Radioactive nuclides and decay

O.Cremonesi
INFN - Sez. Milano Bicocca

SUMMER INSTITUTE: USING PARTICLE PHYSICS TO UNDERSTAND AND IMAGE THE EARTH

JULY 14, 2014 - GSSI - L'AQUILA, ITALY

Nuclear decays

The atomic nucleus

- The symbol of an atomic nucleus is

$$ {}^{A}_{Z}X_{N} $$

where
  - Z = atomic number (number of protons)
  - N = neutron number (number of neutrons)
  - A = mass number (Z + N)
  - X = chemical element symbol

- Each nuclear species with a given Z and A is called a **nuclide**.
- Z characterizes a chemical element.
- The dependence of the chemical properties on N is negligible.

- Nuclides with the same neutron number are called *isotones* and the same value of A are called *isobars*.
- The nuclear charge is $+e$ times the number (Z) of protons.

- Hydrogen's **isotopes**:
  - **Deuterium**: Heavy hydrogen. Has a neutron as well as a proton in its nucleus.
  - **Tritium**: Has two neutrons and one proton.

- The nuclei of the deuterium and tritium atoms are called *deuterons* and *tritons*.
- Atoms with the same Z, but different mass number A, are called **isotopes**.

Nuclear Stability

- The **binding energy of any nucleus** ${}^{A}X$ is the energy required to separate the nucleus into free neutrons and protons

$$B\left({}^{A}_{Z}X\right)=\left[Nm_n+ZM\left({}^{1}\mathrm{H}\right)-M\left({}^{A}_{Z}X\right)\right]c^2$$

- The **binding energy of a nucleus against dissociation** into any other possible combination of nucleons (e.g. R and S) is

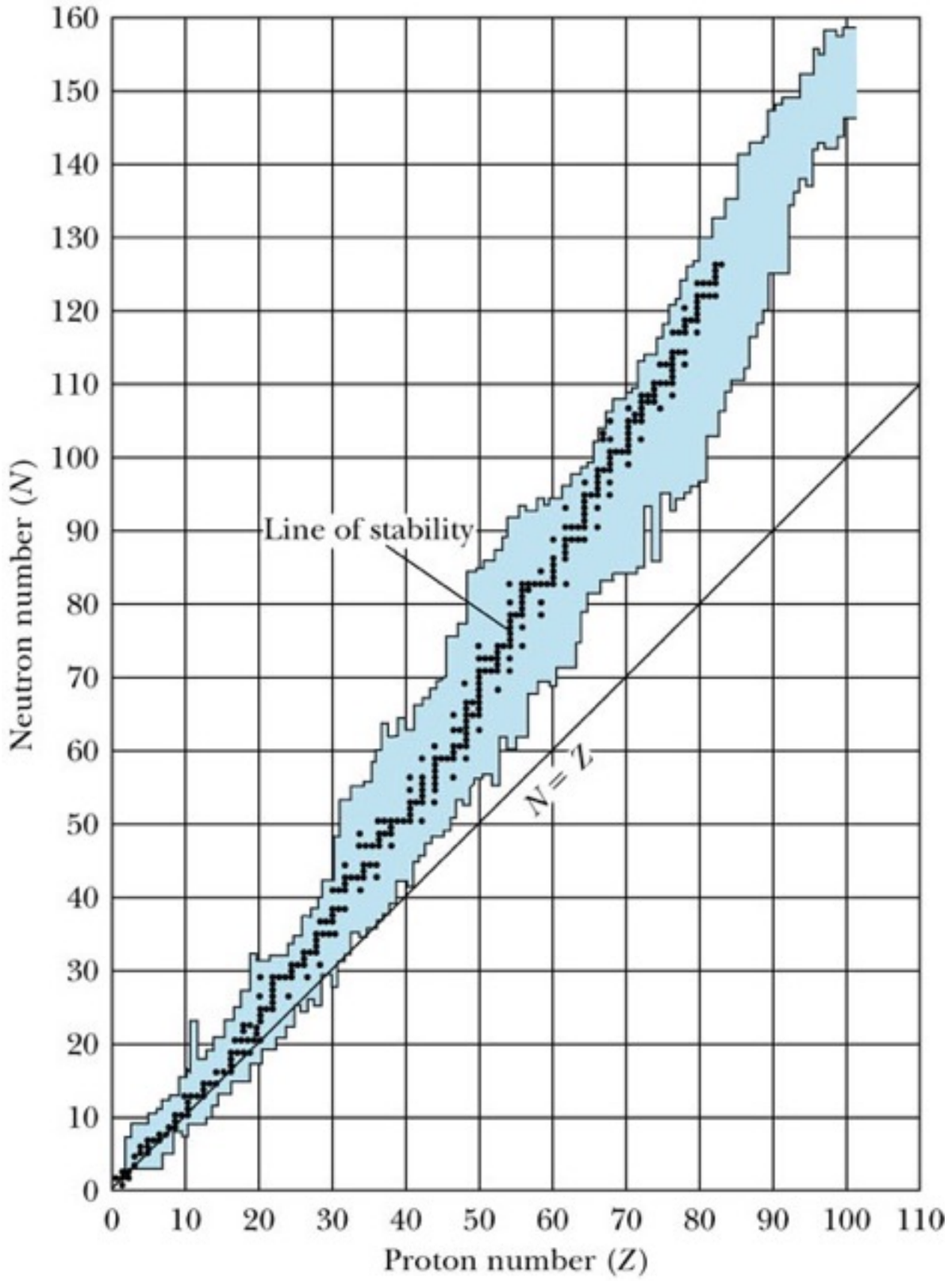
$$B=\left[M(R)+M(S)-M\left({}^{A}_{Z}X\right)\right]c^2$$

- **B<0 → unstable nucleus**

- **Proton (or neutron) separation energy:**
  The energy required to remove one proton (or neutron) from a nuclide.

- Nuclides: all stable and unstable nuclei that are long-lived enough to be observed.

- More than 3000 known nuclides. Only 266 are stable



Nuclear Stability

- The line representing the stable nuclides is the **line of stability**.
  - It appears that for $A \leq 40$, nature prefers the number of protons and neutrons in the nucleus to be about the same $Z \approx N$.
  - However, for $A \geq 40$, there is a decided preference for $N > Z$ because the nuclear force is independent of whether the particles are *nn*, *np*, or *pp*.

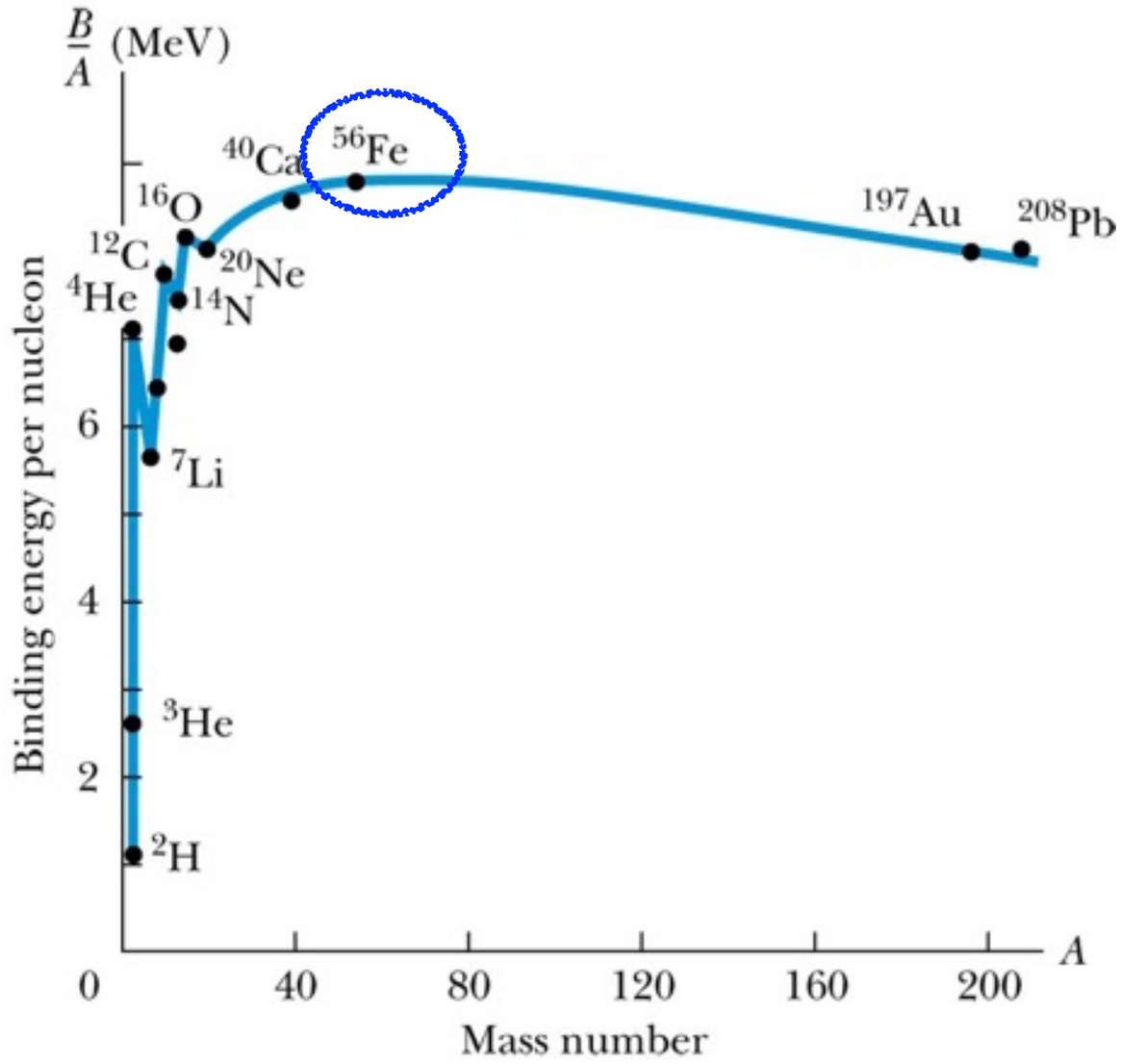
- As the number of protons increases, the Coulomb force between all the protons becomes stronger until it eventually affects the binding significantly.

- Most stable nuclides have both even Z and even N (even-even nuclides), e.g. ^{4}He

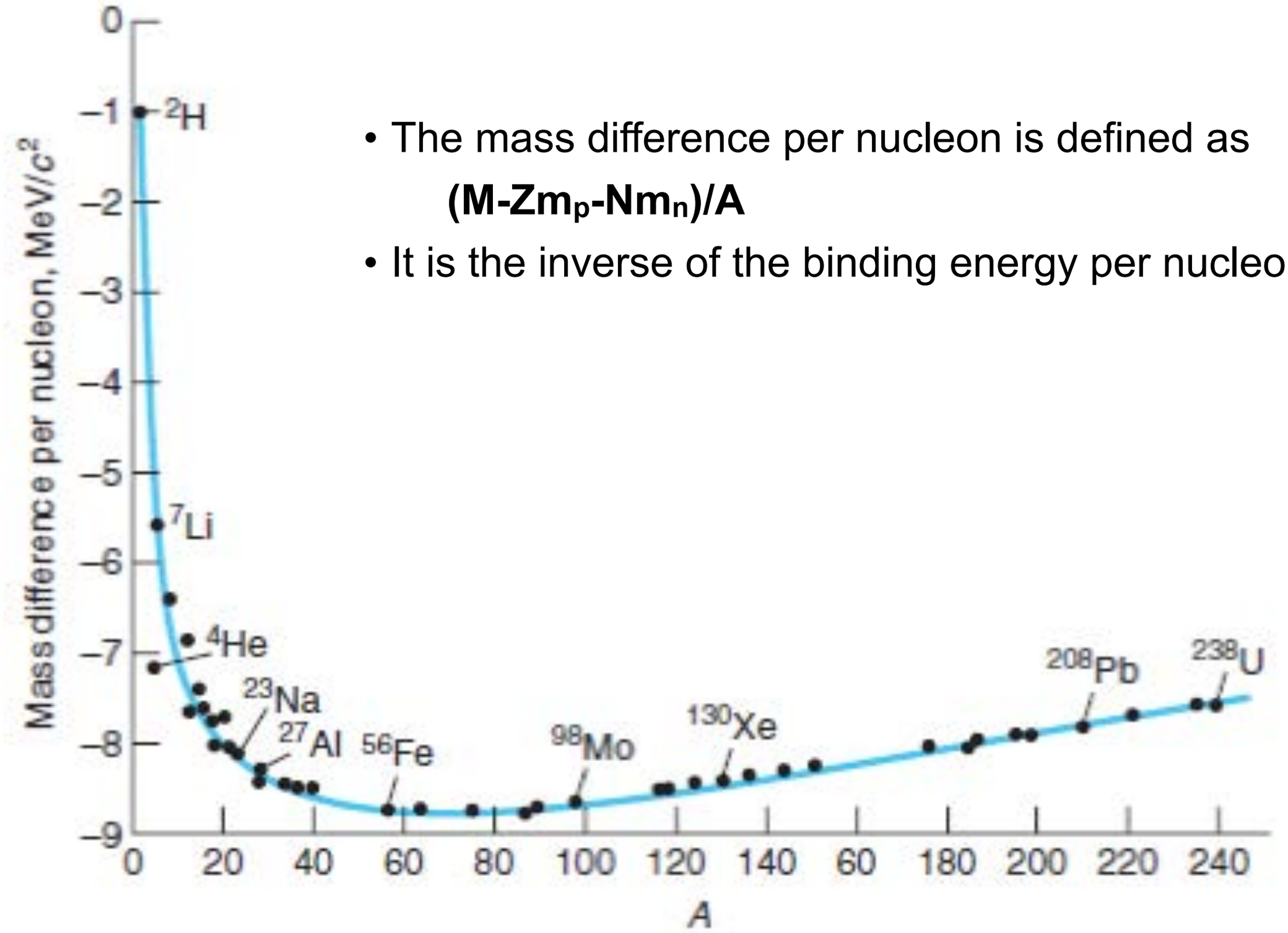
- Only four stable nuclides have odd Z and odd N (odd-odd nuclides): ^{2}H, ^{6}Li, ^{10}B and ^{14}N

Binding Energy per nucleon

- Compares the relative stability of different nuclides.
- It peaks near A = 56.
- The curve increases rapidly, demonstrating the saturation effect of nuclear force.
- Sharp peaks for the even-even nuclides ^{4}He, ^{12}C, and ^{16}O tight bound.



Mass difference



- The mass difference per nucleon is defined as

  $(M-Zm_p-Nm_n)/A$

- It is the inverse of the binding energy per nucleon

Semi-empirical mass formula

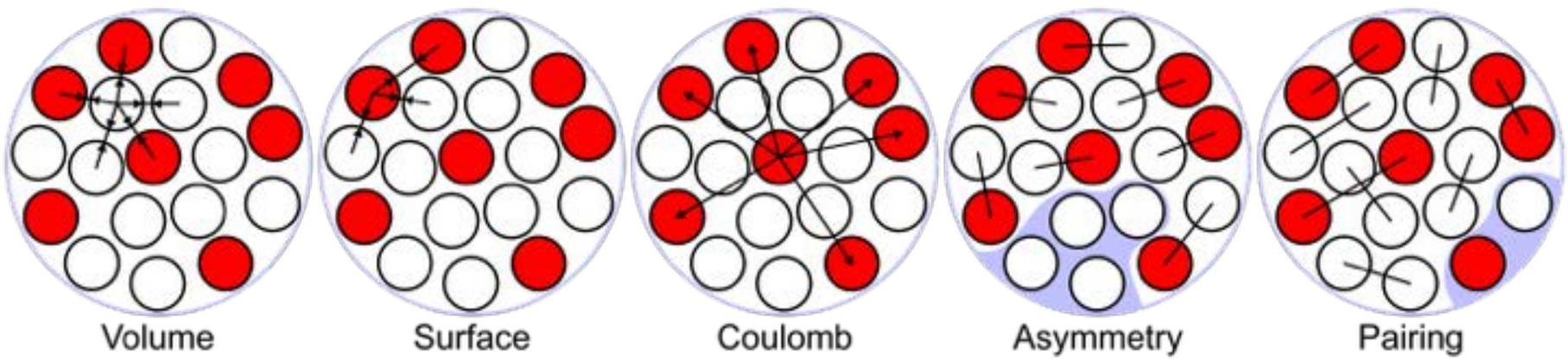
- The Bethe–Weizsäcker formula approximates the mass (and various other properties) of an atomic nucleus from its number of protons and neutrons
- It is based partly on theory and partly on empirical measurements
- The theory is based on the liquid drop model proposed by George Gamow
- The mass of an atomic nucleus is given by

$$m = Zm_p + Nm_n - \frac{E_B}{c^2}$$

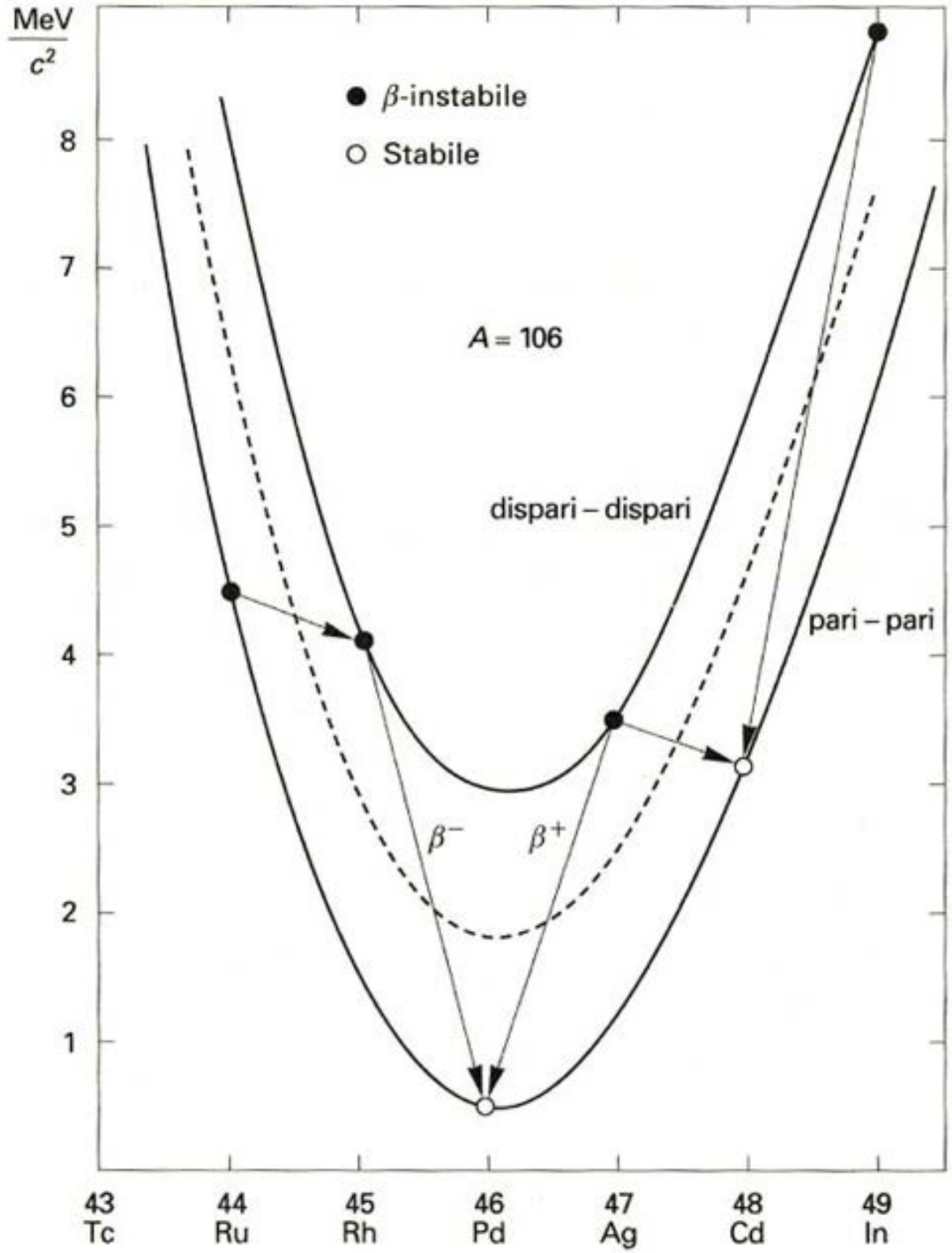
where m_p and m_n are the rest mass of a proton and a neutron, respectively, and E_B is the binding energy of the nucleus.

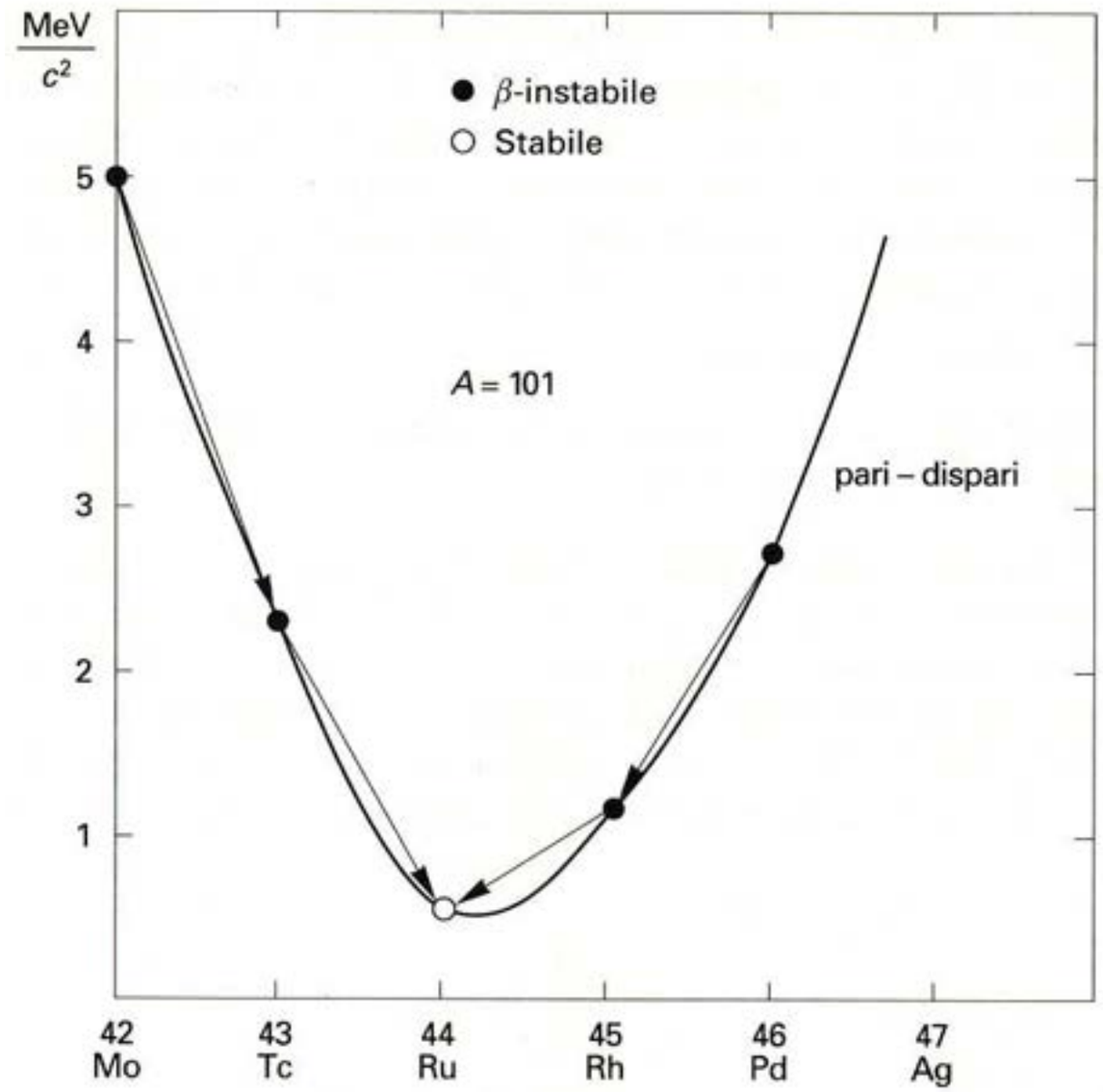
- The semi-empirical mass formula states that the binding energy will take the following form:

$$E_B = a_V A - a_S A^{2/3} - a_C \frac{Z^2}{A^{1/3}} - a_A \frac{(A-2Z)^2}{A} \pm \delta(A,Z)$$



Isobaric curves





Radioactive decay

- When all the relevant conservation laws are observed
  - Mass-energy
  - Linear momentum
  - Angular momentum
  - Electric charge
  - Number of nucleons.

  a nucleus can transform (decay) into a lighter one
- Marie Curie and her husband Pierre discovered polonium and radium in 1898.
  - The simplest decay form is that of a gamma ray, which represents the nucleus changing from an excited state to lower energy state.
  - Other modes of decay include emission of α particles, *β* (– and +) particles, protons, neutrons, and fission.
- The decays per unit time (**activity**) is defined as

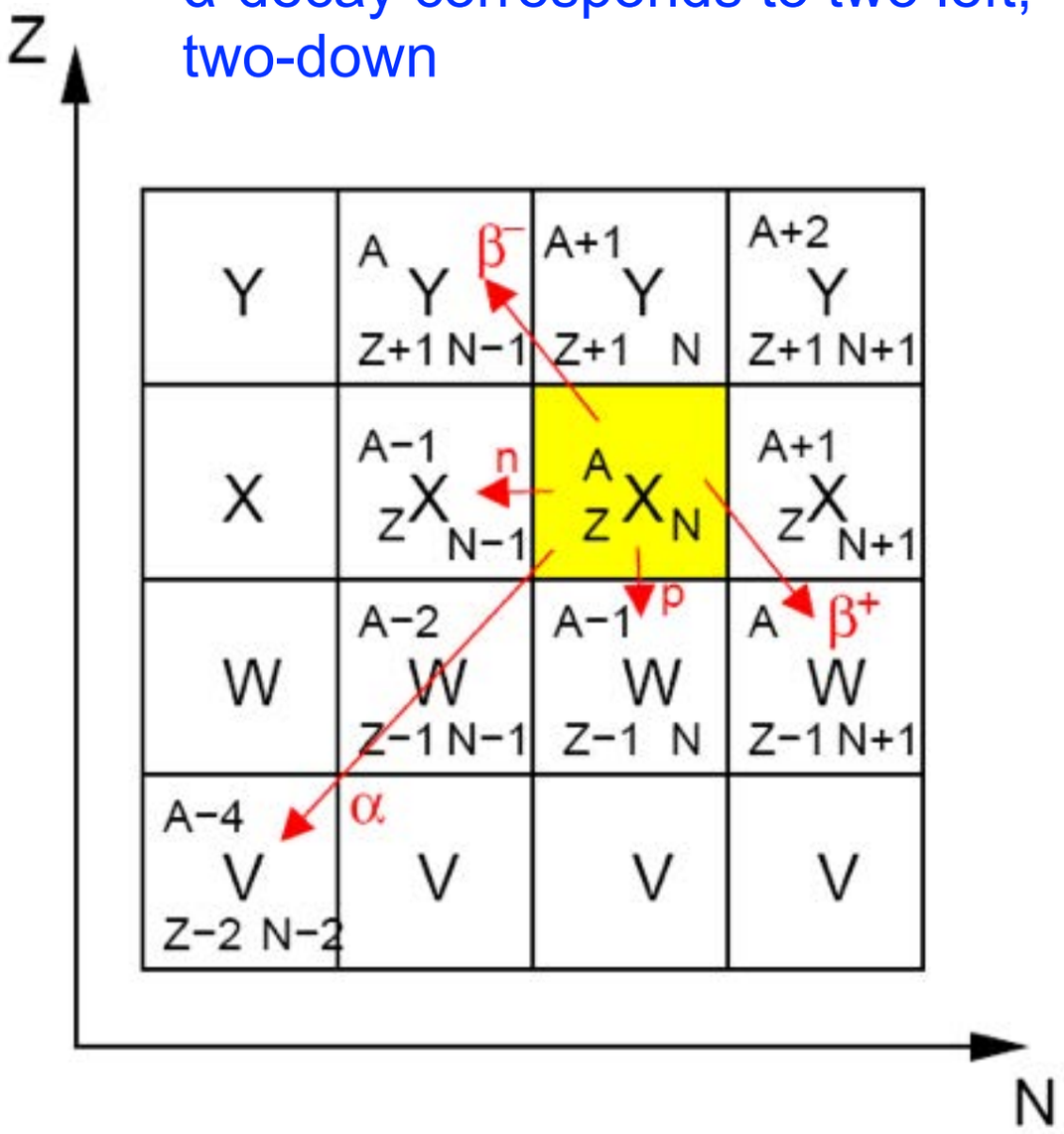
$$\text{Activity} = -\frac{dN}{dt} = R$$

where *dN* / *dt* is negative because total number *N* decreases with time.

Radioactive decay of the nucleus

- There are three main decay mechanism (plus several other less frequent):
  - **α decay:** emission of an α particle results in a smaller nucleus (A-4, Z-2).
  - **β decay:** Two types (plus or minus)
    - A neutron transforms into a proton, an electron and an antineutrino.
    - A protron transforms into a neutron with the emission of a neutrino and a positron.
    - β decay conserves the mass number A
- **γ decay:** Relaxation of the nuclide from an excited state with the emission of a (gamma-)photon.

… the different radioactive decays can easily be connected with movements in the chart, e.g. α-decay corresponds to two left, two-down



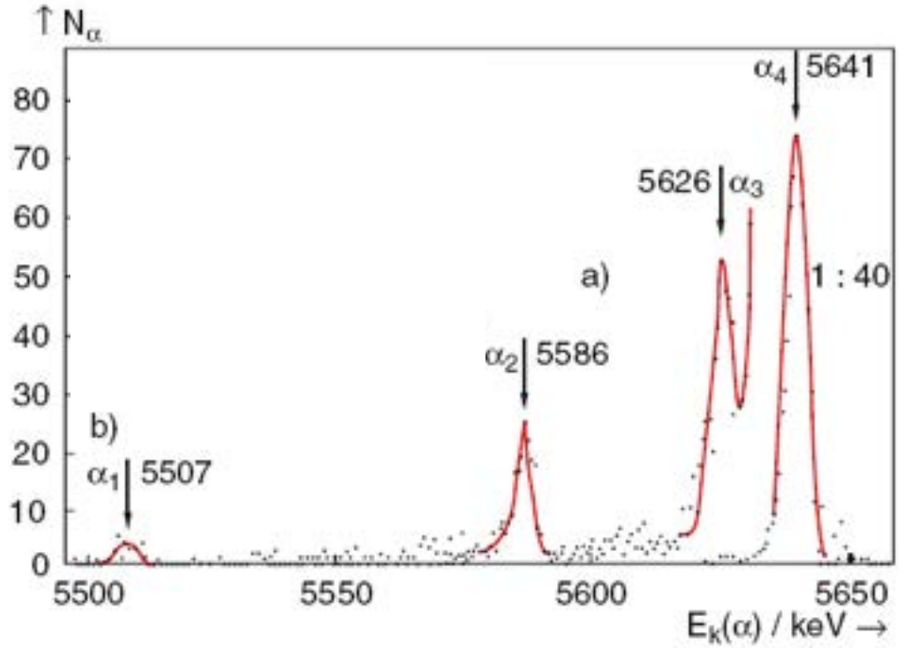
The alpha-decay of the nucleus

- α particles are spontaneously emitted from a nucleus, giving rise to the reaction

$$ {}^{A}_{Z}X \xrightarrow{\alpha} {}^{A-4}_{Z-2}Y $$

- The energy spectrum of the emitted α-particles shows discrete lines (2 body decay)
- The energy of emitted α-particles and "daughter" nuclei follows conservation laws of energy and momentum
- The energy of the emitted α particles is smaller than the electromagnetic energy barrier – short penetration length in materials
- The larger is the penetration R_α of the α particles the higher is the decay rate constant λ

$$ \log \lambda = A + B \cdot \log R_\alpha $$

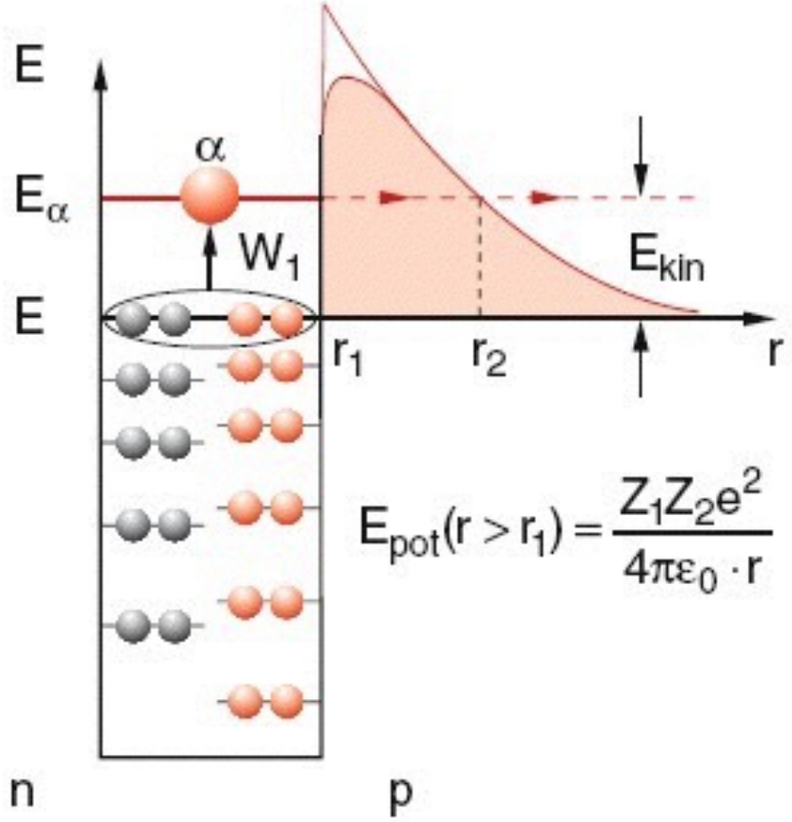


$$ p(M_{A-4}) + p(\alpha) = 0 $$

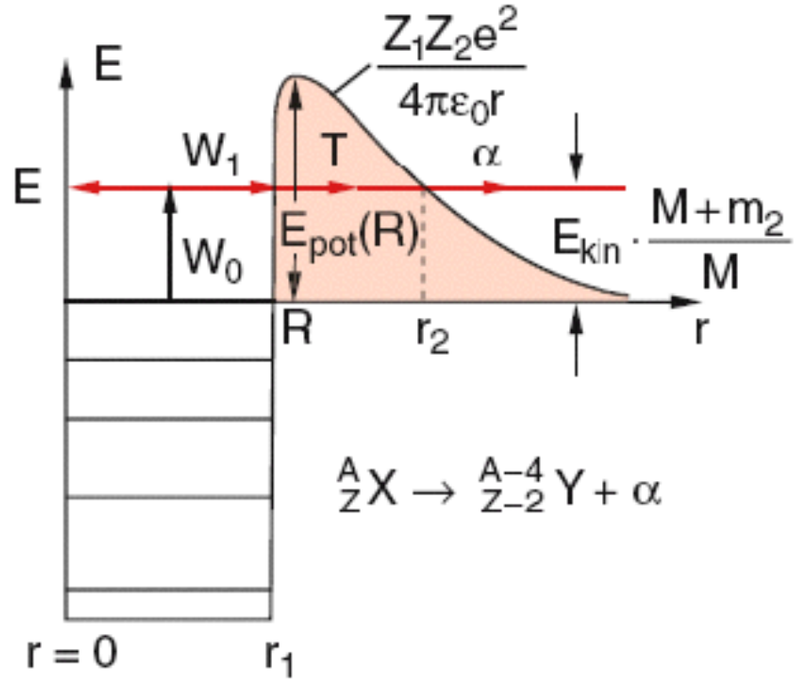
$$ \Rightarrow E_{kin}(\alpha) = E \cdot \frac{M}{M+m} $$

The alpha-decay of the nucleus

- The binding energy pro nucleon of the highest lying nucleons among 5.5 – 6 MeV
- Two neutrons and two protons have a probability of joining in a more stable α-particle
- The larger binding energy of the α-particle (28 MeV; /MeV per nucleon) is compensated by reducing the binding energy of the α-particle to the nucleus, about ~5 MeV above the nucleon levels



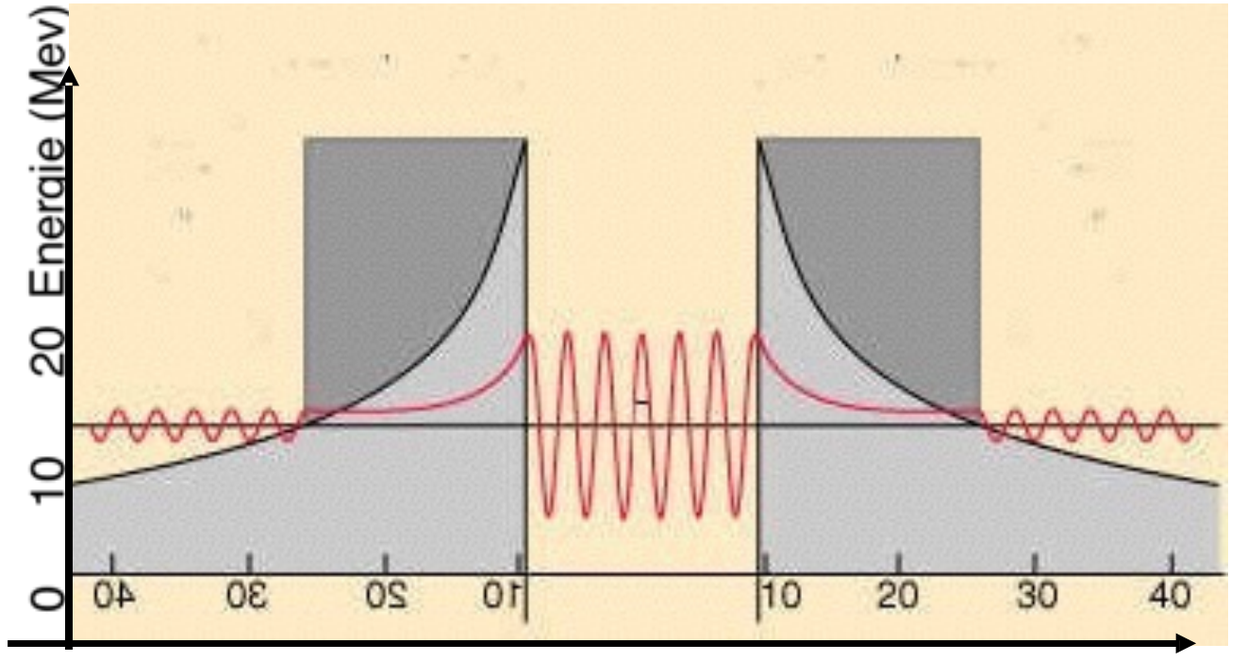
- The lowest binding energy of the α-particle is not enough to escape from the potential well
- The α-particle escape by tunnel effect
- The higher is the energy level of the α-particle, the larger is the decay rate constant



The alpha-decay of the nucleus

The highest is the energy of the α-particle, the lower and the thinner is the tunnel barrier → larger tunneling rate → larger decay rate

Isotop	E_α/MeV	$t_{1/2}$	T_α
$^{212}_{84}$Po	8,78	0,3 µs	$1{,}3 \cdot 10^{-13}$
$^{224}_{88}$Ra	5,7	3,64 d	$5{,}9 \cdot 10^{-26}$
$^{228}_{90}$Th	5,42	1,91 a	$\sim 3 \cdot 10^{-28}$
$^{238}_{94}$Pu	5,5	$8{,}8 \cdot 10^{1}$ a	$\sim 10^{-29}$
$^{230}_{90}$Th	4,68	$7{,}5 \cdot 10^{4}$ a	$\sim 10^{-32}$
$^{235}_{92}$U	4,6	$7{,}1 \cdot 10^{8}$ a	$\sim 10^{-36}$



- Outside of the nucleus, α-particle and remaining nucleus are accelerated by Coulomb repulsion
- The kinetic energy of the system is the difference of masses between initial and final nuclei

$$E = \left[M\left({}^{A}_{Z}\mathrm{X}\right) - M\left({}^{A-4}_{Z-2}\mathrm{Y}\right) - m(\alpha)\right] \cdot c^2$$

and it is distributed following

$$E_{\mathrm{kin}}\left({}^{A-4}_{Z-2}\mathrm{Y}\right) = \frac{m}{M+m} \cdot E \qquad E_{\mathrm{kin}}(\alpha) = \frac{M}{M+m} \cdot E$$

The beta-decay of the nucleus

- **β^- particles (electrons) are spontaneously emitted from a nucleus, resulting in nuclei with higher Z but same A**

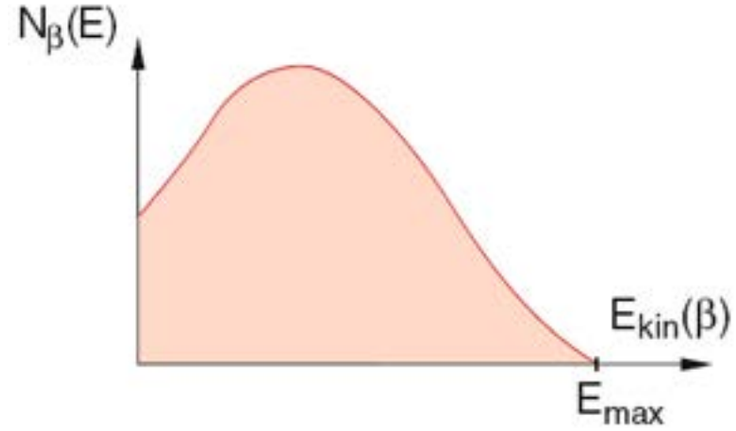
$$^{208}_{81}\mathrm{Tl} \xrightarrow{\beta^-} {}^{208}_{82}\mathrm{Pb}$$

$$^{225}_{88}\mathrm{Ra} \xrightarrow{\beta^-} {}^{225}_{89}\mathrm{Ac}$$

- In other cases positrons (β^+ particles) are emitted, resulting in nuclei with lower Z but same A

$$^{15}_{8}\mathrm{O} \xrightarrow{\beta^+} {}^{15}_{7}\mathrm{N}$$

- The spectrum of the emitted β-particles shows a continuum distribution of energy and momentum
- **The β particles and the "daughter" nuclei do not apparently follow conservation of energy and momentum**
- The maximum energy E_{max} that the β-particles can have do follow conservation of energy



The (continuous) spectrum of ν is complementary to the electron spectrum:

$$E_\nu + E_e \sim Q$$

- An additional particle must be emitted that accounts for energy and momentum conservation: **the neutrino ν**

The beta-decay of the nucleus

There are three types of β-decays occurring spontaneously:

- β⁻-decays

$$n \rightarrow p + e + \bar{\nu} \longrightarrow {}^{A}_{Z}X \rightarrow {}^{A}_{Z+1}Y + e + \bar{\nu}$$

This process may occur in both free neutrons with a life time of 887 s, and in neutrons in nuclei

example $^{101}_{42}Mo \rightarrow ^{101}_{43}Tc + e^- + \bar{\nu}_e$

NEUTRINO and ANTI-NEUTRINO have continuous spectrum

- β⁺-decays

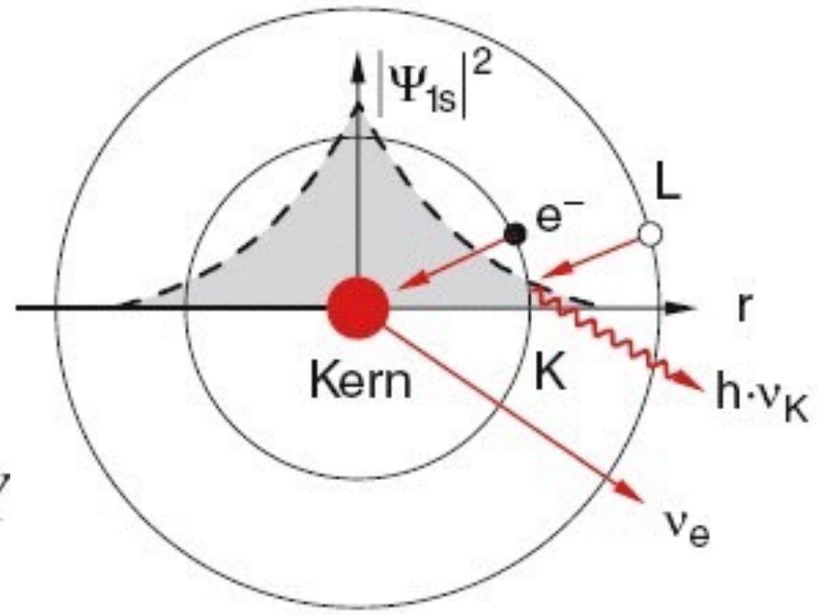
$$p \rightarrow n + e^+ + \nu \longrightarrow {}^{A}_{Z}X \rightarrow {}^{A}_{Z-1}Y + e^+ + \nu$$

