Application of the bateman equation to analyze disequilibrium in the ^{232}Th and ^{238}U chains in low-background detector materials

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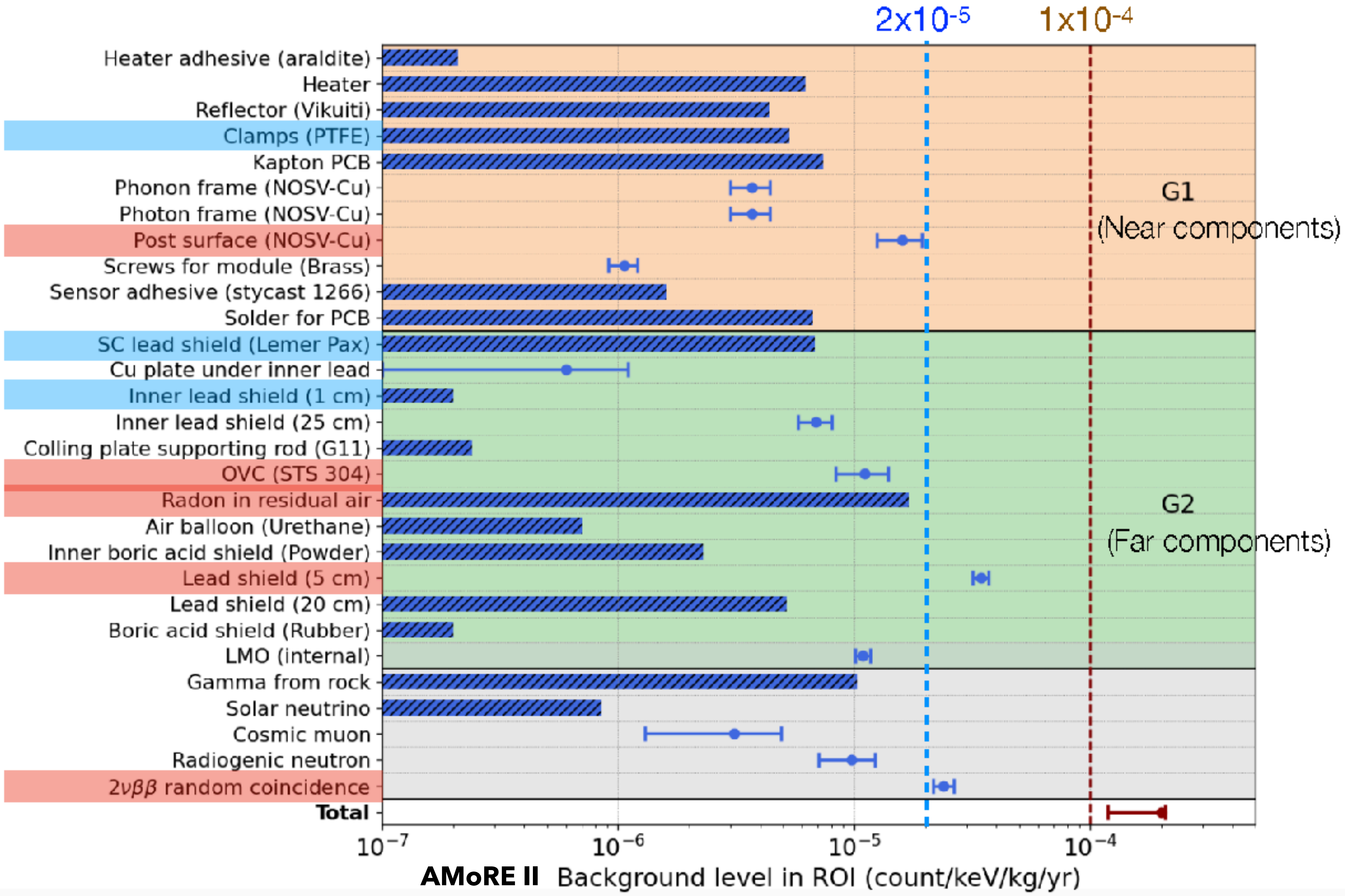
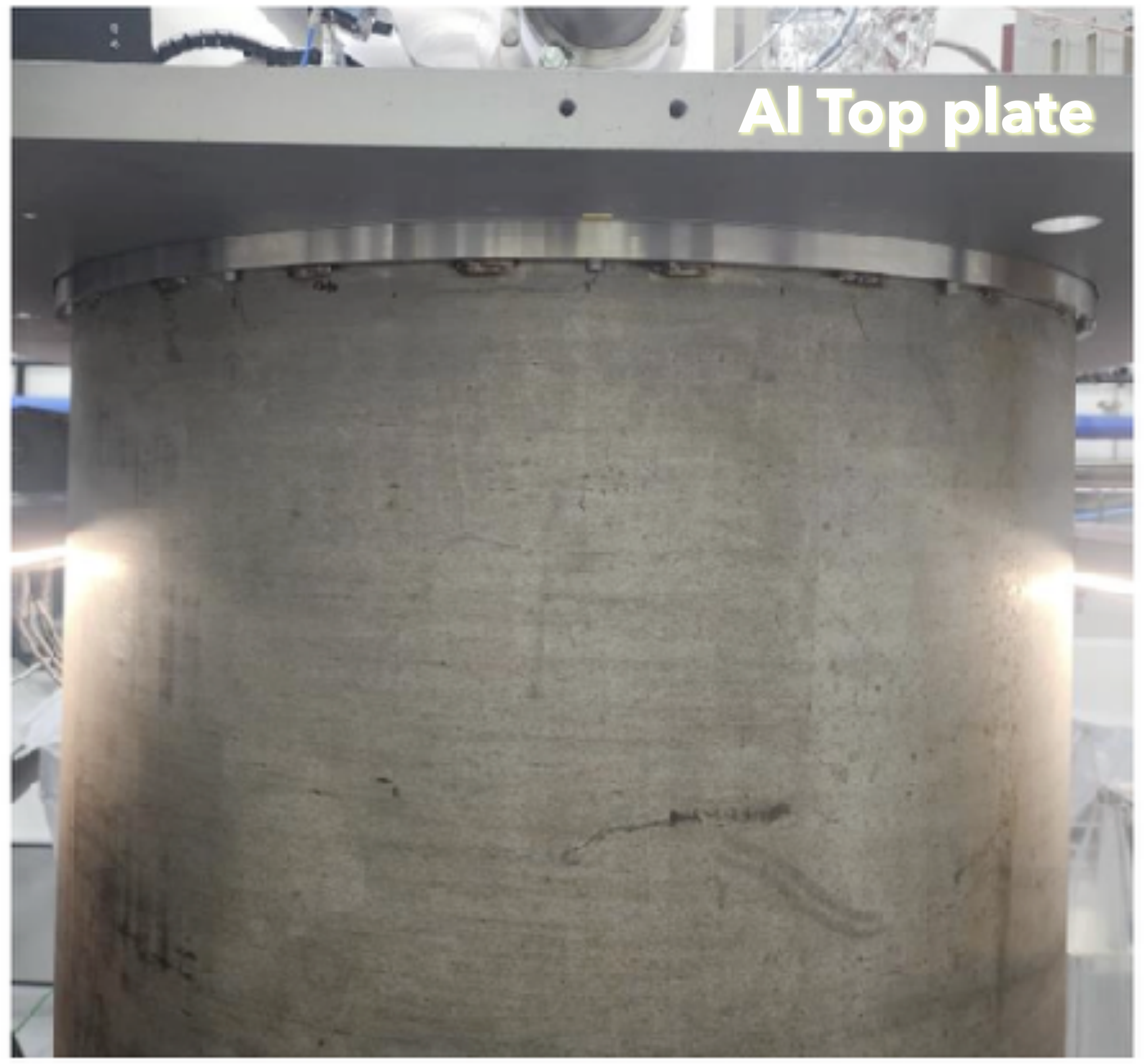
Introduction

- need finite measurements
- need to lower the BG level



Introduction

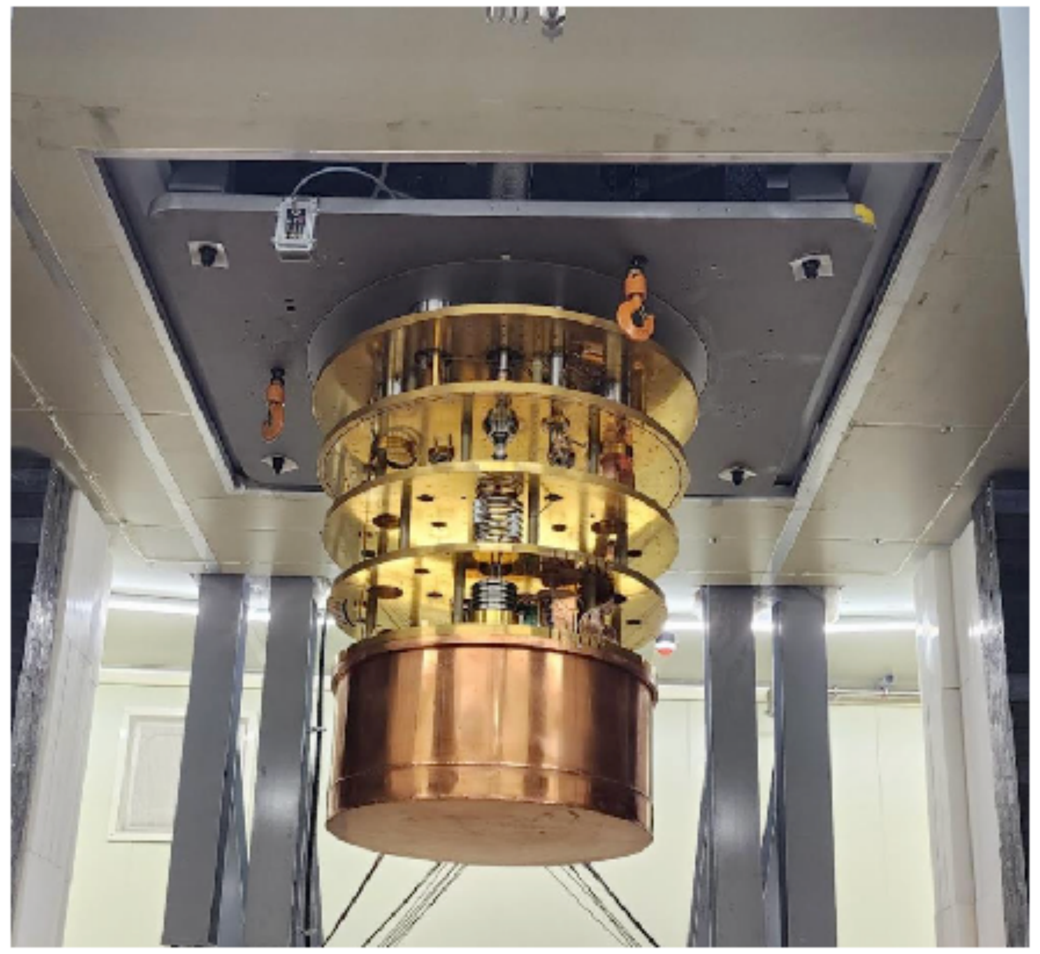


- Background modeling step:
  Simulation indicated that the square **Al top plate** covering the cylinder OVC could **contribute to detector backgrounds**.
- Need for assay:
  To quantify this contribution, the plate was measured with an HPGe detector.
- Unexpected observation:
  The results revealed **large disequilibrium between ^{228}Ac and ^{228}Th**.
- Cross-check:
  Additional ICP-MS measurements of U/Th were performed
  → It was found that the measured **^{228}Th activity was much higher than what the Th concentration** would suggest.

