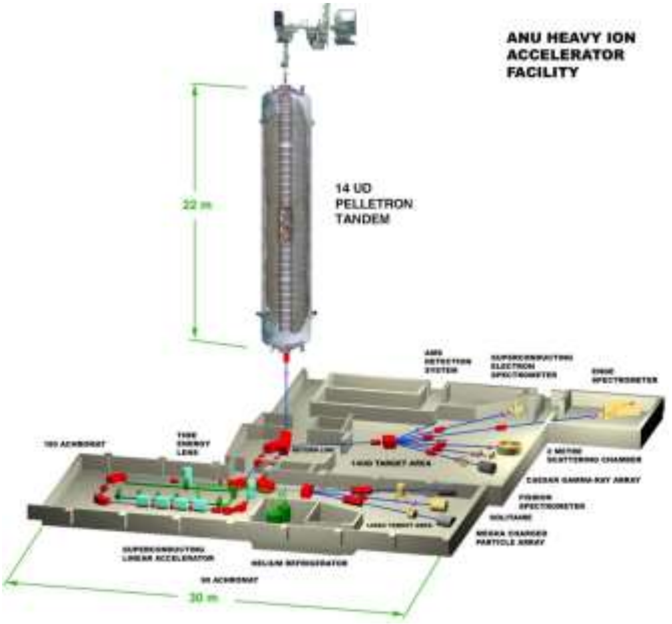
Single atom counting of live interstellar radionuclides in natural archives

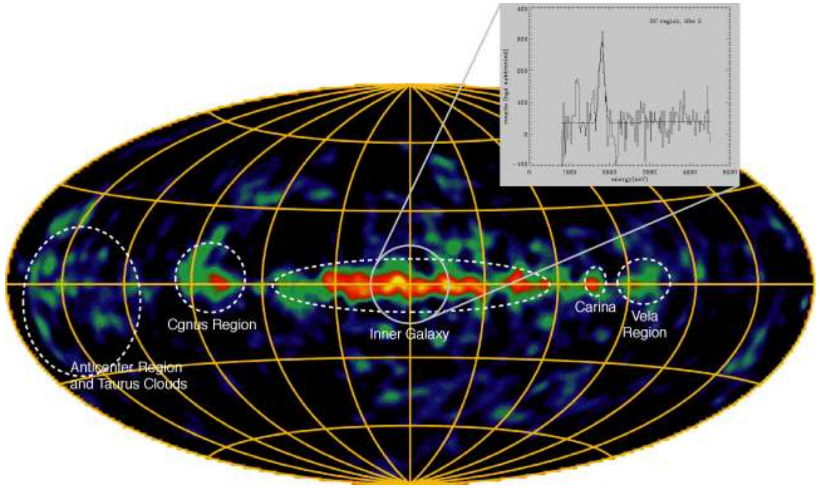
Johannes Lachner

Accelerator Mass Spectrometry & Isotope Research

Helmholtz-Zentrum Dresden-Rossendorf

www.hzdr.de/ams





Live radioactivities – fingerprints of ongoing nucleosynthesis from satellite observations

Superposition of the two ^{60}Fe emission lines (1173 keV & 1333 keV) recorded on board INTEGRAL

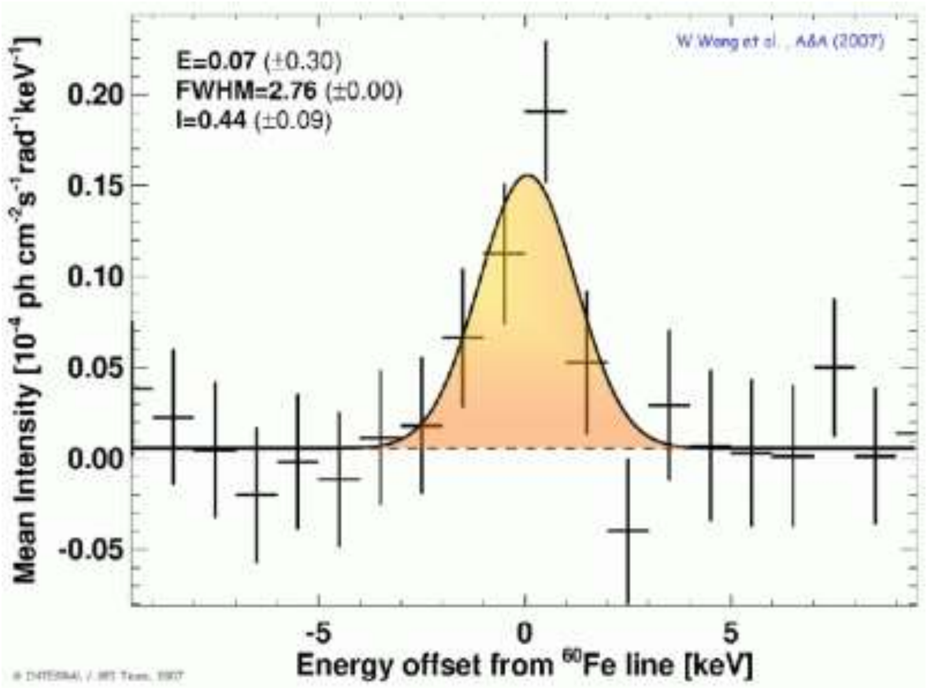
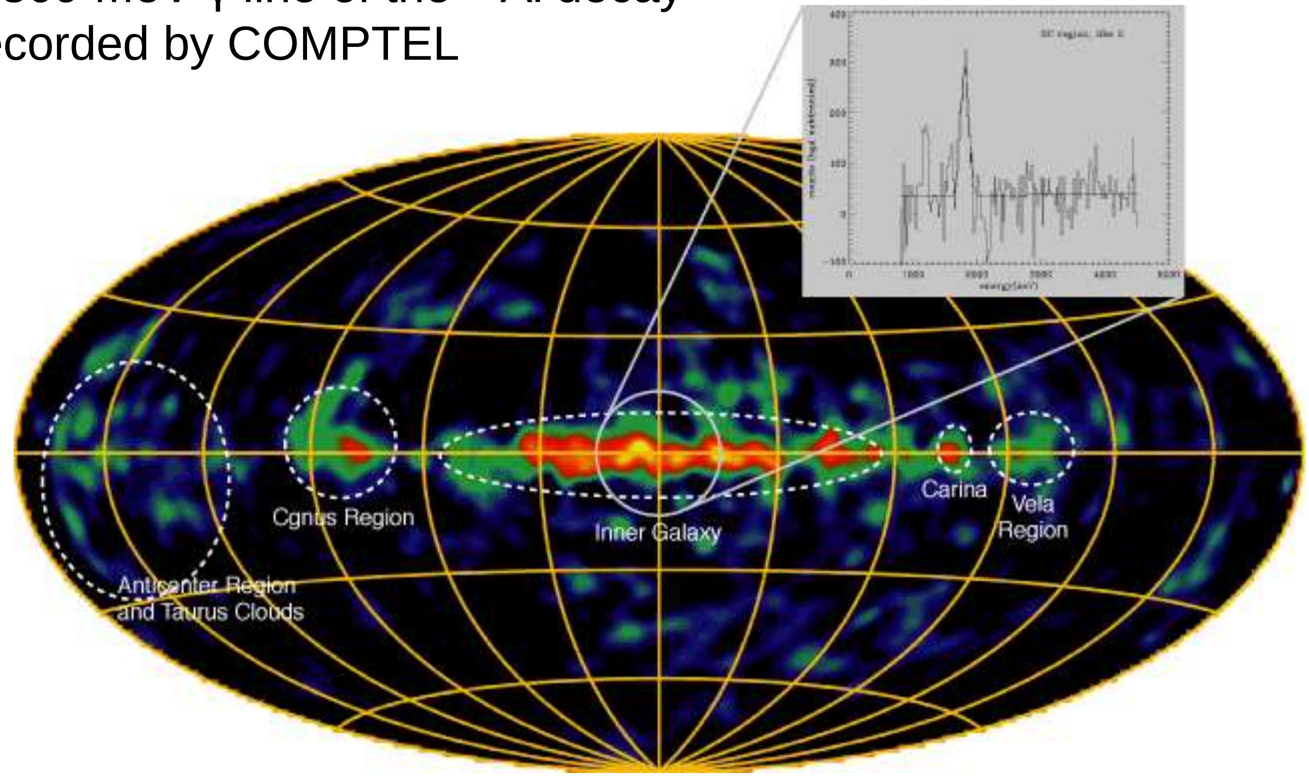
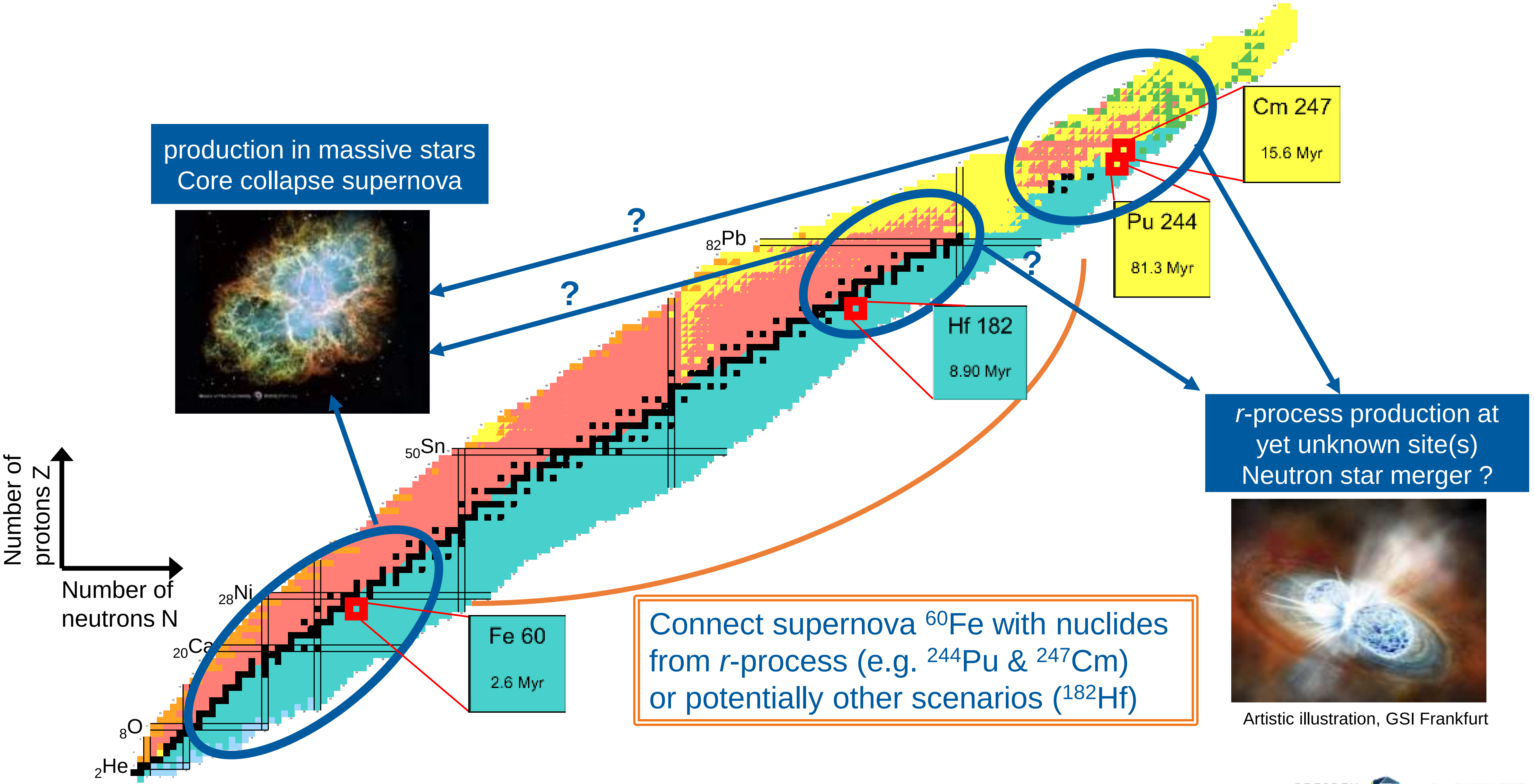


image of the galaxy in intensity of the 1.809 MeV γ line of the ^{26}Al decay recorded by COMPTEL



