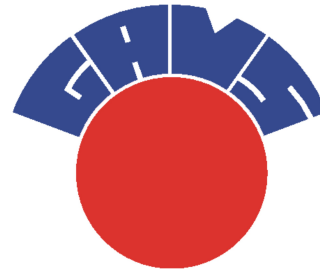
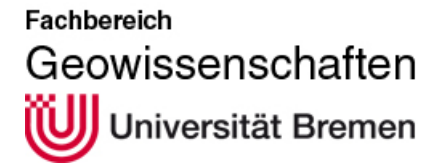


^{60}Fe supernova-searches with AMS



Peter Ludwig¹
Russbach School
March 2014



Shawn Bishop¹, Ramon Egli², Valentyna Chernenko¹,

Thomas Faestermann¹, Leticia Fimiani¹, Jose Gomez¹, Karin Hain¹, Gunther Korschinek¹
Nicolai Famulok¹, Thomas Frederichs³, Marianne Hanzlik⁴, Silke Merchel⁵, Georg Rugel⁵

¹ Physik Department, Technische Universität München

² Zentralanstalt für Meteorologie und Geodynamik, Wien

³ Fachbereich Geowissenschaften, Universität Bremen

⁴ Fakultät für Chemie, Technische Universität München

⁵ Helmholtz Zentrum Dresden Rossendorf

- Accelerator Mass Spectrometry (AMS)
 - Basics
 - AMS isotopes
 - Applications
- AMS Setups at MLL in Garching
- Recent example: Search for supernova ^{60}Fe
 - in ferromanganese crusts
 - in microfossils
 - on the moon
- Summary and outlook

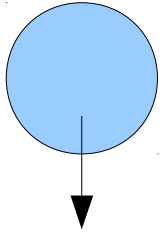
Accelerator Mass Spectrometry (AMS) is a high-sensitivity ion counting technique, primarily used for determination of isotopic ratios involving long-lived radionuclides

- I will not be talking about ^{14}C table top machines !
- Tandem-accelerator based systems → complete suppression of molecular background
- High energies (100-200 MeV) → nuclear physics particle identification techniques
- Sensitivity can reach down to isotopic ratios of 10^{-16}

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- High energies (100-200 MeV) → nuclear physics particle identification techniques
- Sensitivity can reach down to isotopic ratios of 10^{-16}
- Only few milligrams of sample material required (however: destructive!)
- Isotopic ratios measured → systematic errors (ion source efficiency, ...) cancelled
- Challenge: Suppression of isobaric background, e.g. ^{60}Ni , ^{60}Fe or ^{53}Cr , ^{53}Mn , ...
- AMS Facility used for all measurements in this study: Maier-Leibnitz-Laboratory Garching

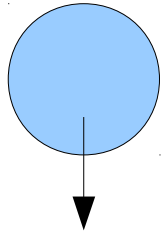
Sample (Element X)



Radioisotope ${}^n\text{X}$

Tiny fraction: ${}^n\text{X}/\text{X} = 10^{-12} \dots 10^{-17}$

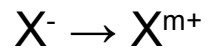
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Radioisotope ${}^n\text{X}$

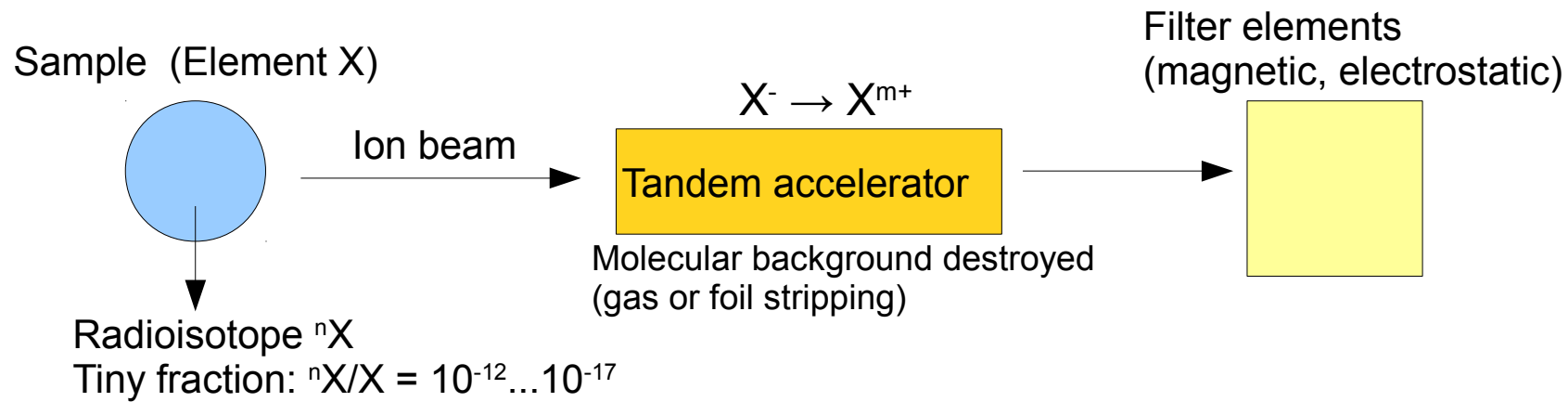
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Ion beam

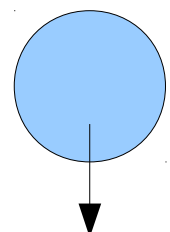


Tandem accelerator

Molecular background destroyed
(gas or foil stripping)

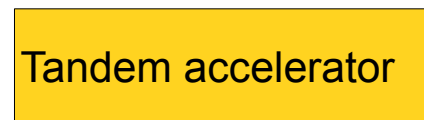


Sample (Element X)



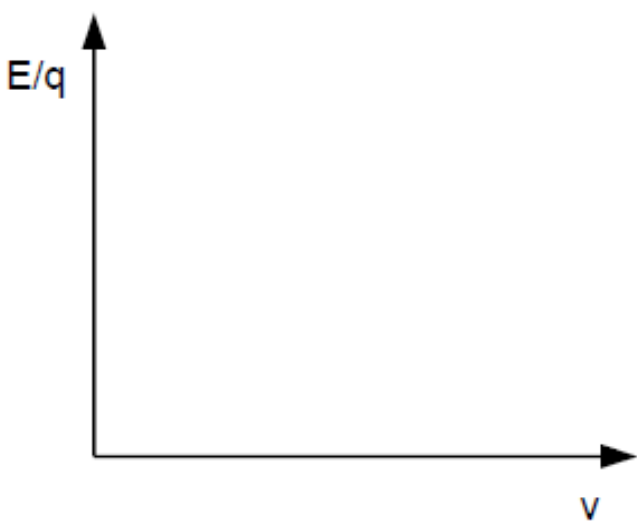
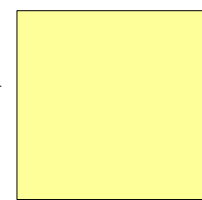
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Ion beam



$\text{X}^- \rightarrow \text{X}^{m+}$
Molecular background destroyed
(gas or foil stripping)

Filter elements
(magnetic, electrostatic)



1.) Magnetic analyzer
(typically 90 degree dipole magnet)

— $E(v) = \frac{1}{2} q r B \cdot v \sim v$

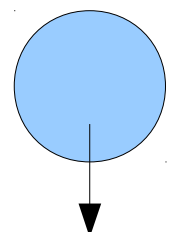
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3.) Wien-Filter

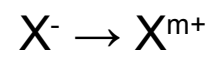
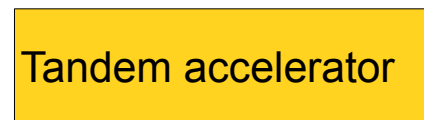
— $v = \frac{U}{B d} \sim const$

Sample (Element X)



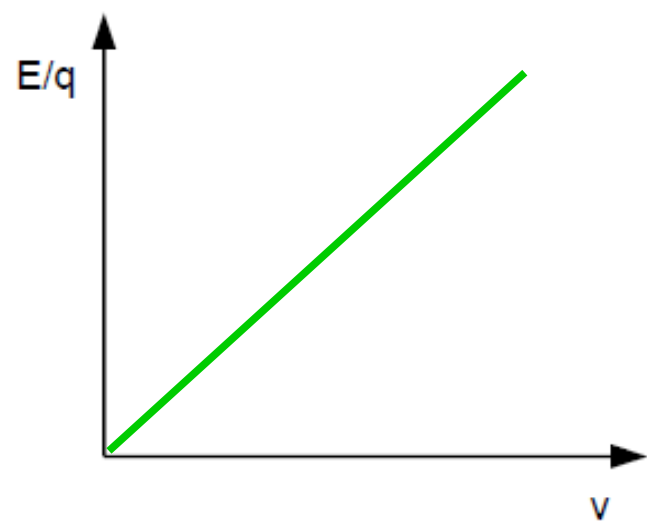
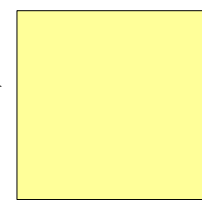
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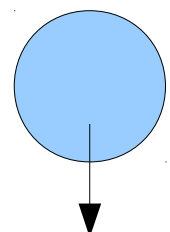
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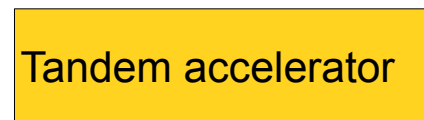
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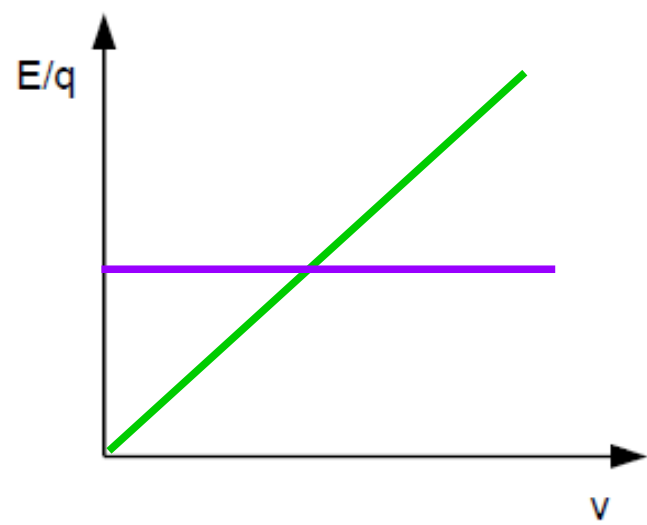
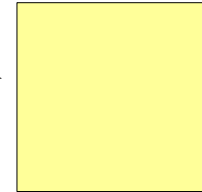
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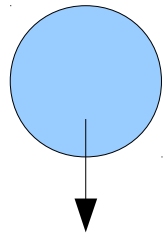
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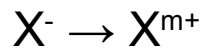
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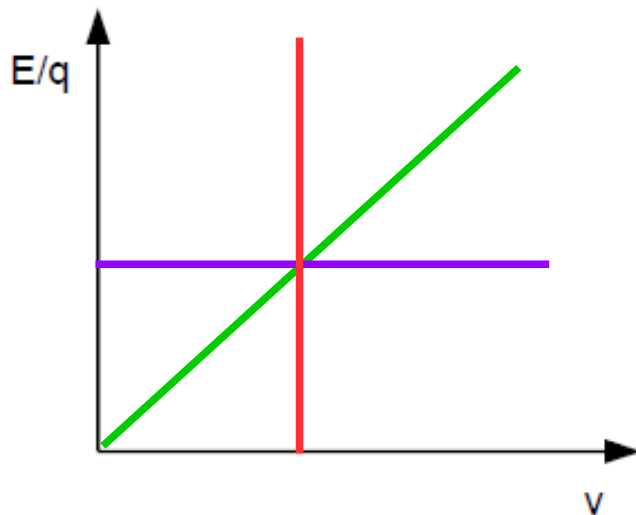
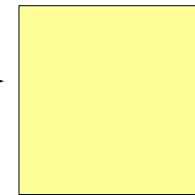
Ion beam



Tandem accelerator

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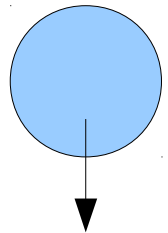
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Sample (Element X)

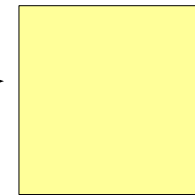


Ion beam

Tandem accelerator

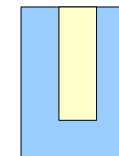
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Filter elements
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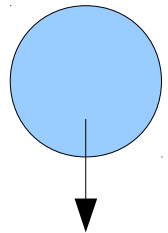
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Faraday cup:
Number of stable X

Sample (Element X)

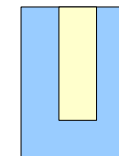
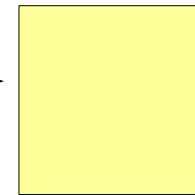


Ion beam

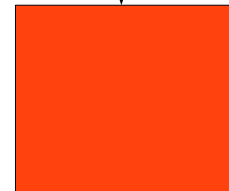
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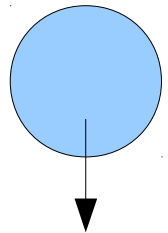
Faraday cup:
Number of stable X



Detector (Particle Identification) e.g. TOF, E, E loss
→ number of ${}^n\text{X}$

Radioisotope ${}^n\text{X}$
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Sample (Element X)



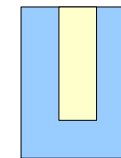
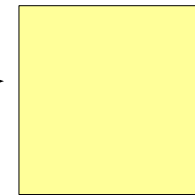
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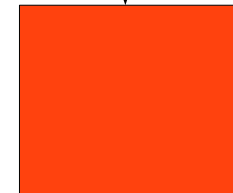
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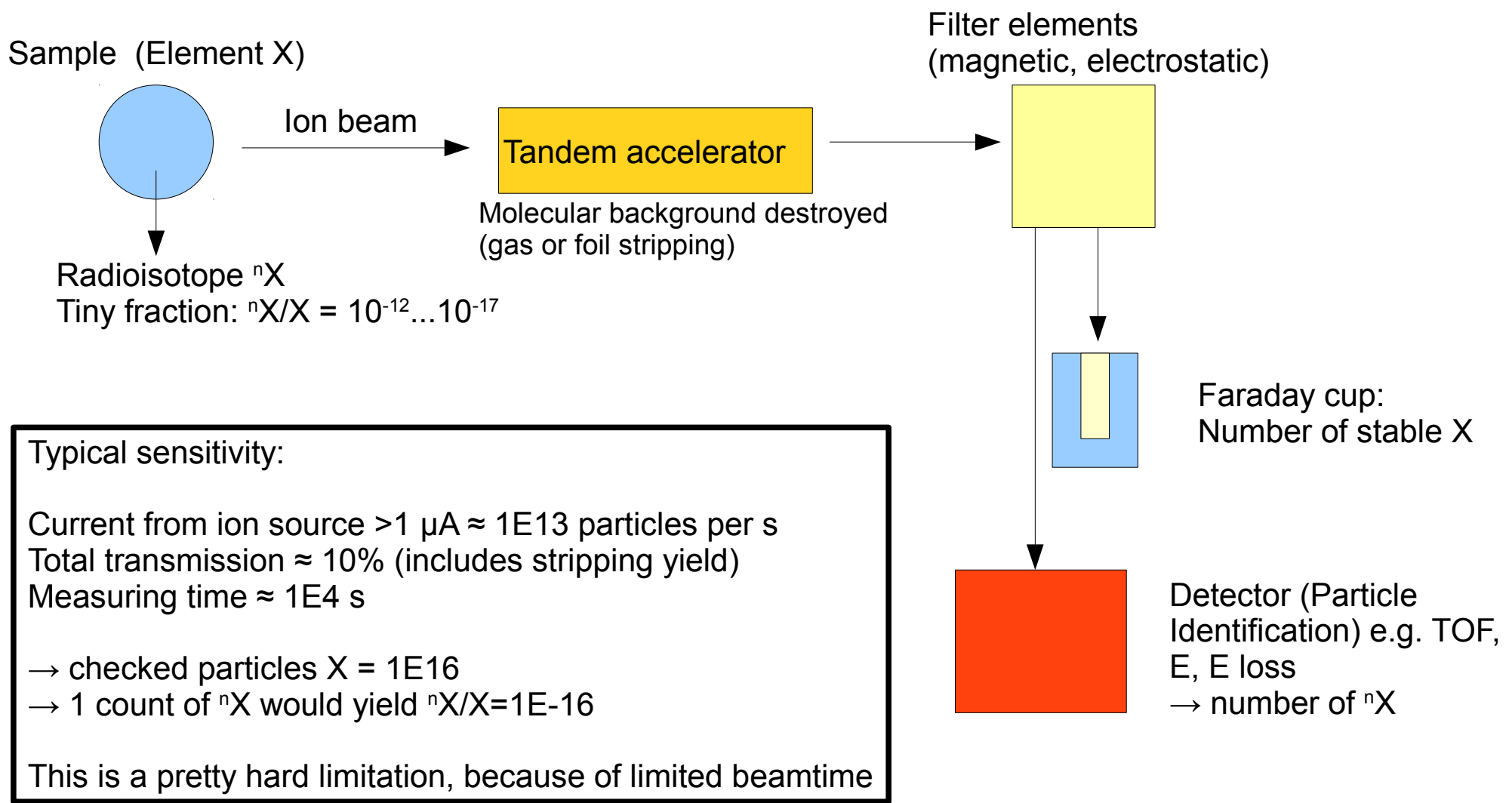
Faraday cup:
Number of stable X



Detector (Particle
Identification) e.g. TOF,
E, E loss
→ number of ${}^n\text{X}$

Problem: Transmission from Faraday cup to detector

- Normally, AMS does **not** measure **absolute** !
- compare number of counts to a standard sample
- Give concentration **relative to standard sample**



Can I measure my favourite
isotope with AMS?

Half-life:

$T_{1/2} > 100 \text{ Ma} \rightarrow$ primordial background

$T_{1/2} < \text{weeks} \rightarrow$ better use decay counting

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e.g. ^{53}Mn and $^{53}\text{Cr} \rightarrow$ setup needs isobar suppression

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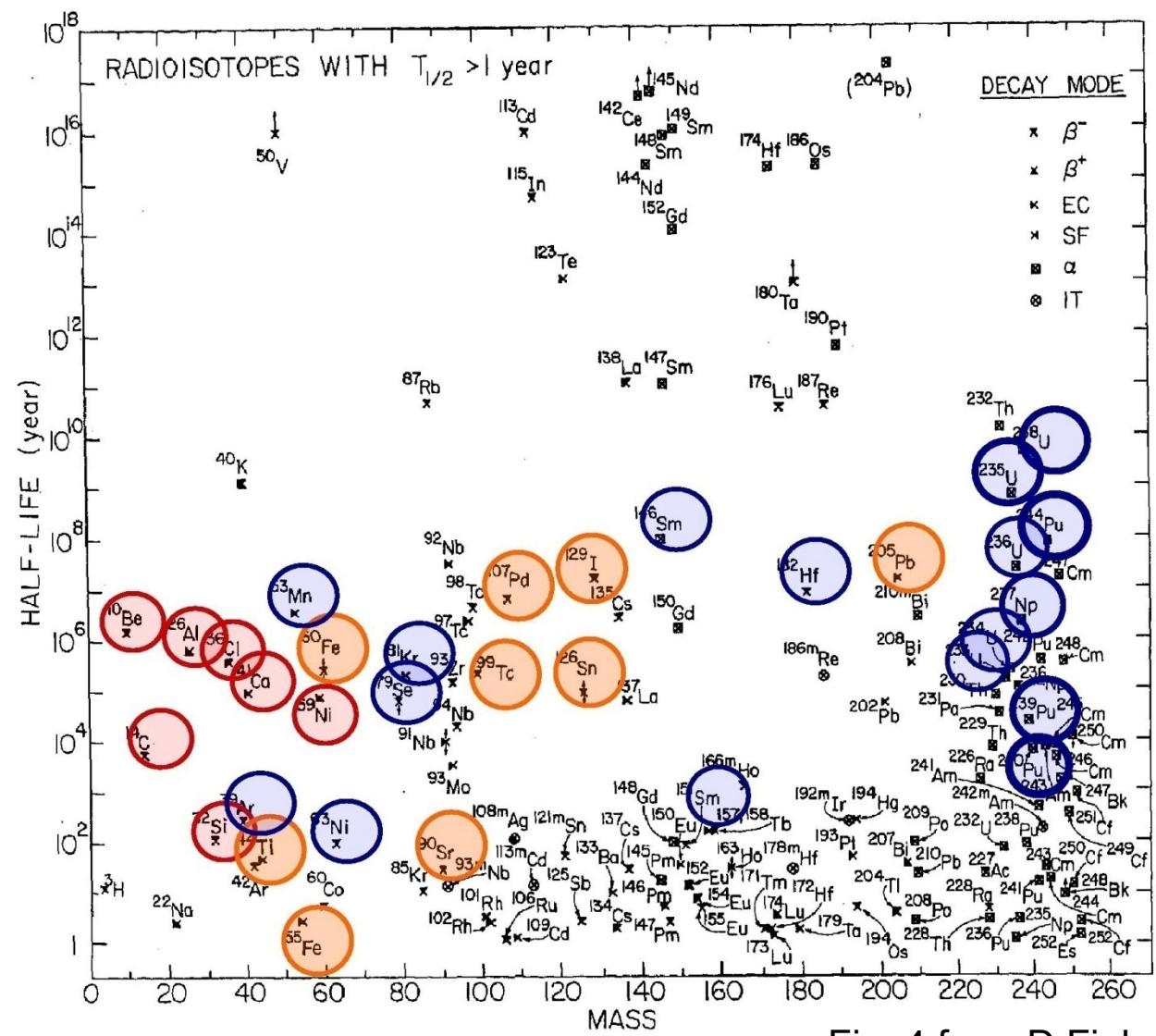
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Standard material available?
Since AMS measures relative, standard
Necessary to obtain concentrations

AMS isotopes



**1981
AMS-2**

$^{10}\text{Be}, ^{14}\text{C}, ^{26}\text{Al},$
 $^{32}\text{Si}, ^{36}\text{Cl},$
 $^{41}\text{Ca}, ^{59}\text{Ni}$

**1996
AMS 3—7
+9 new
isotopes**

**2008
AMS 8 - 11
+17 more
new isotopes**

Fig. 4 from D.Fink, NIM B 268, 2010
modified/updated from W. Kutschera 1981

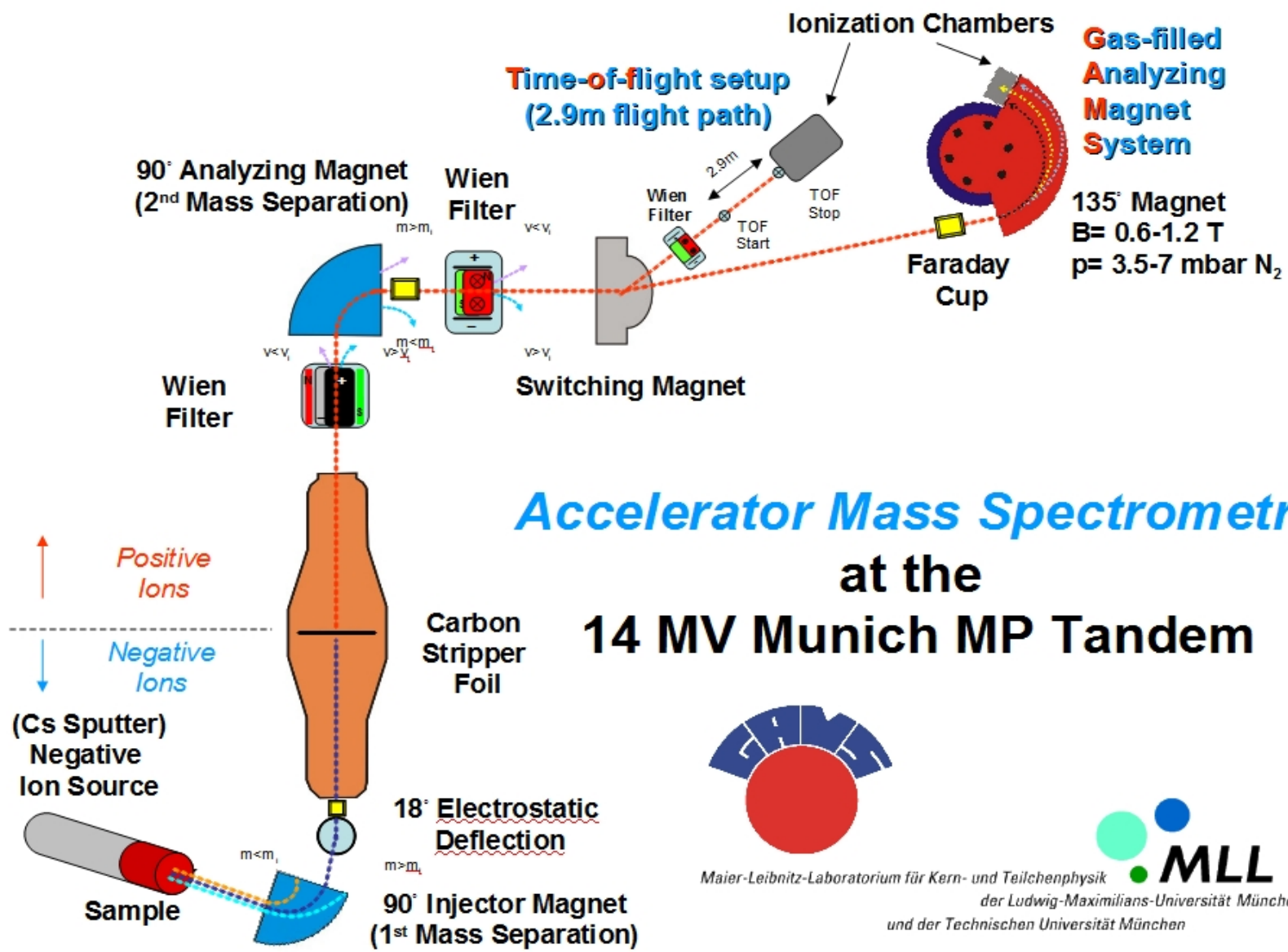
AMS is multidisciplinary

- Environmental and Geo-science
 - atmosphere, oceanography, glacier, **climate**, groundwater, erosion, dating paleoceanography, ...
- Material science
 - fusion research, active waste management
- human metabolism and medical application
 - **dosimetry**, pharmacology, ...
- Chemistry
 - tracing molecules and elements, ...
- Extraterrestrial
 - meteorites (e.g. Lunar and Martian origin)
 - **interplanetary dust, SN**, related cross sections, ...
- Physics
 - **nuclear astrophysics**, super- asymmetric fission, ...

