Discovery of Supernova ^{60}Fe in the Earth’s Microfossil Record

A Cosmic Message in a Bottle

The ^{60}Fe Team

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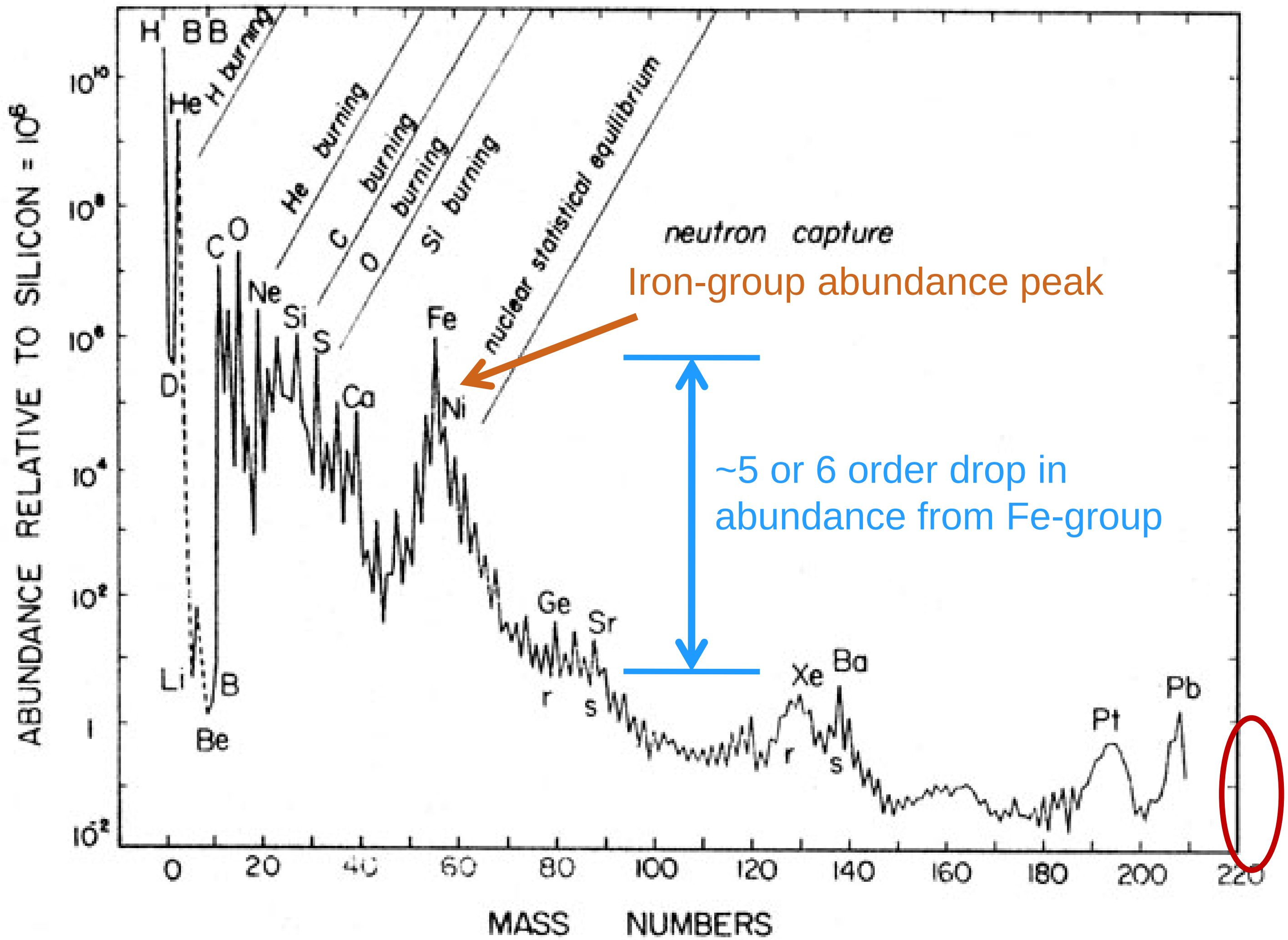
Outline

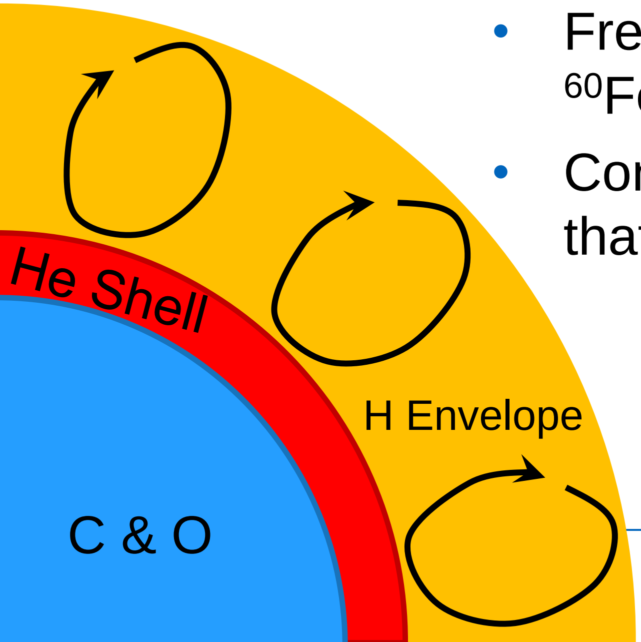
- Astrophysics Introduction
  - The **real** motivation (where is the r-process happening?)
  - A Cosmic Site of ^{60}Fe Production
- Accelerator Mass Spectrometry
- Terrestrial ^{60}Fe Reservoirs
  - Ferromanganese Crust (discovered)
  - ODP Sediment Core: Magnetofossils
  - $^{60}Fe/Fe$ Results from M-fossils
- Conclusion & Future Ideas (time permitting)

Where in Nature are Elements Beyond Fe-Peak Made?



Cosmic Sites of ^{60}Fe Production

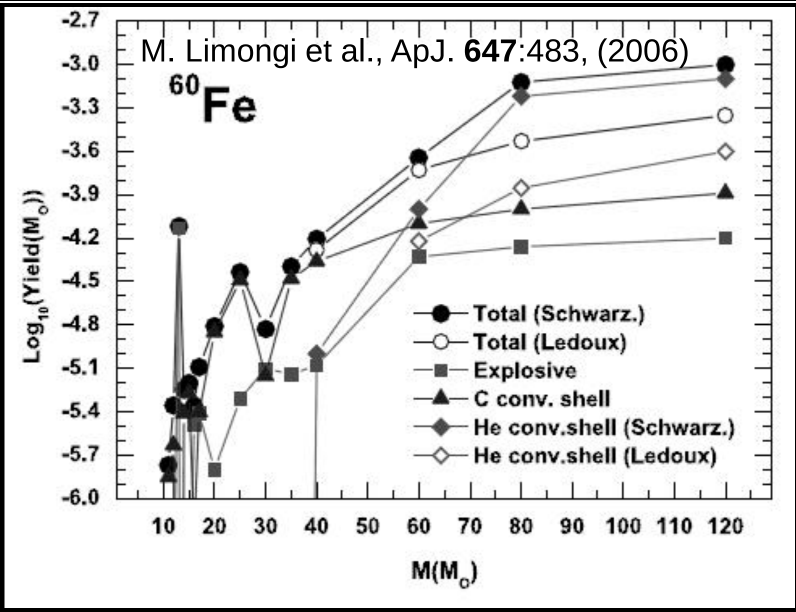
- Stars with masses > 10 solar masses
- Conclusion of core He-burning → He-burning shell → T ~ 4 x 10^8
- Temp. drives the reaction sequence: $^{14}\mathrm{N}(\alpha,\gamma)^{18}\mathrm{F}(\beta^+\nu)^{18}\mathrm{O}(\alpha,\gamma)^{22}\mathrm{Ne}$
- Followed by: $^{22}\mathrm{Ne}(\alpha,n)^{25}\mathrm{Mg}$
- Free neutrons drive an s-process in the shell → ^{60}Fe production

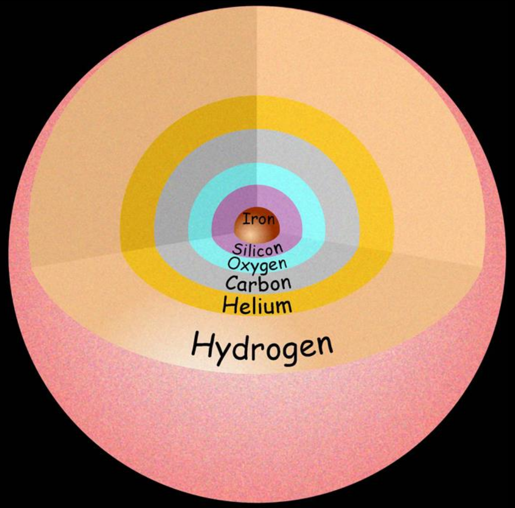


- $^{12}C(^{12}C,\alpha)^{20}Ne$ occurs in core
- Free α's undergo reaction sequence above → ^{60}Fe production
- Convection carries ^{60}Fe to lower temperatures, so that some survives against further n-capture

Core Collapse Supernova

- Shock wave from core bounce slams into carbon and He shells
- Shells are shock heated
  - Heating drives the $^{22}\mathrm{Ne}(\alpha, n)^{25}\mathrm{Mg}$ reaction to faster rate
  - Shells are also expanding (explosively)
  - Neutron capture process as before occurs, but much faster
  - Neutron capture rates are comparable, or faster, than expansion rate of shell
- ^{60}Fe synthesized in these shells ejected into space (5000 km/s)

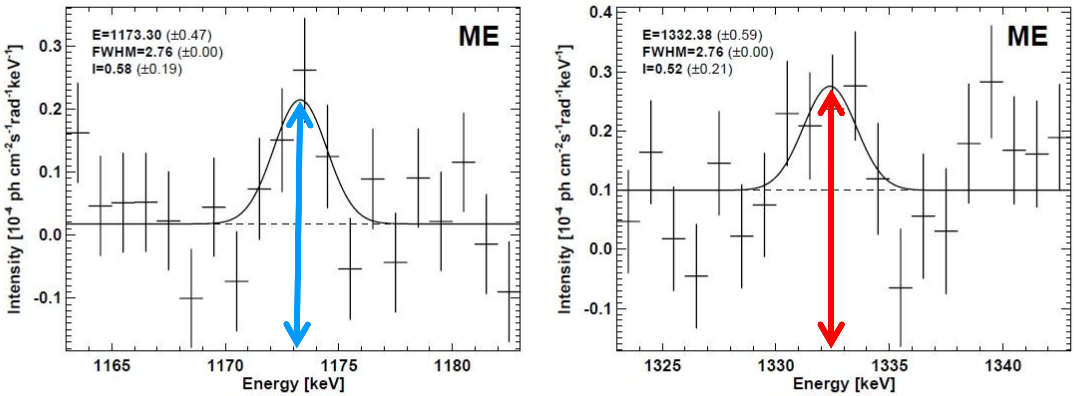




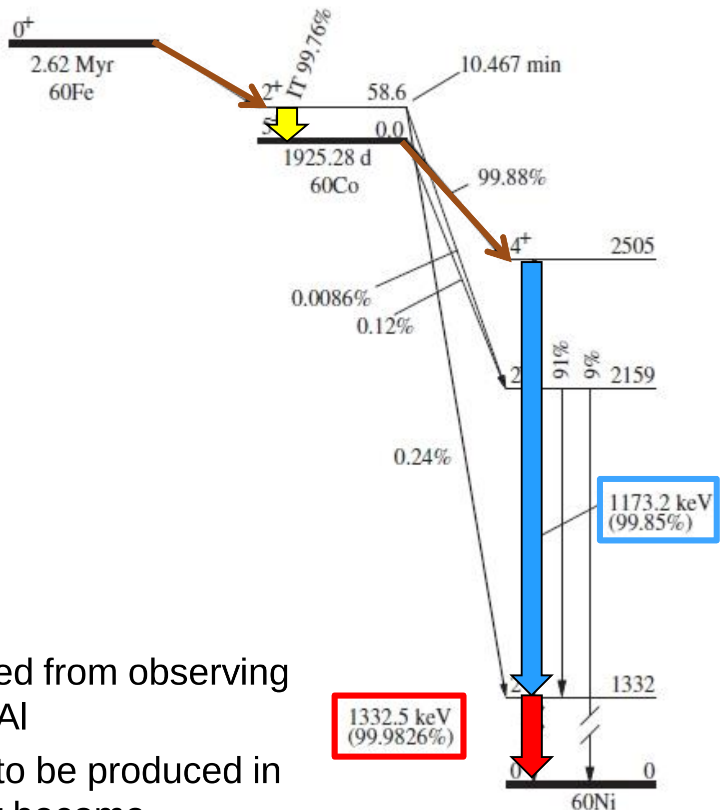
^{60}Fe Astrophysics Points

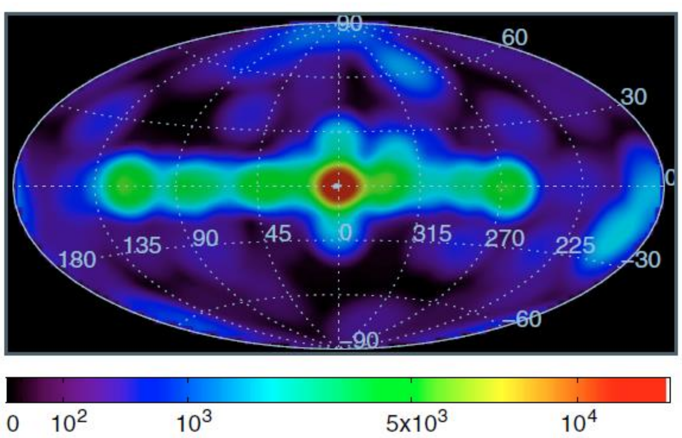
G. Rugel et al., PRL **103** (2009)

