Analytic Progress in Accelerator Mass Spectrometry

ETH zürich

Laboratory of Ion Beam Physics (LIP)

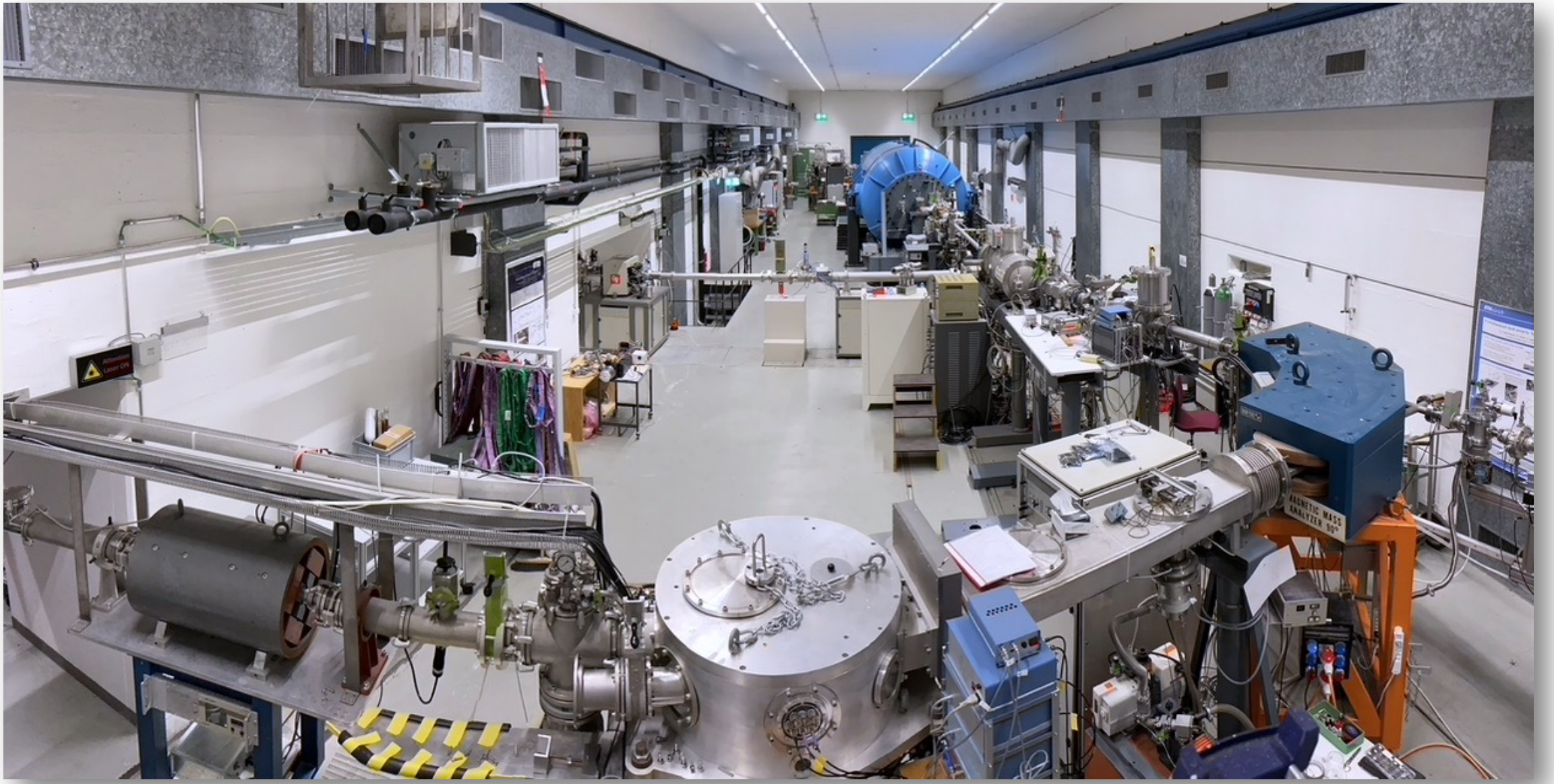
Hans-Arno Synal

2. December 2021, PSI Colloquium

related opportunities for applications with long-lived radionuclides

6 MV Tandem: “good old times in nuclear physics”

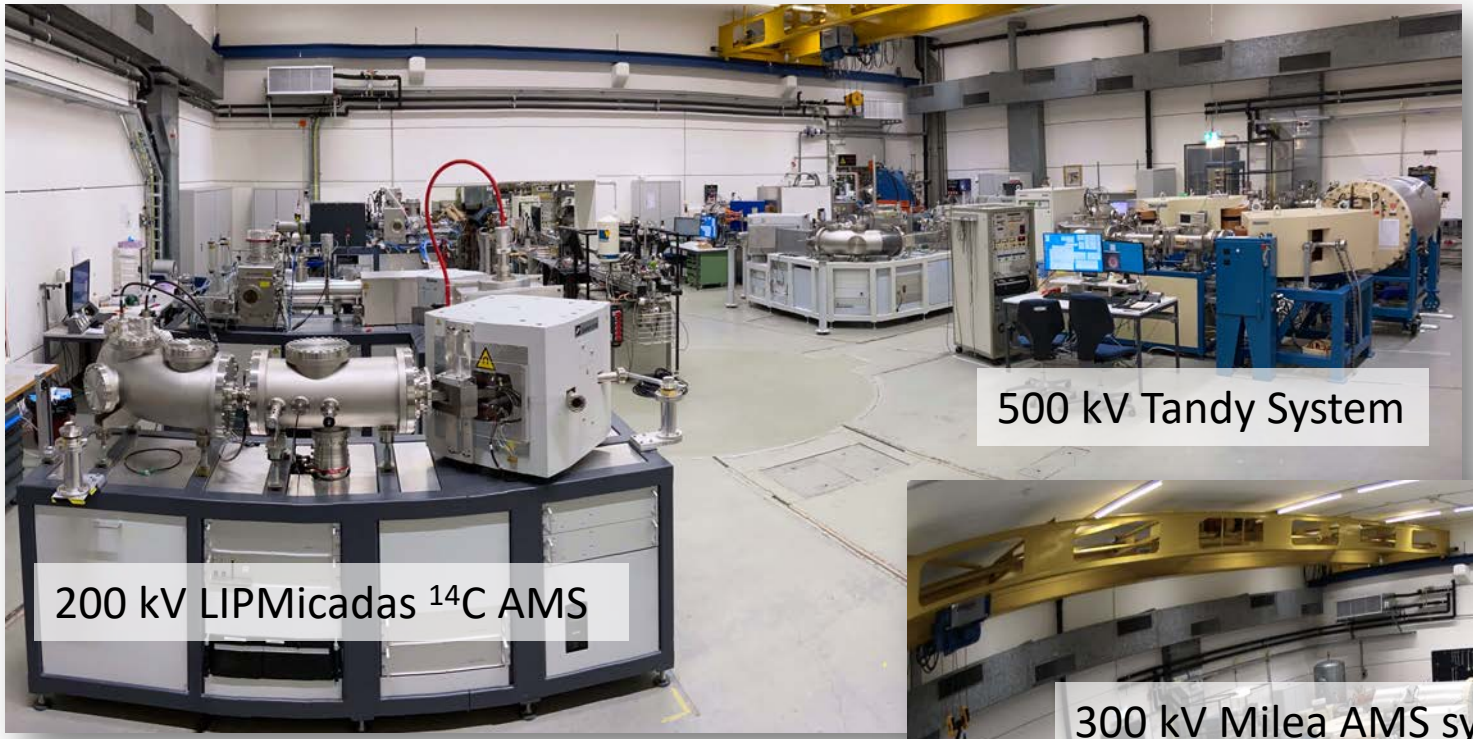
ETH zürich

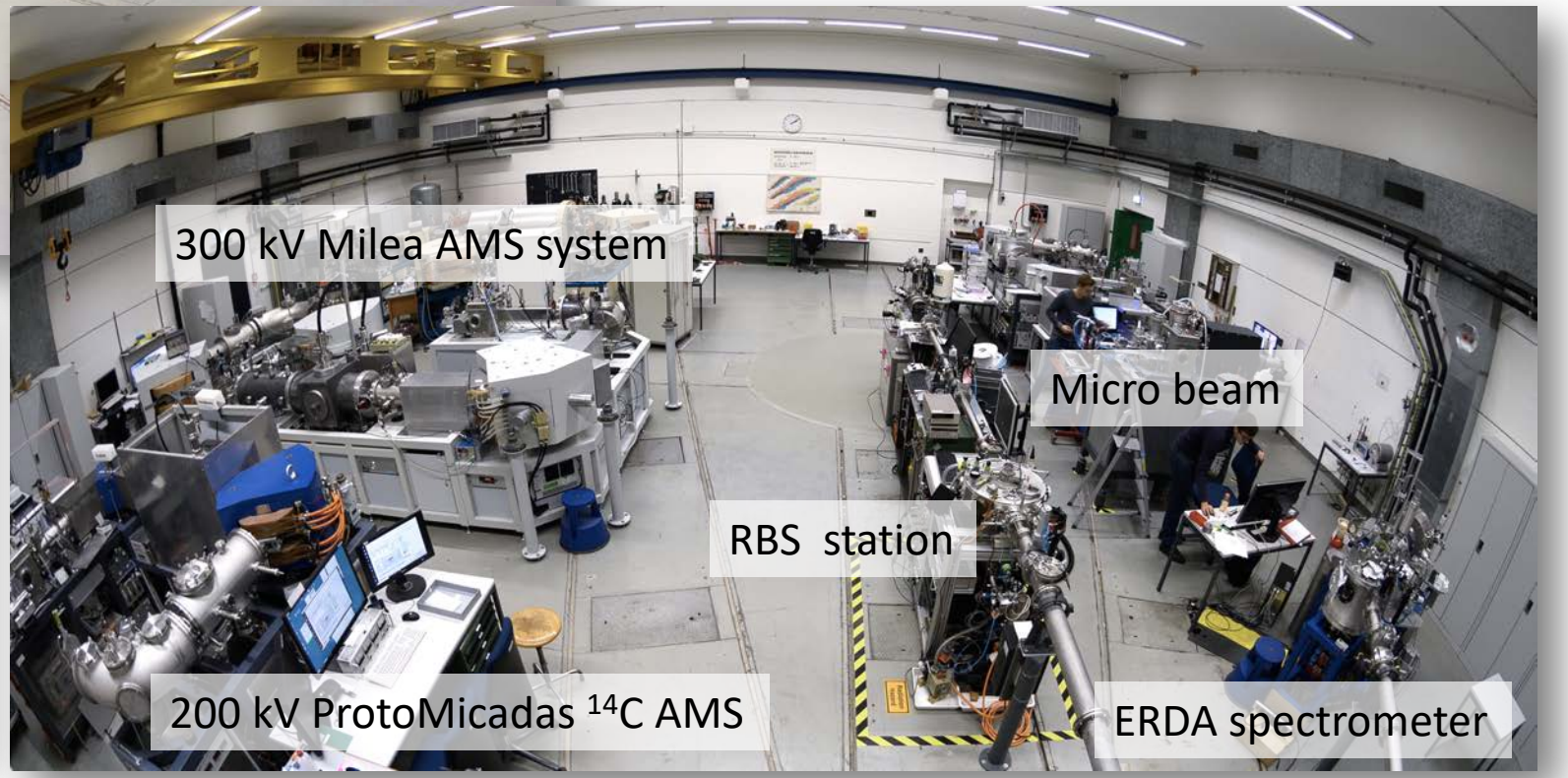


1984-2008 jointly operated between SIN/PSI-ETHZ

Versatile instrumentation for AMS and Ion Beam Analyses

ETH zürich





LIP organization

ETH zürich

D-PHYS

Institute of Particle Physics & Astrophysics

Board of trustees (contract period 4 years)

- ETH (D-PHYS, D-ERDW)
- Swiss Federal Institute of Material Sciences and Technology (Empa)
- Paul Scherrer Institut (PSI)
- Swiss Federal Institute of Aquatic Science and Technology (Eawag)
- Swiss Federal Institute for Forest, Snow and Landscape Research (WSL)

ETH zürich

EMPA
Materials Science & Technology

PAUL SCHERRER INSTITUT
PSI

eawag
aquatic research

WSL

37 Employees on 32 FTE; 2020 budget: 4.06 MCHF; ETHZ: 21%; Partner: 14%; Grants: 18 %; Drittmittel: 47%

AMS: a unique detection technique

ETH zürich

Accelerator Mass Spectrometry

AMS

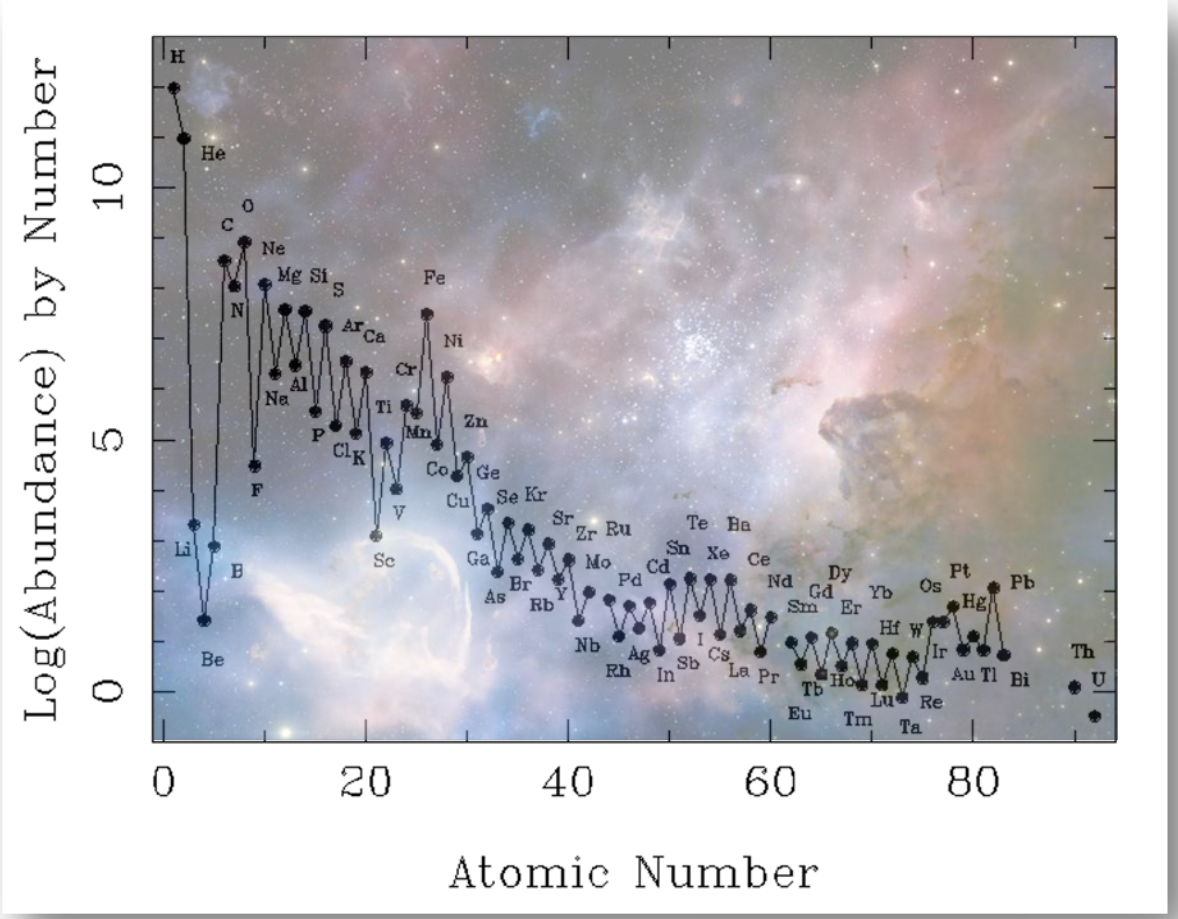
important AMS radionuclides

^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl, ^{41}Ca, ^{129}I, actinides

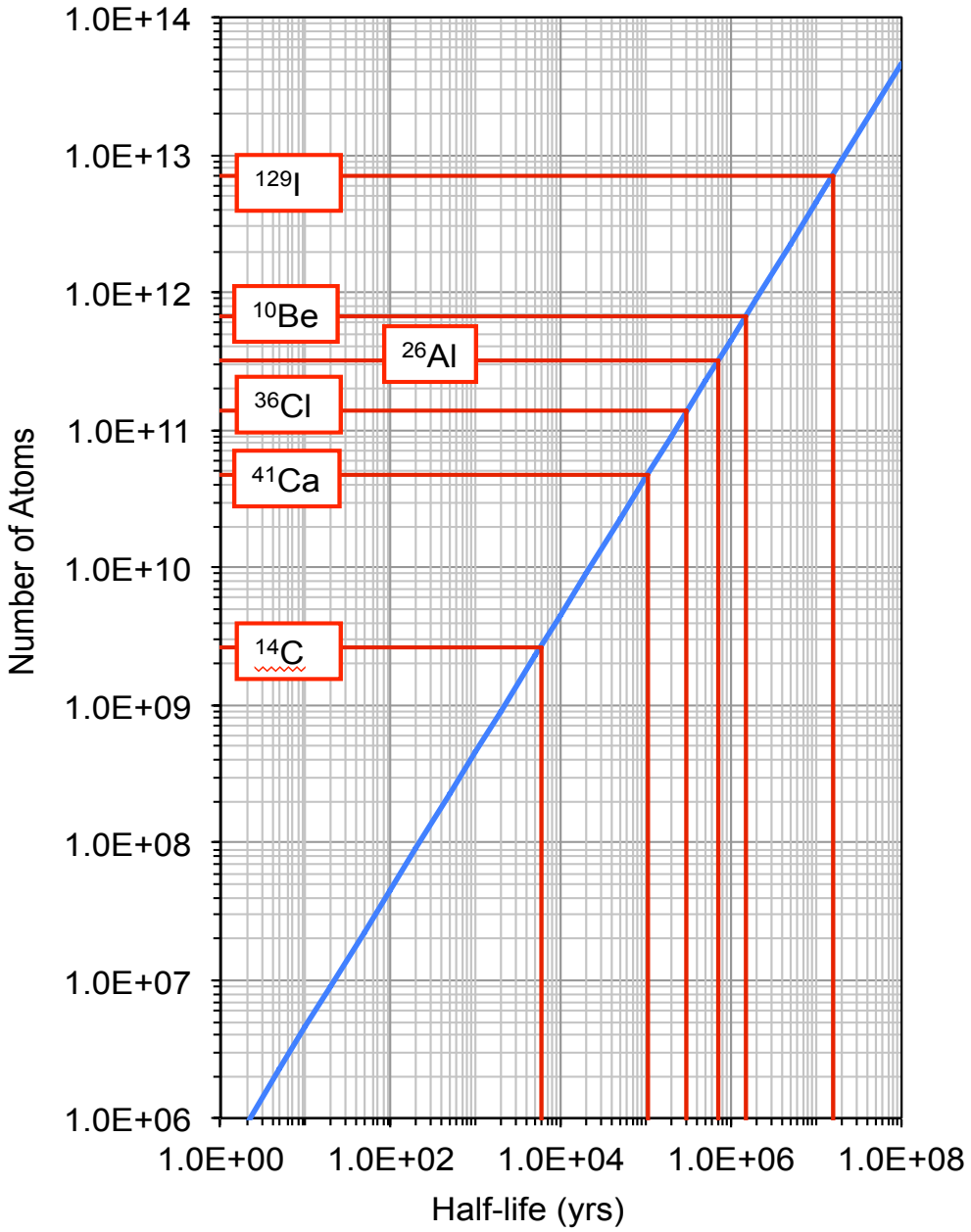
Half-lives: 5’700 -12’700’000 yrs

- Single atom detection capabilities
- High accuracy (‰)
- Dynamic range (10^4)
- Extreme sensitivity

Isotope ratios: 10^{-9} - 10^{-15}

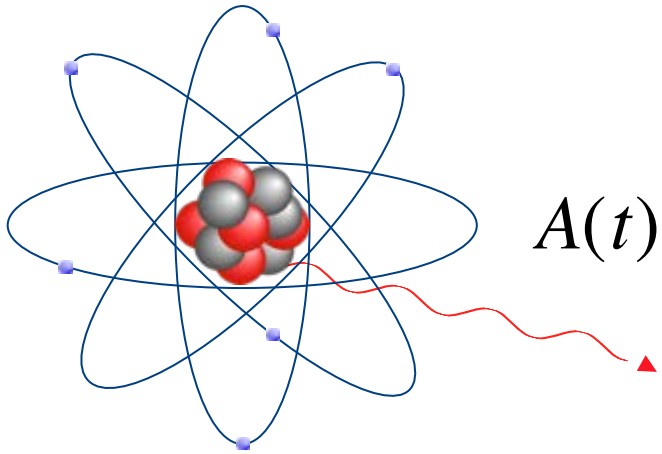


Decay counting



How many Atoms we need
for a good measurement (1%) ?

$$N = \frac{A}{\lambda}$$



$$A(t) = A_0 \cdot e^{-\lambda t}$$

N: Number of atoms

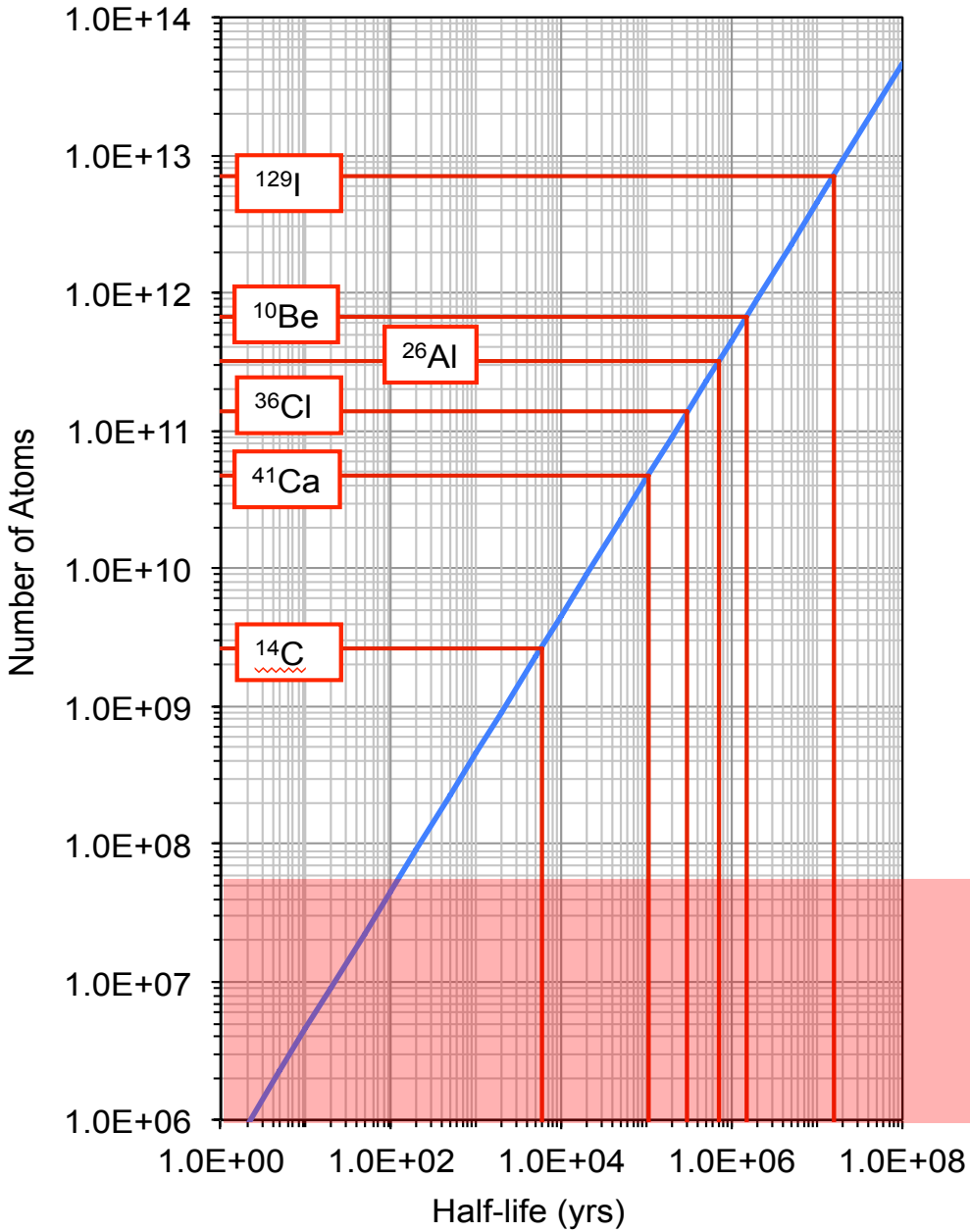
A: Activity

λ: Decay constant

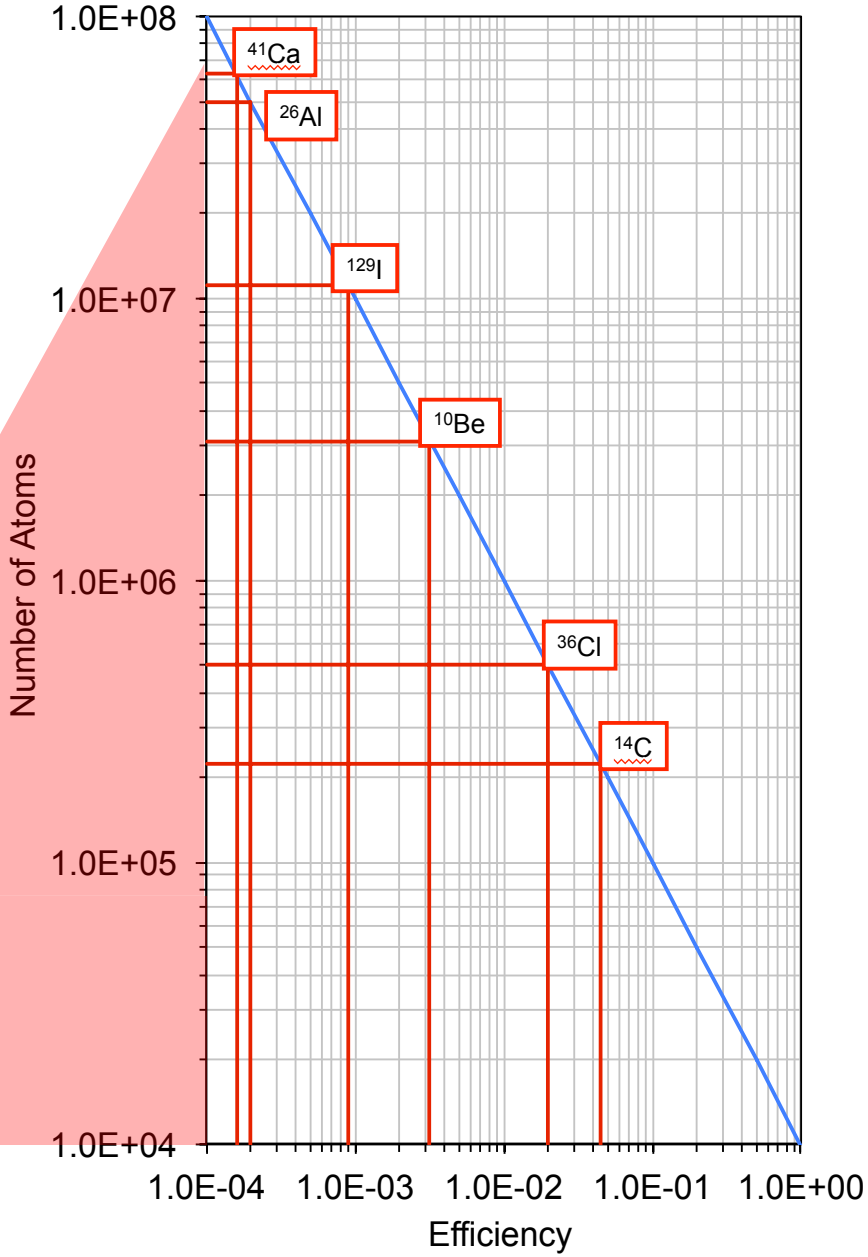
Reasonable assumptions:

