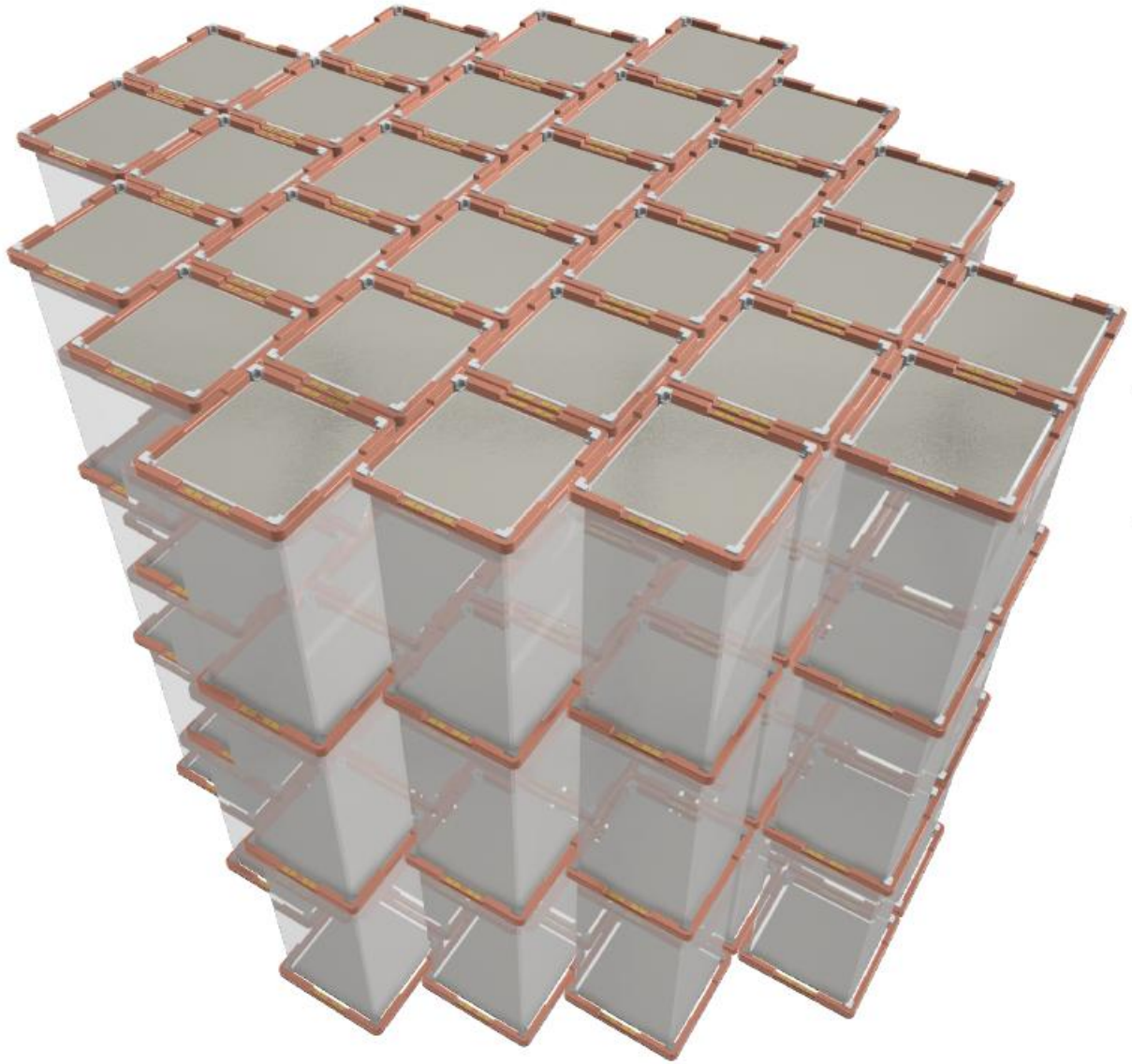
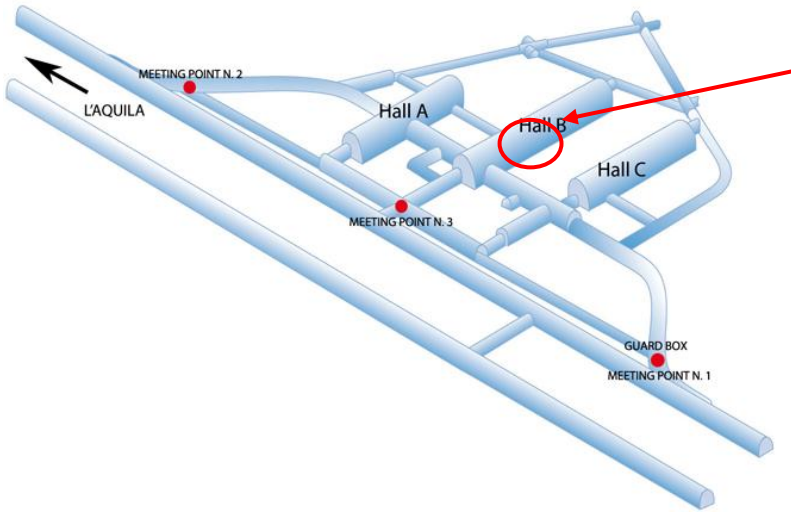
Rapid low-background radioassay techniques for ^{210}Pb and ^{3}H in materials for rare-events searches

The RES-NOVA detector

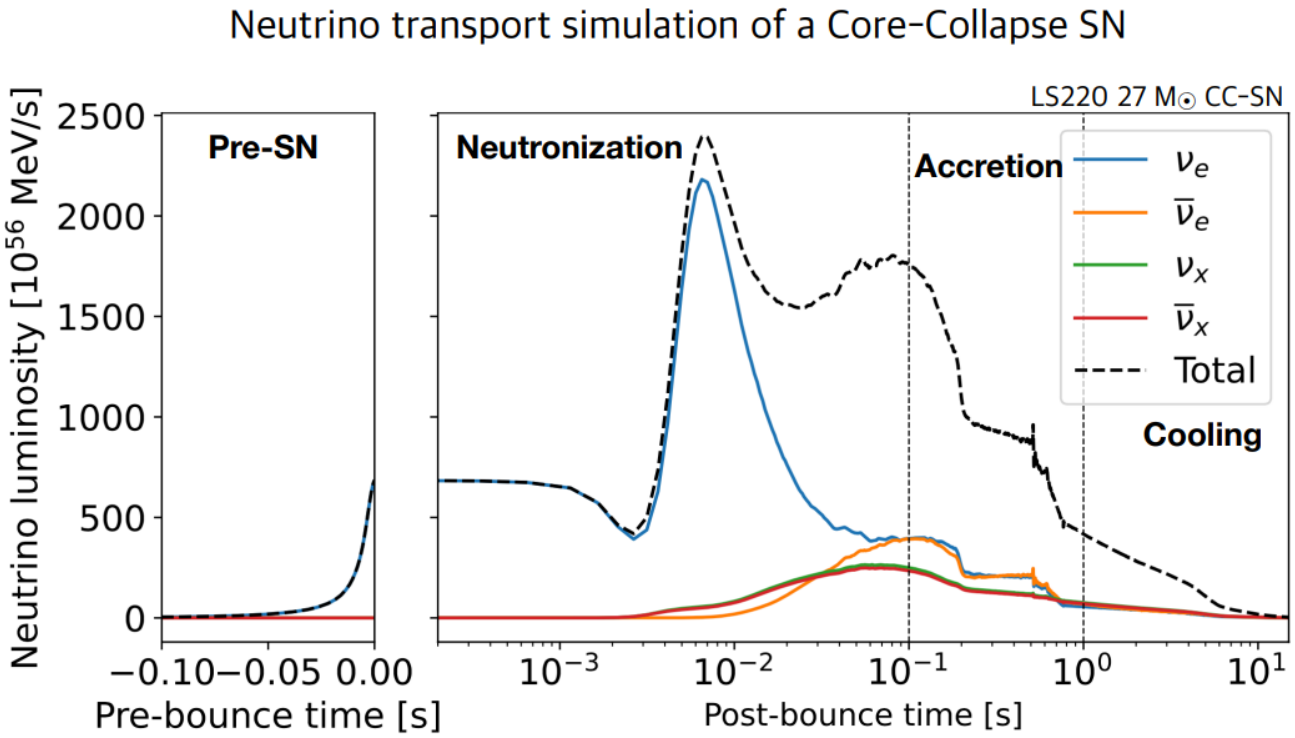


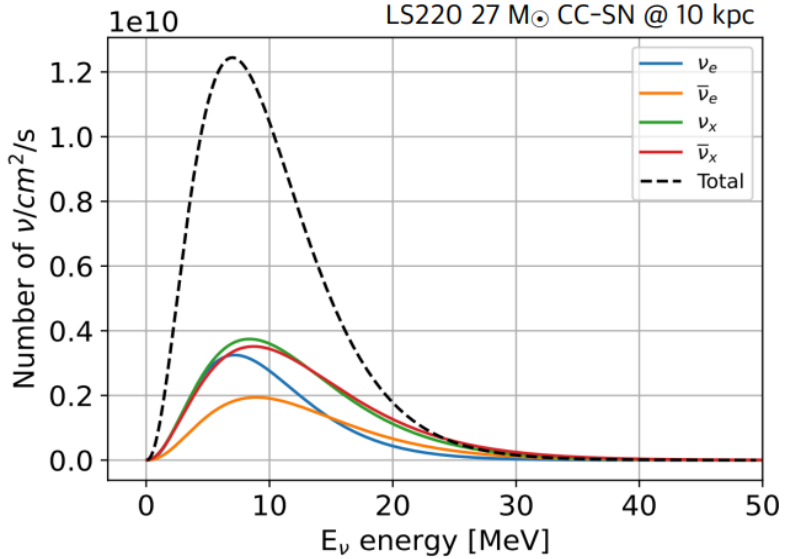
- Modular detector
- Total target mass: 170 Kg
- Cryogenic detector: 10 mK
- Energy threshold: 1 keV
- Archaeological lead target ($PbWO_4$)
- Main scientific purpose:
  - Observation of neutrinos (all flavors!) from Supernovae bursts.
- Secondary scientific purposes:
  - Observation of Dark Matter candidates (WIMPS)
  - Observation of solar neutrinos