Maier-Leibnitz Laboratory (MLL) Garching



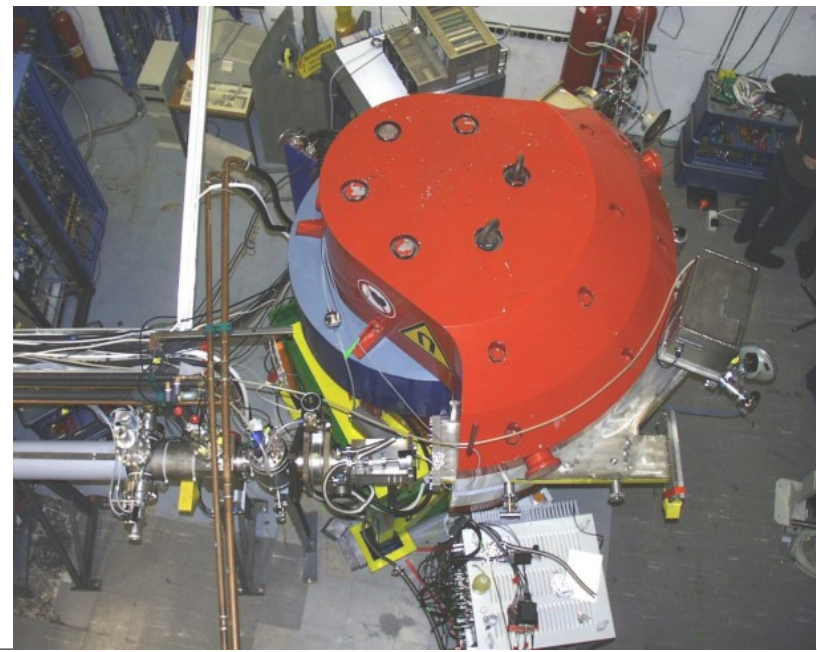
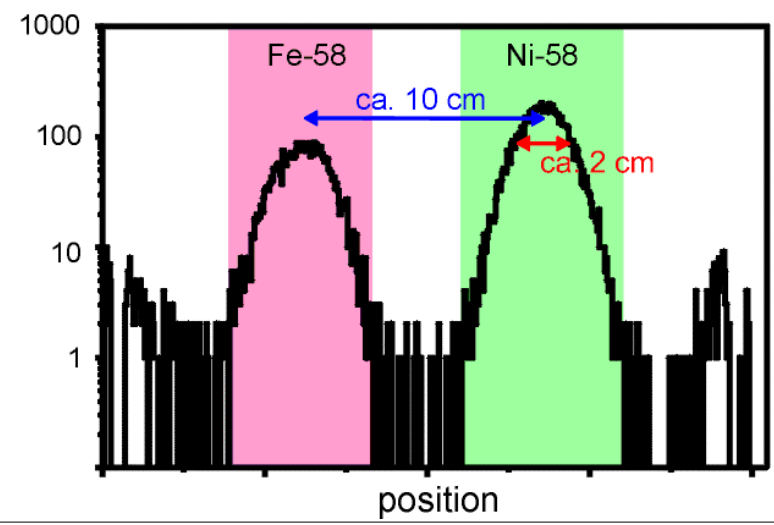
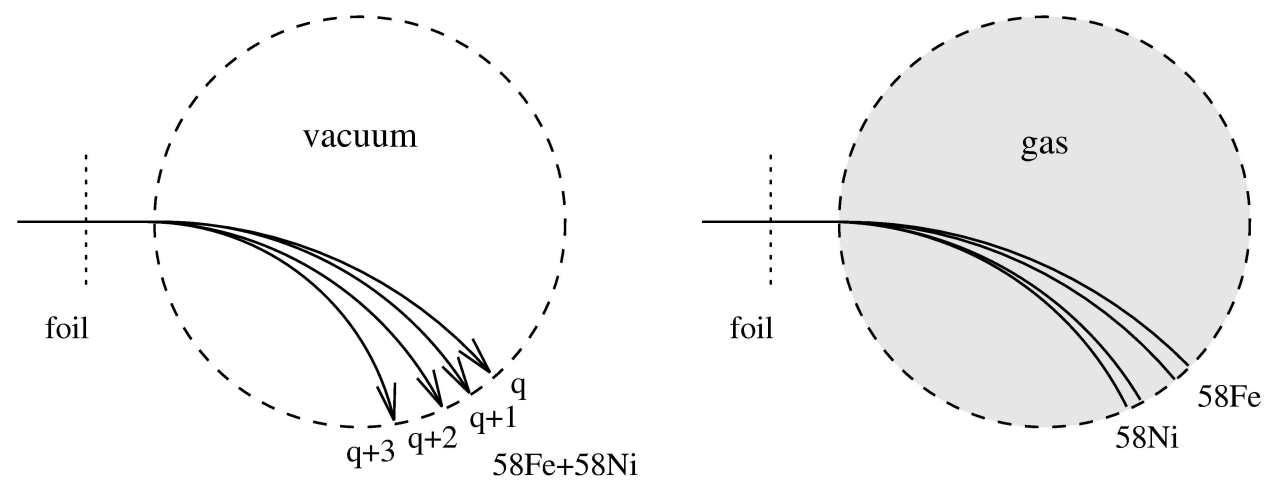
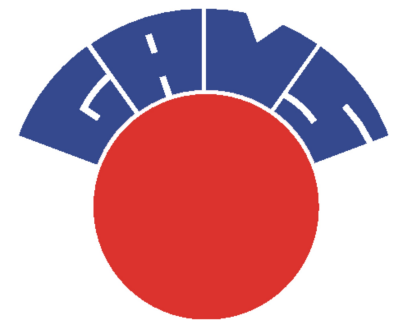
AMS in Garching



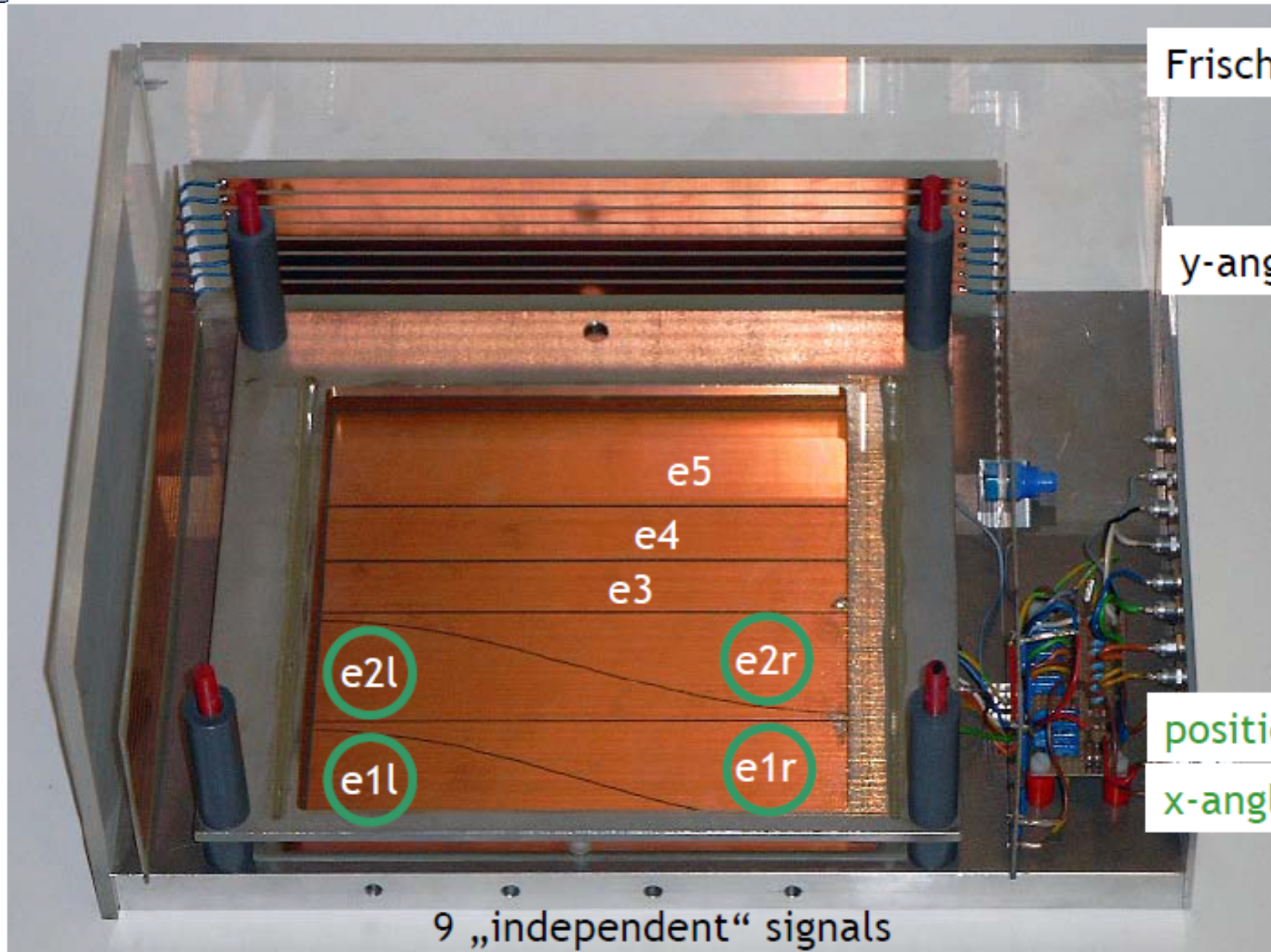
Maier-Leibnitz-Laboratorium für Kern- und Teilchenphysik
 der Ludwig-Maximilians-Universität München
 und der Technischen Universität München

Isobar suppression: GAMS

Challenge: Isobar separation of ^{60}Ni → use of the Gas-filled-Analyzing-Magnet-System (GAMS)



Ionization chamber



Frisch grid: et

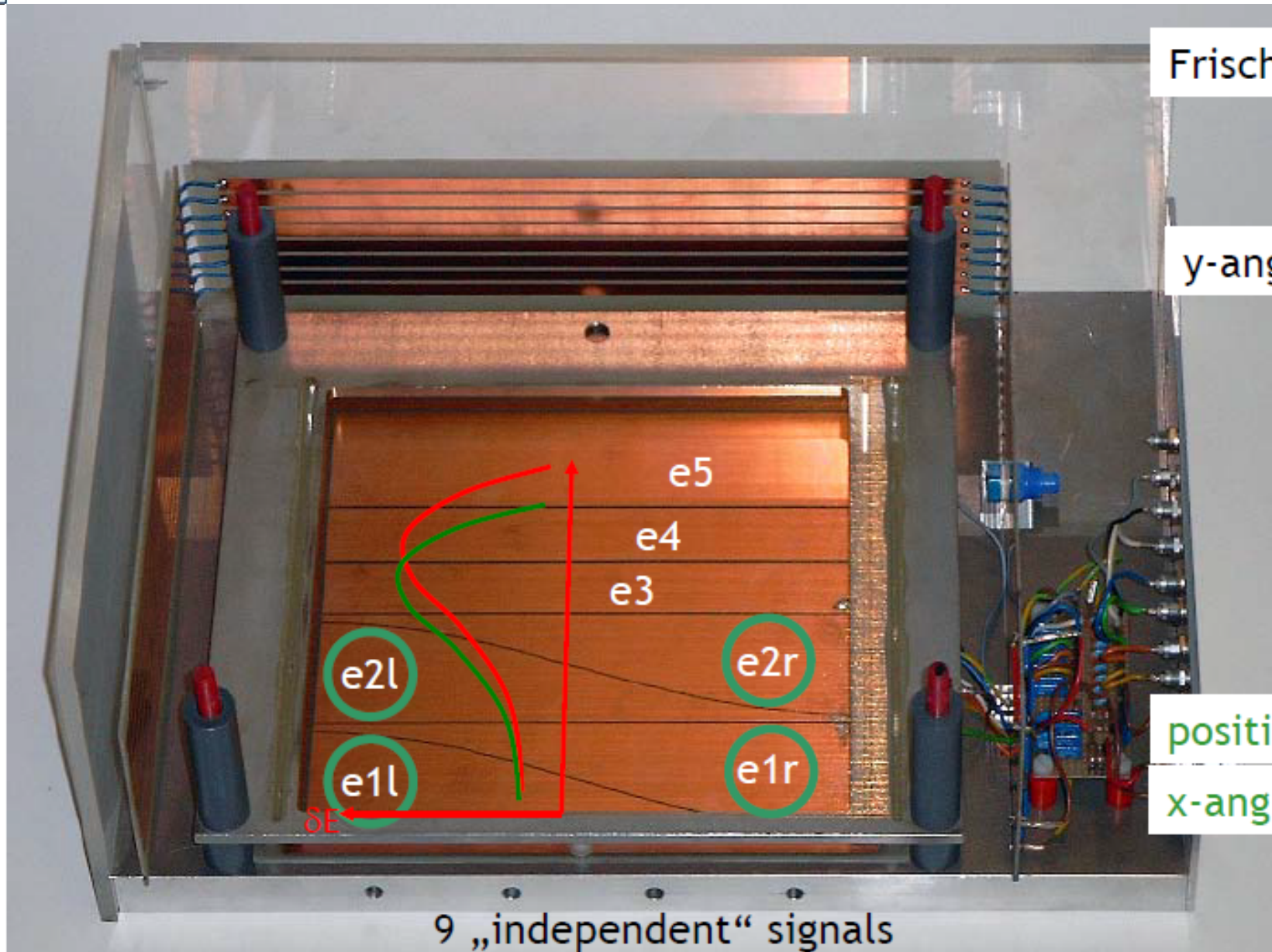
y-angle: dt

position: p

x-angle: dp

9 „independent“ signals

Ionization chamber



Frisch grid: et

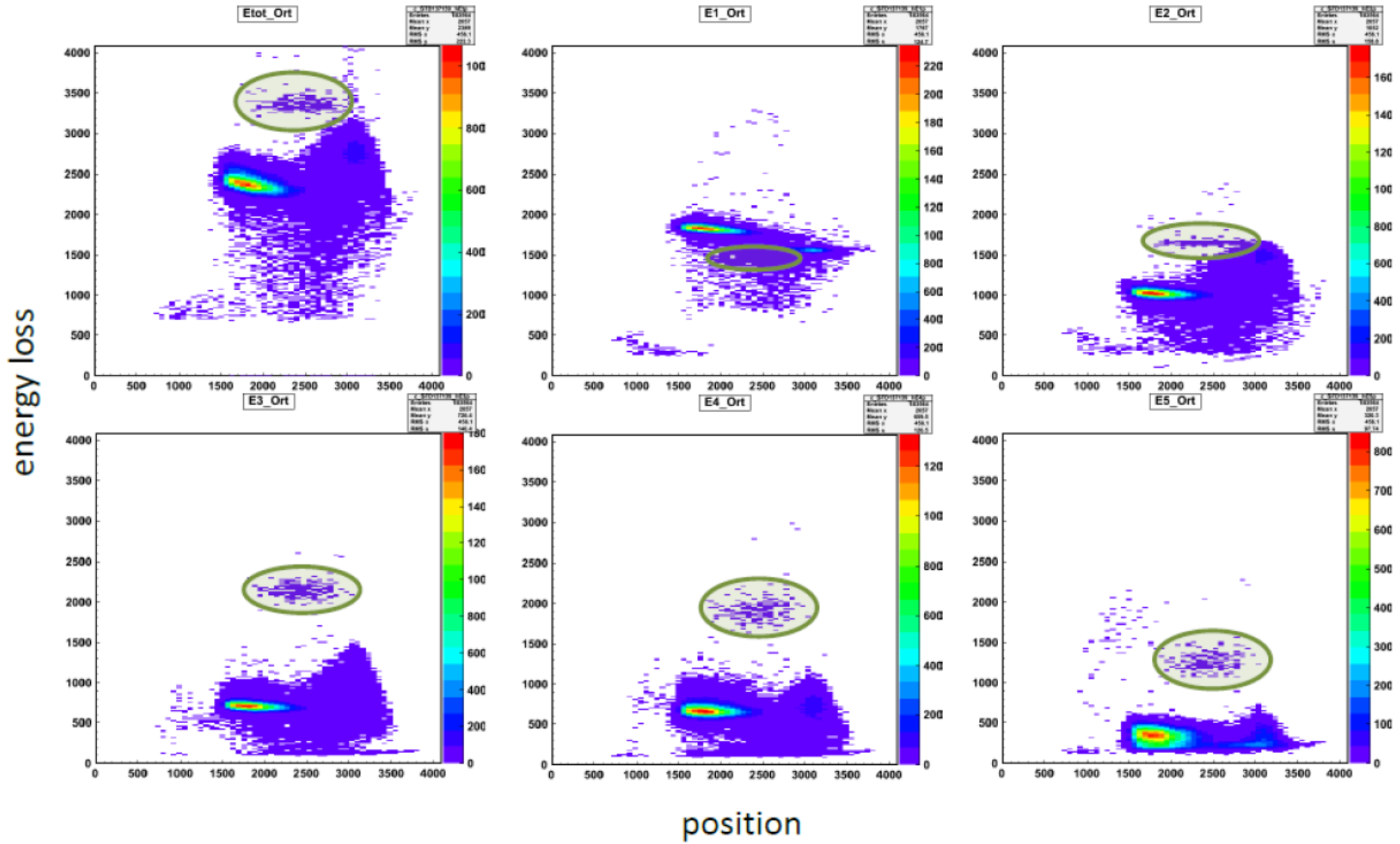
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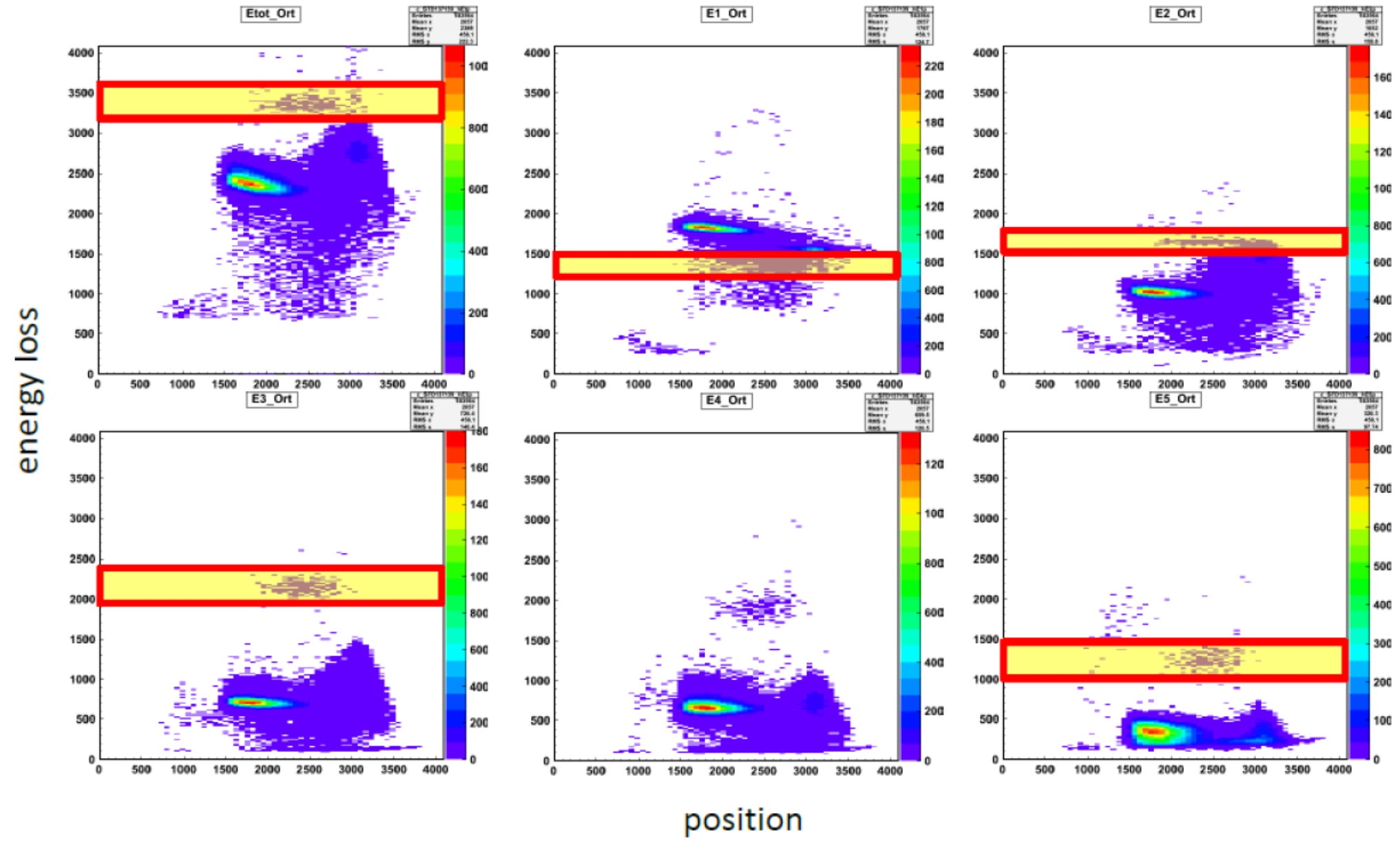
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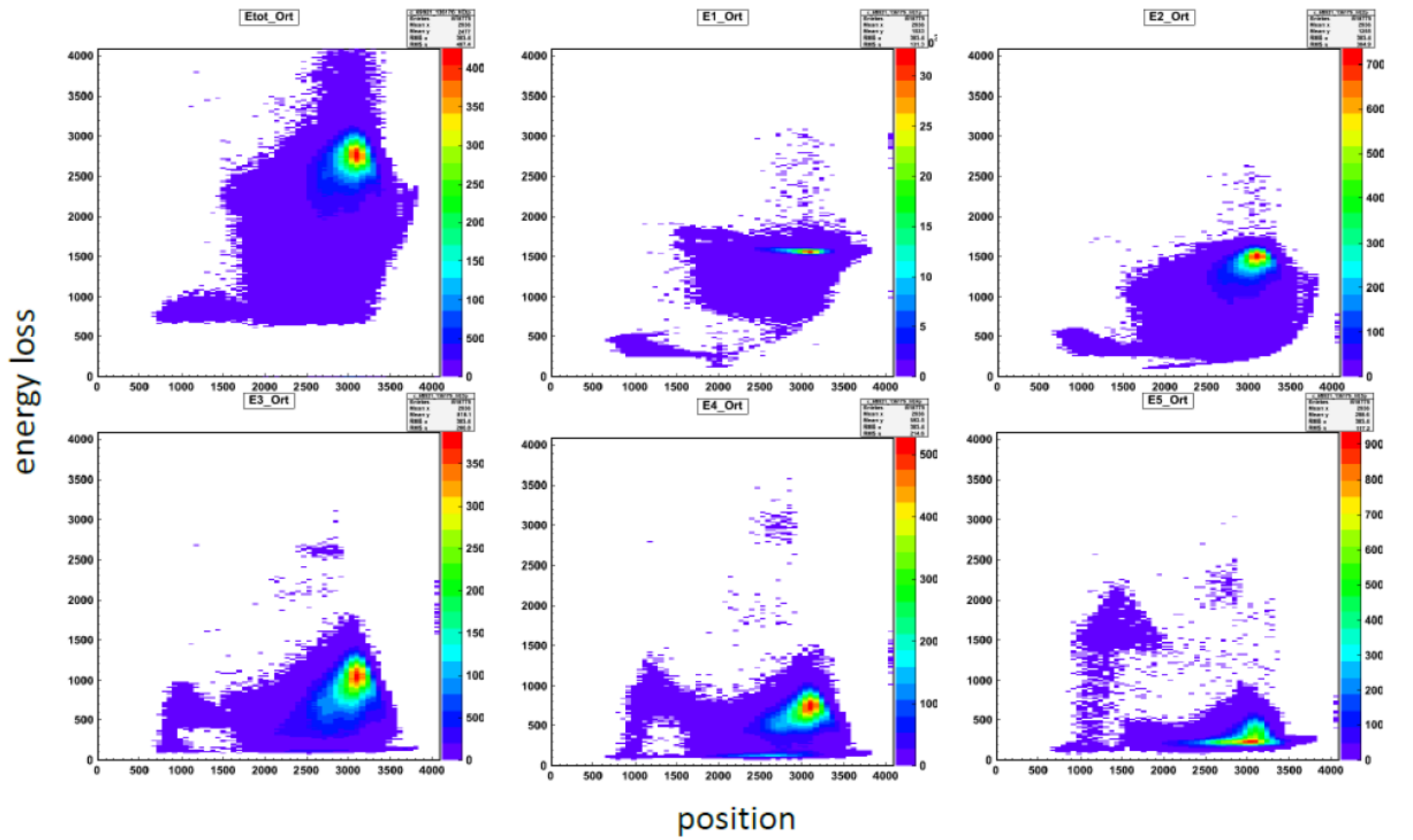
9 „independent“ signals

60Fe standard sample $60\text{Fe}/\text{Fe} \sim 1\text{E-}12$

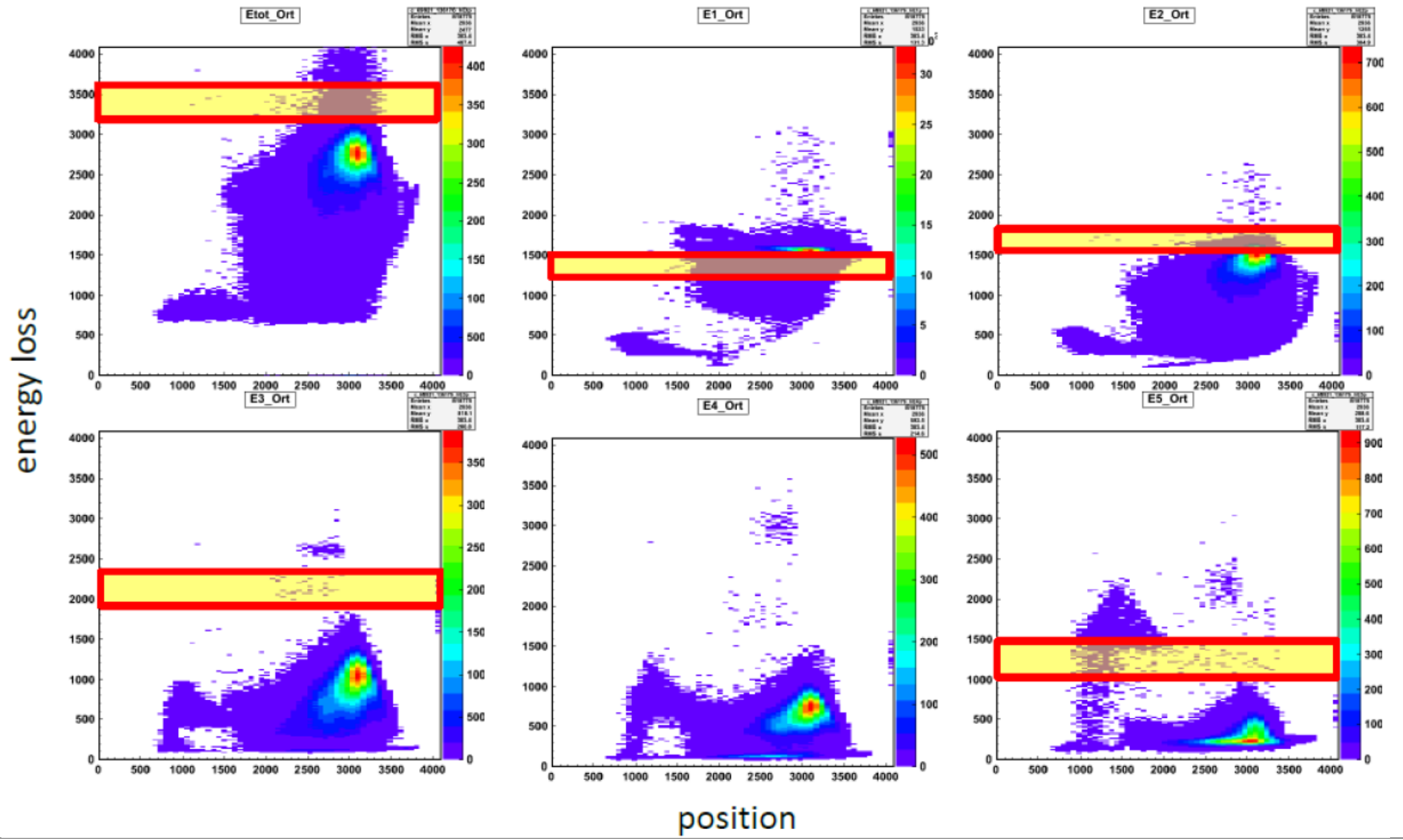




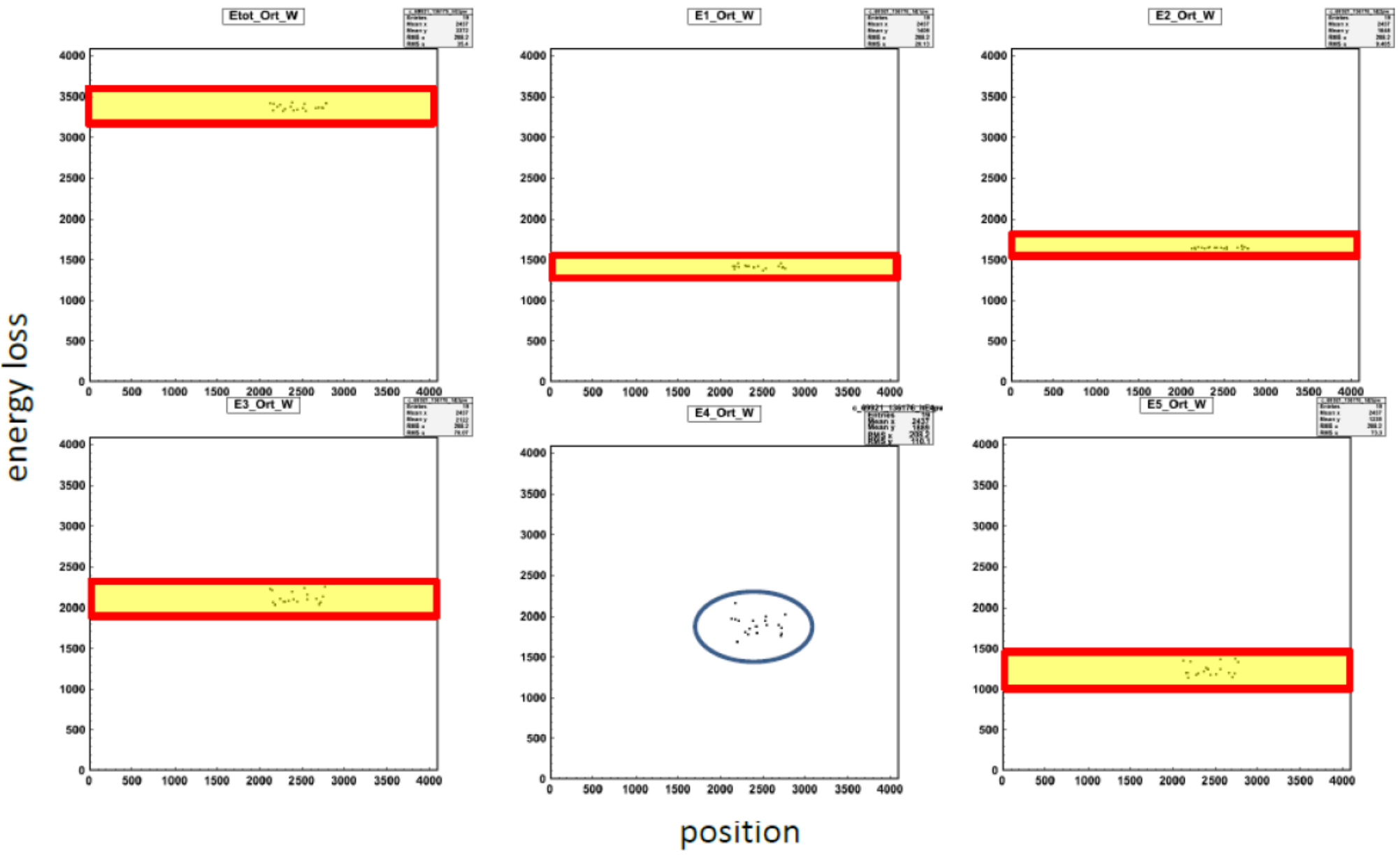
Real sample $^{60}\text{Fe}/\text{Fe}=?$



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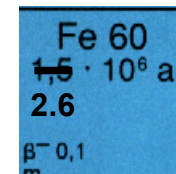
AMS measurements with GAMS

isotope	application	ion in ion out	isobar	isobar Supp.	sensitivity
²⁶ Al	astroph. x-section manganese crusts	AlO ⁻ Al ⁷⁺	²⁶ Mg	10 ⁹	few 10 ⁻¹⁵
³⁶ Cl	groundwater, meteorites	Cl ⁻ Cl ¹⁰⁺	³⁶ S	10 ⁹ – 10 ¹⁰	< 10 ⁻¹⁵
⁴¹ Ca	Hiroshima teeth, meteorites	CaH ₃ ⁻ , CaF ₃ ⁻ Ca ⁹⁺	⁴¹ K	10 ⁹ – 10 ¹⁰	< 10 ⁻¹⁵ 10 ⁻¹⁴ CaF
⁴⁴ Ti	astroph. x-section	TiO ⁻ Ti ⁹⁺	⁴⁴ Ca	10 ⁹ – 10 ¹⁰	few 10 ⁻¹⁵
⁵³ Mn	crusts, in-situ, meteorites, sediments, moon rocks	⁵³ MnO ⁻ , ⁵³ MnF ⁻ ⁵³ Mn ¹¹⁺	⁵³ Cr	10 ⁹	few 10 ⁻¹⁵
⁵⁹ Ni	nuclear waste, meteorites, astroph. x-section	⁵⁹ Ni ⁻ ⁵⁹ Ni ¹²⁺	⁵⁹ Co	10 ⁹	few 10 ⁻¹⁴
⁶⁰ Fe	crusts, meteorites, moon rock, nuclear waste	⁶⁰ FeO ⁻ ⁶⁰ Fe ¹¹⁺	⁶⁰ Ni	>10 ¹¹	few 10 ⁻¹⁶
⁶³ Ni	Hiroshima copper, neutron dosimetry astroph. x-section	⁶³ Ni ⁻ ⁶³ Ni ¹²⁺	⁶³ Cu	10 ⁸ – 10 ⁹	few 10 ⁻¹⁴
⁷⁹ Se, ⁹³ Mo, ⁹³ Zr, ¹⁰⁷ Pd	astroph. x-section, half-life, nuclear waste	various	⁷⁹ Br, ⁹³ Nb, ¹⁰⁷ Ag	10 ⁶	few 10 ⁻¹² ? few 10 ⁻¹⁰ ? ?