- Measurement time: 10^6 s (12 days)
- Minimum count rate: 0.01 cps
- Detection efficiency: 100 %

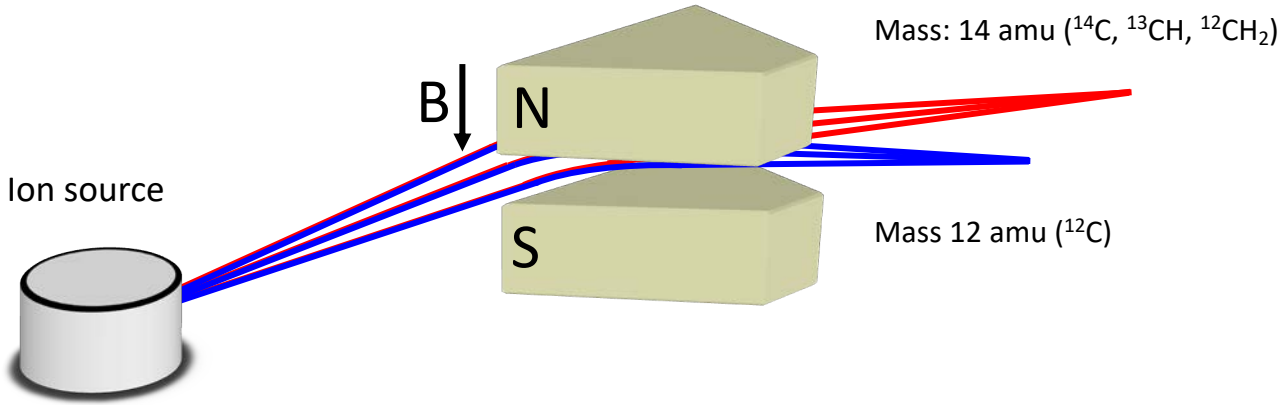
Counting atoms



At least 4 orders of magnitude better!!!

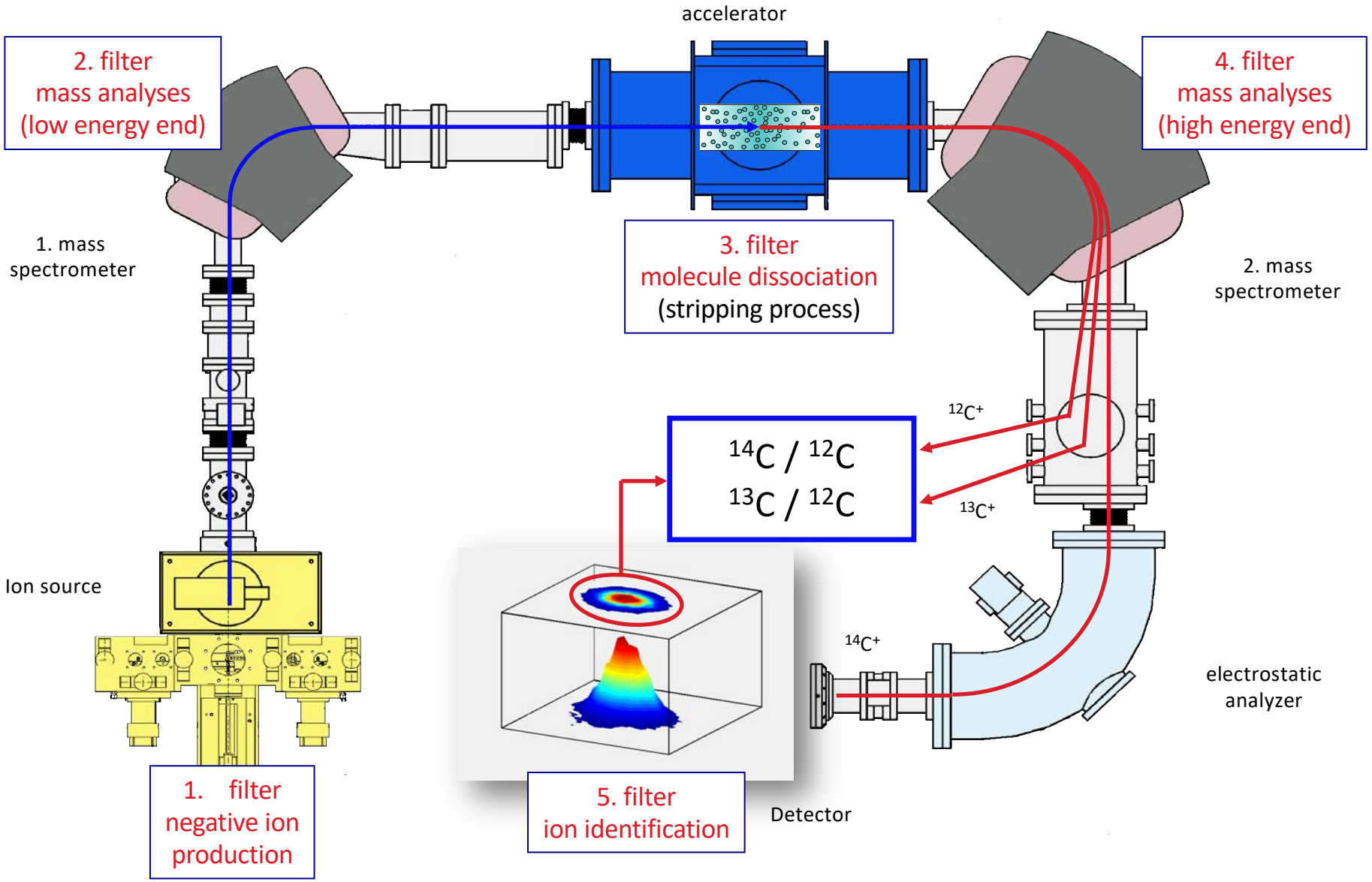


Mass spectrometric separation



isobar/ molecule	mass (MeV/c^2)	Δmass (MeV/c^2)	m/Δm
^{14}C	13044.0422		
^{14}N	13043.8861	0.1561	83562
$^{12}CH_2$	13055.6004	11.5582	1129
^{13}CH	13051.4364	7.3942	1766

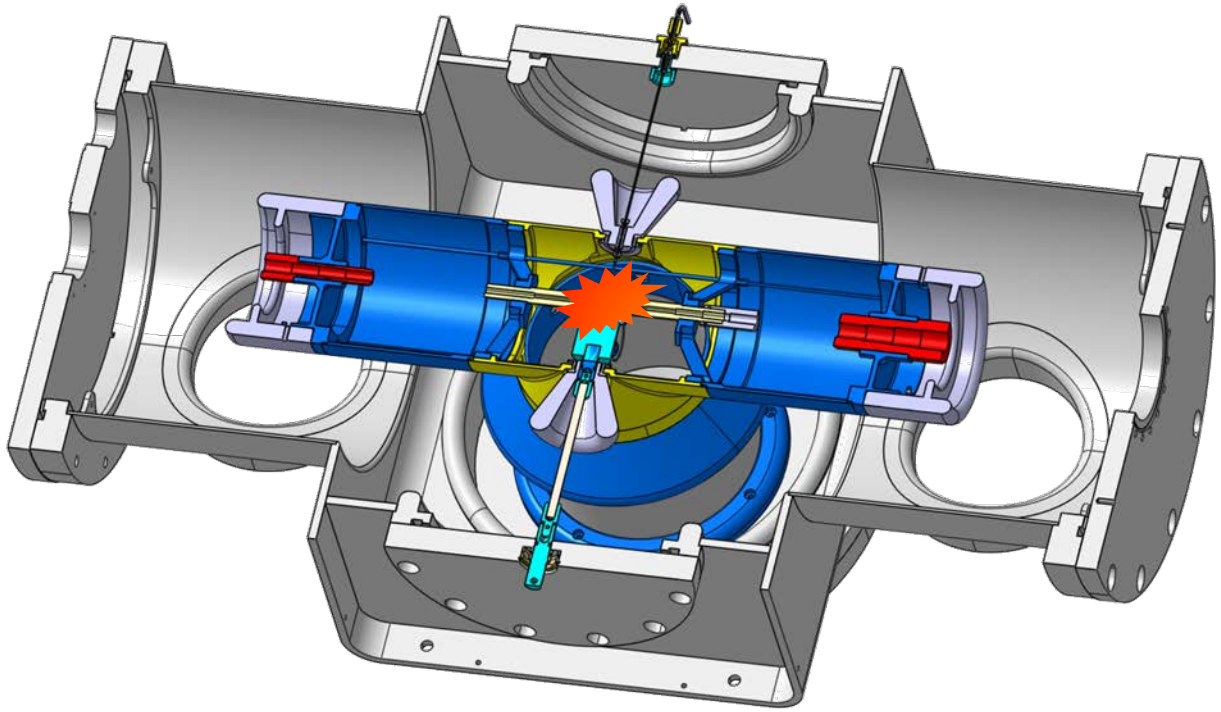
State-of-the-Art AMS system



Molecule dissociation

atomic & molecular ions: $^{14}C, ^{13}CH, ^{12}CH_2$

mass 14 amu



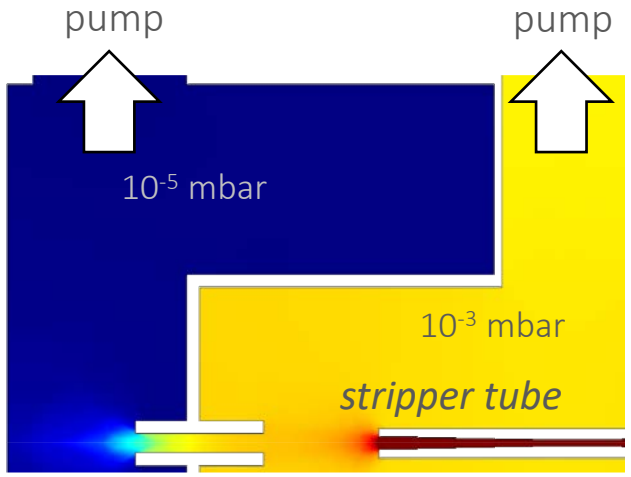
only atomic ions $^{14}C, ^{13}C, ^{12}C$, H

mass:
- 14 amu
- 13 amu
- 12 amu
- 1 amu

Optimizing performance of AMS instruments

ETH zürich

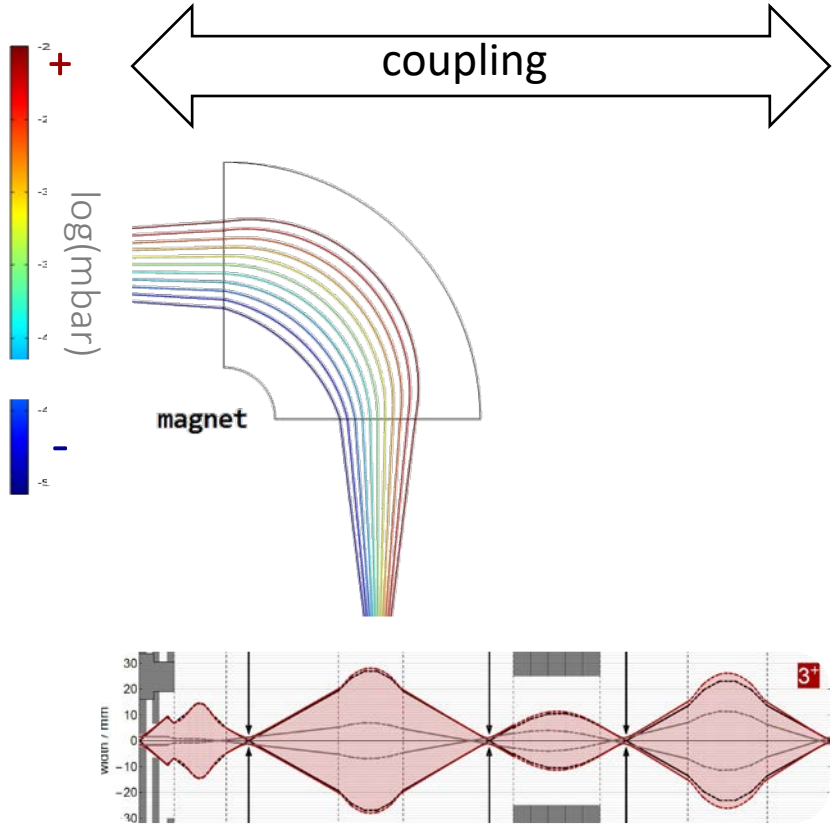
Gas flow model



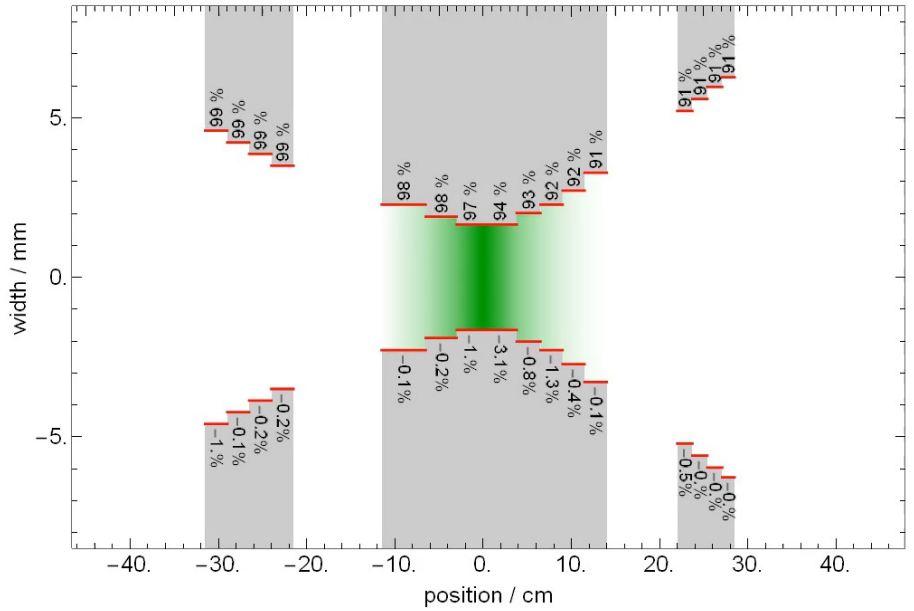
Determine and optimize:

- Gas density distribution
- Total gas flow
- Direct injection into adjacent tubes

Ion optics/ray tracing



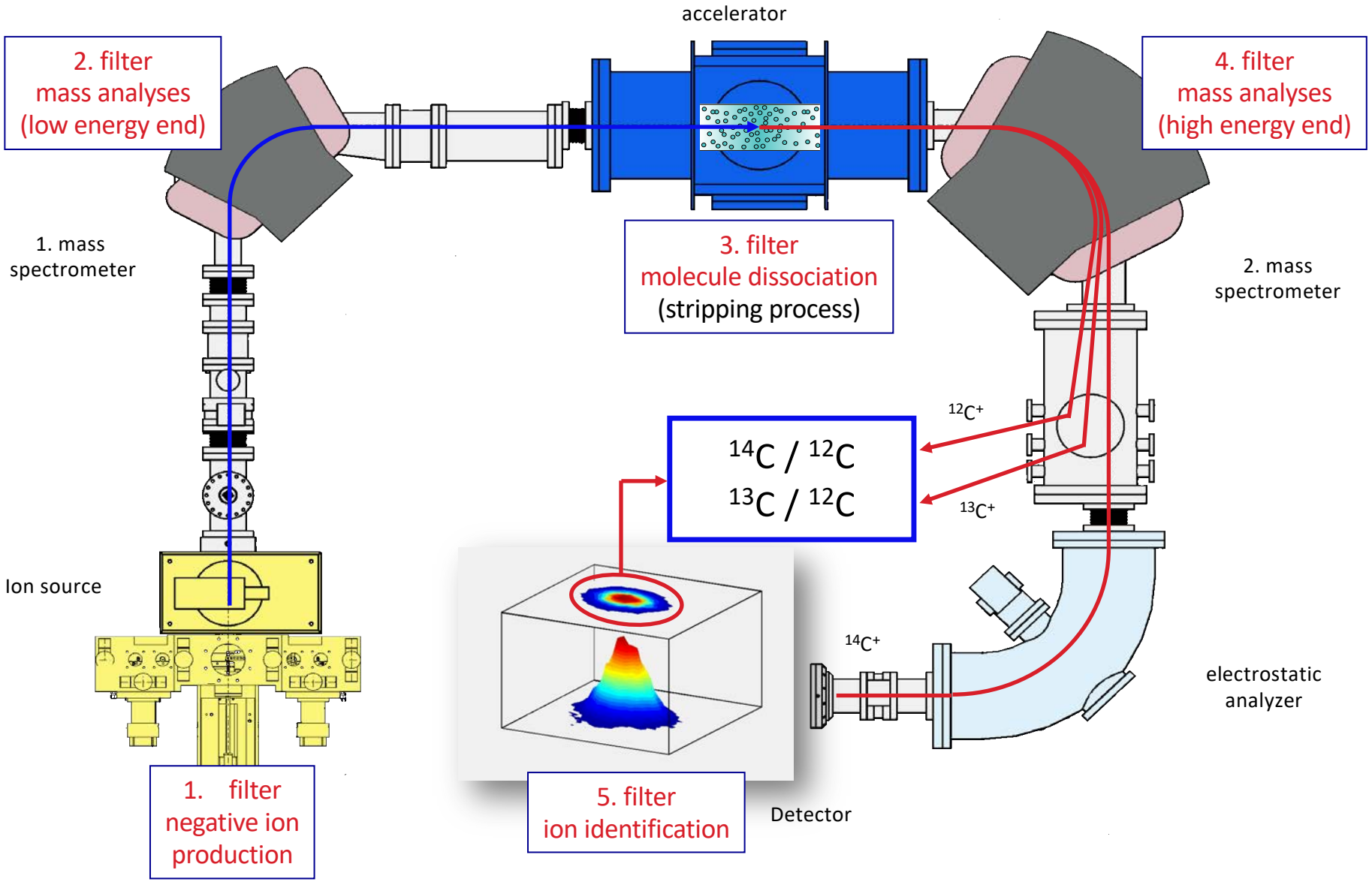
Nuclear scattering simulation



Monte Carlo simulation

- Arbitrary geometries
- Inhomogeneous density profiles

State-of-the-Art AMS system



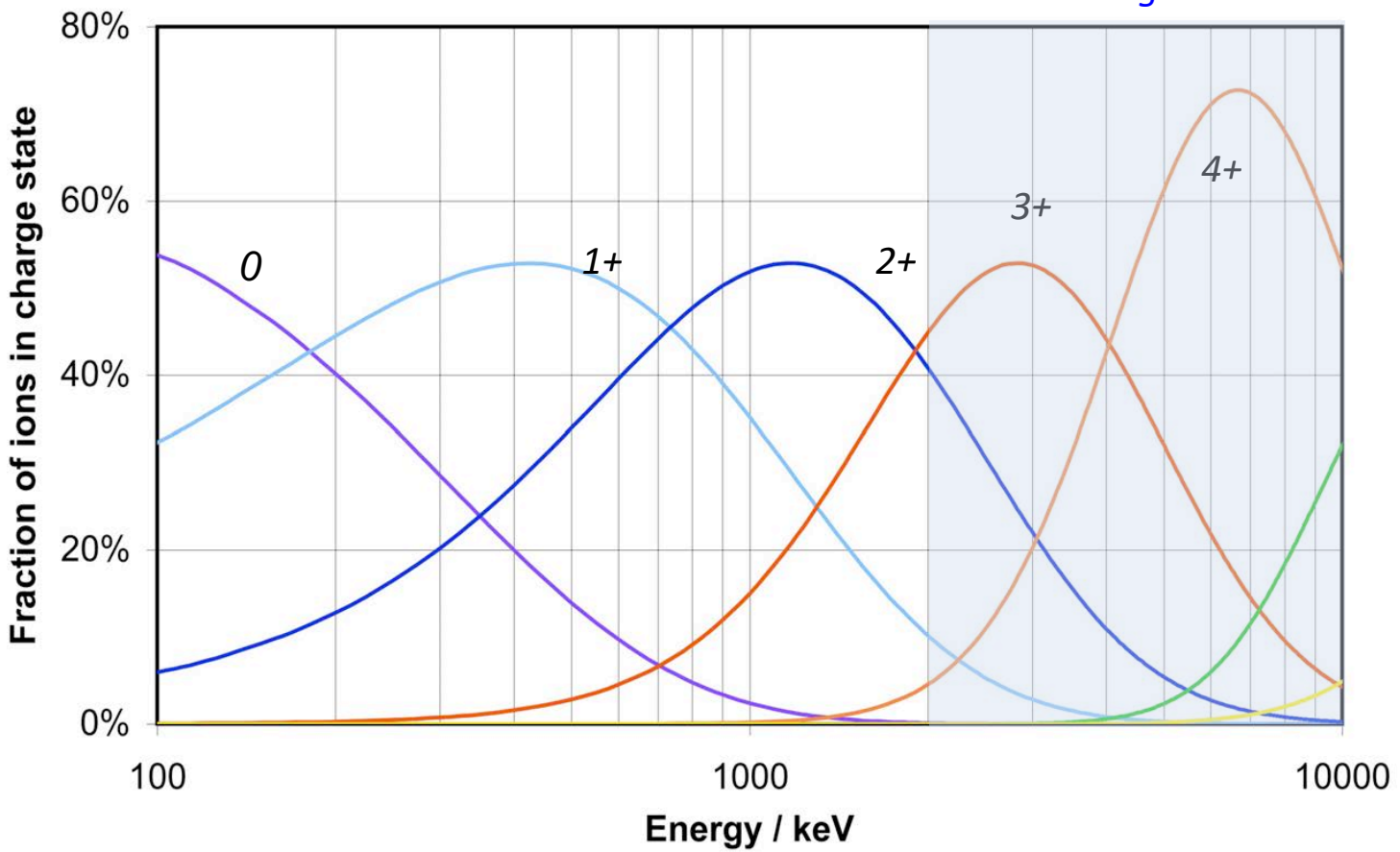
Charge state yield with ion energy

ETH zürich

Traditional AMS
2.5 - 9 MV

Coulomb disintegration

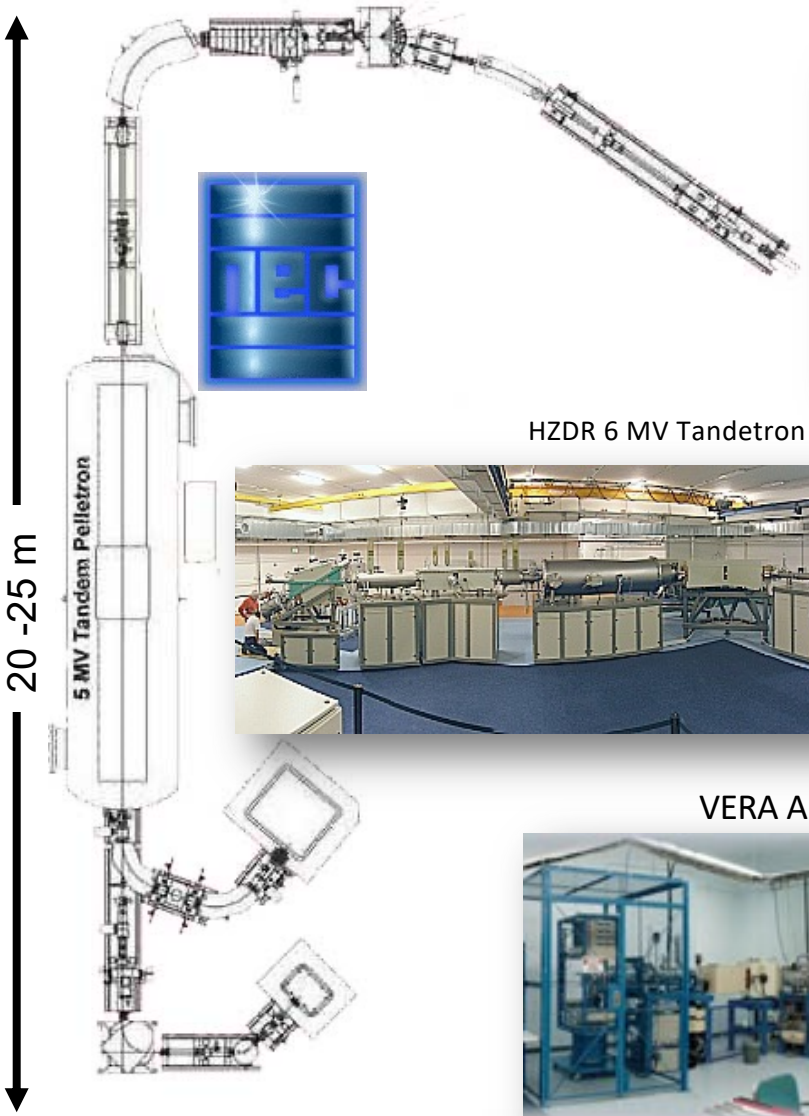
Argon gas stripping



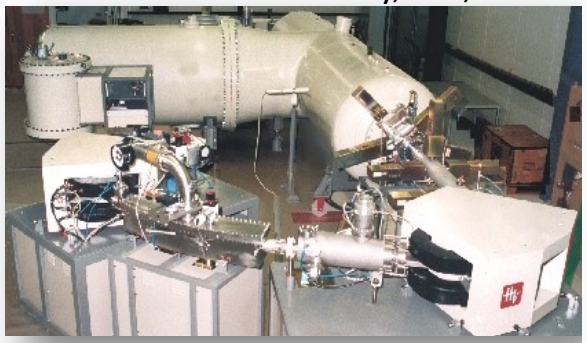
"golden rule in AMS": light molecules are unstable if charge state 3+ of higher is reached

