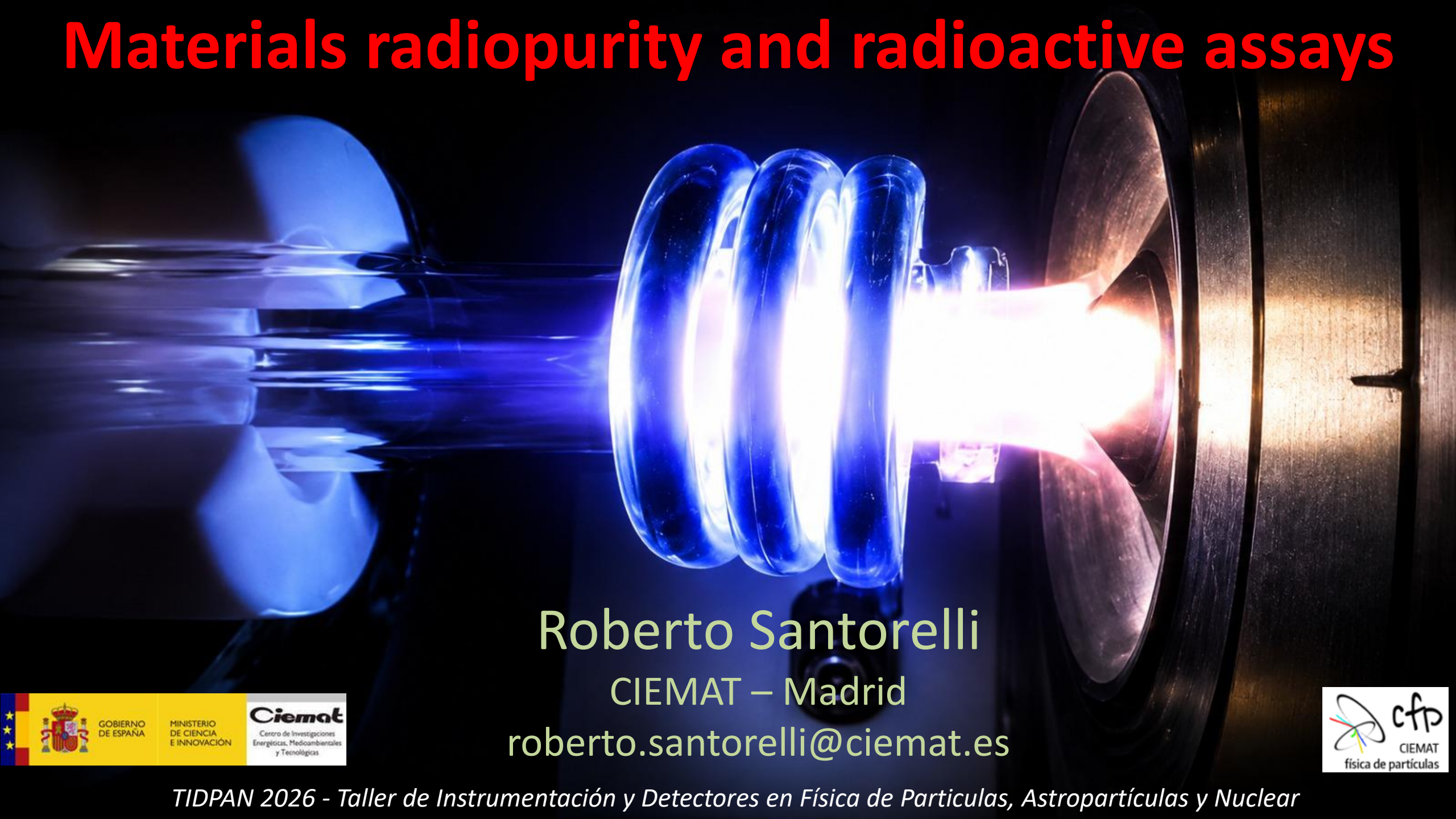
Materials radiopurity and radioactive assays



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Objectives of this lecture

- Figure out the importance of **material assays and radiopurity-**related aspects for low-background techniques
- Know the most important **sources of natural** radioactivity for rare event search experiments
  - Identify the main **primordial** radionuclides relevant for low-background experiments
  - *Grasp the principles of the **cosmogenic** production of radionuclides*
  - ***Anthropogenic** contaminants*
- Understand the general framework of the problems posed by **Radon**
- Know the principle of the most commonly used **assay techniques**

Suggested references

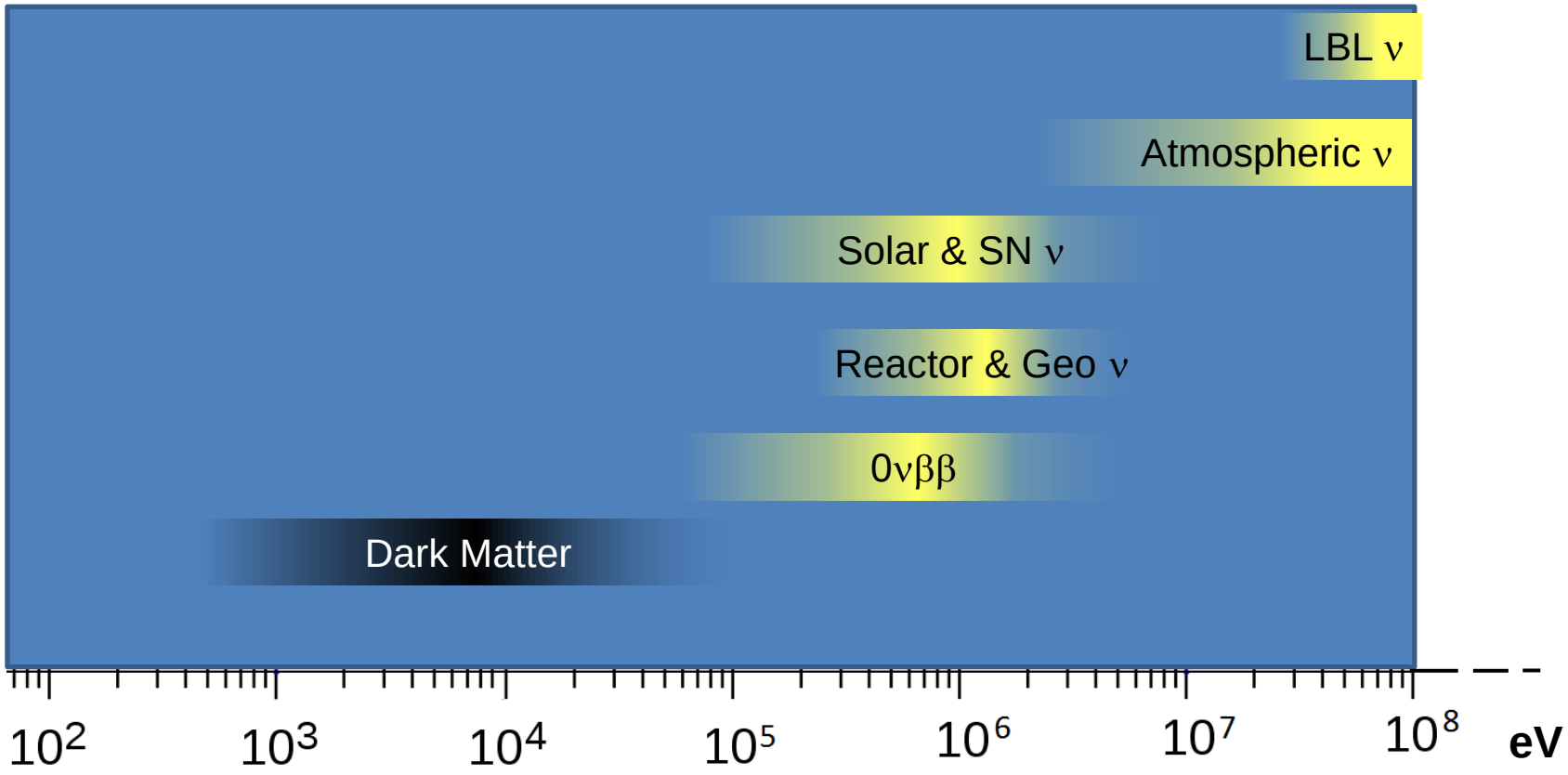
- Heusser "*Low-Radioactivity Background Techniques*"
  Ann. Rev. Nucl. Part. Sci. 45: 543 (1995).
- Formaggio J. A., Martoff C. J., "*Backgrounds to sensitive experiments underground*", Ann. Rev. Nucl. Part. Sci. 54, 361-412 (2004).
- S. Cebrián, "*Cosmogenic Activation in Double Beta Decay Experiments*", Universe 6 (2020) 10, 162 .
- M. Wojcik, G. Zuzel, "*Review of high-sensitivity Radon studies*", *Int.J.Mod.Phys.A* 32 (2017) 30, 1743004.
- M. F. L'Annunziata, "*Handbook of Radioactivity Analysis*", Elsevier.
- G.R. Gilmore, "*Practical gamma-ray spectrometry by Gordon Gilmore*", John Wiley & Sons.
- Several "*material-assay*" papers from different low-background experiments (DarkSide, XENON, EXO, LZ, LEGEND, NEXT, etc) .

What you are expected to know:

- Principles of radioactive decay
- Principles of nuclear radiation
- Principles of radiation interaction with matter

Rare events searches: No worries!... We are going to quickly summarize together some general aspects related to the rare-events searches and low-background techniques

Rare-event searches: solar and SN neutrinos, double-beta decay, Dark Matter particles…



Typical signal energy scale

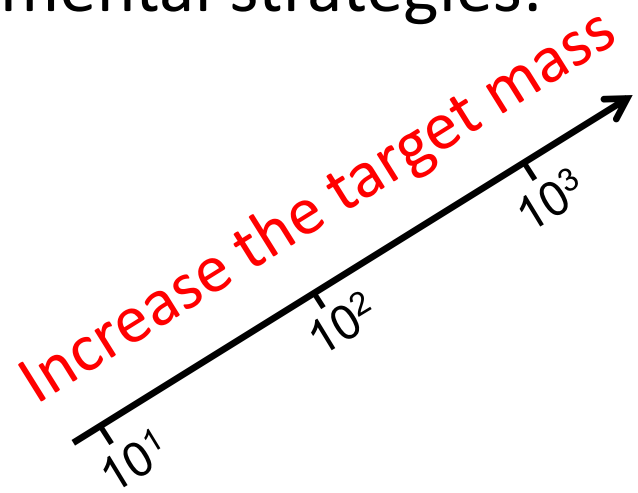
Rare-event searches: solar and SN neutrinos, double-beta decay, Dark Matter particles…

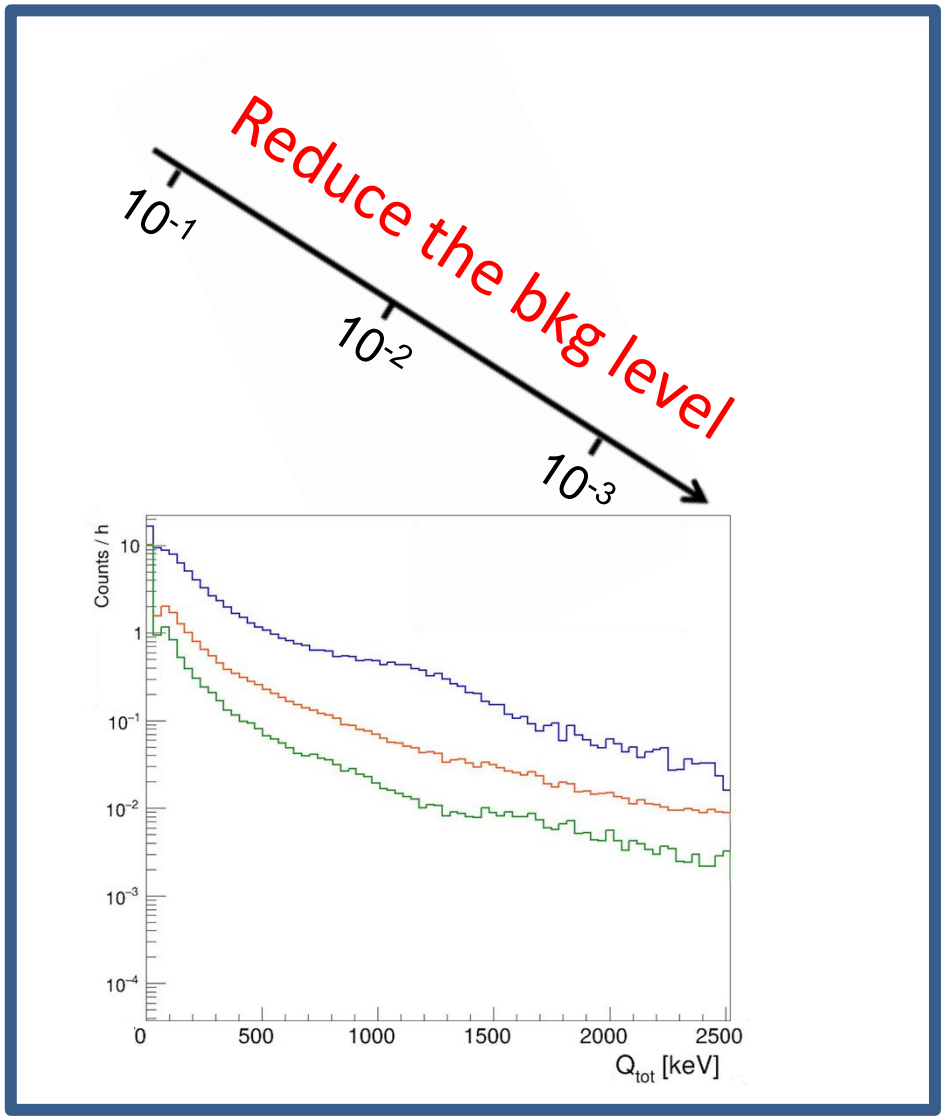
Rates as low as a few events per year are predicted

- *Improved detector technology*
- *Optimized detector design*
- *Improved techniques to extract weak signals over background and noise (e.g. machine learning)*
- *Improved detector modeling, etc…*

Rare-event searches: solar and SN neutrinos, double-beta decay, Dark Matter particles…

Two fundamental strategies:





Backgrounds

- Although the background sources in rare-event search experiments depend on the specific experiment, the main sources are generally:
  - cosmic rays
  - radiation from the materials surrounding the detector
  - radon in the experimental hall

➢**These backgrounds can be shielded and vetoed (more or less efficiently)**

Setting the stage: material-induced backgrounds

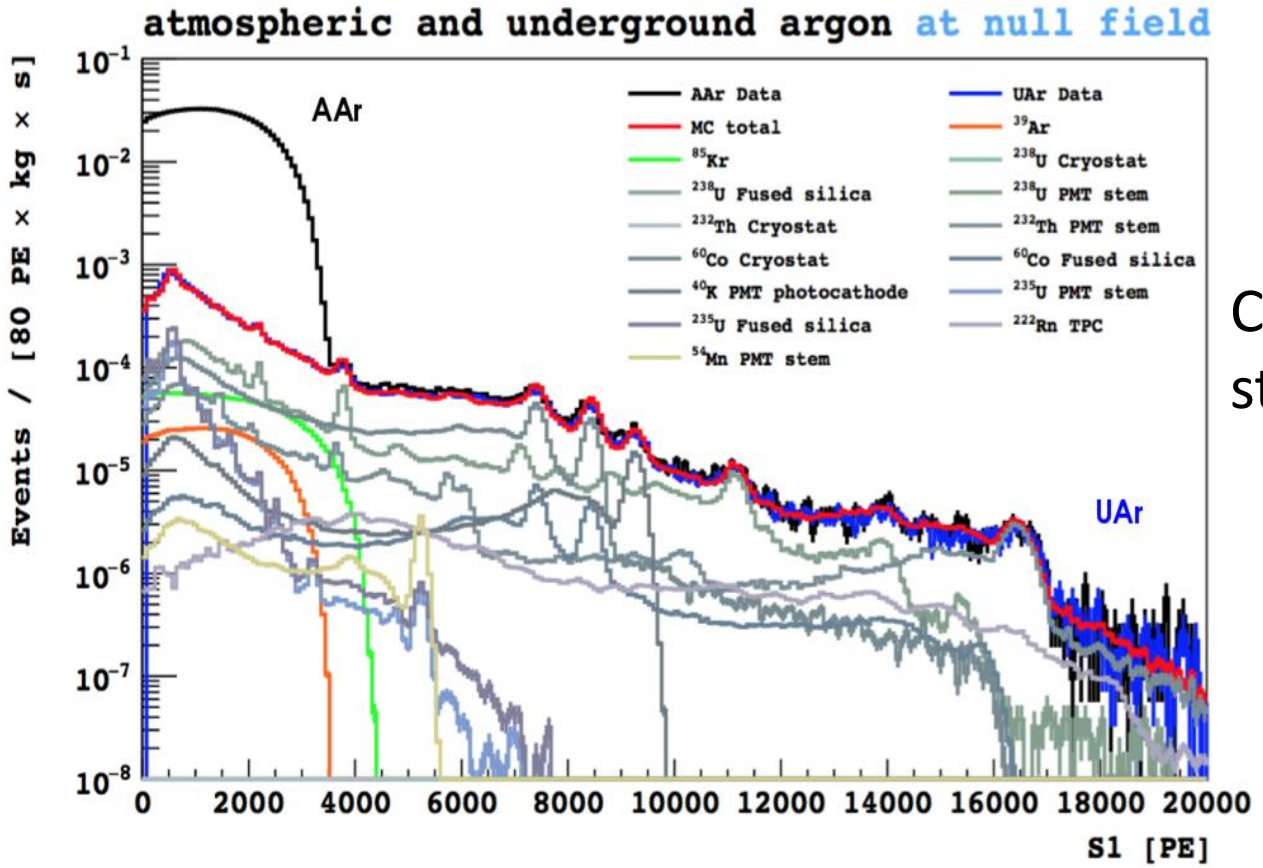
- **α, β, γ, from the detector materials**, especially those surrounding the active volume, can produce the ultimate background for the UG experiments
- It is very difficult to mit … nd
  Limited impact of acti… overburden ineffective…
- A simple and extremely … e extent) is to minimize the amount of material …

- The **radiopurity** of the d… l for achieving the sensitivity required for r…

Material radiopurity: general aspects

In rare-event searches, the background must be carefully assessed: all detector components have to be assayed

DS-50 @ LNGS



Cryostat, sensors, support structure, Ar…

Radiopurity assessment strategy

- **Dedicated measurements to ensure the radiopurity of the materials used in the detector's construction:**
  - ➢ **Quantify the content of radionuclides**

    Multiple assays methods and facilities: gamma counting, ICPMS, radiochemistry…

- **Simulation and calculation of radiation emission**
  - ➢ **quantify their impact on detector background**

    Calculation of the yields of primary and secondary particles, particle tracking …

Sample, vendor, manufacturing, storage history

- The radiopurity of commercial raw materials from **different suppliers** can vary by orders of magnitude
- The contamination level can differ, even in **batches** of the same material with different geological or chemical histories because of differences in the chemical composition of ores
- **Clean manufactoring**: materials can be contaminated during manufacturing or synthesis through the intentional or unintentional use of radioactively contaminated catalysts, molds, lubricants, or additives.
- Prevent **recontamination** through appropriate storage and handling of clean materials

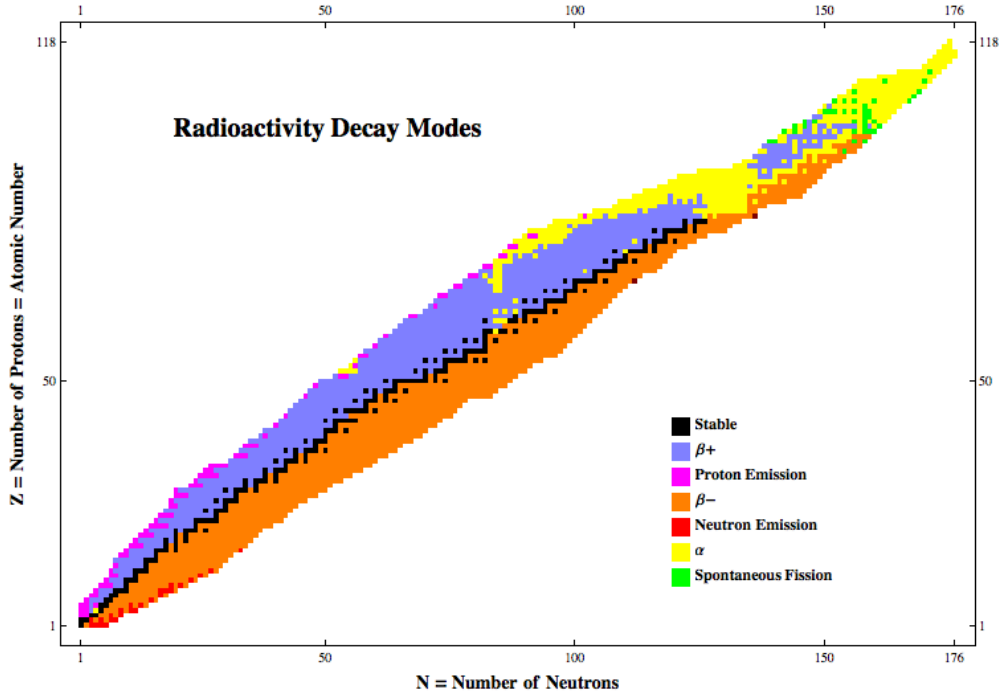
Art?