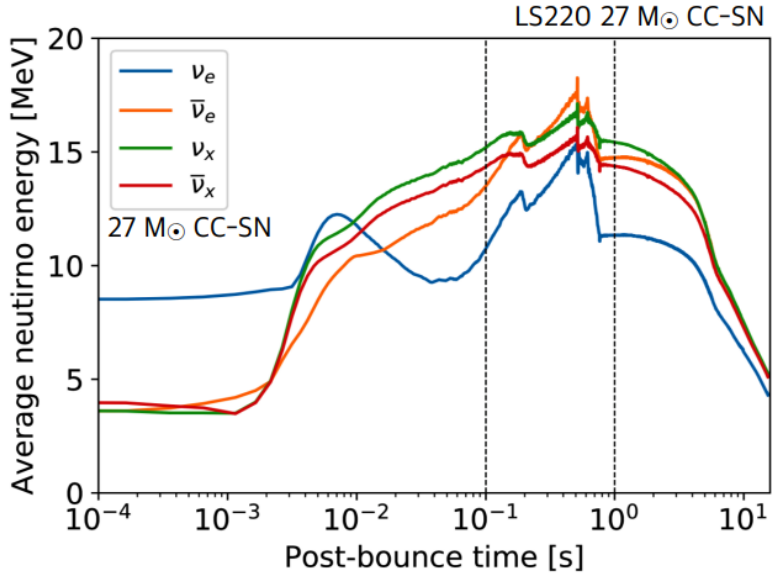


RES-NOVA will be hosted @ LNGS Hall B

Supernova burst





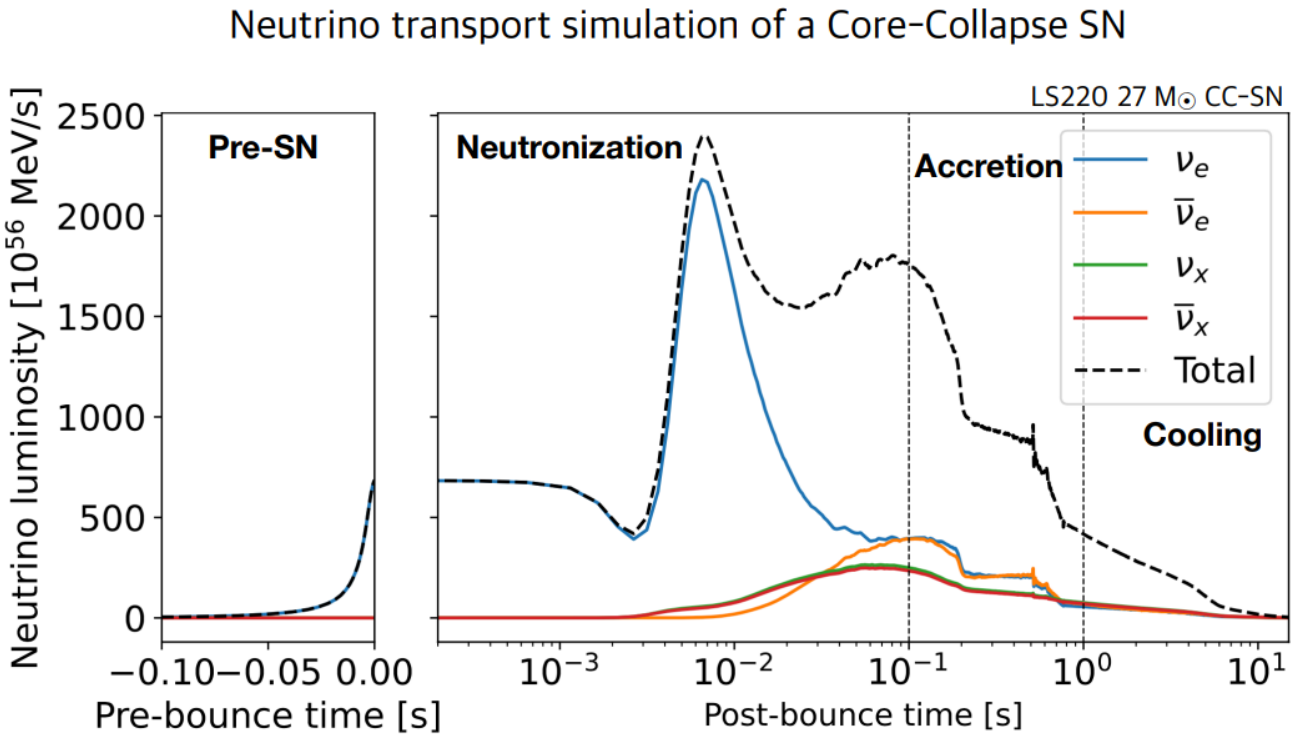


- Supernova: high energy explosion of massive star
- ~90% star binding energy converted in all flavor neutrinos
- ~10% in GW and EM

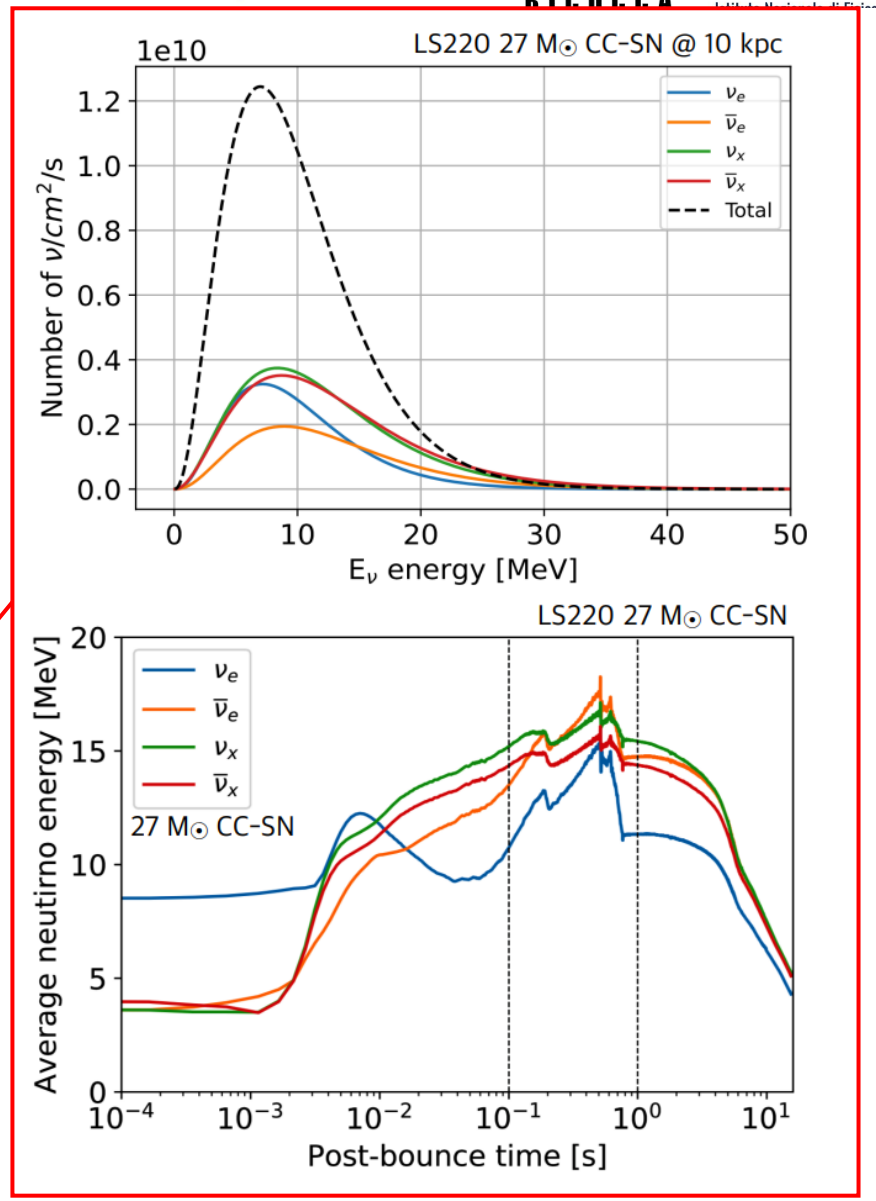
NEURTINOS ARE DIRECT PROBES AND MESSENGERS OF SNe HIDDEN DYNAMICS

A. Mirizzi et al., Riv. Nuovo Cim.39, 1 (2016) [ArXiv:1508.00785]

Supernova burst



SN NEUTRINO DETECTORS NEED ALL FLAVOR SENSITIVITY!

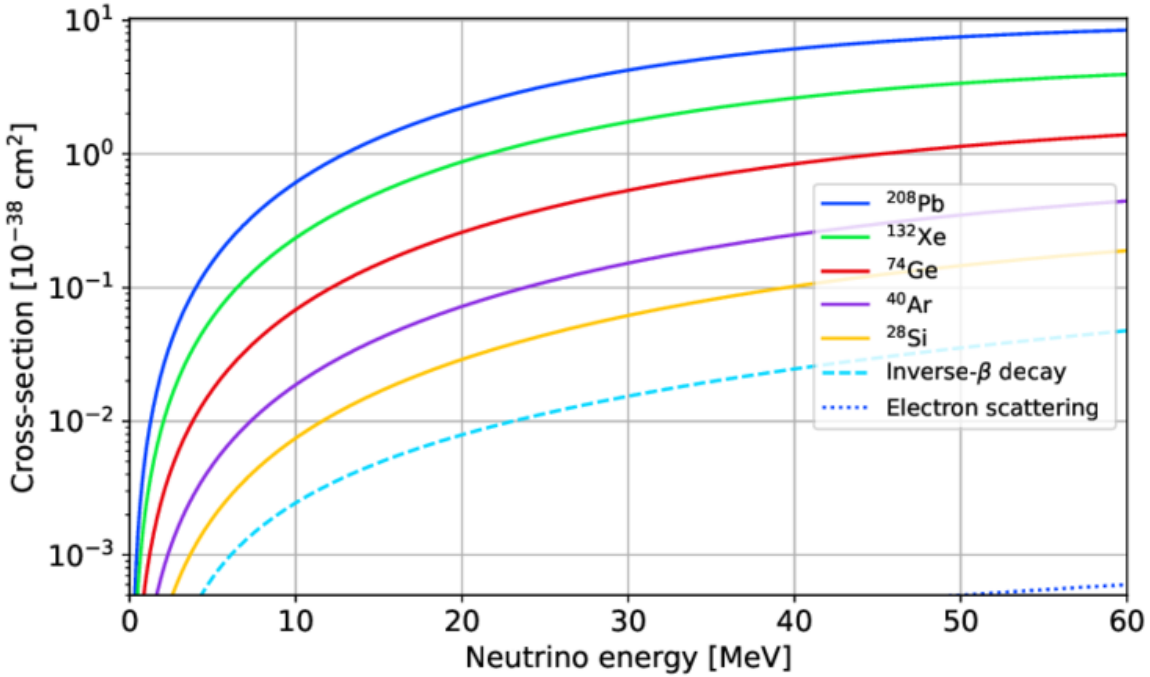


A. Mirizzi et al., Riv. Nuovo Cim.39, 1 (2016) [ArXiv:1508.00785]

CEνNS

Coherent Elastic Neutrino-Nucleus Scattering

L. Pattavina, N. Ferreiro Iachellini and I. Tamborra, *Neutrino observatory based on archaeological lead*, *Phys. Rev. D* **102** (2020) 063001 [arXiv:2004.06936] [INSPIRE]

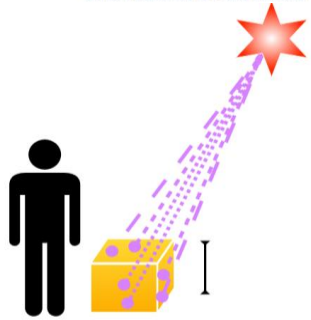


$$\sigma_{CE\upsilon NS} = \frac{G_F^2}{4\pi} F^2(q^2) E_\upsilon^2 Q_W^2$$

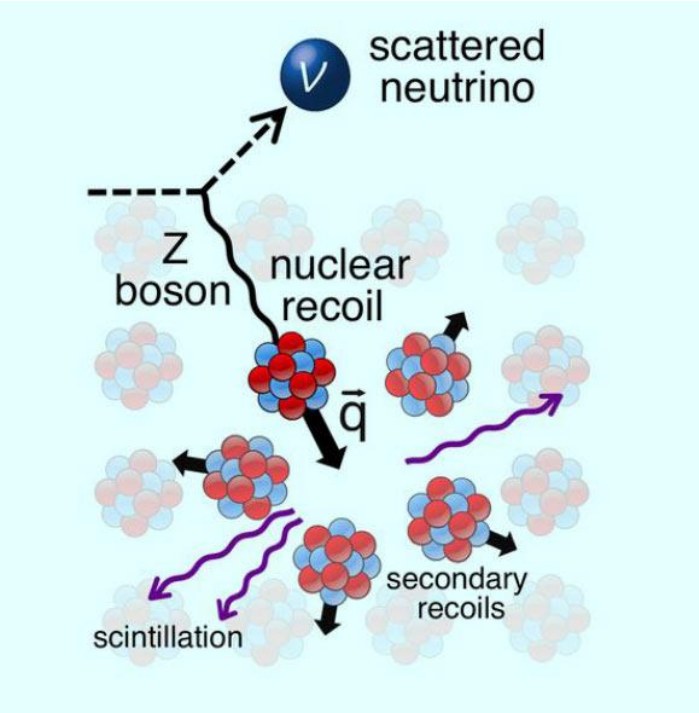
$$Q_W = \mathrm{N} - \mathrm{Z}(1 - 4sin^2\theta_w)$$

$$\sigma_{CE\upsilon NS} \alpha N^2$$

Lead cross-section 2 orders of magnitude bigger than IBD



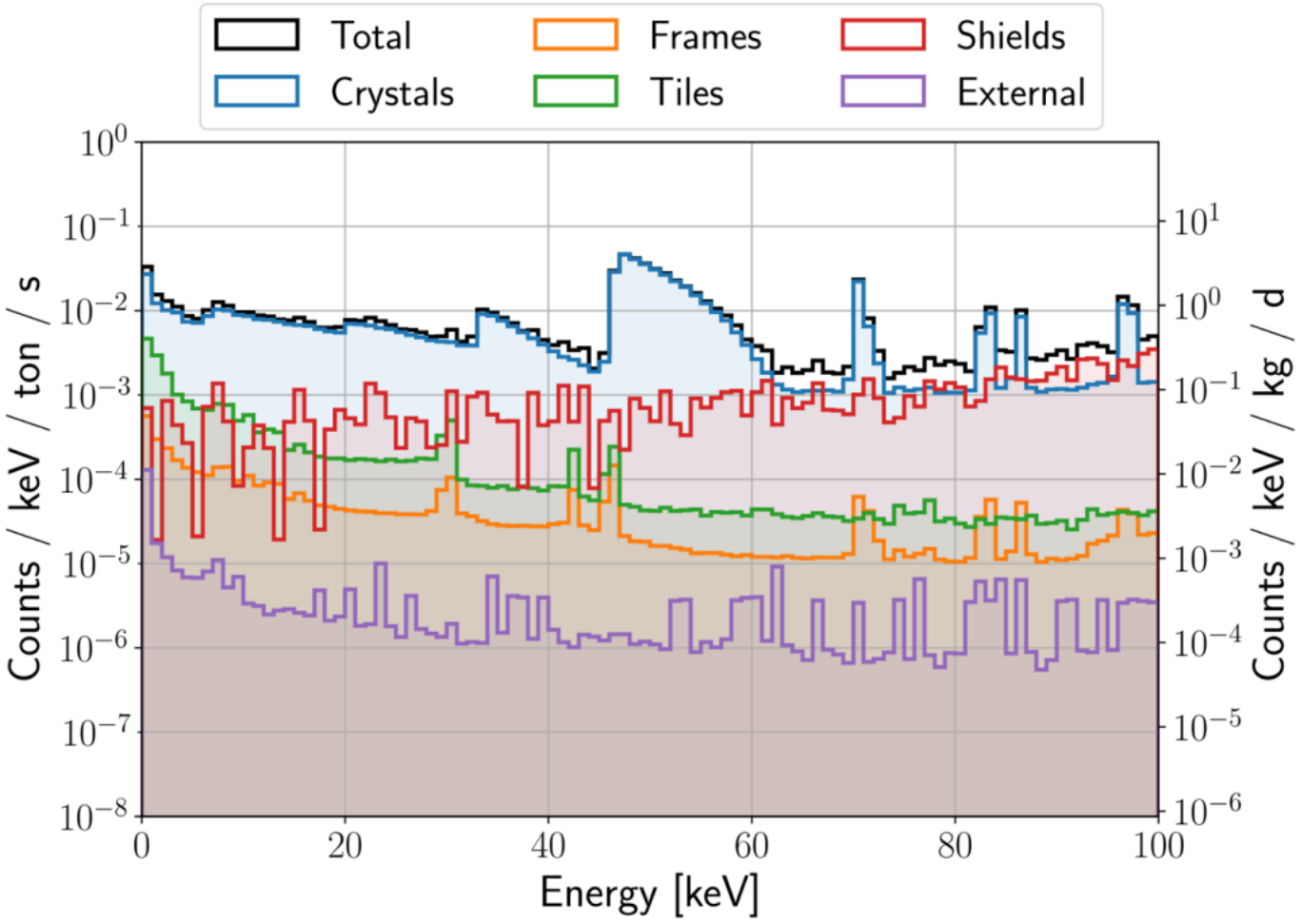
Tabletop detector!



