Muon beams for environmental analysis: determining both element contents and subsoil redox conditions over soccer-playground-size areas

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Abstract: Negative muons can activate nuclei of almost all elements after thermalization, capture and reaction by weak interaction. While muons can penetrate some 100 m of solid rock or soil (low-energy ones a few m limited by decay), γ radiation from the secondary nuclides ($T_{1/2}$ between 10 s and some days) has a range of about 50 cm. Hence a muon beam passed parallel to some soil surface (horizontally or next to a borehole or well) will produce a linear trace of secondarily formed radionuclides which reveal their presence by γ radiation escaping to the nearby surface while the long range of the muon beam permits to analyze surface regions of an area of some 200 m diameter from a single, fixed point of muon production and injection. Reactions among μ^- and non-metal nuclei like ^{15}N, ^{34}S additionally provide information on oxidation state of surface soils (e.g. if water-logged). Estimates of analytical sensitivity are given in the paper. The muons are best produced by a laser-pulse driven electron wakefield accelerator which is small enough for field-site transport and –application, better than using more classical accelerator designs.

Introduction

Muons, the heavier counterparts (but not excited states) of electrons are now known for more than 75 years. People become soon aware of their capability to penetrate very thick layers of matter (superior to every other kind of radiation except of neutrinos in this respect [Cox 2008]) yet efficiently cause nuclear reactions competing with their intrinsic decay (Cohen et al. 1960) as the latter is rather slow ($\tau \approx 2.2$ μs). This stunning combination of properties is due to their being leptons and thus "blind" towards strong interaction, restricting nuclear and particle interactions to the **weak** and electromagnetic forces only; the charge of a proton is cancelled by reaction (weak interaction) with the muon via

$p + \mu^- \rightarrow n + \nu_\mu$ (muon neutrino) [1]

so that the product nucleus charge will be $Z_{target} - 1$. Hence, whereas (strongly interacting) neutrons will pass some decimeters of solid rock or metals at best until reacting, muons will get about 100 times as far, quite similar to the ratio between low-energy-limit coupling constants of strong and electromagnetic interactions. Its similarity and far larger mass than the electron means a negative[2] muon can form chemical bonds among atoms or monocations much like an electron does (Vesman 1967) but equilibrium state will be at far shorter internuclear distances and higher bond dissociation energies – until the muon decays or reacts with one of the nuclei, then there will be strong repulsion to tear the molecule apart at this site.

Upon capture by a proton within some nucleus, muons can

a) penetrate some 100 m of soil or rock before being absorbed (given their kinetic energy is large enough to survive such a range due to time dilatation without decaying before [τ*c ≈ 660 m], for details see below) and
b) μ^- will react with atomic nuclei causing mainly secondary emission of mostly 1 neutron (see below) producing radionuclides in many cases. If $Z_{target} > 88$ fission becomes possible (cp. Mamedov & Grebinnik 1999). Besides of weak interaction, there is an additional pathway into fission by muons, namely "photofission" induced by radiation-less excitation of a nucleus when a muon gets "downward" from 2p or 3s, 3p, 3d states to the 1s level, thereby releasing > 15 MeV for Z > 81 while fission barriers are $\varepsilon_{fiss.} < 7$ MeV for all isotopes Ra and heavier elements. Here the muon keeps bound to the heavier fission fragment by almost 100% until it decays or gets eventually captured.

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[2] the positive (anti-)muon μ^+ will capture an electron to produce some light hydrogen isotope the spin resonance of which can be measured (μ-SR) in the compounds "muonium" will form before μ^+ does undergo decay