“Selecting candidate low-radioactivity materials is as much an art as it is a science”

(*Nucl.Instrum.Meth.A* 839 (2016) 6-11)

Radionuclides

https://www-nds.iaea.org/relnsd/vcharthtml/VChartHTML.html



- For light stable nuclei, the number of neutrons is approximately equal to the number of protons (This reflects the structure of nuclear energy levels, in loose analogy with electron shells)
- For light stable nuclei, N≈Z up to about calcium, (Z=20). For heavier stable nuclei, neutron excess increases with Z
- *Above Z* = 82 (or 83), no stable isotopes of the elements discovered so far

N of parent atoms decreases *exponentially* with time

$$N = N_0 e^{-t/\tau}$$

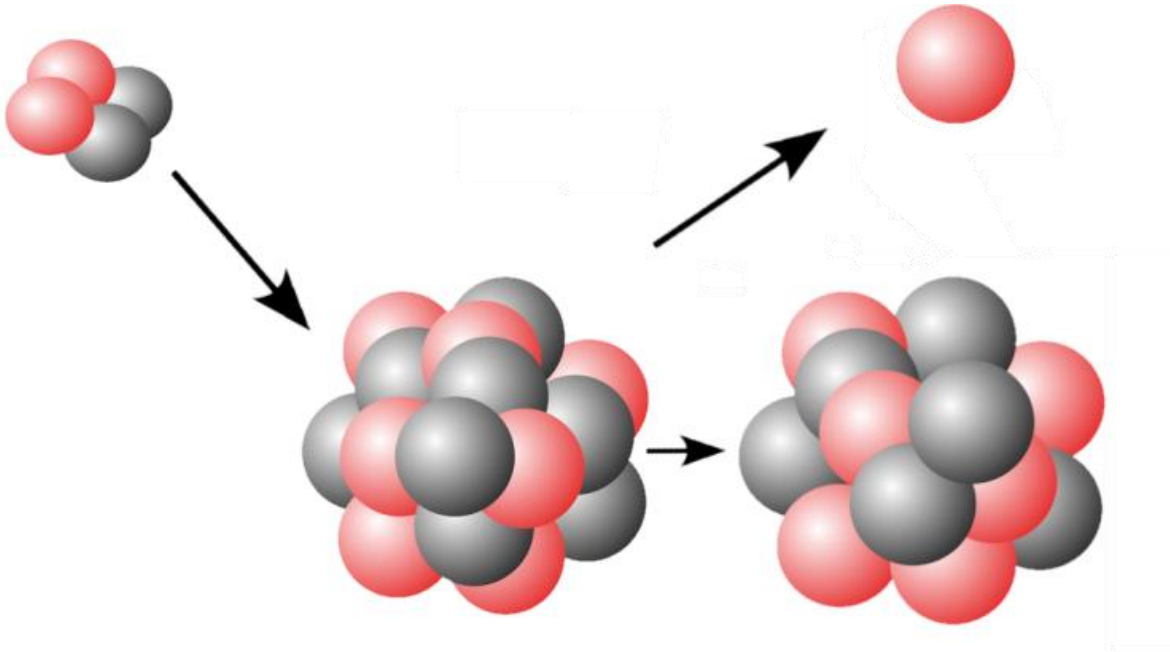
λ is the *decay constant* of a given radionuclide

$\rightarrow A = N\lambda = N/\tau$

The *half-life* $t_{1/2}$ is the time required for half of the radioactive nuclei in a sample to decay

$$t_{1/2} = (\ln 2)/\lambda = 0.693 \cdot \tau$$

α,β,γ,….and neutrons



α-emitters produce secondary neutrons
mainly through
(*α, n*) reactions
+
S.F.

What we have to measure?

How can we measure it?

What we have to measure?

How can we measure it?

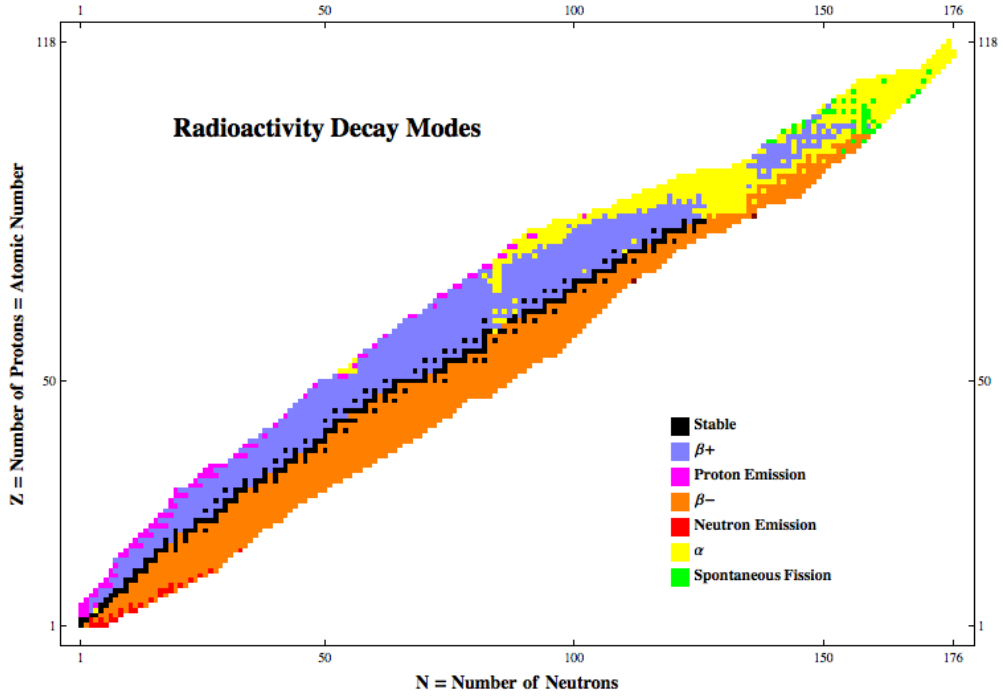
Naturally occurring radionuclides

- **Primordial**

  Found on the Earth and existing in their current form since before Earth was formed

- **Cosmogenic**

  Production of unstable nuclei by cosmic rays entering the Earth's atmosphere



Artificial radionuclides

- **Anthropogenic**

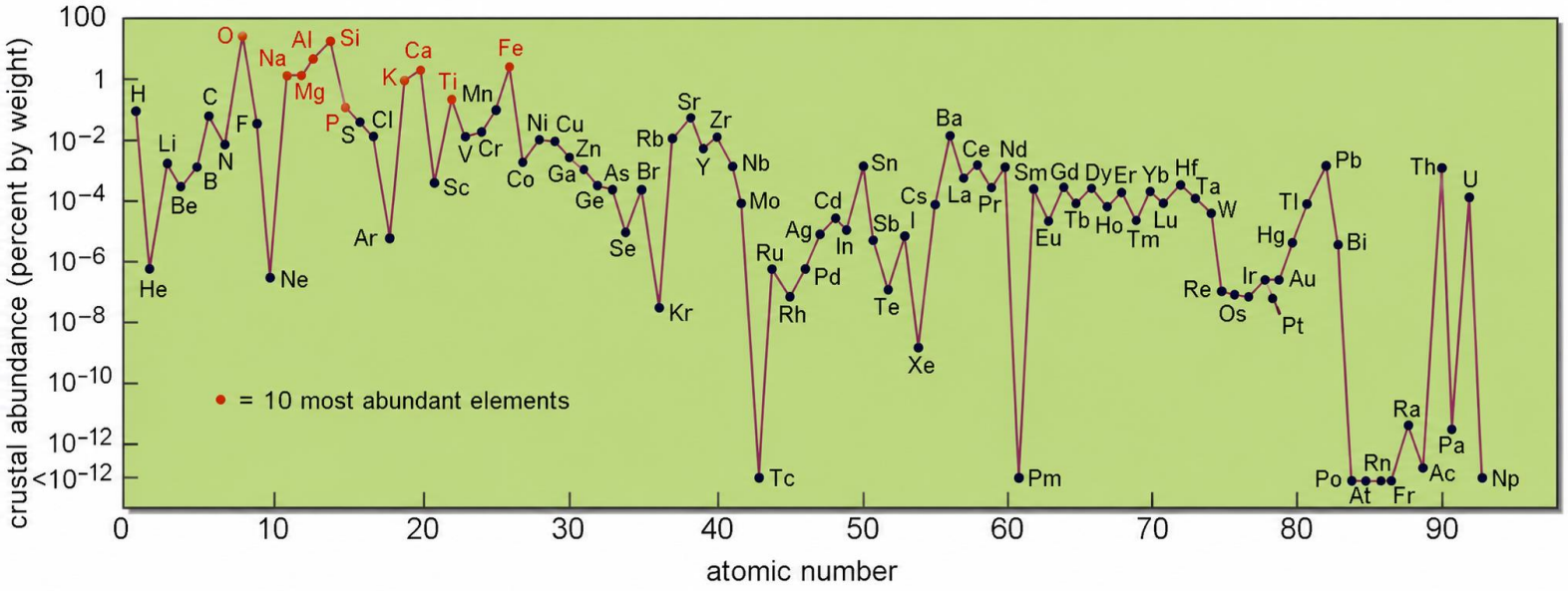
  Nuclear tests, nuclear waste, accelerators…

Relevant primordial radionuclides

For low-background experiments, the relevance of primordial radionuclides is driven by:

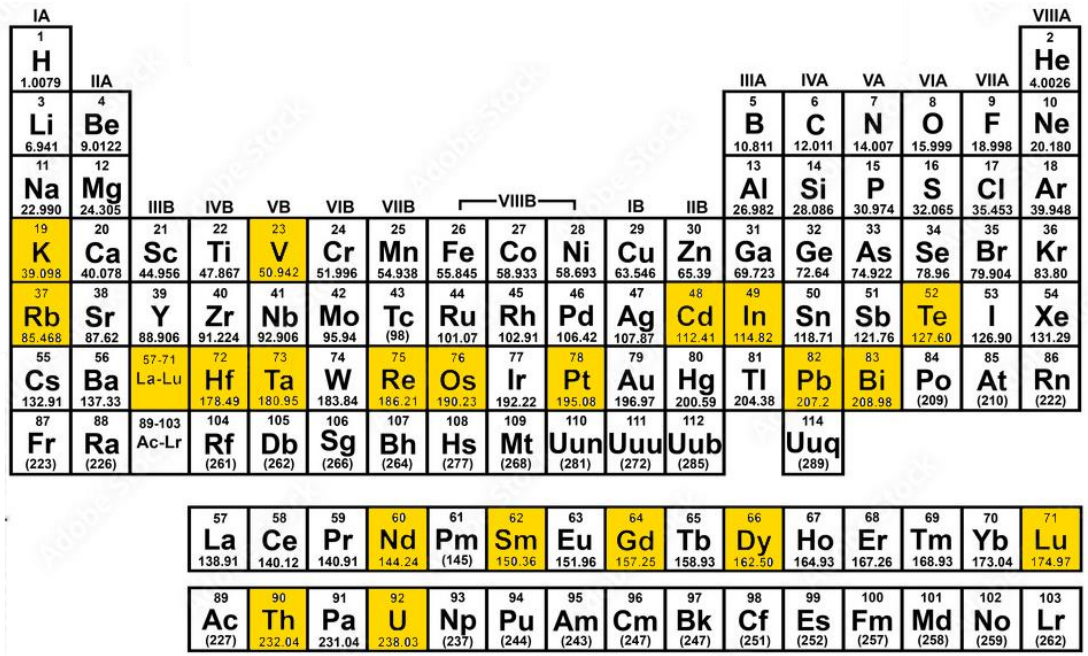
- Abundance of the chemical element in the Earth's crust
- Isotopic abundance of the radioactive isotope

Abundance of the chemical elements in the Earth's upper continental crust



Primordial radionuclides

- There are ≈252 stable nuclides (it depends on the definition of "stability"…)
- On Earth, $\sim 4.5 \times 10^9$ yr after its formation, most of the original unstable nuclei have decayed leaving only those with $\boldsymbol{t_{1/2} > 10^8}$ **yr**
- ≈34 radionuclides that have half-lives long enough to have survived since the formation of the Earth 4.5×10^9 years (isotopes of 28 separate elements)
- These long-lived nuclei undergo either highly forbidden β decays, involving large spin changes, or α decays with Q-values that lead to half-lives in this range



Most of them have half-lives significantly longer than the age of the Universe
(they are practically stable, with an abundance similar to the one of the stable isotopes of their respective elements)

There are only a handful of nuclides with half-lives comparable to the age of the Earth

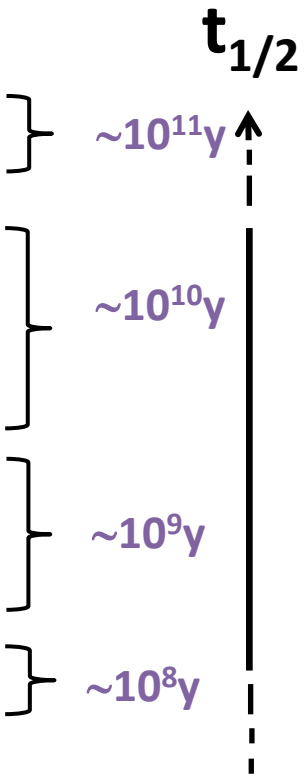
Relevant primordial radionuclides

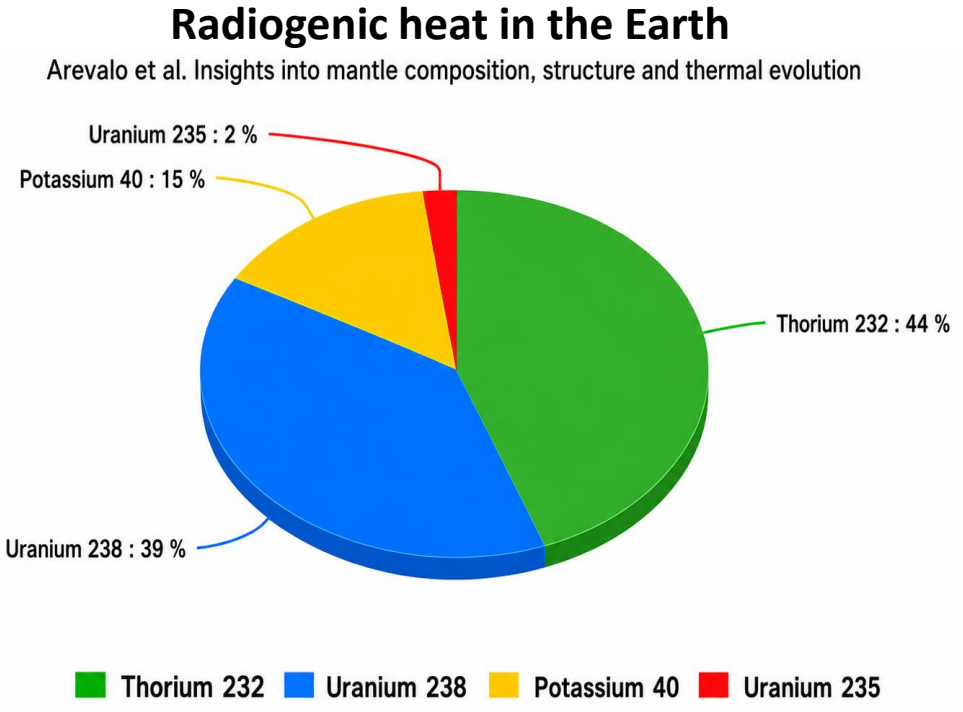
For low-background experiments, the relevance of primordial radionuclides is driven by:

- Abundance of the chemical element in the Earth's crust
- Isotopic abundance of the radioactive isotope
- Half-life: long enough to survive since Earth formation, but short enough to produce measurable activity