Since the proton has a lower mass than the neutron, this process occurs only in protons inside a nucleus (i.e. not for free protons)

example $^{101}_{45}Rh \rightarrow ^{101}_{44}Ru + e^+ + \nu_e$

- Electron capture

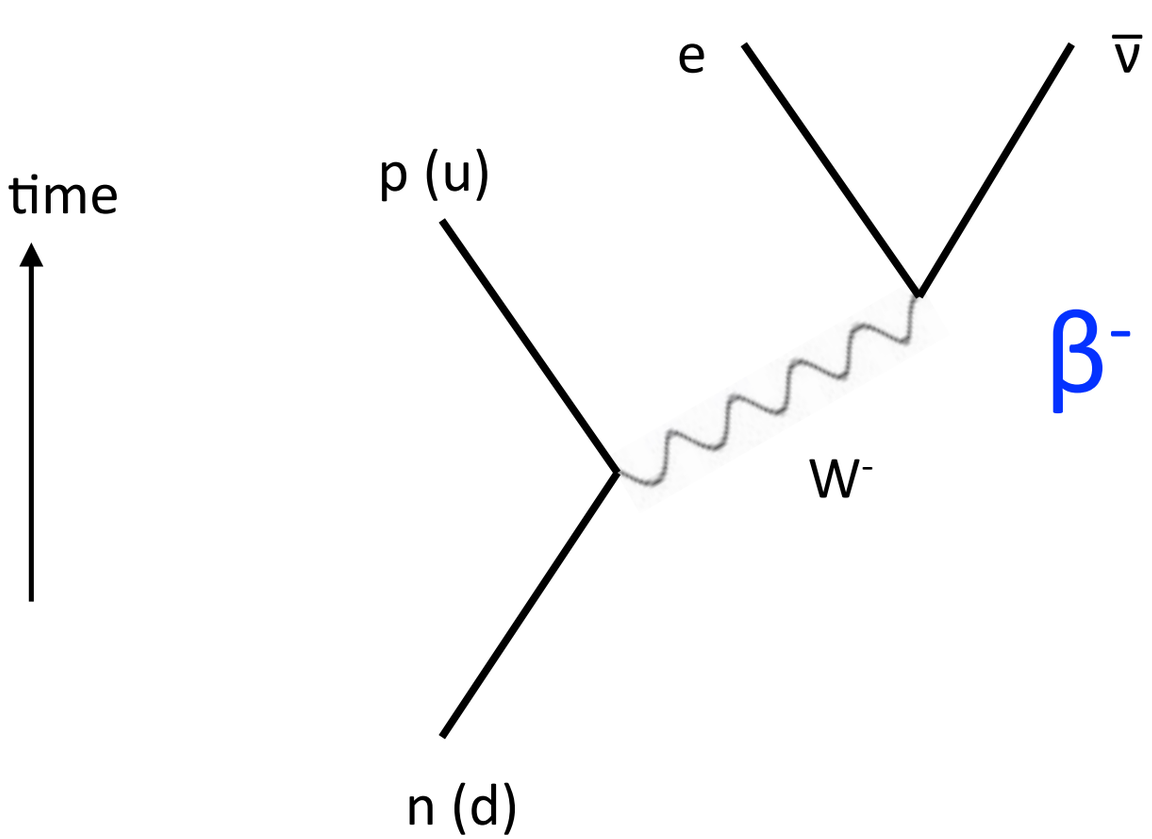
$$ {}^{A}_{Z}X + e \rightarrow {}^{A}_{Z-1}Y \longrightarrow$$

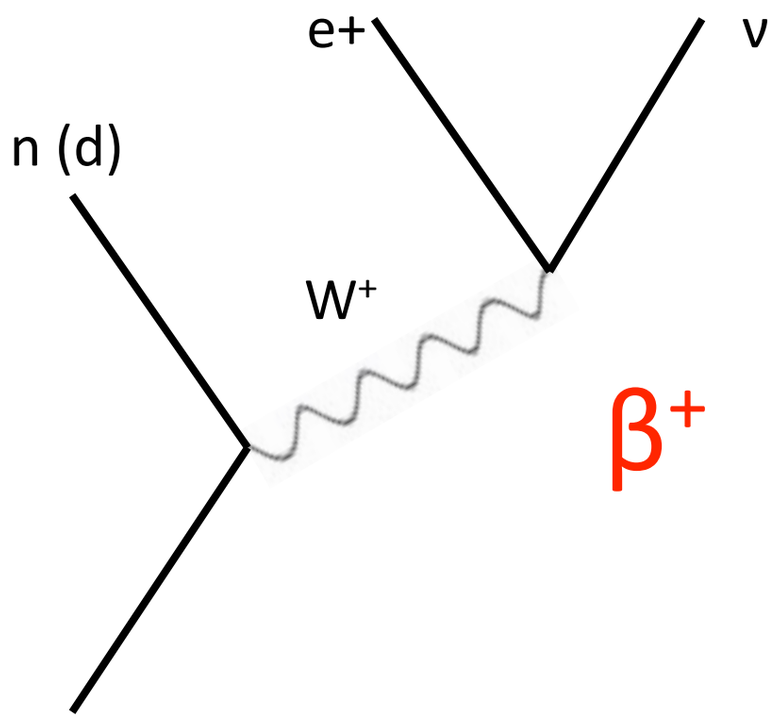


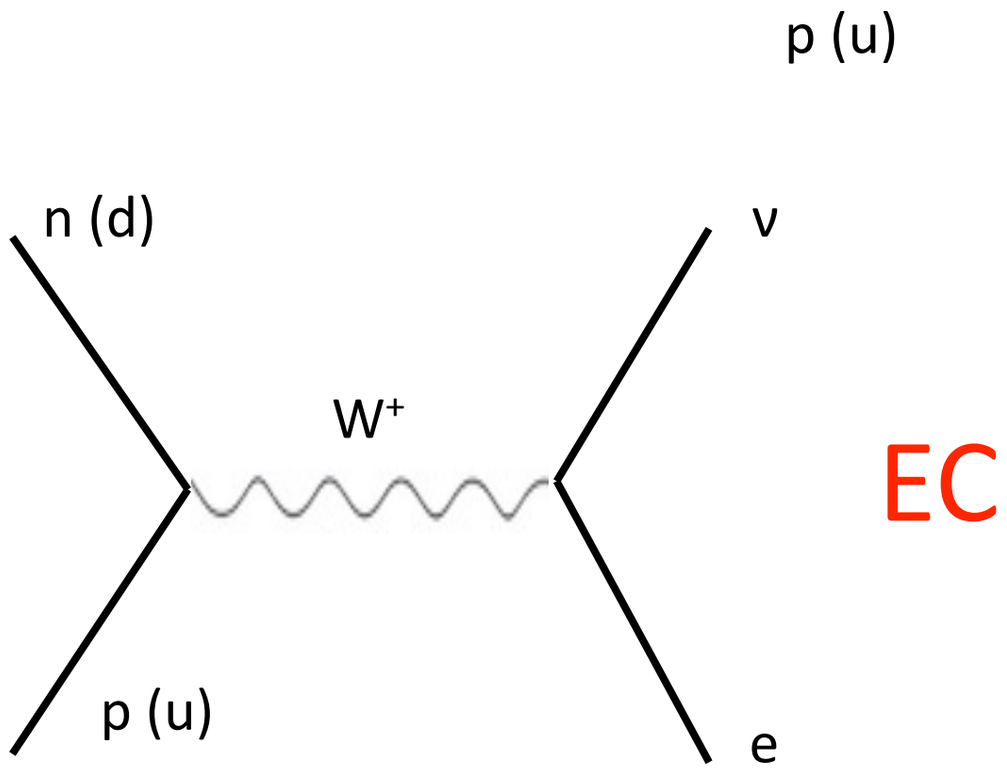
example $^{7}Be + e^- \rightarrow ^{7}Li + \nu_e$

NEUTRINO is monochromatic!

The β family

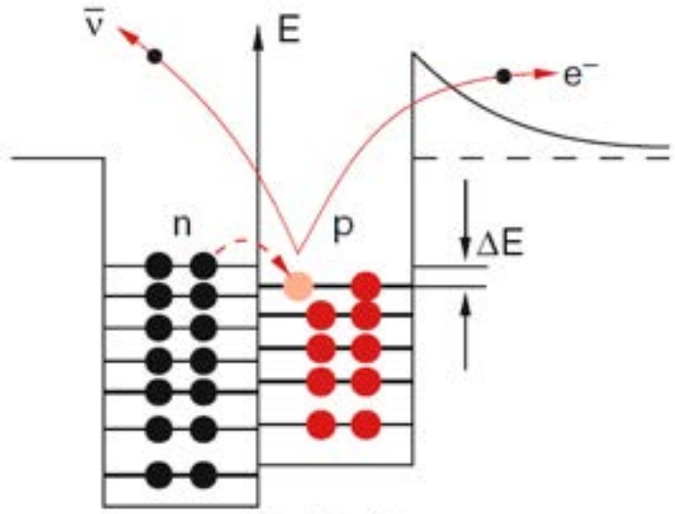




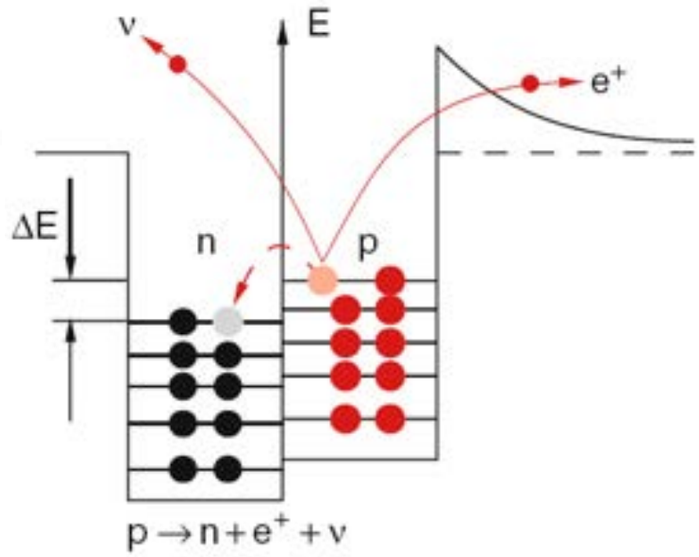


The beta-decay of the nucleus

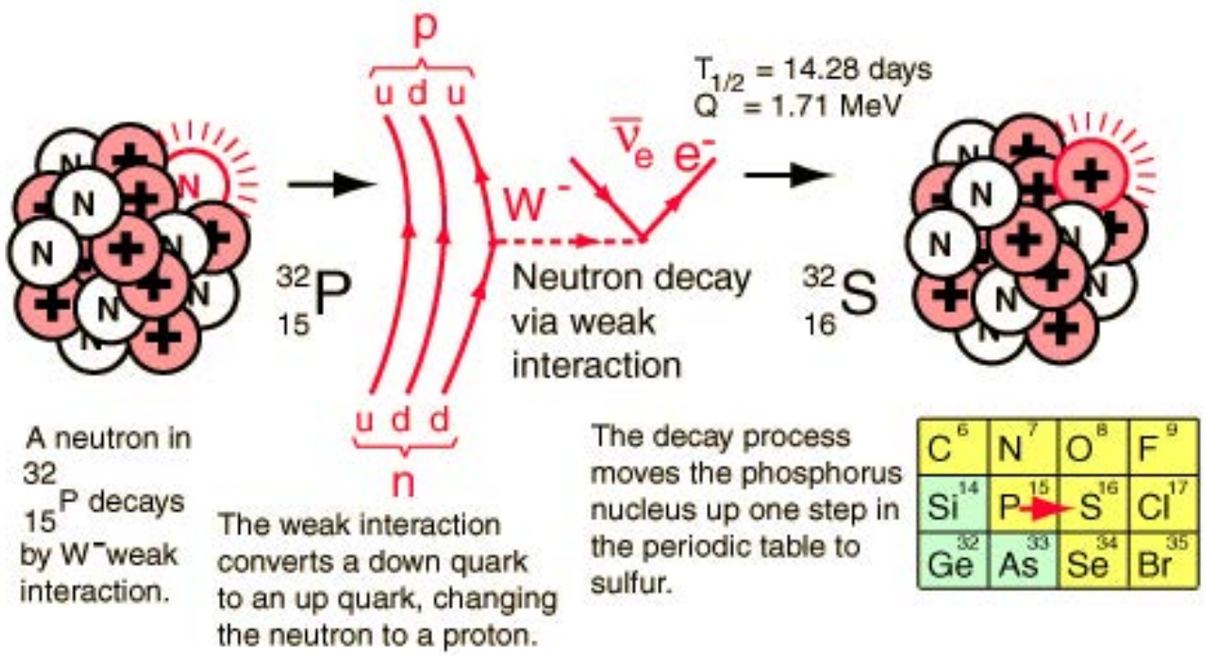
- For β-decay to occur, the product nuclei must have less mass to compensate the mass of the β- particle; the mass of the neutrino is negligible (possible zero)



β⁻-decay



β⁺-decay



- The β-decay also reveals the existence of another interaction in the nucleus, the weak interaction, which accounts for the conversion of a “down” quark into a “up” quark

Nuclear stability upon β decay

Let us consider nuclei with equal mass number A (isobars).
The Weizsäcker-formula can be transformed into

$$M(A,Z) = \alpha A - \beta Z + \gamma Z^2 + \frac{\delta}{A^{1/2}}$$

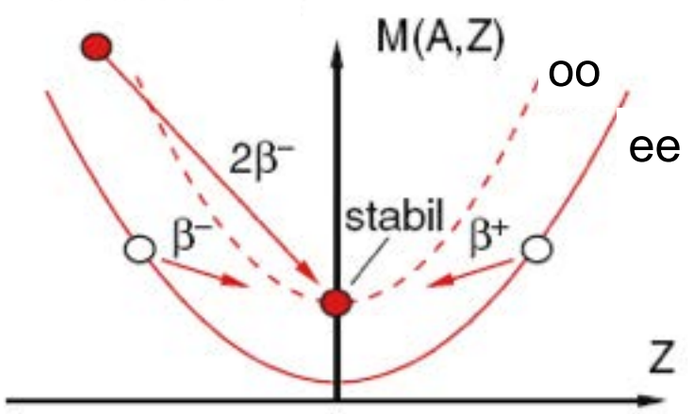
where the coefficients are

$$\begin{aligned}
\alpha &= m_n - a_v + a_s A^{-1/3} + \frac{a_a}{4} \\
\beta &= a_a + (m_n - m_p - m_e) \\
\gamma &= \frac{a_a}{A} + \frac{a_c}{A^{1/3}} \\
\delta &\quad \text{as before (-11.2 or 0 or +11.2 MeV)}
\end{aligned}$$

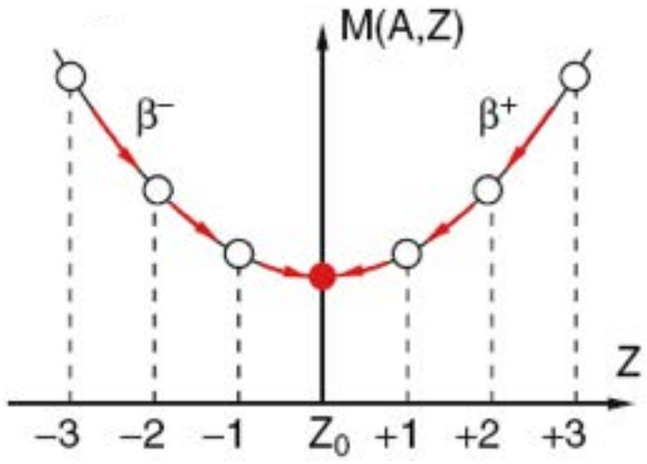
Parabolic behavior respect to Z

Spin-saturation term:
Odd A: One parabola
Even A: Two parabolas, one for (Z,N))=(u,u) and other for (Z,N))=(g,g)

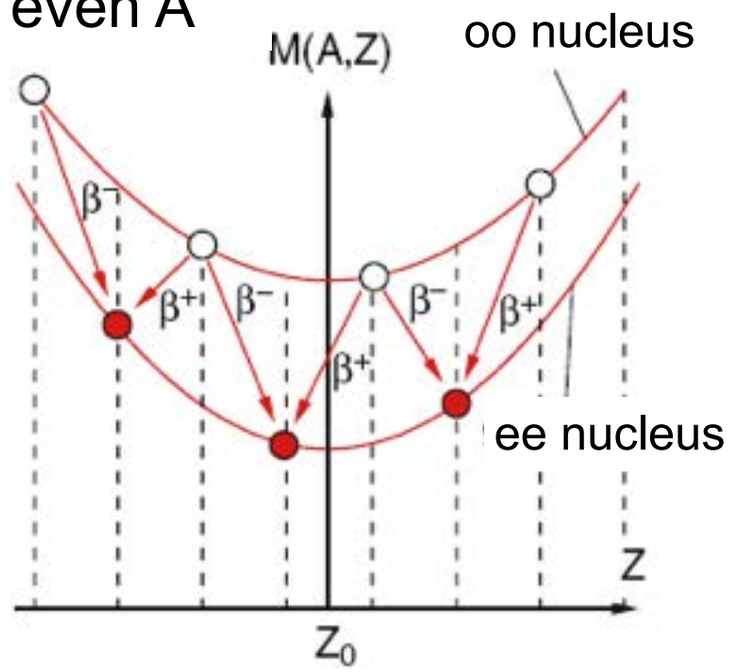
even A



odd A



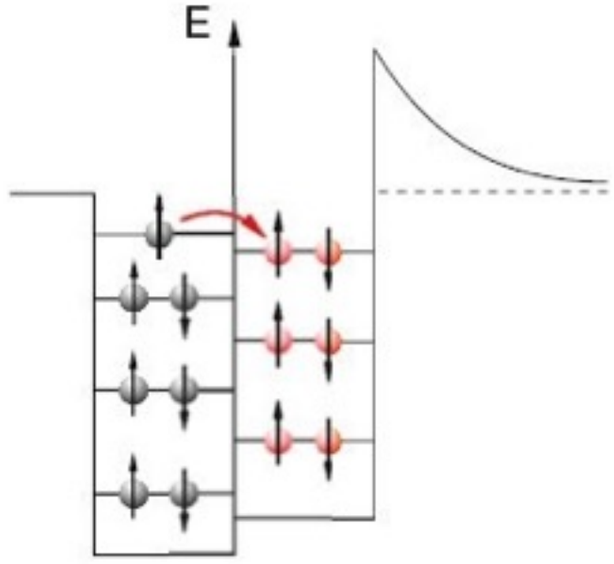
even A



Nuclear stability upon β decay

- The instability of o,o nuclei is due to both, Pauli principle and Coulomb interactions in the nucleus.
- The beta-decay of a proton or a neutron has to relax the nucleus energy-mass an amount

$$\Delta E = E_{\mathrm{f}} - E_{\mathrm{i}}$$
$$= (M_{\mathrm{X}} - M_{\mathrm{Y}} + m_{\mathrm{e}})c^2 + E_{\mathrm{kin}}(\mathrm{e}, \overline{\nu})$$



- The minimum of the parabolas is found by derivation of the Weiszäcker Fromula, at **Z=β/2γ.**
- The nucleus with the smallest mass is stable with respect to β decay

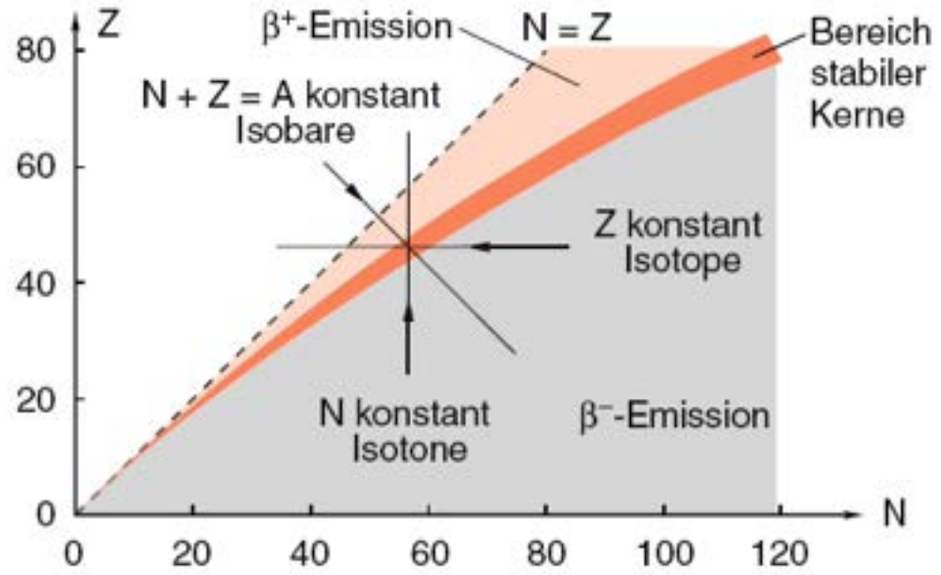


Abb. 3.4. Bereich stabiler Kerne in einem *Z*, *N*-Diagramm. Nach T. Mayer-Kuckuk: *Kernphysik* (Teubner, Stuttgart 1995)

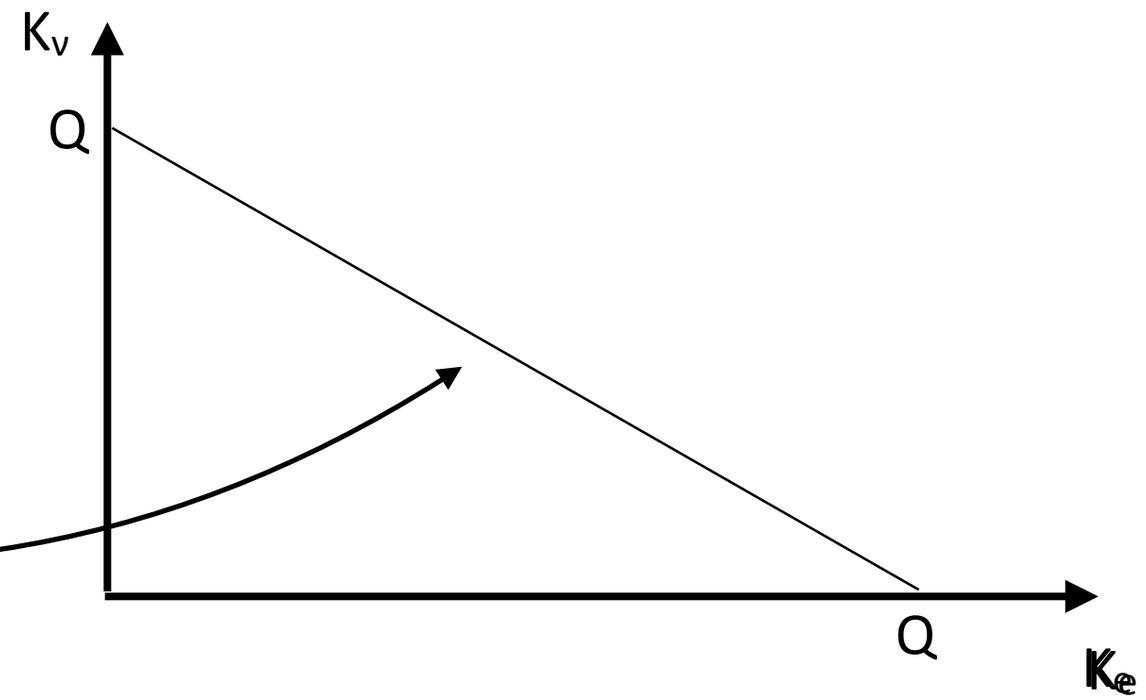
Neutrino spectra

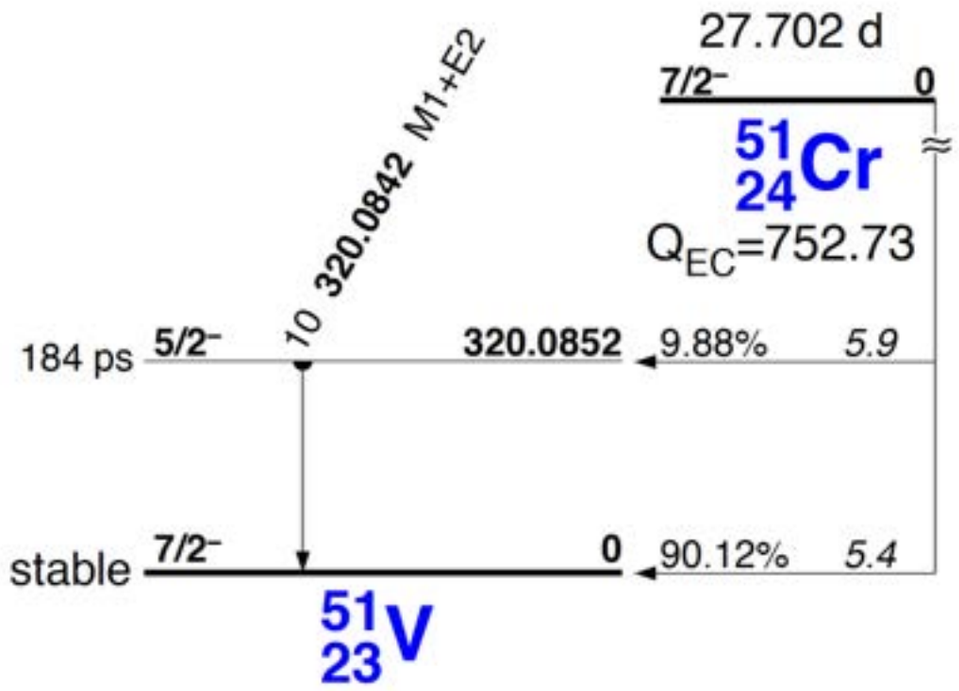
1 **Beta decay:** (A,Z) → (A,Z+1) + e + $\bar{\nu}$

The daughter nucleus accounts for momentum conservation but has negligible kinetic energy

All the available energy is democratically shared by electron and neutrino:

$K_e + K_\nu = Q$





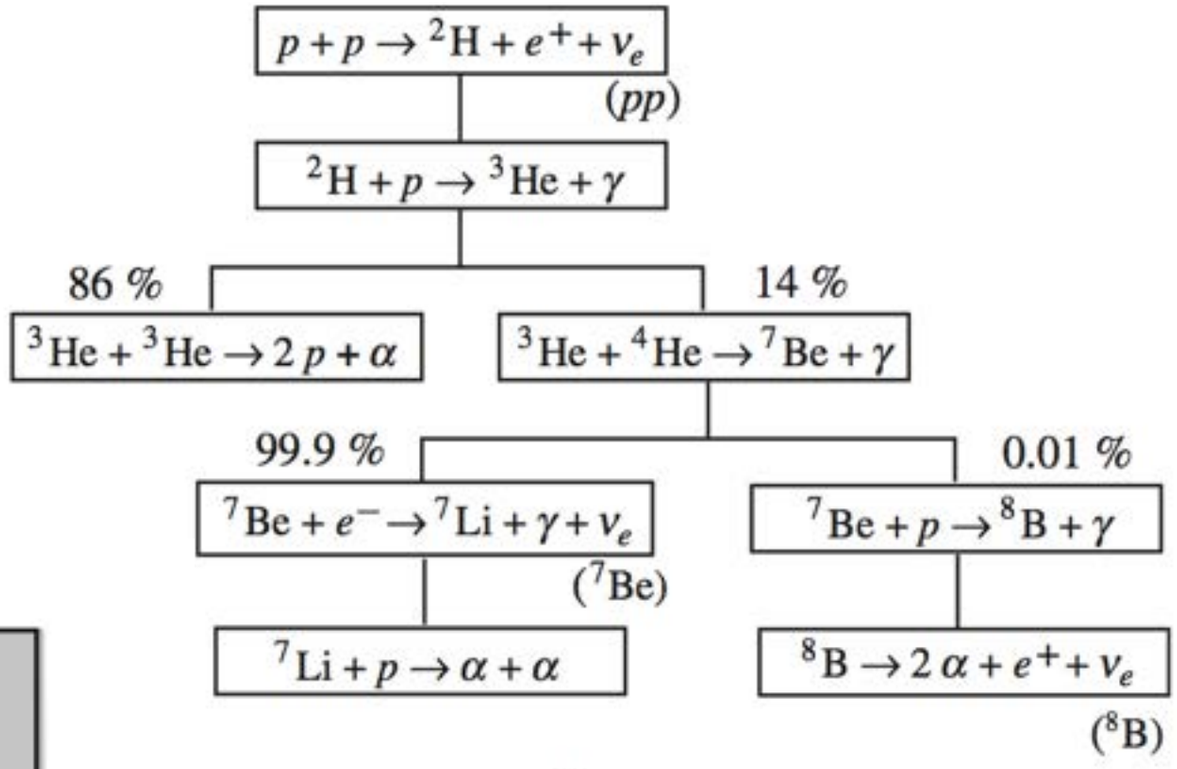
2 **Electron capture:** (A,Z) + e → (A,Z-1) + ν

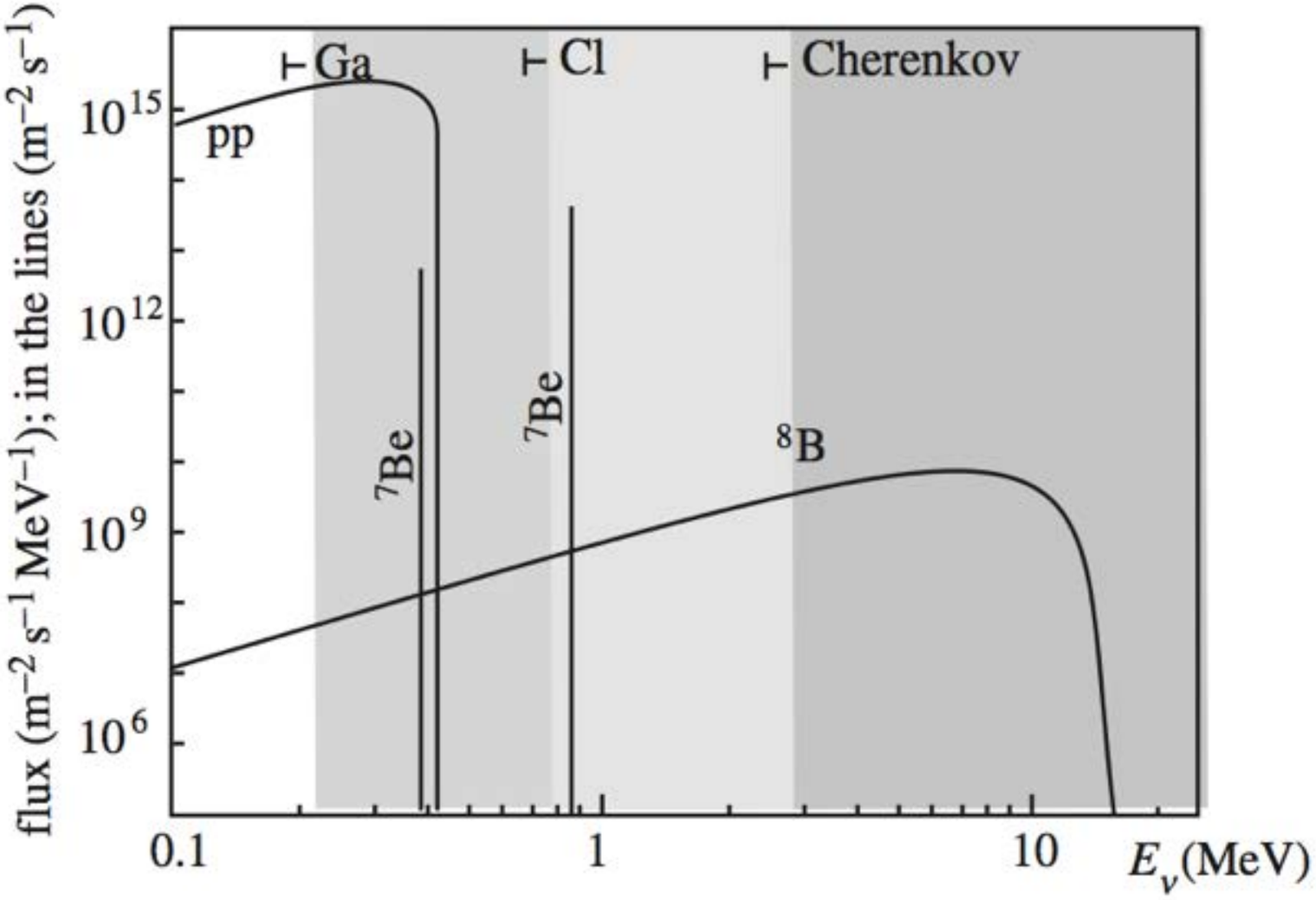
All the available energy (apart from the binding energy of the captured atomic electron) is taken by the neutrino:

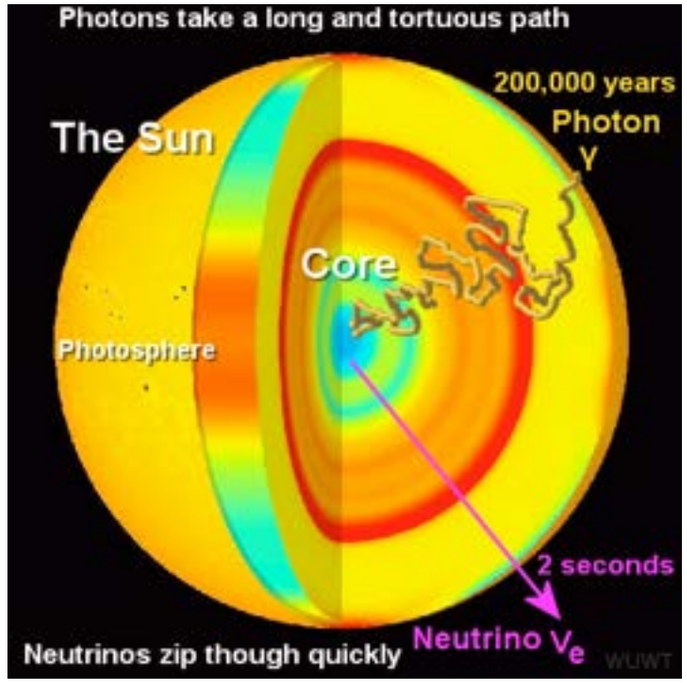
$K_\nu = Q - B$

An example: solar neutrinos

- Solar energy production: the pp cycle



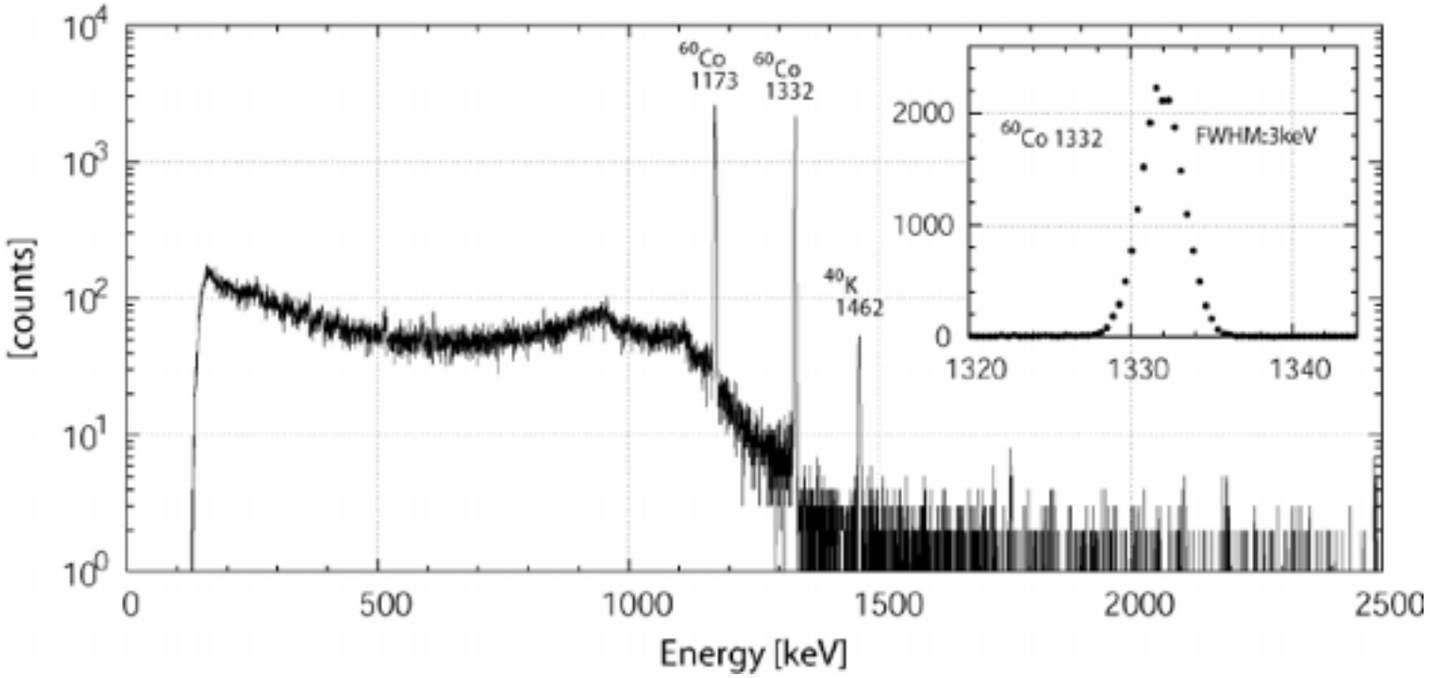


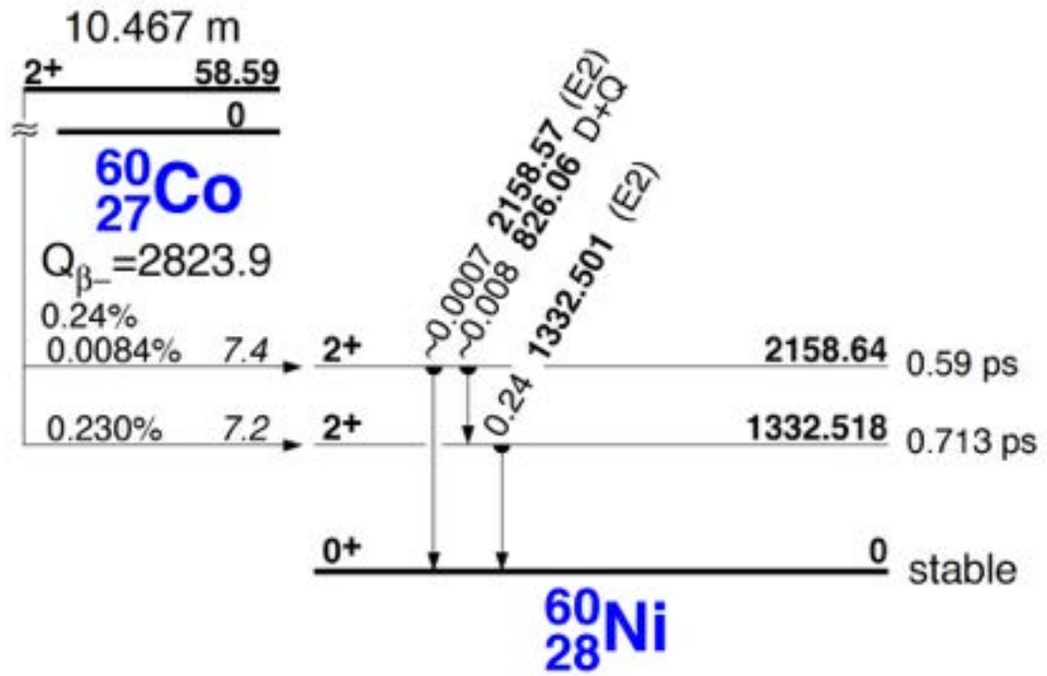


The gamma-decay of the nucleus

- The gamma decay is the de-excitation of an excited nuclear state by emitting electromagnetic radiation (γ-rays).
- It is analogous to the emission of optical radiation or X-rays by the electron shells of excited atoms.
- A and Z of the core remain constant.

