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Gamma-ray Spectrometry

Radioactive decay modes and half-life

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Decay modes

1) Decays with emission of nucleons (α, p, fission, ..)
2) Different modes of beta decay (β^-, β+, $\beta\beta\nu\nu$, $\beta\beta 0\nu$,..)
3) Transitions between states of the same nucleus

Mode of decay	Participating particles	Daughter nucleus
Decays with emission of nucleons:		
Alpha decay	An alpha particle ($A = 4$, $Z = 2$) emitted from nucleus	$(A - 4, Z - 2)$
Proton emission	A proton ejected from nucleus	$(A - 1, Z - 1)$
Neutron emission	A neutron ejected from nucleus	$(A - 1, Z)$
Double proton emission	Two protons ejected from nucleus simultaneously	$(A - 2, Z - 2)$
Spontaneous fission	Nucleus disintegrates into two or more smaller nuclei and other particles	—
Cluster decay	Nucleus emits a specific type of smaller nucleus (A_1, Z_1) smaller than, or larger than, an alpha particle	$(A - A_1, Z - Z_1) + (A_1, Z_1)$

From: Wikipedia

Different modes of beta decay:		
β^- decay	A nucleus emits an electron and an electron antineutrino	$(A, Z+1)$
Positron emission (β^+ decay)	A nucleus emits a positron and an electron neutrino	$(A, Z-1)$
Electron capture	A nucleus captures an orbiting electron and emits a neutrino; the daughter nucleus is left in an excited unstable state	$(A, Z-1)$
Bound state beta decay	A nucleus beta decays to electron and antineutrino, but the electron is not emitted, as it is captured into an empty K-shell; the daughter nucleus is left in an excited and unstable state. This process is suppressed except in ionized atoms that have K-shell vacancies.	$(A, Z+1)$
Double beta decay	A nucleus emits two electrons and two antineutrinos	$(A, Z+2)$
Double electron capture	A nucleus absorbs two orbital electrons and emits two neutrinos – the daughter nucleus is left in an excited and unstable state	$(A, Z-2)$
Electron capture with positron emission	A nucleus absorbs one orbital electron, emits one positron and two neutrinos	$(A, Z-2)$
Double positron emission	A nucleus emits two positrons and two neutrinos	$(A, Z-2)$

From: Wikipedia

Transitions between states of the same nucleus:		
Isomeric transition	Excited nucleus releases a high-energy photon (gamma ray)	(A, Z)
Internal conversion	Excited nucleus transfers energy to an orbital electron and it is ejected from the atom	(A, Z)

Main message: Many strange decay modes exist.

Also many strange productions modes exists, when e.g. high energy cosmic rays interact with matter spallation reactions occur.