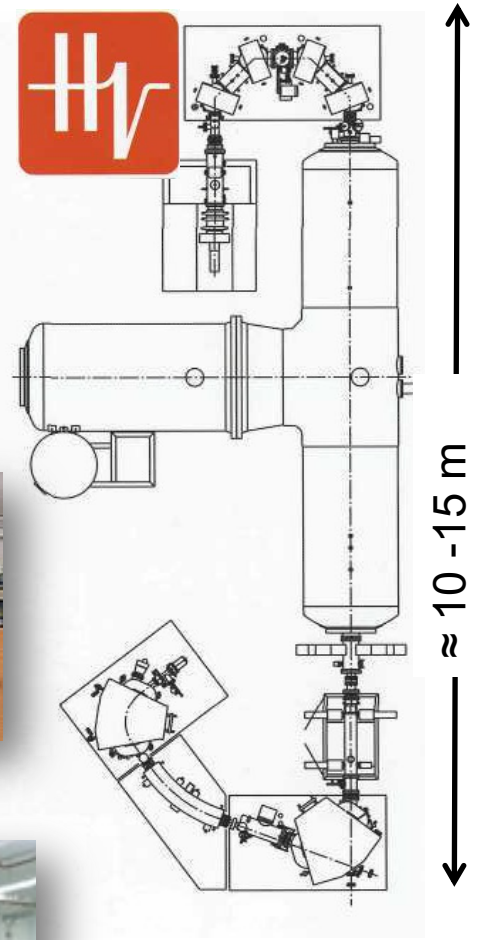
Traditional 3-6 MV AMS systems

ETH zürich



Leibniz AMS 3 MV facility, Kiel, GER





HZDR 6 MV Tandetron AMS facility, Rossendorf, GER

VERA AMS 3 MV facility, Vienna, Austria

Charge state yield with ion energy

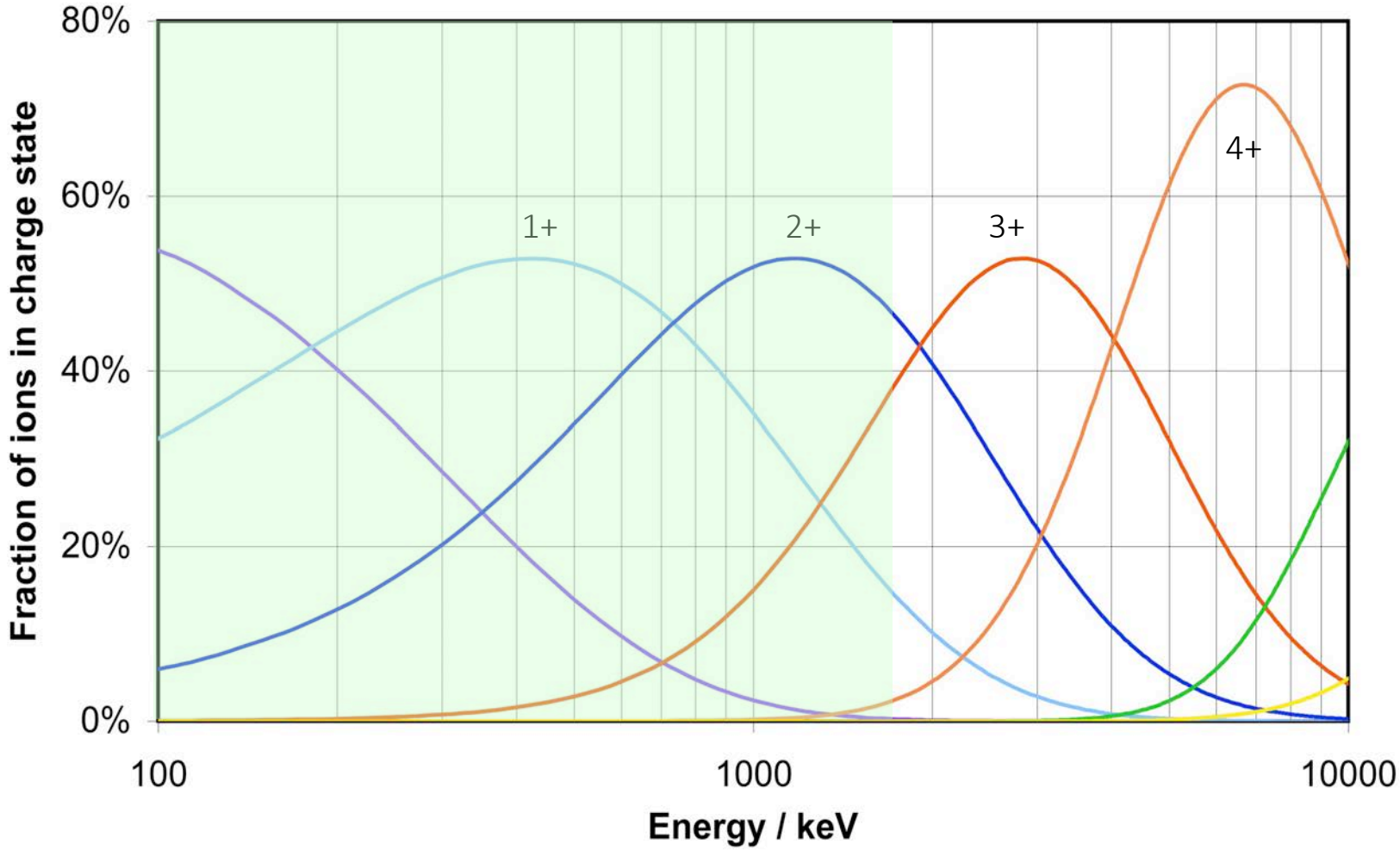
ETH zürich

Compact AMS
0.2 - 1 MV

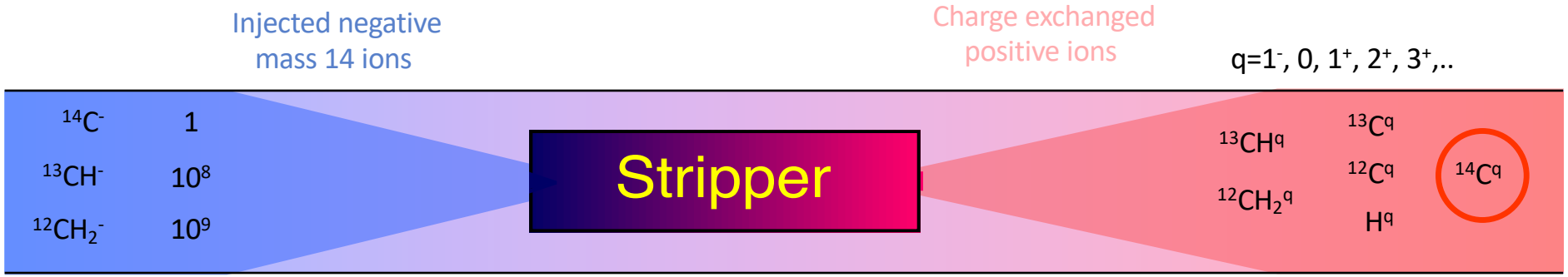
Traditional AMS
2.5 - 9 MV

Multiple ion gas collisions

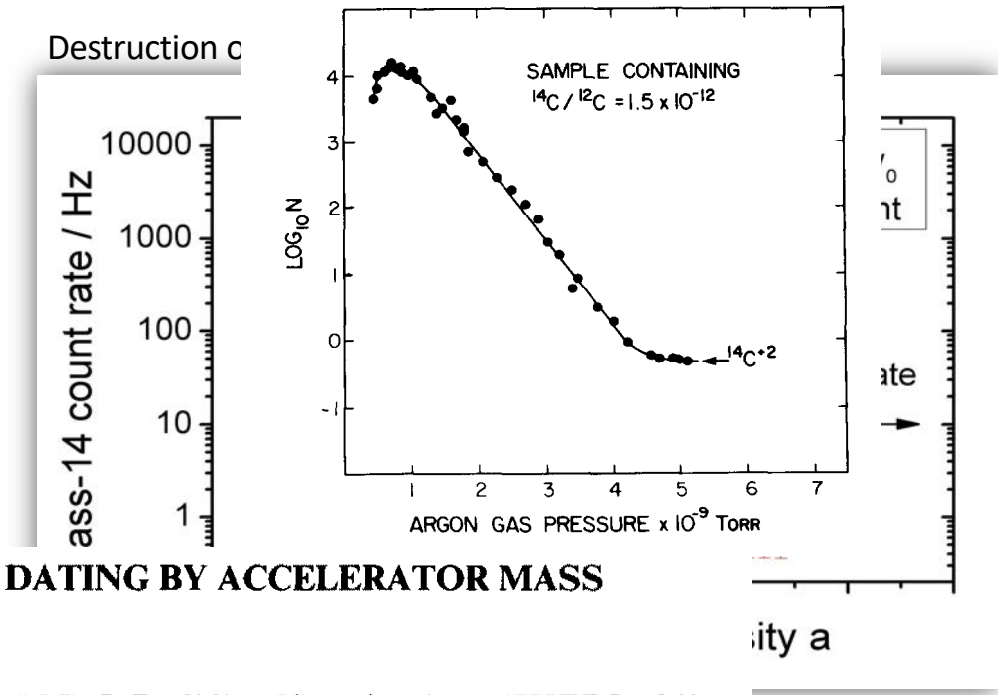
Coulomb disintegration



Stripping Process



- Electron-loss
- Electron capture
- Break-up of molecules
- Energy straggling
- Angular straggling



