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Muons from mobile accelerators in large-scale environmental analysis

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Where we start from

IHI Zittau; Dresden Tech (TUD)

Site of old lignite pits → man-made lake landscape, strongly mixed, chemically still active topsoils

Background in environmental and technical analytical chemistry, electrochemistry; interested in „more physical methods“ to cover larger areas by **one screening**, rather than analyzing samples from (n + 1,001) bore-holes

Which kinds of data do natural (soil, bio-mass) samples reveal without prior impact of chemicals or ionizing radiation? → activation by cosmic

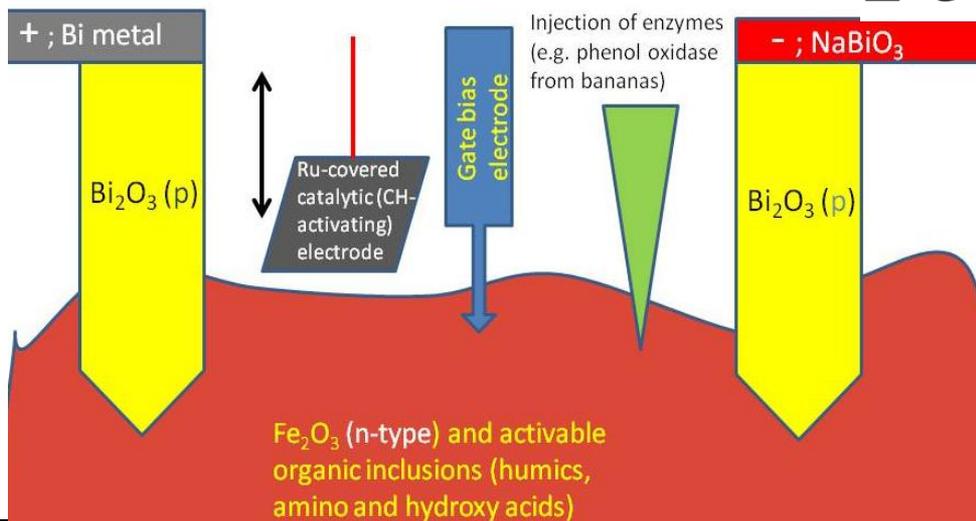


Lake Olbersdorf, residual matter mound



Fe(III) oxide precipitations from aq. Fe^{2+} trickling to the surface from a perturbed soil arrangement; **(down right)**: I am **not** going to speak about this today: making signal amplification right from soil heterogeneity, Fe richness, SC properties!

Zittau and N part of Lusatian Mountains (D/Cz.R.); back centre: Lake Olbersdorf inundated lignite pit, **residue heaps**



Why muons, and which ones?

Muons can penetrate thick samples but are suitable to detect fairly small cavities therein

μ^+ almost behave like protons in condensed media, forming chemical bonds (e.g. $(\mu\text{-OH; muonic water})$) and get accessible to ESR during the μ s before they decay

μ^- are kind of „heavier electrons“ (lepton universality), get trapped by cations or neutral molecules after cooled down to some 40 eV ($v \approx 260 \text{ km/s}$) interacting with heavy atoms, $> 75 \text{ eV}$ for very light ones, only then take part in chemical binding –