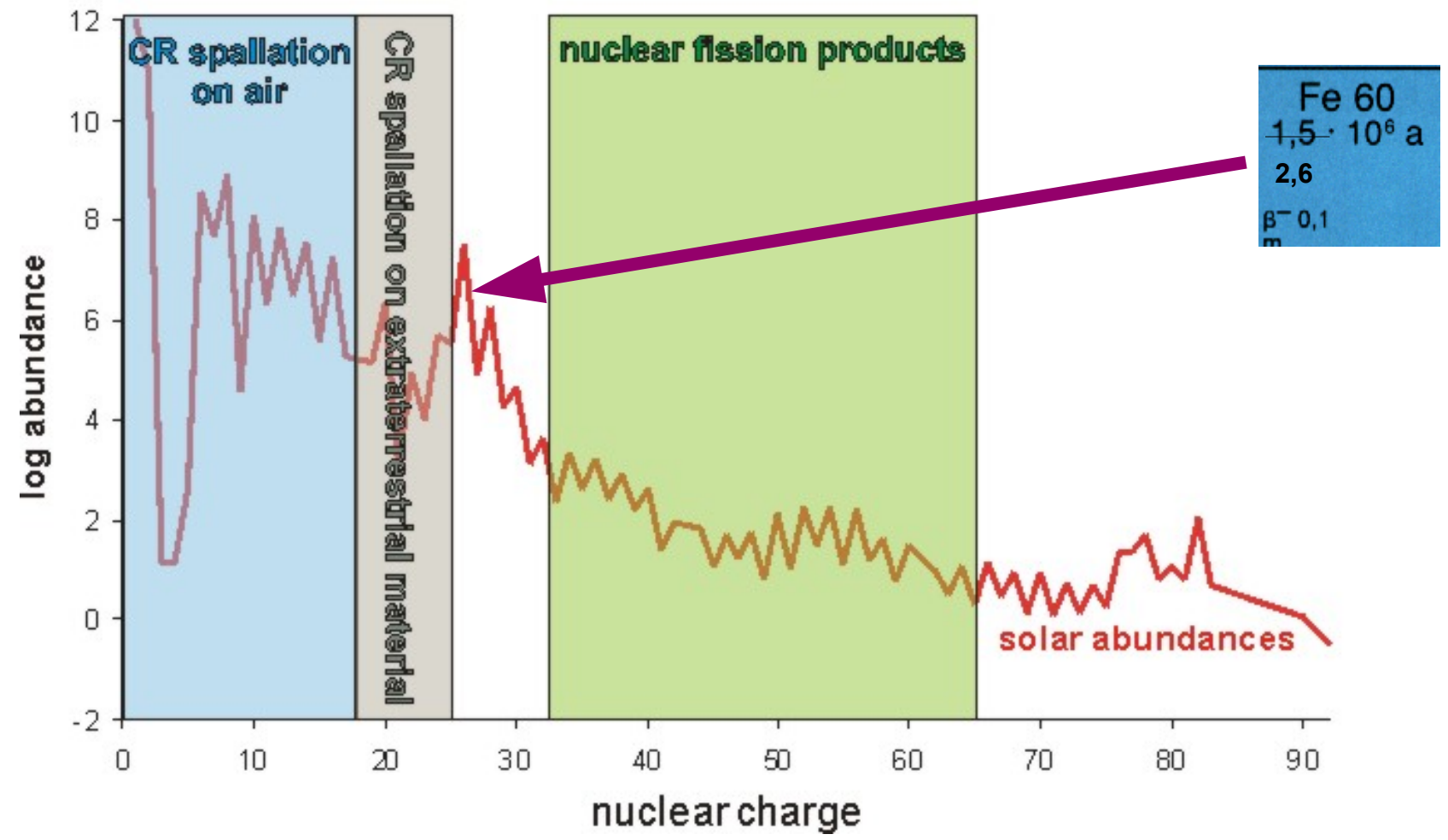
Choosing an isotope



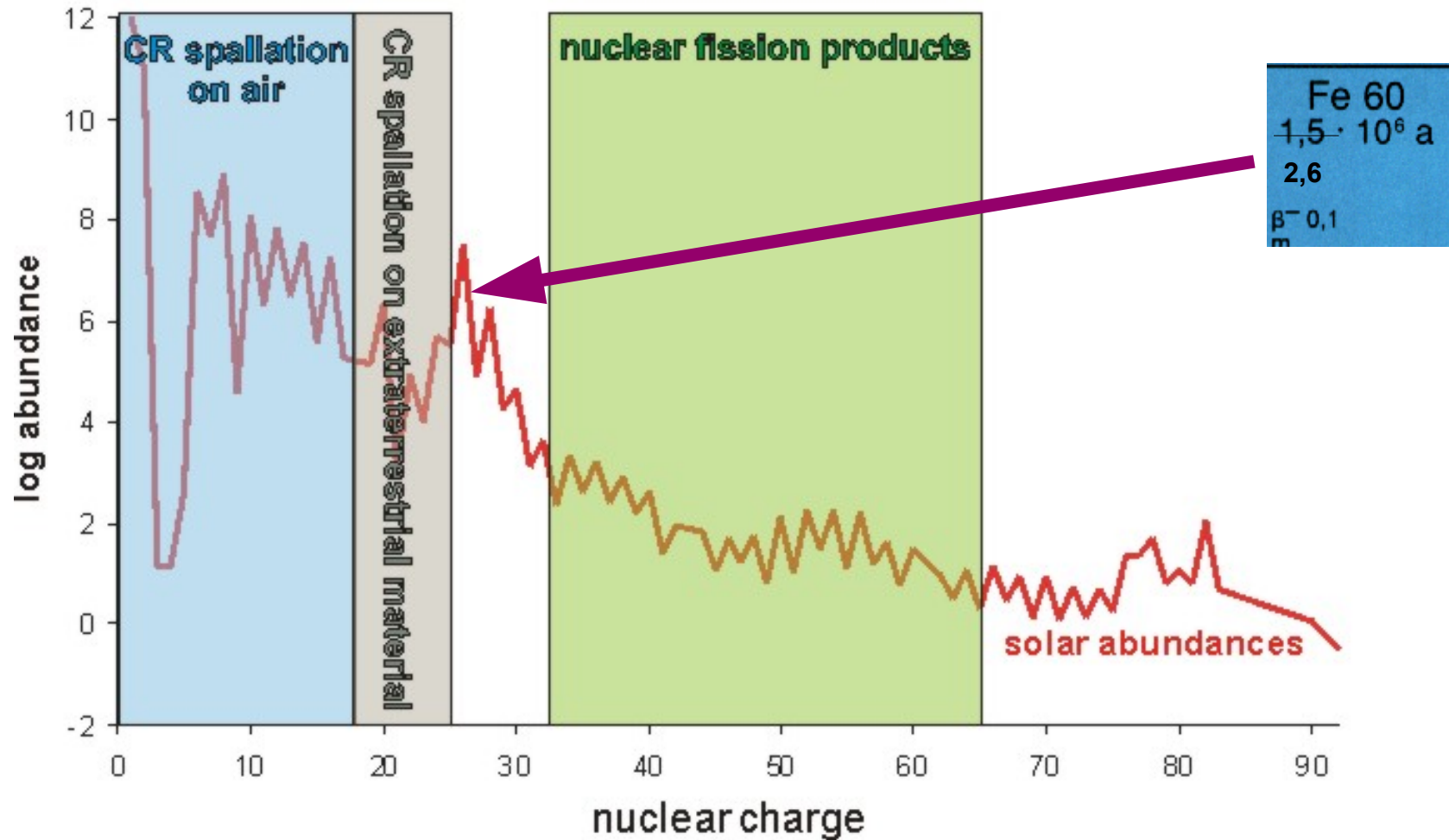
However, I can only talk about ONE of those isotopes today:



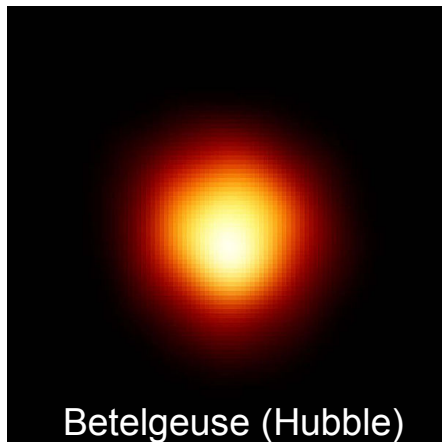
Why ^{60}Fe ?



Why ^{60}Fe ?



- Unique situation:
- 1) Isobar ^{60}Ni has $\Delta Z = 2$ (good for AMS!)
 - 2) Expected background in terrestrial samples $^{60}\text{Fe}/\text{Fe} < 10^{-16}$
 - 3) Half-life on the order of Myr (interesting SN ages)

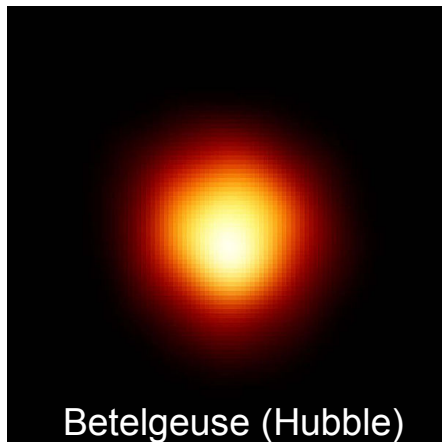
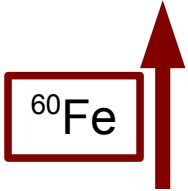


Betelgeuse (Hubble)

Massive star forming ^{60}Fe



Supernova explosion

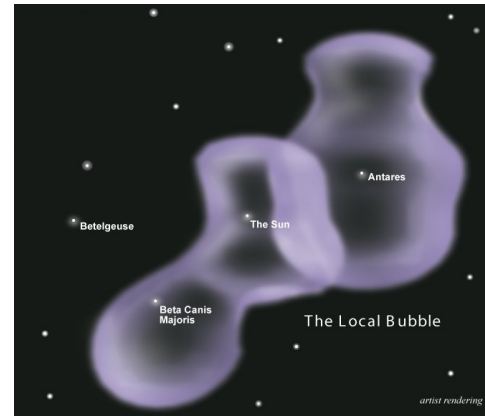
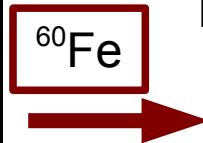


Betelgeuse (Hubble)

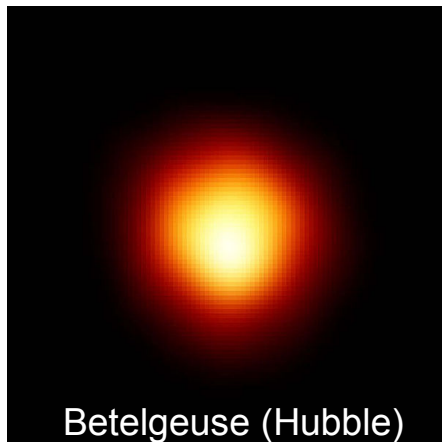
Massive star forming ^{60}Fe

Motivation

Interstellar medium near Earth:
 Local Bubble



Supernova explosion



Betelgeuse (Hubble)

Massive star forming ^{60}Fe

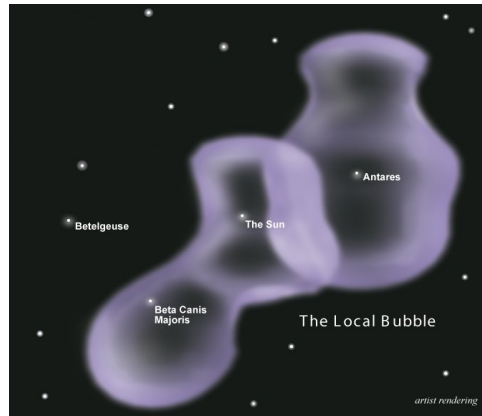
Motivation



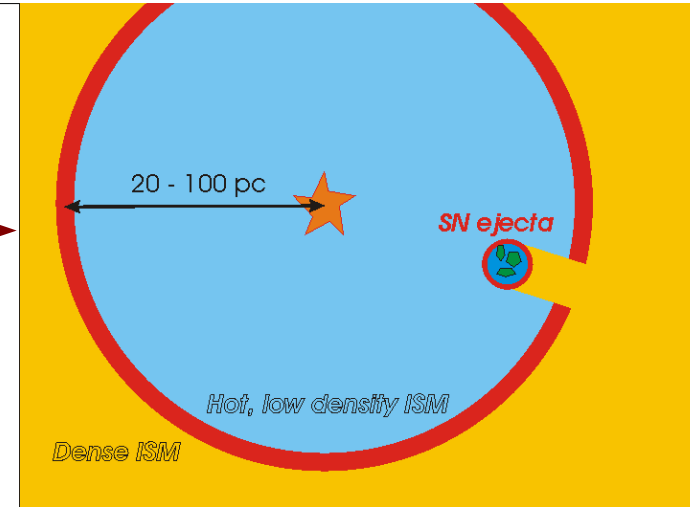
Supernova explosion

^{60}Fe

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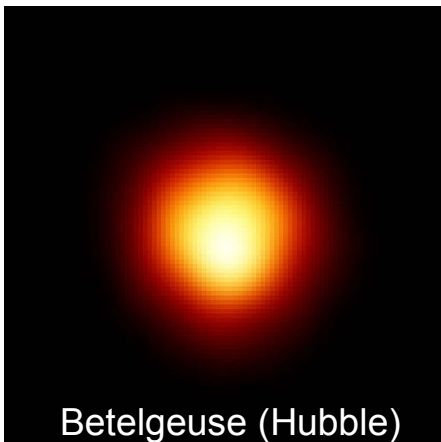


^{60}Fe



Solar system can „pick up“ supernova debris

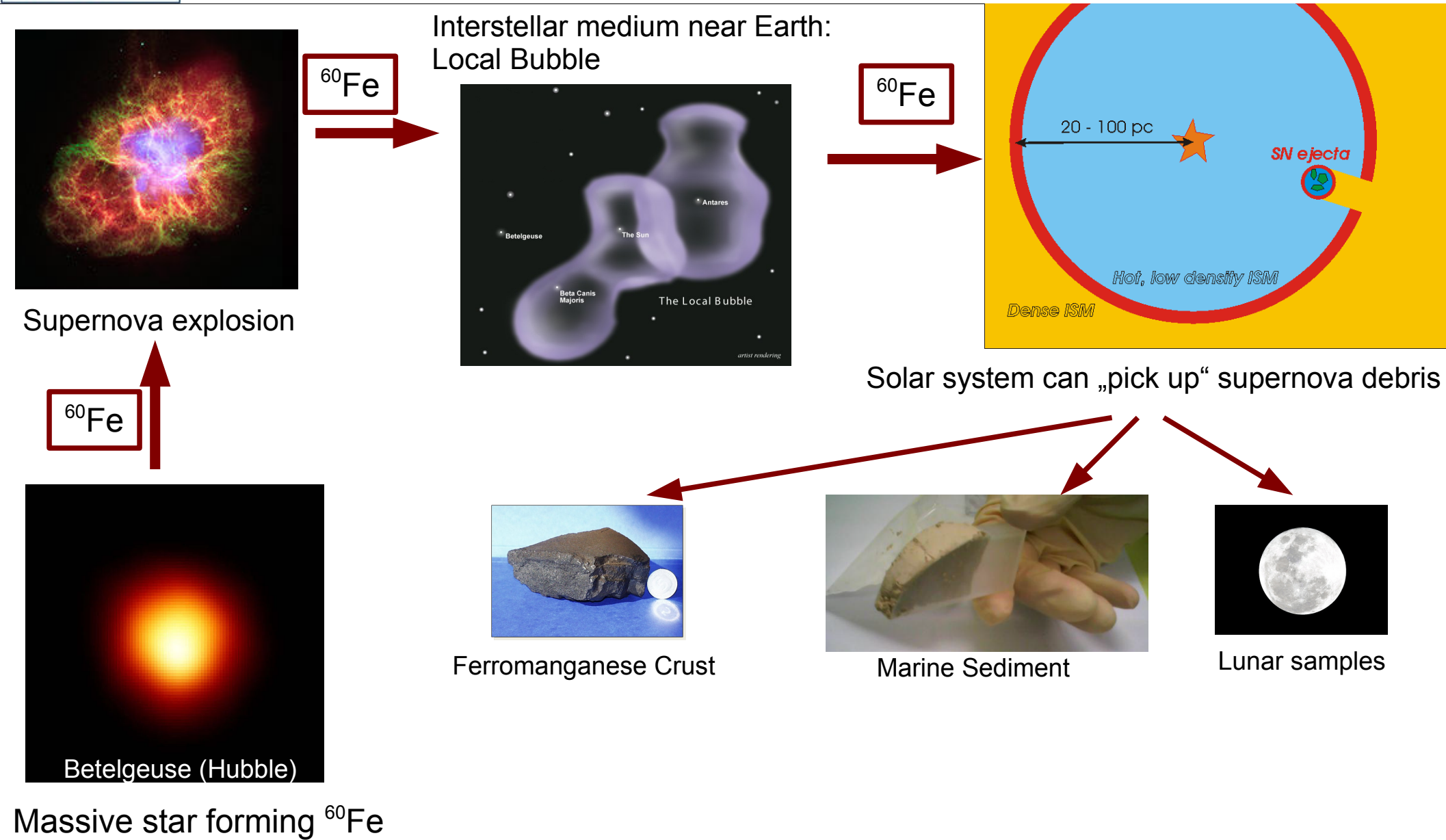
^{60}Fe



Betelgeuse (Hubble)

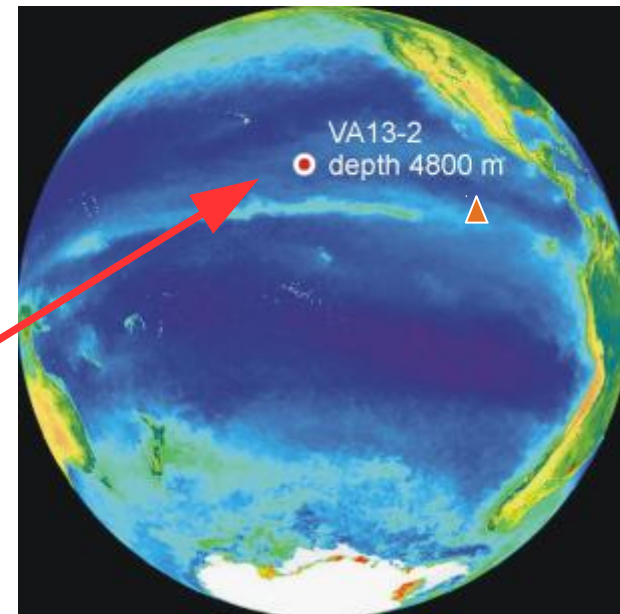
Massive star forming ^{60}Fe

Motivation



Earlier measurement: Ferromanganese Crust

Ferromanganese crust from
equatorial pacific
(9°18'N, 146°03'W), depth 4830 m



- Slow growing (few mm per Ma)
- Can be dated by ^{10}Be measurements

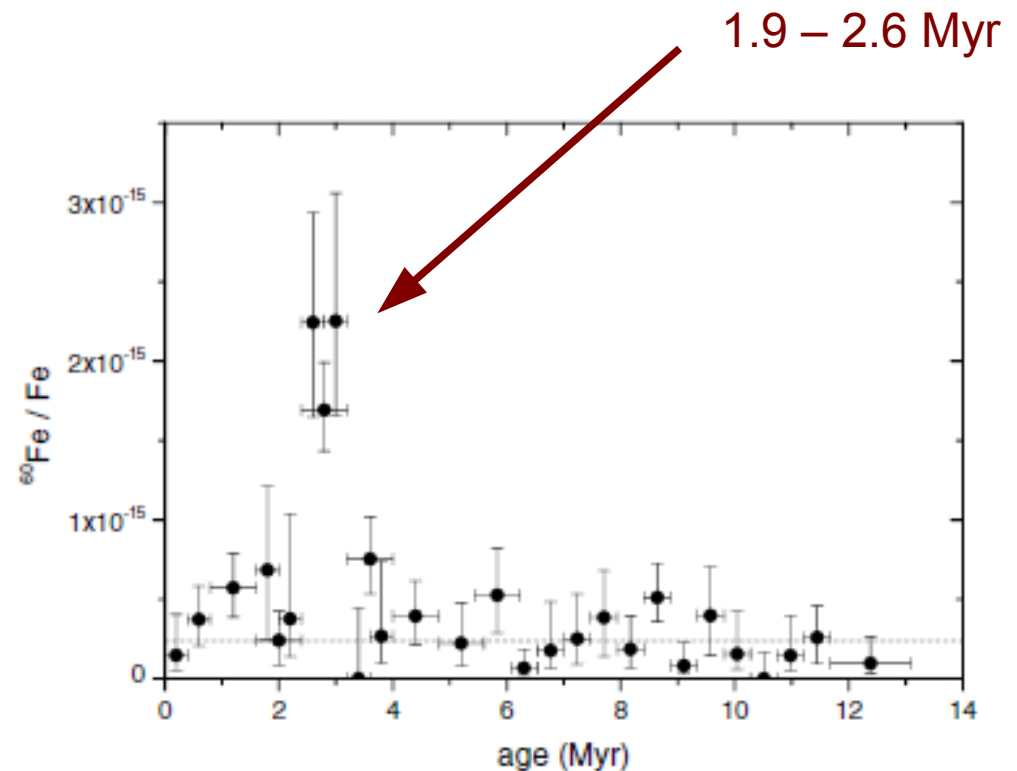
Ferromanganese crust from
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Earlier measurement: Ferromanganese Crust

- Indication for a supernova signal already found using the GAMS setup
- Possible SN event 2-3 Ma ago at a distance ~40 pc
- Goal: Confirmation of this finding in slow growing sediment

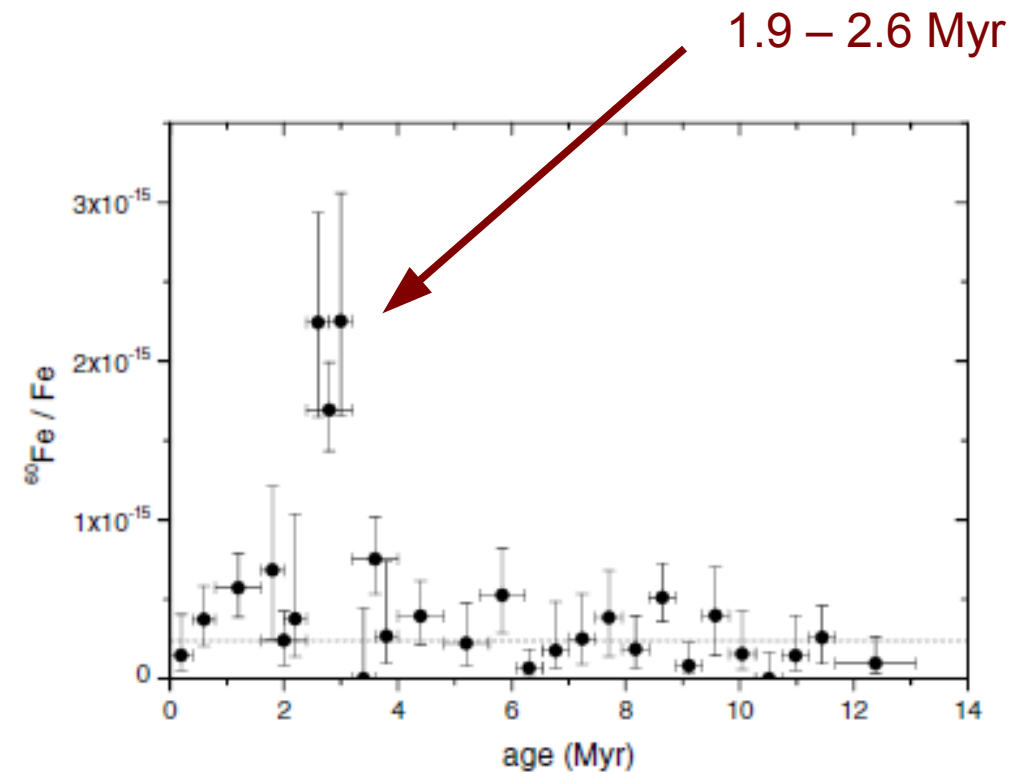
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Knie et. al., PRL **93**, 171103 (2004)

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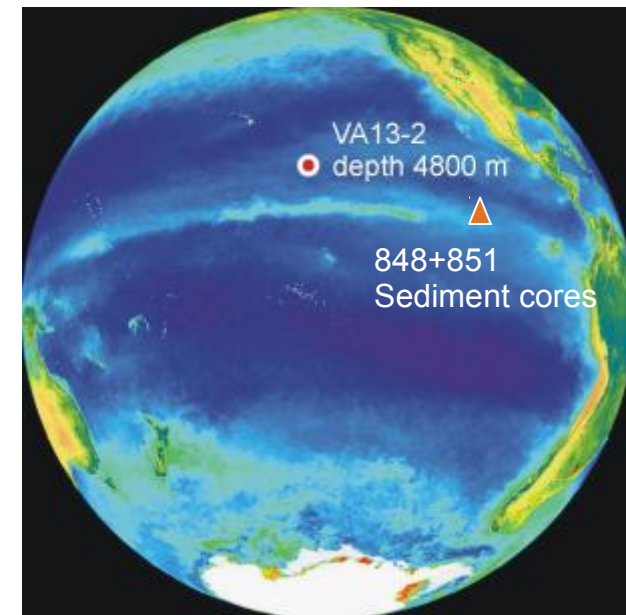
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Knie et. al., PRL **93**, 171103 (2004)

Search for ^{60}Fe in ocean sediment:

- Samples from two drill cores from ODP (Ocean Drilling Program) were obtained, 8 kg of material total – leg 138 – Cores 848 + 851
- Goal: measure depth profile of $^{60}\text{Fe}/\text{Fe}$ with resolution ~ 30.000 years in the age range 1.8 – 3.8 Myr
- Dating available from magnetic field reversal (among others), however, independent dating currently underway using ^{10}Be and ^{26}Al at DREAMS in Dresden



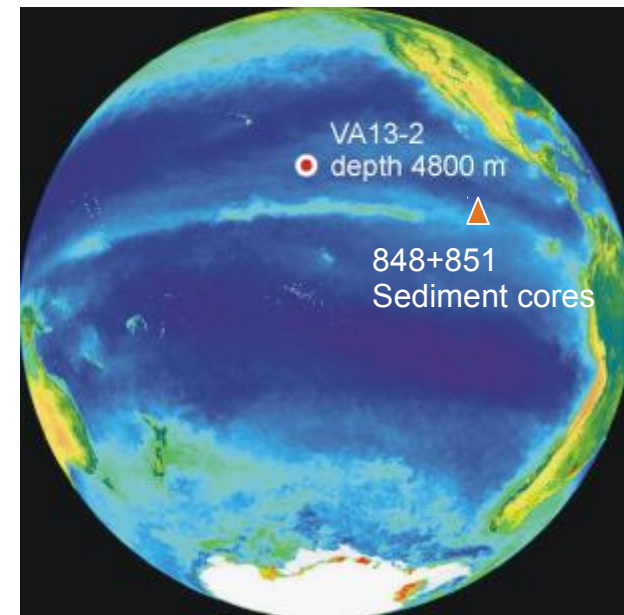
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- Problems:
 - Which Fe-bearing minerals could contain the ^{60}Fe signal?
 - How to prevent dilution of signal?