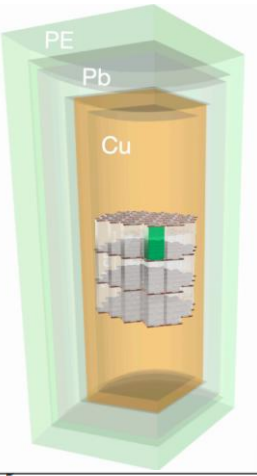
- Lead in active volume!
- Low background needed
- **Archaeological lead**

RES-NOVA background

Total detector energy response

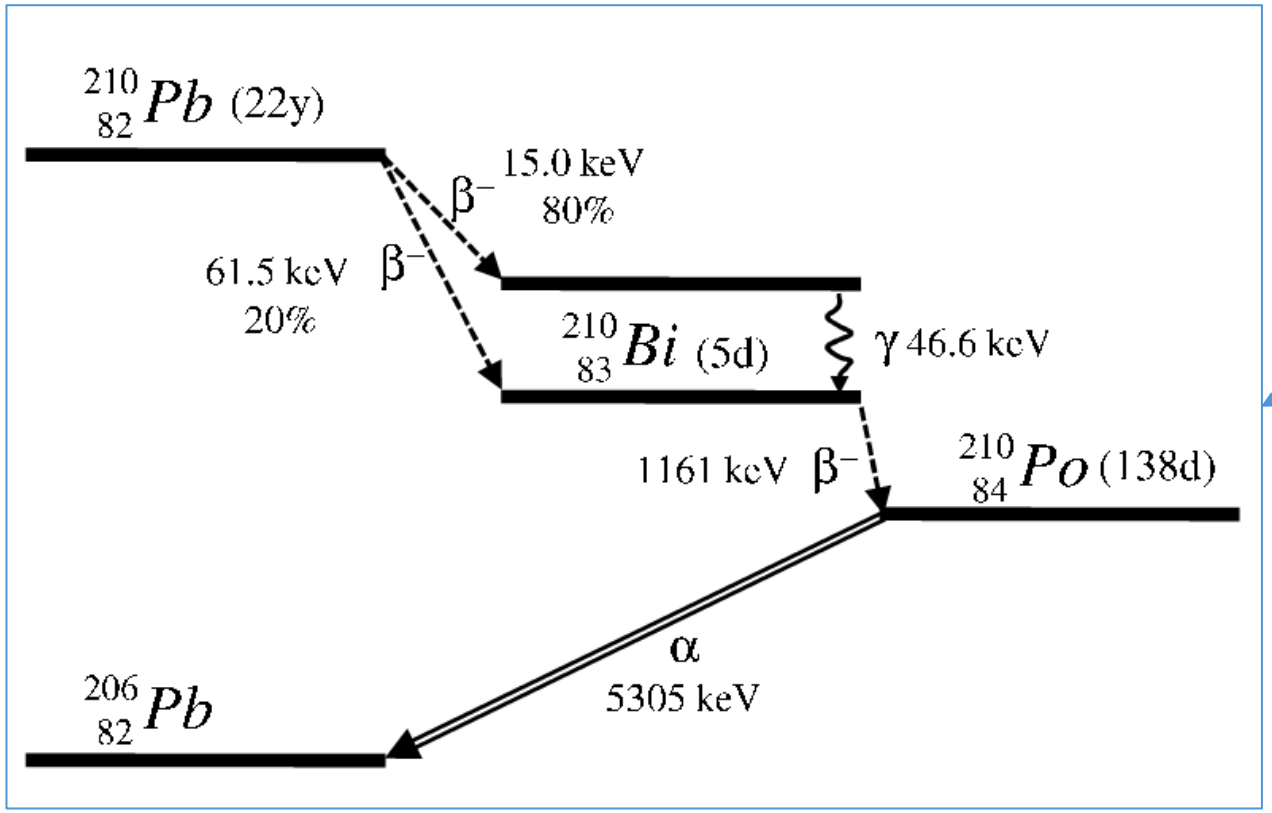


Location	Component	Source	Activity [mBq/kg] ([mBq/cm^2])
Detector	$PbWO_4$	^{232}Th	$< 2.3 \times 10^{-1}$
		^{238}U	$< 7.0 \times 10^{-2}$
		^{210}Pb	$< 7.1 \times 10^{-1}$
		^{40}K	$< 9.0 \times 10^{-2}$
	Cu structure	^{232}Th	$< 4.1 \times 10^{-5}$
		^{238}U	$< 1.7 \times 10^{-5}$
	PTFE holders	^{232}Th	$< 6.1 \times 10^{-3}$
		^{238}U	$< 2.2 \times 10^{-2}$
Shields	Polyethylene	^{232}Th	$< 9.4 \times 10^{-2}$
		^{238}U	< 0.23
	Pb shield	^{232}Th	< 2.3
		^{238}U	< 3.5
		^{210}Pb	3×10^{5}
	Cu shield	^{232}Th	$< 4.1 \times 10^{-2}$
		^{238}U	$< 1.7 \times 10^{-2}$
Surface	$PbWO_4$	^{210}Pb - 10 μm	7.4×10^{-6}
		^{210}Pb - 0.01 μm	8.7×10^{-5}
	Cu structure/Tiles PTFE holders	^{232}Th - 10 μm	$(1.2 \pm 0.1) \times 10^{-5}$
		^{238}U - 10 μm	$(8.3 \pm 0.7) \times 10^{-6}$
		^{210}Pb - 10 μm	1.2×10^{-4}
		^{232}Th - 0.01 μm	$(1.4 \pm 0.1) \times 10^{-6}$
		^{238}U - 0.01 μm	$(1.2 \pm 0.1) \times 10^{-6}$
		^{210}Pb - 0.01 μm	$(4.1 \pm 0.1) \times 10^{-4}$
	Cu shield	^{210}Pb - 0.01 μm	1×10^{-2}
External		neutrons	3.9×10^{-6} cm^{-2}s^{-1}
		muons	3.1×10^{-8} cm^{-2}s^{-1}
		gammas	4.0×10^{-1} cm^{-2}s^{-1}



RES-NOVA Coll. New dark matter direct search based on archaeological Pb, Phys.Rev.D 111 (2025) 10, 103050

RES-NOVA background



All the decay chain in secular equilibrium with half-life of 22 y

^{210}Pb is a natural occurring radioactive material in lead (10^4 Bq/ton for commercial Pb)

Location	Component	Source	Activity [mBq/kg] ([mBq/cm^2])
Detector	$PbWO_4$	^{232}Th	$< 2.3 \times 10^{-1}$
		^{238}U	$< 7.0 \times 10^{-2}$
		^{210}Pb	$< 7.1 \times 10^{-1}$
		^{40}K	$< 9.0 \times 10^{-2}$
	Cu structure	^{232}Th	$< 4.1 \times 10^{-5}$
		^{238}U	$< 1.7 \times 10^{-5}$
	PTFE holders	^{232}Th	$< 6.1 \times 10^{-3}$
		^{238}U	$< 2.2 \times 10^{-2}$
Shields	Polyethylene	^{232}Th	$< 9.4 \times 10^{-2}$
		^{238}U	< 0.23
	Pb shield	^{232}Th	< 2.3
		^{238}U	< 3.5
		^{210}Pb	3×10^5
	Cu shield	^{232}Th	$< 4.1 \times 10^{-2}$
		^{238}U	$< 1.7 \times 10^{-2}$
Surface	$PbWO_4$	^{210}Pb - 10 μm	7.4×10^{-6}
		^{210}Pb - 0.01 μm	8.7×10^{-5}
	Cu structure/Tiles PTFE holders	^{232}Th - 10 μm	$(1.2 \pm 0.1) \times 10^{-5}$
		^{238}U - 10 μm	$(8.3 \pm 0.7) \times 10^{-6}$
		^{210}Pb - 10 μm	1.2×10^{-4}
		^{232}Th - 0.01 μm	$(1.4 \pm 0.1) \times 10^{-6}$
		^{238}U - 0.01 μm	$(1.2 \pm 0.1) \times 10^{-6}$
		^{210}Pb - 0.01 μm	$(4.1 \pm 0.1) \times 10^{-4}$
	Cu shield	^{210}Pb - 0.01 μm	1×10^{-2}
External		neutrons	3.9×10^{-6} cm^{-2}s^{-1}
		muons	3.1×10^{-8} cm^{-2}s^{-1}
		gammas	4.0×10^{-1} cm^{-2}s^{-1}

RES-NOVA Coll. New dark matter direct search based on archaeological Pb, Phys.Rev.D 111 (2025) 10, 103050

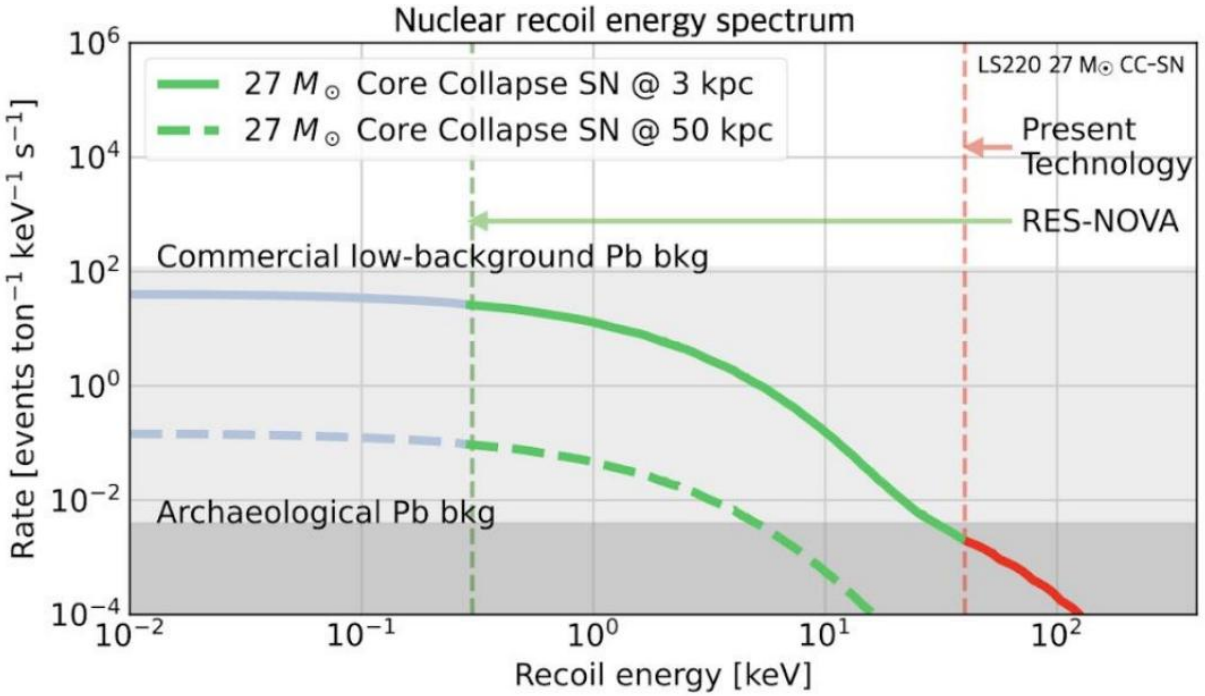
Is there any solution?