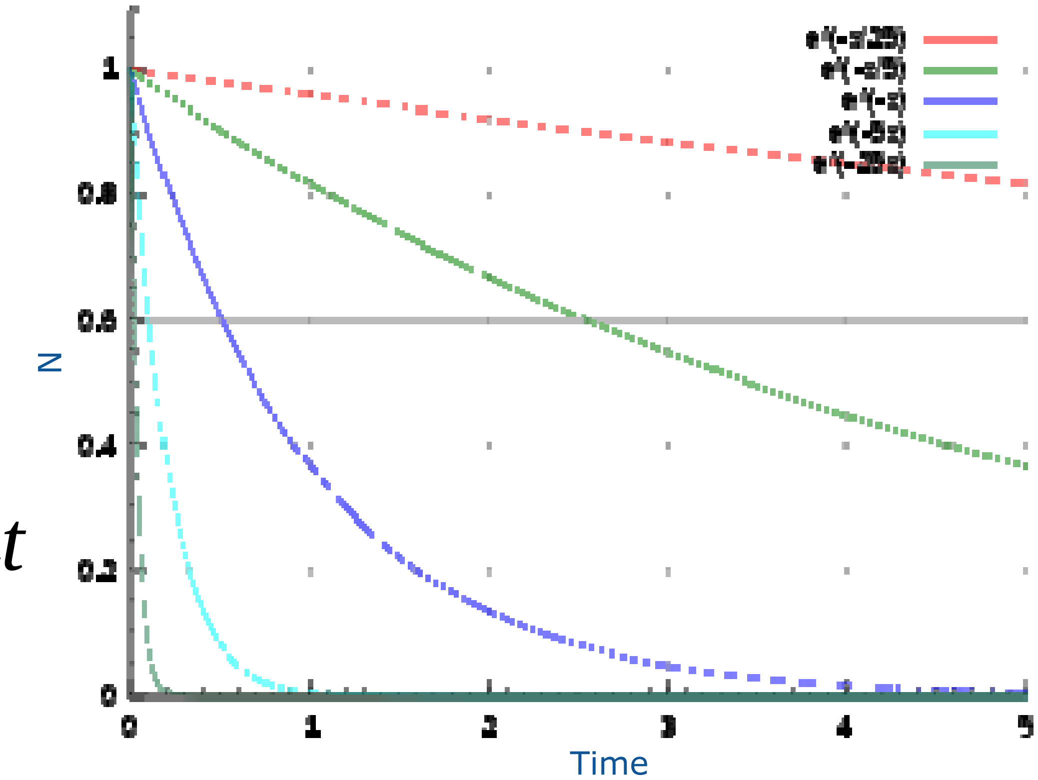
From: Wikipedia

Decay constant, λ

Definition of Exponential decay
When the decrease is proportional to its value

$$\frac{dN}{dt} = -\lambda N$$

$$N(t) = N_0 e^{-\lambda t}$$



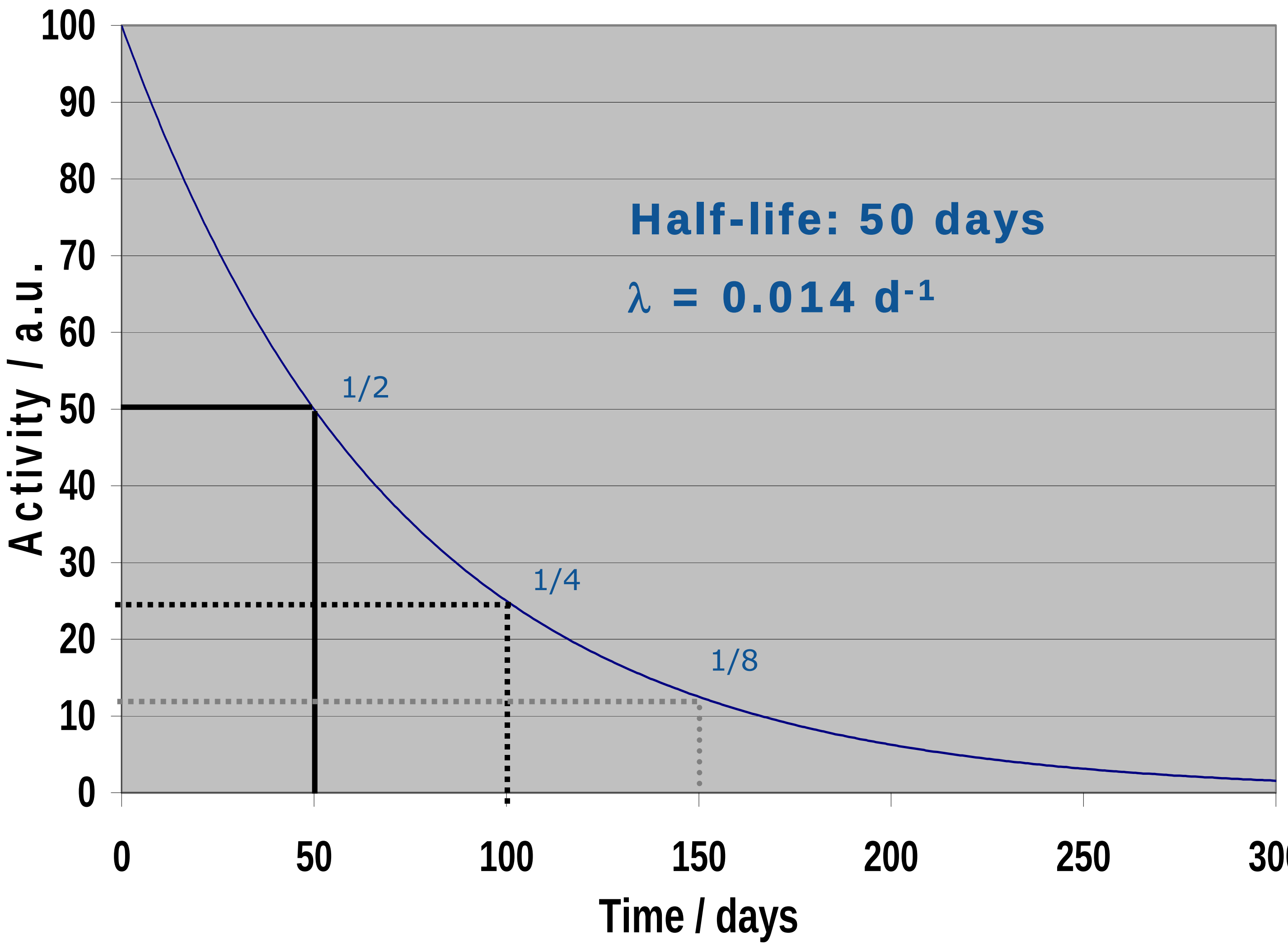
Half-life, $T_{½}$

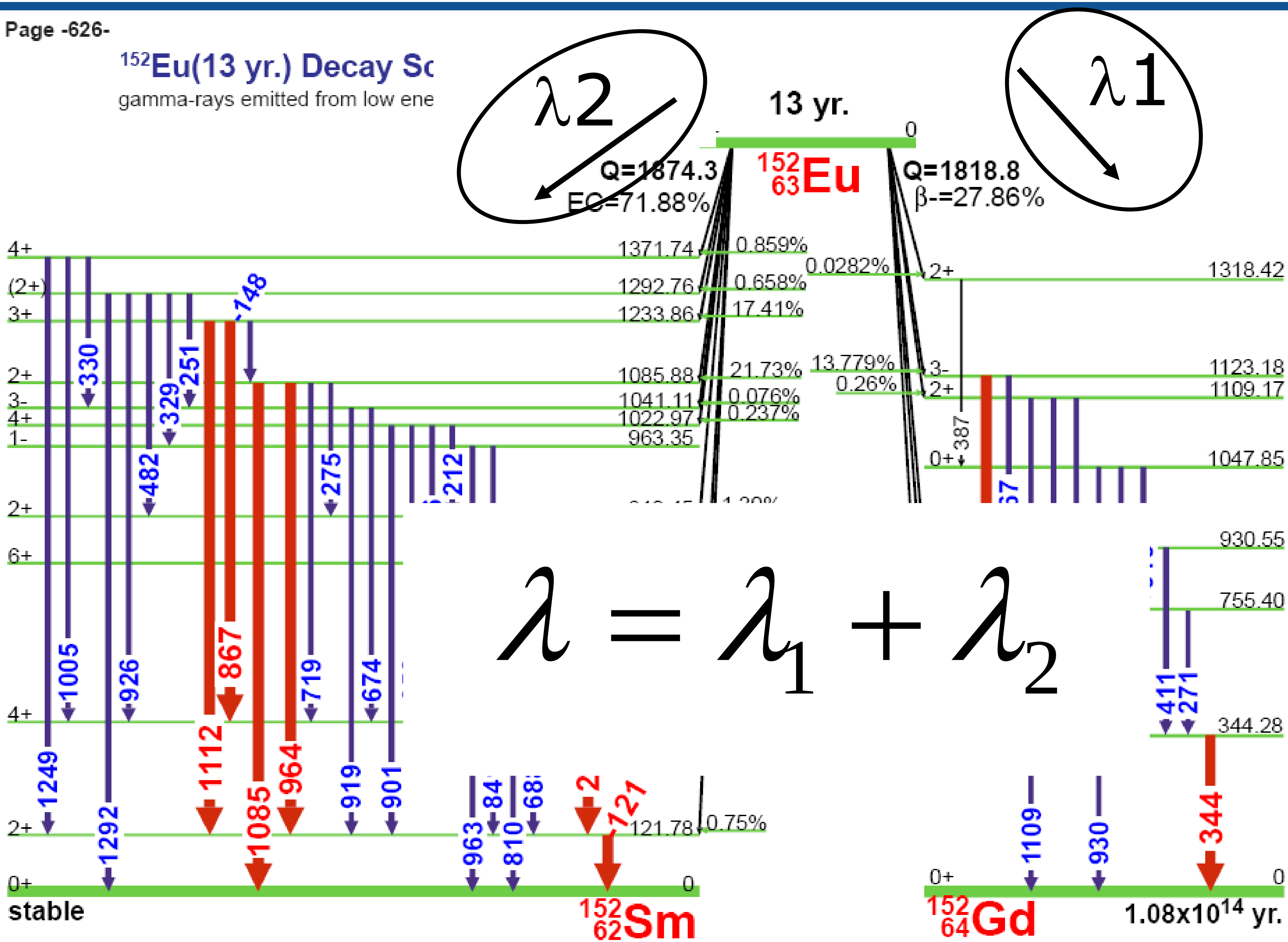
$$T_{1/2} = \frac{\ln(2)}{\lambda}$$

Decay constant

$$\tau = \frac{1}{\lambda}$$

Mean lifetime





$$\lambda = \lambda_1 + \lambda_2$$

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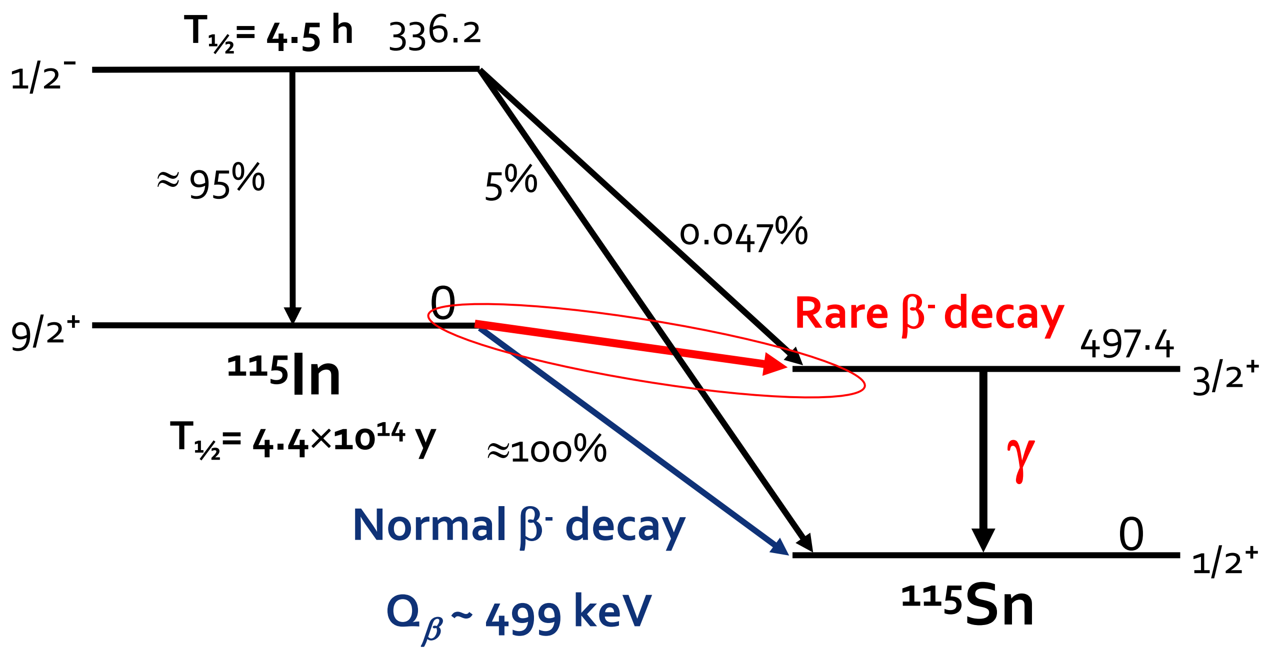
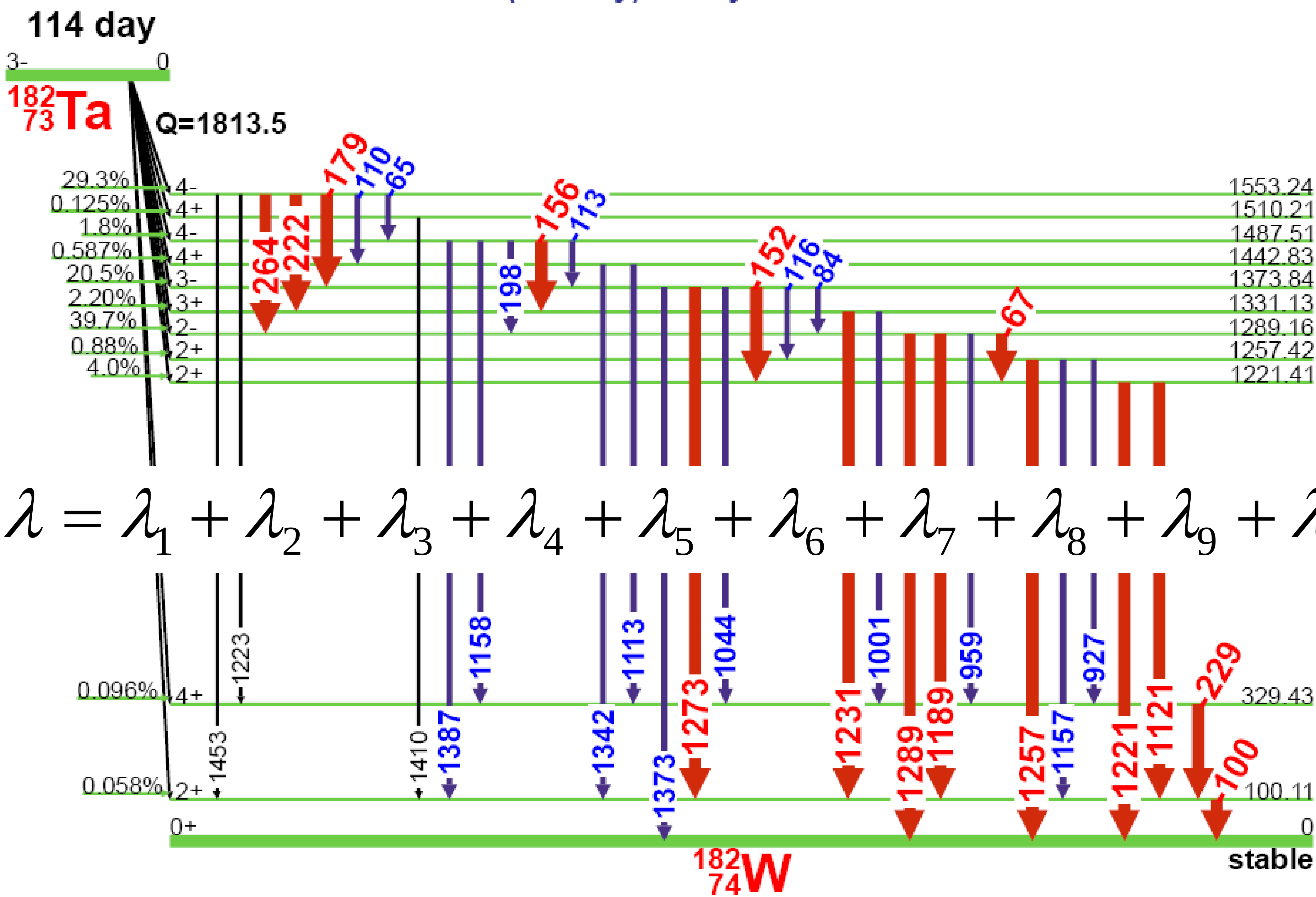
^{115}In rare β$^-$ decay half life

The lowest β$^-$ decay energy Q_β known to man

155(24) eV

(^{187}Re 2.47 keV), (^{3}H 18.6 keV)





$$\lambda = \lambda_1 + \lambda_2 + \lambda_3 + \lambda_4 + \lambda_5 + \lambda_6 + \lambda_7 + \lambda_8 + \lambda_9 + \lambda_{10}$$

How to measure long half-lives?

$$A = \frac{dN}{dt} \Rightarrow \lambda = \frac{A}{N}$$

Determine the number of atoms using

1) Mass spectrometry

   or

2) Knowledge about isotopic composition (perhaps natural)

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Serial decay

$$A \rightarrow B \rightarrow C(stable)$$

A: Like before

B: $$\frac{dN_B}{dt} = -\lambda_B N_B + \lambda_A N_{A0} e^{-\lambda_A t}$$

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Chain of "D" decays

$$A_1 \rightarrow A_2 \rightarrow A_3 \rightarrow A_4 \rightarrow ....A_{D-1} \rightarrow D(stable)$$

Recursive problem: $$\frac{dN_j}{dt} = -\lambda_j N_j + \lambda_{j-1} N_{(j-1)0} e^{-\lambda_{j-1} t}$$

General solution: "<u>Bateman's equations</u>"

$$\frac{dN_D}{dt} = N \sum_{i=1}^{D} C_i e^{-\lambda_i t} \qquad \frac{\prod^{D} \lambda}{\prod_{j=1,i\neq j} (\lambda_j - \lambda_i)}$$

Important to master for radiochemists!!!