Primary
Transported in by wind/water
Large grains (μm)
 ^{60}Fe only on surface

Secondary
Formed in situ
Small grains (20-200 nm)
e.g. magnetite crystals
(Fe_3O_4)

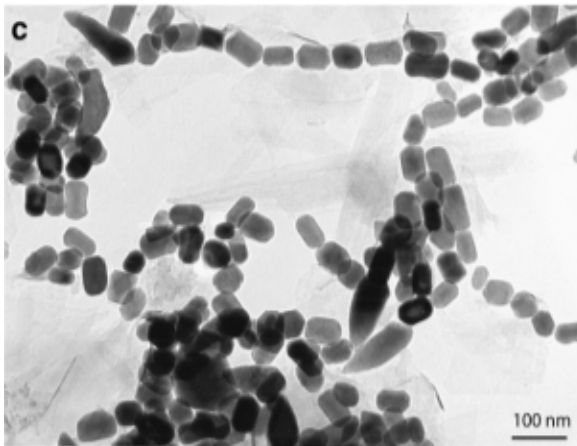
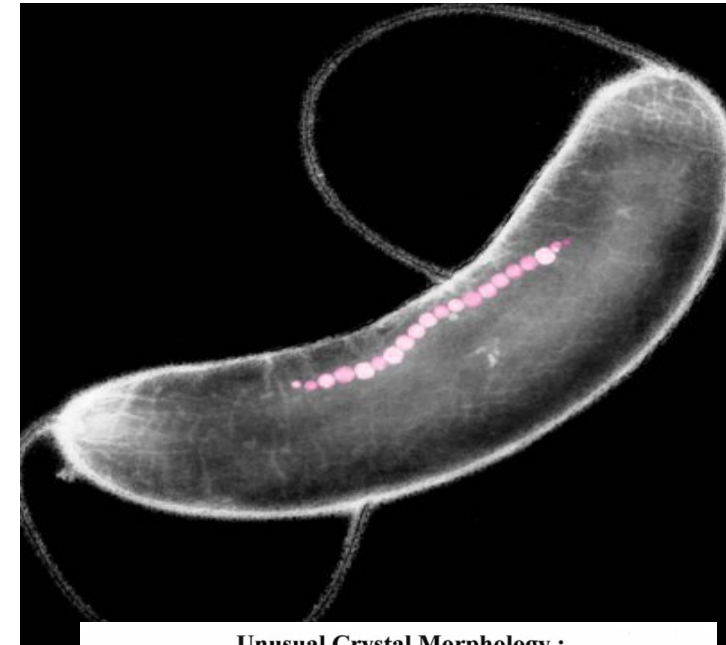
^{60}Fe



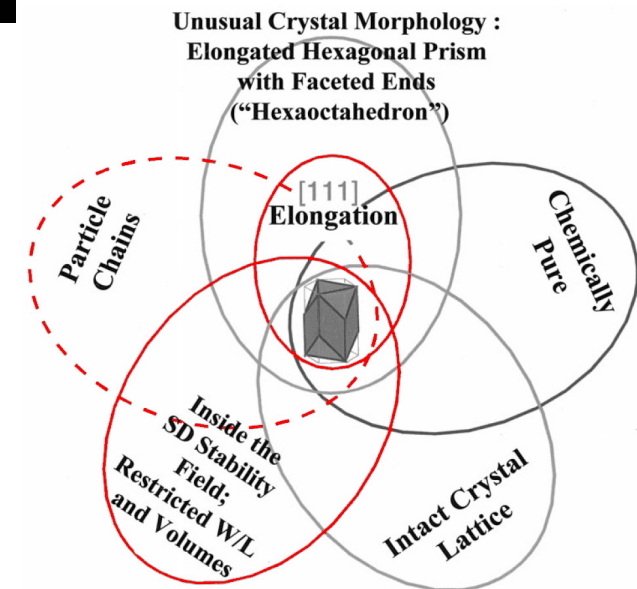
Magnetotactic bacteria

Intracellular formation of biogenic magnetite crystals:

- Magnetotactic bacteria live in sediment just below the surface-water interface
- Bacteria build up chains of magnetite grains (20-80 nm) for orientation in earth's magnetic field (magnetotaxis) → secondary !
- In ^{60}Fe -enriched ocean water, bacteria are forced to build magnetosomes with ^{60}Fe -rich iron
- Magnetic signature can be preserved over geologically significant timescales if the magnetosome chains survive sedimentation.
- Magnetic signature resembles non-interacting single domain (SD) particles → characterization using magnetic measurements is possible because of their unique properties



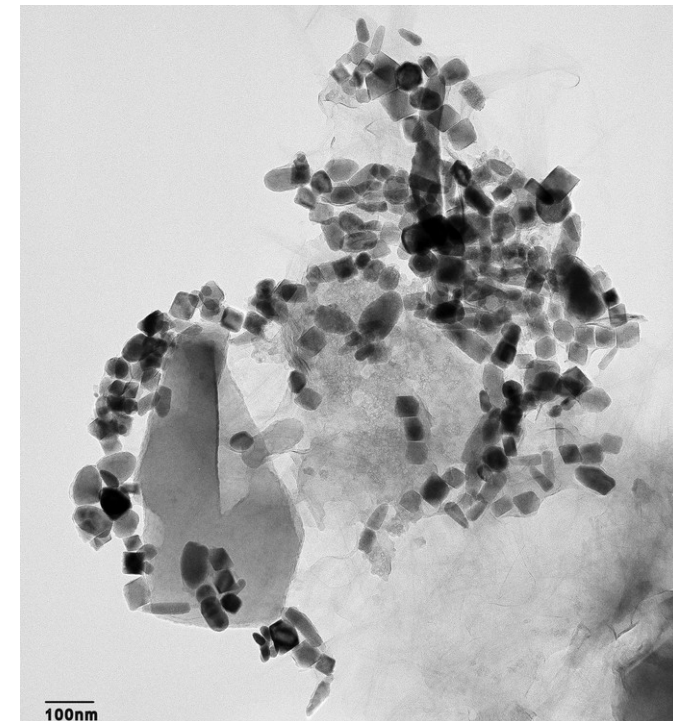
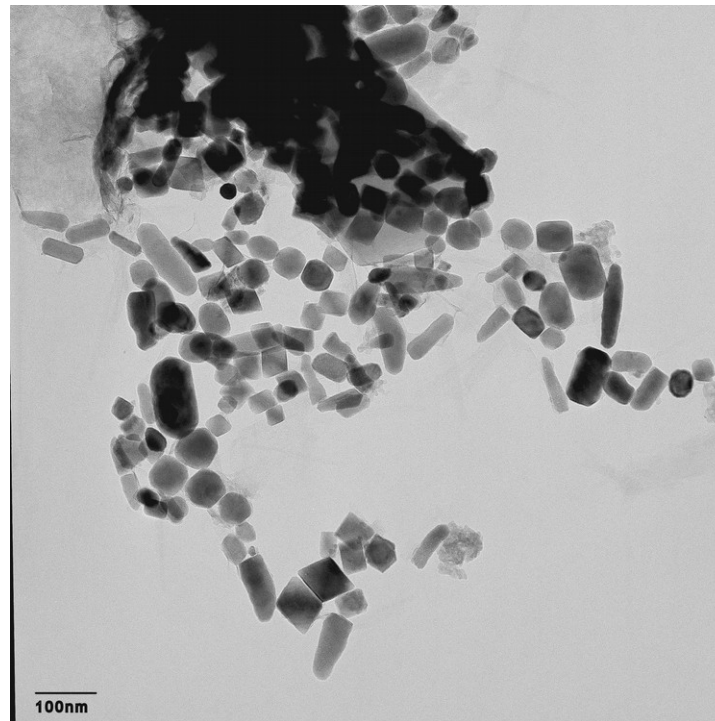
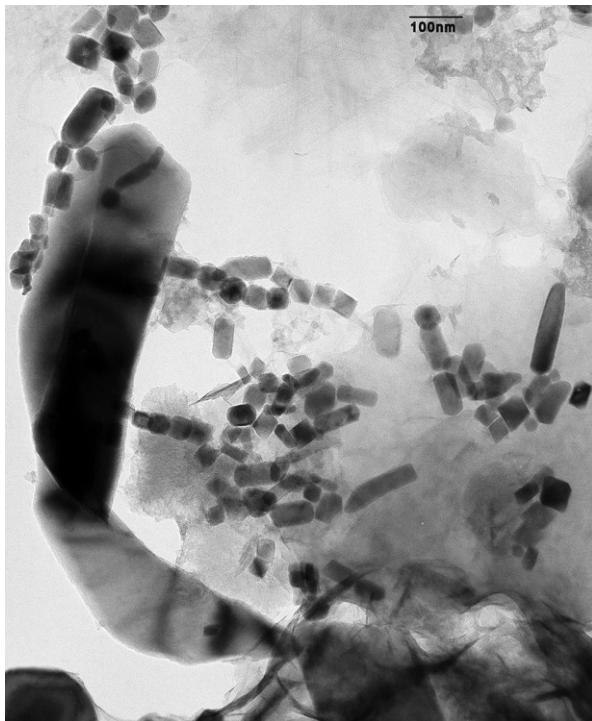
Sediment sample – 150 Myr after sedimentation → chain structure still visible



TEM on Sediment

Transmission electron microscopy (TEM):

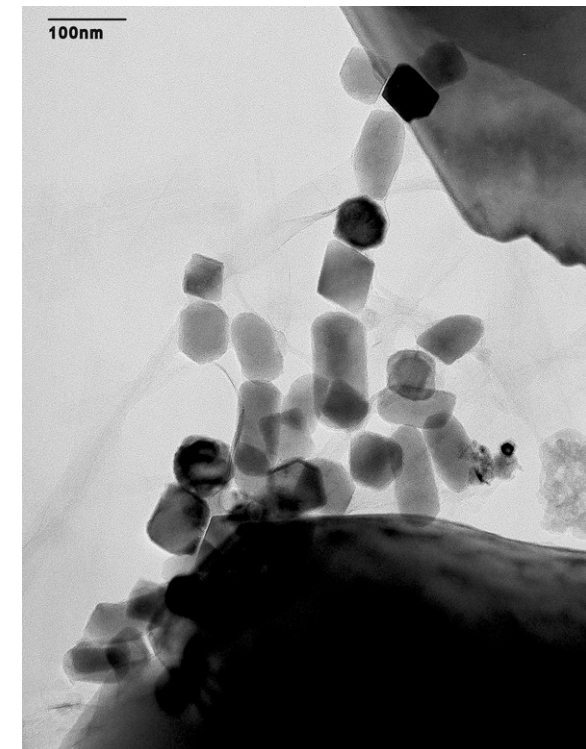
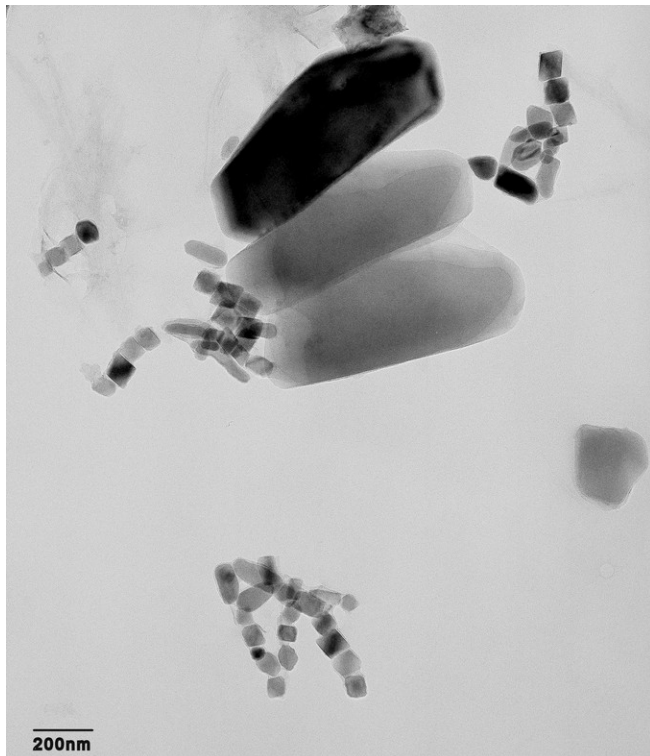
- ◆ After magnetic extraction on one of our sediment samples
- ◆ High abundance of small magnetite grains as chain fragments and clusters



TEM on Sediment

Interpretation:

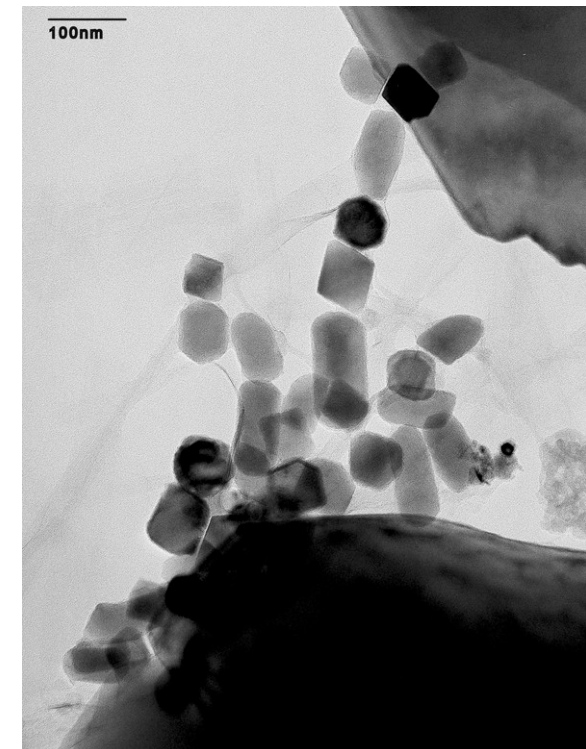
- ◆ Is this magnetite? → zoom in → diffraction analysis OR perform EDX, both say: YES
- ◆ It it biogenic? Both shape and size say YES
- ◆ How much is it? → roughly 10% (mass) of the magnetic extract
→ fits very well with estimate from preliminary ARM/IRM magnetic measurements



TEM on Sediment

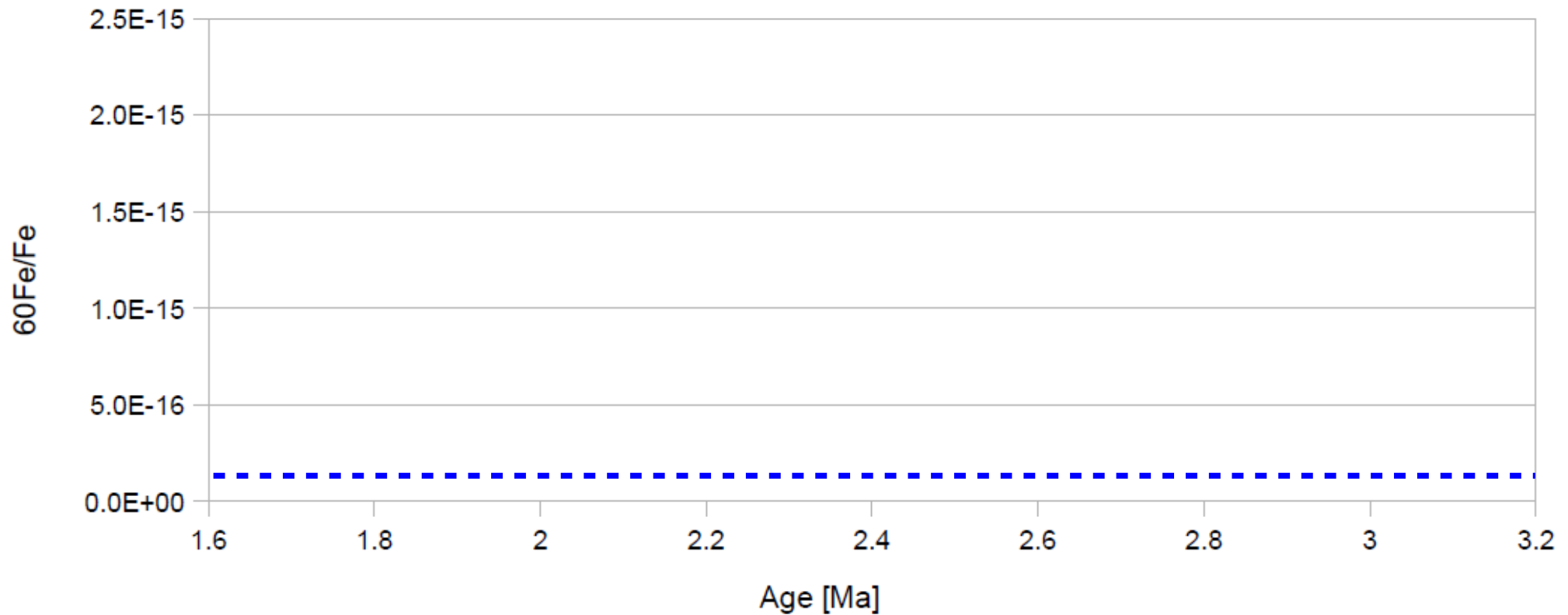
How to get small grained magnetite out to make AMS samples?

- Citrate Bicarbonate Dithionite technique (CBD)
 - very mild leaching (dissolves only < 200 nm)
 - 30 g of sediment yield about 5 mg of Fe_2O_3 AMS sample
 - about 1 week of chemistry needed (not shown here, just to mention...)



AMS results core 848

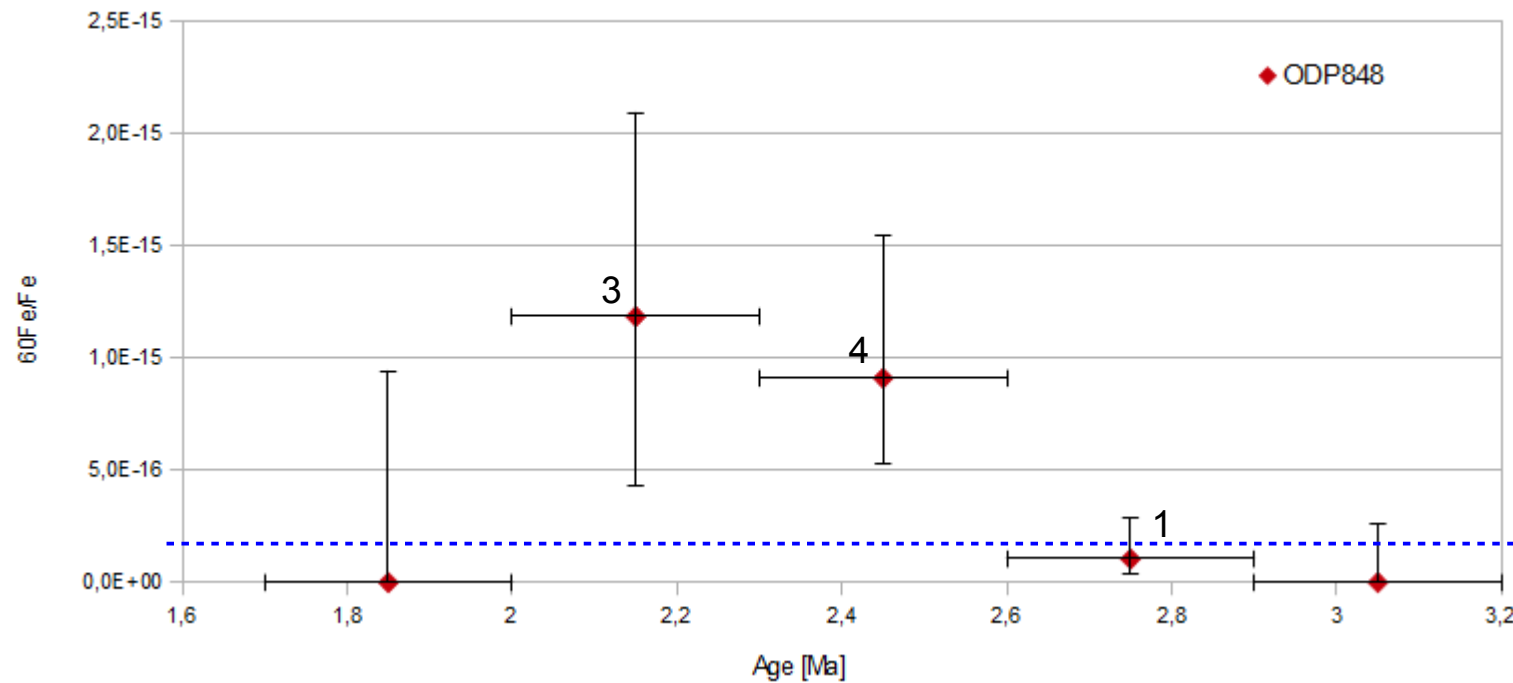
Sediment core 848 - $^{60}\text{Fe}/\text{Fe}$ results



--- Blank level ($1-\sigma$): $^{60}\text{Fe}/\text{Fe} = 1.7\text{e}-16$
From chemistry blank (0 counts)

AMS results core 848

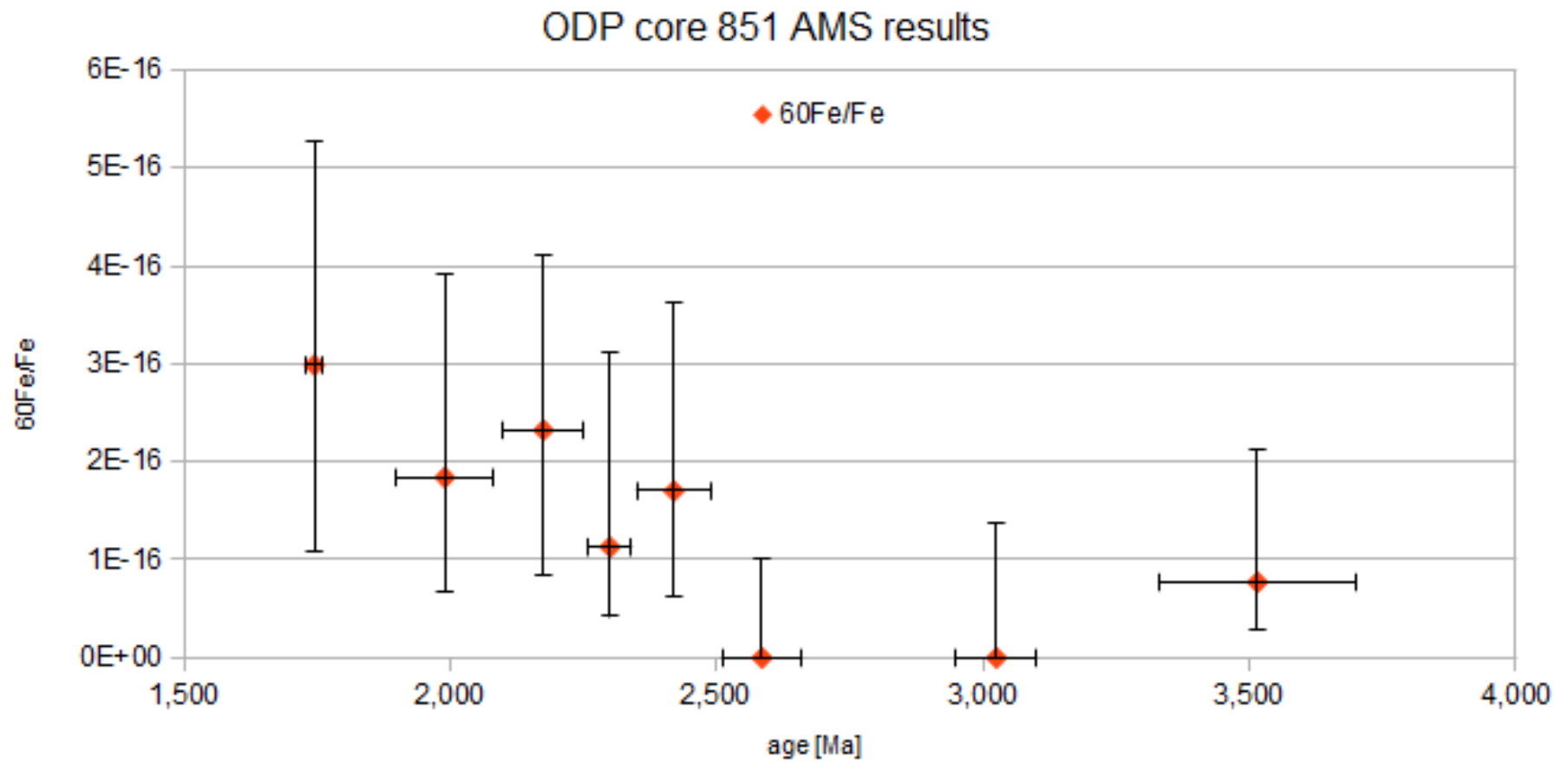
Sediment core 848 – 3 samples grouped each – 8 counts total



----- Blank level (1- σ): $^{60}\text{Fe}/\text{Fe} = 1.7\text{e-}16$
From chemistry blank (0 counts)

Core 851 preliminary results

- ◆ Data analysis still ongoing, better statistics will be available soon
- ◆ The signal is not yet significant enough → more beamtimes necessary
- ◆ 12 counts total detected so far



CORE 848

- ▶ Core sediment used up except for aliquots
- ▶ 90% of AMS samples already measured
- ▶ 8 counts of ^{60}Fe detected in the range 1.9 – 2.7 Ma
- ▶ Average concentration over 1.9 – 2.6 Ma:
 $^{60}\text{Fe}/\text{Fe} \sim 1 \times 10^{-15}$
- ▶ Rough estimation of total number of incident ^{60}Fe atoms can be made
→ local interstellar fluence
 $\Phi_{\text{LIF}} \sim \text{few } 10^7 \text{ } ^{60}\text{Fe cm}^{-2}$

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CORE 851

- ▶ Only ~30% of core measured yet
- ▶ 13 counts of ^{60}Fe detected, among those 12 in the range 1.7-2.5 Ma
- ▶ Average concentration in that range only $2-3 \times 10^{-16}$
- ▶ This is very low → better blank level needed and of course, more counts ^{60}Fe
- ▶ Idea: try magnetic extraction instead of chemical to reduce dilution → currently being set up

CORE 848

- ▶ Core sediment used up except for aliquots
- ▶ 90% of AMS samples already measured
- ▶ 7 counts of ^{60}Fe detected in the range 1.9 – 2.7 Ma
- ▶ Average concentration over 1.9 – 2.6 Ma:
 $^{60}\text{Fe}/\text{Fe} \sim 1 \times 10^{-15}$
- ▶ Rough estimation of total number of incident ^{60}Fe atoms can be made
→ local interstellar fluence
 $\Phi_{\text{LIF}} \sim \text{few } 10^7 \text{ } ^{60}\text{Fe cm}^{-2}$

CORE 851

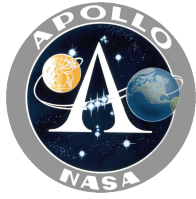
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Most important result so far: **^{60}Fe input** seems to have been **rather long (> 500 kyr)**. This was previously unknown from crust measurements because of low time resolution

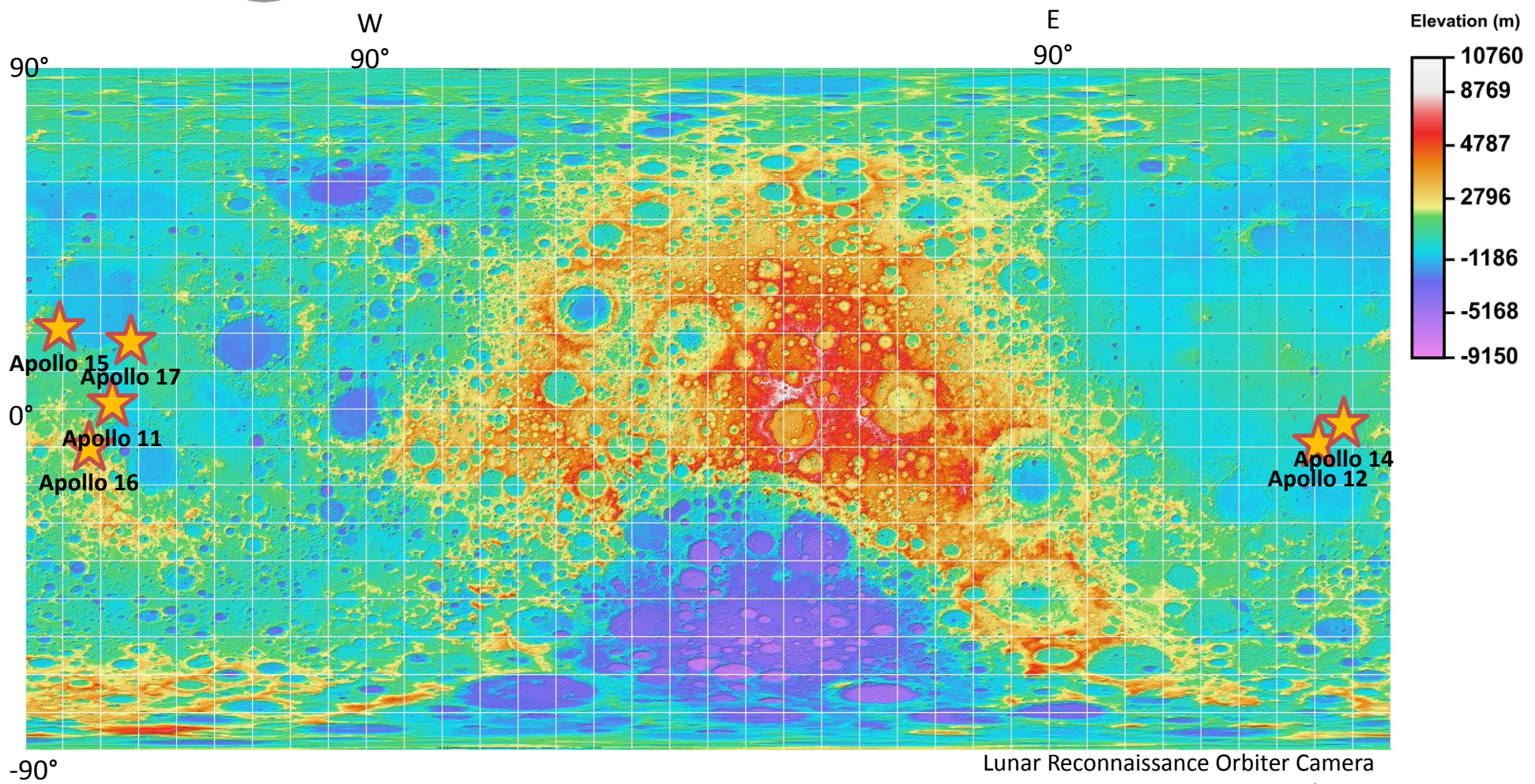
^{60}Fe (and ^{53}Mn) in lunar samples



Apollo Landing Sites



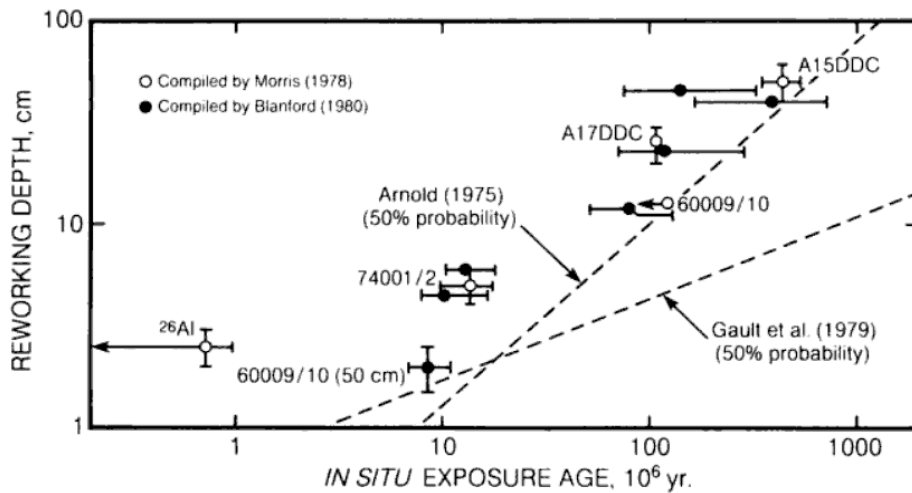
◆ ^{60}Fe and ^{53}Mn Samples from Apollo 12, 15, and 16 obtained



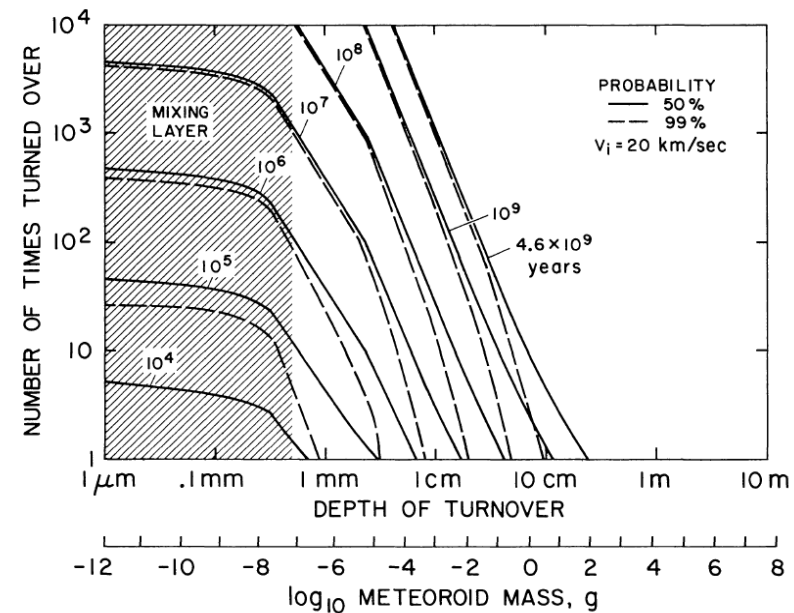
Lunar Reconnaissance Orbiter Camera
 Arizona State University, November 2011

On the moon...