- **The γ-rays have a high penetration through matter**
- They are relatively weakly ionizing and not to get distracted by electric or magnetic fields.
- They are emitted as a line spectrum which is characteristic of the emitting core

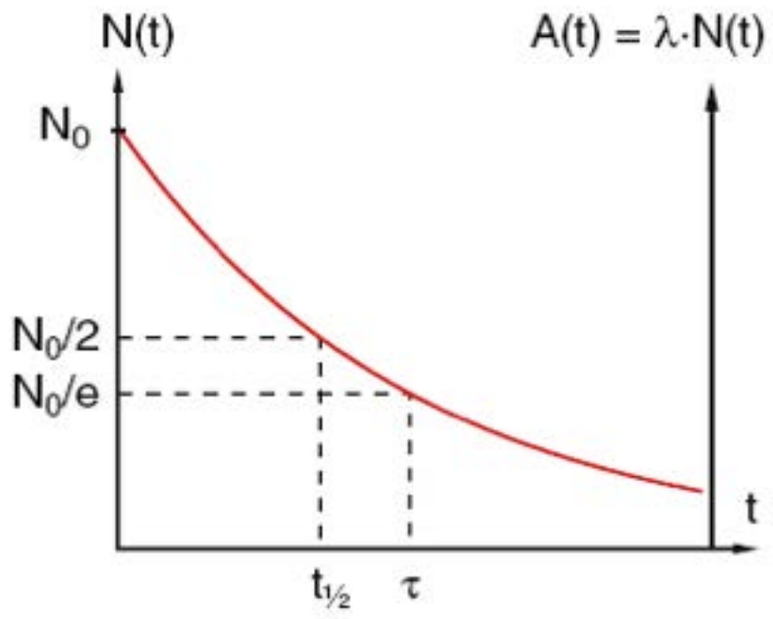




Radioactive decay law

- The decay of any given unstable nucleus is a stochastic process
- It follows the Poisson statistics
- We define the probability per unit of time that an unstable nucleus decays as the ***decay constant*** λ
- **Decay rate equation**: For an ensemble of N nuclei, the number of particles dN decaying in an time interval dt is λN

$$\frac{dN}{dt} = -\lambda N \longrightarrow N(t) = N_0 e^{-\lambda t}$$



- $\tau = 1/\lambda$ is the life time $\mathbf{N(\tau)=N_0/e}$
- $\mathbf{t_{1/2}=\tau\ ln\ 2}$ is the half life $\mathbf{N(t_{1/2})=N_0/2}$
- **Nuclear Activity** of an ensemble of radioactive nuclei is the number of particles per unit of time that decay at a time t
- $\mathbf{A(t)=\lambda\ N(t)}$

Natural radioactive nuclei

Element	Symbol	Strahlugsart, Energie /MeV	$t_{1/2}$/a
Tritium	$^{3}_{1}H$	β^- 0,0286	12,3
Kalium	$^{40}_{19}K$	β^- 1,35	$1,5 \cdot 10^9$
Rubidium	$^{87}_{37}Rb$	β^- 0,275	$5 \cdot 10^{10}$
Iod	$^{129}_{53}I$	β^- 0,15	$1,7 \cdot 10^7$
Cäsium	$^{135}_{55}Cs$	β^- 0,21	$3,0 \cdot 10^6$
Blei	$^{205}_{82}Pb$	α 2,6	$\approx 1,4 \cdot 10^{16}$
Polonium	$^{209}_{84}Po$	α 4,87	103
Radium	$^{226}_{88}Ra$	α 4,77	1620
Thorium	$^{230}_{90}Th$	α 4,5–4,7	$8 \cdot 10^4$
Uran	$^{234}_{92}U$	α 4,6–4,8	$2,5 \cdot 10^5$
	$^{235}_{92}U$	α 4,3–4,6	$7,1 \cdot 10^8$
	$^{238}_{92}U$	α 4,2	$4,5 \cdot 10^9$

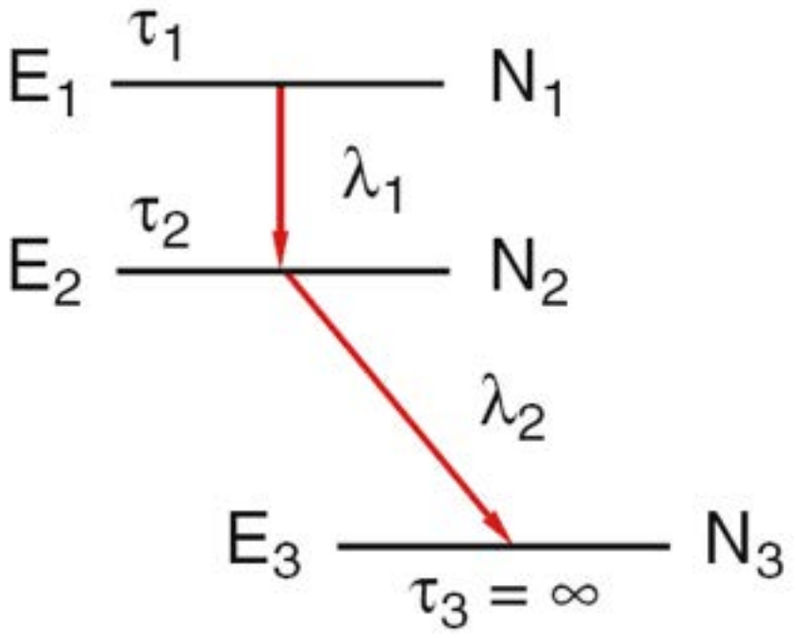
Cascade decays

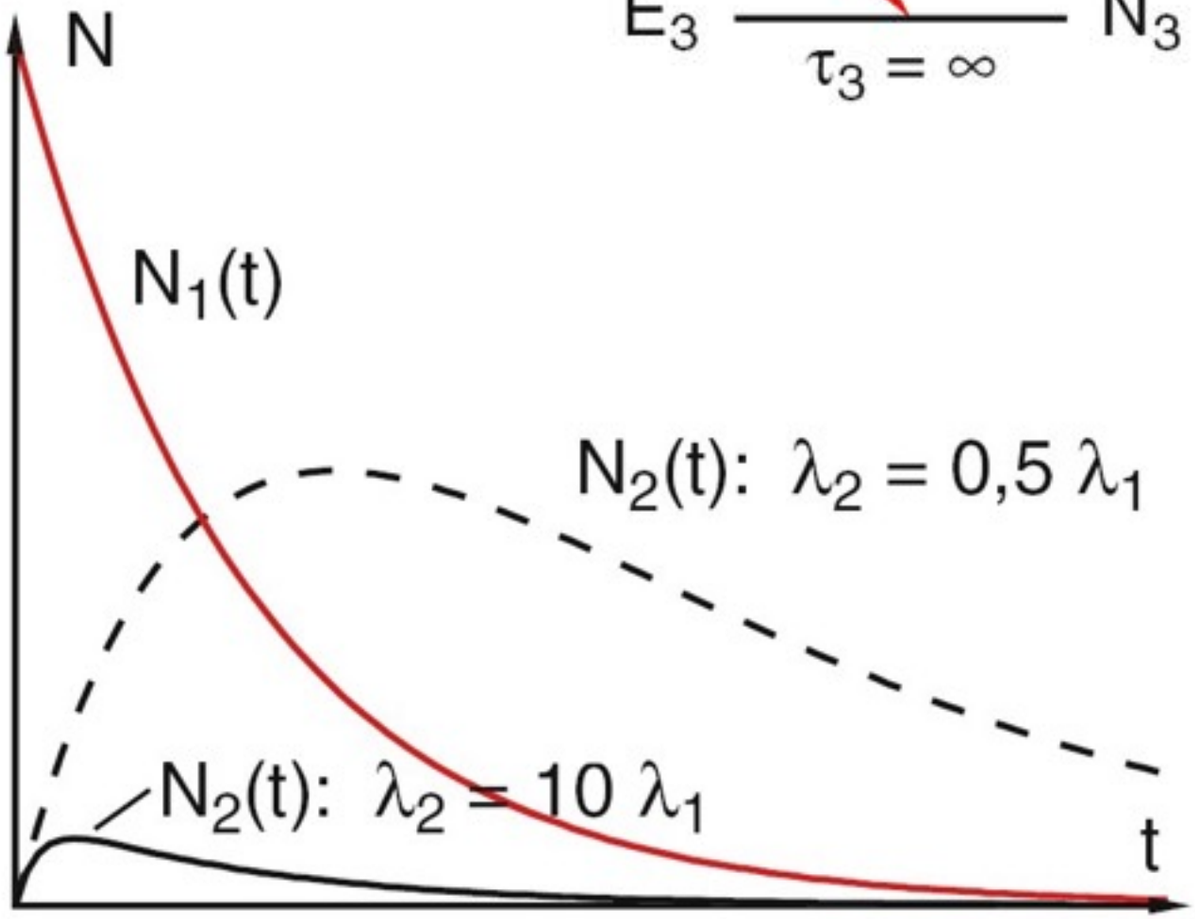
Many nuclei decay into unstable nuclei, which then undergo further radioactive decay: cascade decay

$$\begin{cases} \dfrac{dN_1}{dt} = -\lambda_1 N_1 \,, \\ \dfrac{dN_2}{dt} = +\lambda_1 N_1 - \lambda_2 N_2 \,, \\ \dfrac{dN_3}{dt} = +\lambda_2 N_2 \,, \end{cases}$$

$$N_2(0) = N_3(0) = 0$$

Assume 3 is stable

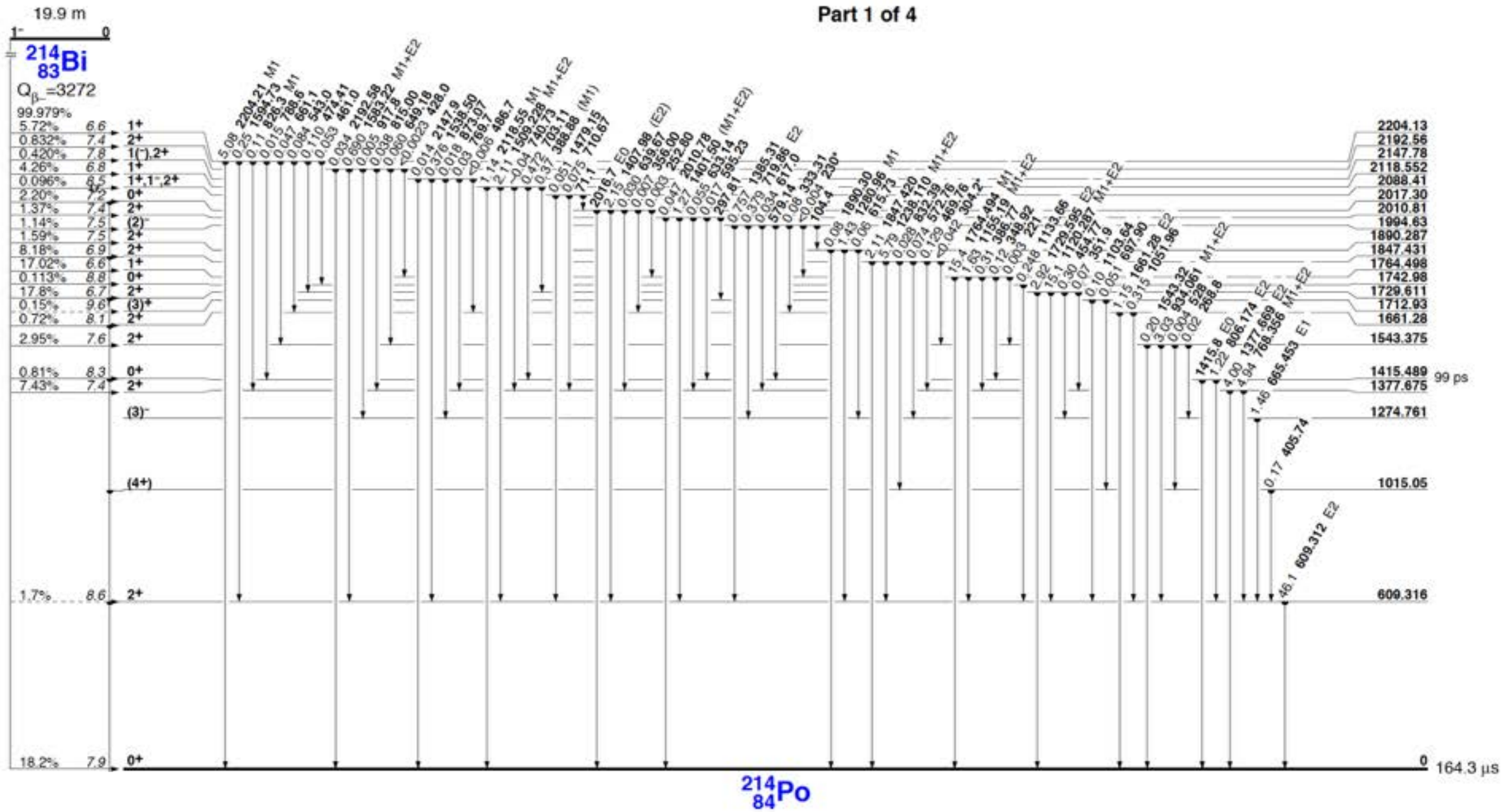




Cascade decays

While

- α decays are predominantly on the ground state
- β (family) decays are on a number of excited states



Secular equilibrium (1/2)

- Occurs in a radioactive decay chain **when the half-life of the daughter radionuclide B is much shorter than the half-life of the parent radionuclide A**.
- As a result **the decay rate of A, and hence the production (and decay) rate of B, are approximately constant (and equal)**, because the half-life of A is very long compared to the timescales being considered.
- The **quantity of radionuclide B builds up until the number of B atoms decaying per unit time becomes equal to the number being produced per unit time**
- The quantity of radionuclide B then reaches a **constant, equilibrium value.**
- Assuming the initial concentration of radionuclide B is zero, full equilibrium usually takes several half-lives of radionuclide B to establish.

- The quantity of radionuclide B when secular equilibrium is reached is determined by the quantity of its parent A and the half-lives of the two radionuclide.
- This can be seen from the time rate of change of the number of atoms of radionuclide B:

$$\frac{dN_B}{dt} = \lambda_A N_A - \lambda_B N_B$$

where λ_A and λ_B are the decay constants of radionuclide A and B, related to their half-lives $t_{1/2}$ by $\boldsymbol{\lambda = \ln(2)/t_{1/2}}$, and N_A and N_B are the number of atoms of A and B at a given time.

Secular equilibrium (2/2)

- **Secular equilibrium occurs when $dN_B/dt=0$ or**

$$N_B = \frac{\lambda_A}{\lambda_B} N_A$$

- Over long enough times, comparable to the half-life of radionuclide A, the secular equilibrium is only approximate
- N_A decays away according to

$$N_A(t) = N_A(0)e^{-\lambda_A t}$$

and the "equilibrium" quantity of radionuclide B declines in turn.

- For times short compared to the half-life of A ($\lambda_A t \ll 1$) the exponential can be approximated as 1.

- **Secular equilibrium is rarely accomplished in nature.**
- **It is frequently broken by mechanical (e.g. ore cracks) or chemical (e.g. metallurgy) actions**

Radioactive nuclei

Radioactive nuclei

Classification:

Naturally occurring	Fossile radioactivity	τ longer than T_{earth}
	Continuously produced	Nuclear reactions (e.g. cosmic rays)
Artificial (anthropogenic)	Fission fragments	Fission reactors
	Nuclear reactions	Accelerators

Natural radioactive nuclides

- Some of the most common naturally occurring **isolated** radioactive nuclides

Nuclide	t1/2(y)	Abundance	Daughter
^{14}C	5730	1.35E-10	^{14}N
^{40}K	1.25E+09	0.0117	^{40}Ar
^{87}Rb	4.88E+10	27.83	^{87}Sr
^{113}Cd	9E+15	12.2	^{113}In
^{115}In	4.4E+14	95.7	^{115}Sn
^{138}La	1.1E+11	0.09	^{138}Ce
^{144}Nd	2.3E+15	23.8	^{140}Ce
^{147}Sm	1.06E+11	15.0	^{143}Nd
^{176}Lu	3.59E+10	2.59	^{176}Hf
^{187}Re	4.30E+10	62.60	^{187}Os

All living people are somewhat radioactive, e.g. depending on how much NaCl they use (it is obtained from mines, where there is some KCl present as well

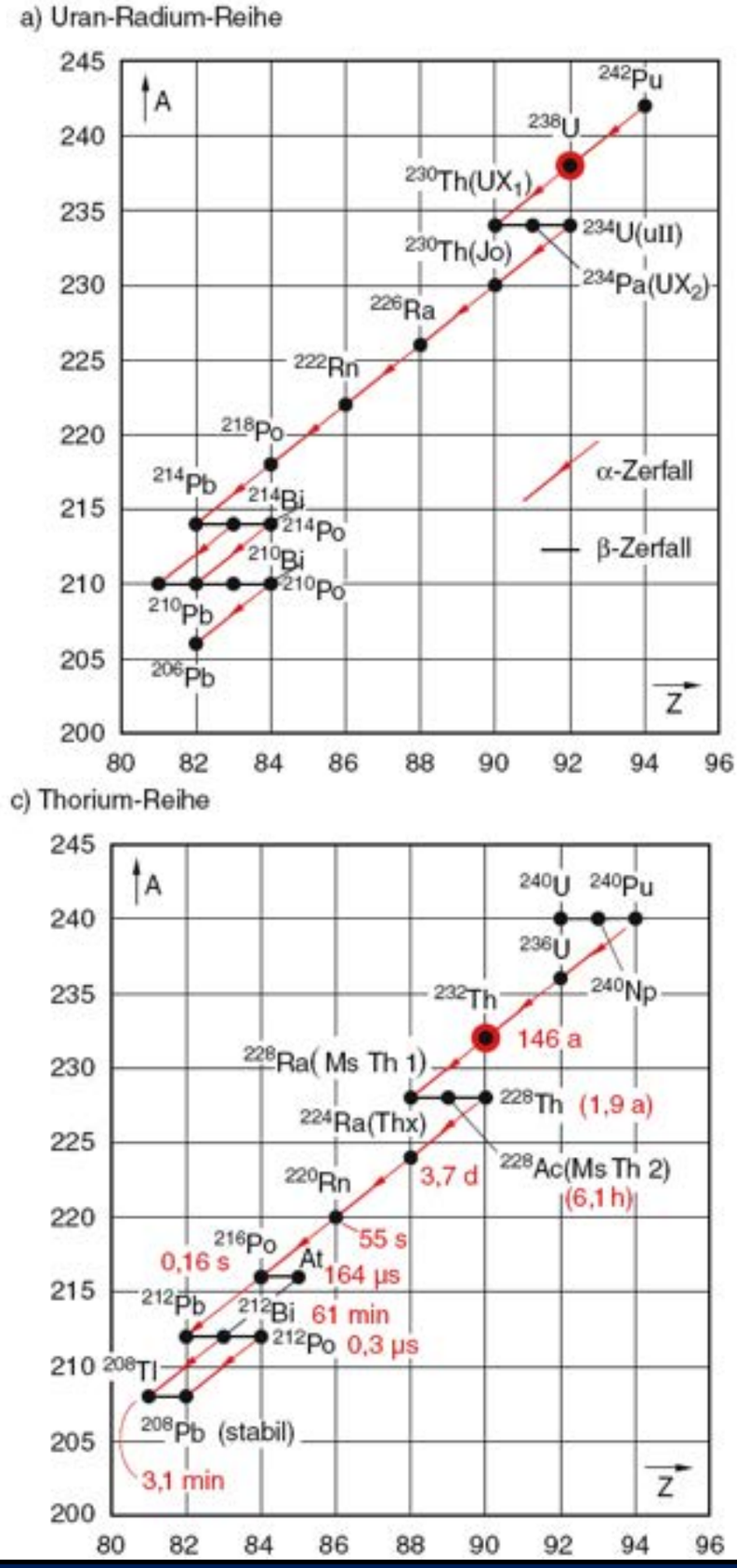
- There are **only four paths (chains)** that the heavy naturally occurring radioactive nuclides may take as they decay.
- All four paths lead to different types of isotopes of Pb

Mass number	Series name	Parent	$t_{1/2}$(y)	End product
4n	Thorium	^{232}Th	1.40E+10	^{208}Pb
4n+1	Neptunium	^{237}Np	2.14E+06	^{209}Bi
4n+2	Uranium	^{238}U	4.47E+09	^{206}Pb
4n+3	Actinium	^{235}U	7.04E+08	^{207}Pb

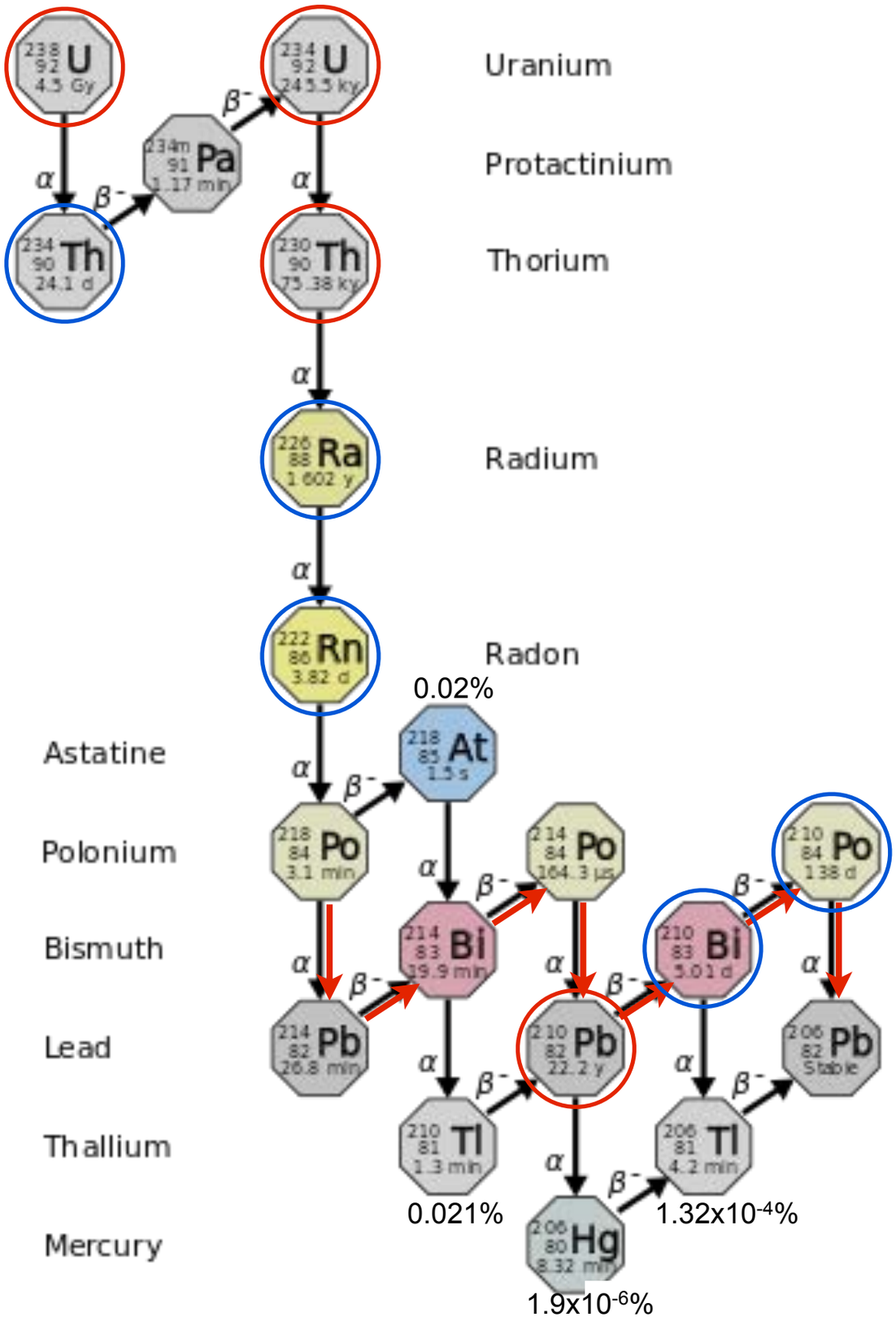
The 4 radioactive series

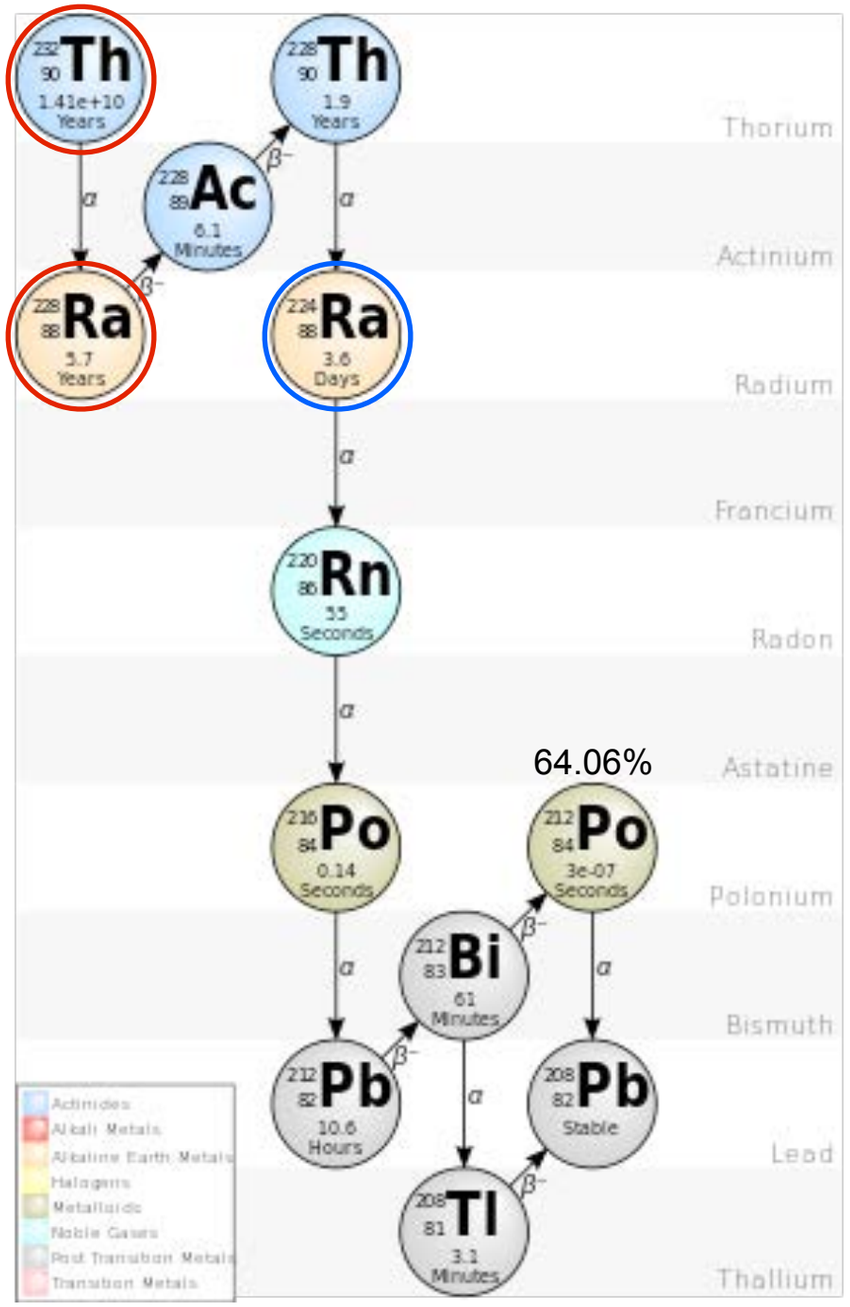
- **All heavy radioactive elements align in four decay-series.**
- Each starts with a heavy element, unstable, and produced during collisions of stars
- Each series ends in stable isotopes of lead or Bismuth
- The isotopes above the red-points in the chain have a very short lifetime, and cannot be found in the earth.

\

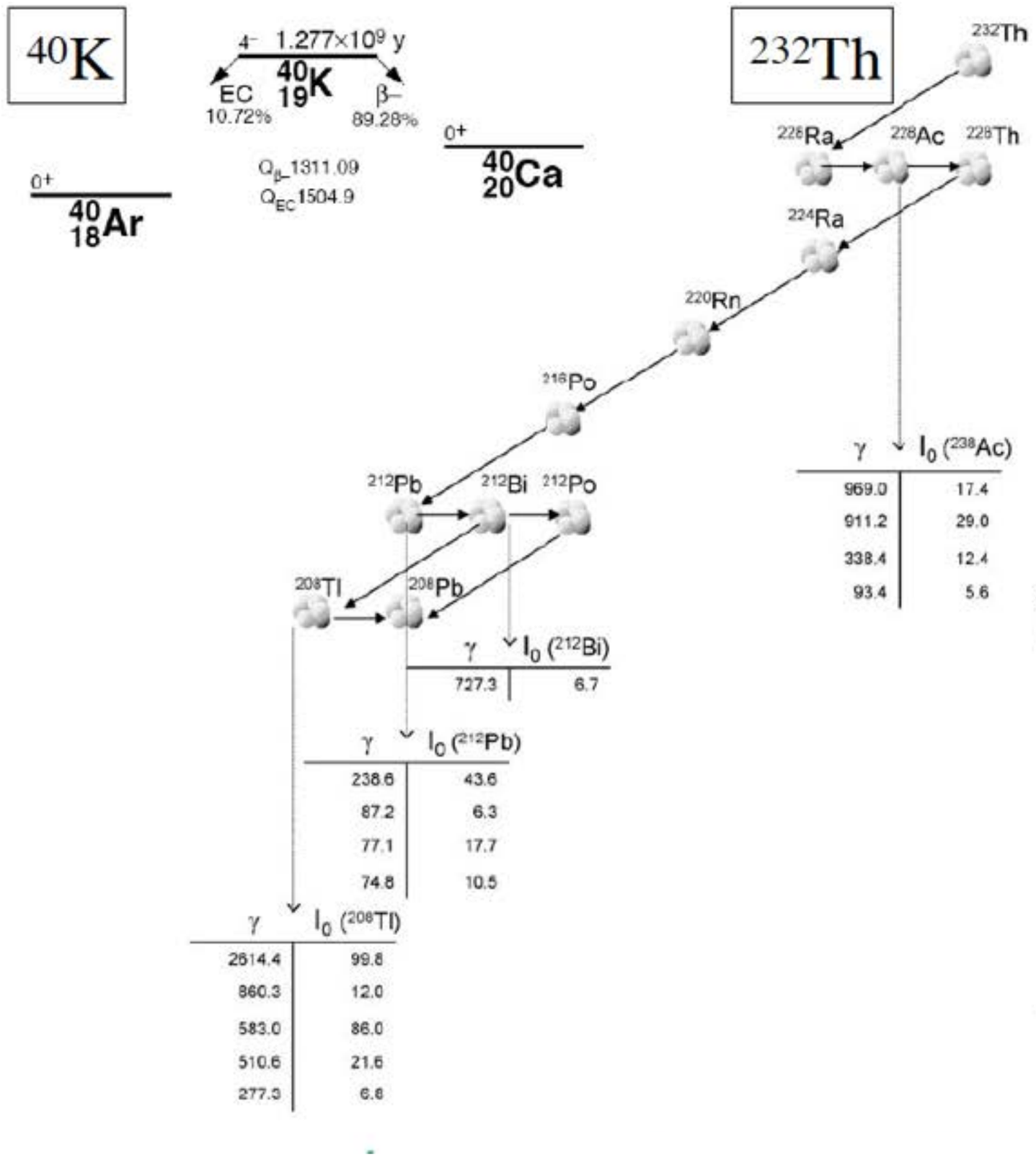


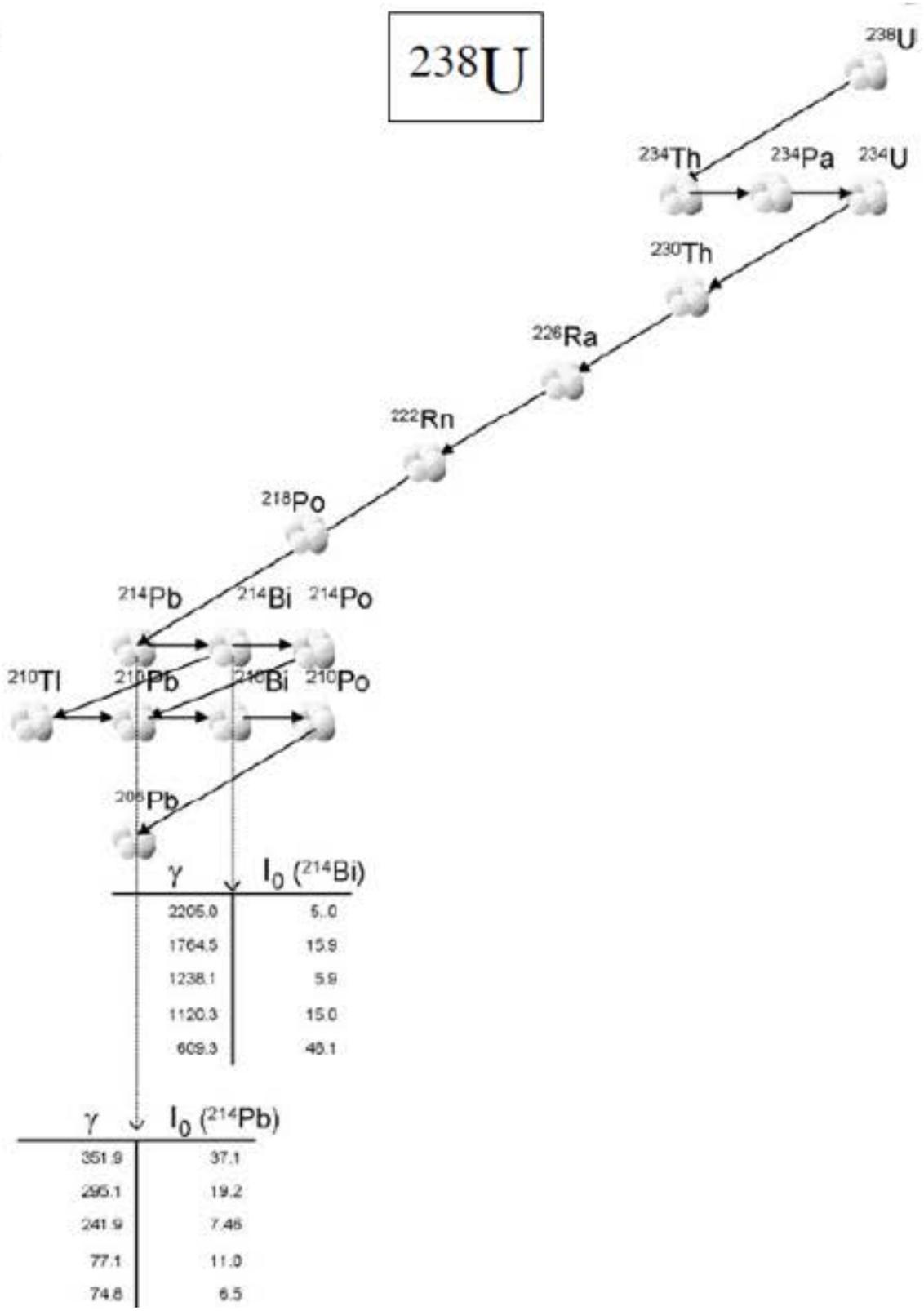
Natural series



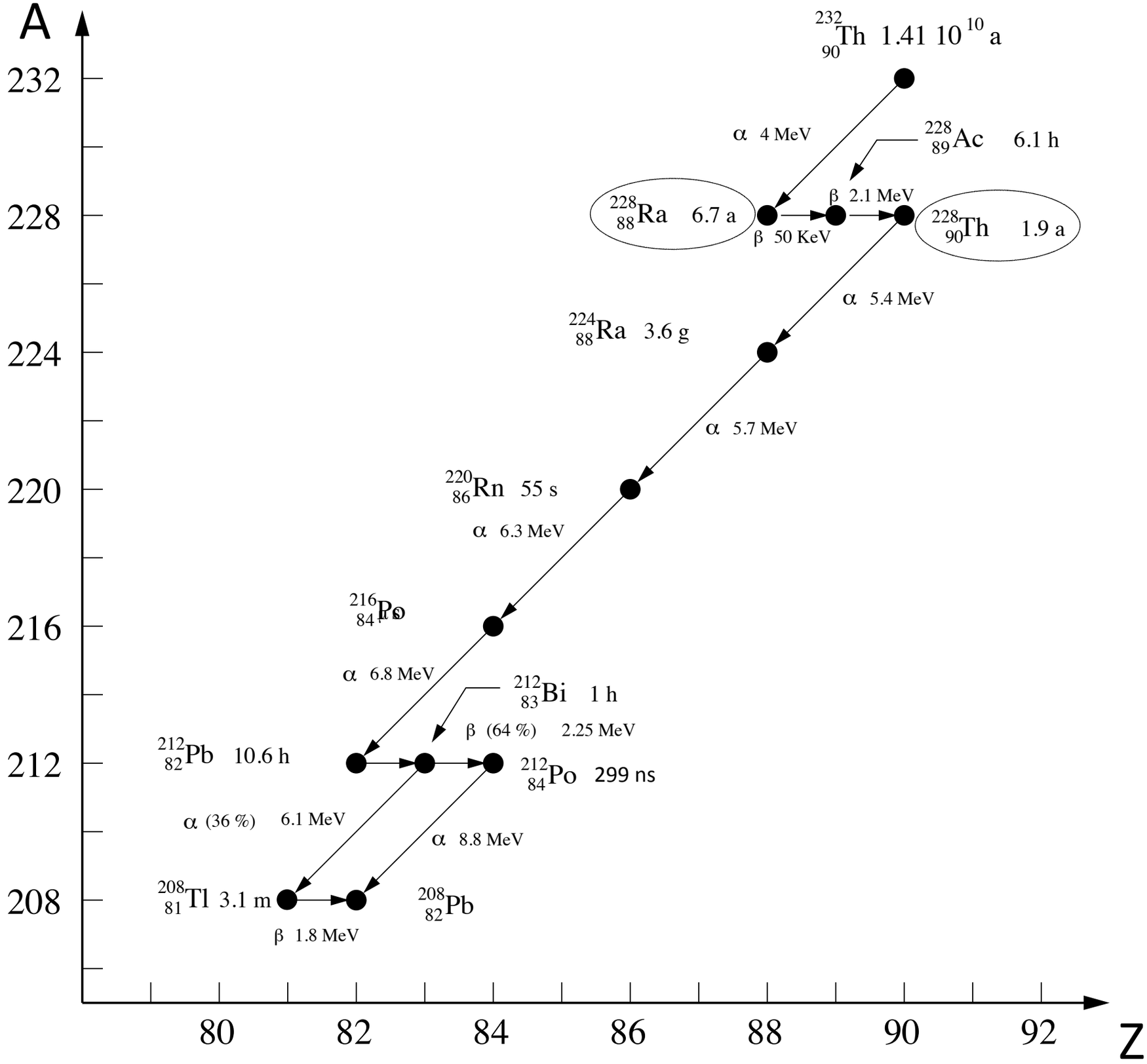


Fossile radioactivity

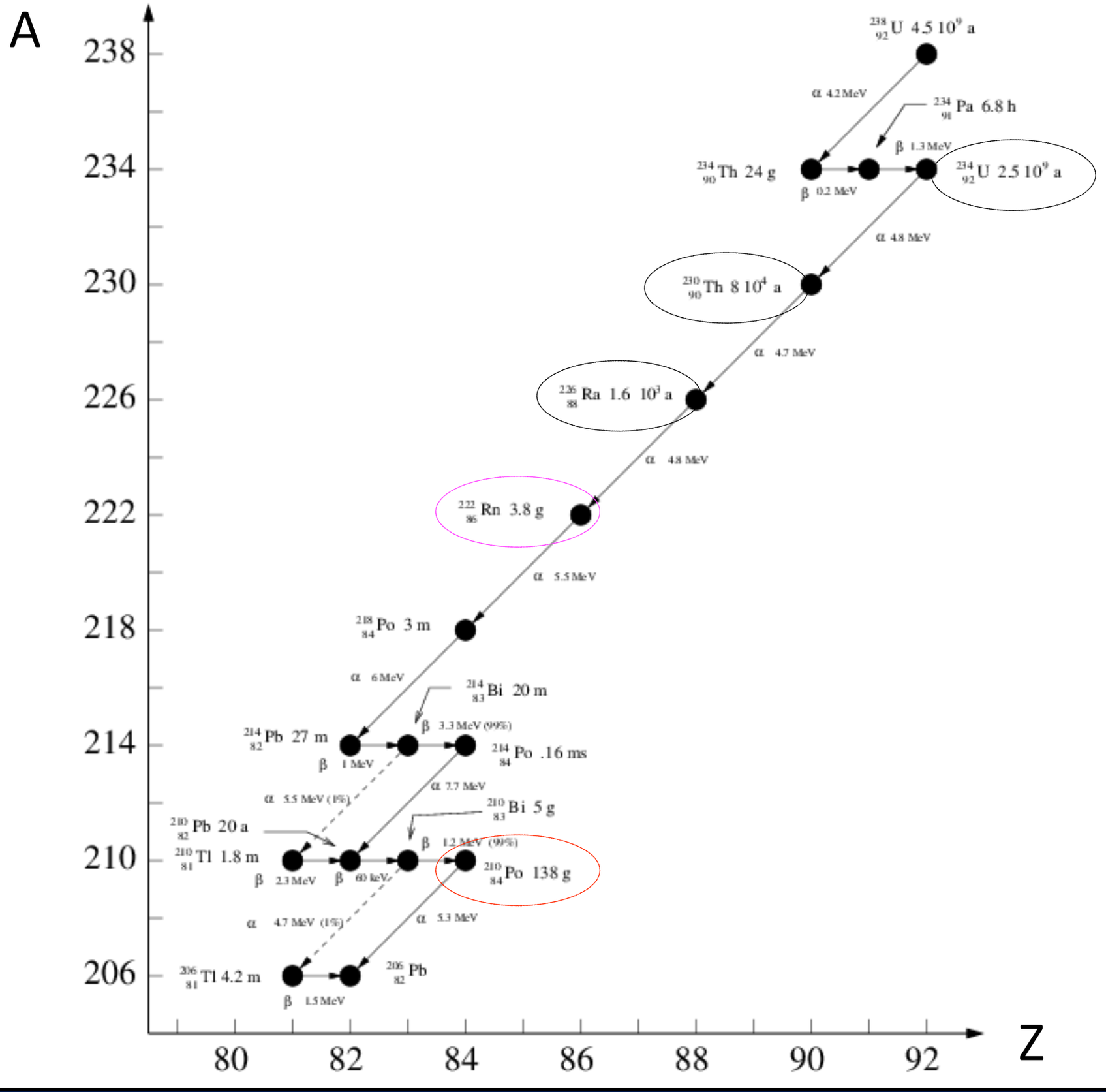




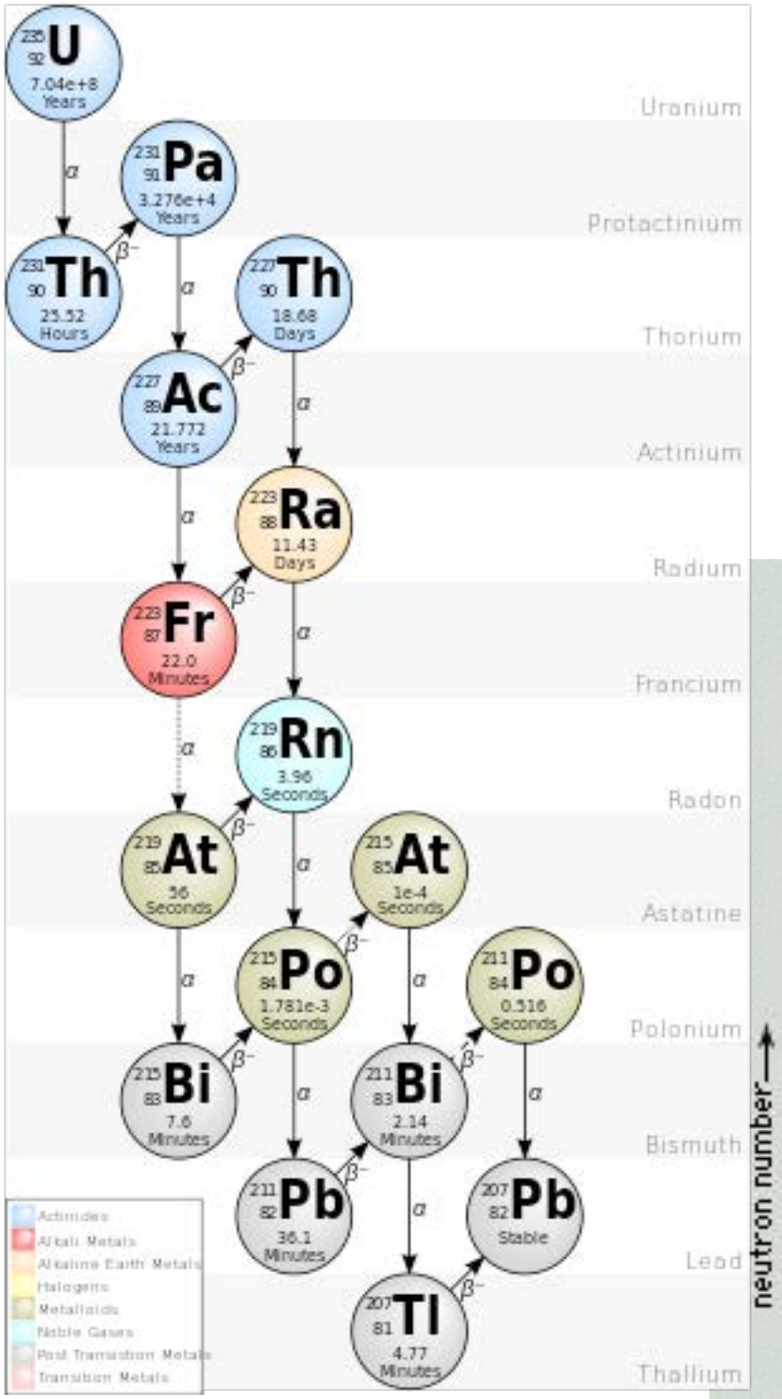
Thorium series



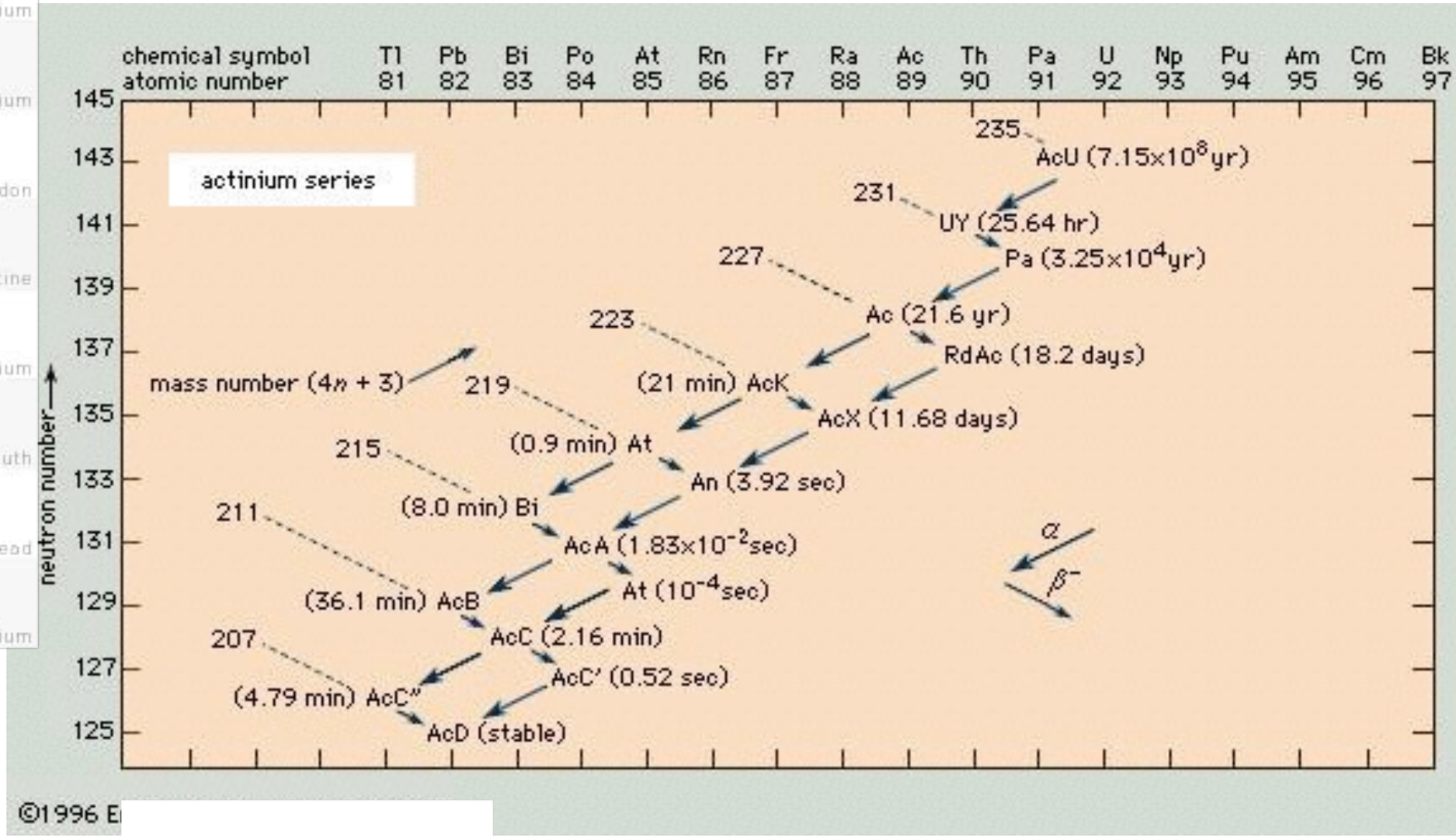
Uranium series



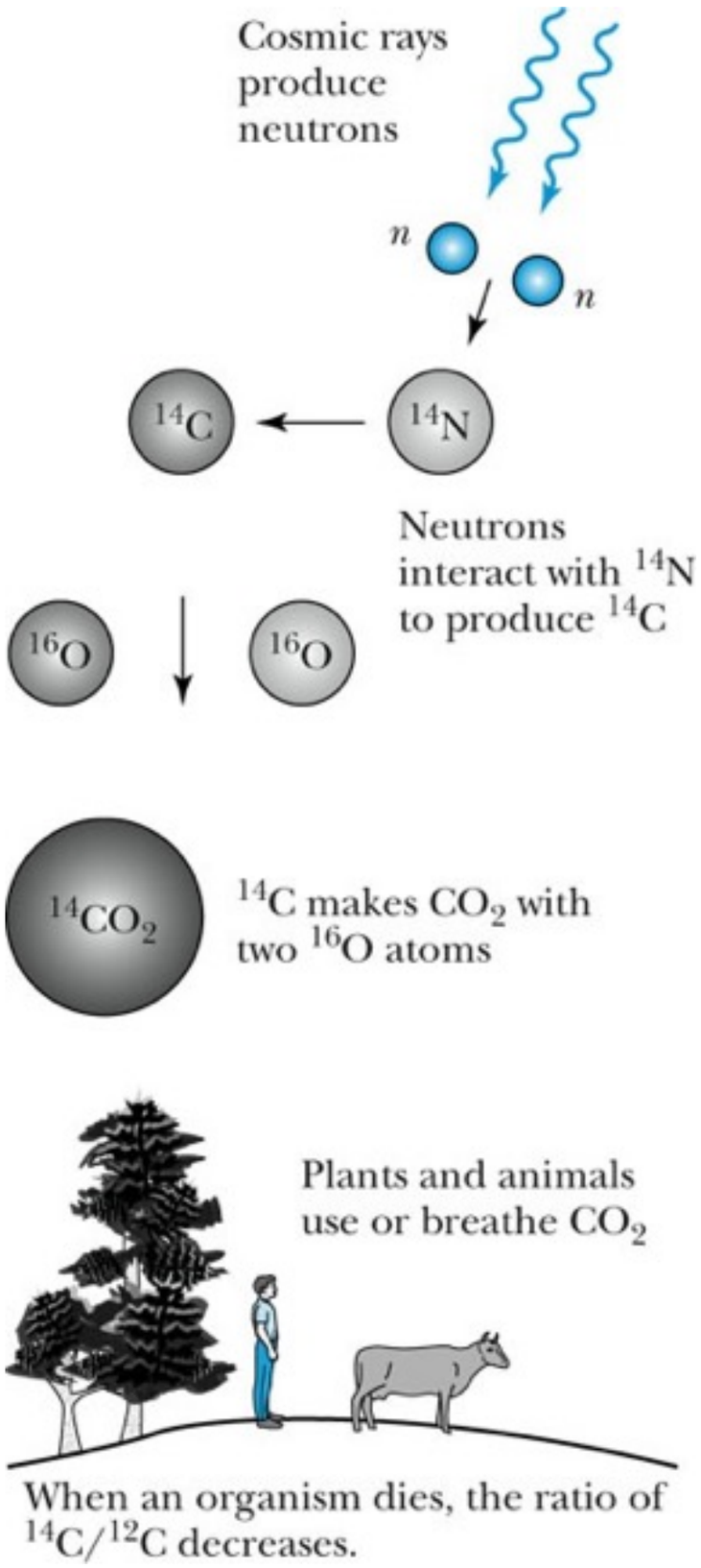
^{235}U



Less abundant (~ factor 20) with respect to Uranium series



Radioactive Carbon Dating



- Radioactive ^{14}C is produced in our atmosphere by the bombardment of ^{14}N by neutrons produced by cosmic rays.

$$n + {}^{14}\mathrm{N} \rightarrow {}^{14}\mathrm{C} + p$$

- When living organisms die, their intake of ^{14}C ceases, and the ratio of ^{14}C / ^{12}C decreases as ^{14}C decays.
- Because the half-life of ^{14}C is 5,730 years, it is convenient to use the ^{14}C / ^{12}C ratio to determine the age of objects over a range up to 45,000 years ago.
- The period just before 9000 years ago had a higher ^{14}C / ^{12}C ratio by factor of about 1.5 than it does today.