THE $^{12}CH_2^{2+}$ MOLECULE AND RADIOCARBON DATING BY ACCELERATOR MASS SPECTROMETRY

H.W. LEE, A. GALINDO-URIBARRI *, K.H. CHANG, L.R. KILIUS and A.E. LITHERLAND

ISOTRACE Laboratory, University of Toronto, Toronto, Ontario M5S 1A7, Canada

Schulze-König, T., NIMB 269 (2011) 34-39

The Generation of Compact AMS Systems

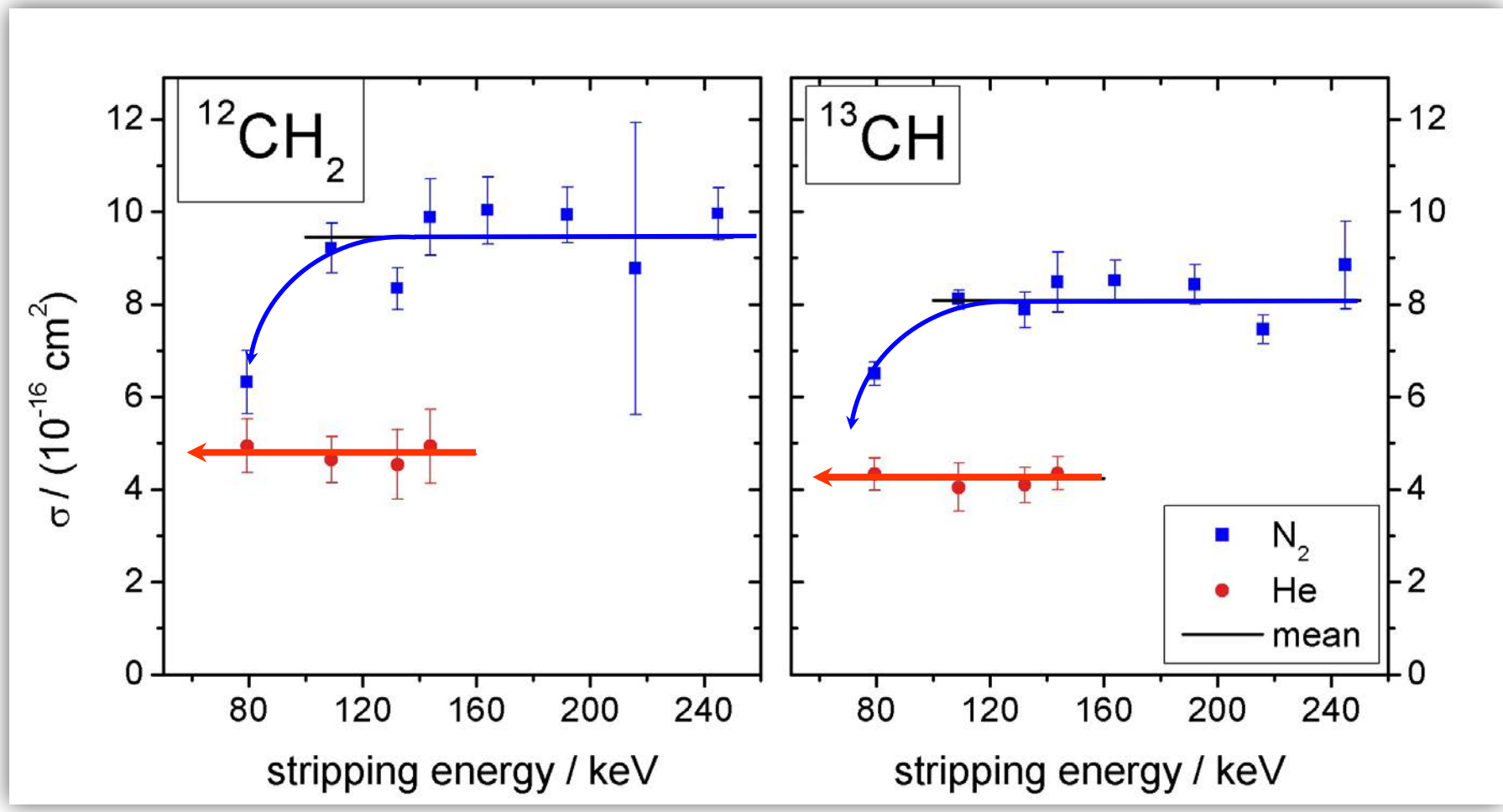
ETH zürich

The first compact AMS system (1998) using charge state 1^+

Commercial systems are on the market from NEC and HVEE

Cross section for molecule dissociation

He stripper gas

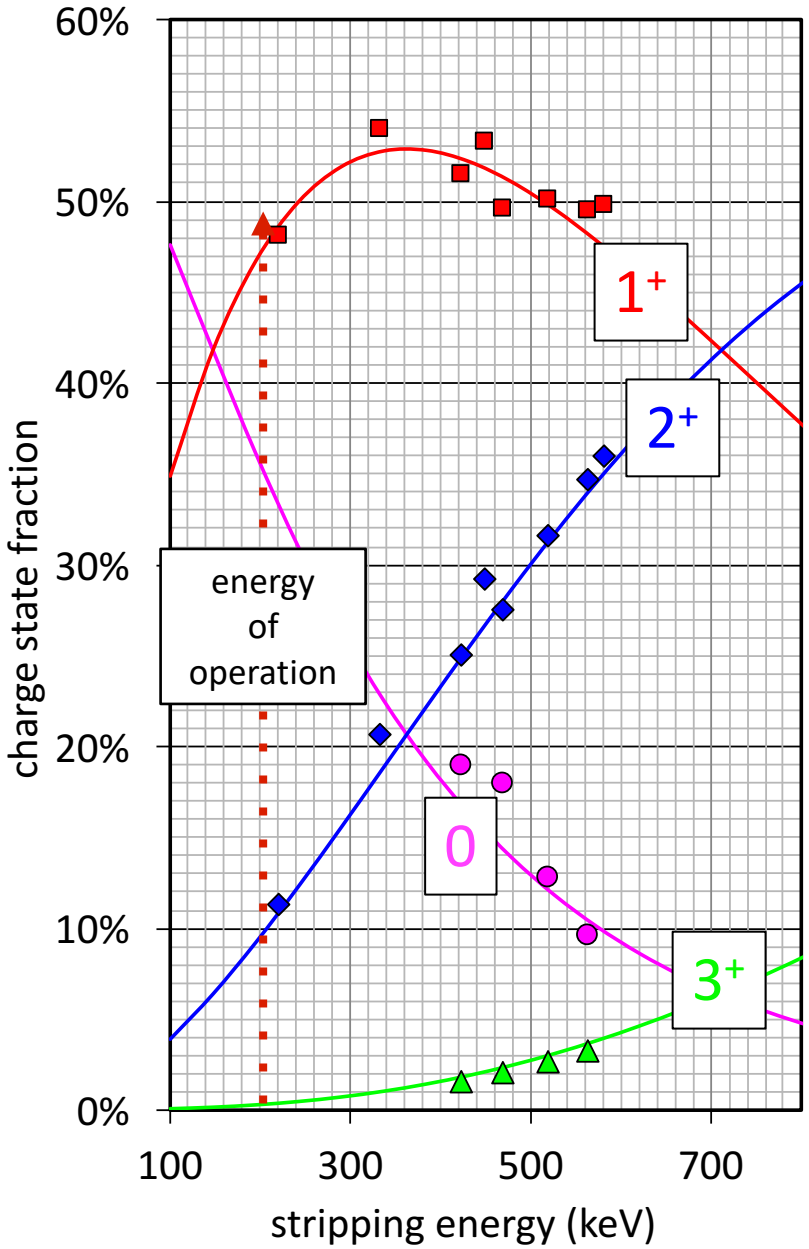


He areal density of ≈ 0.5µg / cm^2 should be sufficient to get rid of molecules

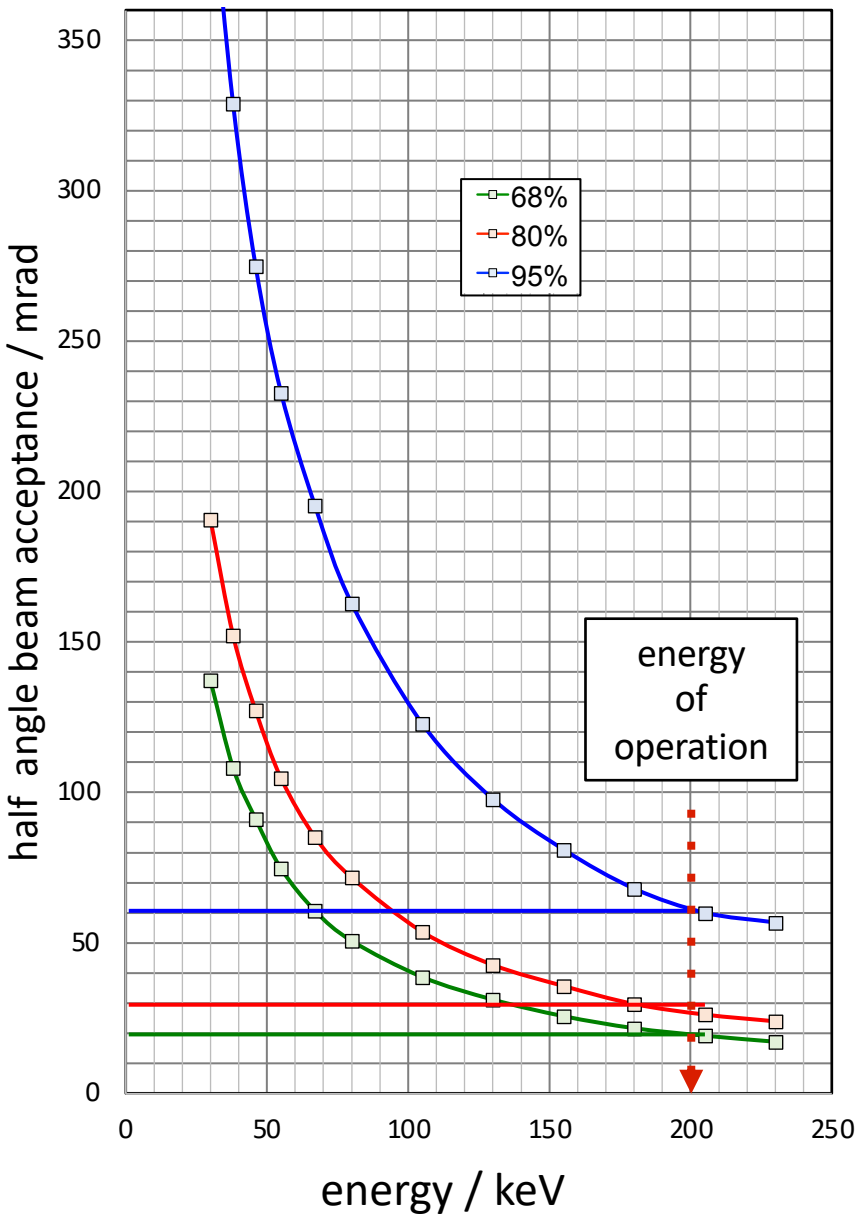
Physical properties of Ar/N_2 stripping

ETH zürich

charge state yield during stripping in Ar

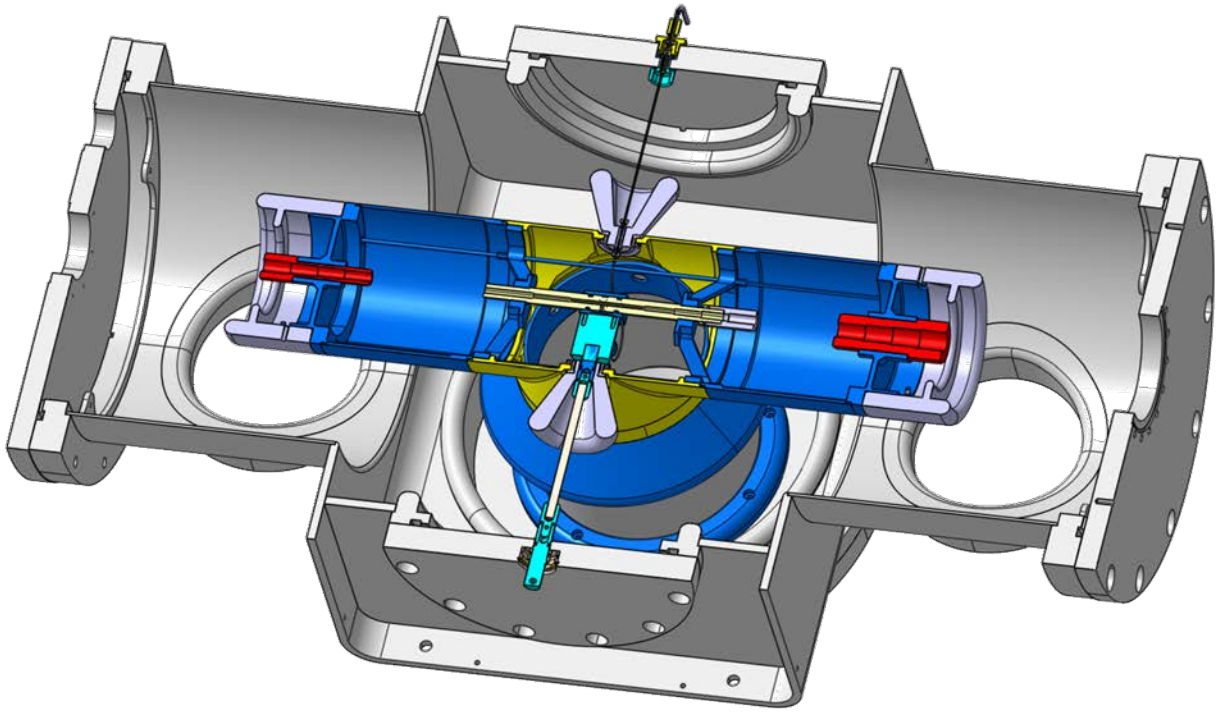


angular straggling during stripping in N_2

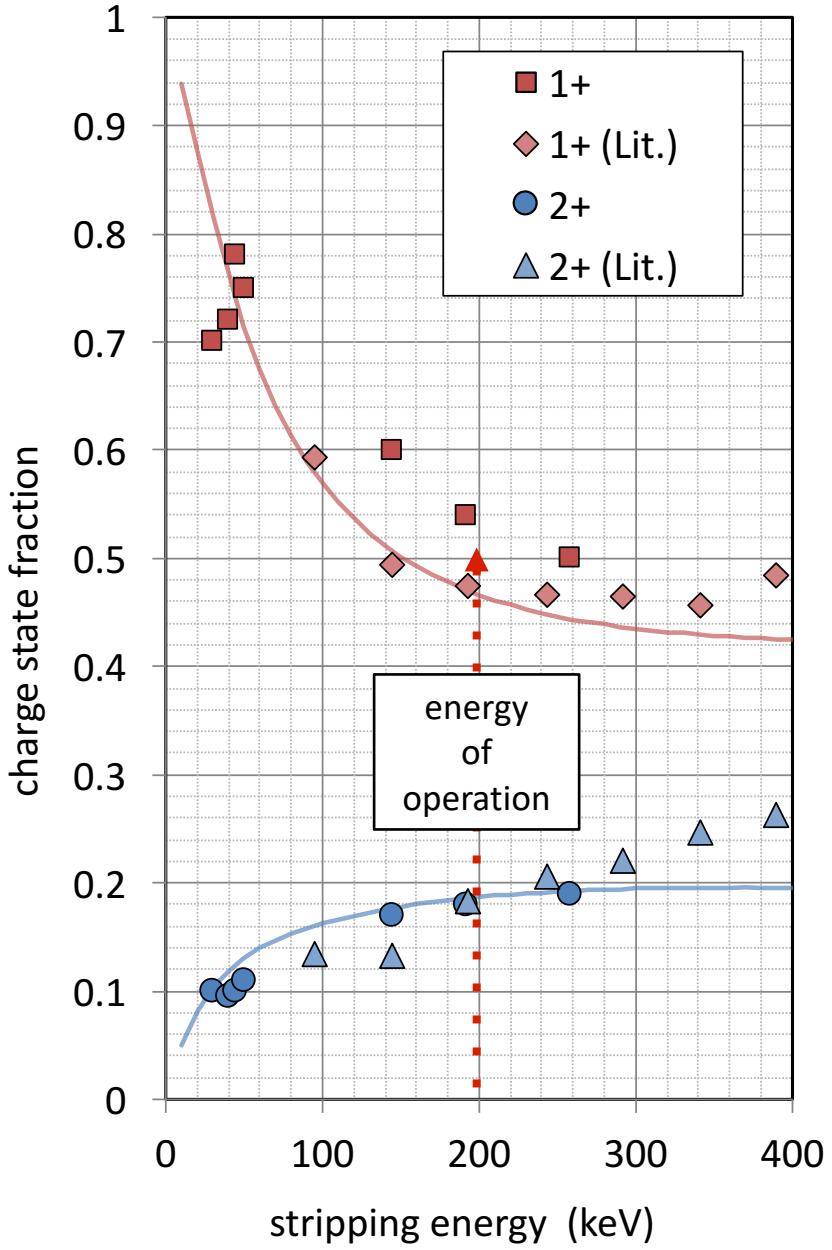


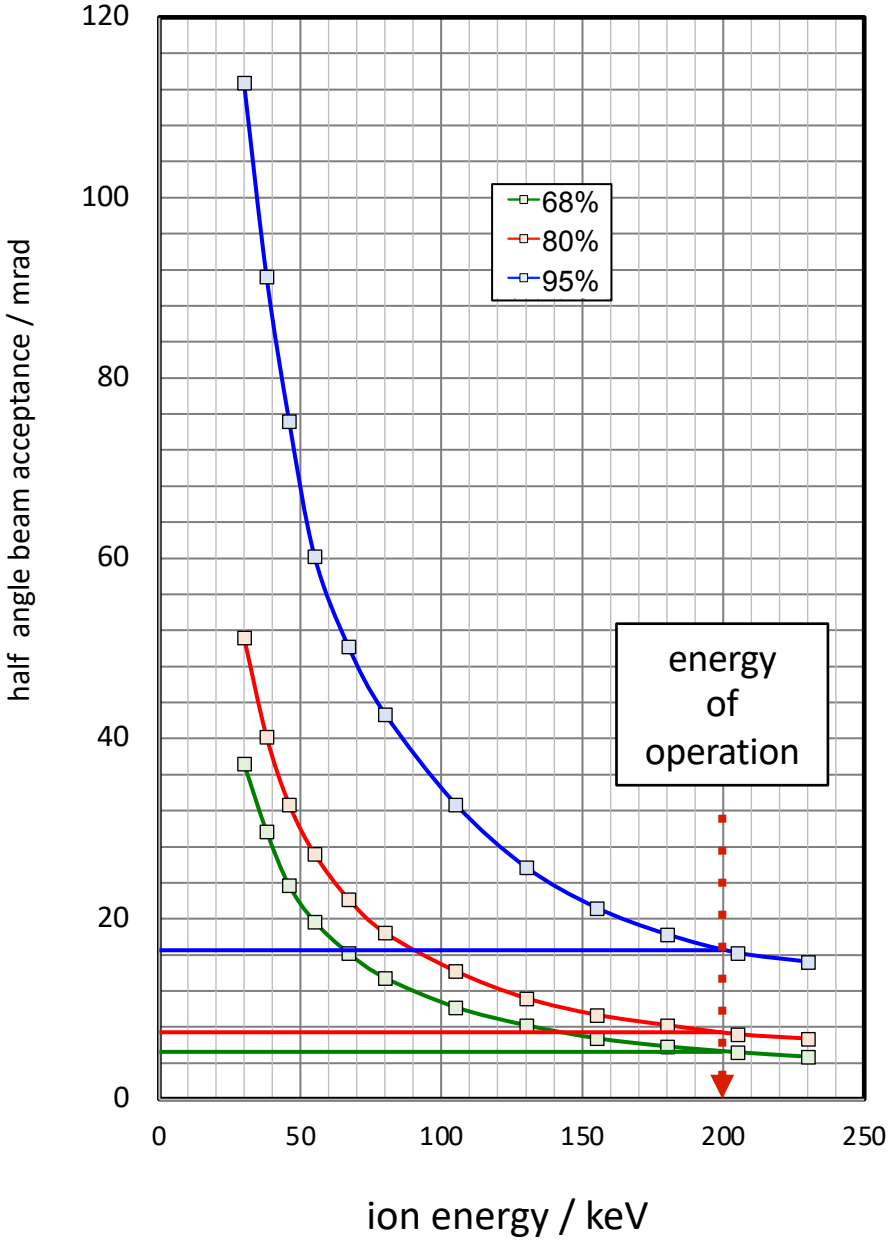
Vacuum insulated High Voltage platform

ETH zürich



Physics of He-stripping at low energies



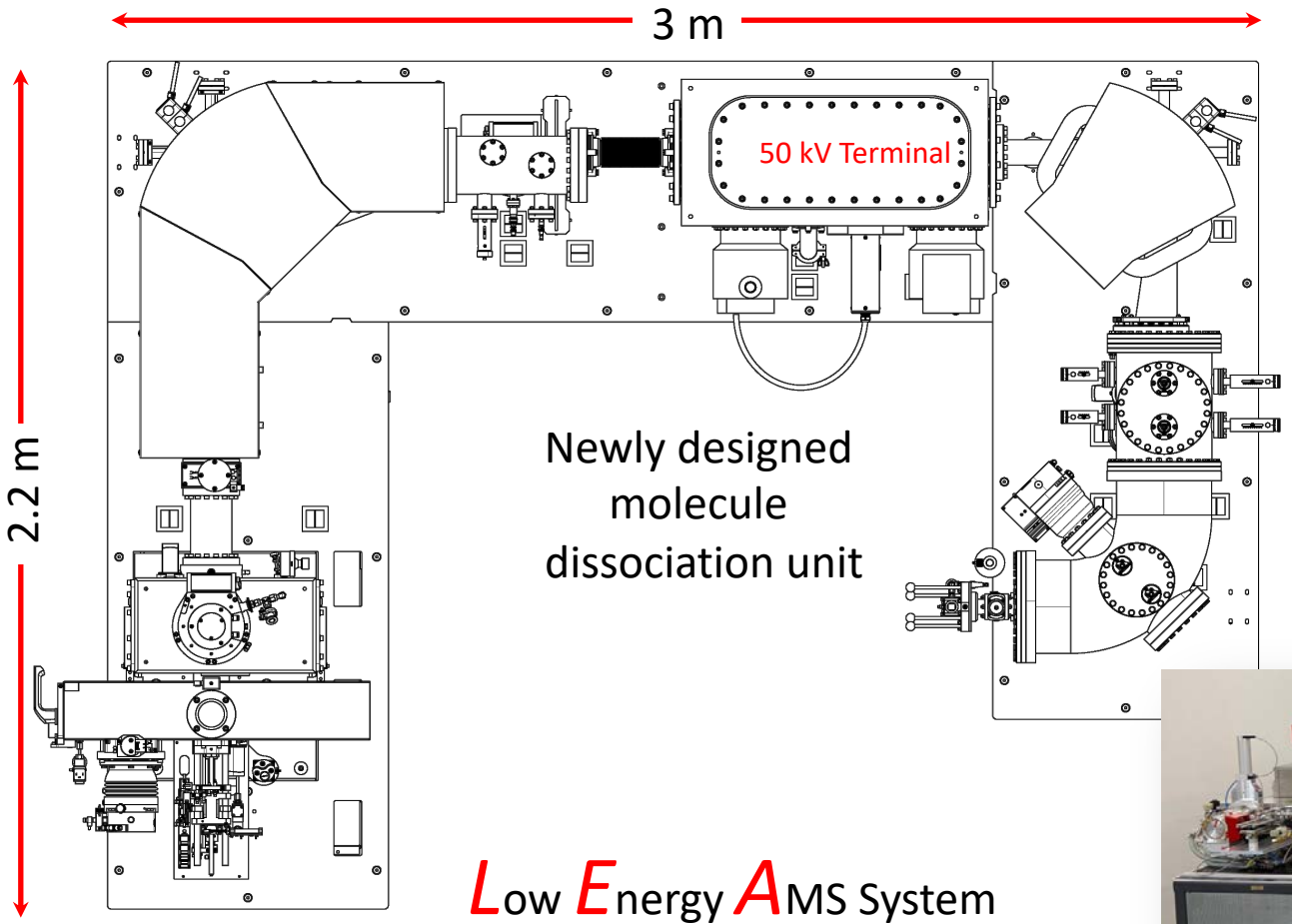


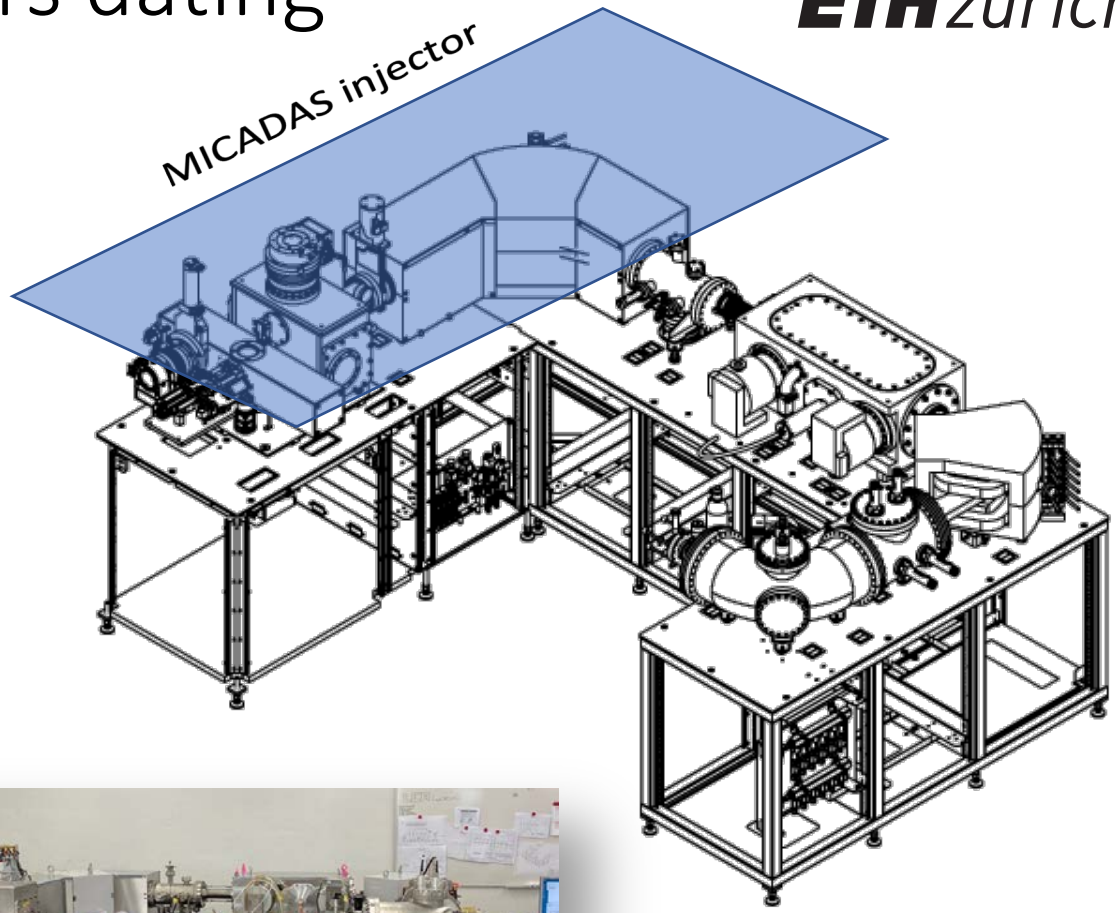
MICADAS: high-performance ^{14}C AMS system

ETH zürich

LEA: A 50kV AMS system for 50kyears dating

ETH zürich





*L*ow *E*nergy *A*MS System

Ionplus
engineering scientific instruments

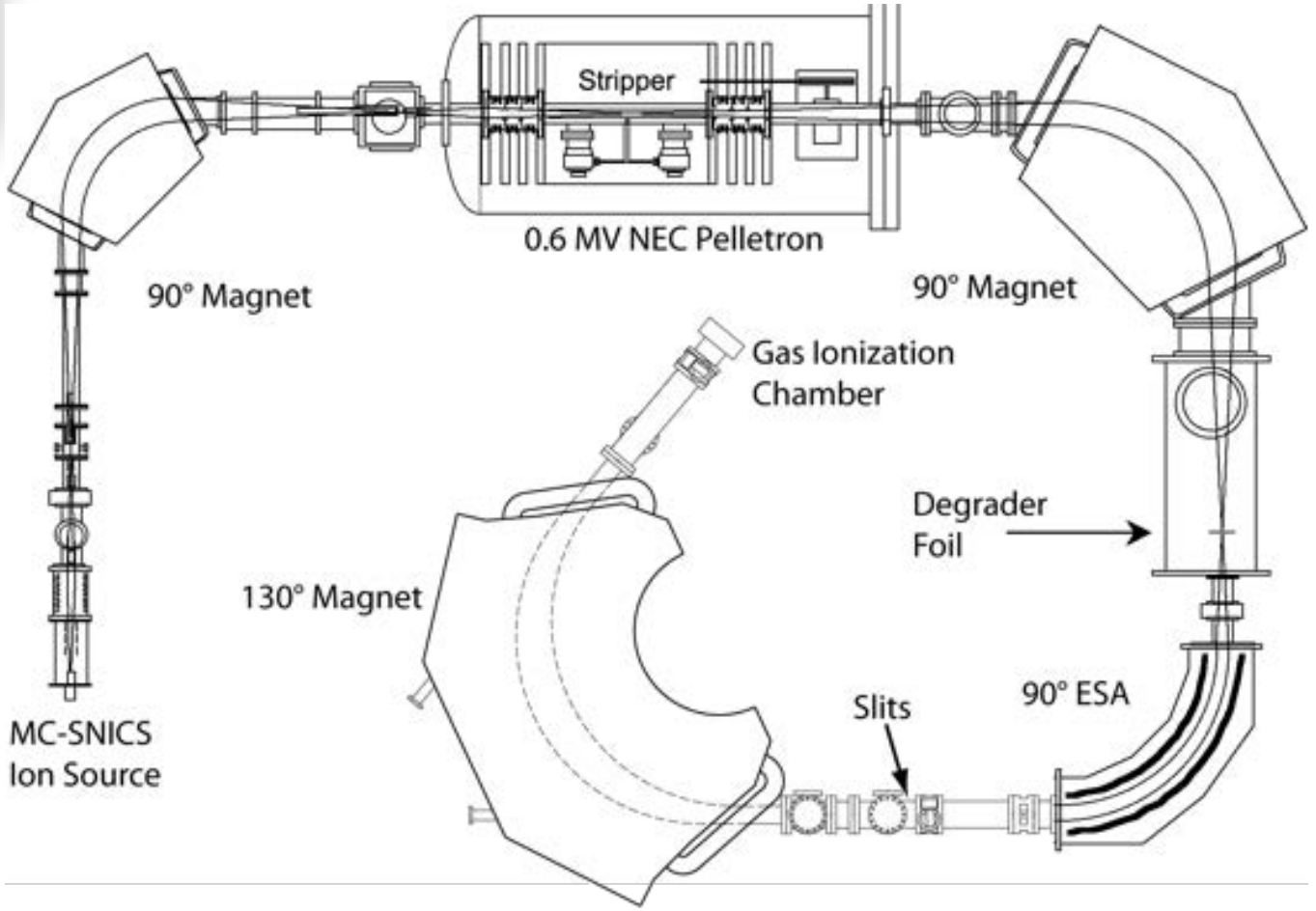
Design protected under European patent: 2559055 licensed to Ionplus AG

Compact multi-isotope AMS systems

- ^{10}Be
- ^{14}C
- ^{26}Al
- ^{41}Ca
- ^{129}I
- Actinides (U, Pu, Pa, Np,…)

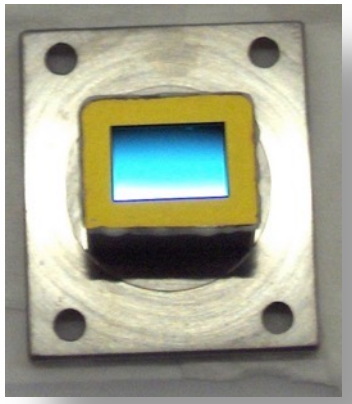
- *Progress*
  in detection technology

- *Better understanding of Stripping processes*

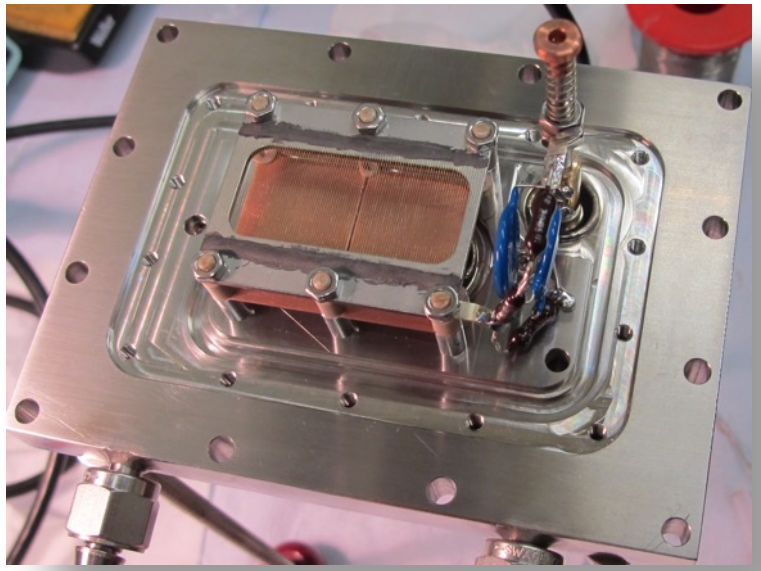


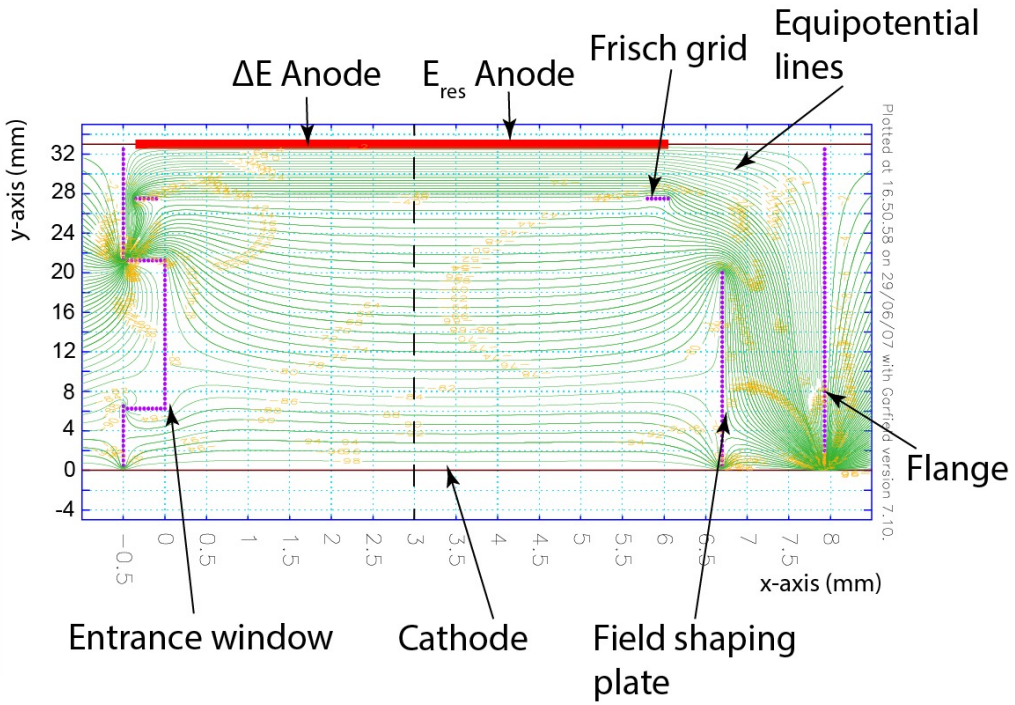
Particle identification: Gas ionization detector

