Accelerator Mass Spectrometry, an unique technique in quite different fields of Science: from the Earth to the Stars.

G. Korschinek

Fachbereich Physik, E15
Technische Universität München

1. What is AMS?

2. Historical

3. AMS at the tandem accelerator (Garching)

4. Selected examples

5. Next?

where are we doing our measurements?

Germany
A
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Munich:
Beer
Oktoberfest
BMW
FC-Bayern
A
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TU-Munich, LMU,
MPQ, MPE, MPA,
MPP, ESO, …
FC Bayern
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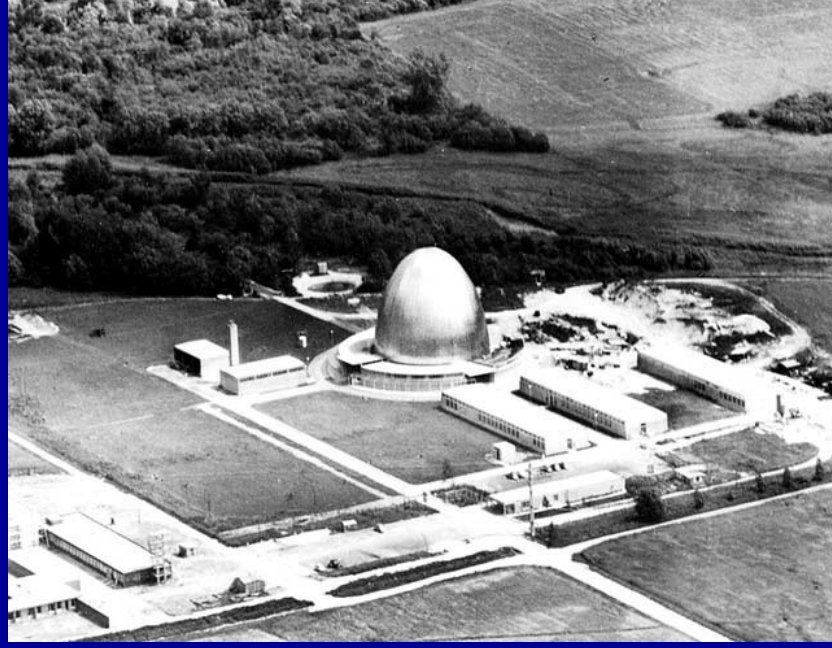
Where are we doing our research?

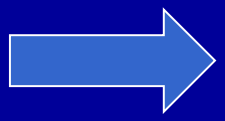


1. What is AMS?

Accelerator **M**ass **S**pectrometry (**AMS**) does, like conventional mass spectrometry, count the individual atoms.

a. The difference is because of the use of an accelerator (mostly a Tandem) where a stripping process is involved. This provides a complete suppression of molecular background.

b. Because of the final energy after acceleration, nuclear counting techniques serve for rigorous reduction of isobaric background. (negative ion formation in the ion source helps in some cases; e.g. ^{14}C as ^{14}N does not form negative ions.)



This yields an ultra sensitive method!

2. Historical

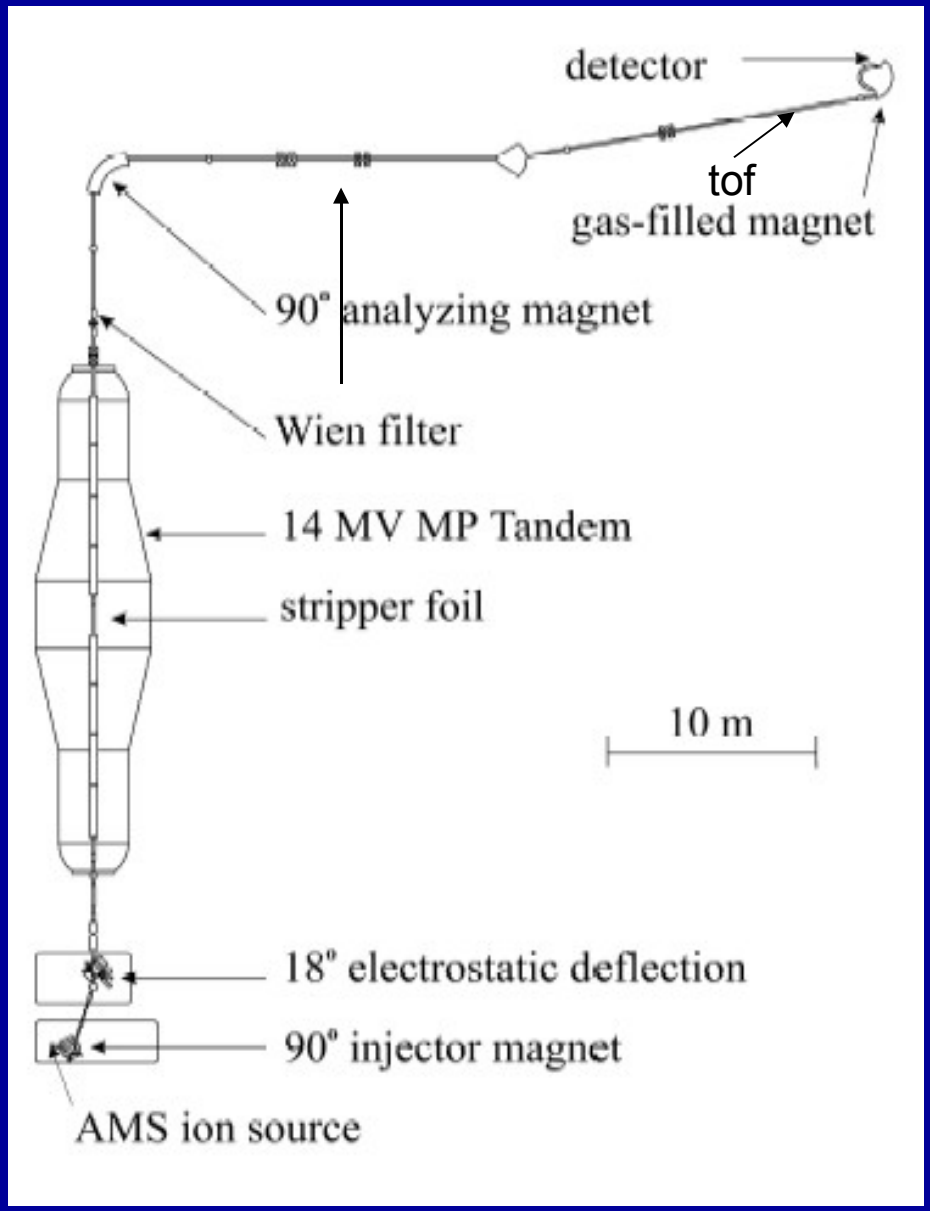
1939 Alvarez and Cornog discovered accidentally ^{3}He (cyclotron).

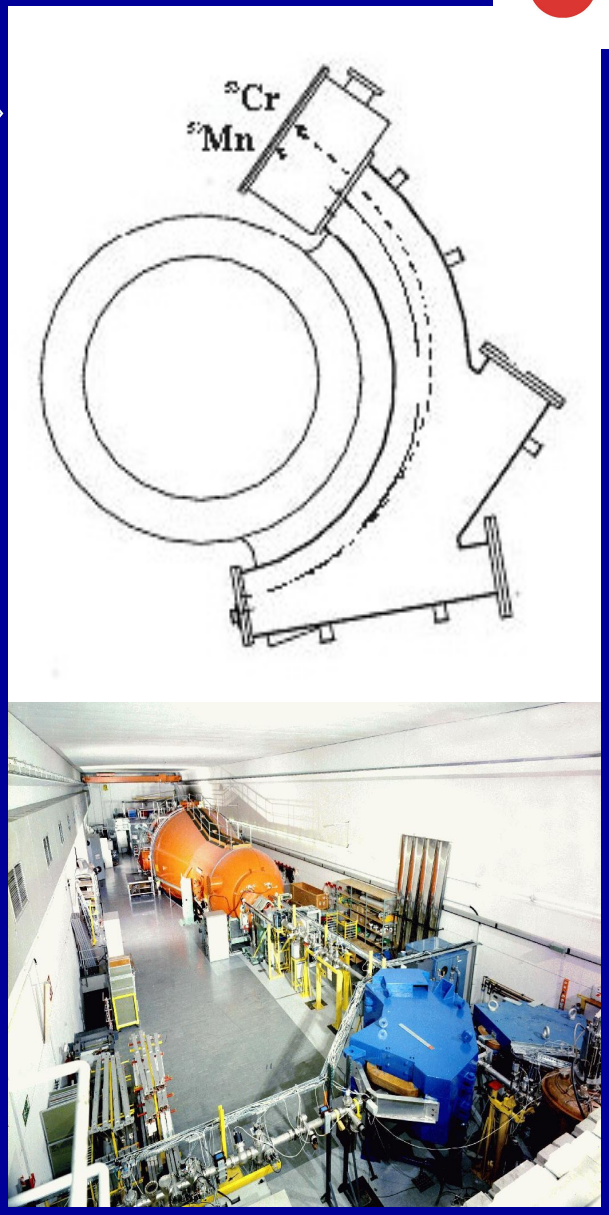
From 1939 until 1977, for around 40years, dark and silent!

1977 Muller proposed to measure ^{14}C or ^{10}Be by means of a cyclotron.
Independently, Gove, Litherland, and Purser suggested the use of a Tandem accelerator for ^{14}C measurements.

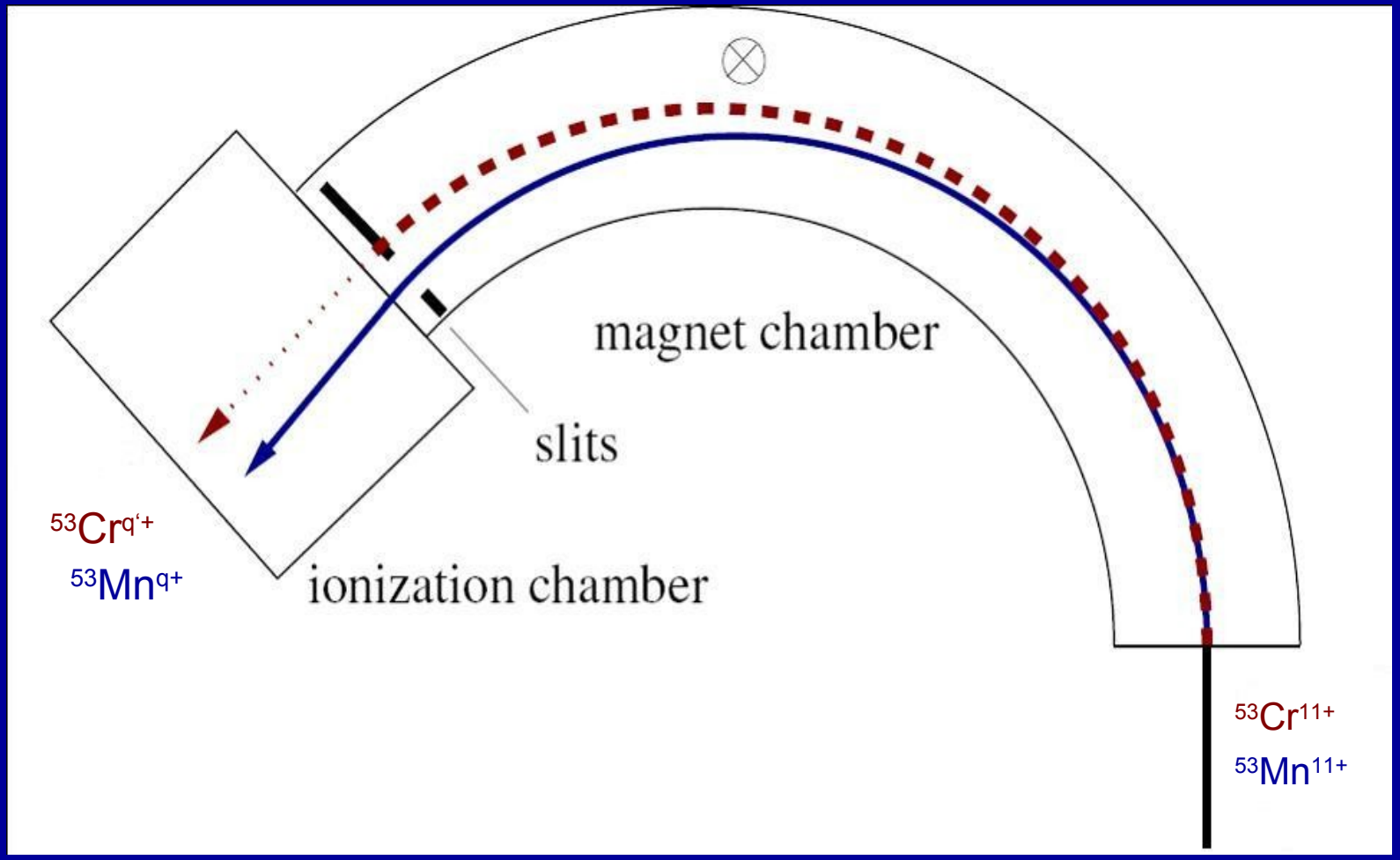
2008 Around 80 AMS laboratories worldwide. From small table top accelerators until large facilities with more than 10MV terminal voltage.
Isotopes of interest from ^{3}H until the heaviest nuclides like ^{244}Pu.
The majority of AMS laboratories is dedicated to ^{14}C and ^{10}Be.

3. AMS at the tandem accelerator (Garching)

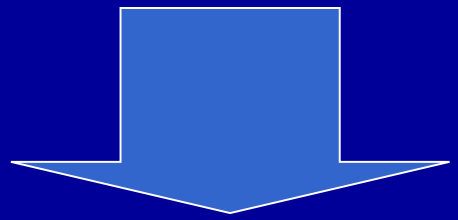




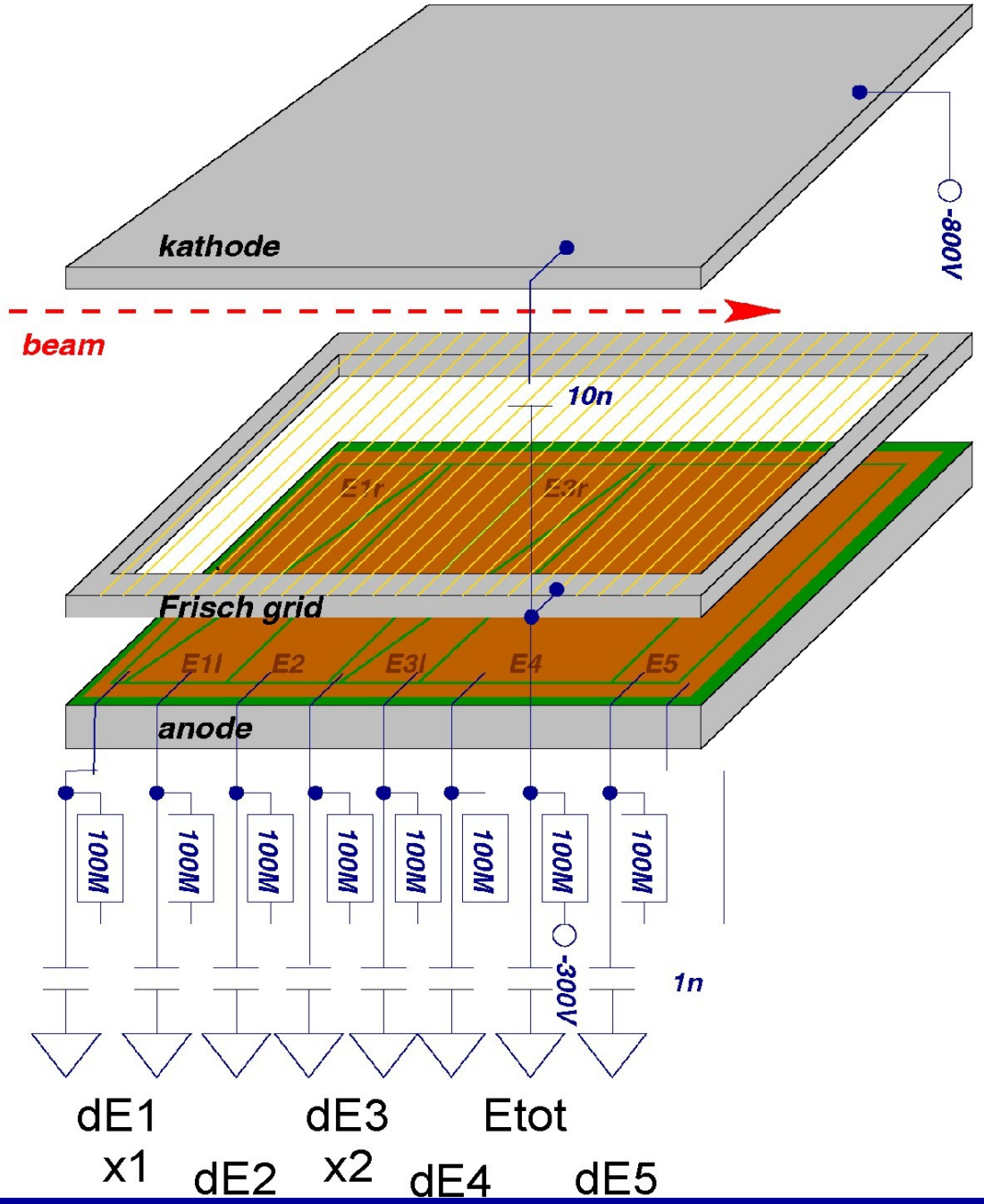
GAMS


magnet chamber
slits
$^{53}Cr^{q'+}$
$^{53}Mn^{q+}$
ionization chamber
$^{53}Cr^{11+}$
$^{53}Mn^{11+}$

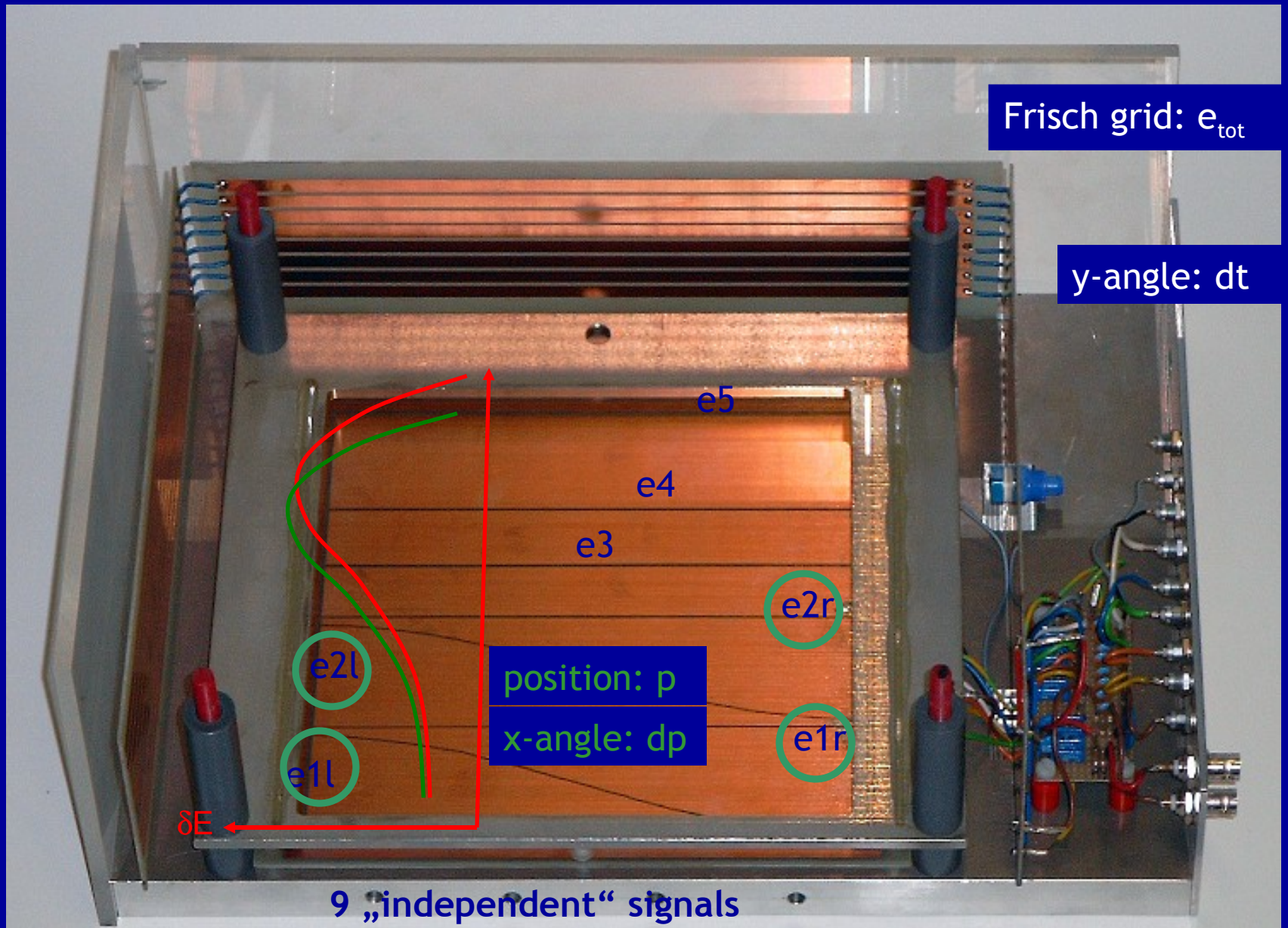
ionization chamber



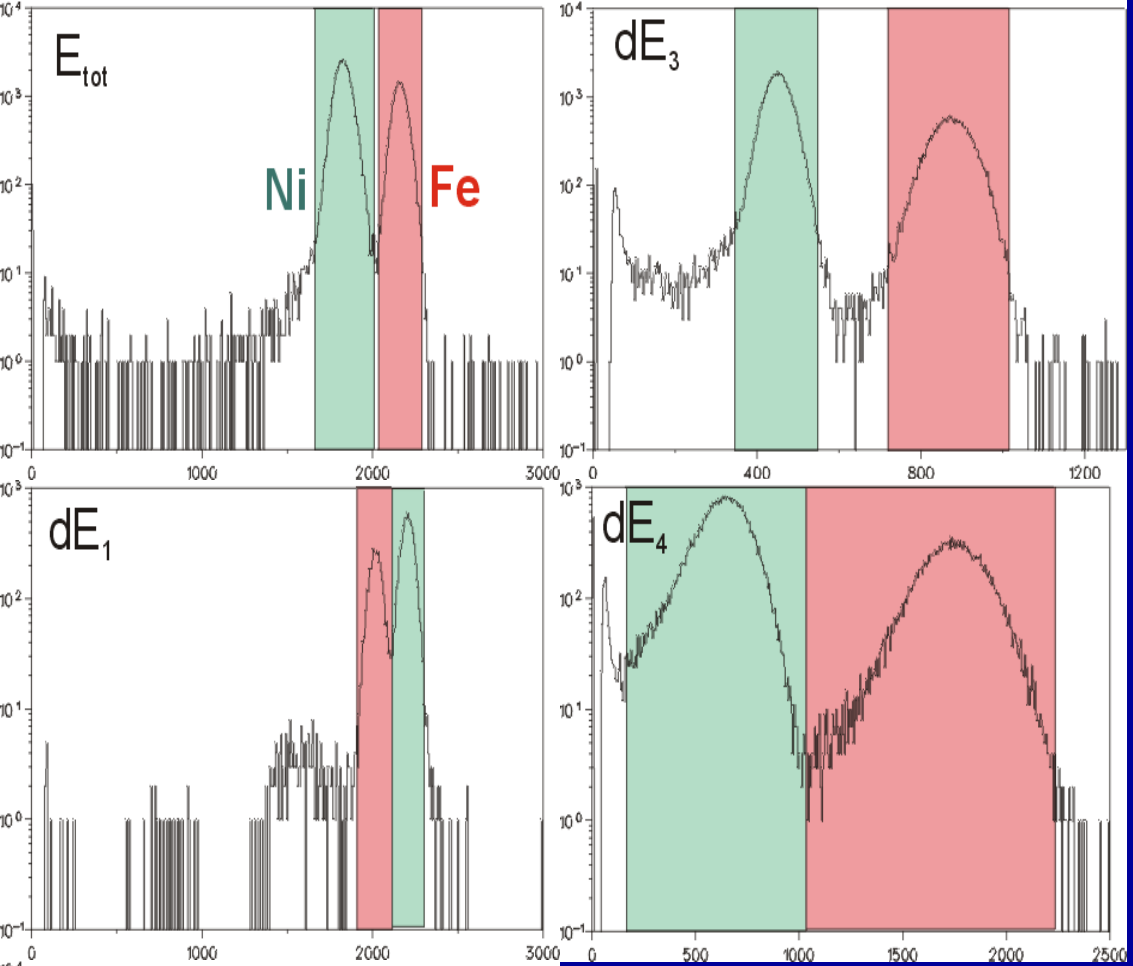
additional isobar separation!