- Half-life = 2.62 Myr
- Gamma-rays observed with INTEGRAL satellite of European Space Agency

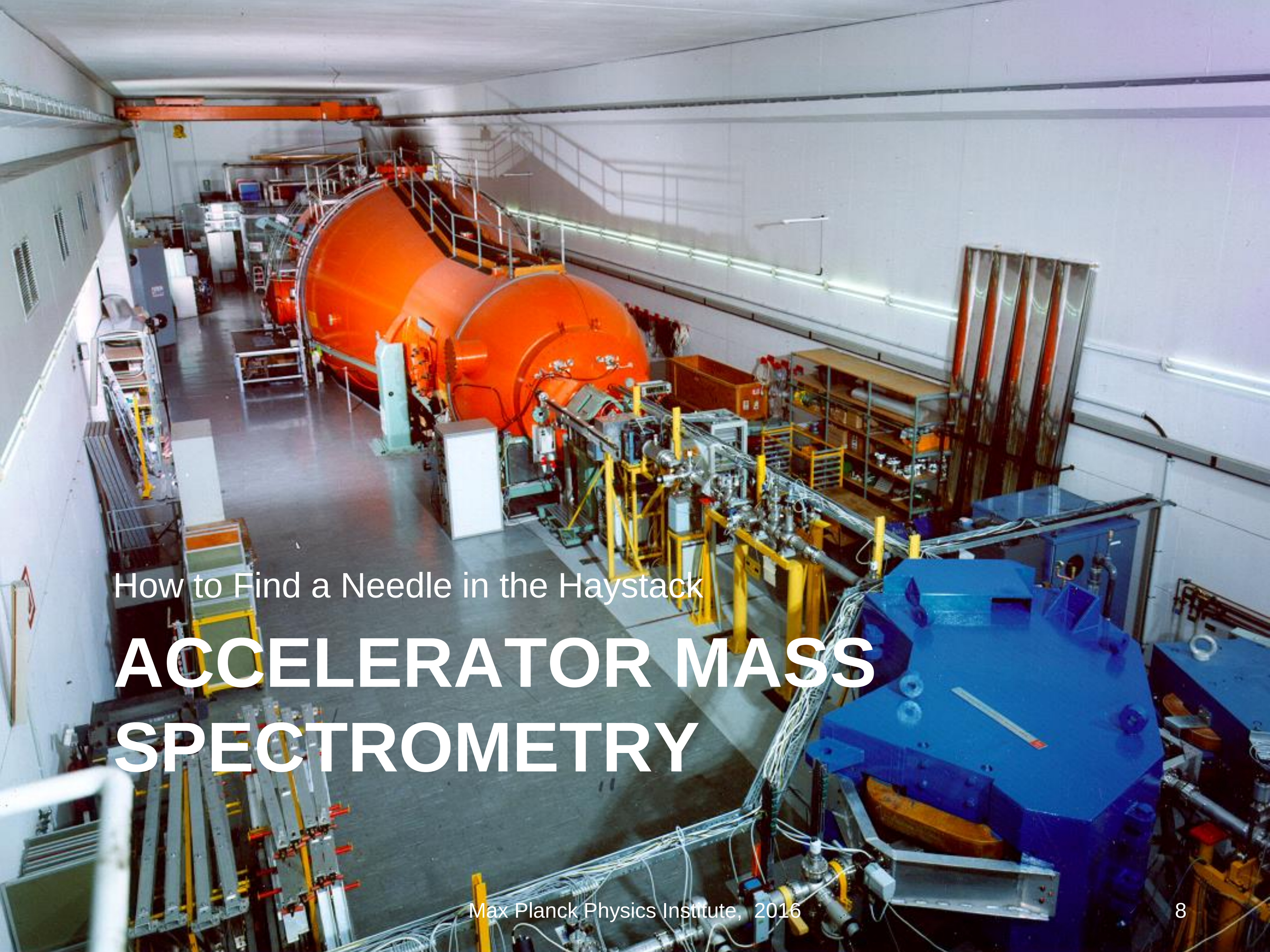


Wang et al., Astron. & Astrophys. **469** (2007)





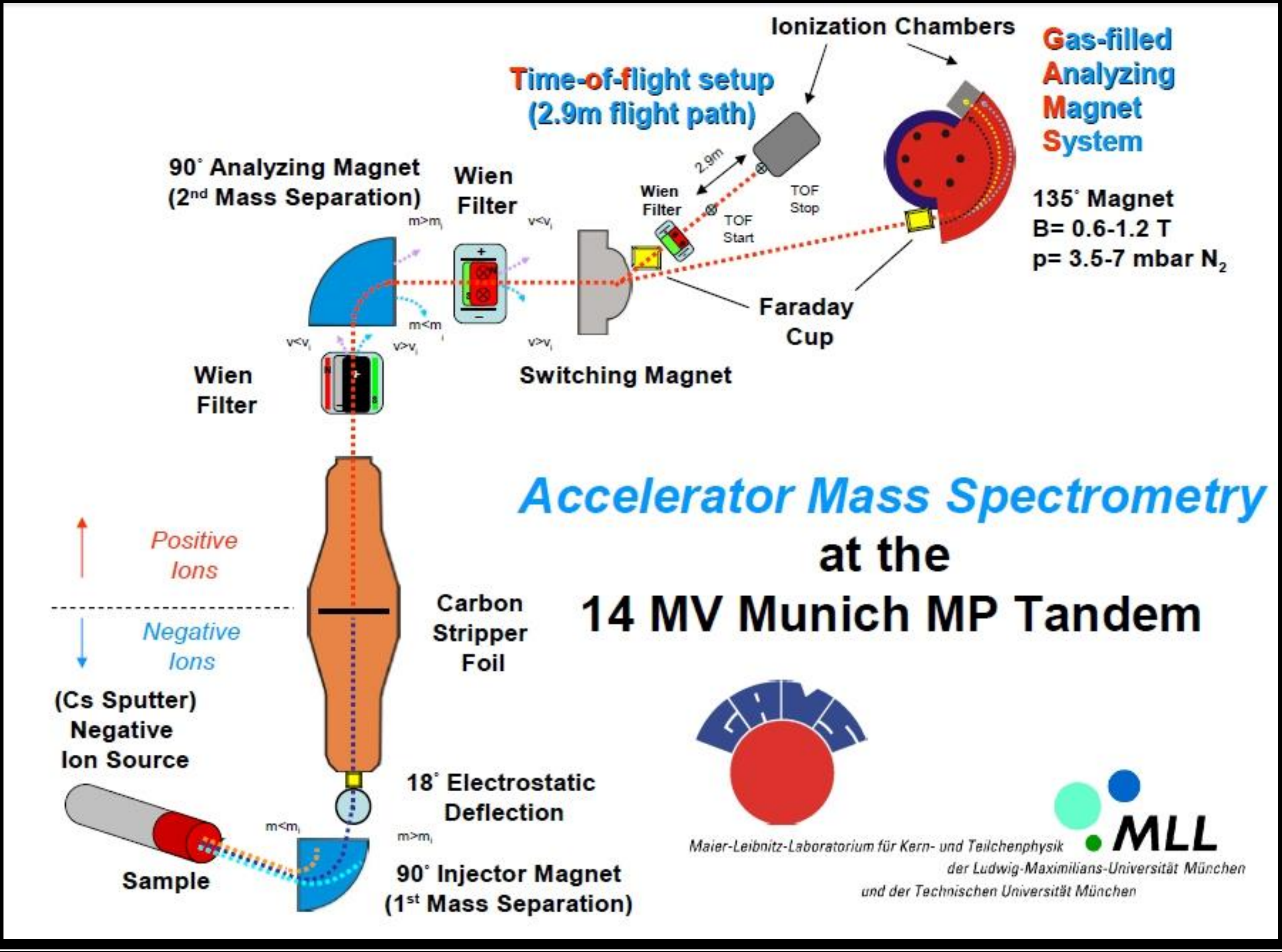
- ^{60}Fe data obtained from observing same region as ^{26}Al
- ^{26}Al also known to be produced in massive stars that become supernovae
- Finding ^{60}Fe in same places as ^{26}Al => observational confirmation ^{60}Fe comes from massive stars and SN