DRESDEN concept HZDR

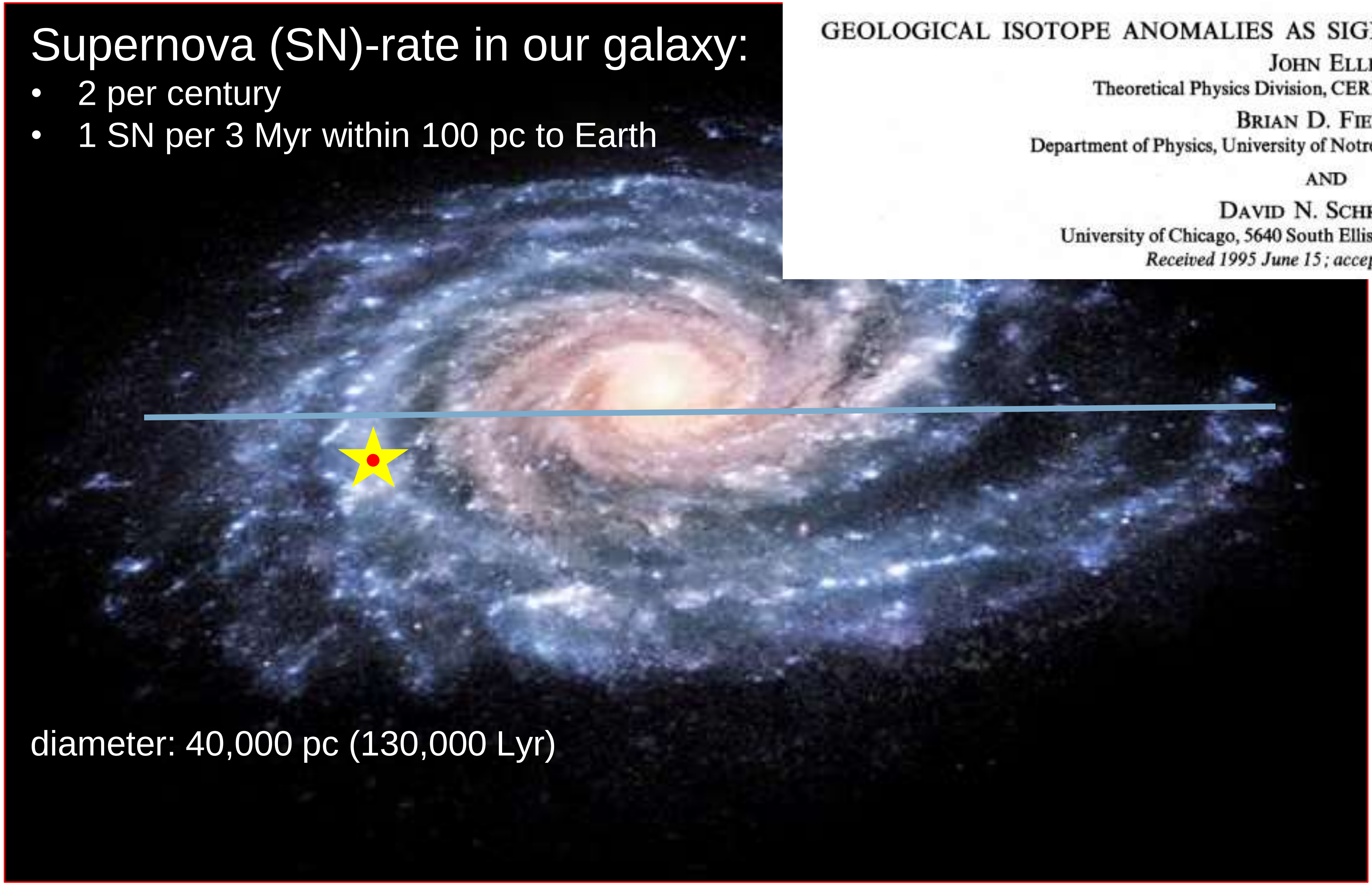
Interstellar radionuclides constrain site of the *r*-process



Artistic illustration, GSI Frankfurt

Long-lived radionuclides „survive“ long-range transport

Suggested radionuclide candidates with Myr lifetimes



GEOLOGICAL ISOTOPE ANOMALIES AS SIGNATURES OF NEARBY SUPERNOVAE

JOHN ELLIS
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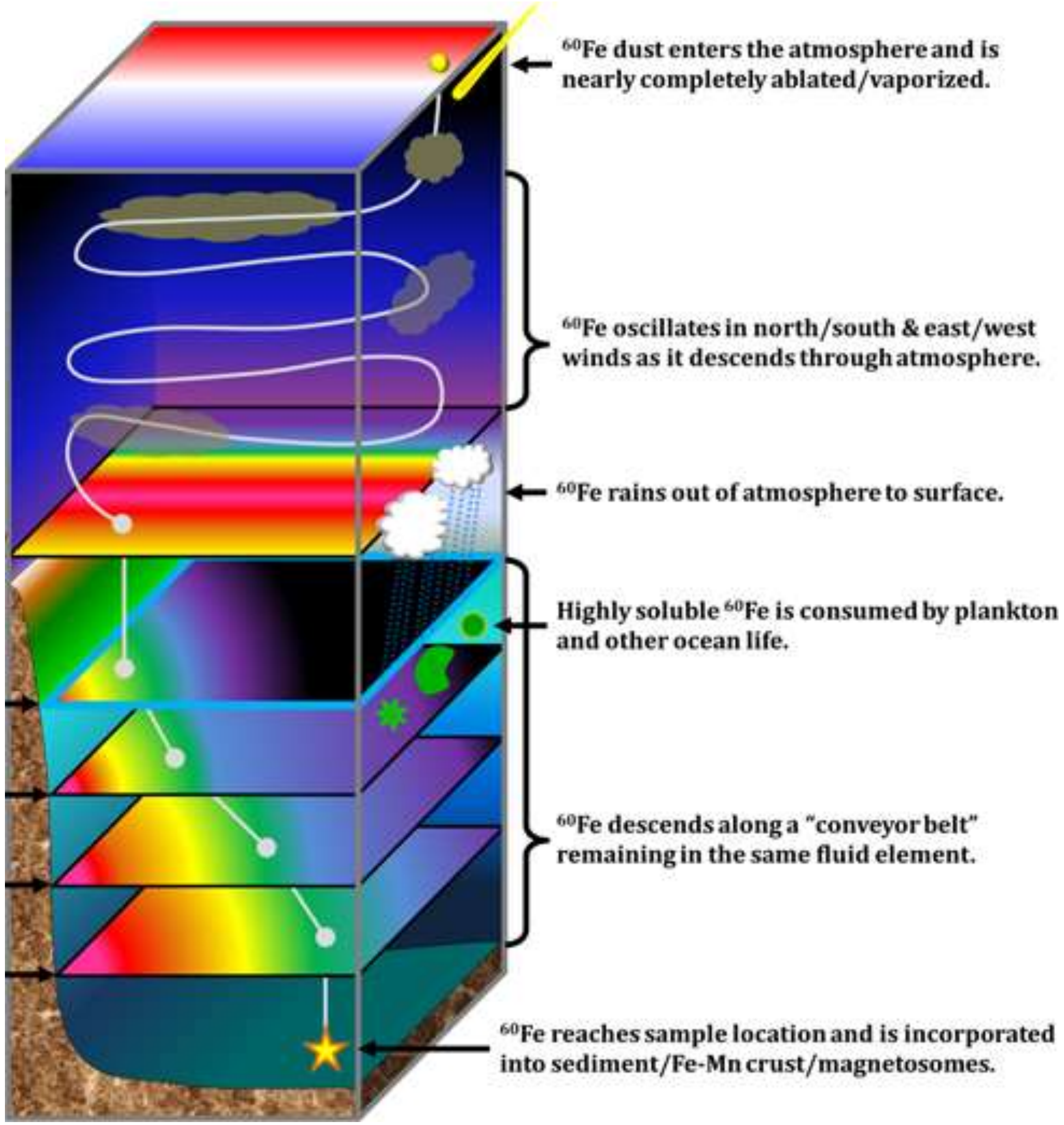
Received 1995 June 15; accepted 1996 May 21

^{10}Be, ^{26}Al, ^{53}Mn, ^{60}Fe, ^{129}I, ^{182}Hf, ^{244}Pu, ^{247}Cm, …

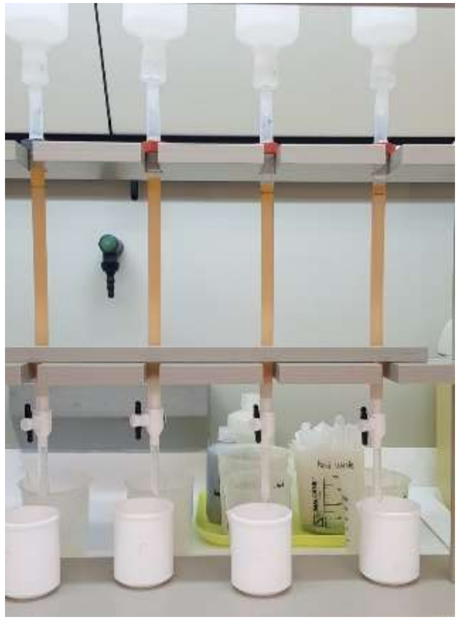
Transport processes on Earth

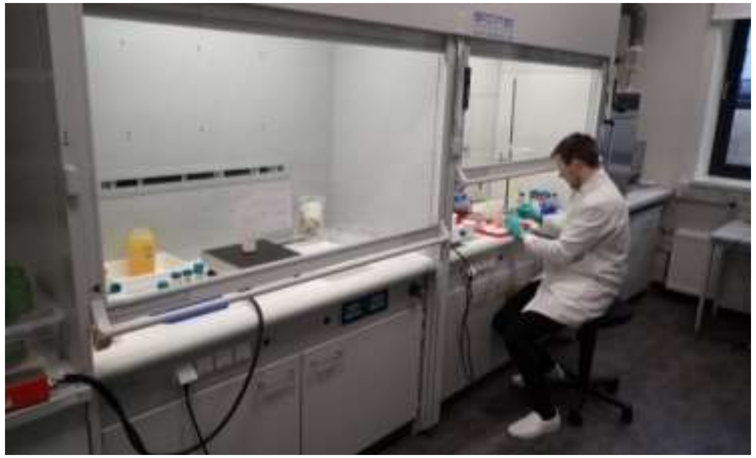
- dust enters atmosphere & gets vaporized
- atoms attach to particles and get rained down
- deposition on ground or in ocean
- transport through ocean currents
- deposition in sediment or other marine „archive“