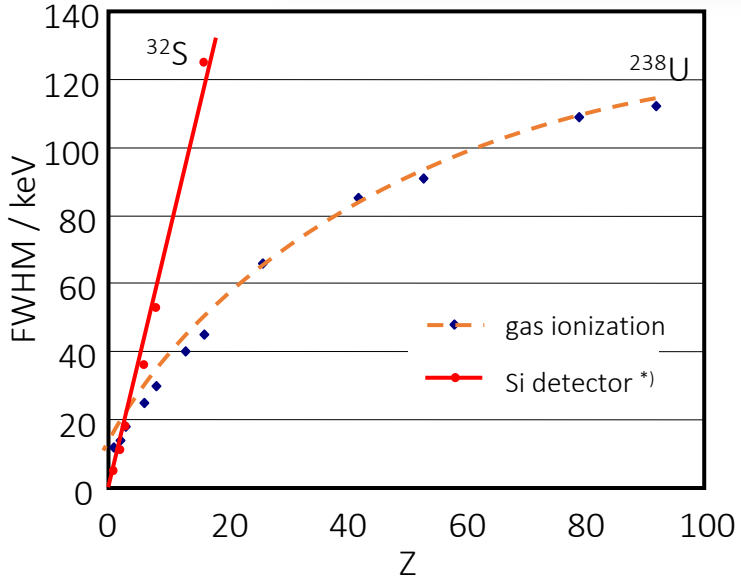
Detector window

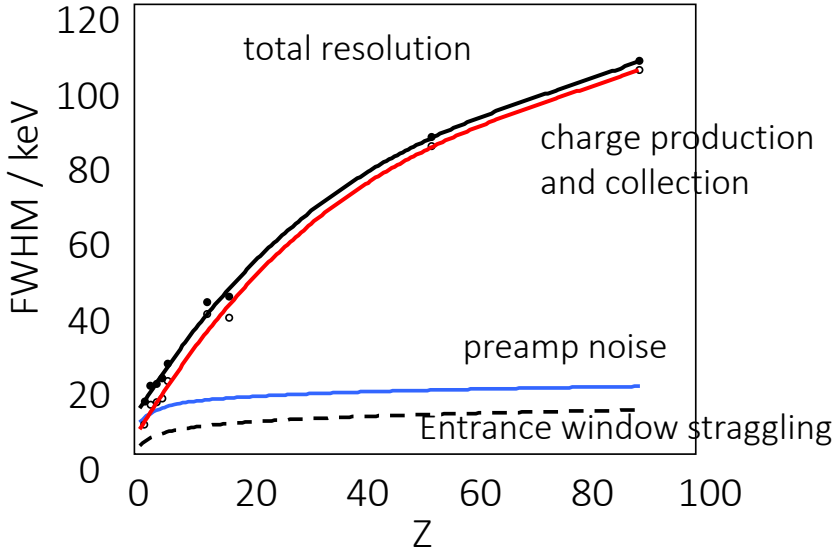


Si_3N_4 membranes

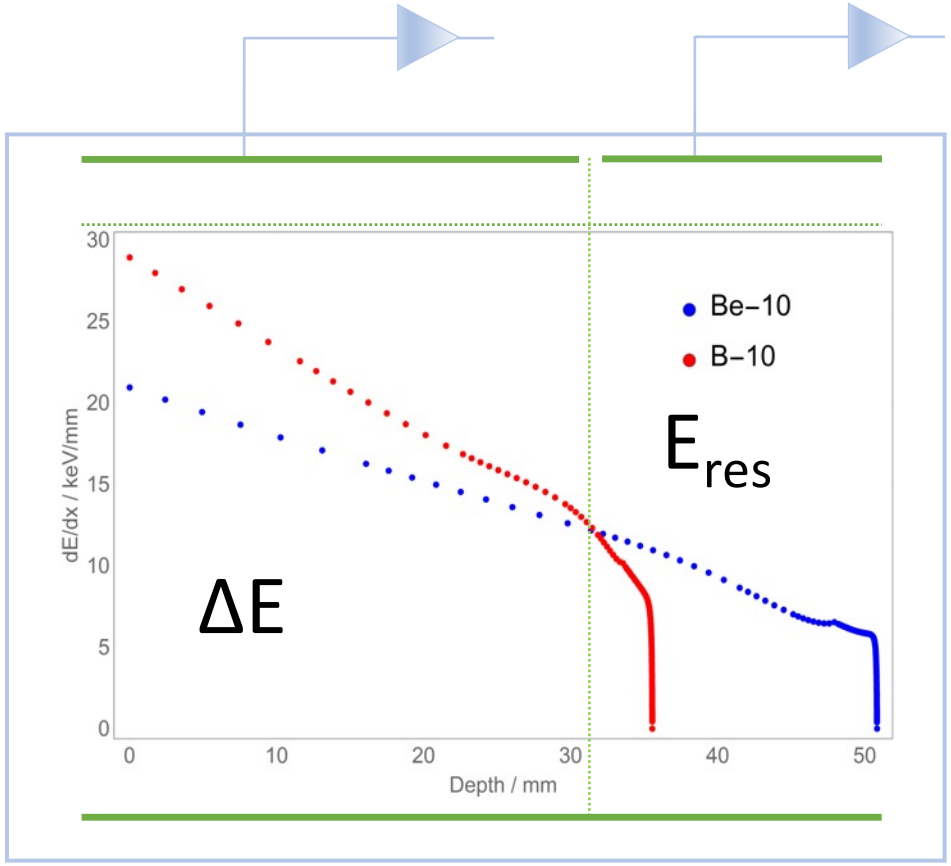


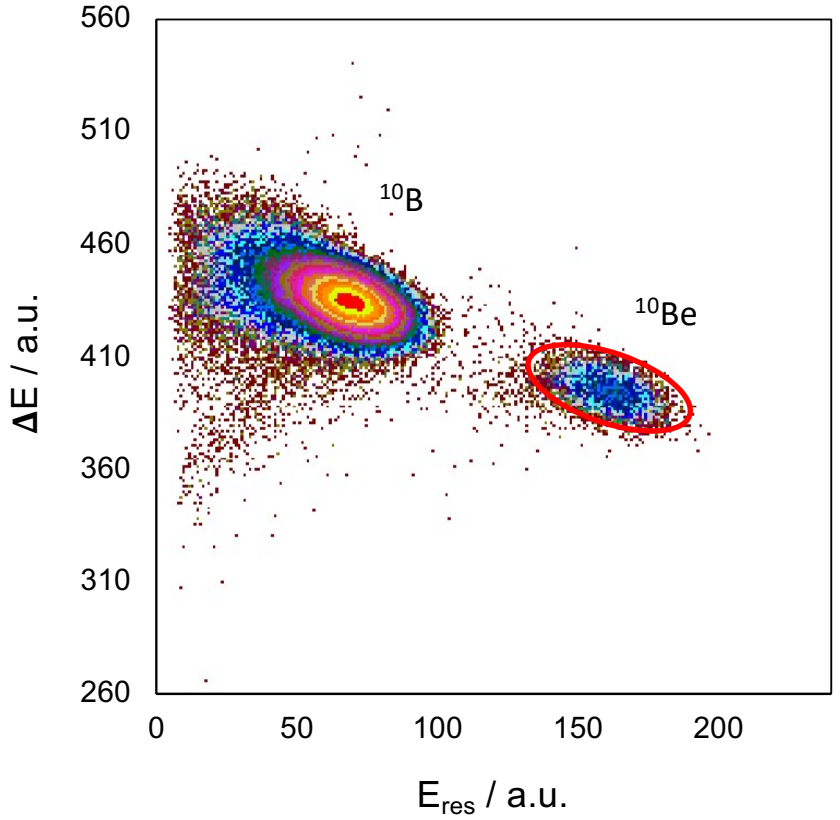






Particle Identification (ΔE-E_{res}) isobar suppression





Enhancing actinide yields @ low energies

ETH zürich

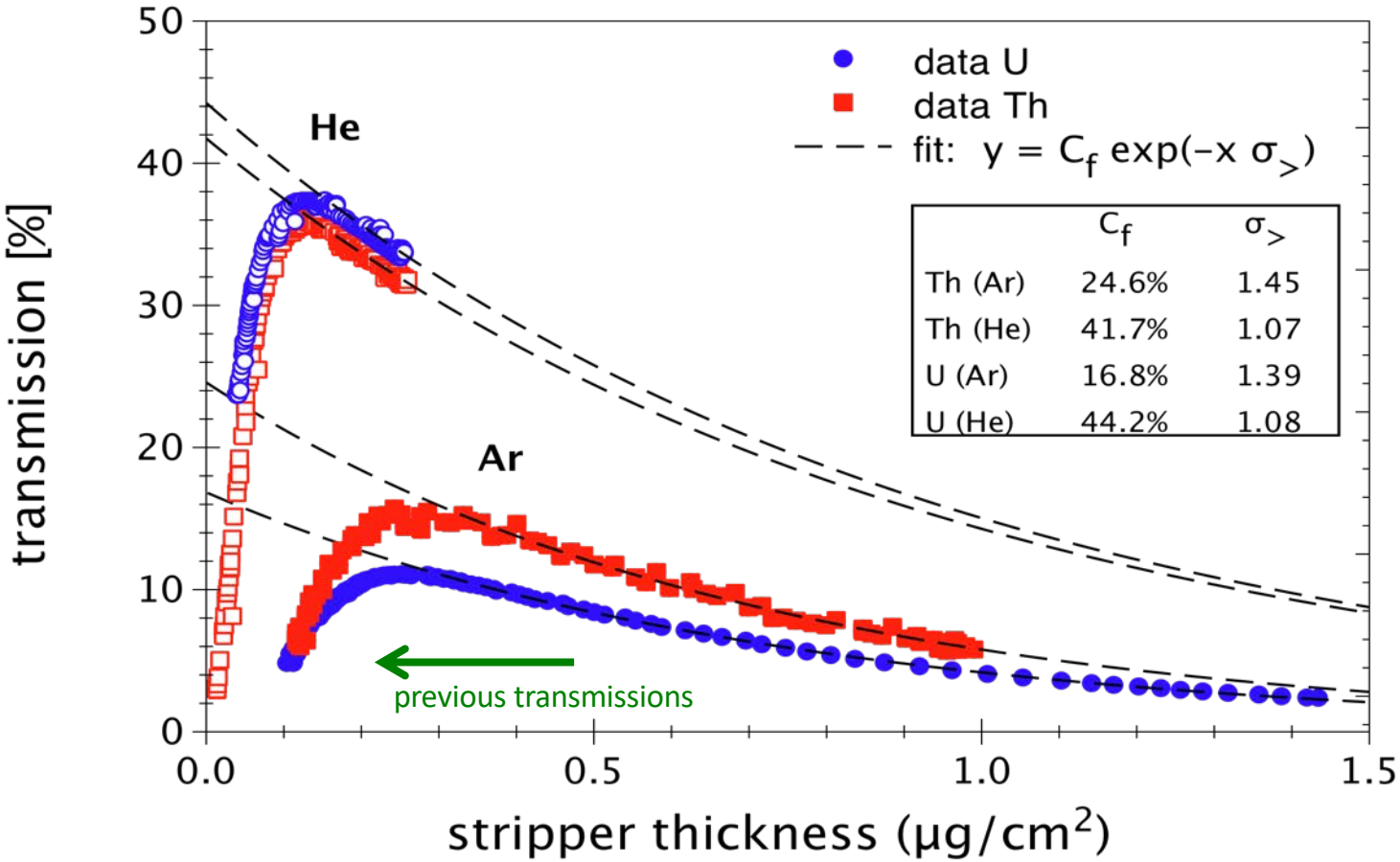
Stripping yield 300 keV

He instead of Ar:

factor 3-4 more > 30% transmission

previous transmissions

at other facilities: ≤ 5% transmission

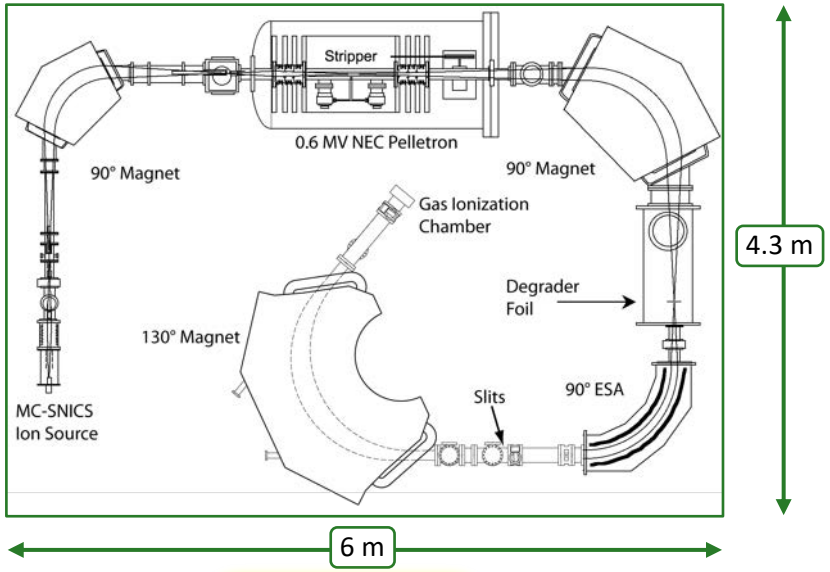


	C_f	$\sigma_>$
Th (Ar)	24.6%	1.45
Th (He)	41.7%	1.07
U (Ar)	16.8%	1.39
U (He)	44.2%	1.08

Universal AMS: ^{10}Be, ^{14}C, ^{26}Al, ^{41}Ca, ^{129}I, and actinides

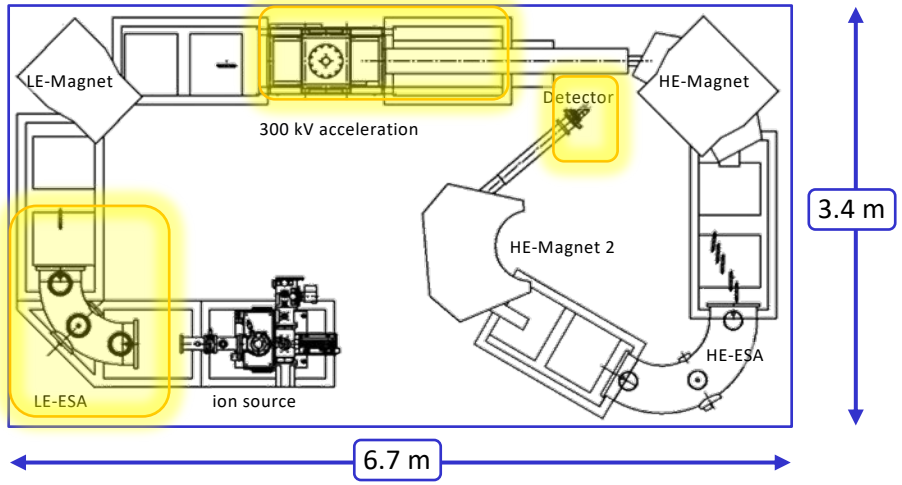
ETH zürich

Compact universal AMS



- ^{10}Be 1+,2+ → effective: 3+,5+
- ^{14}C 1+
- ^{26}Al 2+
- ^{41}Ca 2+ → effective: 5+
- ^{129}I 2+
- Actinides (U, Pu, Pa, Np,..) 3+

New system (preview)



- Additional ESA on injection side
- Compact acceleration & optimized stripper
- Quadrupole lens triplet after acceleration
- Increased angular acceptance on HE side

Physical properties of molecule dissociation

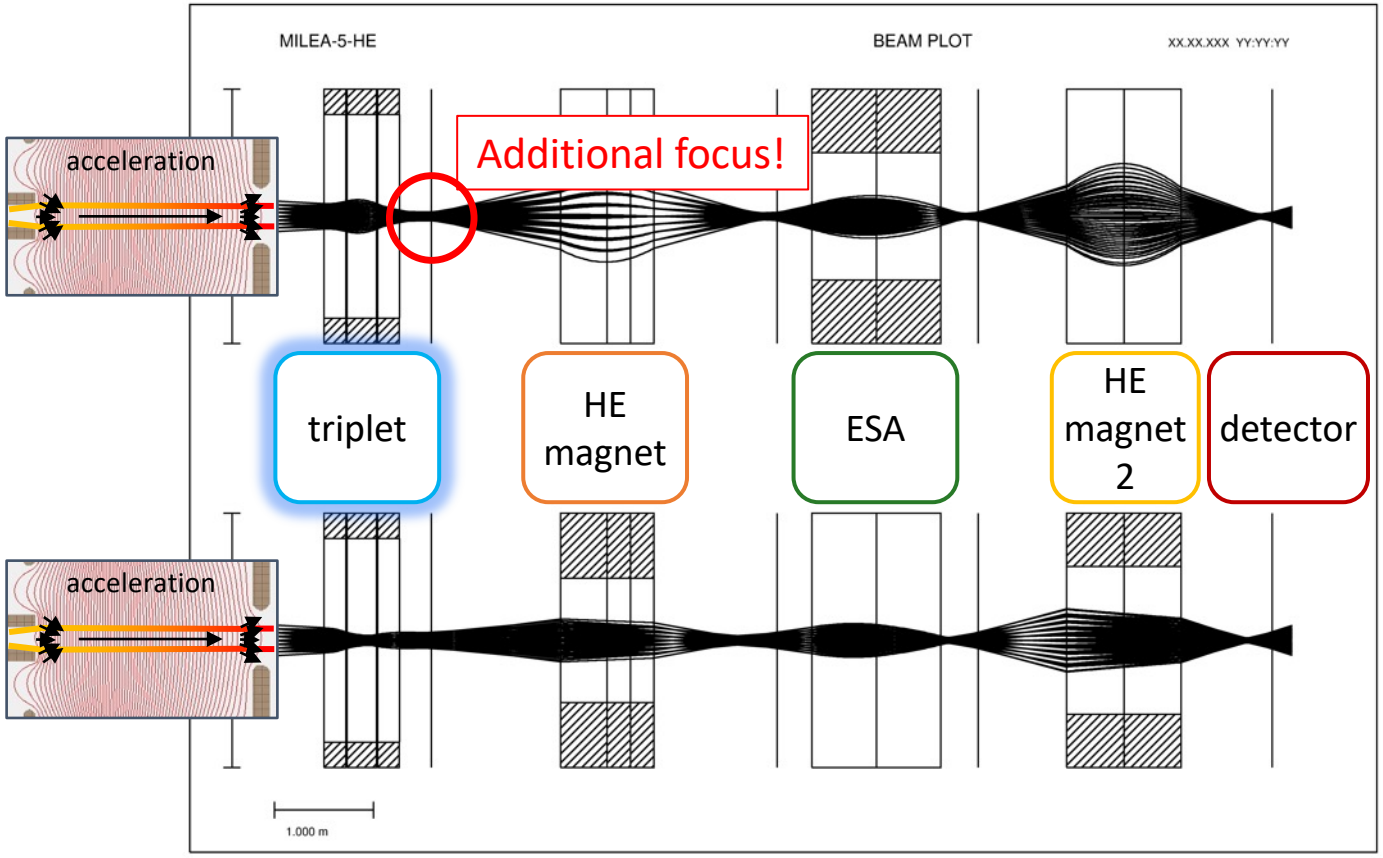
ETH zürich

Because focusing effect of acceleration depends on particle charge state

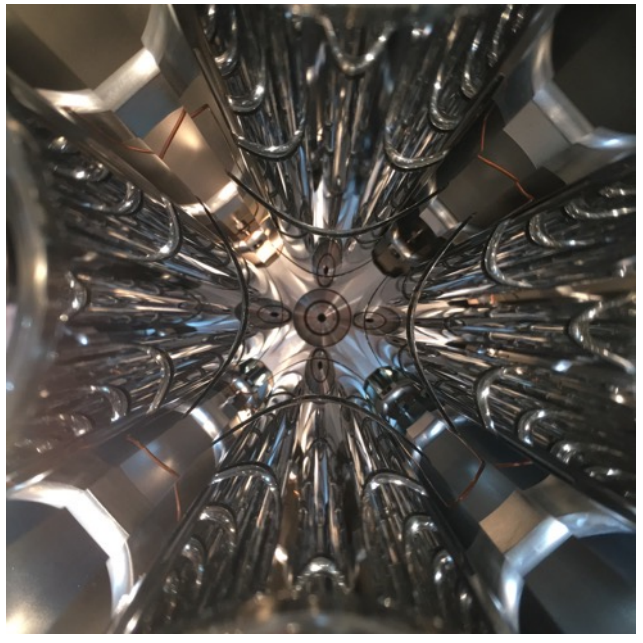
✓ Can be corrected by using a quadrupole lens system

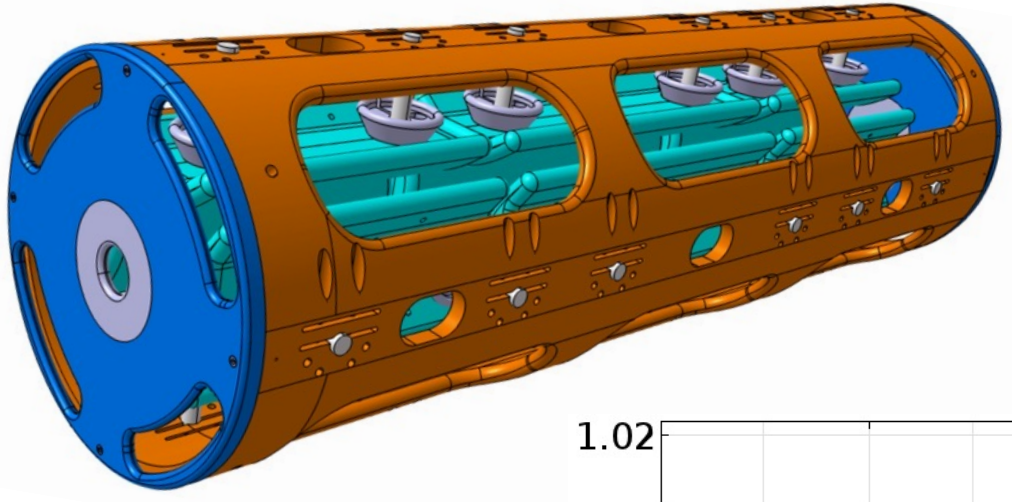
horizontal

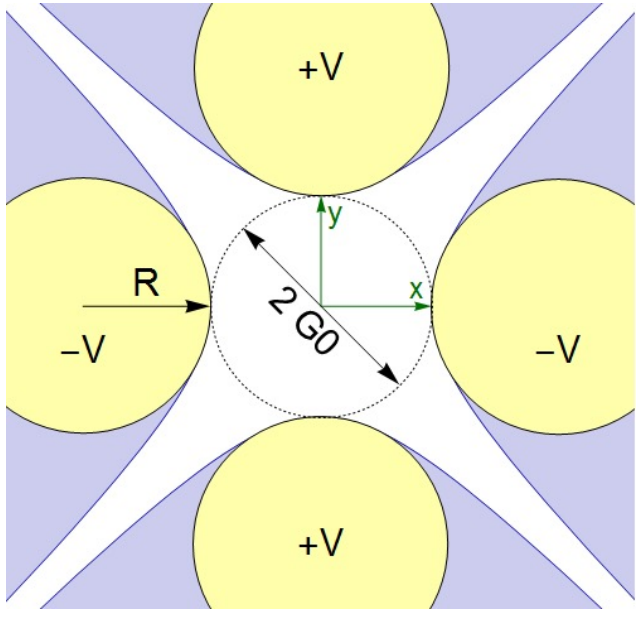
vertical

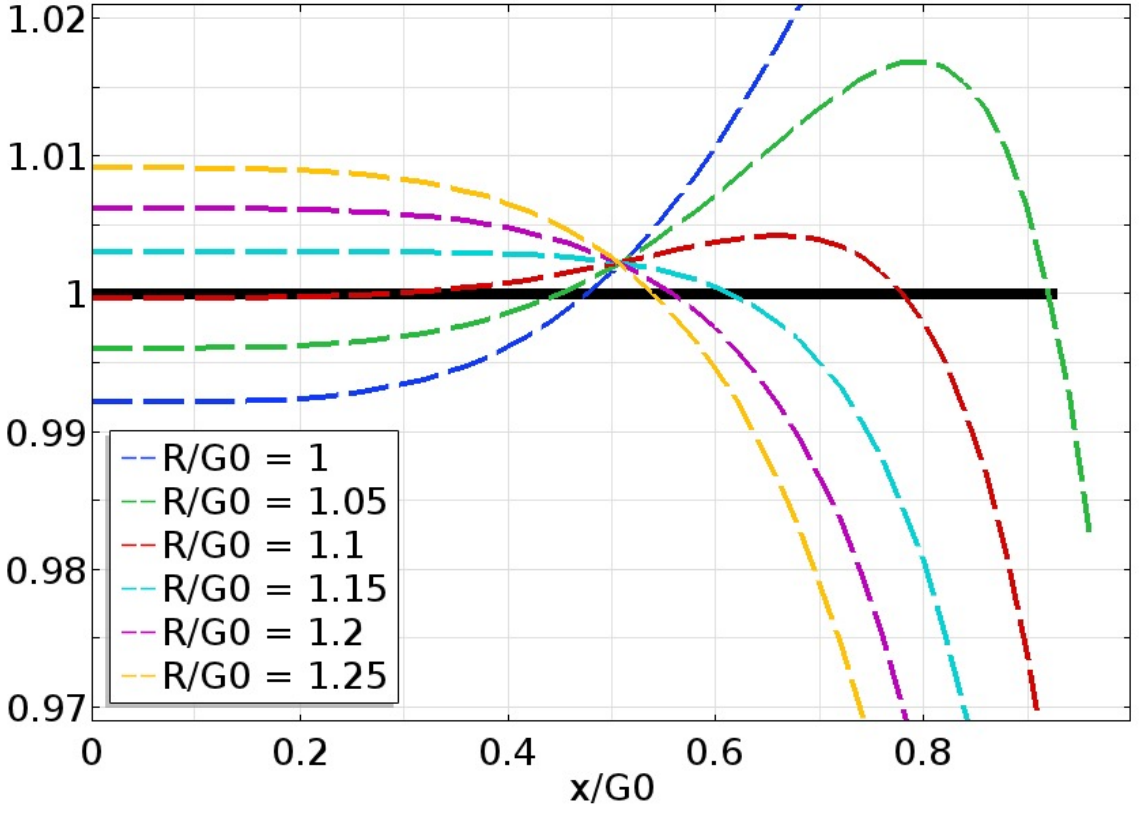


1+,2+,3+,5+



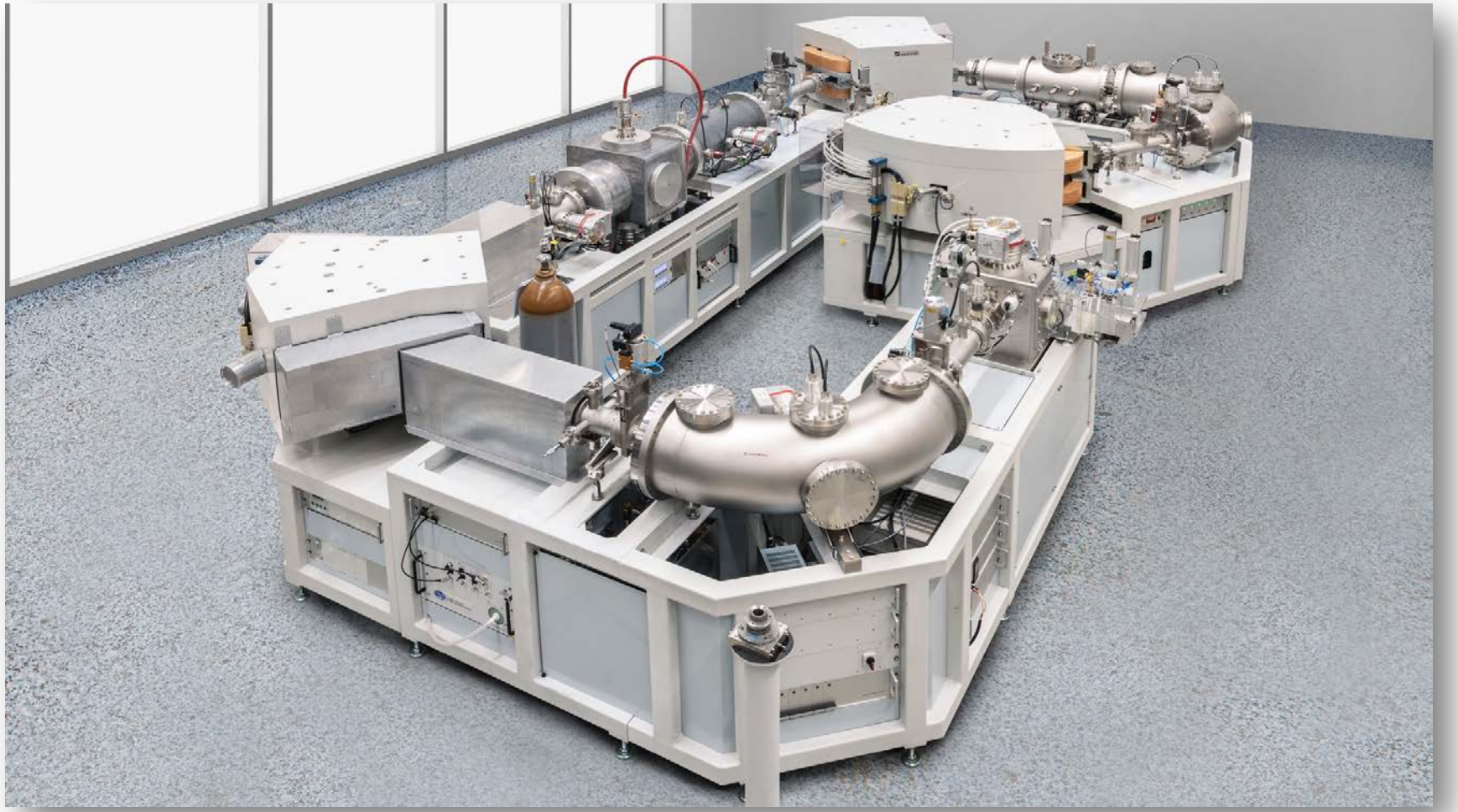



+V
R
y
x
2 G0
–V
–V
+V


1.02
1.01
1
0.99
0.98
0.97
0
0.2
0.4
0.6
0.8
x/G0
R/G0 = 1
R/G0 = 1.05
R/G0 = 1.1
R/G0 = 1.15
R/G0 = 1.2
R/G0 = 1.25

Multi Isotope Low Energy AMS system (MILEA)