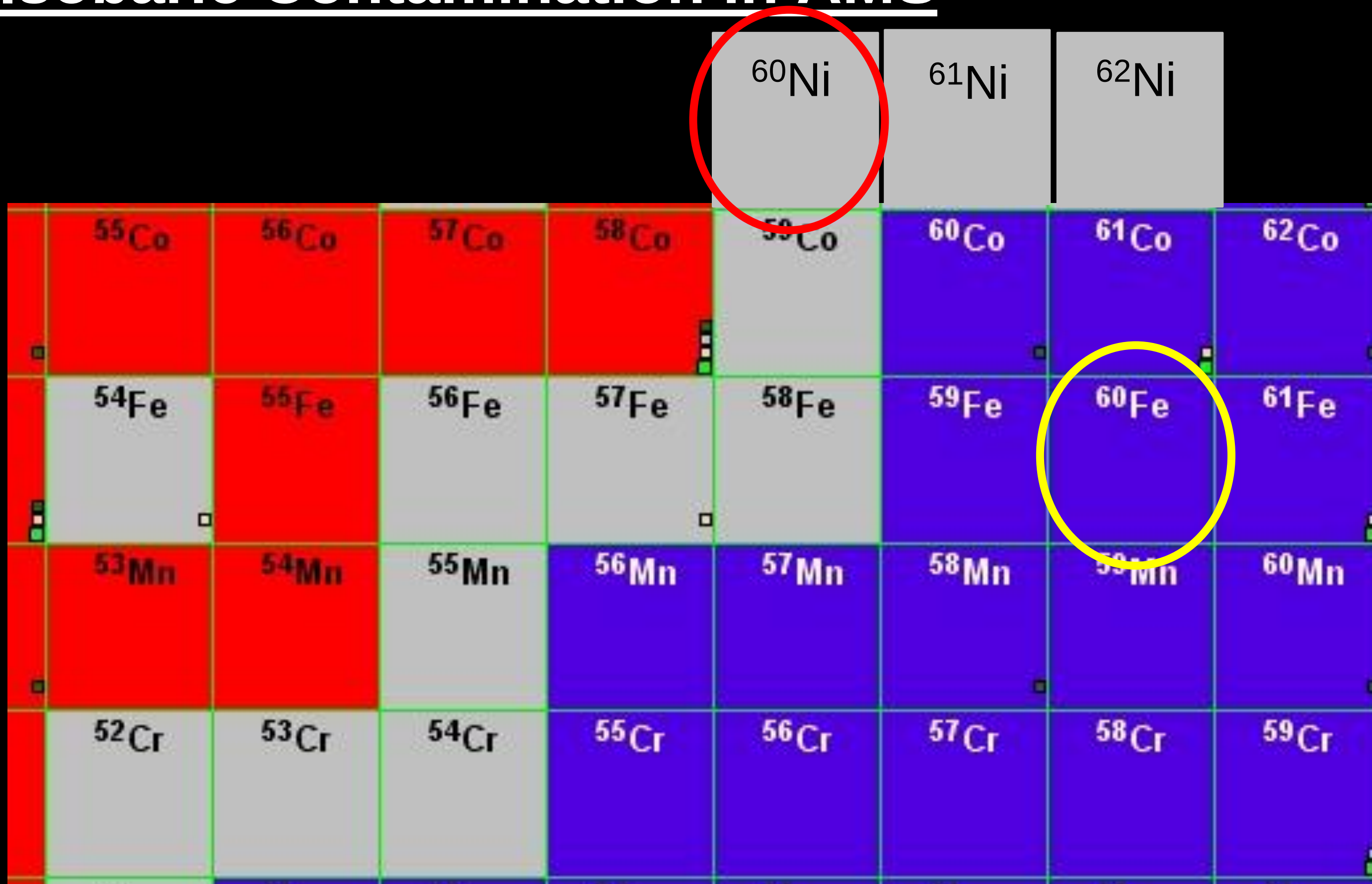
How to Find a Needle in the Haystack

ACCELERATOR MASS SPECTROMETRY

AMS Facility: Schematic



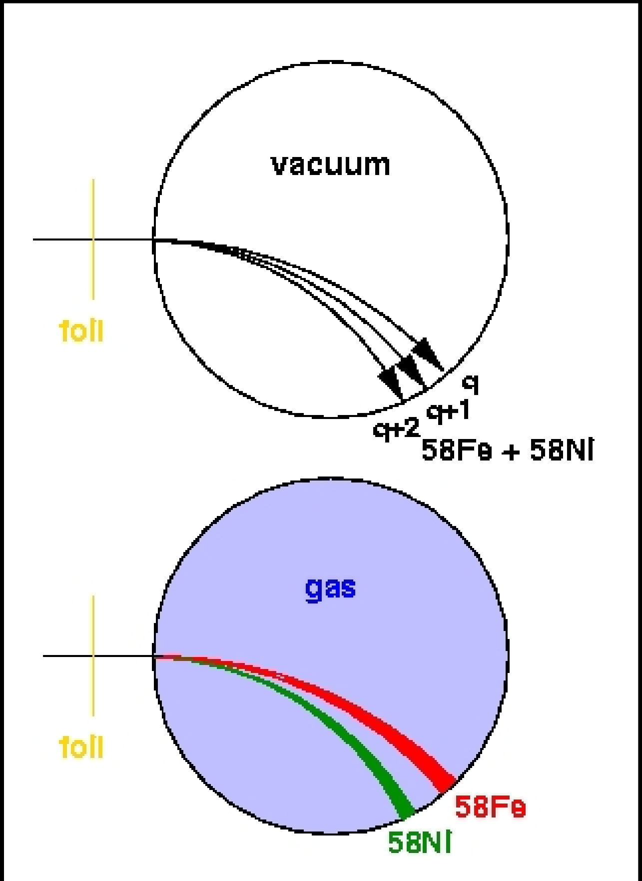
^{60}Ni Isobaric Contamination in AMS



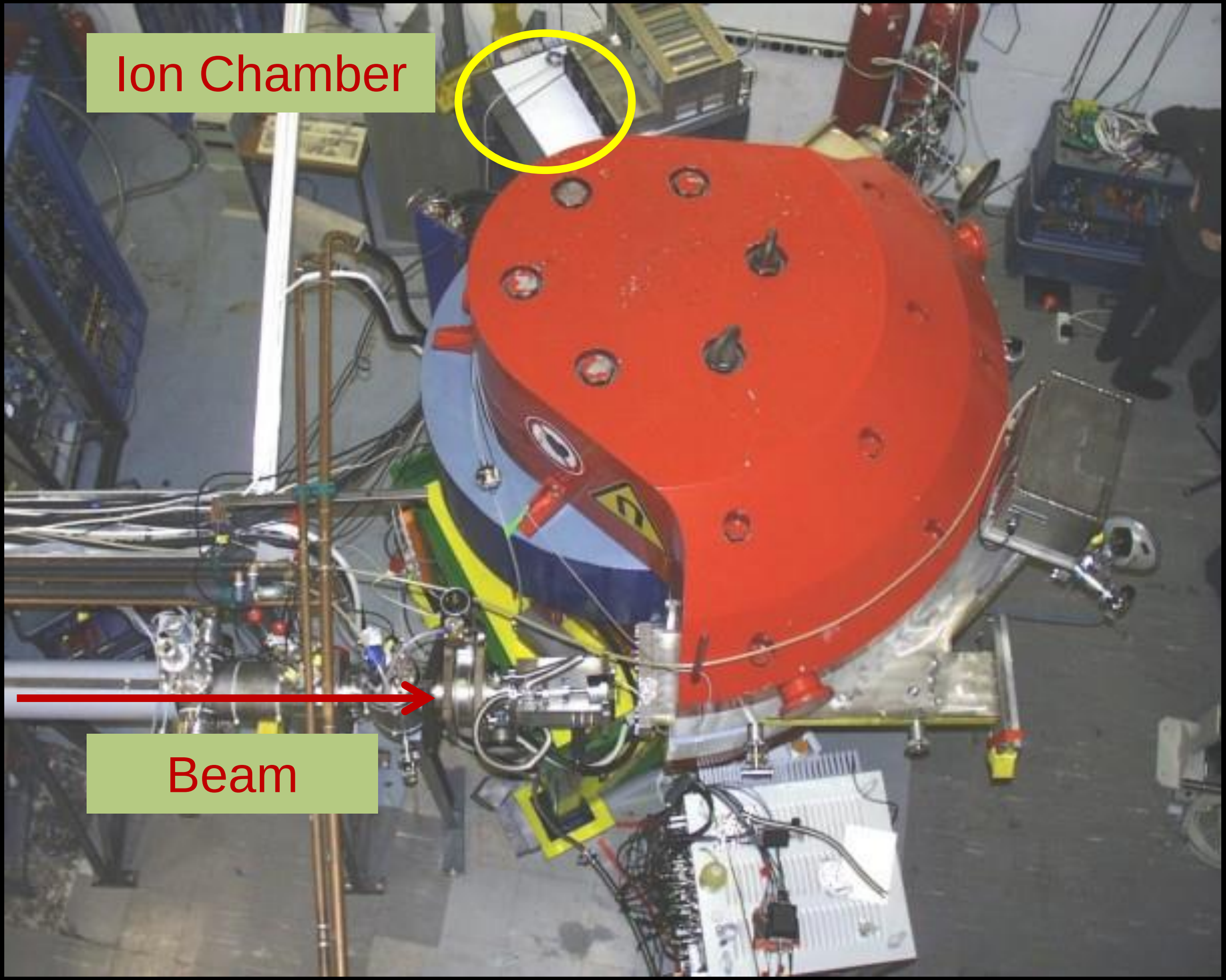
The "Trick" for Background Suppression

- Magnet selects charge:mass ratio
  - $$B\rho = \frac{Mv}{q}$$
- For same ion energy from Tandem:
  - Kinetic E is same:
    $$\Rightarrow \frac{v_1}{v_2} = \sqrt{\frac{M_2}{M_1}}$$
  - If masses are same, cannot separate
- Magnet filled with few mbar N_2 gas
  - Then, q depends on *atomic number*:

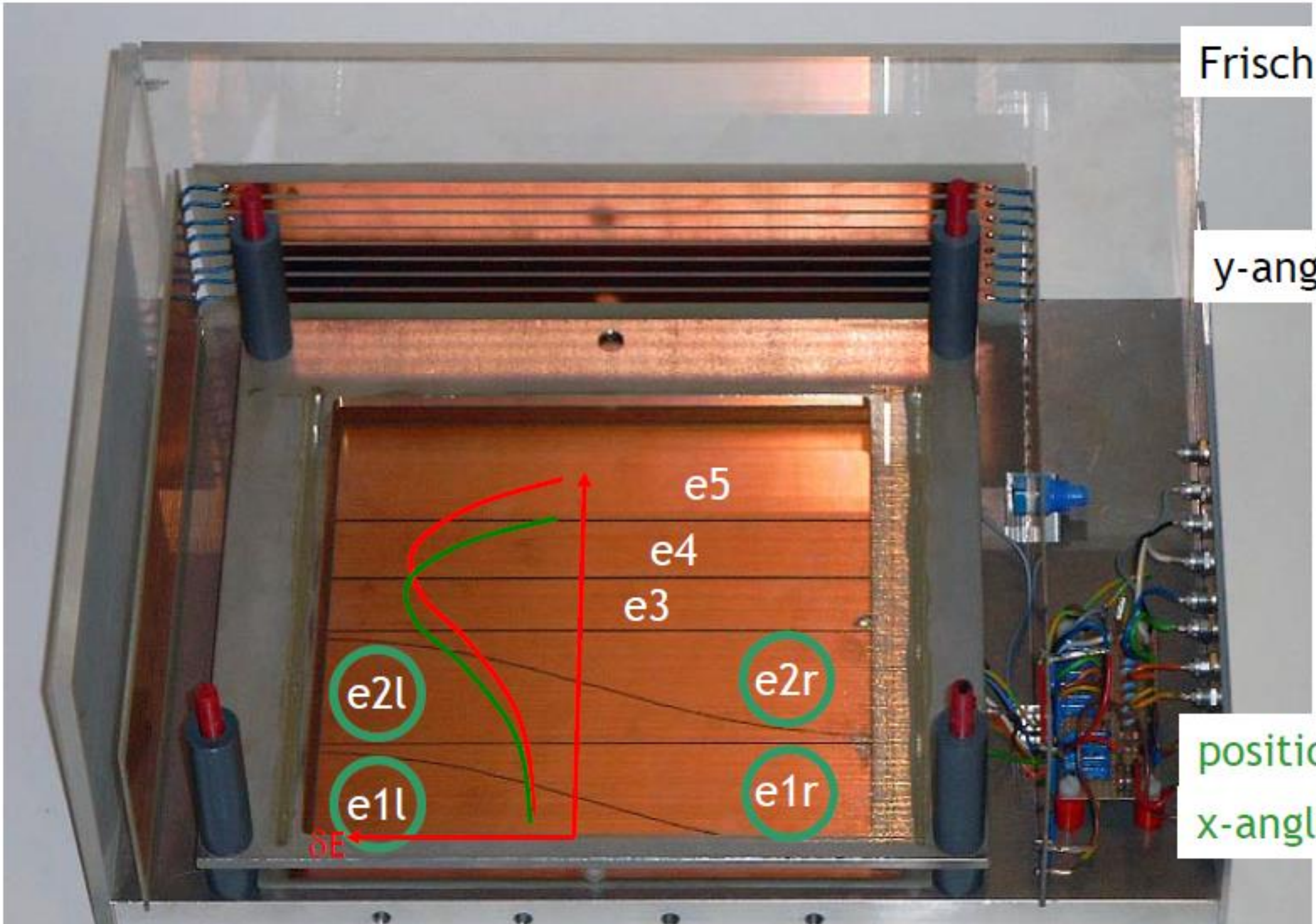
$$\langle q \rangle \propto vZ^{0.4} \quad \Rightarrow B\rho \propto \frac{M}{Z^{0.4}}$$