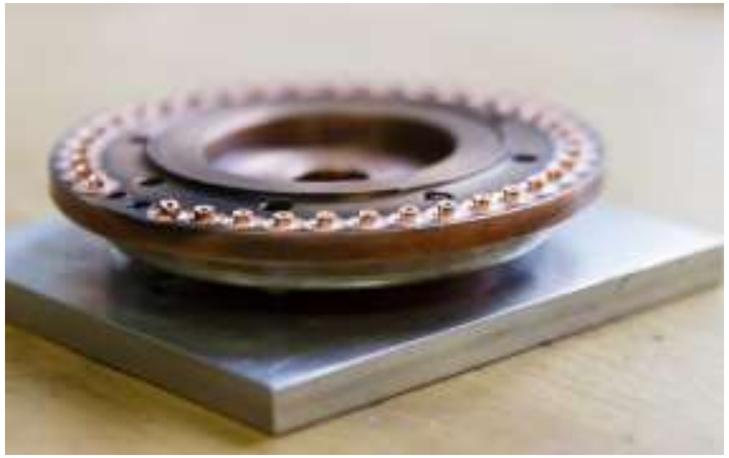


Fry, Fields & Ellis 2016

From the cosmos to the lab

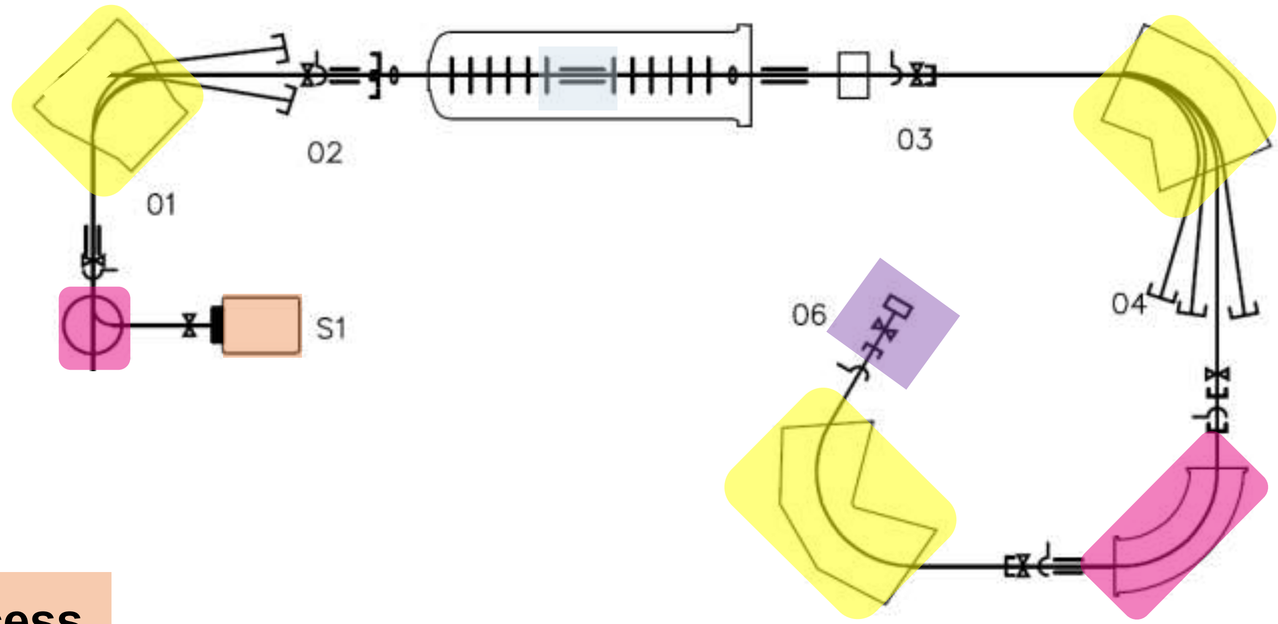






Technique: Accelerator Mass Spectrometry

- ✓ generate, transport and detect many ions of the isotope of interest
  → efficiency typically 0.01-1%
- ✓ suppress molecular and atomic isobars
  → efficiency typically >99.99999%
- ✓ essentially background-free detection at concentrations of 10^{-21} at/g,



The classic AMS toolbox:

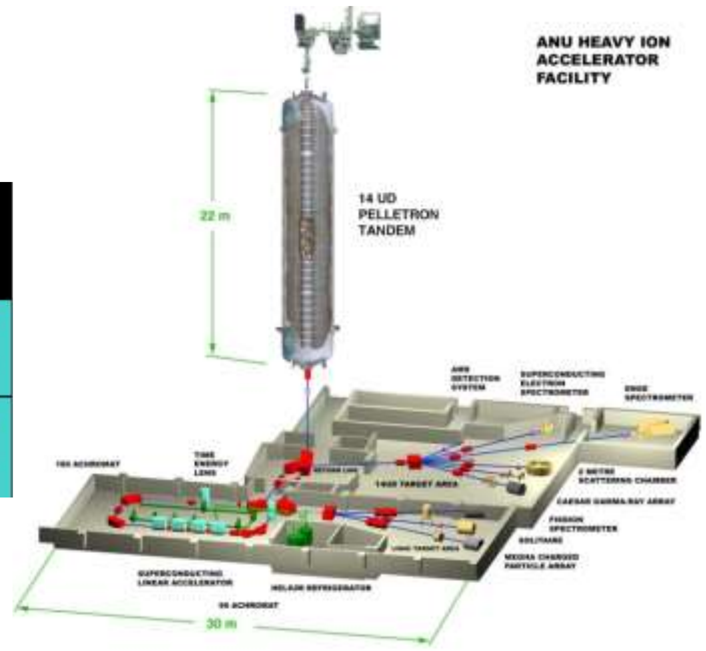
- **element selective negative ionization process**
- **stripping process**
- **mass selection with magnetic and electrostatic filters: isotope ratios in range 10^{-10} to 10^{-17}**
- **single ion identification at increased beam energies: single atom counting**

AMS: examples of ^{60}Fe and ^{244}Pu

^{60}Fe

- big facilities (> 12 MV accelerator voltage) required to provide sufficient isobar suppression at high ion beam energies
- particular efforts at the laboratories at Munich and Canberra

Ni 60 26.2231	Ni 61 1.1399	Ni 62 3.6345
Co 59 100	Co 60 5.2714 yr	Co 61 1.649 h
Fe 58 0.282	Fe 59 44.500 d	Fe 60 2.6 Myr



^{244}Pu

- developments in the AMS community for environmental Pu detection
- no isobaric interference
  going for highest efficiency in particle transport at compact facilities

Pu 239 24.11 kyr	Pu 240 6.561 kyr	Pu 241 14.329 yr	Pu 242 375 kyr	Pu 243 4.9553 h	Pu 244 81.3 Myr

Australian National University

ANSTO
Science. Ingenuity. Sustainability.

DRESDEN concept HZDR

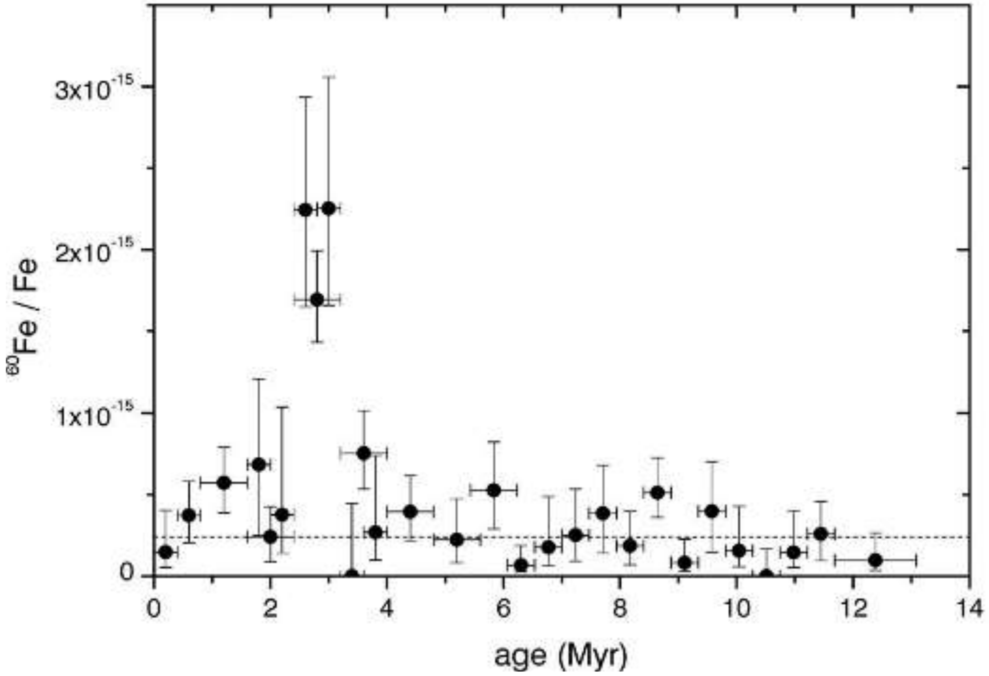
Successful detection of ^{60}Fe

Ni 60	Ni 61	Ni 62
26.2231	1.1399	3.6345
Co 59	Co 60	Co 61
100	5.2714 yr	1.649 h
Fe 58	Fe 59	Fe 60
0.282	44.500 d	2.6 Myr

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]



The ^{60}Fe peak: a continued success story

Ni 60 26.2231	Ni 61 1.1399	Ni 62 3.6345
Co 59 100	Co 60 5.2714 yr	Co 61 1.649 h
Fe 58 0.282	Fe 59 44.500 d	Fe 60 2.6 Myr

LETTER

doi:10.1038/nature17196

Recent near-Earth supernovae probed by global deposition of interstellar radioactive ^{60}Fe

A. Wallner[1], J. Feige[2]†, N. Kinoshita[3], M. Paul[4], L. K. Fifield[1], R. Golser[2], M. Honda[5], U. Linnemann[6], H. Matsuzaki[7], S. Merchel[8], G. Rugel[8], S. G. Tims[1], P. Steier[2], T. Yamagata[9] & S. R. Winkler[2]

RESEARCH ARTICLE | PHYSICAL SCIENCES

Time-resolved 2-million-year-old supernova activity discovered in Earth's microfossil record

Peter Ludwig, Shawn Bishop, Ramon Egli, Valentyna Chernenko, Boyana Deneva, Thomas Faestermann, Nicolai Famulok, Leticia Fimiani, José Manuel Gómez-Guzmán, Karin Hain, Gunther Korschinek, Marianne Hanzlik, Silke Merchel, and Georg Rugel -10 Authors Info & Affiliations