^{234}Th	^{234m}Pa	^{226}Ra(^{238}U)	^{40}K	^{228}Ac	^{228}Th	Unit
13.6 ± 0.7 Bq	14.6 ± 0.8 Bq	4.42 ± 1.14	22.24 ± 5.79	35.84 ± 3.12	**976.56 ± 49.13**	mBq/kg
U: 1070 ppb (13.3 Bq/kg)				Th : 42.5 ppb (**172** mBq/kg)		

HPGe detector and ICP-MS measurement result

Introduction



- **Follow-up study:**
  A previously measured **Al supporter** sample was re-examined about **three and a half years later.**

^{234}Th	^{234m}Pa	^{226}Ra(^{238}U)	^{228}Ac	^{228}Th	Unit, yr
25.2 ± 1.3 Bq	21.1 ± 1.3 Bq	**< 8.31**	**55.6 ± 7.3**	**1523 ± 78**	mBq/kg (2021)
21.9 ± 1.1 Bq	22.5 ± 1.3 Bq	**337.4 ± 18.0**	**159.0 ± 11.4**	**480.9 ± 25.2**	mBq/kg (2025)

Al supporter measured at 2021.05.07 and at 2025.01.31

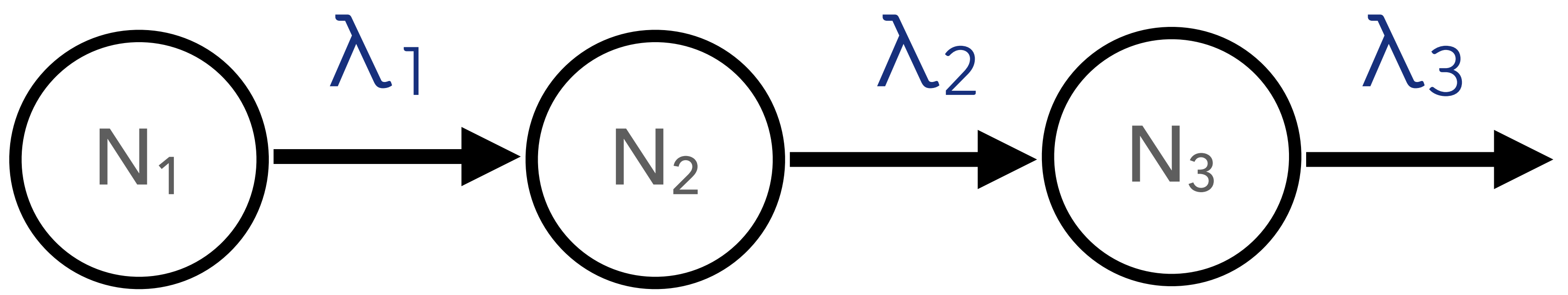
- **Consequence:**
  This discrepancy motivated a systematic study using the **Bateman equation** to explain the time evolution of chain activities.

			Activities (mBq/kg)		Background level (x10^{-5} ckky)
	Material	**Method**	**U-238 (Ra-226)**	**Th-232 (Th-228)**	
Al top plate	Aluminum	HPGe/ICPMS	4.4(11)	977(49)	< 0.29(90% C.L)

From Dr. Eunju Jeon's poster [Updated background simulation and detector design for AMoRE-II]

Bateman equation

General sequential decay : Bateman equation



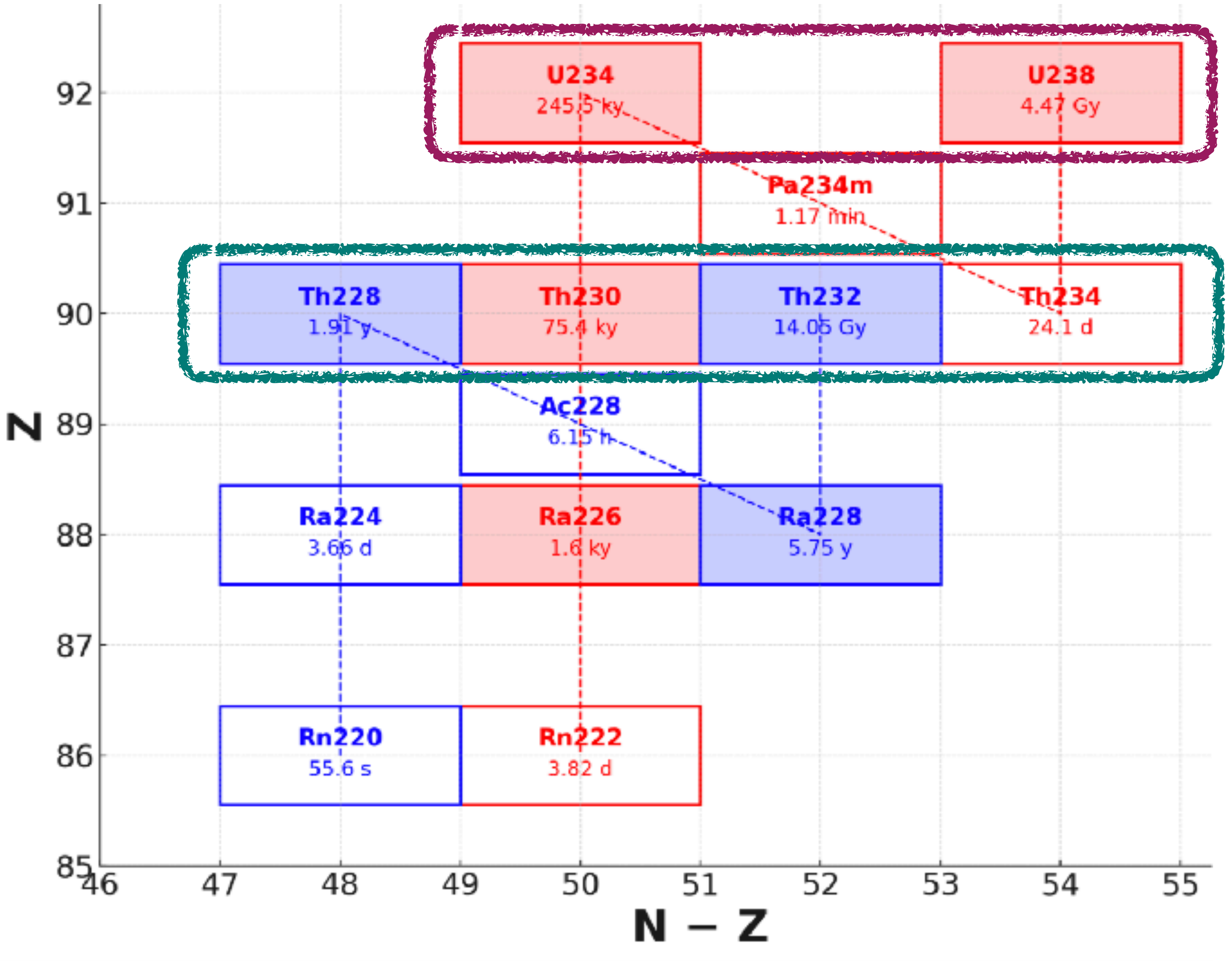
Assume, $N_1(0) > 0, \quad N_{i>1}(0) = 0$

The number of atoms of the n-th isotope at time t is given by:

$$N_n(t) = N_1(0) \sum_{i=1}^{n} \left(\frac{e^{-\lambda_i t} \prod_{j=1, j \neq i}^{n} \lambda_j}{\prod_{j=1, j \neq i}^{n} (\lambda_j - \lambda_i)} \right)$$

This equation describes the number of daughter nuclei produced at a given time in a sequential decay process.