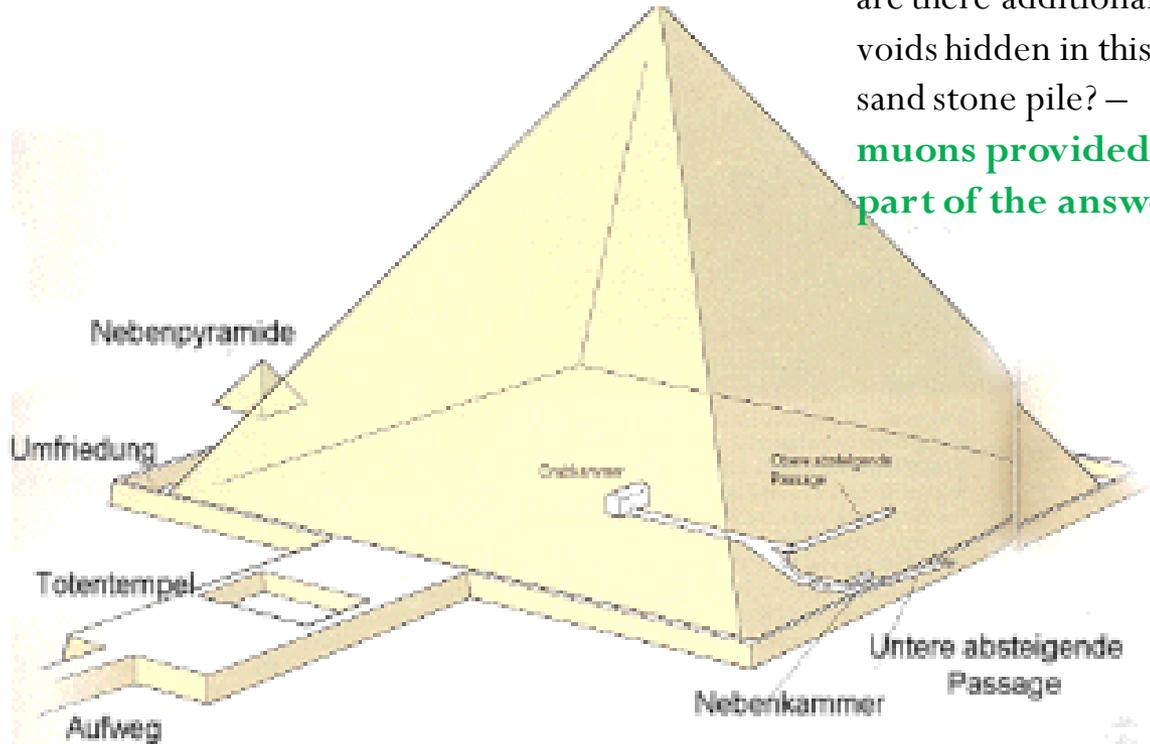
but that's **not** the entire story!



Pharaoh Chephren's large pyramid: was he ever buried there, and

are there additional voids hidden in this sand stone pile? –

muons provided part of the answer!



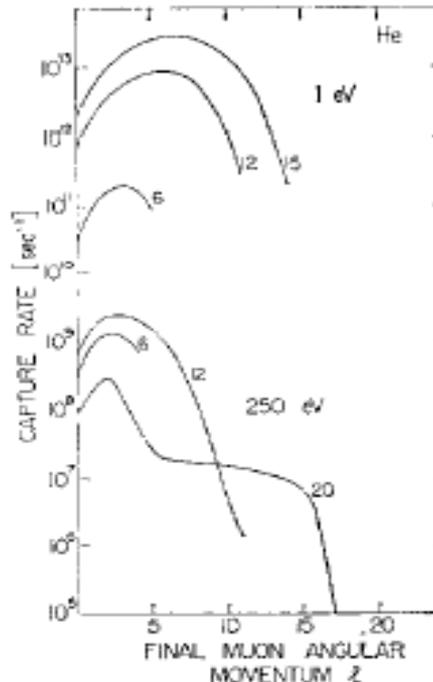
Inverse β decay: electron capture (comparison)

- Electrons (single or pairs [e.g. with ^{78}Kr , ^{130}Ba , ^{152}Gd – besides of α decay in latter: 10^{14} a]) can be trapped by protons (**up** quarks therein) by way of weak interaction, but
- For xs orbitals even, **electron** probability to be inside the nucleus at a given time is small
- Muons are 207 times heavier than electrons \rightarrow average muon/nucleon [μ^-/u quark]-distance 207 times smaller than size of electronic K, L orbitals \rightarrow muon density in nucleus $(207)^3 \approx 10^7$ times larger than with electrons, moreover: what would take 10 s with inv. β decay, will be done within $\ll 1 \mu\text{s}$ by μ^- because
- much more energy is to be gained/released by muon capture hence probability of corresponding weak interaction per period of time is much larger

\rightarrow once negative muons were trapped to a single atom, they are likely to react with the nucleus although short-lived