Letter | Published: 06 April 2016

The locations of recent supernovae near the Sun from modelling ^{60}Fe transport

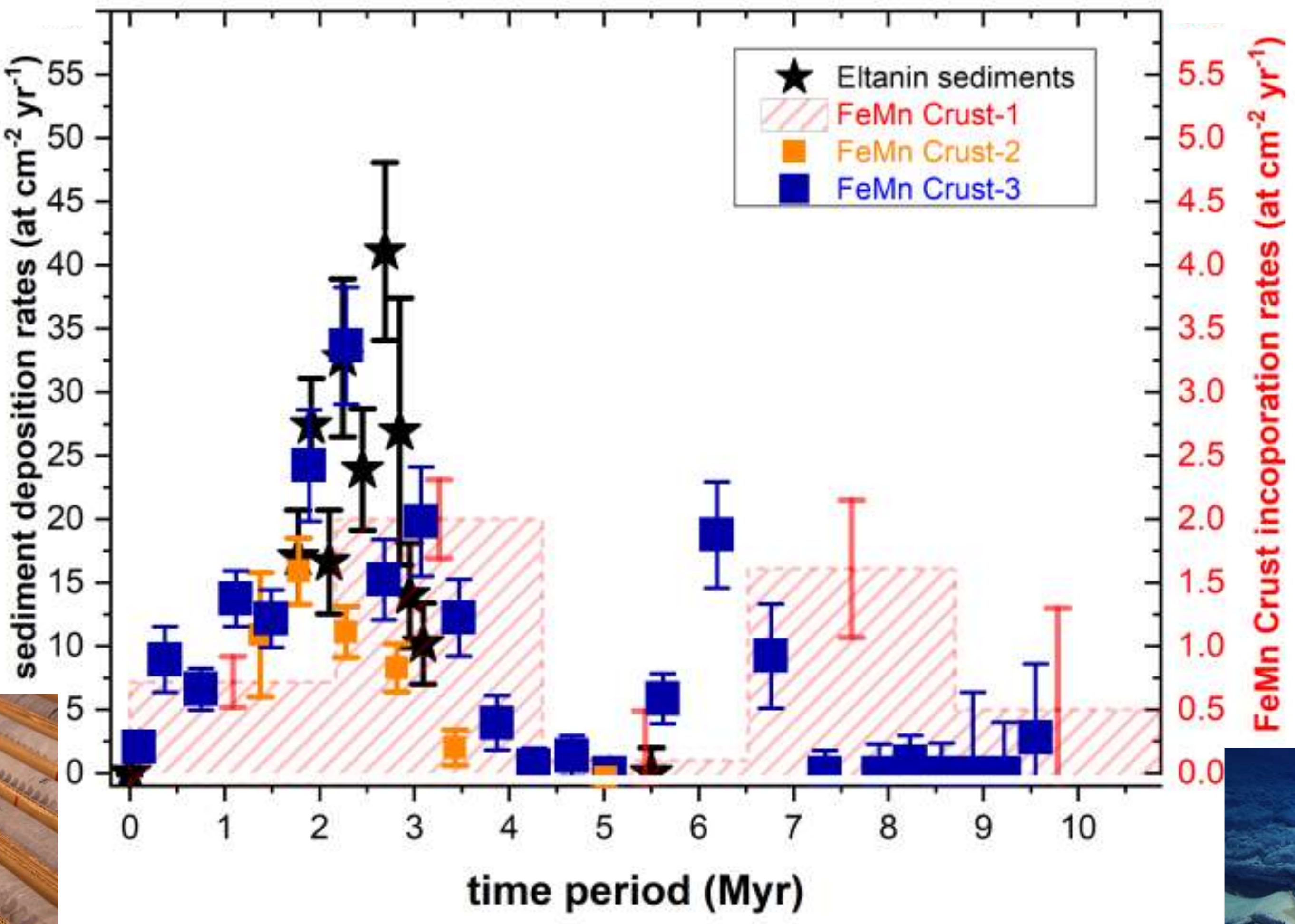
D. Breitschwerdt, J. Feige, M. M. Schulreich, M. A. de. Avillez, C. Dettbarn & B. Fuchs

Nature **532**, 73–76 (2016) | Cite this article

Detection of ^{60}Fe over the last million years

Ni 60	Ni 61	Ni 62
26.2231	1.1399	3.6345
Co 59	Co 60	Co 61
100	5.2714 yr	1.649 h
Fe 58	Fe 59	Fe 60
0.282	44.500 d	2.6 Myr

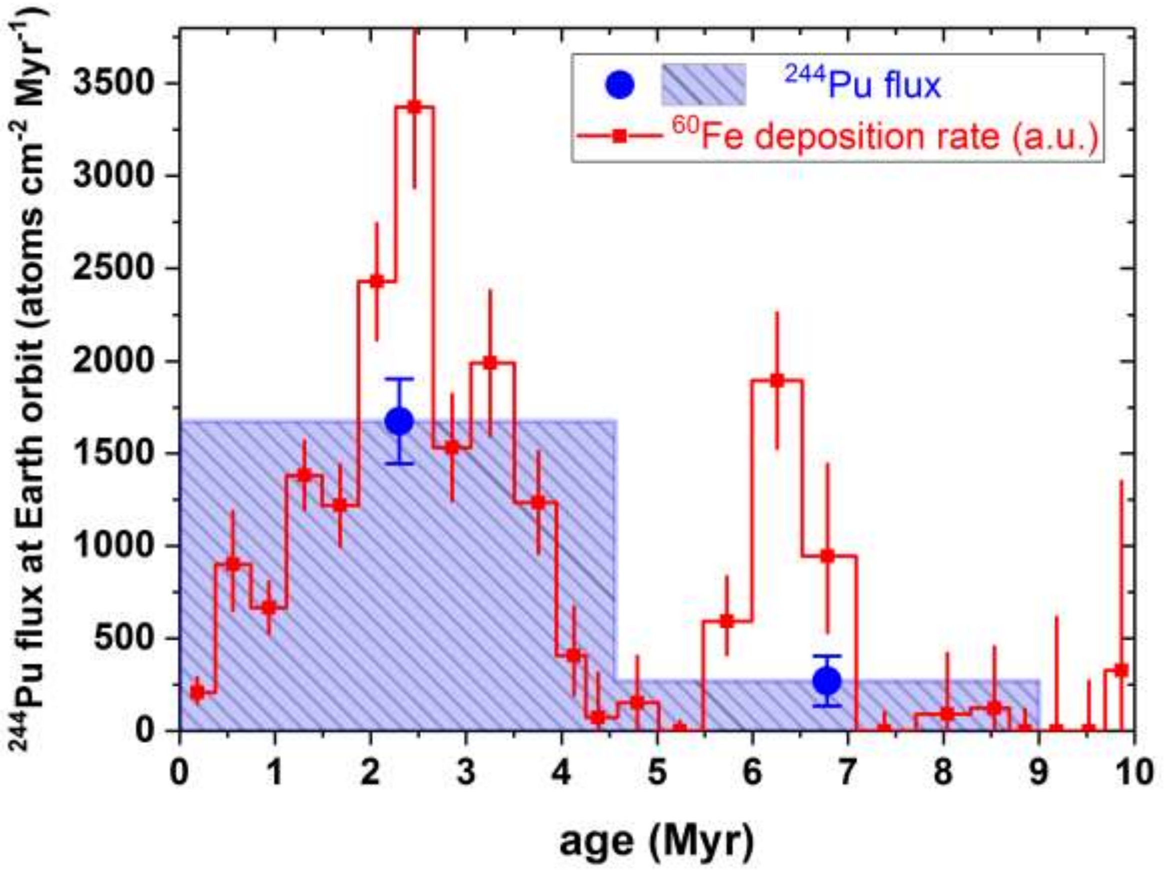
- temporally extended signature
  → multiple events?
- a 2nd earlier influx !



Wallner et al. Nature 2016; Science 2021

Candidate for *r*-process fingerprint: ^{244}Pu

- actinides deliver pure *r*-process signature
- long(er) half-life 80Myr: view further into past
- no significant terrestrial remains from primordial production (Lachner et al., 2012, Wu et al., 2022)
- small influx of live ^{244}Pu, correlates with SN-^{60}Fe?
- ^{244}Pu abundance lower than expected if SNe dominate *r*-process nucleosynthesis
- production of ^{244}Pu within the past few 100 Myr
- difficulty: anthropogenic ^{244}Pu!
  strongly contaminated top mm (!) of
  FeMn crust after sample retrieval
  visible in shorter lived Pu isotopes



Pu 239	Pu 240	Pu 241	Pu 242	Pu 243	Pu 244
24.11 kyr	6.561 kyr	14.329 yr	375 kyr	4.9553 h	81.3 Myr

NUCLEAR ASTROPHYSICS

^{60}Fe and ^{244}Pu deposited on Earth constrain the r-process yields of recent nearby supernovae

A. Wallner[1,2]*, M. B. Froehlich[1], M. A. C. Hotchkis[3], N. Kinoshita[4], M. Paul[5], M. Martschini[1]†, S. Pavetich[1], S. G. Tims[1], N. Kivel[6], D. Schumann[6], M. Honda[7]‡, H. Matsuzaki[8], T. Yamagata[8]