Connection of ^{238}U and ^{232}Th chain in material

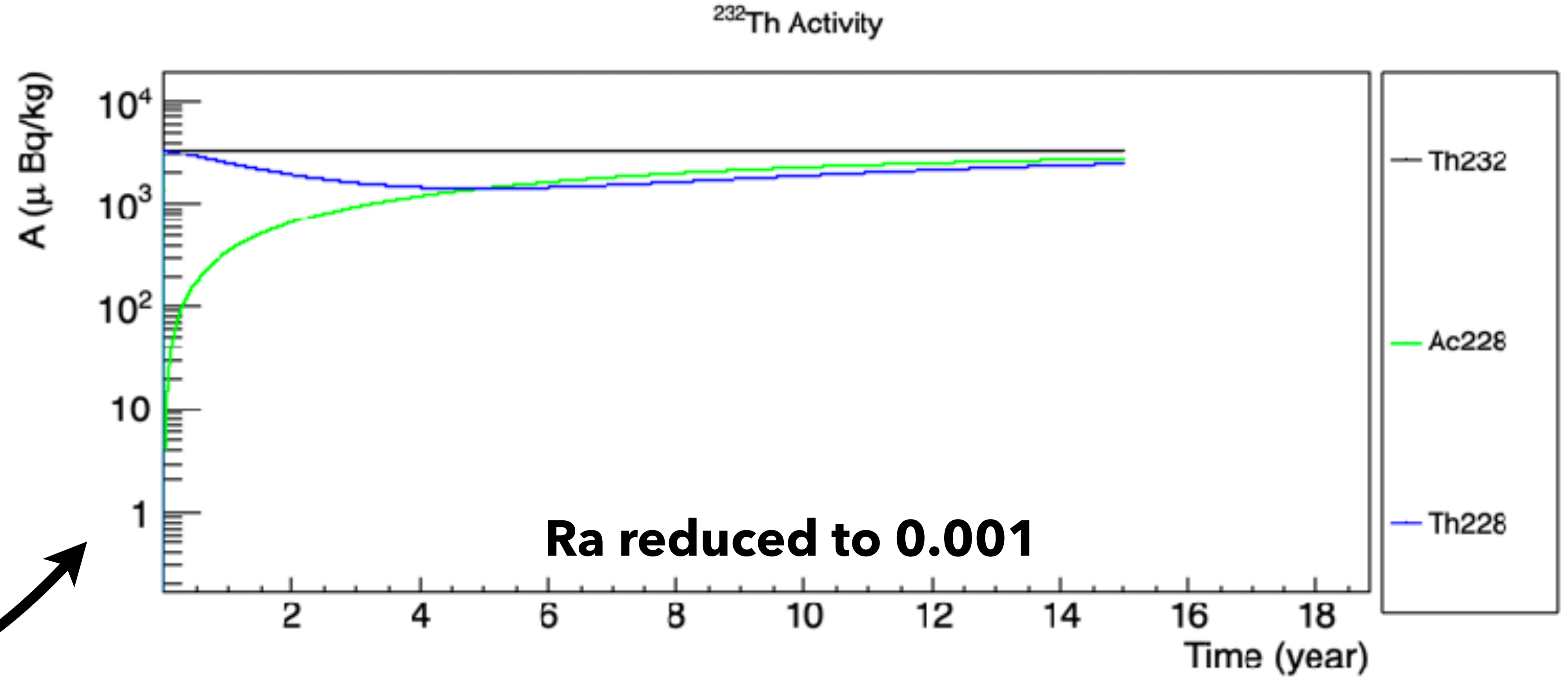


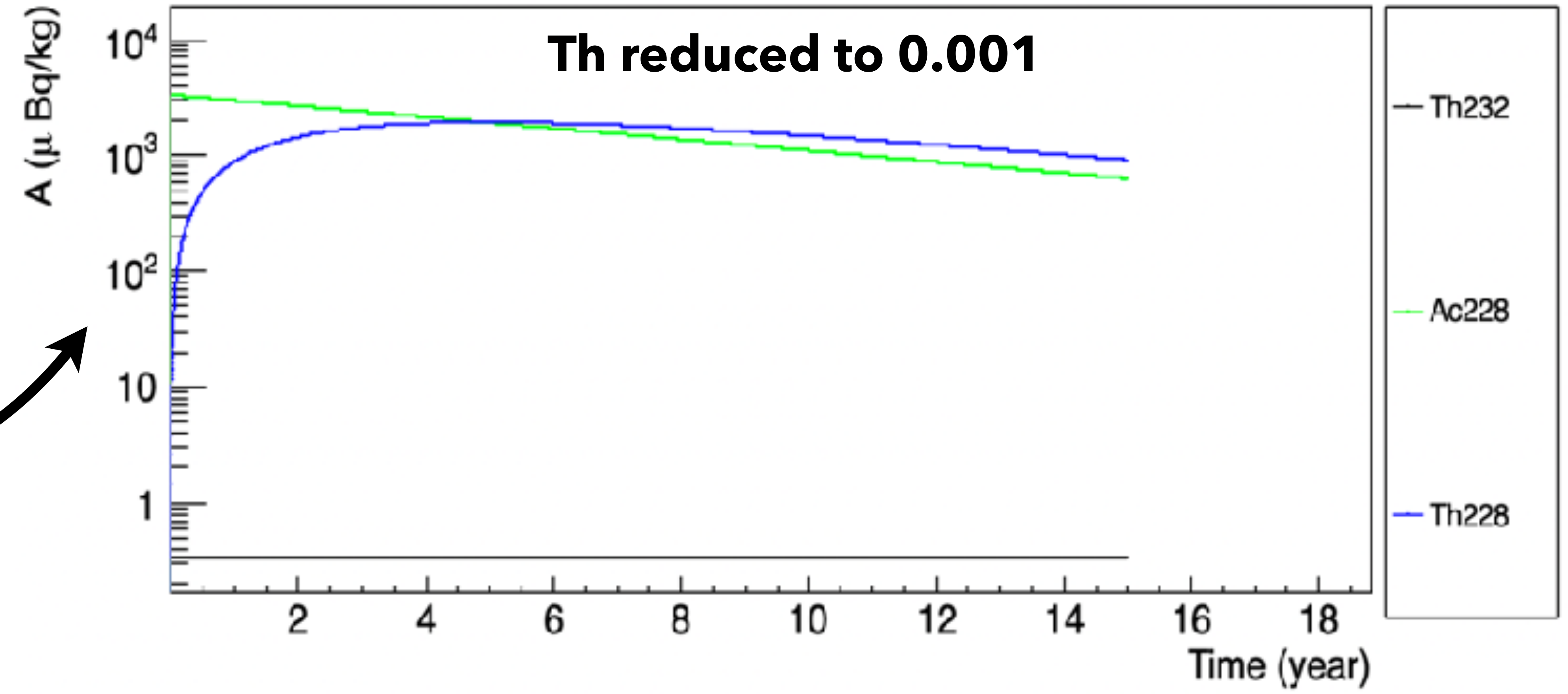
^{238}U and ^{232}Th chain diagram
(Solid square : over 1 yr half-life isotope)
Plot by Gemini

- There are only three elements with long half-lives - U, Th and Ra - that remain after chemical process.
- Changes of Th and Ra affect the radioactivities of both Th and U chains in correlation, which means **^{232}Th(Th chain) and ^{230}Th(U chain) change in the same ratio**.
- The ^{238}U chain reaches secular equilibrium on the order of 500 k years, while the ^{232}Th chain does so in about 40 years.

Why is the activity of ^{228}Th higher than ^{232}Th ?

- Aluminum data shows ^{228}Th has higher activity than ^{232}Th and higher than ^{228}Ac(^{228}Ra) more than 20 times.
- Ra reduction alone can't explain lower ^{232}Th
  - ^{232}Th> ^{228}Th
  - ^{228}Th can't be higher than ^{228}Ac as observed in the experimental results.
- Th reduction gives ^{228}Ac higher than ^{228}Th
  ^{228}Ac > ^{228}Th





Why ^{226}Ra increased in Al supporter sample

$$N_2(t) = \frac{\lambda_1 N_1(0)}{\lambda_2 - \lambda_1}(e^{-\lambda_1 t} - e^{-\lambda_2 t}) \rightarrow A_2(t) = \frac{\lambda_2 A_1(0)}{\lambda_2 - \lambda_1}(e^{-\lambda_1 t} - e^{-\lambda_2 t})$$

$$\lambda_2 \gg \lambda_1, \quad A_2(t) \approx A_1(0)\lambda_2 t \approx A_1(0)\frac{t}{T_{1/2}}$$

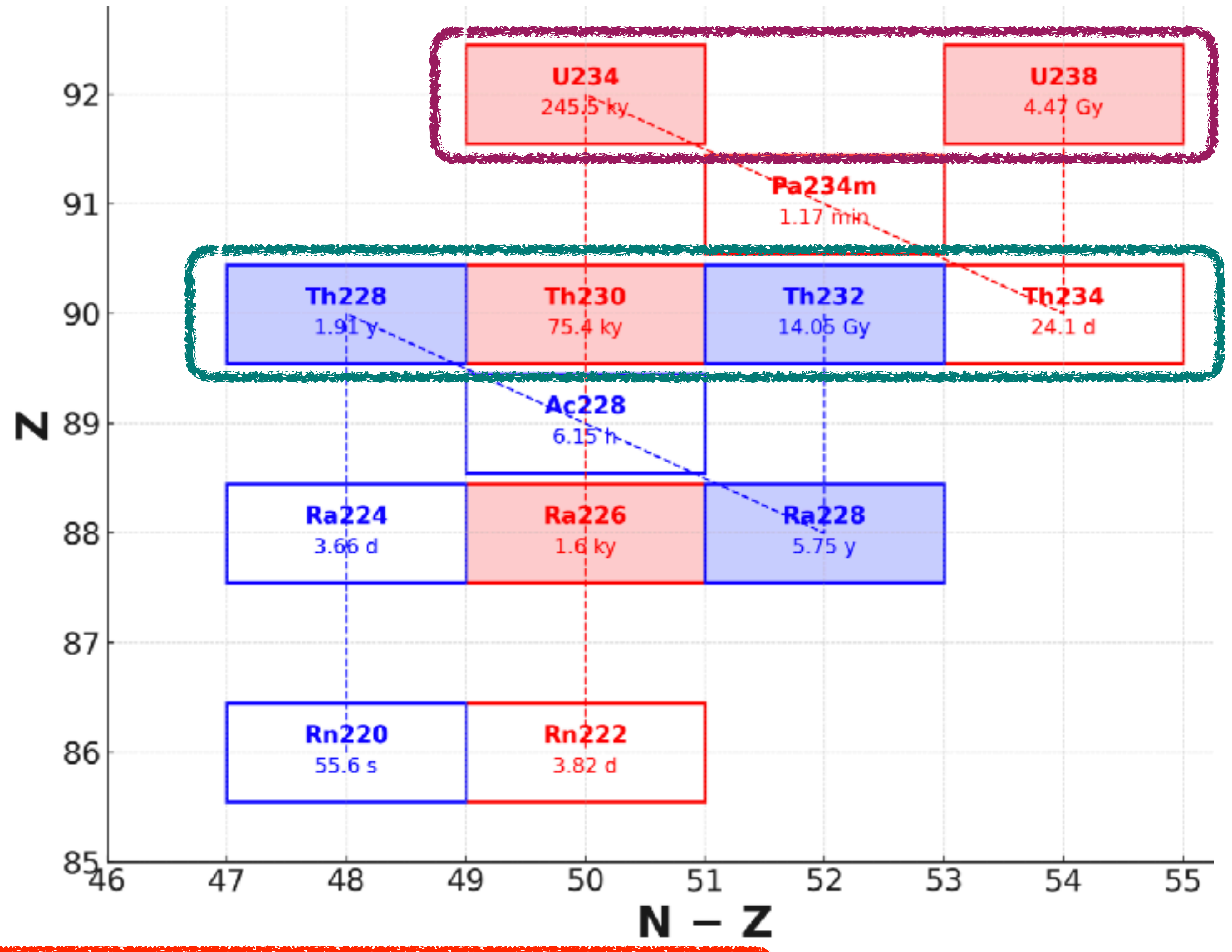
This is the activity produced by mother isotope

$$A_2'(t) = A_1(0)\lambda_2 t$$

$A_2'(t)$ is the activity produced from A_1

Measurement activity of A_2 is

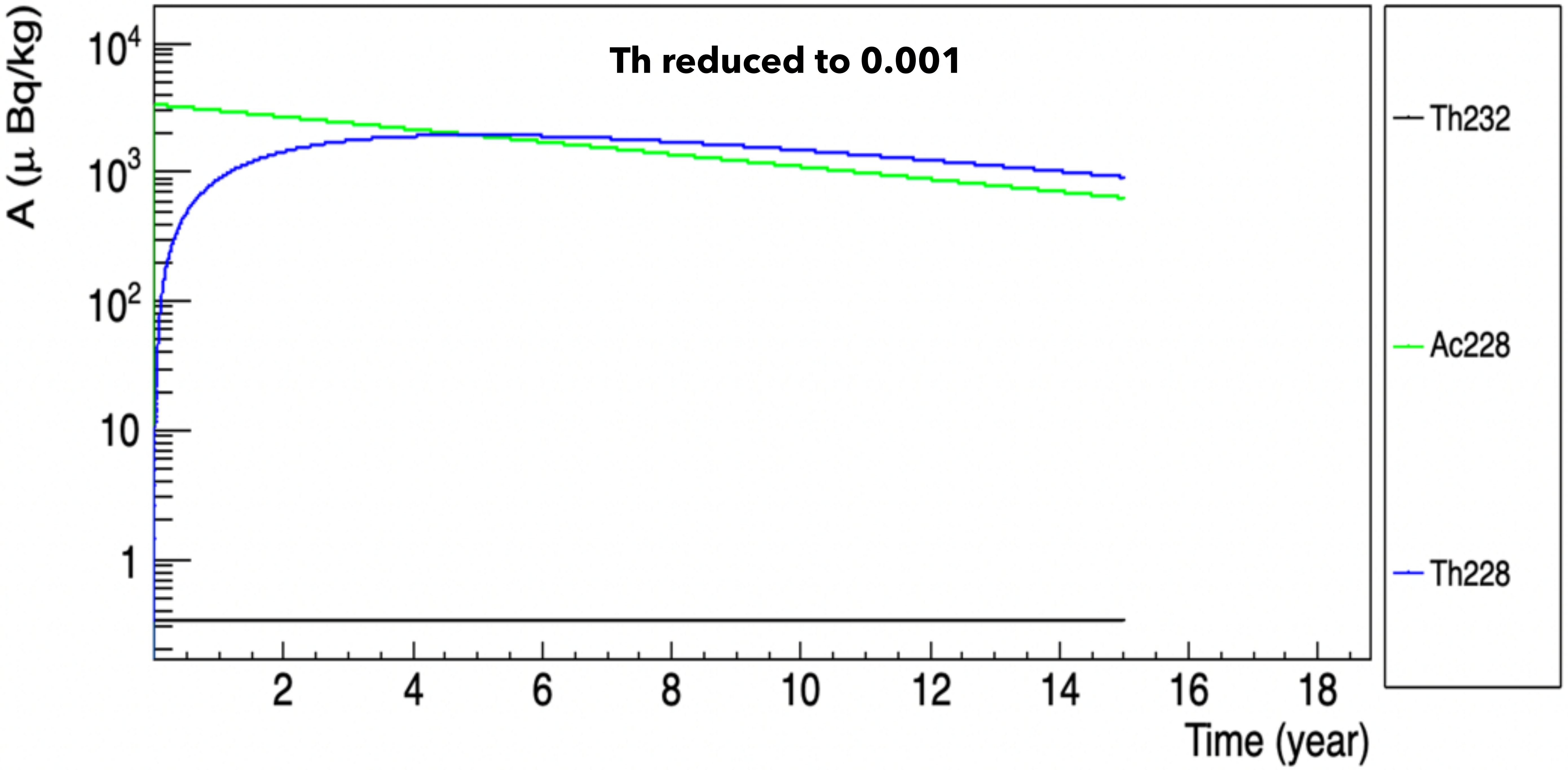
$$A_2(t) = A_2(0)e^{-\lambda_2 t} + A_1(0)\lambda_2 t$$



$$A_2(t) \sim A_2(0)(1 - \lambda_2 t) + A_1(0)\lambda_2 t \;= A_2(0) + [A_1(0) - A_2(0)]\lambda_2 t$$