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Serial decay

$$N_B = \frac{N_{A0}\lambda_A}{\lambda_B - \lambda_A}\left(e^{-\lambda_A t} - e^{-\lambda_B t}\right)$$

$$A_B = \frac{A_{A0}\lambda_B}{\lambda_B - \lambda_A}\left(e^{-\lambda_A t} - e^{-\lambda_B t}\right)$$

More complex if N_{0B} or A_{0B} is not negligible,
but in principle add the term $N_{0B}e^{-\lambda_B t}$ or $A_{0B}e^{-\lambda_B t}$

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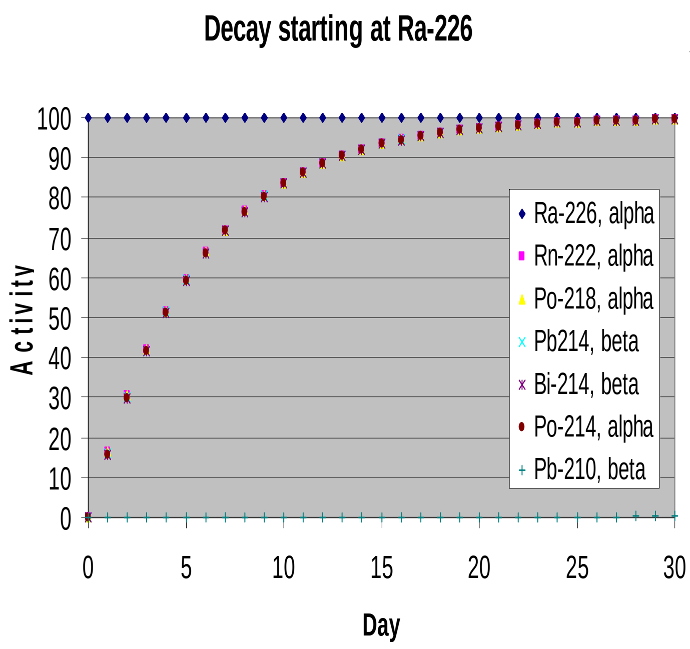
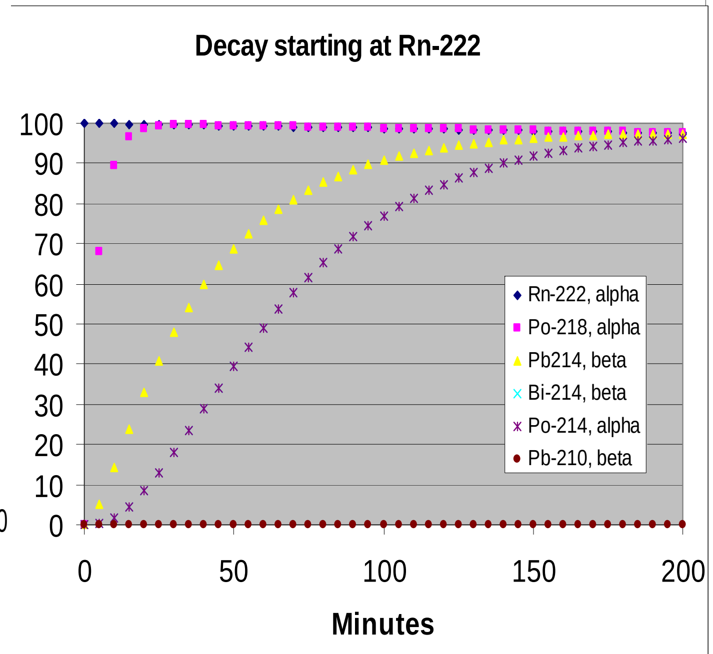
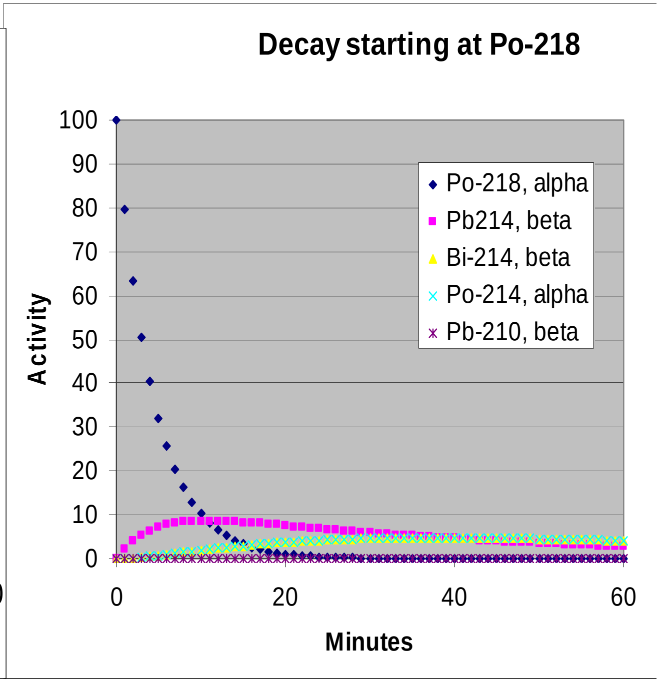
Serial decay

$$A_B = \frac{A_{A0}\lambda_B}{\lambda_B - \lambda_A}\left(e^{-\lambda_A t} - e^{-\lambda_B t}\right)$$

Can look quite differently







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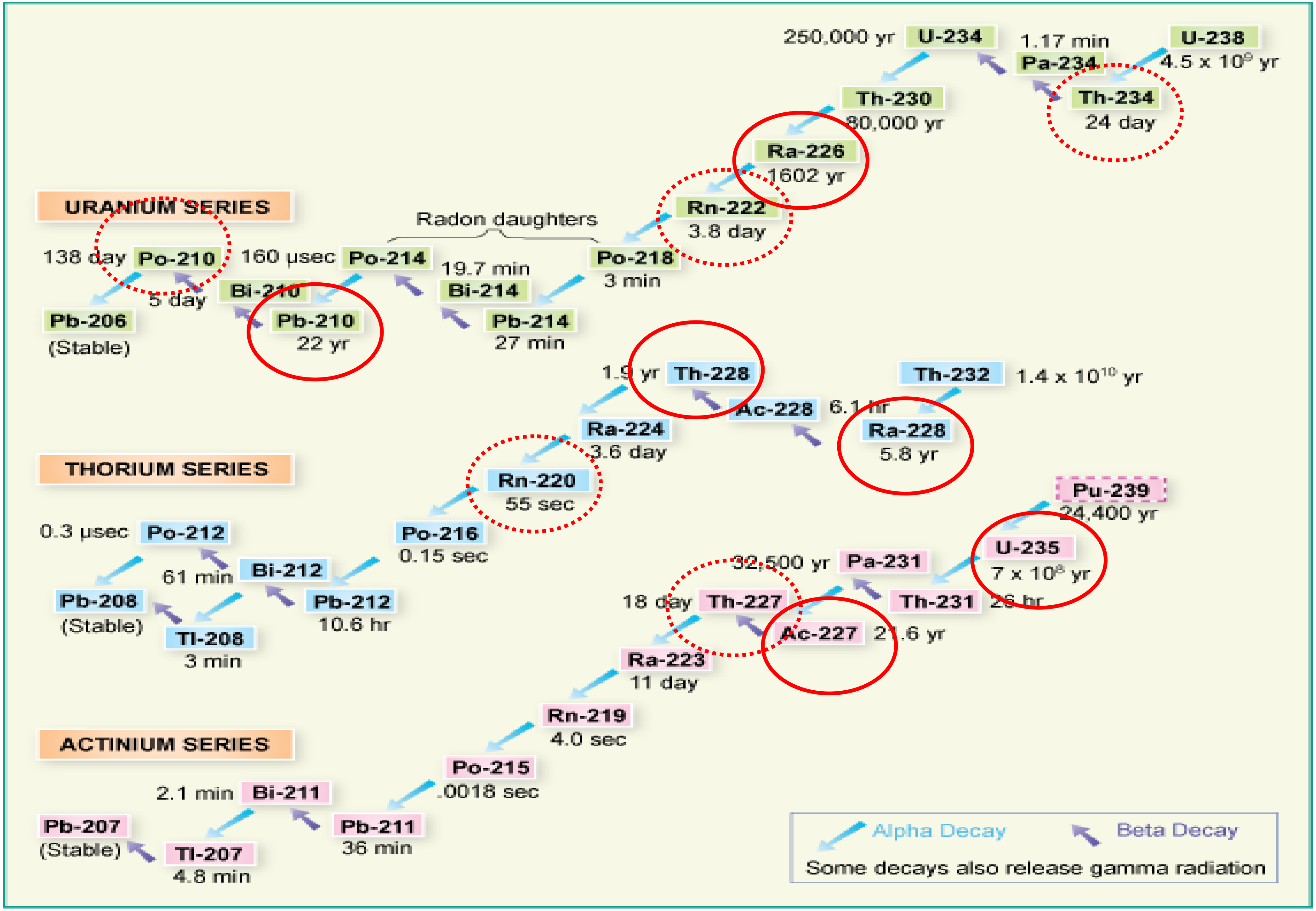
Equilibrium

3 cases

Secular equilibrium

Transient equilibrium

No equilibrium



URANIUM SERIES
THORIUM SERIES
ACTINIUM SERIES
U-238
4.5 x 10⁹ yr
Th-234
24 day
Pa-234
1.17 min
U-234
250,000 yr
Th-230
80,000 yr
Ra-226
1602 yr
Rn-222
3.8 day
Radon daughters
Po-218
3 min
Pb-214
27 min
Bi-214
19.7 min
Po-214
160 µsec
Pb-210
22 yr
Bi-210
5 day
Po-210
138 day
Pb-206
(Stable)
Th-232
1.4 x 10¹⁰ yr
Ra-228
5.8 yr
Ac-228
6.1 hr
Th-228
1.9 yr
Ra-224
3.6 day
Rn-220
55 sec
Po-216
0.15 sec
Pb-212
10.6 hr
Bi-212
61 min
Po-212
0.3 µsec
Tl-208
3 min
Pb-208
(Stable)
Pu-239
24,400 yr
U-235
7 x 10⁸ yr
Th-231
26 hr
Pa-231
32,500 yr
Ac-227
21.6 yr
Th-227
18 day
Ra-223
11 day
Rn-219
4.0 sec
Po-215
.0018 sec
Pb-211
36 min
Bi-211
2.1 min
Tl-207
4.8 min
Pb-207
(Stable)
Alpha Decay
Beta Decay
Some decays also release gamma radiation

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Secular equilibrium

- *Mother half-life >> daughter*
- *The apparent half-life of the daughter = the half-life of the mother*
- *Total activity is doubled*

=> Use correct half-life when calculating activity!!!