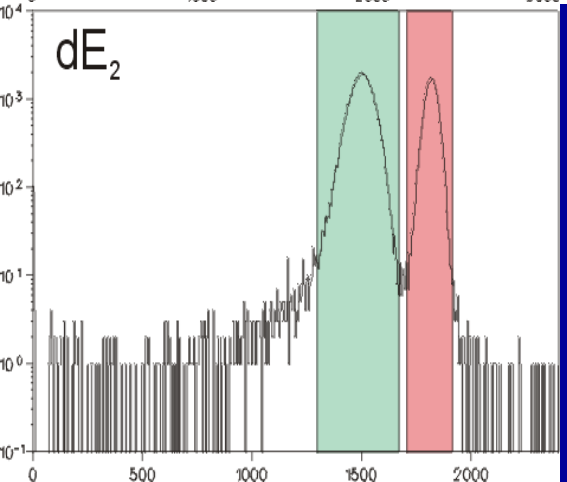


Ionization chamber



9 „independent“ signals



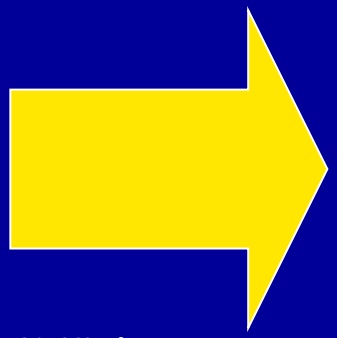


+ dE_5
+ x angle
+ y angle
+ pile up rejection

Efficiency: ~ 10^{-4}

Sensitivity: down to 10^{-16} isotopic ratios

ratio with respect to the bulk ~ 10^{-20}



21500km^3

minute traces of long-lived
radio-isotopes
(natural or anthropogenic origin)

4. Examples

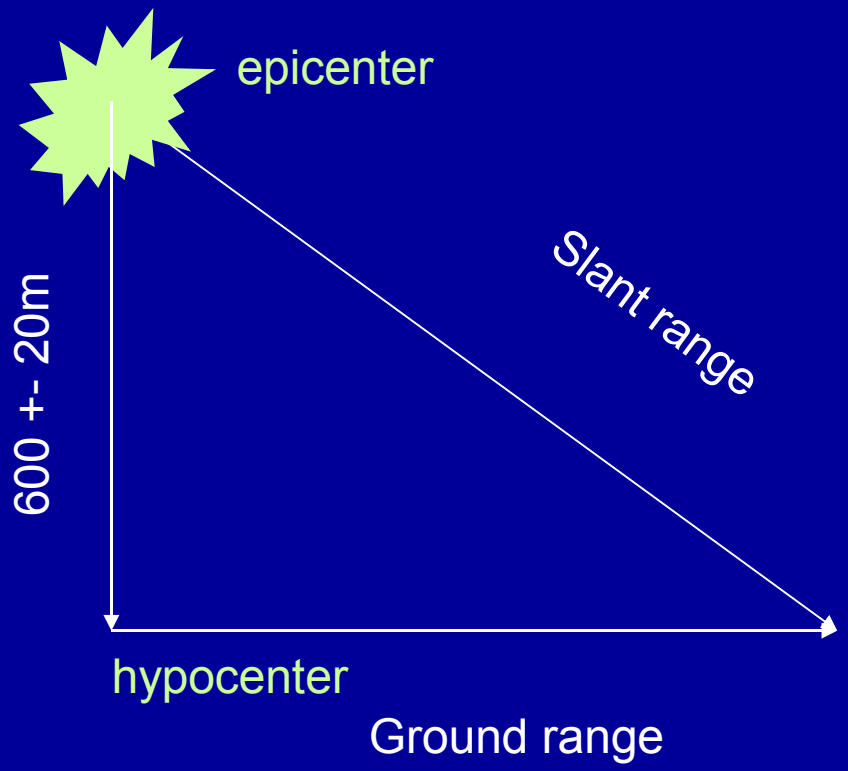
- Environmental and Geo-science
  - atmosphere, oceanography, glacier, climate, groundwater, erosion, paleo-oceanography, paleo-tectonic
- Material science — fusion research, active waist management
- Human metabolism and medical application
  - dosimetry, pharmacology, ….
- Chemistry — tracing molecules and elements, …
- Extraterrestrial
  - meteorites (e.g. Lunar and Martian origin) interplanetary dust. SN condensate. Related cross sections ….
- Physics
  - nuclear astrophysics, super- asymmetric fission …

- **Dosimetry system**

Thermal and fast neutron fluencies in Hiroshima and Nagasaki due to the atomic bombs (study of 120 000 individuals).

Geometry at the time of explosion

(Hiroshima, energy yield: 16 +- 2kt, DS02)



position

^{36}Cl

^{41}Ca

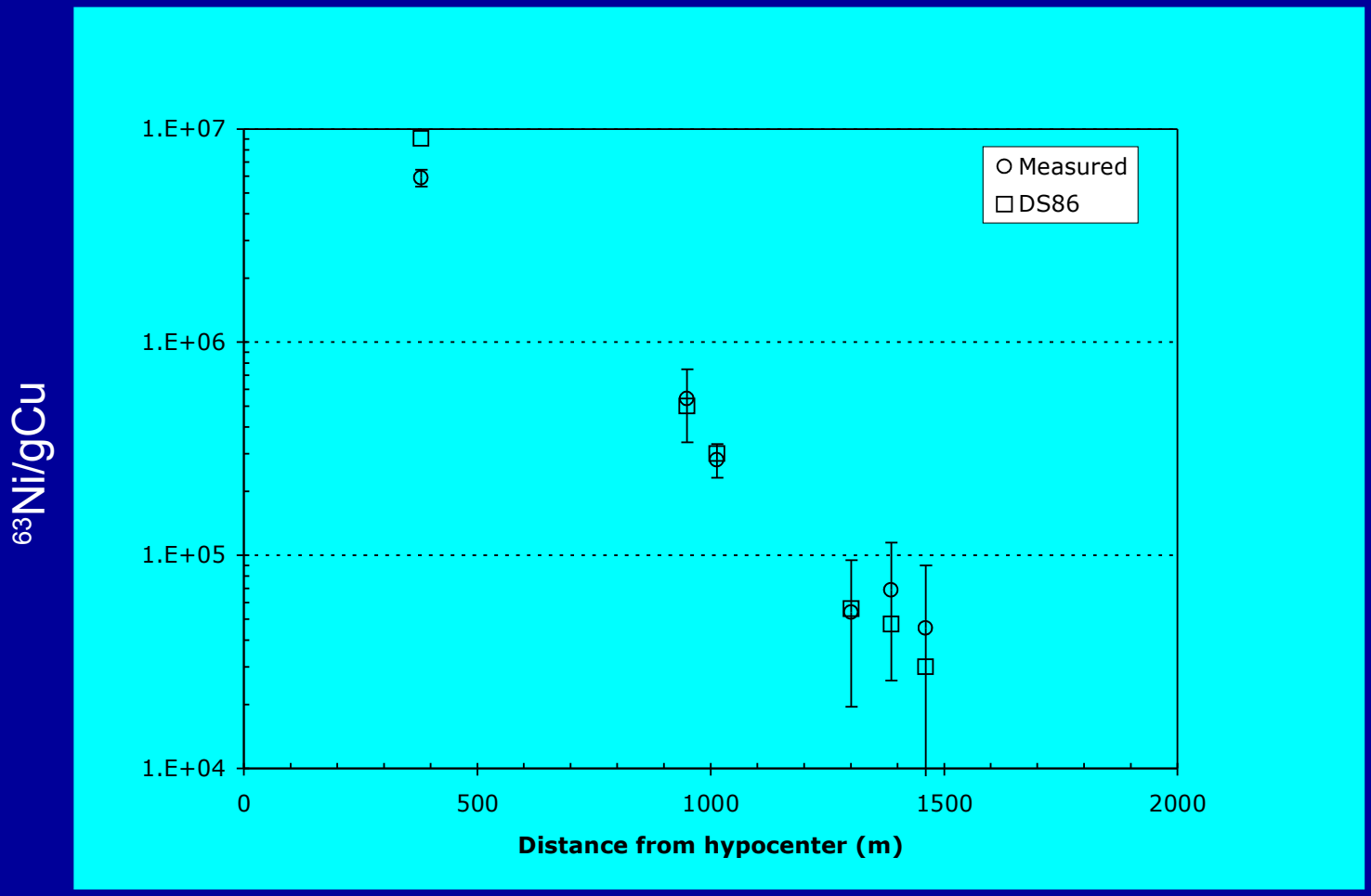
^{63}Ni

Thermal neutron monitor: ^{36}Cl (3.01×10^{5}a, target ^{35}Cl),
^{41}Ca (1.04×10^{5}a, target ^{40}Ca)

Fast neutron monitor: ^{63}Ni (100a, target ^{63}Cu), $^{63}Cu(n_f,p)^{63}Ni$

Typical sample for ^{63}Ni

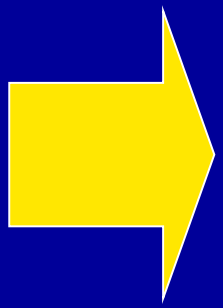
(^{63}Ni)



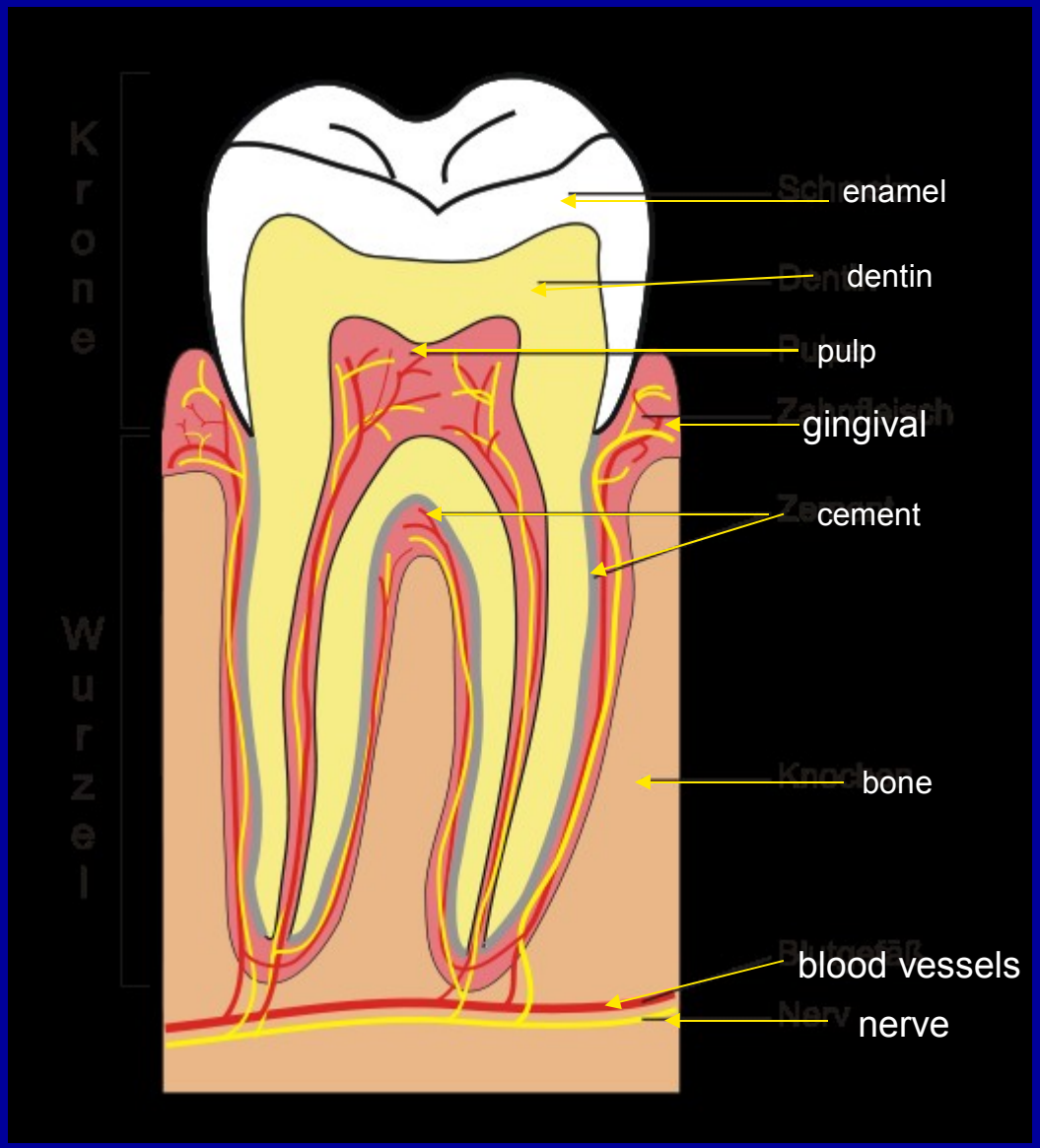
Straume et al. Nature, 430 (2004) 483

Questions: individuals and local variations of neutron flux? (location, shielding, secondary production, ..)

We need a dosimeter for each individual!



^{41}Ca ?


Krone
Wurzel
enamel
dentin
pulp
gingival
cement
bone
blood vessels
nerve

^{41}Ca !

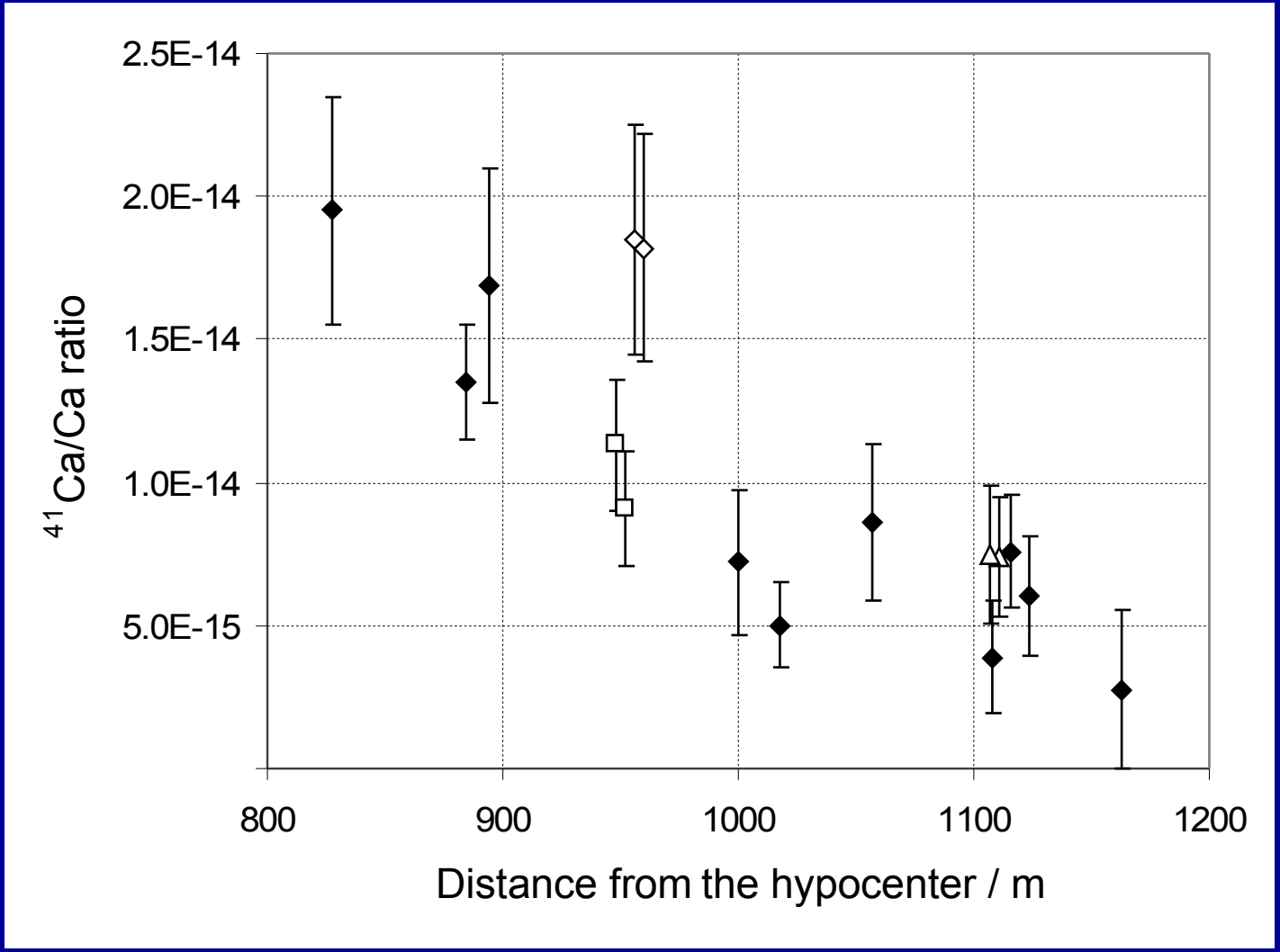
^{41}Ca in tooth enamel!

$^{40}Ca(n_{th},\gamma)^{41}Ca$

16 tooth enamel samples from 13 atomic bomb survivors.
100mg enamel corresponds ≈ 36mg Ca.

Background because of natural ^{41}Ca level?
(from 6 control samples of distant survivors)
Near equator, sea level, high erosion rate, and sea food! → 1.7×10^{-15} $^{41}Ca/Ca$

(^{41}Ca)


2.5E-14
2.0E-14
1.5E-14
1.0E-14
5.0E-15
^{41}Ca/Ca ratio
800
900
1000
1100
1200
Distance from the hypocenter / m

In collaboration with:

W. Rühm, Germ. Res. Center f. Environ. Health

N. Nakamura, RERF Hiroshima

S. Egbert, Science Appl. Intern. Corp. US

•Micrometeorites on Earth.

- Interplanetary dust deposit on Earth.
- Cosmic ray intensity during the last million of years.
- ^{53}Mn as a proxy for interstellar ^{60}Fe.