Archaeological lead

Archaeological lead samples held captive @ LNGS

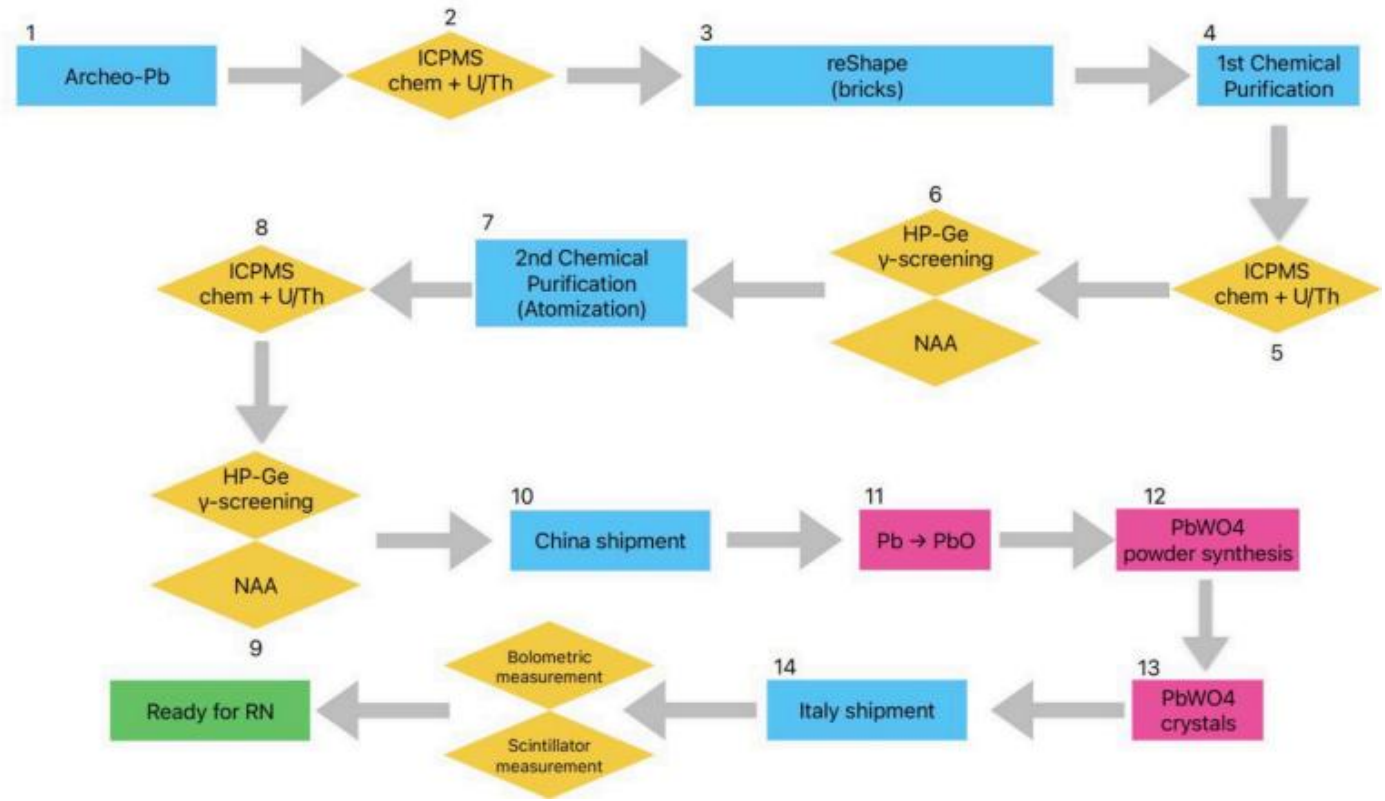
The perks of being stuck in a sunken ship:

- Very old samples! (> 2000 y)
- Natural shielding from cosmogenic activation
- Far “cleaner” (in a radiological way) than normal lead (^{210}Pb expected to be < 1mBq/Kg)
- There is more where they came from!



Archaeological lead

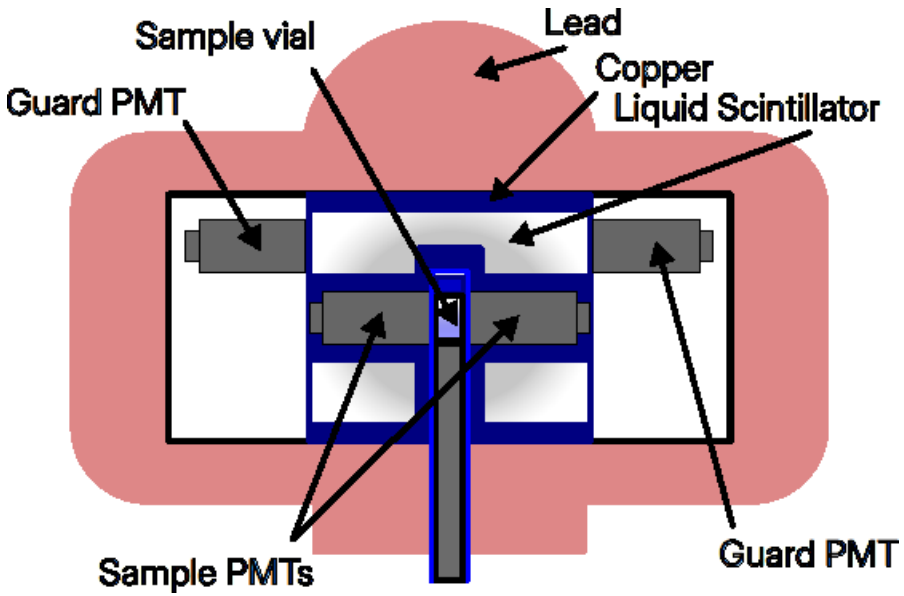
Archaeological lead samples held captive @ LNGS



Chemical purification chain from Pb to $PbWO_4$ crystals

For each step, contamination can be introduced in archaeological lead samples!

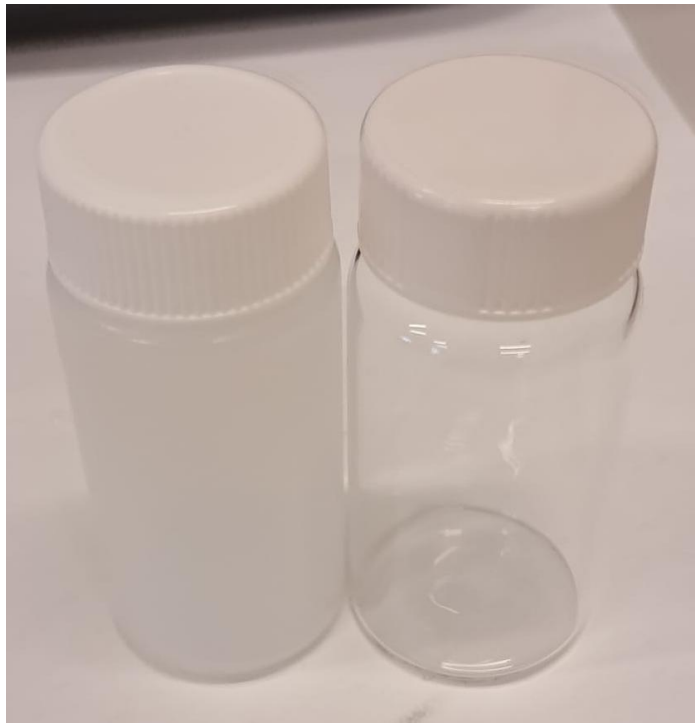
LS detection with Quantulus 1220



Commercial detector!

- Lead casing for cosmic ray protection
- Active veto system
- PMTs for coincidence measurements
- Accepted/rejected spectra splitting
- α/β discrimination via Pulse Shape Analysis (PSA)
- Liquid Scintillator Counting (LSC)
- Cherenkov Counting

Set-up for a ^{210}Pb LSC campaign



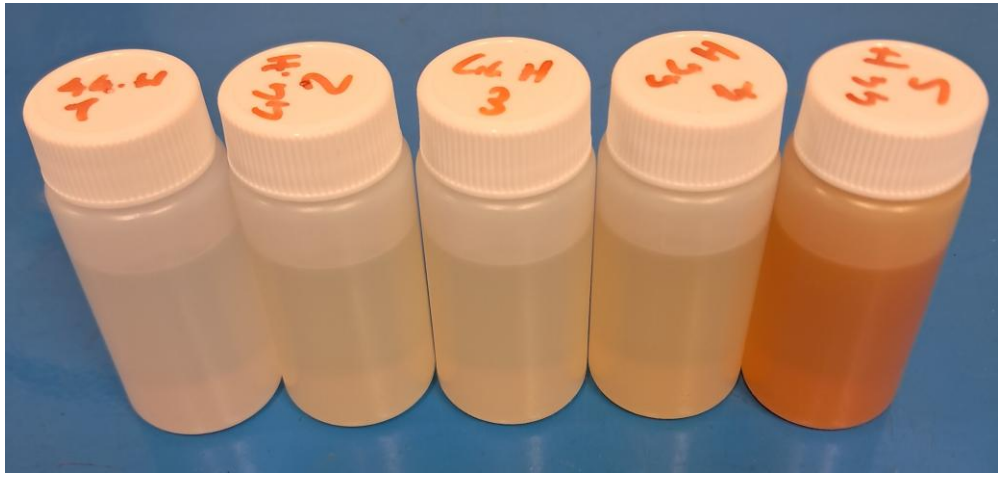
2 Quantulus vials made of PTFE (left) or glass (right)

1. Finding the optimal detection set-up
2. Finding the optimal PSA set-up to allow correct α/β discrimination
3. Calibration curve with sample spiked with known activity of ^{210}Pb to identify ROIs and check detector linearity.
4. Samples measurements.

WHAT IS A QUANTULUS SAMPLE?
20 ml vials containing a cocktail of mixed LS and liquid samples.
Usual LS to sample ratio are:
15 ml LS + 5 ml liquid sample
12 ml LS + 8 ml liquid sample (the one we used)

Set-up for a ^{210}Pb LSC campaign

Each measurement campaign requires a new calibration process, since chemical conditions of samples may strongly differ even between “intuitively similar” samples (such as atomized Pb and metallic Pb).



From left to right: samples with the same archaeological Pb mass but increasing HNO_3 concentration

A sample of atomized Pb (left) and metallic Pb (right) with same Pb concentration

Preparation of lead samples

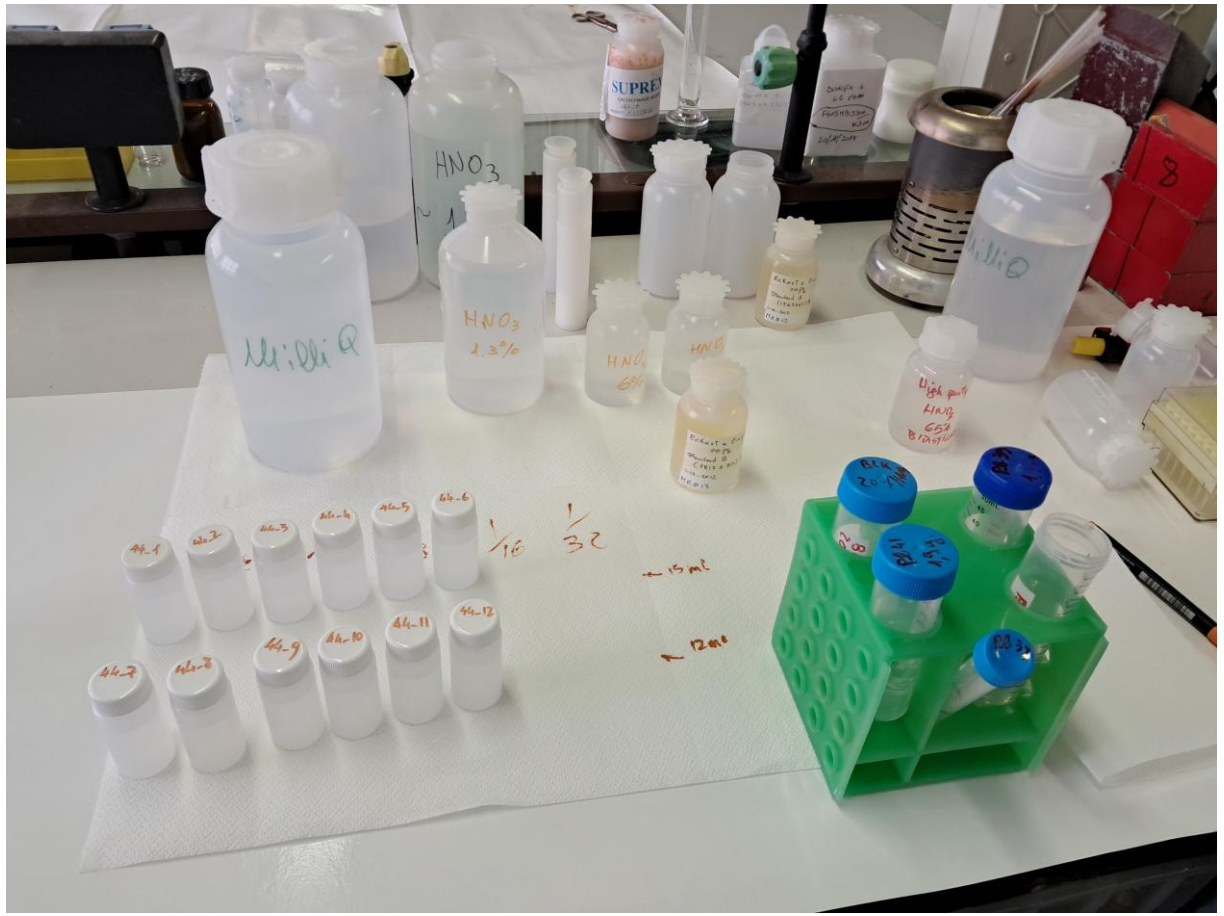
2-3 g of lead are dissolved in 20-30 ml of 20% concentrated HNO3 solution