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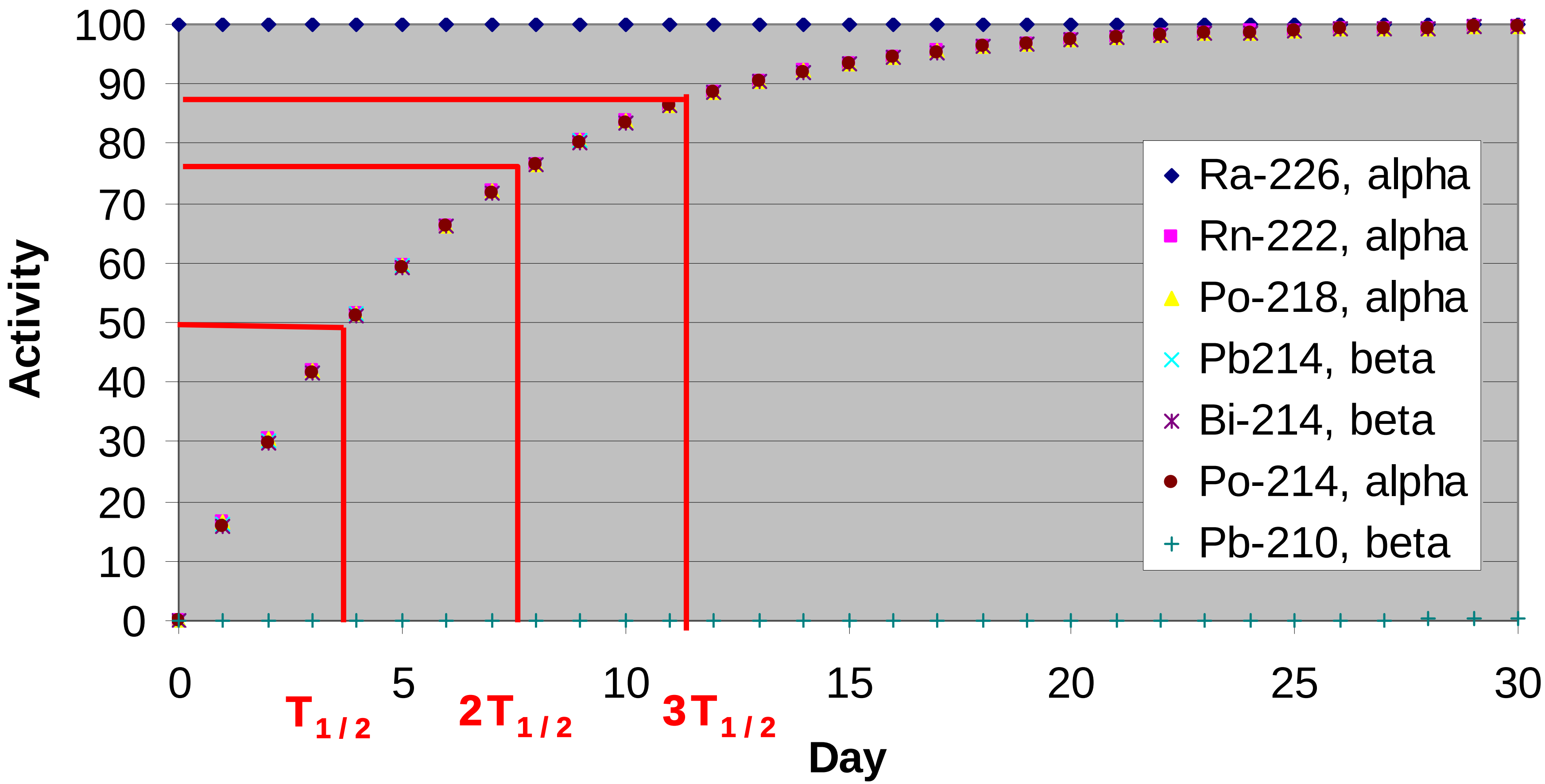
Decay starting at Ra-226

$T_{1/2}$ ^{222}Rn: 3.8 days



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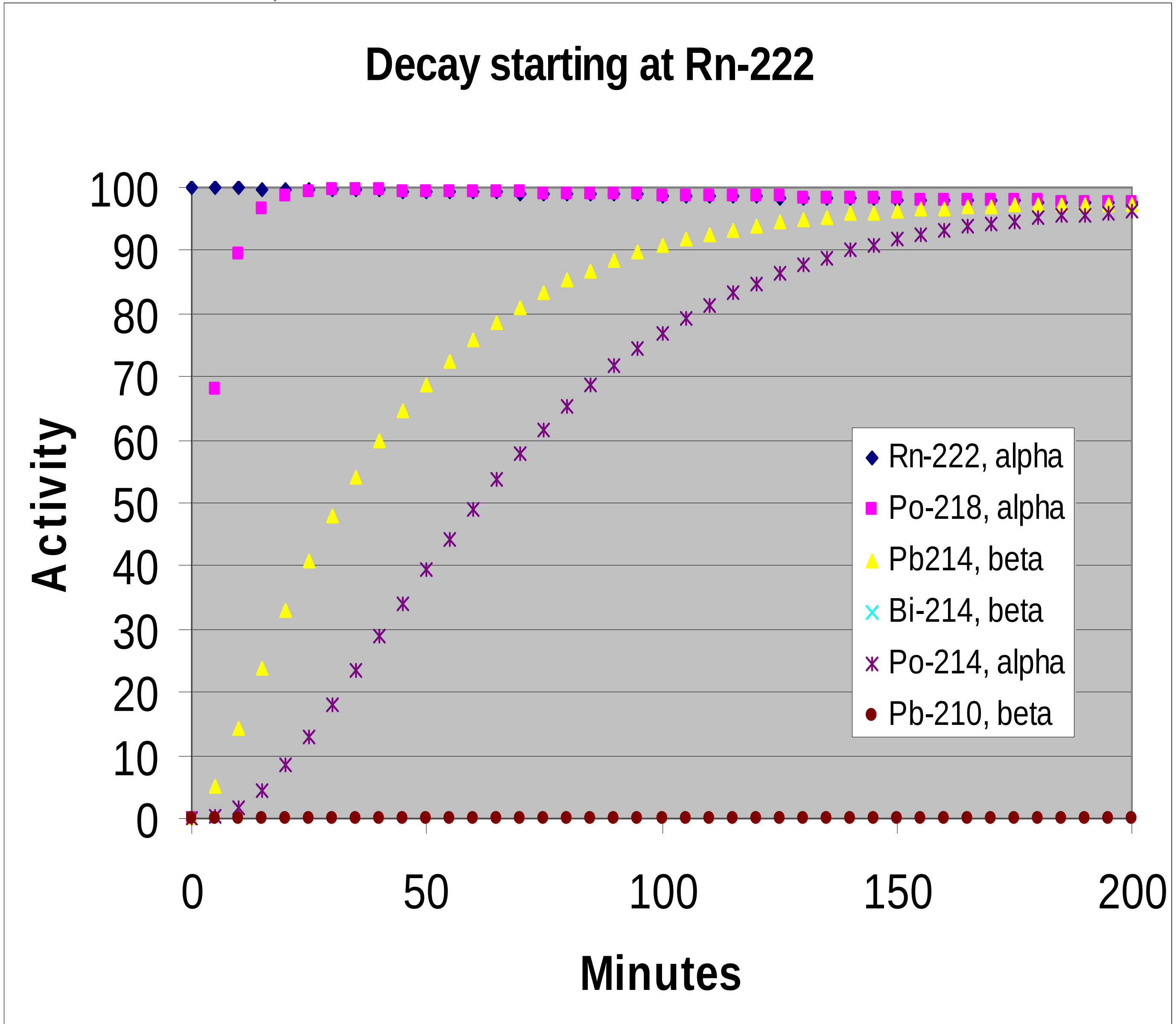
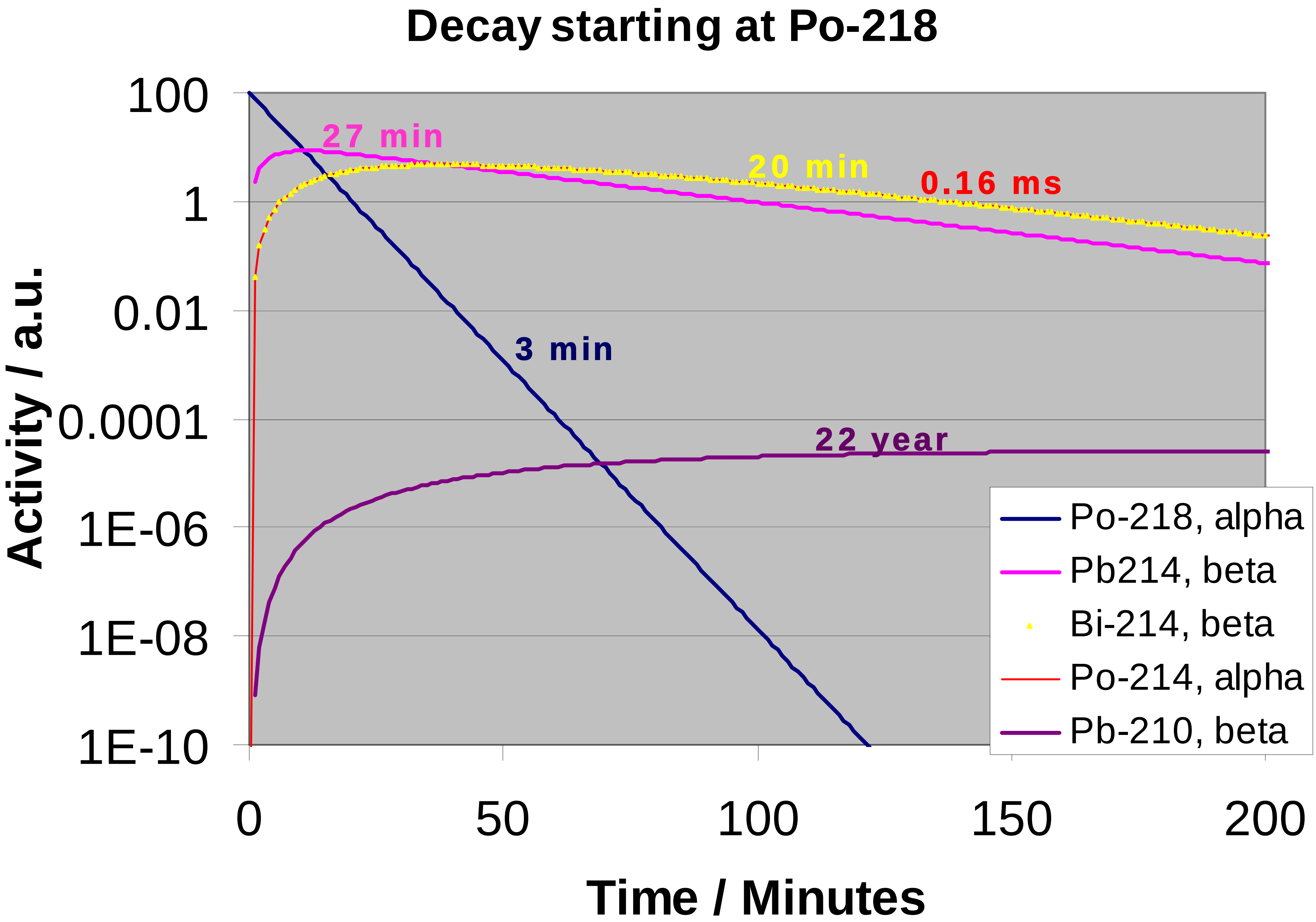
Common case of secular equilibrium

$$^{90}Sr \xrightarrow{\text{28.8 years}} {}^{90}Y \xrightarrow{\text{2.7 days}} {}^{90}Zr \text{ (stable)}$$