- **micrometeorites (cosmic dust)**
  (between 800t and 40 000t per year)

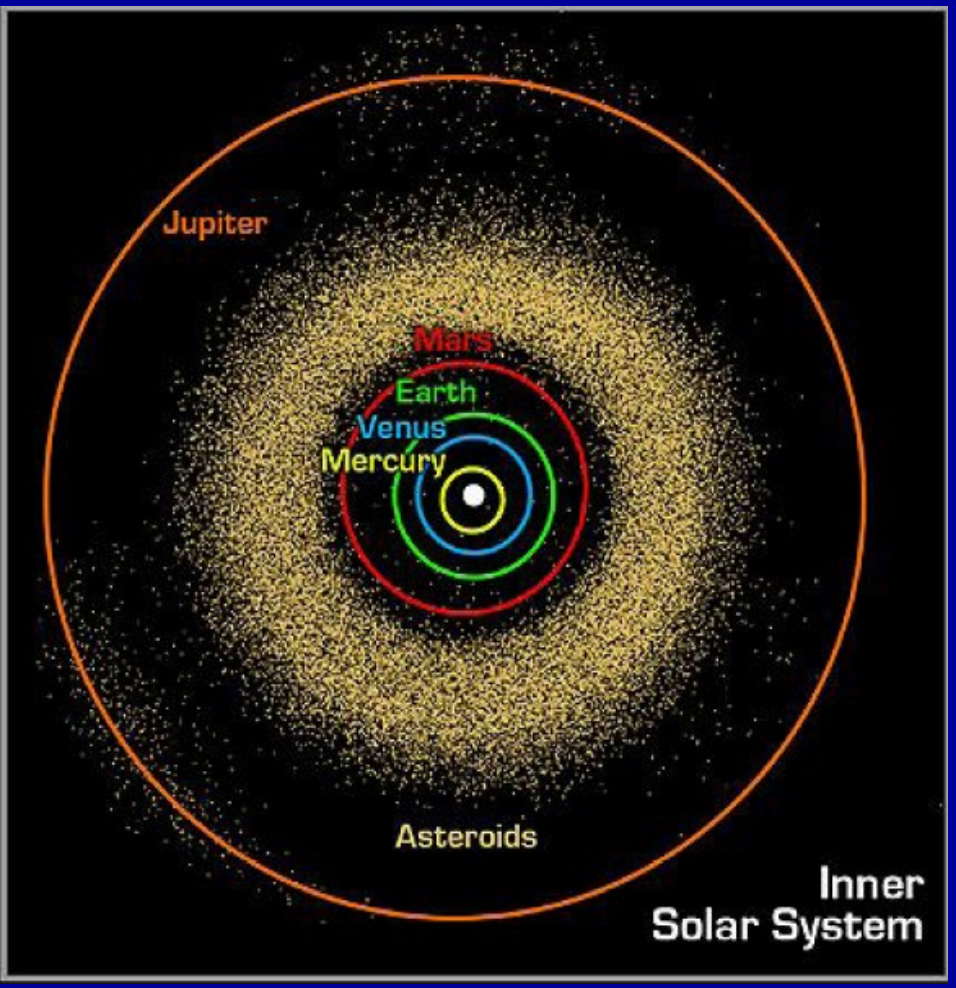
Sources:

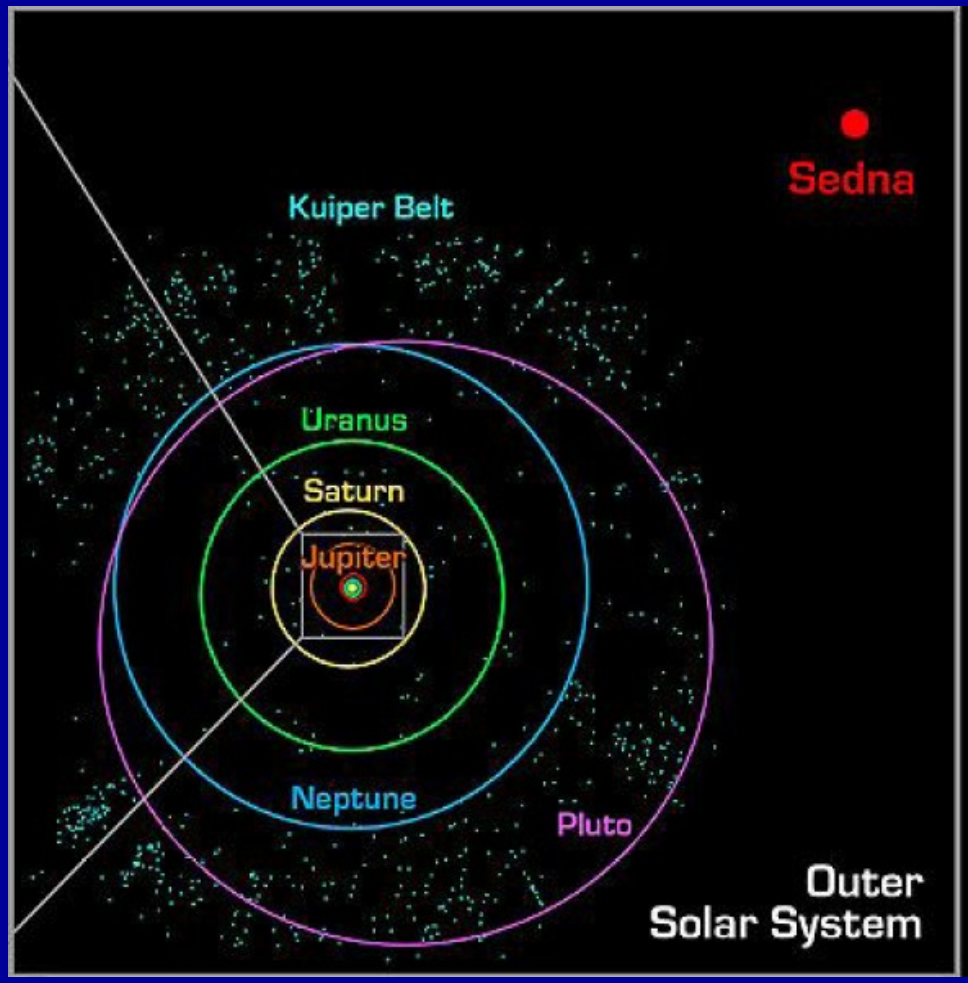
-cometary origin (Kuiper belt)

-asteroids

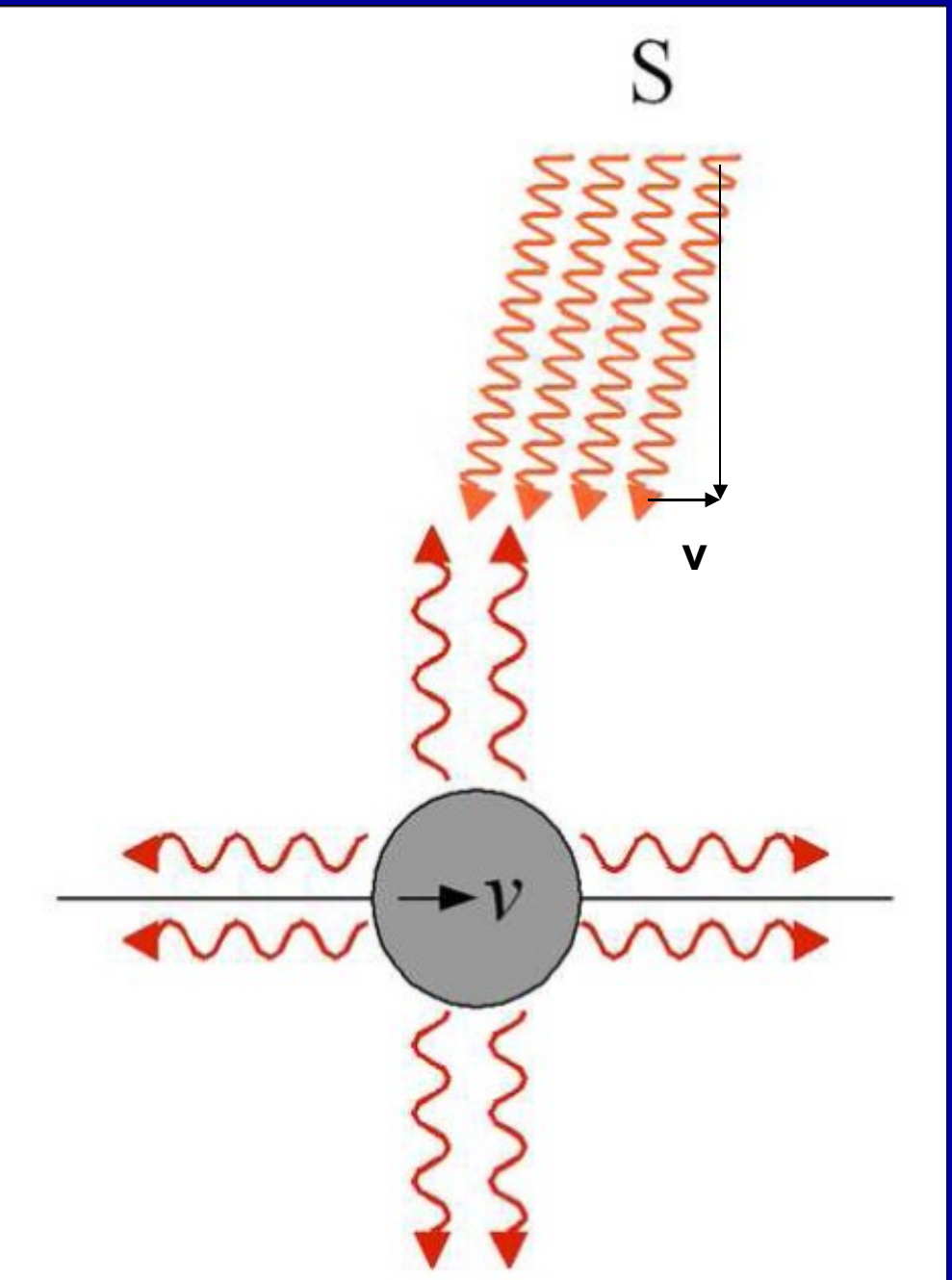
-melt off from large objects during entrance into the atmosphere

Cosmic dust sources: asteroid belt and cometary cloud

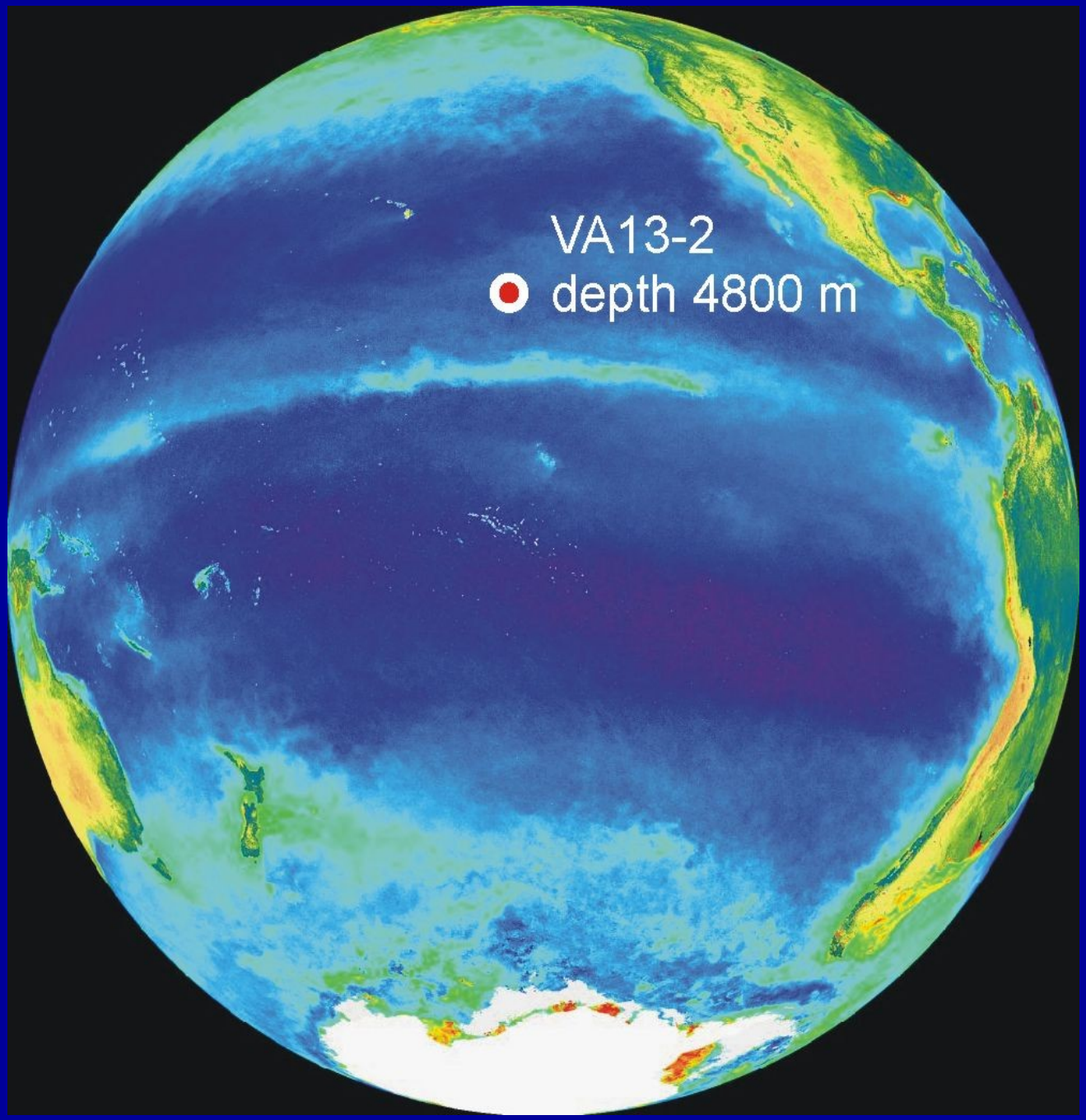




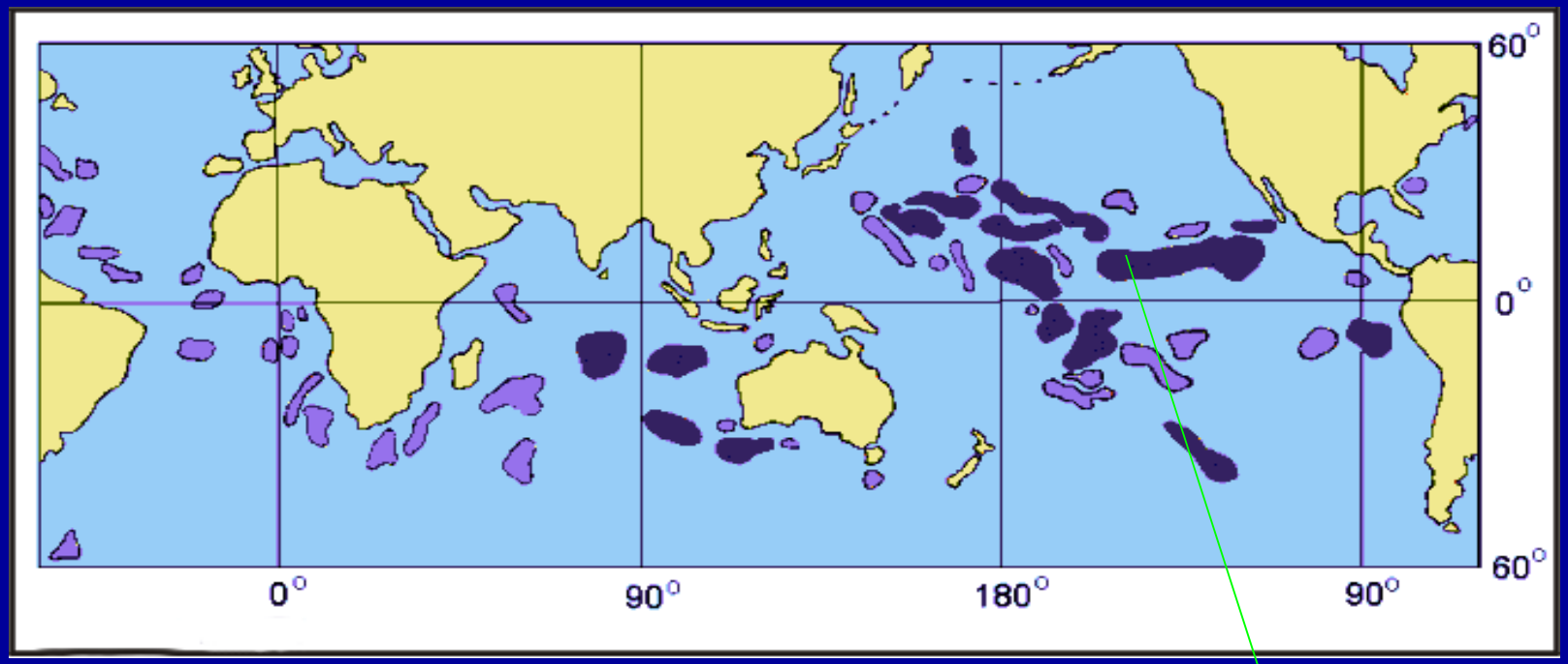
Anisotropically illumination
Poynting-Robertson effect

