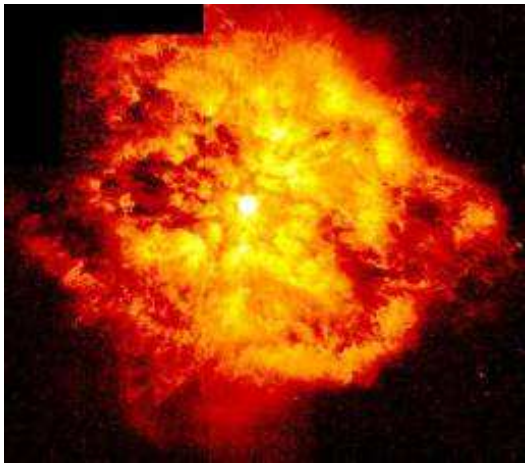
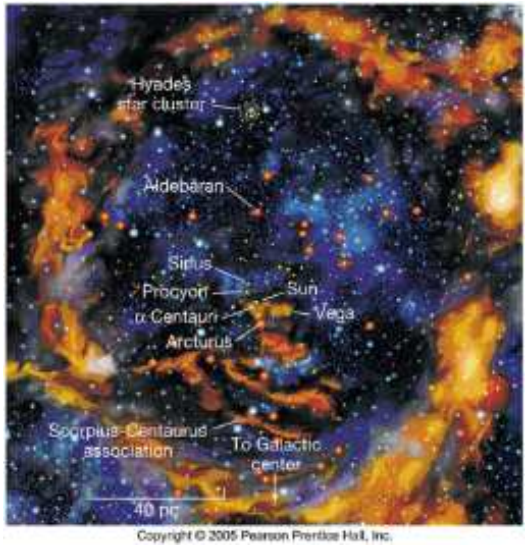
Nuclear Data in AMS:
from nuclear (astro) physics to the environment



The Australian National University (ANU)



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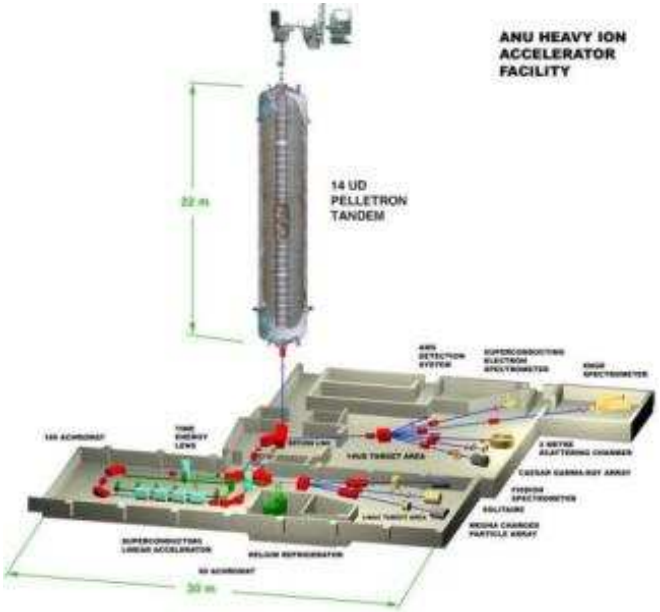

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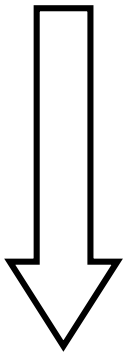
Some radionuclides measured with AMS

fundamental physics



applied sciences

^{55}Fe	2.7 years
^{3}H	12.3
^{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
^{79}Se	280 000
^{36}Cl	301 000
^{26}Al	720 000
^{10}Be	1 388 000
^{60}Fe	2 600 000
^{53}Mn	3 600 000
^{182}Hf	8 900 000
^{129}I	17 000 000
^{236}U	23 000 000
$^{239\text{-}244}$Pu, ^{247}Cm	- 81 000 000

new: ^{55}Fe, ^{68}Ge, ^{93}Zr, ^{106}Pd, ^{135}Cs, ^{146}Sm, ^{202}Pb, ^{210}Bi, ^{226}Ra, ^{229}Th, stable…

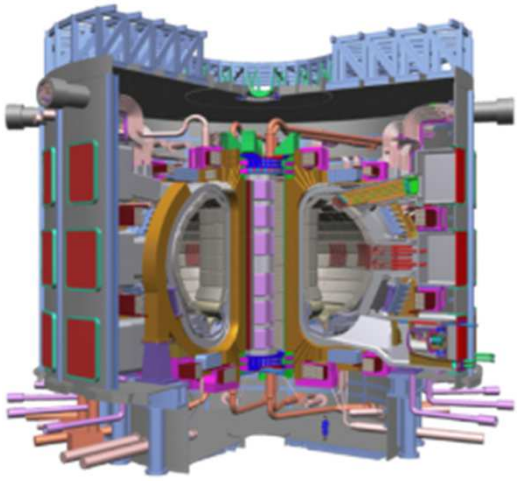
Some recent activities using AMS

I. **Nuclear Astrophysics**: Radionuclides of **extraterrestrial** origin

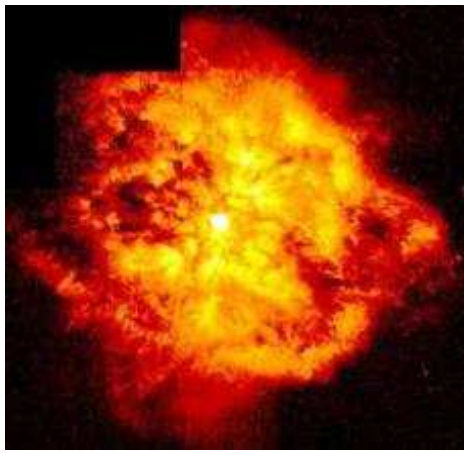
II. **Nuclear Astrophysics**: Studying nucleosynthesis in stars (cross sections)

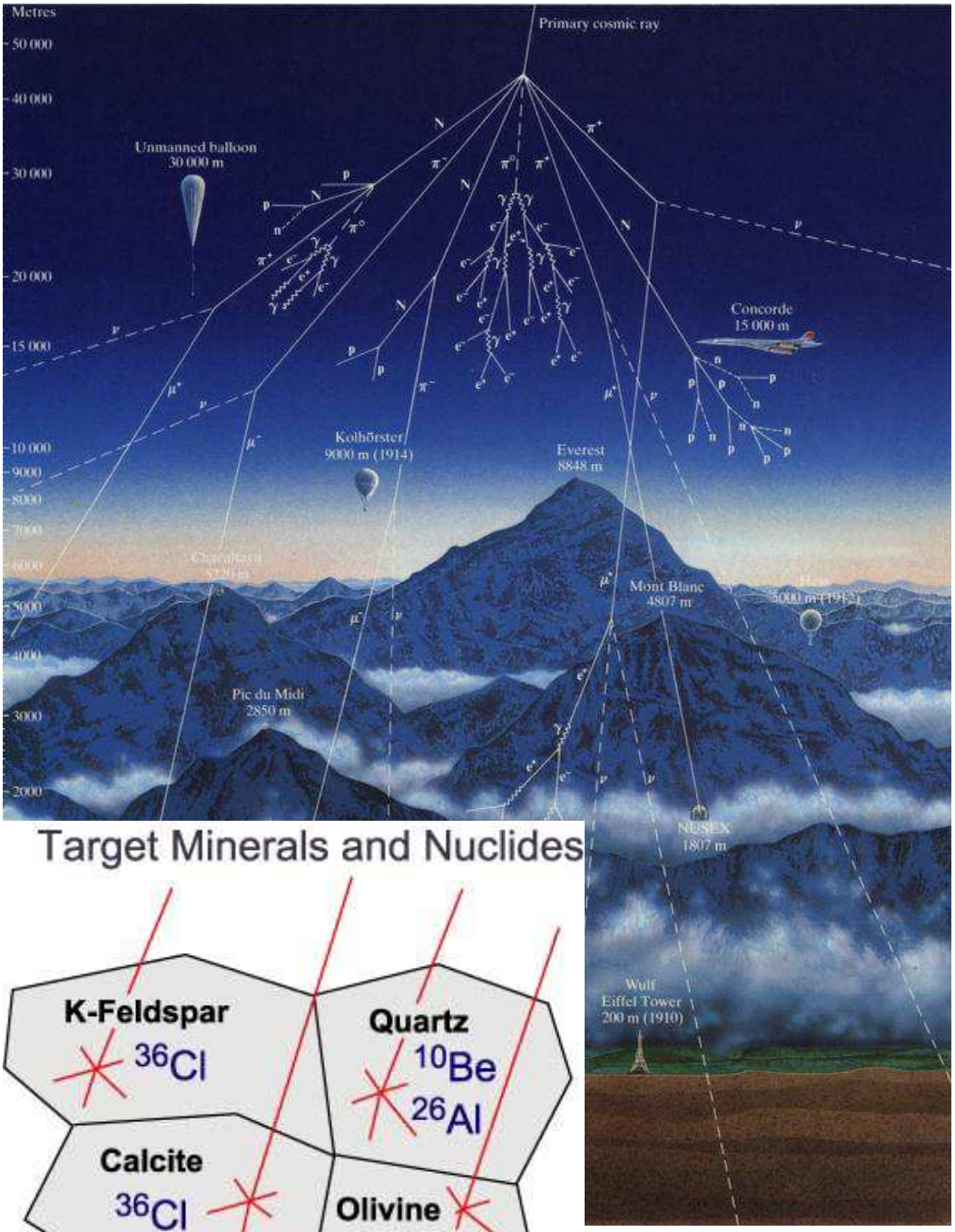
III. **Nuclear Physics**: Radionuclides in the environment

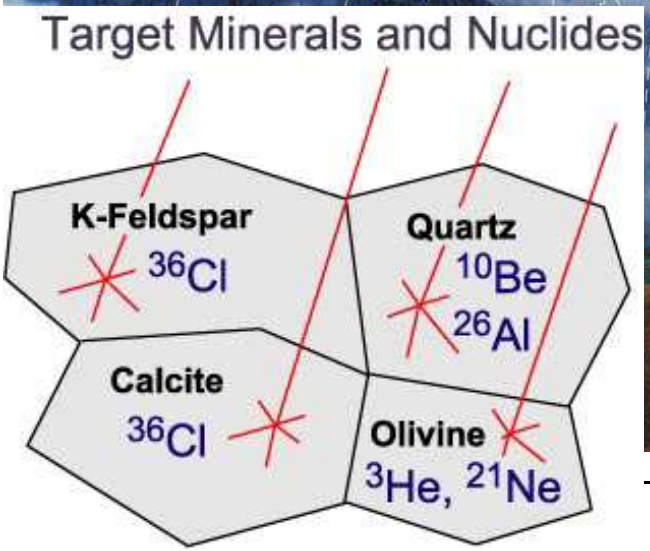
IV. **Nuclear Physics**: Cross-section measurements and production rates for nuclear applications (fusion-ITER, fission-GenIV, ADS) – longlived nuclides



atom counting of radionuclides







Cosmogenic nuclides

e.g. ^{7}Be, ^{10}Be, ^{14}C, ^{21}Ne, ^{26}Al, ^{36}Cl from O, N, Ar

Earth and Space Science

- *Terrestrial cosmogenic nuclide production*
- *Atmospheric processes*
- *Paleoclimatology*
- *Geomorphology*
- *Neotectonics*
- *Glaciology*
- *Hydrology (ground water dating)*
- *Cosmic ray exposure of meteorites*
- *Solar variability*

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Energy and Mineral Resources application

- *Dating of petroleum fluids*
- *Methane-rich fluid migration in shallow geosphere*
- *Geothermal Energy*
- *Waste disposal*

Biological Sciences

- *Tracing organic molecules with ^{14}C*
- *Measuring bone resorption rates with ^{41}Ca*
- *Studying the effectiveness of adjuvants in vaccines with ^{26}Al*

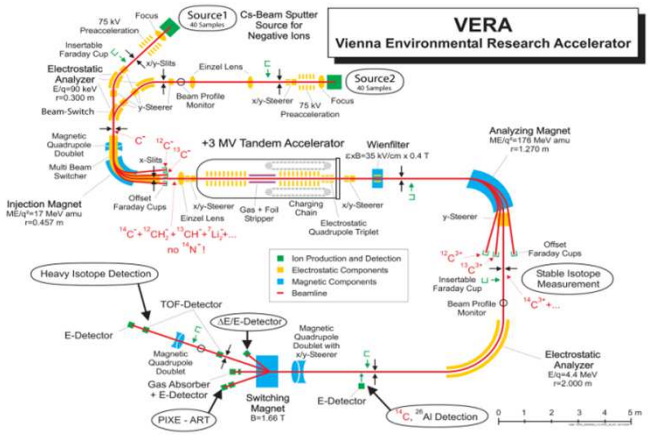
Health physics:

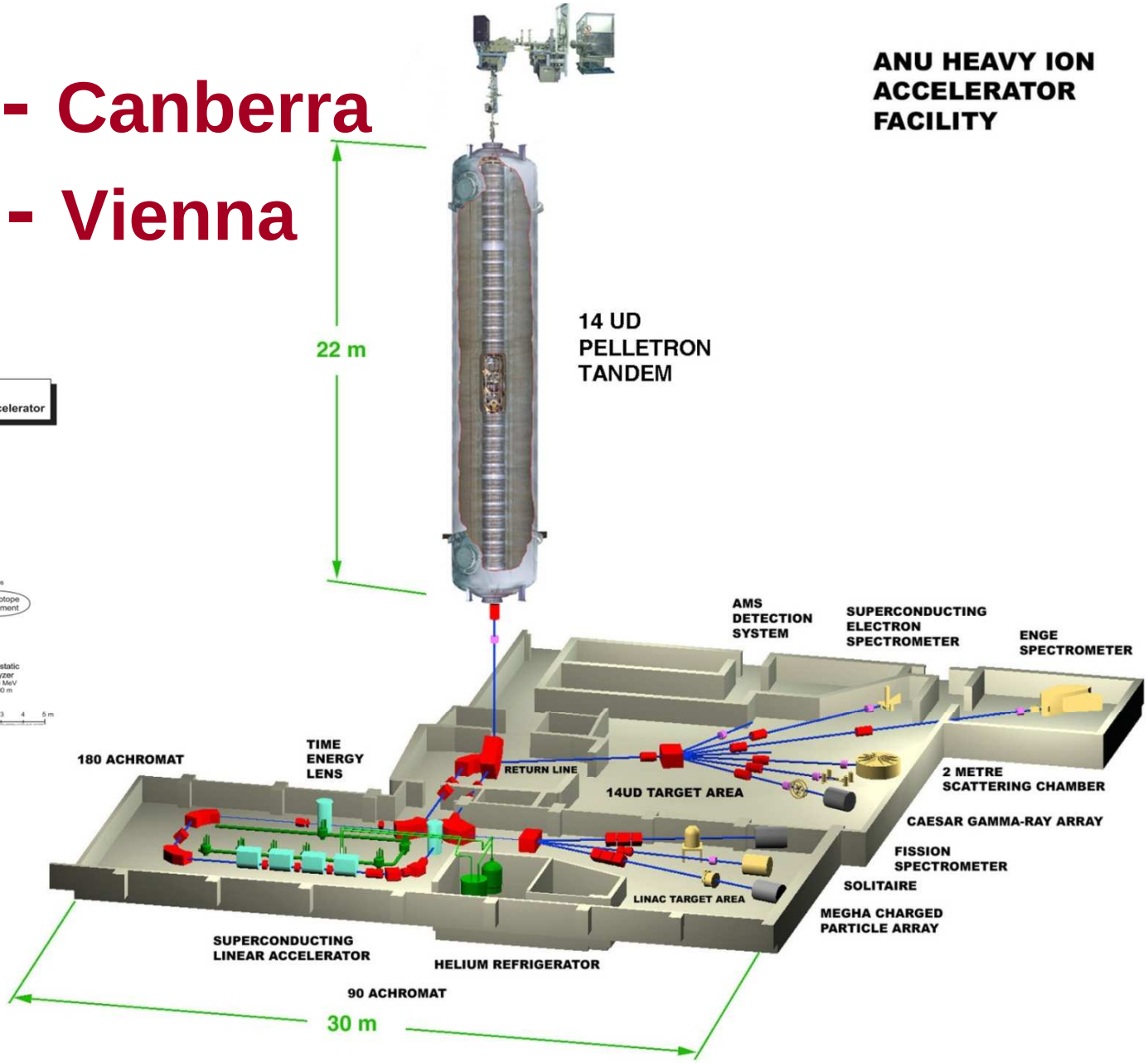
- *Reconstruction of Hiroshima and Chernobyl dosimetry*

Nuclear safety and safeguard

- *Environmental monitoring to detect signatures of undeclared nuclear activities*
- *Environmental monitoring around nuclear power reactors and reprocessing plants*

ANU (15 MV) - Canberra
VERA (3 MV) - Vienna





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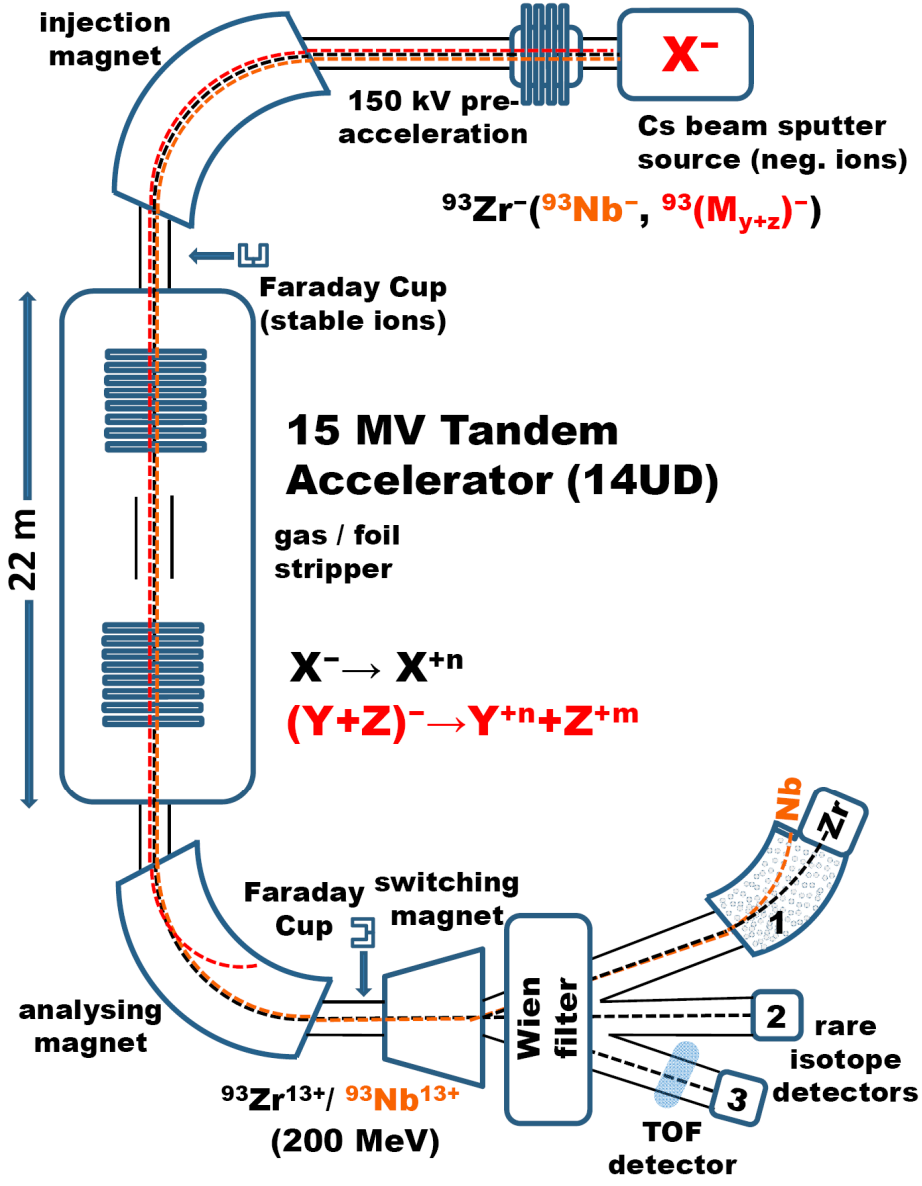
ANU's AMS facility

- *AMS determines isotope ratios – atom counting technique*
- *$^{14}C/^{12}C$ – radiocarbon dating*
- *highest sensitivity: 10^{-12} – 10^{-16}*
- *no isobaric background (<-> ICPMS) (molecules are completely destroyed)*
- *isotopic background clearly identified*



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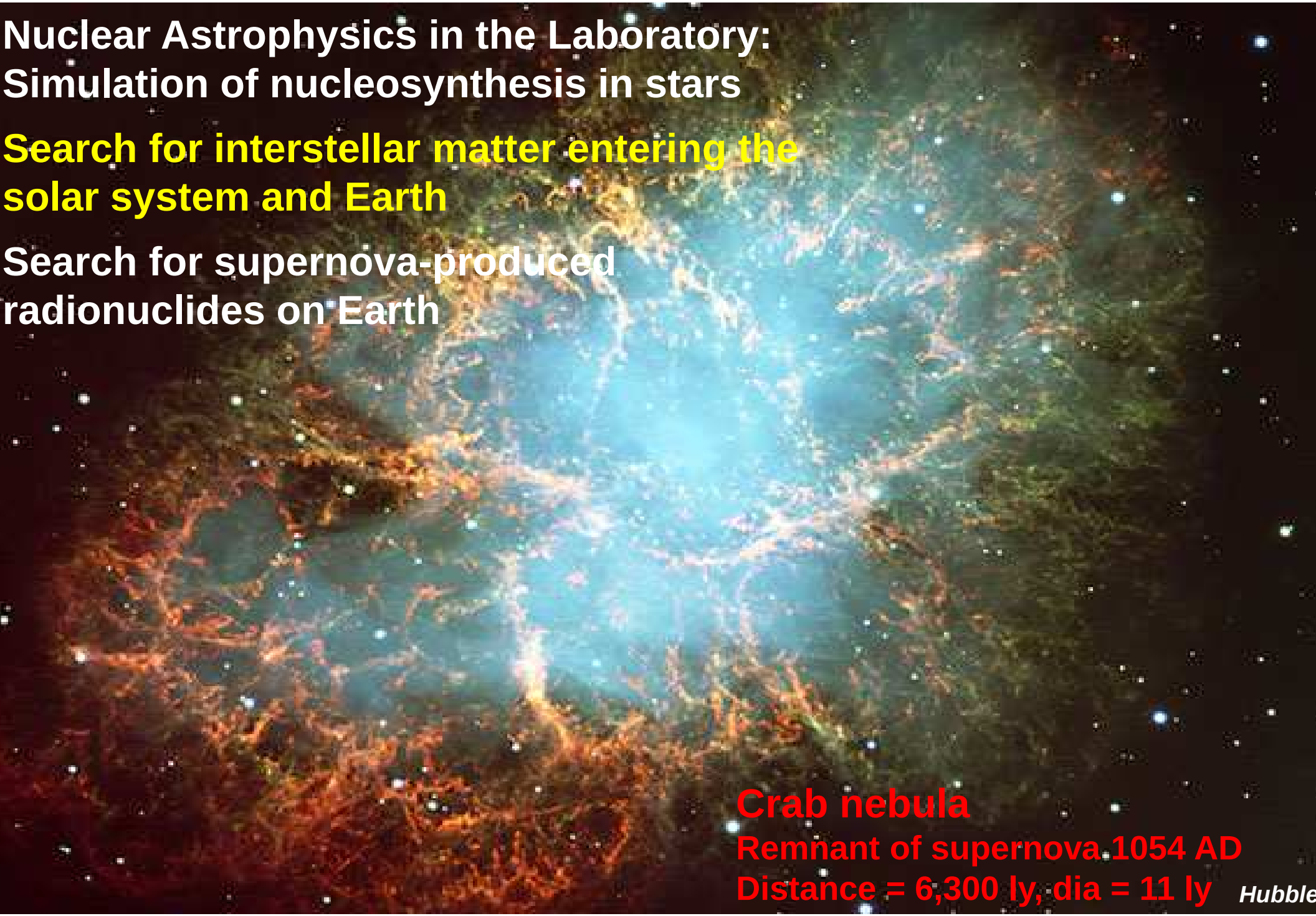
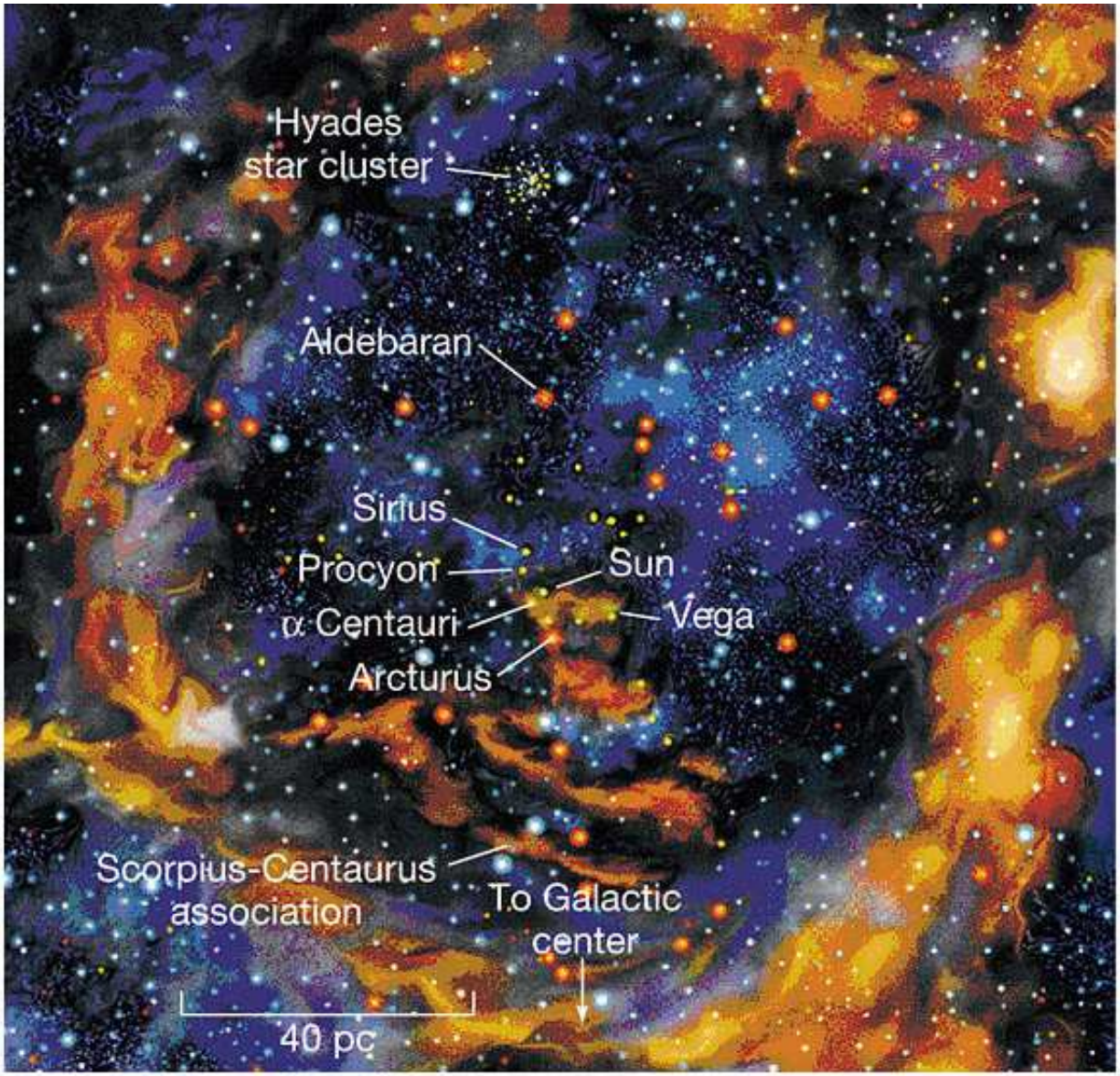