Starting with radon-222

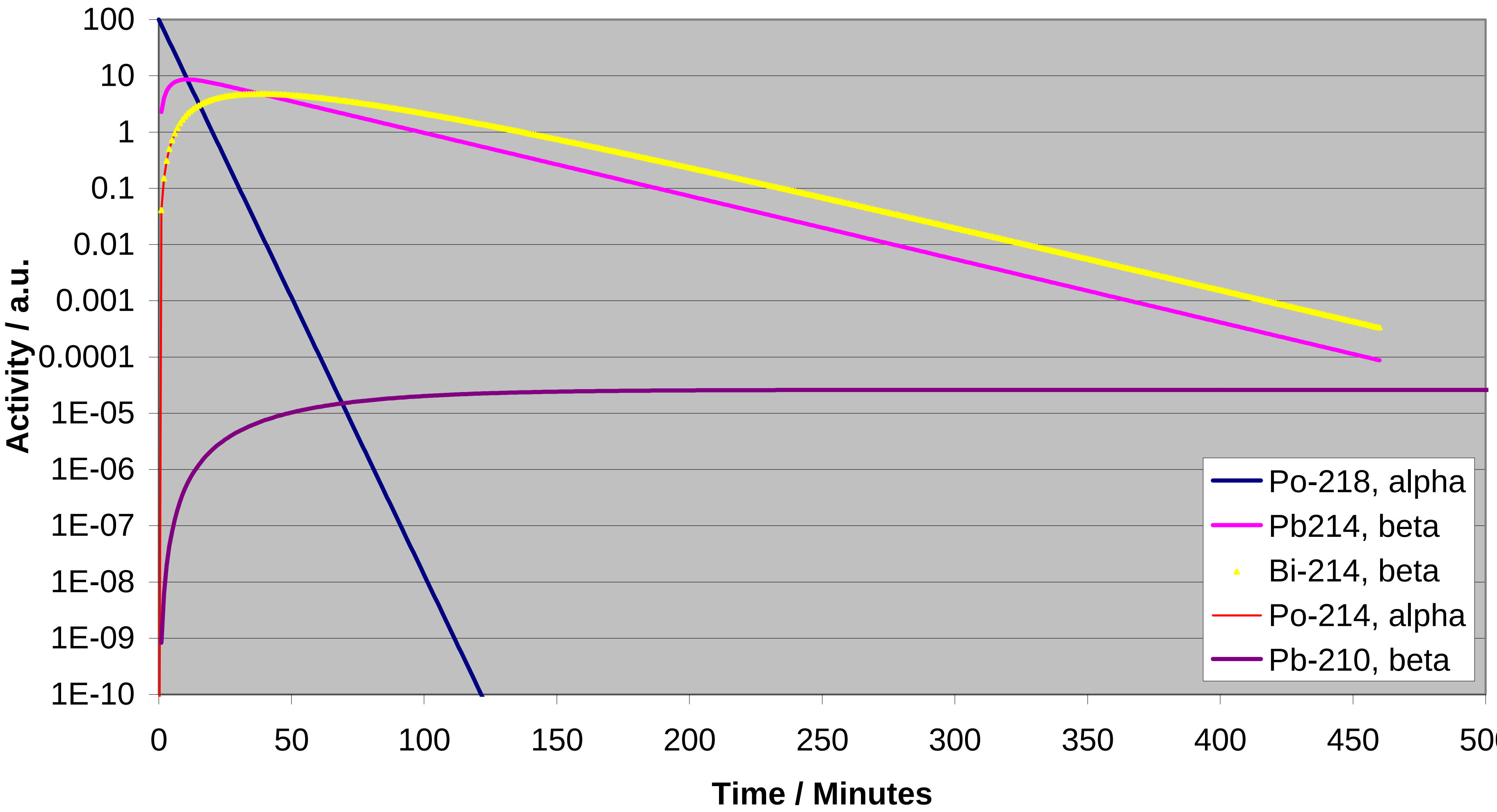
Half-life 3.8 days





Decay starting at Po-218
100
1
0.01
0.0001
1E-06
1E-08
1E-10
Activity / a.u.
0
50
100
150
200
Time / Minutes
27 min
20 min
0.16 ms
3 min
22 year
Po-218, alpha
Pb214, beta
Bi-214, beta
Po-214, alpha
Pb-210, beta

Decay starting at Po-218



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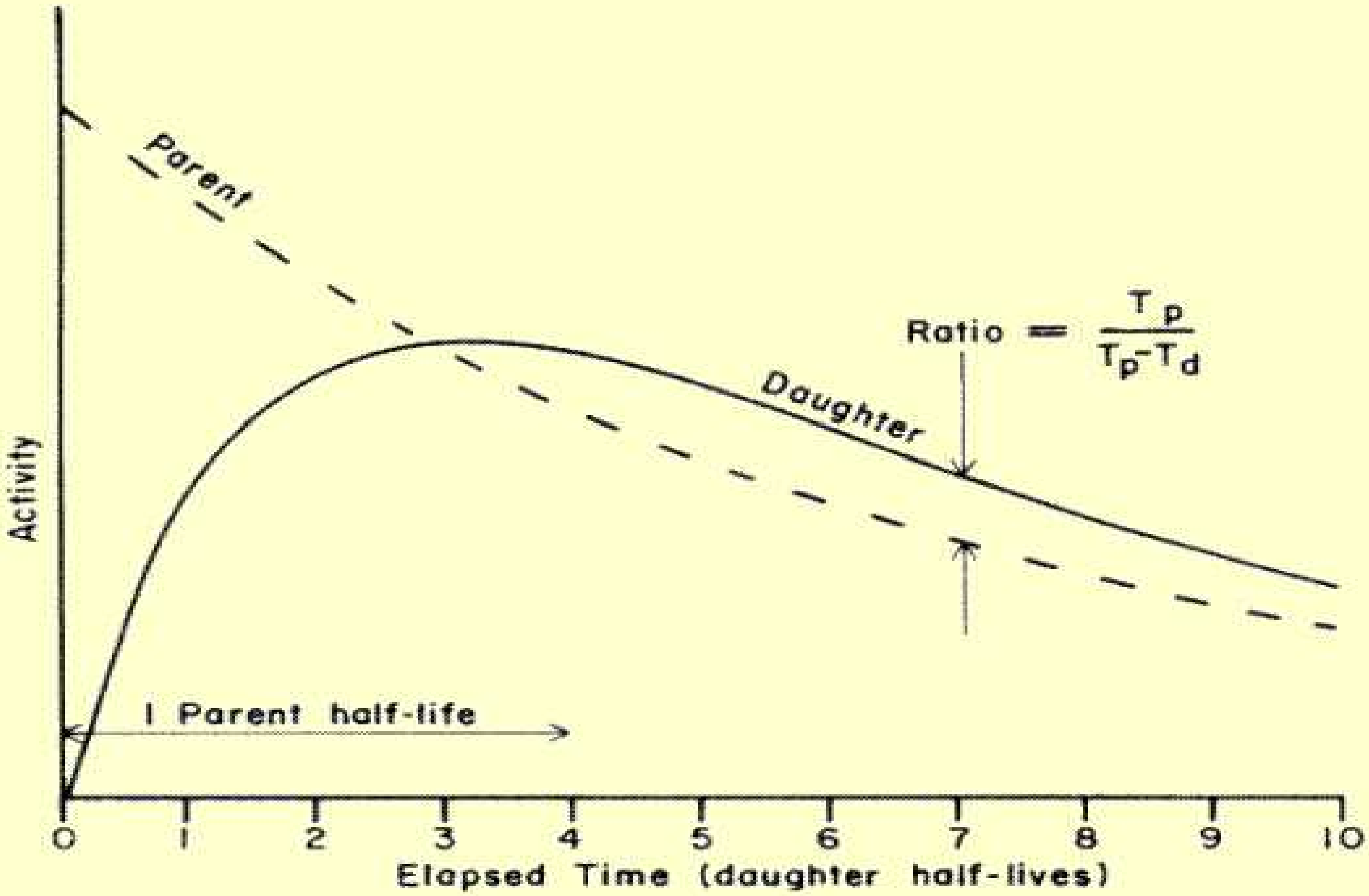
Transient equilibrium

- *Mother half-life > daughter half-life (ratio between 1 and 10000 or so)*
- *The apparent half-life of the daughter = the half-life of the mother*
- *Total activity is NOT EXACTLY doubled. Equilibrium factor:* $\dfrac{\lambda_B}{\lambda_B - \lambda_A}$


Parent
Daughter
Ratio = $\frac{T_p}{T_p - T_d}$
Activity
I Parent half-life
0 1 2 3 4 5 6 7 8 9 10
Elapsed Time (daughter half-lives)