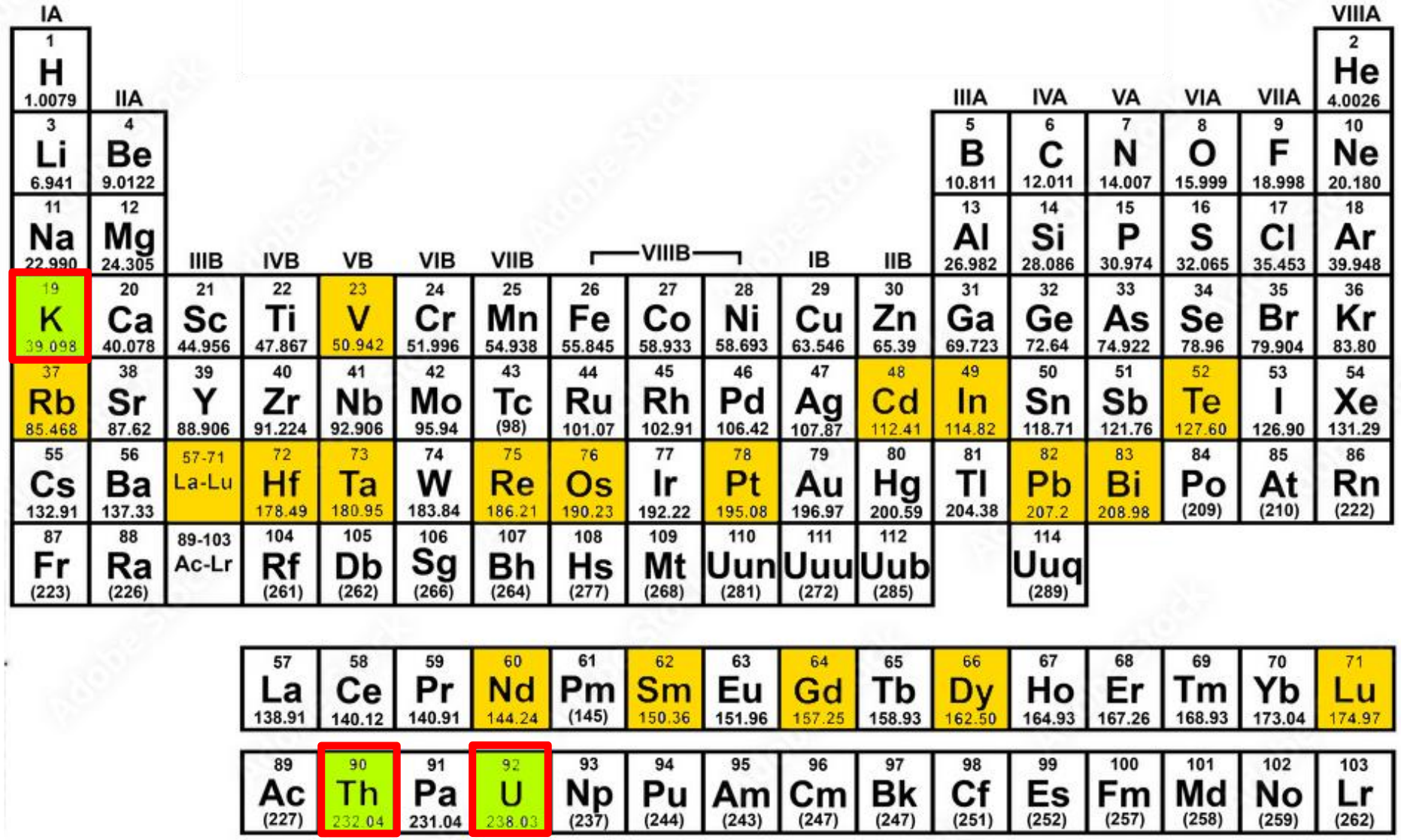
decay	half-life (years)	isotopic abundance (percent)	activity ($Bq\,kg^{-1}$) (element)	activity ($Bq\,kg^{-1}$) (crust)
$^{147}Sm \rightarrow {}^{143}Nd\,\alpha$	1.06×10^{11}	15.1	1.3×10^{5}	9×10^{-1}
$^{87}Rb \rightarrow {}^{87}Sr\,e^{-}\bar{\nu}_e$	4.75×10^{10}	27.83	8.8×10^{5}	8.0×10^{1}
$^{187}Re \rightarrow {}^{187}Os\,e^{-}\bar{\nu}_e$	4.15×10^{10}	62.6	1.1×10^{6}	8×10^{-4}
$^{176}Lu \rightarrow {}^{176}Hf\,e^{-}$	3.78×10^{10}	2.61	5.5×10^{4}	4×10^{-2}
$^{232}Th \rightarrow {}^{228}Ra\,\alpha$	1.405×10^{10}	100	4.05×10^{6}	3.5×10^{2}
$^{238}U \rightarrow {}^{234}Th\,\alpha$	4.468×10^{9}	99.275	1.2×10^{7}	4.7×10^{2}
$^{40}K \rightarrow {}^{40}Ca\,e^{-}\bar{\nu}_e$ 89% $\rightarrow {}^{40}Ar\,\nu_e$ 11%	1.28×10^{9}	0.0117	3.0×10^{4}	6.3×10^{2}
$^{235}U \rightarrow {}^{231}Th\,\alpha$	7.038×10^{8}	0.72	5.7×10^{5}	1.7×10^{1}
$^{146}Sm \rightarrow {}^{142}Nd\,\alpha$	1.03×10^{8}	$<10^{-7}$	<1	$<10^{-4}$

"Fundamentals In Nuclear Physics From Nuclear Structure to Cosmology" - Springer





U, Th, and K dominate because they combine natural abundance, suitable half-lives, and radiologically relevant decay emissions.



^{40}K

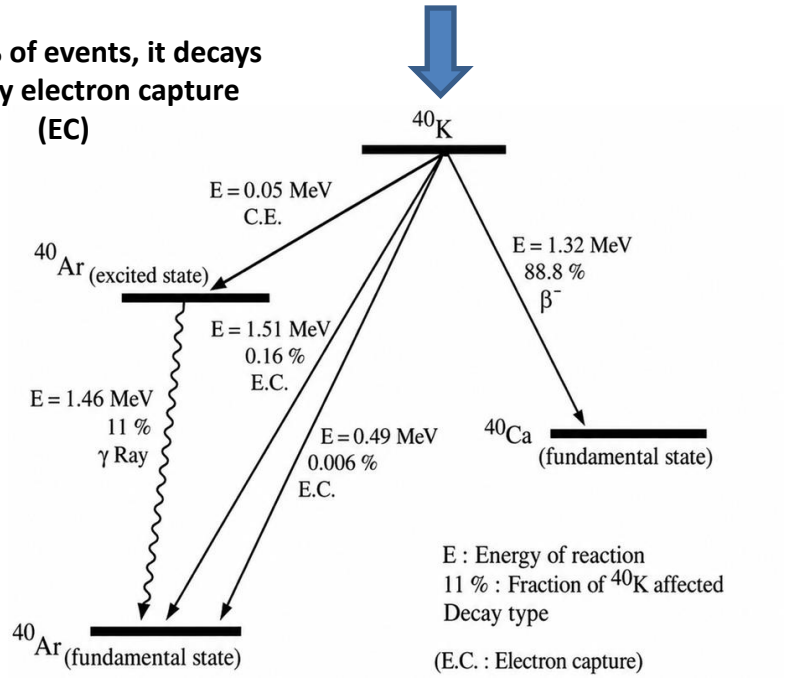
Potassium is one of the most common elements on the Earth's crust

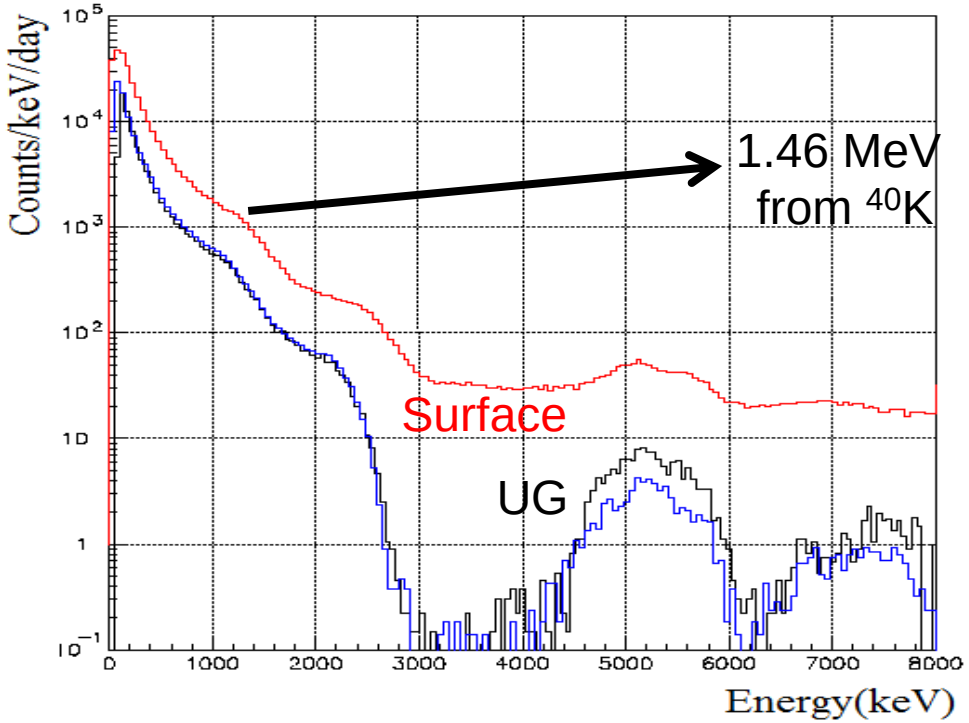
^{40}K emits detectable gamma rays

Stable	Radioactive	Stable
93.269%	0.0117%	6.739%
$^{39}_{19}K$	$^{40}_{19}K$	$^{41}_{19}K$

Half-life = 1.2×10^9 years

In 10.72% of events, it decays to ^{40}Ar by electron capture (EC)





- K content of the body is 0.2 %
- ^{40}K in humans → primary source of radiation from our body (practically constant)

For a 70-kg person, the amount of ^{40}K will be about 4.26 kBq

^{238}U, ^{235}U, ^{232}Th

They do not decay directly to a stable state: each initiates a **decay chain**

Altogether, these chains include ≈42 radionuclides

The mass number of the members may be expressed as four times an appropriate integer (n) plus the constant for that series. Three naturally occurring series are:

Thorium series (4n): ^{232}Th ($t_{1/2}$ =1.4·10^{10} y) → 10 radionuclides → ^{208}Pb (^{232}Th ≈100% ^{nat}Th)

Uranium series (4n+2) : ^{238}U ($t_{1/2}$= 4.5·10^{9} y) → 17 radionuclides → ^{206}Pb (^{238}U ≈99.3% ^{nat}U)

Actinium series (4n+3): ^{235}U ($t_{1/2}$= 7·10^{8} y) → 15 radionuclides → ^{207}Pb (^{235}U ≈0.7% ^{nat}U)

The neptunium series with Np-237 (with a half-life of 2 million years) 4n+1 series do not occur naturally, because the half-life of the longest lived isotope in the series is short compared to the age of the Earth.

The Np decay chain has no long-lived nuclei heavier than ^{209}Bi.

→ *The chain thus consists of a single decay ^{209}Bi → $^{205}Tl_{(stable)}$ ($t_{1/2}$ = 1.9 × 10^{19} y)*

^{238}U, ^{235}U, ^{232}Th

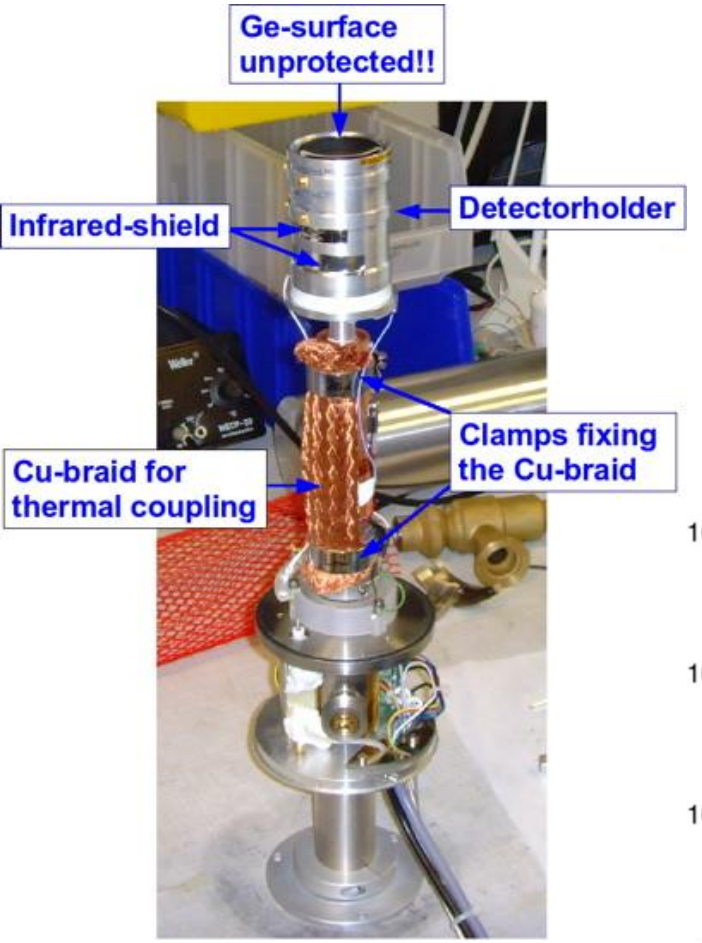
α decay

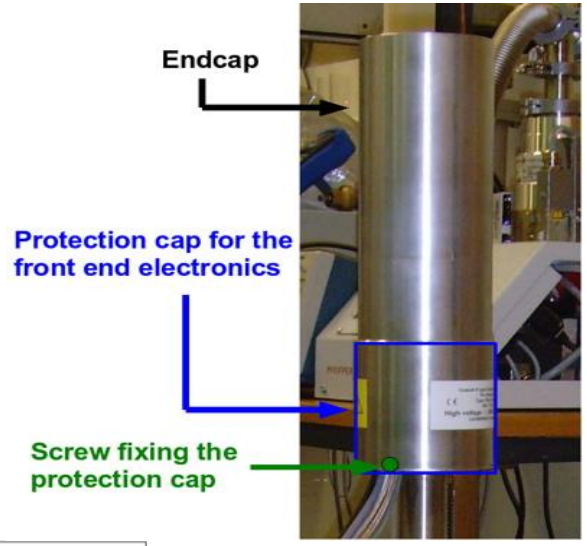
β decay

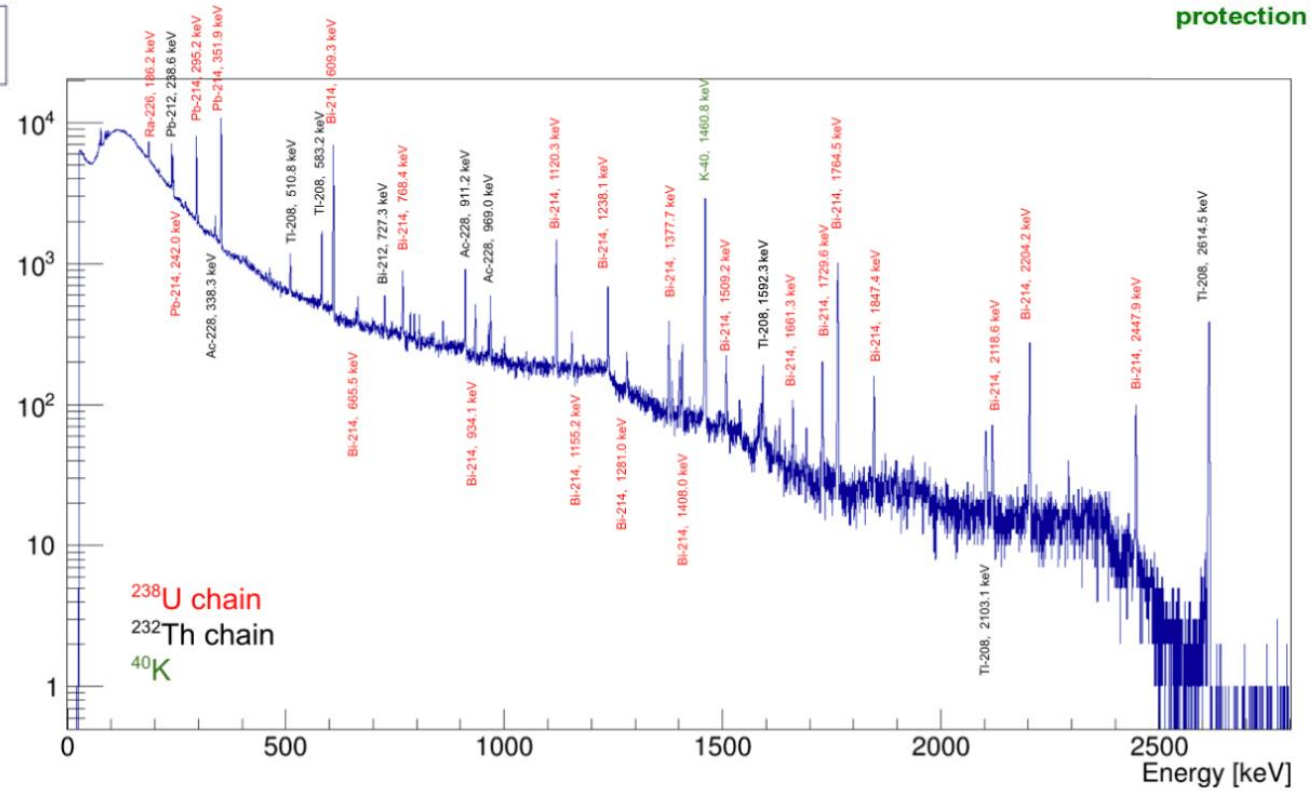
α or β decay

uranium series	actinium series	thorium series
$^{238}U_{92}$ 4.468 Gyr		
$^{234}Th_{90}$ 24.10 d		
$^{234}Pa_{91}$ 1.17 m		
$^{234}U_{92}$ 245.5 kyr	$^{235}U_{92}$ 0.7038 Gyr	
$^{230}Th_{90}$ 75.38 kyr	$^{231}Th_{90}$ 25.52 h	$^{232}Th_{90}$ 14.05 Gyr
$^{226}Ra_{88}$ 1600 y	$^{231}Pa_{91}$ 32760 y	$^{228}Ra_{88}$ 5.75 y
$^{222}Rn_{86}$ 3.8235 d	$^{227}Ac_{89}$ 21.773 y	$^{228}Ac_{89}$ 6.15 h
$^{218}Po_{84}$ 3.10 m	$^{227}Th_{90}$ 18.72 d	$^{228}Th_{90}$ 1.9116 y
$^{214}Pb_{82}$ 26.8 m	$^{223}Ra_{88}$ 11.435 d	$^{224}Ra_{88}$ 3.66 d
$^{214}Bi_{83}$ 19.9 m	$^{219}Rn_{86}$ 3.96 s	$^{220}Rn_{86}$ 55.6 s
$^{214}Po_{84}$ 164.3 µs	$^{215}Po_{84}$ 1.781 ms	$^{216}Po_{84}$ 0.145 s
$^{210}Pb_{82}$ 22.3 y	$^{211}Pb_{82}$ 36.1 m	$^{212}Pb_{82}$ 10.64 h
$^{210}Bi_{83}$ 5.013 d	$^{211}Bi_{83}$ 2.14 m	$^{212}Bi_{83}$ 60.55 m
$^{210}Po_{84}$ 138.376 d	$^{207}Tl_{81}$ 4.77 m	64% $^{212}Po_{84}$ 0.299µs
		36% $^{208}Tl_{81}$ 3.053 m
$^{206}Pb_{82} > 10^{20}$ yr	$^{207}Pb_{82} > 10^{20}$ yr	$^{208}Pb_{82} > 10^{20}$ yr

γ-spectroscopy with HPGe detectors







However….