Another result of this process of weak interaction, like with [β^+mp] or [β^-xn] (m = 1 or 2, x = 1 – 3) processes, is **delayed nucleon emission**. While in (subsequent to) β decay this does occur only if $T_{1/2}$ < 2 min, this is much more common in muon (μ^-) capture as substantially more energy (E ≈ 100 MeV except for neutrino losses) is released in the latter process. Average neutron yield upon negative muon capture is about $0.03*A^{⅓}$ [Huff & Tombrello 1974] where A denotes the atomic mass of the nucleus. While the neutrons are not directly measured this secondary (delayed) emission influences the yield of radioactive products which can be detected as simple muon capture by weak interaction translates into transforming an (usually fairly abundant) even-even nuclide into an odd-odd daughter which is always radioactive except for the lightest representatives of this series and a few very long-lived nuclides like ^{50}V, ^{176}Lu, or ^{180}Ta which are very rare in addition. On the contrary, odd-even nuclides tend to be stable so that delayed 1-neutron emission means production of non-active isotones. Accordingly most common isotopes of the most abundant elements like O, Mg, Cl, Ca, Ti, Mn, Fe (A ≈ 50 for the latter five, that is, 1n-path dominating) will produce but modest yields of radioactive daughters (e.g. $^{46\text{-}49}$Sc [half-lives between 84 days and about one hour], $^{52;53}$V [a few min], or ^{57}Mn [90 s]) which are sufficiently short-lived for sensitive detection even though the actual distribution is fairly broad. E.g., for Si, perceptible activity will be mainly ^{28,29}Al from the two heavy silicon isotopes which add up to some 7% of the natural mixture. The only exceptions among the more common Z ≤ 30 elements in soil are Ti and Zn which readily produce short-lived radionuclides. Accordingly, muon-capture activation detection thresholds are lower (better, more sensitive) here, e.g. in Ti-containing sands or clays or along the calcite/smithsonite ($ZnCO_3$) series.

Production of muons, beams thereof

Besides of using cosmic radiation muons, they can be readily prepared in fairly strong beams by devices of acceptable size and energy consumption. Corresponding accelerators (i.e., electron microtrons) now can be placed on a truck and thus become mobile and suitable for outdoor (field) use. They operate at some 300 MeV primary energy in rest frame and produce ≥ 10^5 μ/s; table-top dimensions of appropriate closed-loop electron accelerators did appear feasible more then 10 years ago already (Nagamine et al. 2002). While cyclotrons of a few m diameter producing protons of several hundred MeV are available (one of them right at Dubna), and might be transported along a power supply on some lorry, it is much more convenient to produce muons using a wakefield electron accelerator powered by a pulsed very-high power laser. There, focused several J/< 100 fs pulses from a Ti:sapphire laser serve to produce a bunch of relativistic electrons (up to about 1 GeV) which follow the exploding plasma to gain energy if radiation is focused to > 10^{19}W/cm^2. When they hit some solid, π mesons are formed which then decay into muons. A wakefield accelerator can be run in a fast sequence of such pulses producing an almost constant muon beam for our purposes (the repetition frequence of electron bunches can be > 1 Hz while average lifetimes of nuclides produced by μ^- capture are between some 10 s and a few days (tab.1). The entire system is much more compact than a cyclotron (“desktop accelerator”).

This muon flux rate will determine spatial resolution and detection limits along the flux tube defined by jet collimation and distance, allowing for elastic scattering which will broaden the beam somewhat. Nevertheless, even cosmogenic muons are sufficiently parallel and undergo so little elastic scattering that cavities of a few meters size could be detected within a body of rock (here, sandstone) as large as (and, in fact, identical to) Pharaoh Chufu´s large pyramid located at Giza (Egypt). Saturation, and thus a signal which becomes constant over time will occur at irradiation times similar to the half-lives of the produced radionuclides, that is, several minutes for most suitable nuclides (see below) with conventional detection limits of some 10^{-2} to 10^{-4} Bq/sample volume. Just a small percentage of γ radiation will be emitted upward towards the detector window and penetrate ≤ 50 cm of overlying soil to get detected (some 3 - 10%).