This reaction as a rule produces **radionuclides**

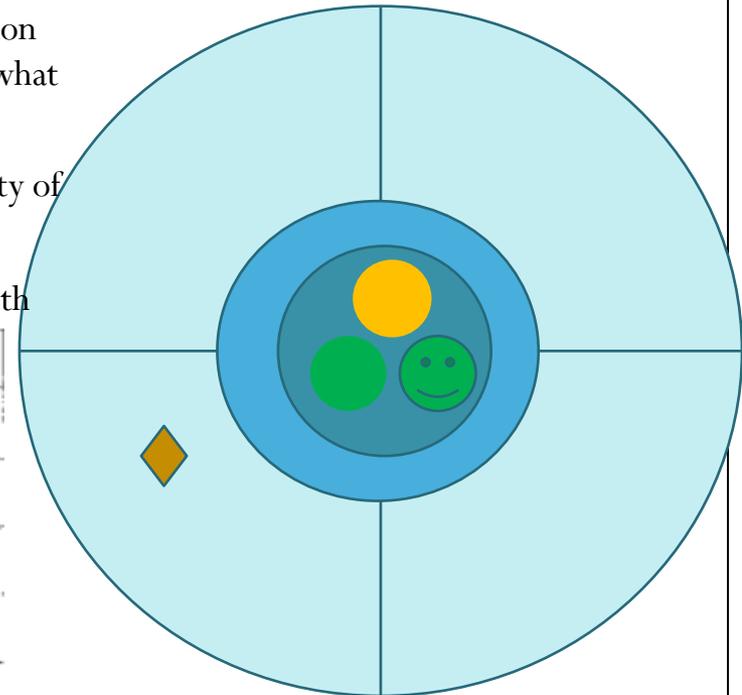
Quench, capture energy and final angular momentum distribution for muon quenching by ^4He ; subsequent nuclear reaction likely only for $L = 0, 2, 4 \dots$ (best: $L = 0$ [s orbitals, right picture])



$$(\text{lepton})^- + \mathbf{u} \text{ q.} \rightarrow \nu + \mathbf{d} \text{ q.}$$

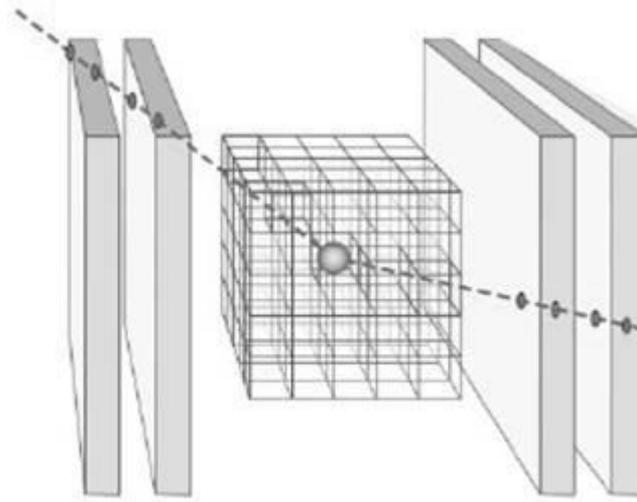
$$= (\text{lepton})^- + p \rightarrow \nu_{(L)} + n$$

(λ^0);
always **exothermic** for $L = \mu$ or τ



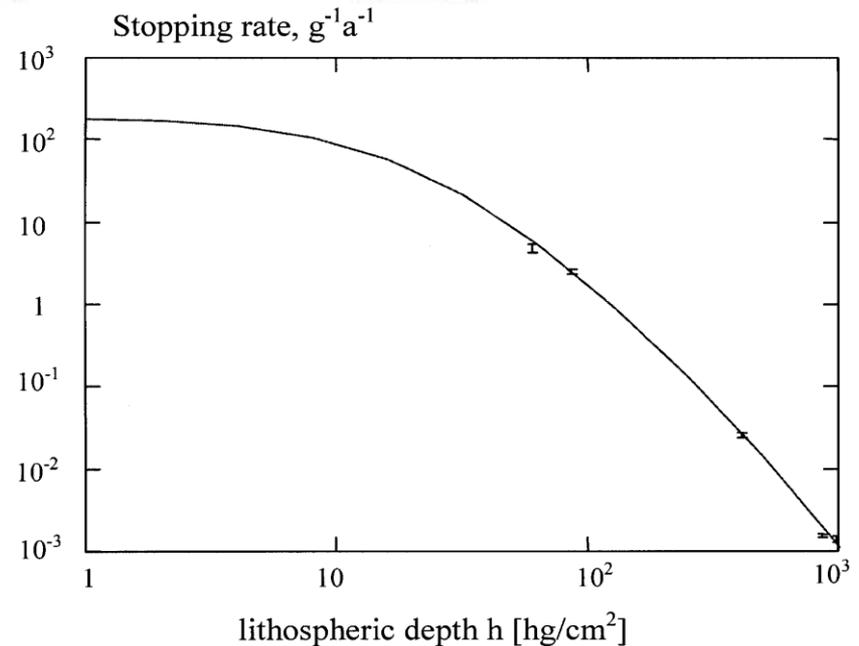
The probability to find a muon (brown crystal) in 1s or 2s..., states somewhere increases from outside when approaching the nucleus (quarks shown) [light to dark blue shades]