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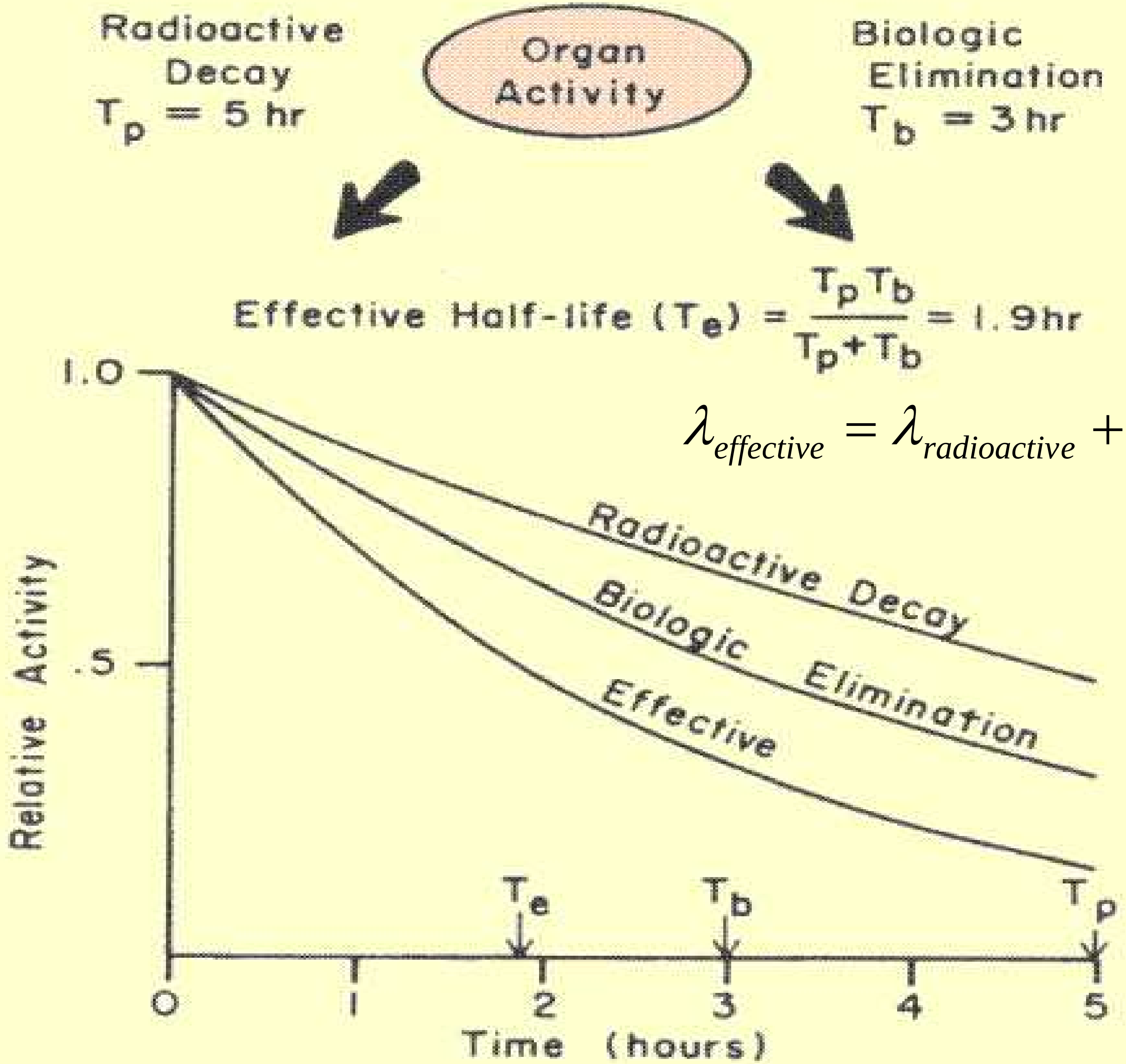
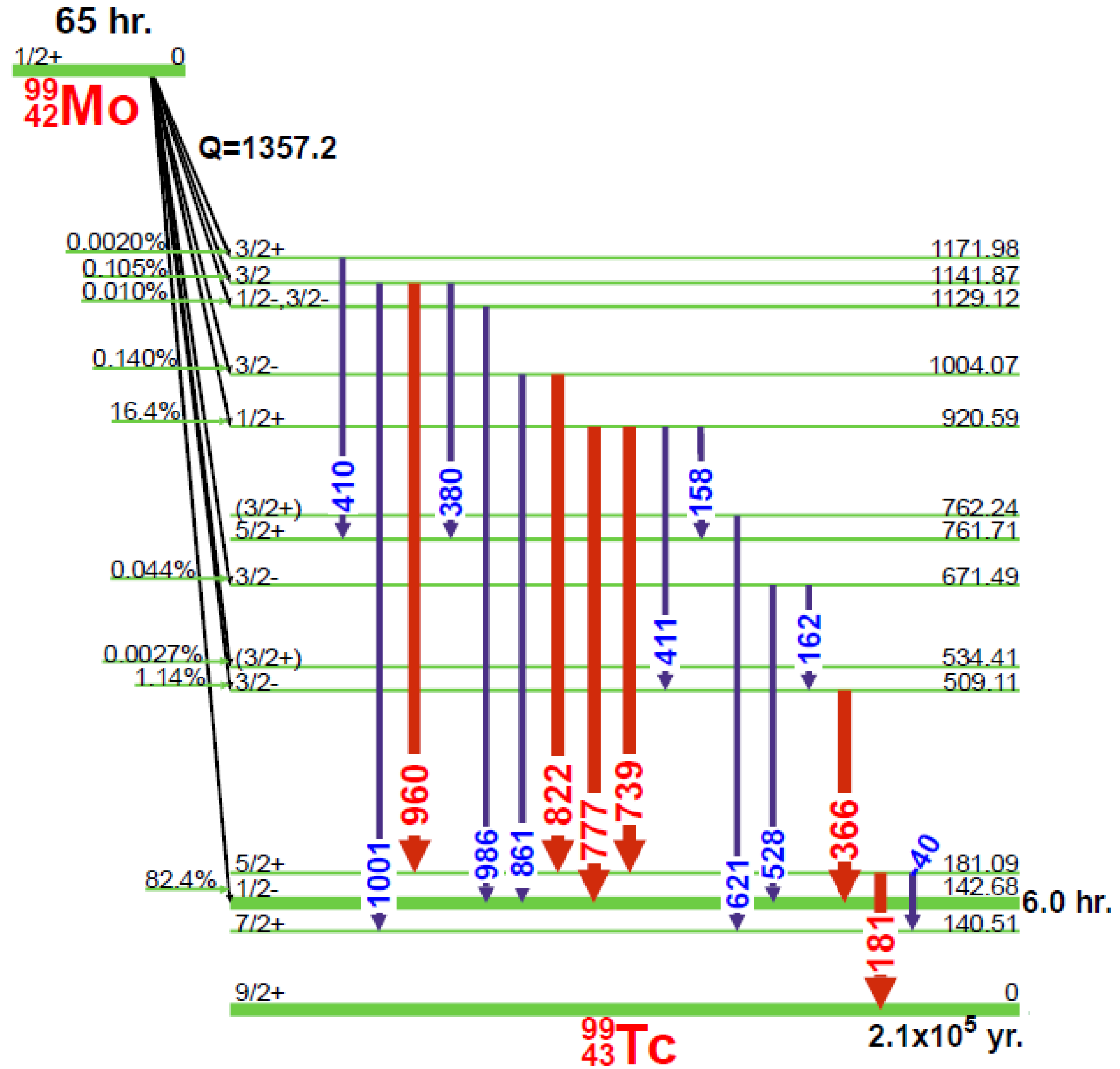
No equilibrium

- *Mother half-life < daughter half-life*



$$\lambda_{effective} = \lambda_{radioactive} + \lambda_{biological}$$

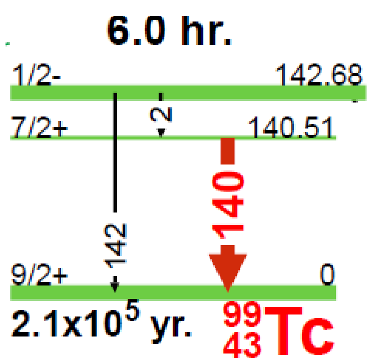
^{99}Mo(65 hr.) Decay Scheme



Iγ (140 keV) = **89,6%**

In equilibrium

^{99m}Tc*(6.0 hr.)



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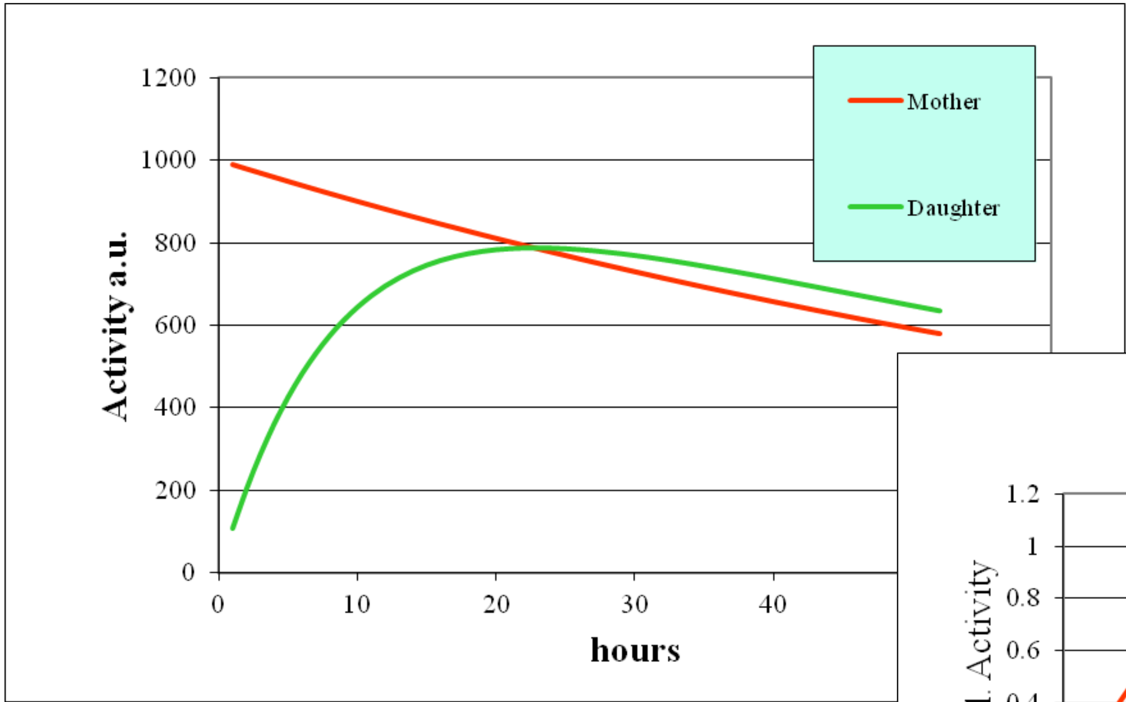
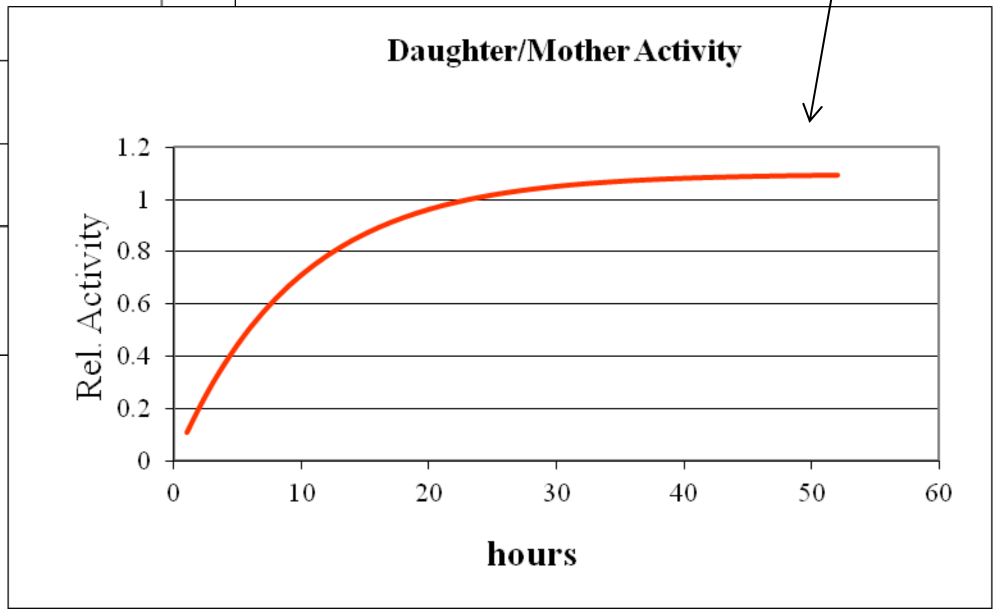
Mo-99 and Tc-99m