Many of the products are short-lived radionuclides which produce intense γ-radiation capable of penetrating several decimeters of superior (overlying) soil themselves. Hence a (negative) muon beam passing through the ground parallel to the surface would be optimal for determining chemical composition of topsoil over an area of some 30,000 m^2 in one session. It just needs a gamma spectrometer moved on the surface while the beam is circulating underneath, being deflected by magnetic means to follow a path just parallel to the soil surface (spontaneous decay of muons will protect the environment and people in farer distance even if due to terrain features the jet will eventually cross the surface from below). The rest-frame flight range of muons would become 100 m at a kinetic energy of some 2 MeV, being an average value during moderation of muons due to all scattering (which is small), diffraction and producing bremsstrahlung (again, much less than with electrons as muons are more massive). The relationship between soil cover/pathway through soil and muon nuclear reaction product yield for different energies is as follows where 1 hg/cm^2 ≈ 45 cm of densely packed soil:

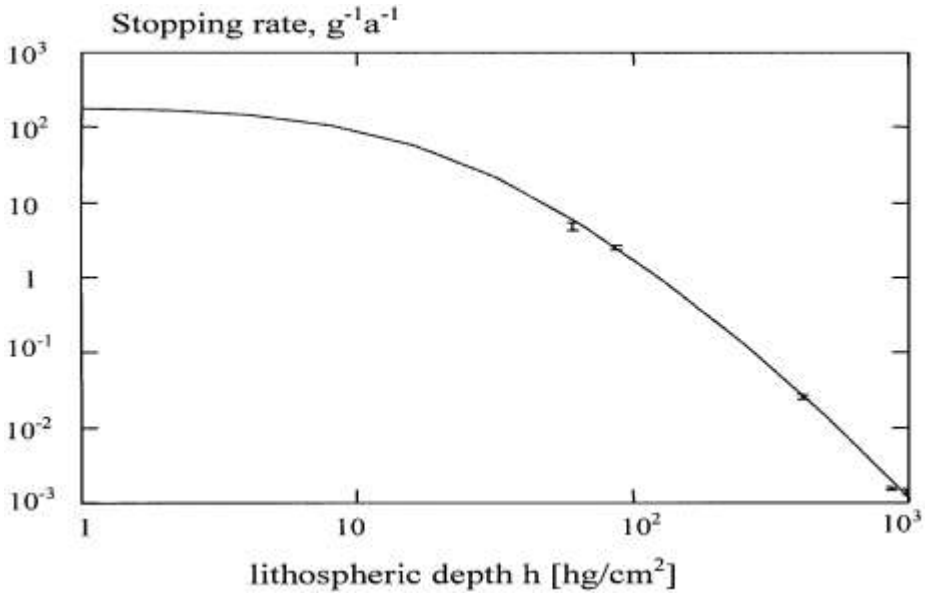

Fig.1: Relative penetration of CR muons into sediment

The rates given here correspond to natural cosmic-ray muon fluxes rather than the products of a local accelerator. It can be seen that for ranges of about 5 m (that is, a measuring circle of some 80 m^2) rates are rather constant whereas just 1% of the original rate would be obtained at a source distance of some 40 m which presumably is the limit of meaningful detection sensitivity; accordingly, the measure area from one starting point would be about the size (but not shape) of a soccer playground (5,000 m^2). If 0.01% would do (for the most common elements which can be activated in this manner) the range would extend to some 250 m or 20 ha (!). Of course, collisions with protons will soon decrease kinetic energy such as the muon nuclear reaction yields close to the source will not be quite the same than in larger distances. Rather than being fully thermalized, most μ^- get captured by atoms or cations at E_{kin} ≈ 40 eV (Vogel et al. 1975). Muon range until decaying at kinetic energies just above those typical for absorption by atoms is about 60 cm.

Muon capture by small molecules (e.g. water, CO_2, N_2, H_2S, CH_4 [gas inclusions, bubbles, cavities in soil], glycine) will probably make it dissociate, producing soft X-rays from the atoms involved. First a given bond will be contracted by the valence muon in two- or three-atom molecules to some small

fraction of the common length of electron-based, then, when the muon either propagates closer to one of the involved nuclei (L or K states) or simply decays, pronounced repulsion would result, dissociating the molecule along the bond where this muon had been located, e.g.