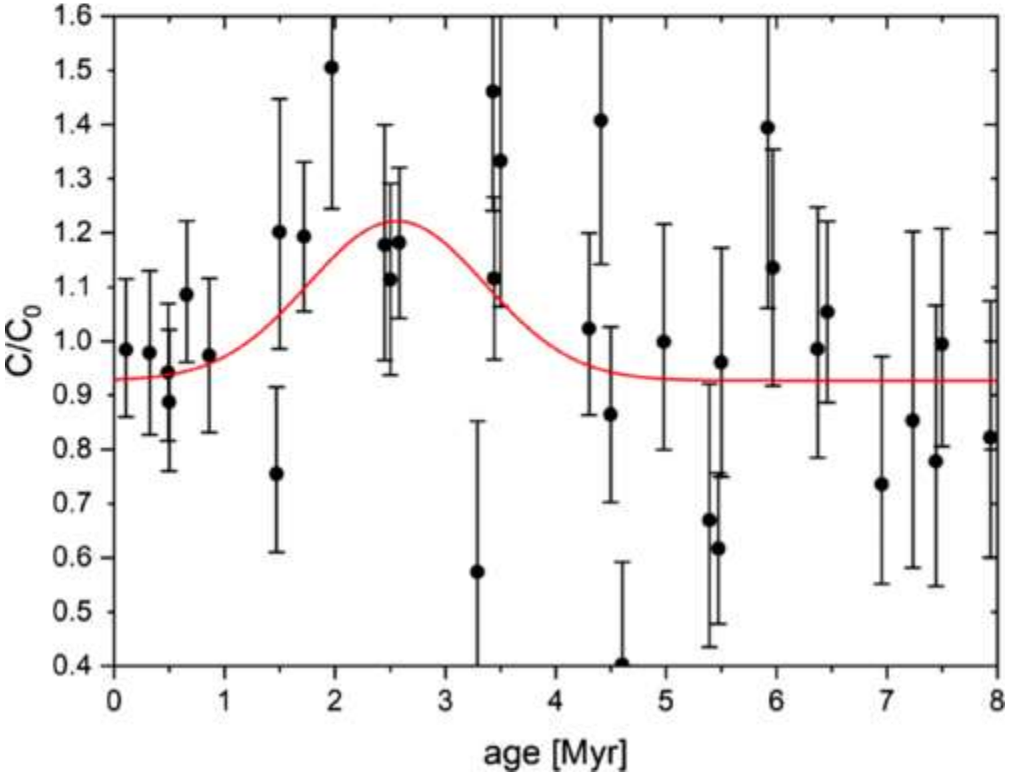
DRESDEN concept HZDR

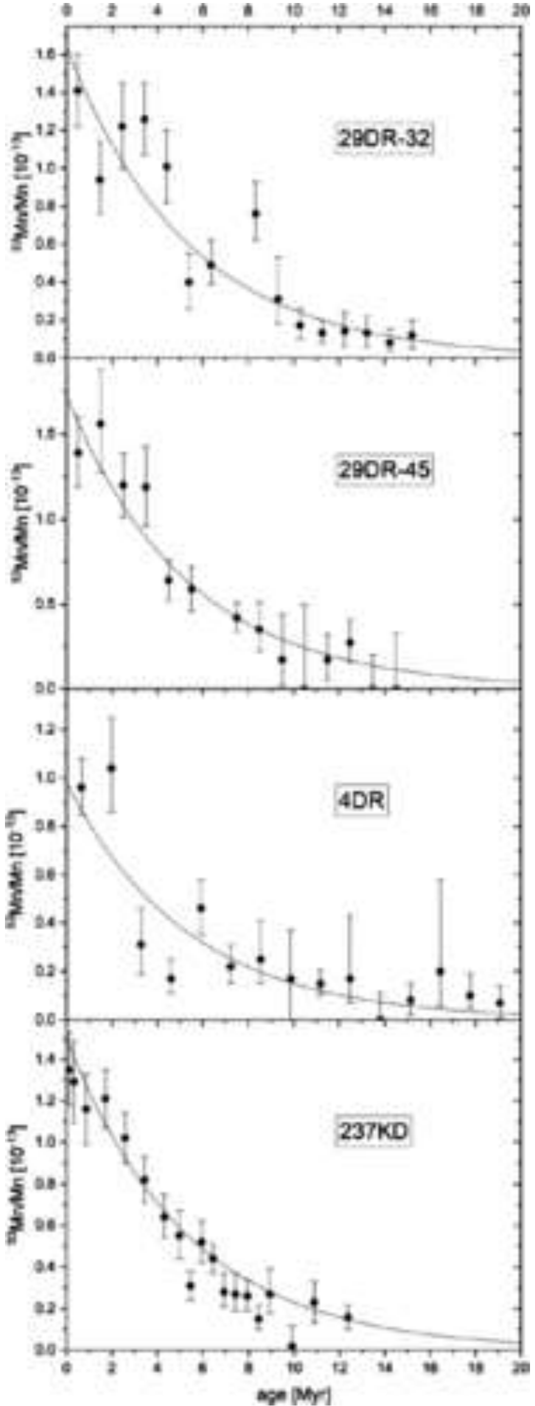
Another long-lived candidate: ^{53}Mn

$^{53}Mn/^{55}Mn$ ratios from deep-sea FeMn crust (Korschinek et al., PRL 2020)

^{53}Mn mainly formed by cosmic ray reactions (mostly on iron)

surplus of ^{53}Mn coincidental with ^{60}Fe





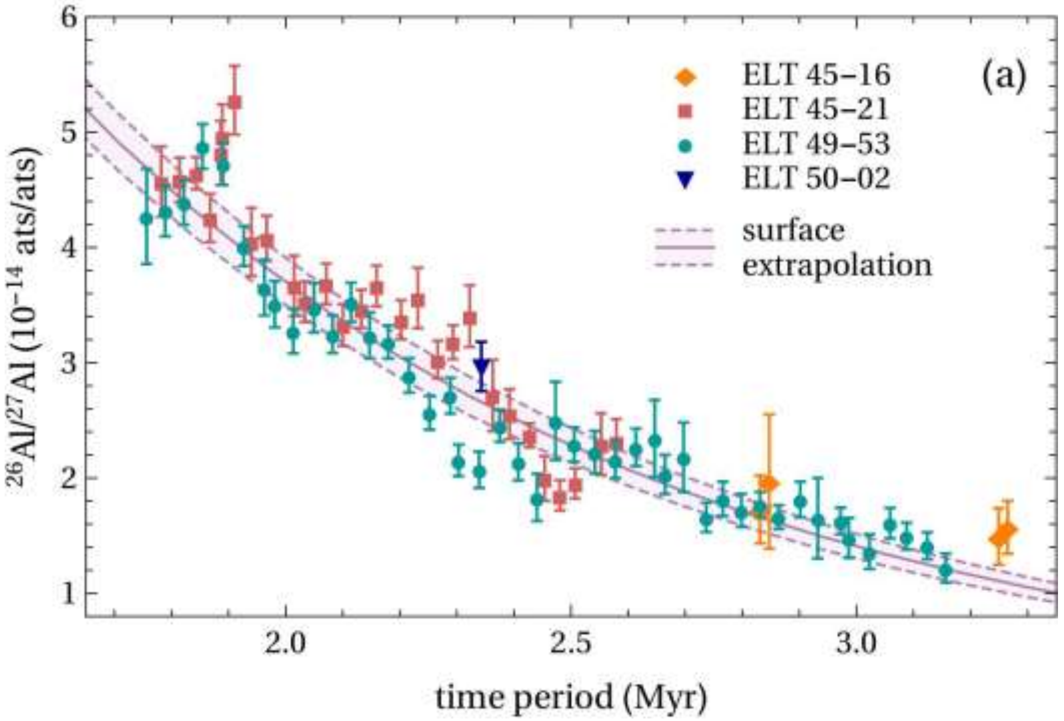
Another long-lived candidate: ^{26}Al

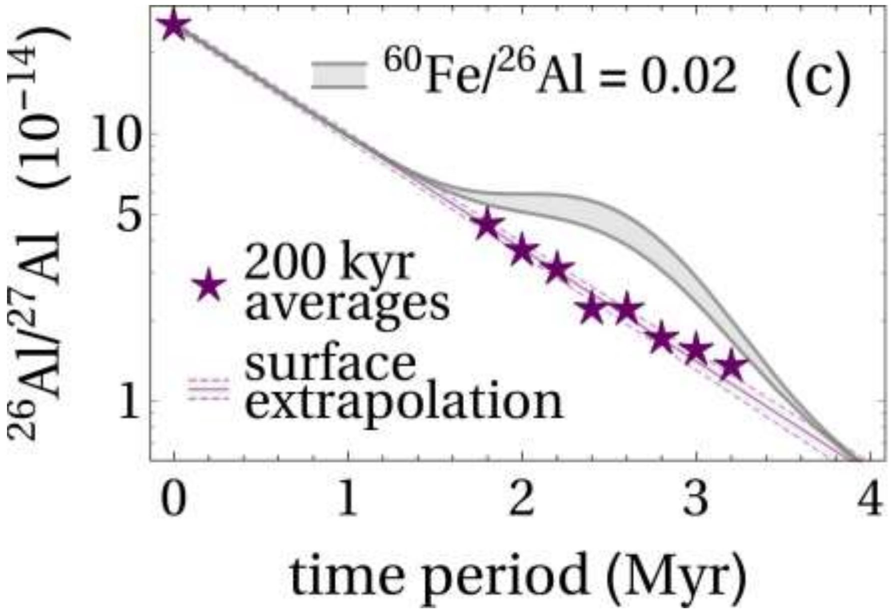
^{26}Al/^{27}Al ratios from deep-sea sediment cores
(Feige et al., PRL 2018)

assume that ^{60}Fe and ^{26}Al are transported equally to the solar system within dust particles

data set provides lower limits: $^{60}\mathrm{Fe}\,/^{26}\mathrm{Al} = 0.18^{+0.15}_{-0.08}$

from γ-ray observations: $^{60}\mathrm{Fe}\,/^{26}\mathrm{Al} = 0.18 \pm 0.04$
(Wang et al., 2020)





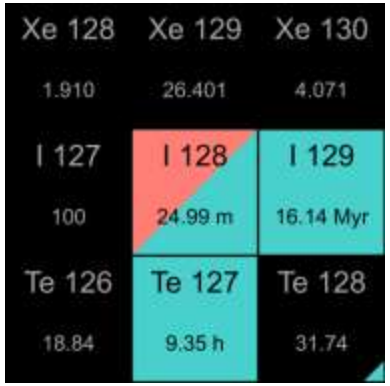
Al 26 717 kyr	Al 27 100
Mg 25 10.011	Mg 26 11.025

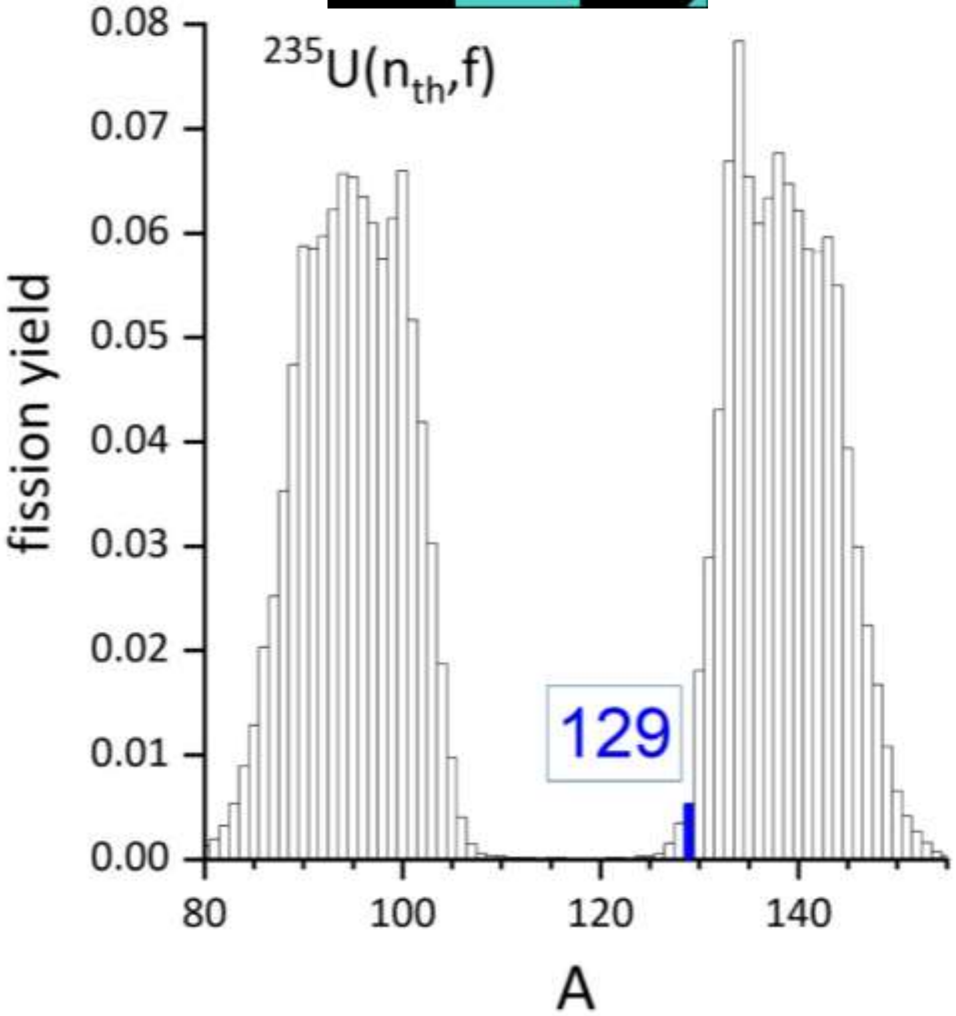
DRESDEN concept HZDR

Another long-lived candidate: ^{129}I

- perfect AMS radionuclide:
  good negative ion formation
  stable isobar does not form negative ions
- long half-life: $T_{1/2}$=16 Myr