Make muons, pass them to the ground



A muon beam may be deflected at both voids and sites where exceptionally light or heavy atoms „gather“ (e.g. organic inclusions)

- Size, kind of μ source: GeV protons, about car-sized
- Electromagnetic deflection
- Range of muons, range of secondary γ photons \rightarrow measurements must be made parallel to some surface (grass-roots level, along a cliff or around a borehole)
- Lifetime of, penetration by „moderately relativistic“ muons \rightarrow beam will pass several 100 m ($\tau_0 * c \approx 660$ m) \rightarrow an area of several times 50 m in diameter can be sampled from a single point of muon injection \rightarrow rapid analysis at high spatial resolution
- Activation of U, Pu by simple capture producing the typical prompt Moseley-type γ radiation rather than causing nuclear reactions which give away hidden nuclear materials



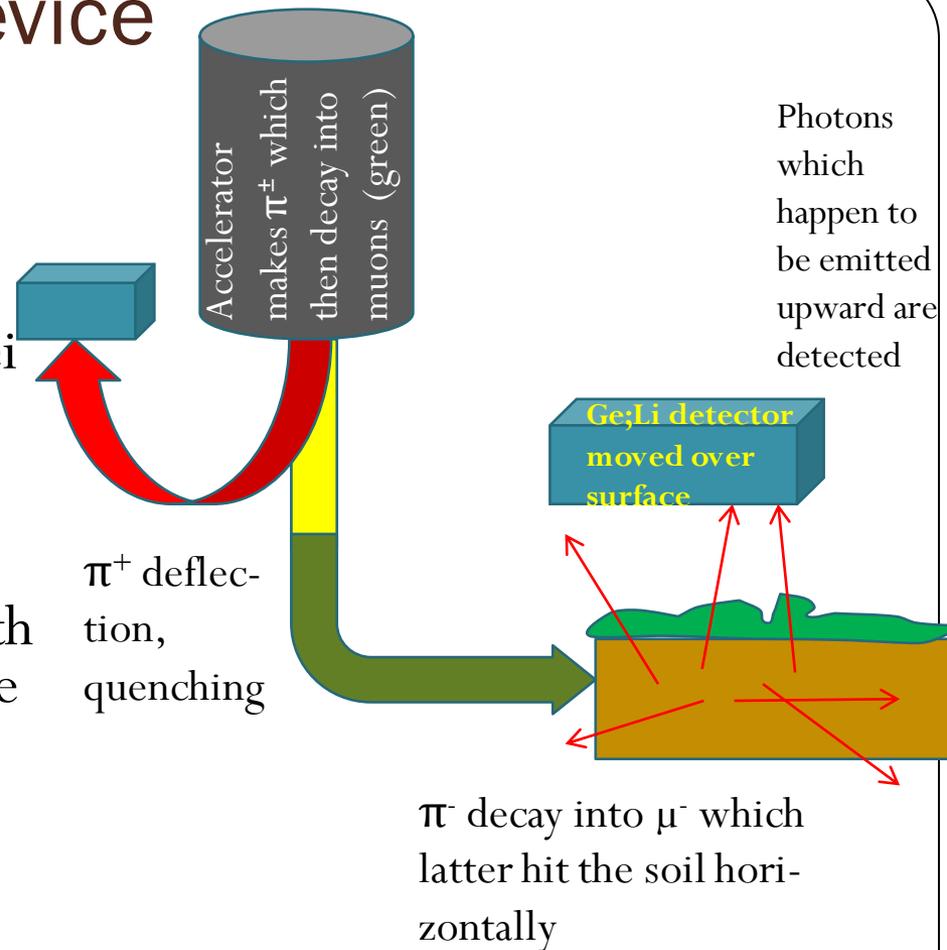
Range, capture rate of cosmic rad. μ^- vs. amount of overlying sediment matter, rocks: decline starts at ≈ 1 kg/cm² ≈ 4.5 m layer, reasonable yields still at 20 kg/cm² $\approx 75 - 80$ m layer; average primary energy higher than in soilborne irradiation