In these chains there are alpha and beta emitters, most of which do not emit detectable gamma rays:

- Gamma spectroscopy alone is not sufficient

Equilibrium in the chain

Equilibrium means that the **activities** of the parent and daughter nuclides are the same

- If no progeny atoms are present at time $t = 0$, the number of progeny atoms, N_2, at any later time t is:

$$N_2(t) = [\lambda_1/(\lambda_2 - \lambda_1)]N_0(e^{-\lambda_1 t})$$ Bateman equation

N_0 is the number of parent atoms present at time $N_1(t = 0)$
λ_1 is the decay constant of the parent,
λ_2 is the decay constant of the progeny

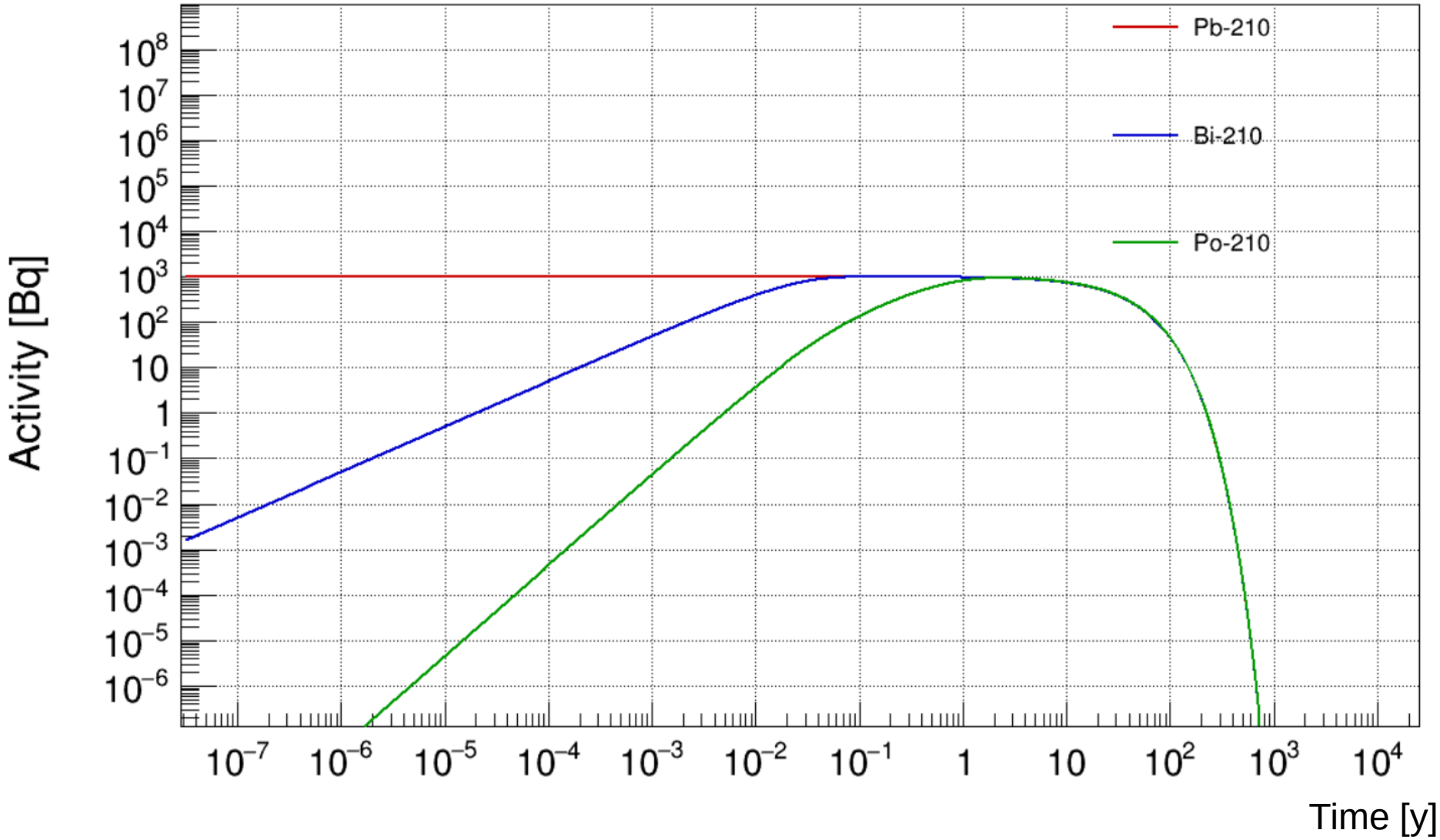
- If $(N_2)_0$ progeny atoms are present at time $t = 0$, the expression for $N2$ is written

$$N_2(t) = (N_2)_0 e^{-\lambda_2 t} + [\lambda_1/(\lambda_2 - \lambda_1)]N_0(e^{-\lambda_1 t} - e^{-\lambda_2 t})$$

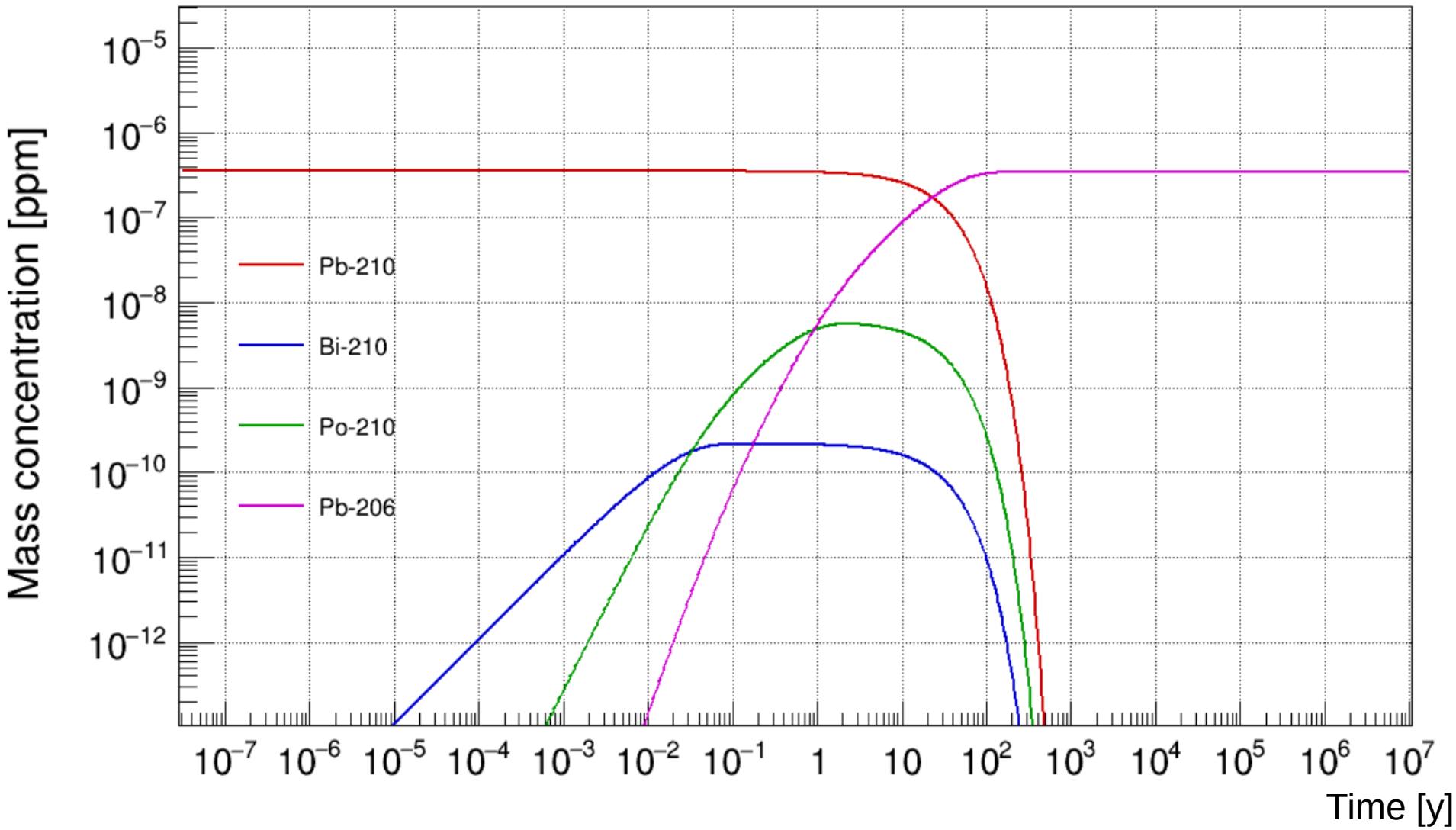
The apparent half-life of the progeny reflects the simultaneous production and decay of the progeny.
The ratio of activities $A1/A2$ for X and Y, respectively, is

$$A_1/A_2 = (\lambda_2 - \lambda_1)/\lambda_2$$

Example: Activity Evolution



Example: Abundance Evolution

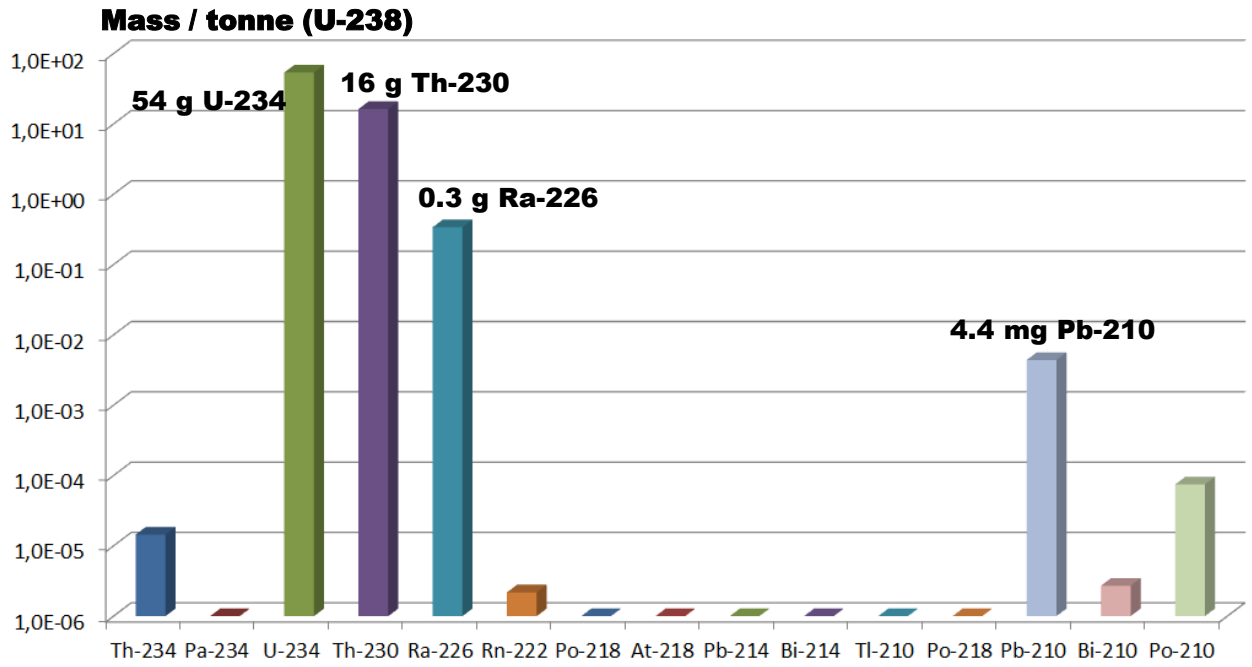


Secular equilibrium

"**Secular equilibrium**": the half-life of the parent nuclide is very long and the half-life of the daughter nuclide is considerably shorter than that of the parent

A radioactive nuclide decays at the same rate at which it is being produced:
→ the number of atoms present remains constant (over a certain period of time)

Over the 4.5 billion years of the Earth's history, all U and Th decay chains have reached equilibria between the parent nucleus and the various descendants (assuming that the system has been closed for millions of years)



- Minerals that contain uranium and thorium also contain virtually all daughter nuclides in their decay chains, provided the system has remained closed for a sufficiently long time
- In secular equilibrium, all nuclides in the chain have the same activity
- The mass of each daughter nuclide present in the mineral is proportional to its half-life

Disequilibrium in natural decay chains

Equilibrium in the decay series ***is rarely maintained*** in most surface and near-surface geological environments, as a result of physical or chemical processes:

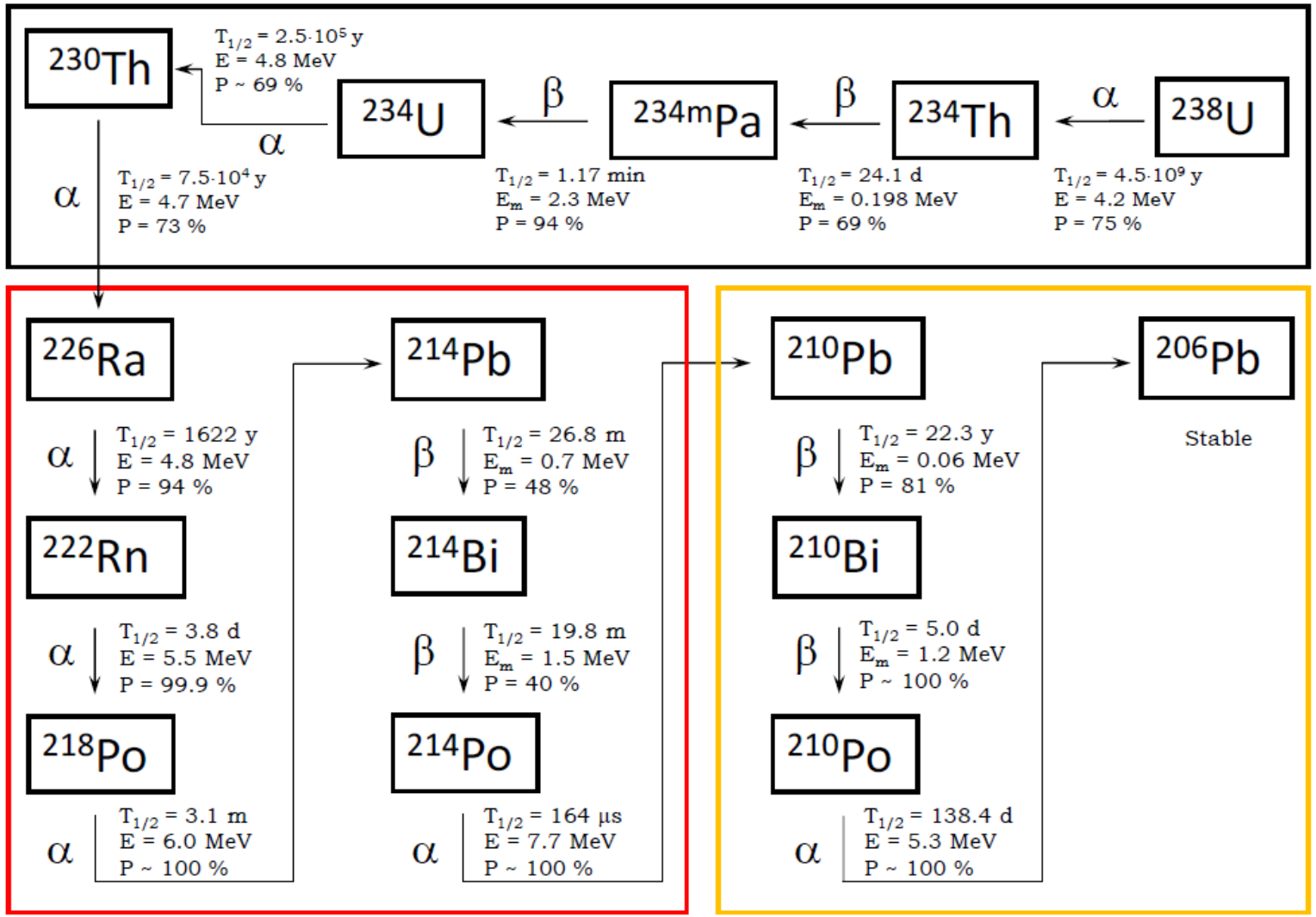
- chemical, artificial or natural processes (e.g. dissolution into groundwater) can remove one or more members of the chain.

If, for example, uranium is dissolved from a primary uranium-bearing mineral by oxidation, the remaining radionuclides in the ^{238}U series will be supported by its most long-lived radionuclide which is ^{230}Th. If the dissolved uranium is then precipitated somewhere out of the system a new equilibrium will begin to develop.

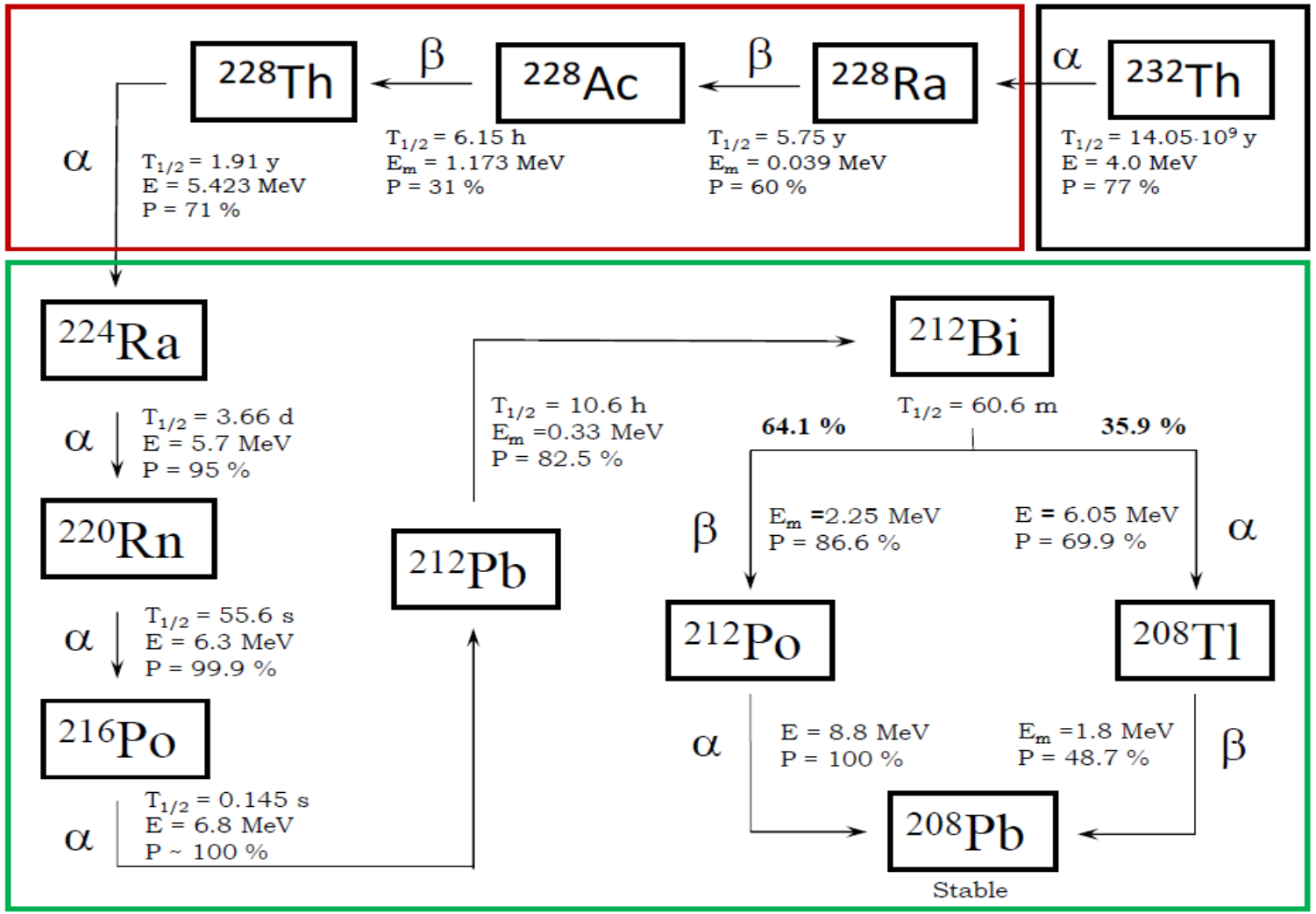
The time required to attain the equilibrium is governed by the most long-lived daughter radionuclide in the series, ^{230}Th in the case of ^{238}U series. The half-life of ^{230}Th is 75000 years and this time is required to reach 50% of equilibrium.