- ✓ Net sedimentation rates are small: U60Fe~100%
- ✓ Ni concentrations are in general low: low in-situ production of 60Fe
- ✗ Gardening of the lunar surface: ~2-3 cm reworking depth in 10Myr
- ✗ Hard to reach

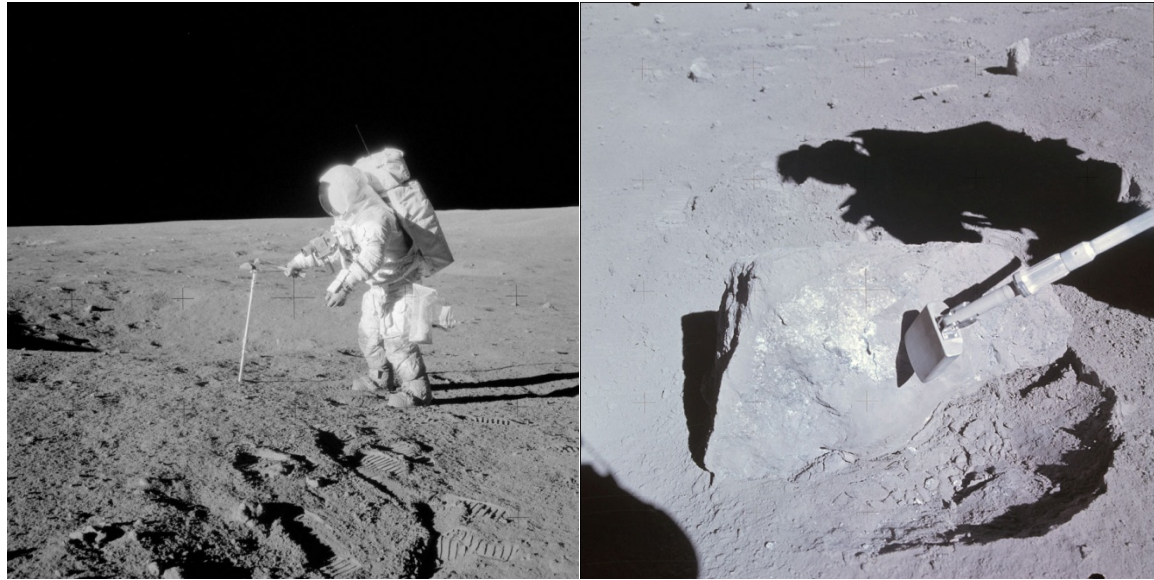


McKay et al. Lunar Sourcebook, Cambridge University Press, pp. 285-356



Gault et al., Proceedings of the 5th Lunar Conference (1974)

Sample history and production of ^{60}Fe



Potential origins:

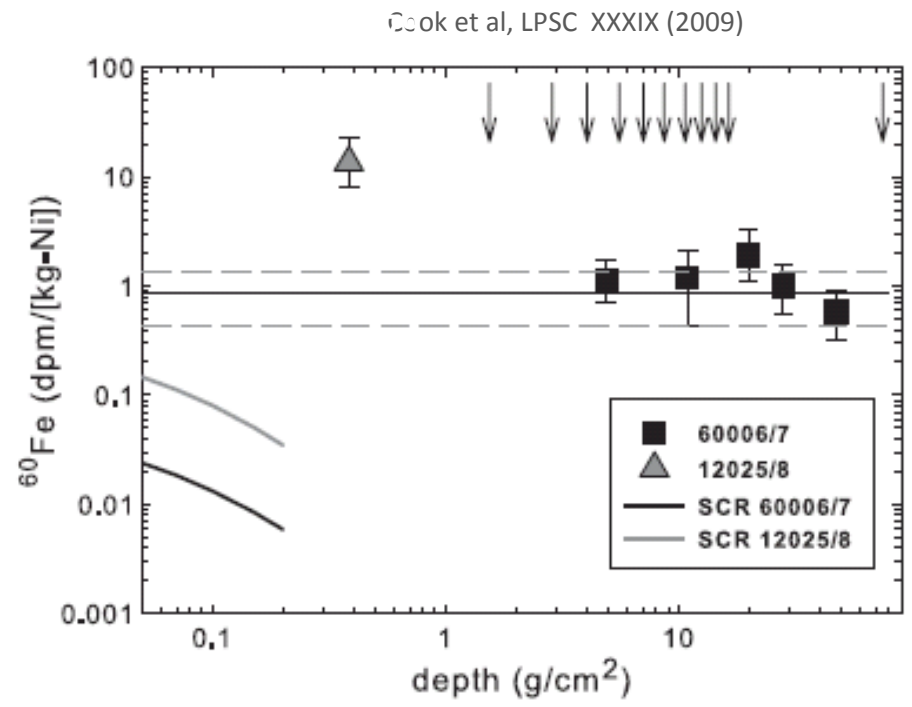
- ✓ Galactic Cosmic Rays
- ✓ Solar Cosmic Rays
- ✓ Deposition of SN debris.

$P_{60\text{Fe-GCR}} = (0.88 \pm 0.44) \text{ dpm/kg Ni}$

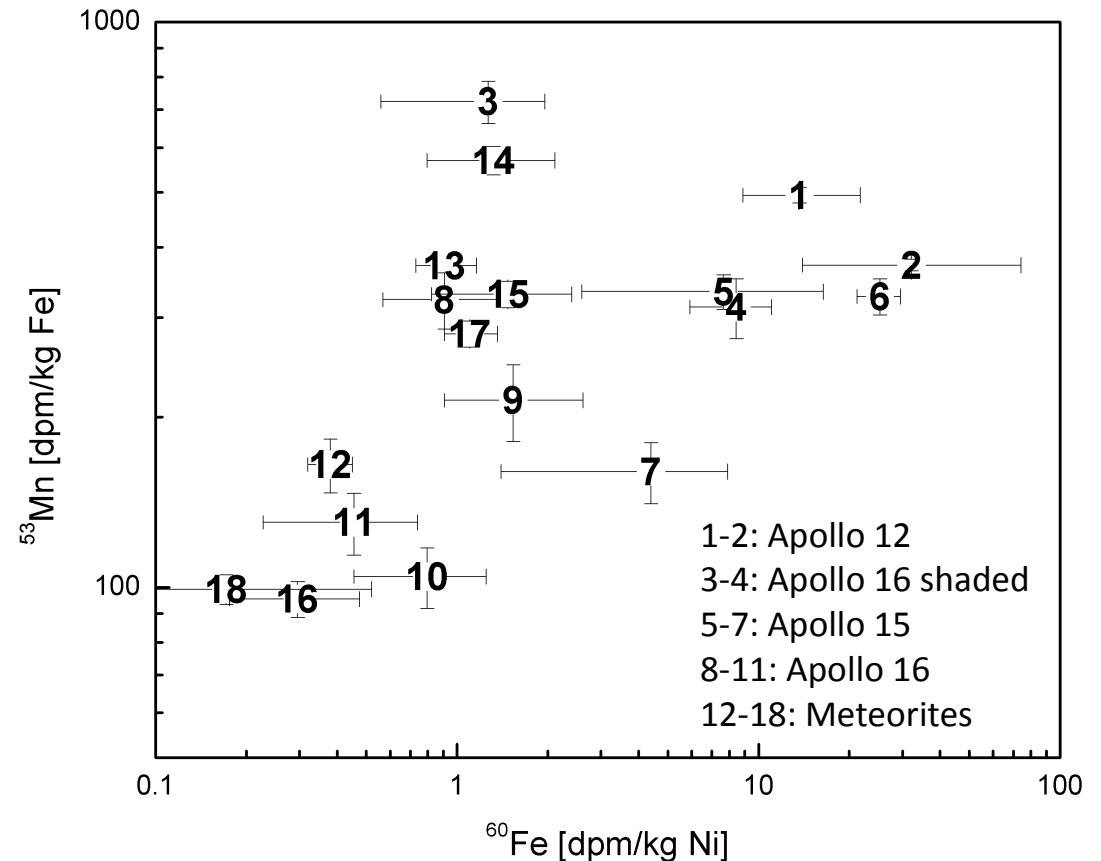
Knie et al, Planet.ary Science Letters 34, 729 (1999)

$P_{60\text{Fe-SCR}} = 0.07 - 0.45 \text{ dpm/kg Ni on the surface}$

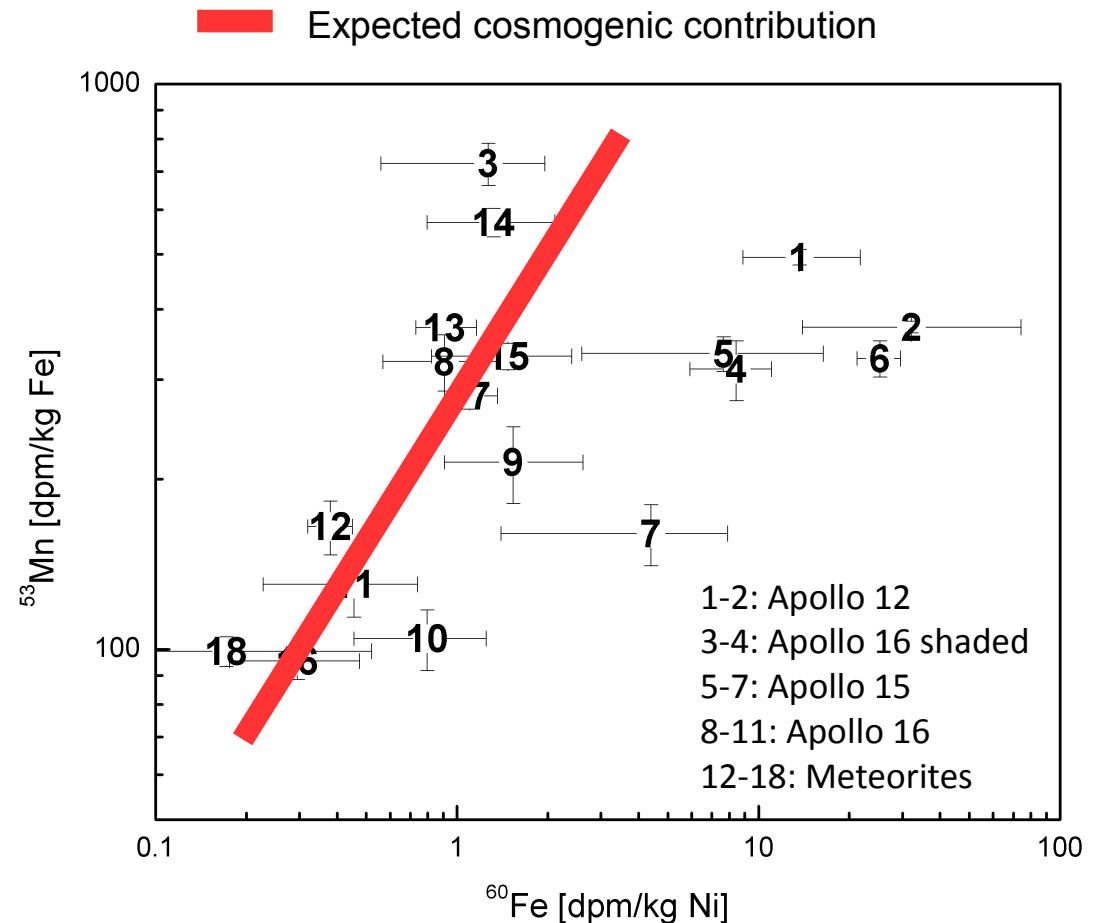
Cook et al, LPSC XXXIX (2009)



- Plot shows both radioisotopes, 53Mn and 60Fe divided by mass of their respective most likely source element under irradiation from cosmic rays
- Enhancement in both isotopes for some samples visible

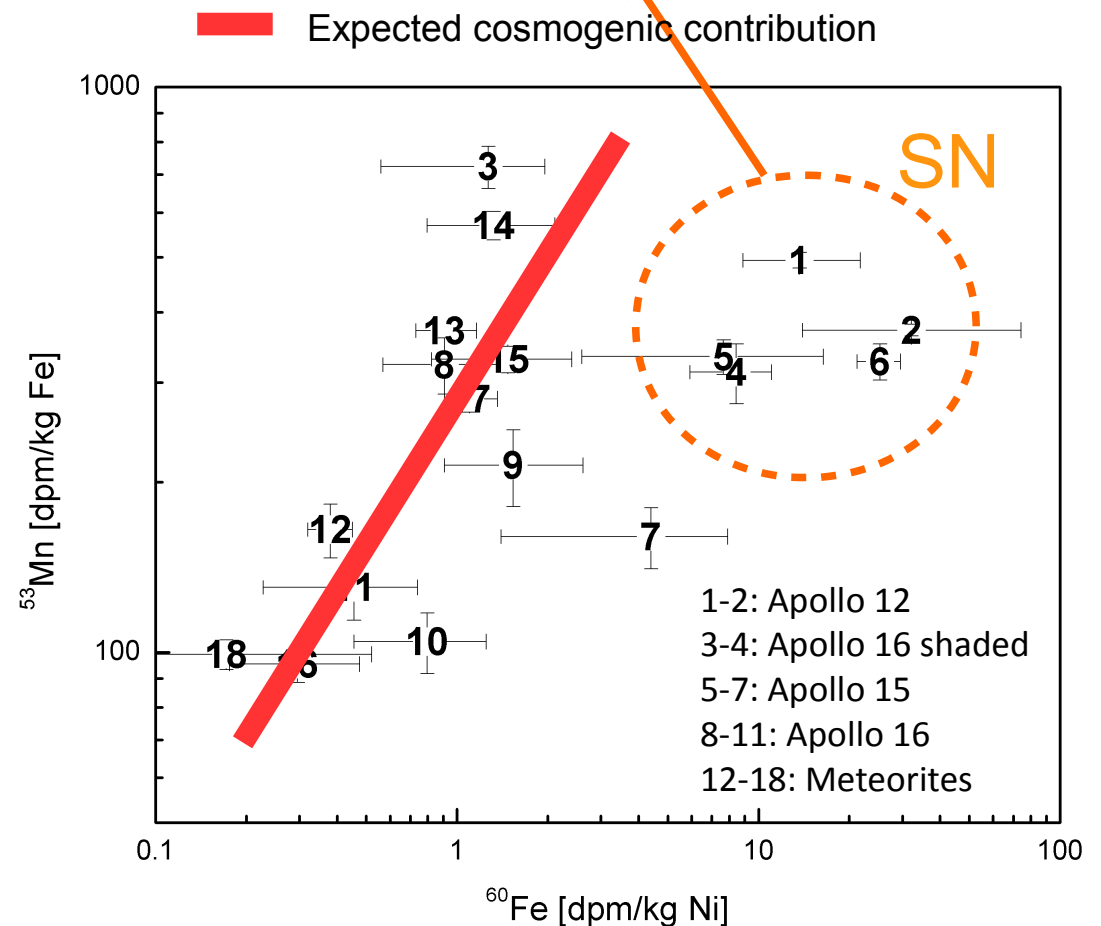


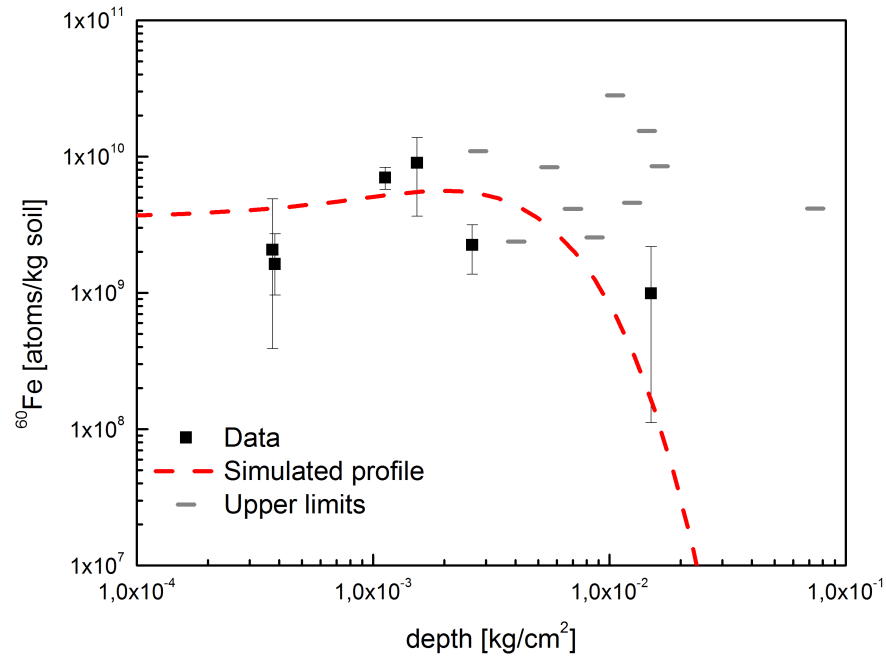
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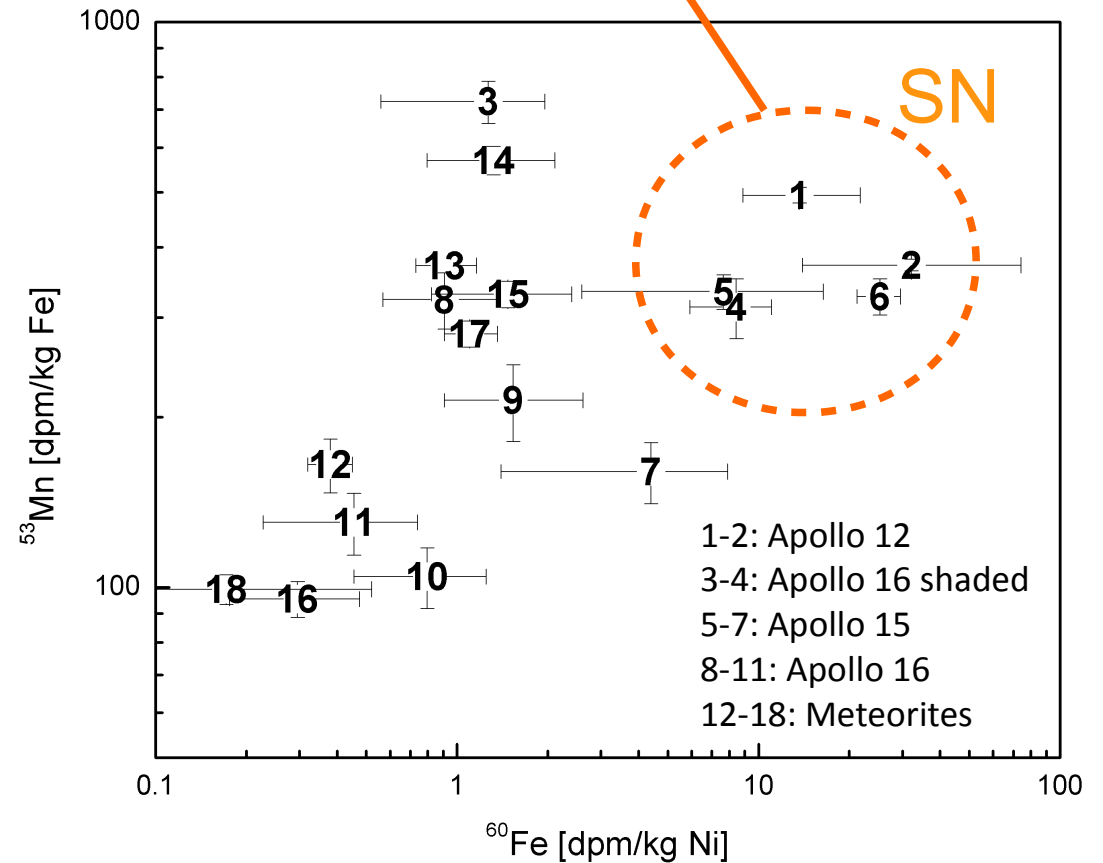
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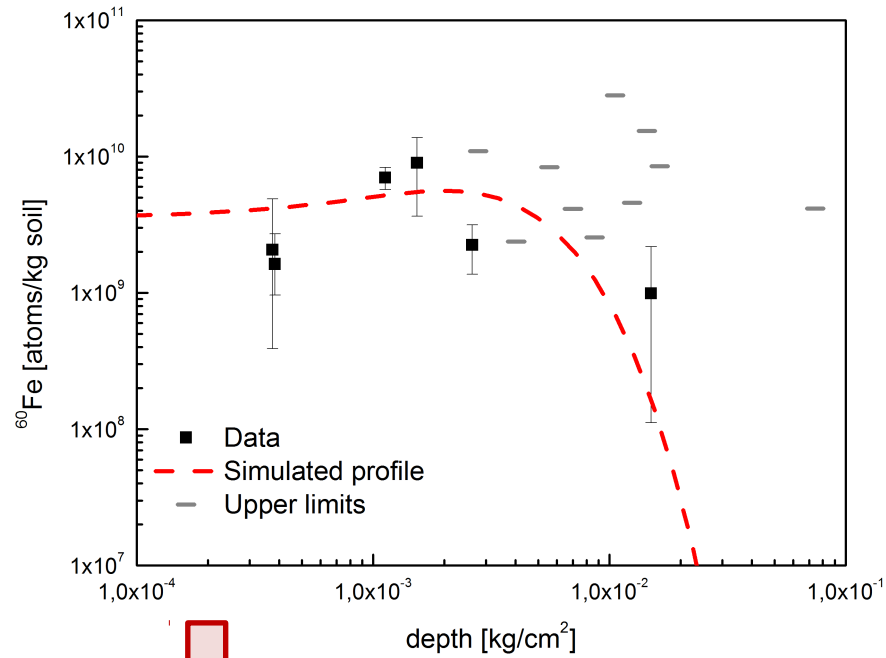
Excess in 60Fe in 5 samples interpreted as supernova input





Excess in ⁶⁰Fe in 5 samples interpreted as supernova input

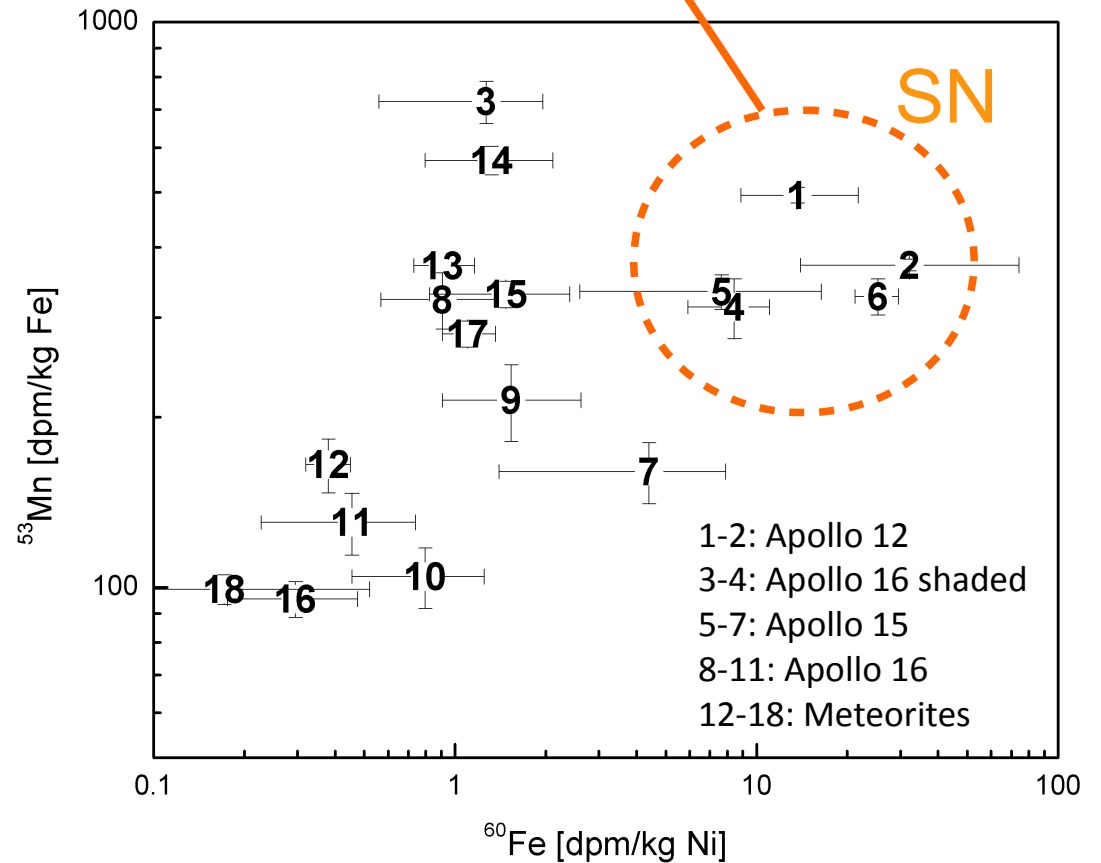




Ingetrate to obtain
Local interstellar fluence:

$$\Phi_{^{60}\text{Fe,LIS}} \approx 3.5 \times 10^7 \frac{\text{at}}{\text{cm}^2}$$

Excess in ^{60}Fe in 5 samples interpreted as supernova input

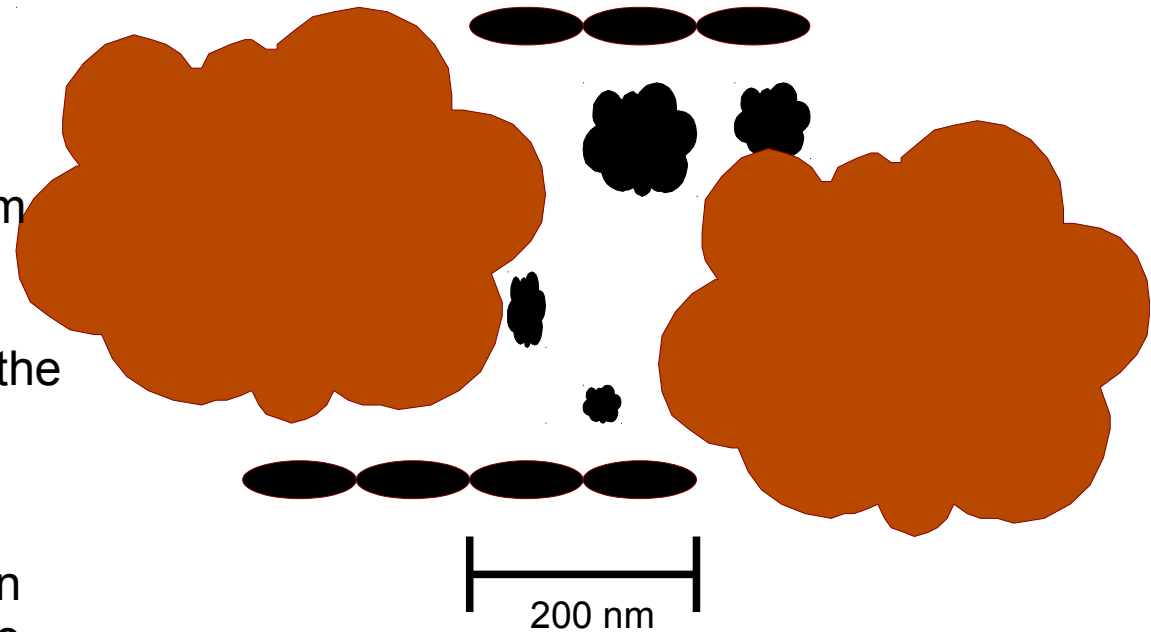


Summary

- AMS is an ultrasensitive technique for isotope ratio measurement
- Sensitivities reaching down to and below $1\text{E-}16$
- Applications in nuclear astrophysics
- Setup at MLL in Garching with 2 AMS beamlines
- GAMS beamline for isobar suppression (intermediate A)
- New results: ^{60}Fe signature found in sediment (~ 20 counts so far but: more statistics required \rightarrow further beamtimes
 \rightarrow favors long input time (~ 500 kyr)
- Lunar samples scanned for ^{53}Mn and ^{60}Fe
- Enhancement of ^{60}Fe found in 5 samples \rightarrow SN input
 $\rightarrow \phi_{\text{LIS}} \sim 3.5 \times 10^7 \text{ at/cm}^2$ (not decay-corrected)

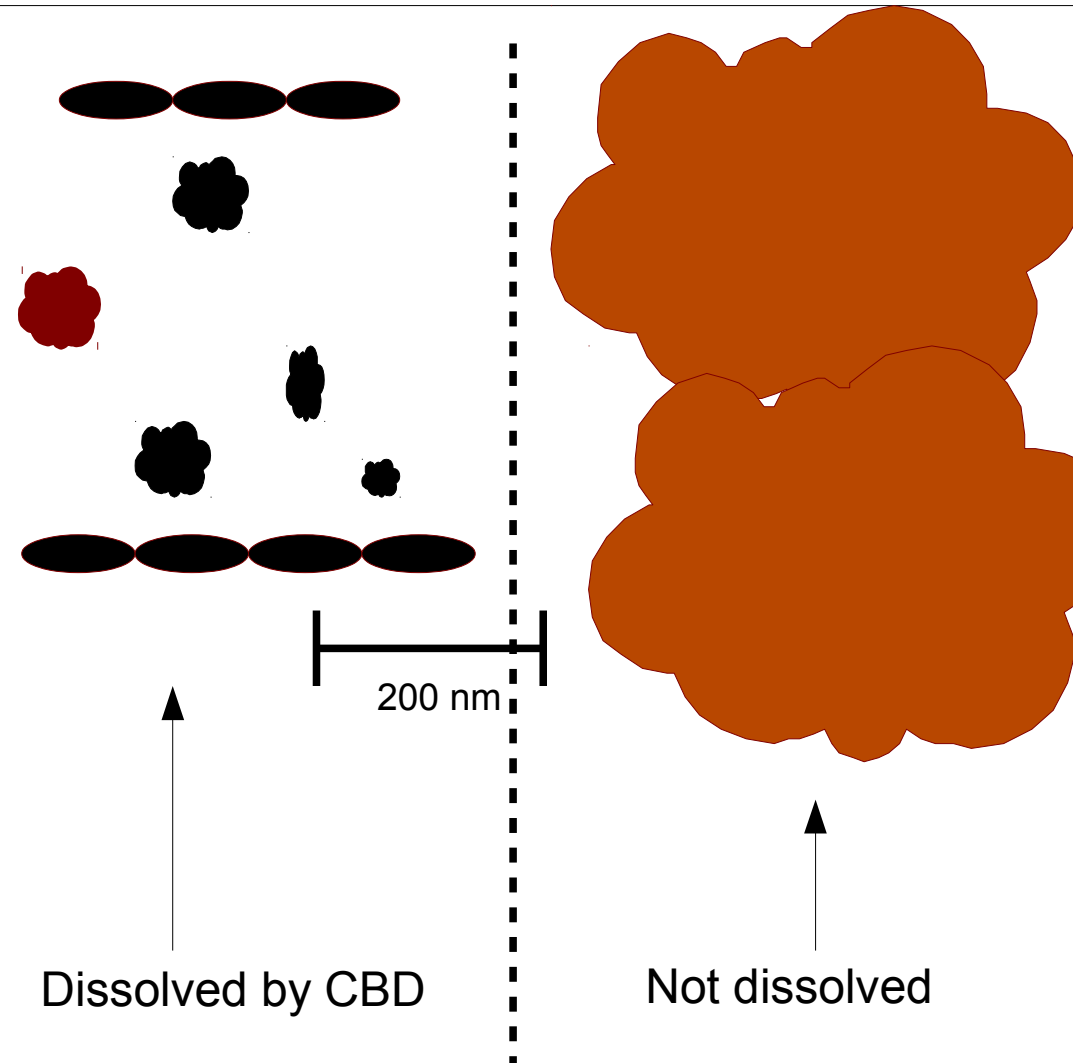
Thank you for your attention

- Fe content of drill core material ~1-2%
- ^{60}Fe expected in small iron oxide (e.g. magnetite) grains: size $\ll 1\mu\text{m}$ in the form of bacteria fossils and others
- Primary iron oxide (brown) would dilute the signal contained in secondary minerals (black)
- Chemical extraction of small grained iron oxides using Citrate-bicarbonate-dithionite (CBD) extraction method: Dissolves mainly small grained material
 - Dithionite: Strong reducing agent ($\text{Fe III}^+ \rightarrow \text{Fe II}^+$)
 - Citrate: extracts and chelates Fe II^+
 - Sodium-Bicarbonate: pH buffer