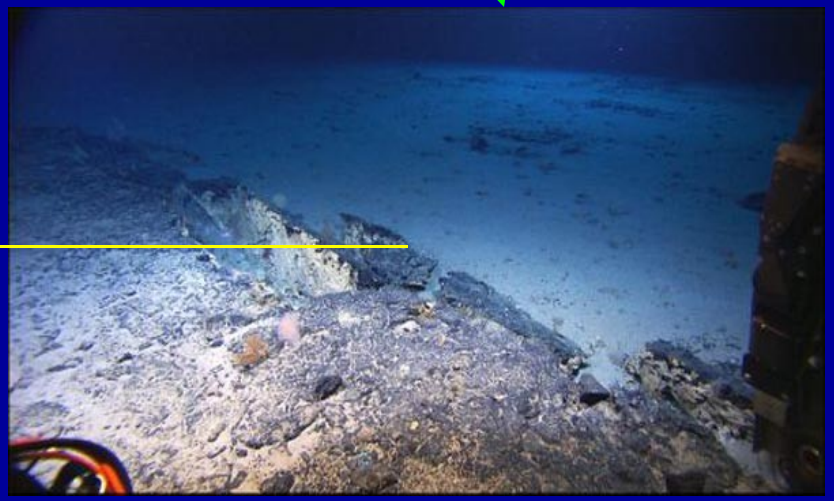


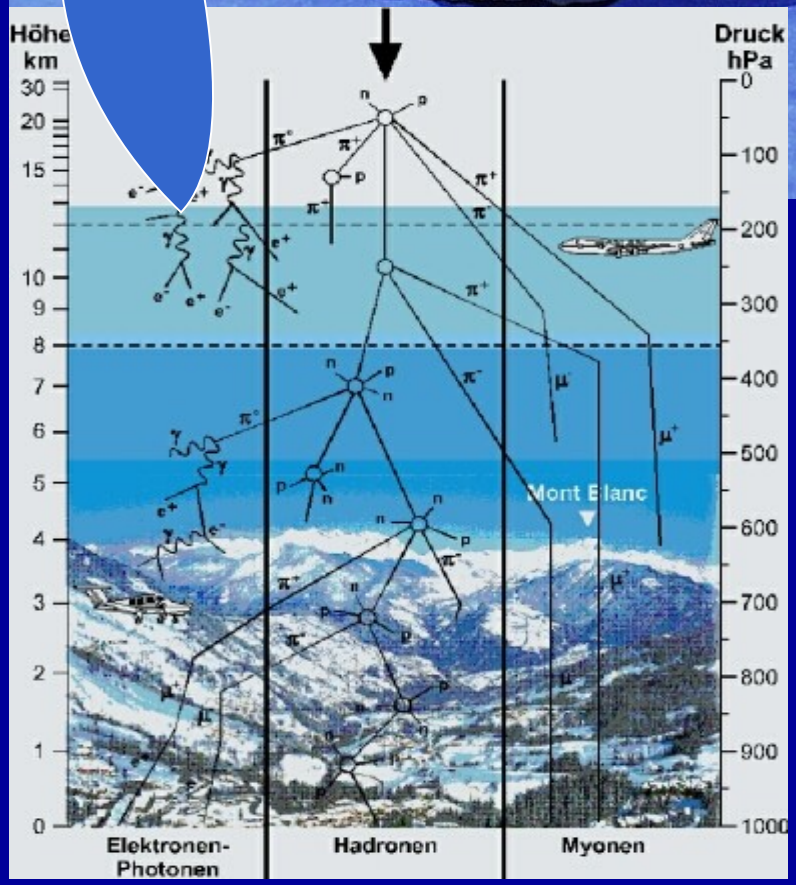
Poynting-Robertson effect


VA13-2
depth 4800 m

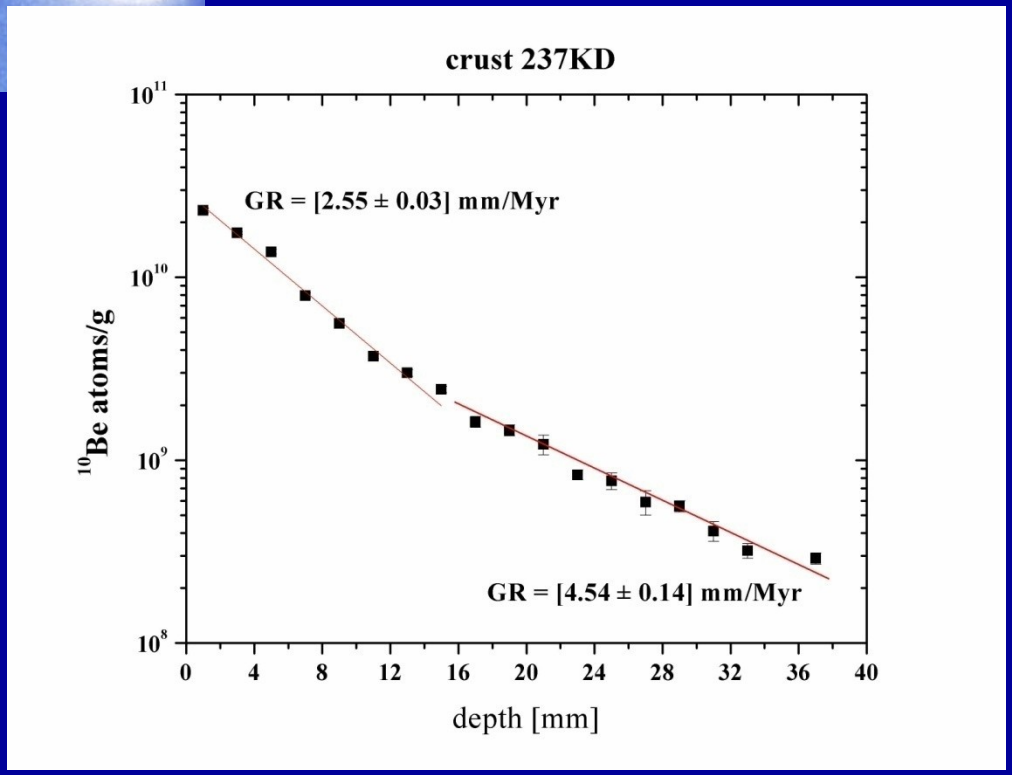
Ferromanganese crust





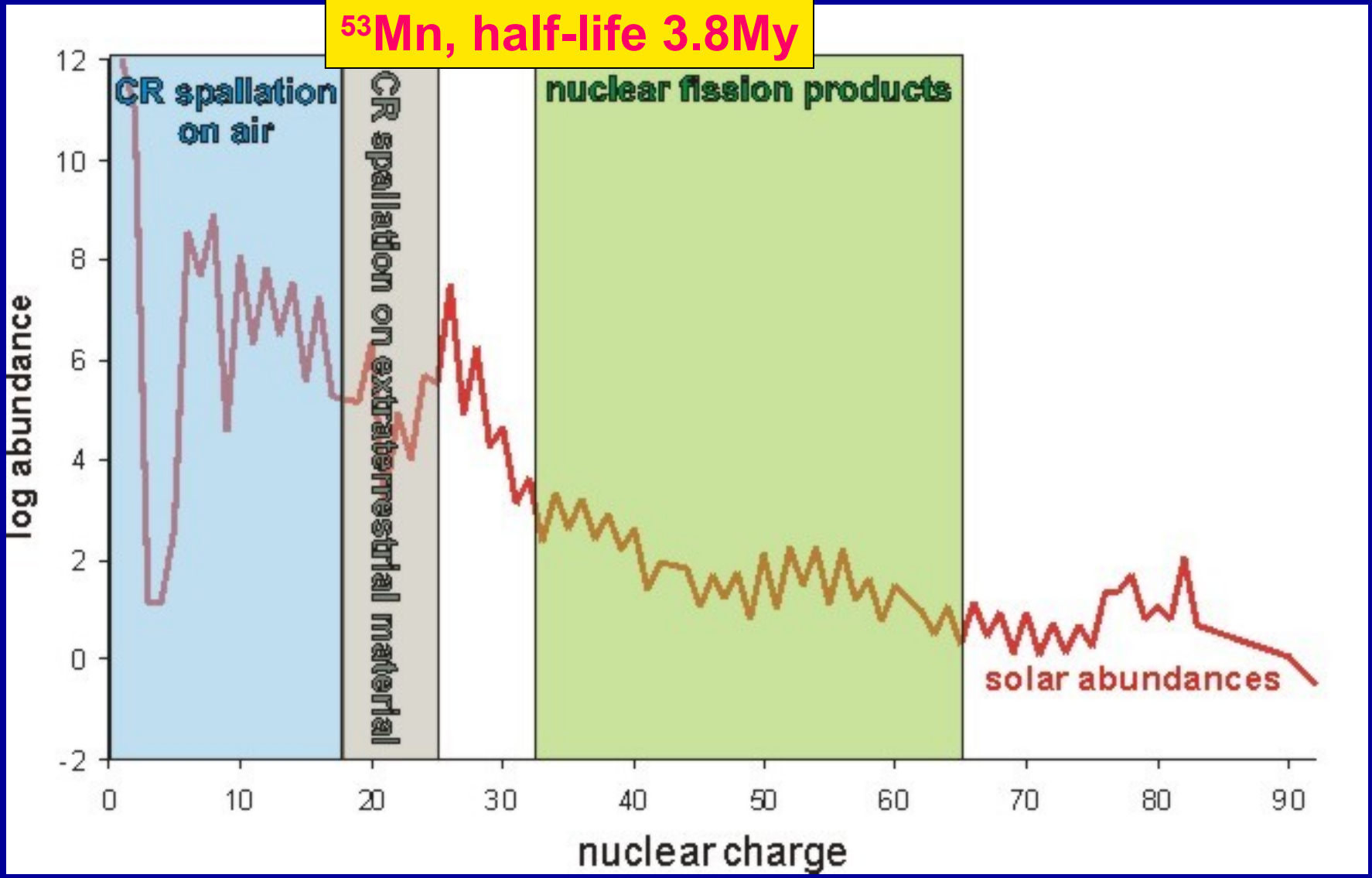


^{10}Be, $T_{1/2}$=1.387My

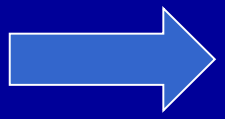


Which isotope exhibits a clear signature of cosmic dust?

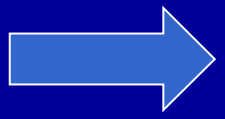
^{53}Mn, half-life 3.8My

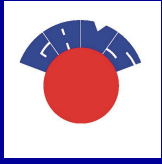


cosmic rays (mostly protons) interact with the dust particles (spallation on Fe, Ni, ..).

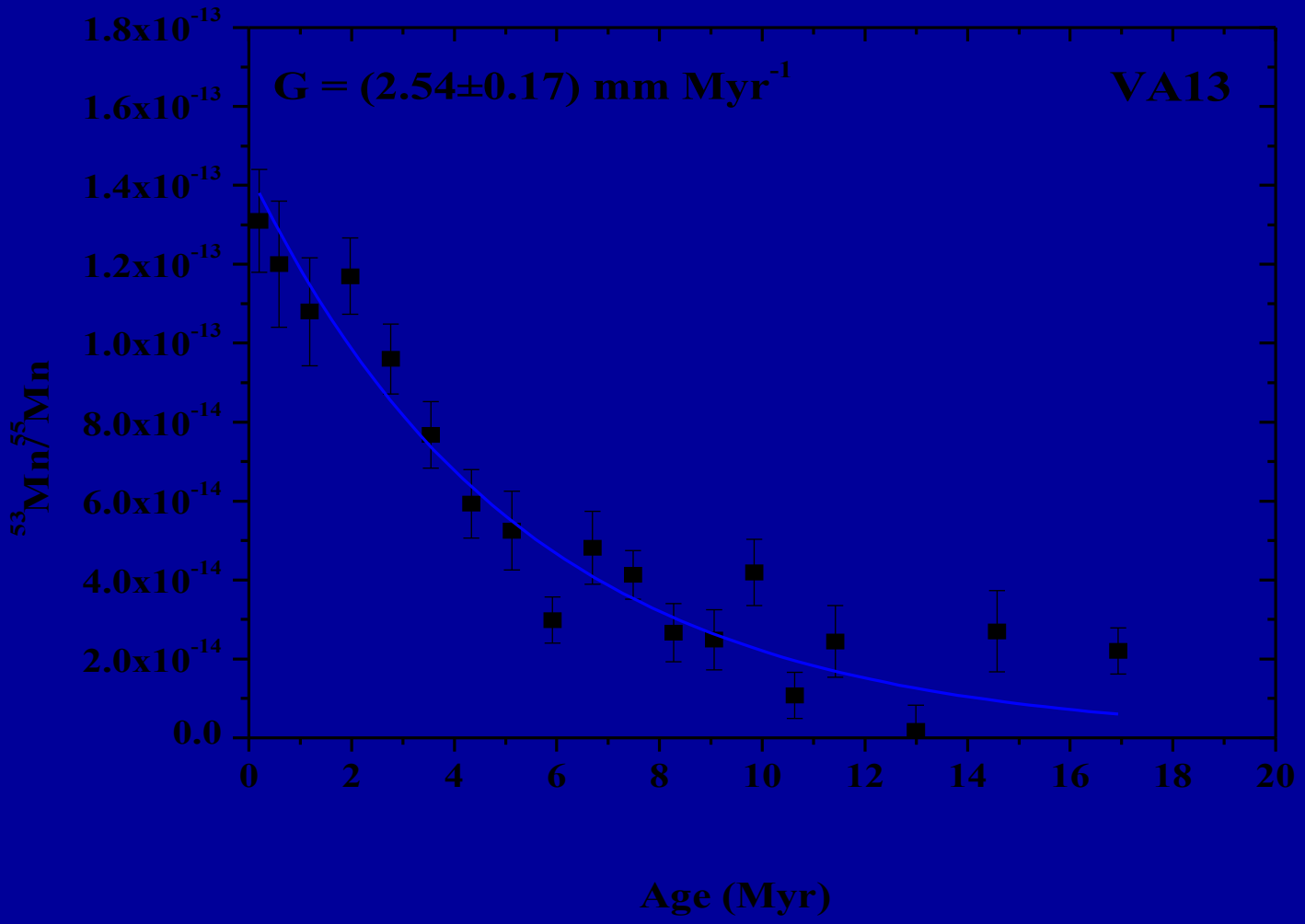
 …….^{53}Mn ($T_{1/2}$=3.8My)…….

^{53}Mn not produced in the atmosphere, and only marginally on Earth's surface!

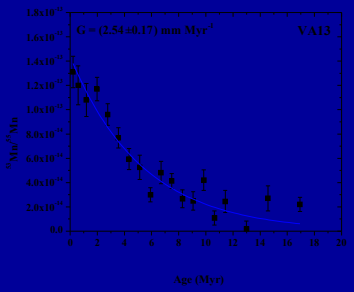
 unique signature for interplanetary dust flux!



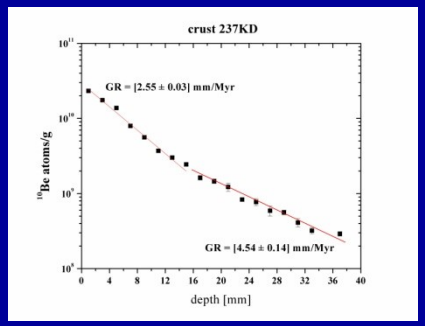
Co54 193.23 ms 0+ * EC	**Co55** 17.53 h 7/2- EC	**Co56** 77.27 d 4+ EC	**Co57** 271.79 d 7/2- EC	**Co58** 70.82 d 2+ * EC	**Co59** 7/2- 100	**Co60** 5.2714 y 5+ *
Fe53 8.51 m 7/2- * EC	**Fe54** 0+ 5.8	**Fe55** 2.73 y 3/2- EC	**Fe56** 0+ 91.72	**Fe57** 1/2- 2.2	**Fe58** 0+ 0.28	**Fe59** 44.503 d 3/2- β-
Mn52 5.591 d 6+ * EC	**Mn53** 3.74E+6 y 7/2- EC	**Mn54** 312.3 d 3+ EC,β-	**Mn55** 5/2- 100	**Mn56** 2.5785 h 3+ β-	**Mn57** 85.4 s 5/2- β-	**Mn58** 3.0 s 0+ * β-
Cr51 27.702 d 7/2- EC	**Cr52** 0+ 83.789	**Cr53** 3/2- 9.501	**Cr54** 0+ 2.365	**Cr55** 3.497 m 3/2- β-	**Cr56** 5.94 m 0+ β-	**Cr57** 21.1 s 3/2-,5/2-,7/2- β-

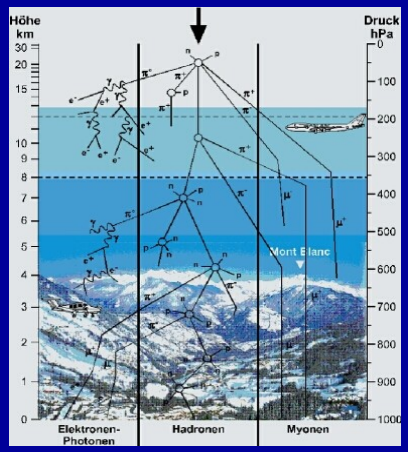


^{53}Mn ($T_{1/2}$= 3.8Ma)



$$N_{53Mn}(t) \sim \Phi_{cosmic\ rays}(t) \times M_{cosmic\ dust}(t)$$



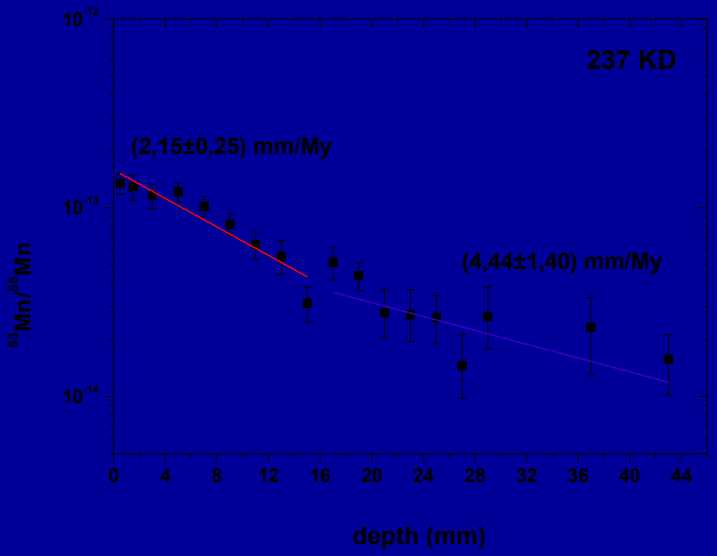


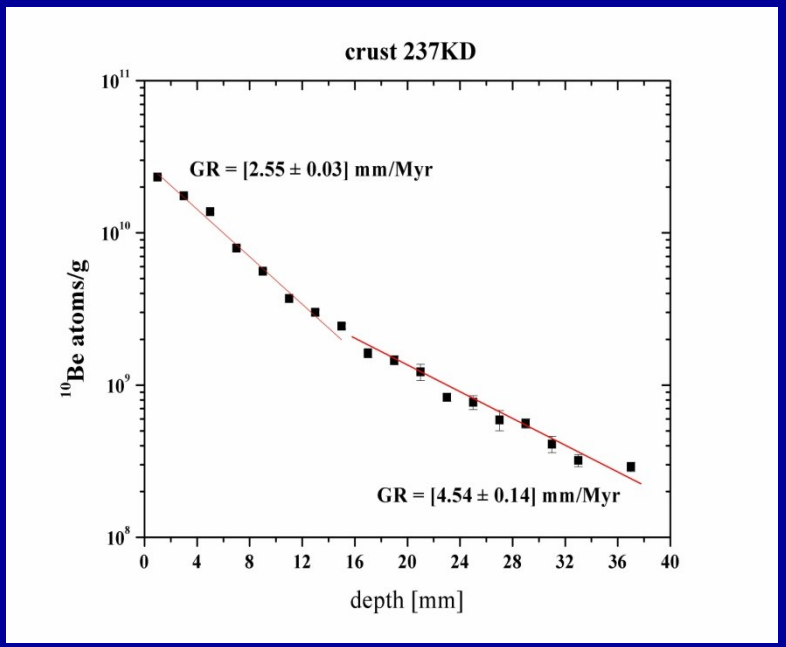
$$N_{10Be}(t) \sim \Phi_{cosmic\ rays}(t) \times M_{atmosphere}$$

$$N_{53Mn}(t) / N_{10Be}(t) \sim M_{cosmic\ dust}(t) / M_{atmosphere}$$

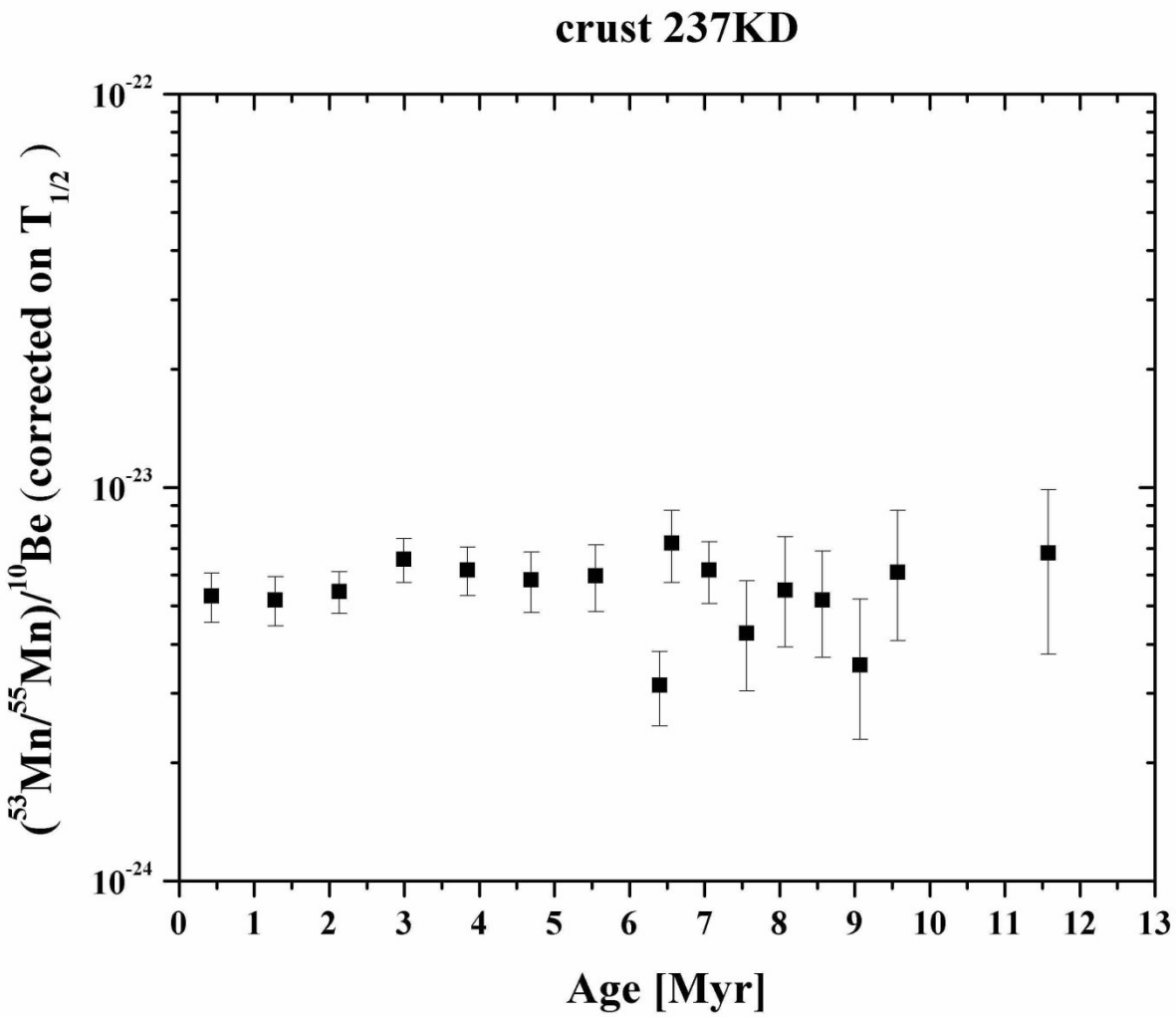
Cosmic rays plus cosmic dust → ^{53}Mn

Cosmic rays plus atmosphere → ^{10}Be





cosmic dust fluency



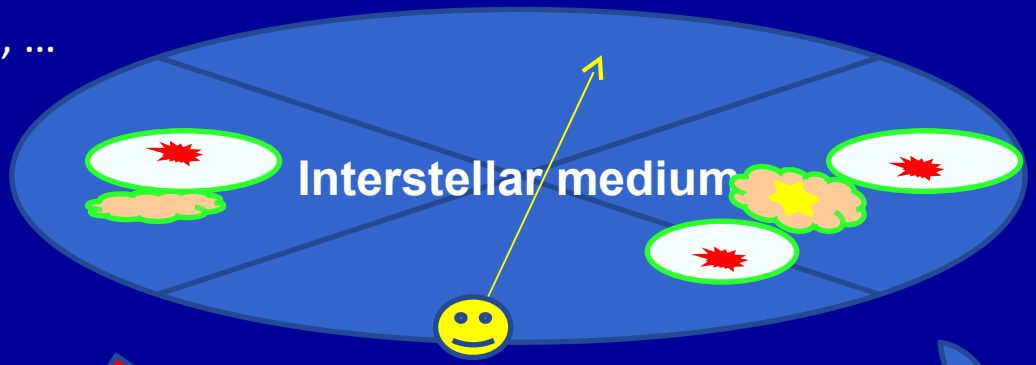
- Interstellar ^{60}Fe on Earth

The cosmic metabolism and the merit of radio nuclides

$^{26Al}T_{1/2} = 7 \times 10^5$ y

$^{60Fe}T_{1/2} = 2.62 \times 10^6$ y

^{26}Al, ^{44}Ti, ^{53}Mn, ^{60}Fe, …



Nucl. synth.