The disequilibria can be utilized in dating geological events.

^{238}U chain


230Th
T1/2 = 2.5·10^5 y
E = 4.8 MeV
P ~ 69 %
234U
α
234mPa
β
234Th
β
238U
α
α
T1/2 = 7.5·10^4 y
E = 4.7 MeV
P = 73 %
T1/2 = 1.17 min
Em = 2.3 MeV
P = 94 %
T1/2 = 24.1 d
Em = 0.198 MeV
P = 69 %
T1/2 = 4.5·10^9 y
E = 4.2 MeV
P = 75 %
226Ra
α
T1/2 = 1622 y
E = 4.8 MeV
P = 94 %
222Rn
α
T1/2 = 3.8 d
E = 5.5 MeV
P = 99.9 %
218Po
α
T1/2 = 3.1 m
E = 6.0 MeV
P ~ 100 %
214Pb
β
T1/2 = 26.8 m
Em = 0.7 MeV
P = 48 %
214Bi
β
T1/2 = 19.8 m
Em = 1.5 MeV
P = 40 %
214Po
α
T1/2 = 164 µs
E = 7.7 MeV
P ~ 100 %
210Pb
β
T1/2 = 22.3 y
Em = 0.06 MeV
P = 81 %
210Bi
β
T1/2 = 5.0 d
Em = 1.2 MeV
P ~ 100 %
210Po
α
T1/2 = 138.4 d
E = 5.3 MeV
P ~ 100 %
206Pb
Stable

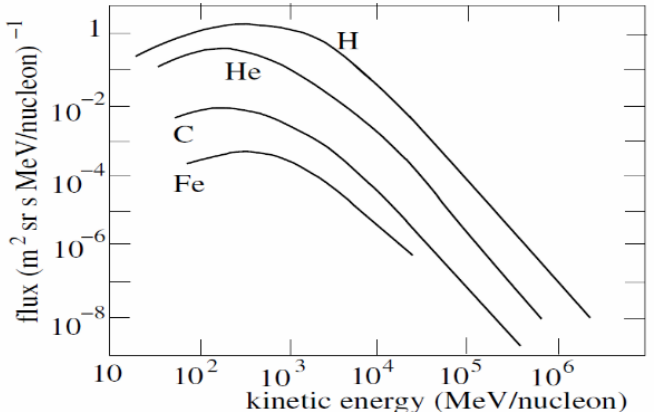
^{232}Th chain


232Th
α
T1/2 = 14.05·10^9 y
E = 4.0 MeV
P = 77 %
228Ra
β
T1/2 = 5.75 y
Em = 0.039 MeV
P = 60 %
228Ac
β
T1/2 = 6.15 h
Em = 1.173 MeV
P = 31 %
228Th
α
T1/2 = 1.91 y
E = 5.423 MeV
P = 71 %
224Ra
α
T1/2 = 3.66 d
E = 5.7 MeV
P = 95 %
220Rn
α
T1/2 = 55.6 s
E = 6.3 MeV
P = 99.9 %
216Po
α
T1/2 = 0.145 s
E = 6.8 MeV
P ~ 100 %
212Pb
T1/2 = 10.6 h
Em =0.33 MeV
P = 82.5 %
212Bi
T1/2 = 60.6 m
64.1 %
35.9 %
β
Em =2.25 MeV
P = 86.6 %
E = 6.05 MeV
P = 69.9 %
α
212Po
208Tl
α
E = 8.8 MeV
P = 100 %
Em =1.8 MeV
P = 48.7 %
β
208Pb
Stable

Cosmogenic radionuclides

Cosmic rays produce radioactive nuclei via their interactions with nuclei in the atmosphere and in the Earth's crust

The primary components of cosmic radiation are high-energy alpha particles and protons, which induce nuclear reactions when they impact on the nuclei of the atmospheric atoms (especially oxygen, nitrogen and argon)



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

$^{14}N + n \rightarrow {}^{12}C + {}^{3}H$

In the atmosphere, the most common radioactivity produced is that of ^{14}C
^{14}C is mixed throughout the atmosphere and enters the food chain through CO_2 ingesting plants

These radionuclides are isotopes of lighter elements, and their half-lives vary widely

Nuclide	Half-life	Nuclide	Half-life
^{3}H	12.3 a	^{7}Be	53 d
^{10}Be	$2.5 \cdot 10^6$ a	^{14}C	5730 a
^{22}Na	2.62 a	^{26}Al	$7.4 \cdot 10^5$ a
^{32}Si	710 a	^{32}P	14.3 d
^{33}P	24.4 d	^{35}S	88 d
^{36}Cl	$3.1 \cdot 10^5$ a	^{39}Ar	269 a

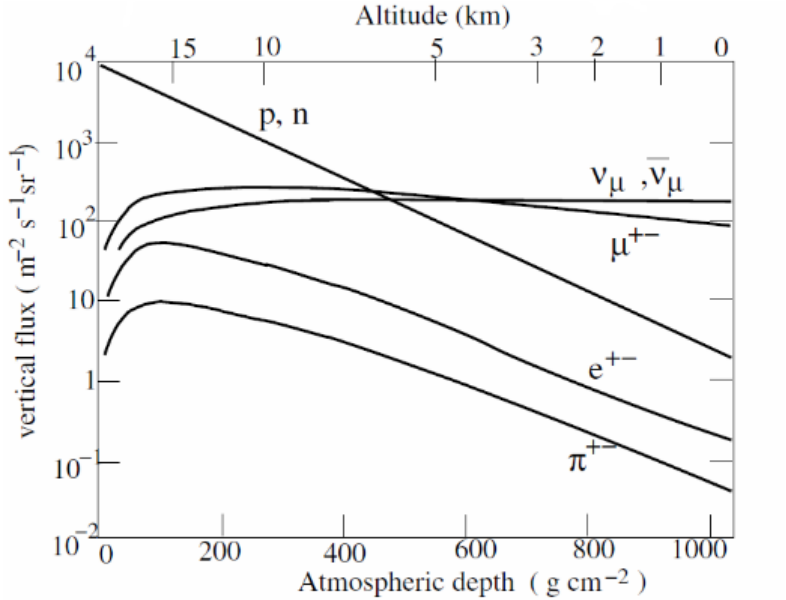
The total production rate R of a radioisotope is obtained by summing over all target nuclei i and over all cosmic-ray particle components j:

$$R = \sum_i N_i \sum_j \int \Phi_j(E)\sigma_{ij}(E)dE$$

N_i is the number of target nuclei
$\varphi_j(E)$ is the energy-dependent cosmic-ray flux
$\sigma_{ij}(E)$ is the production cross section for isotope i induced by particle j
(integral accounts for the contribution of all incident particle energies)

Cosmogenic radionuclides

There is, however, great variation at different heights of the atmosphere and at different latitudes.



Fundamentals in Nuclear Physics - From Nuclear Structure to Cosmology – Basdevant, Rich, Spiro

- The total intensity of these secondary's reaches a maximum at an atmospheric depth of about 150 g/cm (~13.7 km altitude) and then falls off gradually
- Most of the cosmogenic radionuclides are attached to aerosol particles and are deposited on the ground. Some, however, are gaseous, such as ^{39}Ar (a noble gas), and thus stay in the atmosphere.

At sea level (and obviously U.G) the muon flux dominates

- *Muon-induced spallation*: a high-energy muon emits a hard photon (virtual or real), which interacts photonuclearly with the nucleus and fragments it, producing radioactive residual isotopes
- *Electromagnetic/photonuclear showers*: muon-induced bremsstrahlung photons and $e^{\pm}$ cascades generate hard photons that activate nuclei through photonuclear reactions such as (γ,n)
- *Muon-induced neutrons*: secondary neutrons produced as by-products of muon-induced spallation or photonuclear reactions in electromagnetic showers, which then activate nuclei via reactions such as (n,γ), (n,p), or (n,2n)
- *Negative-muon capture*: stopped μ^- are captured by nuclei, converting a proton into a neutron and leaving the nucleus excited, possibly producing radioactive isotopes

Cosmogenic radionuclides

For example neutron "**spallation**" reactions:

- A neutron with kinetic energy greater than ~20 MeV can remove two neutrons for a germanium nucleus,

  e.g. $n + {}^{70}Ge \rightarrow 3n + {}^{68}Ge$ $t_{1/2}({}^{68}Ge) = 270.7\text{day}$

  This reaction results in an activity of ~0.3 mBq kg^{-1} in Ge crystals produced at the Earth's surface
  This is the most important source of intrinsic radioactivity in the HPGe crystals

- A fast neutron with kinetic energy above the (n,2n) threshold can remove one neutron from atmospheric argon,

  e.g. $n + {}^{40}Ar \rightarrow 2n + {}^{39}Ar$ $t_{1/2}({}^{39}Ar) = 269\text{ yr}$

  This cosmogenic production leads to an activity of ~1 Bq kg^{-1} in atmospheric argon
  This is the dominant intrinsic radioactive background of Ar_{Atm} in low-background LAr detectors

Cosmogenic radionuclides

- The **activation** by the hadronic component can reach specific radioactivity levels higher than the residual contamination from primordial nuclides.

- Since the intensity of cosmic radiation is constant over the long-term, the production of cosmogenic radionuclides is rather constant.

- Materials have to be **stored underground** (the production of cosmogenic radionuclides stops and the activity decays away).

- *Materials extracted UG should be largely depleted in cosmogenic isotopes.*

Rates of production of long-lived radionuclides at sea level (atoms/kg/day)

Copper

	^{46}Sc	^{48}V	^{54}Mn	^{56}Co	^{57}Co	^{58}Co	^{59}Fe	^{60}Co
Half-life[27,108]	83.787(16)	15.9735	312.19(3)	77.236	271.81(4)	70.85(3)	44.494	5.2711(8)
units	d	d	d	d	d	d	d	y
Measurement [202]	2.18 ± 0.74	4.5 ± 1.6	8.85 ± 0.86	9.5 ± 1.2	74 ± 17	67.9 ± 3.7	18.7 ± 4.9	86.4 ± 7.8
Measurement [184]	$2.33^{+0.95}_{-0.78}$	$3.4^{+1.6}_{-1.3}$	$13.3^{+3.0}_{-2.9}$	$9.3^{+1.2}_{-1.4}$	$44.8^{+8.6}_{-8.2}$	$68.9^{+5.4}_{-5.0}$	$4.1^{+1.4}_{-1.2}$	$29.4^{+7.1}_{-5.9}$
ACTIVIA (MENDL-2P) [36]	3.1		12.4	14.1	36.4	38.1	1.8	9.7
ACTIVIA [36,184]	3.1		14.3	8.7	32.5	56.6	4.2	26.3
COSMO [184]	1.5	3.1	13.5	7.0	30.2	54.6	4.3	25.7
ACTIVIA [46]	4.1		30.0	20.1	77.5	138.1	10.5	66.1
ACTIVIA [99]	3		16	9	34	60	2	29
GEANT4 [46]	1.2		12.3	10.3	67.2	57.3	8.8	64.6
TALYS [94]			16.2		56.2			46.4
MENDL+YIELDX [43]	2.7		27.7	20.0	74.1	123.0	4.9	55.4
CONUS [99]	3		14	10	50	76	5	92

SS

Isotope	^{7}Be	^{46}Sc	^{48}V	^{54}Mn	^{56}Co	^{58}Co
Half-life (d) [27,108]	53.22(6)	83.787(16)	15.9735	312.19(3)	77.236	70.85(3)
Measurement [202]	389 ± 60	19.0 ± 3.5	34.6 ± 3.5	233 ± 26	20.7 ± 3.5	51.8 ± 7.8
GEANT4 [46]	0.05	8.8		230	16	90
ACTIVIA [46]	2.05	18		191	131	13

Lead

	^{194}Hg	^{202}Pb	^{207}Bi
Half-life (y) [27,108]	444	$5.25\ 10^{4}$	32.9
Measurement [205]	8.0 ± 1.3	120 ± 25	<0.17
TALYS [205]	16	77	

(Universe 6 (2020) 10, 162)

Artificial radionuclides

Sources of artificial radionuclides:

- *nuclear weapons production and explosions;*
- *nuclear energy production;*
- *radionuclide production by reactors and accelerators (medical application etc…)*

Artificial radionuclides form the **largest group of radionuclides**, comprising more than two thousand nuclides.

The explosion clouds of the most powerful tests entered the upper part of the atmosphere, the stratosphere, from where the radioactive pollutants have deposited on a global scale.

- *Cs-137 ($T_{1/2}$=30.2 y), along with iodine-131, xenon-133, and strontium-90, has been released into the environment by nuclear weapons tests and major nuclear accidents.*

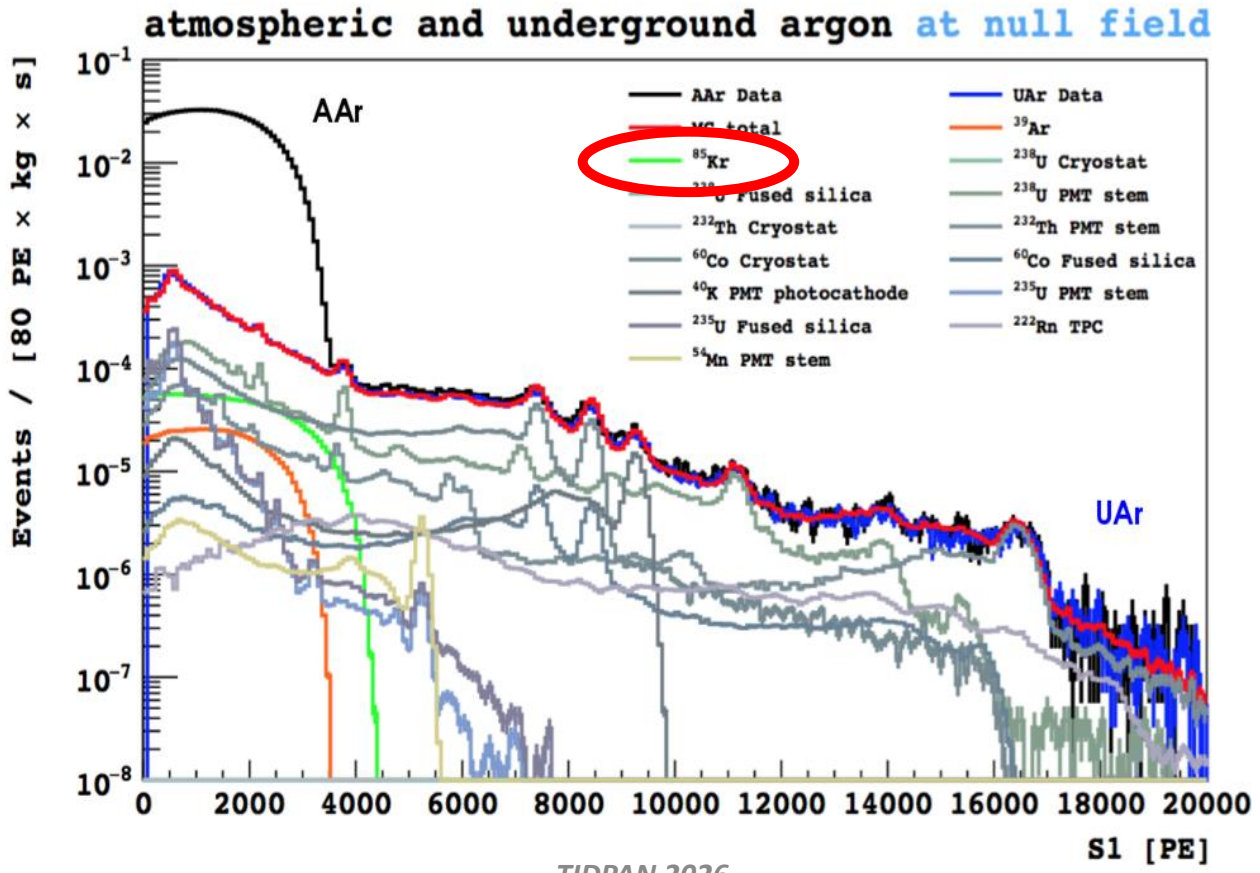
- *The mean Cs-137 contamination in Eastern Europe following the Chernobyl disaster was several kBq/m^2.*

Artificial radionuclides

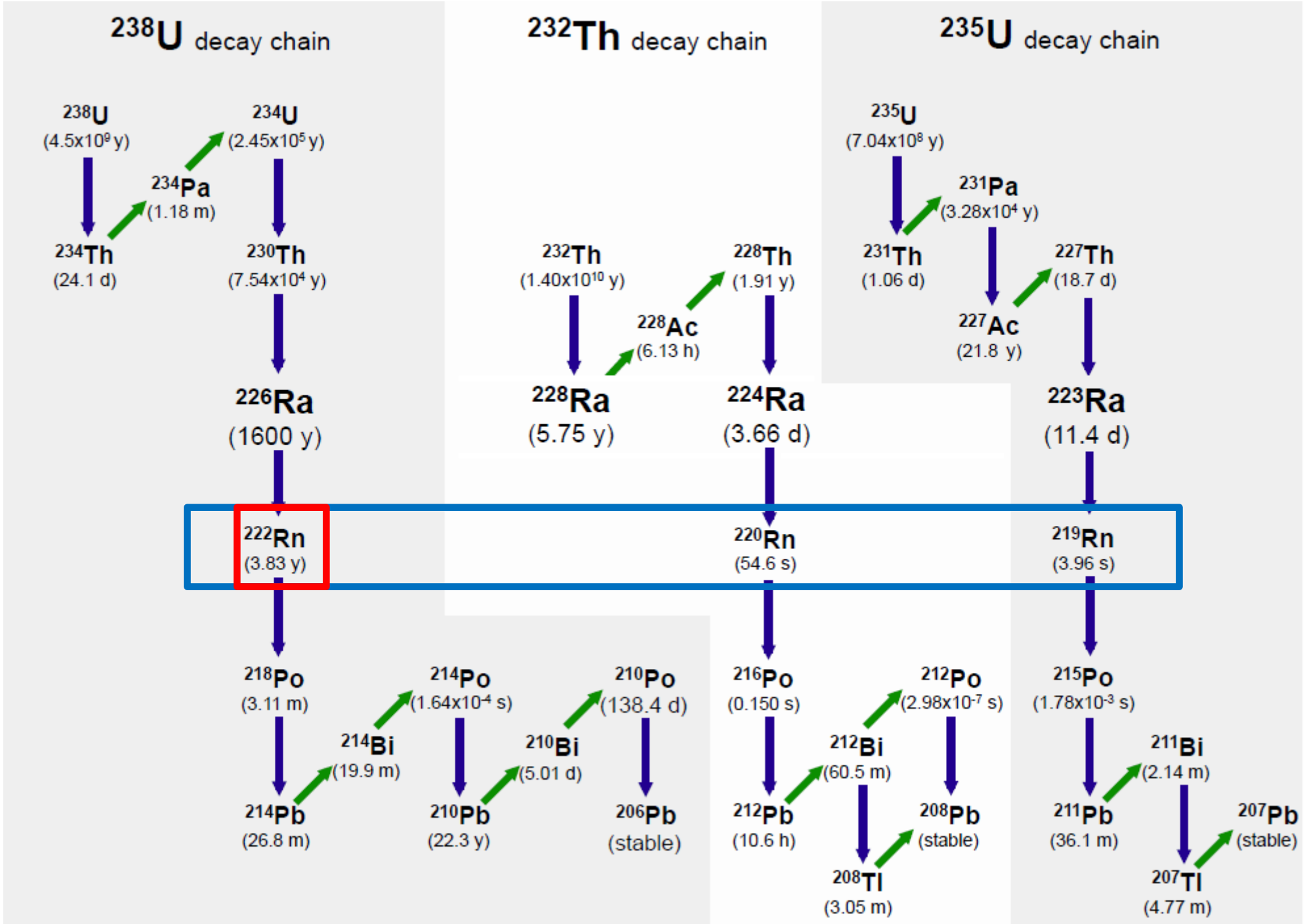
Kr-85 ($T_{1/2}$=10.8 y) is one of the seven common medium-lived fission products

Kr-85 is produced in small quantities by cosmic rays interactions with stable Kr-84 in the atmosphere. Natural sources maintain a steady-state inventory of about 0.09 PBq in the atmosphere.