↓

The solution is heated in an ultrasonic bath (50°C)

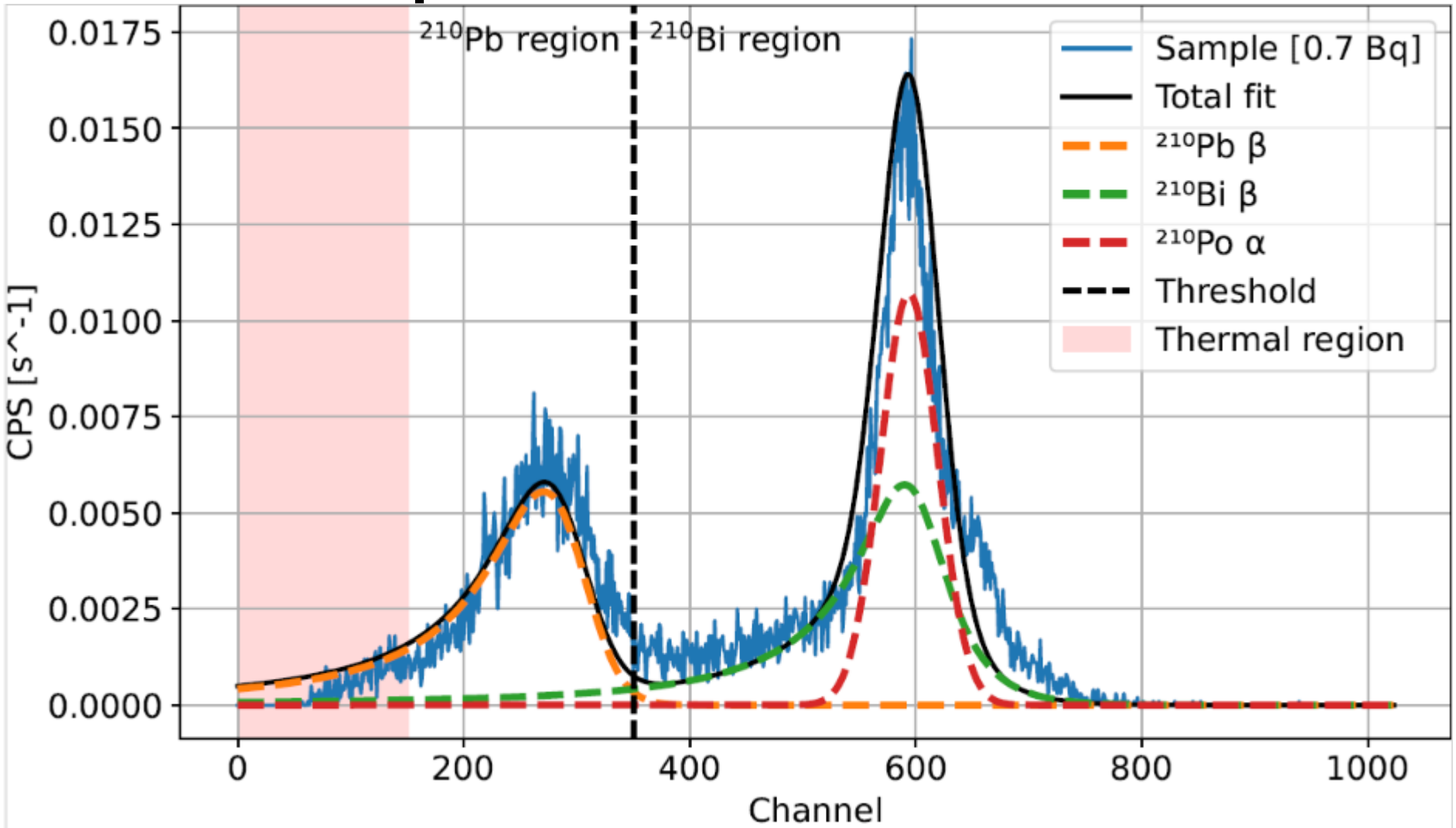
↓

8 ml of dissolved lead samples are mixed with 12 ml of ULTIMAGoldAB

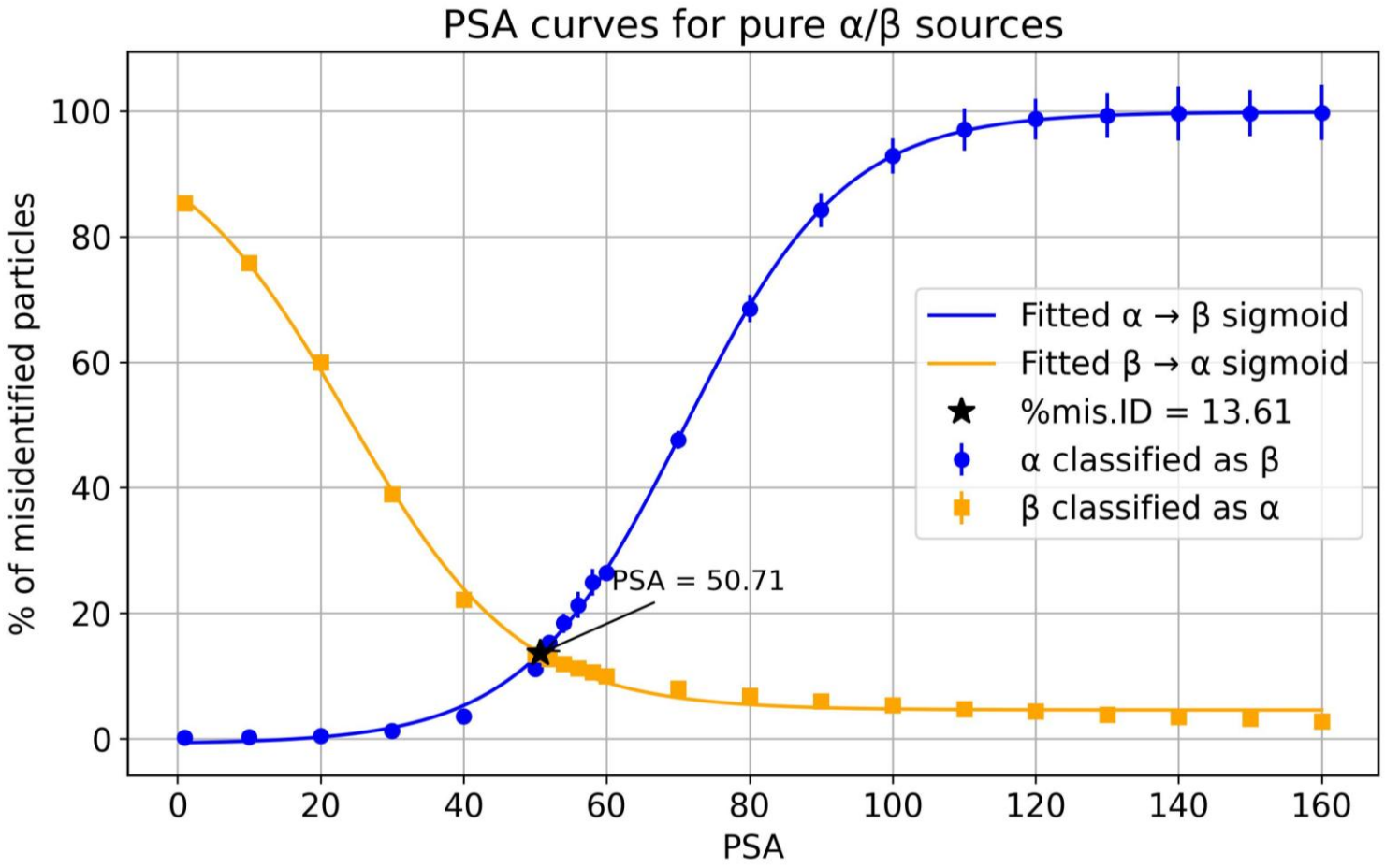


NOTE: This is the **optimized sample preparation procedure**. Different Pb concentrations, HNO_3 concentrations, and LS-to-sample ratios were tested before selecting these conditions.