ETH zürich



A versatile AMS system for:

^{10}Be, ^{14}C,

^{26}Al, ^{41}Ca,

^{129}I, ^{236}U,

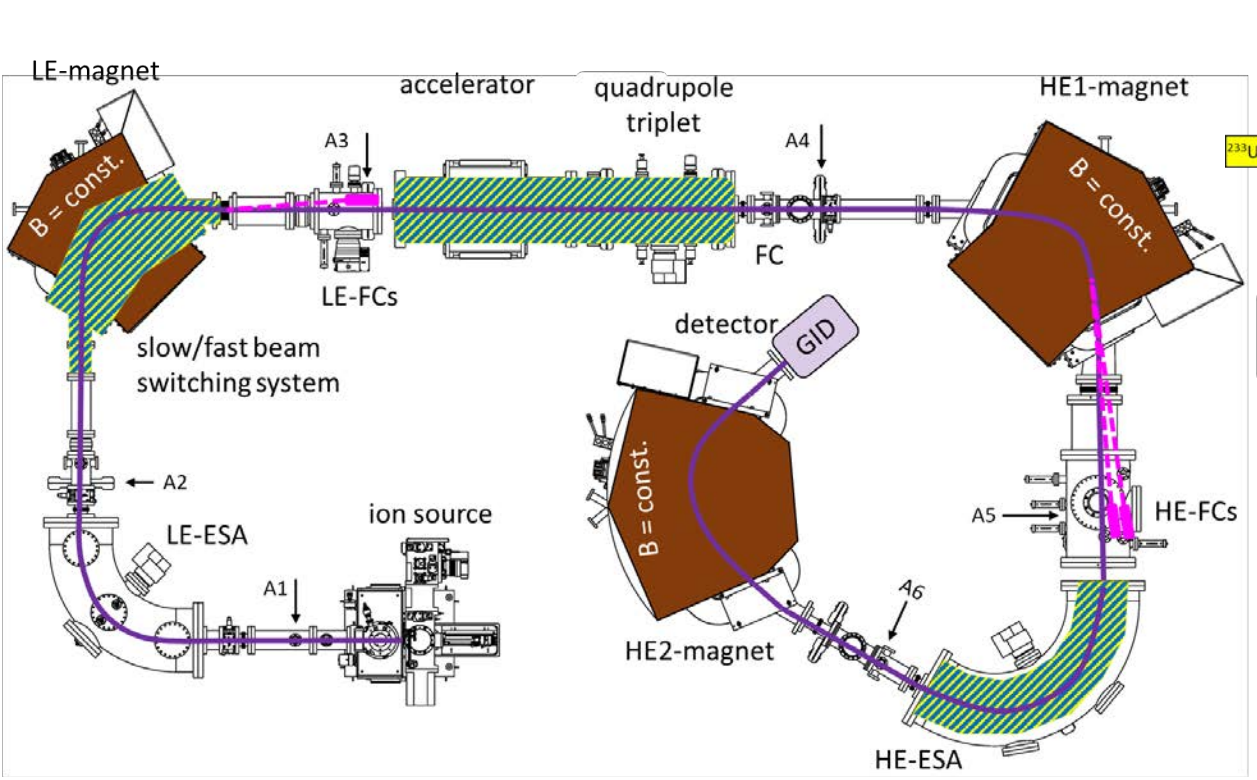
$^{239,240,242,244}Pu$,

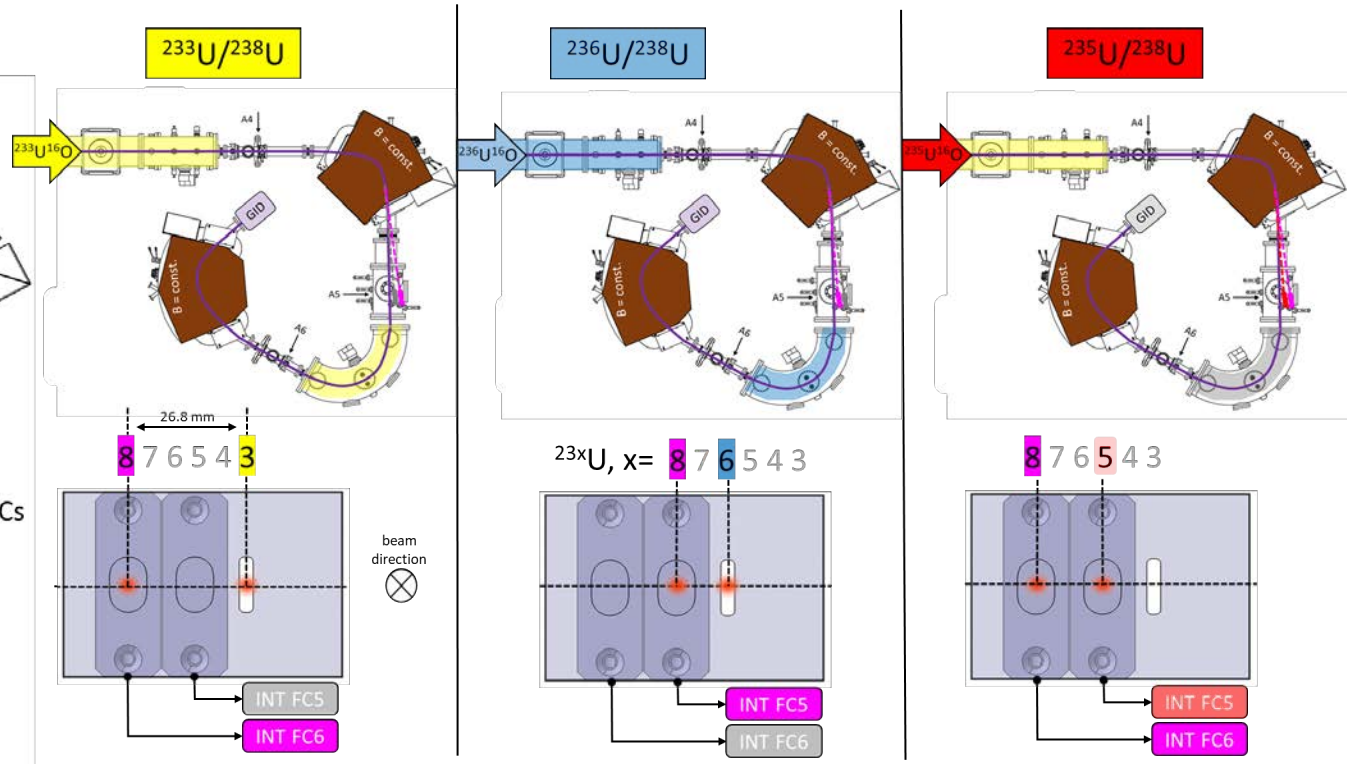
and other actinides

Ionplus

engineering scientific instruments

Actinide measurement sequence: uranium





high total detection efficiency
$^{236}U/^{238}U$ background
multiple isotope ratio measurement capability:

of up to 1% for U and Pu
in the 10^{-14} range
$^{233}U/^{238}U$; $^{236}U/^{238}U$; $^{235}U/^{238}U$

Dissemination of LIP technologies

ETH zürich

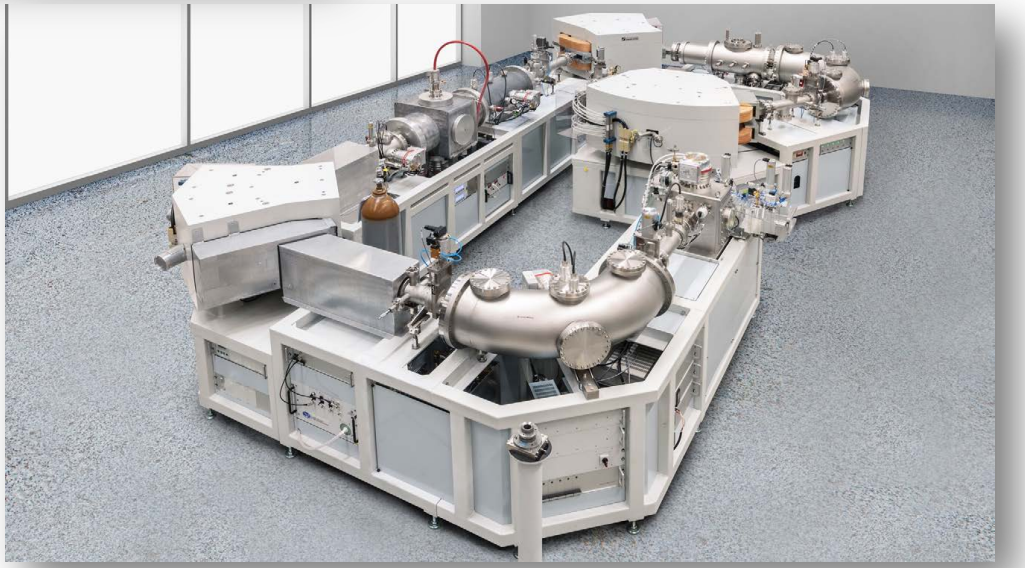
Founded: February 2013

commercializing LIP
technical developments

Today: World market leader for
compact AMS systems
25 Employees
Annual turnover: >10 MioCHF

MICDAS AMS system

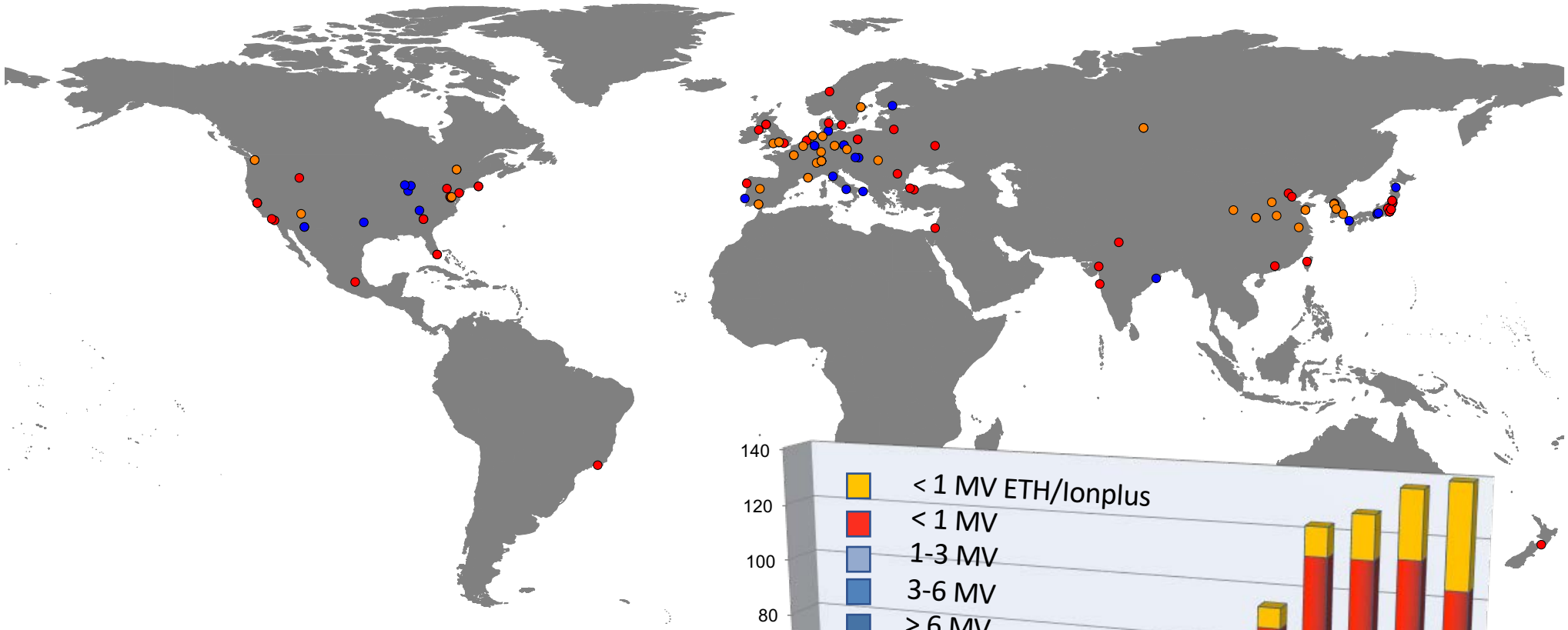
MILEA AMS system

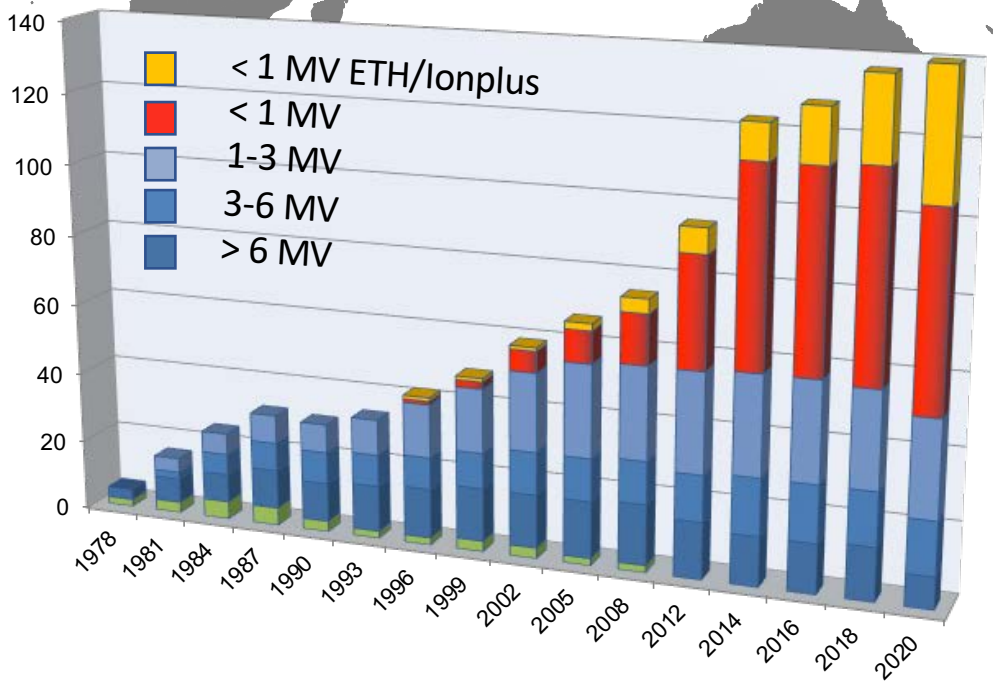


spinoff ETH zürich

Accelerator Mass Spectrometry worldwide

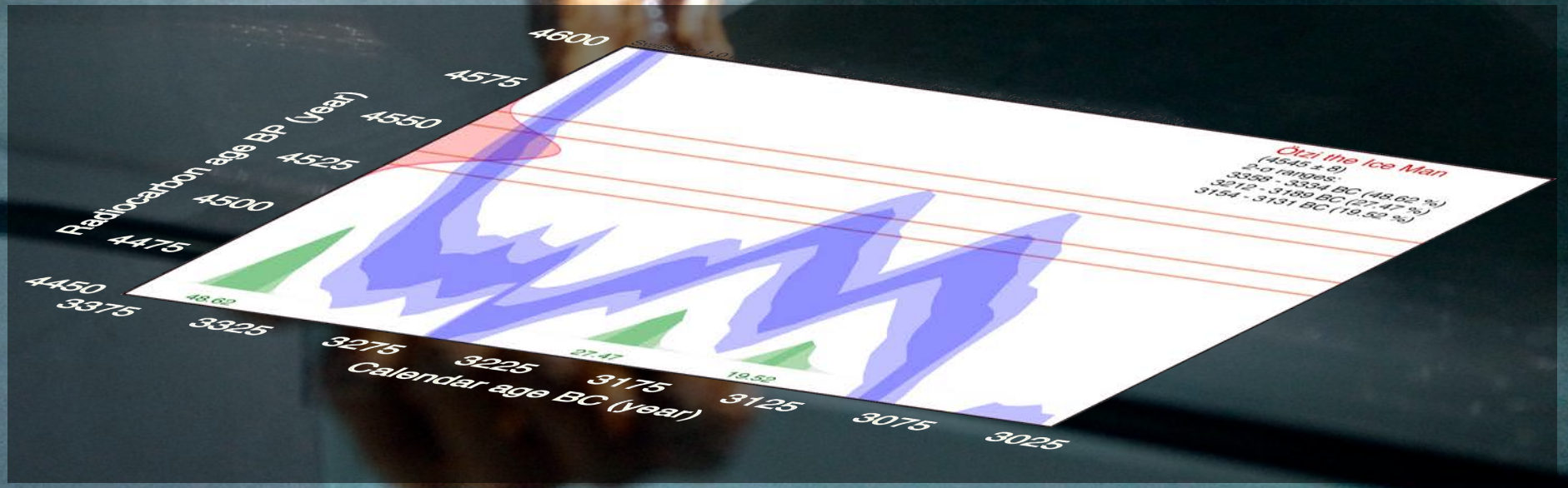
ETH zürich



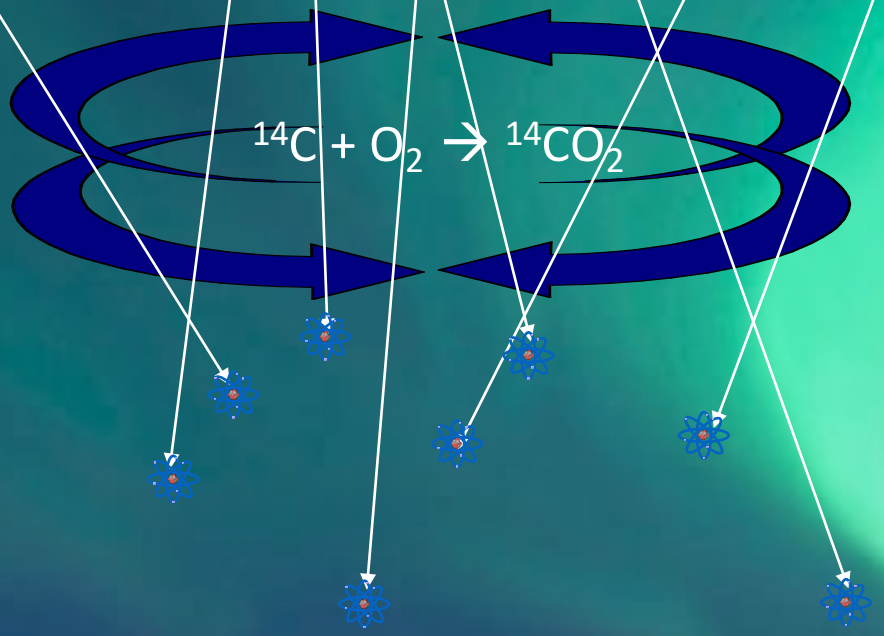


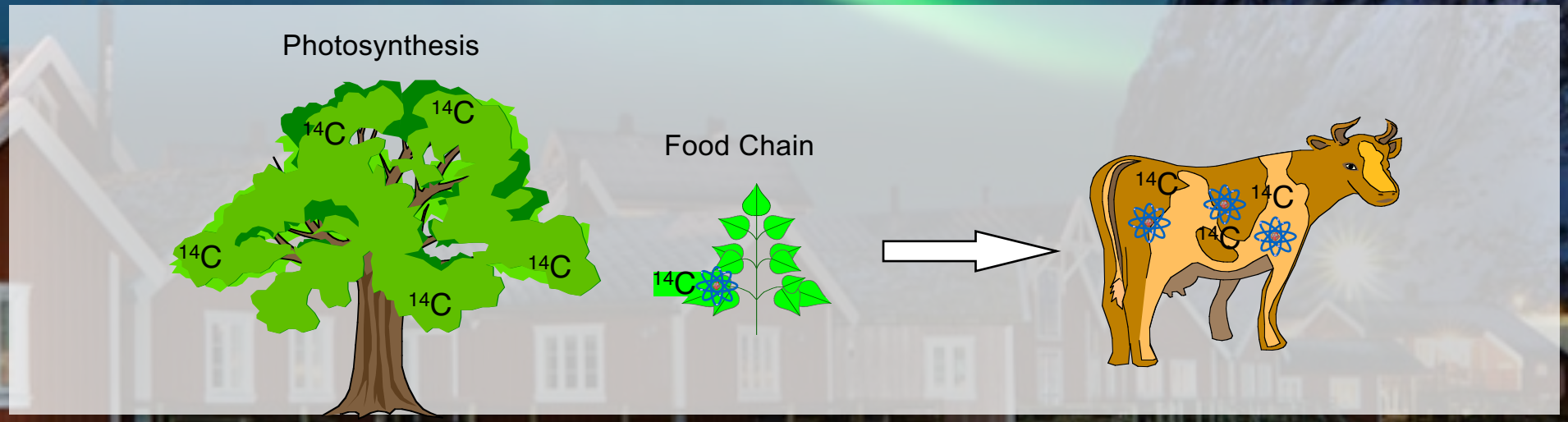
- Traditional AMS systems (3+ and higher) (50)
- ETH technology-based systems (1+ / 2+) (55)
- Systems build by LIP or Ionplus (35)