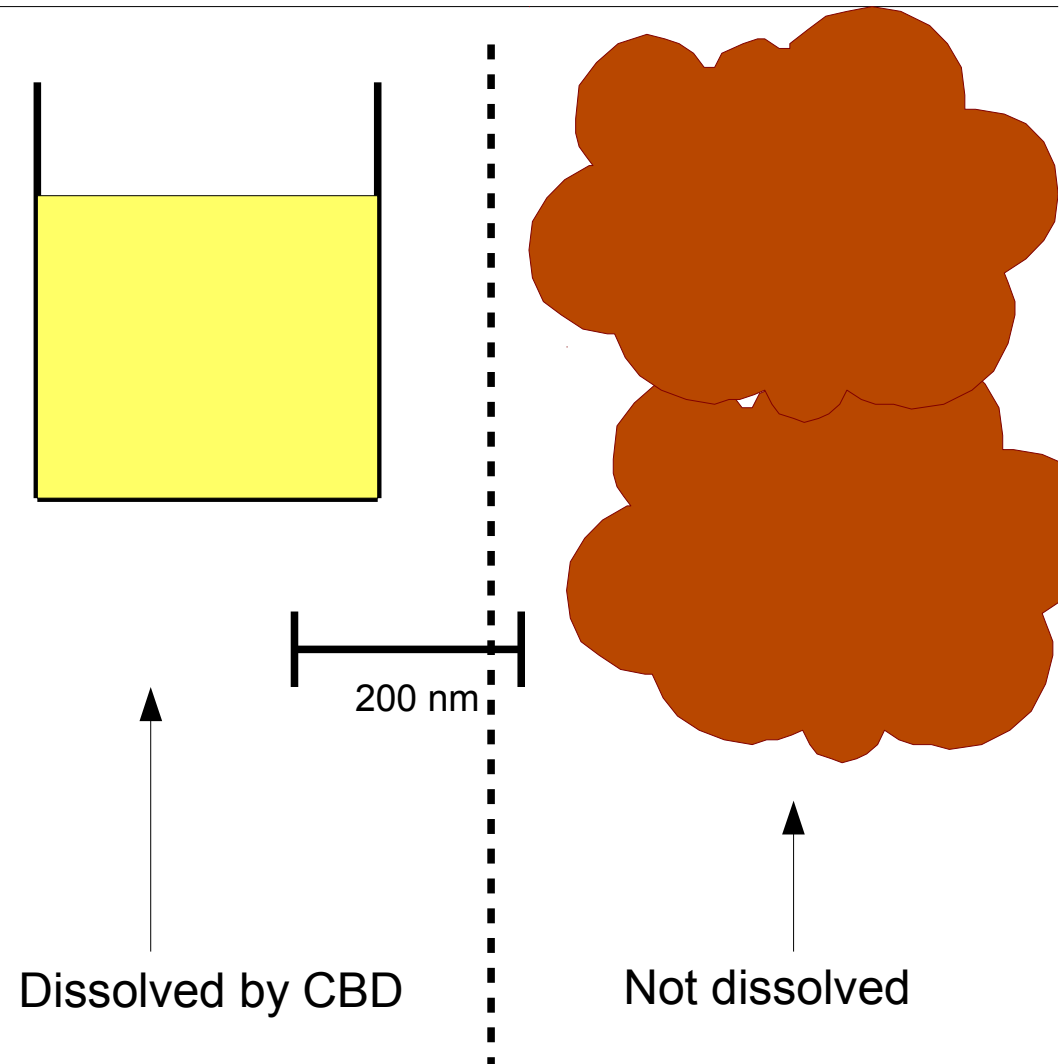
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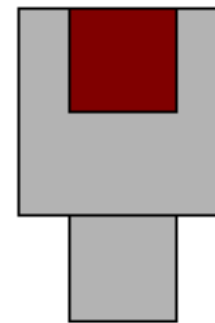
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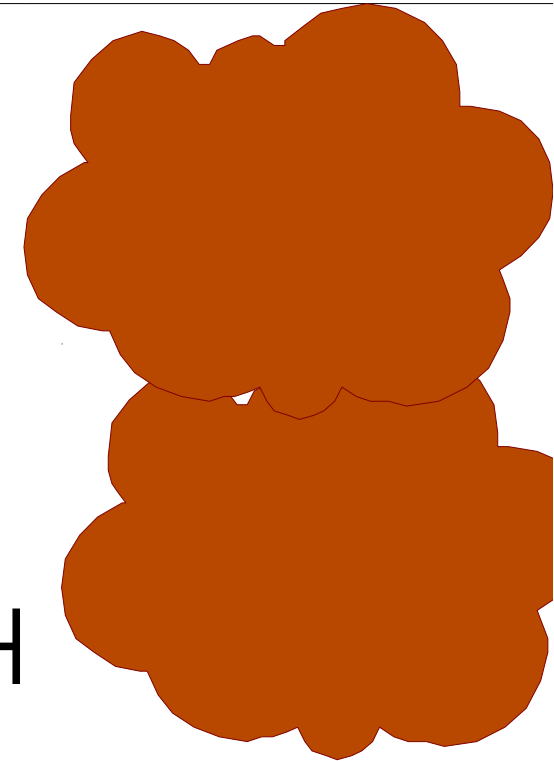


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~ 5 mg Fe₂O₃
AMS sample

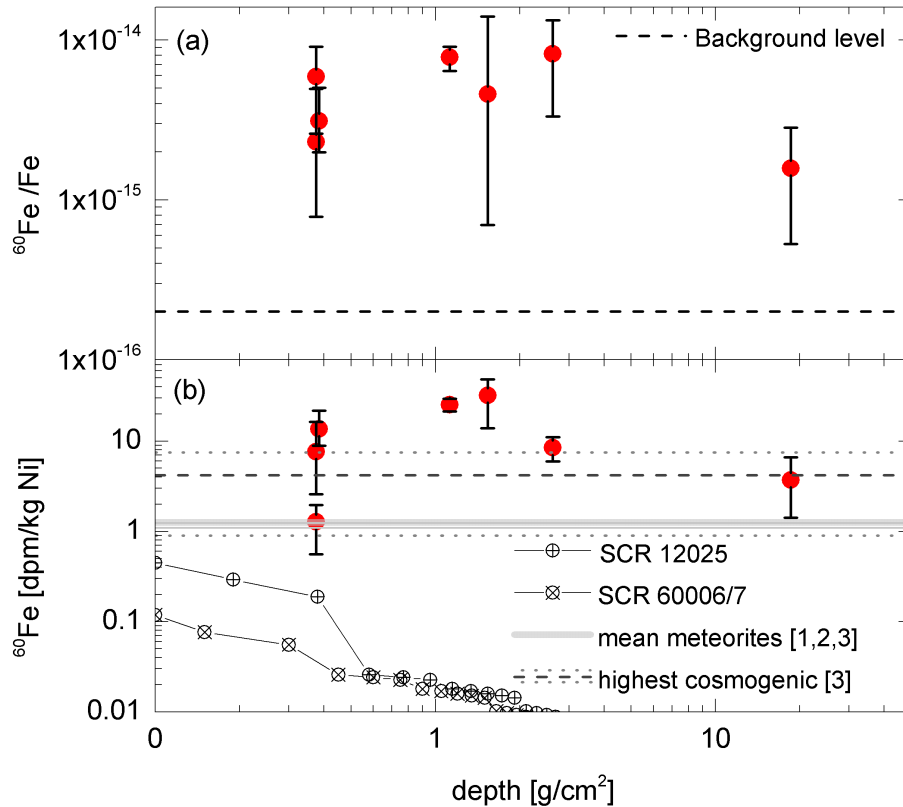


200 nm



Not dissolved

New ^{60}Fe measurements



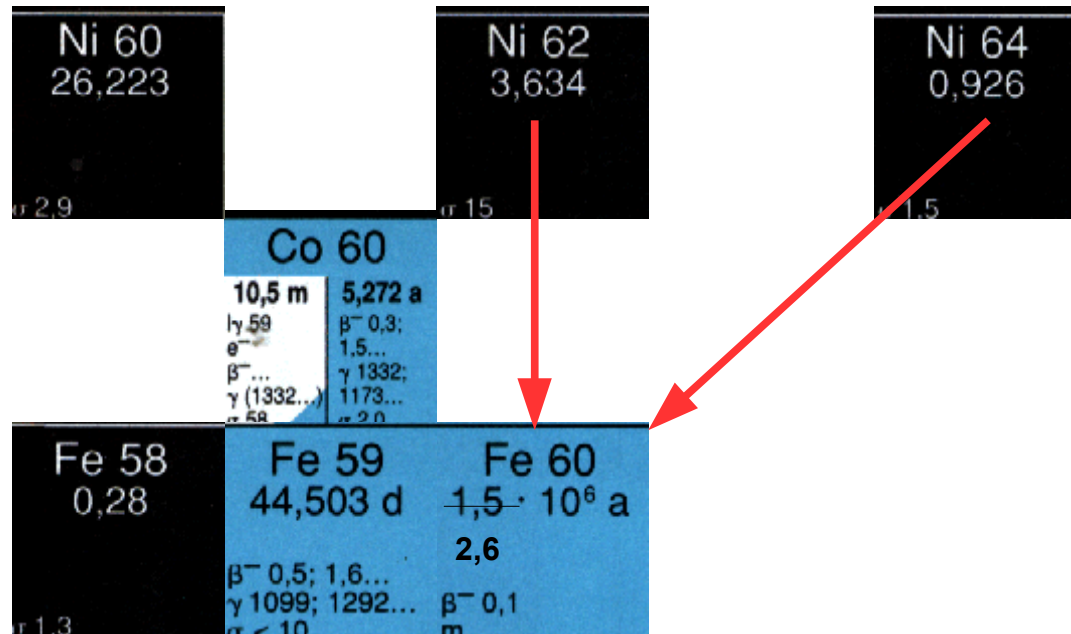
◆ New ^{60}Fe measurements show ^{60}Fe counts in 7 samples

◆ To distinguish from cosmic ray production look at $^{60}\text{Fe}/\text{kg}(\text{Ni})$

→ 3 samples show significantly higher values than highest cosmogenic production observed in meteorites

→ identification as SN input possible

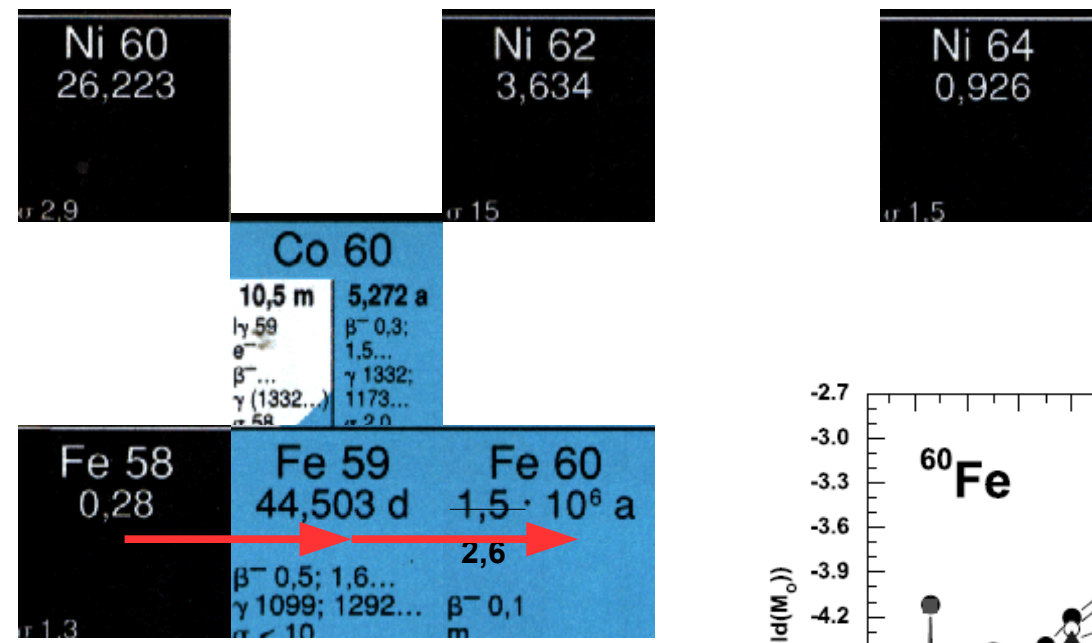
^{60}Fe production: Spallation



Cosmic ray spallation:

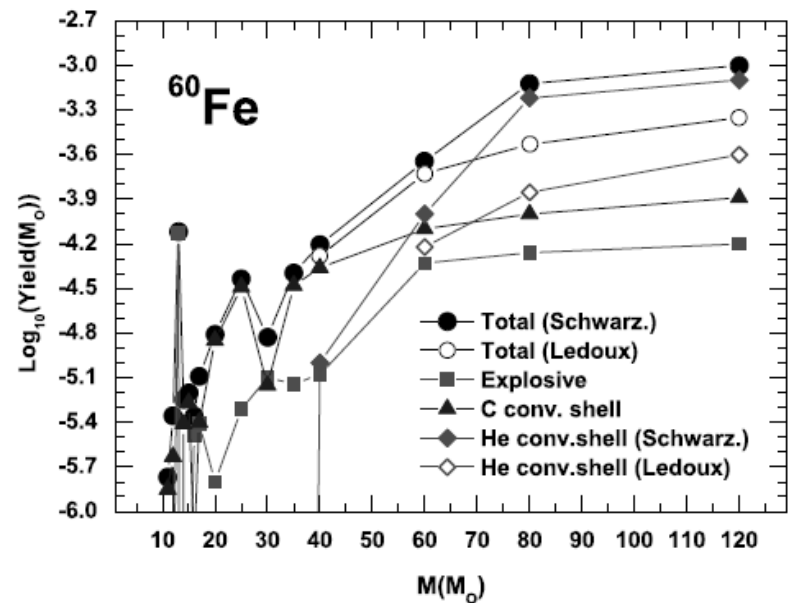
- ^{60}Fe production in cosmic ray spallation mostly on Ni targets
- On Earth, atmosphere prevents large build-up - background $^{60}\text{Fe}/\text{Fe} < 10^{-16}$
- In meteorites and lunar samples, this is not given

^{60}Fe production: Stellar



Production in stars:

- 2 Neutron captures (s-process) on ^{58}Fe
- Shell He burning in massive stars ($M > 40 M_{\text{sun}}$)
- Shell C burning in massive stars ($M < 40 M_{\text{sun}}$)
- Explosive synthesis in SN when shockwave passes through shells \rightarrow small contribution

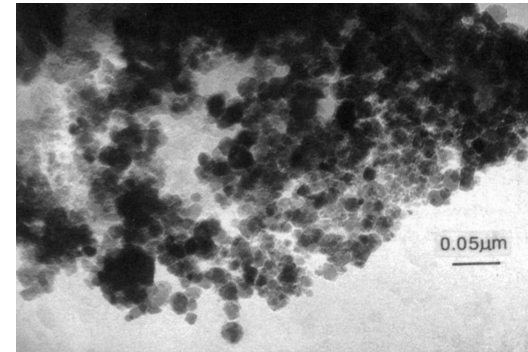
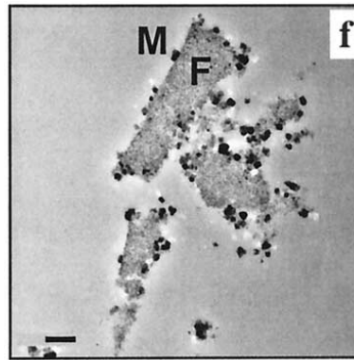
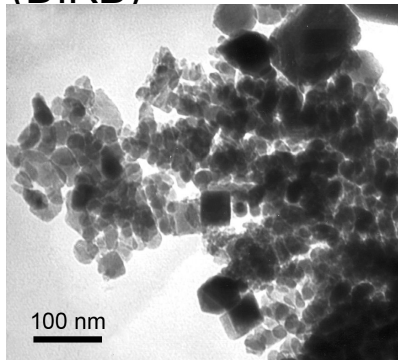


Limongi, M., & Chieffi, A. 2006, ApJ, 647, 483

Magnetite in sediment

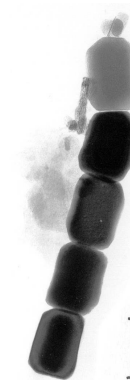
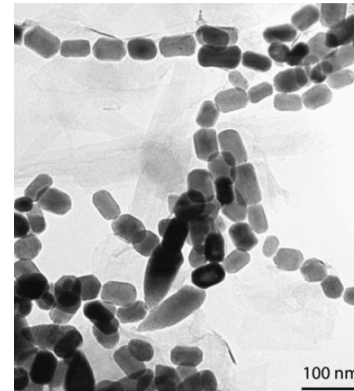
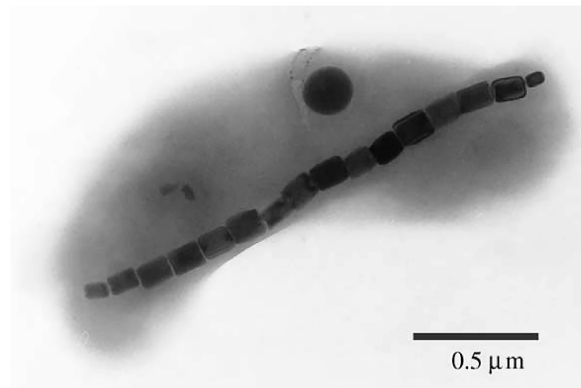
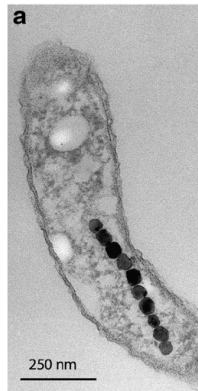
“Extracellular magnetite”

Secondary magnetite precipitated from redox reactions, either *inorganically*, or by mediation of *dissimilatory metal reducing bacteria* (DIRB)



“Biogenic magnetite”

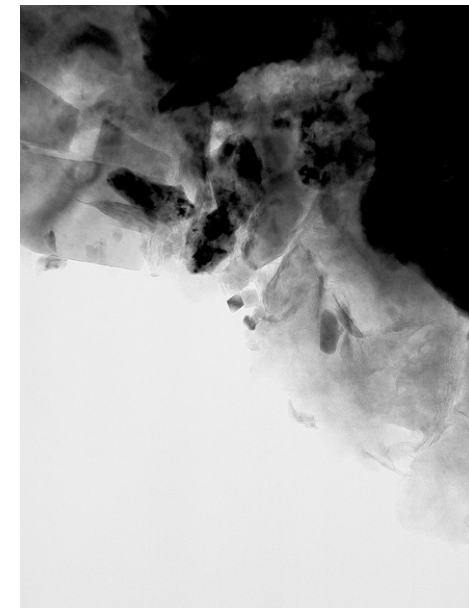
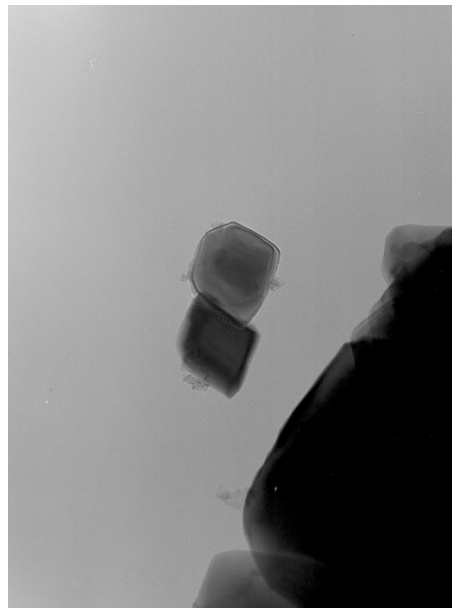
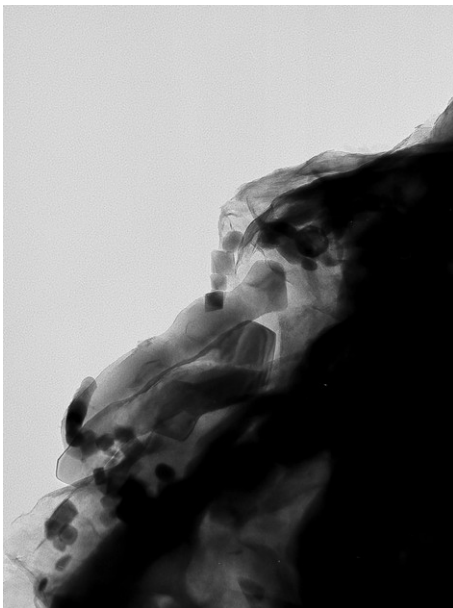
Magnetosomes and magnetosome chains produced by *magnetotactic bacteria* (MB)



Do we have biogenic magnetite in our sediment?

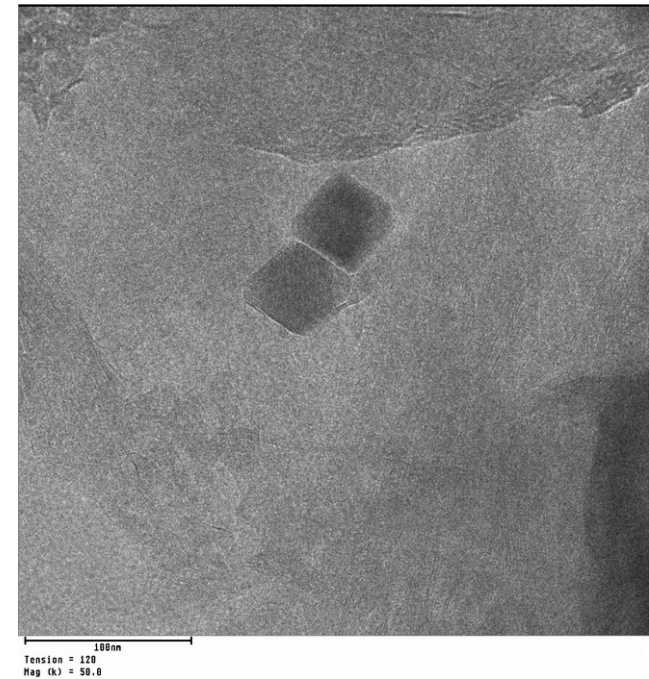
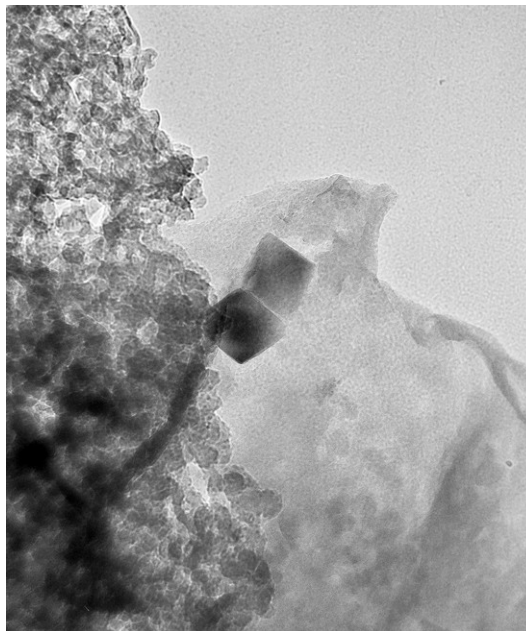
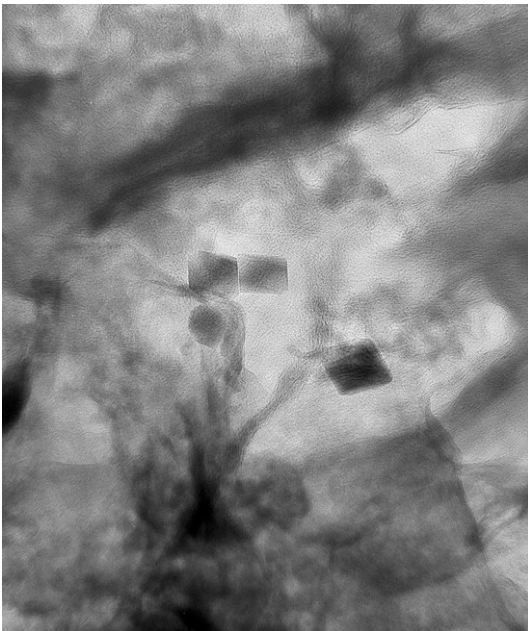
Transmission electron microscopy (TEM):

- ♦ Setup at TUM Chemistry (Marianne Hanzlik)
- ♦ First try: TEM on untreated sediment
→ extremely difficult and time consuming
- ♦ Results show at least some biogenically-looking magnetite particles (magnetite identified by diffraction analysis)



Transmission electron microscopy (TEM):

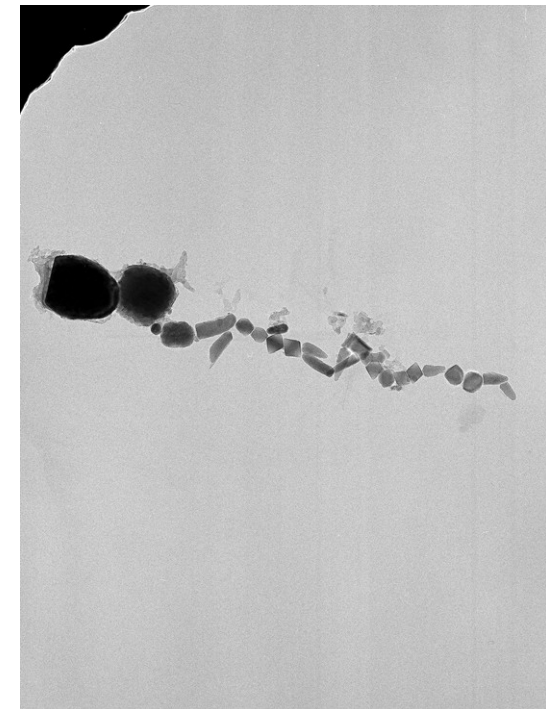
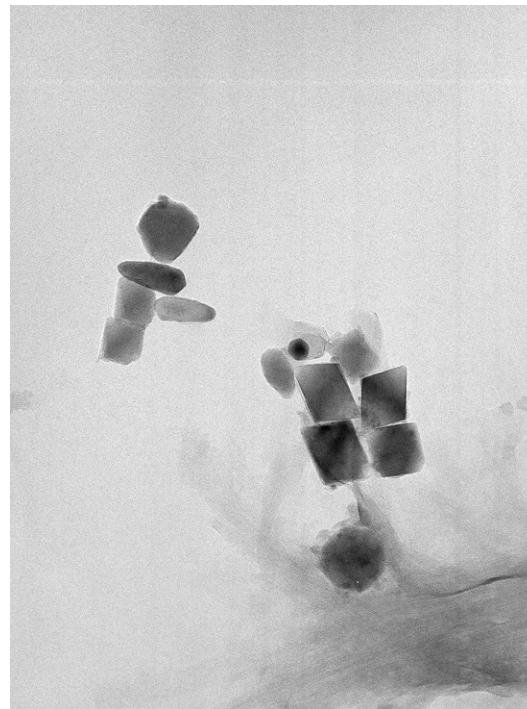
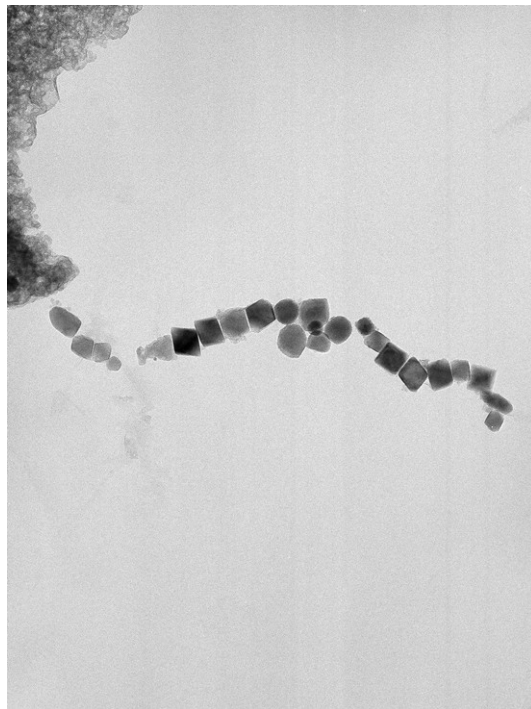
- ♦ Second try: pre-treatment of sediment with AcOH (20%) for 1 hour
→ reduction of sample mass by 80% → calcite matrix dissolved
- ♦ Still extremely time-consuming and difficult



TEM on Sediment

Transmission electron microscopy (TEM):