The Fukushima accident released an estimated 44–84 PBq of Kr-85 in the atmosphere




DANGER
RADON GAS


238U decay chain
232Th decay chain
235U decay chain
238U (4.5x10⁹ y)
234U (2.45x10⁵ y)
234Pa (1.18 m)
234Th (24.1 d)
230Th (7.54x10⁴ y)
226Ra (1600 y)
222Rn (3.83 y)
218Po (3.11 m)
214Po (1.64x10⁻⁴ s)
214Bi (19.9 m)
210Po (138.4 d)
210Bi (5.01 d)
214Pb (26.8 m)
210Pb (22.3 y)
206Pb (stable)
232Th (1.40x10¹⁰ y)
228Th (1.91 y)
228Ac (6.13 h)
228Ra (5.75 y)
224Ra (3.66 d)
220Rn (54.6 s)
216Po (0.150 s)
212Po (2.98x10⁻⁷ s)
212Bi (60.5 m)
212Pb (10.6 h)
208Pb (stable)
208Tl (3.05 m)
235U (7.04x10⁸ y)
231Pa (3.28x10⁴ y)
231Th (1.06 d)
227Th (18.7 d)
227Ac (21.8 y)
223Ra (11.4 d)
219Rn (3.96 s)
215Po (1.78x10⁻³ s)
211Bi (2.14 m)
211Pb (36.1 m)
207Pb (stable)
207Tl (4.77 m)

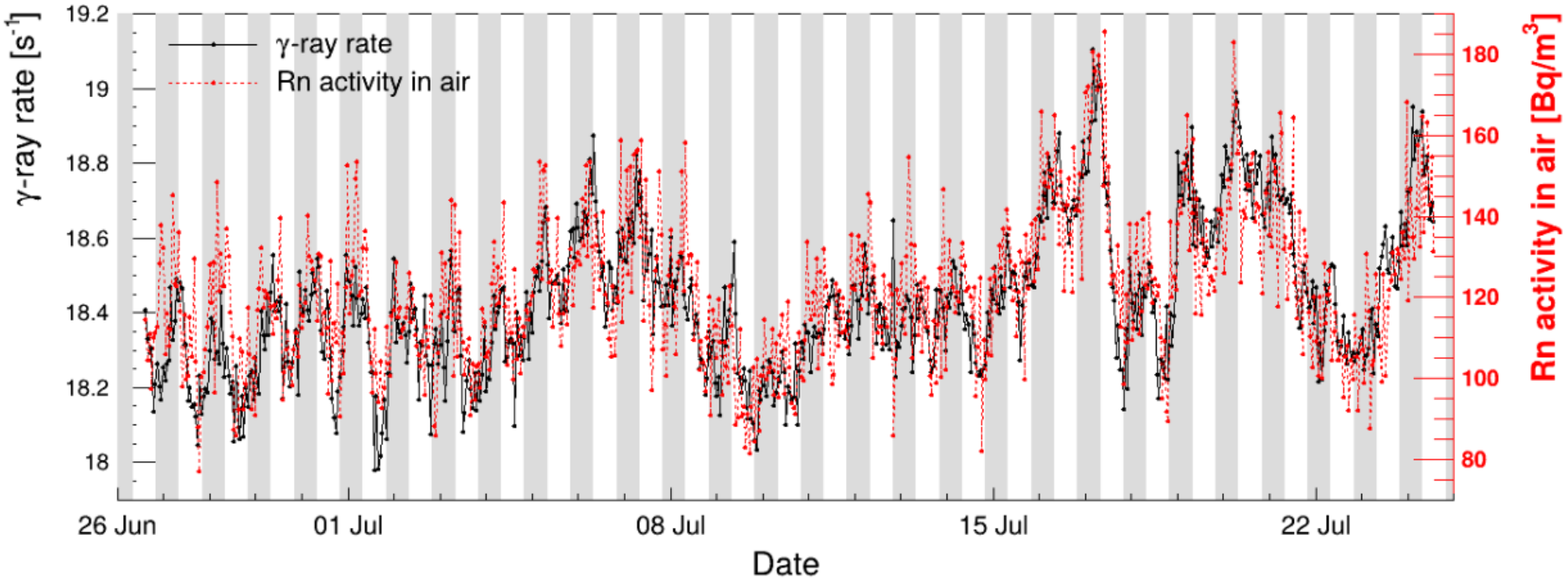
^{222}Rn problem

Why is Rn a problem?

Radon is a noble gas and can escape both solid and liquid materials.

- ^{220}Rn, ^{219}Rn, and especially ^{222}Rn (together with their daughters) emit several high-energy gammas
- Rn daughters plate out electrostatically onto surfaces. This effect is strongly enhanced on statically charged surfaces such as plastics or glass
- ^{222}Rn feeds the long-lived isotope ^{210}Pb ($T_{1/2}$=22 y)

Techniques based on charcoal adsorption have been used which suppress Radon (by about a factor of 10^4-10^5).



- Rn emanates constantly from the rock.
- **^{222}Rn concentration is significantly higher in an underground facility than on surface.**
- *It is mandatory to protect a the material surface against Rn (particularly UG)*

What we have to measure?
U, Th, K, Cs, Co…

How can we measure it?

How radiopure?

1 Bq	^{238}U /kg	$\cong$ 81×10^{-9} g_U/g	(81 ppb U)
1 Bq	^{232}Th /kg	$\cong$ 246×10^{-9} g_{Th}/g	(246 ppb Th)
1 Bq	^{40}K /kg	$\cong$ 32.3×10^{-6} g_K/g	(32.3 ppm K)

- One banana is a source of about ≈ 20 Bq of ^{40}K → ≈ 100 Bq/kg
- ^{40}K in humans → primary source of radiation from our body (practically constant)
  - K content of the body is 0.2 %
  - Natural abundance of 0.0117 % of K_{Nat}
  - K-40 specific activity ≈2.6e5 Bq g^{-1}

  For a 70-kg person, the amount of ^{40}K will be about 4.26 kBq (≈ 60 Bq/kg)
- ^{14}C for a 70-kg person would be about 3.08 kBq (≈44 Bq/kg)

For a low-background detector, a human body is a radioactive object: per kilogram, we are about one hundred thousand times hotter than the materials we are trying to select.

radiopurity.org

Search | Submit | Edit | Settings | Login

copper

Total results: 86

Grouping	Name	Isotope	Amount	Isotope	Amount	
ILIAS UKDM	Copper	Th-232	0.004 ppb	U-238	0.005 ppb	...
ILIAS UKDM	Copper	Th-232	0.5 ppb	U-238	0.5 ppb	...
ILIAS ROSEBUD	Copper, OFHC					...
ILIAS UKDM	Copper C101	Th-232	0.5 ppb	U-238	0.5 ppb	...
ILIAS UKDM	Rexalite, copper removed	Th-232	1 ppb	U-238	3 ppb	...
EDELWEISS (2011)	Copper, Apical, cables					...
ILIAS UKDM	Copper alloy swarf	Th-232	0.5 ppb	U-238	0.5 ppb	...
ILIAS Edelweiss	Copper, Cuc2 Plate, CARLIER					...
XENON100 (2011)	Copper, Norddeutsche Affiinerie			U-238	11 mBq/kg	...
XENON100 (2011)	Copper, Norddeutsche Affinerie			U-238	70 uBq/kg	...
EDELWEISS (2011)	Copper, screens, support					...
EDELWEISS (2011)	Copper, CuC2, detector casings			U-238	1.4 mBq/kg	...
ILIAS Edelweiss	Adhesive tape, Copper					...

Common assay techniques

- Gamma counting with HPGe crystals
- Mass spectroscopy
  - ICP-MS
  - GD-MS
  - LA-ICP-MS
- Neutron Activation Analysis
- Emission spectroscopy
  - XRF
  - ICP-AES
- Radio-chemical methods (^{210}Po extraction)

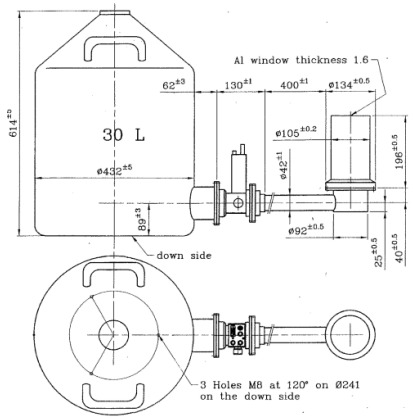
Gamma spectroscopy with HPGe

- Gamma rays produced by the great majority of radionuclides
- The typical range for γ-rays emitted by nuclei is 50 keV - 10 MeV (endcap - size of the crystals)
- Gamma spectrometry is a **non-destructive technique**
- Gamma spectrometry using HPGe semi-conductor detectors is one of the best techniques to identify and quantify radionuclides: **very good energy resolution**

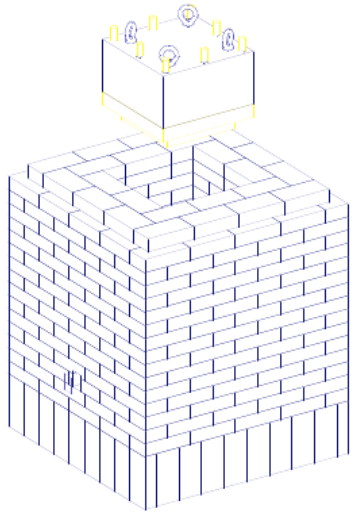
- Operating Ge detectors not possible at room temperature (leakage current due to the small bandgap energy)
- The germanium detector preamp is normally included as part of the cryostat package.
- All of the cryostat materials around the detector should be as low Z as possible to reduce photon scatter. Hence, aluminum, magnesium, beryllium, Teflon, and Mylar are used whenever possible.

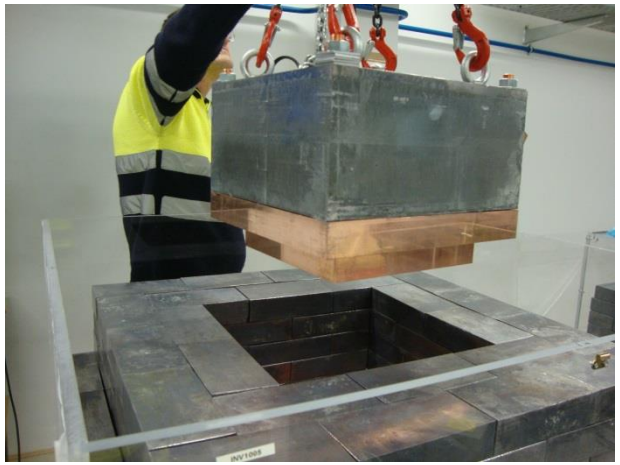
Courtesy of LSC

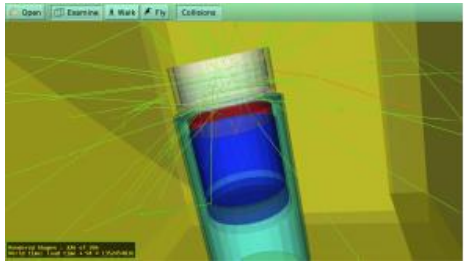
HPGe shielding

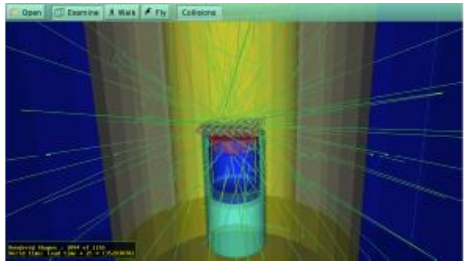


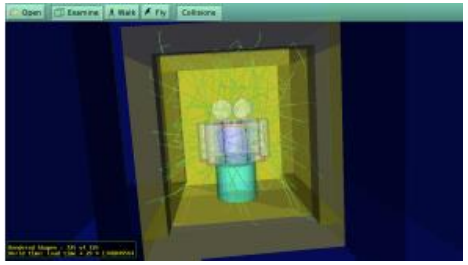
- 10 cm Cu-OF
- 20 cm Pb (~660 bricks or “donuts”)
- Methacrylate Rn box.
- Door



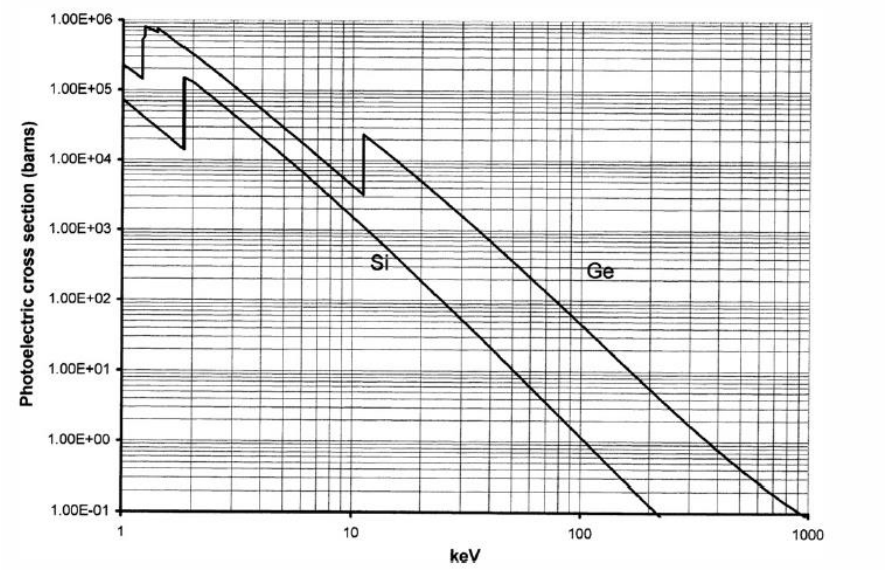




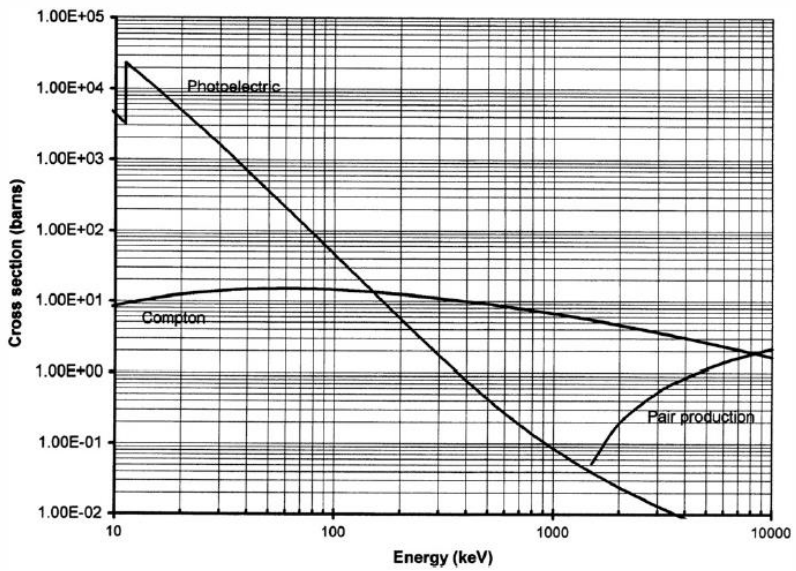




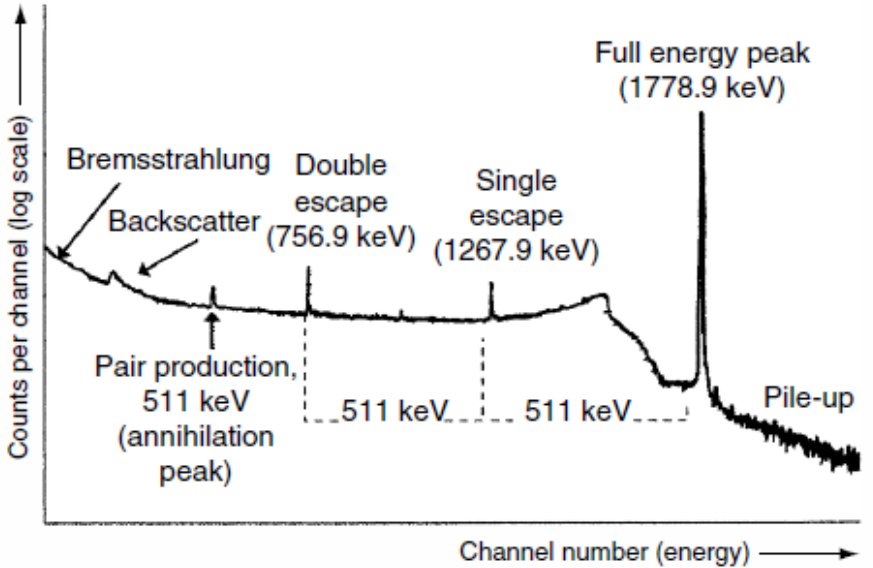
Gamma spectroscopy with HPGe



Photoelectric cross section (barns) of Si (lower curve) and Ge (higher curve) as a function of energy (keV).



Compton, photoelectric, and pair production cross section of Ge for high-energy g-rays.



Courtesy of LSC

Ultra-Low Background Measurements Service- Hall C

7 HPGe p-type coaxial (2 kg)-LSC
1 SAGe Well model GSW275L-LSC
1SAGe Well model GSW400-Poland

7 mounted and taking data: Asterix, GeLatuca, GeOroel, GeTobazo, GeAnayet, GeAspe, GeRysy.