Size, construction of device

- Table-top electron- or proton accelerator with loop system (e some 300 MeV, p about 1 GeV)

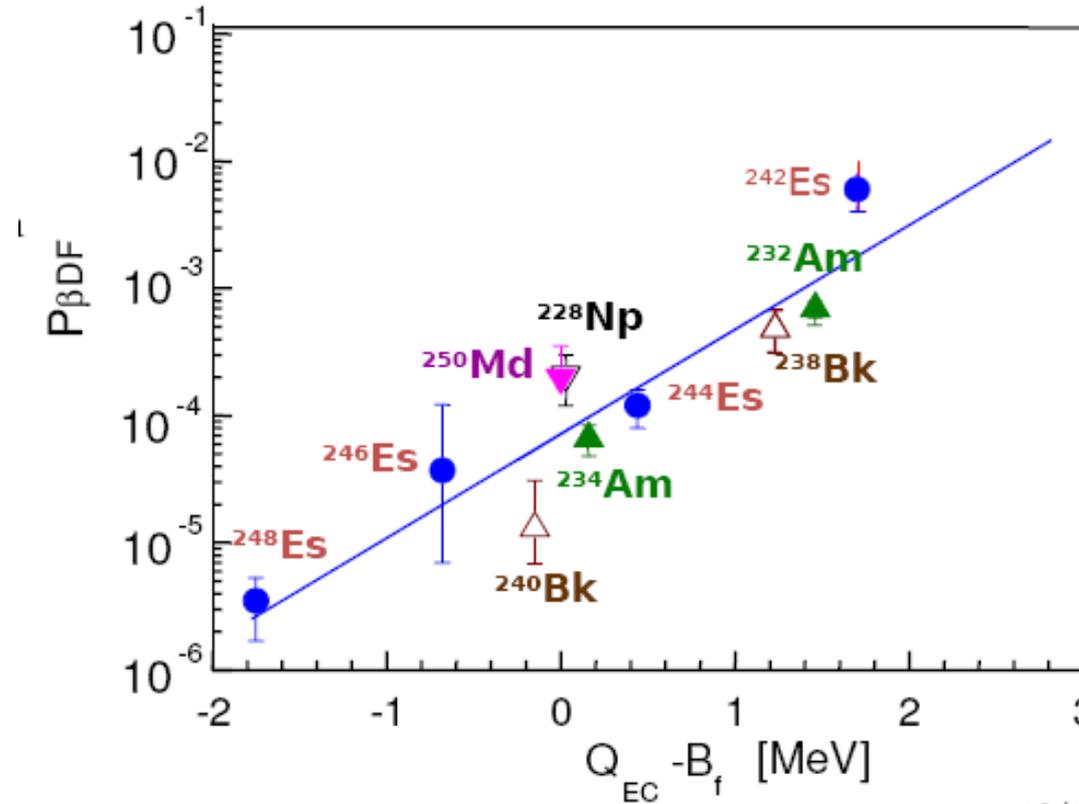
First, π^\pm are formed/kicked out of nuclei which

- decay into μ on some 1 m way-length
- electromagnetic separation of μ and π mesons (remaining π rapidly react with matter in a very thin layer, while μ^+ are discarded), then the
- μ^- beam is directed parallel to the surface
- γ detector is moved around on the surface, looking „downward“
- Classical translation of γ spectra into element compositions, like in INAA



Kinds of nuclear reactions secondary to (delay due to) μ^- capture by nuclei (w.i.)

- Pure γ emission (daughter nuclide is stable but formed in an excited state), to be distinguished from Auger-Moseley emissions shifted into γ by muon mass
- n- or p emission
- Fission (cp. β dF, disc. at Dubna on ^{232}Am ; β dF may occur rarely if energy gain $<$ fission barrier)
- Fission is also possible by a closely orbiting muon (1s- or 2s states) pulling opposite (proton) charges in nucleus forth and back periodically, making the nucleus vibrate (lepton, purely electromagnetic effect)
- n emission prevails in moderately heavy nuclei, commonly just one neutron is detached (excitation energy ≈ 10 MeV; $\rightarrow {}^m\text{E}^Z \rightarrow {}^{m-1}\text{E}'^{Z-1}$, e.g. $^{68}\text{Zn} \rightarrow ^{67}\text{Cu}$ ($T_{1/2} = 62$ h; $\gamma = 185$ or 93 keV); pattern of products from common topsoil, sand, pig iron, carbonate etc. minerals (that is, C, N, O, Si, S, Ti, Ca, Mg or Fe isotopes)