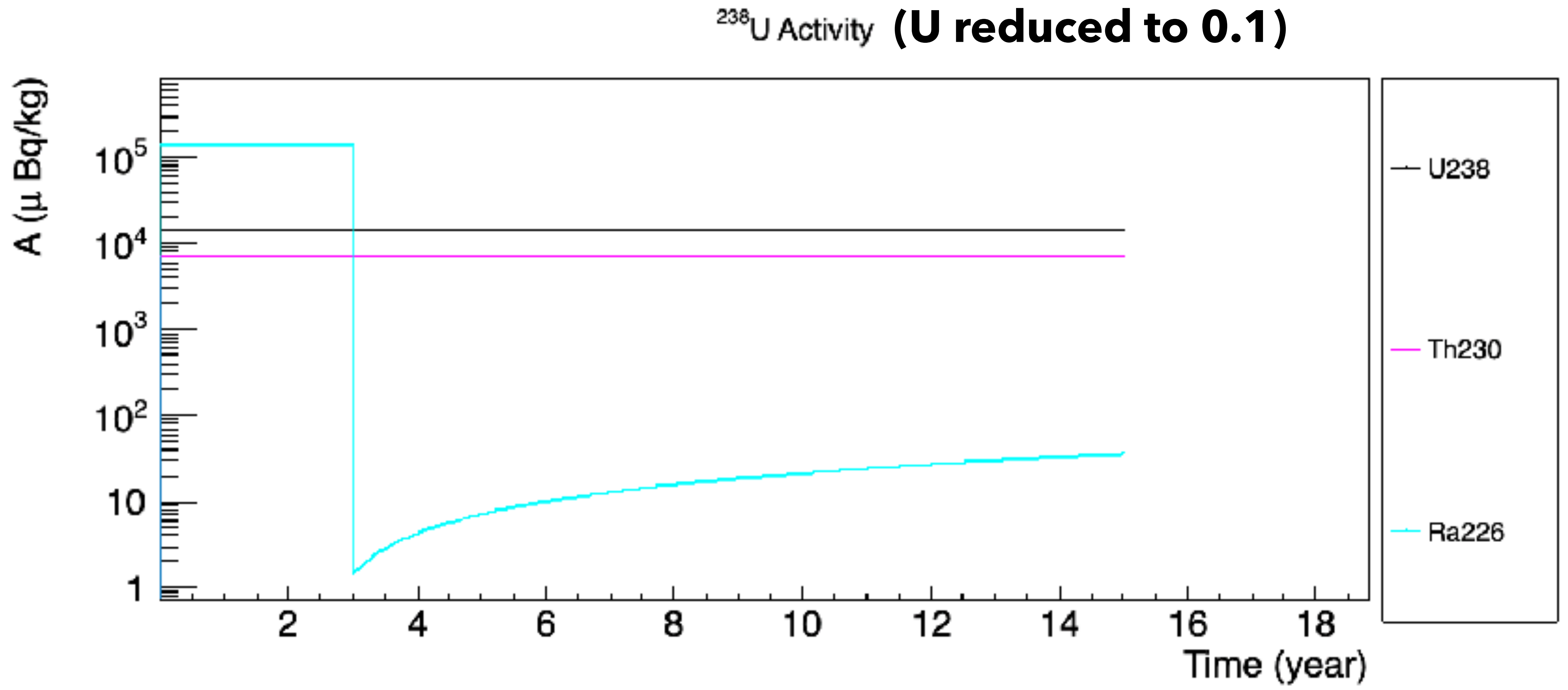
- By substituting the values of ^{230}Th and ^{226}Ra, it can be seen that **the lower the initial ^{226}Ra value** after purification, the more **rapidly it regrows** over time.