Obelix and GeLaraca (SAGe)-no shielding

HPGe: Figure of Merit

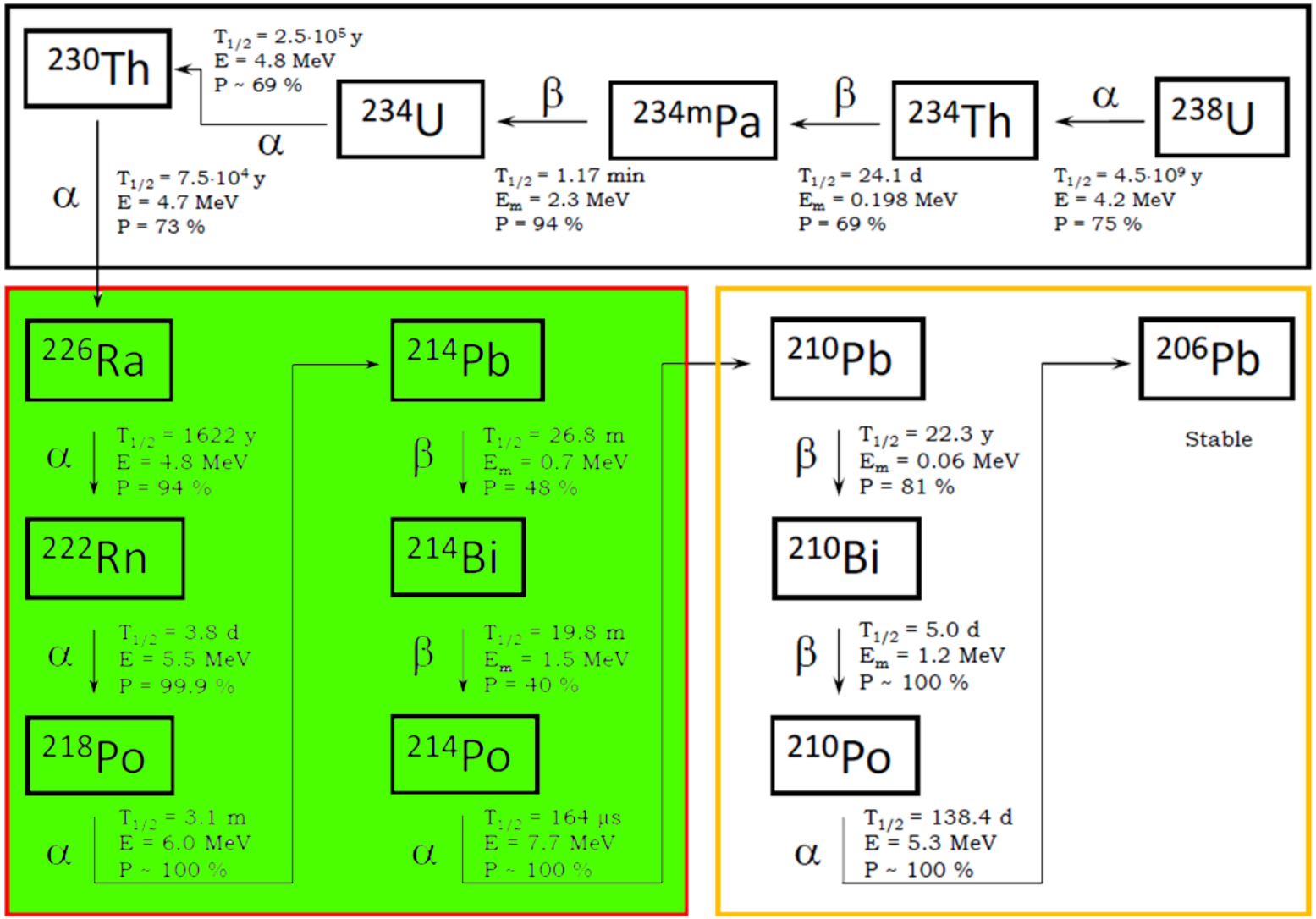
Advantages:

- The best technique to assay the γ-emitters: K-40, middle primordial decay chains, cosmogenics … etc
- Relatively simple sample preparation
- Non-destructive technique
- Very good sensitivity achievable (<< mBq/kg)

Limitations:

- Actual sensitivity strongly depends on the mass of the sample (not suitable for small samples)
- Stringent detector stability and background requirements
- Only sensitive to γ-emitters
- Several weeks are typically necessary to assay clean samples
- Detailed Monte Carlo simulation to calculate the efficiency as a function

Middle Chain HPGe



MASS SPECTROMETRY (MS)

- Inductively coupled plasma mass spectrometry (ICP-MS)
- Glow discharge mass spectrometry (GD-MS)
- Laser ablation mass spectrometry (LA-ICP-MS)
- Secondary ion mass spectrometry (SIMS),
- Resonance ionization mass spectrometry (RIMS),
- Accelerator mass spectrometry (AMS)

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INDUCTIVELY COUPLED PLASMA MASS SPECTROMETRY (ICP-MS)

It allows the determination of **elements** with atomic mass ranges 7 to 250 (Li to U)

Concentration at very low level in a wide variety of samples

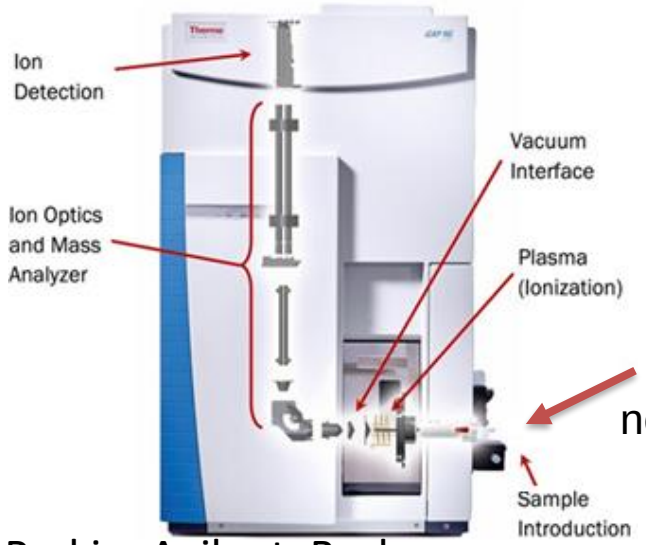
High resolution ICP-MS (HR-ICP-MS), uses a magnetic field and electrostatic analyzer, providing very low detection limits, in the $pg \cdot L^{-1}$ range

Destructive analysis: a **previous sample digestion** is required in order to obtain a liquid solution:

- Acid digestion uses a combination of different acids in order to obtain a total simple digestion (strong mineral acids: Nitric Acid, Hydrochloric Acid, Hydrofluoric Acid, Perchloryc Acid…)
- Dry ashing: Especially suitable for samples with a high organic matter content (organic matter can be completely eliminated)
- Need to avoid contamination during sample processing (acids, tools, crucible…)

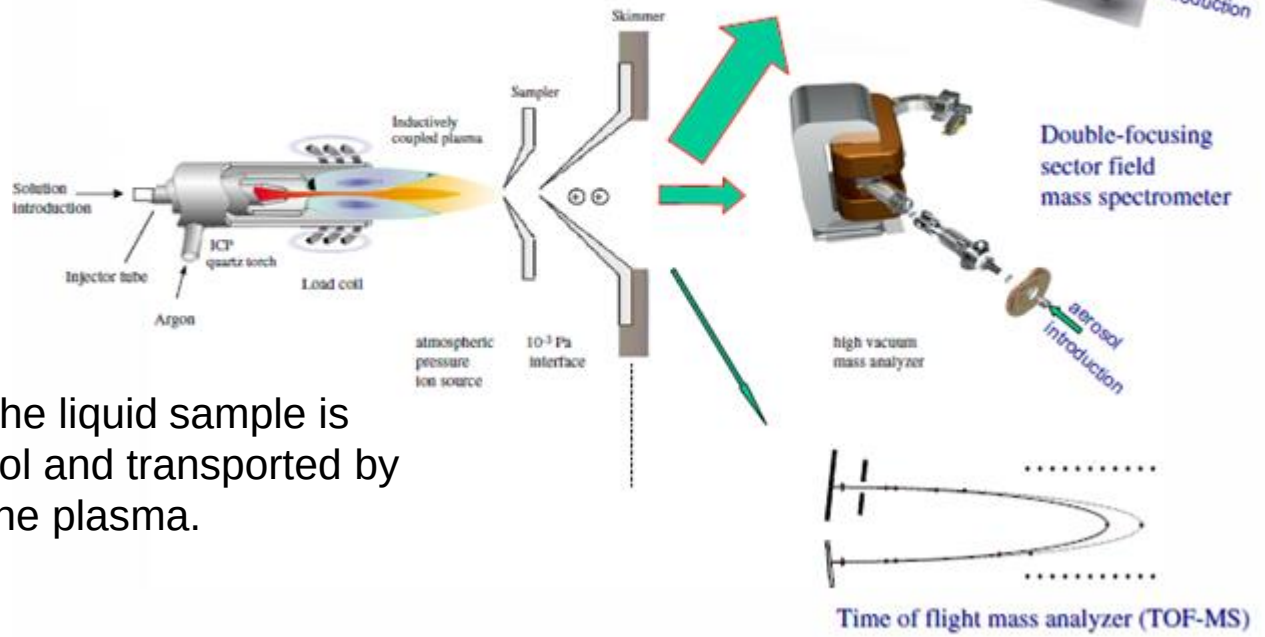
Major advantage of ICP-MS: In comparison to other alternative solid-state mass spectrometric techniques is advantageous because liquid solutions can be easily and **rapidly analyzed**

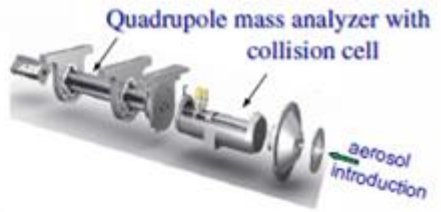
INDUCTIVELY COUPLED PLASMA MASS SPECTROMETRY (ICP-MS)



Perkin, Agilent, Bruker….

Sample introduction: the liquid sample is nebulized into a fine aerosol and transported by argon gas into the plasma.





ICP source: the inductively coupled argon plasma atomizes and ionizes the elements in the sample, producing mostly positive ions.

Interface region: sampler and skimmer cones extract ions from the atmospheric-pressure plasma into the low-pressure region.

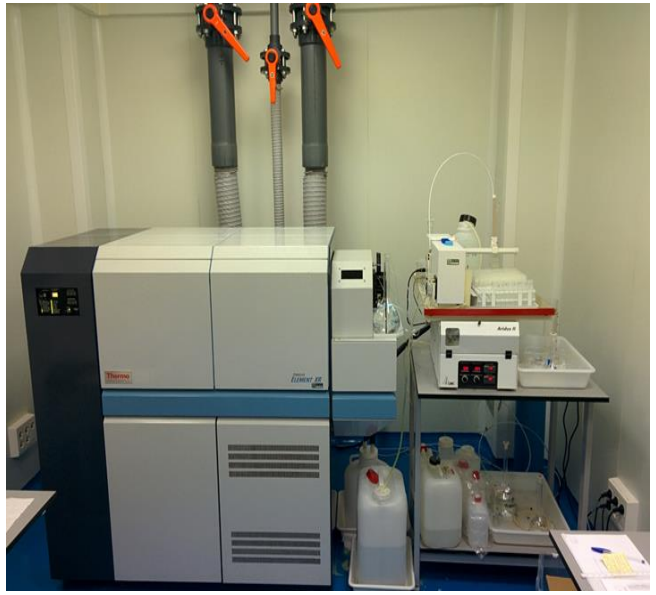
Ion optics: electrostatic lenses focus and guide the ion beam, while removing photons, neutrals and unwanted particles.

Mass analyzer: ions are separated according to their mass-to-charge ratio, using quadrupole, sector-field or time-of-flight analyzers.

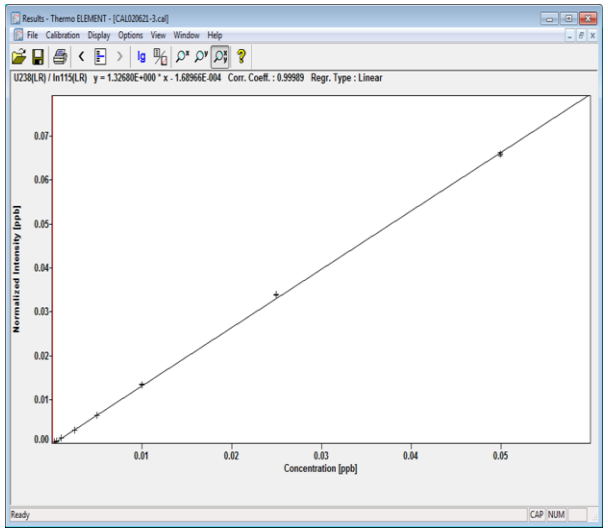
Detector: the selected ions are counted, giving elemental concentrations down to sub ppt levels for U, Th and other trace contaminants.

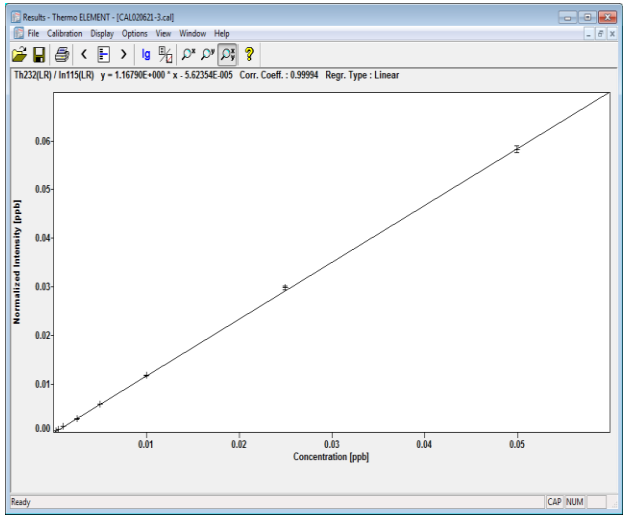
ICP-MS

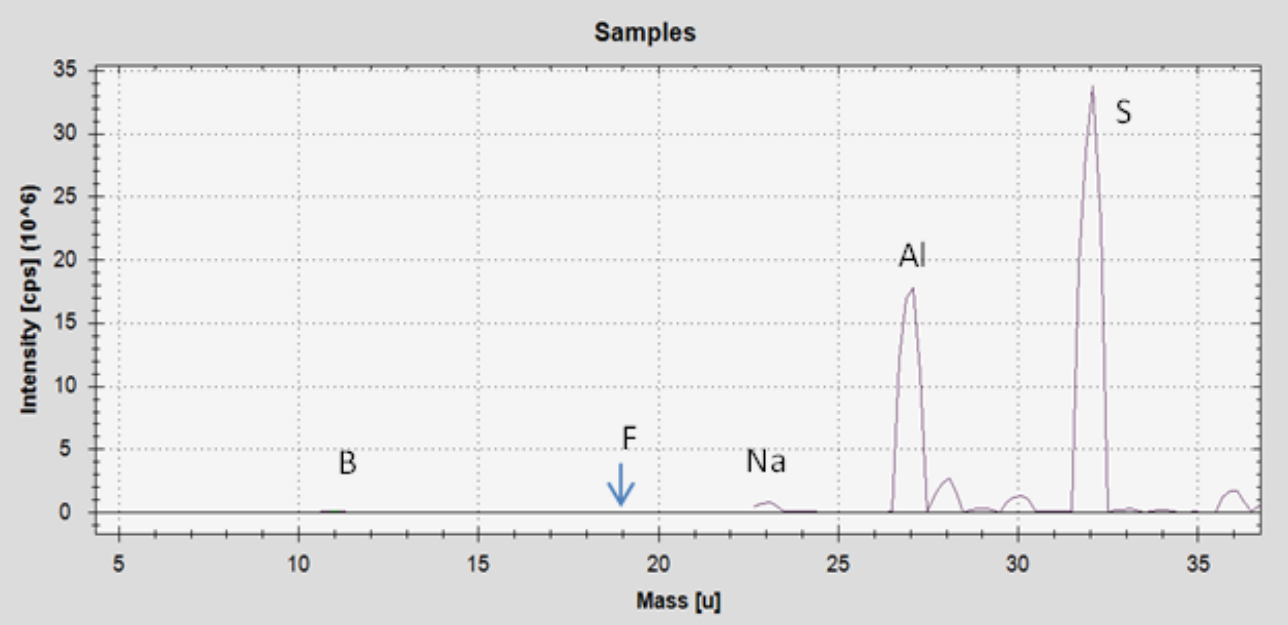
HR-ICP-MS Thermo Element



^{238}U AND ^{232}Th CALIBRATION

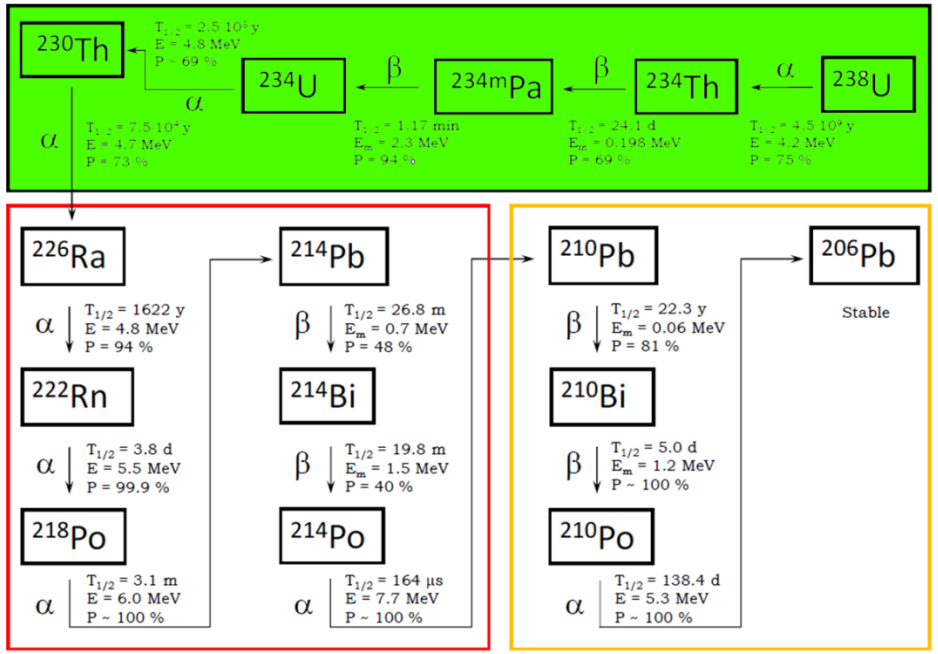


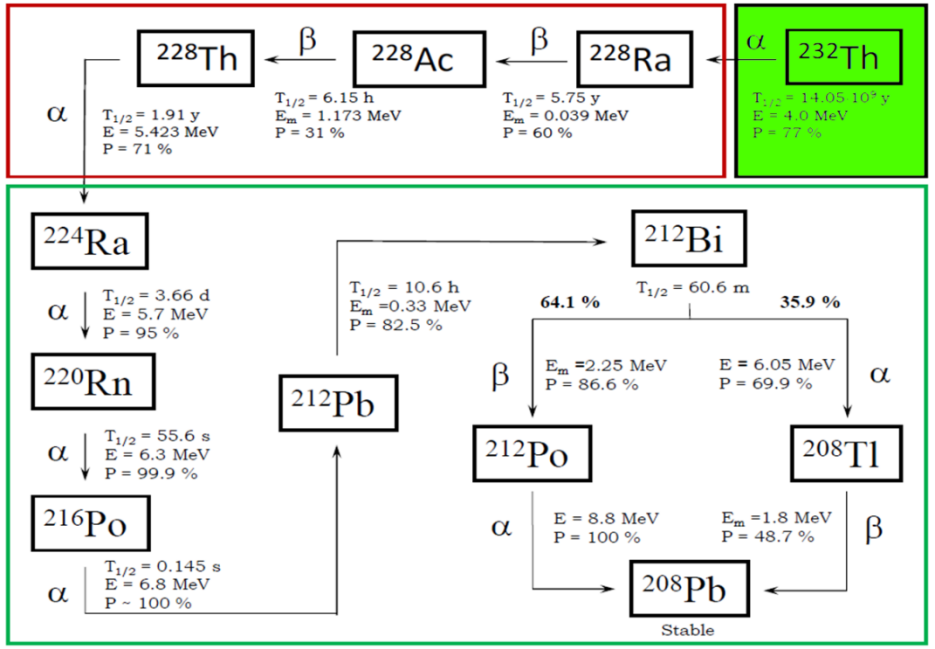




	Th (ng/g)	U (ng/g)
LOQ	0.052	0.580

Spectroscopic interferences (molecular ion formation) can be decreased or overcome by the use of a collision cell.