Ratio = 1.1



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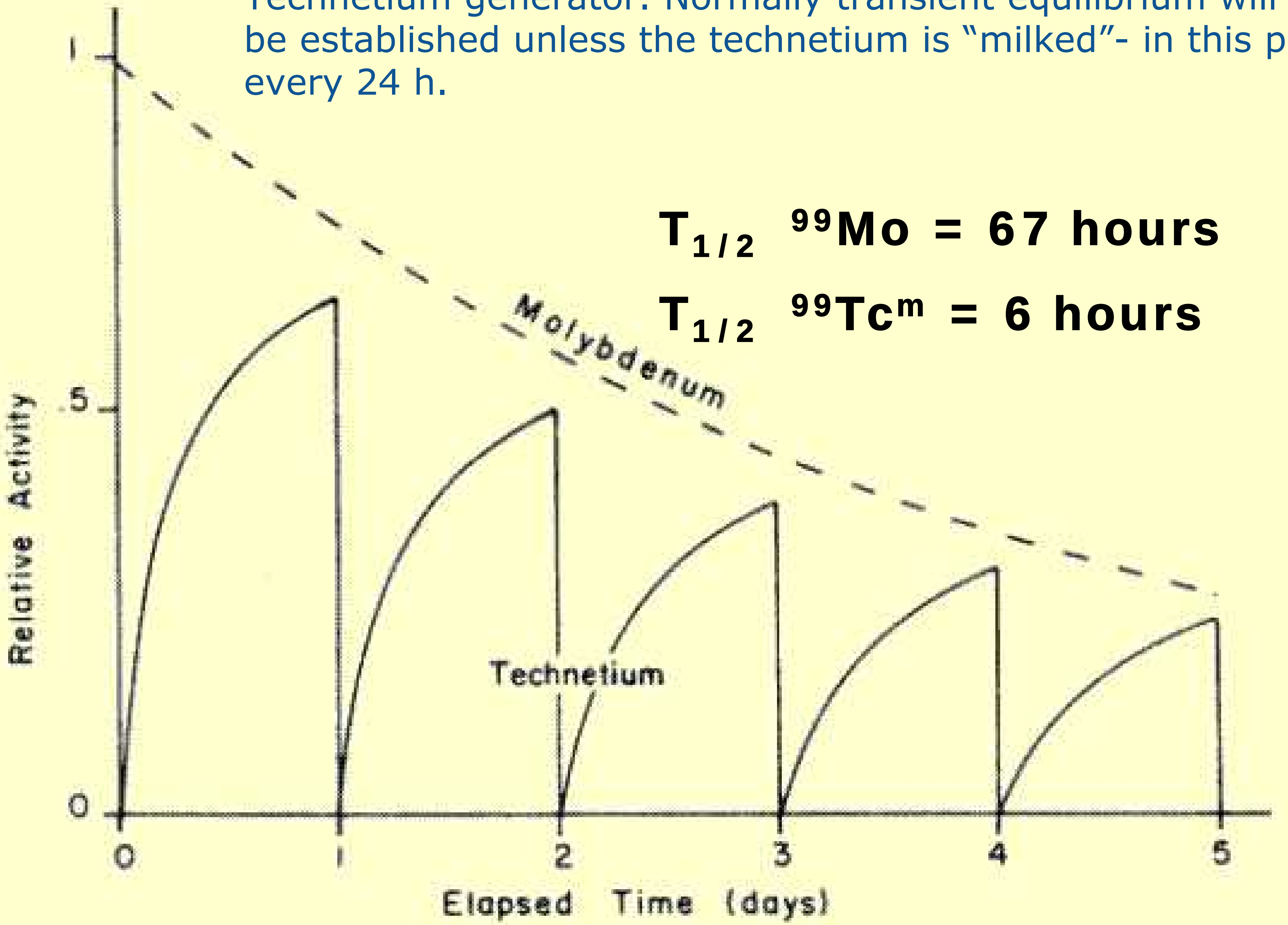
Technetium generator. Normally transient equilibrium will be established unless the technetium is “milked”- in this plot every 24 h.

$T_{1/2}$ ^{99}Mo = 67 hours

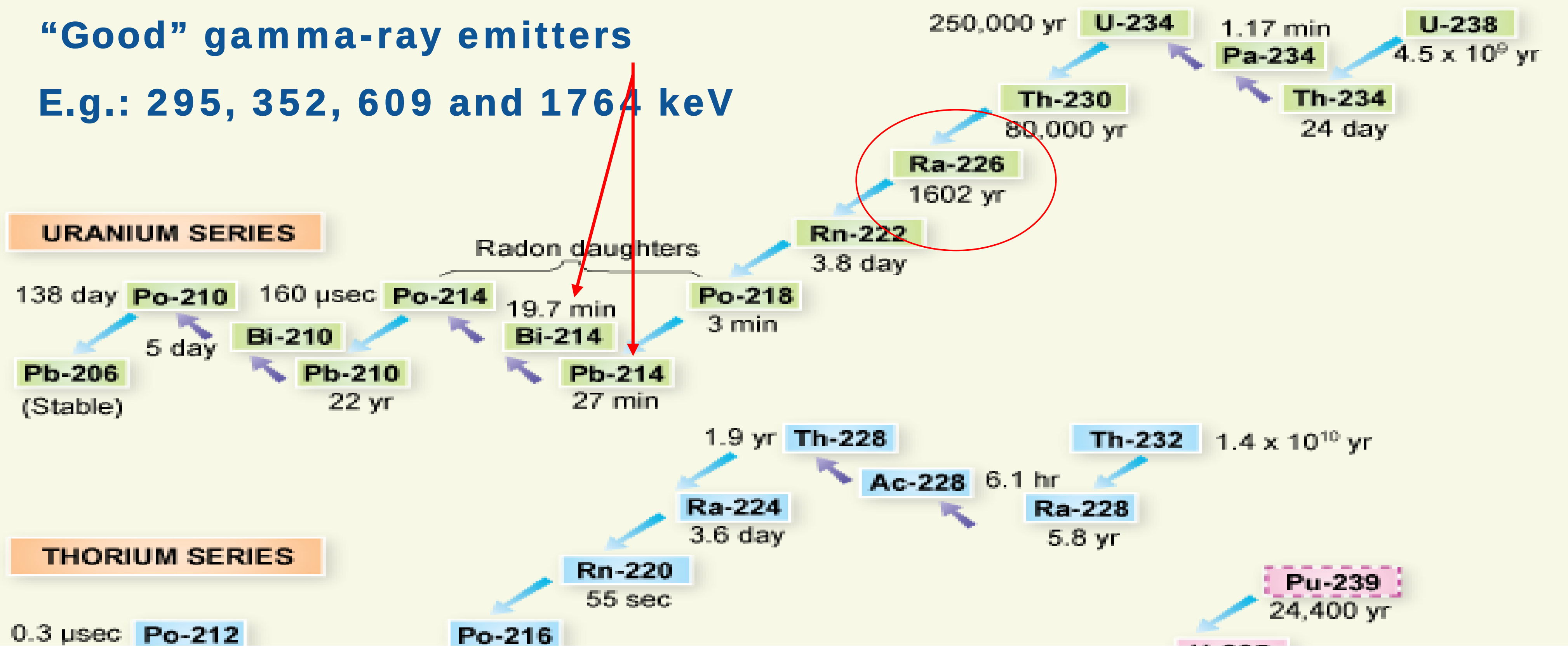
$T_{1/2}$ ^{99}Tc^{m} = 6 hours



Measurement of Ra-226 using gamma-ray spectrometry

"Good" gamma-ray emitters

E.g.: 295, 352, 609 and 1764 keV



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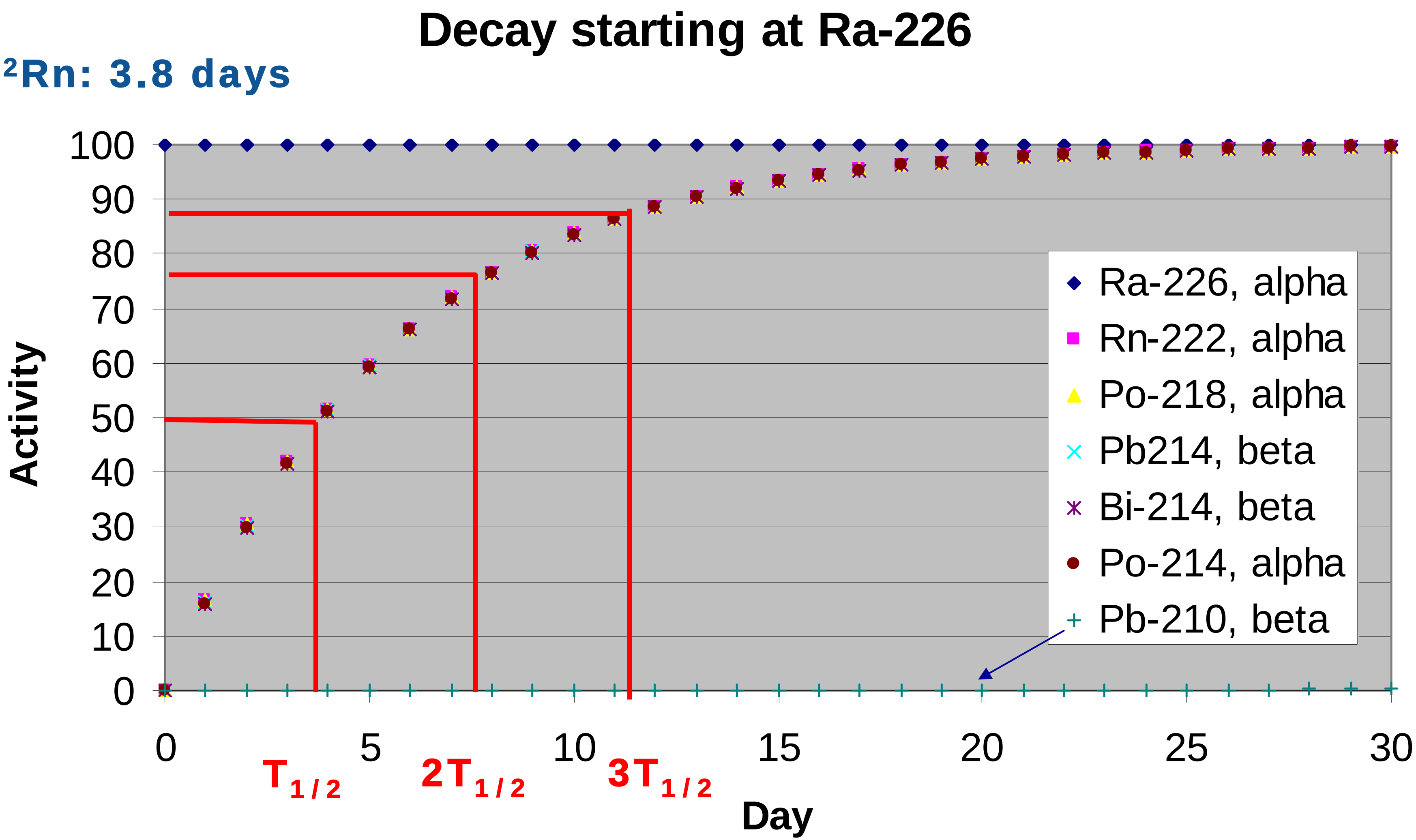
Secular or transient?