$CO_2 + \mu^- \rightarrow CO^* + [\mu\text{-}O]^-$ [2]

Thus, possibly, additional information might be obtained over various energy ranges from fluorescence of such molecules including water, glycin, other soil organics like humic acids, Ca carbonates, -silicates, -clays (also containing Si, Al, Ti, K etc.) or a salt like NaCl, KSCN. Within some non-elementary (i.e. multielement) molecule (i.e., rather than N_2 or graphite), μ^- capture probabilities are fairly proportional to $\mathbf{Z^{1.15}}$ of the involved elements, implying that hydrogen in water or organic soil components or HCO_3^- is poorly contributing (here, μ^- capture would produce 1 – 2 [with D] neutrons which in turn will give rise to other activation). Thus there

1) will be a "bias" in the secondary (nuclear) reactions, and
2) chemical binding including oxidation states can be inferred to some extent, i.e. sulfate ions will differ in local response from sulfides, carbonates from organic matter and so on, allowing to estimate the state of oxidation of samples beneath the surface (fig.2)

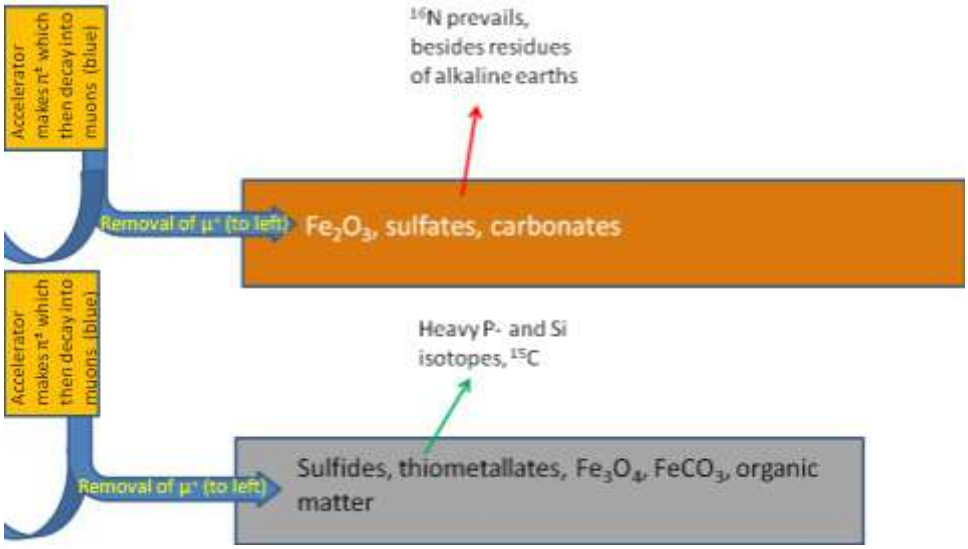

Fig. 2: Radionuclides mainly produced by negative muons penetrating oxidizing (top) and reducing (below) soils, sediments. Thus oxidation state of near-surface sediments can be estimated

In an oxidized soil, O will be the most common anion-forming element while under reducing conditions S will prevail, with some also redox-dependent background by C, Cl[3], N. Accordingly, the absorption will be larger and the spatial range correspondingly shorter (chemical bonds do not matter; a molecule can be treated as a loose association of atoms when μ^- interaction [capture] is concerned). Rather than directing the beam parallel to the surface which requires rather flat terrain, it may proceed perpendicularly along (besides of) some borehole or well or canyon through which the detector is put downward. Then, measurements may extend to some 50 – 100 m below the surface.

[3] the only detection pathway for chlorine would the heavy isotope-no neutron one which yields ^{37}S ($T_{1/2}$ = 5 min); likewise nitrogen (nitrate, amino acids around roots or below moss pads, hydroxamates made by fungi and soil bacteria) would be detected only via ^{15}C formation ($T_{1/2}$ = 2.45 s).