Beta-delayed fission yields vs. energy excess during decay: a rather rare event which suggests most heavy nuclei will behave well during μ^- capture, rather than undergo fission and make „atypical“ daughter nuclides (for comparison only)

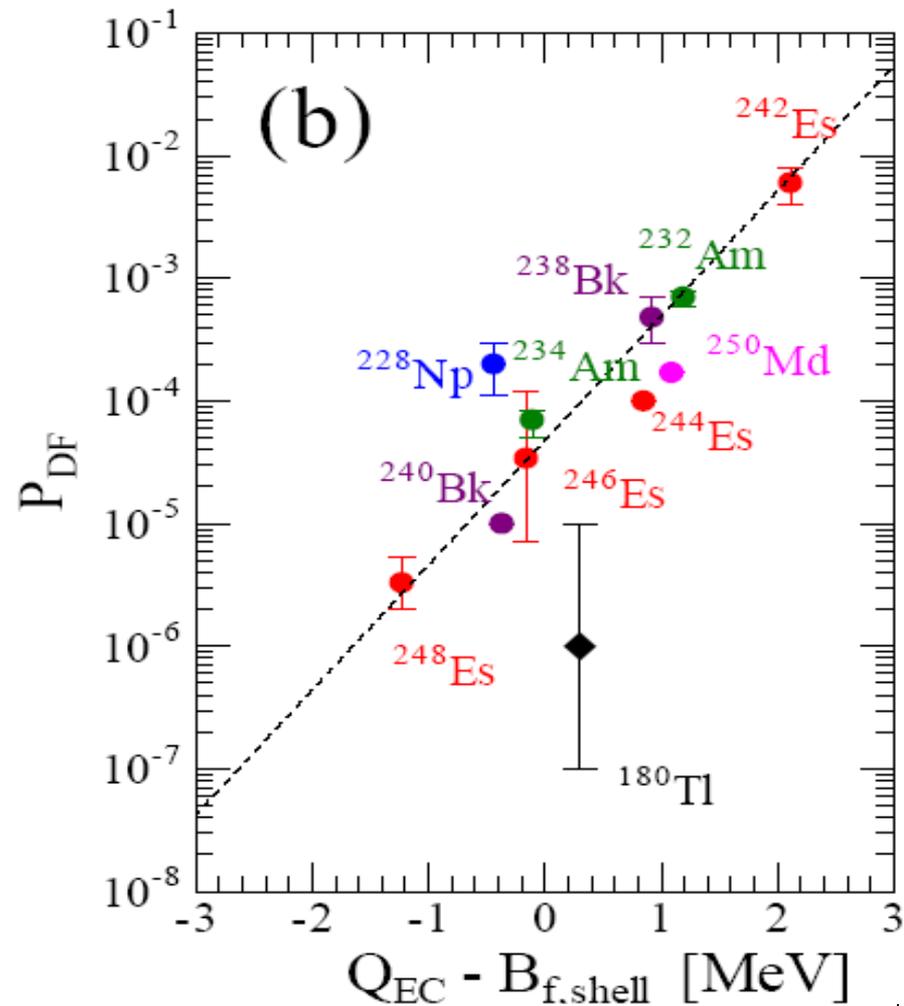
Examples of emitting nuclides

- Available from most elements by one pathway or another
- Positron emitters, annihilation γ from Cd, Sn, Ba, Ce, Nd, and Hg $\rightarrow \gamma$ (341; 511 keV) useful for summarizing toxicological indications (Cd, Hg, Ba)! [table]
- Fission products
- „common“ β^- emitters even from very light elements