$^{235/238}U$, ^{244}Pu, …

Molecular cloud

Star formation

$^{244Pu}T_{1/2} = 8.1 \times 10^7$ y

$^{235U}T_{1/2} = 7 \times 10^8$ y

$^{238U}T_{1/2} = 4.5 \times 10^9$ y

Signatures of recent (≈10 Myr), nearby (≈100 pc) SNe in the local interstellar environment

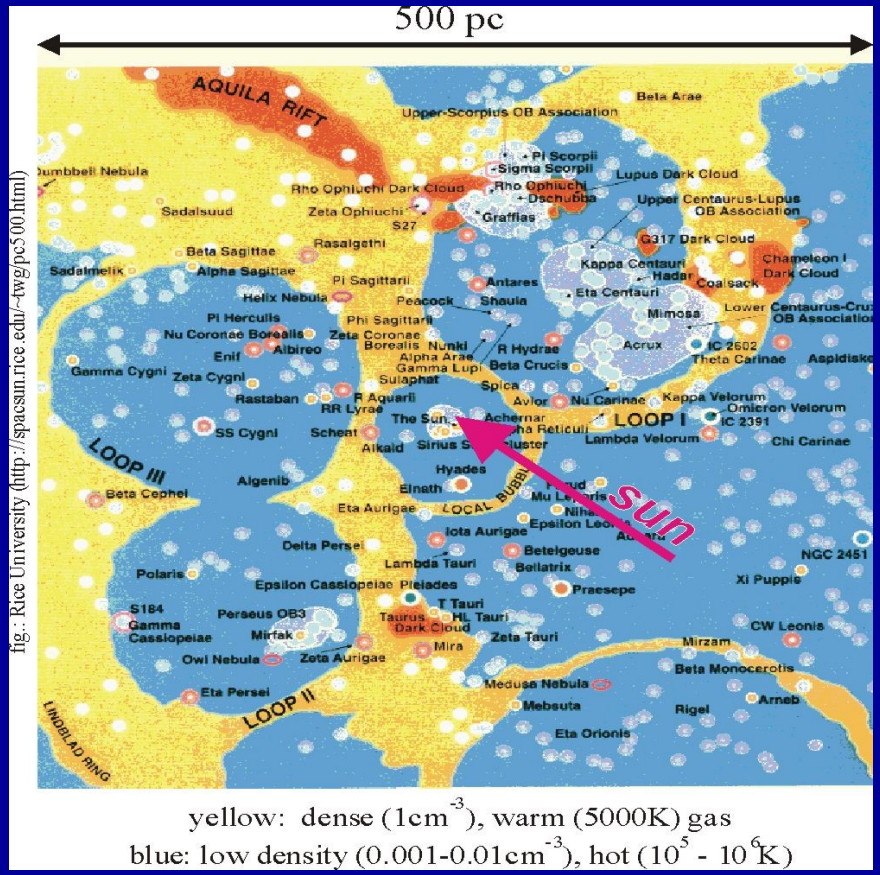
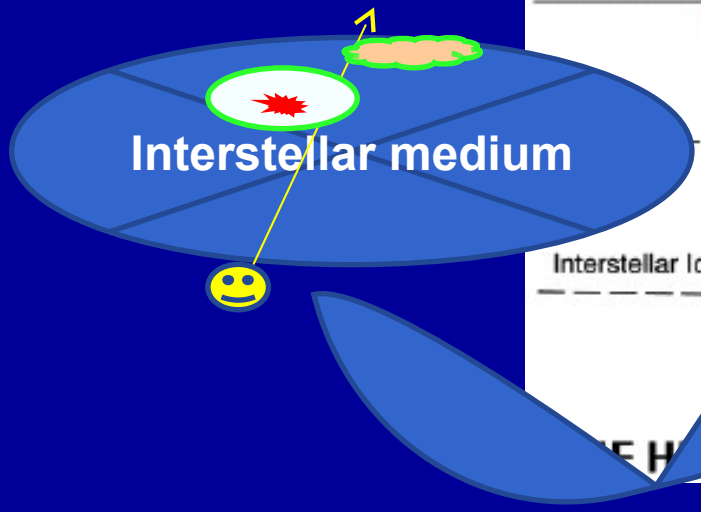


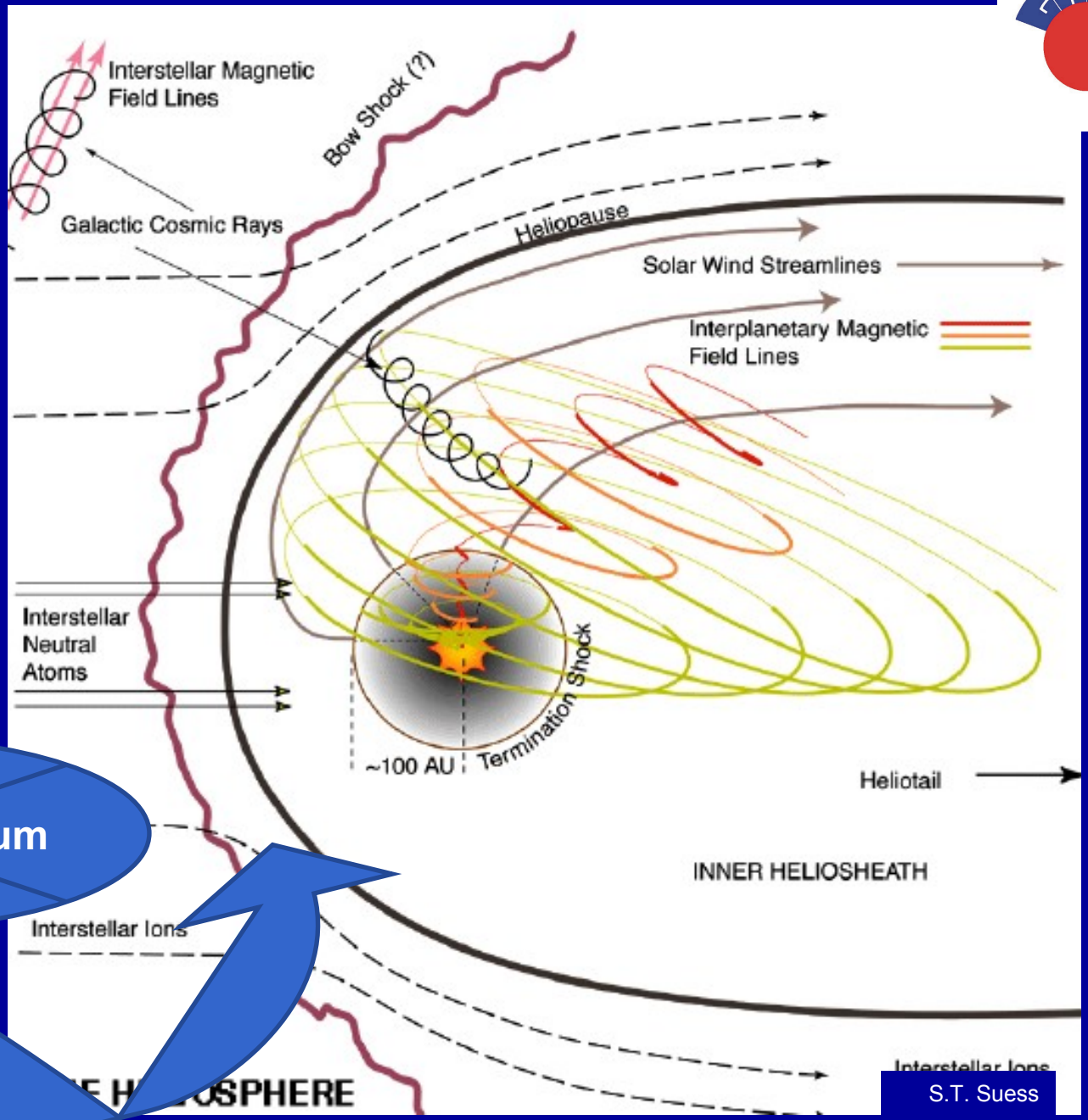
fig.: Rice University (http://spacsun.rice.edu/~twg/pc500.html)

yellow: dense (1cm^{-3}), warm (5000K) gas
blue: low density (0.001-0.01cm^{-3}), hot (10^5 - 10^6K)

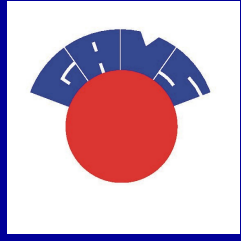
- **Existence of the local bubble:**
  2-3 SN in the solar vincinity,
  last SN **2-5 Myr** ago.
  Smith & Cox, ApJS 134(2001)283
- **Ionization of the "Local Fluff":**
  27 % HII, 35 % HeII
  ⇨ too high for equilibrium
  ⇘ ionized by a SN shock
  ⇘ onset of the recombination: **3-4 Myr**
  Barstow et al., MNRAS 286(1997)58
- **Antiprotons in cosmic rays:**
  CR propagation models produce too few antiprotons (B/C correct)
  ⇘ fresh, local CR component?
  Moskalenko et al., ApJ 586(2003)1050
- **Isotope anomalies on Earth due to direct deposition of SN ejecta?**
  Ellis et al., ApJ 470(1996)1227,
  Korschinek et al., Radiocarbon 38(1996)68

Interstellar matter (ISM) within the solar system.





S.T. Suess



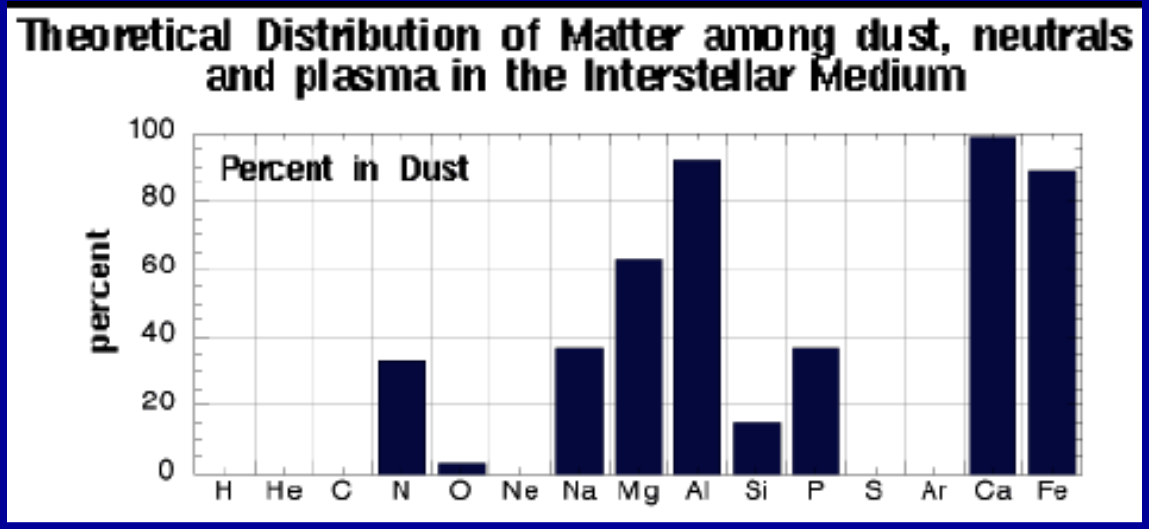
Direct deposition:

Conditions for solar wind penetration

$$P_{ej} \geq P_{SW}$$

$$D_{crit} \approx 20pc \quad (\rho_{ISM} \approx 1cm^{-3})$$

Or!



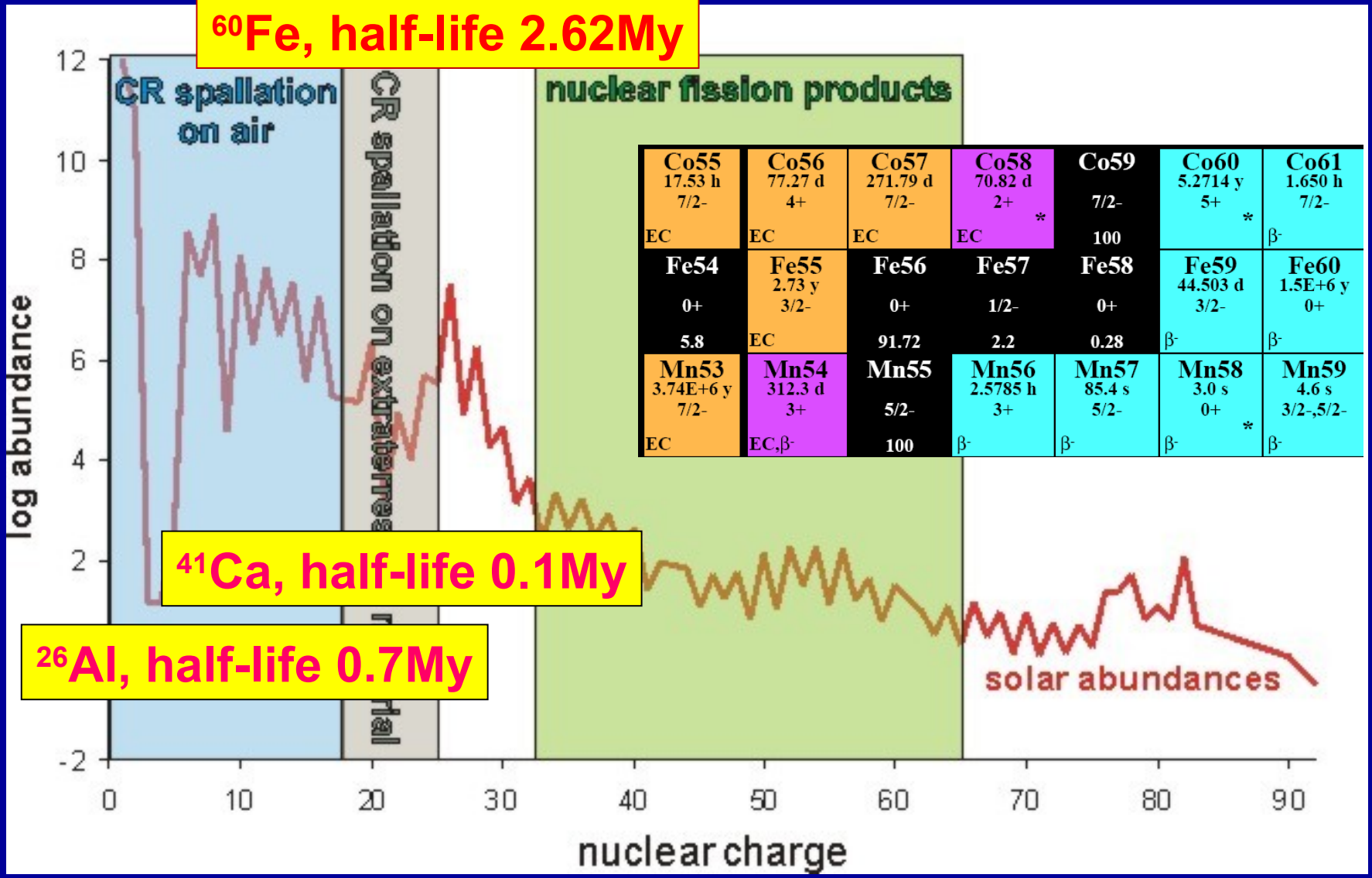
^{26}Al, T1/2 = 0.7 My

^{41}Ca,T1/2 = 0.1 My

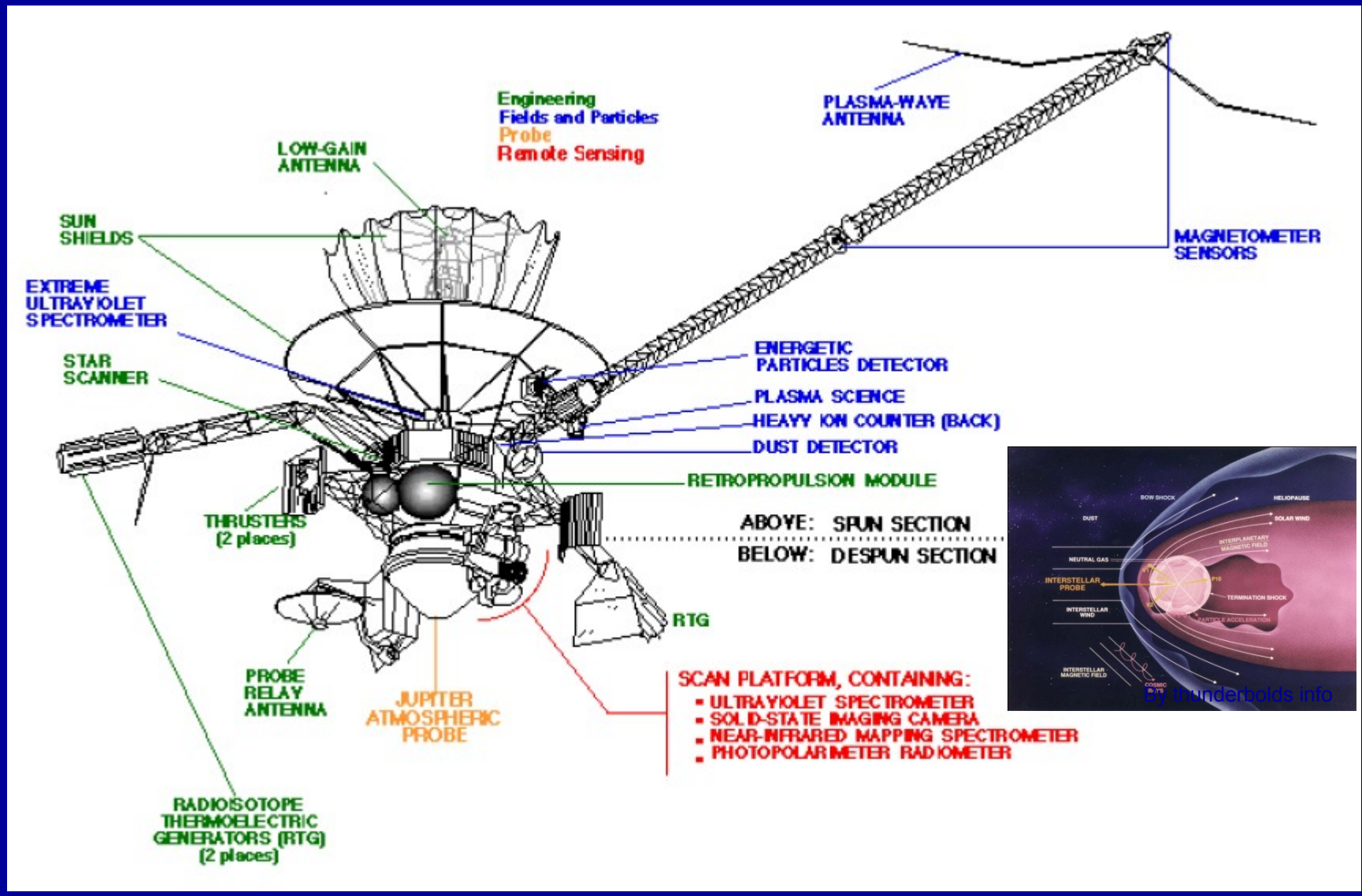
^{60}Fe, T1/2 = 2.62 My

Paulette C. Liewer et al. 1999, JPL NASA

Which isotope exhibits a clear signature?

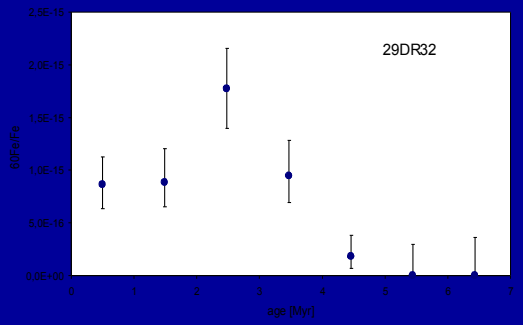


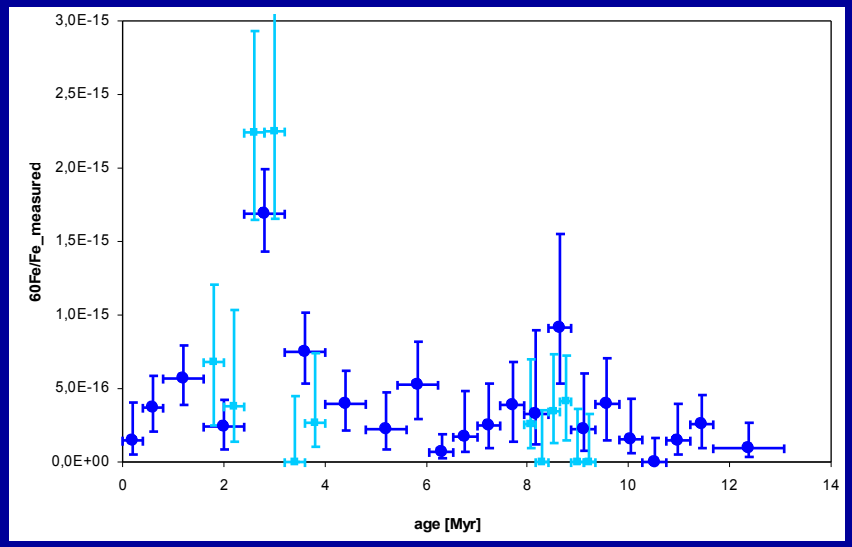
Galileo, Ulysses, Cassini ⟹ ISD grains (mean ≈ 1.6×10^{-16} kg), flux = 2.5×10^{-5} m^{-2} s^{-1} (Altobelli et al., 2003)



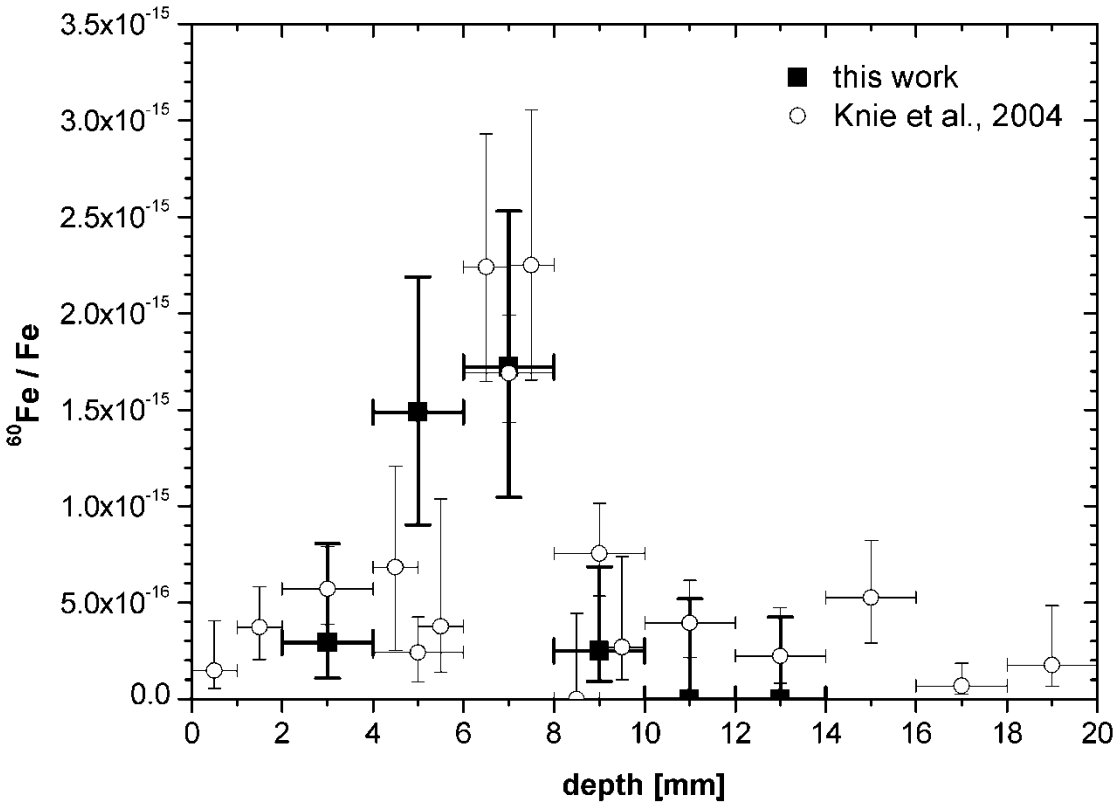
Where can we find ISM?