Nuclear Astrophysics in the Laboratory:
Simulation of nucleosynthesis in stars
Search for interstellar matter entering the solar system and Earth
Search for supernova-produced radionuclides on Earth
Crab nebula
Remnant of supernova 1054 AD
Distance = 6,300 ly, dia = 11 ly
Hubble

Extraterrestrial Radionuclides on Earth



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Can we find isotopic fingerprints of the ISM (interstellar medium) in terrestrial archives?

Radionuclides contain time information as they can serve as radioactive clocks!

^{60}Fe: $t_{1/2}$ = 1.5 / 2.6 / ?3? Ma

Ni 55 209 ms β+ 7.7… γ (2919; 2976; 3303)	Ni 56 6.075 d ε; no β+ γ 158; 812; 750; 480; 270…	Ni 57 36.0 h ε β+ 0.8… γ 1378; 1920; 127…	Ni 58 68.0769 σ 4.6 σn, α <0.00003	Ni 59 7.5 · 10⁴ a ε; β+ … no γ ; σ 77.7 σn, α 14; σn, p 2 σabs 92	Ni 60 26.2231 σ 2.9	Ni 61 1.1399 σ 2.5 σn, α 0.00003	Ni 62 3.6345 σ 15	Ni 63 100 a β− 0.07 no γ σ 20
Co 54 1.48 m | 193.2 ms β+ 4.3 γ 411; 1130; 1407 β+ 7.3… γ (2561)	Co 55 17.54 h β+ 1.5… γ 931; 477; 1409…	Co 56 77.26 d ε; β+ 1.5… γ 847; 1238; 2598; 1771; 1038…	Co 57 271.79 d ε γ 122; 136; 14	Co 58 8.94 h | 70.86 d Iγ (25) e⁻ σ 140000 ε β+ 0.5; 1.3 γ 811 σ 1900	Co 59 100 σ 20.7 + 16.5	Co 60 10.5 m | 5.272 a Iγ 59 e⁻ β⁻… γ (1332…) σ 58 β⁻ 0.3; 1.5… γ 1332; 1173… σ 2.0	Co 61 1.65 h β⁻ 1.2… γ 67; 909…	Co 62 14.0 m | 1.5 m β⁻ 2.9… γ 1173; 1163; 2003… β⁻ 4.1… γ 1173; 2302; 1129…
Fe 53 2.5 m | 8.51 m Iγ 701; 1328; 1011; 2340… β+ 2.6… γ 378; (1620…)	Fe 54 5.845 σ 2.3 σn, α 1E-5	Fe 55 2.73 a ε no γ σ 13 σn, α 0.01	Fe 56 91.754 σ 2.8	Fe 57 2.119 σ 1.4	Fe 58 0.282 σ 1.3	Fe 59 44.503 d β⁻ 0.5; 1.6… γ 1099; 1292… σ 13	Fe 60 1.5 · 10⁶ a β⁻ 0.1 m	Fe 61 6.0 m β⁻ 2.6; 2.8… γ 1205; 1027; 298…
Mn 52 21 m | 5.6 d β+ 2.6… γ 1434… Iγ 378 ε β+ 0.6… γ 1434; 936; 744…	Mn 53 3.7 · 10⁶ a ε no γ σ 70	Mn 54 312.2 d ε γ 835 σ <10	Mn 55 100 σ 13.3	Mn 56 2.58 h β⁻ 2.9… γ 847; 1811; 2113…	Mn 57 1.5 m β⁻ 2.6… γ 14; 122; 692…	Mn 58 65.3 s | 3.0 s β⁻ 3.9… γ 811; 1323… Iγ 72; e⁻ β⁻ 6.1… γ 1447; 2433…	Mn 59 4.6 s β⁻ 4.4; 4.8… γ 726; 473; 571…	Mn 60 1.77 s | 0.28 s β⁻ 5.7; 6.1… γ 823; 1969… Iγ 272 β⁻ 8.2… γ 823; 1150; 1532
Cr 51 27.70 d ε γ 320 σ <10	Cr 52 83.789 σ 0.8	Cr 53 9.501 σ 18	Cr 54 2.365 σ 0.36	Cr 55 3.50 m β⁻ 2.6 γ (1528…)	Cr 56 5.9 m β⁻ 1.5 γ 83; 26	Cr 57 21.1 s β⁻ 5.1… γ 83; 850; 1752; 1535…	Cr 58 7.0 s β⁻ γ 683; 126; 290; 520… m	Cr 59 1.05 s β⁻ γ 1238; 1900; 112; 663…

Half-life value of ^{60}Fe

- nucleosynthesis in our galaxy is observed through γ-rays (^{26}Al, ^{60}Fe, …)
- proof of ongoing nucleosynthesis (on time scale of Myrs)
- origin in massive stars: production / destruction complex: ^{58}Fe(n,γ)^{59}Fe(n,γ)^{60}Fe(n,γ)^{61}Fe
- early solar system: decay product ^{60}Ni – surplus in meteorites; heat source
- late SN injection of fresh ^{60}Fe into protoplanetary disk (Bizzarro et al. 2007, Wasserburg et al. 2006)
- extraterrestrial ^{60}Fe was found in ocean crust material (recent SN origin?)

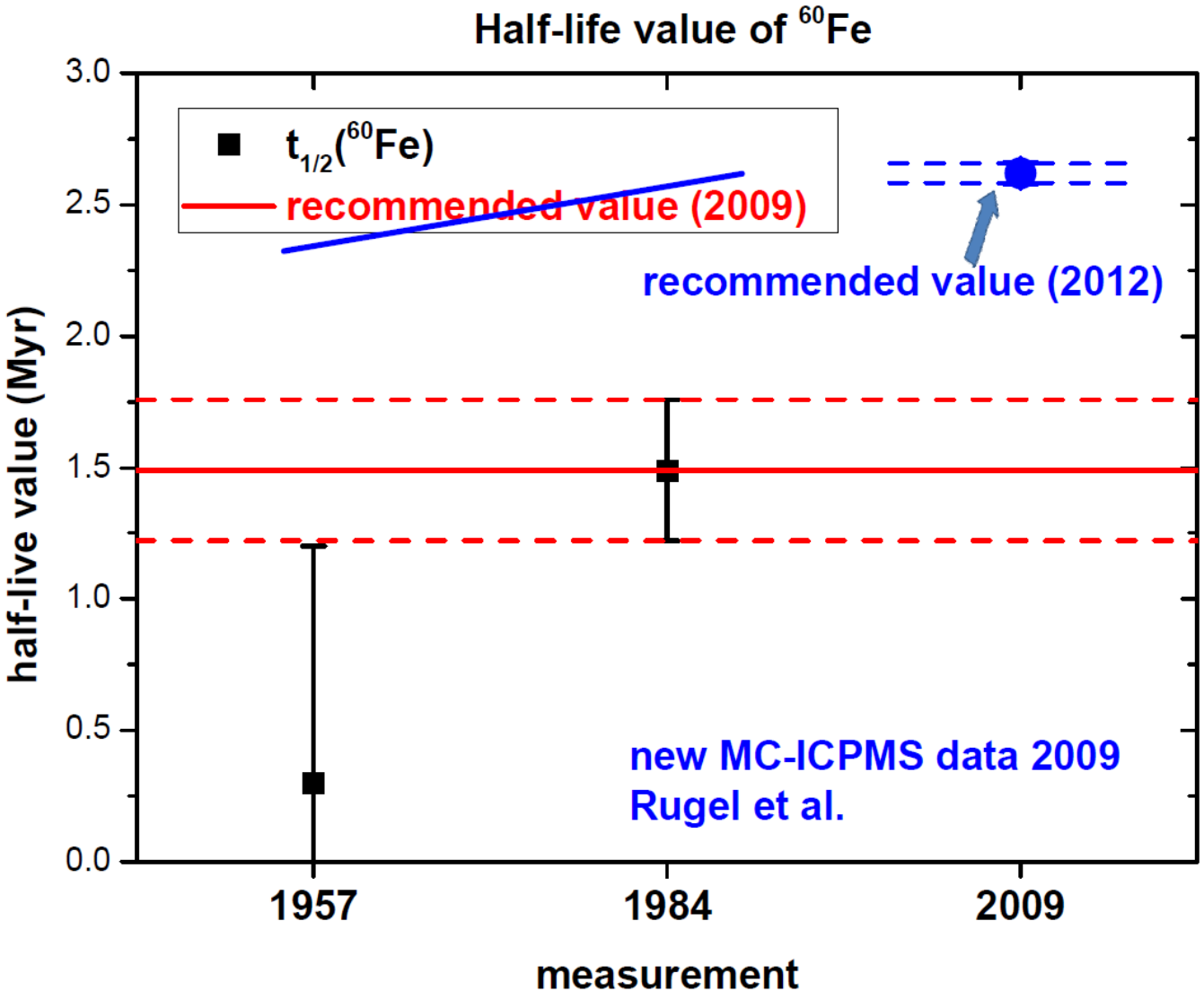
New Measurement of the ^{60}Fe Half-Life

G. Rugel, T. Faestermann, K. Knie,* G. Korschinek, and M. Poutivtsev
Technische Universität München, D-85748 Garching, Germany

D. Schumann, N. Kivel, I. Günther-Leopold, R. Weinreich, and M. Wohlmuther
Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland
(Received 25 March 2009; published 14 August 2009)