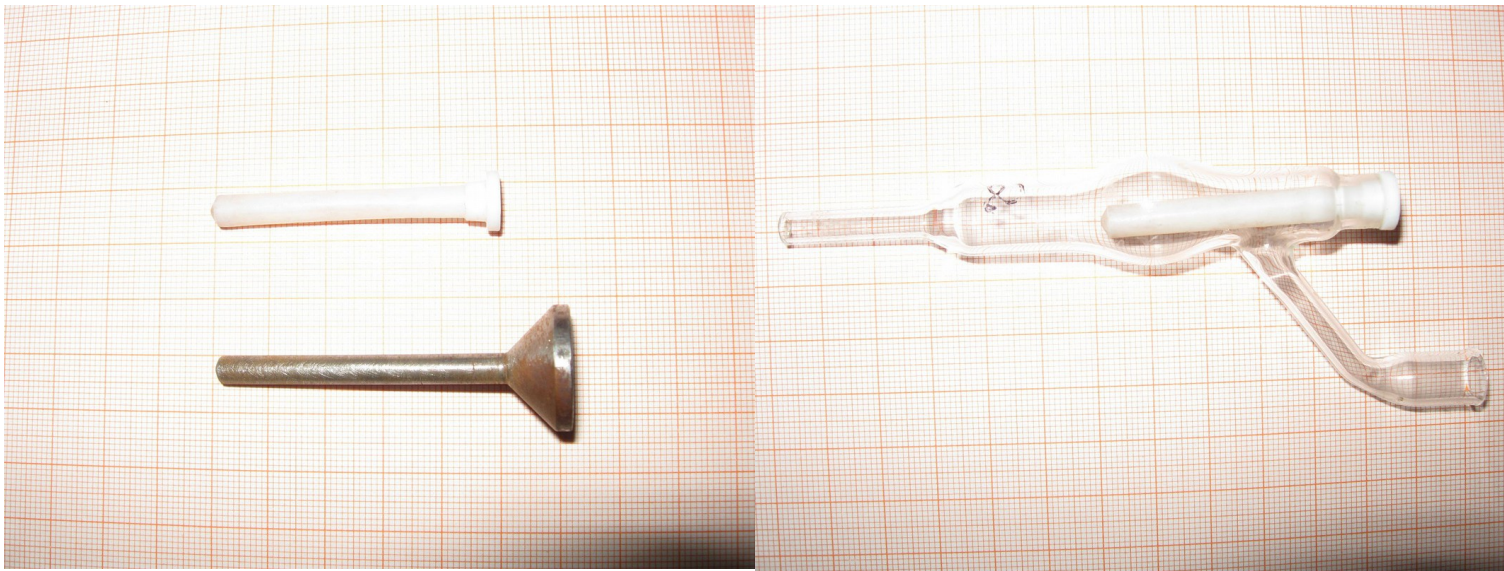
- ♦ Third try: Insert magnetic finger into sample from try #2
- ♦ Much easier to find magnetite formations: chains and clusters can be seen
- ♦ **HOWEVER:** Are these originally present in the sediment? Or just formed during magnetic extraction? → impossible to interpret



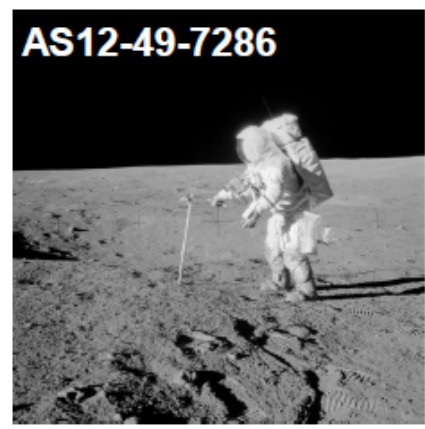
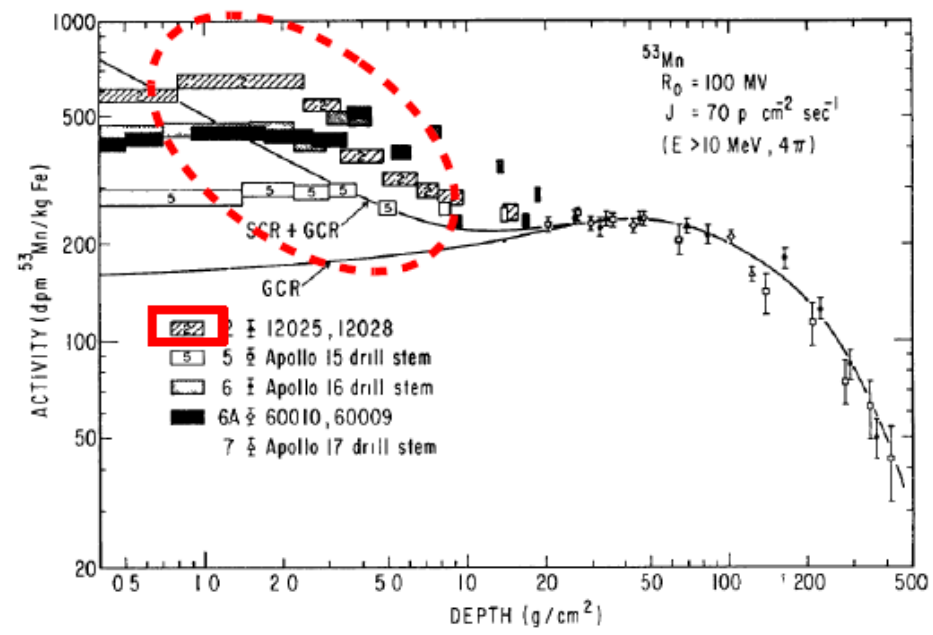
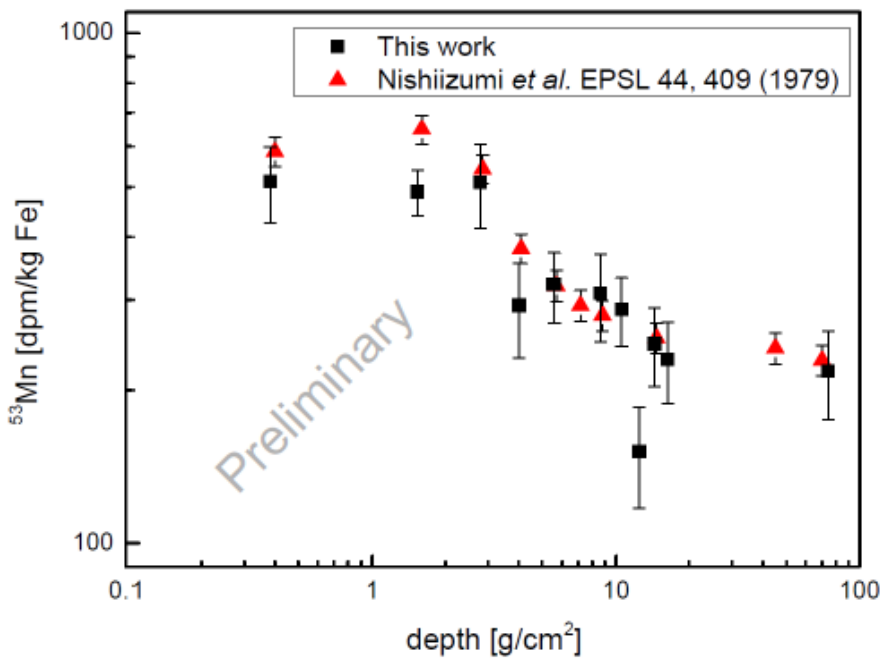
- ♦ Attempt to obtain more sediment of core 848 in SN region
 - Currently being discussed, samples may be available

- ♦ Try alternative extraction method:
 - Dissolve calcite matrix with AcOH (20%)
 - Perform magnetic extraction
 - Optionally then CBD extraction
 - AMS

The magnetic extraction is currently being set up in Garching as a Bachelor's Thesis



53Mn in lunar samples



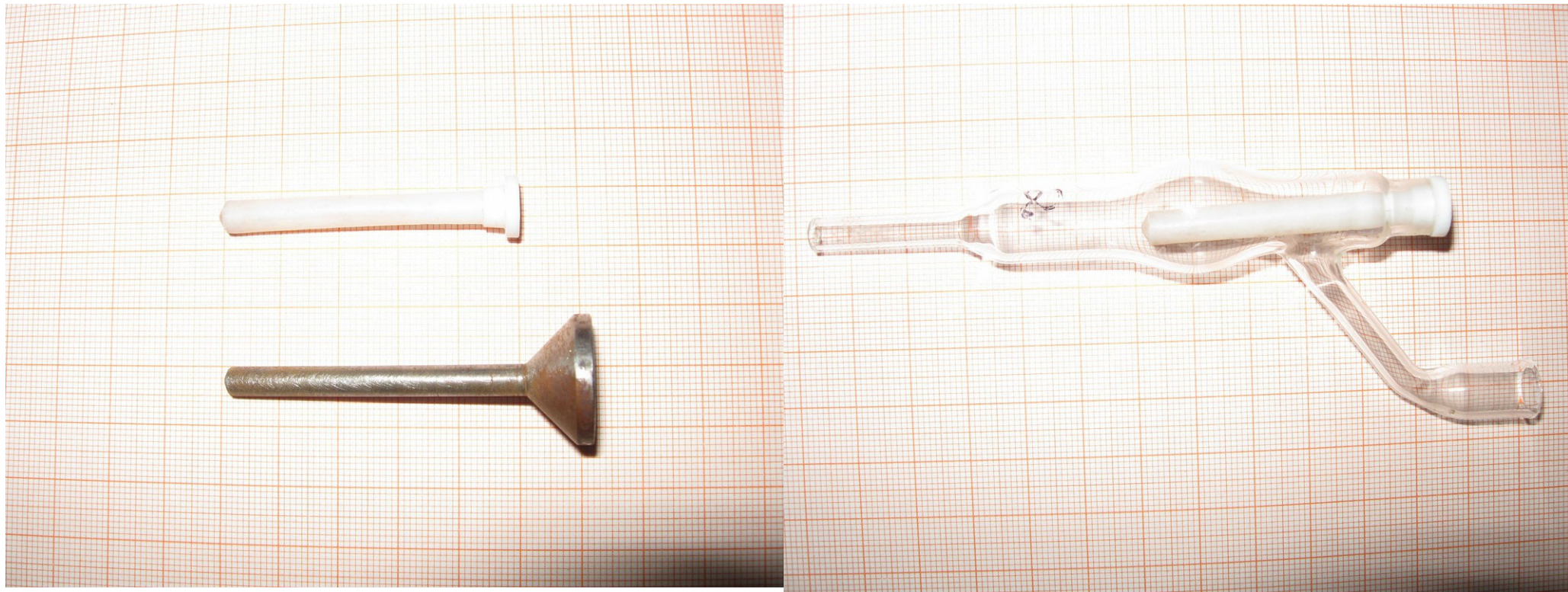
Excess of ⁵³Mn?



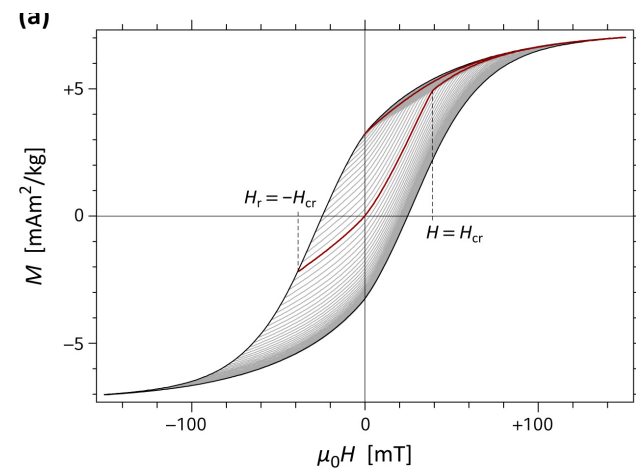
- ? Mixing of the regolith
- ? Addition of irradiated material
- ? Deposition of SN debris
- ? ...

Transmission electron microscopy (TEM):

- ◆ Fourth try: Perform magnetic extraction directly on sediment
- ◆ VIDEO



Use an Alternate Gradient Field Magnetometer (in Bremen) to record a set of 450 first-order reversal curves (FORCs), 4-5 times → total time ~1 day per sample



Chemical isolation of iron

Procedure for ~30g sediment



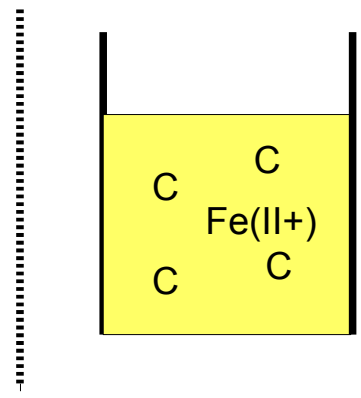
Magnetosomes in Sediment:
 $Fe_3O_4 = Fe(II+) + Fe(III+)$

1h, 40 °C

CBD extraction
 $Fe(III+) \rightarrow Fe(II+)$



Filtration: remove undissolved sediment



Chemical isolation of iron

Procedure for ~30g sediment



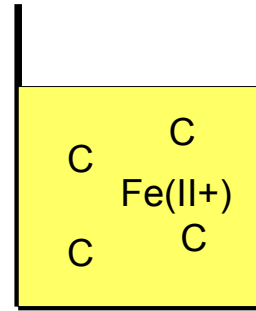
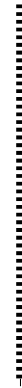
Magnetosomes in Sediment:
 $Fe_3O_4 = Fe(II+) + Fe(III+)$

1h, 40 °C →

CBD extraction
 $Fe(III+) \rightarrow Fe(II+)$



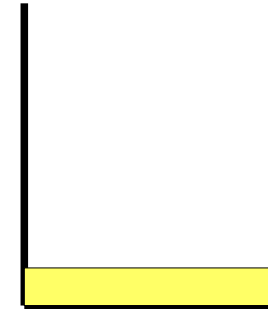
Filtration: remove undissolved sediment



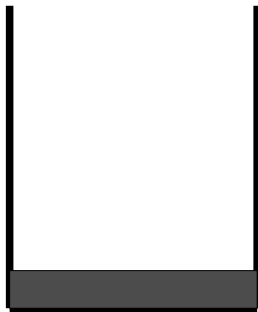
Fe(II+) Chelate
 In 200ml H₂O



evaporation



300°C
 → Citrate
 decomposes



Chemical isolation of iron

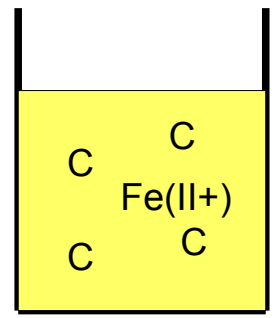
Procedure for ~30g sediment



Magnetosomes in Sediment:
 $Fe_3O_4 = Fe(II+) + Fe(III+)$

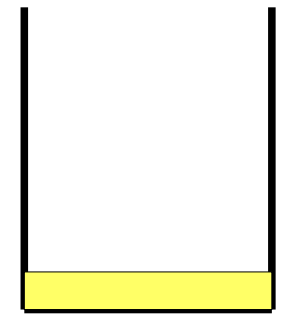
1h, 40 °C

CBD extraction
 $Fe(III+) \rightarrow Fe(II+)$



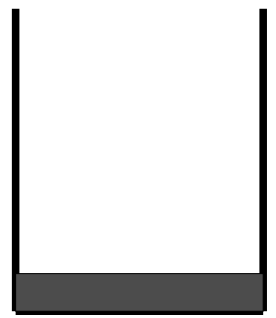
Fe(II+) Chelate
In 200ml H₂O

evaporation

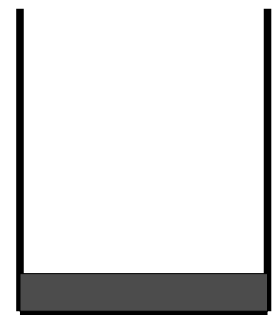


Filtration: remove undissolved sediment

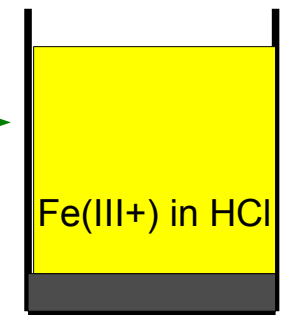
300°C
→ Citrate
decomposes



+HNO₃
 $Fe(II+) \rightarrow Fe(III+)$



+HCl → Fe(III+)
Extracted
+ centrifuge



Chemical isolation of iron

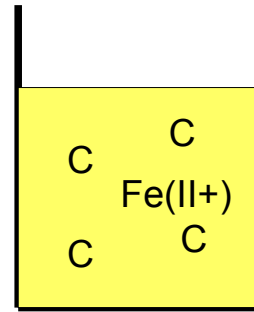
Procedure for ~30g sediment



Magnetosomes in Sediment:
 $Fe_3O_4 = Fe(II+) + Fe(III+)$

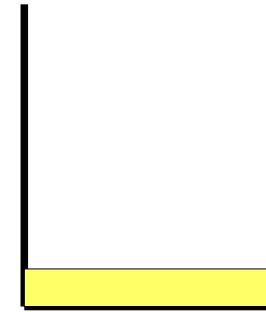
1h, 40 °C

CBD extraction
 $Fe(III+) \rightarrow Fe(II+)$



Fe(II+) Chelate
In 200ml H₂O

evaporation

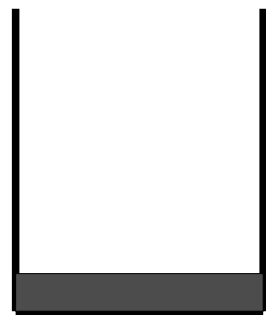


Filtration: remove undissolved sediment



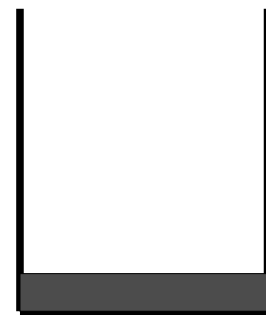
300°C

→ Citrate decomposes



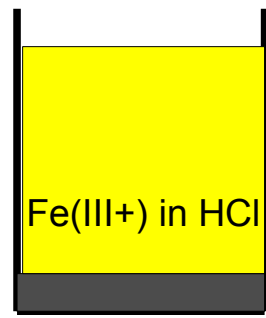
+HNO₃

$Fe(II+) \rightarrow Fe(III+)$



+HCl → Fe(III+)

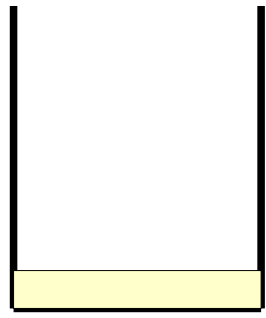
Extracted
+ centrifuge



Fe(III+) in HCl

Evaporation

Until dry
→ white residue



Chemical isolation of iron

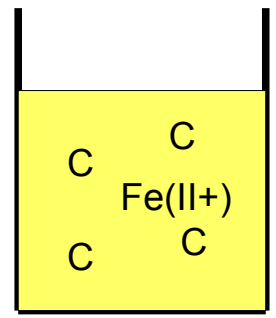
Procedure for ~30g sediment



Magnetosomes in Sediment:
 $Fe_3O_4 = Fe(II+) + Fe(III+)$

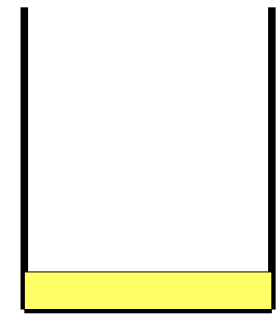
1h, 40 °C

CBD extraction
 $Fe(III+) \rightarrow Fe(II+)$



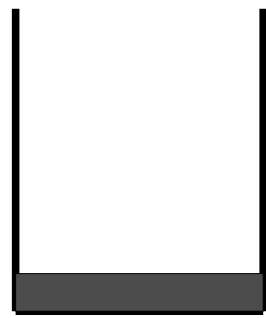
Fe(II+) Chelate
 In 200ml H₂O

evaporation

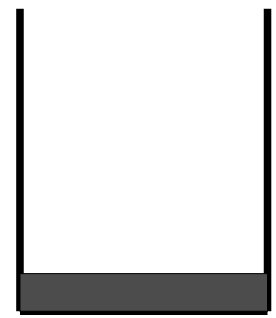


Filtration: remove undissolved sediment

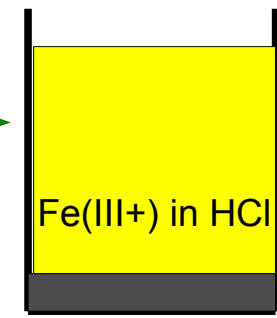
300°C
 → Citrate decomposes



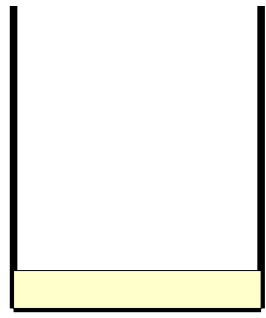
+HNO₃
 $Fe(II+) \rightarrow Fe(III+)$



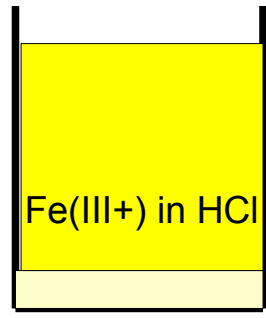
+HCl → Fe(III+)
 Extracted
 + centrifuge



Evaporation
 Until dry
 → white residue



20 ml Hcl
 centrifuge



Chemical isolation of iron

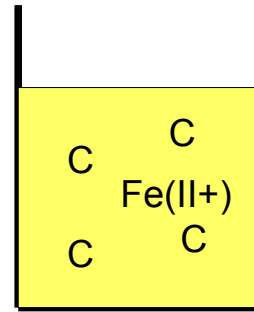
Procedure for ~30g sediment



Magnetosomes in Sediment:
 $Fe_3O_4 = Fe(II+) + Fe(III+)$

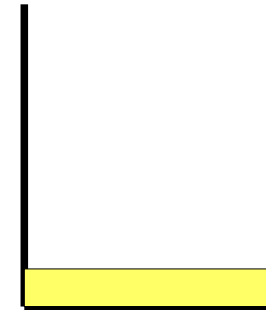
1h, 40 °C

CBD extraction
 $Fe(III+) \rightarrow Fe(II+)$



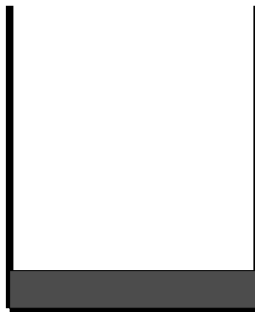
Fe(II+) Chelate
In 200ml H₂O

evaporation

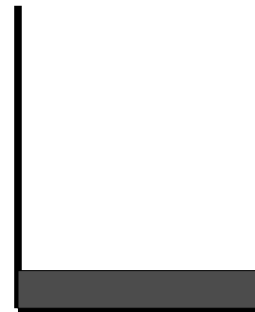


Filtration: remove undissolved sediment

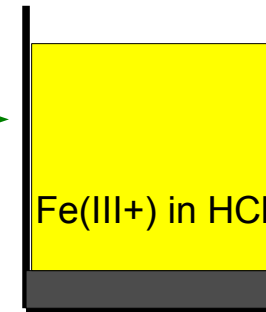
300°C
→ Citrate
decomposes



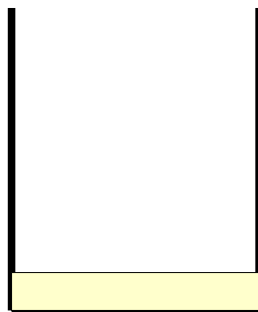
+HNO₃
 $Fe(II+) \rightarrow Fe(III+)$



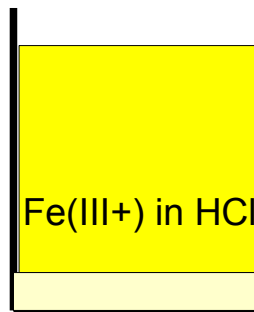
+HCl → Fe(III+)
Extracted
+ centrifuge



Evaporation
Until dry
→ white residue

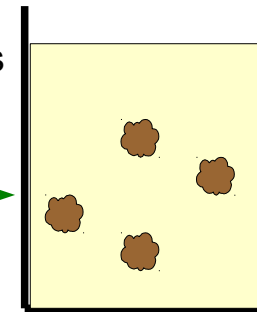


20 ml Hcl

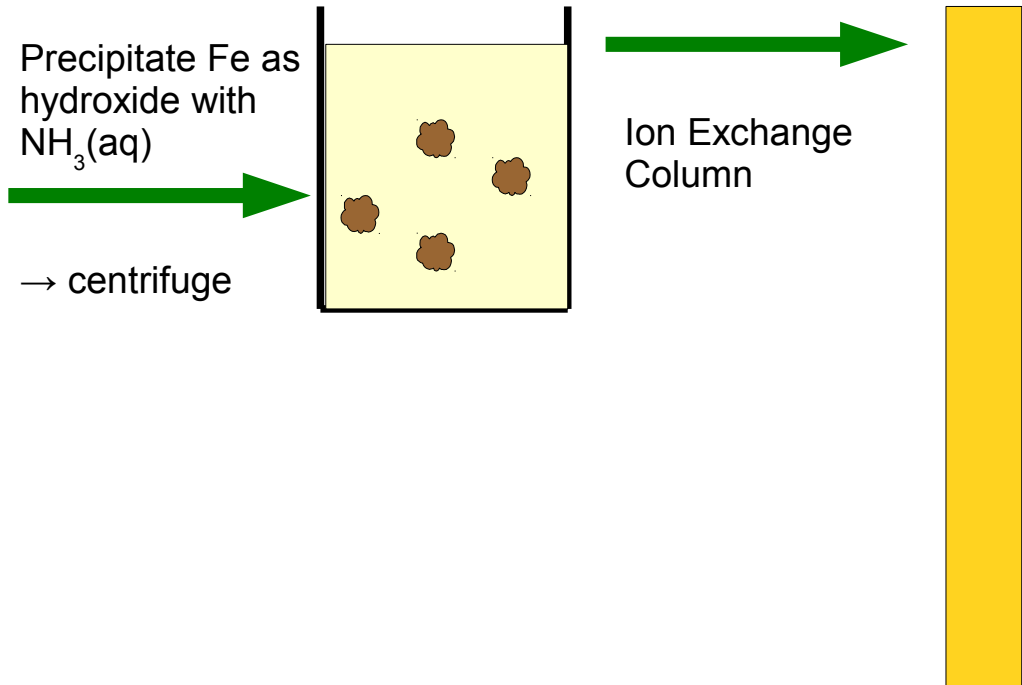


Precipitate Fe as
hydroxide with
NH₃(aq)