^{60}Fe in crusts





Leaching of the crust



The Moon collects 100% of the dust!

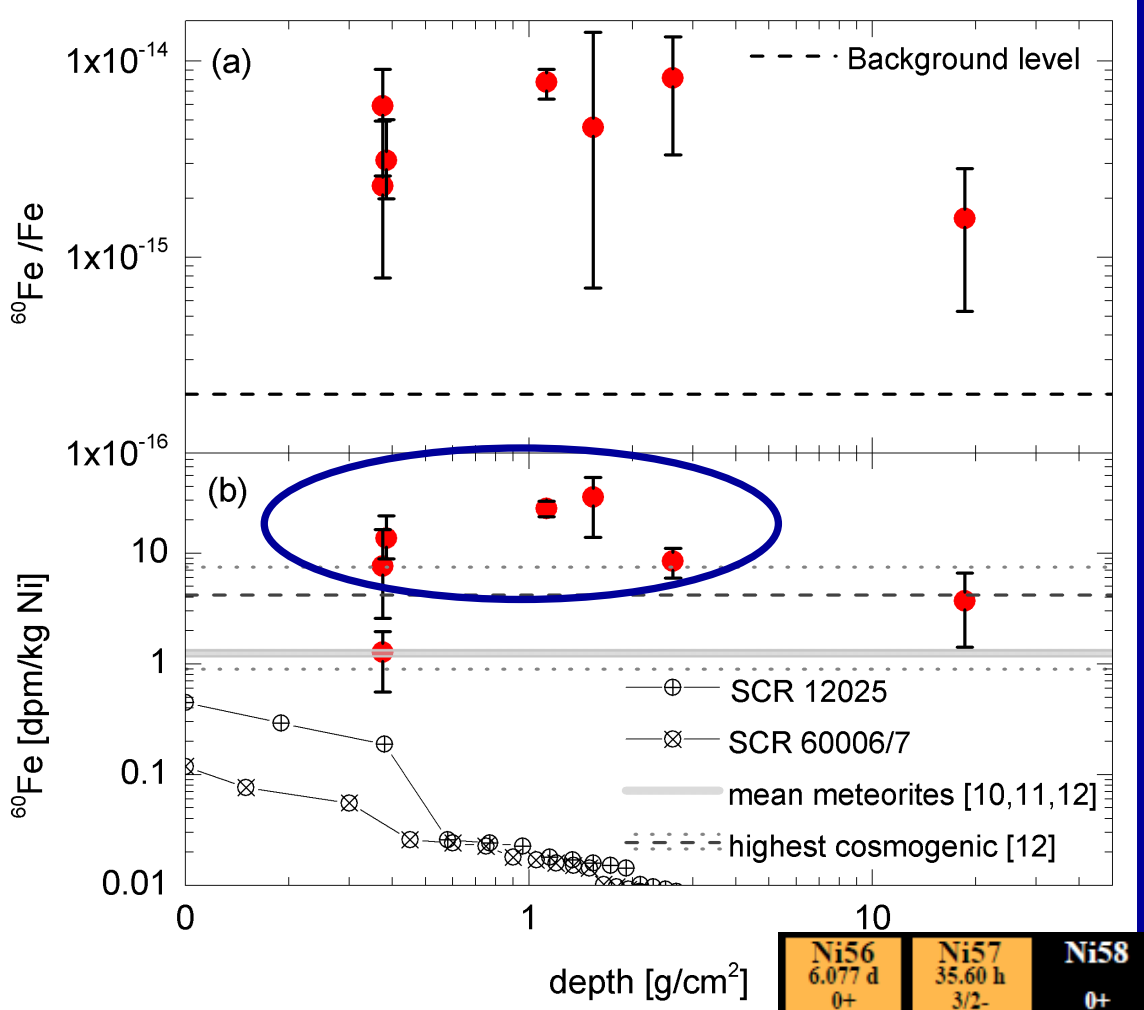
In collaboration with:

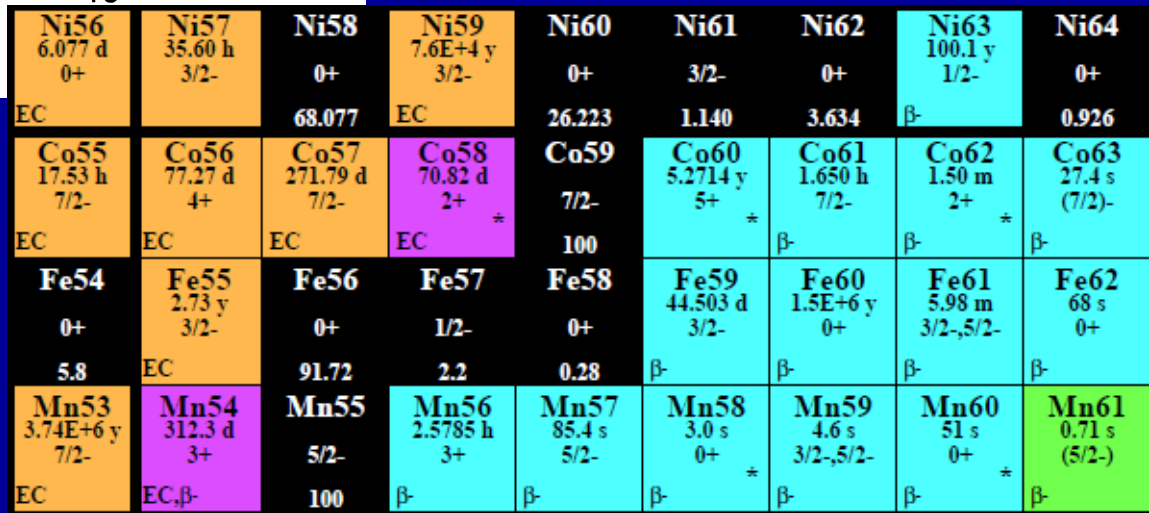
G. Herzog et al.
Rutgers University

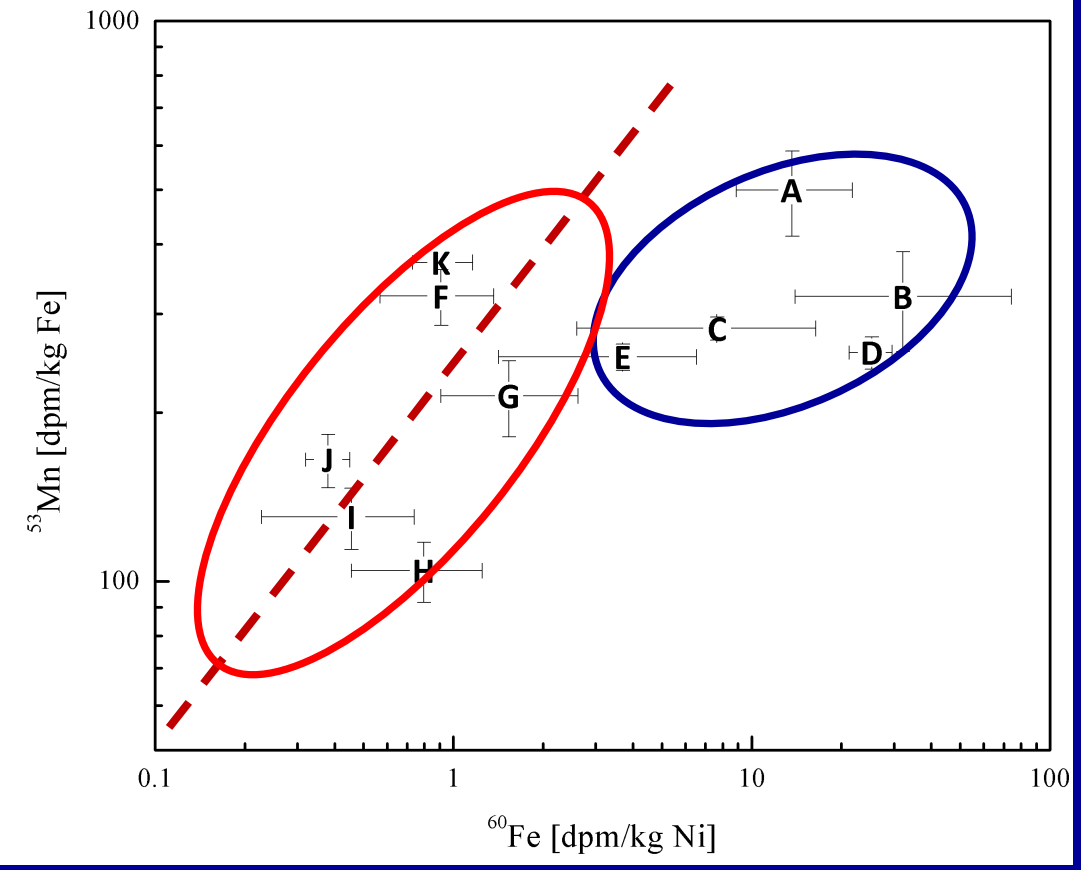
NASA

The Moon as a storage:

a. No dilution by sedimentation
b. 100% uptake
c. but gardening effect
d. cosmic ray activation
   (background!)
e. poor sampling


(a)
Background level
1x10-14
1x10-15
1x10-16
60Fe /Fe
(b)
10
1
0.1
0.01
60Fe [dpm/kg Ni]
SCR 12025
SCR 60006/7
mean meteorites [10,11,12]
highest cosmogenic [12]
0
1
10
depth [g/cm2]


Ni56 6.077 d 0+ EC
Ni57 35.60 h 3/2-
Ni58 0+ 68.077
Ni59 7.6E+4 y 3/2- EC
Ni60 0+ 26.223
Ni61 3/2- 1.140
Ni62 0+ 3.634
Ni63 100.1 y 1/2- β-
Ni64 0+ 0.926
Co55 17.53 h 7/2- EC
Co56 77.27 d 4+ EC
Co57 271.79 d 7/2- EC
Co58 70.82 d 2+ * EC
Co59 7/2- 100
Co60 5.2714 y 5+ * β-
Co61 1.650 h 7/2- β-
Co62 1.50 m 2+ * β-
Co63 27.4 s (7/2)- β-
Fe54 0+ 5.8
Fe55 2.73 y 3/2- EC
Fe56 0+ 91.72
Fe57 1/2- 2.2
Fe58 0+ 0.28
Fe59 44.503 d 3/2- β-
Fe60 1.5E+6 y 0+ β-
Fe61 5.98 m 3/2-,5/2- β-
Fe62 68 s 0+ β-
Mn53 3.74E+6 y 7/2- EC
Mn54 312.3 d 3+ EC,β-
Mn55 5/2- 100
Mn56 2.5785 h 3+ β-
Mn57 85.4 s 5/2- β-
Mn58 3.0 s 0+ * β-
Mn59 4.6 s 3/2-,5/2- β-
Mn60 51 s 0+ * β-
Mn61 0.71 s (5/2-) β-



Ni56 6.077 d 0+ EC	Ni57 35.60 h 3/2-	Ni58 0+ 68.077	Ni59 7.6E+4 y 3/2- EC	Ni60 0+ 26.223	Ni61 3/2- 1.140	Ni62 0+ 3.634	Ni63 100.1 y 1/2- β-	Ni64 0+ 0.926
Co55 17.53 h 7/2- EC	Co56 77.27 d 4+ EC	Co57 271.79 d 7/2- EC	Co58 70.82 d 2+ * EC	Co59 7/2- 100	Co60 5.2714 y 5+ * β-	Co61 1.650 h 7/2- β-	Co62 1.50 m 2+ * β-	Co63 27.4 s (7/2)- β-
Fe54 0+ 5.8	Fe55 2.73 y 3/2- EC	Fe56 0+ 91.72	Fe57 1/2- 2.2	Fe58 0+ 0.28	Fe59 44.503 d 3/2- β-	Fe60 1.5E+6 y 0+ β-	Fe61 5.98 m 3/2-,5/2- β-	Fe62 68 s 0+ β-
Mn53 3.74E+6 y 7/2- EC	Mn54 312.3 d 3+ EC,β-	Mn55 5/2- 100	Mn56 2.5785 h 3+ β-	Mn57 85.4 s 5/2- β-	Mn58 3.0 s 0+ * β-	Mn59 4.6 s 3/2-,5/2- β-	Mn60 51 s 0+ * β-	Mn61 0.71 s (5/2-) β-

5.Next:Radioisotopes of Interest

Isotope	Half-Life (t1/2)	
3H	12.3 a	
10Be	1.5 Ma	
14C	5730 a	
26Al	720 ka	
32Si	140 a	
36Cl	301 ka	
41Ca	103 ka	
44Ti	60 ka	
53Mn	3.7 Ma	
55Fe	2.7 a	
60Fe	2.6 Ma	
59Ni	76 ka	
63Ni	100 a	
79Se	480 ka	
90Sr	28.5 a	
107Pd	6.5 Ma	
126Sn	240 ka	
129I	15.7 Ma	
146Sm	100 Ma	(α)
148Gd	74.8 a	(α)
150Gd	1.8 Ma	(α)
154Dy	3 Ma	(α)
182Hf	9 Ma	
193Pt	50 a	
194Hg	520 a	
205Pb	15 Ma	
226Ra	1600 a	(α)
229Th	7880 a	(α)
230Th	75.4 ka	(α)
231Pa	32.8 ka	(α)
232U	69 a	(α)
233U	159 ka	(α)
234U	246 ka	(α)
235U	704 Ma	(α)
236U	23 Ma	(α)
238U	$4.5*10^9$ a	(α)
237Np	2.14 Ma	(α)
238Pu	88 a	(α)
239Pu	24 ka	(α)
240Pu	6.6 ka	(α)
241Pu	14.4 a	(α)
242Pu	375 ka	(α)
244Pu	80 Ma	(α)
???Xy	?? a	

This list is not complete!

colleagues:

T. Faestermann, (TUM), Germany
L. Fimiani, (TUM), Germany
K. Knie, (TUM, now GSI), Germany
M. Poutivtsev, (TUM), Germany
W. Rühm, Helmholtz, Germany
G. Rugel, (TUM), Germany
A. Wallner, (TUM, now ANU/Australia), Germany

+ collaborating institutions

Thank you!

- **Climate in the past**

Exposure Dating

(exposure of a surface to cosmic rays)

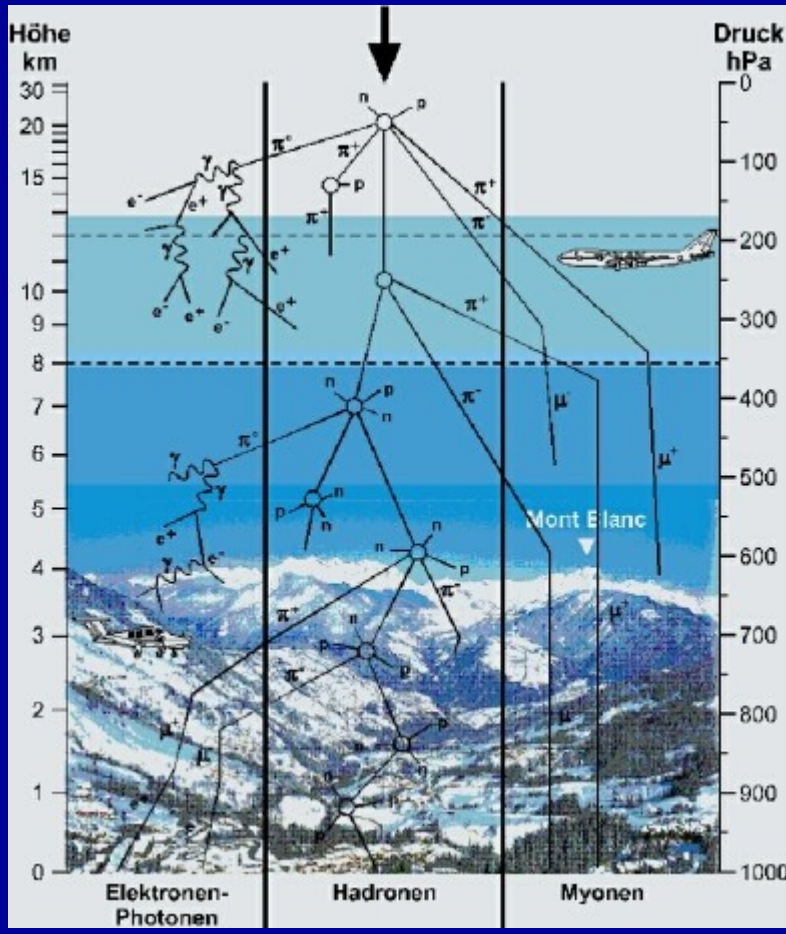
Production by cosmic radiation

Cosmic radiation:

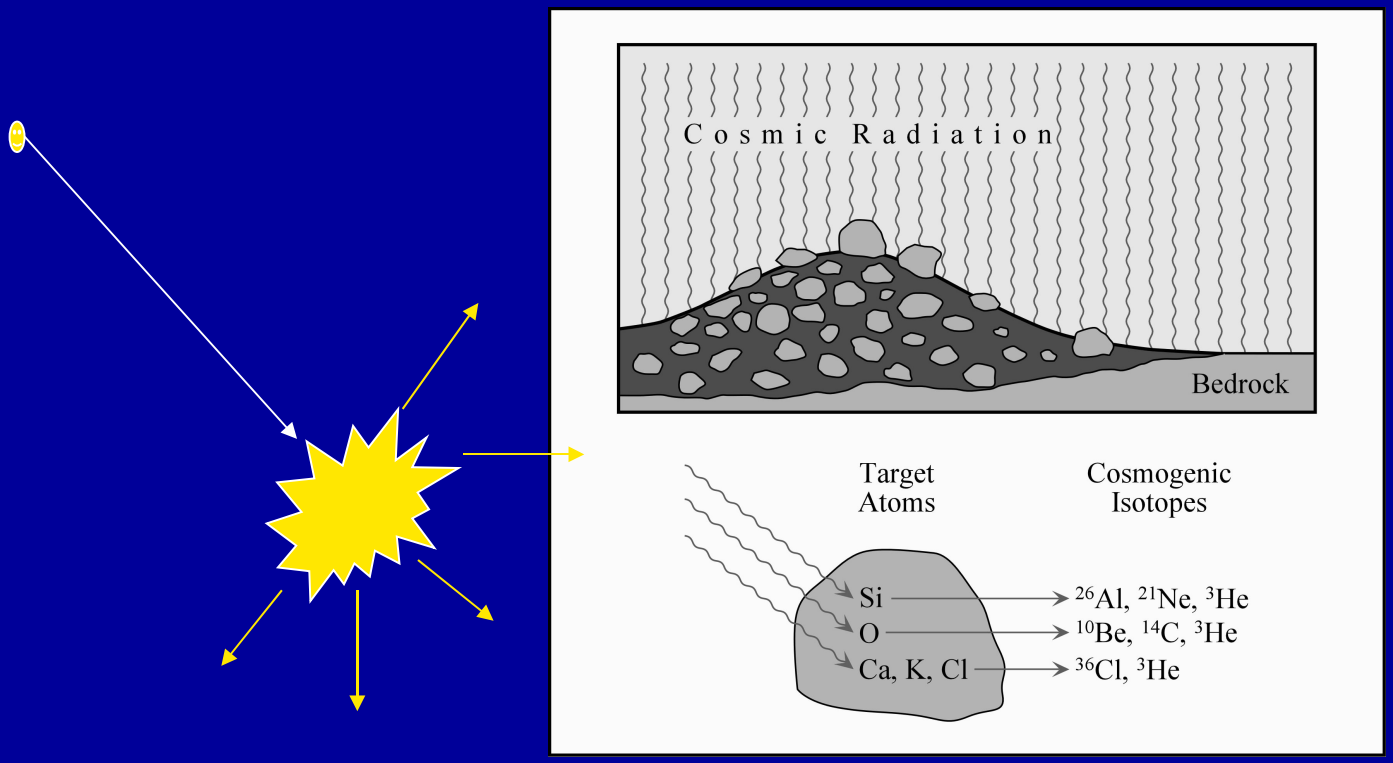
- Protons and neutrons
- Muons
- Electrons and photons
- Neutrinos

Cosmic radiation depends on the latitude and on the altitude

Cosmic rays produce isotopes by nuclear processes like spallation, neutron capture etc.