Example: ^{14}C dating

An archaeologist finds a 25.0-g piece of charcoal in the ruins of an ancient city. The sample shows a ^{14}C activity of 250 decays/min. How long has the tree from which this charcoal came been dead?

Solution First calculate the decay constant for ^{14}C, which has a half-life of 5730 yr.

$$\lambda = \frac{0.693}{T_{1/2}} = \frac{0.693}{(5730\ \text{yr})(3.16 \times 10^7\ \text{s/yr})}$$
$$= 3.83 \times 10^{-12}\ \text{s}^{-1}$$

The number of ^{14}C nuclei can be calculated in two steps.

(1) The number of ^{12}C nuclei in 25.0 g of carbon is

$$N(^{12}C) = \frac{6.02 \times 10^{23}\ \text{nuclei/mol}}{12.0\ \text{g/mol}}(25.0\ \text{g})$$
$$= 1.26 \times 10^{24}\ \text{nuclei}$$

Knowing that the ratio of ^{14}C to ^{12}C in the live sample was 1.3×10^{-12}, we see that the number of ^{14}C nuclei in 25.0 g *before* decay is

$$N_0(^{14}C) = (1.3 \times 10^{-12})(1.26 \times 10^{24})$$
$$= 1.6 \times 10^{12}\ \text{nuclei}$$

Hence the initial activity of the sample is

$$R_0 = N_0\lambda = (1.6 \times 10^{12}\ \text{nuclei})(3.83 \times 10^{-12}\ \text{s}^{-1})$$
$$= 6.13\ \text{decays/s} = 370\ \text{decays/min}$$

(2) We can now calculate the age of the charcoal, using Equation which relates the activity R at any time t to the initial activity R_0:

$$R = R_0 e^{-\lambda t} \quad \text{or} \quad e^{-\lambda t} = \frac{R}{R_0}$$

Because it is given that $R = 250$ decays/min and because we found that $R_0 = 370$ decays/min, we can calculate t by taking the natural logarithm of both sides of the last equation:

$$-\lambda t = \ln\left(\frac{R}{R_0}\right) = \ln\left(\frac{250}{370}\right) = -0.39$$

$$t = \frac{0.39}{\lambda} = \frac{0.39}{3.84 \times 10^{-12}\ \text{s}^{-1}}$$
$$= 1.0 \times 10^{11}\ \text{s} = 3.2 \times 10^3\ \text{yr}$$

Time Dating Using Lead Isotopes

- A plot of the abundance ratio of 206Pb / 204Pb versus 207Pb / 204Pb can be a sensitive indicator of the age of ores.
- It can be used to date rocks that formed from about 1 million years to over 4.5 billion years ago with routine precisions in the 0.1–1 percent range.
- Such technique has been used to show that meteorites and the earth are 4.55 billion years old.Uranium–lead
- The dating method relies on two separate decay chains, the uranium series from 238U to 206Pb, with a half-life of 4.47 billion years and the actinium series from 235U to 207Pb, with a half-life of 710 million years.
- These uranium to lead decay routes occur via a series of alpha (and beta) decays, in which 238U with daughter nuclides undergo eight total alpha and six beta decays whereas 235U with daughters only experience seven alpha and four beta decays.

- **Method:**

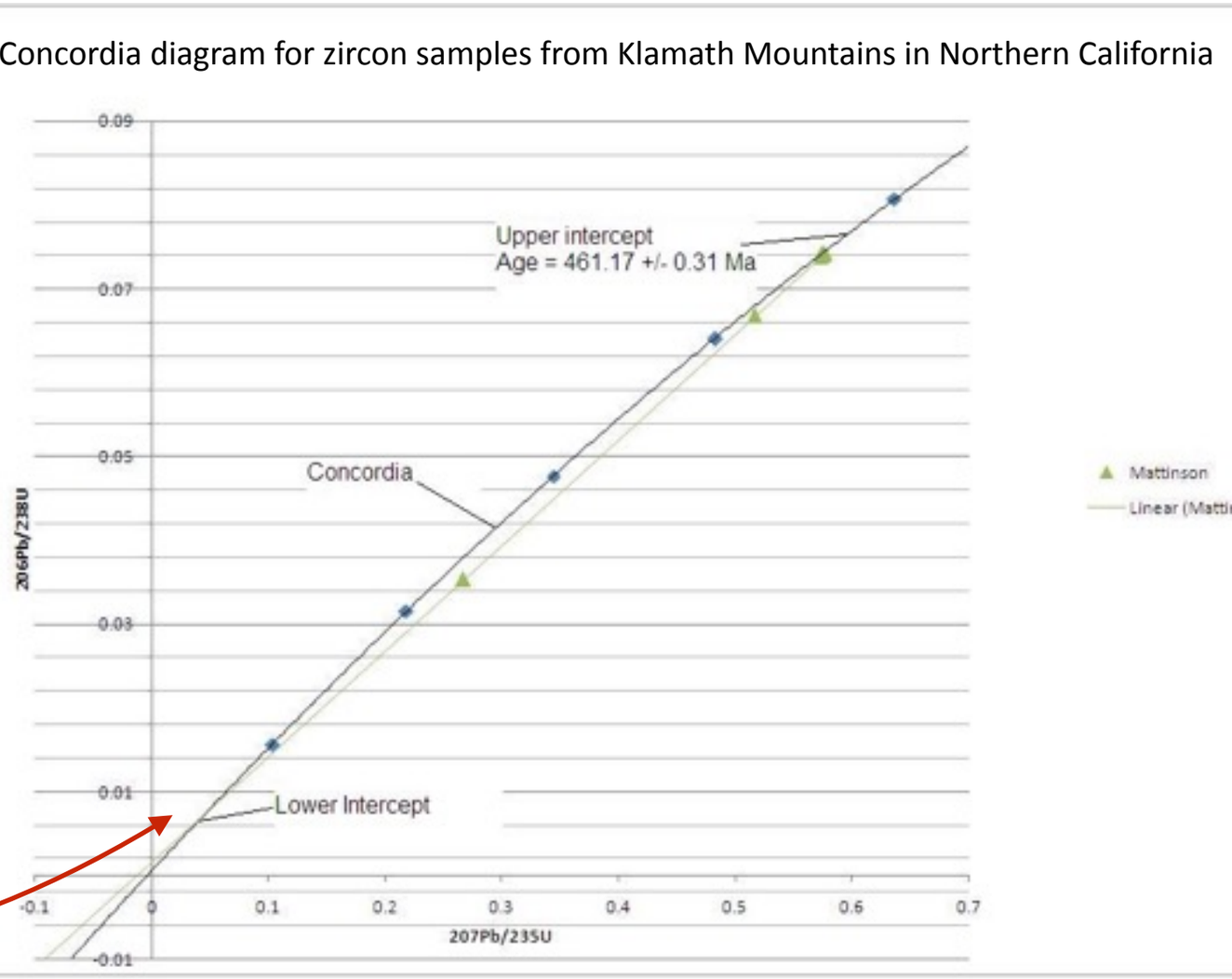
  $N_U = (N_U+N_{Pb})\ \exp(-\lambda_U t)$

  $N_{Pb}/N_U = \exp(-\lambda_U t) - 1$

  Separately for ^{206}Pb and ^{207}Pb

- **Build Concordia plot**

Concordia diagram for zircon samples from Klamath Mountains in Northern California



Examples

The ^{226}Ra nucleus undergoes alpha decay according to Equation 13.12. Calculate the Q value for this process. Take the atomic masses to be 226.025 406 u for ^{226}Ra, 222.017 574 u for ^{222}Rn, and 4.002 603 u for $^{4}_{2}$He, as found in Appendix B.

Solution Using Equation 13.16, we see that

$$Q = (M_X - M_Y - M_\alpha) \times 931.494 \frac{\text{MeV}}{\text{u}}$$

$$= (226.025\,406\ \text{u} - 222.017\,574\ \text{u} - 4.002\,603\ \text{u}) \times 931.494 \frac{\text{MeV}}{\text{u}}$$

$$= (0.005\,229\ \text{u}) \times \left(931.494 \frac{\text{MeV}}{\text{u}}\right) = 4.87\ \text{MeV}$$

A sample of the isotope ^{131}I, which has a half-life of 8.04 days, has an activity of 5 mCi at the time of shipment. Upon receipt of the ^{131}I in a medical laboratory, its activity is 4.2 mCi. How much time has elapsed between the two measurements?

Solution We can make use of Equation 13.10 in the form

$$\frac{R}{R_0} = e^{-\lambda t}$$

Taking the natural logarithm of each side, we get

$$\ln\left(\frac{R}{R_0}\right) = -\lambda t$$

$$(1) \qquad t = -\frac{1}{\lambda} \ln\left(\frac{R}{R_0}\right)$$

To find λ,

$$(2) \qquad \lambda = \frac{0.693}{T_{1/2}} = \frac{0.693}{8.04\ \text{days}}$$

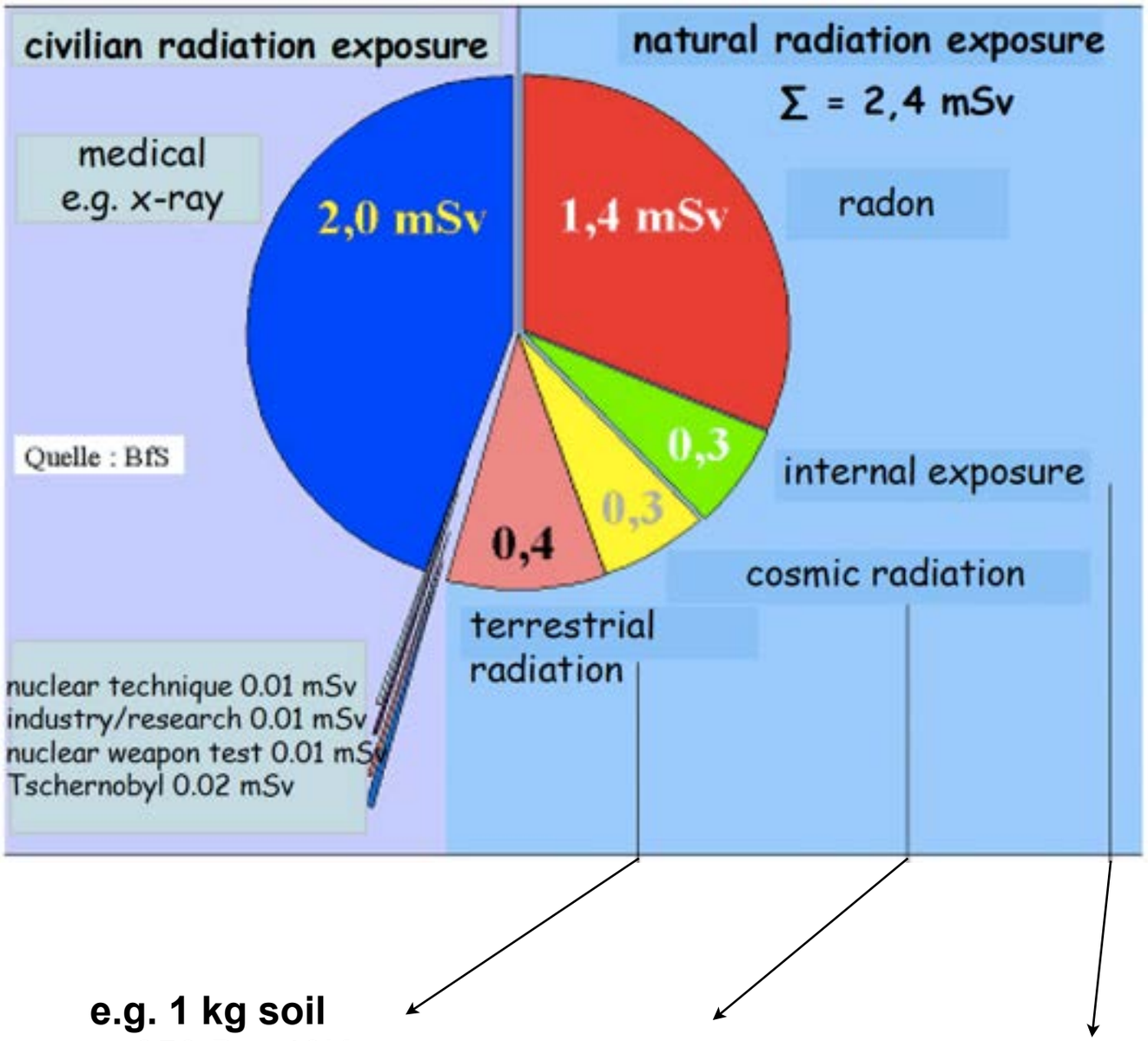
Substituting (2) into (1) gives

$$t = -\left(\frac{8.04\ \text{days}}{0.693}\right) \ln\left(\frac{4.2\ \text{mCi}}{5.0\ \text{mCi}}\right) = 2.02\ \text{days}$$

Sources of human radiation

average annual radiation

(Germany: total: 4.5 mSv)



Just an example:

- Borexino liquid scintillator
- concentration ^{232}Th ≈ 4x10-13 fraction
  ^{238}U ≈ 6x10-15 fraction
  (< 0.3 pBq/kg)
- **So 300t of LS correspond to 2 ng of soil**

e.g. 1 kg soil

- 850 Bq 40K
- 44 Bq 232Th
- 36 Bq 238U

at flight altitude

- ca x 40

e.g. about
60 Bq/kg 40K

Screening techniques

Units

Conversion Bq/kg g/g:

- M = sample mass (kg)
- R = Rate (activity)
- a_{exp} = Specific activity

$$\frac{R}{M} = \frac{\lambda N}{M} = a_{exp}[\frac{Bq}{kg}]$$

$$\frac{m}{M} = \frac{NA}{N_A M} = C[\frac{g}{g}]$$

$$\frac{a_{exp}}{\lambda}\frac{A}{N_A} = C$$

$$k = \frac{A}{\lambda N_A} = \frac{A\tau}{N_A}$$

^{238}U:

- τ = 4.47e9 y/ln(2) = 6.45e9 y = 2.035e17sec
- A/N_A = 238/6.022E23*1E-3kg
- k = 8.04e-8

^{232}Th:

- τ = 1.405e10 y/ln(2) 2.026e10 y = 6.397e17 sec
- A/N_A = 232*/6.022E23 g
- k = 2.46e-7

^{40}K: 3.7e-10
^{210}Pb: 3.5e-16
^{60}Co: 2.4e-17

$$\frac{Bq}{kg}k = \frac{g}{g}$$

“Common” assay techniques

- Goal: assess radioactive traces in materials
- Method:
  - detect decay debris → daughter nucleus, α, β, γ
  - decay can be “natural” or induced

    - Direct γ counting with Ge detectors [~10^{-12} gU(Th)/g]
    - Neutron Activation Analysis (NAA) [~10^{-14}-10^{-15} gU(Th)/g]
    - Mass Spectroscopy [~10^{-12}-10^{-13} gU(Th)/g]
      - Inductively Coupled Plasma-Mass Spectroscopy (ICP-MS)
      - Gas Discharge-Mass Spectroscopy (GD-MS)
    - Emission Spectroscopy
      - X-Ray Fluorescence
      - Inductively Coupled Plasma-Emission Spectroscopy (ICP-ES)
    - Target-specific procedures

Example:

- Extraction of ^{224}Ra and ^{226}Ra using manganese oxide (MnOx) and hydrous titanium oxide (HTiO) compounds to assay water with sensitivity of ~10^{-16} g/g. [NIM A501, 386 (2003), NIM A501, 399 (2003)]

Radio assay techniques

Method	Suited for	Sensitivity U/Th
Ge-spectroscopy*	γ emitting nuclides	10-100 μBq/kg
Rn emanation assay	^{226}Ra, ^{228}Th	0.1-10 μBq/kg
Neutron activation	primordial parents	0. 01 μBq/kg
Liquid scintillation counting	a, emitting nuclides	1 mBq/kg
Mass spectrometry (ICP-MS; A-MS)	primordial parents	1-100 μBq/kg
Graphite furnace Atom Adsorption Sp	primordial parents	1-1000 μBq/kg
Roentgen Exitation Analysis	primordial parents	10 mBq/kg
Alpha spectroscopy	^{210}Po, a emitting nuclides	1 mBq/kg

* needs counting time of several weeks to month

Equilibrium breaking is a serious issue:

- **Ge-spectroscopy (high energy resolution ...but small BR's)**
- **α-spectroscopy (only calorimetric approach)**

see e.g.: Borexino Collaboration, Arpesella, C. et al., Measurements of extremely low radioactivity levels in Borexino, Astrop. Phys. 18 (2002) 1-25

Most sensitive techniques

Gamma rays spectroscopy

- Mainly on solids
- Sensible to gamma emitters
- Huge amount of material, few tens of kg
- Long running time measurement many weeks
- Well established technique

Neutron Activation Analysis

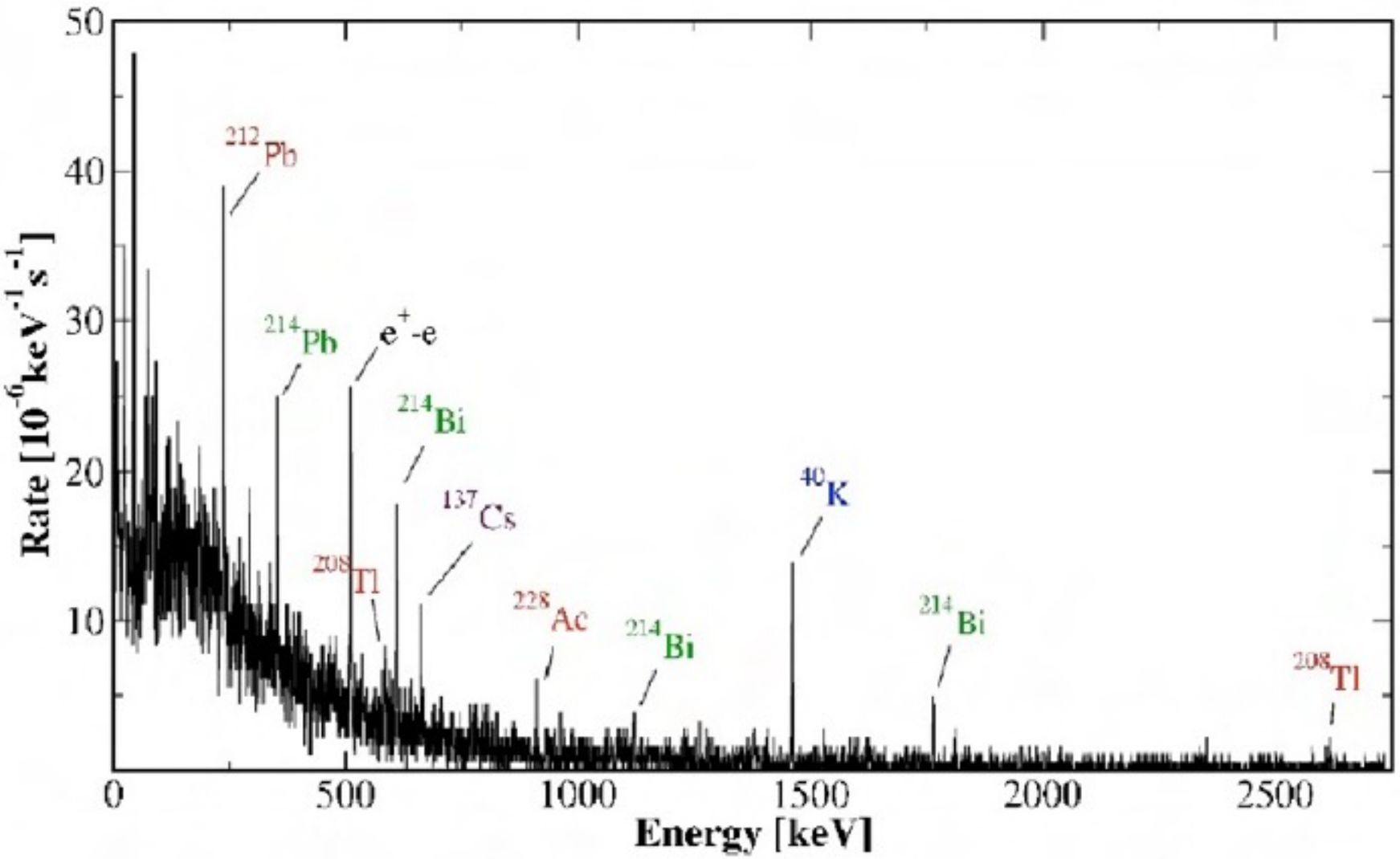
- Solids and Liquids
- Sensitivity on primordial
- Moderate amount of material, few tens of g
- Medium running time, few weeks
- Technique under development for low level contaminations

HR ICP-MS

- Liquids/dissolved Solids
- Sensitivity on primordial
- Small amount of material, less then 1 g
- Short running time, days
- Techniques under fast evolution

HPGe systems

An example:



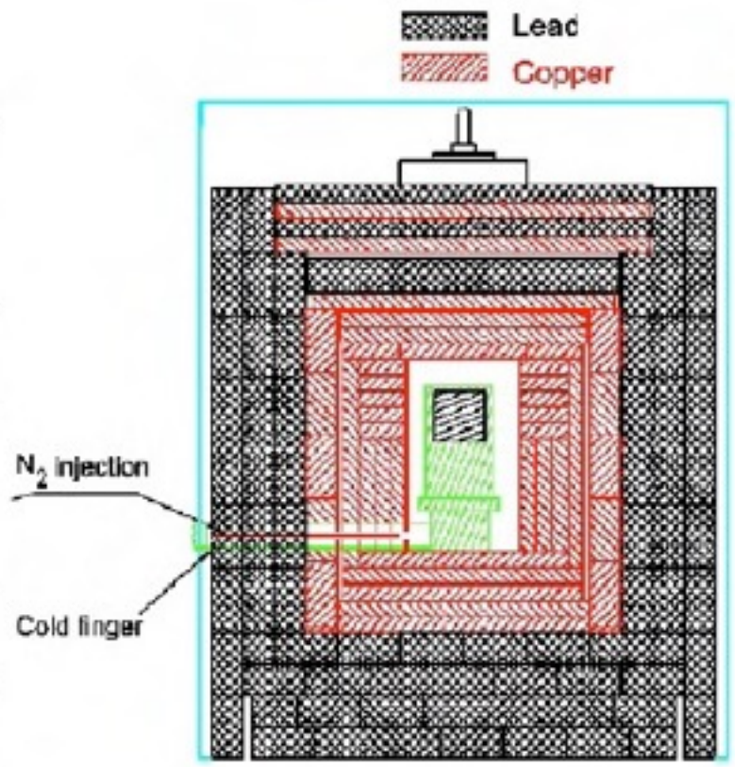
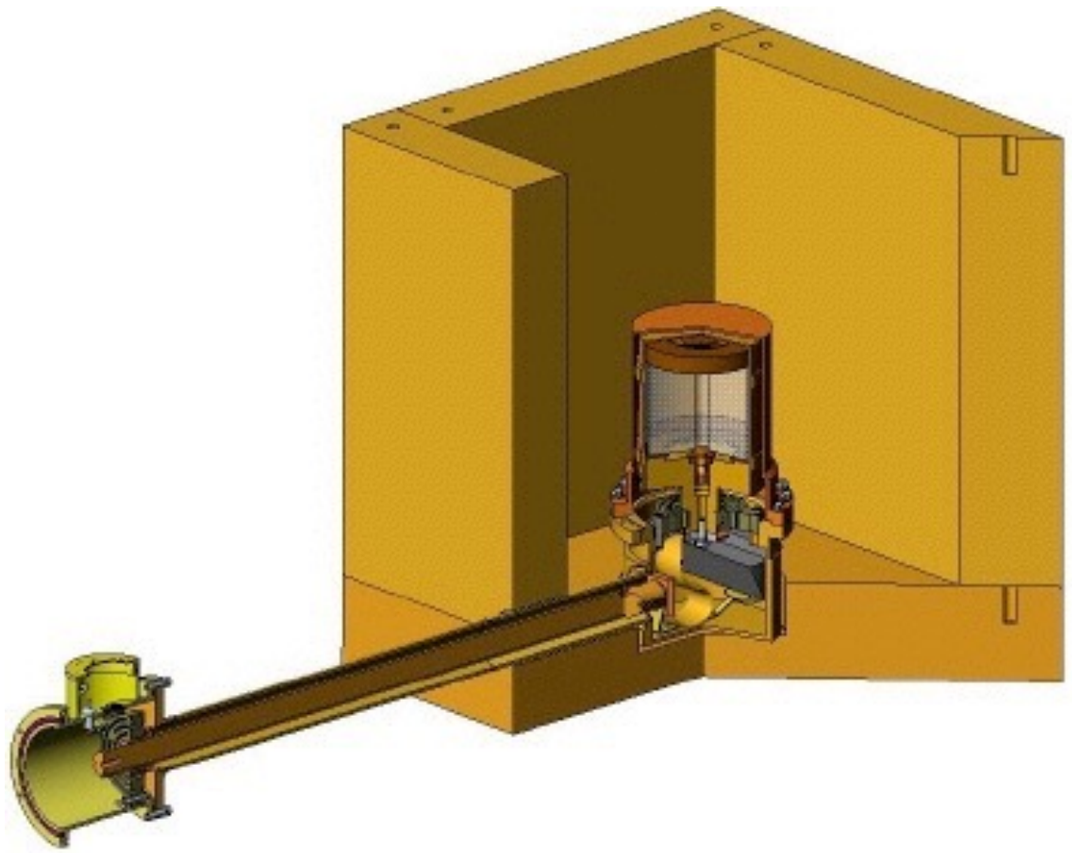


Fig. 3. Underground germanium background spectrum, measured with no sample for 672 h. Gamma lines from natural activities, namely the ^{232}Th (^{208}Tl, ^{228}Ac, ^{212}Pb) and ^{238}U (^{214}Bi, ^{214}Pb) chains, ^{40}K, as well as the ^{137}Cs line from an artificial activity, are indicated.

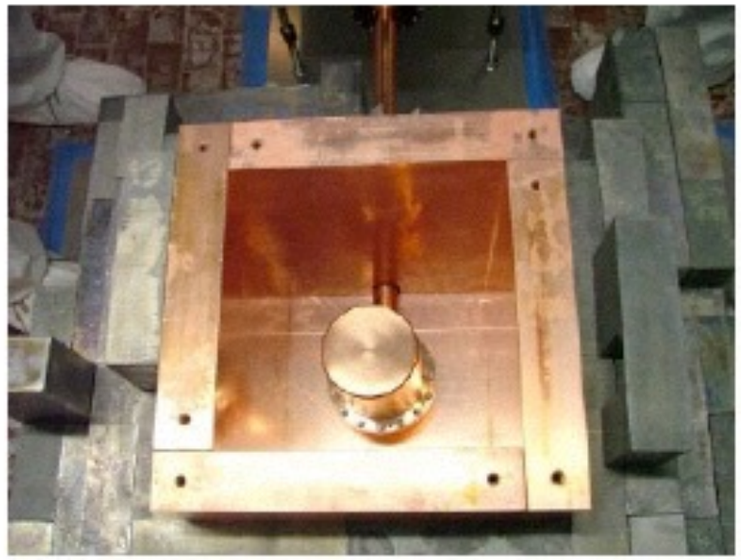
Nucl. Instrum. Meth. A591:490-509,2008.

GeMPI



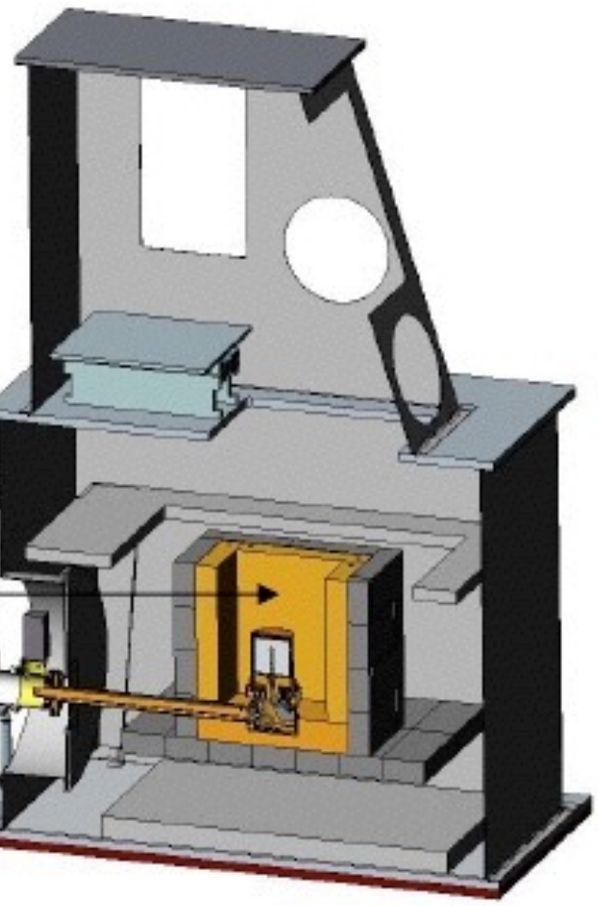
Ge spectrometer of MPI-K at LNGS since 1997 (still state of the art)

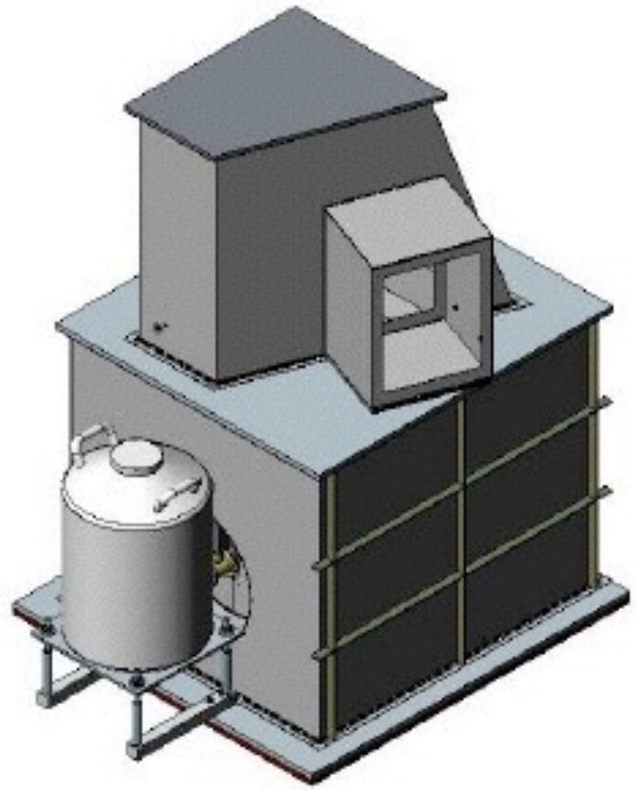
High purity copper directly placed underground after electrolysis



15 l sample chamber

holds e.g. 125 kg Cu
or 157 kg Pb



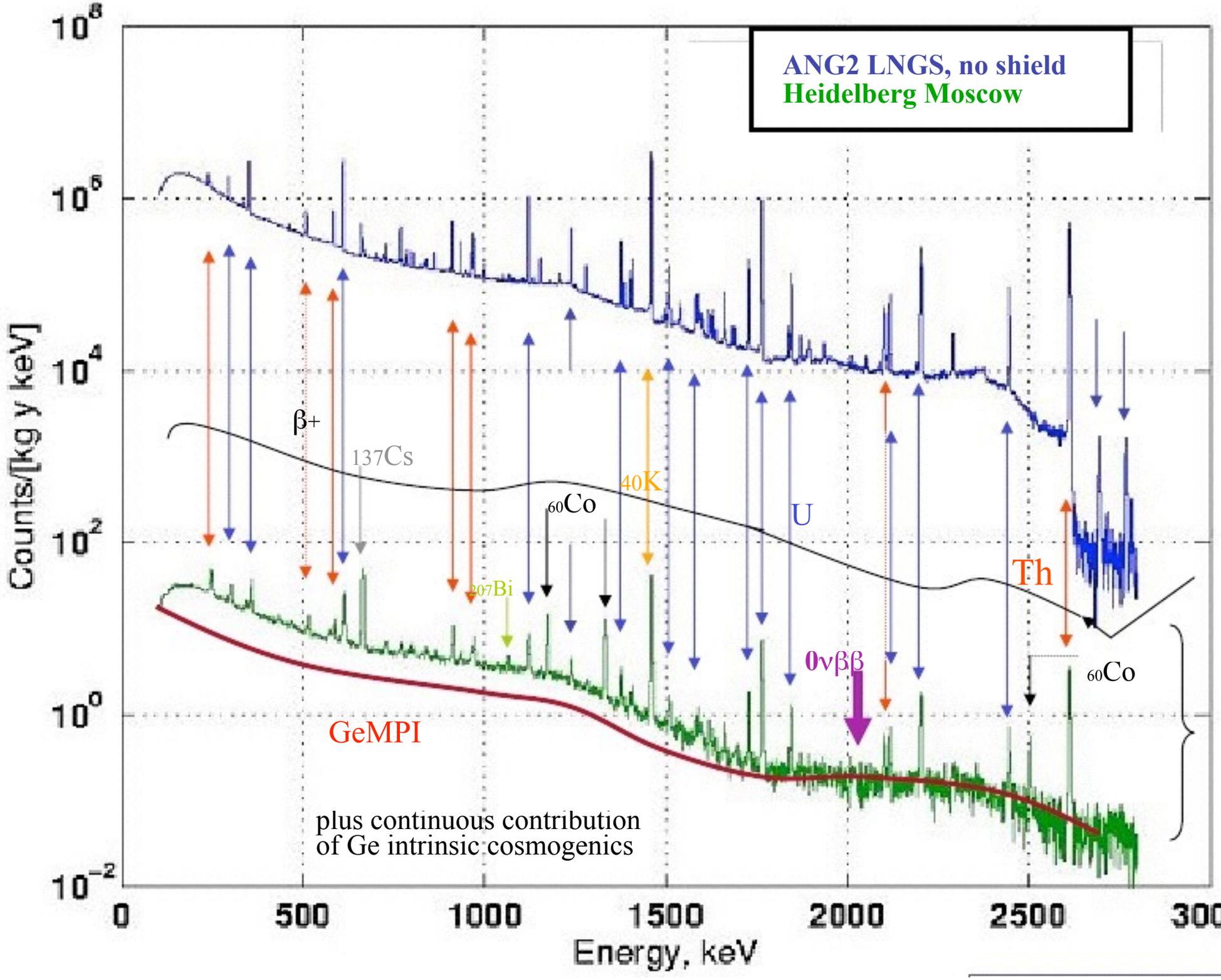


Example: sample output

```
sample:          plastic scintillator slabs, MPI-K-HD old NE 102A
weight:          7.30 kg
live time:               1963974 s
detector:                GeMPI

radionuclide concentrations:

Th-232:
Ra-228:          (9.2 ± 0.4) mBq/kg              <==>    (2.3 ± 0.1) E-9 g/g
Th-228:          (9.3 ± 0.3) mBq/kg              <==>    (2.3 ± 0.1) E-9 g/g

U-238:
Ra-226           (0.96 ± 0.13) mBq/kg    <==>    (8 ± 1) E-11 g/g
Pa-234m          < 12 mBq/kg             <==>    < 9.9 E-10 g/g

U-235:           < 0.57 mBq/kg           <==>    < 1.0 E-9 g/g

K-40:            (2.2 ± 0.7) mBq/kg              <==>    (7 ± 2) E-8 g/g

Cs-137:          < 0.18 mBq/kg

upper limits with k=1.645,
uncertainties are given with k=1 (approx. 68% CL);

Ra-228 from Ac-228;
Th-228 from Pb-212 & Bi-212 & Tl-208;
Ra-226 from Pb-214 & Bi-214;
U-235 from U-235 & Ra-226/Pb-214/Bi-214
```

only directly visible if Ra is depleted

Going underground



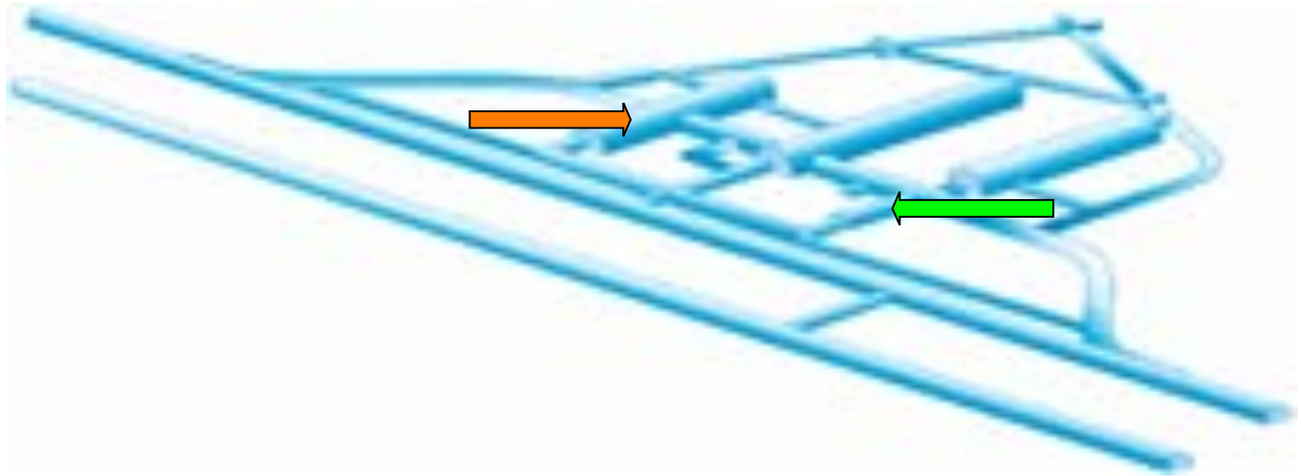
reached at Heidelberg

difference besides higher radiopurity due to: insufficient veto, neutron and muon activation with longer life time than the blocking time and interaction of delayed neutrons

HPGe best installations

LNGS Facility for Low Level Radioactivity Measurements

Laboratori Nazionali del Gran Sasso



- HPGe installed underground
- Shielded with selected materials
- Continuous antiradon flux system

type	volume [cm³]	rel. efficiency	FWHM [keV]
GeBer n-type	235	56%	2.0
GeMi p-type	403	86%	1.9
GePV p-type	363	91%	1.8
GsOr p-type	414	96%	1.9
GeMPI p-type	413	102%	1.9
GePaolo p-type	518	113%	2.0
GeCris p-type	465	120%	2.0
GeMulti p-type	4×225	4×96%	2.0

Intrinsic contamination of (purest) materials

Material	Activity (μBq/kg)				2.6 MeV γ attenuation (10^{-6})
	^{226}Ra (U)	^{228}Th (Th)	^{40}K	various	
lead	≤ 29 a)	≤ 22 a)	270 a)	≤ 4 x 10^3 ^{210}Pb b)	28.5cm
copper	≤ 16 a)	9 c) ≤ 29 a)	≤ 88 a)	≤ 10 60Co a)	40.9 cm
steel	130 a)	≤ 40 a)	50 a)	140 60Co a)	46.2 cm
water	≤ 1 *	0.04 d) 0.008 c)	≤ 2 d)		324 cm
LS (PC)	10^{-6} f)	≤ 10^{-6} f)	10^{-3} f)		373 cm
LN2	≤ 0.3 *		10^{-3} ^{39}Ar g)	0.04 ^{85}Kr g)	443 cm
LAr	600*			106 ^{39}Ar b)	276 cm
LXe				1 ^{85}Kr i)	120 cm

a) GeMPI; b) bolometric Milano; c) Ge PNNL; d) ^{232}Th by ICP-MS Ispra; e) ^{232}Th by NAA TU München; f) ^{226}Ra + ^{228}Th by Bi-Po Borexino; * ^{222}Rn MPI-K; g) Rare Gas MS MPI-K h) PC Bern, WARP; i) XMASS

High sensitivity RNAA

High sensitivity measurements on natural contaminants mean

$$n + K^{41} \rightarrow K^{42} \xrightarrow[12.36h]{\beta^-} Ca^{42} \qquad \sigma_{th} = 1.2\ b$$

$$n + Th^{232} \rightarrow Th^{233} \xrightarrow[22.3m]{\beta^-} Pa^{233} \xrightarrow[27.0d]{\beta^-} U^{233} \qquad \sigma_{th} = 6.1\ b$$

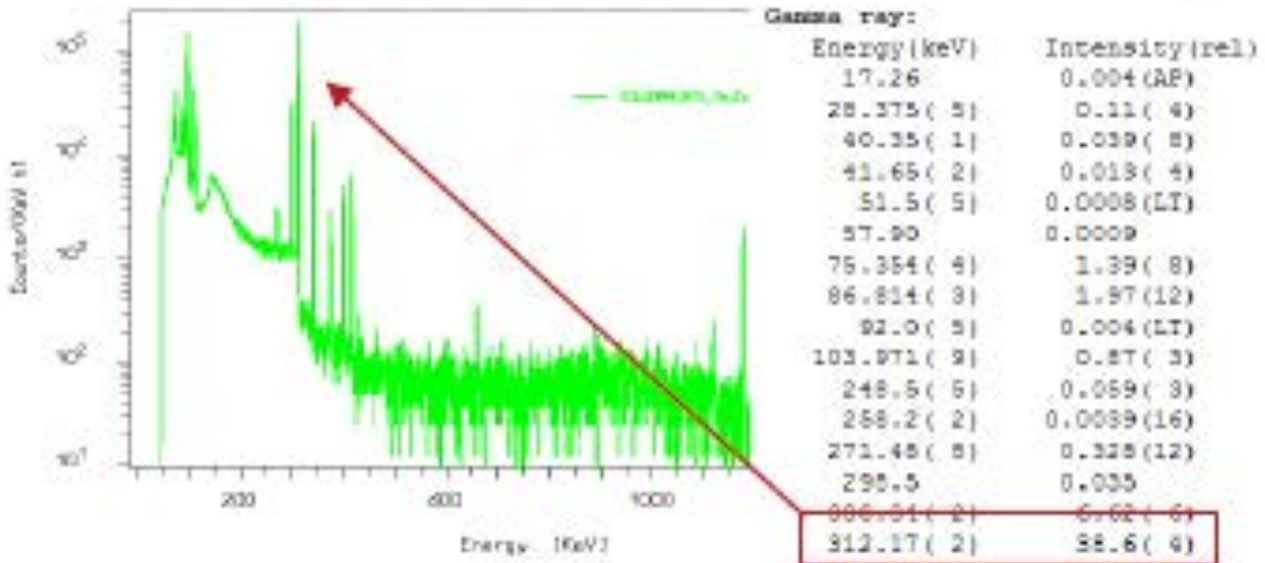
$$n + U^{238} \rightarrow U^{239} \xrightarrow[23.5m]{\beta^-} Np^{239} \xrightarrow[2.36d]{\beta^-} Pu^{239} \qquad \sigma_{th} = 2.3\ b$$