PRL **103,** 072502 (2009) PHYSICAL REVIEW LETTERS week ending 14 AUGUST 2009

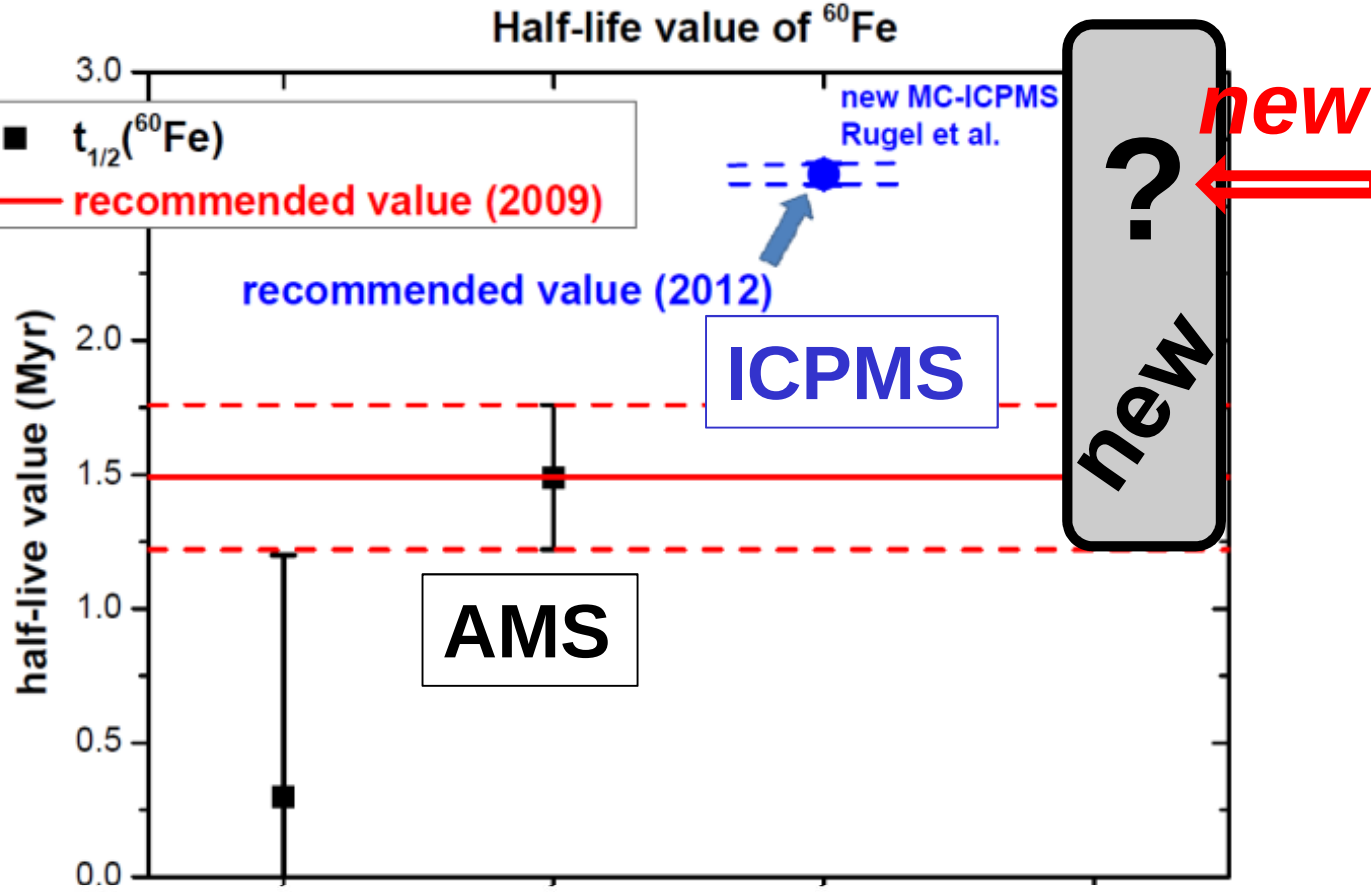
Half-life value of ^{60}Fe



Half-life value of ^{60}Fe:

$$A_{60} = \lambda_{60} \frac{^{60}Fe}{^{55}Fe} \cdot {}^{55}Fe$$

AMS

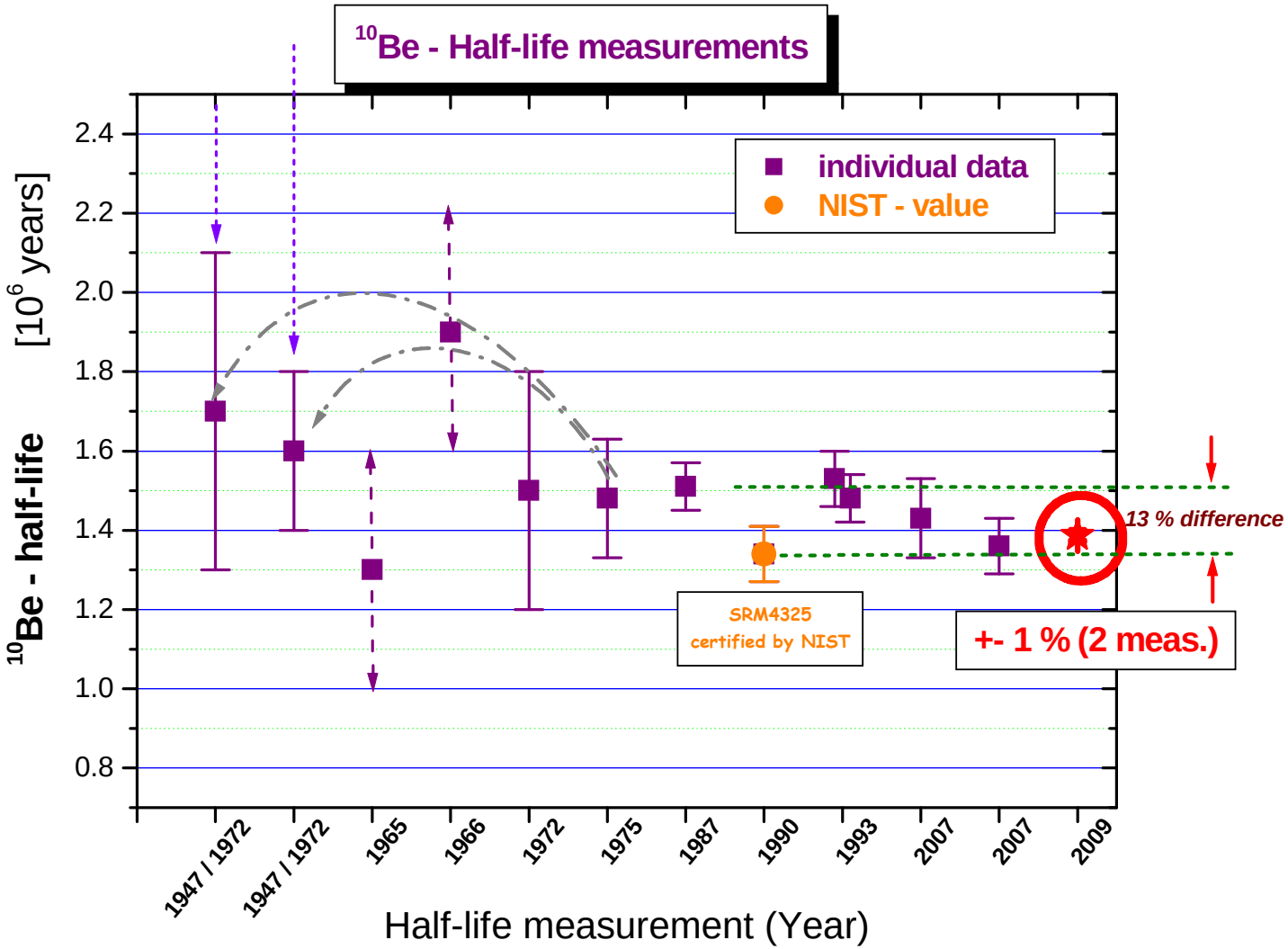


PRL 114, 041101 (2015) PHYSICAL REVIEW LETTERS week ending 30 JANUARY 2015

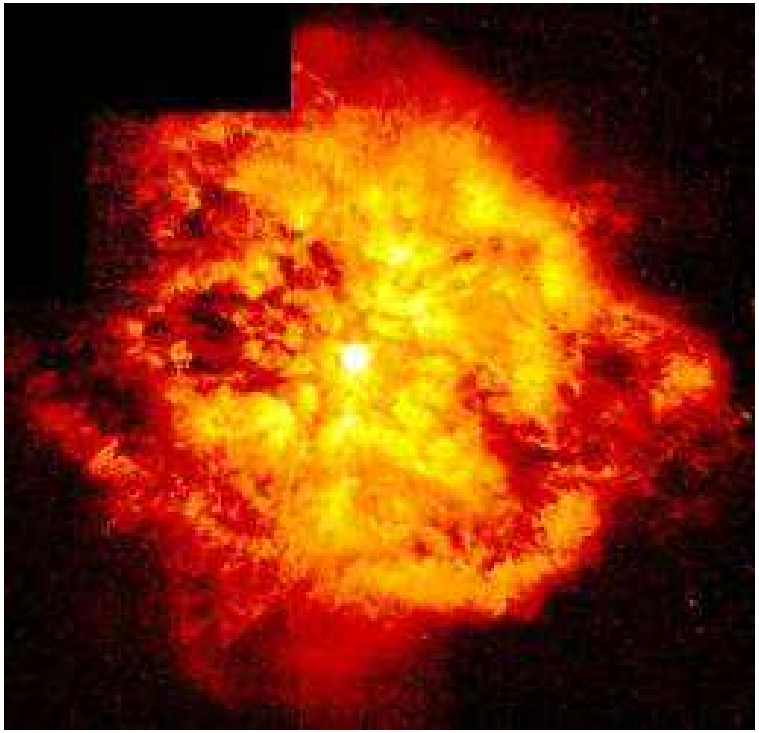
Settling the Half-Life of ^{60}Fe: Fundamental for a Versatile Astrophysical Chronometer

A. Wallner,[1,2] M. Bichler,[3] K. Buczak,[2,3] R. Dressler,[4] L. K. Fifield,[1] D. Schumann,[4] J. H. Sterba,[3] S. G. Tims,[1] G. Wallner,[5] and W. Kutschera[2]

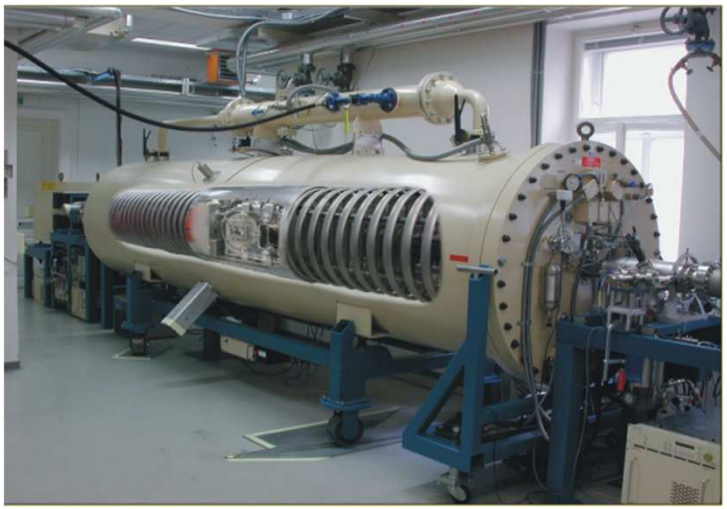
^{10}Be: Half-life and standards



(II) Nucleosynthesis in the laboratory



(I) sample exposure



(II) AMS: analysis

stellar nucleosynthesis → laboratory studies

e.g. activation

Activation (I) → AMS-Measurement (II)

fusion / fission / ADS products / nuclear astrophysics

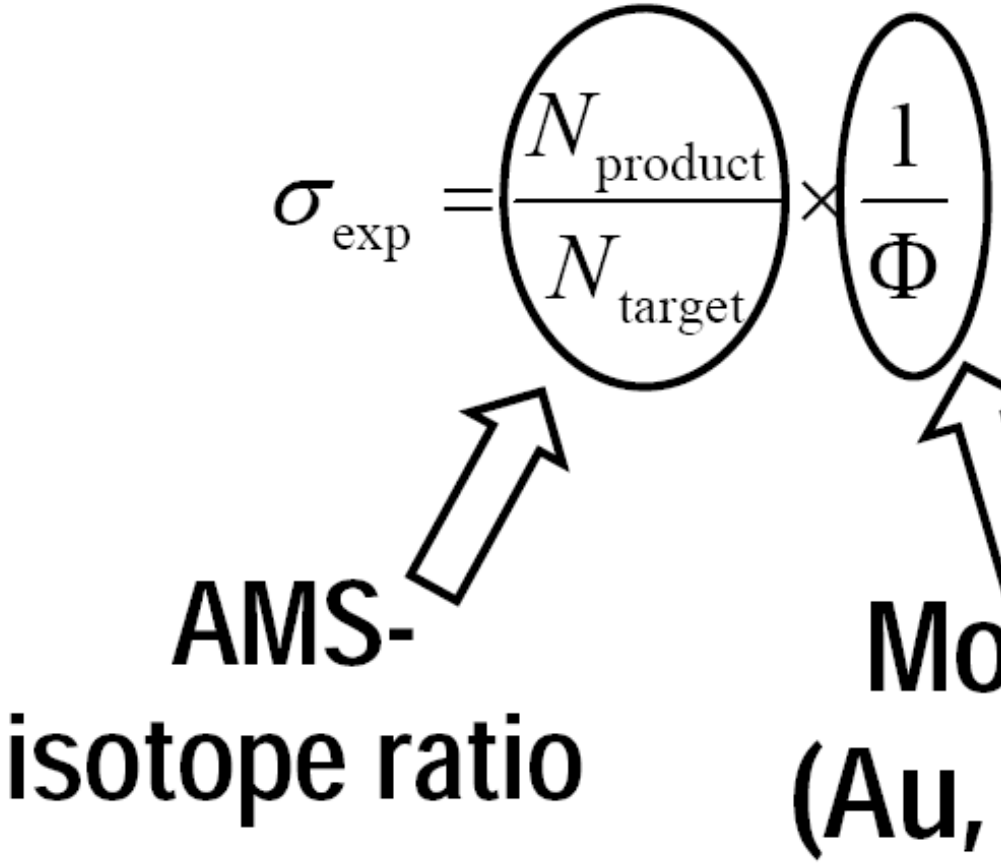
Recent neutron irradiations:

- **IRMM Geel:** 7 MV Van de Graaff: ($T(p,n)^3He$: E_n = 0.5 – 2.5 MeV)
- **IRMM Geel:** 7 MV Van de Graaff: ($D(d,n)^3He$: E_n = 2 – 5 MeV)
- **KIT Karlsruhe:** 3.7 MV Van de Graaff: $^7Li(p,n)^7Be$: 25 keV Maxwell-

II) atom counting of the reaction products using AMS

- **TU Dresden**
- **Atominstitut Vienna:** research reactor, thermal neutrons
- **IKI Budapest:** research reactor, thermal neutrons; cold neutrons

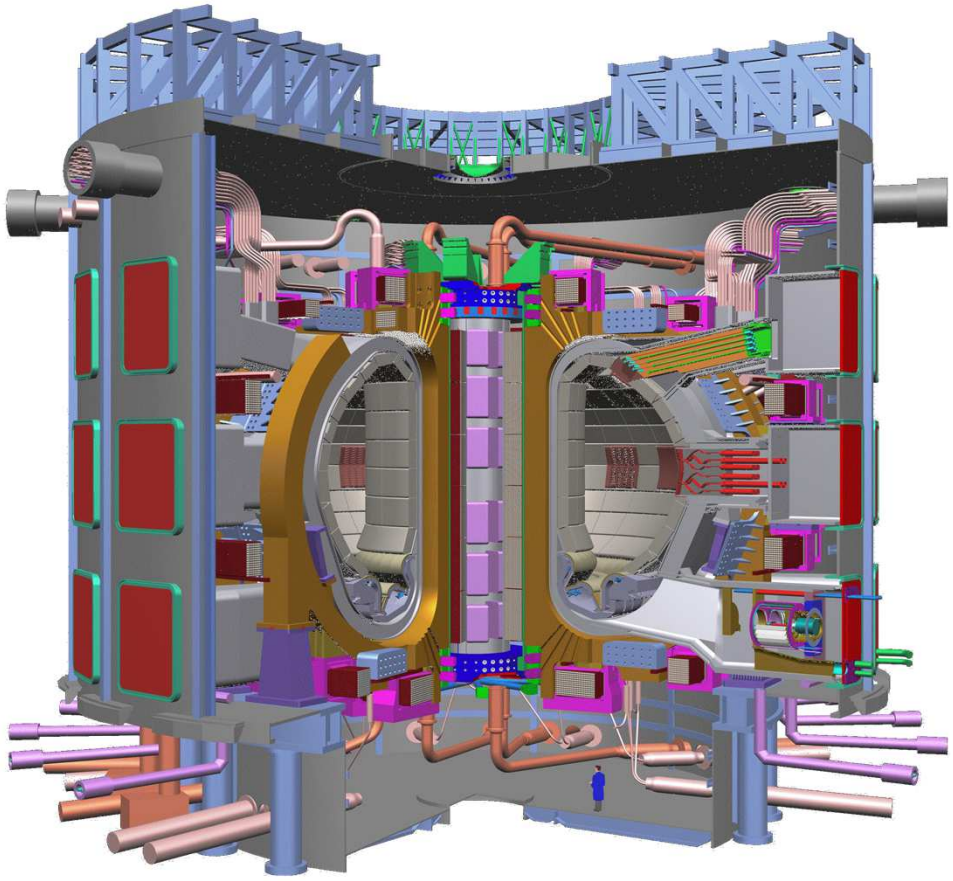
How to derive cross sections?



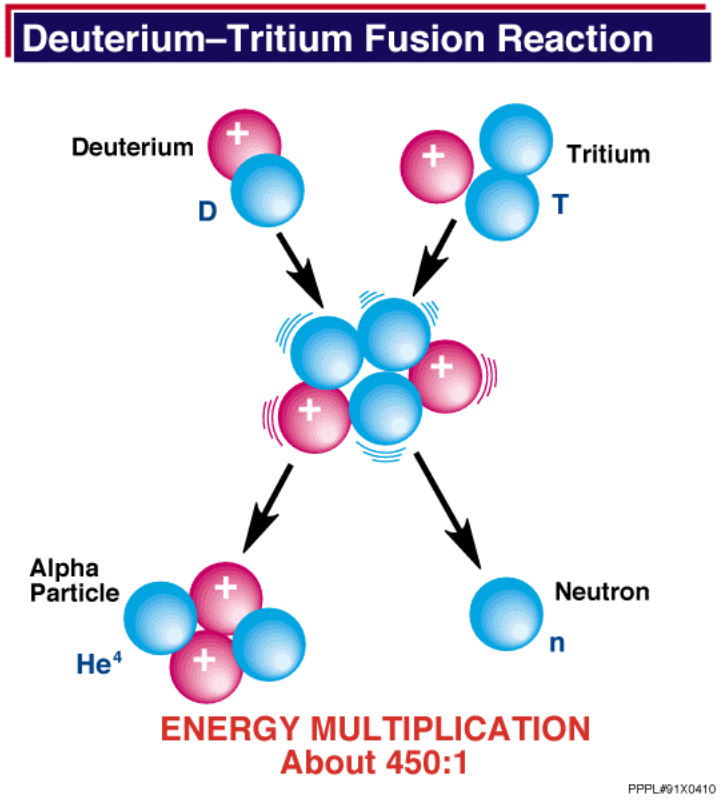
U

AMS monitor reaction (Au)

Production of long-lived nuclides in a fusion environment



ITER: DT-fusion



LONG-TERM RADIOACTIVE WASTE FROM FUSION REACTORS:

S. FETTER, E.T. CHENG and F.M. MANN

coincides with AMS nuclides!

Table 2

Specific activity limits for class "C" disposal of activated metal radionuclides with 5 y < $t_{½}$ < 10^{12} y[a]

Radio-nuclide	Halflife	SAL (Ci/m^3)[b]	Ot
H-3	12.3 y	TMSA[c]	TMSA
Be-10	1.6 My	5.E+03	7,0
C-14	5.7 ky	6.E+02 - 6.E+03	80 (
Al-26	720. ky	9.E-02	
Si-32	104. y	6.E+02 - 4.E+03	60
Cl-36	301. ky	1.E+01 - 1.E+02	
Ar-39	269. y	2.E+04	
Ar-42	33. y	2.E+04	0.8
K-40	1.3 Gy	2.E+00	
Ca-41	103. ky	1.E+04 - 3.E+04	
Ti-44	47. y	2.E+02	0.0
Mn-53	3.7 My	TMSA	60
Fe-60	100. ky	1.E-01	0.0
Co-60	5.3 y	3.E+08	TMSA
Ni-59	75. ky	9.E+02	220
Ni-63	100. y	7.E+05 - 7.E+06	7,000
Se-79	65. ky	5.E+01 - 5.E+02	
Kr-81	210. ky	3.E+01	
Kr-85	10.7 y	TMSA	
Rb-87	48. Gy	TMSA	
Sr-90	28.5 y	8.E+05 - 7.E+06	70,000
Zr-93	1.5 My	TMSA	20
Nb-91	680. y	2.E+02	
Nb-92	36. My	2.E-01	
Nb-93m	13.6 My	TMSA	
Nb-94	20. ky	2.E-01	0.2
Mo-93	3.5 ky	4.E+03	
Tc-97	2.6 My	4.E-01 - 4.E+00	
Tc-98	4.2 My	1.E-02 - 8.E-02	

Long-term radioactivity – e.g. Eurofer

K. Seidel et al., 2003

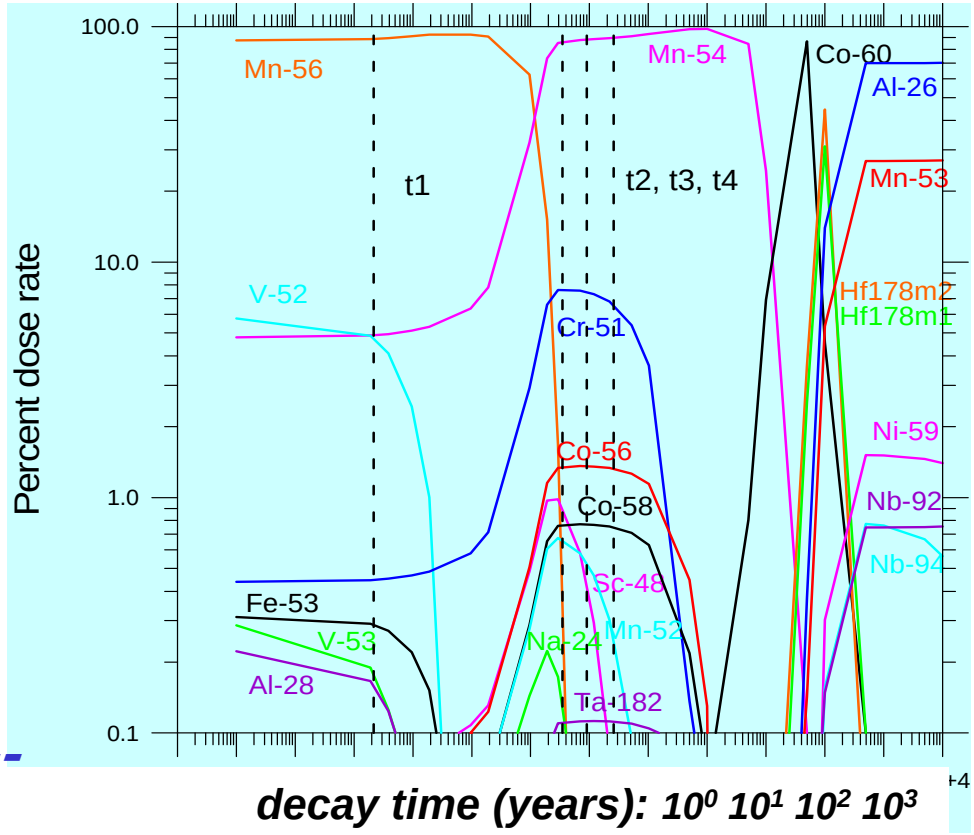
dominant nuclides after 1000 years:

^{26}Al: 70% of the dose rate produced by ^{27}Al(n,2n)

^{53}Mn: 27% of the dose rate produced by ^{54}Fe(n,np+d).

-- small Al content of the Eurofer sample --

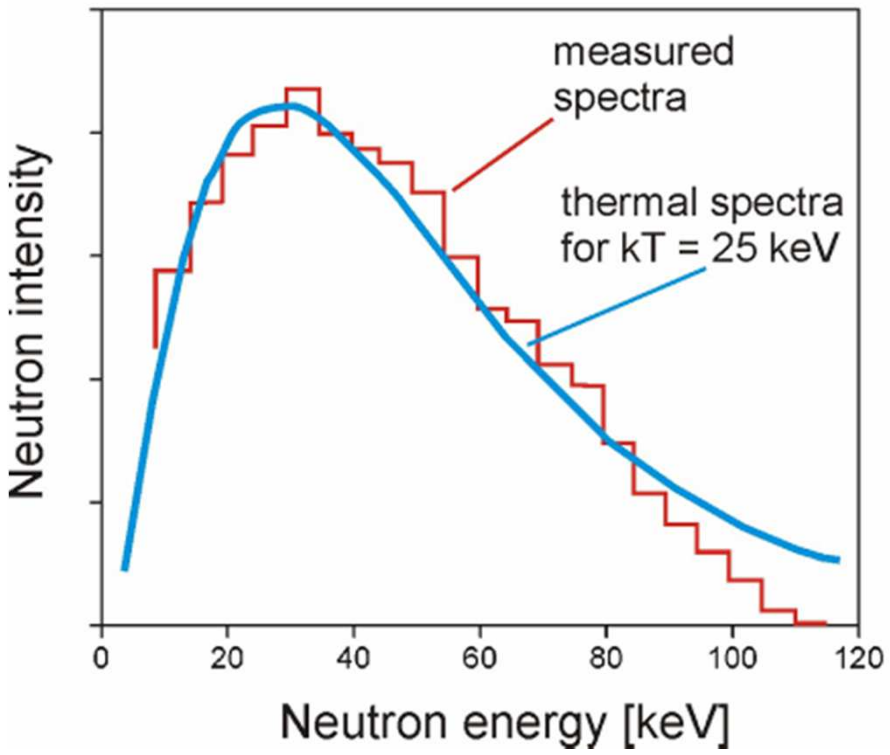
Uncertainties of calculated values estimated:
30% for ^{26}Al –> new data
60% for ^{53}Mn –> new measurements



Radioactivity mainly by 14 MeV neutrons and by thermal neutrons

Stellar reactions →
Maxwellian averaged cross sections

e.g. KIT(F. Käppeler et al.): neutr



produce therm

red giant: T_6 =

→ measur
directly

+ 120/180/5

fluence

$^{9}Be(n,\gamma)^{10}Be$
$^{13}C(n,\gamma)^{14}C$
$^{14}N(n,p)^{14}C$
$^{35}Cl(n,\gamma)^{36}Cl$
$^{40}Ca(n,\gamma)^{41}Ca$
$^{54}Fe(n,\gamma)^{55}Fe$
$^{235}U(n,\gamma)^{236}U$
$^{238}U(n,\gamma)^{239}U$