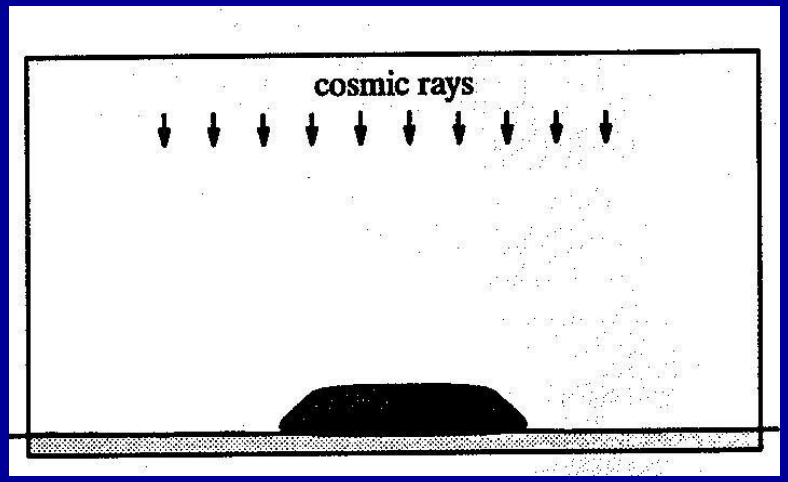


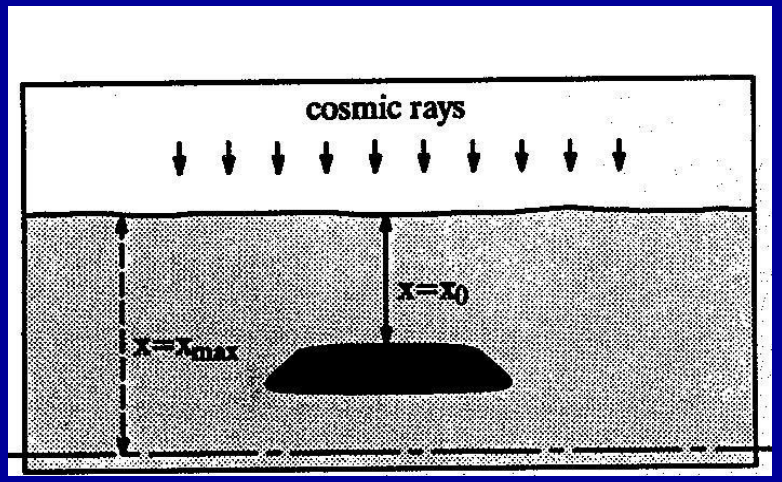
Spallation



- High-energy protons/neutrons hit the target
- Some nuclides split in fragments
- Fragments can be: ^{3}He, ^{10}Be, ^{26}Al,…

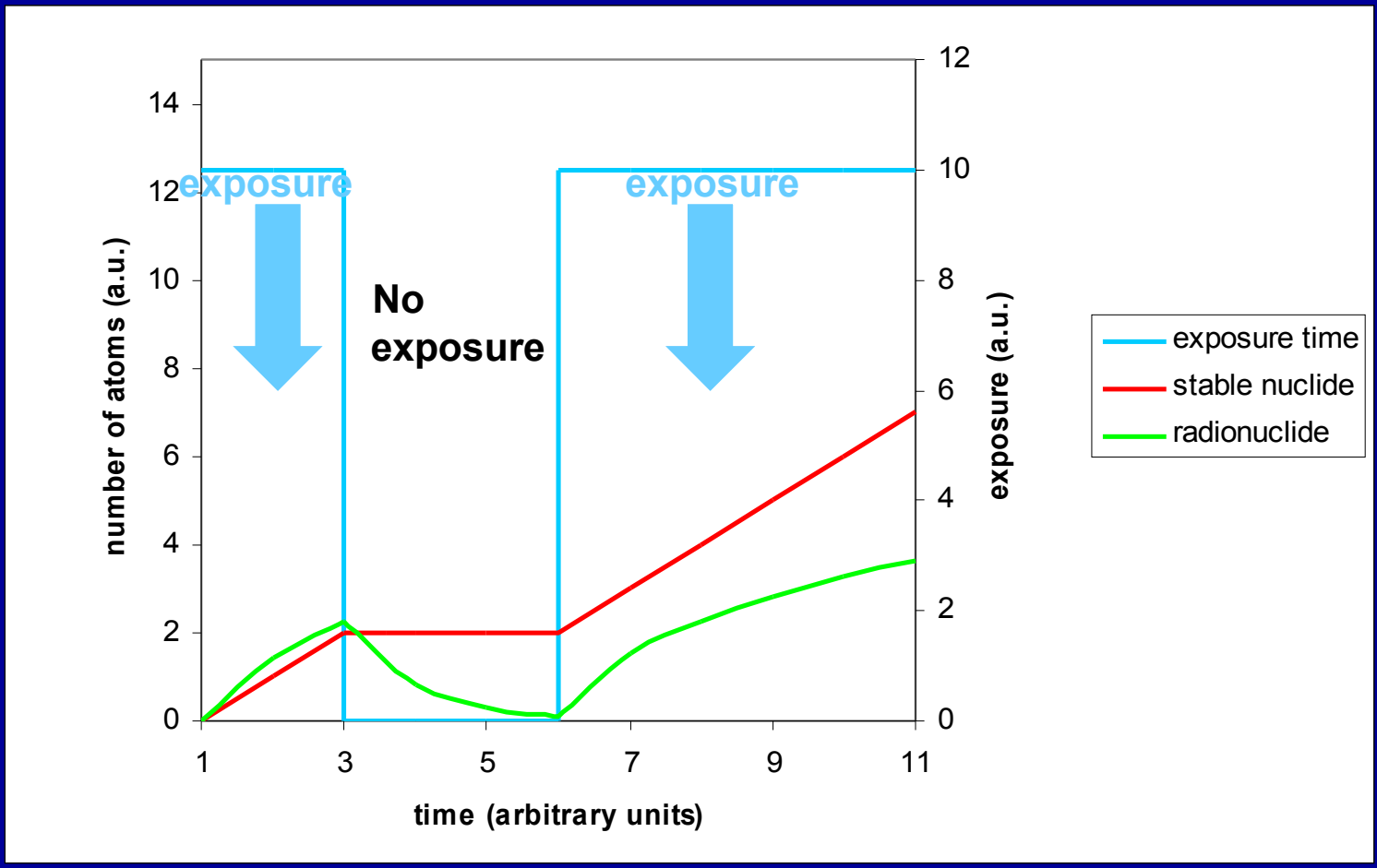
Shielding





- Radionuclides are mainly formed on surface because the cosmic ray flux is attenuated by the depth
- By any shielding of the rock (e.g. ice) the production of the nuclides is stopped

Difference:
Noble gas - Radionuclide



SED with different nuclides

SED=surface exposure dating

Stable: ^{3}He, ^{21}Ne, …

- Only information of the total exposure age

Unstable: ^{10}Be, ^{26}Al, ^{36}Cl, ^{53}Mn…

- Half-life

^{10}Be = 1.4 My

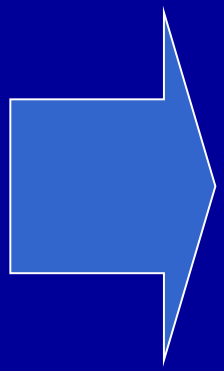
^{27}Al = 0.7 My

^{36}Cl = 0.3 My

^{53}Mn = 3.8 My

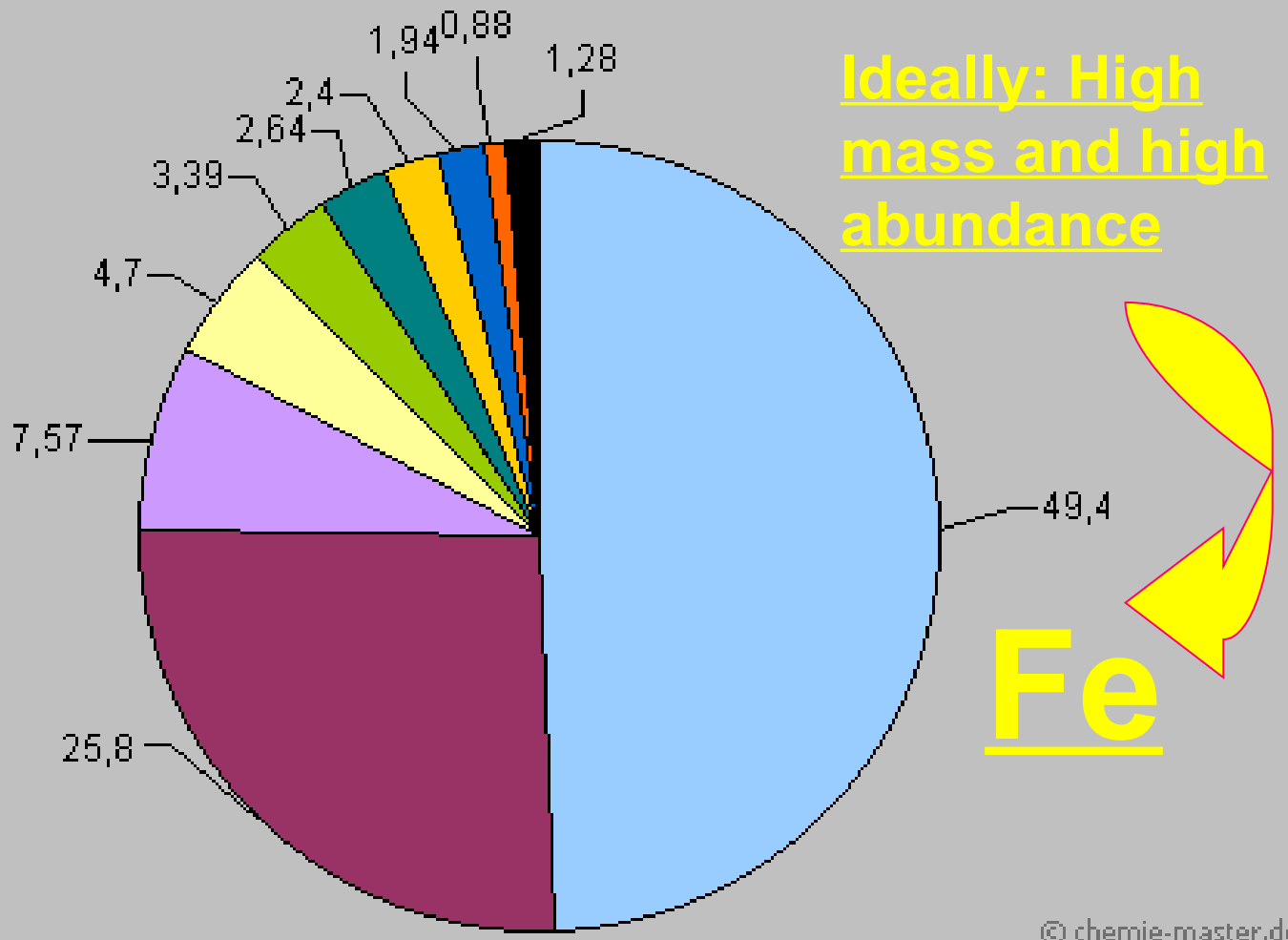
- Depending on the mineral
- Many possible target elements
- Mobility?

The merit of ^{53}Mn in comparison to isotopes like ^{3}He, ^{10}Be, ^{21}Ne, ^{26}Al, and ^{36}Cl is:



One (target) element (abundant!) and one production channel! Long half-life of $T_{1/2}$=3.8My

Composition of major chemical elements in the Earths' crust



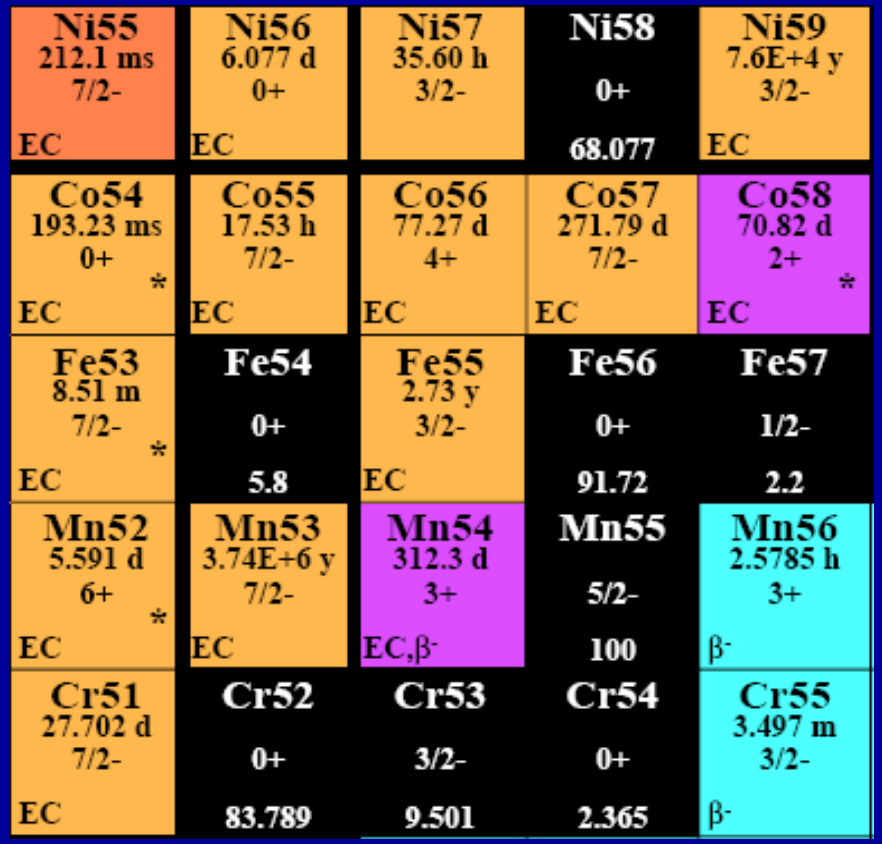
Ideally: High mass and high abundance

Fe

© chemie-master.de

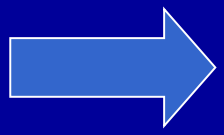
Production of ^{53}Mn

- ^{53}Mn is produced to 99% by spallation on iron on the earth‘s surface
- Remaining 1%: nickel and copper
- Mn production is dependent on the location (altitude, latitude) of exposure



Difficulty in measuring ^{53}Mn:

Mn does not form negative ions; but Cr does!



therefore:

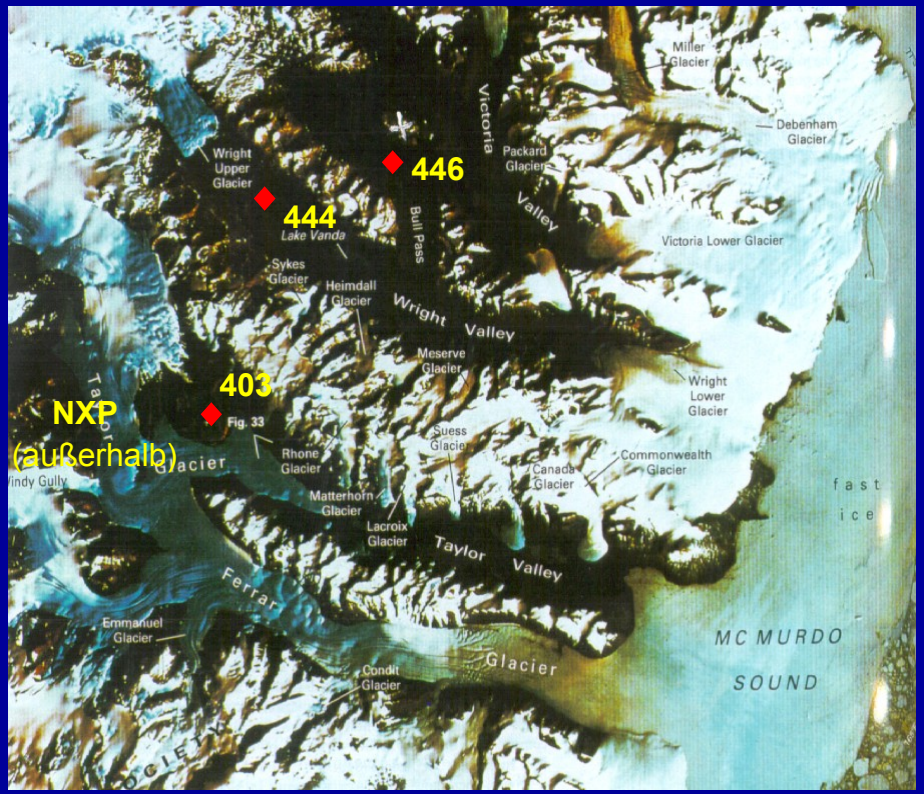
MnO^- (but CrO^- similar like MnO^-)

but we found: $MnF^-/CrF^- \approx 10/1$

Sample origin

Antarctica

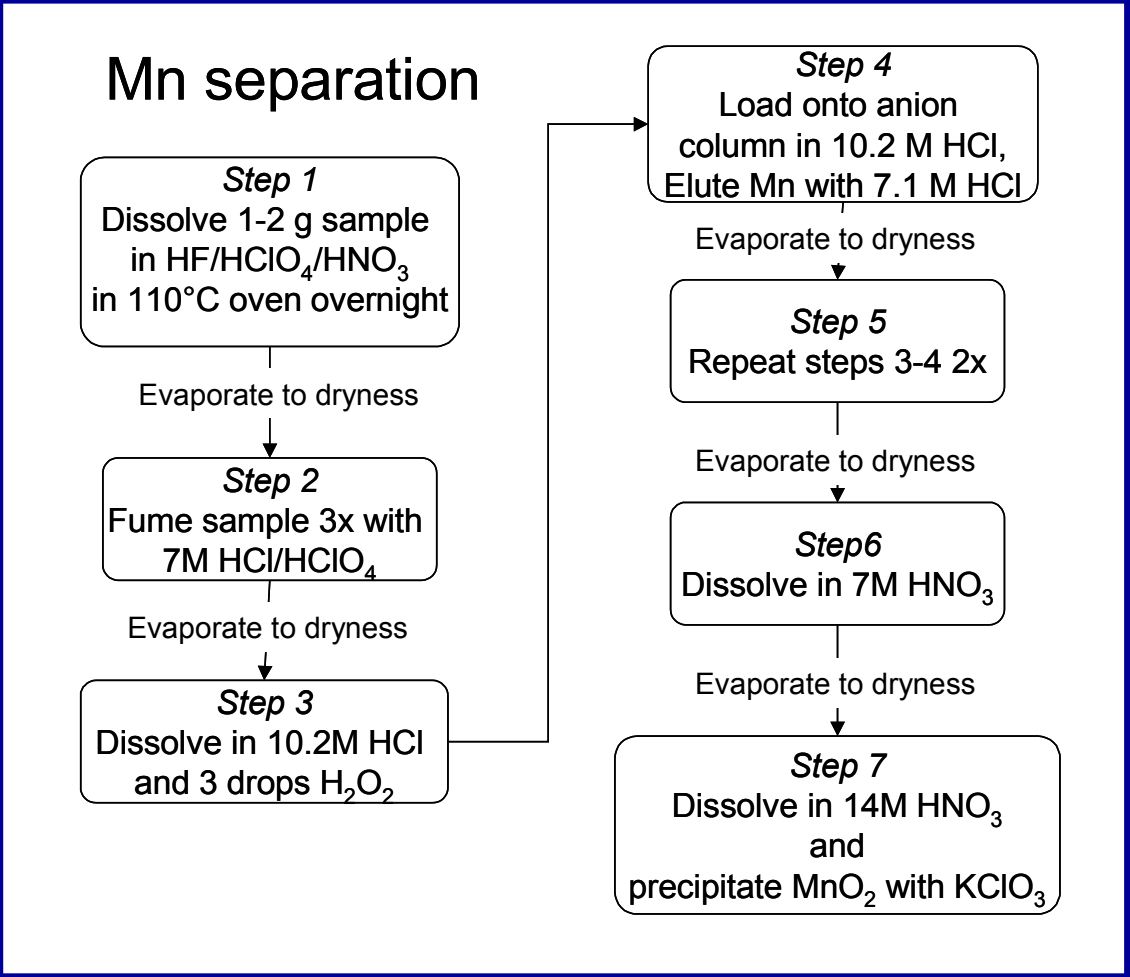
Sample origin



Map of Dry Valley Region, Antarctica
with sample location

Dolerite boulders, Mt. Feather, Antarctica
(NXP sample location)

Chemistry


Mn separation
Step 1
Dissolve 1-2 g sample in HF/HClO4/HNO3 in 110°C oven overnight
Evaporate to dryness
Step 2
Fume sample 3x with 7M HCl/HClO4
Evaporate to dryness
Step 3
Dissolve in 10.2M HCl and 3 drops H2O2
Step 4
Load onto anion column in 10.2 M HCl, Elute Mn with 7.1 M HCl
Evaporate to dryness
Step 5
Repeat steps 3-4 2x
Evaporate to dryness
Step6
Dissolve in 7M HNO3
Evaporate to dryness
Step 7
Dissolve in 14M HNO3 and precipitate MnO2 with KClO3

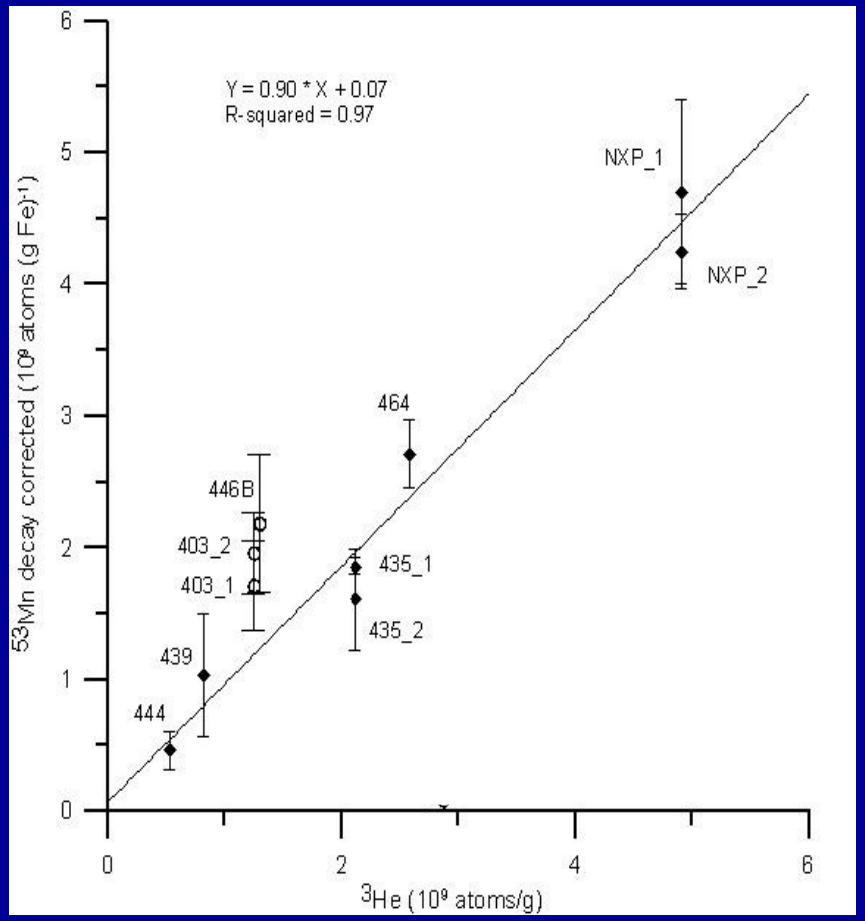
Results

- Linear dependency
- Production rates for manganese:
  - By means of ^{53}Mn and the exposure ages of neon (exp)
  - Calculations with a model (th)

Production rates at sea level and high latitudes (Antarctica):

$P_{53}(exp)=110\pm13$ atoms yr^{-1} $(g\ Fe)^{-1}$

$P_{53}(th) =120\pm18$ atoms yr^{-1} $(g\ Fe)^{-1}$

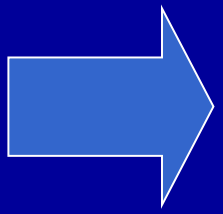


Surface exposure dating with ^{53}Mn:

- Rocks dated from 1.2My to 7.6My
- Determination of Earth surface processes including exposure and shielding periods (timescale 10 My)

Future application of ^{53}Mn in Antarctic

How old is the Antarctic ice shield?