Dating of the Öztal Ice Man

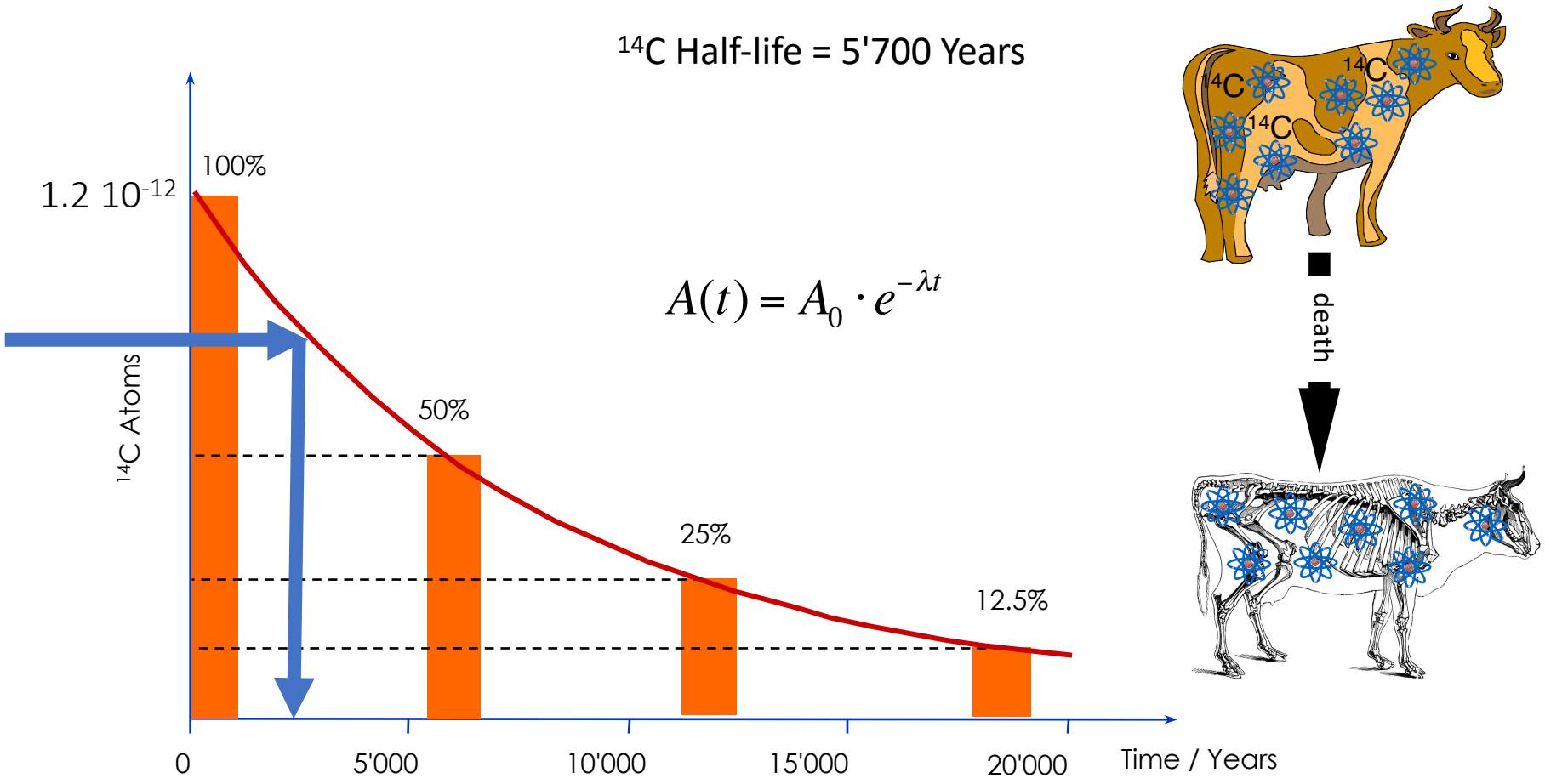


^{14}C production and CO_2 Cycle



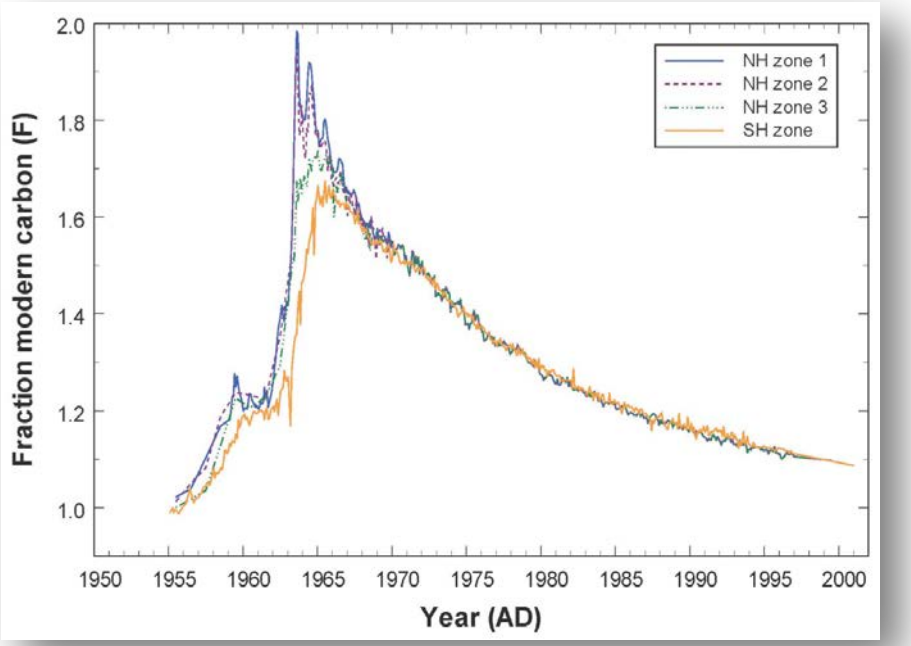


Radiocarbon age



Precondition for dating

- ^{14}C Production variable with time?
  Intensity variation of cosmic radiation
  - modulation of Earth magnetic field
  - Solar activity
- variation of CO_2 concentration in atmosphere!
  - Release of fossil CO_2 (Suess-Effect)
  - Variation of CO_2 exchange from different reservoirs (Climate)
- No additional ^{14}C-sources!
  - Atomic bomb testing
  - Nuclear activities

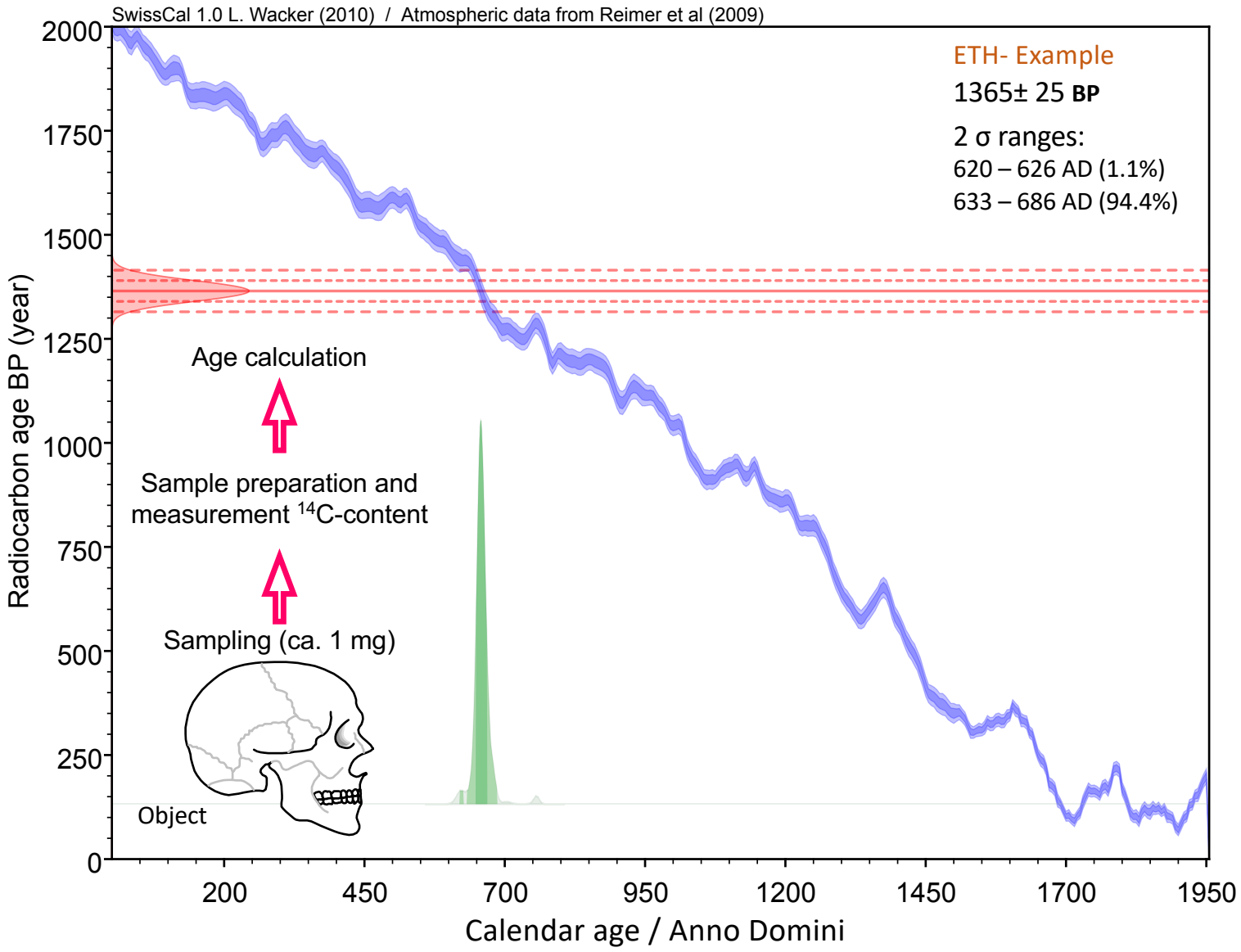


Tree ring chronologies

- Same growing pattern can be matched.
- Time range can be extended over the lifetime of one tree.

Making an absolute time scale by counting individual tree-rings

Radiocarbon Dating Procedure



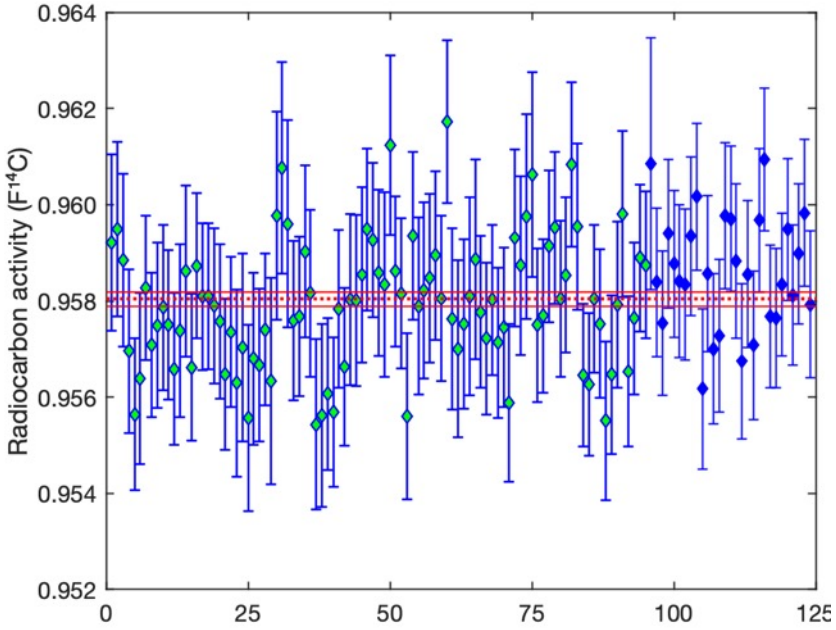
Refining radiocarbon calibration data

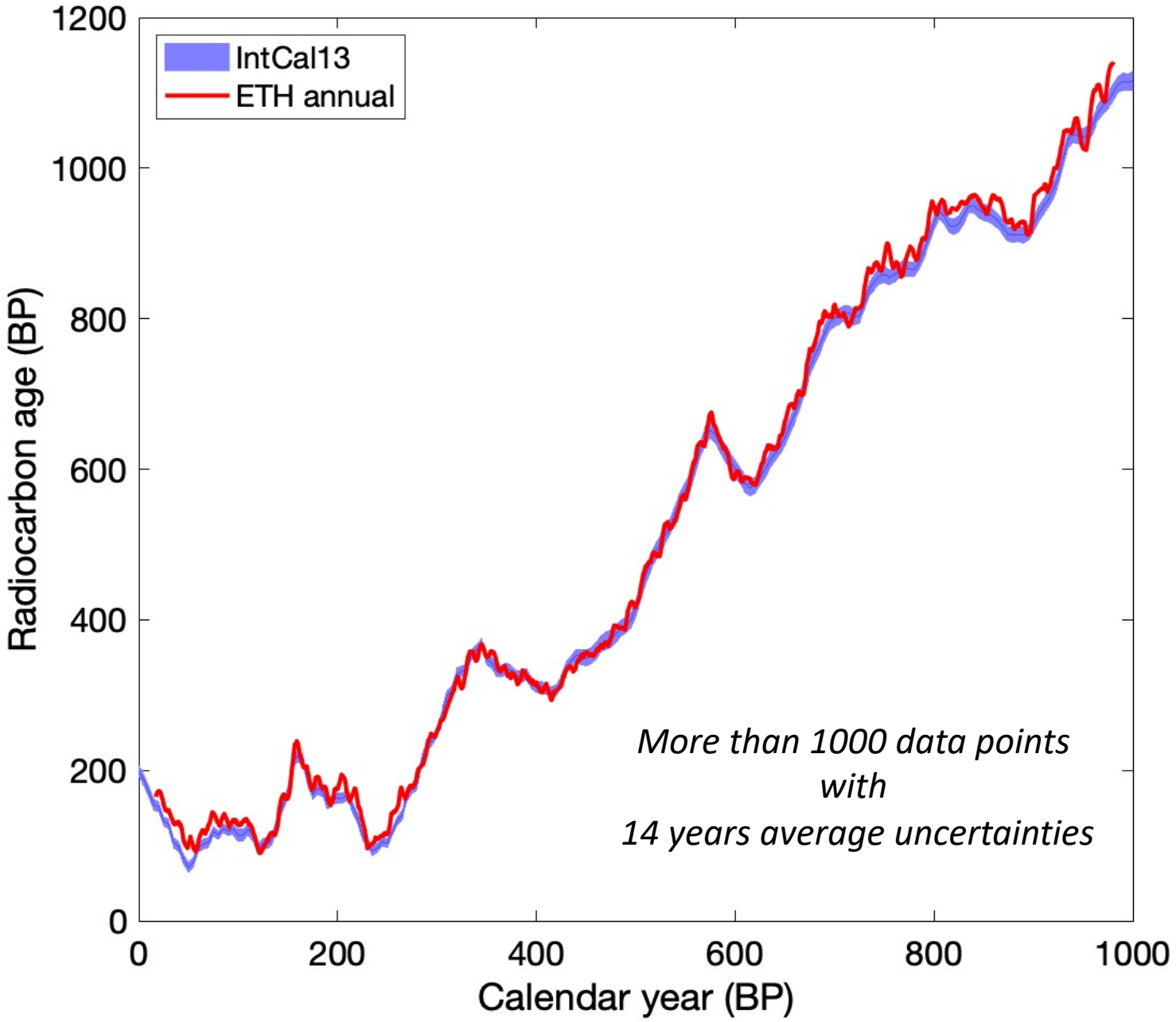
ETH zürich

Results of reference material: 1515AD

Mean Radiocarbon Age:
344.3 ± 1.3 BP

sample scatter: 12 years
slightly less than
average uncertainty: 14 years

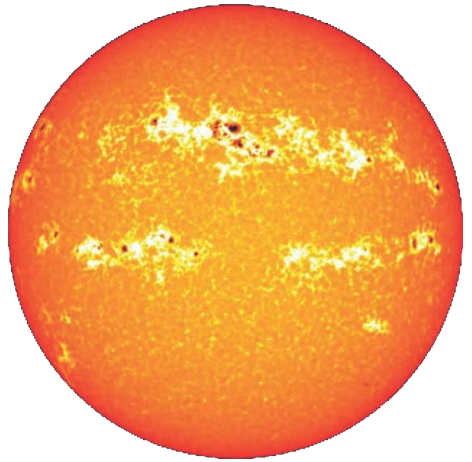


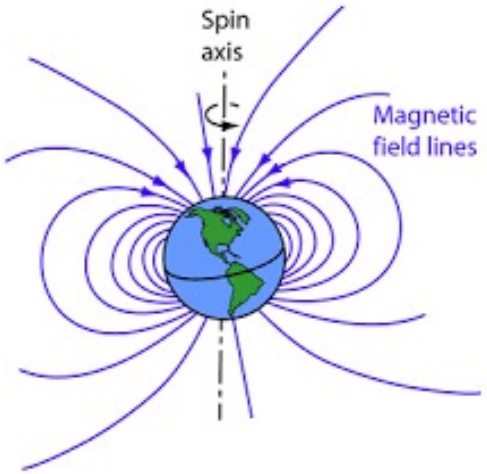


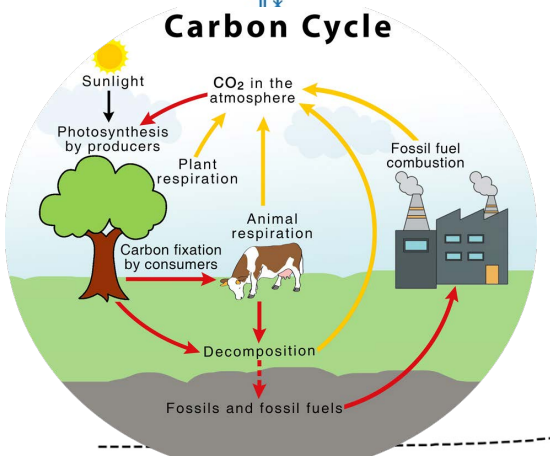
L. Wacker, ISBN 978-3-906897-30-1 (2019) 29

ETH zürich

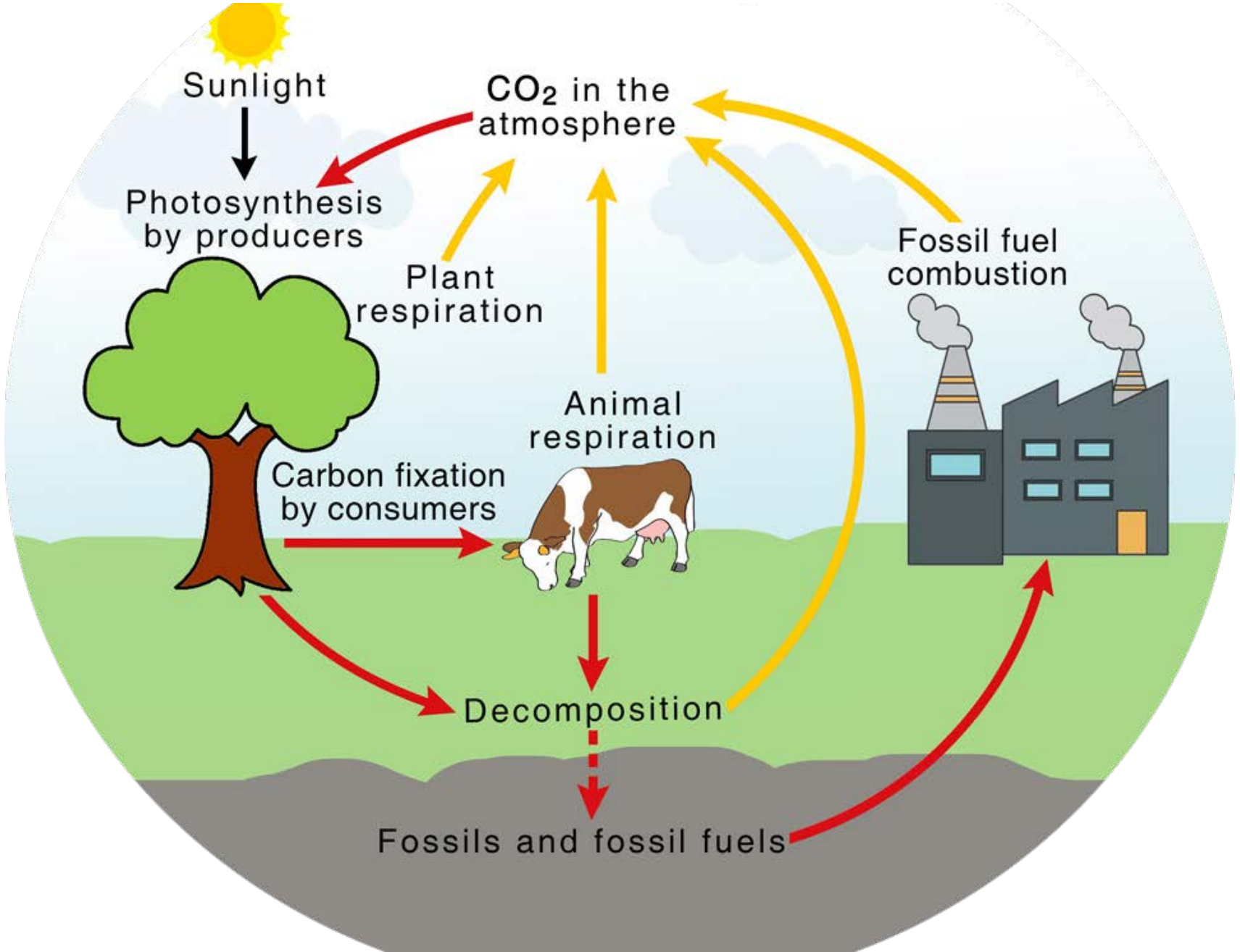
Solar activity



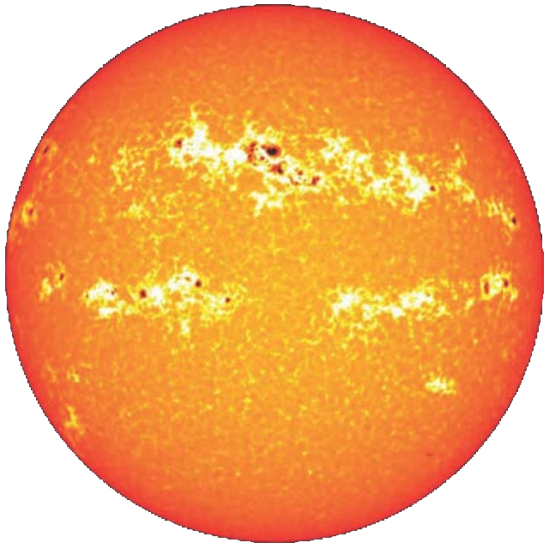


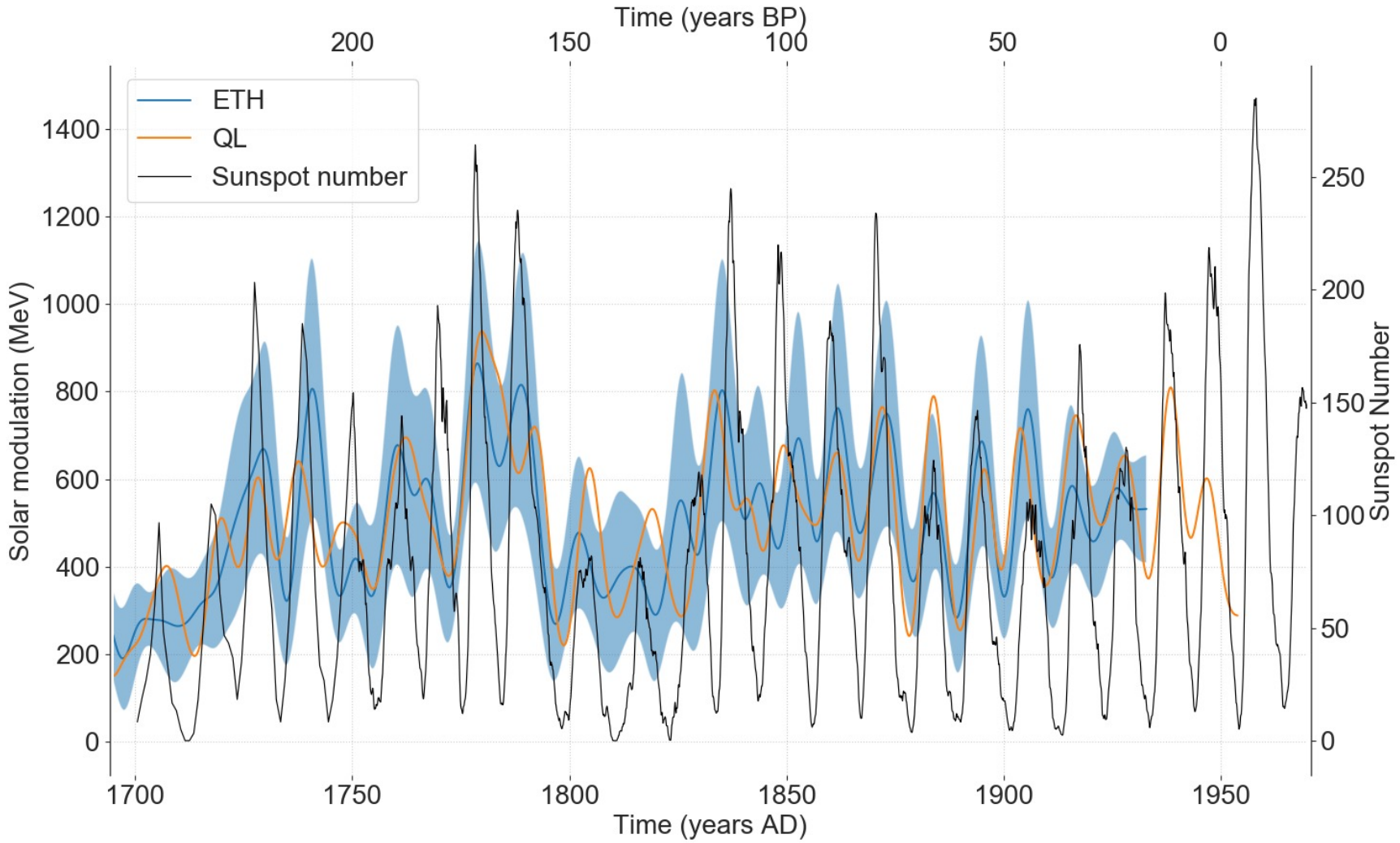


Box model: Global carbon cycle



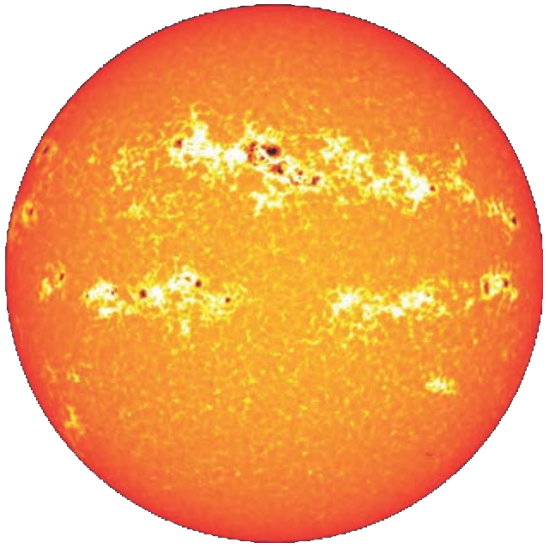
Comparison: direct observation - reconstruction