element	Short-lived isotopes of (Z – 1); half-life	Natural abundance of M	Natural abundance of (M + 1; M + 2)	β^+ emission, nuclide
Cd	¹⁰⁵ Ag 41.3 h; ¹⁰⁶ Ag 24 min		¹⁰⁶ , ¹⁰⁸ Cd about 1% each	β^+ in ¹⁰⁵ Ag, ^{106m} Ag; extremely hard γ in ¹¹² Ag (3.1 h; 1387 keV) and ¹¹⁵ Ag (20 min; 2156 keV)
Sn	¹¹⁰ In 4.9 h; ^{110m} In 69 min; ^{111m} In 7.7 min; ¹¹⁴ In 72 s;...		¹¹² Sn 1.0%	¹¹⁰ In β^+
Ba	¹²⁹ Cs 32.06 h; ¹³⁰ Cs 29.2 min; ^{134m} Cs 2.9 h; ^{135m} Cs 53 min		¹³⁰ , ¹³² Ba about 0.1% each	Almost purely β^+ in ¹²⁹ , ¹³⁰ Cs (¹³¹ , ¹³² Cs but too long-lived for efficient detection)
Ce	¹³⁵ La 19.5 h; ¹³⁶ La 9.9 min; ¹⁴⁰ La 40.2 h; ¹⁴¹ La 3.9 h; ¹⁴² La 91 min		¹³⁶ Ce 0.19%	¹³⁵ , ¹³⁶ La (100% each)
Nd	¹⁴⁰ Pr 3.4 min; ¹⁴² Pr 19.1 h; ^{142m} Pr 14.6 min		¹⁴² Nd 27.1%	¹⁴⁰ Pr β^+ 100%
Hg	^{196m} Au 8.1 s; ^{197m} Au 7.73 s; ²⁰⁰ Au 48.4 min; ²⁰¹ Au 26 min; ²⁰² Au 28.8 s; ²⁰³ Au 53 s; ²⁰⁴ Au 40 s	¹⁹⁶ Hg 0.15%	¹⁹⁸ Hg 10.1%	γ deexcitation daughter ¹⁹⁶ Au (6.17 h): β^+ = 92.8%; plus fission products

„Good“ vs. „poor“ absorbers

- Water content of sample and muon moderation efficiency
- Production of β^+ emitters from certain p-rich nuclides (precursors ...) \rightarrow annihilation radiation among the γ spectrum
- Particularly hard γ radiation emitters in spectrum
- C-, N-, S- vs. O capture efficiency \rightarrow how is the chance to detect oxidation state of sample, or even content of organic matter?
- μ dF of heavy nuclei, mainly Pb: an analytical nuisance or a chance to enhance sensitivity? (rate of μ dF [cp. β dF: always $\ll 1\%$ except for ^{242}Es ?]; exper. muonogenic fission yield: $13 \pm 5\%$ in ^{238}U , some 45% in ^{239}Pu , $< 2\%$ for ^{232}Th (cp. thermal neutrons))
- Typical traces of Hg, Tl, Bi and lighter Pb isotopes are detected only due to fission (look to right!)



β dF yields increase by **factor 10 with every MeV $\Delta \epsilon B_f$** \rightarrow about 3% at + 3MeV; efficient excess some 5 MeV at Pb ($M \neq 208$), $\rightarrow y_{\mu\text{dF}} > 50\%$; Tl, Bi..., $(M/Z)_{\text{prod}} \approx 2.6$ in μ dF products with $Z_{\text{start}} \geq 80 \rightarrow$ few s to min; no perturbation of Sn, Ba, REE detection, but fission-less detection of Pb only as ^{206}Tl (4.2 min, $\gamma = 803$ keV)