Neutron cross section are reasonable
Half lifetimes are long enough to measure the produced daughters

NAA: copper

- Copper is one of the reference materials for low background experiments
- It is a very pure materials and will be use for many parts of detector construction
- The most interesting radioactive contaminant is ^{232}Th

$$^{232}Th(n,\gamma)^{233}Th \xrightarrow[22,6\ m]{\beta^-} {}^{233}Pa \xrightarrow[27\ d]{\beta^-} {}^{233}U$$



Gamma ray:

Energy(keV)	Intensity(rel)
17.26	0.004(AP)
28.375(5)	0.11(4)
40.35(1)	0.039(8)
41.65(2)	0.018(4)
51.5(5)	0.0008(LI)
57.90	0.0009
75.354(4)	1.39(8)
86.814(3)	1.97(12)
92.0(5)	0.004(LI)
103.971(9)	0.87(3)
248.5(5)	0.059(3)
258.2(2)	0.0039(16)
271.48(5)	0.328(12)
298.5	0.035
300.31(2)	6.62(6)
312.17(2)	38.6(4)
340.51(3)	4.47(4)
375.45(4)	0.679(8)
398.62(8)	1.390(12)
415.76(4)	1.745(16)

Copper: Activation of the sample

Copper neutron activation

Activation of the Copper

$$^{232}Th(n,\gamma)^{233}Th \xrightarrow[22,6\ m]{\beta^-} {}^{233}Pa \xrightarrow[27\ d]{\beta^-} {}^{233}U$$

^{64}Cu $t_{1/2}$ 12.7 h

^{66}Cu $t_{1/2}$ 5.1 m

After irradiation samples are very radioactive

Waiting time 1 month after irradiation

Activation of the trace elements in the Copper

^{75}Se $t_{1/2}$ 120 d

^{192}Ir $t_{1/2}$ 74d

^{198}Au $t_{1/2}$ 2,7d

^{124}Sb $t_{1/2}$ 60d

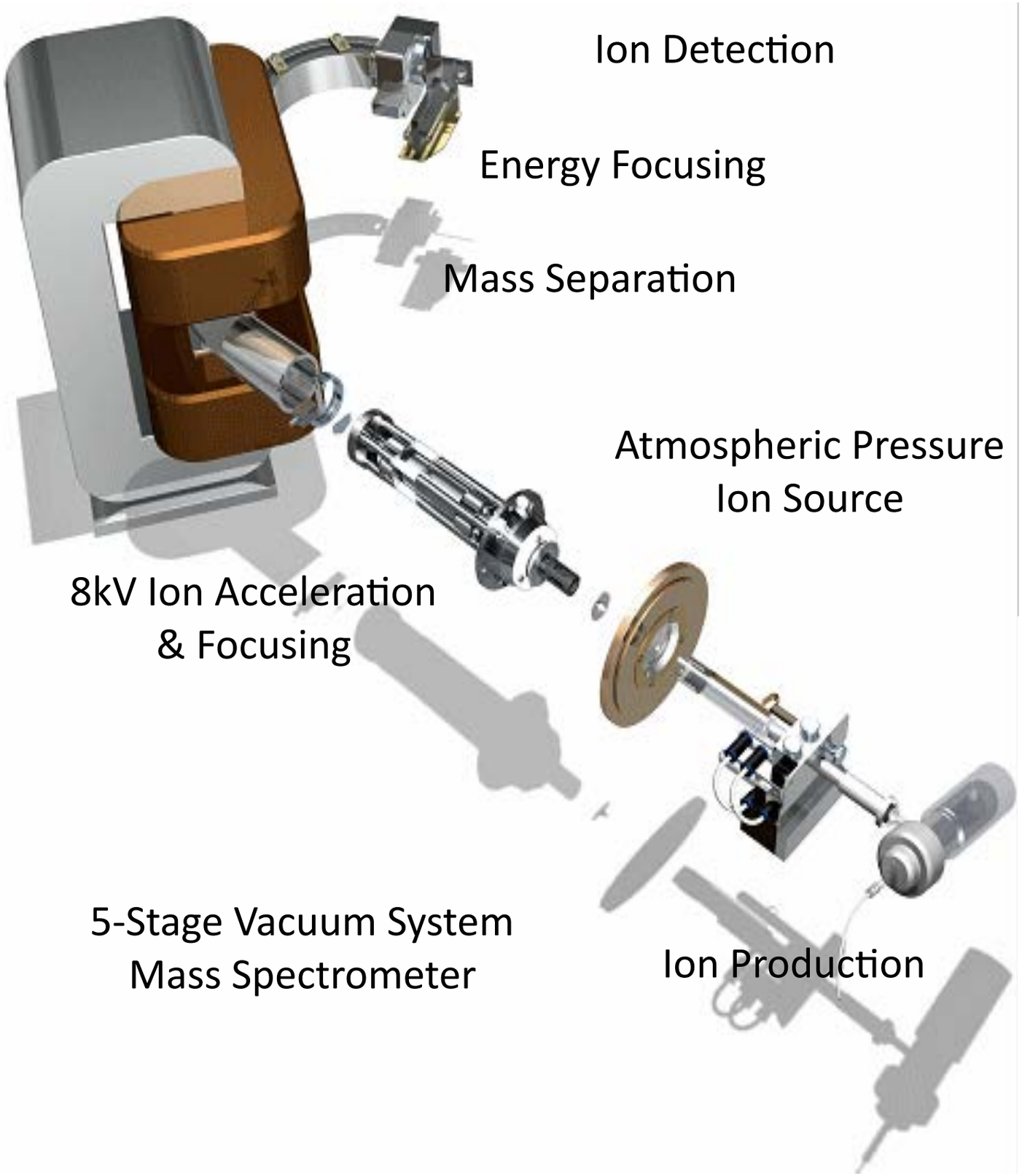
^{110m}Ag $t_{1/2}$ 250d

^{60}Co $t_{1/2}$ 1925d

There are lot of interferences

Separation of the activated element with Ion Exchange Resin

Inductively Coupled Plasma Mass Spectrometry



Ion Detection

Energy Focusing

Mass Separation

Atmospheric Pressure
Ion Source

8kV Ion Acceleration
& Focusing

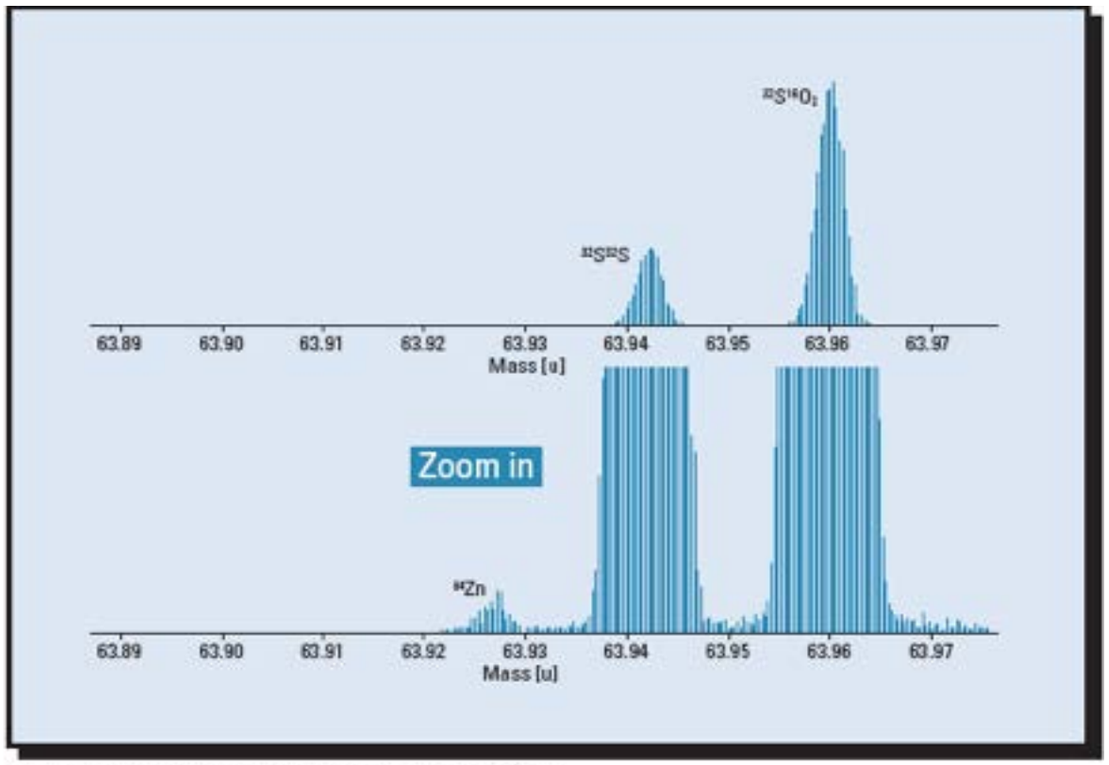
5-Stage Vacuum System
Mass Spectrometer

Ion Production

HR ICP MS Double Focusing

Specifications and Installation Requirements

Sensitivity (Concentric Nebulizer)	> 1 x 10^9 counts per second (cps)/ppm In	
Detection Power	< 1 ppq for non-interfered nuclides	
Dark Noise	< 0.2 cps	
Dynamic Range	> 10^9 linear with automatic gain calibration (ELEMENT 2) > 10^{12} linear with automatic gain calibration (ELEMENT XR)	
Mass Resolution	300, 4000, 10,000 (10 % valley, equivalent to 5 % height); 600, 8000, 20,000 (FWHM)	
Signal Stability	< 1 % RSD over 10 minutes < 2 % RSD over 1 hour	
Scan Speed (magnetic)	m/z 7 to 240 to 7 < 150 ms	
Scan Speed (electric)	1 ms/jump, independent of mass range	
Oxide and Doubly Charged Ions	ratio	measured
	BaO^+/Ba^+	< 0.002
	Ba^{2+}/Ba^+	< 0.03



Zinc in H_2SO_4 (10 % w/w), High Resolution

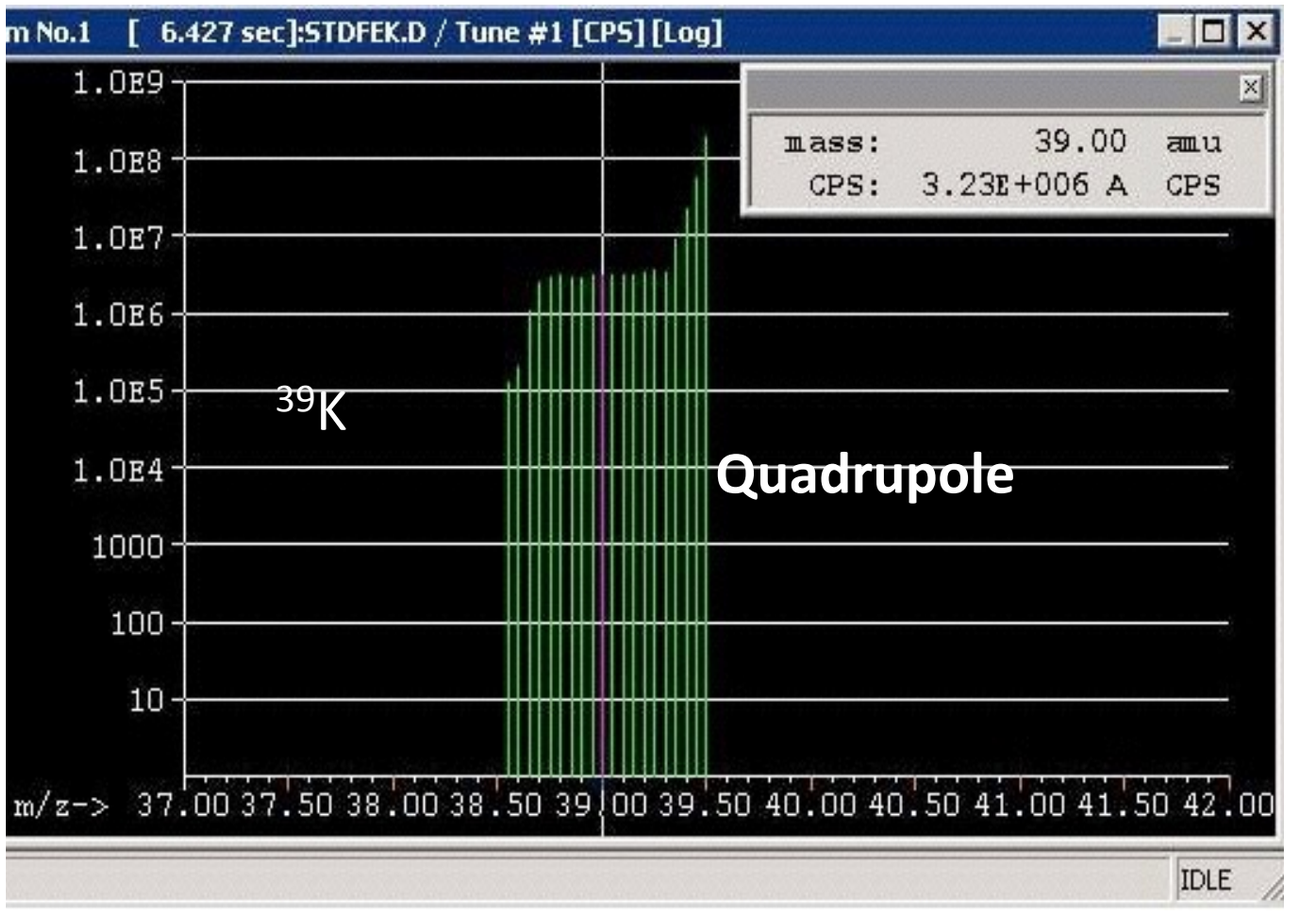
ICP MS for radioactivity measurements

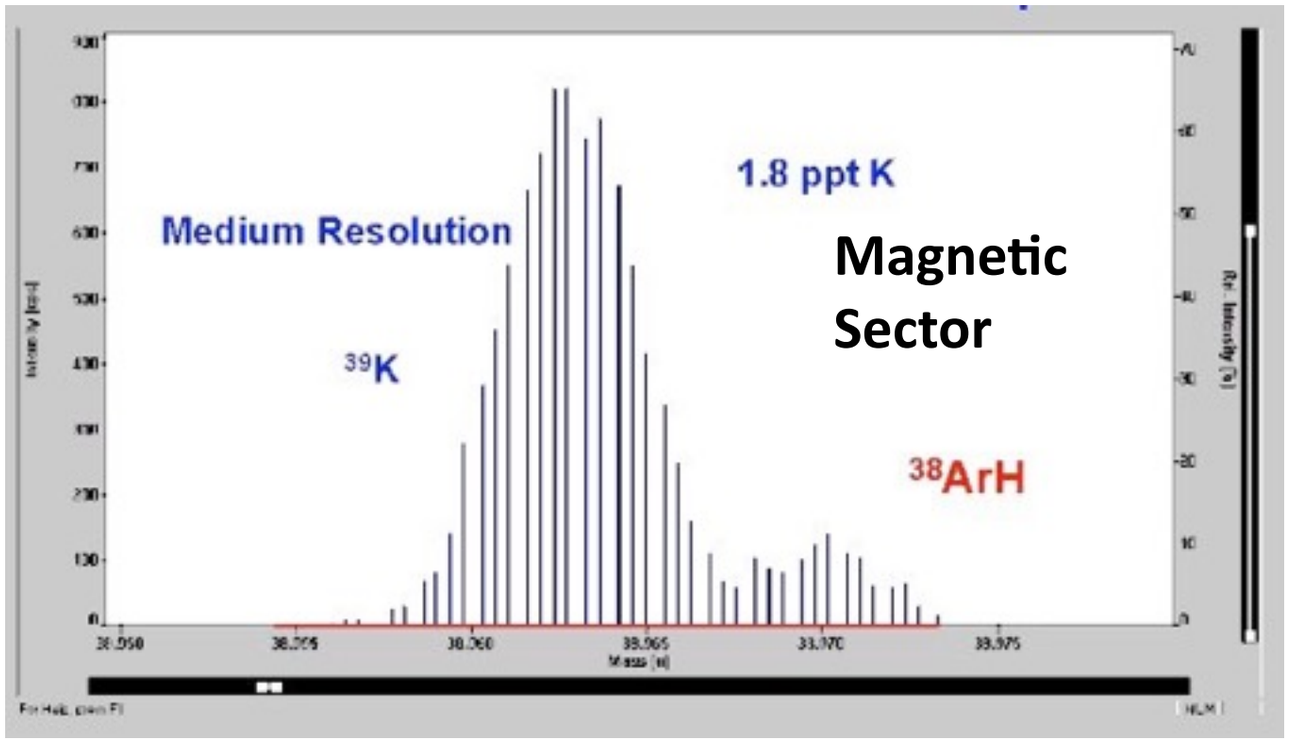
There are various ICP MS instruments:

- quadrupole
- magnetic sector
- multicollector

Effective sensitivity

- quadrupole ~ 10^{-12} g/g
- magnetic sector ~ 10^{-15} g/g
- multicollector ~ 10^{-15} g/g but
- ~ 10^{-6} isotopic ratio



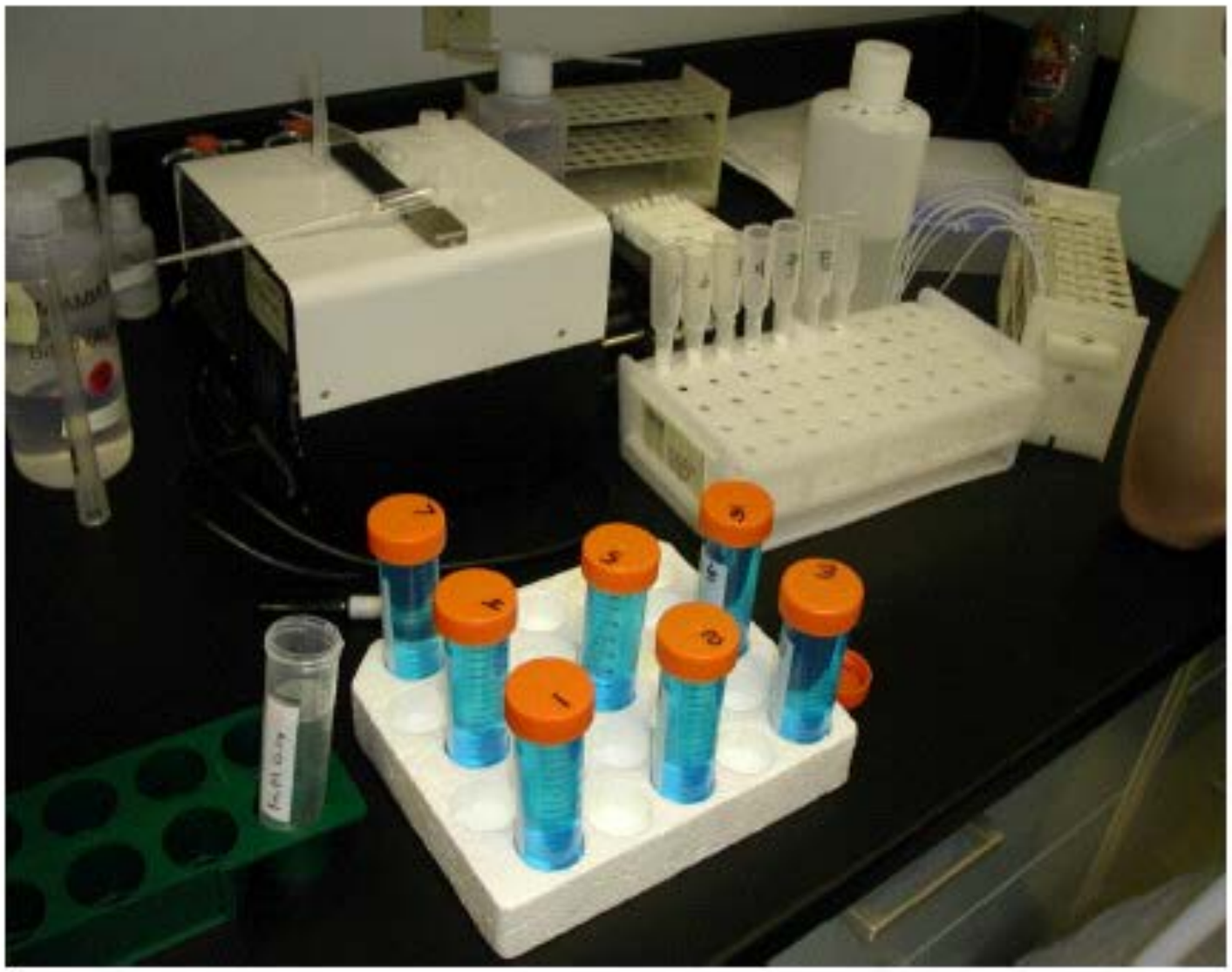


Quadrupole ICP MS can show interferences

- We want to measure ^{39}K
  …but $^{38}Ar^{1}H$ have the same mass
- Real sensitivity ~ 10^{-6} g/g

ICP MS for radioactivity measurements

- It is possible to perform high sensitivity measurement on copper
- Also in this case the pre-concentration of the interesting elements is important



- Analysis of 7 aliquots from a single copper sample dissolved in nitric acid

- 10 ml columns loaded with 0.8 ml of TRU resin using Millipore LC 10 µm filter to retain resin

- Work performed manually on bench top with open columns

ICP MS for radioactivity measurements

In this case the purity of the acids used is very critical!

1. Condition column
   - 1.0ml of 2.5M nitric acid

2. Load the sample
   - 50ml total volume comprised of 25ml of 15M nitric acid, 25ml DI water, and 0.1ml of 10^{-12} g/ml ^{229}Th tracer

3. Wash the column
   - 2.0ml of 2.5M nitric acid

4. Strip column
   - 5 aliquots of 0.5ml 1mM Bioxalate
   - Save and combine aliquots
   - Acidify with 0.025 ml of 15M nitric acid
   - Analyze solution by ICP/MS

5. Wash the column
   - 30 ml DI water to remove any bioxalate

ICP MS for radioactivity measurements

	Ave of µBq ^{232}Th/kg in Blanks	µBq ^{232}Th/kg of Starting Anode Cu	µBq ^{232}Th/kg of PNNL Electroformed Cu
Column 1	1.0	1.7	1.6
Column 2	0.5	1.6	1.2
Column 3	0.6	1.4	0.9
Column 4	0.5	1.5	2.0
Column 5	0.5	1.8	1.5
Column 6	0.4	1.0	1.3
Column 7	0.6	1.3	0.9
Ave	0.6	1.5	1.3
Std Dev	0.2	0.2	0.4
% Std Dev	34.9	16.8	30.2

Ratio between starting to electroformed copper was expected to be much larger!

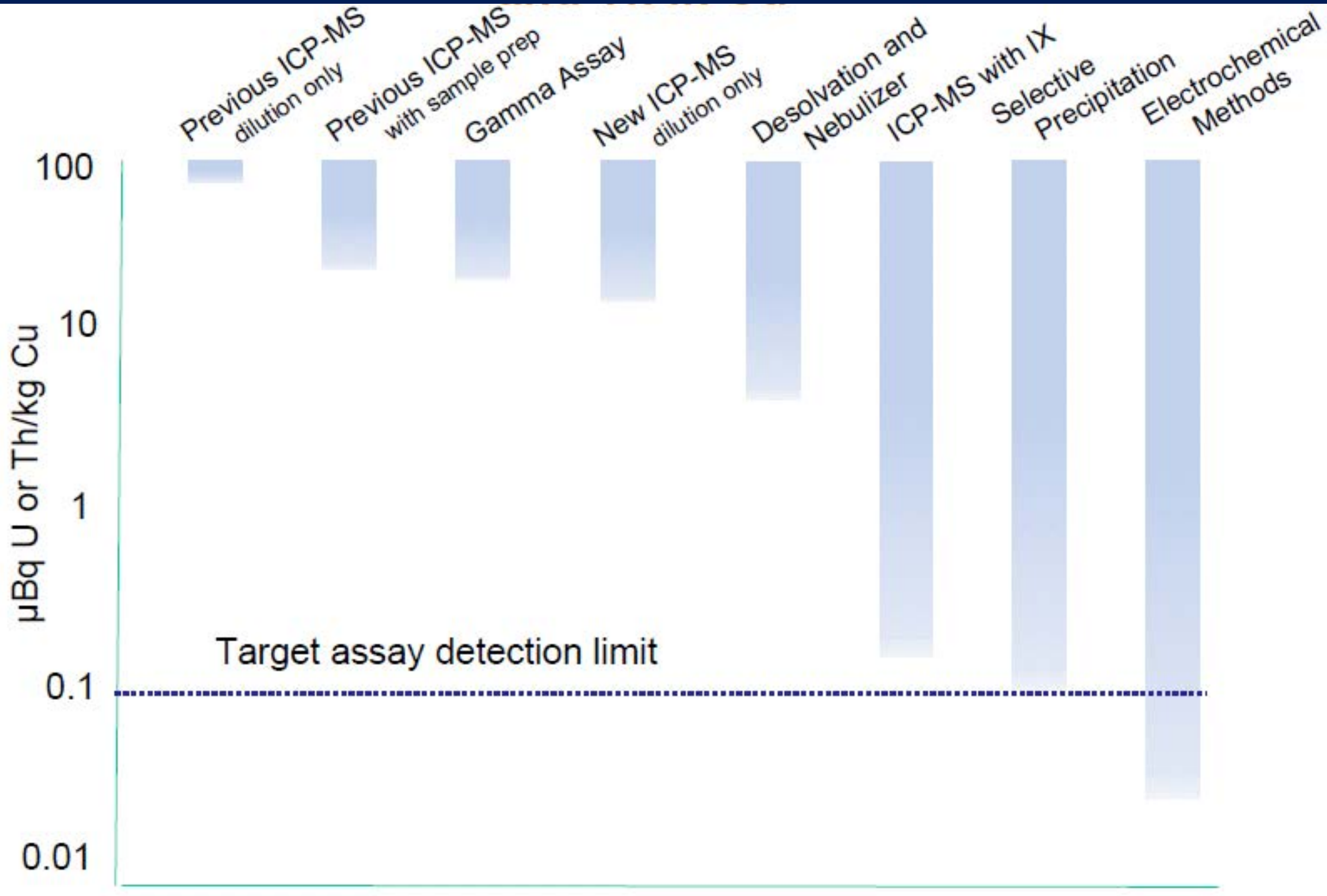
Analysis of electroforming bath solution using precipitation techniques found ave 77µBq ^{232}Th/liter

Indicated rejection ratio of ~10^2-10^3 which is consistent with Journal of Radioanalytical and Nuclear Chemistry 277(1):103, 110. doi:DOI: 10.1007/s10967-008-0716-5

- ~0.6 µBq ^{232}Th/kg Cu DL (0.15 x 10^{-12} pgTh/gCu)

Problem can be found on the evaluation of systematic errors

ICP MS evolution

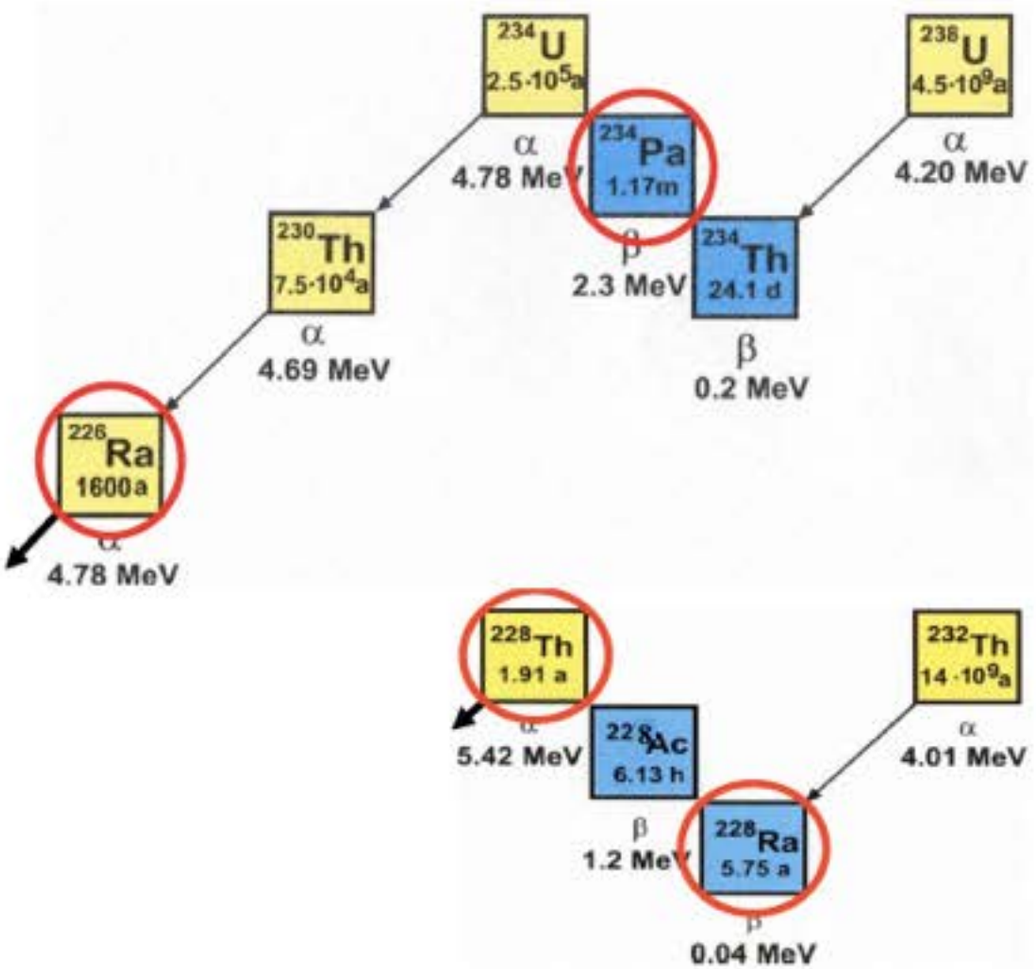


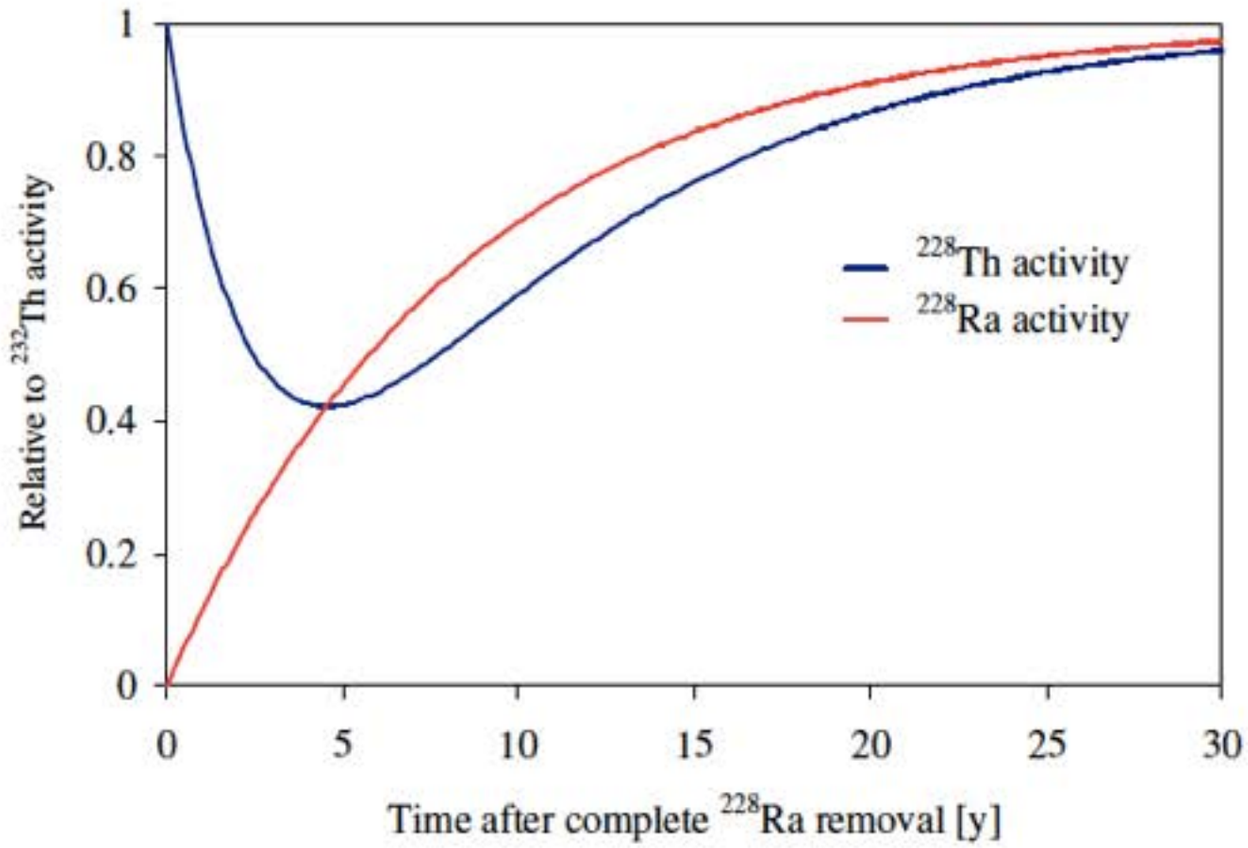
Disequilibrium in natural chains

Radium is very reactive

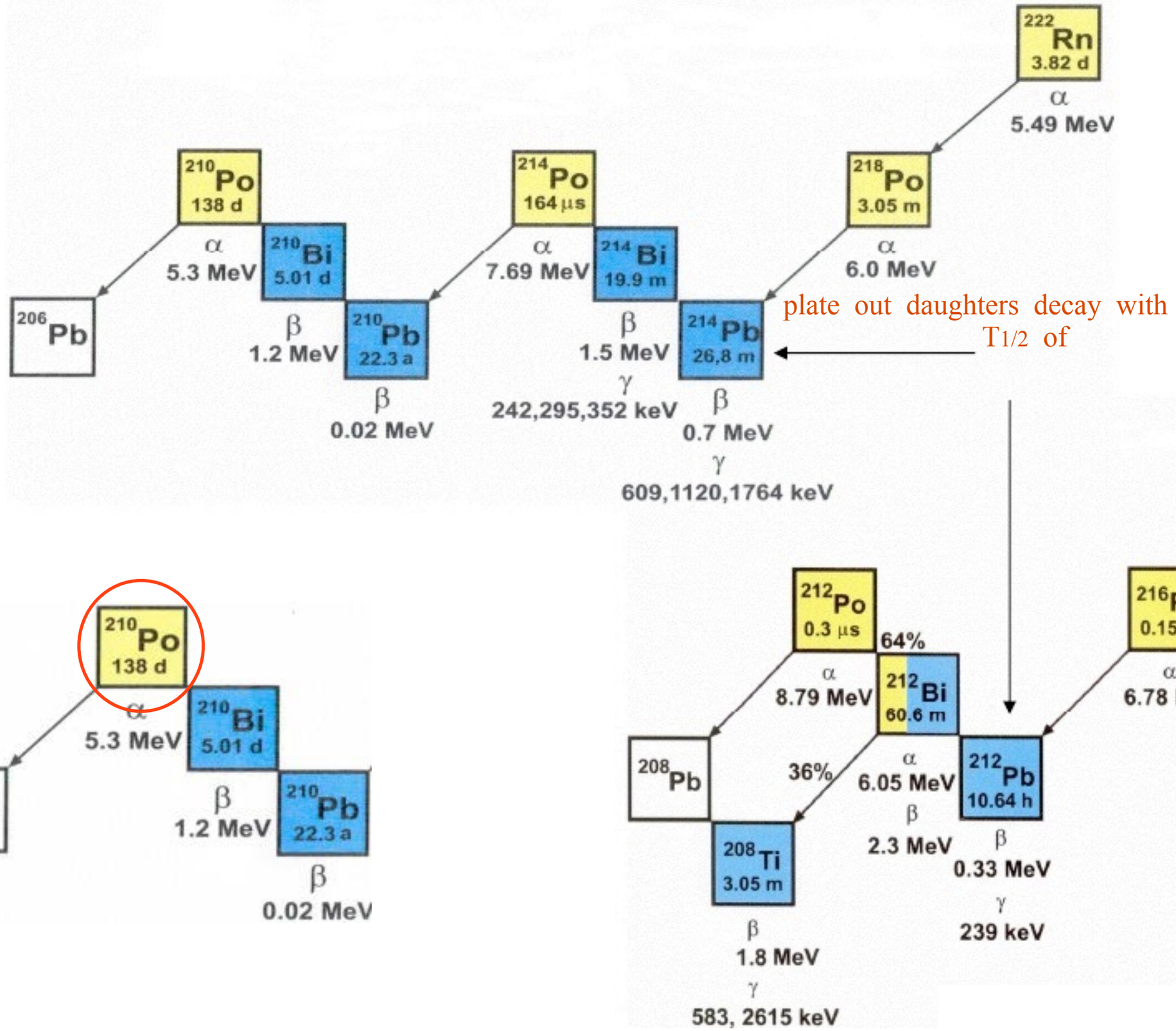
- It can be easily extracted or introduced in a material
- Chain disequilibrium

sample	activity of U/Th progenies [mBq/kg] and the irratios					
	^{234m}Pa	^{226}Ra	^{234m}Pa/^{226}Ra	^{228}Th	^{228}Ra	^{228}Th/^{228}Ra
WTS steel	121±18	0.99±0.15	122±26	3.7±0.4	1.7±0.2	2.18±0.35
pre WW1 steel	5.7±1.4	0.15±0.02	38±11	0.46±0.07	0.47±0.05	0.98±0.18





Radon and its progenie



Radon

A very dangerous enemy:

^{220}Rn ($\tau 1/2$=56 sec)... → ^{212}Pb ($\tau_{1/2}$=11 h)

^{222}Rn ($\tau 1/2$=3.8 d)... → ^{210}Pb ($\tau_{1/2}$=22 y)

^{222}Rn concentration is higher in an underground facility than on surface.

- Kamioka mine tunnel: ~1200 Bq m-3 [Phys. Lett. 452, 418 (1999)]
- SNO underground lab: 123±13 Bq m-3or 60,000 atoms/l.
- Surface ~1/10 to 1/20 as much

Why Rn is a problem?

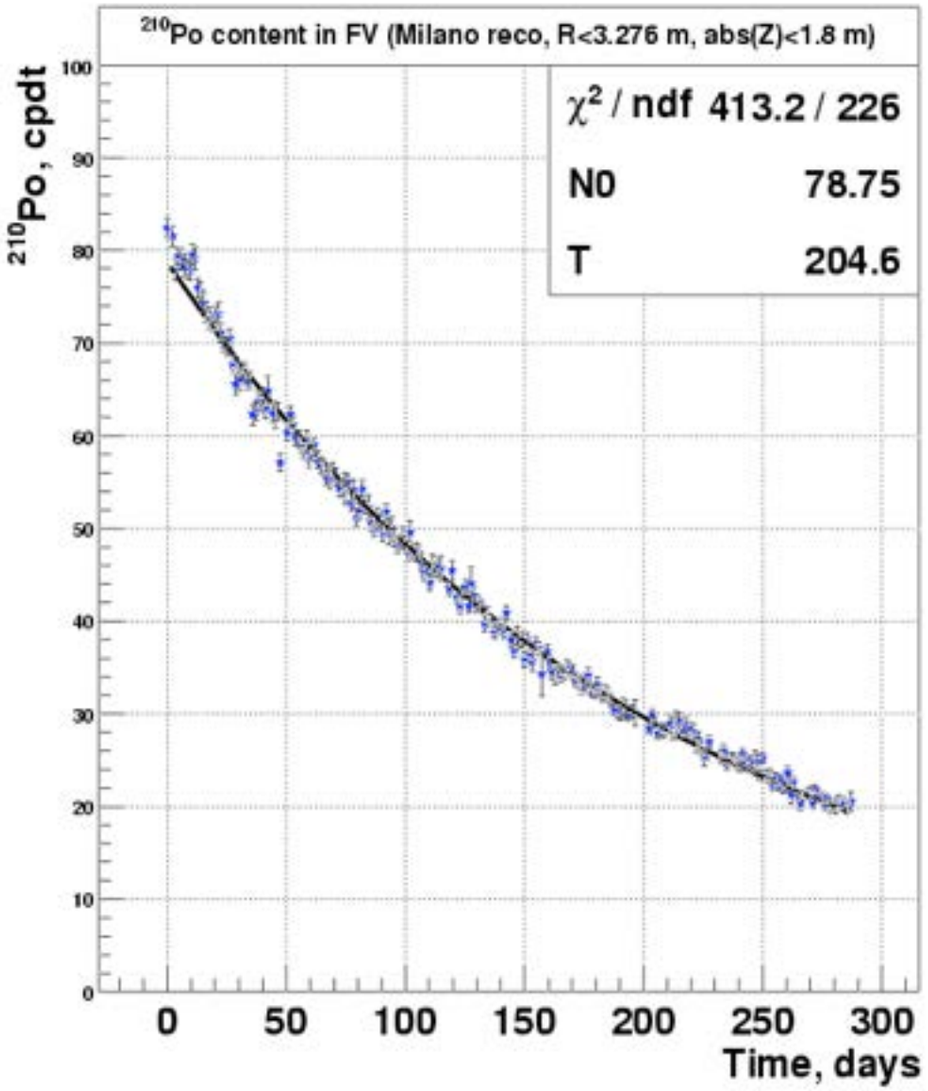
- Rn daughters plate out electrostatically onto surfaces
- Some fraction might be transferred to active detector volume by leaching

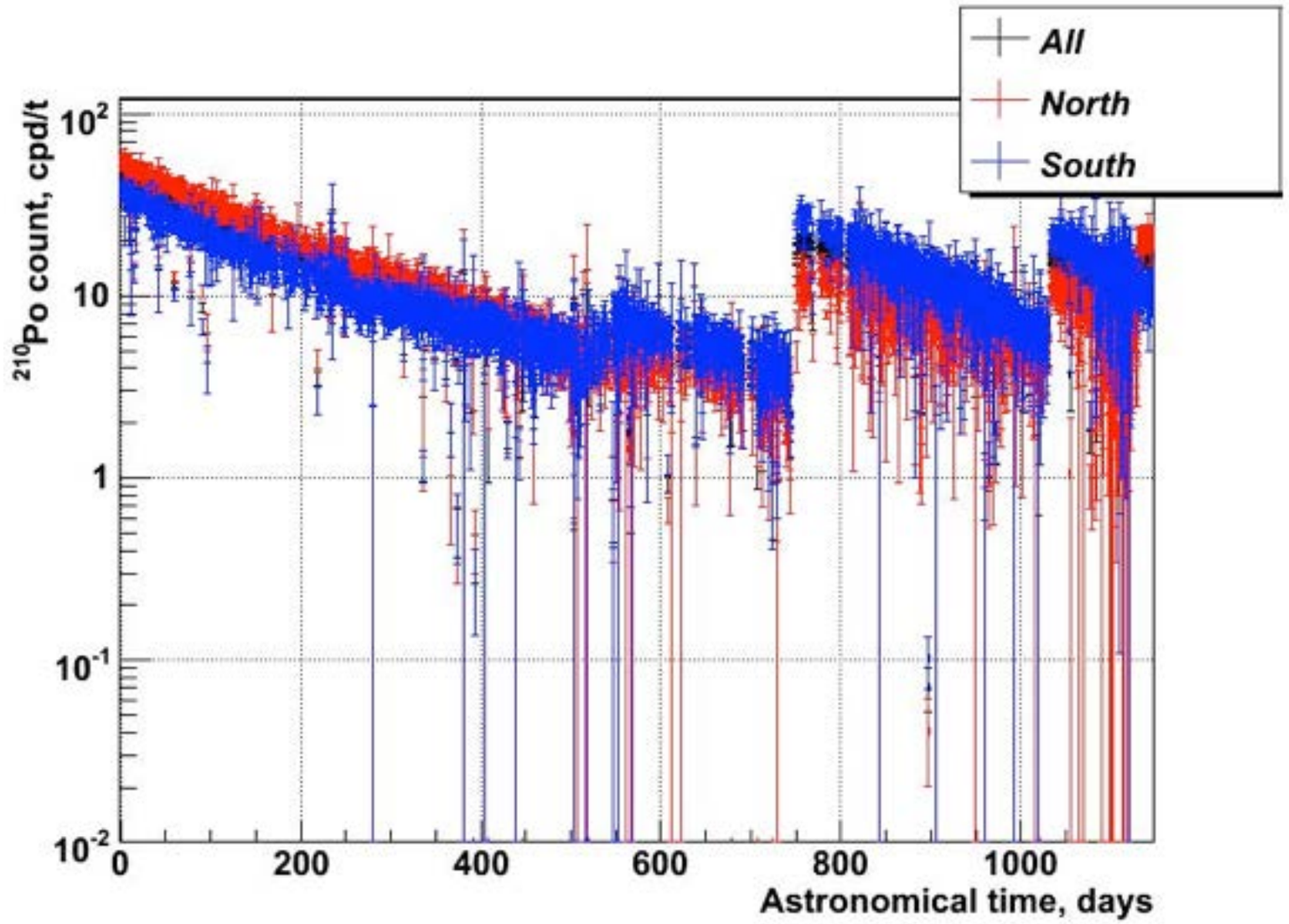
Techniques based on charcoal adsorption have been used which suppress Radon by about a factor of 104-105.