Intermission: visualizing Quantulus spectra



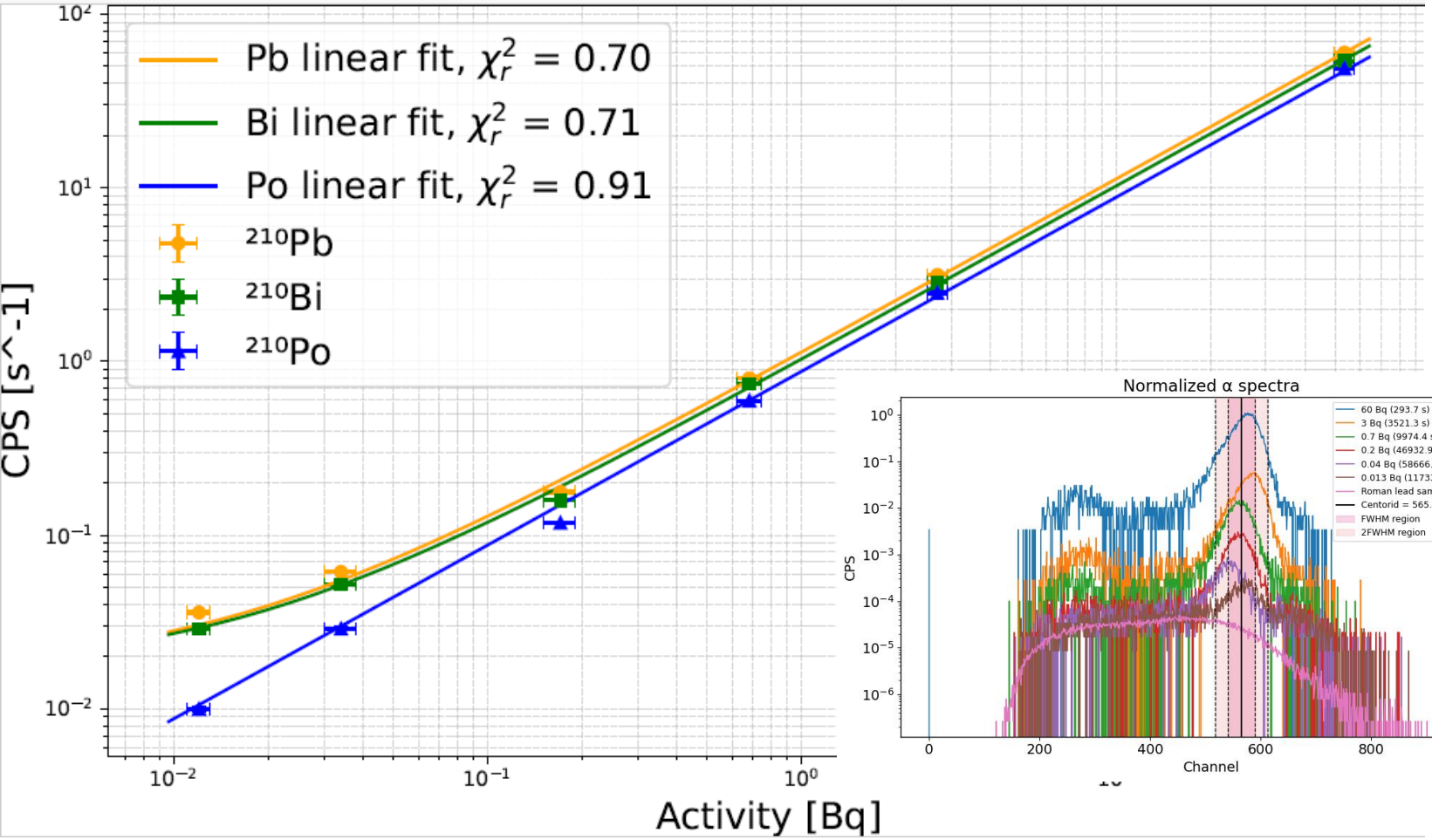
PSA Set-Up

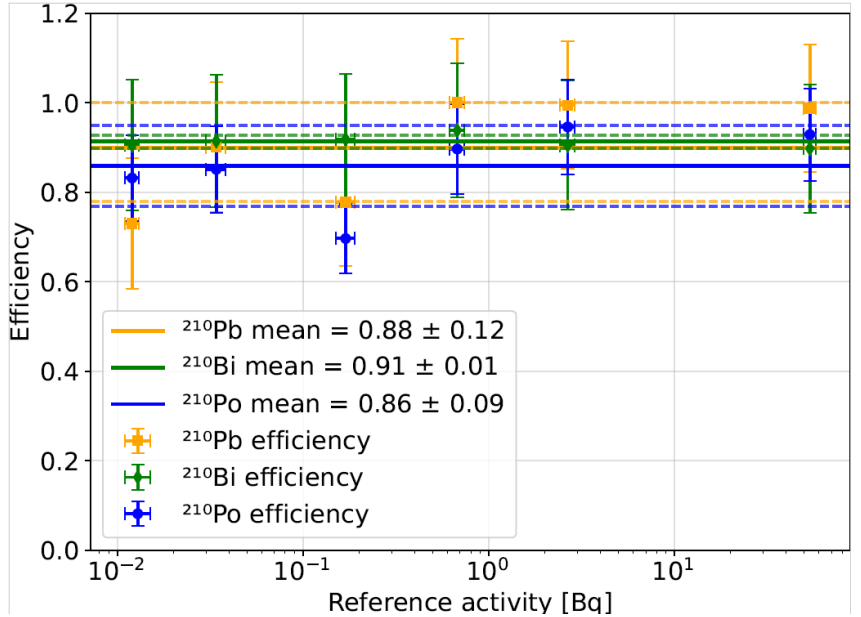


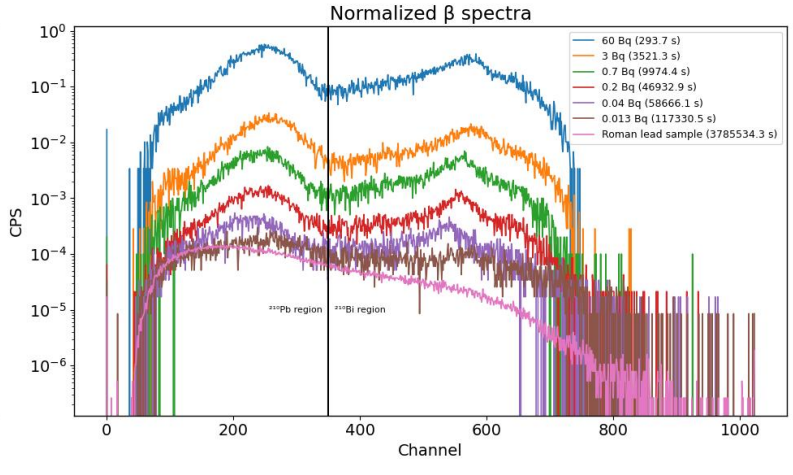
PSA SCANNING PROCEDURE

1. Preparation of 2 PSA samples chemically identical to optimal sample.
2. Spiking both samples with pure α/β emitter.
3. Measurement of both samples while scanning all possible PSA values
4. Determination of optimal setup through:
   - minimization of total mis-identification
   - symmetry of mis-identification

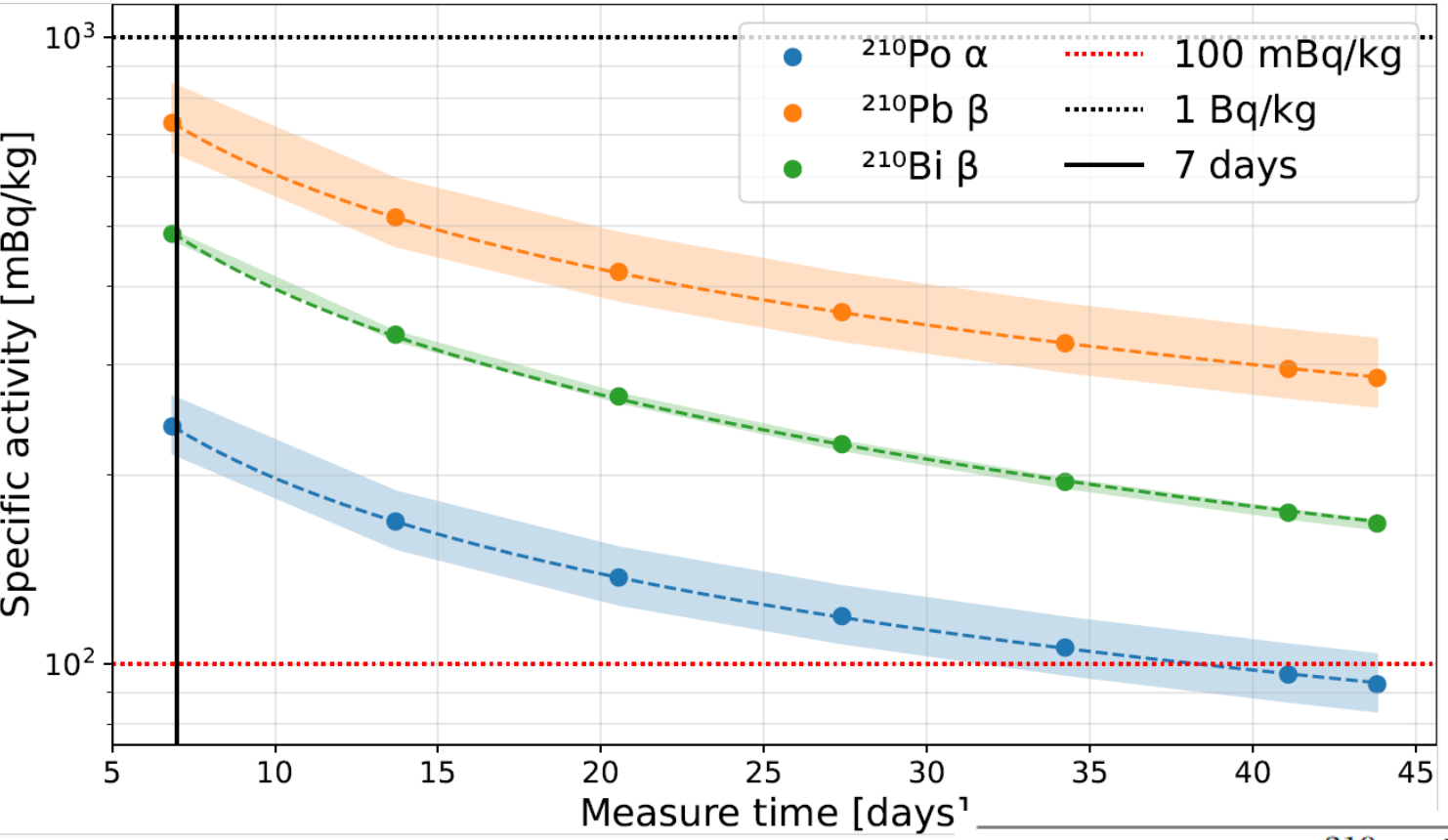
Linearity, efficiency and ROI







Results on metallic archaeo-Pb



4 samples:

1. Archaeolead 1: 0.79 g
2. Archaeolead 2: 0.79 g
3. High-purity Pb (CSC Pure Technologies): 0.75 g
4. Opera Pb: 0.76 g

$$\mathrm{DL}\left[\frac{\mathrm{Bq}}{\mathrm{kg}}\right] = \frac{k_\alpha\,\sigma_s}{m\,\epsilon\,T\,\mathcal{B}}$$

Simultaneous monitoring and measurement of the full ^{210}Pb decay chain!

Sample	^{210}Pb [mBq/kg]	^{210}Bi [mBq/kg]	^{210}Po [mBq/kg]	Time [d]
Archaeolead 1	< 720	< 661	< 203	7
	< 300	< 249	< 103	42
Archaeolead 2	< 731	< 485	< 239	7
	< 286	< 167	< 93	44
High–purity Pb	< 802	< 636	< 258	7
OPERA Pb	$(67 \pm 7) \times 10^3$	$(58 \pm 7) \times 10^3$	$(27 \pm 8) \times 10^3$	7

Quantulus paper

Eur. Phys. J. C (2026) 86:698
https://doi.org/10.1140/epjc/s10052-026-15922-7

THE EUROPEAN PHYSICAL JOURNAL C

Regular Article - Experimental Physics

A rapid low-background assay of ^{210}Pb in archaeological lead

M. Consonni[1,2,a], M. Clemenza[1,2,b], E. Di Stefano[2,3], N. Ferreiro Iachellini[1,2], F. Filippini[1,2,3], A. Gardini[4,5], G. Grosso[4], L. Pattavina[2,1], R. Della Pergola[3], S. Quitadamo[1,2], E. Sala[2,6], F. Saliu[2,3], A. Salvini[4,5], L. Trombetta[1,2]

[1] Dipartimento di Fisica, Università di Milano-Bicocca, Piazza della Scienza 3, 20126 Milan, Italy
[2] INFN Sezione di Milano-Bicocca, Piazza della Scienza 3, 20126 Milan, Italy
[3] DISAT, Università di Milano-Bicocca, Piazza della Scienza 1, 20126 Milan, Italy
[4] Laboratorio Energia Nucleare Applicata, Via Aselli 41, 27100 Pavia, Italy
[5] INFN Sezione di Pavia, Via Bassi 6, 27100 Pavia, Italy
[6] Center for Underground Physics, Institute for Basic Science, Daejeon 34126, Korea

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Abstract In this work, we present a fast and highly efficient method for the measurement of ^{210}Pb in metallic archaeological lead using the commercial low-background liquid scintillation counter Wallac Quantulus 1220 installed at the University of Milano–Bicocca (Italy). By combining an optimized chemical preparation with pulse-shape analysis

ground events generated by environmental radioactivity and cosmic rays.

Natural radioactivity is unavoidably present in all materials used in detector construction, making it challenging to achieve the ultra-low-background conditions required to detect such elusive signals. Moreover, the massive shielding

Come and check the RES-NOVA collaboration!
https://res-nova.unimib.it/

DOI: 10.1140/epjc/s10052-026-15922-7

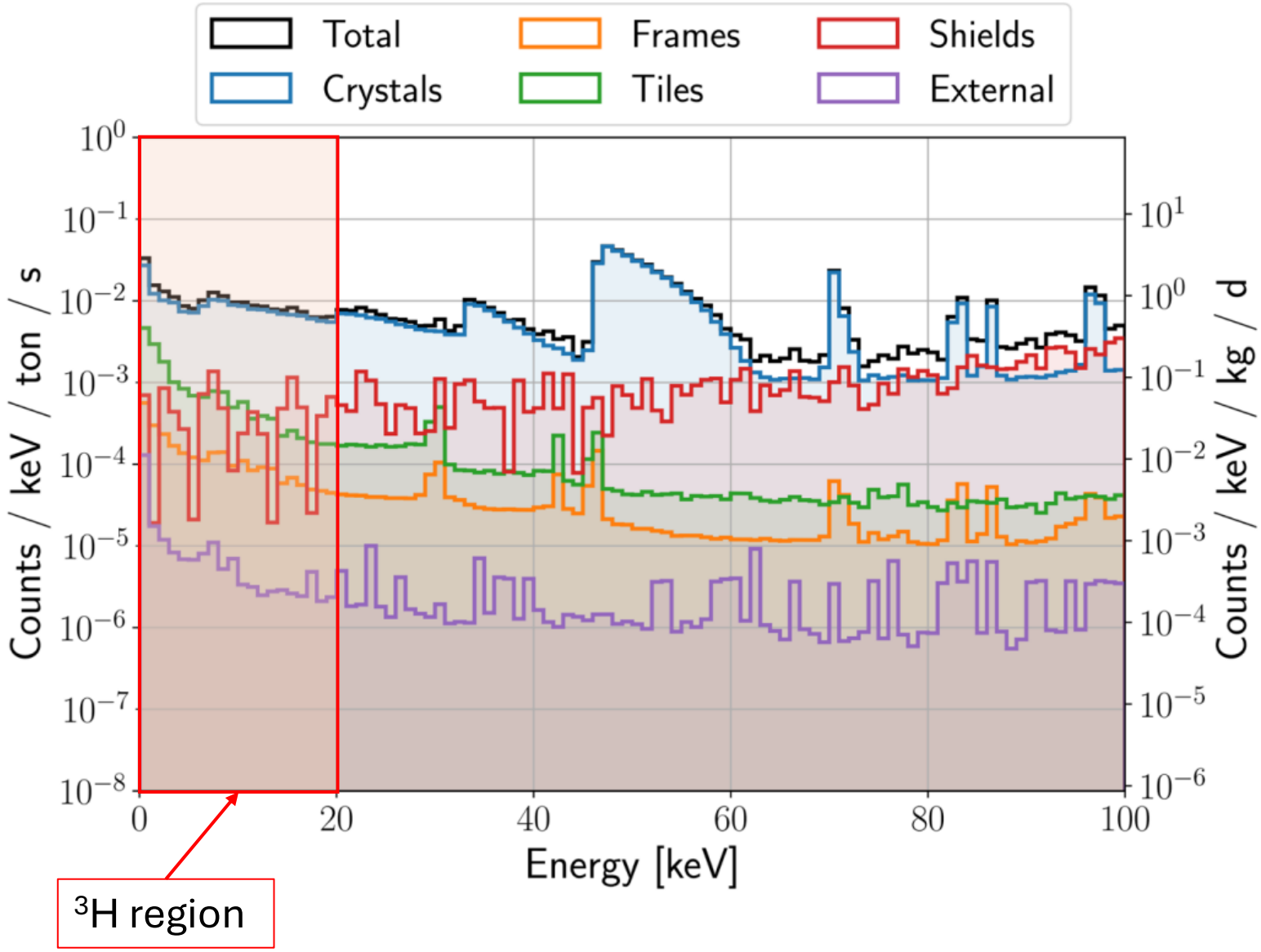
RES-NOVA cosmogenic background

Cosmogenic radionuclides are an additional source of background in rare-event detectors.