$T_{1/2}$ ^{222}Rn: 3.8 days



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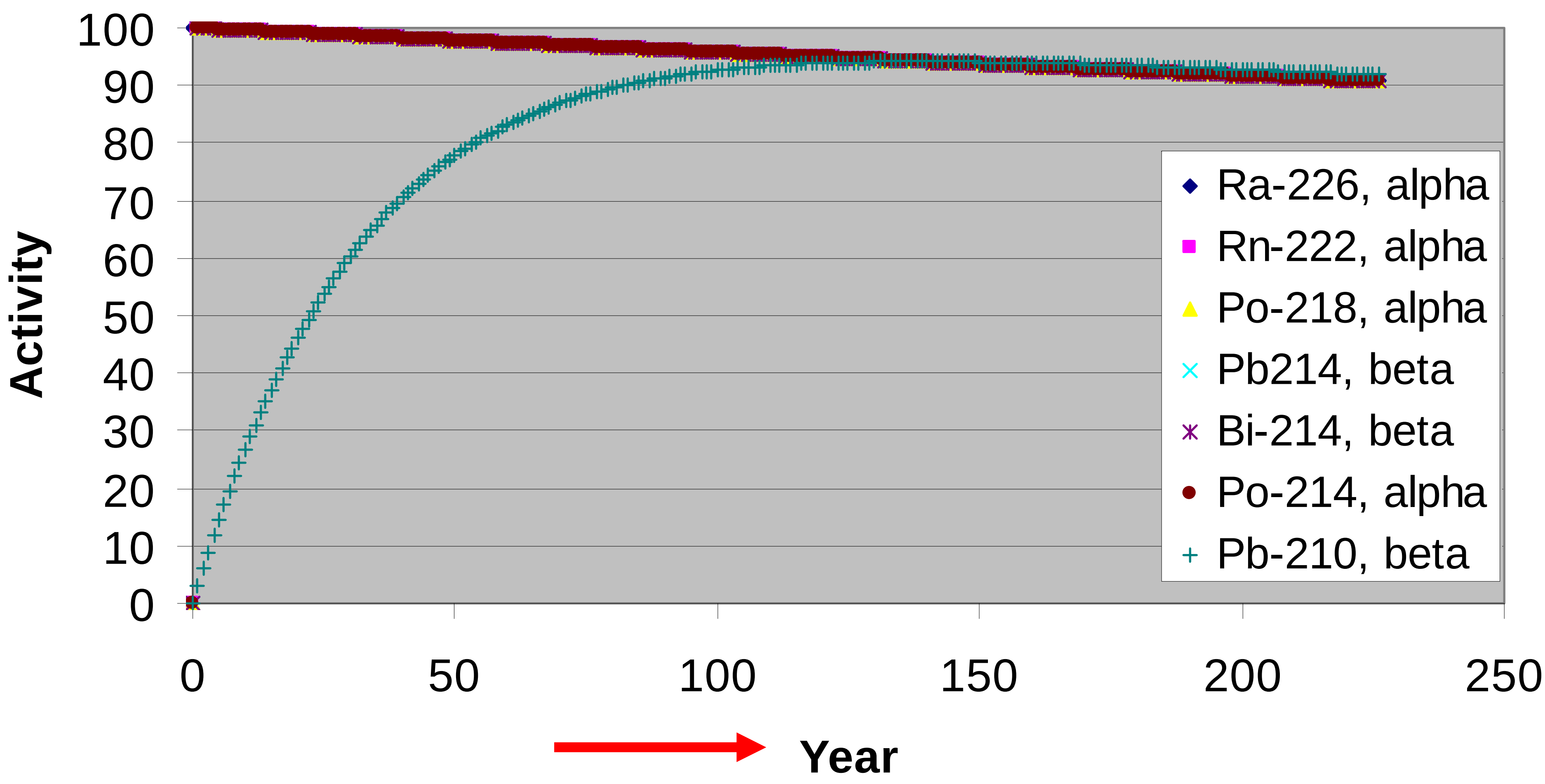
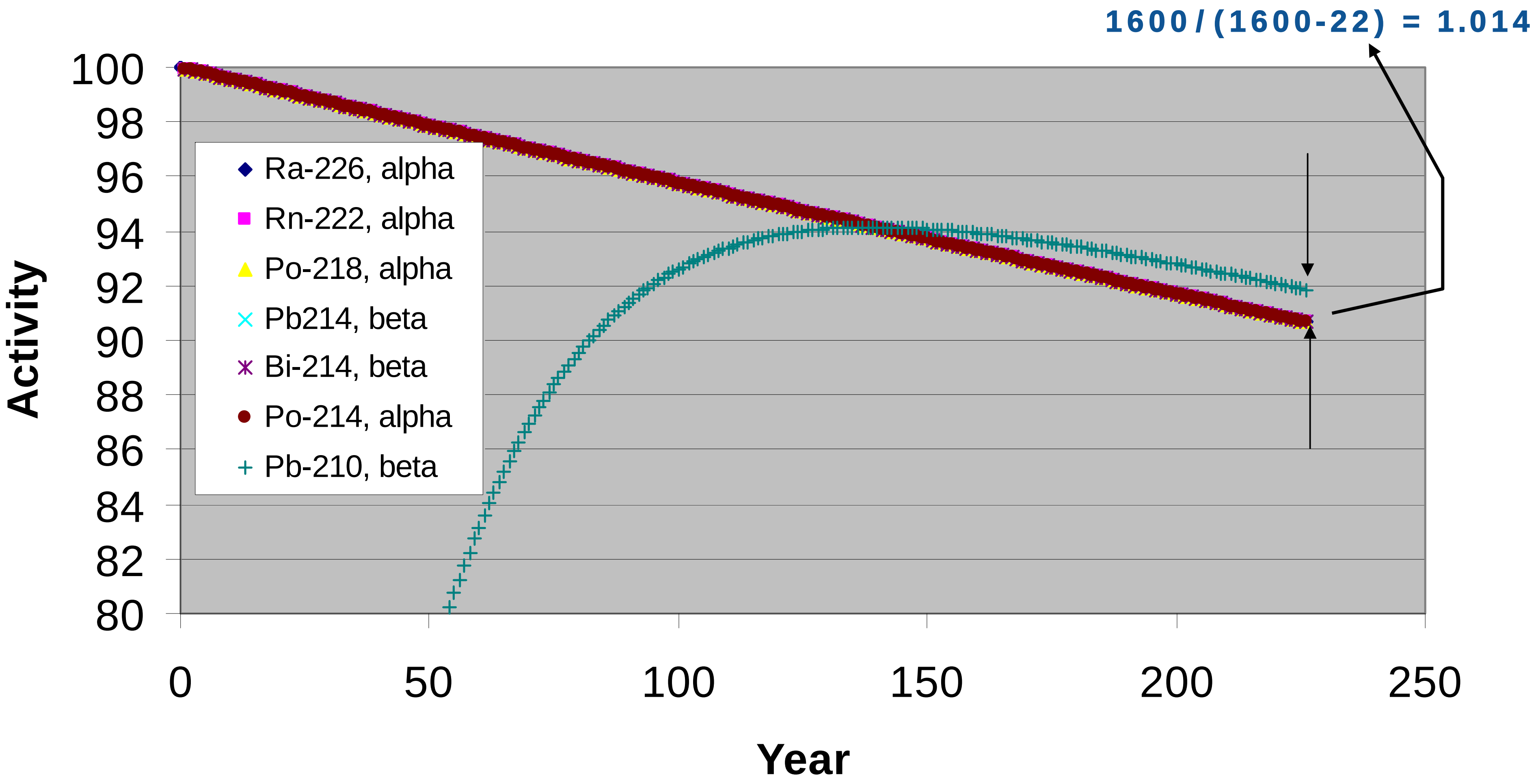
Decay starting at Ra-226



Decay starting at Ra-226



Quantification of Ra-226 and U-235 in a soil sample

- Soil contains many natural radionuclides and results in a complex gamma-ray spectrum.
- Other methods are better than Gamma-ray spectrometry (GS) for determining U and Ra but GS is often used due to simple procedure and low cost.
- Difficult to use the 186.2 keV line from Ra-226 due to interference from U-235 (185.7 keV) – in a typical soil sample the two peak areas are fairly equal*.
- Difficult to detect 144 keV line and 163 keV lines from U-235 (lower emission probability).
- Need to use the 295, 352, 609 and/or 1764 keV lines (and more if detected) from Pb-214 and Bi-214.

****If the U-238 and Ra-226 activities are equal and there is a natural isotopic abundance of U, the ratio of peak counts (Ra226/U235) will be 1.27.***

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Quantification of Ra-226 and U-235 in a soil sample

- **Difficult to use the 186.2 keV line from Ra-226 due to interference from U-235 (185.7 keV)**
- **Difficult to detect 144 keV line and 163 keV lines from U-235.**
- **Need to use the 295, 352, 609 and/or 1764 keV lines (and more if detected) from Pb-214 and Bi-214.**

1) **Fill a radon-tight container to the rim with your sample (e.g. dried soil)**
2) **Make sure equilibrium is established: Keep the sample in a sealed radon-tight container for about 2 weeks (or so). – *if you have time you can always measure the increase of activity from radon-daughters during this time***
3) **Calculate the Ra-226 activity based on the 295, 352, 609 and/or 1764 keV lines**
4) **Calculate the number of counts in the 186.2 keV peak based on the Ra-226 activity – subtract these counts from the total 186 keV peak area in order to obtain the counts for the 185.7 keV peak.**
5) **A good final check is to look at the ratio between U-238 and U-235**

Muon activation in 2 kg germanium

"only" based on Mn-54, Co-57, Co-58, Co-60, Zn-65, Ge-68, Ge-77 and As-73