For example, when Fe^{2+} is leached from lignite and ferrous water penetrates next to the surface in small springs and filtration plains on a descending area (producing Fe contents in sediment ranging from about 1.5 to 40% within 10 m distance) while conspicuously red Fe_2O_3 might form at the most Fe-rich sites. When a muon beam is passed through such a plain, local variations in chemistry would readily be spotted by differences in absorption and patterns of produced radionuclides. With somewhat reduced Fe-rich phases (say, magnetite Fe_3O_4 or titanoilmenite), muons might also couple with the local magnetic field produced by them, influencing their rates and modes of capture as the energy levels when they experience ultimate binding to some atom are fairly small. In addition oxidizing and reducing conditions are represented by prevalence of carbonates and sulfates vs. sulfides, thiometallates and organic compounds. In the former, oxidizing case, O does contribute to muon reactions while H in the latter hardly does. The technical key question would be whether recent small accelerators producing muons via $\pi^{\pm}$ mesons can be mobile, e.g. transported on a lorry and located into a hole or natural pit/depression/trench/bed of creek by some crane. On the opposite, gamma detectors would be standard technology (Ge:Li, f.e.). It is required to have the originally formed $\pi^{\pm}$ mesons (τ = 26 ns) converted (decayed) into μ (cp. fig. 2) before the soil sample is hit to avoid other kinds of nuclear reactions like

^{27}Al (π^-, π^+) ^{27}Na ($\tau_{1/2}$ = 0.30 s and 9.5 min for ^{27}Mg; βn pathway is negligible [0.13%] for ^{27}Na).

The γ rays produced during decay of the nuclides produced from the soil isotopes during μ irradiation partly pass to the surface for detection. Secondary quanta could be produced by XFS, additionally, increasing sensitivity of detection but somewhat reducing the depth from which the elementary composition can be determined. First feasibility studies should be done on rather homogeneous binary or ternary samples (e.g. quartz sand [≈ SiO_2] at a beach, pig iron lagerstätten, rock salt layers [irradiating parallel to existing cavities or along waste salt pile cliffs]) which extend over dozens of meters, measuring activation of principal compounds (Si, Fe, Na, Cl, and O, respectively, making ^{16}N, $^{28\text{-}30}Al$, ^{23}Ne, ^{37}S, and $^{57;58}Mn$) and looking for other activities not attributable to these only then.

Fission rates/yields by muon (i.e., μ^-) capture along either pathway are of % orders (Olanders et al. 1980 for uranium) and thus substantially higher than those for **electron**-capture-induced fission (εdf) in e.g. ^{228}Np or ^{232}Am which make up about 0.1% of decays in these nuclides (often, much less while the highest reported εdf branching yield is about 0.7% with ^{242}Es). Of course the electromagnetic process by radiationless nuclear excitation is the faster one but can release just some 15 MeV while the larger fission barriers of nuclei with Z << 88 can only be overcome by using most of the muon´s rest mass, that is, via the weak-interaction channel converting the muon into ν_μ. This is important in detection and determination of elements heavier than latest REEs Yb or Lu: fission products are less specific than those of simple μ^- capture but may take up another negative charge after electromagnetic fission as the muon is most likely to be retained by one of fission fragments (commonly, the heavier one) and will eventually react with it (rather than simply decay). Electromagnetically induced fission commonly prevails over that induced by weak interaction particularly in distinctly non-spherical nuclei.

Tab.1: radionuclides produced by negative muon irradiation of various elements. For γ energies, see the nuclide chart (e.g., Seelmann-Eggebert et al. 1981).