- too much background production on our planet:
  - atmospheric production by GCR not clearly established (10^{-4} - 10% relative to ^{26}Al)
  - fission isotope:
    anthropogenic contamination: continued release from reprocessing plants
    continuous production from spontaneous fission of ^{238}U
- high solubility of I in (ocean) water:
  long residence time of IO_3^- (340 kyr, Broecker & Peng 1982)
  → signals will get washed out

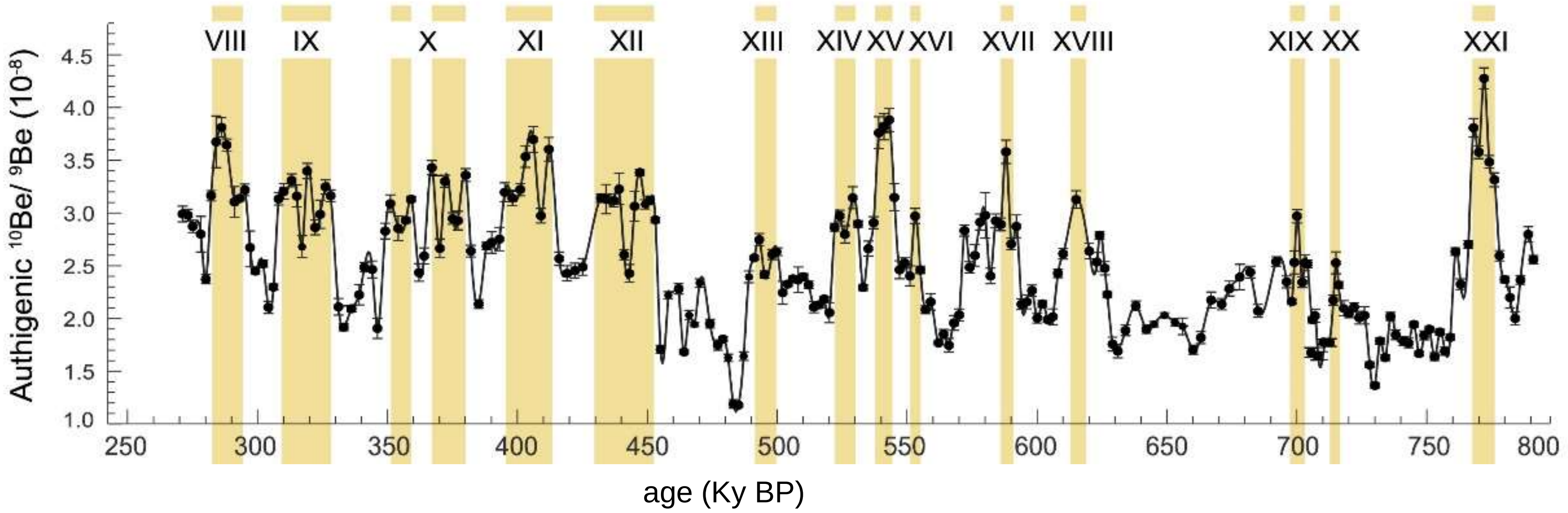




A radionuclide with many signatures: ^{10}Be

B 10	B 11
19.65	80.35
Be 9	Be 10
100	1.387 Myr

- primary isotope for dating archives on Million year ranges via decay curves
- short-termed spikes in concentration appear due to
  - geomagnetic events (field excursions & reversals)
  - solar proton events
- production signal also modulated by solar magnetic field



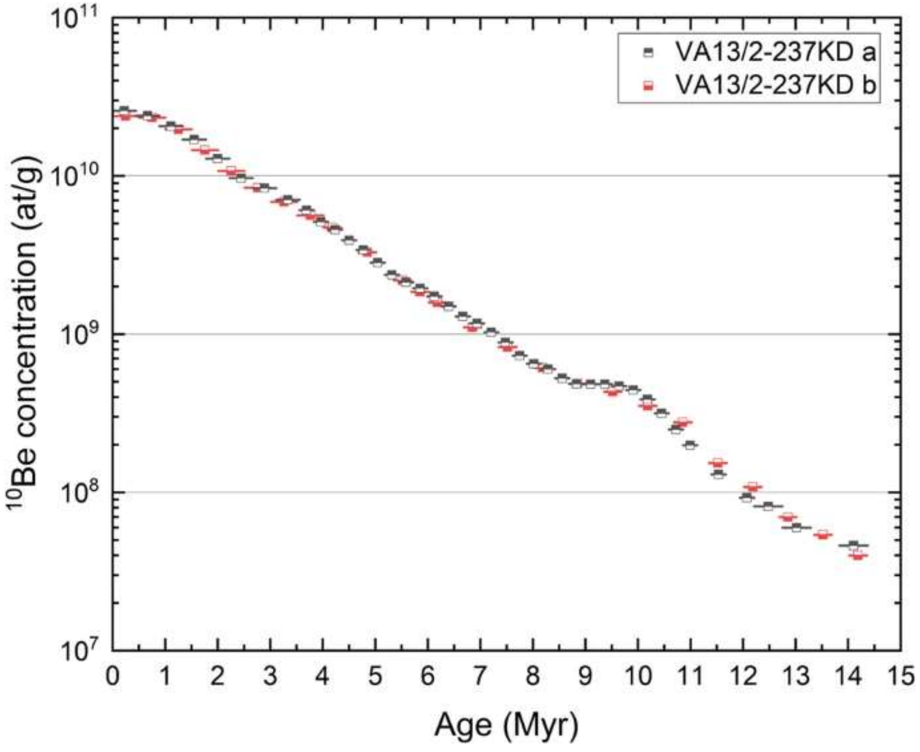
Ménabréaz et al., 2014

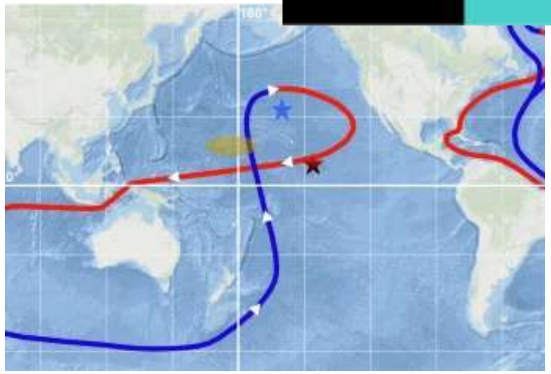
New insights from ^{10}Be

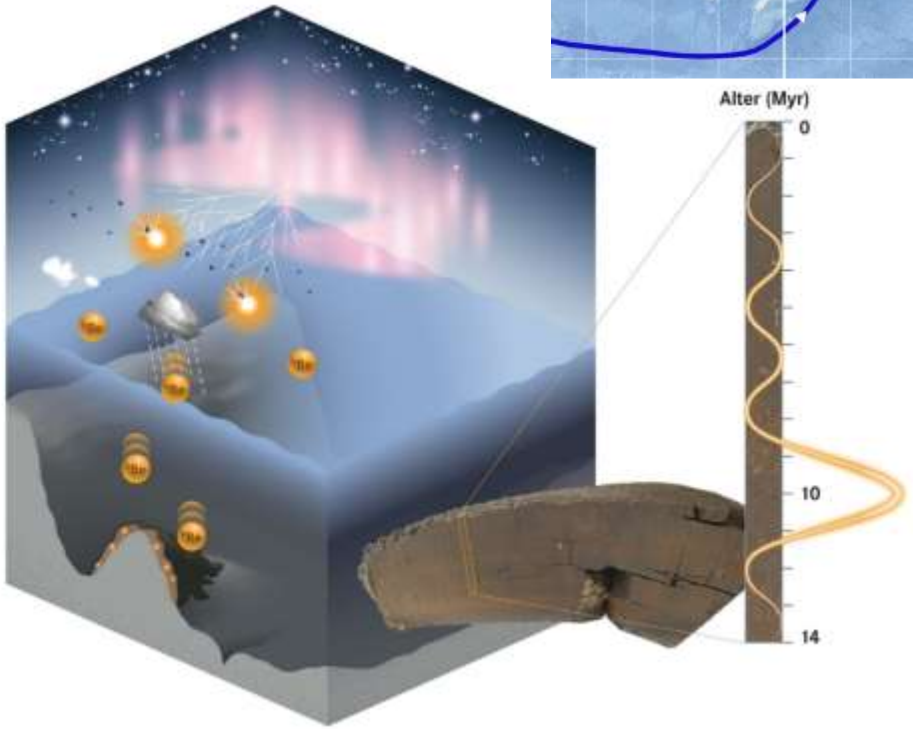
B 10	B 11
19.65	80.35
Be 9	Be 10
100	1.387 Myr

Koll et al., Nature Comm. 2025

- significant change of otherwise long-time stable ocean transport pattern?
- increased production of ^{10}Be in atmosphere?