Activation studies for RES-NOVA
To investigate cosmogenic activation, PWO crystals and archaeological Pb were neutron-irradiated at ChipIR (ISIS).

Here: preliminary results on ^{3}H in activated PWO

Poster session: activation studies of archaeological PWO
(*hopefully, you already noticed!)*



Measurement of ^{3}H in activated PWO

1. Optimal detection set-up → 12 ml LS + 8 ml liquid sample mix.
2. NO PSA CALIBRATION NEEDED.
3. Calibration curve with sample spiked with known activity of ^{3}H.
4. Measurements of samples (PRELIMINARY).

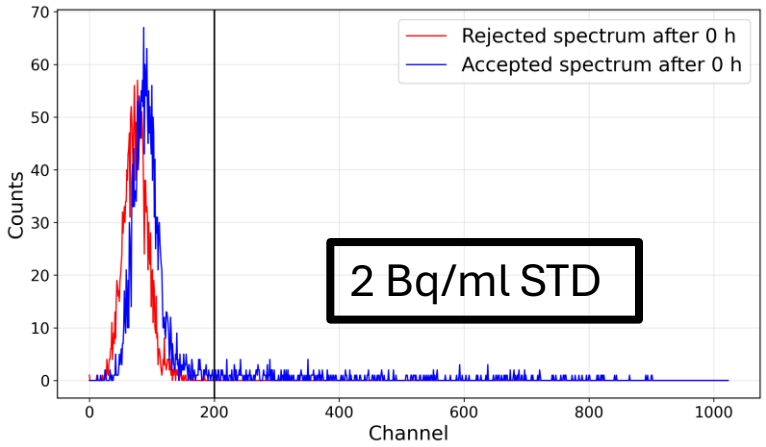
ISSUES:

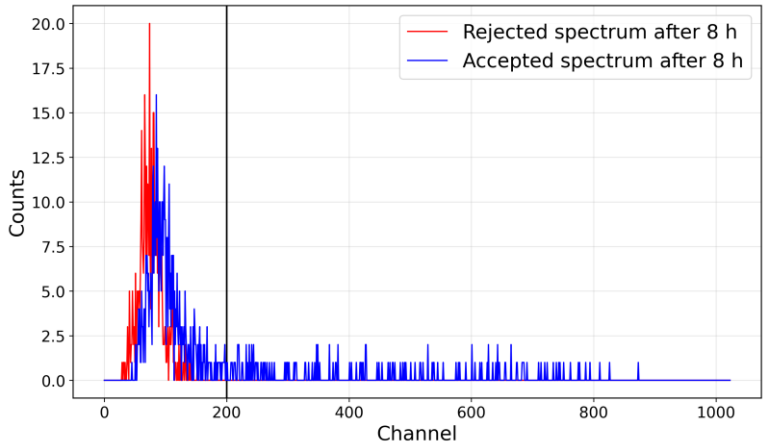
1. Phase separation of NaOH solvent with LS → detection efficiency temporal dependance.
2. PWO Blank samples slightly ^{3}H contaminated, hard to correctly measure.
3. Overall low detection efficiency expected.

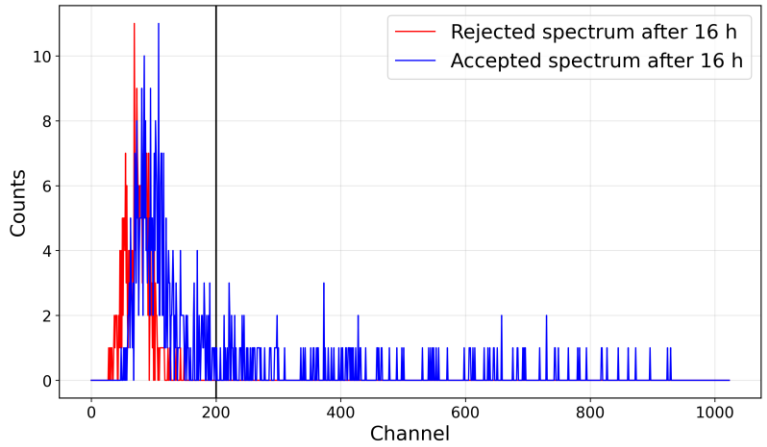
PWO sample few minutes after preparation