A novel method for studying neutron-induced
reactions

in collaboration with:

- KIT (Karlsruhe): F. Käppeler, I. Dillmann
- IRMM / Geel: A. Plompen, A. Krasa
- IKI Budapest: T. Belgya, L. Szentmiklosi
- TU Vienna: M. Bichler
- TSL Uppsala: A. Prokofiev et al.
- ANU (Canberra): L.K. Fifield, S. Tims, M. Srncik
- VERA (Vienna): K. Buzcak, F. Quinto, P. Steier, S. Winkler, C. Lederer
- ETH Zurich: M. Christl, J. Lachner
- ANSTO: M. Hotchkis
- Univ. Heidelberg: J. Lippold
- IAEA: R. Capote , V. Semkova

AMS

neutrons

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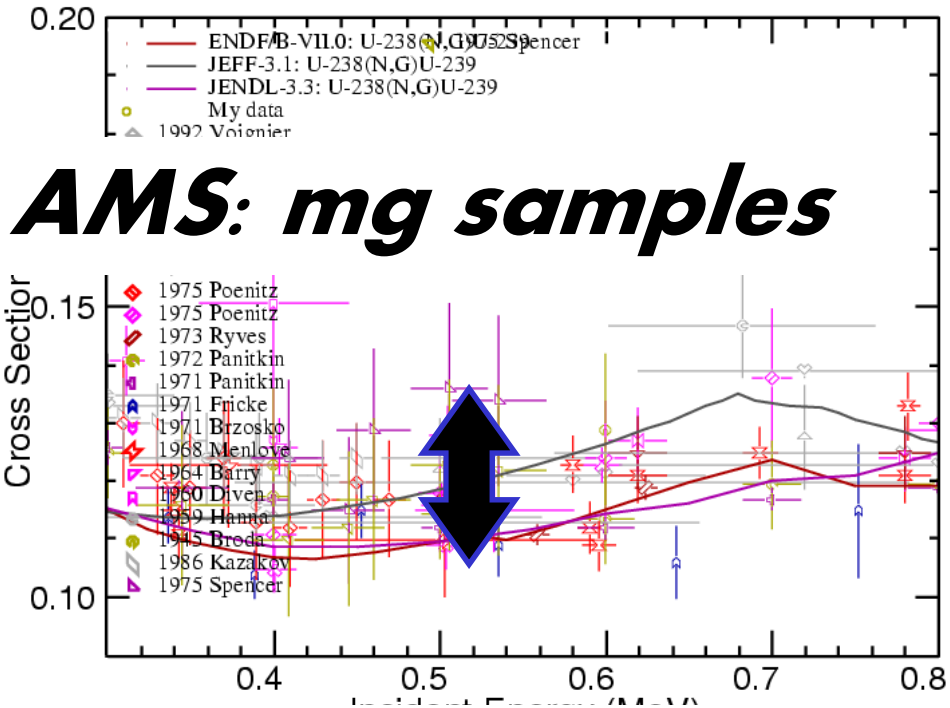
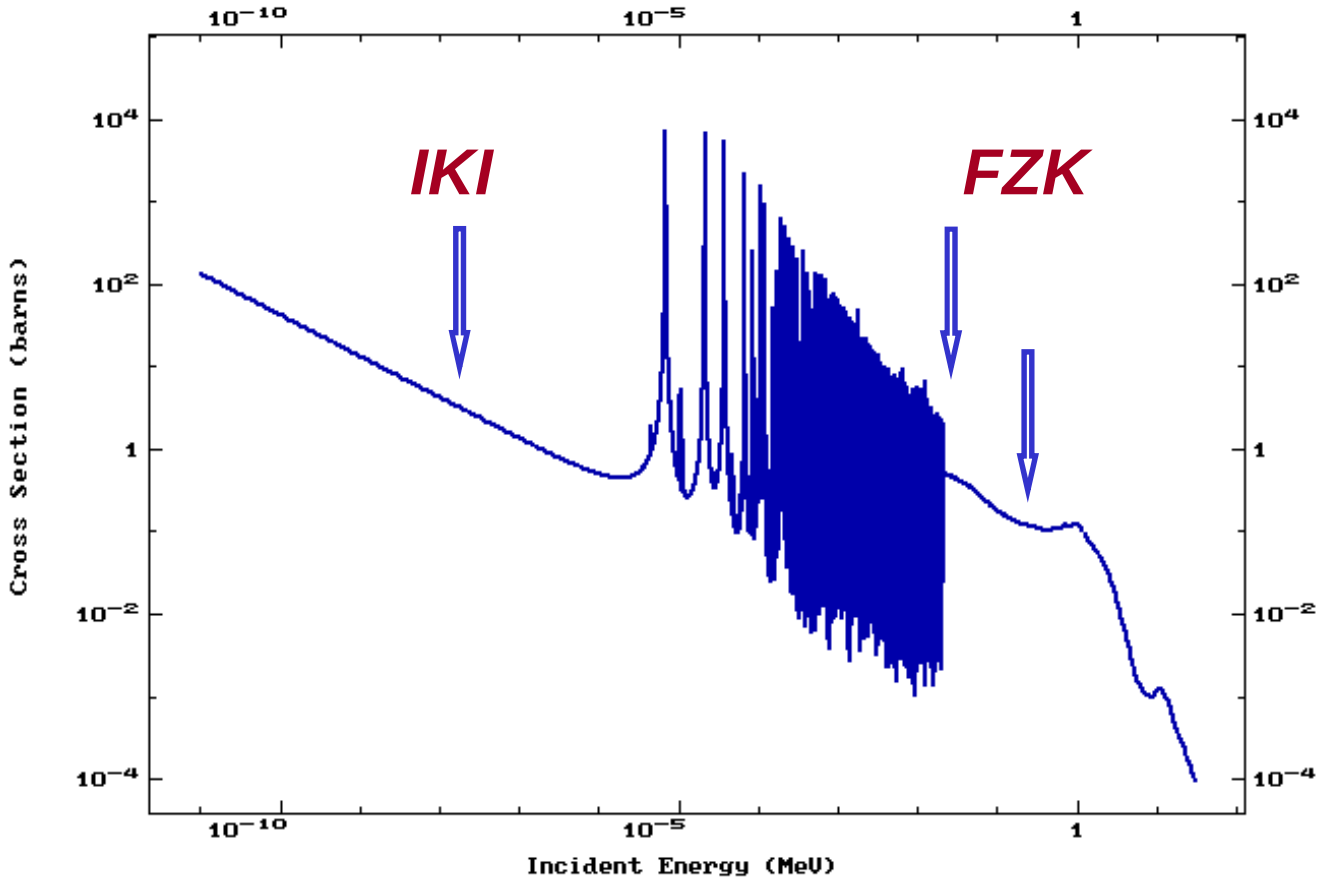
Motivation: $^{235,238}U(n,\gamma)/^{232}Th(n,\gamma)$: E_n > thermal

- ratio of capture to fission of the fissile isotopes
- existing data via TOF and detections of prompt γ-rays;
- multiple scattering corrections due to large sample size in previous exp.? (e.g. 2 gram)
- HPRL (NEA) listed = highest priority
- large discrepancies above thermal E_n
- k_{eff} in reactor technology: even 5-10% discrepancy in cross section too much
- very few exp. data above thermal energies
- activation + AMS is an independent method; no influence from fission channel



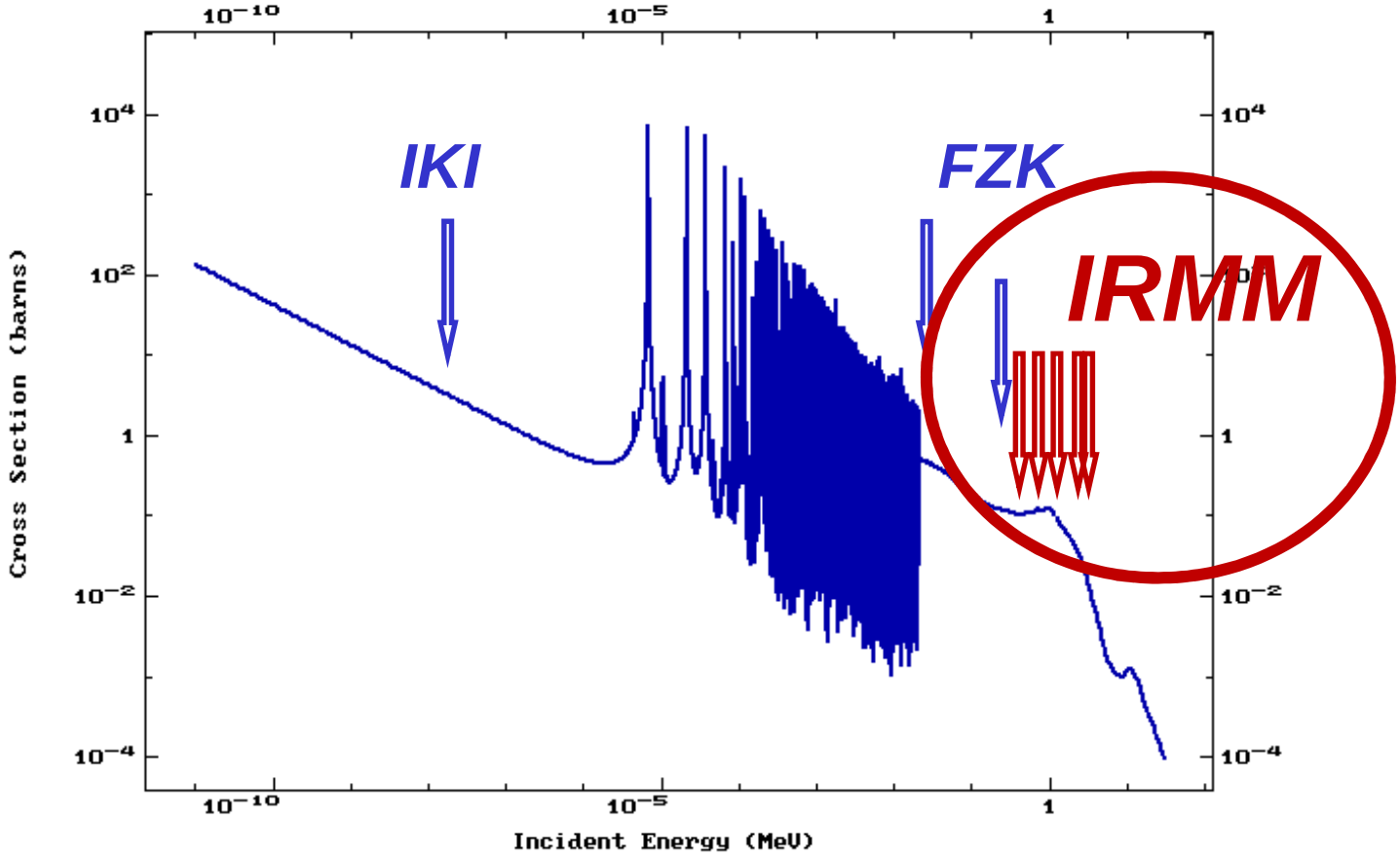
- *new method* → *does not rely on previous drawbacks*
  → *simple direct method!*

$^{238}U(n,\gamma)^{239}U$ -> ^{239}Np -> ^{239}Pu (24 kyr)



$^{235}U(n,\gamma)^{236}U$ (23 Myr) - simultaneously

$^{238}U(n,\gamma)^{239}U$ -> ^{239}Np -> ^{239}Pu (24 kyr)