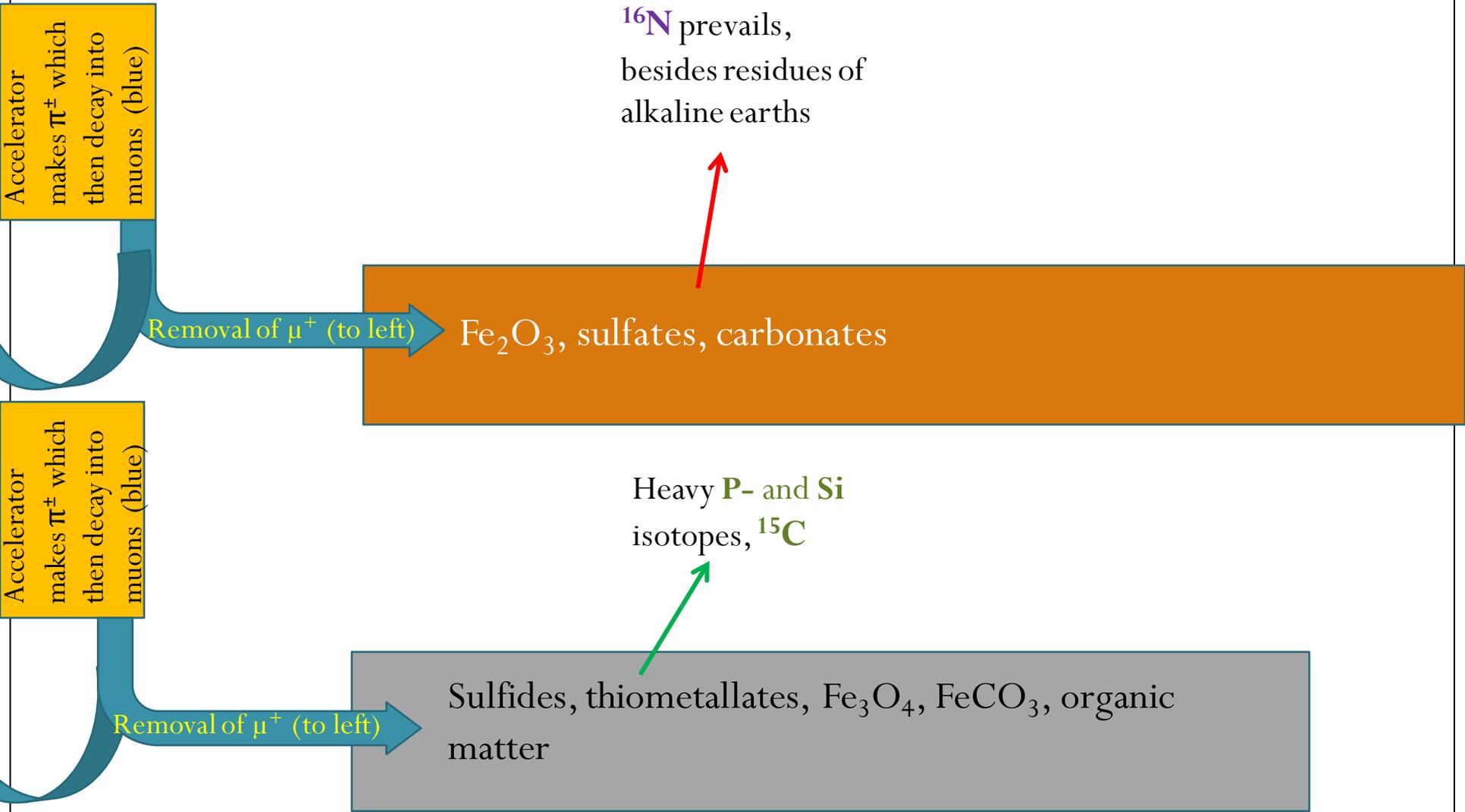
Safety considerations: where can it be done?

- We do not do anything else than is caused by cosmic radiation anyhow though at elevated rates
- Control of beam: loss of electromagnetic deflection means directing beam perpendicularly downward into massive bedrock
- Life-times, amounts of radionuclides formed (few GBq along 100m/ 20 cm diameter flux tube for minutes to weeks)
- Effects on local biota?! - μ SR **with low energy μ^+ is used for making pictures [μ -RT], not destroying tumour cells, even in human brain tissue!**



Slightly rough forest terrain in which such measurements may be made (for illustration only, displaying biogenic Fe oxide deposits above a reducing bottom environment)

Oxidizing vs. reducing soil conditions: muons feel and tell the difference!



Estimating analytical sensitivities

- Several γ /min at given energy readily detectable by Ge;Li detector, < 10% of photons directed into upward segment, some 70 - 80% absorption in thicker soil layers \rightarrow about 2 Bq must be produced (≈ 0.1 Bq/kg in 30*30*20 cm activated volume)
- 10^5 muons/s back in 2002, today some 10^7 ; equal-intensity pathway some 10 m, high relative yields in abundant isotopes, but annihilation γ from rare ones ($\leq 1\%$ except for ^{198}Hg) only, average radionuclide lifetime several times 10^3 s means
- Saturation activation after some min by $\approx 10^9$ μ^- total dose in column (some 10^7 in sampling segment of flux tube)
- ppm or upper-ppb determination levels should be reached for elements which produce min- to hour-lifetime nuclides from fairly abundant isotopes
- \rightarrow **competes with ICP-OES, inferior to ICP-MS but you need no drilling and no preparation/digestion of hundreds or thousands of individual samples, and you won't destroy information on local structures, redox conditions, organic contents!!**

Thank you for your kind attention –
spassiba!