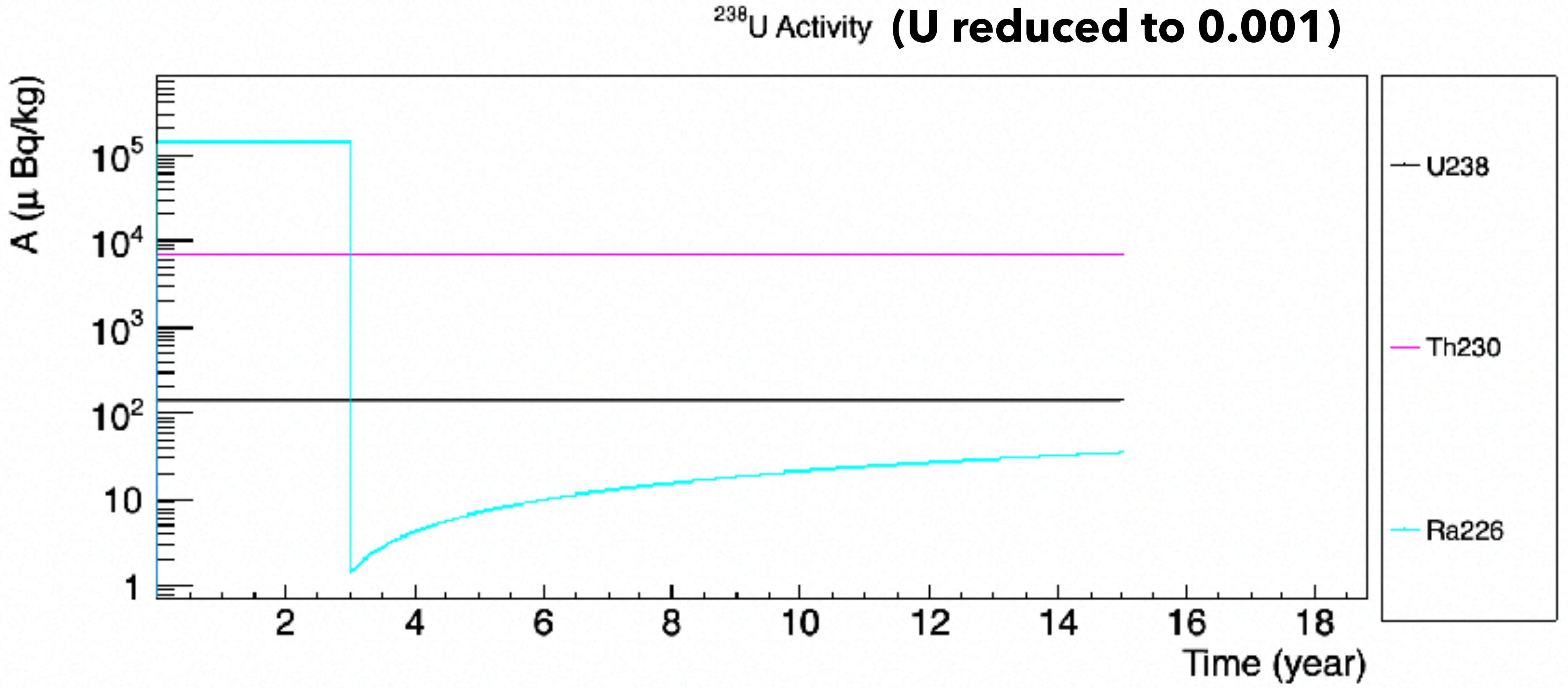
How to fit the data

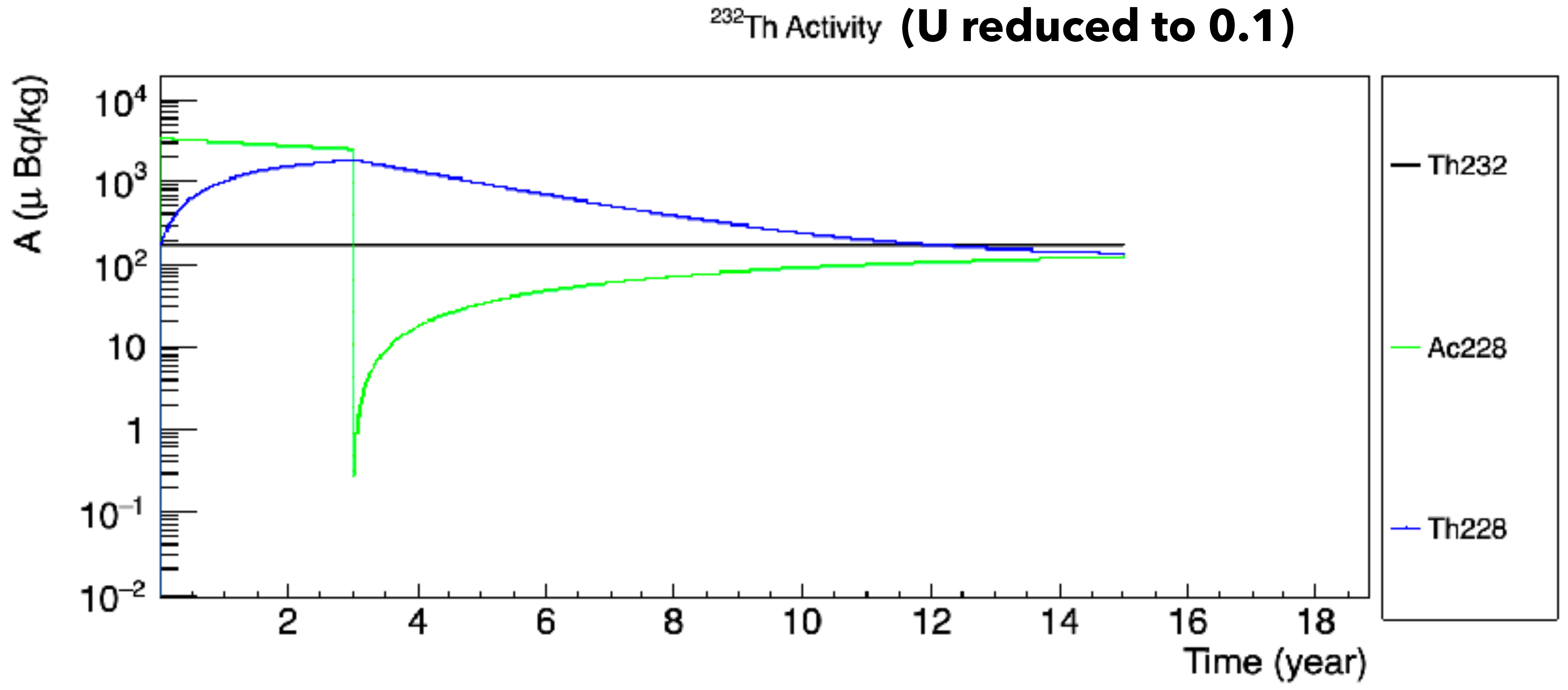


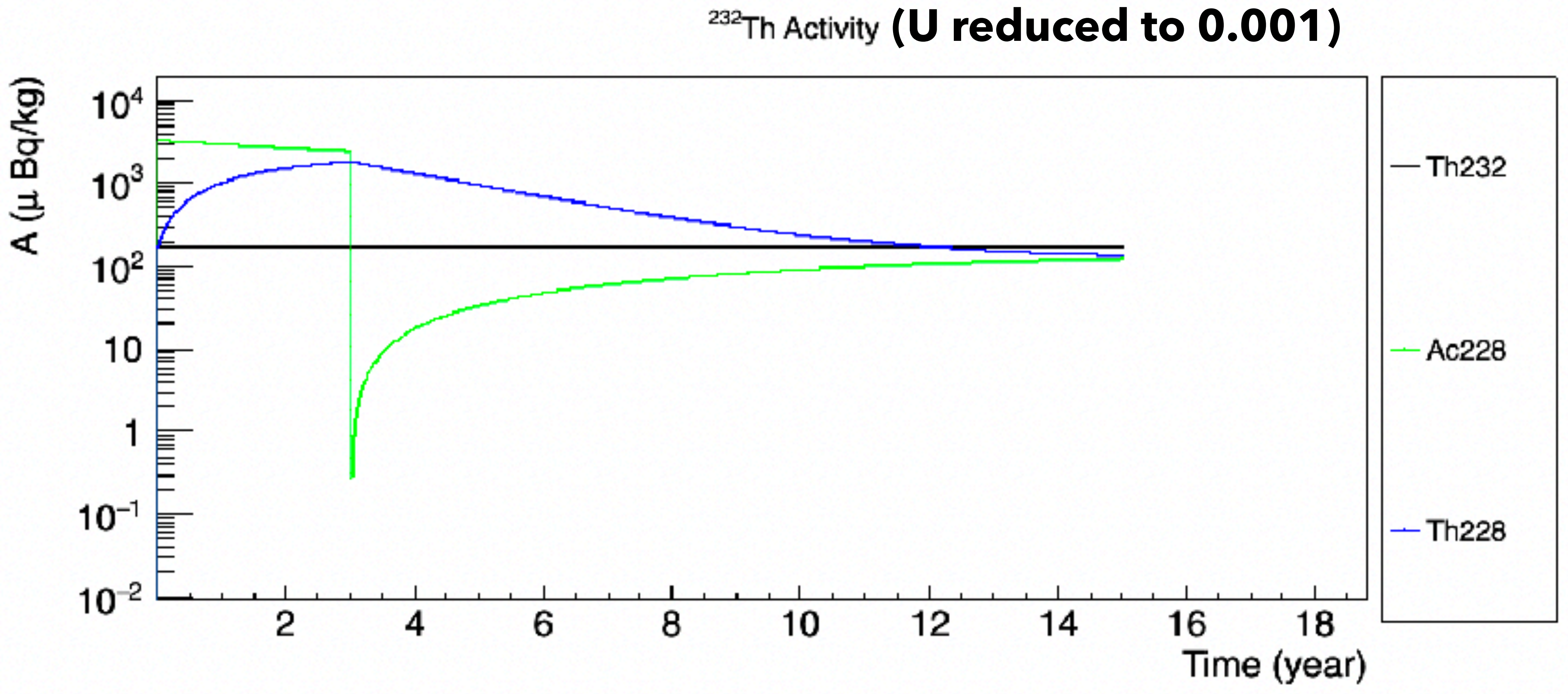
- When Th is reduced, ^{228}Th increase from ^{228}Ra decay while ^{232}Th stay reduced.
- When ^{228}Th activity increased sufficiently, reduce Ra !
- **Multiple reductions can fit the data**.