$^{232}Th(n,\gamma)^{233}Th$ -> ^{233}Pa -> ^{233}U (159 kyr)- simultaneously

New Precision Measurements of the $^{235}U(n, \gamma)$ Cross Section

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Los Alamos

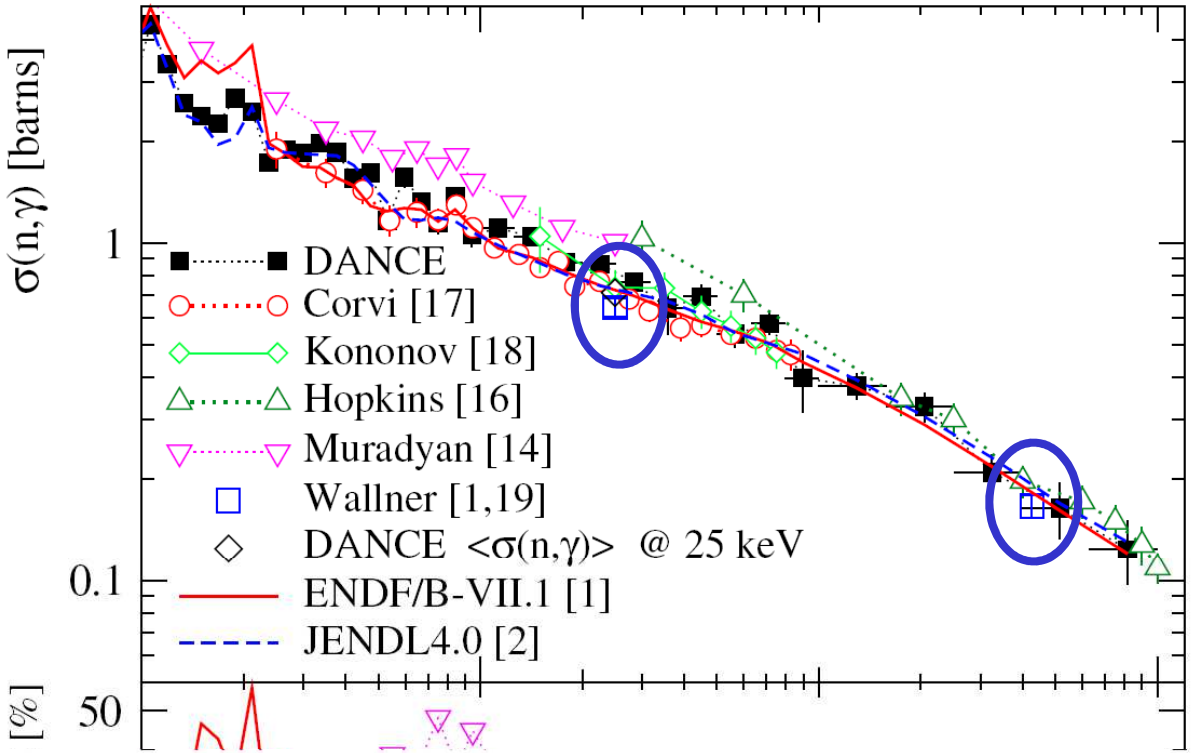
25 keV – Maxwell-Boltzmann:

JANDEL - 2012: $\sigma_{U\text{-}5} = 0.70 \pm 0.06$

ENDF/B-VII.1 $\sigma_{U\text{-}5} = 0.679$

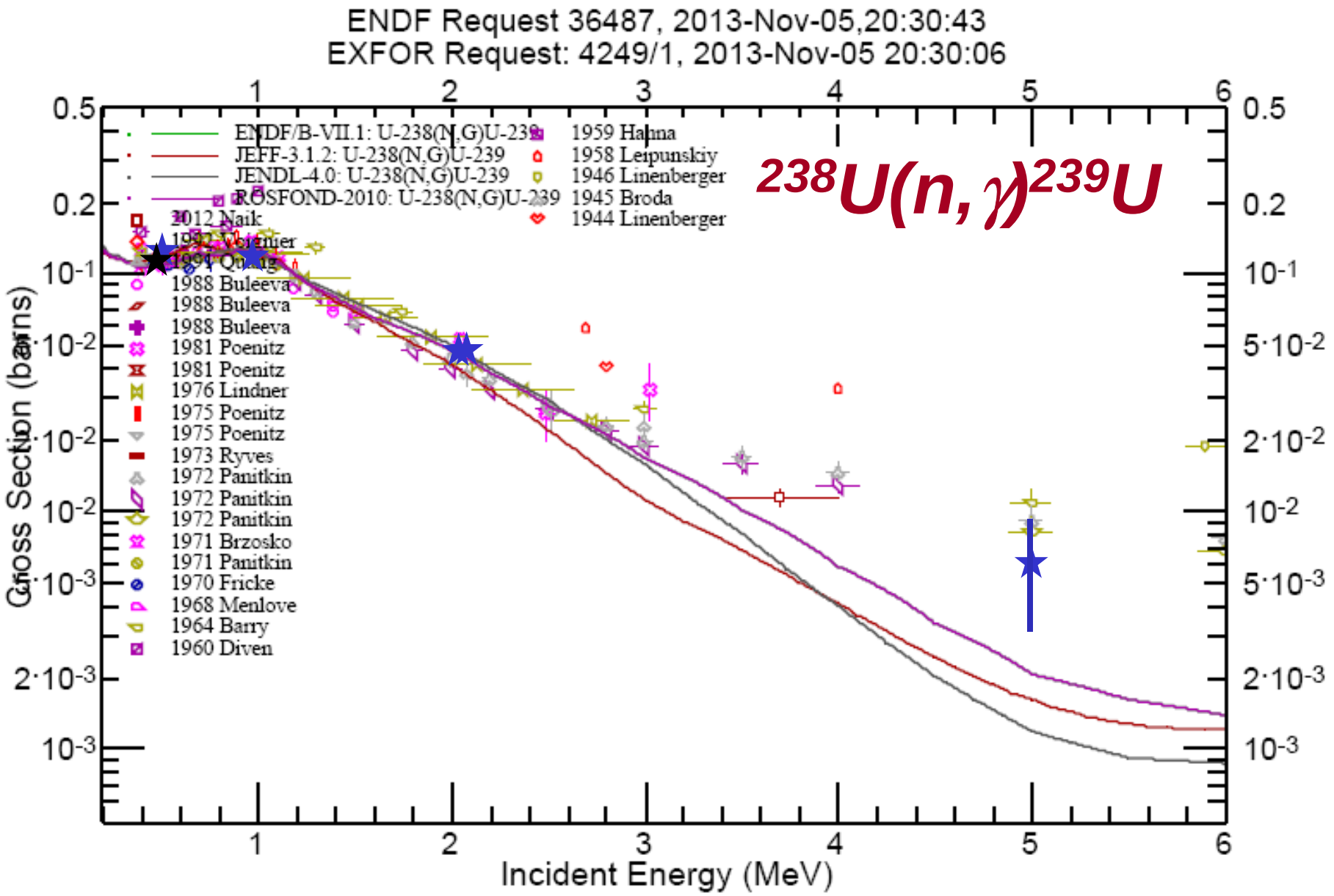
this work (AMS): $\sigma_{U\text{-}5} = 0.646 \pm 0.038$

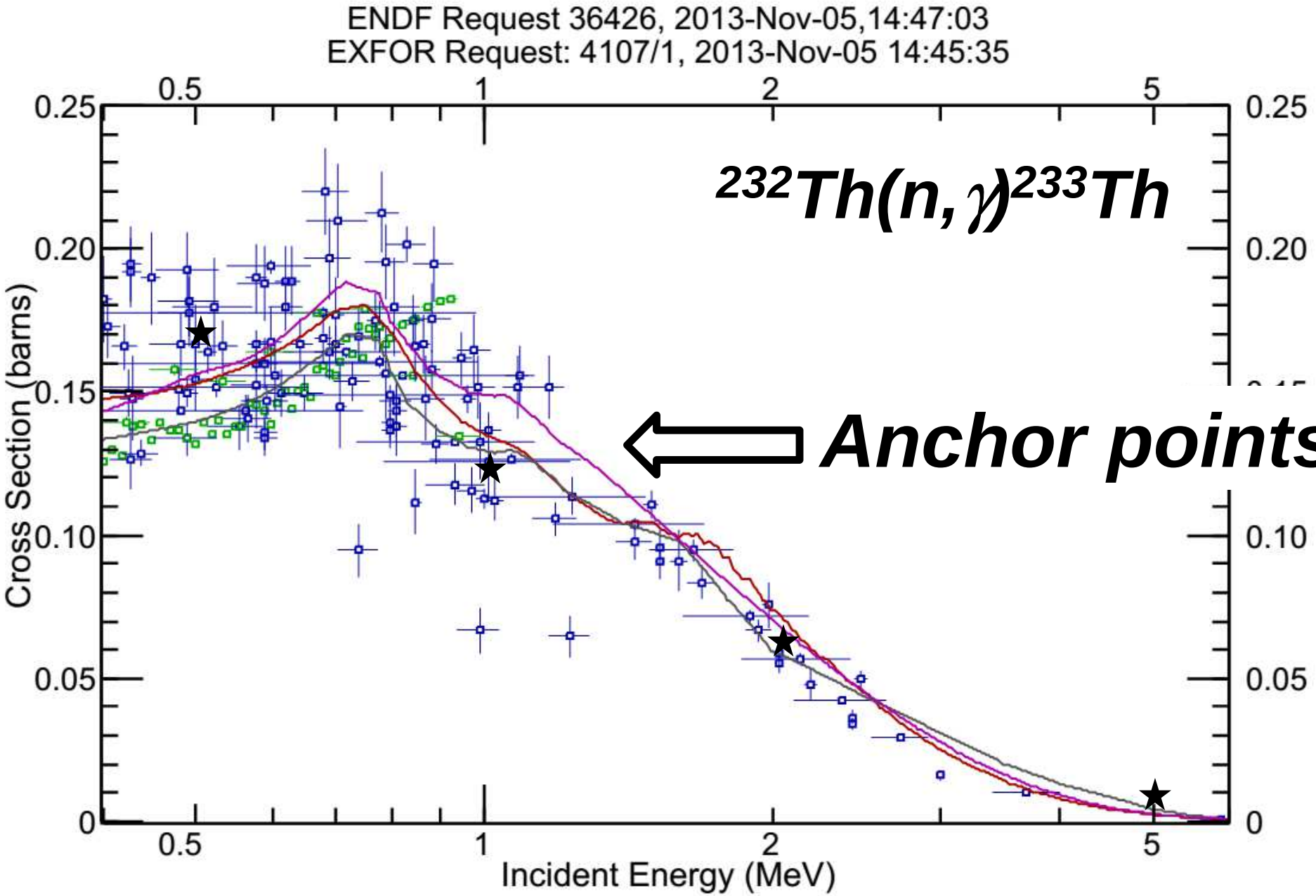
AMS vs JANDEL: $\sigma_{U\text{-}5}$ ***(25 keV) = 0.92 ± 0.10***



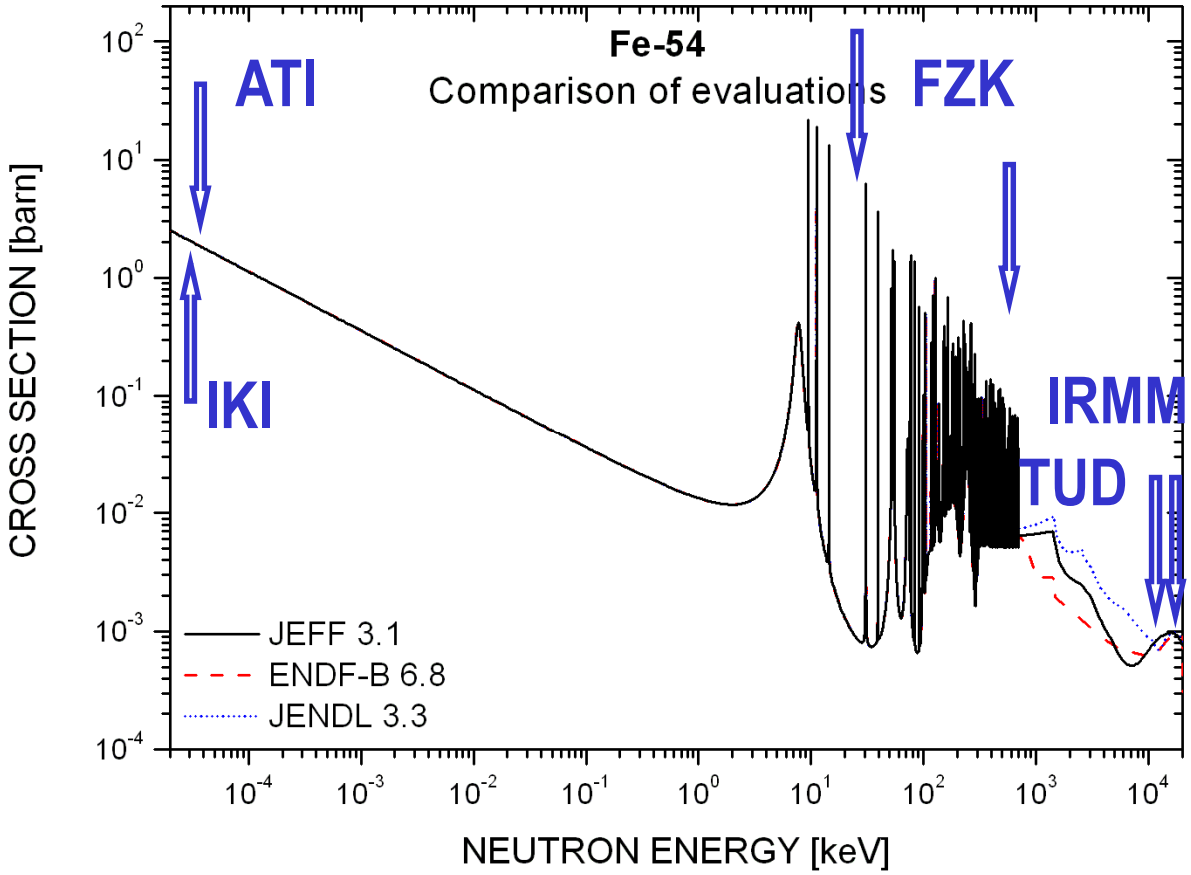
Novel Method to Study Neutron Capture of ^{235}U and ^{238}U Simultaneously at keV Energies

A. Wallner,[1,2,*] T. Belgya,[3] M. Bichler,[4] K. Buczak,[2,4] I. Dillmann,[5,6] F. Käppeler,[5] C. Lederer,[2,7,†] A. Mengoni,[8] F. Quinto,[2,‡] P. Steier,[2] and L. Szentmiklosi[3]


ENDF Request 36487, 2013-Nov-05,20:30:43
EXFOR Request: 4249/1, 2013-Nov-05 20:30:06
238U(n,γ)239U
ENDF/B-VII.1: U-238(N,G)U-239
JEFF-3.1.2: U-238(N,G)U-239
JENDL-4.0: U-238(N,G)U-239
ROSFOND-2010: U-238(N,G)U-239
2012 Naik
1992 Voronier
1988 Buleeva
1988 Buleeva
1988 Buleeva
1981 Poenitz
1981 Poenitz
1976 Lindner
1975 Poenitz
1975 Poenitz
1973 Ryves
1972 Panitkin
1972 Panitkin
1972 Panitkin
1971 Brzosko
1971 Panitkin
1970 Fricke
1968 Menlove
1964 Barry
1960 Diven
1959 Hanna
1958 Leipunskiy
1946 Linenberger
1945 Broda
1944 Linenberger
Cross Section (barns)
Incident Energy (MeV)


ENDF Request 36426, 2013-Nov-05,14:47:03
EXFOR Request: 4107/1, 2013-Nov-05 14:45:35
0.5
1
2
5
0.25
0.20
0.15
0.10
0.05
0
Cross Section (barns)
Incident Energy (MeV)
$^{232}Th(n,\gamma)^{233}Th$
⟸ Anchor points!