- Pocar et.al. at Princeton developed a pressure swing system
- Lalanne et.al at Modane developed a cryogenic adsorption system

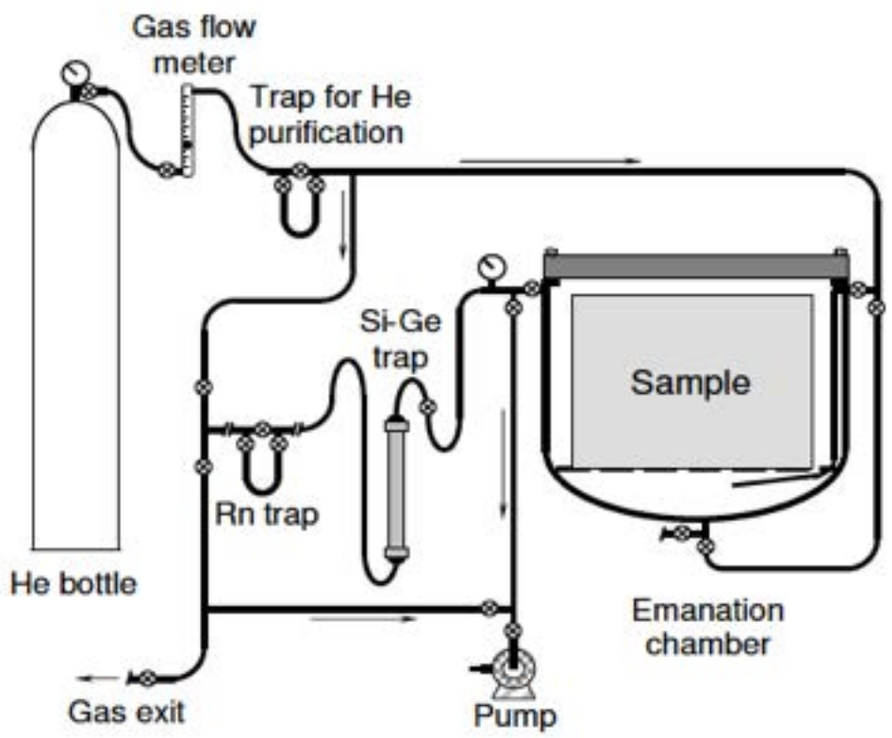
^{210}Po

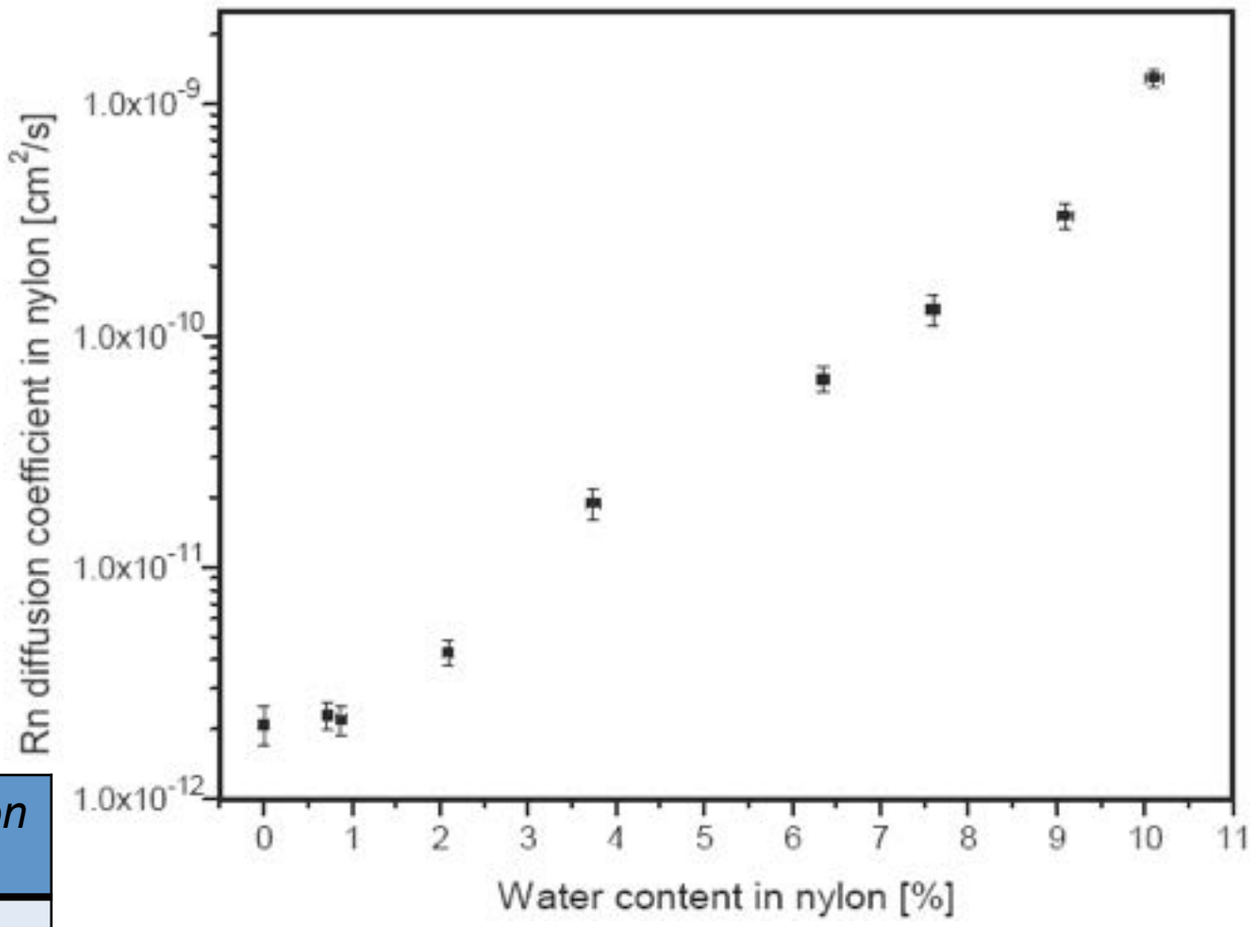
- ^{210}Po belongs to ^{238}U (^{222}Rn) progenie
- It is reactive and shows clear selective affinity with different materials





^{222}Rn (^{226}Ra) assay system





sample	*surface concentration (μ Bq/m²)*	*bulk concentration (μBq/kg)*
steel foil (untreated) (71 m²)	10 ± 1	640 ± 210a
after rinsing with H_2O	5 ± 1	
N2 sparging column (280 m²)	8 ± 1	
Sniamid/Capron foil (0.1 mm, 208 m²)	3 ± 1	100 ± 20b*
Sniamid nylon (0.125 mm,130 m²)	< 0.8	< 21b

U and Ra out of equilibrium

a measured by Ge g spectroscopy (expected emanation rate by a-recoil: 0.2 μBq/m²)
b measured via ^{222}Rn emanation under wet condition (enhanced permeability)
* corresponds to ~ 8 ppt U-equivalent - but ~ 2.5 ppt U measured with ICP-MS

Grzegorz Zusel's smart idea:

- measure dry foil to get signal from
- surface water saturated foil for bulk concentration

^{222}Rn diffusion, permeability and solubility

Material description	Diffusion coeff. D [10^{-10} cm^2/s]	Solubility coeff. S	Permeability coeff. P [10^{-8} cm^2/s]	Diffusion length coeff. d_e [mm]
Polyamidsupronyl	6.1	3.4	0.2	0.1
Plexi	6.2	8.2	0.5	0.1
Kalrez	12	12.1	1.4	0.24
Teflon	14	2.3	0.3	0.3
Butylrubber	49	4.4	2.1	0.5
Rubbersoft	1000	12	120	2.2
Naturalrubber	17000			9.0
PUhard	88	7.9	6.9	0.6
PUsoft	408	5.6	23	1.4
PVChard	140	5.2	7.3	0.8
PVCsoft	420	10	42	1.4
ApiezonMgrease	1640	1.6	26.3	2.8
Silicongrease	21200	18.4	3900	10.0

MPI-K and Krakow

PE – Polyethylene
PU – Polyurethane
PVC – Polyvinylchloride

$$d_e = (D/\lambda_{Rn})^{-2}$$

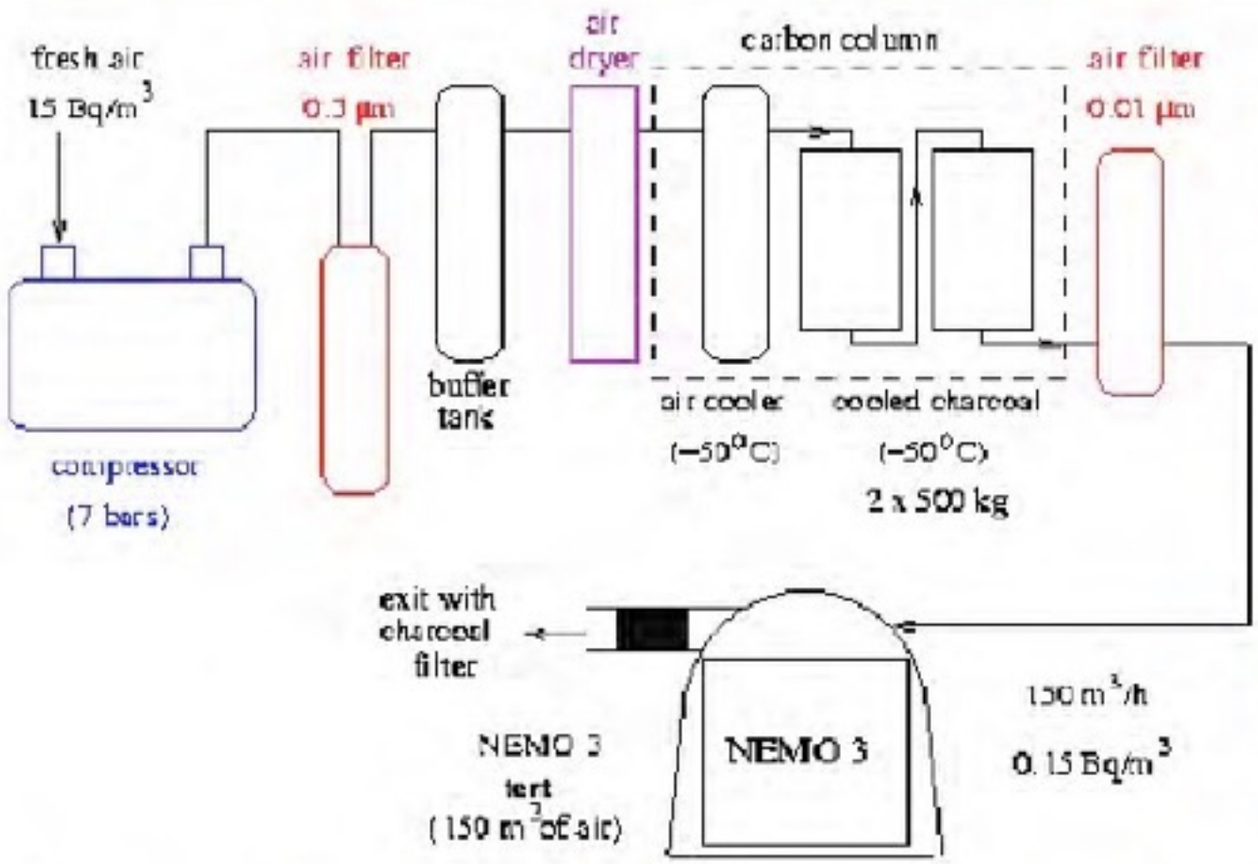
^{222}Rn emanation data

Material	Sample	Rn emanation [mBq/m^2]
Polyurethan 90°	Shore 1 O-ring, 8.4 mm * 715 mm, 0.06 m^2, 150 g	< 0.42
Polyurethan 70°	Shore 5 flat gasket discs, 0.27 m^2	< 0.29
Butyl rubber 65°	5 O-rings, 8.4 mm * 715 mm, 0.3 m^2, 791 g	13 ± 1
Butyl rubber 65 GD	3 flat gasket discs, 0.16 m^2	59 ± 3
Butyl rubber 65 GD	O-ring, 4 mm * 16 cm	< 30
Viton	1 O-ring, 8.4 mm * 715 mm, 0.06 m^2	322 ± 8
Viton GF, Batch 97100801	10 O-rings, 3.3 mm * 38 mm, 0.014 m^2, 23.6 g	75 ± 4
Silicon rubber	10 O-rings, 2.7 mm * 57 mm, 0.015 m^2, 16g	196 ± 4
Teflon-coated silicon rubber 10	O-rings, 3..5 mm * 57 mm, 0.015 m^2, 16g	11 ± 2
Teflon foil	Foil, 28,5 m^2	< 0.009
PCTFE	Material for valve seat	< 0.32
Kalrez	O-ring	7 ± 2
Nitril		190 ± 10
Gylon 3510	1 flat gasket disc, 0.006 m2, 11.8 g	207 ± 4
Busto	1 flat gasket disc	1750 ± 40

MPI-K

Modane

Principe:
Air circulation (150 m^3/h) trough a column of charcoal cooled down at -50°C



The specifications of the radon trapping facility are :

- Compressor (7 bars)
- filtration with oil separator (0.03 µm) and dust separator (0.1 µm)
- Air Dryer with a dew point -70°C for 8.5 bars -30°C at maximum value
- Cooling unit
- Two adsorption columns, with internal diameter of 600 mm and 3 m high
- Charcoal: activated carbon 2x500 kg

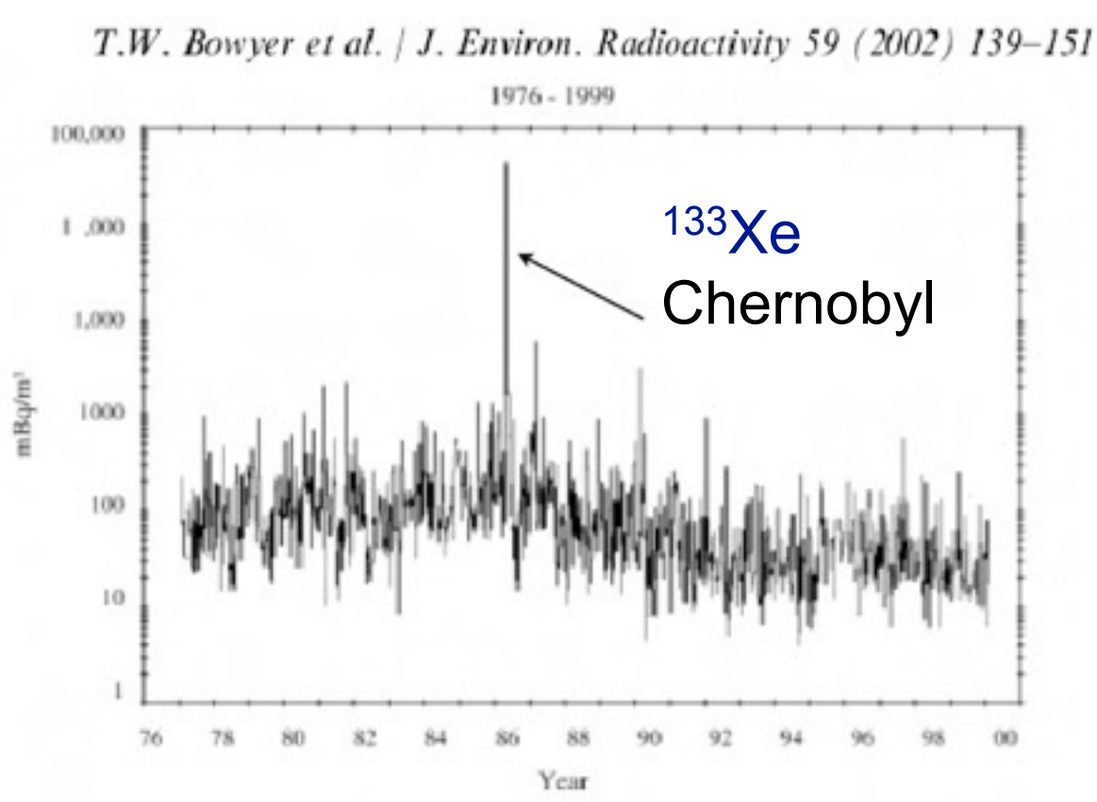
The Rn level at exit of the column: 18 mBq/m^3 ⟹ air sent into the tent

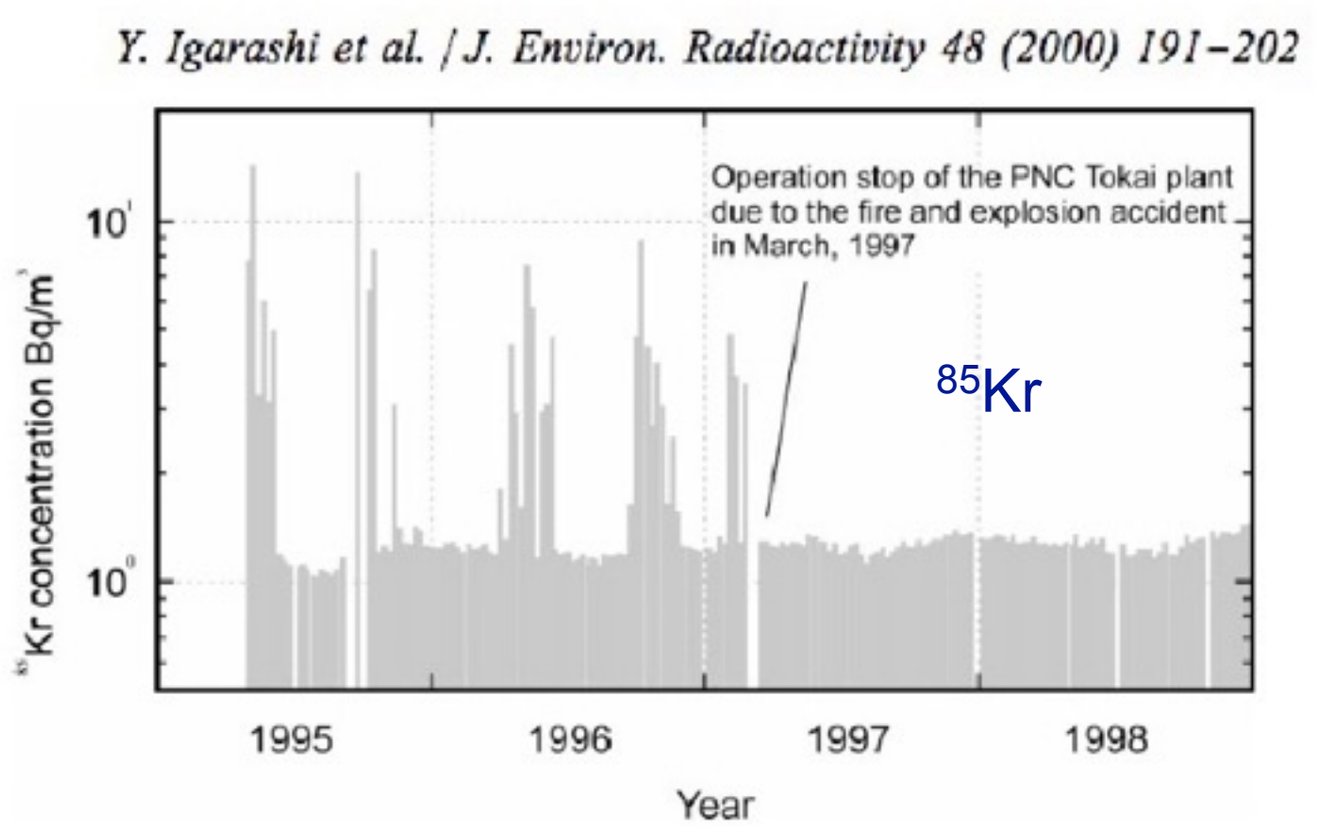
Ancient Lead: ^{210}Pb

lead sample	weight	time	specific activity [∝Bq/kg]				
	[kg]	[d]	^{226}Ra	^{228}Th	^{40}K	^{207}Bi	^{210}Pb
DowRun	144.6	101.7	<29	<22	440±140	98±24	$(2.7\pm0.4)\times10^{7}$
Boliden	144.3	75.0	<46	<31	460±170	<13	$(2.3\pm0.4)\times10^{7}$
roman	22.1	37.2	<45	<72	<270	<19	$<1.3\times10^{6}$
	bolometric measurement: Alesandrello et al.: NIMB142 (1998)163						$<4\times10^{3}$

Airborne radioactivity

- Mainly spallation products from nitrogen and oxygen
- Also airborne radioactivities (^{3}H, ^{14}C, ^{85}Kr, ^{90}Sr, ^{137}Cs) from previous nuclear weapon testings and nuclear power generation.





- Problematic for certain next-generation dark matter (^{39}Ar and ^{42}Ar) and solar neutrino experiments (^{85}Kr).

Airborne radioactivity

TABLE 11 Atmospheric abundance for selected isotopes*

Isotope	Decay (Max. energy)	Half-life	Activity (Bq/kg)	Flux in atmosphere (atoms cm^{-2} s^{-1})
^{3}H	β^- (18.6 keV)	12.35 y	0.25	
^{7}Be	E.C. (478 keV)	53.4 d	—	8.1×10^{-3}
^{10}Be	β^- (556 keV)	1.6×10^{6} y	—	0.036
^{14}C	β^- (156 keV)	5730 y	400–500	2.2
^{22}Na	β^+ (1275 keV)	2.6 y	—	6×10^{-5}
^{26}Al	β^+ (1809 keV)	7.16×10^{6} y	—	1.7×10^{-4}
^{35}S	β^- (167 keV)	7.16×10^{6} y	—	1.4×10^{-3}
^{36}Cl	β^- (709 keV)	3.0×10^{5} y	—	1.1×10^{-3}
^{39}Ar	β^- (565 keV)	269 y	10^{-6}	Negligible
^{42}Ar	β^- (3525 keV from ^{42}K)	32.9 y	$(0.1–7.0) \times 10^{-6}$	Negligible
^{85}Kr	β^- (514 keV γ emission)	10.7 y	0.5–0.7	Negligible

*Values for ^{39}Ar and ^{42}Ar are from theoretical calculations including the effects of nuclear weapons testing. Value for ^{85}Kr is from direct measurements made in the Northern Hemisphere in 1985.

Annu. Rev. Nucl. Part. Sci. 2004. 54:361–41

Formaggio & Martoff Annu. Rev. Nucl. Part. Sci.54 (2004).361

Networks

- Systems with different overhead burden and intrinsic detector backgrounds. Optimize counting efficiency and throughput.

Example:

- The Collaboration of European Low Level Underground Laboratories (CELLAR)
- Some collaborations have their own smaller networks.

Table 1
List of the underground facilities of the CELLAR network members

Institute	Undergrcund laboratory	Depth (m w.e.)	HPGe-detectors Integral counting rate [d^{-1} kg^{-1}] (40–2700 keV)	Detector type	Main activity
ARC Seibersdorf research (Schwaiger et al., 2002)	(Austria)	ca. 1	8200 ± 200	p-type extended range	Environmental radioactivity, CTBT
Max-Planck-Institut für Kernphysik, Heidelberg (Heusser, 1986)	Low-level laboratory (Germany)	15	2012 ± 23	p-type coaxial	Rare events research and detector development
IAEA-MEL (Povinec, 2002)	CAVE (Monaco)	35	840 ± 50	p-type well	Environmental radioactivity
VKTA (Niese et al., 1998)	Felsenkeller (Germany)	110	3870 ± 30	p-type well	Environmental radioactivity
University of Iceland (Theodórsson, 2003)	(Iceland)	350	—	—	Studies of background components in radiation detectors
IRMM (Hult et al., 2003)	HADES (Belgium)	500	260 ± 4	p-type coaxial	Reference measurements
PTB (Neumaier et al., 2000)	UDO in the salt mine Asse (Germany)	2100	277 ± 4	p-type extended range	Reference measurements
LNGS (Arpesella, 1996)	Gran Sasso (Italy)	3800	87 ± 1	p-type coaxial	Radiopurity of construction materials to support to rare event experiments
LSCE (Reyss et al., 1995)	Modane (France)	4800	30 ± 1 186 ± 2	p-type coaxial p-type well	Environmental radioactivity

M. Laubenstein et al. / Applied Radiation and Isotopes 61 (2004) 167–172

Radio-purity database

Welcome to the ILIAS database on radiopurity of materials

This site is part of the ILIAS program, a common European project for the development of the underground science.

Data sources: *UKDM Collaboration (Boulby Mine), ANAIS, CAST, ROSEBUD Collaborations (Canfranc Underground Laboratory), EDELWEISS (Laboratoire Souterrain de Modane), BOREXINO (Gran Sasso)*

The database provides access to radionuclide concentration in materials commonly used in low background experimental set-ups. The measurements have been performed by groups involved in experiments which require extremely low backgrounds.

While this data is an useful guide for material selection, the radionuclide concentration may vary depending on the lot. Therefore, it is necessary to control all used materials when building low background set-ups.

Search radionuclide concentration:

Acrylic Search

http://radiopurity.in2p3.fr/

An example: photomultipliers

- PMT

Material	Measured by	Method	^{238}U (ppb)	^{232}Th (ppb)	^{40}K (ppm)	Comments
PM bulb (Hamamatsu R5912)	Supplier's data	GES?	81	74	97	Measurements reported as Bq/kg or (raw) counts/1000s/100g.
PM bulb (Hamamatsu R7081)	Supplier's data	GES?	537	36	122	
PM tube (Burle 5")	J. C. Barton	GES	< 200	< 200	~ 2000	assume 100g glass.
PM tube, ceramic (EMI) `small'	J. C. Barton	GES	~ 200?	~. 200?	50(50)	U + Th 400 ppm
PM tube, ceramic (EMI) `large'	J. C. Barton	GES	~250?	~250?		U + Th 500 ppm
PM tube, low b/g (EMI)	Supplier's data	GES?	250(40)	200(50)	90(40)	
PM tube, low b/g (EMI 5" 9390B53)	Supplier's data		27(20)	33(8)	74(16)	
PM tube (ET 9823B)	Supplier's data	GES?	245(9)	280(9)	340(17)	
PM tube (ET 9823QB)	Supplier's data	GES?	215(13)	240(13)	1400(110)	quartz window
PM tube, 8" (ET 9354B)	Supplier's data	GES?	30(17)	30(10)	70(17)	low b/g
PM tube (Hamamatsu R1250)	Supplier's data	GES?	307	306	17900	Measurements reported as Bq/kg or (raw) counts/1000s/100g.
PM tube (Hamamatsu R1250)	Supplier's data	GES?	309	306	464	"Low RI" faceplate. Measurements reported as Bq/kg or (raw) counts/1000s/100g.

http://radiopurity.in2p3.fr/

Details

Nuclear β decay

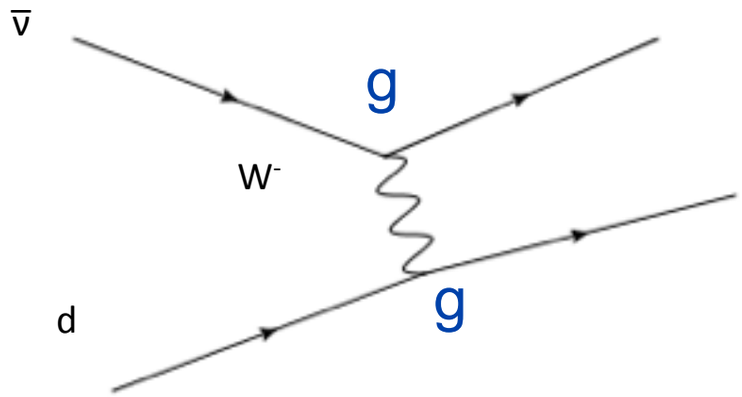
Free neutron decay is the prototype for weak interactions:

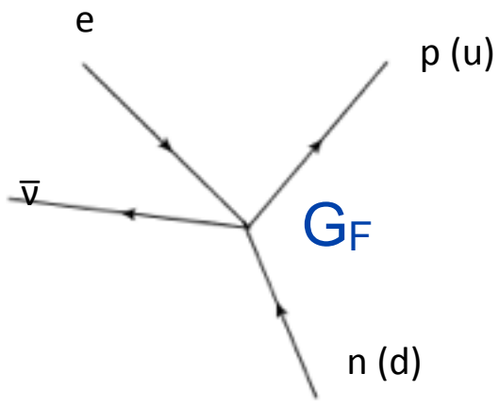
$$n \rightarrow p + e + \bar{\nu}_e$$

or in terms of quarks:

$$d \rightarrow u + e + \bar{\nu}_e$$

Within particle SM it is described by the exchange of a $W^{\pm}$ boson





However, when $q^2 << M_W^2$ it can be approximated as point like (Fermi theory)

Fermi 1934: in analogy with QED, weak interactions are described as a point-like 4 fermion interactions (2 vector currents),with an effective coupling constant G_F $(=g^2/M_w^2)$.

Nuclear β decay

According to Fermi 2nd golden rule the transition probability is given by

$$W = \frac{2\pi}{\hbar} G^2 |M|^2 \frac{dN}{dE_0}$$

where

- E_0 is the available energy in the final state,
- dN/dE0 is the final state density (Phase Space factor)
- |M| is the transition matrix element

In the case of neutron decay (also for allowed and super-allowed β-decays)

M ~ k (costante)

In other terms the angular momentum variation determines the transition properties. Lepton angular momenta have only two possible combinations:

J(Leptoni)=0 → Fermi
J(Leptoni)=1 → Gamow-Teller

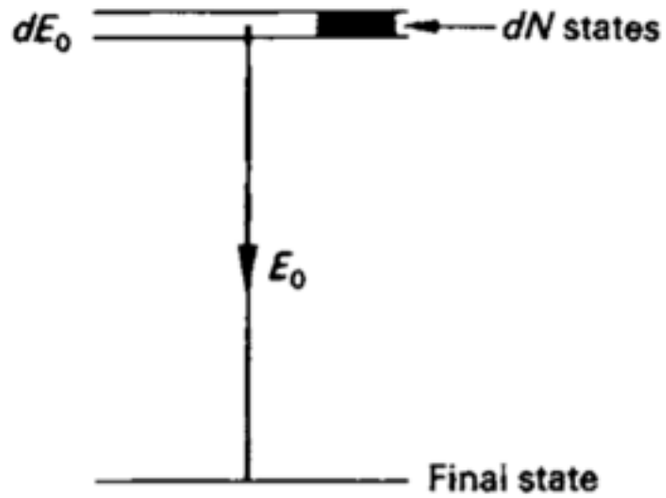
So, is ΔI = 0,1 then no nuclear spin is required and transitions proceed without problems ([super]“allowed”)

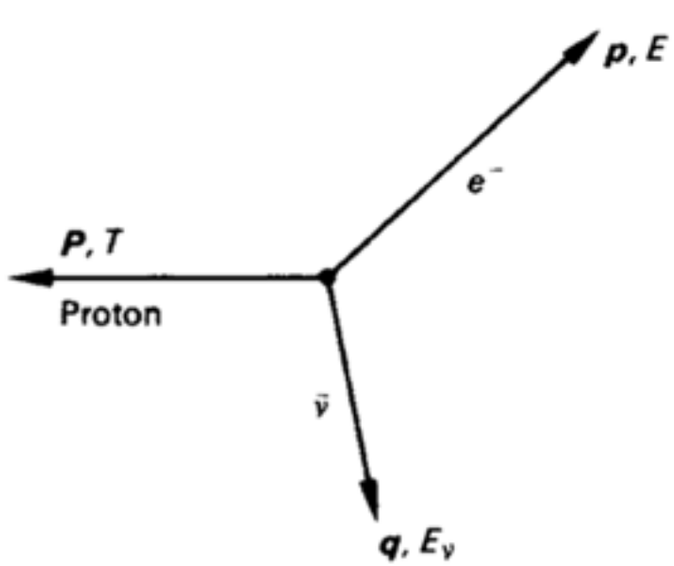
Phase space factor (electron spectrum)

PS is determined by the number of ways in which the available energy can be distributed in the interval $[E_0, E_0+dE_0]$ among final state particles .

If **p**,**q** and **P** are the electron, neutrino and proton momenta and E, E_ν,T their kinetic energies, then by 4-energy conservation (in initial particle rest frame):

$$\mathbf{P} + \mathbf{p} + \mathbf{q} = \mathbf{0}$$
$$E + E_\nu + T = E_0$$





Assuming
- $m_\nu = 0 \rightarrow E_\nu = q$
- $E_0 \sim 1MeV \rightarrow P \sim 1MeV$

If M is the mass of the recoiling nucleon, then its kinetic energy will be:
- $T = P^2/2M \sim 10^{-3}$

i.e. negligible.
Furthermore:
- $E_\nu = E_0 - E = q$

Taking into account that the number of electron states in the interval [p,p+dp] is $\frac{V d\Omega}{(2\pi)^3 \hbar^3} p^2 dp$

Box quantisation

Let's enclose our system in a side L bos. The electron wave function is a plane wave

$$\psi(\vec{r}) = Ne^{i\vec{k}\cdot\vec{r}}$$

Normalization: the probability of finding the electron in volume $V = L^3$ must be equal to 1

$$\int |\psi|^2 d^3\vec{r} = 1 \quad \Rightarrow \quad N = \frac{1}{\sqrt{V}}$$

We must now satisfy the boundary conditions at the box surface:

$$\psi(\vec{r}) = \psi(\vec{r} + L)$$

$$\Rightarrow e^{ik_x L} = e^{ik_y L} = e^{ik_z L} = 1$$

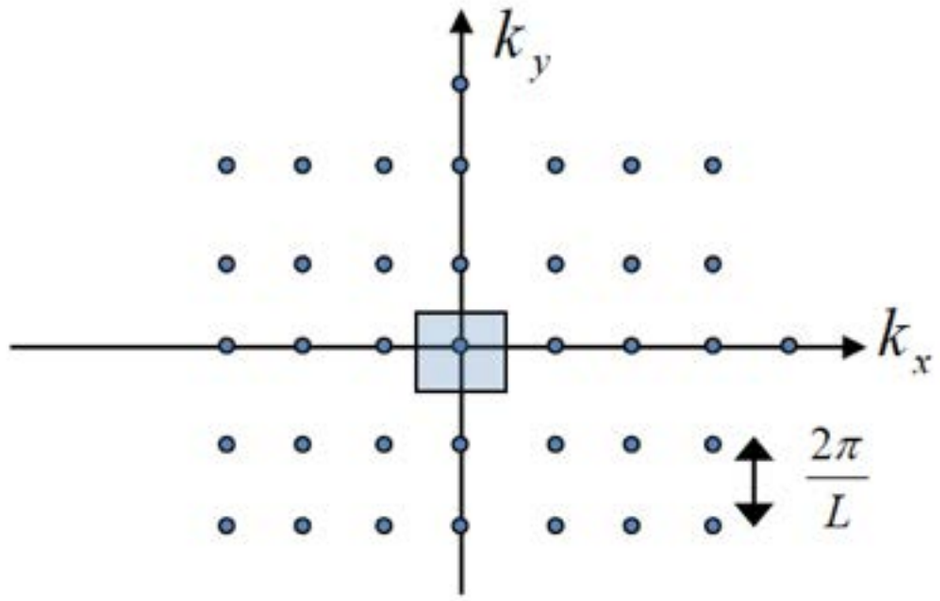
Which implies that $\mathbf{k_m = 2\pi n_m/L}$ with $\mathbf{m=x,y,z}$ and $\mathbf{n_m} \in \mathbb{Z}$

$$\vec{p} = \left(\frac{2\pi\hbar n_x}{L}, \frac{2\pi\hbar n_y}{L}, \frac{2\pi\hbar n_z}{L} \right)$$

Box quantisation (2)

We can now use this result to compute the density of states, i.e. the number allowed **k** (or **p**) values in a momentum region $d^3\mathbf{k}$:

$$dN = \frac{d^3\vec{k}}{singlestatevolume}$$



Each state occupies a volume $(2\pi/L)^3$ in k space, or $(2\pi\hbar/L)^3$ in p space.

The number of states in **p**,**p**+$d^3\mathbf{p}$ is therefore

$$dN = \frac{d^3p}{(2\pi\hbar/L)^3} = \frac{p^2 dp d\Omega}{(2\pi\hbar/L)^3}$$

$$\rho(p) = \frac{dN}{dp} = \frac{p^2 d\Omega}{(2\pi\hbar/L)^3}$$

Solid angle element

$$d\Omega = \sin\vartheta d\vartheta d\varphi$$

Phase space factor (2)

Now, normalizing over a unit volume and integrating over angles:

$$\frac{4\pi p^2 dp}{(2\pi)^3 \hbar^3}$$

Analogously for the neutrino

$$\frac{4\pi q^2 dq}{(2\pi)^3 \hbar^3}$$

Now nucleo momentum is fixed by momentum conservation (it disappears with the corresponding Dirac delta):

$$\frac{(4\pi)^2}{(2\pi)^6 \hbar^6} p^2 q^2 dp dq$$

and q is fixed by energy conservation: $q=E_0-E$
Moreover (natural units) $dq=dE$ and therefore:

Zero massa: dE= dp
Non zero massa: EdE = pdp

$$\frac{dN}{dE} = \frac{1}{4\pi^4} p^2 (E_0 - E)^2 dp$$

which gives the final formula for electron **spectrum** (apart from possible p and q terms in the matrix element):

$$N(p)dp \propto p^2 (E_0 - E)^2 dp \qquad N(E)dE \propto pE(E_0 - E)^2 dE$$

from which we can obtain the **Kurie plot** as $\sqrt{\frac{N(p)}{p^2}} dp$

Nuclear beta decay (2)

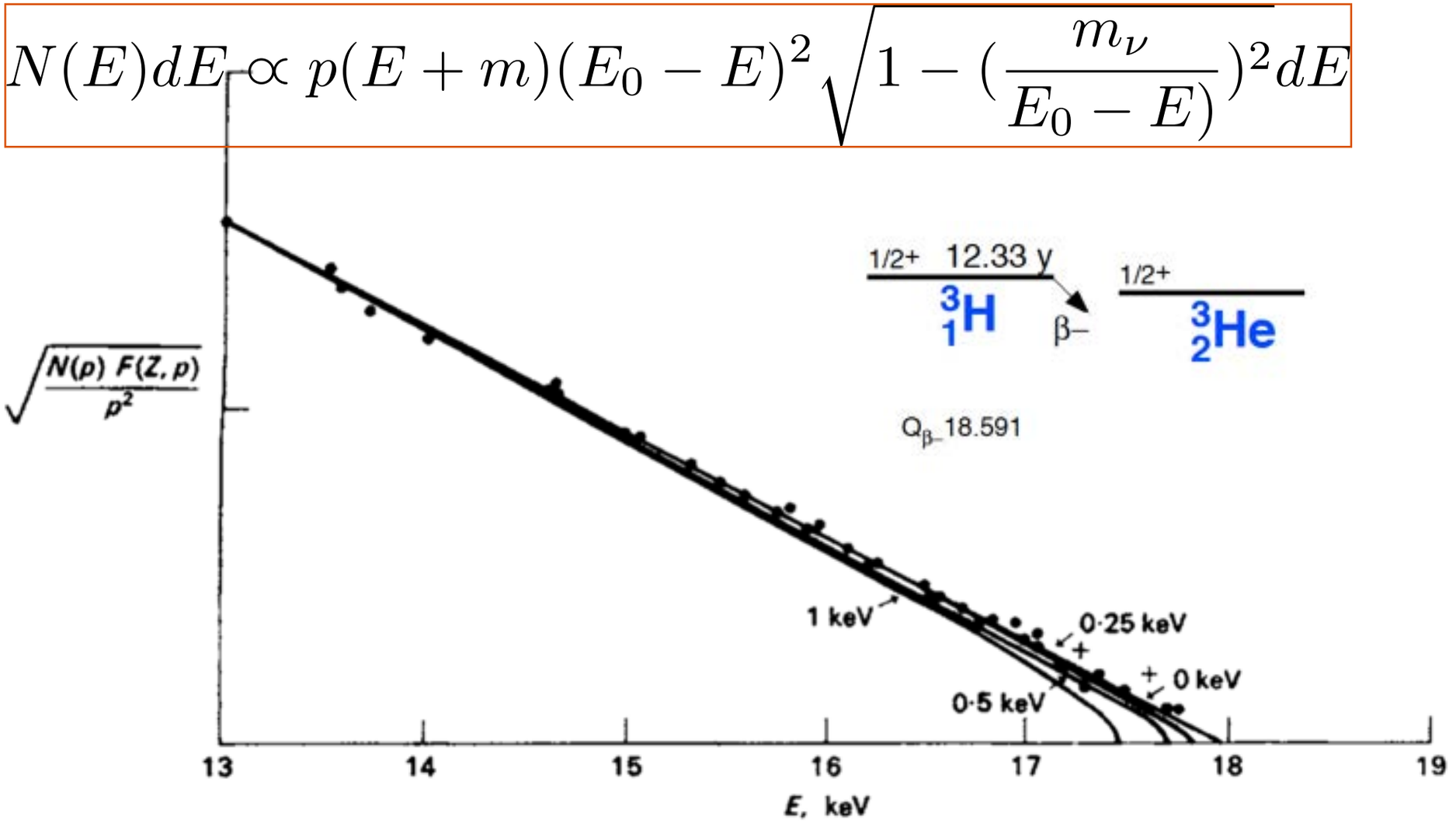
For large variations of the nuclear angular momentum, leptons can't provide the requested spin and orbital components are needed.
These transitions are much slower and known as "prohibited".Their Kurie plot is no more linear and requires proper corrections.
Furthermore, for **finite neutrino masses**, an additional term is required to conserve energy:

$$E_\nu + E + T = E_0 - m_\nu$$

and correspondingly for the spectrum:

$$N(E)dE \propto p(E+m)(E_0-E)^2\sqrt{1-(\frac{m_\nu}{E_0-E})^2}dE$$

where we have taken into account that $E_\nu^2 = q^2 + m_\nu^2$ and therefore $E_\nu dE_\nu = qdq$, as well as $EdE = pdp$.



“Allowed” and “prohibited” transitions

Allowed transitions and the various forbidden transitions are defined by the following selection rules:

$L = 0,1$	$\pi_i\pi_f = +1$	allowed transitions	(2a)
$L = 0,1$	$\pi_i\pi_f = -1$	first-forbidden non unique transitions	(2b)
$L > 1$	$\pi_i\pi_f = (-1)^L$	L^{th}-forbidden non unique transitions	(2c)
	$\pi_i\pi_f = (-1)^{L-1}$	$(L-1)^{th}$-forbidden unique transitions	(2d)

with

$$L = \Delta J = |J_i - J_f|$$

J_i, π_i and J_f, π_f are spins and parities of the initial and final nuclear states, respectively.

“Applications”

Applications

- Very powerful techniques have been developed in the framework of Nuclear Physics Experiments
- This methods have been applied to other scientific fields like
  - **fall out monitoring**
  - **environmental analysis**
  - **archeological and archeometrical studies**
  - **cultural heritage**
  - **…….**

- The Montecarlo simulation can be of fundamental relevance for:
  - **environmental dosimetry**
  - **clinical dosimetry (diagnostic and therapeutics)**

Eccentricities: Napoleon death

MISURE CON ATTIVAZIONE NEUTRONICA SULLA PRESENZA DI ARSENICO NEI CAPELLI DI NAPOLEONE BONAPARTE E DI SUOI FAMIGLIARI

MASSIMO CLEMENZA[1], ETTORE FIORINI[1], LAURA GUERRA[1], COSTANZA HERBORG[2], MASSIMO LABRA[3], EDOARDO ORVINI[2], ADALBERTO PIAZZOLI[4], EZIO PREVITALI[1], FRANCESCO PUGGIONI[5], ANGELA SANTAGOSTINO[3]

[1] Dipartimento di Fisica G.P.S. Occhialini, Università di Milano-Bicocca INFN, Sezione di Milano-Bicocca - Piazza della Scienza 3, 20126 Milano, Italia
[2] Dipartimento di Chimica Generale, Università di Pavia - Via Taramelli 12, 27100, Pavia, Italia
[3] Dipartimento di Scienze dell'Ambiente e del Territorio Università di Milano-Bicocca - Piazza della Scienza 3, 20126 Milano, Italia
[4] Dipartimento di Fisica Nucleare e Teorica dell'Università INFN, Sezione di Pavia - Via Bassi 6, 27100, Pavia, Italia
[5] Centro Studi "In novitate" - Novi Ligure, 15067, Alessandria, Italia

Questa ricerca, svolta da una collaborazione tra fisici, chimici e tossicologi, consiste nella tecnologia sviluppata per ricerche di fisica fondamentale per una ricerca di carattere storico-archeometrico. Utilizzando l'attivazione neutronica tramite un piccolo reattore si è rilevato che la concentrazione di arsenico nei capelli prelevati da Napoleone dopo la sua morte era sì molto elevata rispetto a quella misurata in soggetti viventi attualmente, ma del tutto confrontabile con quella riscontrata non solo nei capelli dell'Imperatore in altri periodi della sua vita, ma anche in quelli del figlio e della prima moglie. Ciò permette di escludere l'ipotesi di un omicidio.