U reduced : Increasing ratio of ^{226}Ra is same

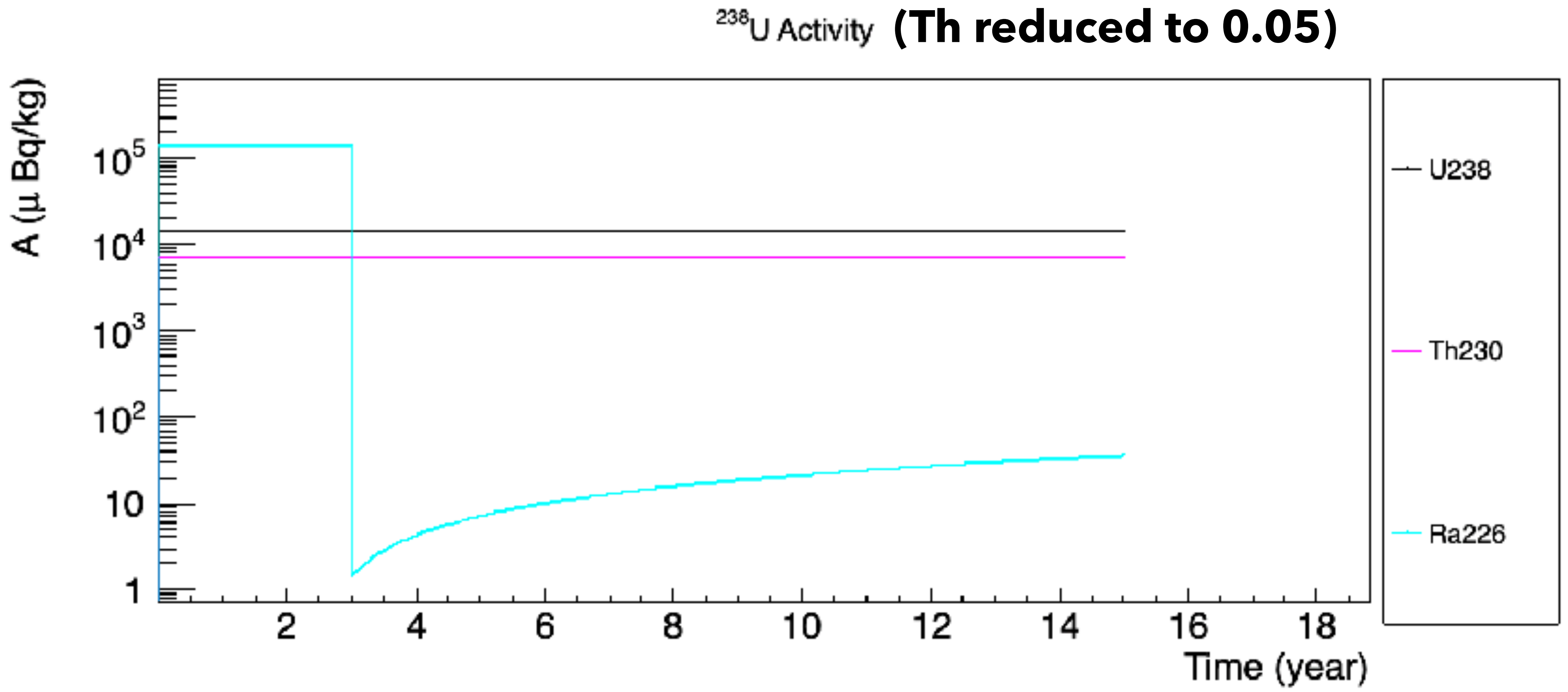


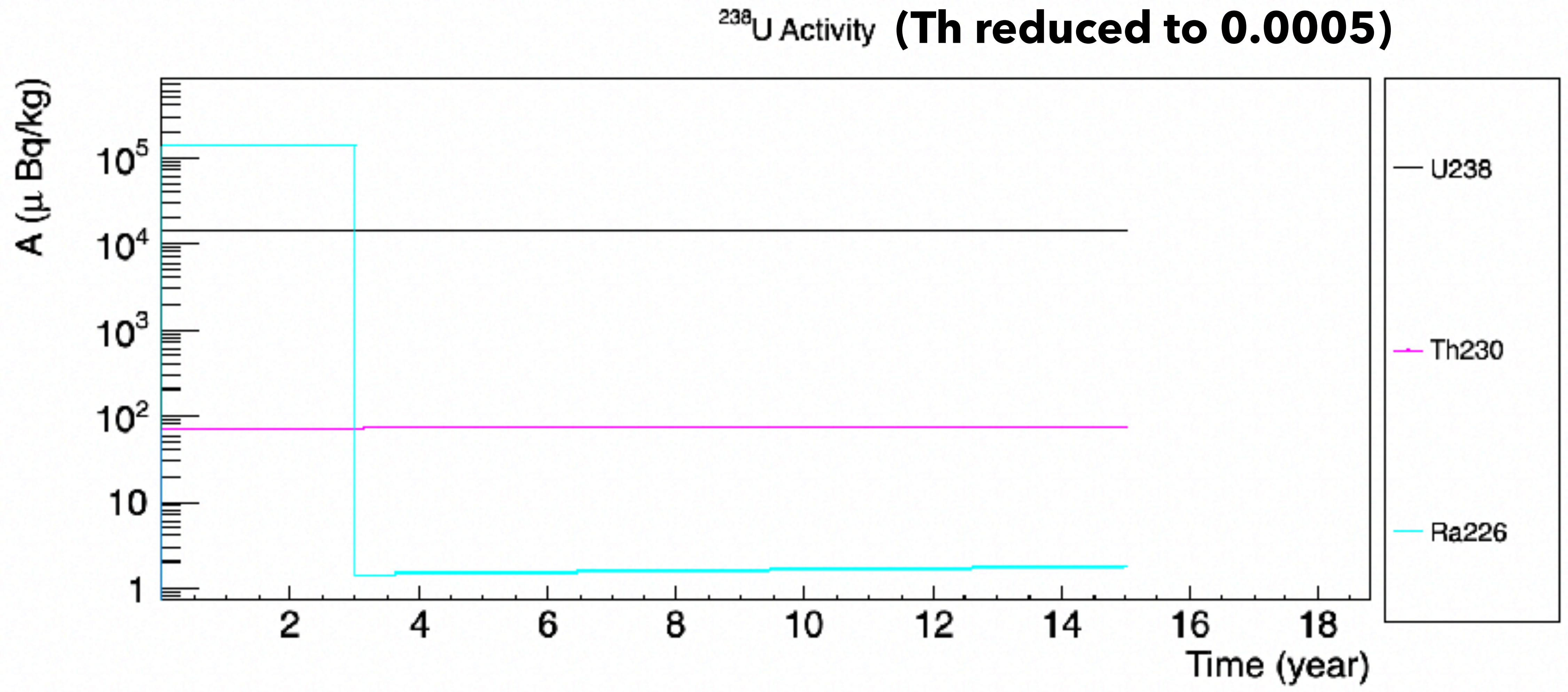


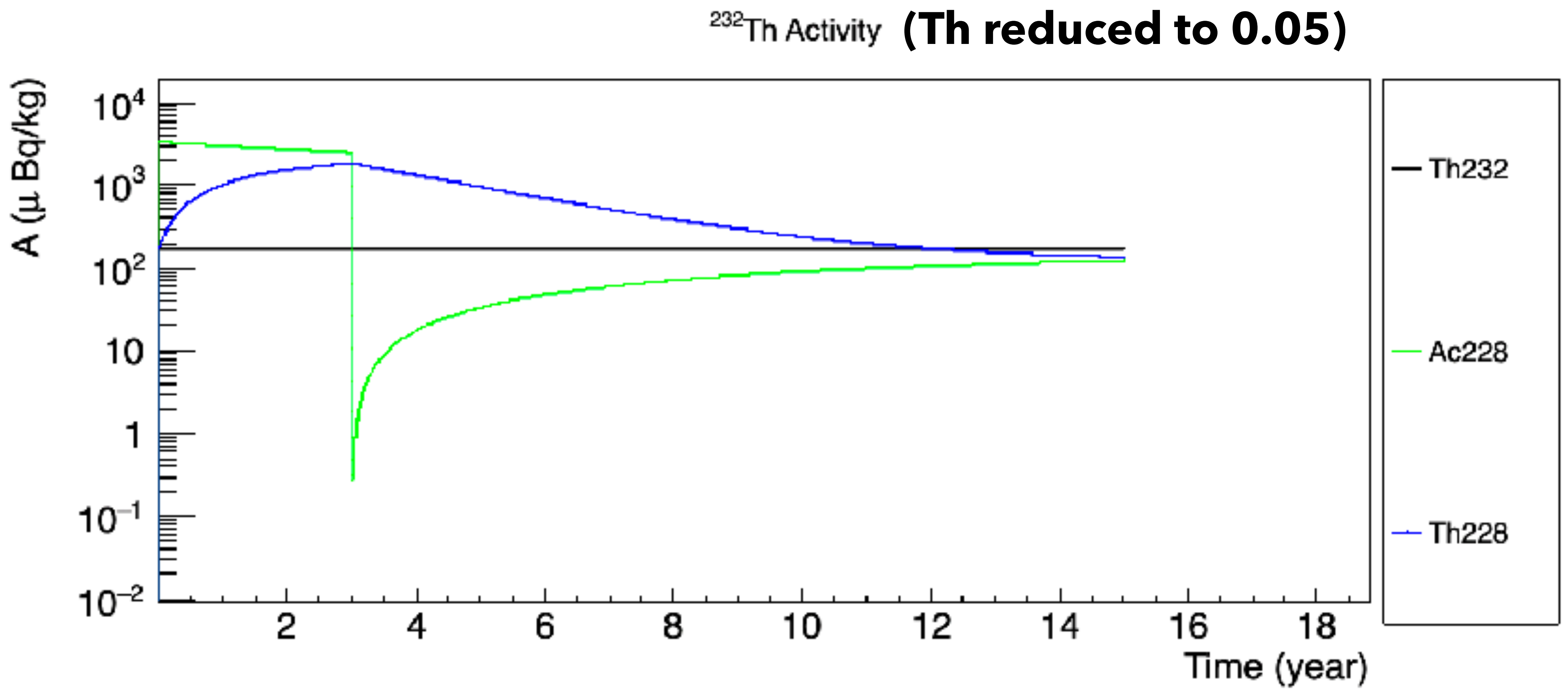


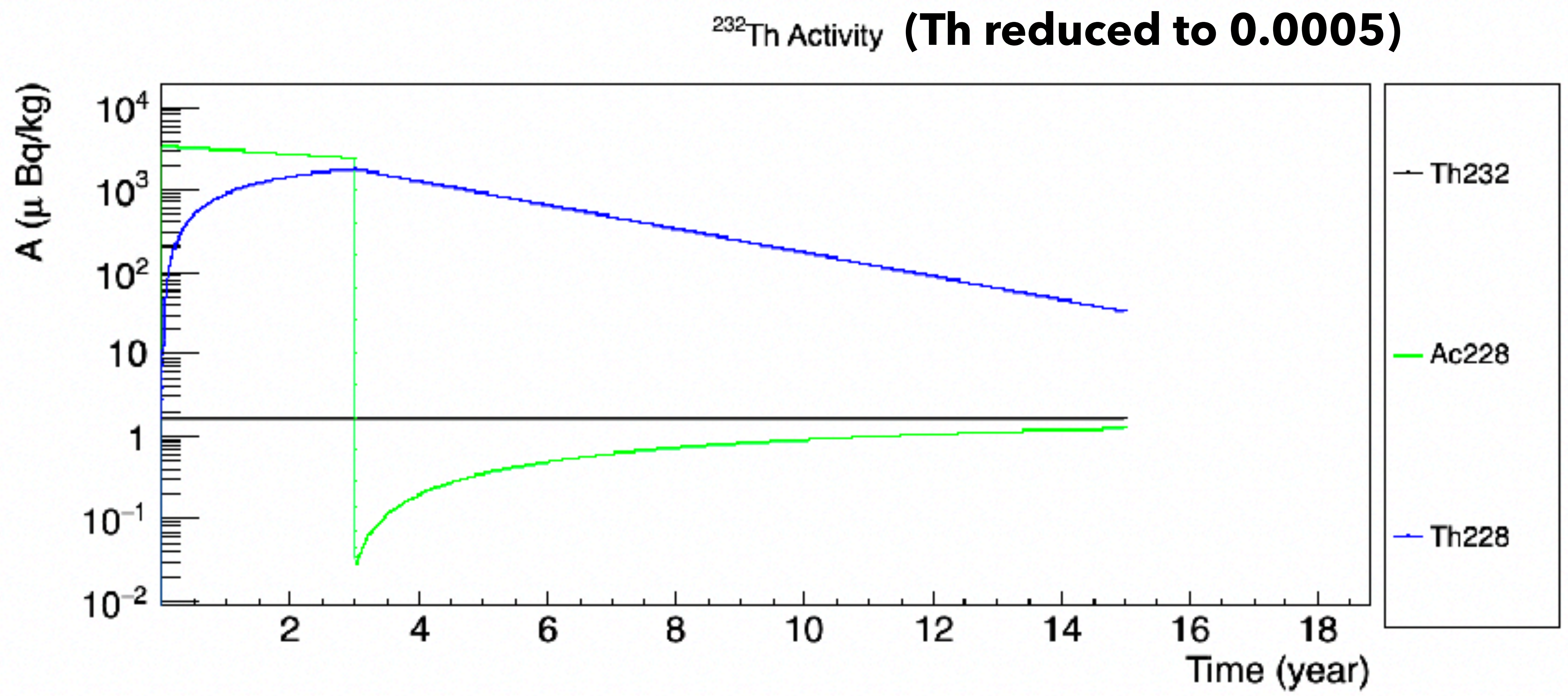


Th reduced : Increasing ratio of ^{226}Ra is changed









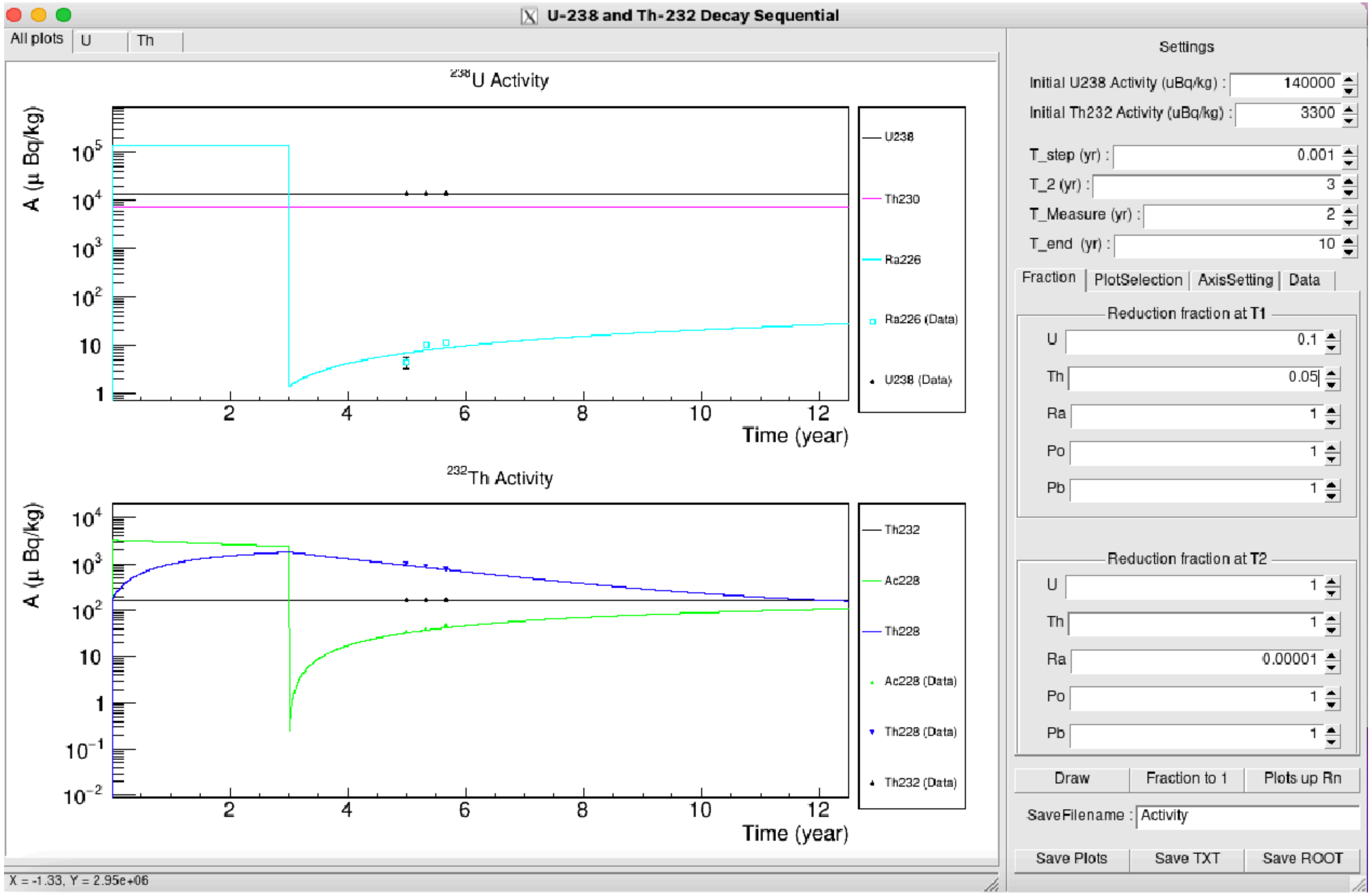
Measurement data over time of Al sample

^{234}Th	^{234m}Pa	$^{226}Ra(^{238}U)$	^{40}K	^{228}Ac	^{228}Th	Sample name	Measured time
25.2 ± 1.3 Bq	21.1 ± 1.3 Bq	< 8.31	< 32.61	55.6 ± 7.3	1523 ± 78	Al supporter	2021.5.7
21.9 ± 1.1 Bq	22.5 ± 1.3 Bq	337.4 ± 18.0	36.9 ± 14.6	159.0 ± 11.4	480.9 ± 25.2		2025.1.31
21.4 ± 1.1 Bq	22.8 ± 1.3	22.0 ± 2.6	23.9 ± 8.5	162.6 ± 10.3	429.6 ± 22.2		2025.8.13
13.6 ± 0.7 Bq	14.6 ± 0.8 Bq	4.42 ± 1.14	22.24 ± 5.79	35.84 ± 3.12	976.56 ± 49.13	Al top plate	2024.10.30
12.9 ± 0.7 Bq	13.8 ± 0.7 Bq	10.41 ± 1.35	28.69 ± 7.11	39.51 ± 3.28	861.30 ± 43.40		2025.2.25
13.2 ± 0.7 Bq	14.5 ± 0.8 Bq	11.69 ± 1.30	25.74 ± 5.32	48.32 ± 3.44	778.18 ± 39.15		2025.7.01
U : 1070 ppb (13.3 Bq/kg)				Th : 42.5 ppb (172 mBq/kg)			2025.02

- Huge differences of ^{238}U vs ^{226}Ra, ^{228}Ac vs ^{228}Th shown
- Activity of ^{228}Ac is smaller than that of ^{228}Th
- Activity of ^{228}Th is higher than that of ^{232}Th

A typical case is shown here.