$^{54}Fe(n,\gamma)^{55}Fe$ ($t_{1/2}$ = 2.75 a) - AMS data

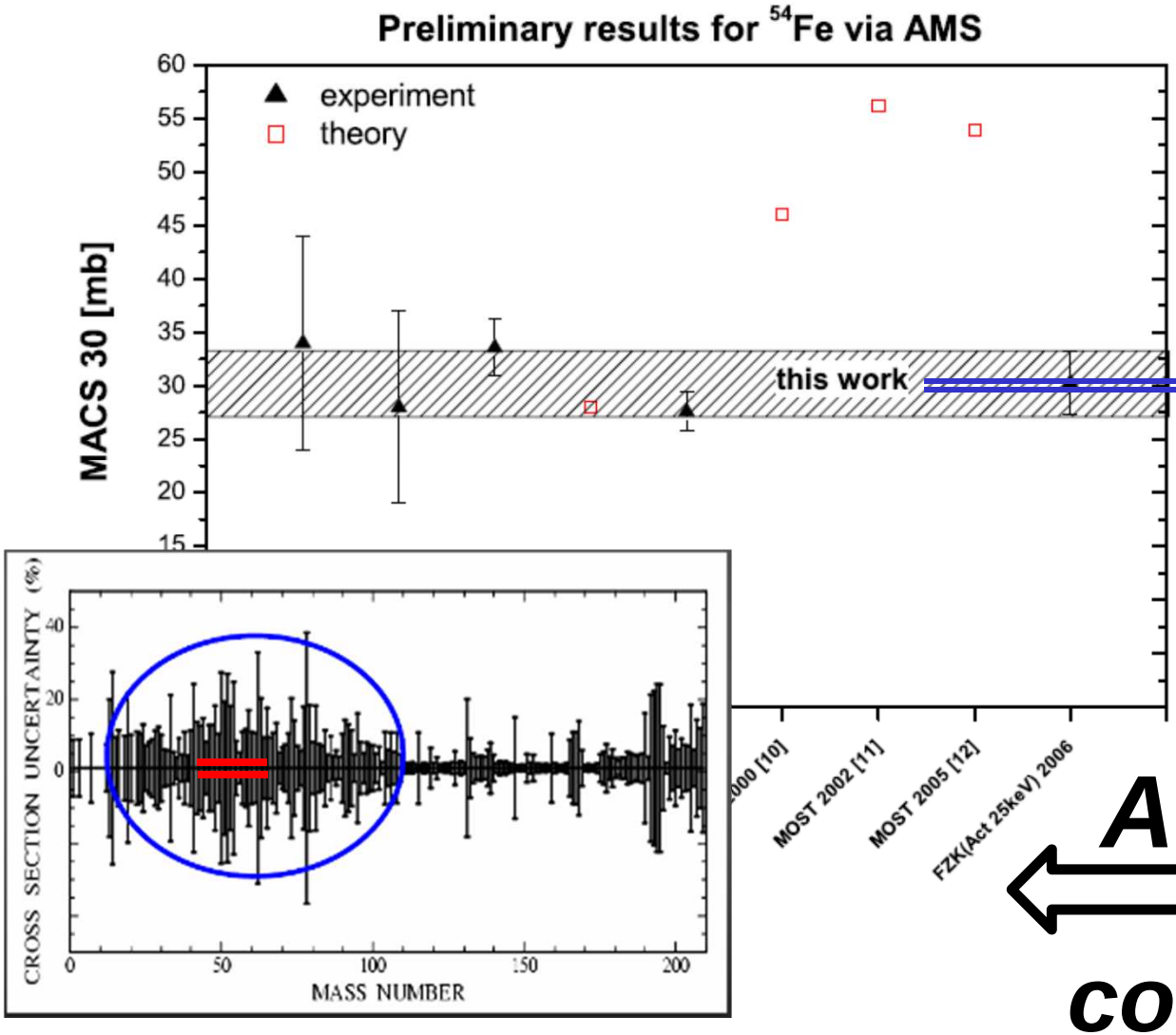


Neutron activations:

→ cold, thermal, MB 25 keV, 500 keV, 14 MeV + ...

→ quasimonoenergetic and broad E-distributions (MB)

thermal value: (2.25 +-0.18) barn (+- 8 %)



stellar: 30 keV

new AMS data

$^{54}Fe(n,\gamma)^{55}Fe$

Anchor point!

complements TOF data

+ thermal value: (+- 2 %)

+ 25 & 500 keV + 14 MeV AMS finished

Pu, U in Environmental Samples

Motivation

- Pu & U isotopic composition: "fingerprint" of different sources (anthropogenic)

  i. atmospheric weapons testing

  ii. direct discharge of radioactive wastes

  iii. accidental release

  iv. minor sources: … low-level radioactive waste disposal, slight leakage from nuclear power plants

- ❑ erosion studies via Pu-spikes from 60ies (soil and sediment transport & movement)
- ❑ Nuclear forensics, uranium exploration, biological uptake of actinides, ocean currents, …

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

and: $^{238,241,242,244}Pu/^{239}Pu$, $^{236}U/^{238}U$ in the environment as sensitive markers

-- fingerprints --

A mahogany plantation near Darwin
– the result of 140 mm of rain in 24 hours
Plutonium-labelled soil in motion

Flood plume, Burdekin R., Nth. Queensland, Australia

Large rivers empty into the lagoon behind the Great Barrier Reef. What has been the effect of humans on rate of delivery of sediment? Pu gives rate of soil loss over past 50 years, ^{10}Be gives long term rate.

Conclusions

- AMS represents an independent method for studying nuclear reactions leading to long-lived radionuclides
- direct method: atom counting as isotope ratio; i.e. no sophisticated data reduction or corrections required
- not a universal method – AMS is used for some 25 radionuclides
- if it can be applied – usually very (the most) sensitive method
- can serve as an important tool to provide some anchor points
- New facilities are capable to produce precise data (0.5-2%) – thus precise standard material required; also more nuclear data required to utilise this potential.

Collaboration- Nuclear astrophysics:

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- IAEA (Vienna): A. Mengoni, R. Capote
- GAMS (TU Munich): T. Faestermann, G. Korschinek, K. Knie, M. Suter, G. Rugel
- Hebew Univ.(Jerusalem): M. Paul
- ETH Zurich: C. Vockenhuber
- MPI Mainz; FZD (Germany): U. Ott, S. Merchel
- Atominstitut (Vienna): M. Bichler

in collaboration (nuclear physics) with:

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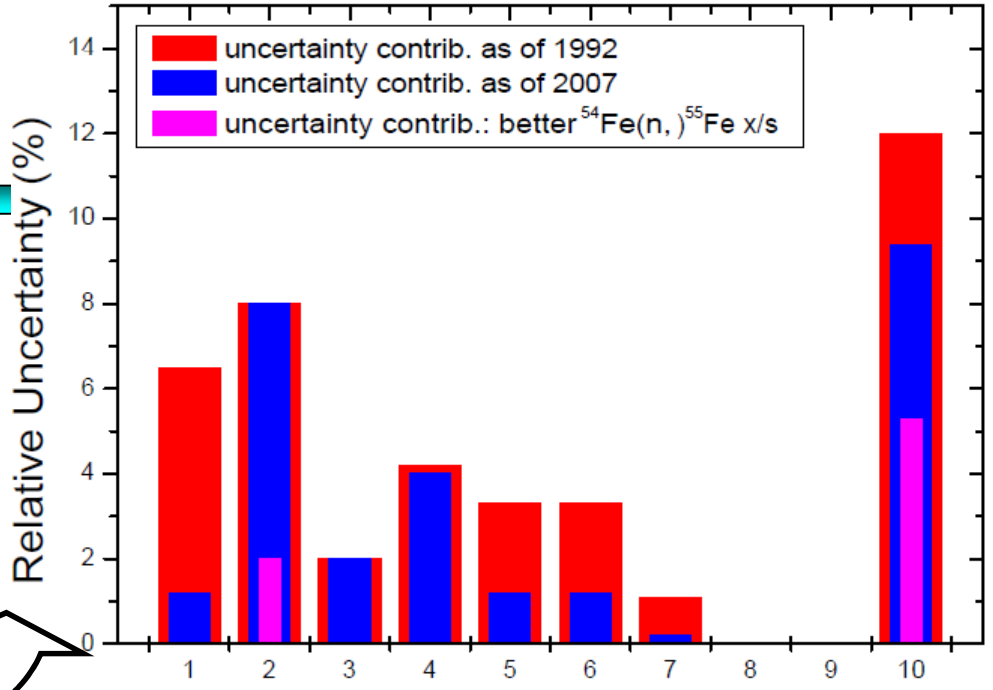
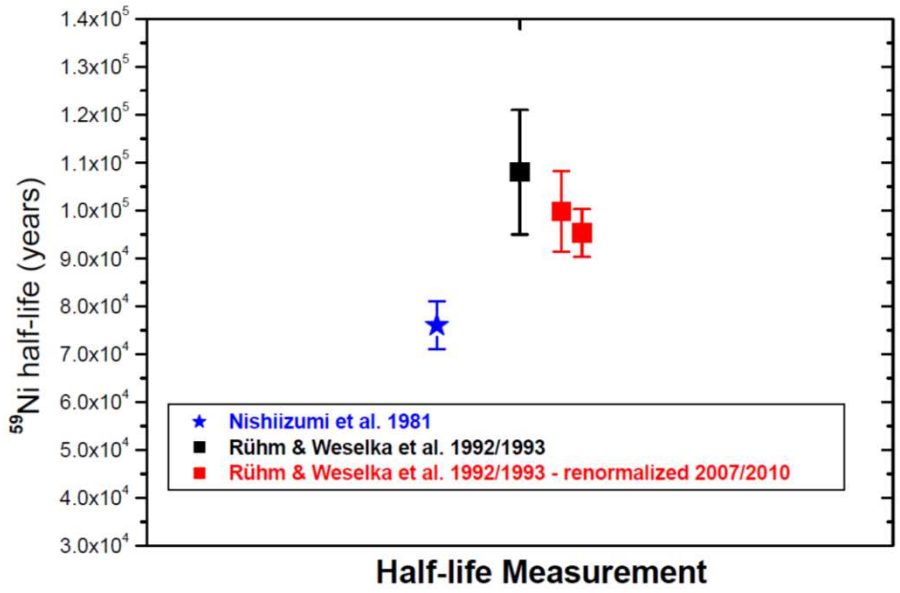
- IRMM / Geel: A. Plompen, A. Krasa
- IKI Budapest: T. Belgya, L. Szentmiklosi
- TSL Uppsala: A. Prokofiev et al.
- VERA (Vienna): P. Steier, S. Winkler, C. Lederer
- ANU (Canberra): L.K. Fifield, S. Tims, M. Froehlich
- ETH Zurich: M. Christl, J. Lachner
- ANSTO: M. Hotchkis
- Univ. Heidelberg: J. Lippold
- IAEA: R. Capote , V. Semkova

Half-life value of ^{59}Ni

Nishiizumi, 1981: $t_{1/2}(^{59}Ni) = (76\,000 \pm 5\,000)$ years

Rühm et al. 1994: $t_{1/2}(^{59}Ni) = (108\,000 \pm 13\,000)$ years



renormalized 2010: $t_{1/2}(^{59}Ni) = (95\,300 \pm 5\,300)$ years