Element	Short-lived isotopes of (Z – 1); half-life	Natural abundance of M	β^+ emission, nuclide and relative yield/ branching ratio (→ 2 or 3 e^+/e^- annihilation γ of typical energy, same half-life)
C	^{12}B, 20 ms	98.9%	
N	^{11}C, 20.4 min ^{15}C, 2.45 s	0.366 % (15)	
O	^{16}N, 7.1 s	99.75%	
Na	^{23}Ne, 37 s	100 %	
Mg	^{25}Na, 59 s; ^{26}Na, 1.08 s	^{25}Mg 10.3 %; ^{26}Mg 11%	
Al	^{27}Mg, 9.46 min	100%	
Si	^{28}Al, 2.24 min; ^{29}Al, 6.56 min; ^{30}Al, 3.6 s	$^{28\text{-}30}$Si 100%	
S	^{34}P 12.4 s; ^{36}P 5.6 s		
Cl	^{37}S 5.05 s	24.6 %	
K	^{41}Ar 110 min	7 %	
Ca	^{44}K 22.1 min; ^{48}K 6.8 s	^{44}Ca 2%, ^{48}Ca 0.2%	
Ti	^{49}Sc 57 min; ^{50}Sc 1.71 min		
Mn	^{55}Cr 3.50 min		
Fe	^{57}Mn 85 s; ^{58}Mn 3.0 s		
Cu	^{65}Ni 2.52 h		
Zn	^{64}Cu 12.7 h; ^{66}Cu 5.1 min		
As	^{75}Ge 83 min; ^{75m}Ge 47.7 s		
Rb	^{87}Kr 76 min		
Sr	^{86m}Rb 61 s; ^{88}Rb 17.8 min		
Y	No useful nuclide formed (^{88}Sr is stable, ^{89}Sr too long-lived [50 d, no γ])		
Zr	^{91m}Zr 49.7 min; ^{92}Y 3.54 h; ^{93}Y 10.2 h; ^{94}Y 18.7 min; ^{95}Y 10.3 min; ^{96}Y 5.34 s		
Mo	^{94m}Nb 6.26 min; ^{97}Nb 72 min; ^{99}Nb 15 s; ^{99m}Nb 2.6 min; ^{100}Nb 1.5 s; ^{100m}Nb 2.99 s		
Sn	^{110}In 4.9 h; ^{110m}In 69 min; ^{111m}In 7.7 min; ^{114}In 72 s; ^{115m}In 4.49 h; ^{116}In 14.1 s; ^{117}In 43 min; ^{118}In 5 s; $^{118m´}$In 8.5 s; ^{119}In 2.4 min; ^{120}In 3.1 s; ^{121}In 23.1 s; ^{122}In 1.5 s; ^{123}In 6.0 s; ^{123m}In 47.8 s; ^{124}In 3.1 s; ^{124m}In 3.7 s		
I	^{127}Te 9.35 h		

Ba	^{129}Cs 32.06 h; ^{130}Cs 29.2 min; ^{134m}Cs 2.9 h; ^{135m}Cs 53 min; ^{136m}Cs 19 s; ^{138}Cs 33.4 min; ^{138m}Cs 2.9 min; ^{139}Cs 9.27 min; ^{140}Cs 63.7 s		Almost purely β^+ in $^{129;130}$Cs (also $^{131;132}$Cs but too long-lived for efficient detection)
La	^{139}Ba 83 min		
Ce			
Nd			
Eu[4]	^{153}Sm 46.3 h		
Yb			
Au	^{197}Pt 19.9 h; ^{197m}Pt 95.4 min		Plus fission products
Hg			Plus fission products
Tl			Plus fission products
Pb	^{206}Tl 4.20 min ^{206m}Tl 3.74 min; ^{207}Tl 4.77 min; ^{207m}Tl 1.33 s; ^{208}Tl 3.05 min		Plus fission products
Th	^{230}Ac 2.03 min; ^{231}Ac 7.5 min		Plus fission products
U	$^{236;237}$Pa some 9 min each		Plus fission products

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[4] Mobilities of europium in soil and biomass and stable levels in seawater sensitively depend on oxidation state (+II or +III) and thus on redox or (in plant leaves etc.) photoredox processes (Fränzle 2010); hence its distribution (or that of μ^- capture product ^{153}Sm) gives additional information besides the above-mentioned relative activities