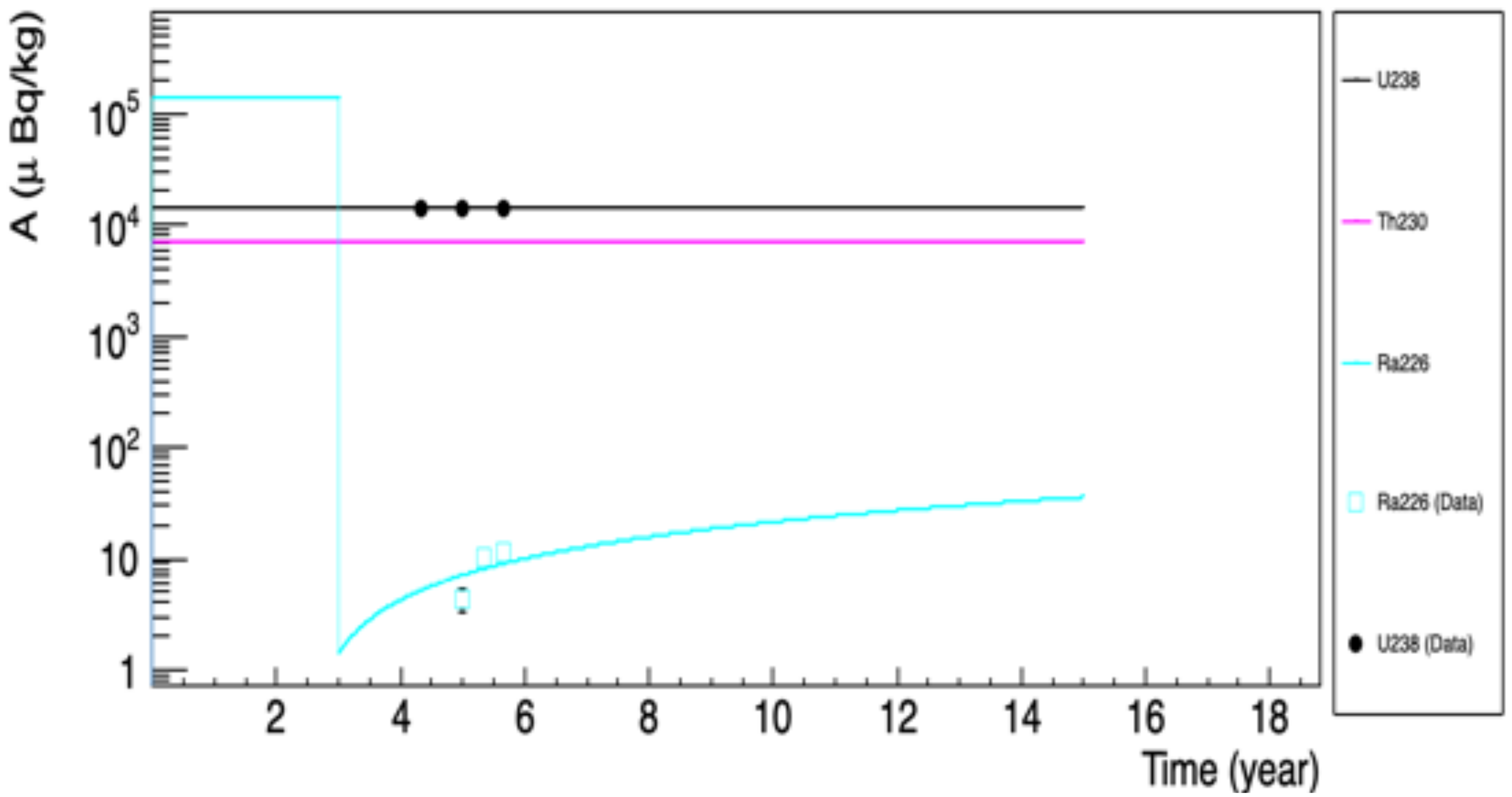
Calculation Bateman Eq. Program in root



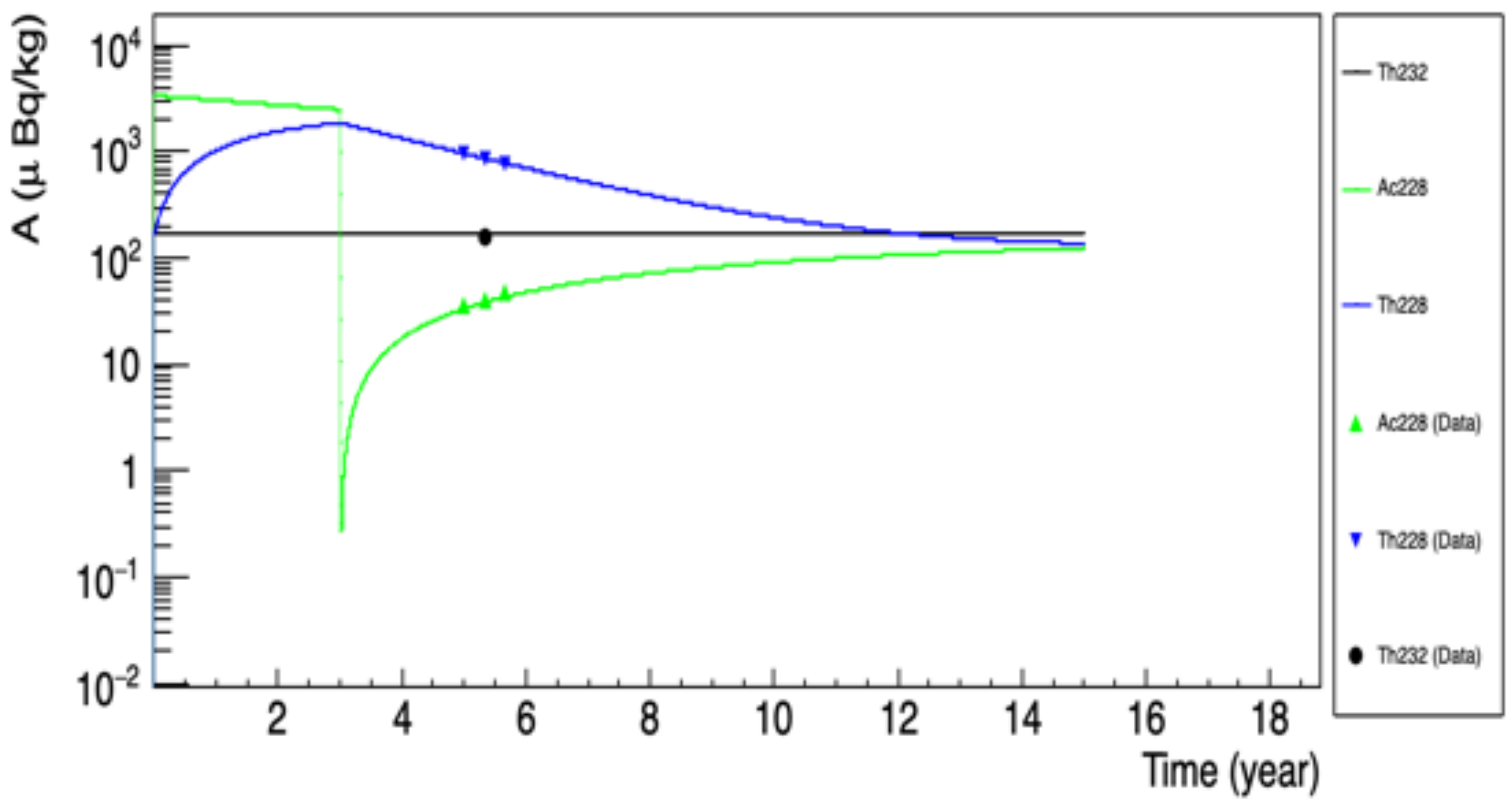
- Initial U238 &Th232 Activity : Activity of equilibrium state
- T_step : 1st purifying date
- T_2 : 2nd purifying date. (After 1st purifying)
- T_Measure : measure time (after 2nd purifying)
- T_end : y-axis range
- Fraction : put number on T1, T2 separately
- PlotSelection : drawing marked isotopes
- AxisSetting : linear or logarithmic on X and Y axes
- Data : put measured data information

Program developed by Dr, Youngsoo Yoon (KRISS)