The “Business” End of the AMS Facility



Segmented Anode Ionization Chamber



Frisch grid: et

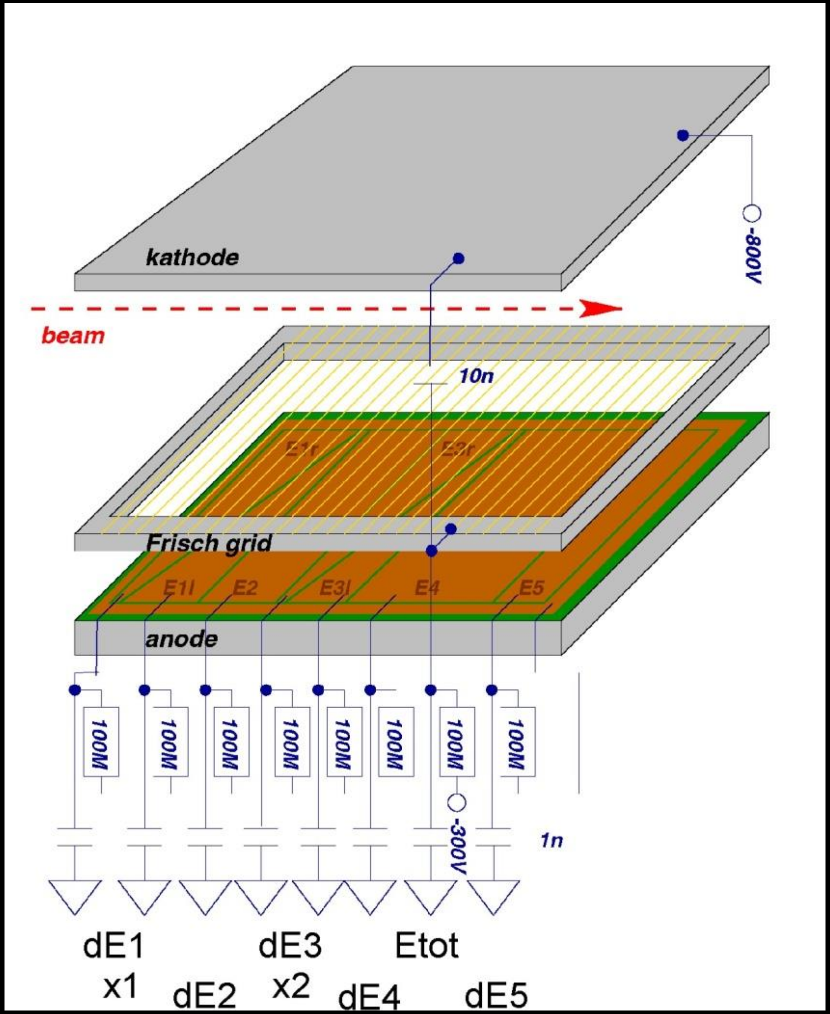
y-angle: dt

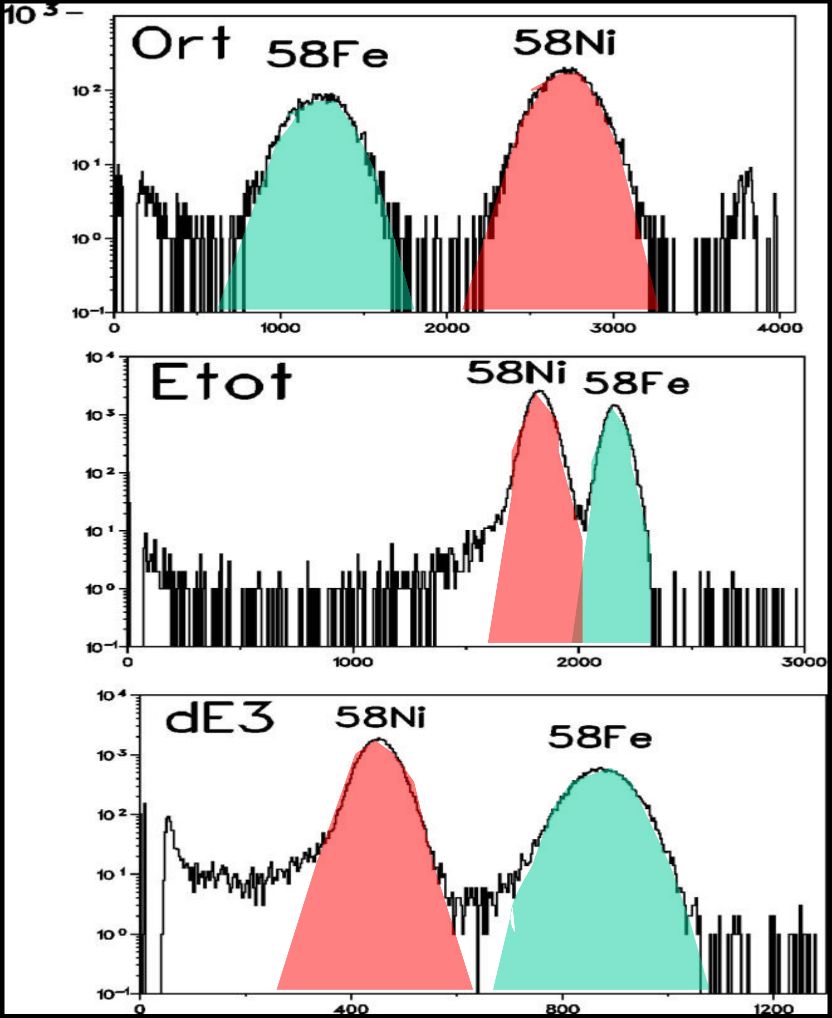
position: p

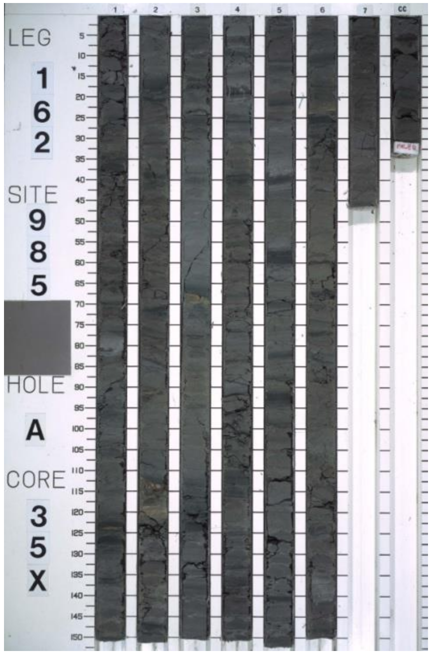
x-angle: dp

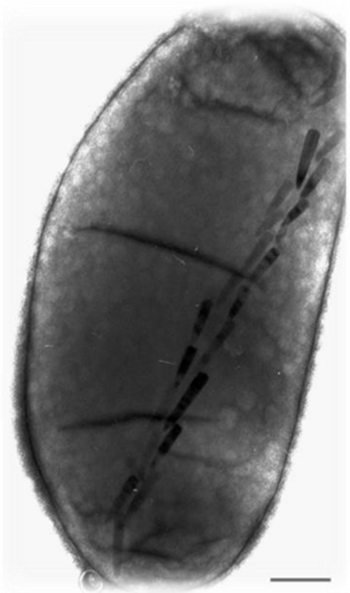
9 „independent“ signals

Final Particle Identification





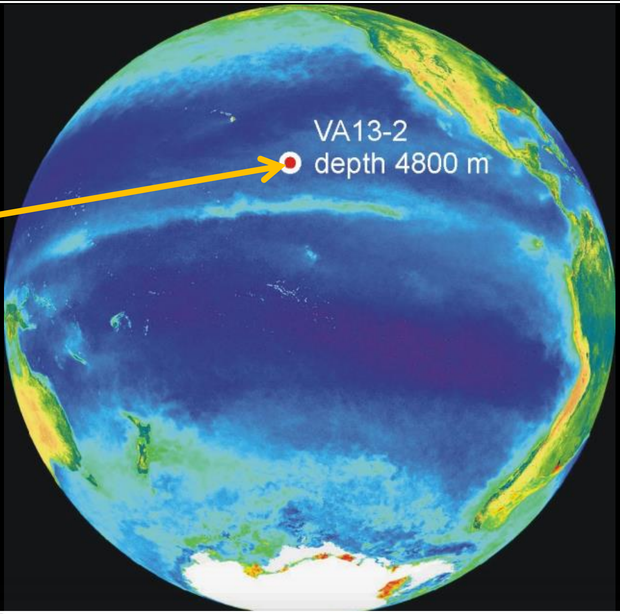


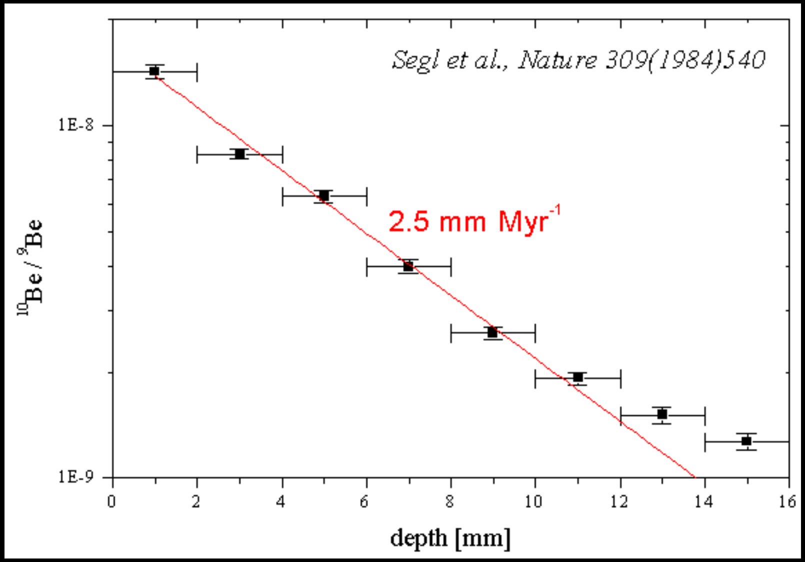


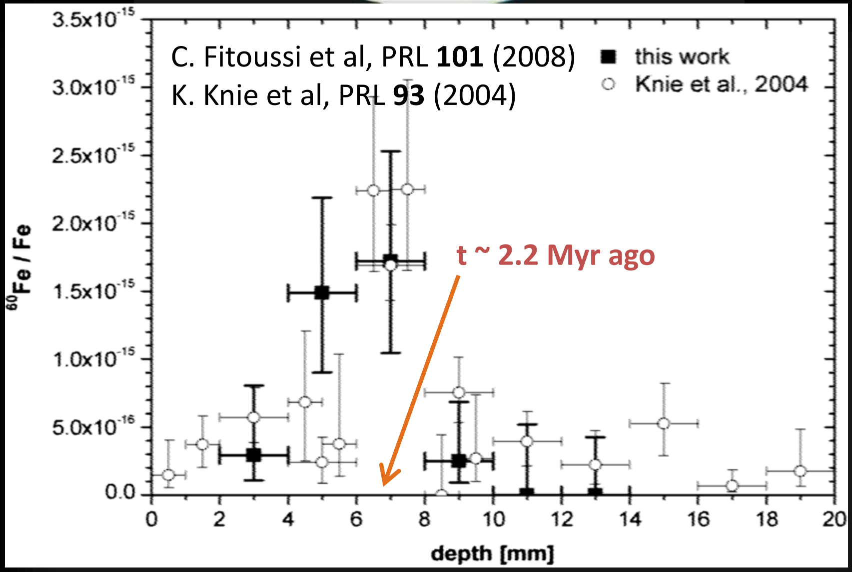
Where can we look?

TERRESTRIAL ^{60}FE RESERVOIRS

The FeMn Crust ^{60}Fe Results

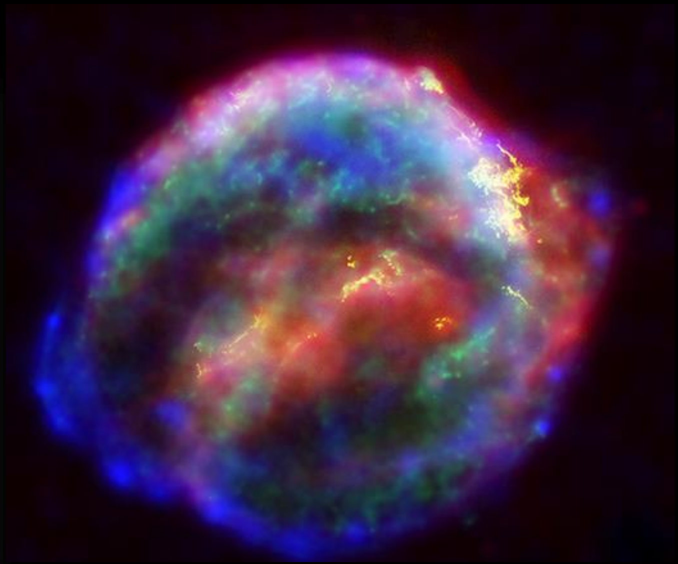


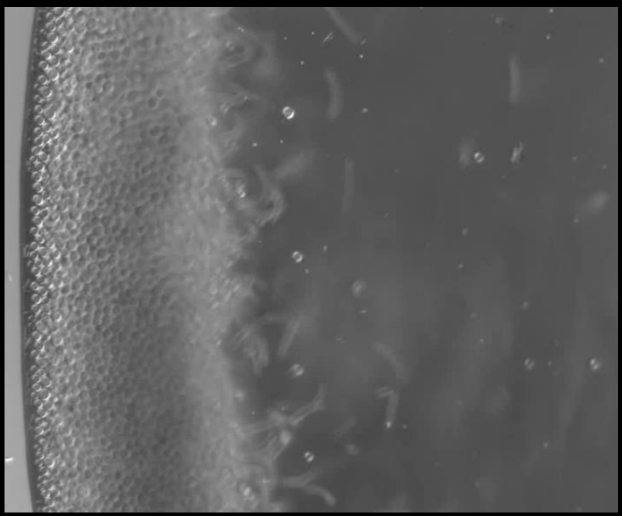




Results from Ferro-Manganese Crust Findings

- Terrestrial ^{60}Fe fluence determined as $\phi_{60} = 2.8 \times 10^8$ atom/cm^2 (after new ^{60}Fe $t_{1/2}$ correction)
- Fluence: $\phi_{60} \propto 1/U_{Fe}$
- Uptake used was 0.6%, but:
  - Could be as large as unity (arguments for this not published)
- If unity, Knie et al. fluence reduced by ~165 times
  - Distance estimate wrong by ~15
- Second attempt made in Norwegian Sea sediment: no success
  - Why? Please ask. ☺
- Motivated to find a different/new ^{60}Fe reservoir for cross checking and constraining uncertainties
- What is the new reservoir?