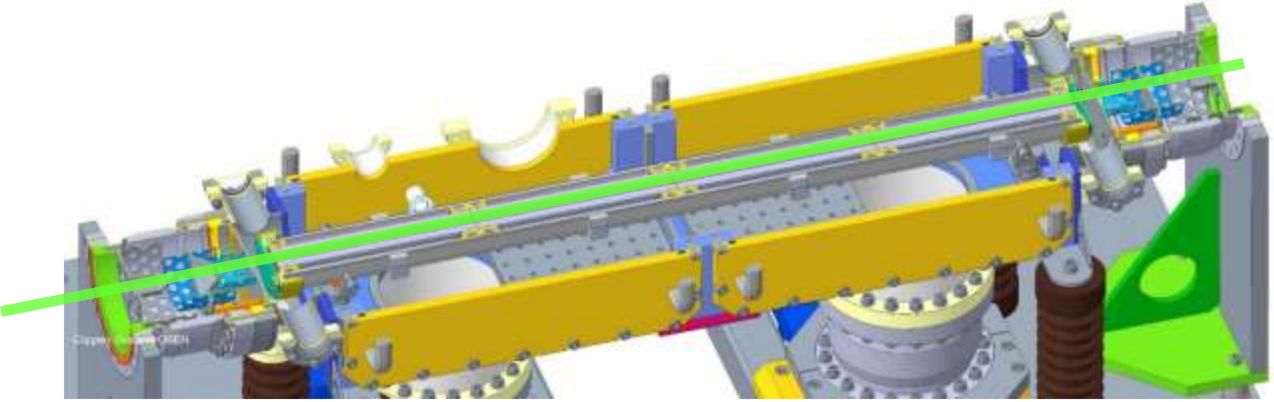


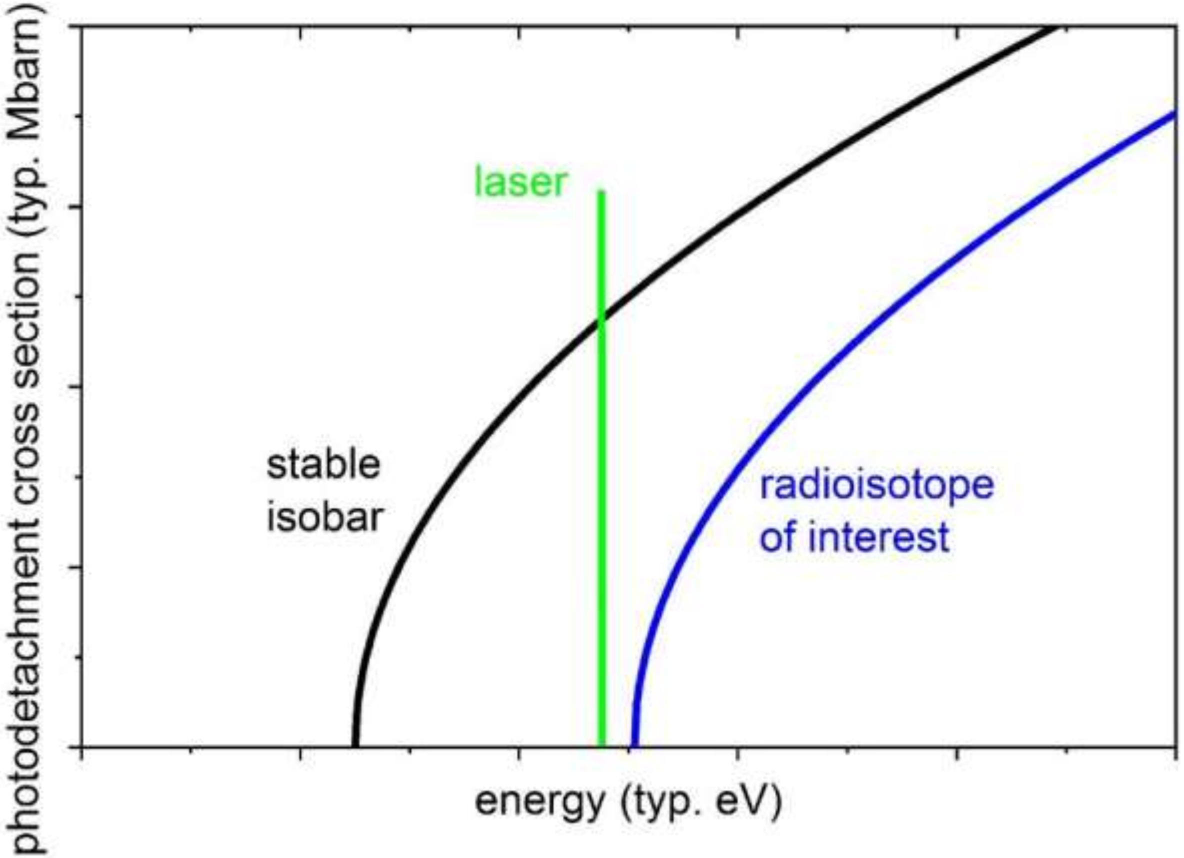


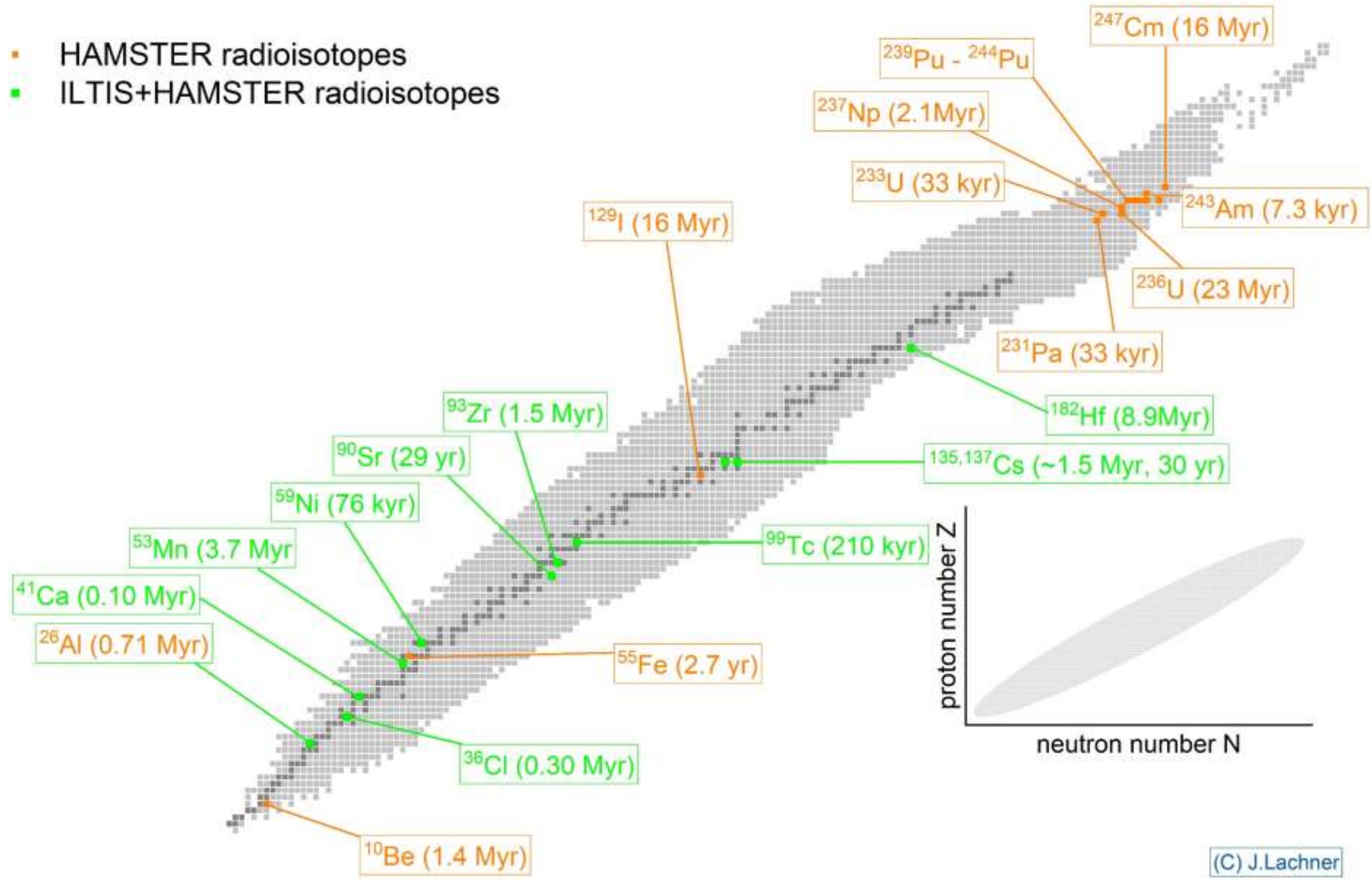
What about ^{182}Hf? AMS 2.0

- new isobar suppression techniques
- add more element selective processes for negative ions before the accelerator: interactions of slow ions with gas and laser light
- promising results for ^{182}Hf (Martschini et al., 2020)

W 182 26.50	W 183 14.31	W 184 30.64
Ta 181 99.98799	Ta 182 114.74 d	Ta 183 5.1 d
Hf 180 35.08	Hf 181 42.39 d	Hf 182 8.90 Myr







HAMSTER radioisotopes
ILTIS+HAMSTER radioisotopes
247Cm (16 Myr)
239Pu - 244Pu
237Np (2.1Myr)
233U (33 kyr)
243Am (7.3 kyr)
129I (16 Myr)
236U (23 Myr)
231Pa (33 kyr)
182Hf (8.9Myr)
93Zr (1.5 Myr)
90Sr (29 yr)
135,137Cs (~1.5 Myr, 30 yr)
59Ni (76 kyr)
53Mn (3.7 Myr
99Tc (210 kyr)
41Ca (0.10 Myr)
26Al (0.71 Myr)
55Fe (2.7 yr)
36Cl (0.30 Myr)
10Be (1.4 Myr)
proton number Z
neutron number N
(C) J.Lachner

^{182}Hf and ^{247}Cm

W 182 26.50	W 183 14.31	W 184 30.64
Ta 181 99.98799	Ta 182 114.74 d	Ta 183 5.1 d
Hf 180 35.08	Hf 181 42.39 d	Hf 182 8.90 Myr

MS 6.3 13. März 2025, 11:30
Investigations on ILIAMS isobar suppression for non-routine AMS isotopes — •MARTIN MARTSCHINI

MS 5.6 Poster
AMS-detection of ^{182}Hf: Characterization of new low-level reference materials and cross-contamination experiments — •LAURENZ WIDERMANN

Cm 246 4.706 kyr	Cm 247 15.6 Myr	Cm 248 348 kyr
Am 245 2.05 h	Am 246 39 m	Am 247 23.0 m
Pu 244 81.3 Myr	Pu 245 10.5 h	Pu 246 10.84 d

MS 4.5; today, 17:00
Challenges in the extraction of ^{182}Hf from geological archives — •SEBASTIAN FICHTER

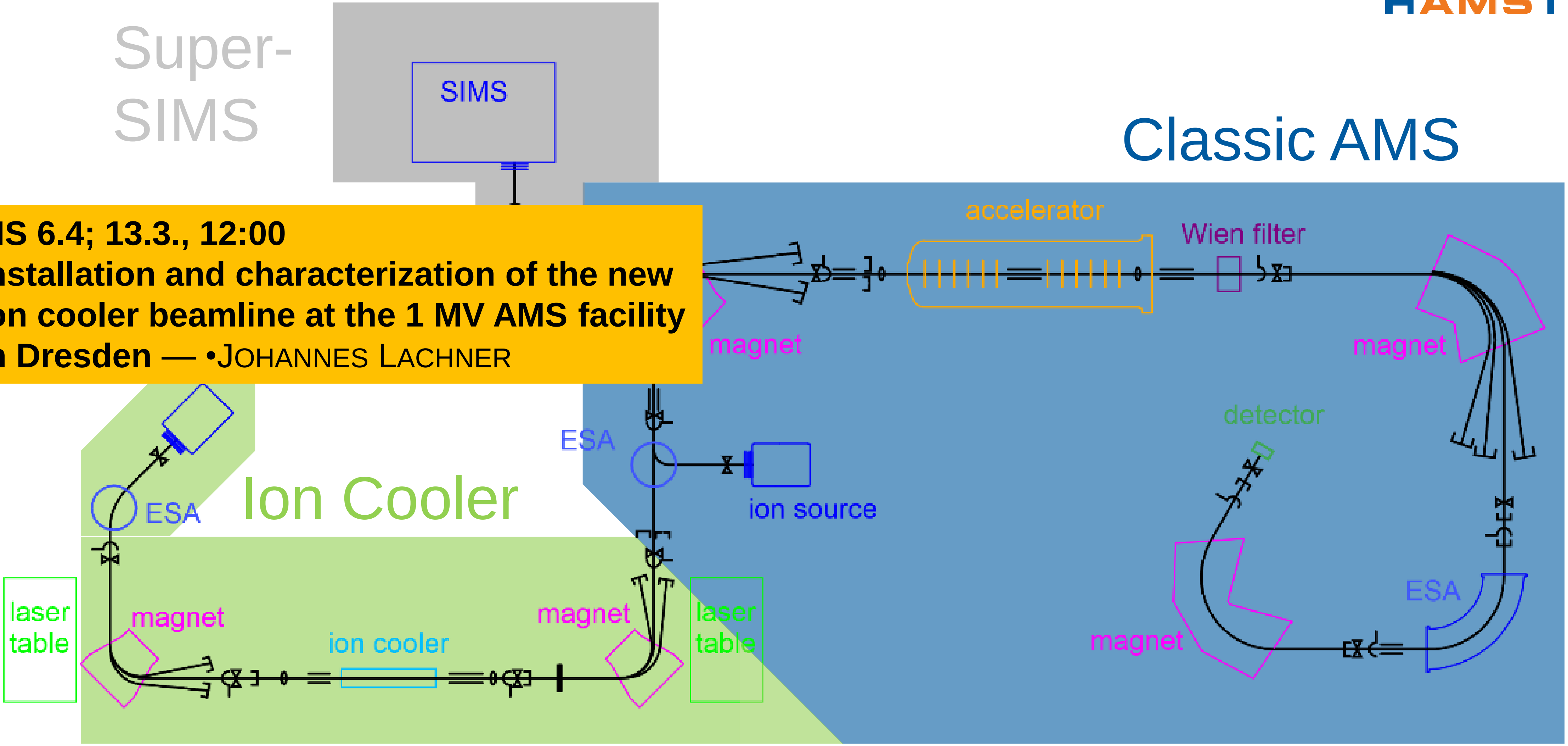
MS 9.5; 13. März 2025, 18:45
Isotopic purification of trans-uranium tracers using RIMS at RISIKO and their characterization with AMS (II) — •DOMINIK KOLL