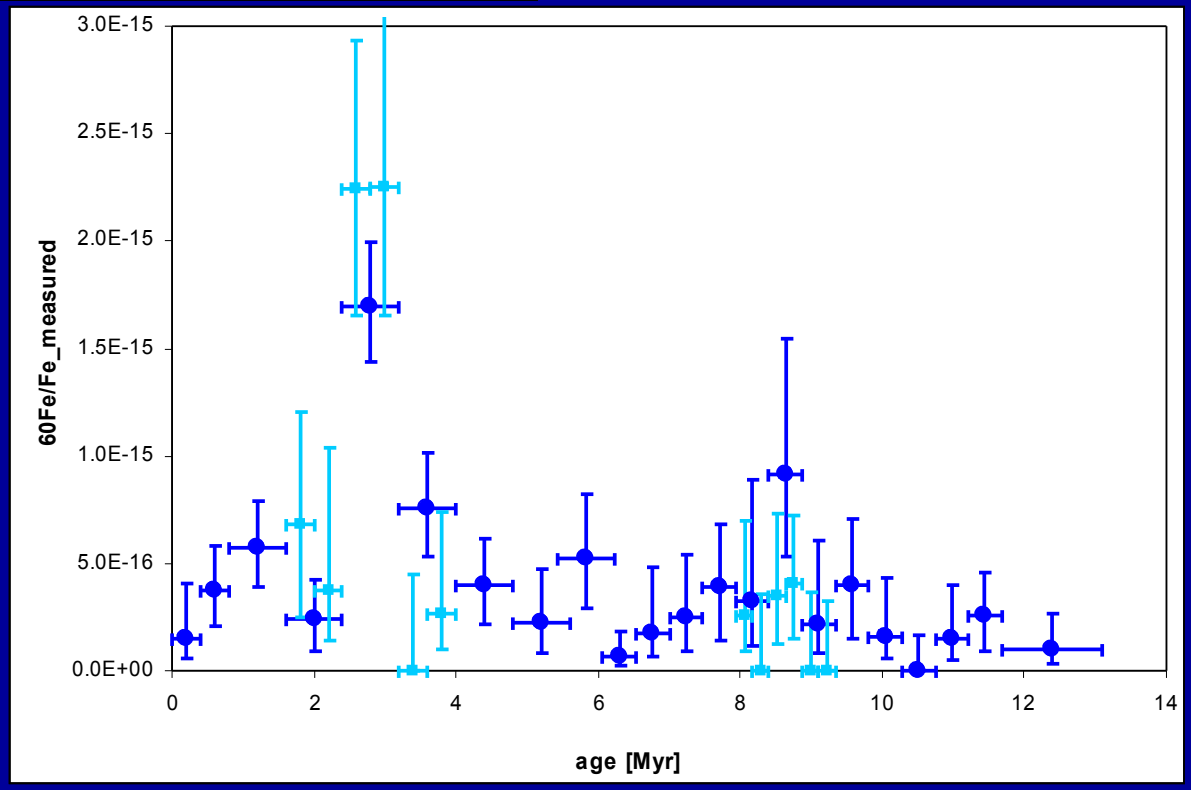
^{53}Mn determination from below the ice shield!
(same for Greenland)

In collaboration with:

J. Schaefer et al. Columbia University

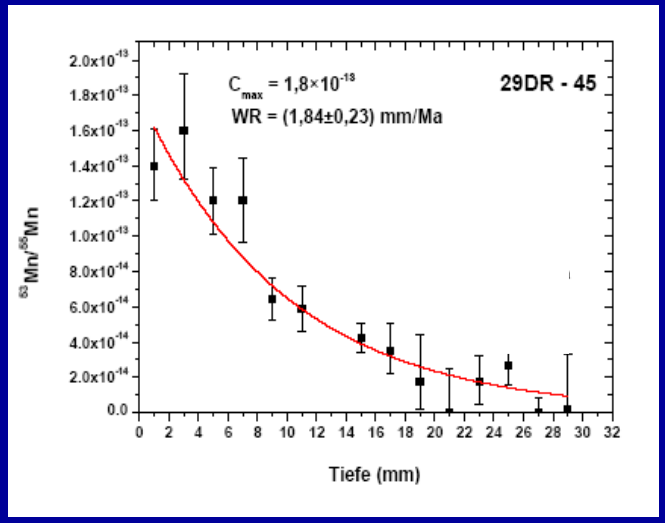
G. Herzog et al. Rutgers University

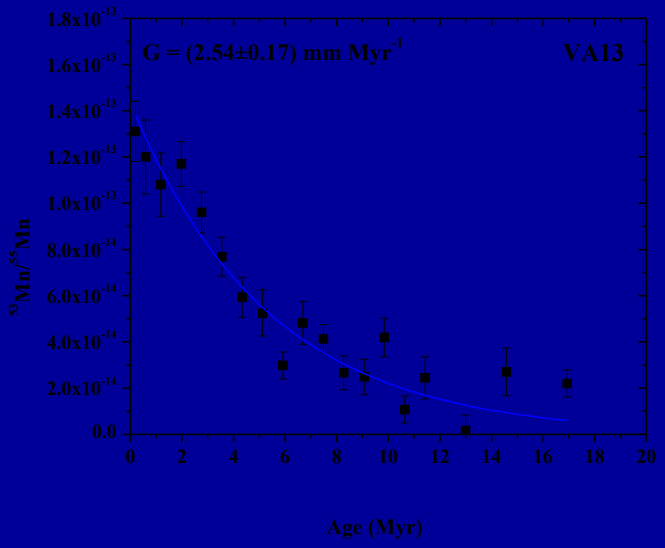
Co55 17.53 h 7/2- EC	**Co56** 77.27 d 4+ EC	**Co57** 271.79 d 7/2- EC	**Co58** 70.82 d 2+ * EC	**Co59** 7/2- 100	**Co60** 5.2714 y 5+ * β-	**Co61** 1.650 h 7/2- β-
Fe54 0+ 5.8	**Fe55** 2.73 y 3/2- EC	**Fe56** 0+ 91.72	**Fe57** 1/2- 2.2	**Fe58** 0+ 0.28	**Fe59** 44.503 d 3/2- β-	**Fe60** 1.5E+6 y 0+ β-
Mn53 3.74E+6 y 7/2- EC	**Mn54** 312.3 d 3+ EC,β-	**Mn55** 5/2- 100	**Mn56** 2.5785 h 3+ β-	**Mn57** 85.4 s 5/2- β-	**Mn58** 3.0 s 0+ * β-	**Mn59** 4.6 s 3/2-,5/2- β-

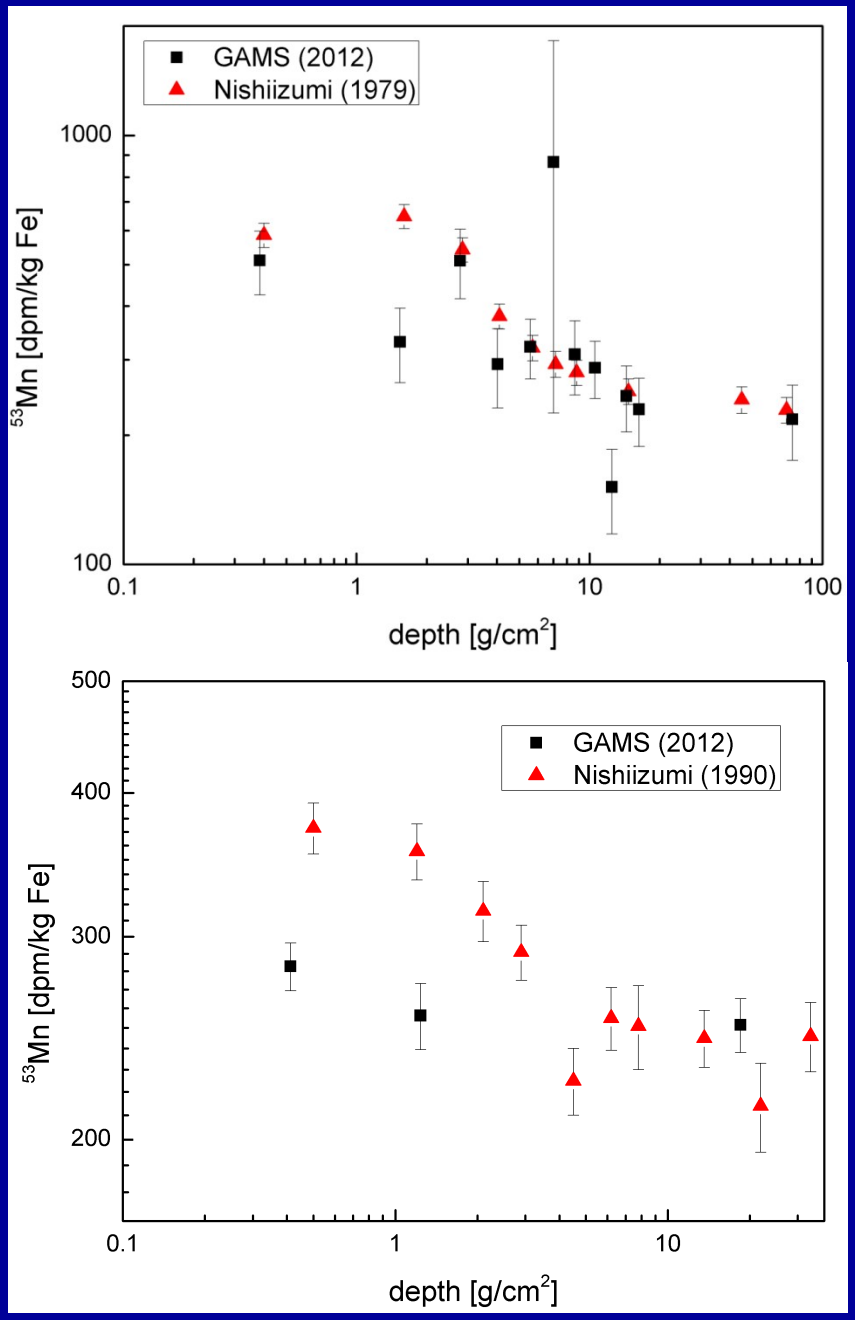


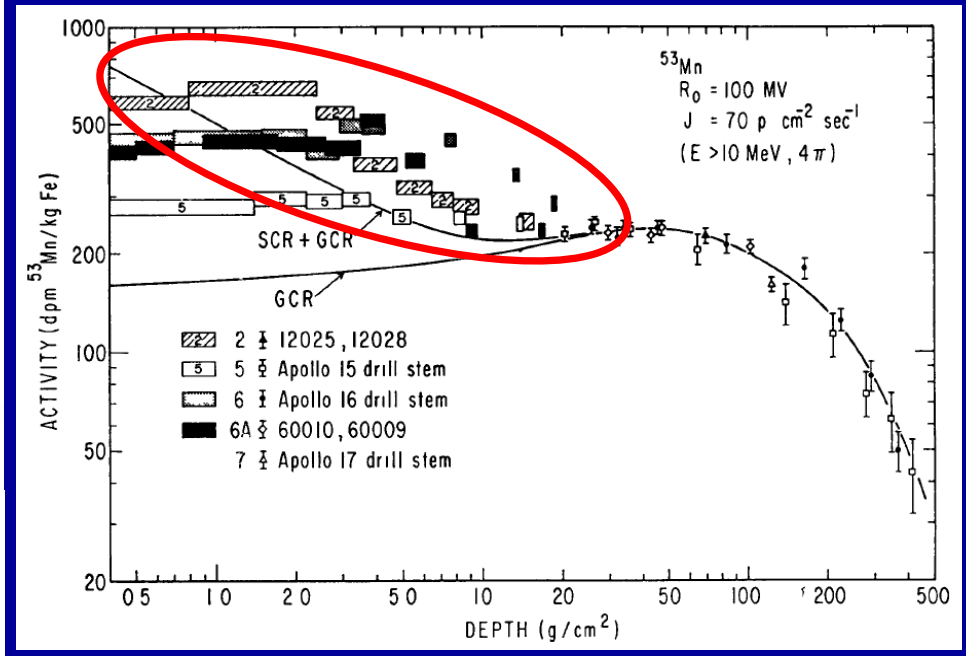
K.Knie et. al. PRL, 2004

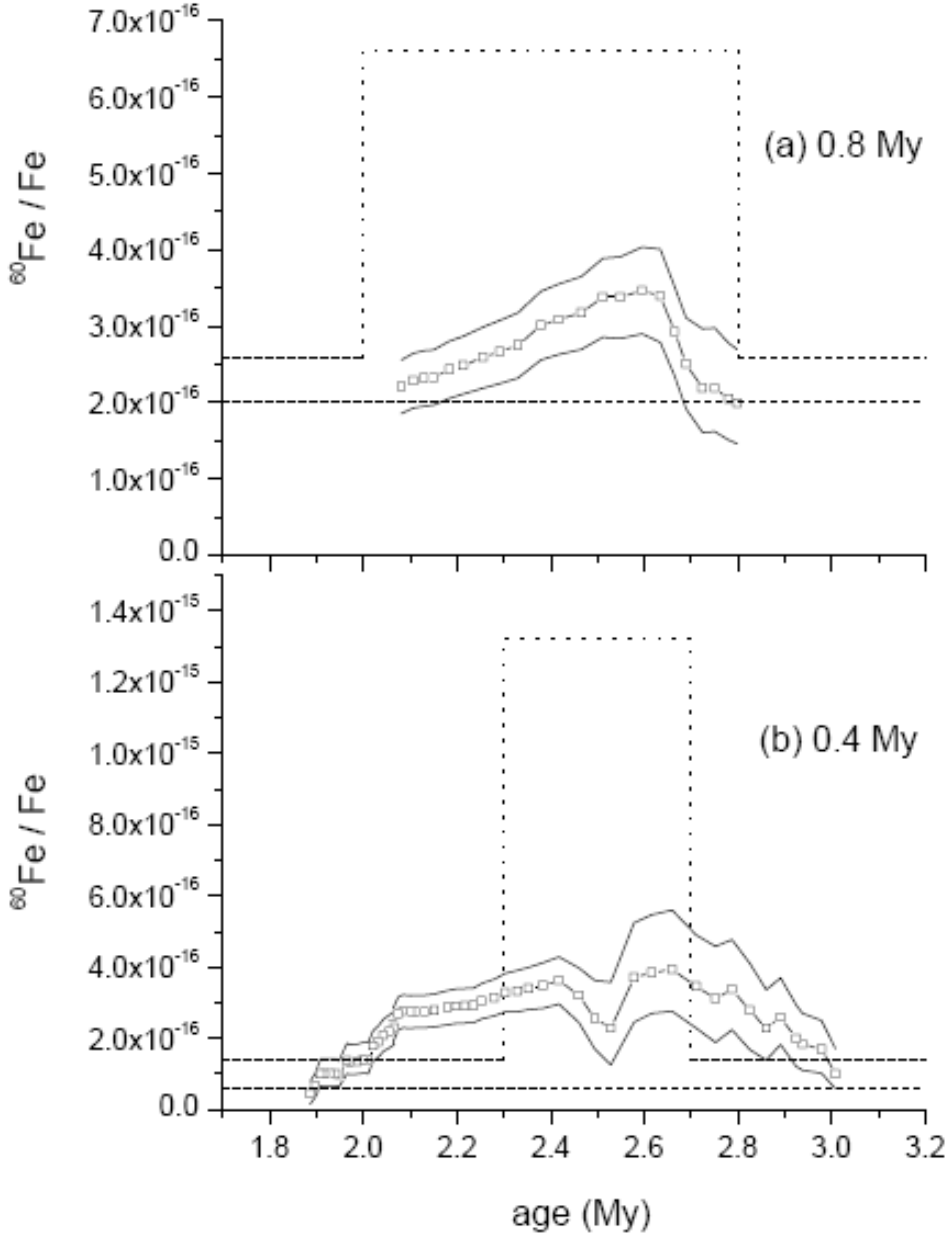
^{53}Mn in another crust

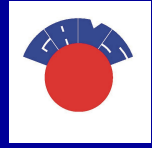





GAMS (2012)
Nishiizumi (1979)
1000
100
0.1
1
10
100
^{53}Mn [dpm/kg Fe]
depth [g/cm^2]
500
400
300
200
GAMS (2012)
Nishiizumi (1990)
0.1
1
10
^{53}Mn [dpm/kg Fe]
depth [g/cm^2]


^{53}Mn
R_0 = 100 MV
J = 70 p cm^{-2} sec^{-1}
(E >10 MeV, 4π)
SCR + GCR
GCR
2 ‡ 12025, 12028
5 ‡ Apollo 15 drill stem
6 ‡ Apollo 16 drill stem
6A ‡ 60010, 60009
7 ‡ Apollo 17 drill stem
ACTIVITY (dpm ^{53}Mn/kg Fe)
DEPTH (g/cm^2)
1000
500
200
100
50
20
0.5
1.0
2.0
5.0
10
20
50
100
200
500


7.0x10-16
6.0x10-16
5.0x10-16
4.0x10-16
3.0x10-16
2.0x10-16
1.0x10-16
0.0
60Fe / Fe
(a) 0.8 My
1.4x10-15
1.2x10-15
1.0x10-15
8.0x10-16
6.0x10-16
4.0x10-16
2.0x10-16
0.0
60Fe / Fe
(b) 0.4 My
1.8
2.0
2.2
2.4
2.6
2.8
3.0
3.2
age (My)



Ionization Chambers

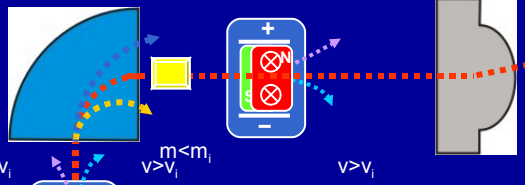
Gas-filled Analyzing Magnet System

90˚ Analyzing Magnet (2nd Mass Separation)

Wien Filter

$m>m_i$

$v<v_i$



$m<m_i$

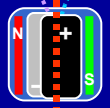
$v<v_i$

$v>v_i$

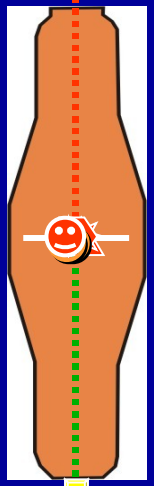
$v>v_i$

Faraday Cup

Wien Filter



Switching Magnet

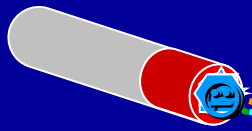


Positive Ions

Negative Ions

Carbon Stripper Foil

(Cs Sputter) Negative Ion Source



18˚ Electrostatic Deflection

$m<m_i$

$m>m_i$

Sample

90˚ Injector Magnet (1st Mass Separation)