There are more things in heaven and earth, Horatio, than are dreamt of in your philosophy. -- Hamlet

SUPERNOVA SIGNATURE OF BIOGENIC ORIGIN

The Hypothesis

- Supernova ^{60}Fe flux arrives in upper atmosphere
  - Molecular, fine grains/dust

- Mixes into atmosphere enters Earth's Fe-cycle
- SN ^{60}Fe (and stable Fe) oxidized
- Forms nano-size oxide grains

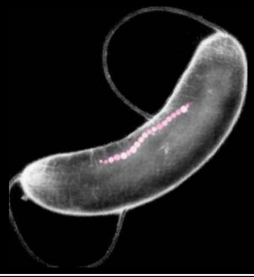
- Nano-oxides reach ocean
- Rapidly dissolves and re-precipitates
- Forms poorly crystalline ferric hydroxides (“rust”)
- Settles into sediment

Ocean

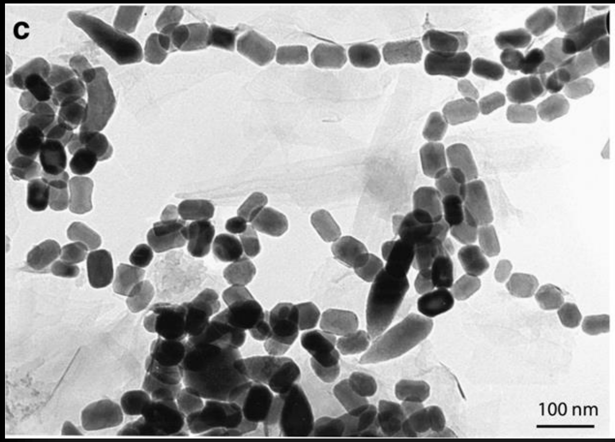
Sediment

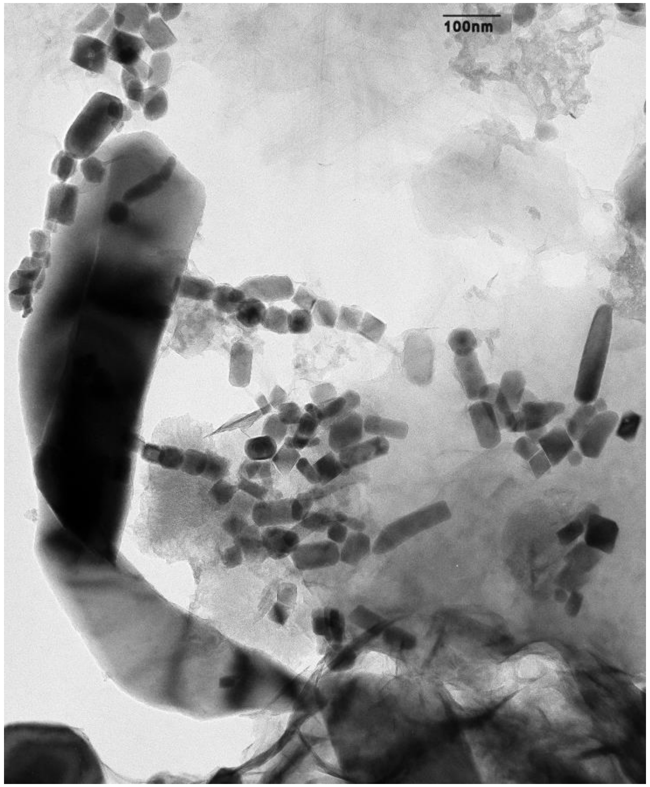
Magnetotactic Bacteria: The Essentials

- Form ~90 nm sized magnetic crystals of **magnetite: Fe_3O_4**
- Live in sediment just below the surface-water interface
- Process ferric hydroxide nano-grains into magnetosomes
- Bulk Fe phases from detrital sources not the primary Fe source → magnetosomes effectively sequester ^{60}Fe
- Bacteria co-move with sediment-water interface as it grows
- Magnetosomes become “magnetofossils”
- Any ^{60}Fe is “locked” inside



150 Myr BP

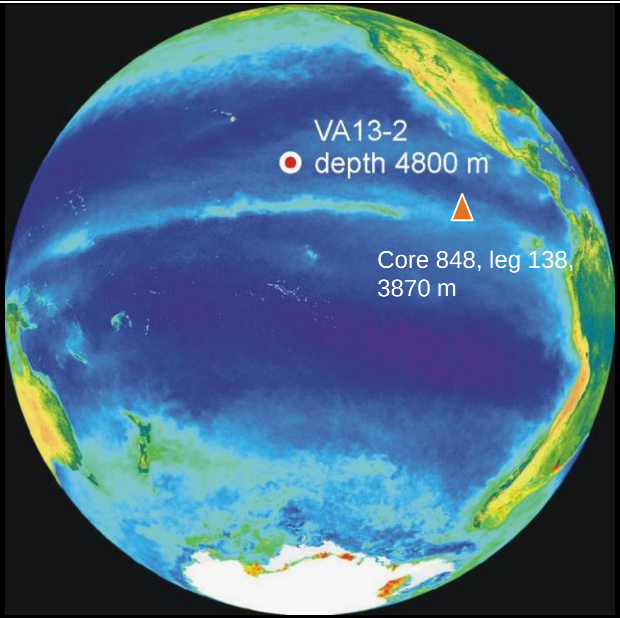


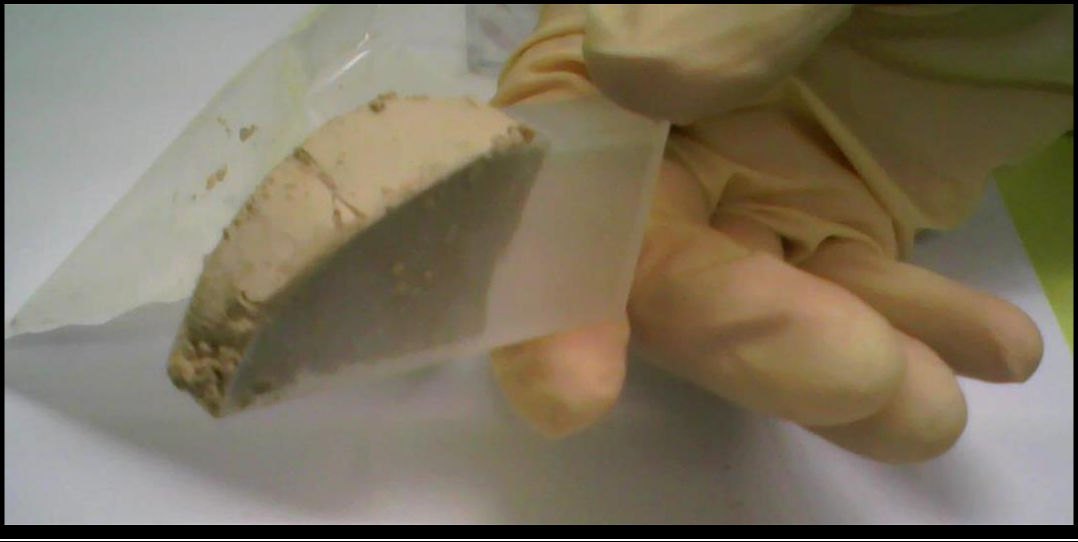


ARE WE REALLY EXTRACTING THESE FROM THE “DIRT”?

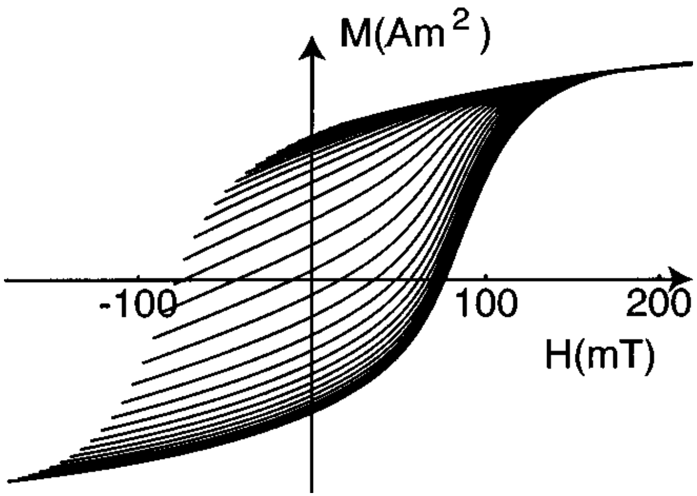
Ocean Drill Core Samples

- Core 848, leg 138, equatorial Pacific
- Water depth: 3870 m
- Predominantly Calcium-carbonate (80%) and SiO_2 (20%)
- Column height obtained: 0 – 3.3 Myr BP
- Location: reduces detrital Fe inputs from continental run-off

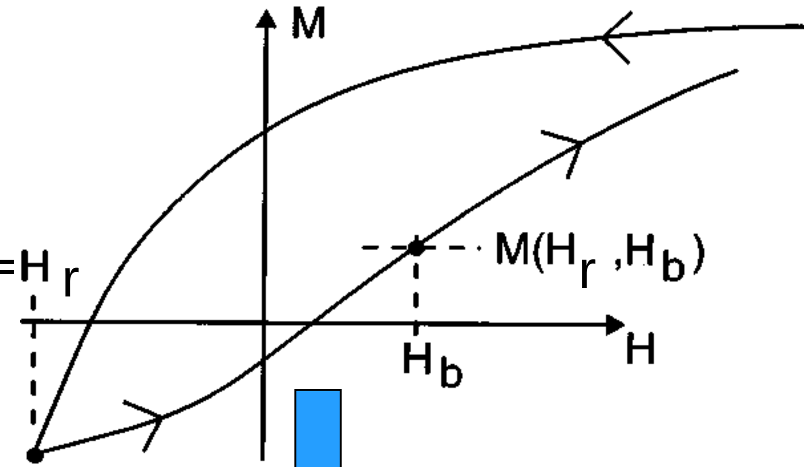




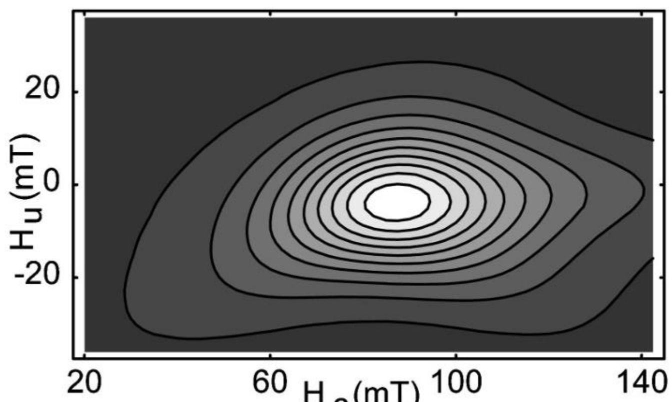
First Order Reversal Curves (FORC's)



Reversal field = H_r



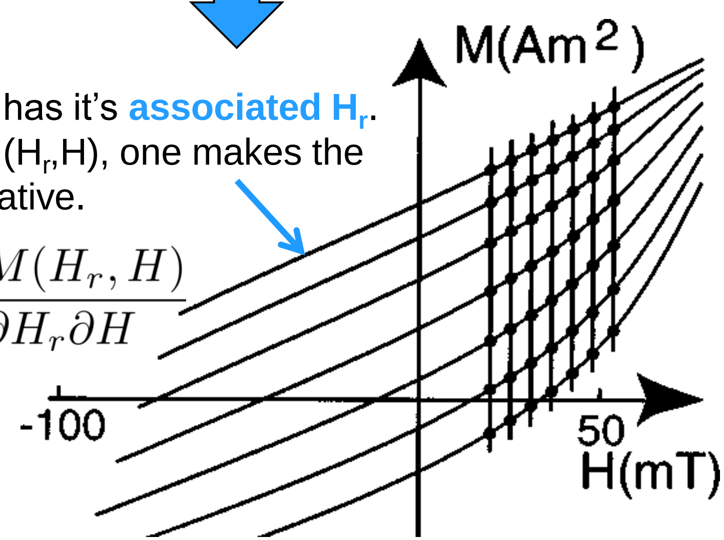
$H_u \equiv (H + H_r)/2$

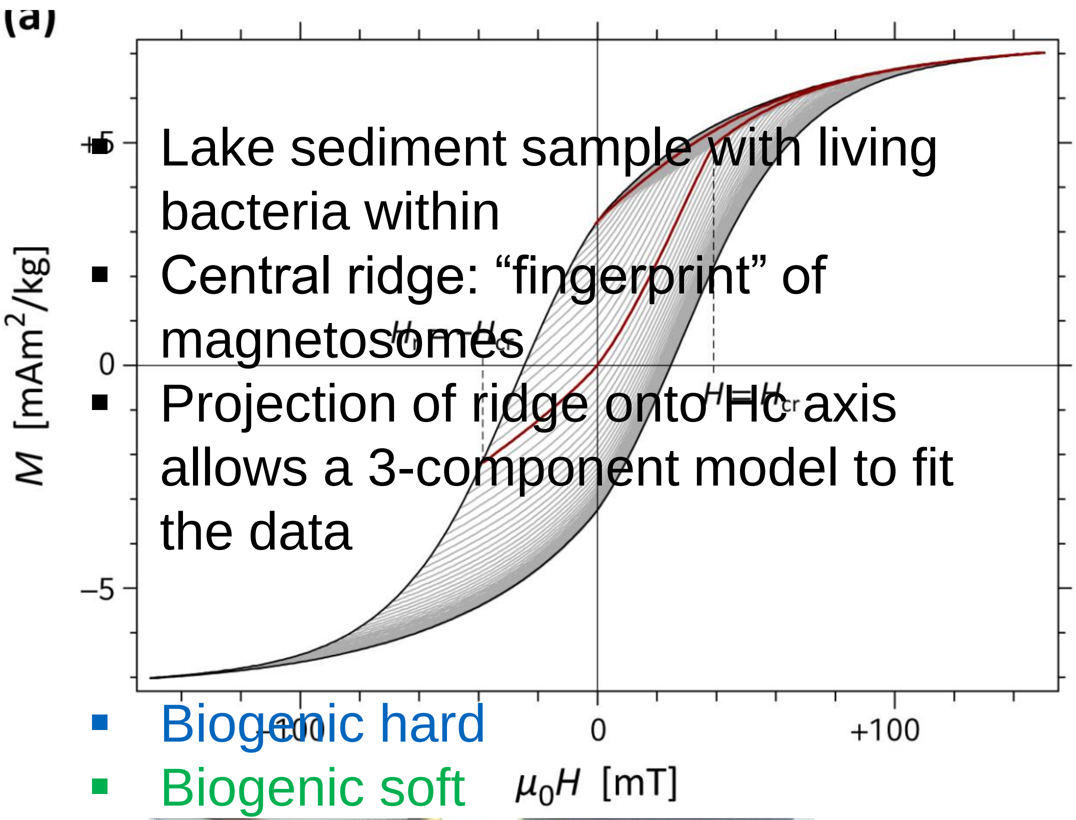


$H_c \equiv (H - H_r)/2$

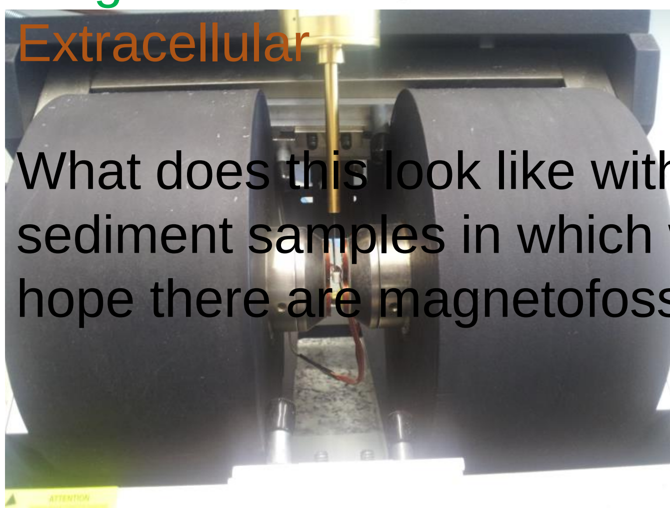
Each curve has it's **associated H_r**. With grid in (H_r,H), one makes the mixed derivative.