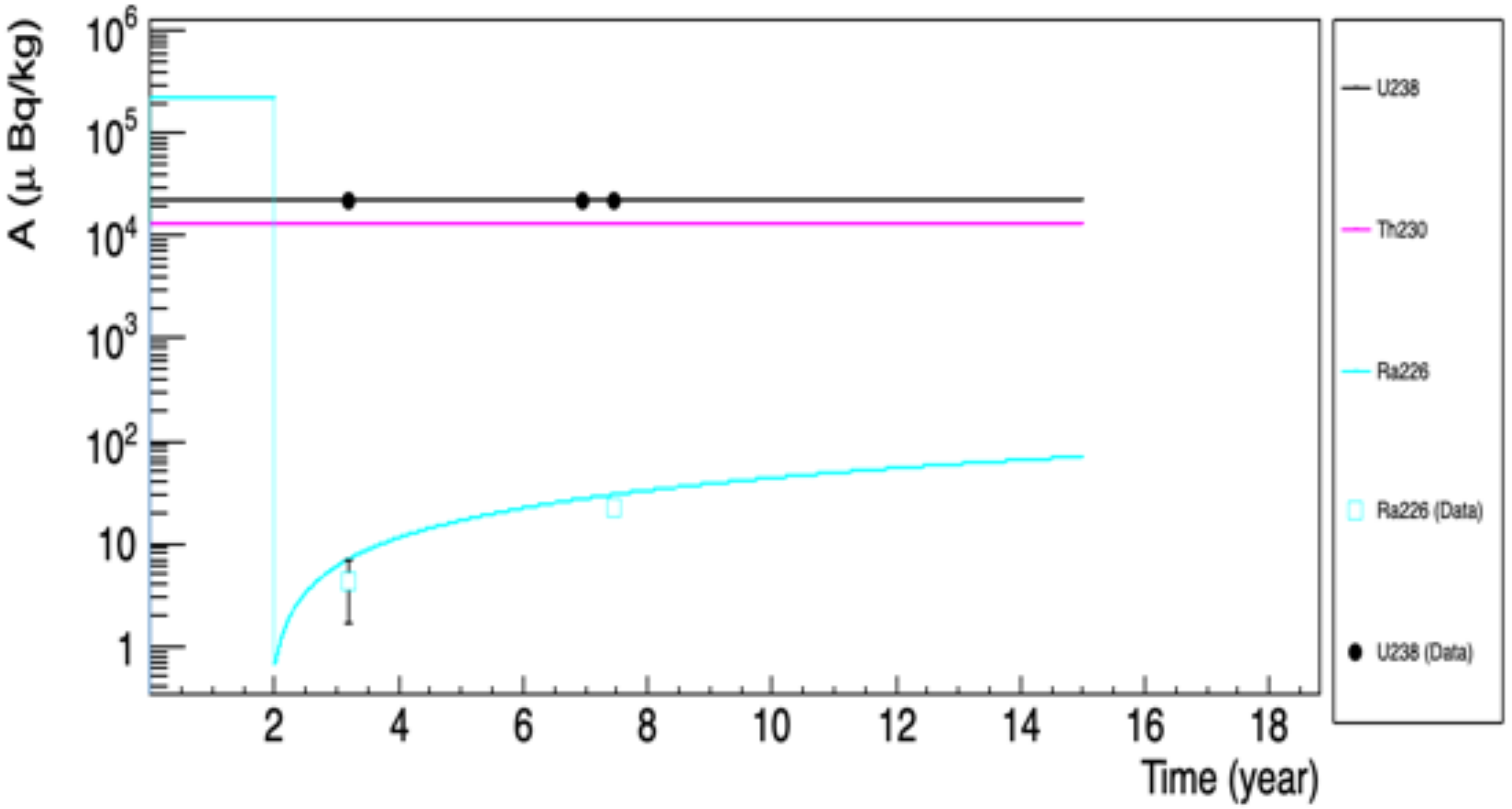
^{238}U Activity of Al top plate



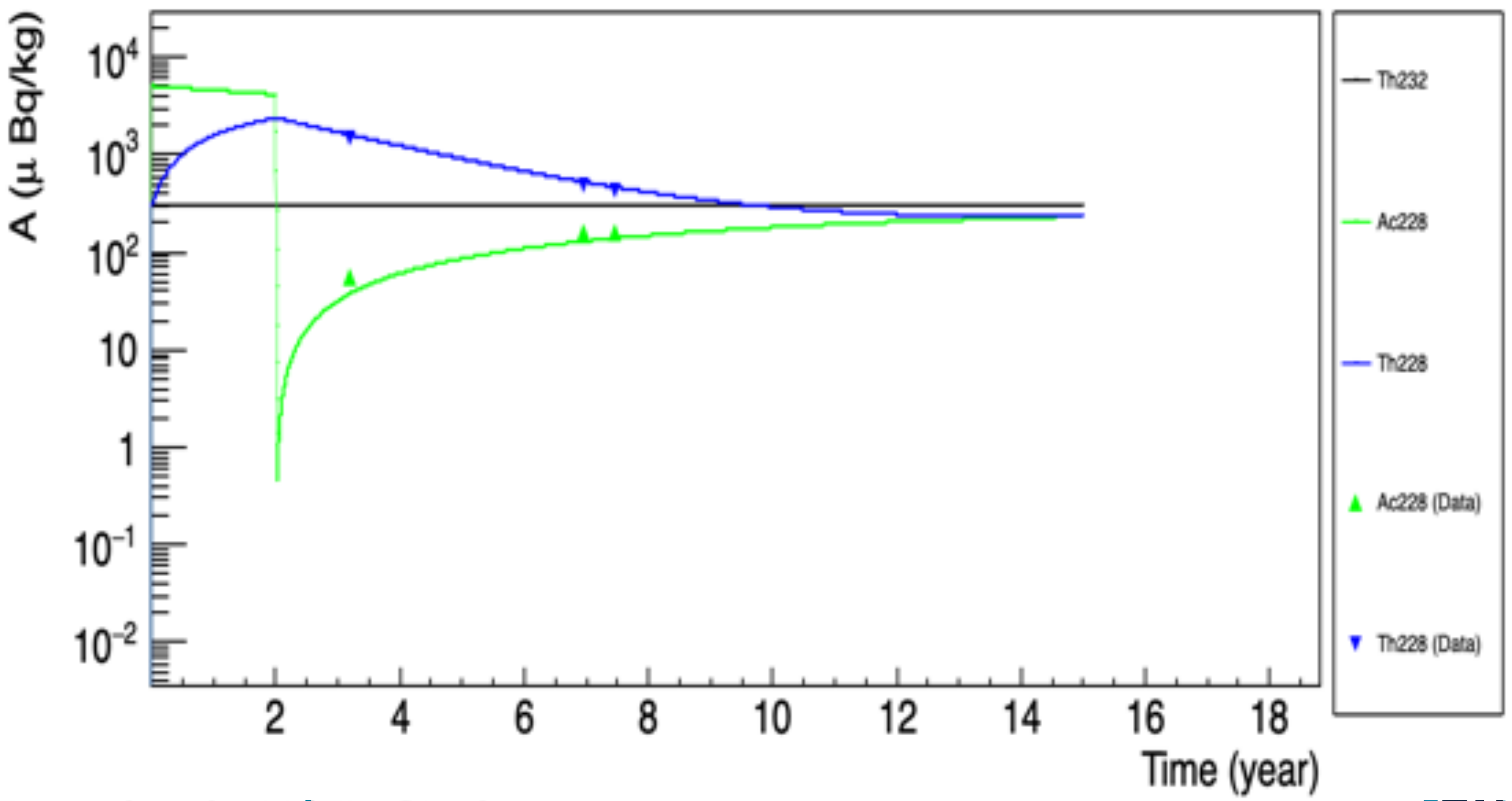
^{232}Th Activity of Al top plate



^{238}U Activity of Al supporter



^{232}Th Activity of Al supporter



Estimation Activities at the start of the AMoRE II

$^{226}Ra(^{238}U)$	^{228}Th	Date	BKG level
4.4	977	2024.10.30	< 0.29(90% C.L)
11.7	778	2025.7.1	
12.2	**700**	**2025.12.31**	

Al top plate

- AMoRE-II will start end of 2025.
- From Bateman Eq. calculating program →
- ^{226}Ra increase, while ^{228}Th decrease
  → meaning the background from **^{226}Ra is underestimated** and that from **^{228}Th is overestimated**.
- **It affects real data signal analysis**.
- We will measure more samples with lower radioactivities than the current samples, and try to model lower radioactivity time evolution. This may turn out to be important for ultimate low background experiments.
- Will measure the current Aluminum sample again in half a year.

Summary

- AMoRE-II background analysis requires **time-evolution studies of disequilibrium radioactivities**.
- Aluminum top plate measurements show **large disequilibrium in U and Th chains**.
- Measurements on ^{234}Pa, ^{226}Ra, ^{232}Th, ^{228}Ac, and ^{228}Th for a long-term time evolution can be explained by a simple model using **Bateman Equation** with a few parameters (initial radioactivities, purification times and ratios, measurement times, etc.).
- This modeling can **predict radioactivity changes** with small uncertainties.
- Although this study used samples with relatively high radioactivity, the same time-evolution mechanism applies. We will extend this study to purer materials such as MoO_3 powder, copper, etc.

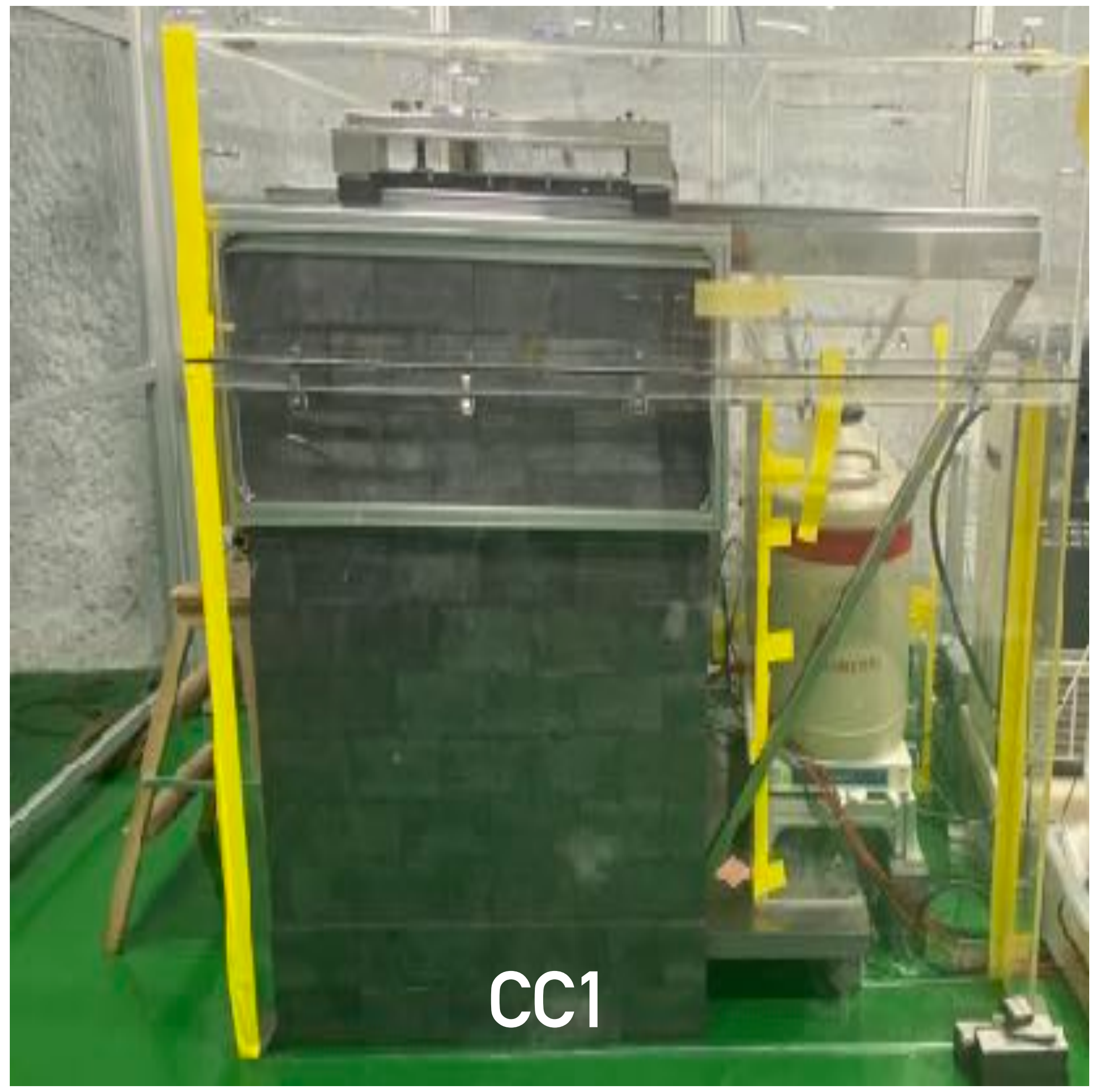
Back up

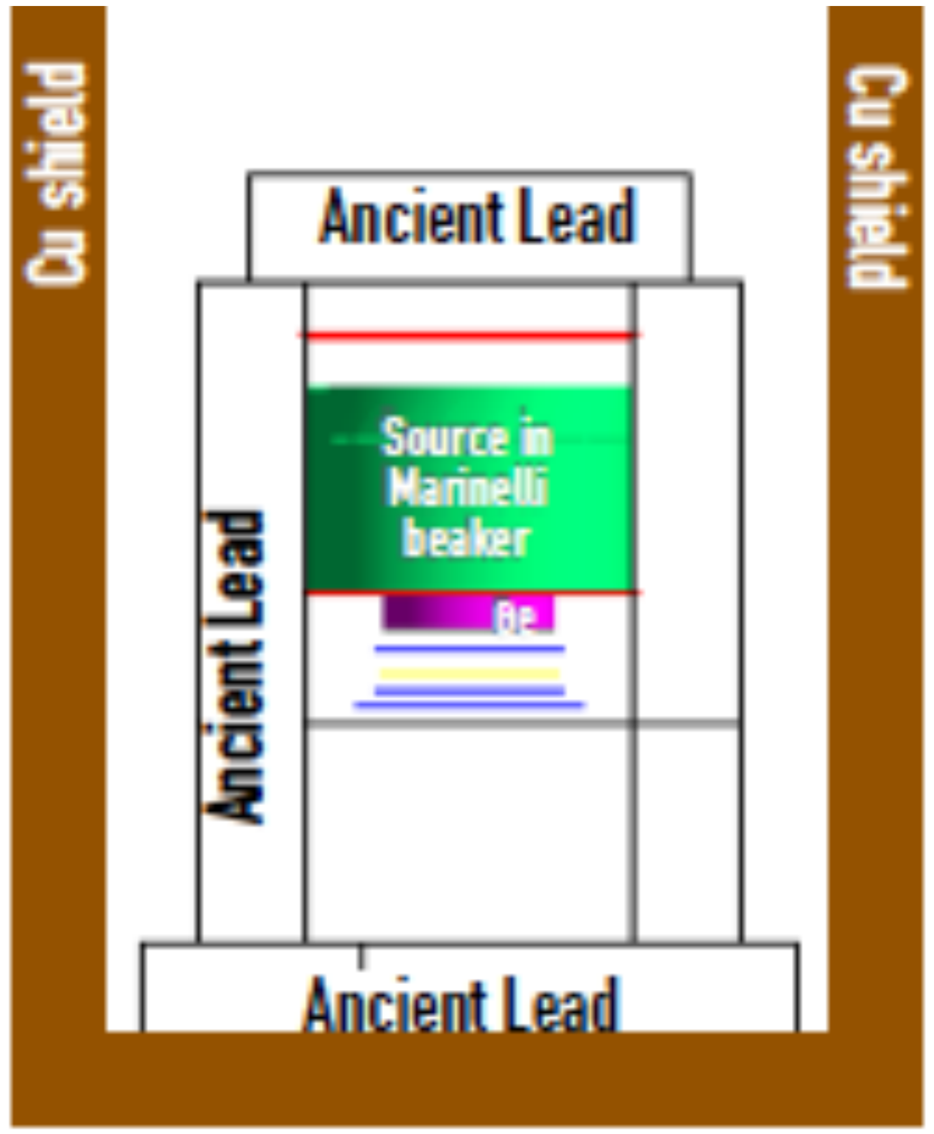
HPGe detector in Yemilab

CC1

- 100% HPGe Canberra
- Dedicated Shielding System
  Top & bottom: Pb (15 cm)+Cu (10 cm)
  Side: Pb (15 cm)+Cu (10 cm)
  Inner : Ukraine Ancient lead 5 cm
- Net count rate of background (50~4000 keV)

mHz	2015	2018	2019	2020	2021	2022	2024
> 50 keV	8.1	7.9	7.8	7.9	8.0	8.2	8.0~11





CC2

- 100% HPGe Canberra
- Dedicated Shielding System
  General Pb(15 cm) + Goslar Pb(5 cm) +
  Cu(10 cm)
- Net count rate of background (50~3500 keV)

mHz	2017	2018	2019	2020	2021	2022	2024	2025
> 50 keV	9.5	7.9	6.8	6.2	6.1	6.1	6.2	6.3