1 Introduzione

I risultati ottenuti negli ultimi decenni nella fisica fondamentale nucleare, subnucleare ed astroparticellare hanno portato a notevoli sviluppi nelle tecniche adottate per la ricerca di eventi nucleari rari [1]. Citiamo in particolare gli eventi prodotti da particelle pesanti con interazione debole quali le cosiddette WIMPS (da *Weakly Interacting Massive Particles*) [2] che potrebbero costituire un terzo circa della massa del nostro Universo. La recente scoperta delle oscillazioni del neutrino [1, 3-7] ha in questi ultimi anni stimolato fortemente le ricerche di eventi rari di bassa energia quali le interazioni di neutrini solari [7] o il decadimento beta doppio che, se trovato, permetterebbe di chiarire la natura del neutrino e di determinarne la massa [8]. In tutti questi esperimenti ed in particolare in quelli sulle WIMPS e sul decadimento beta doppio condotti con rivelatori a semiconduttori o termici [9-11] è indispensabile studiare e ridurre il fondo di

Poisoning?

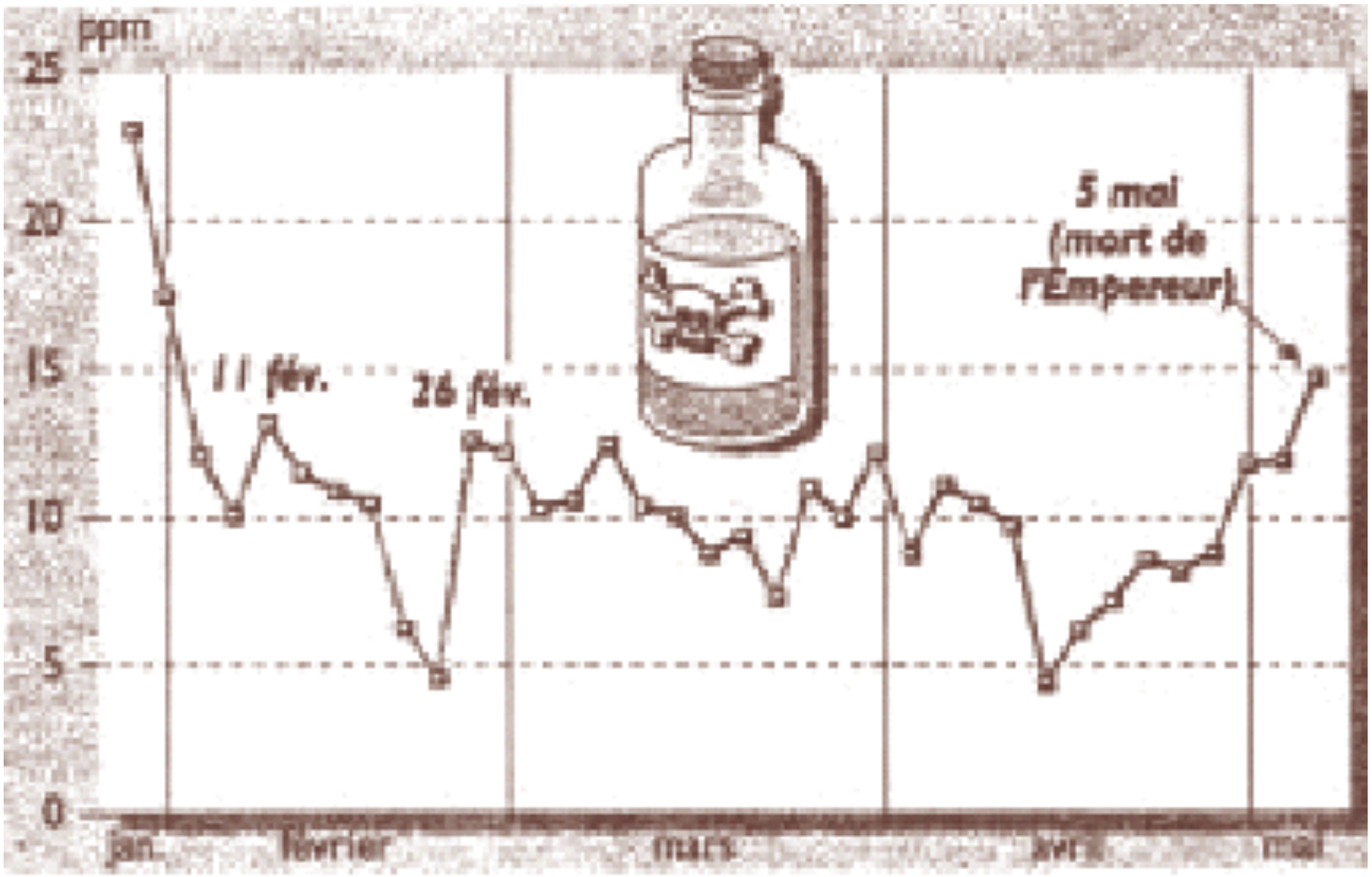
- Based to the symptoms Forshufvud in 1960 hypothesizes arsenic chronic poisoning
- This hypothesis is based on various indications

- In particular, some found symptoms could be interpreted with assumption of As
  - Napoleone surely had many enemies that wanted him dead
    - Gen. Lowe head of the garrison to S. Helen
    - Dr. Antommarchi physician of Napoleon
    - Count of Motolon jealous for the Napoleon relationships with his wife
  - In 1840 the exhumed dead body was in perfect state of maintenance this would be justifiable with heavy assumptions of As

The assumption of As from Napoleon has always been tied up to the poisoning

Poisoning?

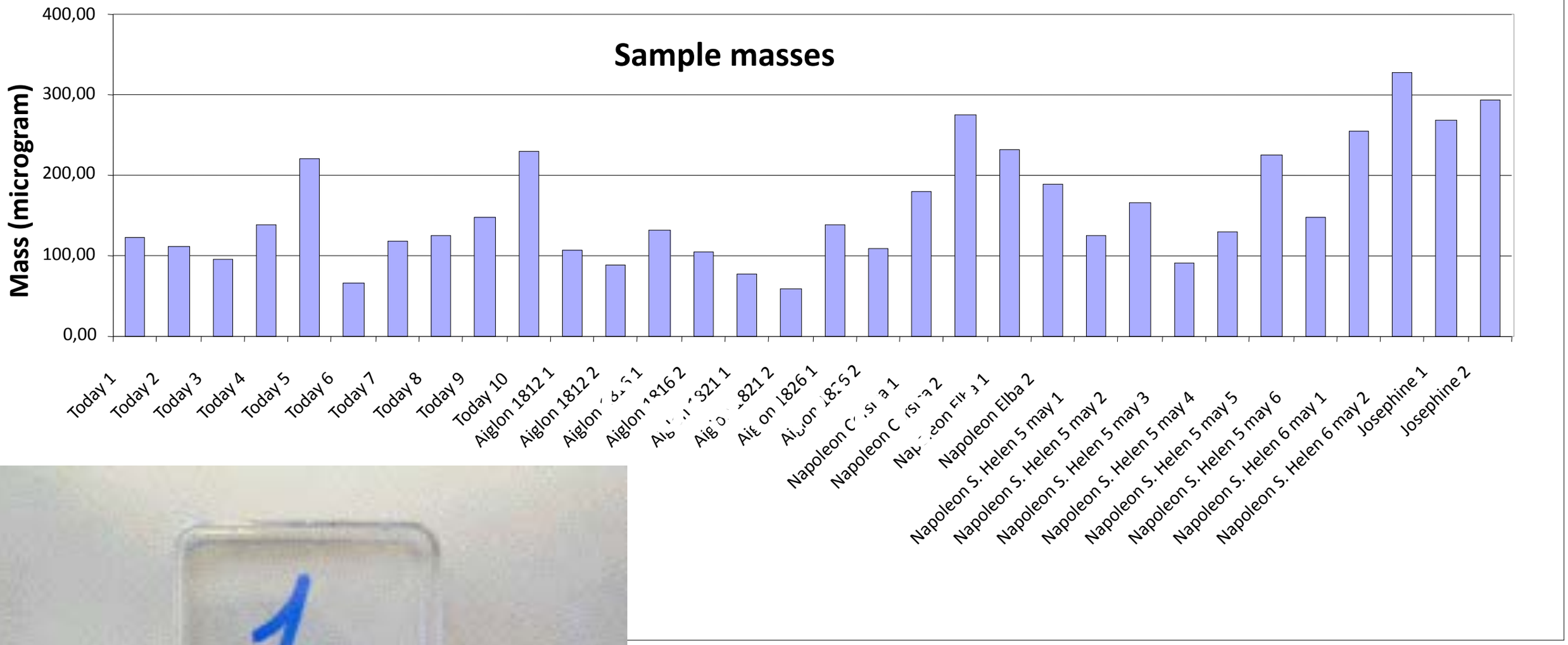
- Sponsored by Ben Weider numerous measurements were done to search As
- Various measurement techniques were used
  - NAA Harwell
  - AAS* FBI

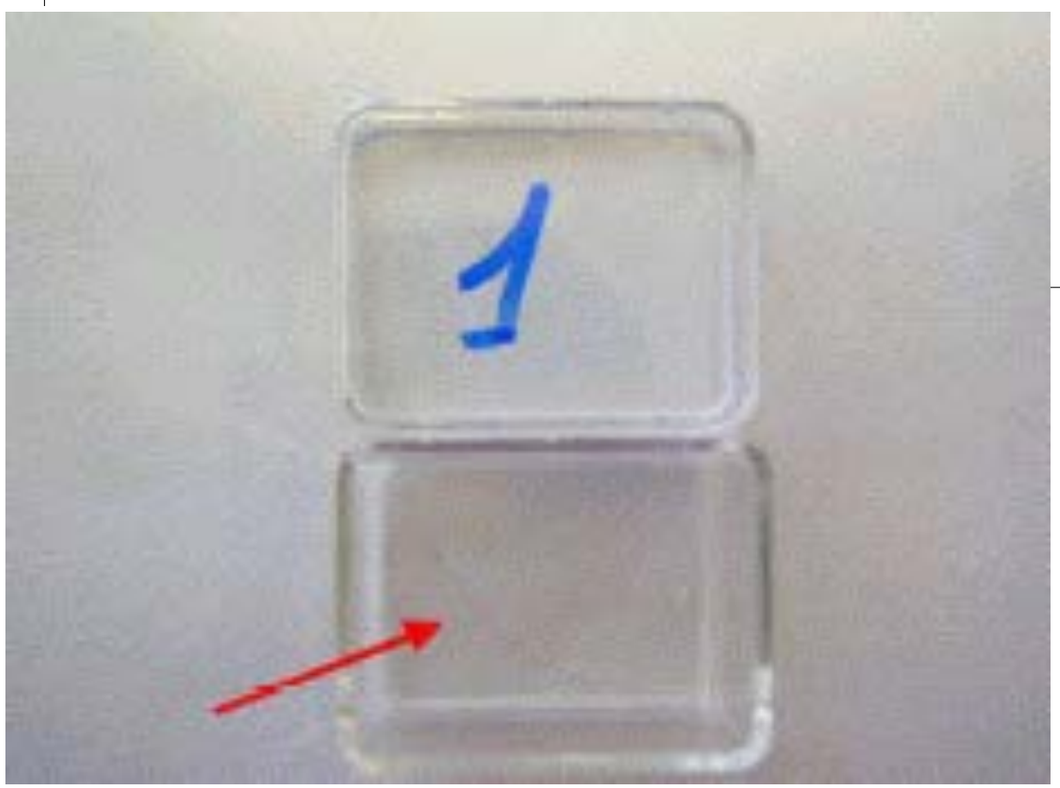


L'analyse du cheveu n°3 de l'Empereur.

* Atomic Absorption Spectroscopy

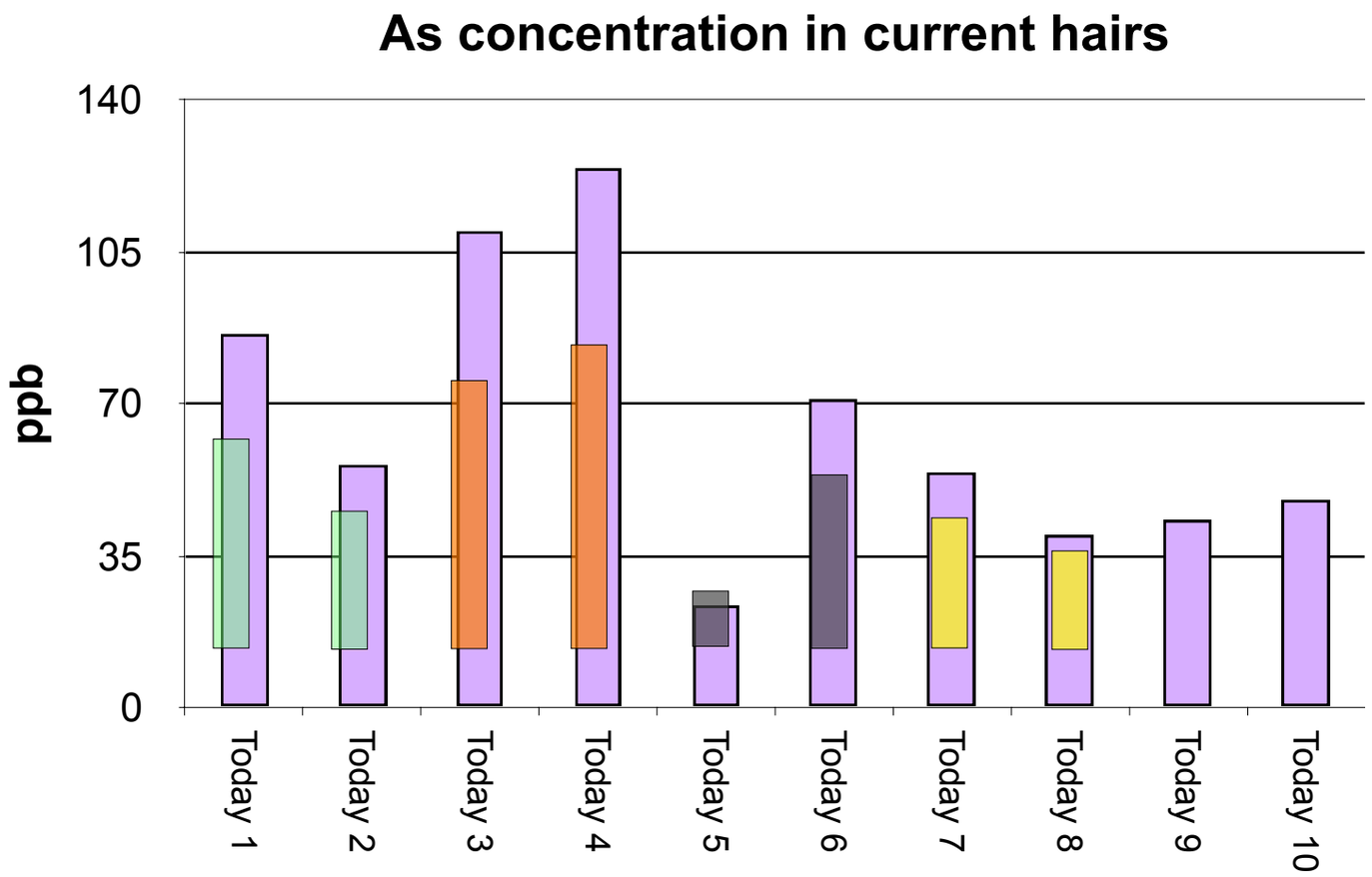
The results





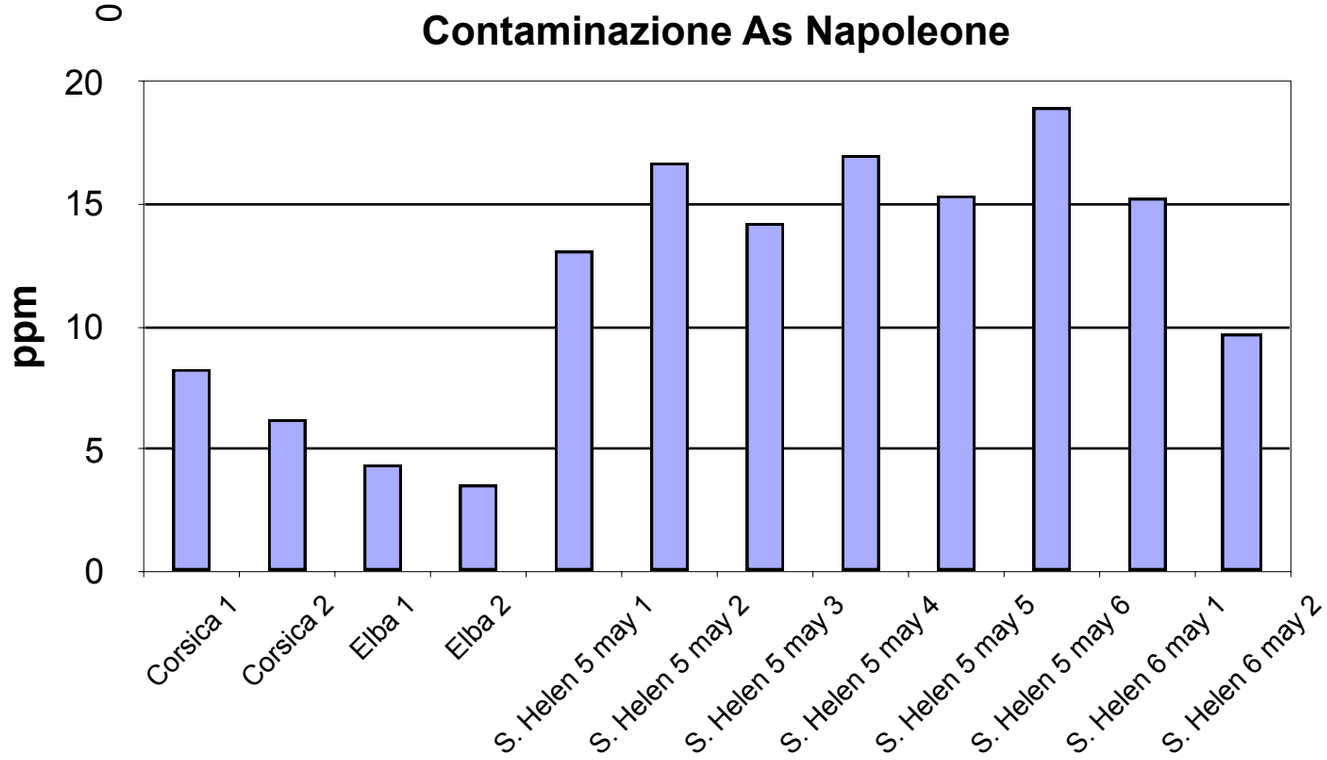
Technical approach to the measurements:
Nuclear Activation Analysis (NAA)

NAA on hair samples



Presently As concentration in hair is on the scale of 0.1 ppm

- Concentration of As in Napoleon hairs is on the scale of 10 ppm
- But….
  - The concentration is practically the same in Napoleon child at Ajaccio and Napoleon in exile in S. Helen island



Archeometry

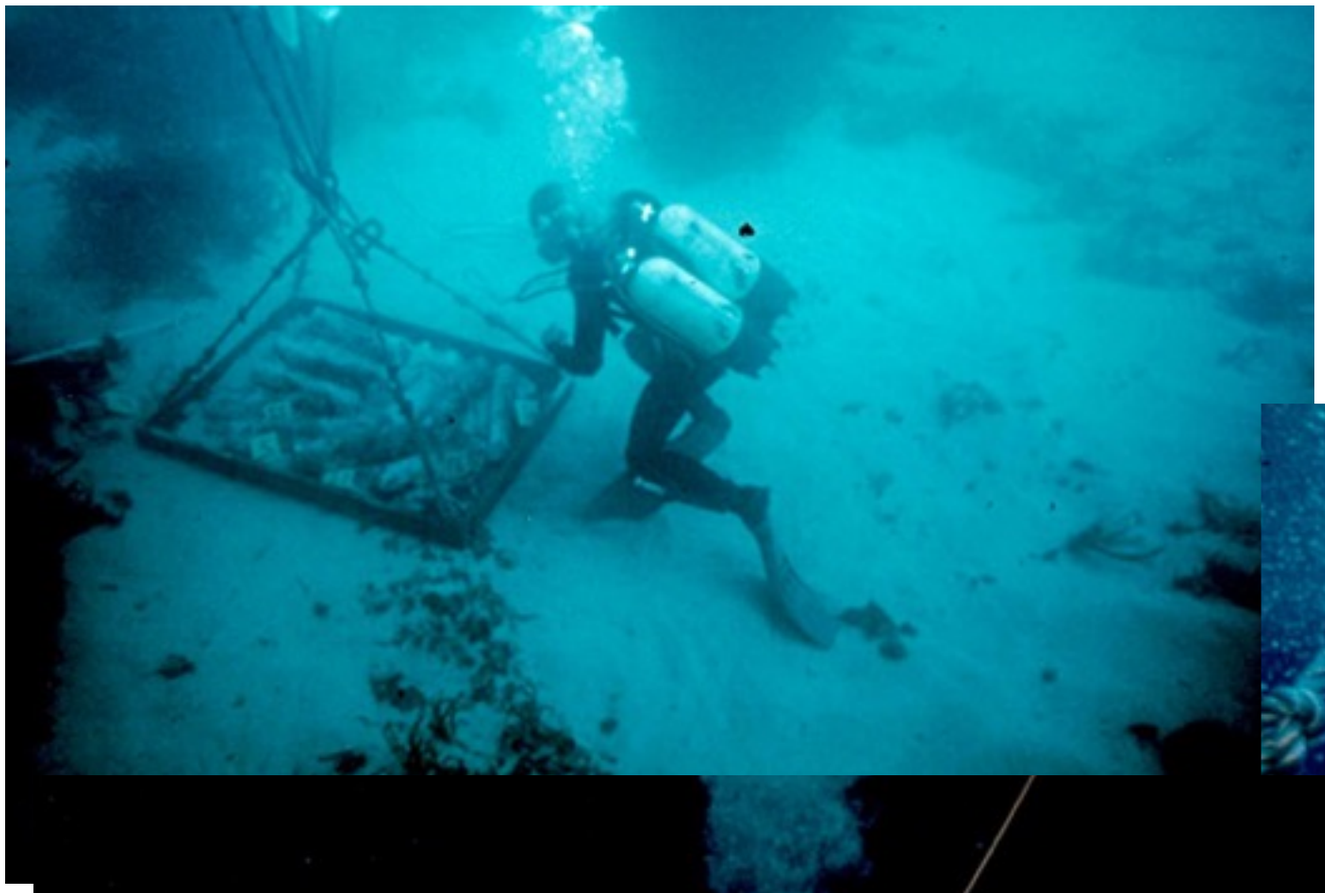
It was the year 1990 .....

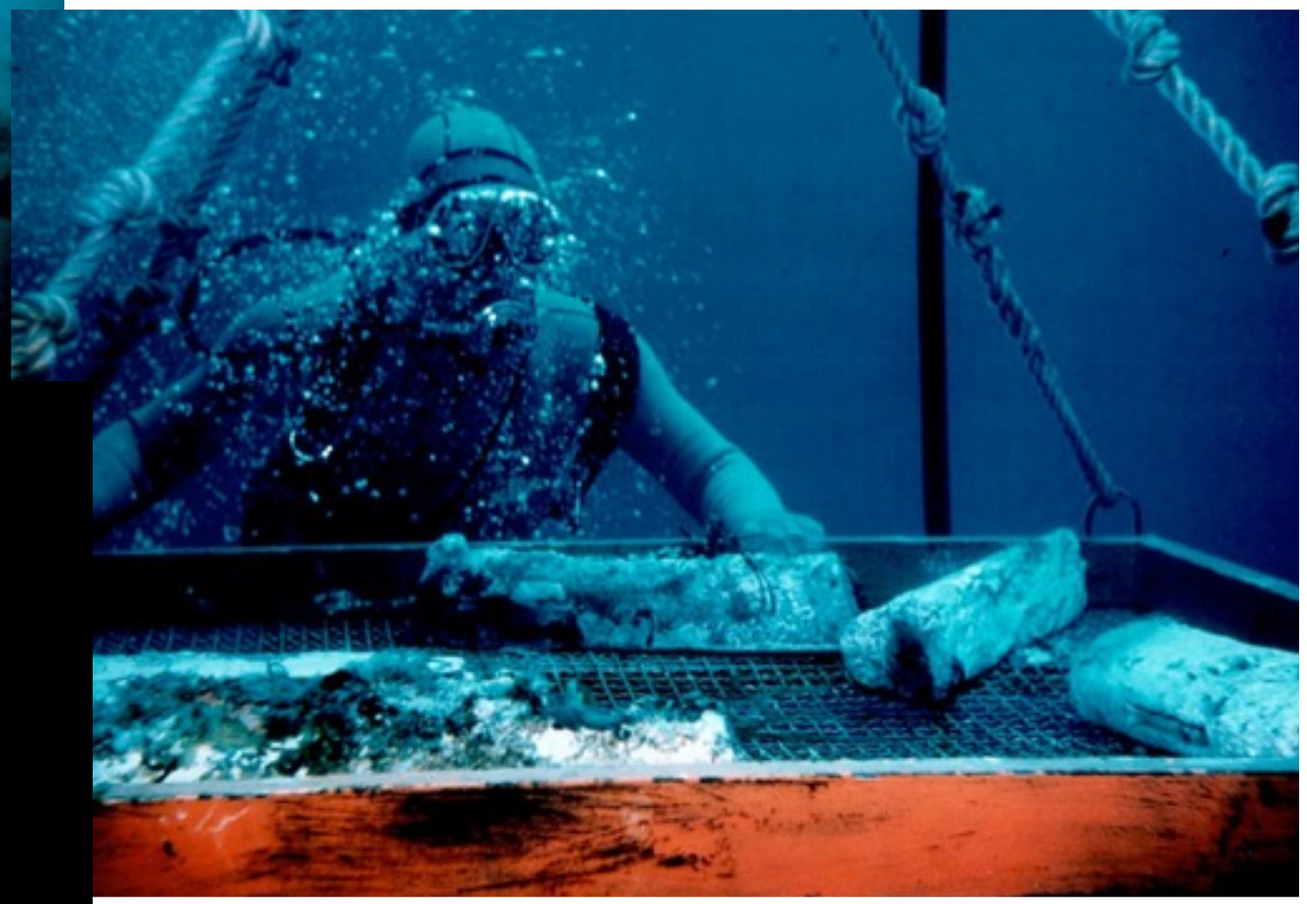
- .... When an ancient roman ship sunk close to Sardinia Island was found
- It carried a cargo of about 1000 ingots of lead (33 tons) from various roman foundries

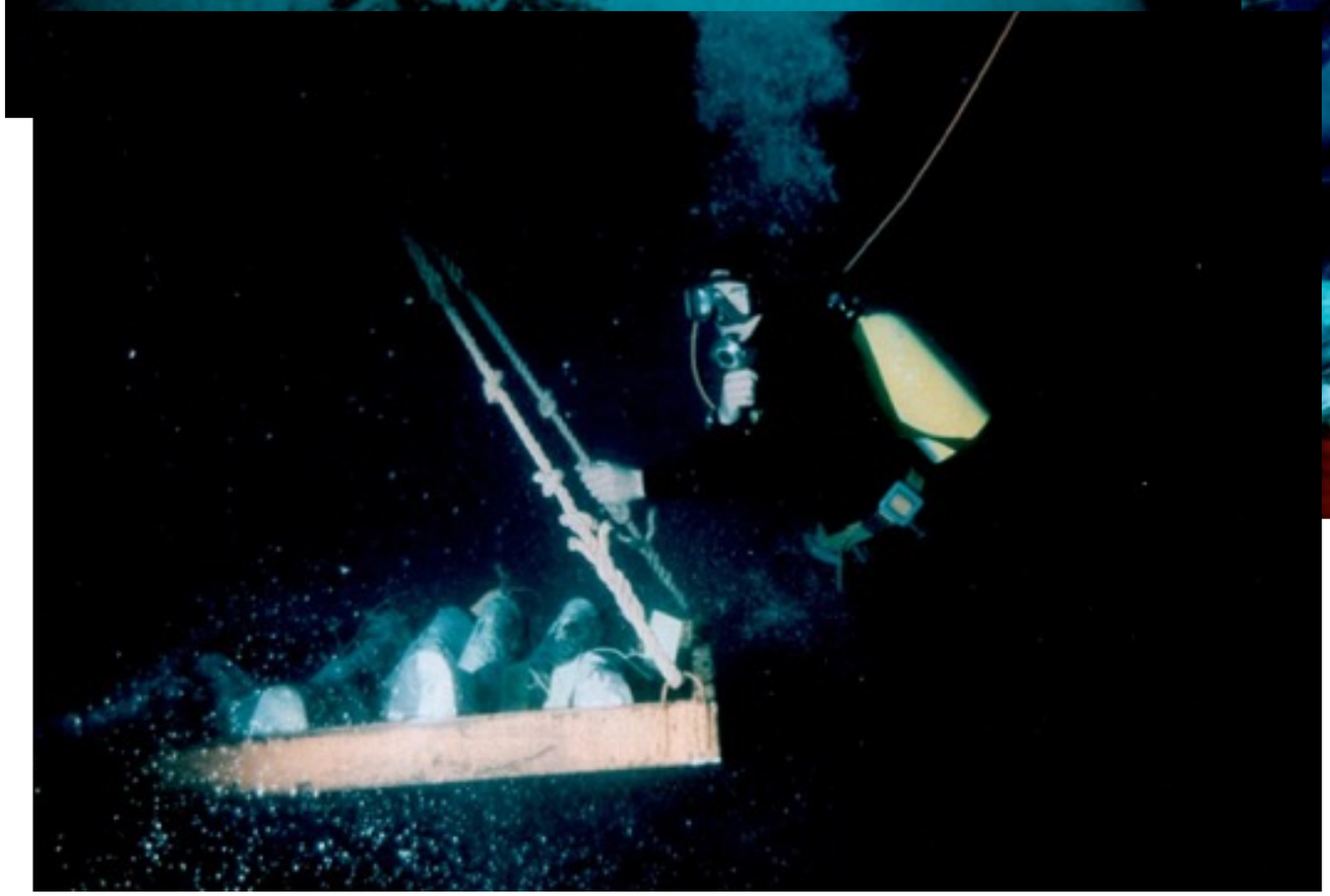
- All the ingots were recovered thanks to the support of INFN

- INFN was interested to use the ancient roman lead for very low background experiments

Malu Entu island lead







Malu Entu island lead

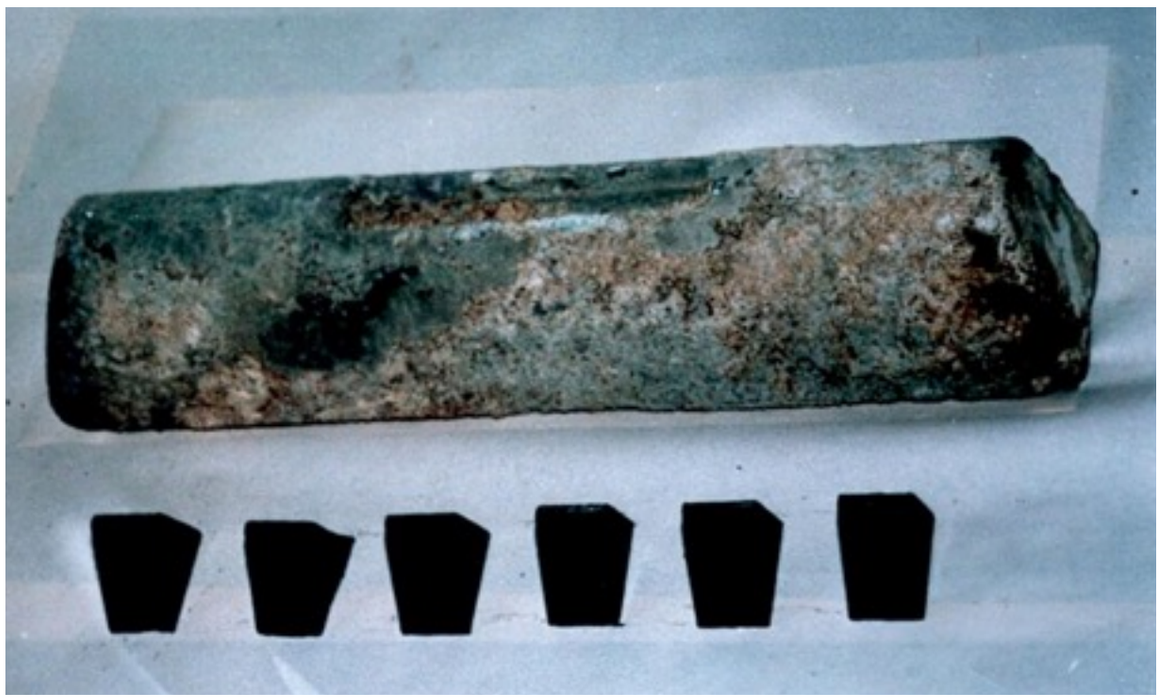
However, there are some questions:

- when the ship sank
- from where the ship comes from
- from where the lead comes from
- where the ship was going ....

On ingots there are some indication

- *socmcpontilienorummf*
- *soc-m-c-pontilienorum-m-f*
- *societas marci e cai pontilienorum marci filiorum*

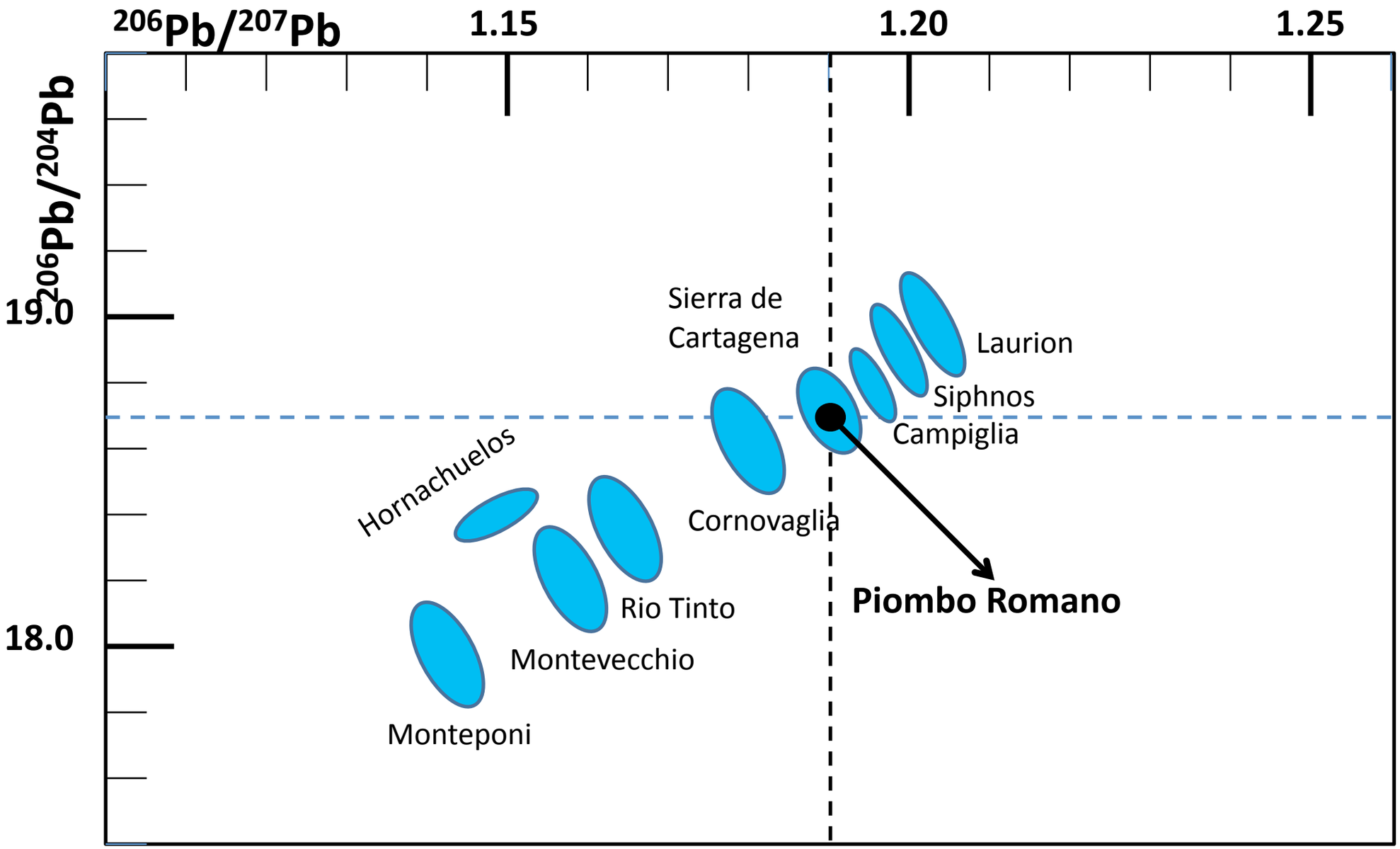
company of Marco and Caio Pontilieni son of Marco



- *lcarulilfhispalimn*
- *l-caruli-l-f-hispali-m-n*
- *luci caruli luci fili hispali menenia tribu*

Lucio Carulo Hispalio son of Lucio tribù Menenia

ICP MS analysis of anctient roman lead



The location of the mine in the Sierra de Cartagena tells us that:

- Confirm that the lead came from Spain
- Identifies a time interval:
  - after 89 BC for historical reasons
  - before 50 BC mines were abandonment

Fall-out measurements

A nuclear reactor accident releases a lot of radioactive nuclei in the atmosphere

Dose coefficients averaged over time spent in body after ingestion

Actinide	ε [nSv/Bq]	FP	ε [nSv/Bq]
^{238}U	44	^{90}Sr	0.34
^{237}Np	110	^{93}Zr	0.86
^{238}Pu	230	^{99}Tc	0.64
^{239}Pu	250	^{126}Sn	5
^{240}Pu	250	^{129}I	110
^{241}Pu	5	^{131}I	22
^{242}Pu	240	^{134}Cs	19
^{241}Am	200	^{135}Cs	2
^{243}Am	200	^{137}Cs	13
^{244}Cm	120		

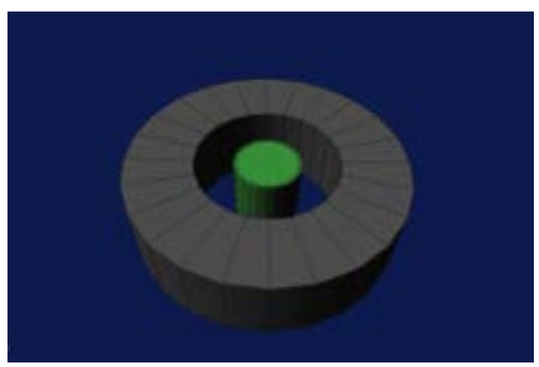
Neutron activation products

Fission products

Sublimation points:

- Iodine: 184 °C (I_2 gas e Particolato)
- Cesium: 671 °C (Particolato)

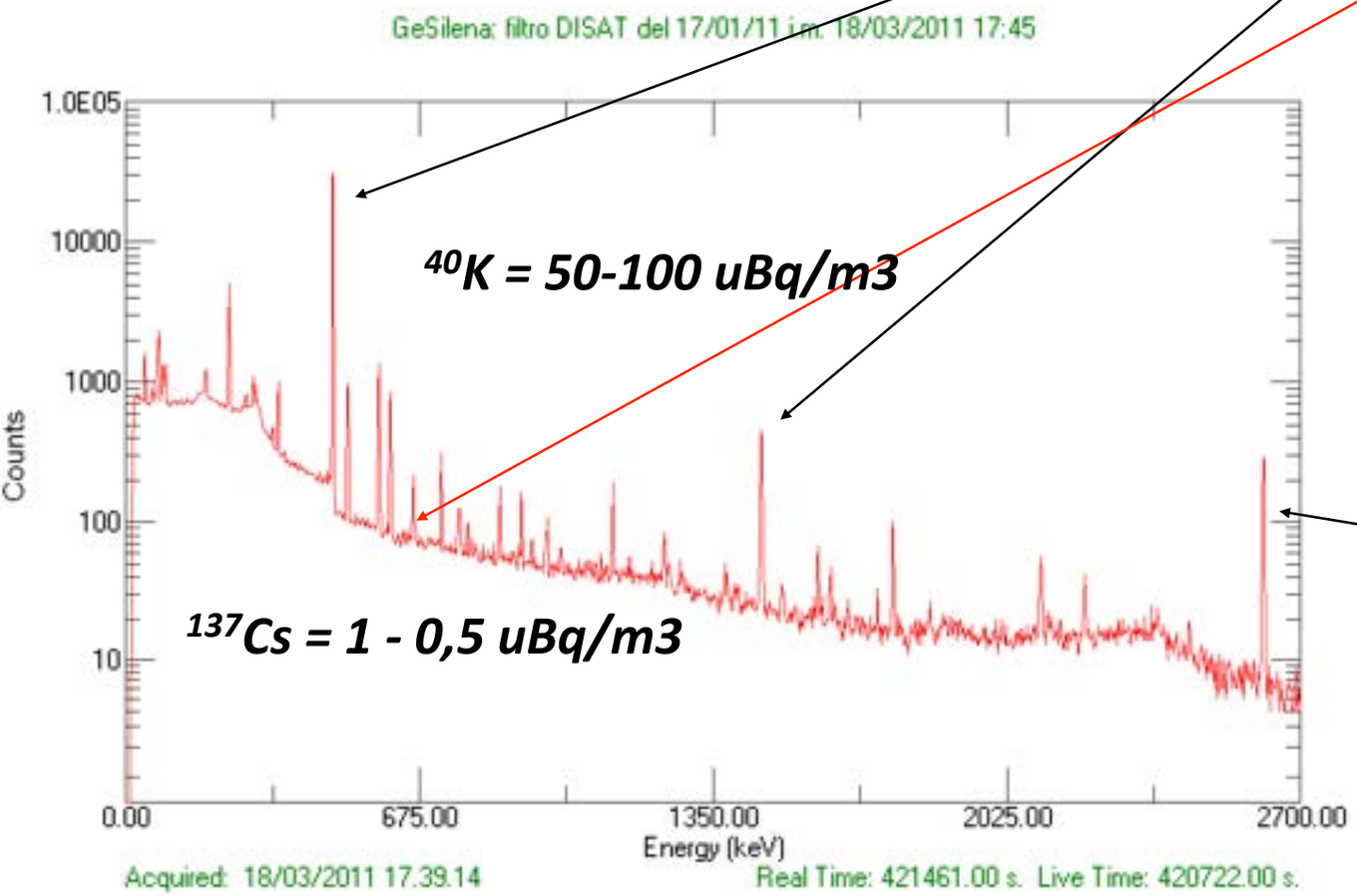
With efficient filters (very high flux) and low background detectors this contaminants can be detected also at large distances



Natural radioactivity in air

^{7}Be = 5-10 mBq/m^3

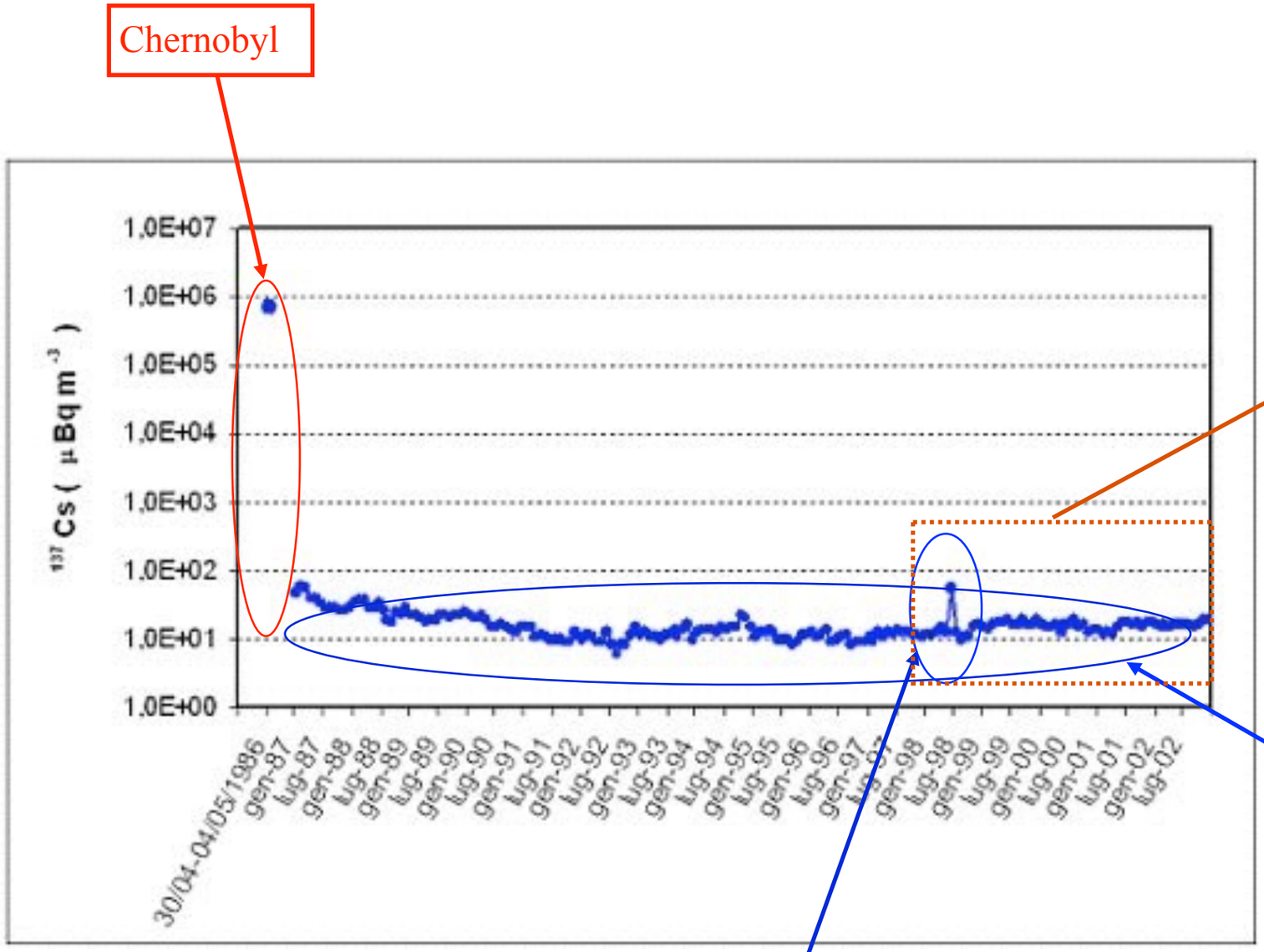
$^{14}N + p \rightarrow {}^{7}Be + 2\ {}^{4}He$
$^{14}N + n \rightarrow {}^{7}Be + {}^{8}Li$
$^{16}O + p \rightarrow {}^{7}Be + {}^{10}B$
$^{16}O + p \rightarrow {}^{7}Be + {}^{7}Li + {}^{3}He$
$^{16}O + n \rightarrow {}^{7}Be + {}^{10}Be$
$^{16}O + n \rightarrow {}^{7}Be + {}^{6}He + {}^{4}He$