$$\rho(H_r, H) = -\frac{1}{2}\frac{\partial^2 M(H_r, H)}{\partial H_r \partial H}$$

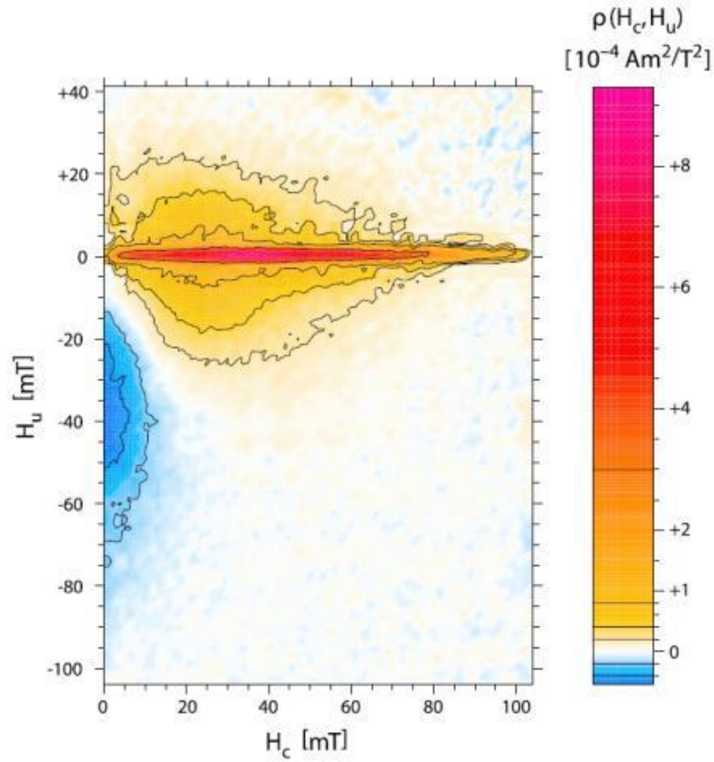


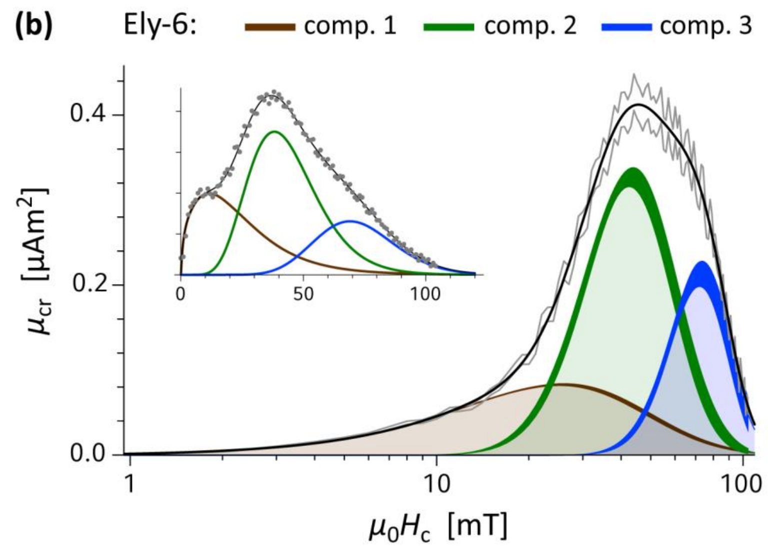


- Lake sediment sample with living bacteria within
- Central ridge: “fingerprint” of magnetosomes
- Projection of ridge onto Hc axis allows a 3-component model to fit the data
- Biogenic hard
- Biogenic soft
- Extracellular

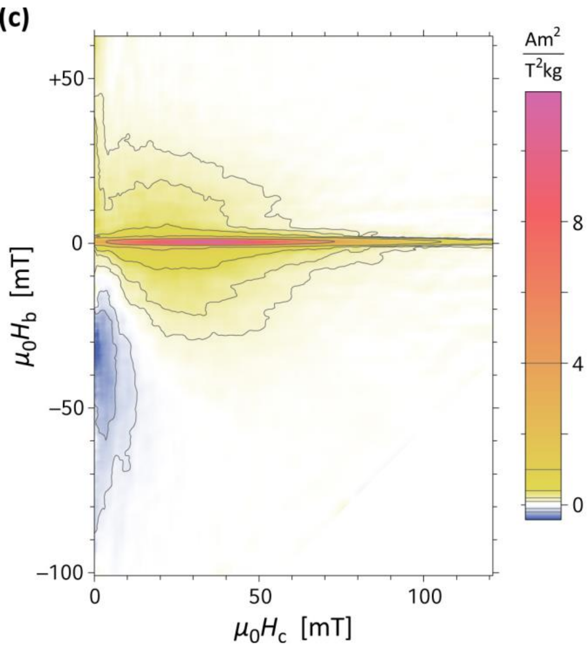


- What does this look like with our sediment samples in which we hope there are magnetofossils?





FORC's of Our Sediment



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<u>Untreated</u>

- 3% by mass Fe
- 60 ppm SD Iron
- Approx. 3% Fe in mag. minerals (biogenic and others)

<u>Treated</u>

60% from mag. minerals
> 27% from bacteria
< 6% from primary

P. Ludwig et al., Glob. Plan. Change **110** (2013)

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Characterization of primary and secondary magnetite in marine sediment by combining chemical and magnetic unmixing techniques

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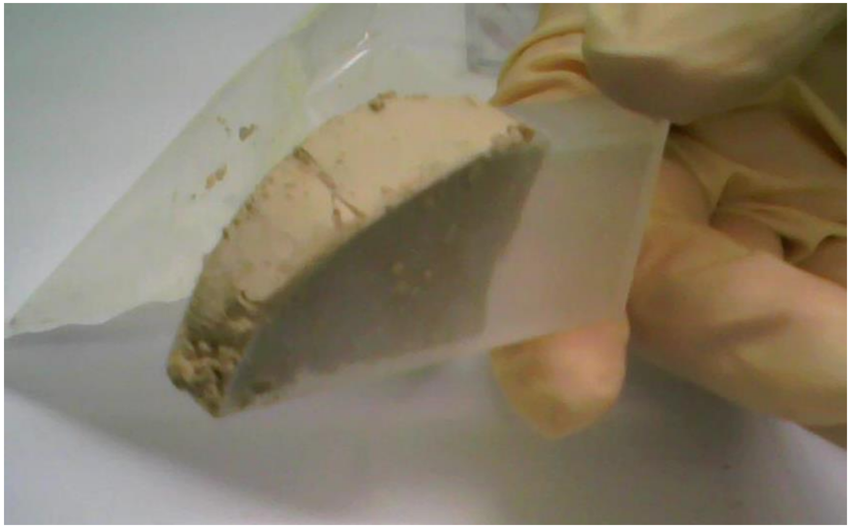
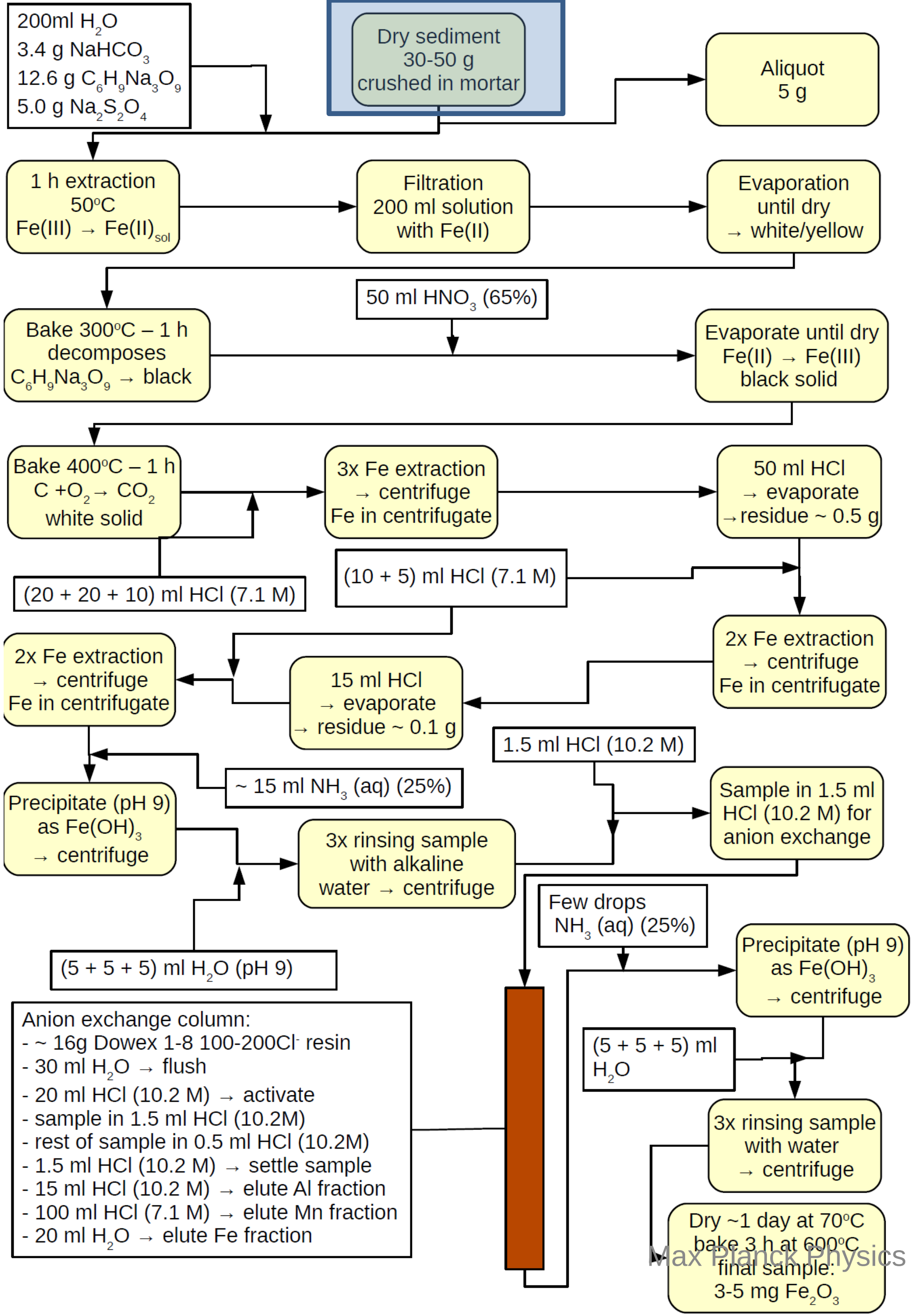
ABSTRACT