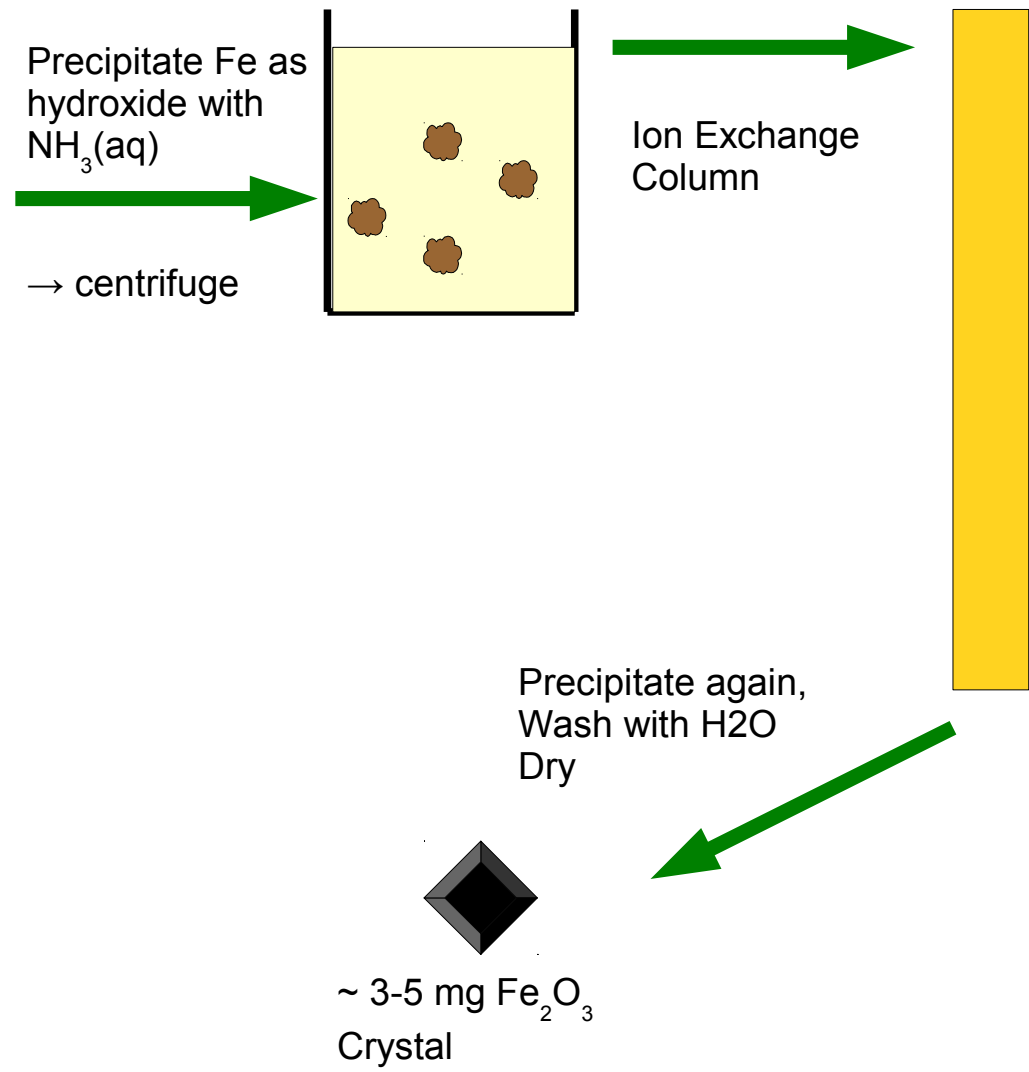
→ centrifuge



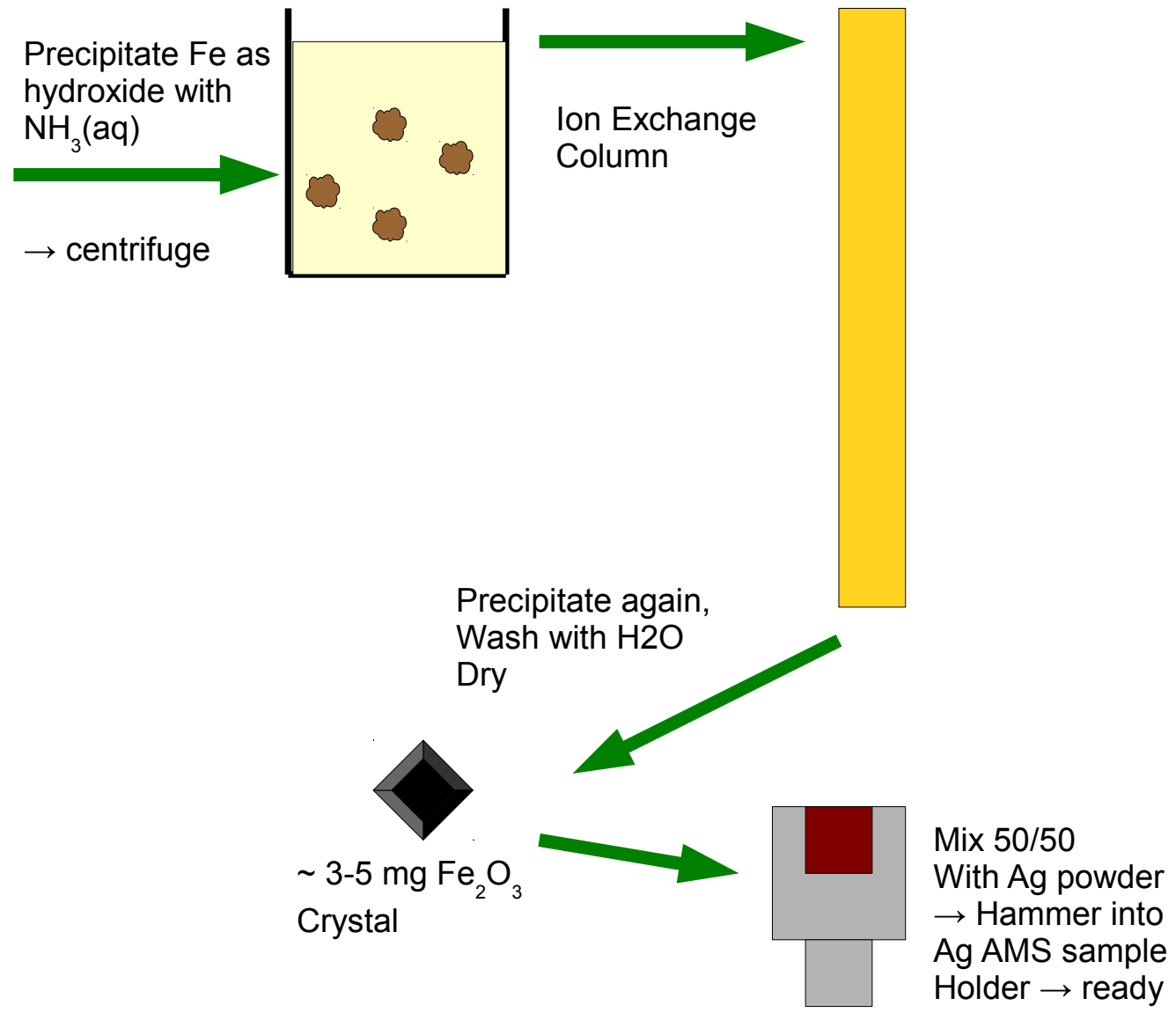
Chemical isolation of iron



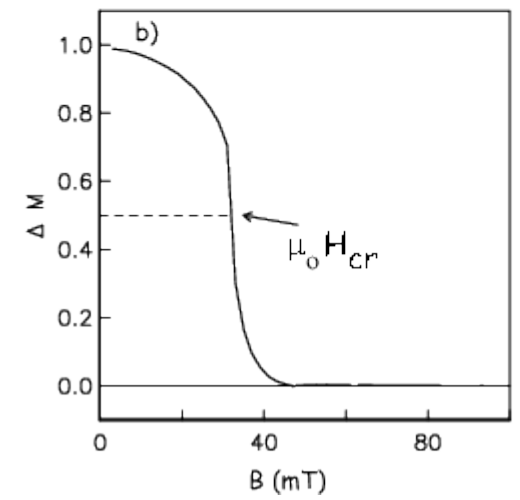
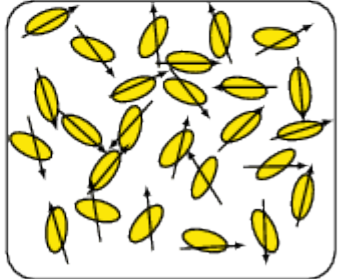
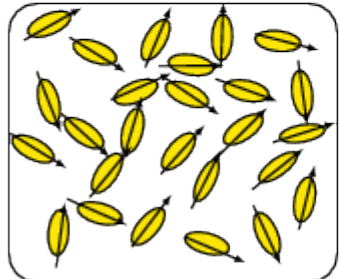
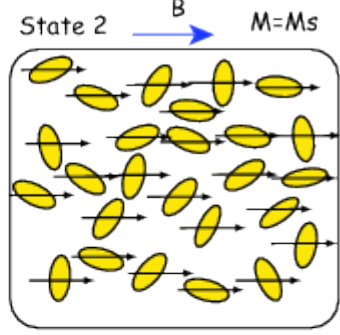
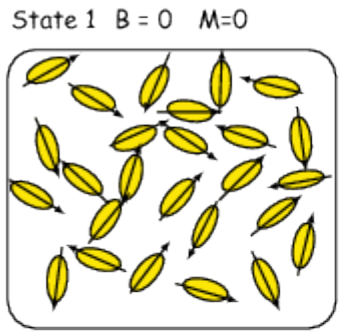
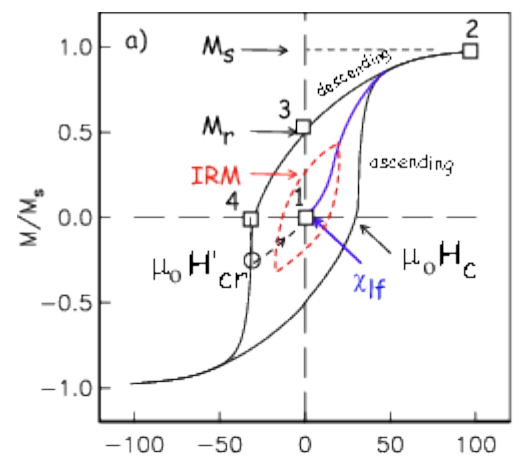
Chemical isolation of iron



Chemical isolation of iron



Hysteresis measurements



B (H) – External mag. Field [mT]

M – Magnetization

M_s – saturation magnetization

M_r (M_{rs}) – remanence magnetization

H_c – Coercivity

H_{cr} – Coercivity of remanence

χ_{lf} – low-field susceptibility

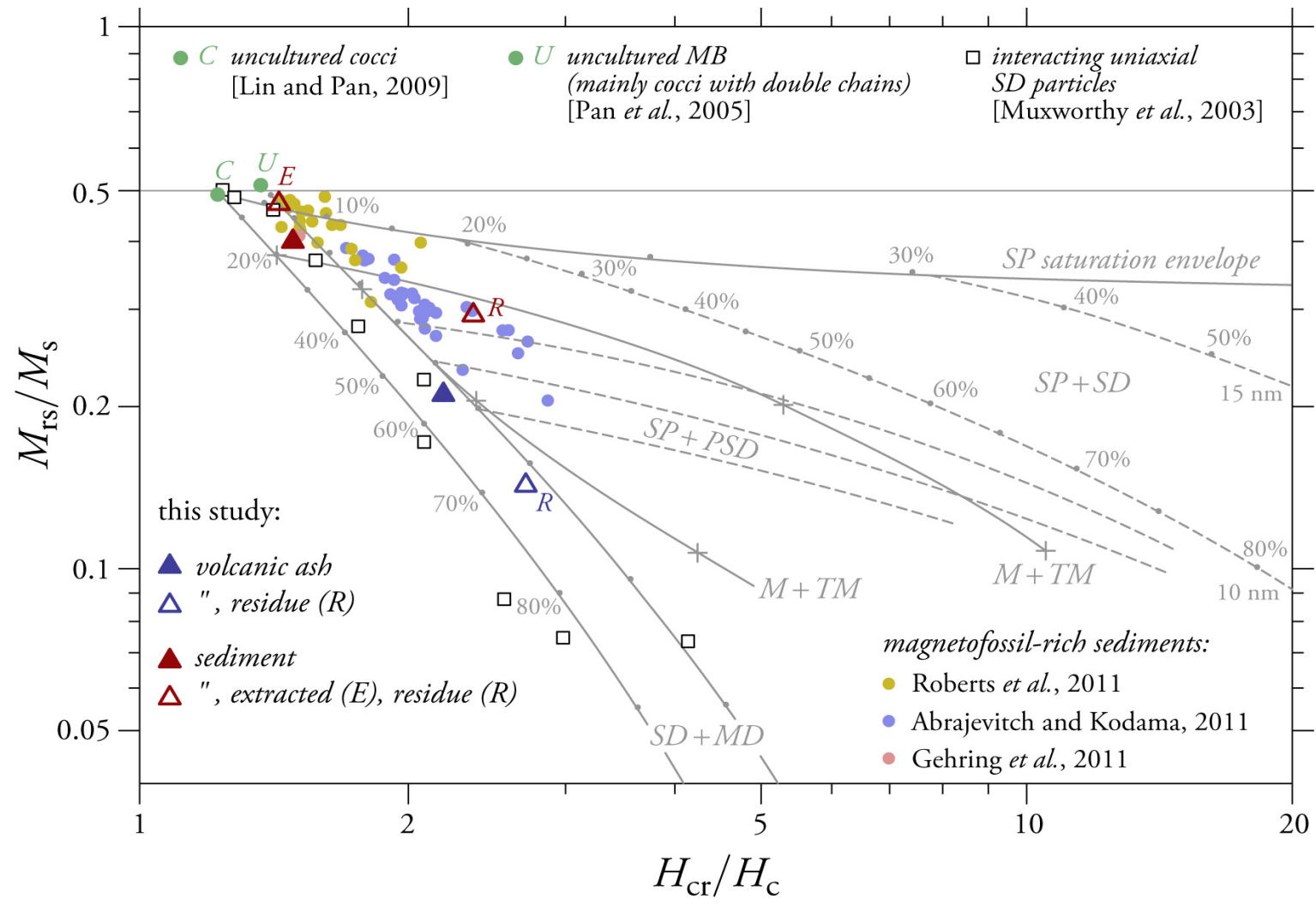
IRM – Isothermal remnant mag.

(a) Hysteresis loop of ferromagnetic material

(b) Difference between descending and ascending curve of (a), giving another measure for H_{cr}

Tauxe, L., Essentials of Paleomagnetism, University of California Press, 2010

Day plot



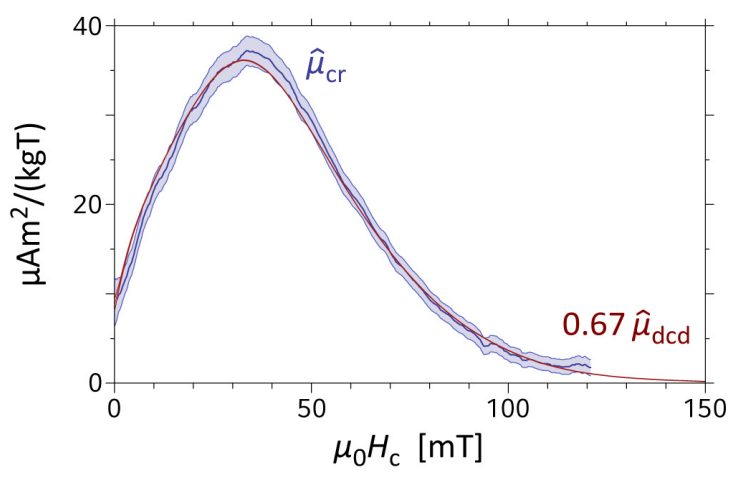
Day plot:

- ◆ Domain-state sensitive
- ◆ Generated from bulk mag. Properties
- ◆ Problem: Non-unique interpretations
- ◆ Hints to high abundance of magnetofossils in our sediment, BUT:
- ◆ For quantitative analysis, this is not enough!

Legend:

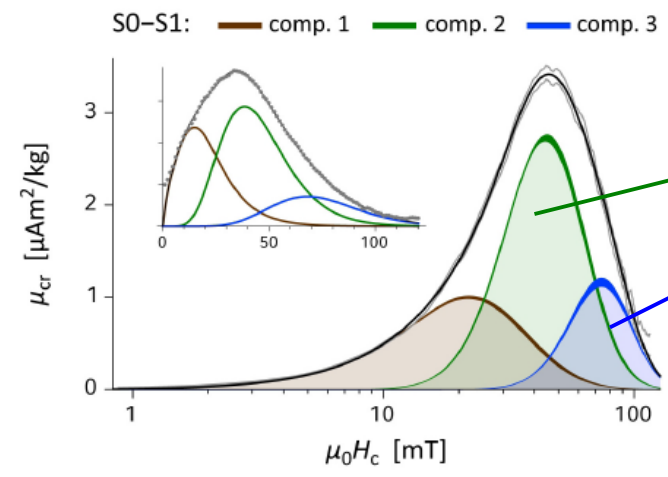
- ◆ SD – single domain
- ◆ MD – multi domain
- ◆ SP – super paramagnetic
- ◆ PSD – pseudo SD
- ◆ M – magnetite
- ◆ TM – Titanomagnetite

AMS sample coercivity distribution



Untreated sediment:

- ◆ ≈ 2% total Fe
- ◆ 60 ppm SD iron
- ◆ ≈ 3% Fe in mag. minerals



CBD extraction

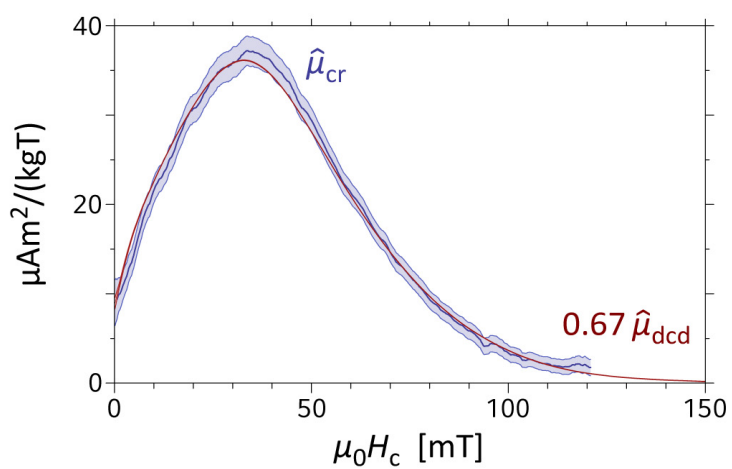
Sample for AMS:

- ◆ 60% from magnetic minerals
- ◆ 40% from non-mag. minerals
- ◆ < 6% from primary mag. minerals
- ◆ >27% from mag. Bacteria

Magnetofossils !!!

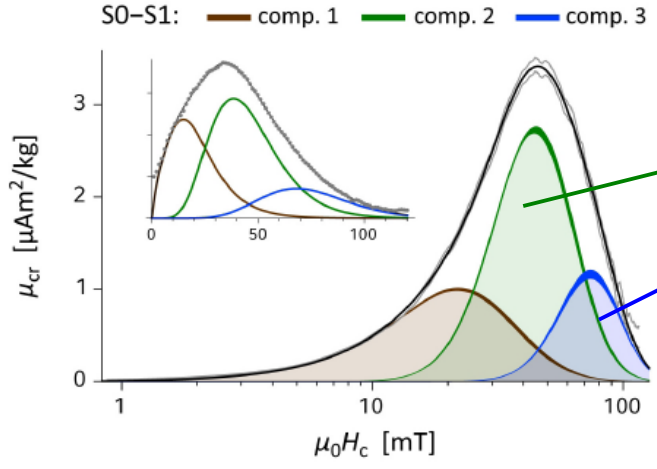


Summary magnetic characterization core 848



Untreated sediment:

- ◆ ≈ 2% total Fe
- ◆ 60 ppm SD iron
- ◆ ≈ 3% Fe in mag. minerals



CBD extraction

Sample for AMS:

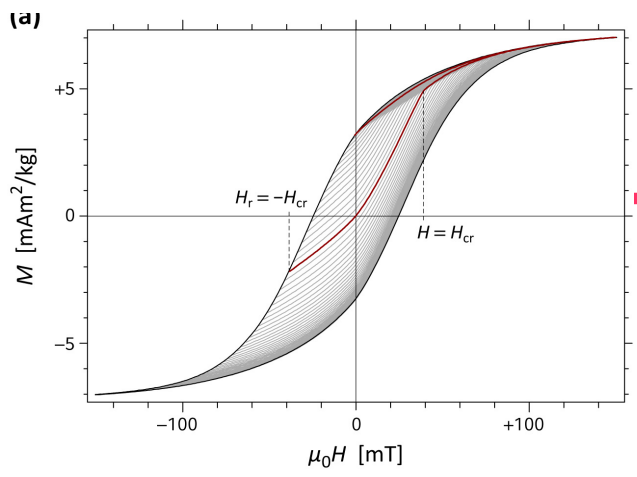
- ◆ 60% from magnetic minerals
- ◆ 40% from non-mag. minerals
- ◆ < 6% from primary mag. minerals
- ◆ >27% from mag. Bacteria

Magnetofossils !!!

Assuming ^{60}Fe only in secondary magnetic minerals (lower limit)
 $^{60}\text{Fe}/\text{Fe}$ is enhanced by a factor of ≈ 250 relative to complete dissolution of Fe.
 Dilution compared to original ^{60}Fe concentration in secondary minerals < 1.9
 → core 848 suited for CBD extraction, assuming roughly constant composition across the core

FORC measurements

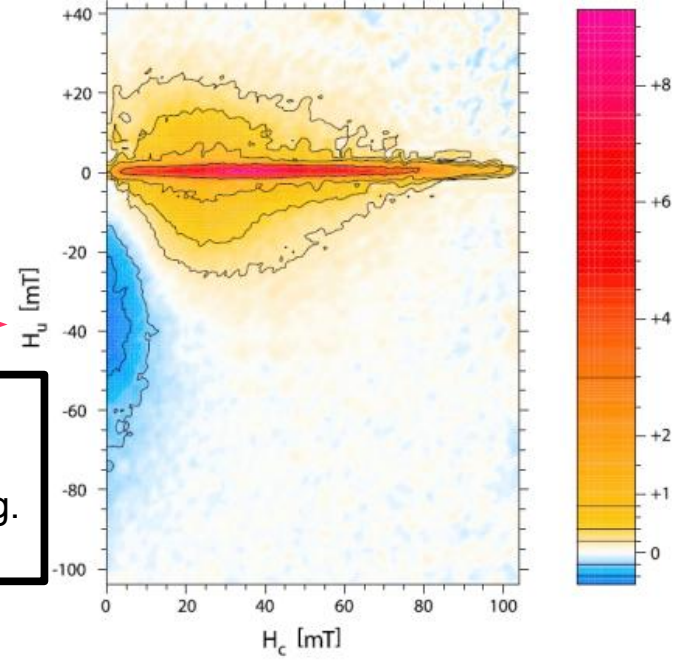
Use an Alternate Gradient Field Magnetometer (in Bremen) to record a set of 450 first-order reversal curves (FORCs), 4-5 times → total time ~1 day per sample



Mathematical transformation: $H-M \rightarrow H_u - H_c$
 where H_u : bias field (from other particles)
 H_c : coercivity (mag. hardness)

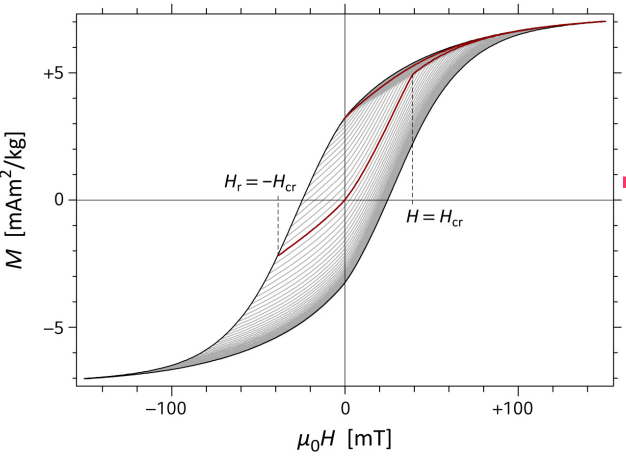
Feature of FORC diagram of SD particles:
 Central ridge: Delta function along $H_u = 0$
 to identify non-interacting SD particles, e.g. magnetofossils

Lake sample containing magnetofossils $\rho(H_c, H_u)$ [$10^{-4} \text{ Am}^2/\text{T}^2$]



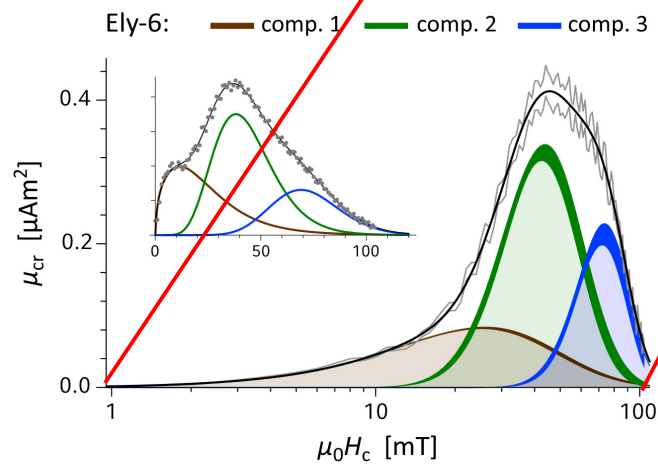
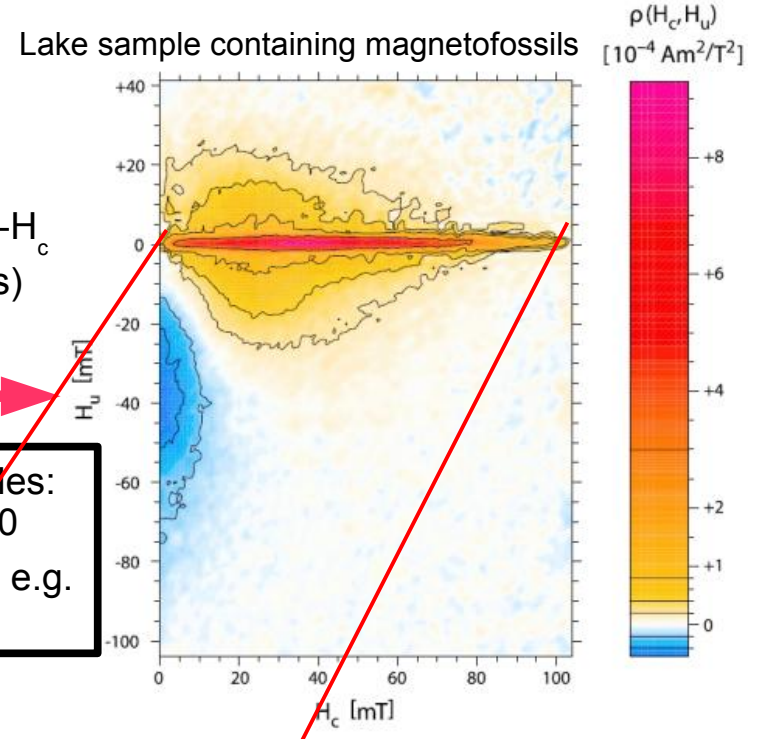
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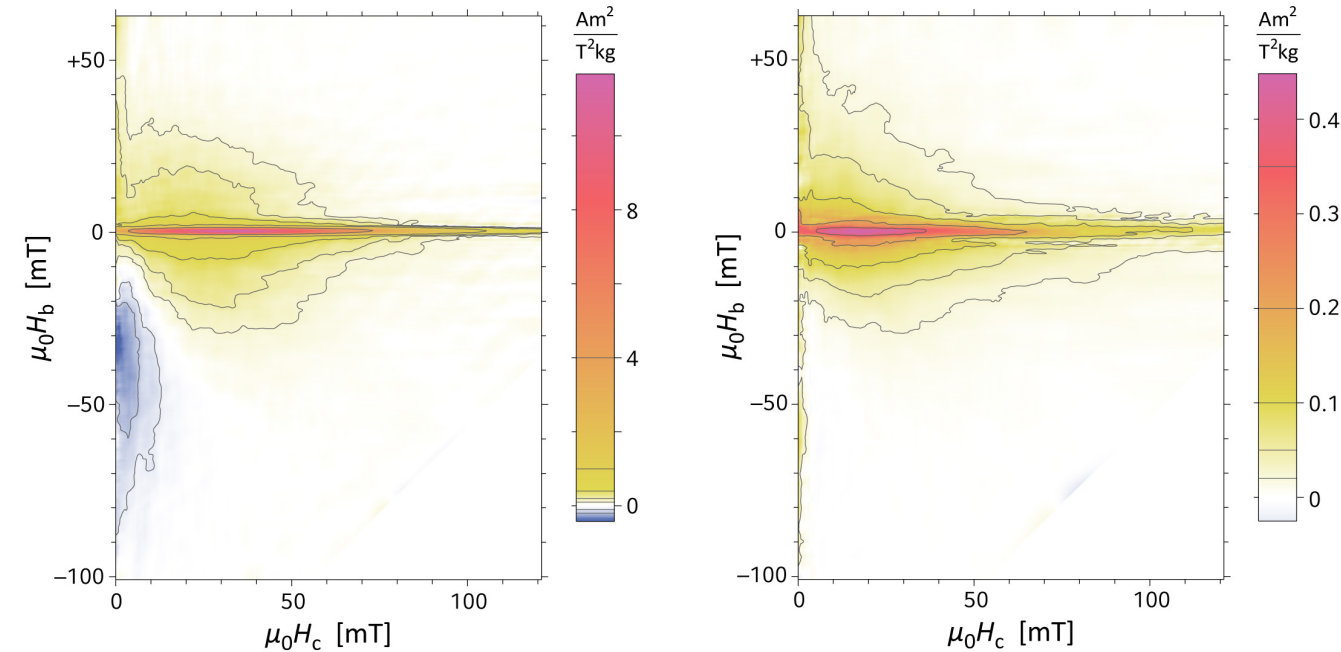
Integrate and fit coercivity distribution:
 For the lake sample – 3 components
 (1) Extracellular (low coercivity)
 (2) Biogenic soft
 (3) Biogenic hard

FORC analysis of sediment

Sediment 848 – 3.2-3.3 Myr
untreated

Sediment 848 – 3.2-3.3 Myr
after CBD extraction

AMS sample



?

Central ridge almost disappears (~5% left) after 1 CBD extraction

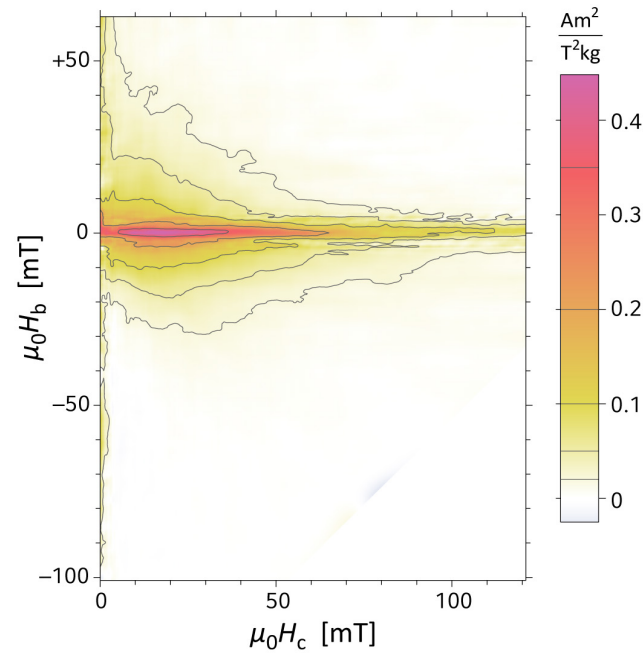
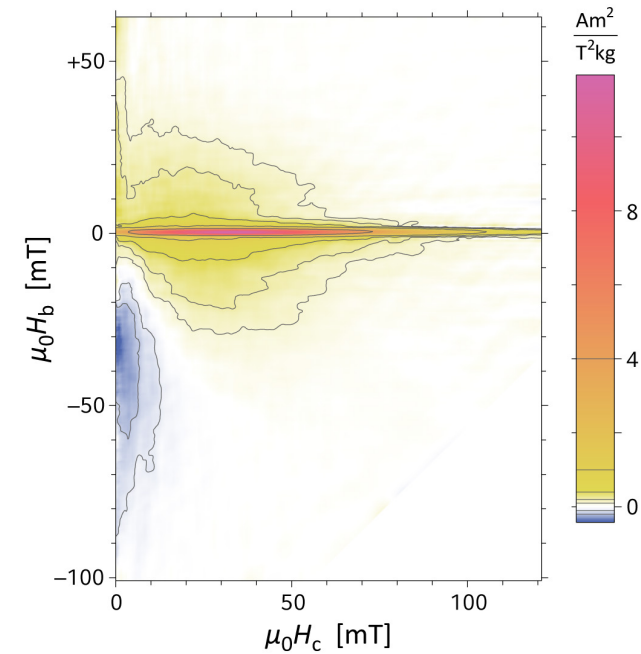
BUT: What's the magnetic signature of the AMS sample?

FORC analysis of sediment

Sediment 848 – 3.2-3.3 Myr
untreated

Sediment 848 – 3.2-3.3 Myr
after CBD extraction

AMS sample



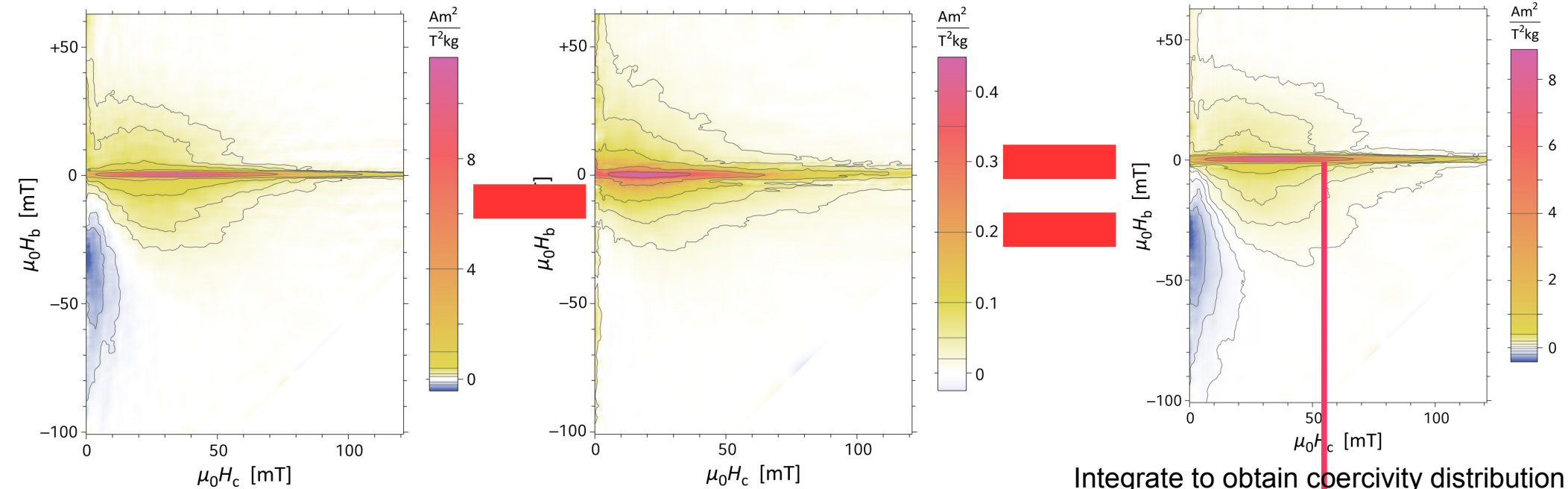
Central ridge almost disappears (~5% left) after 1 CBD extraction

BUT: What's the magnetic signature of the AMS sample?

Sediment 848 – 3.2-3.3 Myr
untreated

Sediment 848 – 3.2-3.3 Myr
After CBD extraction

AMS sample
calculated as difference



Integrate to obtain coercivity distribution

