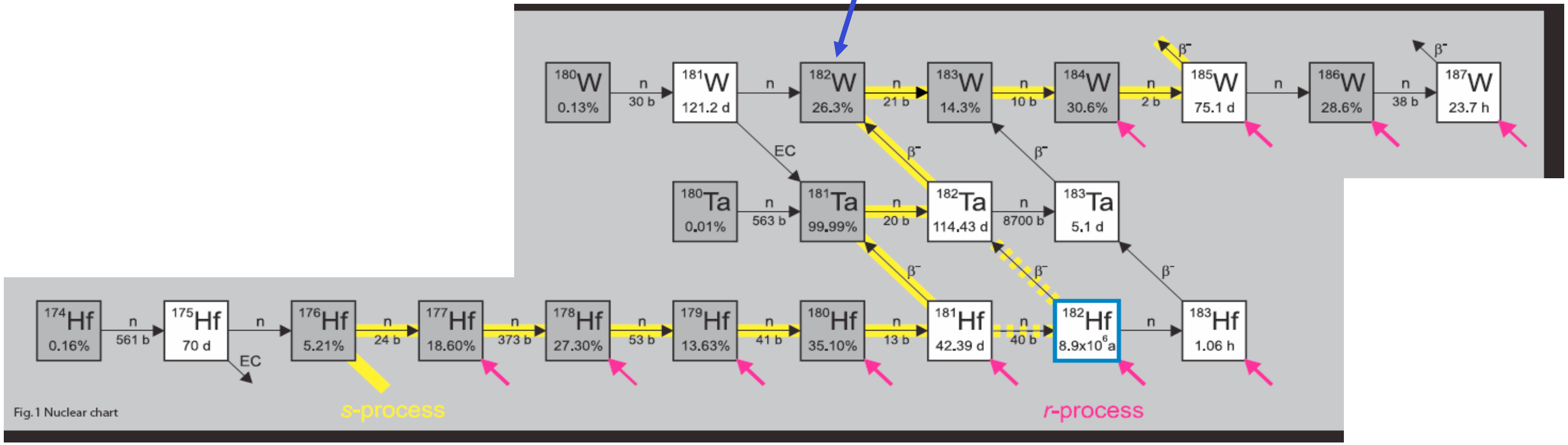


Fig. 1 Nuclear chart

^{244}Pu: Is our Earth too much contaminated to find the faint signal of supernova-produced ^{244}Pu?

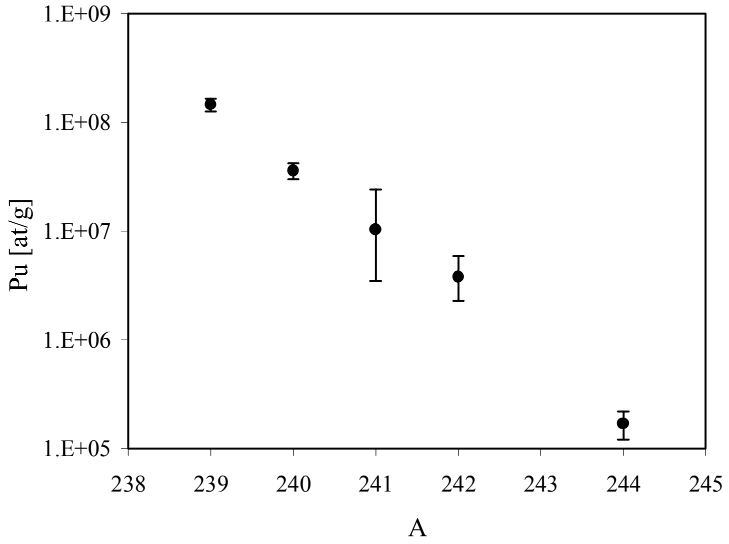


Fig1.
Figure 1 shows the measured concentrations of the plutonium isotopes. The value of ^{241}Pu ($T_{1/2}$ = 14.35 y) is decay corrected for 1960.

	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{244}Pu
cts.	142	58	2	7	12
C [10^6 at/g]	146 ± 19	36 ± 6	$1.5^{+2.0}_{-1.0}$	$3.8^{+2.1}_{-1.5}$	0.17 ± 0.05

Table1.
The measured counts of each Pu isotope are indicated. The deduced concentration with the statistical error is given in 10^6 at/g. In the case of ^{241,242}Pu are the errors calculated according to Feldman and Cousins [ref] for low count numbers.

Manganese nodule

	external ratios“ (rel. to ^{239}Pu[1])
• ^{239}Pu:	1
• ^{240}Pu:	0.25
• ^{241}Pu:	0.01 (0.07 d. corr.)
• ^{242}Pu:	0.026
• ^{244}Pu:	0.001

[1] after: C. Wallner et al., New Astron. Rev. (2003)

And finally:

Why not searching for the unknown such as superheavy elements? An AMS facility can certainly be tuned to regions of the nuclear chart which are far away from known nuclides.