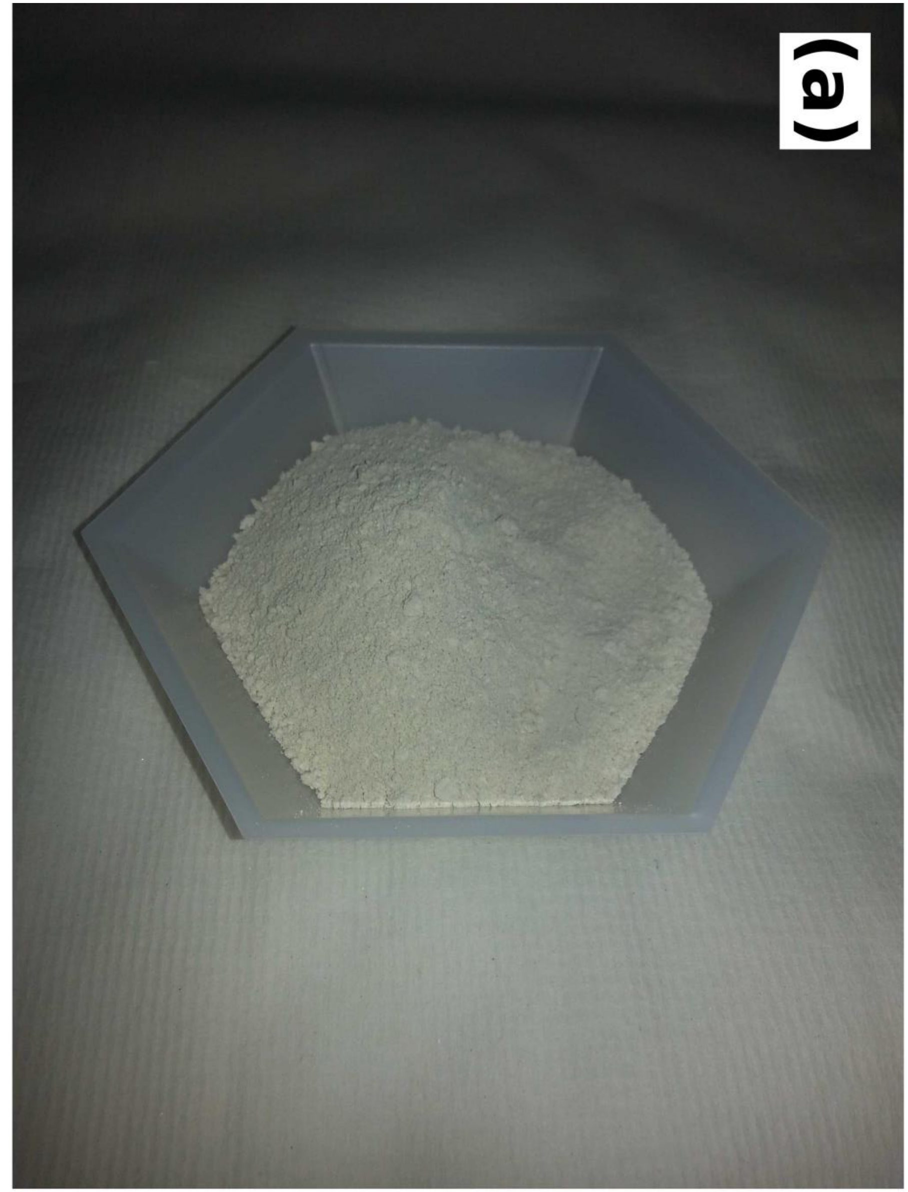
We present a novel technique for quantitative unmixing of primary and secondary ferrimagnetic minerals in sediments. Hysteresis and high-resolution first-order reversal curve (FORC) measurements are performed on sediment samples before and after digestion in a citrate–bicarbonate–dithionite (CBD) solution optimized for maximum selective extraction of secondary fine-grained iron oxides. The difference between magnetic measurements of untreated and CBD-treated sample materials is used to calculate the original magnetic signature of CBD-extractable minerals. A combination of selective chemical extraction and magnetic measurements suited

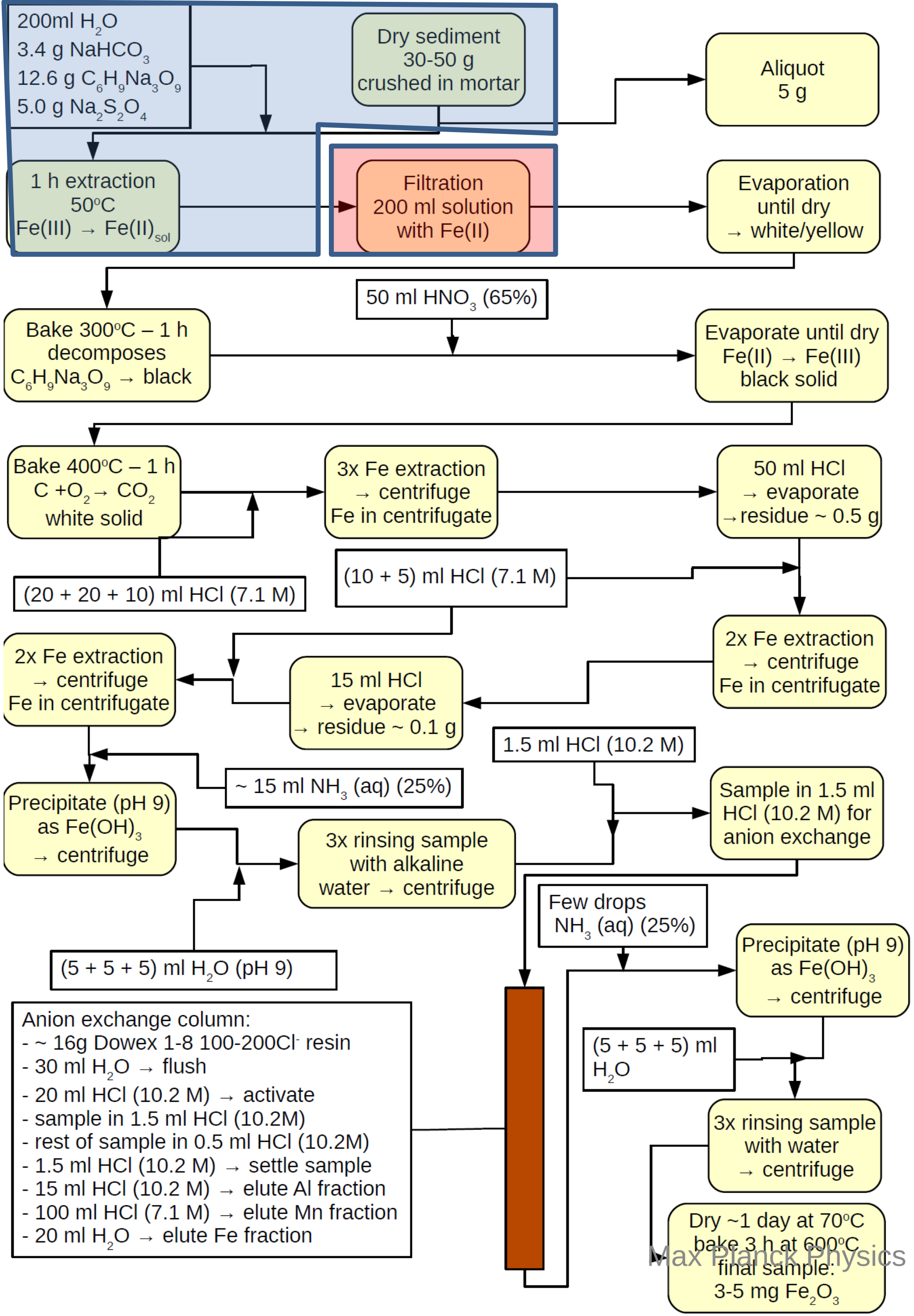


Extracting the Microfossil ^{60}Fe and Measuring It

THE HOW-TO'S


200ml H_2O
3.4 g $NaHCO_3$
12.6 g $C_6H_9Na_3O_9$
5.0 g $Na_2S_2O_4$
Dry sediment 30-50 g crushed in mortar
Aliquot 5 g
1 h extraction 50°C $Fe(III) \rightarrow Fe(II)_{sol}$
Filtration 200 ml solution with Fe(II)
Evaporation until dry → white/yellow
50 ml HNO_3 (65%)
Bake 300°C – 1 h decomposes $C_6H_9Na_3O_9$ → black
Evaporate until dry $Fe(II) \rightarrow Fe(III)$ black solid
Bake 400°C – 1 h $C + O_2 \rightarrow CO_2$ white solid
3x Fe extraction → centrifuge Fe in centrifugate
50 ml HCl → evaporate → residue ~ 0.5 g
(20 + 20 + 10) ml HCl (7.1 M)
(10 + 5) ml HCl (7.1 M)
2x Fe extraction → centrifuge Fe in centrifugate
15 ml HCl → evaporate → residue ~ 0.1 g
2x Fe extraction → centrifuge Fe in centrifugate
1.5 ml HCl (10.2 M)
~ 15 ml NH_3 (aq) (25%)
Precipitate (pH 9) as $Fe(OH)_3$ → centrifuge
3x rinsing sample with alkaline water → centrifuge
Sample in 1.5 ml HCl (10.2 M) for anion exchange
(5 + 5 + 5) ml H_2O (pH 9)
Few drops NH_3 (aq) (25%)
Precipitate (pH 9) as $Fe(OH)_3$ → centrifuge
Anion exchange column:
- ~ 16g Dowex 1-8 100-200Cl[-] resin
- 30 ml H_2O → flush
- 20 ml HCl (10.2 M) → activate
- sample in 1.5 ml HCl (10.2M)
- rest of sample in 0.5 ml HCl (10.2M)
- 1.5 ml HCl (10.2 M) → settle sample
- 15 ml HCl (10.2 M) → elute Al fraction
- 100 ml HCl (7.1 M) → elute Mn fraction
- 20 ml H_2O → elute Fe fraction
(5 + 5 + 5) ml H_2O
3x rinsing sample with water → centrifuge
Dry ~1 day at 70°C bake 3 h at 600°C final sample: 3-5 mg Fe_2O_3


(a)



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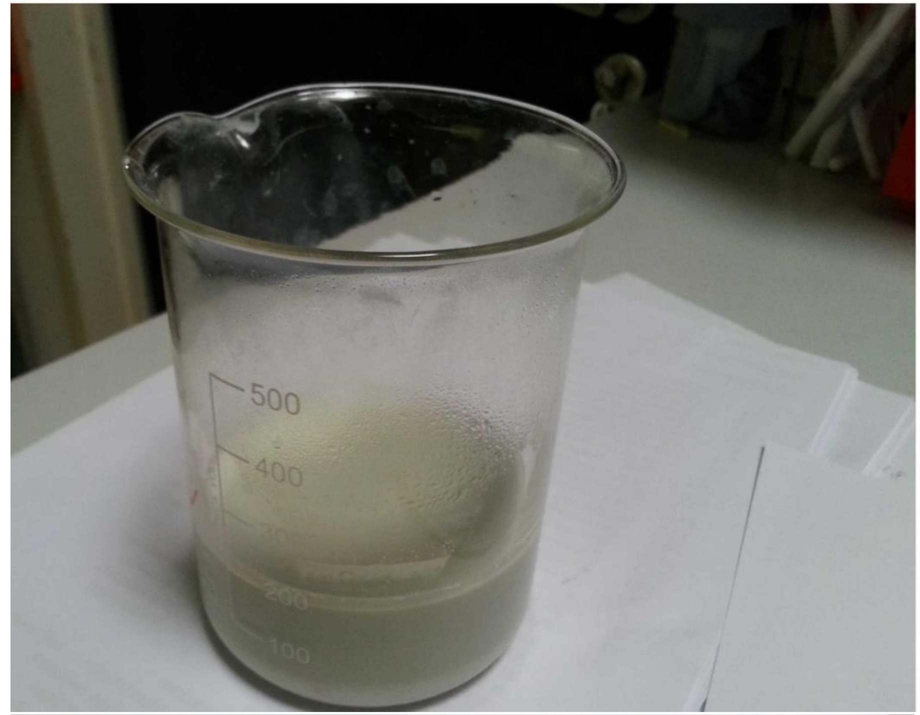
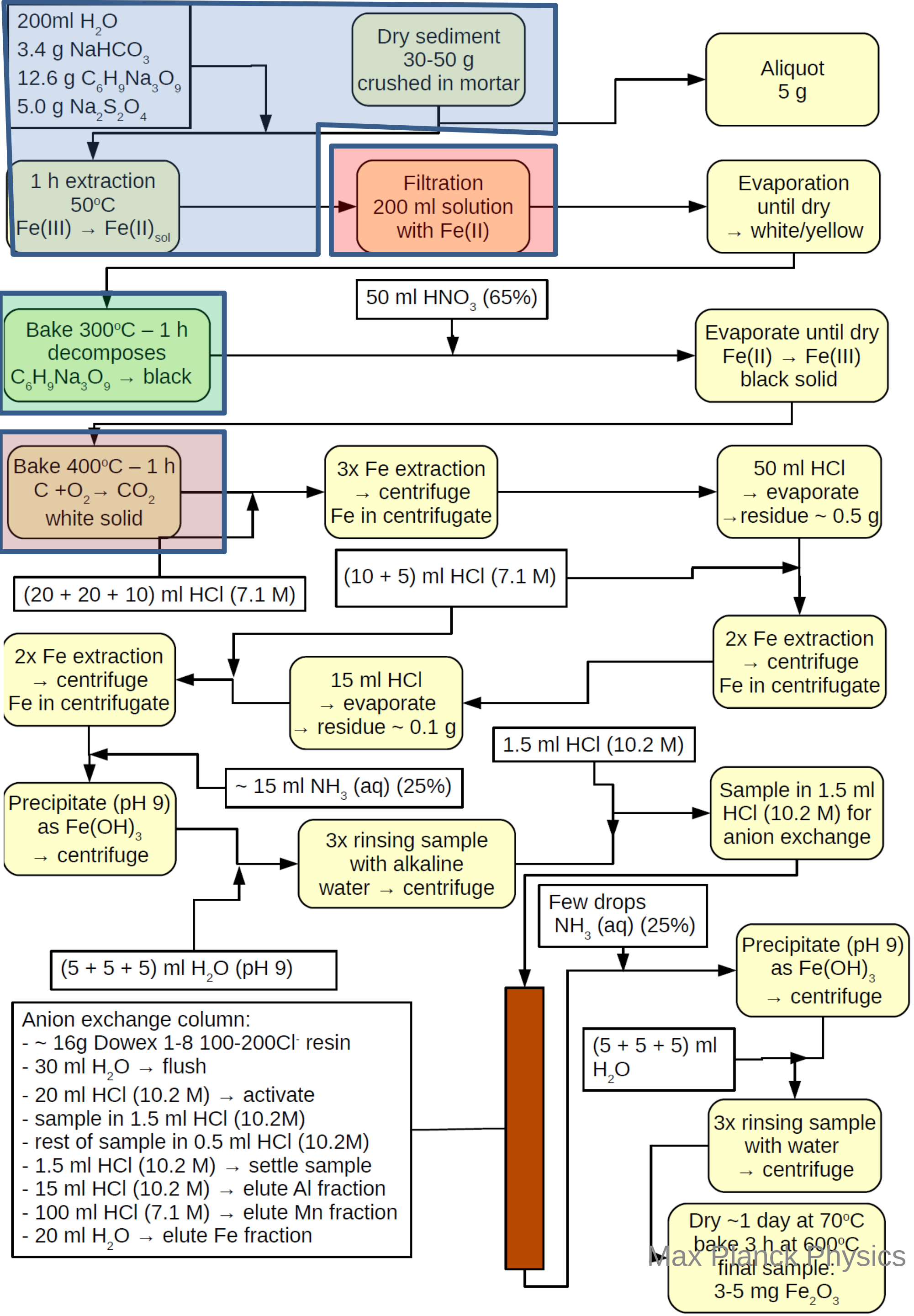