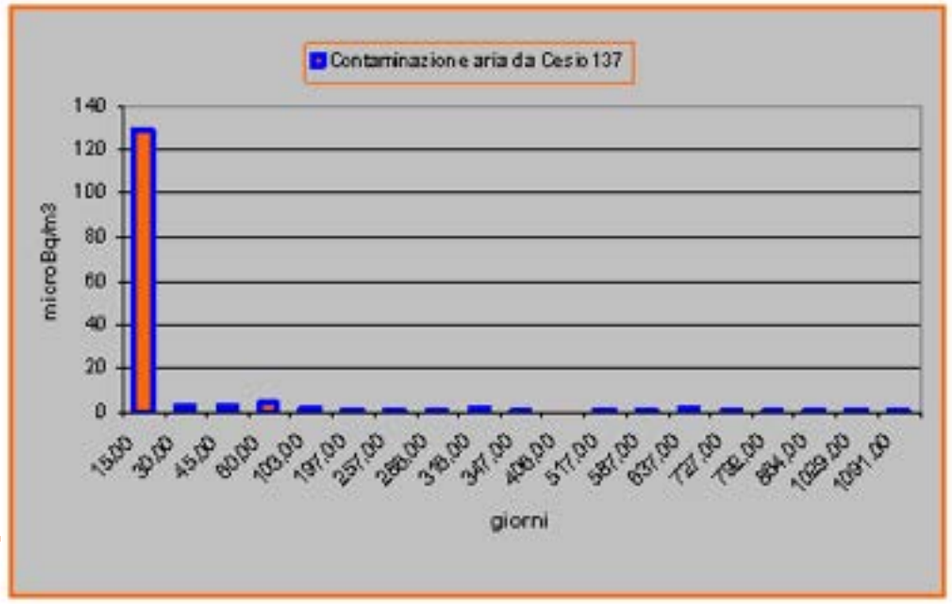


^{222}Rn	$T_{1/2}$	Energia Radiazione Gamma	Provenienza
^{7}Be	53.29 d	478	Cosmogenic
^{40}K	1.3x10^{9} y	1460	Primordial
^{137}Cs	30.1 y	661	Artificial
^{214}Pb	26.8 min	352	^{222}Rn
^{214}Bi	19.9 min	609	^{222}Rn
^{210}Pb	22.3 y	46	^{222}Rn
^{212}Pb	10.64 h	238	^{222}Rn
^{212}Bi	60.55 min	727	^{222}Rn
^{208}Tl	3.053 min	2615	^{222}Rn

Accident monitoring

^{137}Cs average concentration values in air

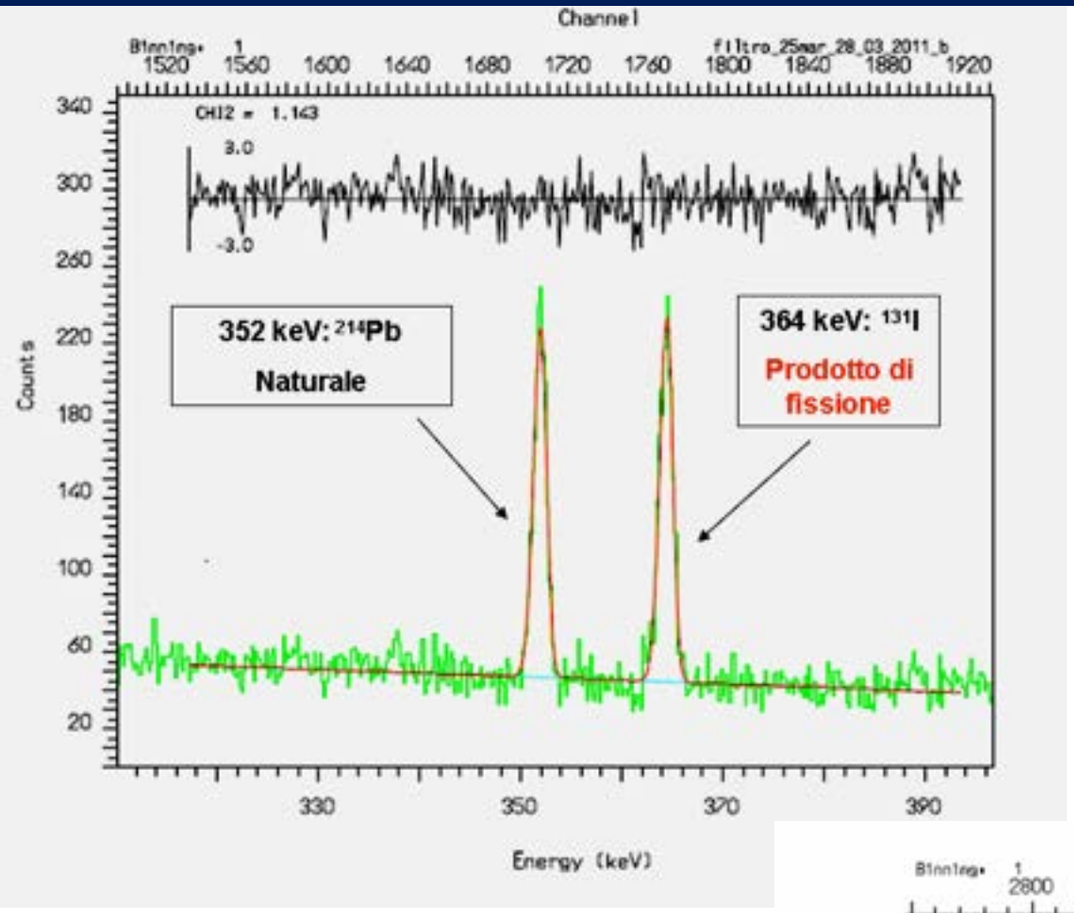


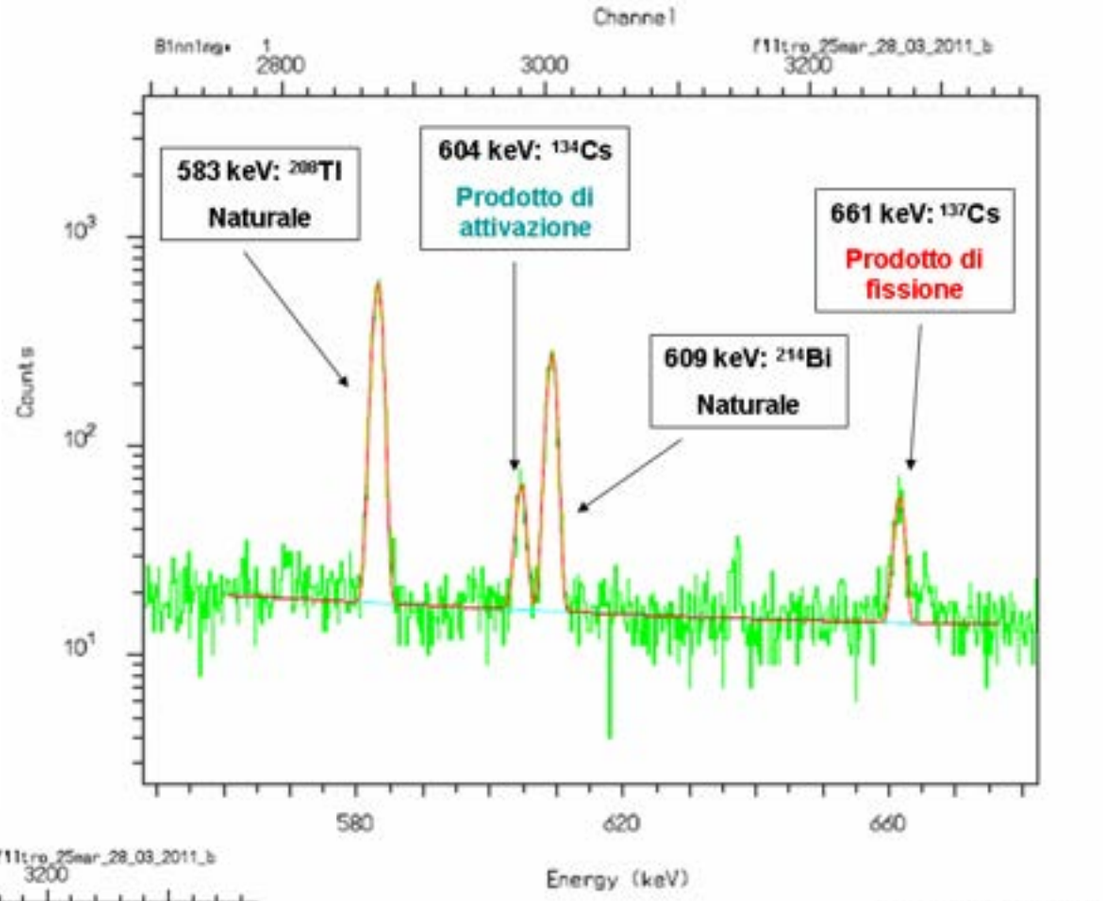


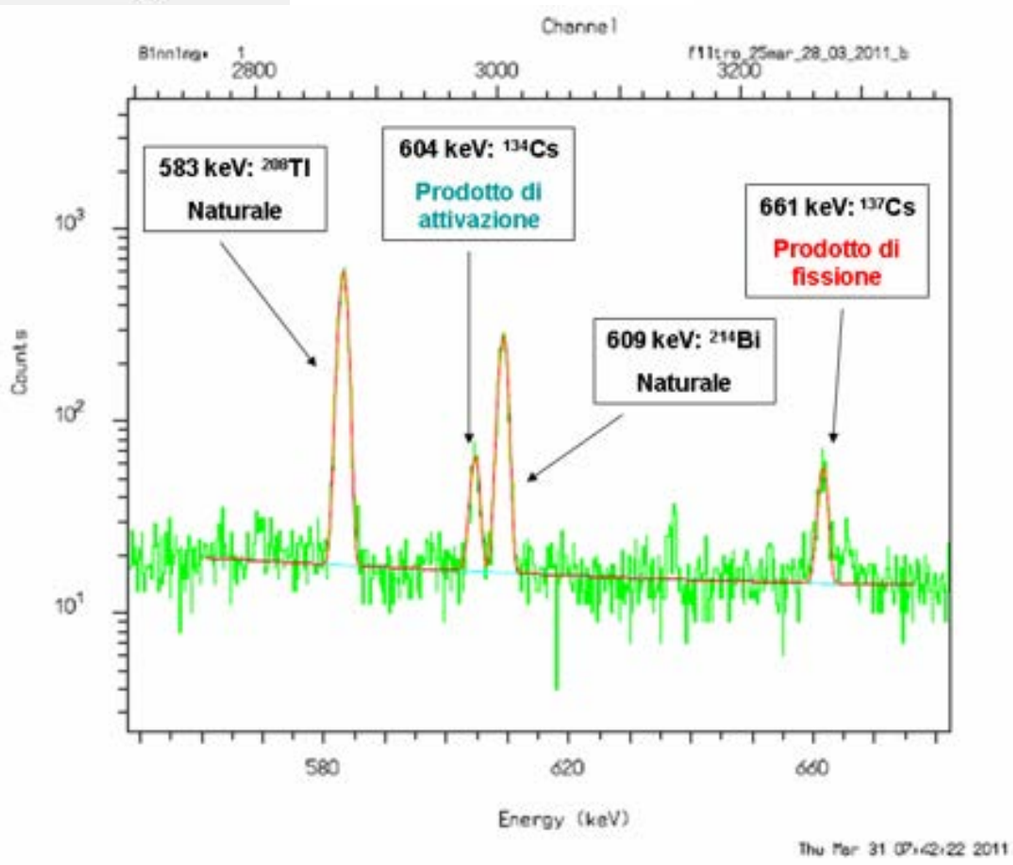
Suspension of soil ^{137}Cs

Accidental fusion of a ^{137}Cs source (Algeciras, Spain)

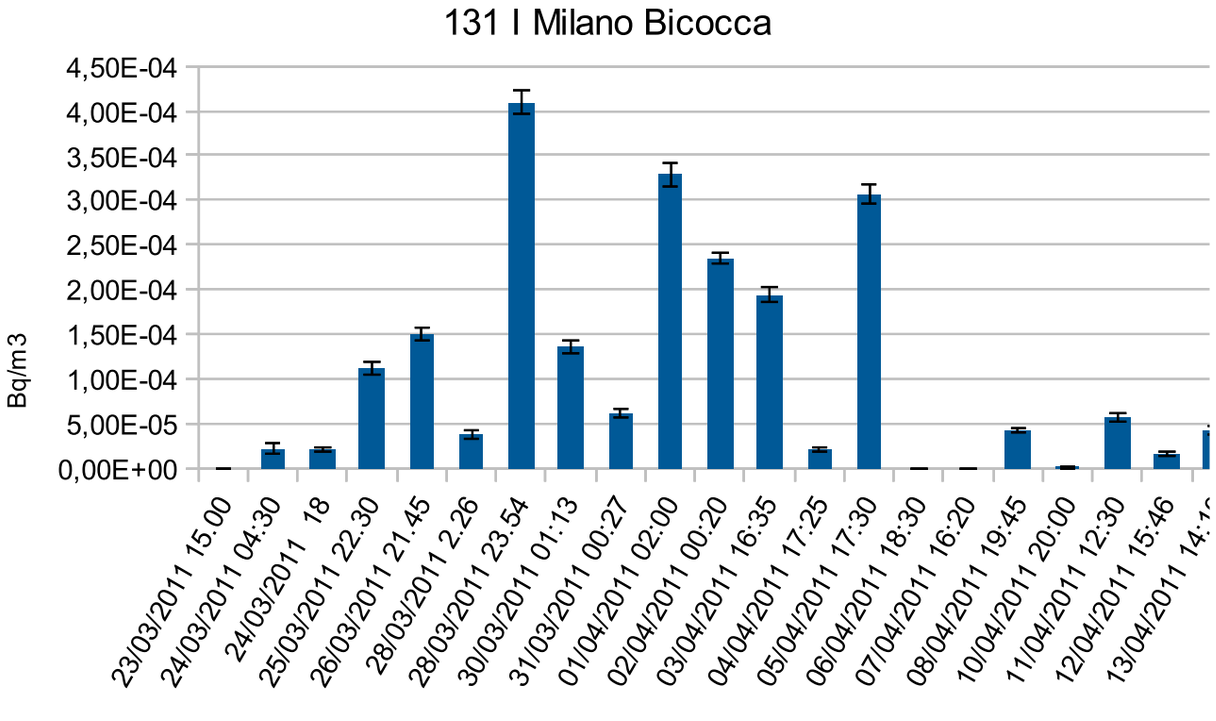
Fukushima fall-out measurements (@

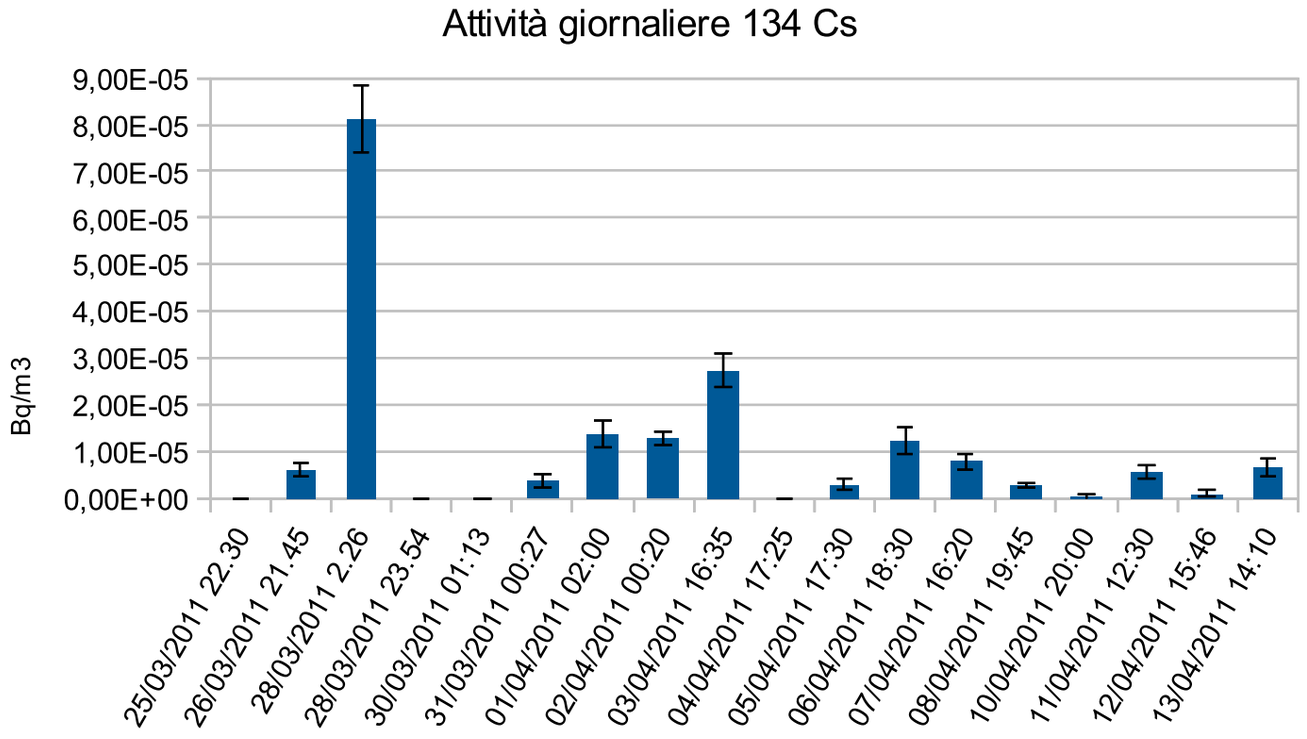


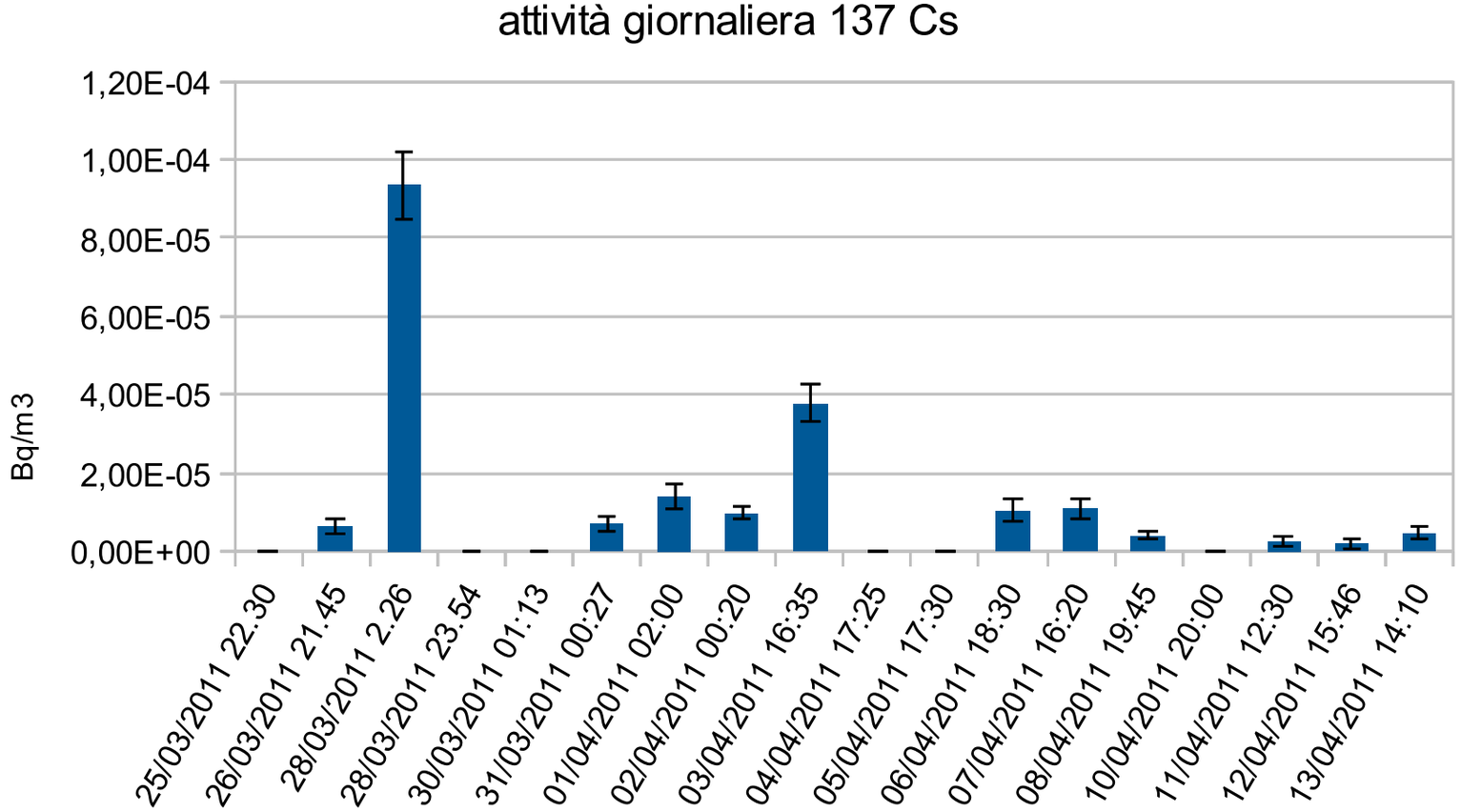




Fukushima fall-out measurements (@



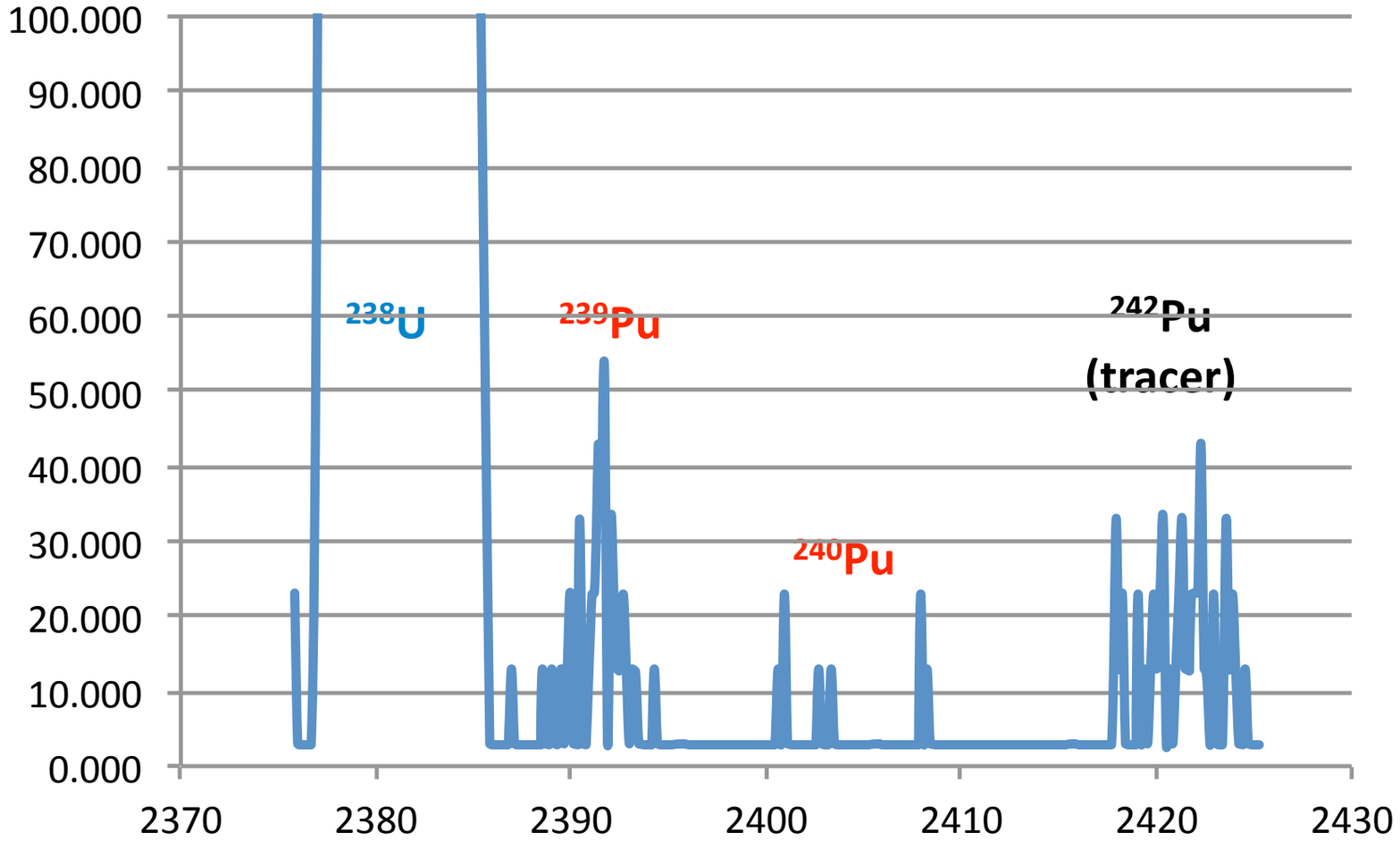




Heavy elements (?)

Measurements of isotopic ratio in soils with HR ICP MS

- Soils from **Brjansk (200 km nord of Chernobyl), Russia – I.A.E.A.375**
- Soil from **Ternate, Varese – A.R.P.A.**



$^{240}Pu/^{239}Pu$	
Brjnsk, Russia	0,28 + 0,05
Ternate (Va)	0,05 + 0,01

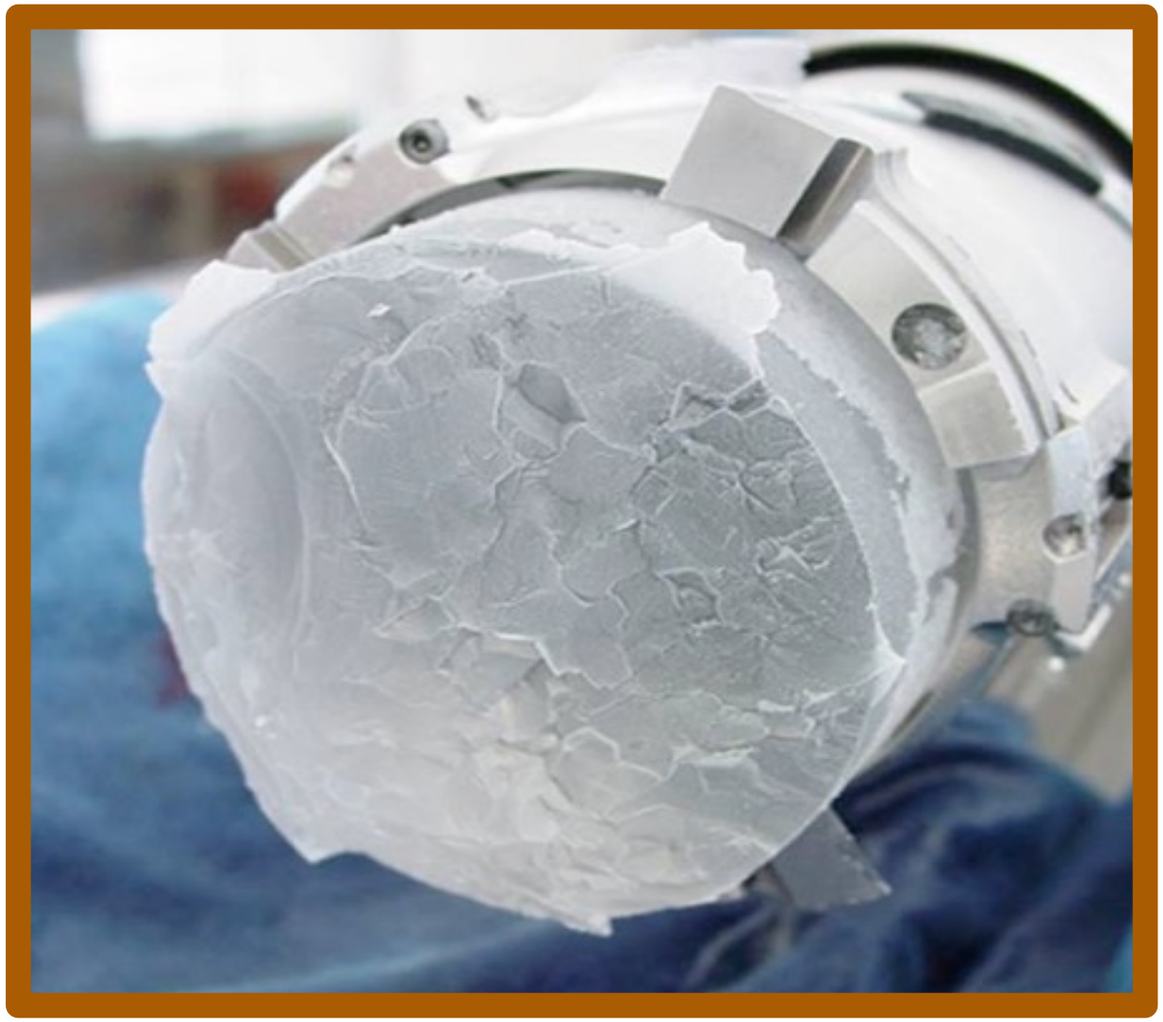
- **Pu from Chernobyl/Fukushima did not arrive to Italy**
- **Pu in north Italy is due to nuclear weapon tests in the atmophere**

Nuclear tests

Ice cores are very important historical archives of climate change and pollution

With a precise analysis it is possible to determine:

- the ages of the various core parts
- the elemental contaminations
- the source of contamination
- possible anthropogenic influences



In order to do these analysis a very high sensitivity in elemental analysis is needed

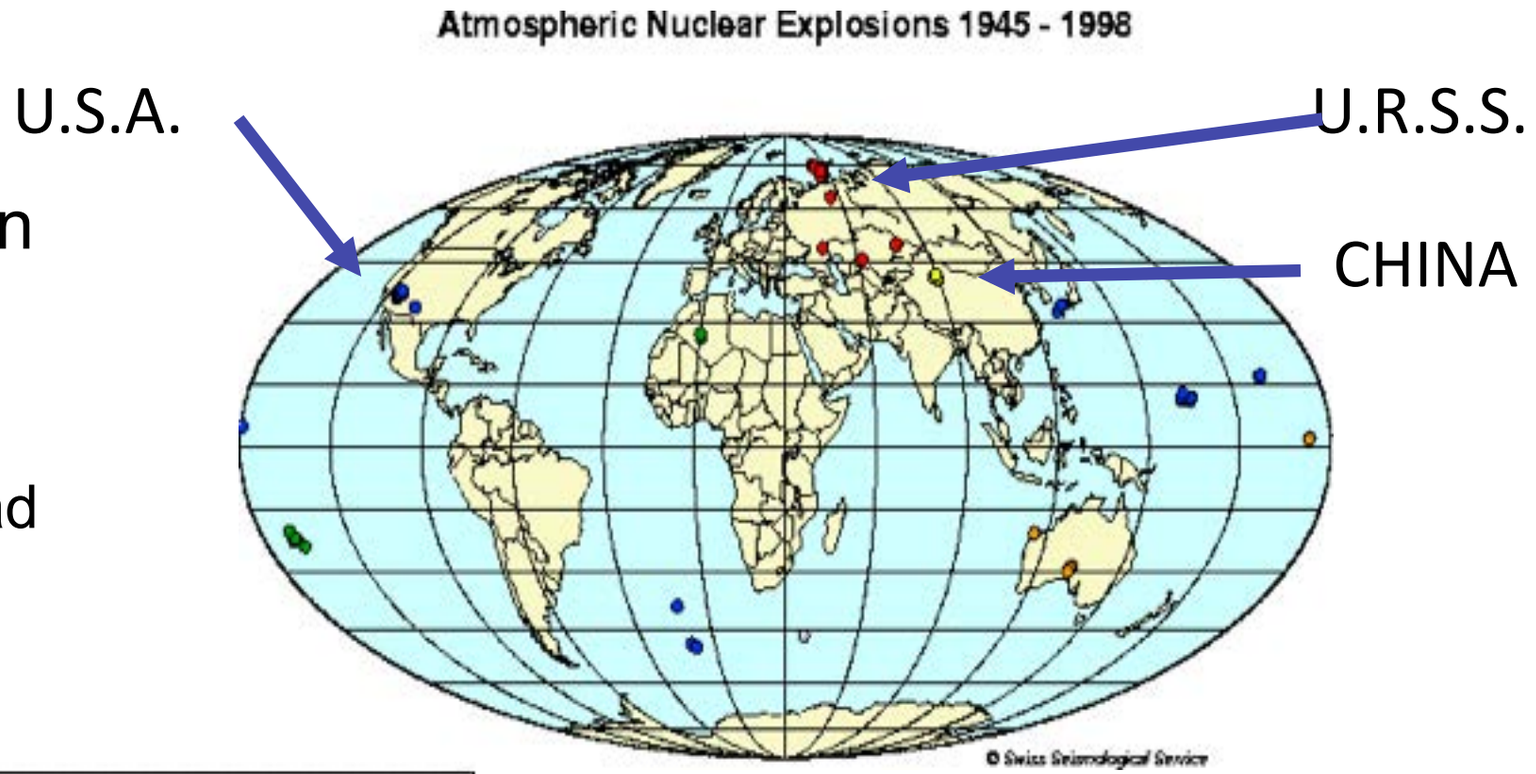
- in Antarctica ice core dust concentration is of the order of 1 ppm
- trace elements in dust shows concentration between 1 ppb and 1 ppm

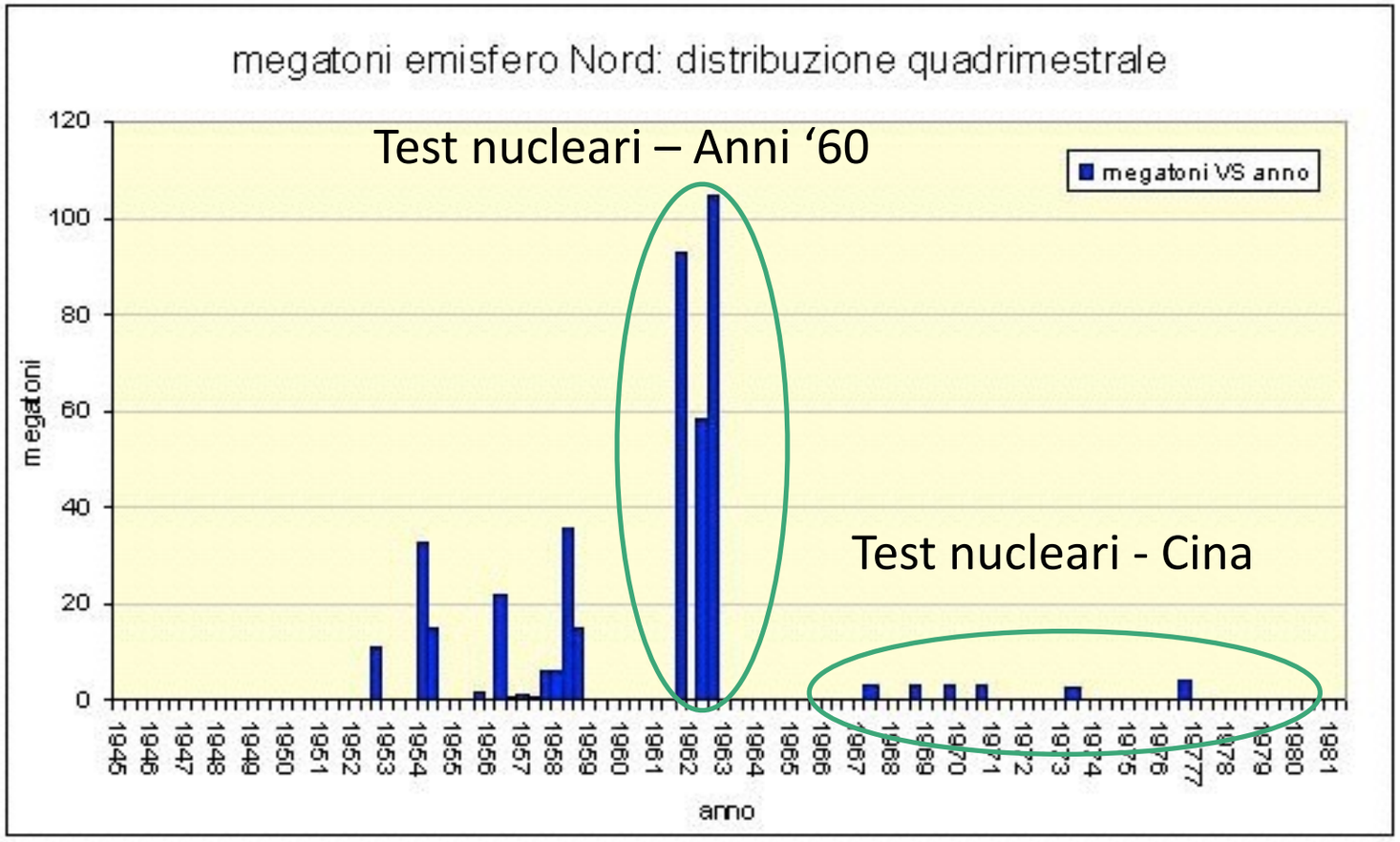
A sensitivity on the scale of 10^{-12} g/g is needed

Nuclear tests

Many test explosion with nuclear weapons were done in the north emisphere

Few countries in various places had test their nuclear arsenal





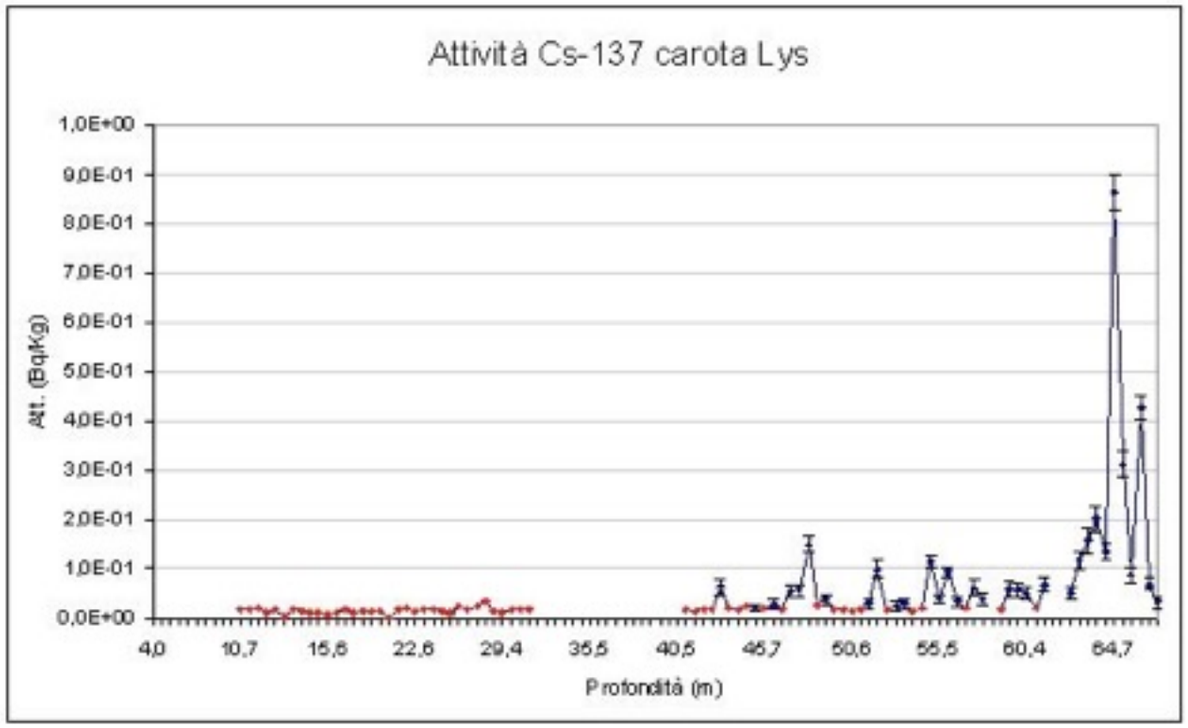
- In 1963 USA e URSS signed for a stop of test in atmosphere of their weapons
- CHINA had stopped his test at the end of the '70

- Can we check if it is real?

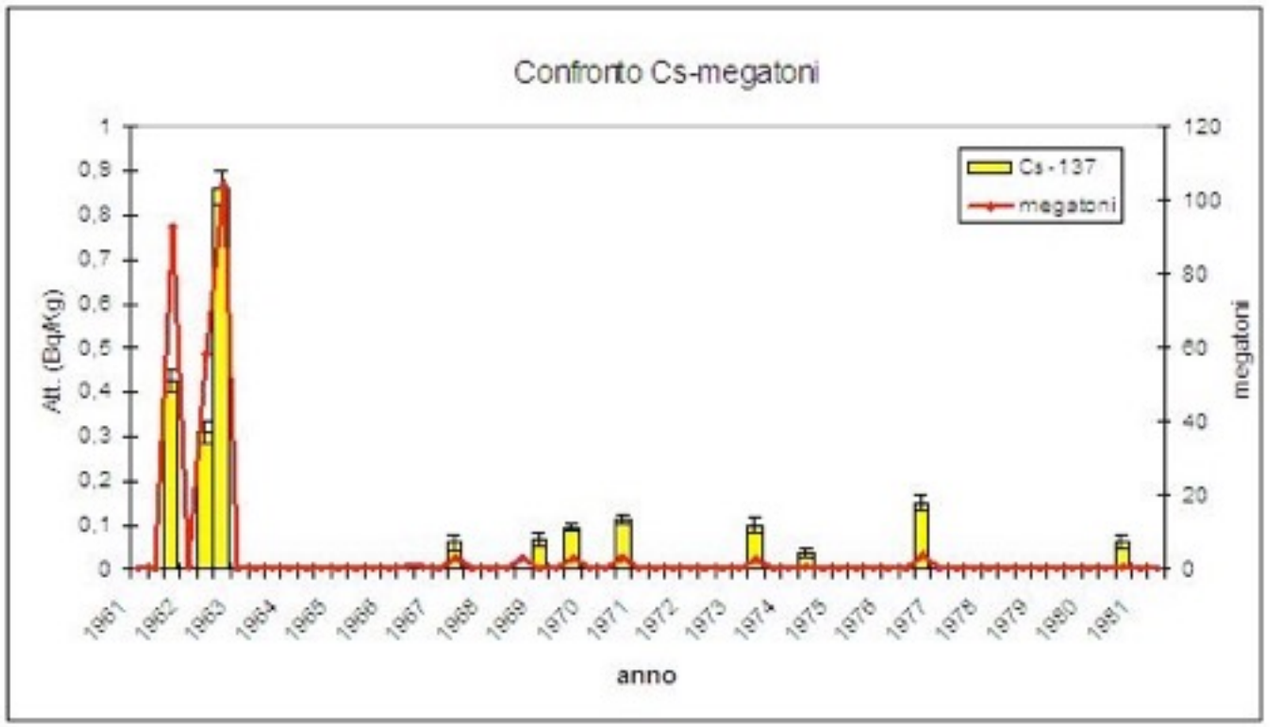
A natural archive

Ice cores collected in the alps are a powerful tool for

- measurements of elements that come from nuclear fall-out
- nuclear weapon test explosions and radioactive fall-out
- identification of ^{137}Cs and ^{90}Sr (sensitivity in the range 10^{-15} g/g is needed)



Measurements of ^{137}Cs contamination in ice core as a function of depth



Correlation between the megatons released by nuclear test explosions and ^{137}Cs in ice cores