MS 6.5; 13. März 2025, 12:15
Photodetachment measurements of negatively charged molecules and element separation at VERA — •T. NIEMEYER

A new 1 MV AMS facility at HZDR: HAMSTER

Helmholtz Accelerator Mass Spectrometer Tracing Environmental Radionuclides

MS 6.4; 13.3., 12:00
Installation and characterization of the new ion cooler beamline at the 1 MV AMS facility in Dresden — •JOHANNES LACHNER

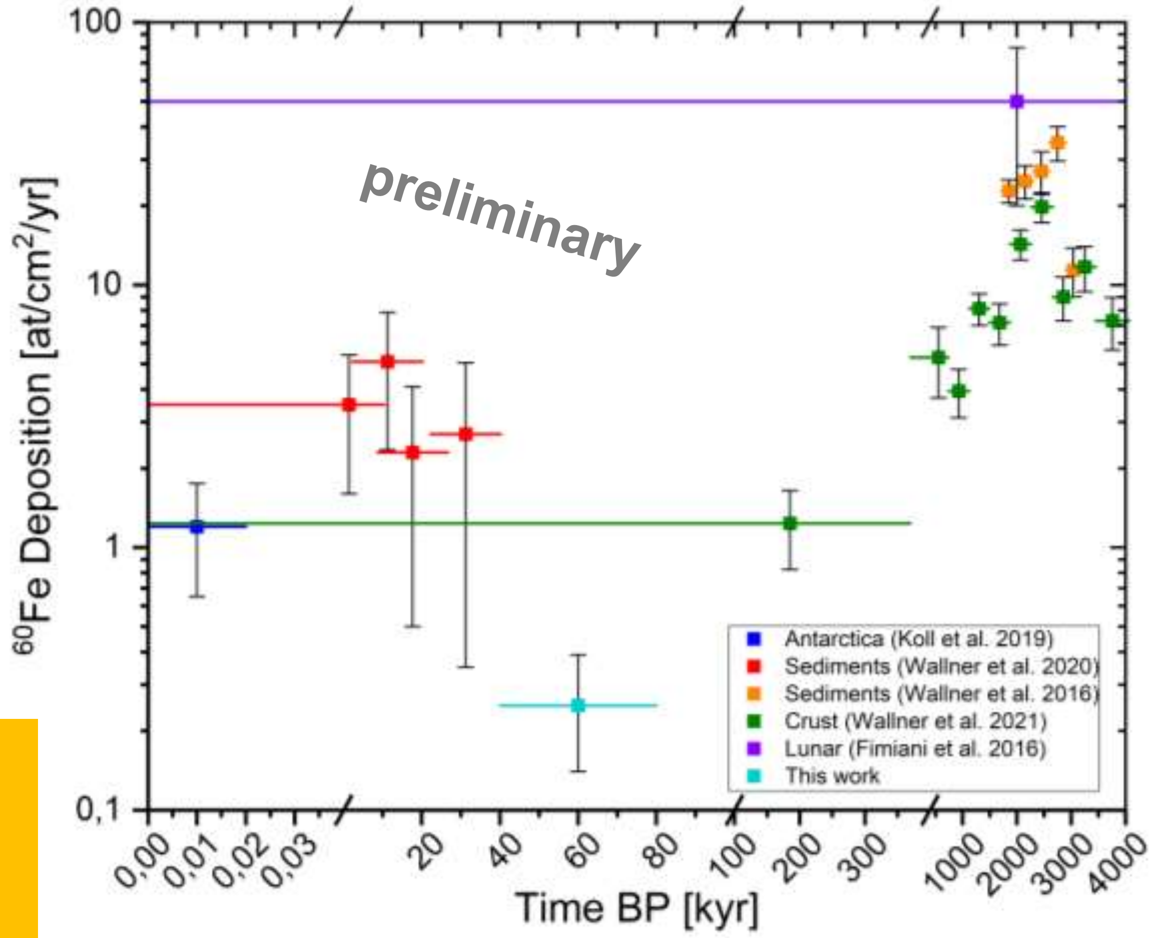


Alternatives to FeMn crusts and marine sediments

Detection of ^{60}Fe in younger sediment or in ice & snow

- recent input of interstellar material?
- ice cores as well-resolved archive of direct precipitation
- interplanetary contribution quantified by $^{60}Fe/^{53}Mn$ ratio
- present day ^{60}Fe deposition on Earth ~1.2 atoms/cm²/yr

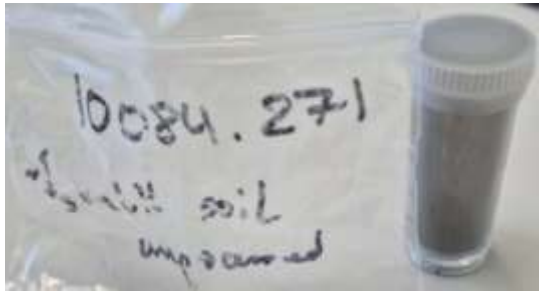
MS 4.3; today, 16:30
Interstellar ^{60}Fe in Antarctic Ice Tracing the Local Interstellar Cloud
— •ANNABEL ROLOFS



Rolofs, 2024; based on Koll et al., 2020

Expanding the time scale: Go to the Moon

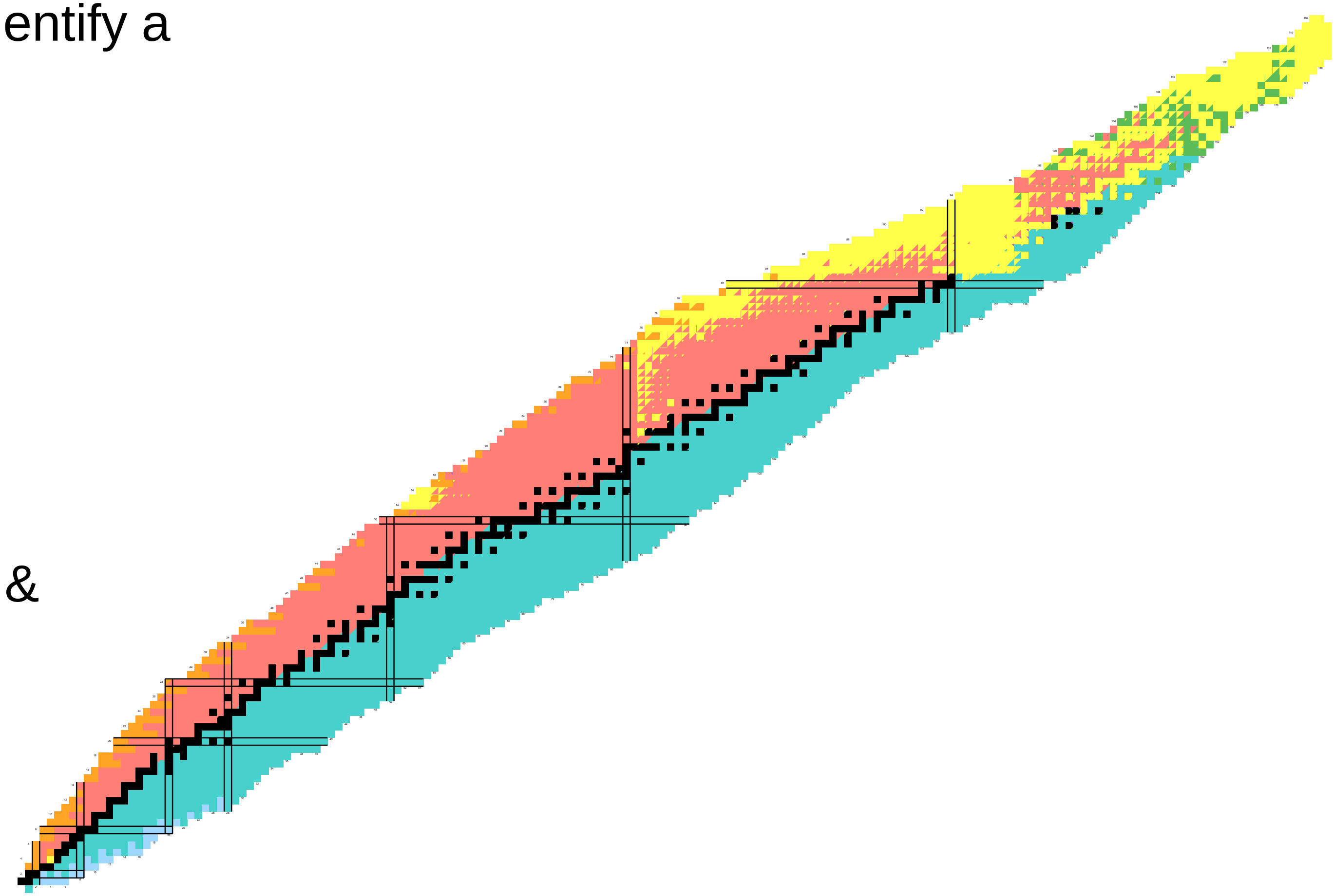
- no geologic activity:
  - no time resolution
  + integration of input over many Myr
- „direct“ collection of cosmic dust:
  smaller sample size sufficient
- easier to find old remnants of ongoing nucleosynthesis



MS 4.3; today, 16:45
Interstellar Radionuclides in Lunar Regolith Tracing Supernova and *r*-Process Events —
•SEBASTIAN ZWICKEL

Summary

- few radionuclides relevant & suitable to identify a cosmic nucleosynthesis fingerprint
- different archives tell us different stories
- AMS as powerful technique to detect live radionuclides in nature
- critical to rule out input from other natural & anthropogenic sources



Special Thanks & Acknowledgments go to

A. Wallner, S. Fichter, **D. Koll**, A. Rolofs, S. Merchel,
G. Rugel, S. Zwickel

HELMHOLTZ ZENTRUM DRESDEN ROSSENDORF

Australian National University

A. Stuchbery, S. Battisson, L. K. Fifield, M. B. Froehlich,
M. Hartnett, S. Pavetich, Z. Slavkovska, S. G. Tims

M. A. Hotchkis, D. Child

ANSTO
Science. Ingenuity. Sustainability.

G. Korschinek, T. Faestermann

J. Feige

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F. Adolphi, M. Hörhold