---------- Summary ----------

Your input : 0.000001 mg 238Uc

ID	NUCL	Nb. Atoms	Ac[Bq]	Mass[g]
16	238U	2.529772e+12	1.243652e-05	1.000000e-09
17	234Th	3.735978e+01	1.243652e-05	1.451975e-20
18	234Pa	1.264919e-03	1.243652e-05	4.915961e-25
19	234U	1.387185e+08	1.243652e-05	5.391072e-14
20	230Th	4.267999e+07	1.243652e-05	1.630286e-14
21	226Ra	9.059165e+05	1.243652e-05	3.400122e-16
22	222Rn	5.927183e+00	1.243652e-05	2.185167e-21
23	218Po	3.283404e-03	1.243652e-05	1.188633e-24
24	218At	6.459207e-09	2.238574e-09	2.338310e-30
25	214Pb	2.884570e-02	1.243428e-05	1.025046e-23
26	214Bi	2.142289e-02	1.243652e-05	7.612691e-24
27	210Tl	2.938889e-07	2.611670e-09	1.024781e-28
28	214Po	2.947269e-09	1.243391e-05	1.047304e-30
29	210Pb	1.262621e+04	1.243652e-05	4.402594e-18
30	210Bi	7.771144e+00	1.243652e-05	2.709695e-21
31	210Po	2.145474e+02	1.243652e-05	7.480941e-20
	Total		1.741113e-04	1.000071e-09
			1.119283e-04 (a)	
			6.218299e-05 (b)	

---------- Summary ----------

Your input : 0.000001 mg 232Thc

ID	NUCL	Nb. Atoms	Ac[Bq]	Mass[g]
5	232Th	2.595325e+12	4.057385e-06	1.000000e-09
6	228Ra	1.062144e+03	4.057385e-06	4.021854e-19
7	228Ac	1.296841e-01	4.057385e-06	4.910543e-23
8	228Th	3.531120e+02	4.057385e-06	1.337061e-19
9	224Ra	1.852267e+00	4.057385e-06	6.890321e-22
10	220Rn	3.256625e-04	4.057385e-06	1.189765e-25
11	216Po	8.493271e-07	4.057385e-06	3.046363e-28
12	212Pb	2.243639e-01	4.057385e-06	7.898076e-23
13	212Bi	2.128074e-02	4.057385e-06	7.491241e-24
14	208Tl	3.856248e-04	1.458224e-06	1.331802e-25
15	212Po	1.121930e-12	2.599161e-06	3.949369e-34
	Total		4.057385e-05	1.000000e-09
			3.100085e-05 (a)	
			9.572994e-06 (b)	

Concentration of the daughters too low for MS

SS sample

Sample preparation

The samples were etched in three phases with 6 ml of ultra pure Hydrochloric acid and 2 ml ultra pure nitric acid. Then the samples were diluted 20 times before ICP-MS measurements

The first etching has been considered as cleaning but however has been measured to look for superficial contamination informations. For samples N 4 and 5 was difficult to control the reaction that was quite violent.

Sample.Ecth	Initial weight [g]	Final weight [g]	Difference [g]	Volume [ml]	Dilution
1.1	13.81	12.28	1.53	15	9.8
2.1	13.84	12.28	1.56	10.5	6.7
3.1	4	2.5	1.5	12.5	8.3
4.1	10.6	9.28	1.32	12.5	9.5
5.1	9.95	8.36	1.59	17	10.7
1.2	12.28	10.81	1.47	17.5	11.9
2.2	12.28	10.78	1.5	15	10.0
3.2	2.5	1.53	0.97	11	11.3
4.2	9.28	8.73	0.55	14	25.5
5.2	8.36	7.61	0.75	15	20.0
1.3	10.81	9	1.81	18	9.9
2.3	10.78	9.11	1.67	19	11.4
3.3	1.53	0	1.53	16	10.5
4.3	8.73	5.93	2.8	12.5	4.5
5.3	7.61	5	2.61	20	7.7

		Reagents
Sample 1	SS-8-VCR-4	2.0 ml HNO3+6.0 ml HCl
Sample 2	SS-8-VCR-3	2.0 ml HNO3+6.0 ml HCl
Sample 3	Flex pipe small	2.0 ml HNO3+6.0 ml HCl
Sample 4	Flex pipe medium	2.0 ml HNO3+6.0 ml HCl
Sample 5	Flex pipe big	2.0 ml HNO3+6.0 ml HCl

Analysis results

The concentrations were evaluated in semiquantitative analysis, based on the response of a single spike solution containing 100 ppt of Th and U added to aliquot of Step 3 . The uncertainty is about 30% of the given values. The contribute of procedure and reagents has been subtracted to the samples.

	Pb	Th	U
	[ng/g]	[ng/g]	[ng/g]
1.1	280	1.4	2.8
2.1	480	1.9	7.0
3.1	455	1.5	6.1
4.1	620	2.5	9.1
5.1	510	2.8	9.6

Table: 1st etching cleaning step measurements

ICP-MS: Figure of Merit

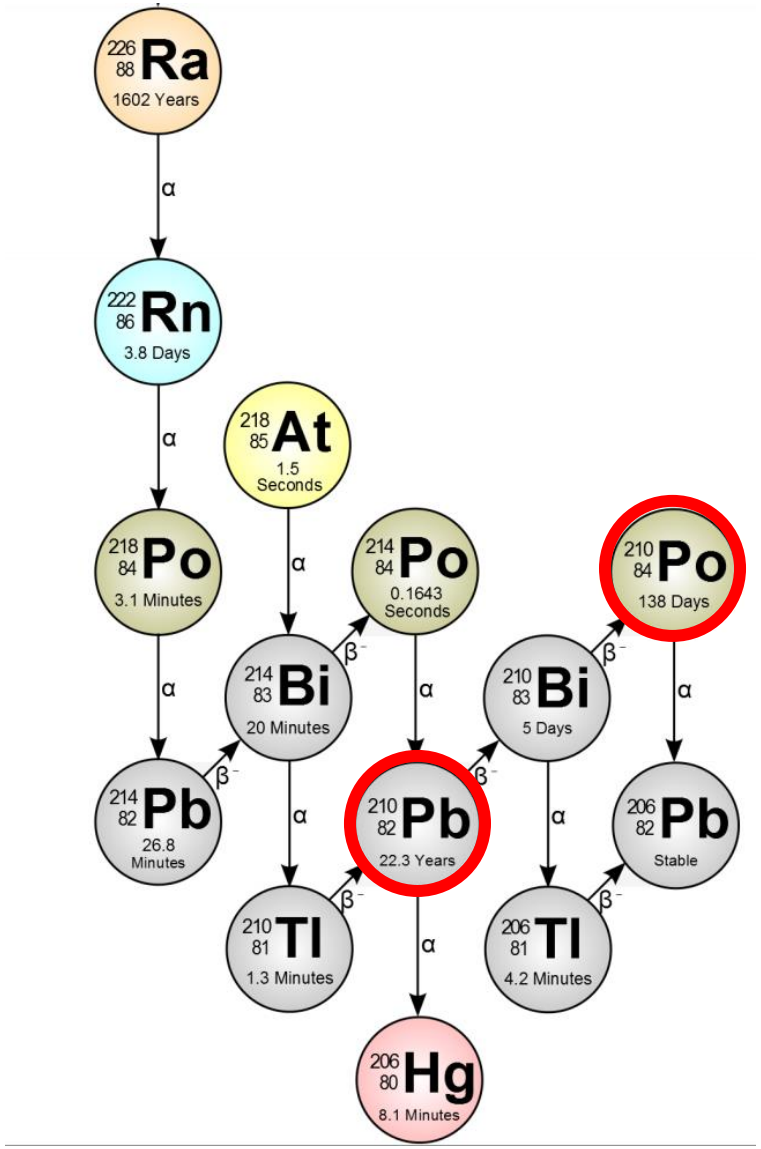
Advantages:

- **Fast analytical technique for simultaneous determination of trace and ultra-trace elements in liquid solution (≈ 1 d)**
- **Excellent sensitivity, accuracy and very low det. limits: ~ 0.1 pg/mL**
- **Little amount of materials needed (small sample)**
- **Isotope ratio measurements**

Limitations:

- Destructive analysis
- Sample preparation for solid samples (full digestion, recuperation, contamination..)
- It only measures the upper chain (It cannot measure K-40)
- Molecular ions interferences with atomic ions (e.g. $^{238}UH^{+}$ and $^{239}Pu^{+}$)

Radio-chemical methods (^{210}Po extraction)



Background component:

- Surface contamination
- Bulk contamination
- (α,n) reaction

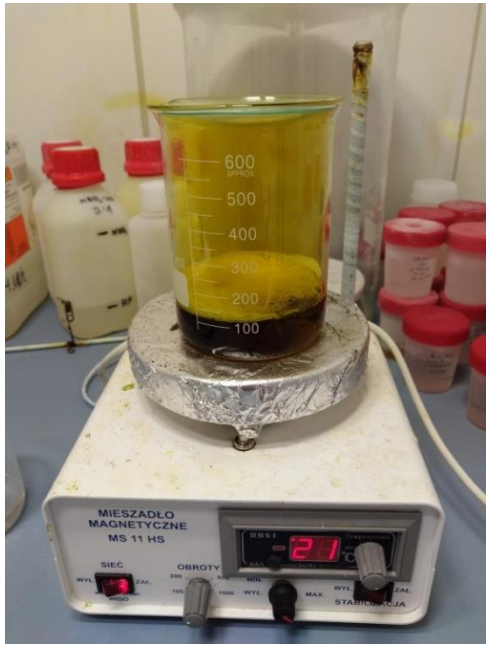
Radioanalytical/Radiochemical problems:

- measurement can be done only by alpha spec
- High volatility of Po – only wet ashing in boiling concentrated acids and acid mixtures (problems with some polymers materials)

Chemical separation of ^{210}Po

Separation method of ^{210}Po:

- Sample etch and dissolution
- Sample pre-concentration and purification (if needed) by co-precipitaion or ion-exchange chromatography (e.g. measurement of bulk ^{210}Po in Cu samples)
- Source preparation by spontaneous deposition



Radiochemical tracers:
^{208}Po (5.115 MeV alphas)
^{209}Po (4.883 MeV alphas)

Reagents:

- HNO_3
- HCl
- H_2O_2 30%
- $N_2H_4 \bullet 2HCl$
- Silver discs

Laboratory equipment:

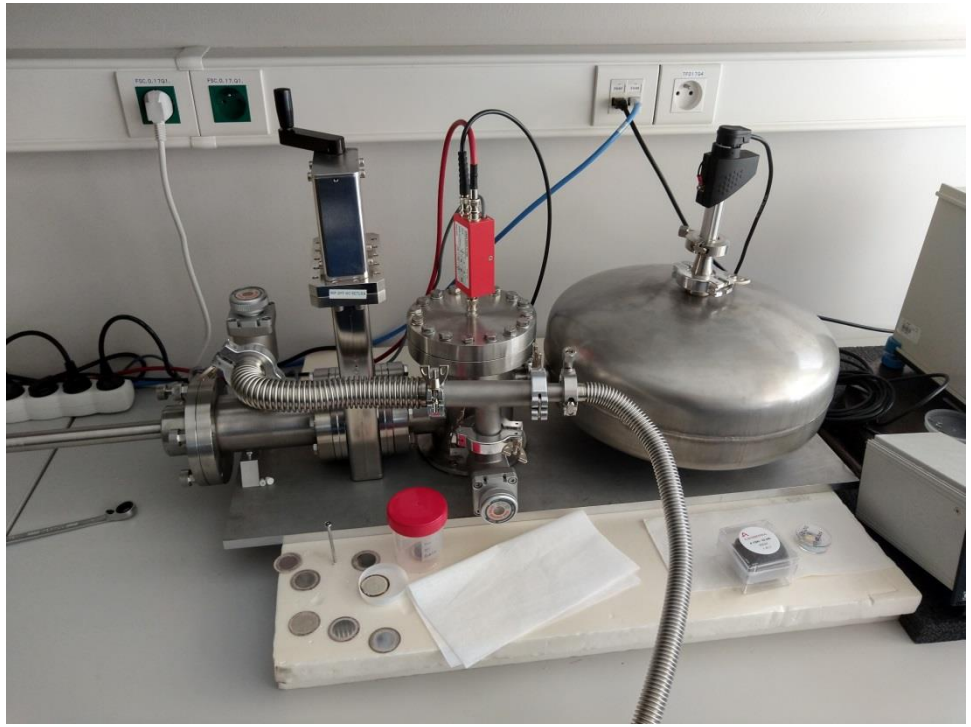
- Glass beakers (250 ml)
- Watch glasses
- Stirring rods with PTFE coating
- Hotplate with stirrer
- Pipette 10-100 µl+ tips
- Protective equipment (glasses, gloves etc.)

Courtesy of T. Mróz
Jagiellonian University in Cracow, Poland

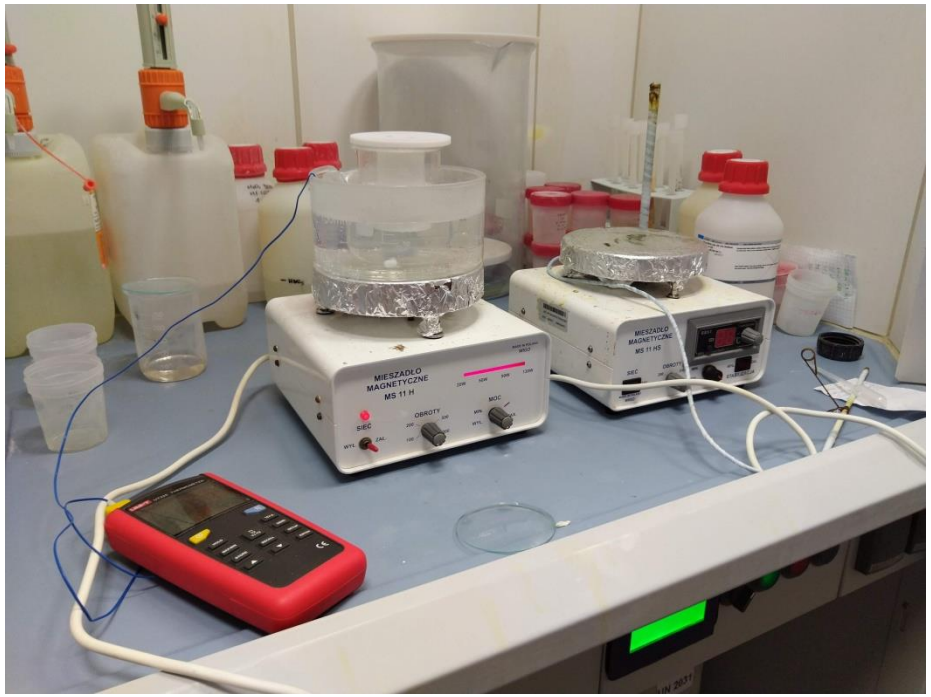
Detection of ^{210}Po – semiconductor alpha spectrometer

- Very good energy resolution (20-30 keV FWHM)
- Detection limits for activity lower than for XiA

- Requires radiochemical separation of ^{210}Po
- Efficiency determined with tracer
- Spectrum resolution depends on the source quality
- Blank runs required to control the background for each procedure



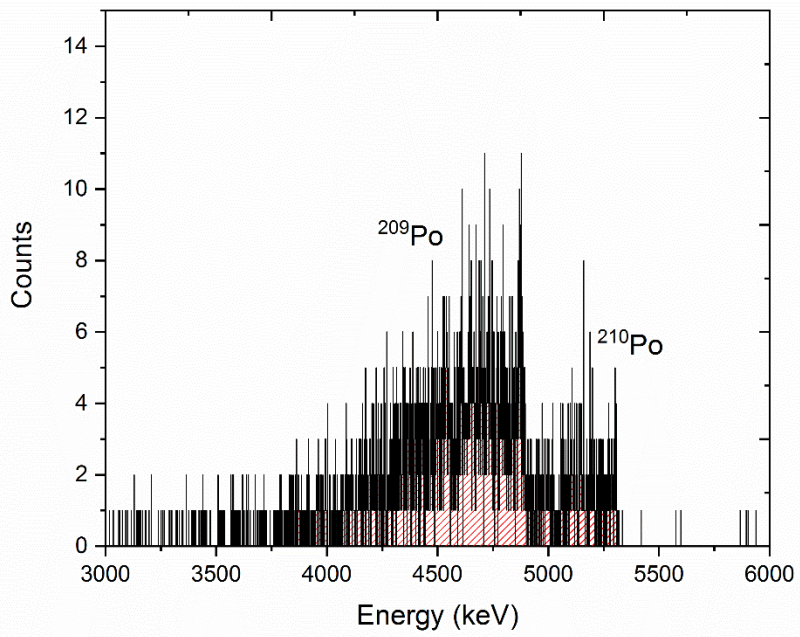
Source preparation



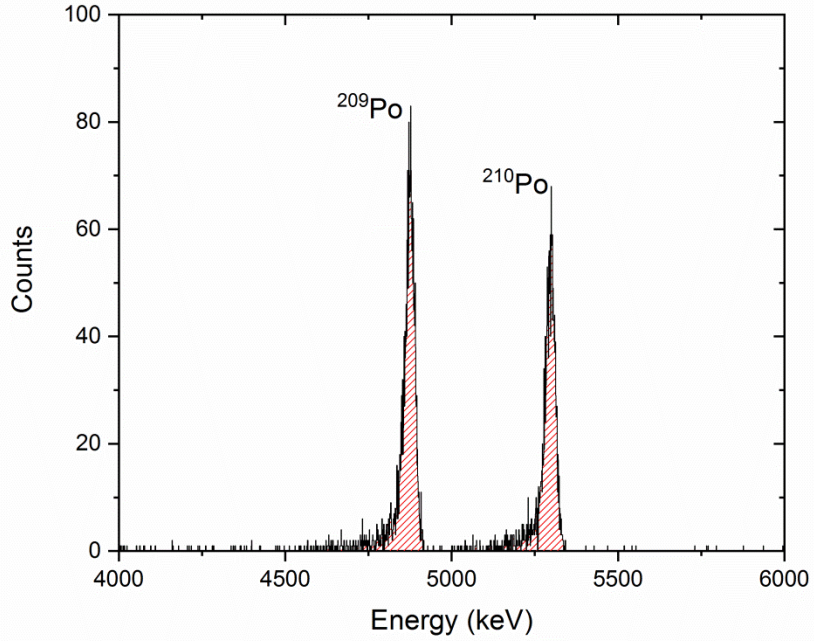
Spontaneous deposition of Po on Ag from diluted HCl solution

Alpha spectrometry sources

Thick layer deposited on source



Thin source (~20 keV FWHM)



Radio-chemical methods : Figure of Merit

Advantages:

- **Is the only technique able to measure the lower chain**

Limitations:

- Sample preparation

OBJECTIVES of this lecture

After this class, you should be able to:

- Know the most important sources of **natural radioactivity.**
- Understand the principles of secular **equilibrium and disequilibrium** of the decay chains.
- Grasp the principles of the **cosmogenic** production of radionuclides.
- Understand the general framework of the problems posed by the **Rn-222.**
- Figure out the **importance of material assays and radiopurity** related aspects for low-background techniques.
- Know the principle of the **most used techniques** for the assays.

Thank you for your kind attention!

Backup