500
400
200
100

(c)
115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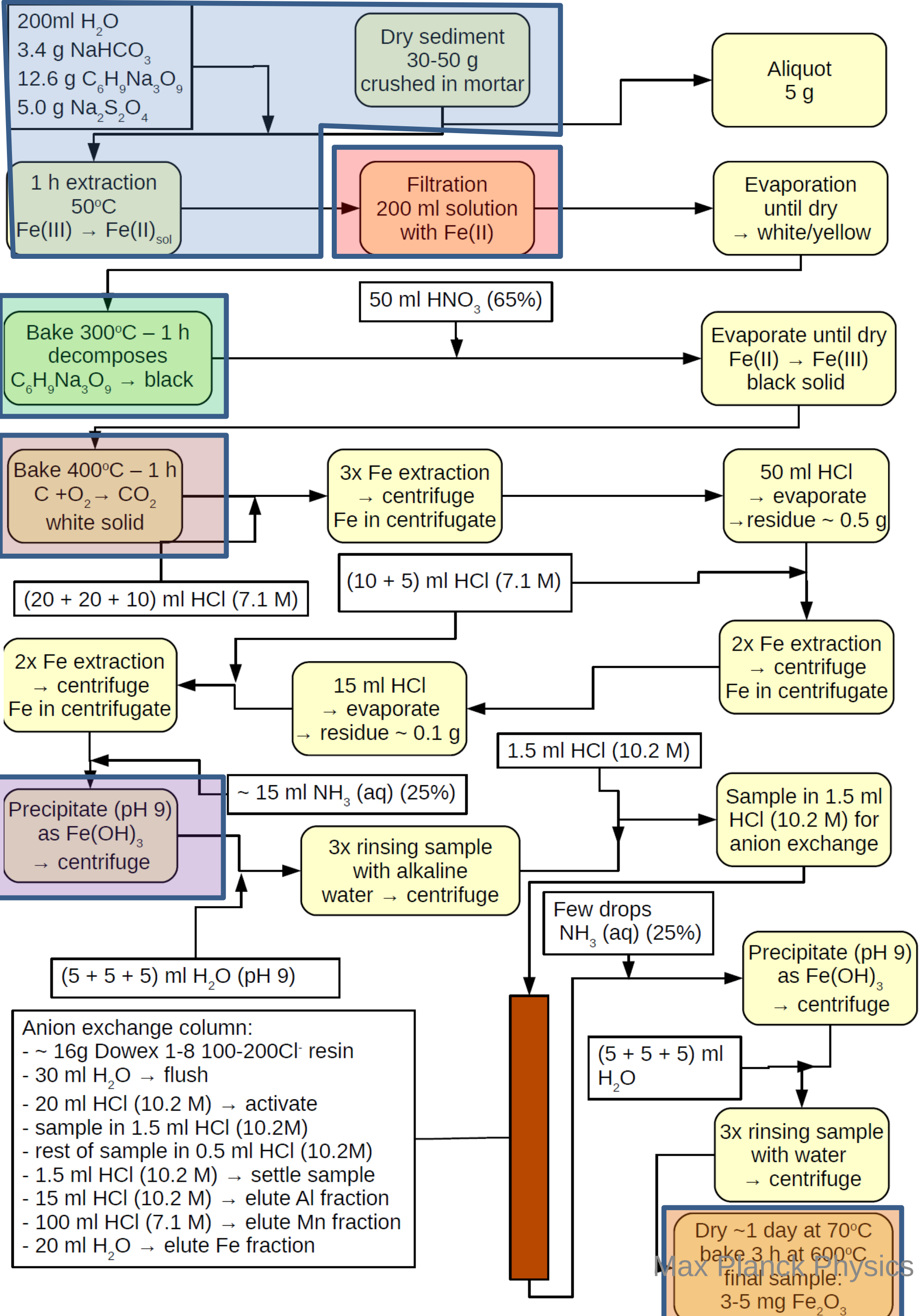
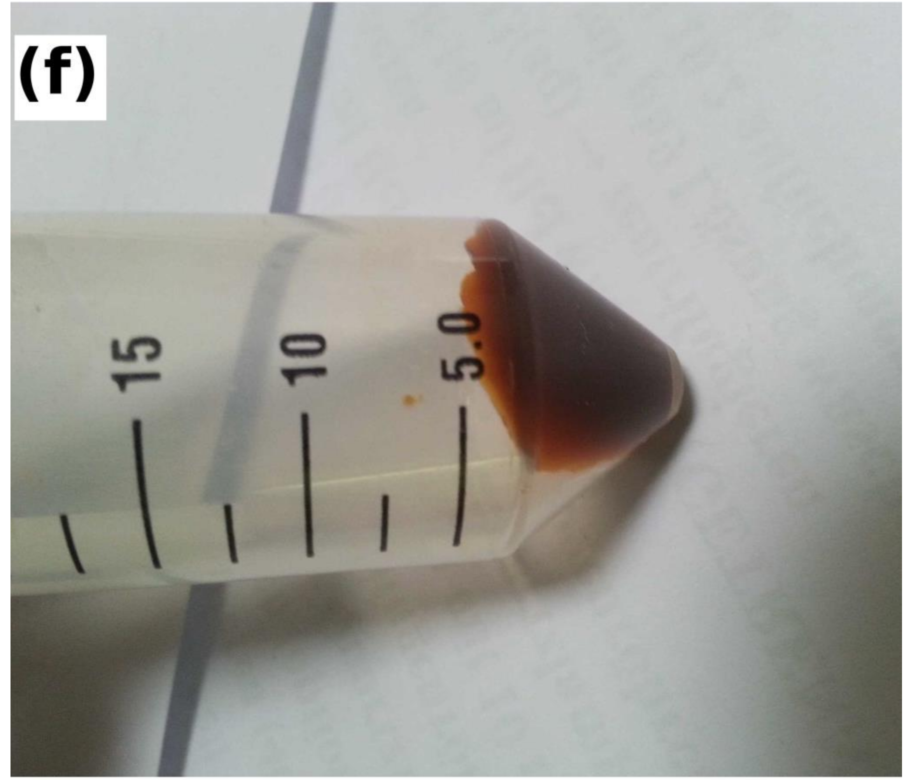
Dry sediment
30-50 g
crushed in mortar
Aliquot
5 g
1 h extraction
50°C
Fe(III) → $Fe(II)_{sol}$
Filtration
200 ml solution
with Fe(II)
Evaporation
until dry
→ white/yellow
50 ml HNO_3 (65%)
Bake 300°C – 1 h
decomposes
$C_6H_9Na_3O_9$ → black
Evaporate until dry
Fe(II) → Fe(III)
black solid
Bake 400°C – 1 h
C + O_2 → CO_2
white solid
3x Fe extraction
→ centrifuge
Fe in centrifugate
50 ml HCl
→ evaporate
→ residue ~ 0.5 g
(20 + 20 + 10) ml HCl (7.1 M)
(10 + 5) ml HCl (7.1 M)
2x Fe extraction
→ centrifuge
Fe in centrifugate
15 ml HCl
→ evaporate
→ residue ~ 0.1 g
2x Fe extraction
→ centrifuge
Fe in centrifugate
1.5 ml HCl (10.2 M)
~ 15 ml NH_3 (aq) (25%)
Sample in 1.5 ml
HCl (10.2 M) for
anion exchange
Precipitate (pH 9)
as $Fe(OH)_3$
→ centrifuge
3x rinsing sample
with alkaline
water → centrifuge
Few drops
NH_3 (aq) (25%)
Precipitate (pH 9)
as $Fe(OH)_3$
→ centrifuge
(5 + 5 + 5) ml H_2O (pH 9)
Anion exchange column:
- ~ 16g Dowex 1-8 100-200Cl⁻ resin
- 30 ml H_2O → flush
- 20 ml HCl (10.2 M) → activate
- sample in 1.5 ml HCl (10.2M)
- rest of sample in 0.5 ml HCl (10.2M)
- 1.5 ml HCl (10.2 M) → settle sample
- 15 ml HCl (10.2 M) → elute Al fraction
- 100 ml HCl (7.1 M) → elute Mn fraction
- 20 ml H_2O → elute Fe fraction
(5 + 5 + 5) ml
H_2O
3x rinsing sample
with water
→ centrifuge
Dry ~1 day at 70°C
bake 3 h at 600°C
final sample:
3-5 mg Fe_2O_3


(d)



200ml H_2O
3.4 g $NaHCO_3$
12.6 g $C_6H_9Na_3O_9$
5.0 g $Na_2S_2O_4$
Dry sediment 30-50 g crushed in mortar
Aliquot 5 g
1 h extraction 50°C Fe(III) → $Fe(II)_{sol}$
Filtration 200 ml solution with Fe(II)
Evaporation until dry → white/yellow
50 ml HNO_3 (65%)
Bake 300°C – 1 h decomposes $C_6H_9Na_3O_9$ → black
Evaporate until dry Fe(II) → Fe(III) black solid
Bake 400°C – 1 h C + O_2 → CO_2 white solid
3x Fe extraction → centrifuge Fe in centrifugate
50 ml HCl → evaporate → residue ~ 0.5 g
(20 + 20 + 10) ml HCl (7.1 M)
(10 + 5) ml HCl (7.1 M)
2x Fe extraction → centrifuge Fe in centrifugate
15 ml HCl → evaporate → residue ~ 0.1 g
2x Fe extraction → centrifuge Fe in centrifugate
1.5 ml HCl (10.2 M)
~ 15 ml NH_3 (aq) (25%)
Precipitate (pH 9) as $Fe(OH)_3$ → centrifuge
Sample in 1.5 ml HCl (10.2 M) for anion exchange
3x rinsing sample with alkaline water → centrifuge
Few drops NH_3 (aq) (25%)
(5 + 5 + 5) ml H_2O (pH 9)
Precipitate (pH 9) as $Fe(OH)_3$ → centrifuge
Anion exchange column:
- ~ 16g Dowex 1-8 100-200Cl⁻ resin
- 30 ml H_2O → flush
- 20 ml HCl (10.2 M) → activate
- sample in 1.5 ml HCl (10.2M)
- rest of sample in 0.5 ml HCl (10.2M)
- 1.5 ml HCl (10.2 M) → settle sample
- 15 ml HCl (10.2 M) → elute Al fraction
- 100 ml HCl (7.1 M) → elute Mn fraction
- 20 ml H_2O → elute Fe fraction
(5 + 5 + 5) ml H_2O
3x rinsing sample with water → centrifuge
Dry ~1 day at 70°C bake 3 h at 600°C final sample: 3-5 mg Fe_2O_3



(f)
15
10
5.0

Fe_2O_3

ODP Drill Core 848 Results

- Width ~800 kyr
- World record blank level sensitivity
- World record $^{60}Fe/Fe$ concentration sensitivity

Data redacted: Under peer review.

Will appear in Proc. Natl. Acad. Sci.

ODP Drill Core
851 Results

Data redacted: Under peer review.

Will appear in Proc. Natl. Acad. Sci.

Summary

- Supernova signal residing in a biogenic reservoir
- First time-resolved SN signal transiting solar system
  - Does the time profile map the density profile of that material?
  - Width of ~800 kyr and shape requires future theoretical explanation
  - Is the shape determined by dynamics of supernova ejecta with interstellar medium?
  - Is the shape determined by terrerstrial “residence times?
  - Combination of these both?
- What’s next? To search for ^{244}Pu (r-process only isotope) to try answering:
  - Does the r-process occur in core-collapse supernova?
  - Geological field expedition to Atacama in Nov. 2016
- This work to appear infed Proc. Nat. Acad. Sci. (PNAS)