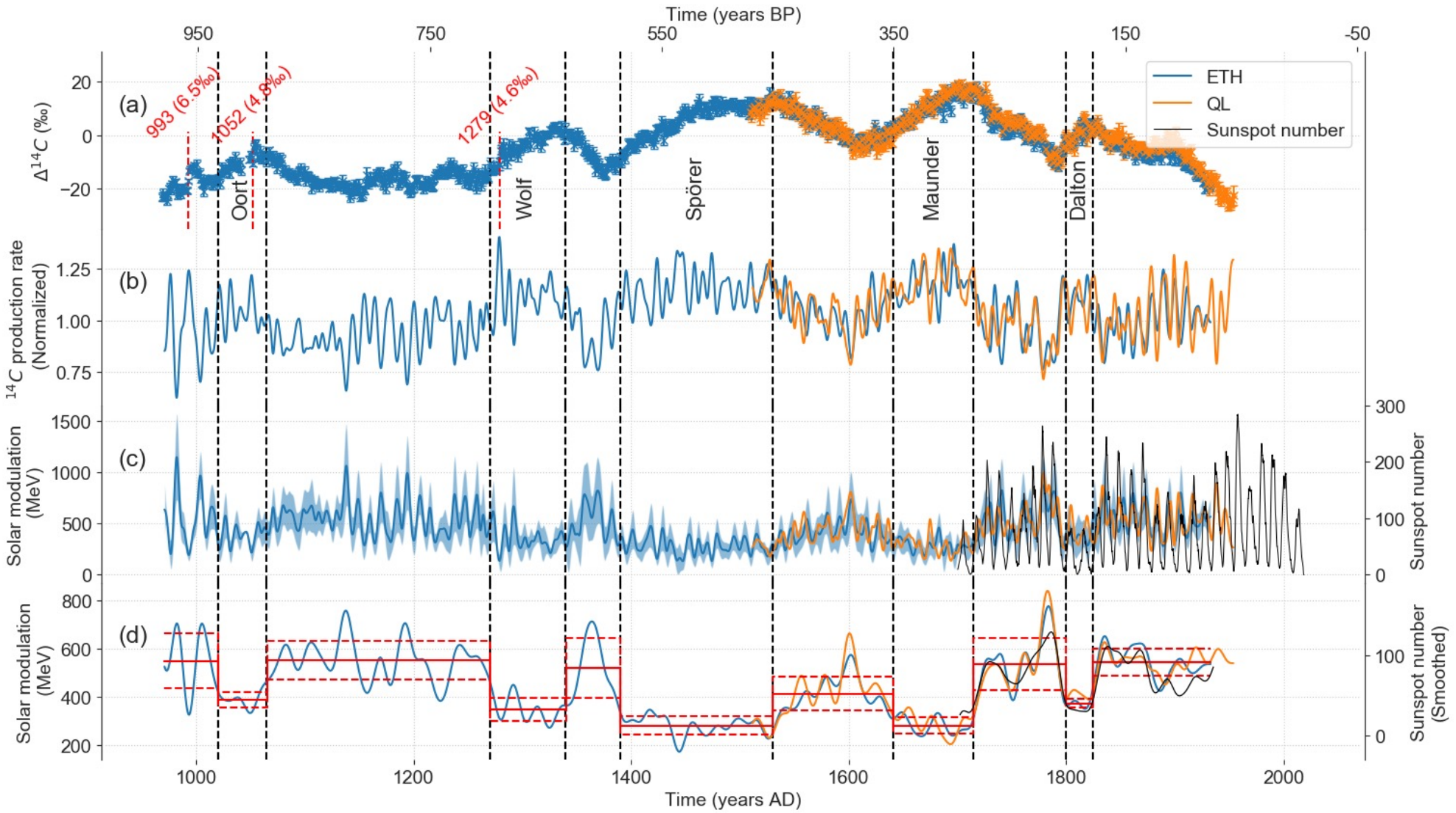




A new sunspot record over 1000 years!

ETH zürich





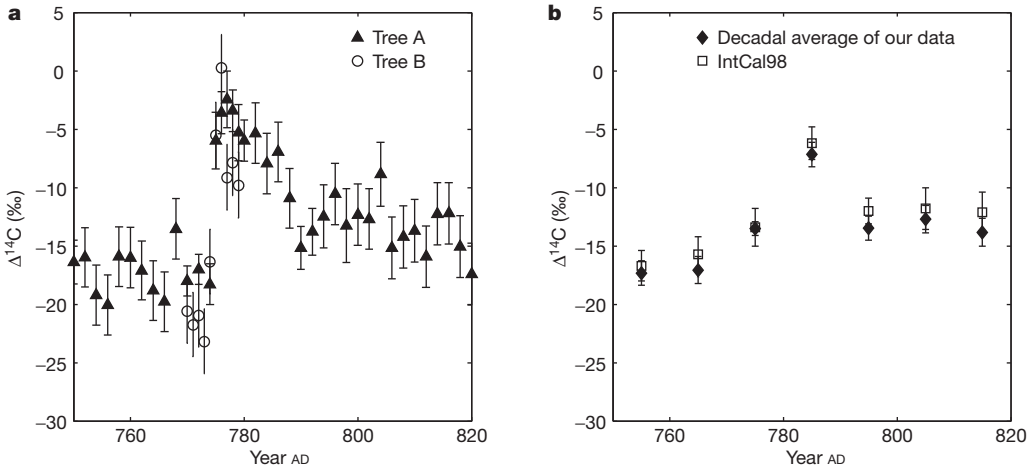
Nature Geoscience **volume 14**, pages 10–15 (2021)

Annually resolved tree ring data

ETH zürich

A signature of cosmic-ray increase in AD 774–775 from tree rings in Japan

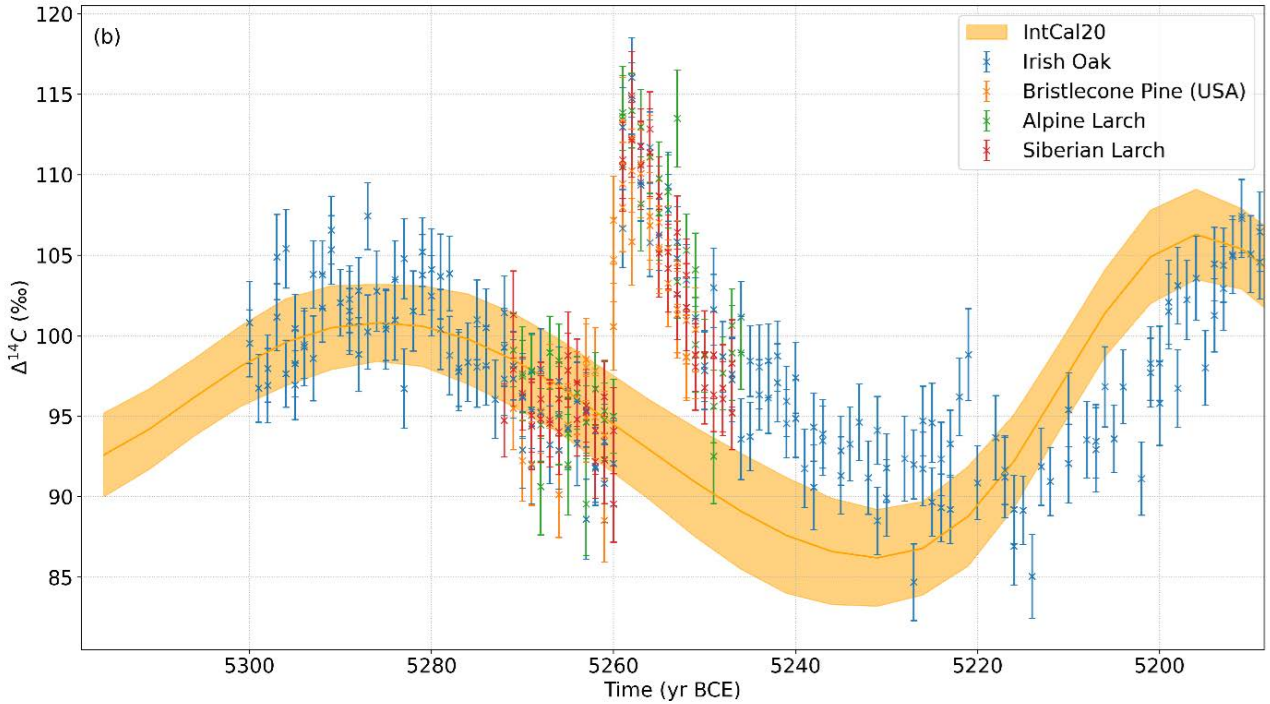
Fusa Miyake[1], Kentaro Nagaya[1], Kimiaki Masuda[1] & Toshio Nakamura[2]

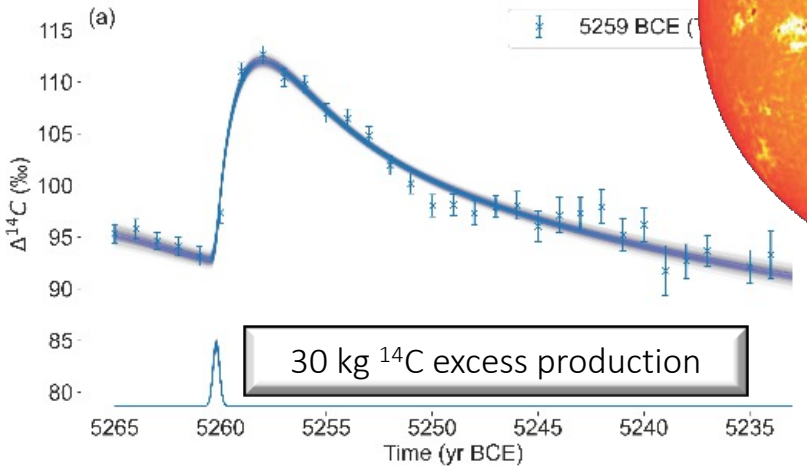


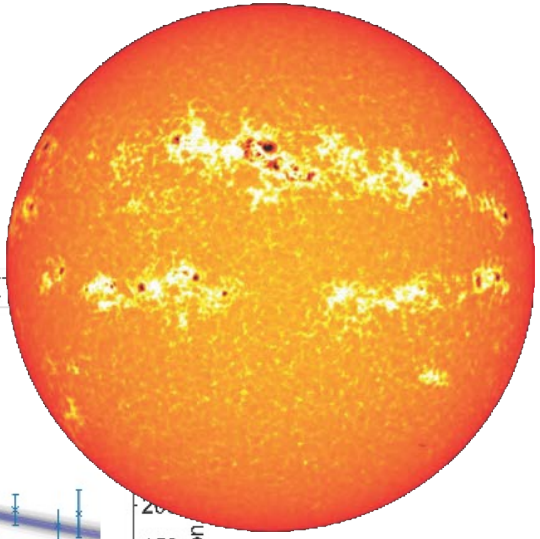
14 JUNE 2012 | VOL 486 | NATURE | 241

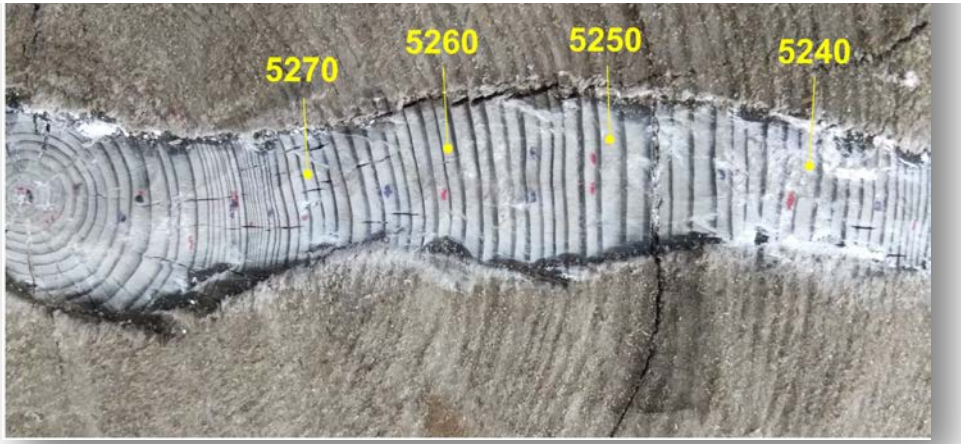
strong solar proton events

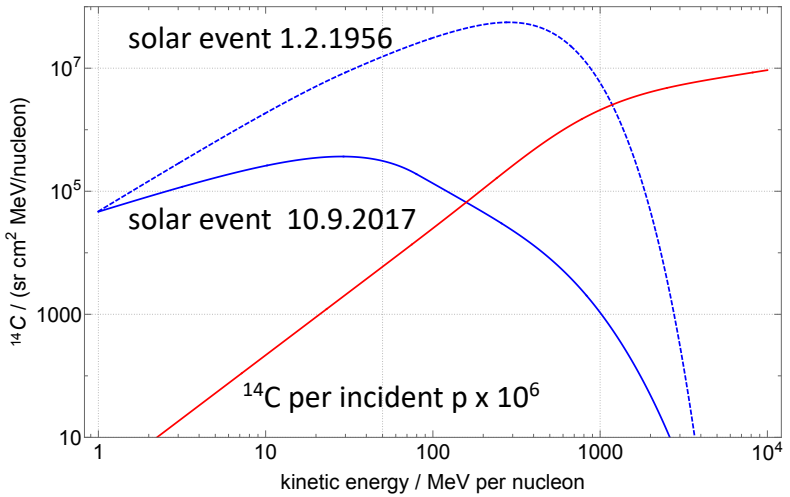
ETH zürich











Cosmic ^{14}C
global production
≈ 0.24 mg/s
≈ 7.47 kg/a

Nicolas Brehm

Natureportfolio DOI:10.21203/rs.3.rs-753272/v1

Switzerland (2021)

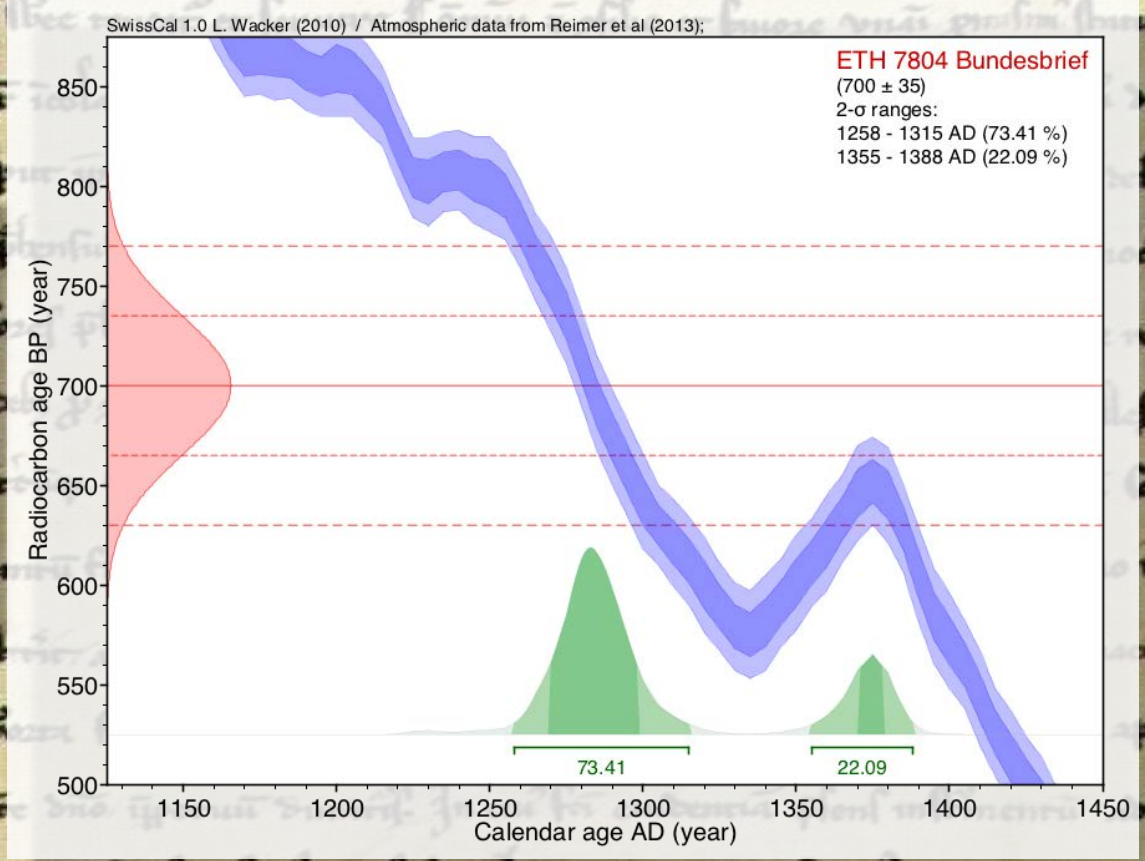
Switzerland (1291)

ETH zürich

Results of the first measurements

700rd year Swiss anniversary (1991)

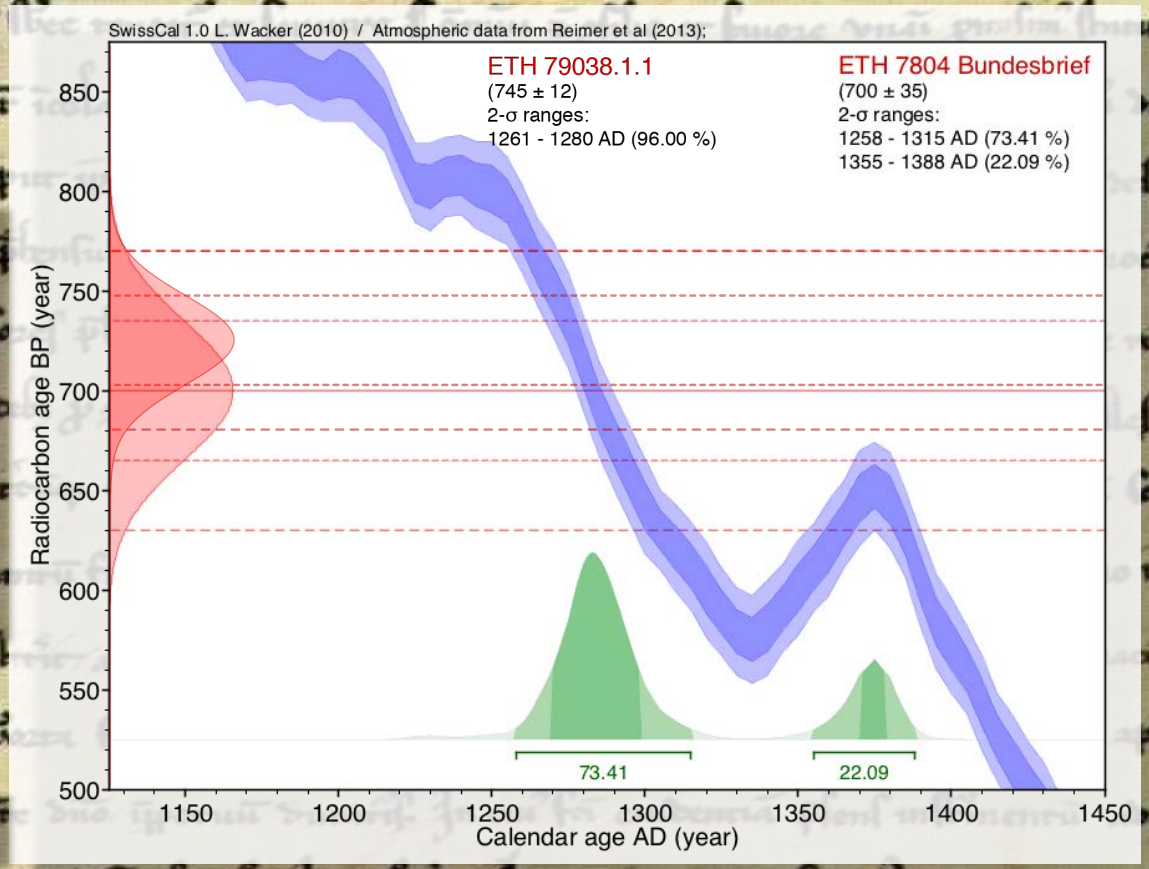
Lab. no.:	sample label	^{14}C age (yrs BP)
ETH 7804-1	BB-1	706±47
ETH 7804-2	BB-2	683±45
ETH 7804-3	BB-3	714±45
ETH 7804	mean	700±35



Re-evaluation of residual material

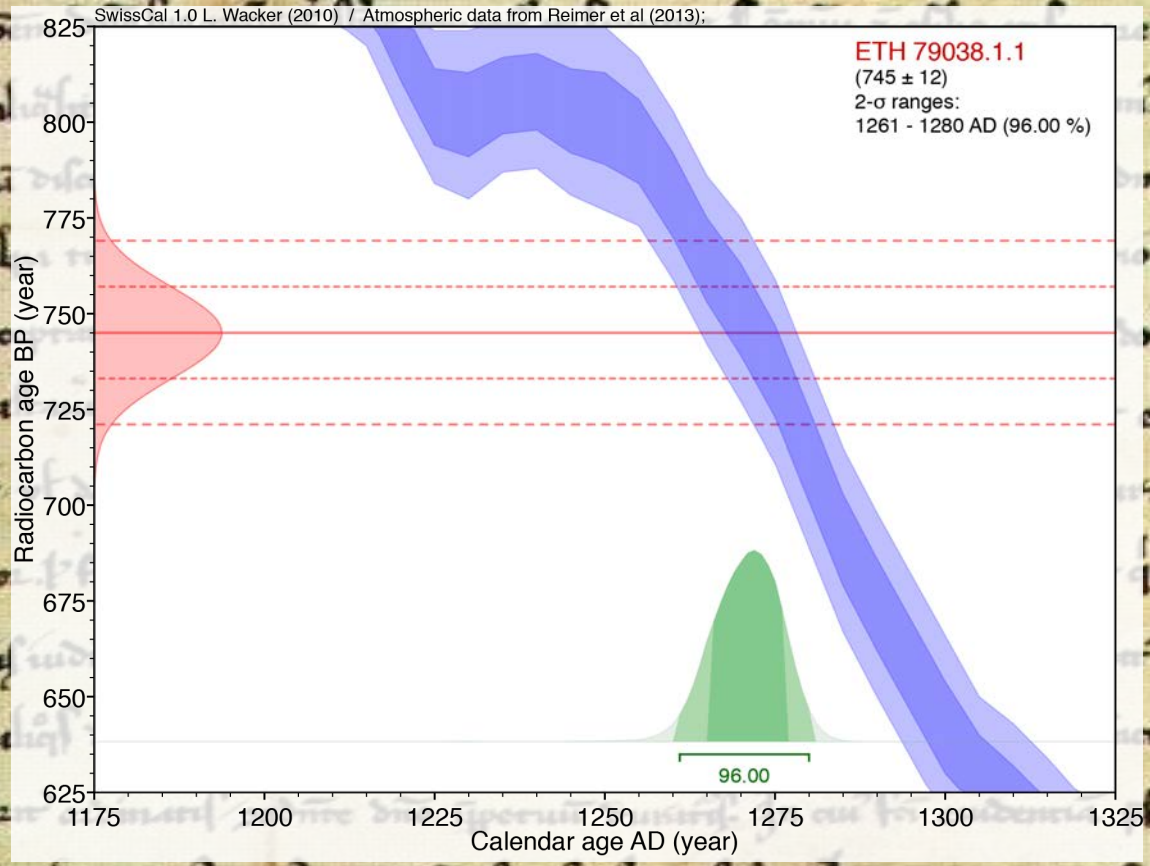
2017 high-precision measurement

Lab. no.:	sample label	^{14}C age (yrs BP)
ETH 7804-1	BB-1	706±47
ETH 7804-2	BB-2	683±45
ETH 7804-3	BB-3	714±45
ETH 7804	mean	700±35
ETH 79038.1.1		745±12



Re-evaluation of residual material

2017 high-precision measurement

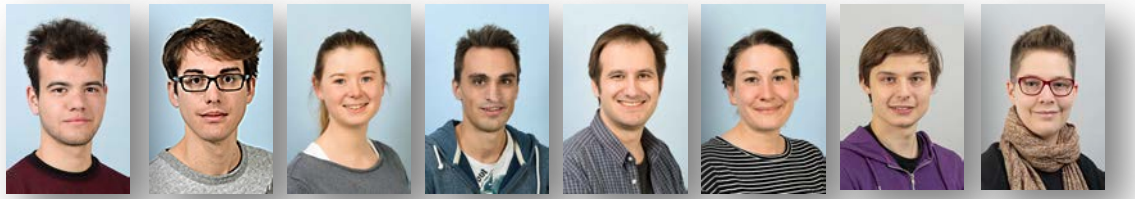


People at LIP

ETH zürich

Senior scientific staff (8)

Ph.D. Students (8)

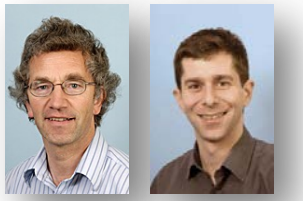


Scientific staff (7)

Administration (1)

Chemistry laboratory technicians (4)

Electronics & IT (2)



Mechanics & engineering (5)

35 people (15 female / 20 male)