PWO sample 5 hours after preparation

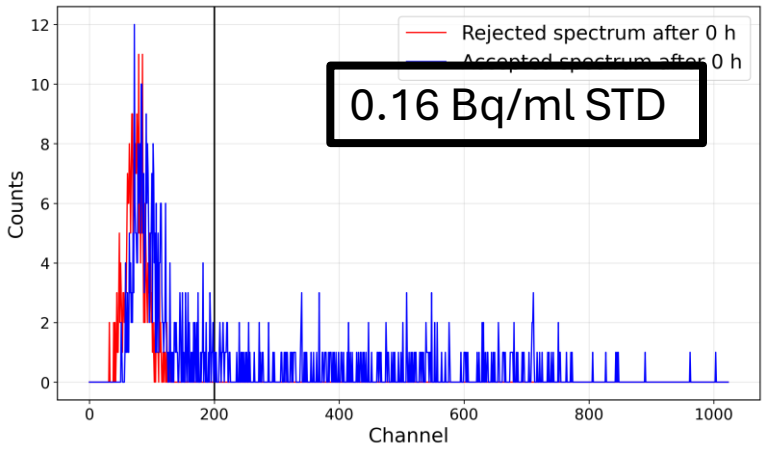
^{3}H ROIs

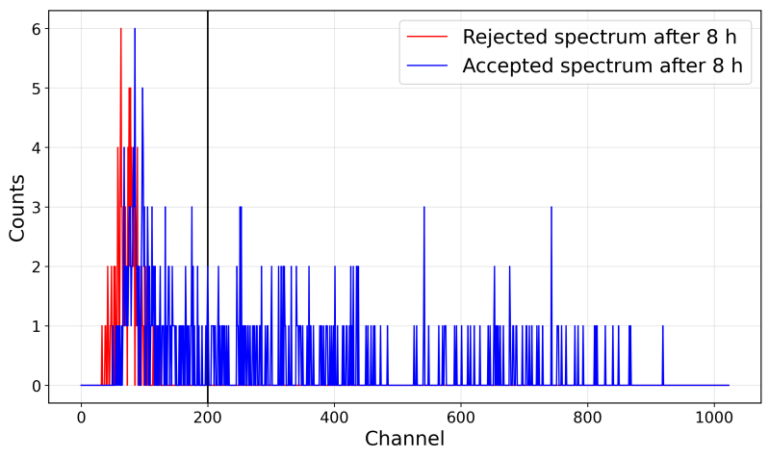


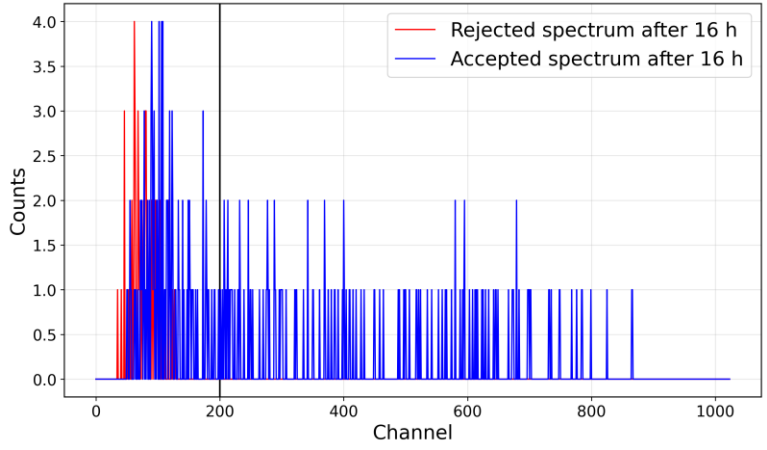




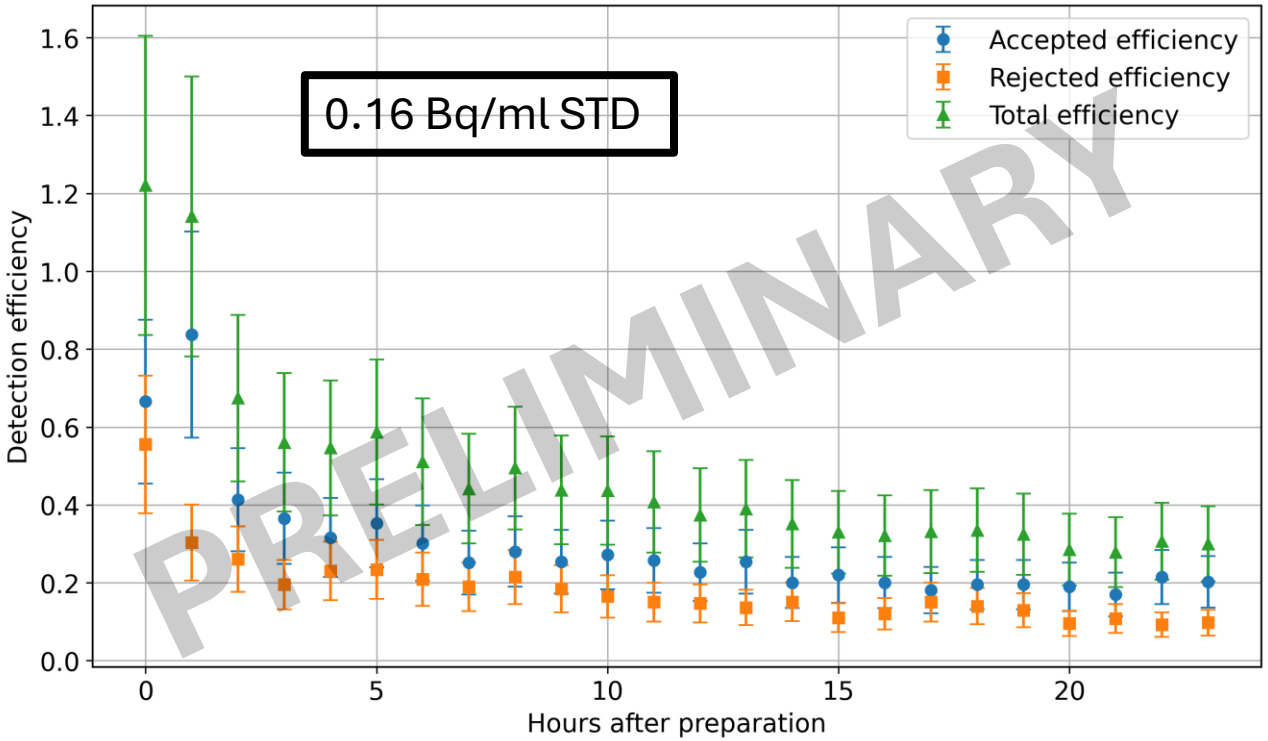
Measure time

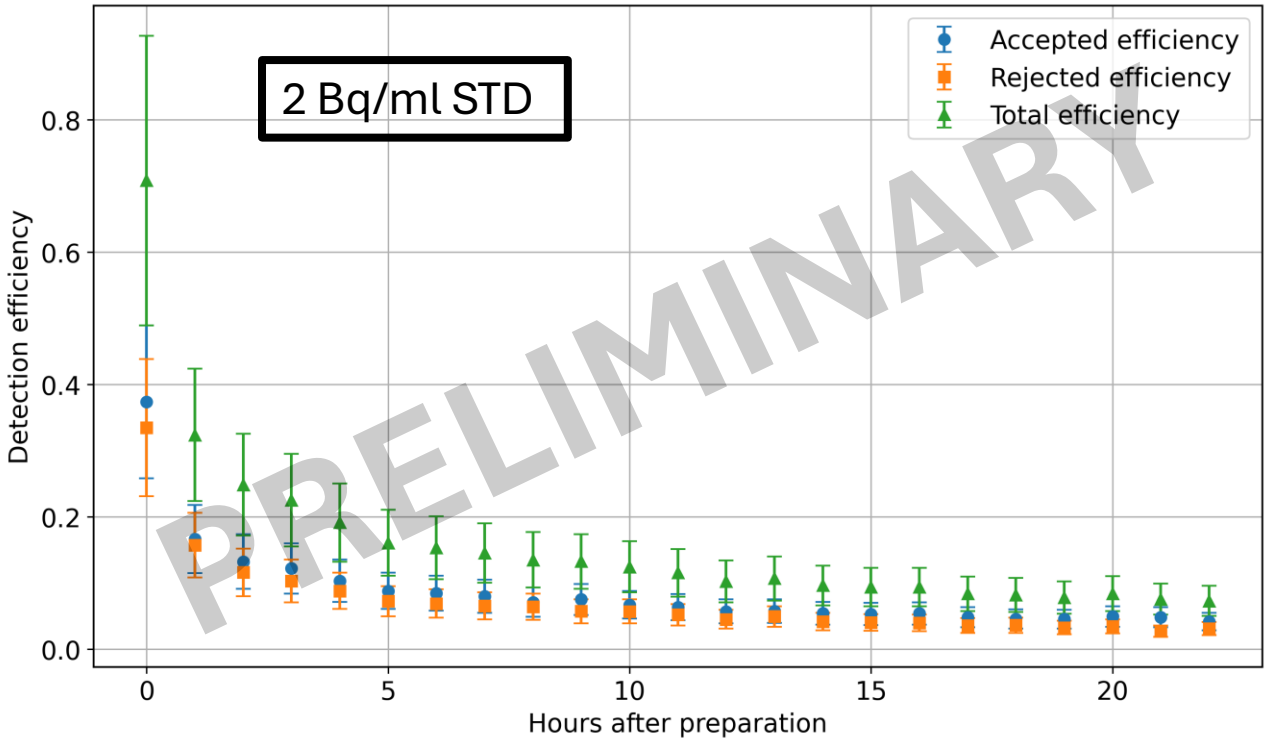






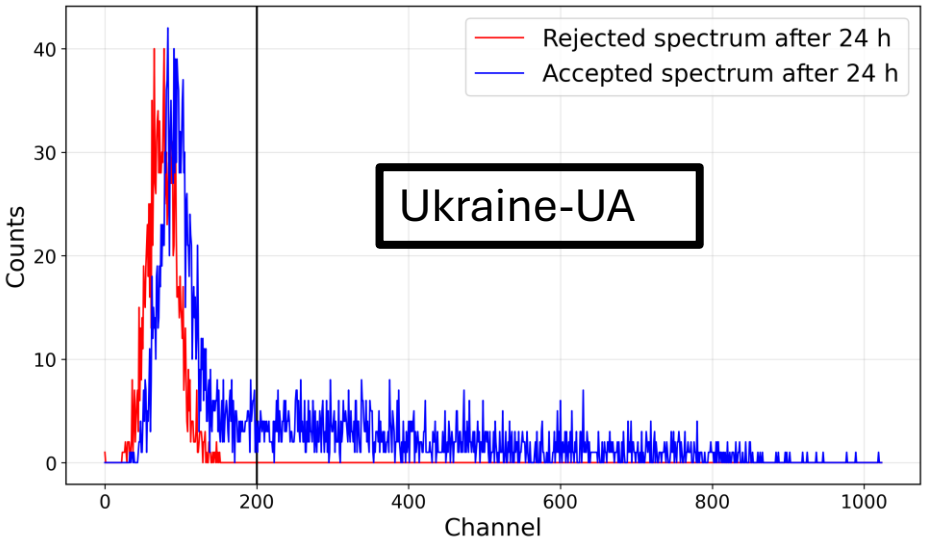
Detection efficiency stability in time

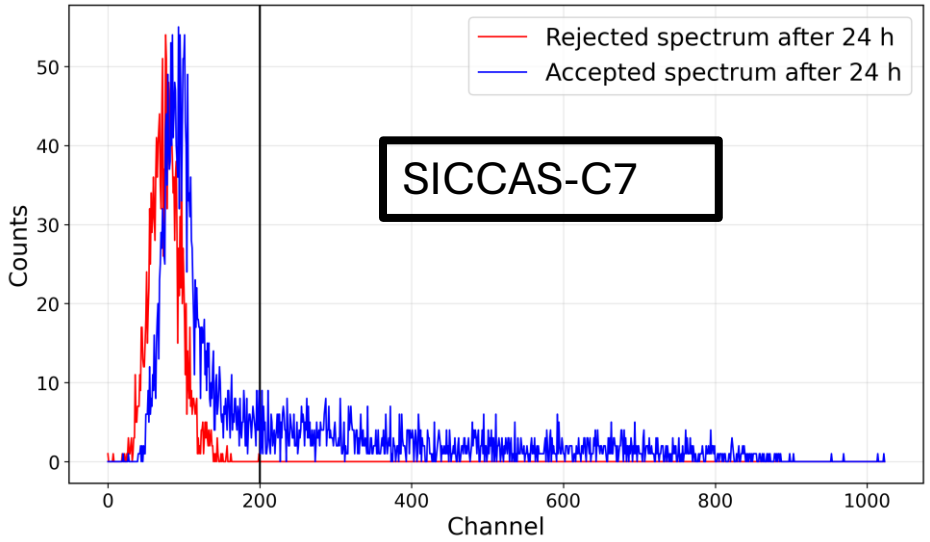


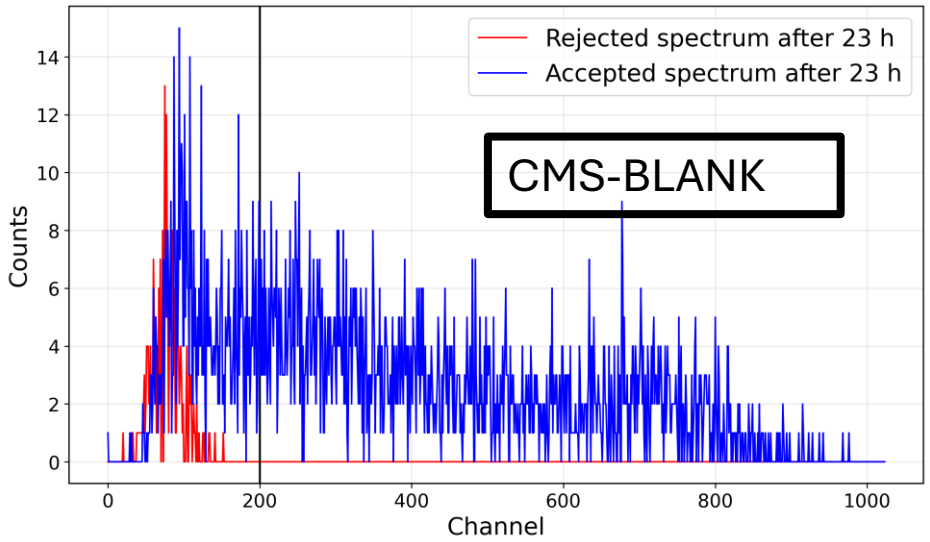


- Phase separation changes detection stability
- Stability reached after 15-20 h from sample preparation

^{3}H measures in PWO crystals



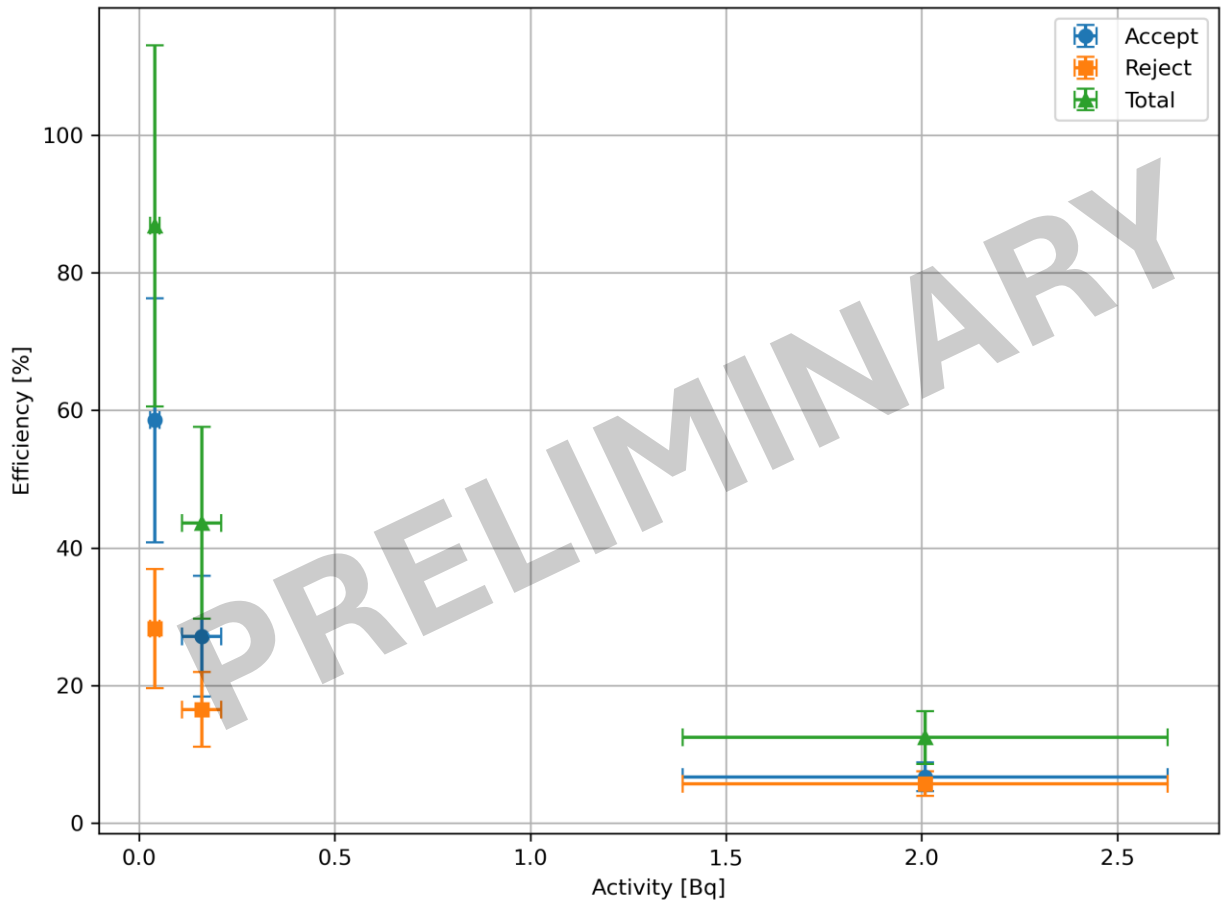




PRELIMINARY ^{3}H results

Sample	Method	Activity [Bq/kg]
C7	Accepted	307.0 ± 95.8
	Rejected	285.1 ± 89.4
	All	296.7 ± 92.2
UA	Accepted	117.5 ± 36.7
	Rejected	110 ± 34.6
	All	114 ± 35.5
CMS		$\sim$10-20

NOTE: the C7 (Siccas) and UA (Ukraine) samples were neutron-activated at ChipIR, ISIS neutron source that mimics the fast atmospheric neutron spectrum with a flux enhanced by a factor 10^9.



A rapid and versatile method for low-background radioassays

RAPID
~1 week for $O(10^2)$ mBq/kg sensitivity

LOW SAMPLE MASS
< 1g of sample

SIMPLE AND ACCESSIBLE
based on commercial LSC detector

SIMULTANEOUS detection of the ^{210}Pb decay chain


Thank you!

Backup slides

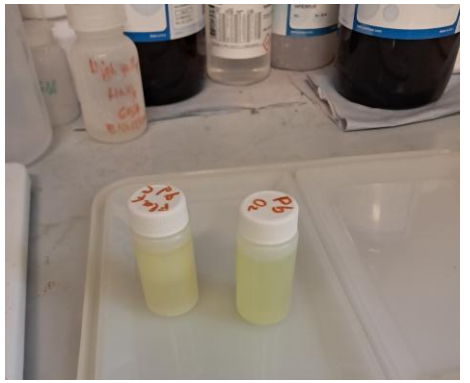
Issues on atomized Pb

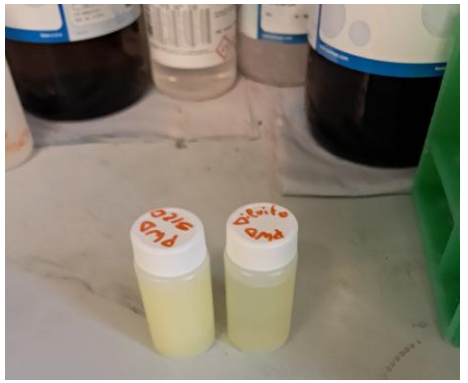
Chemistry issues:
Highly concentrated samples of atomized Pb react with LS, producing yellow-colored samples with strong energy quenching (likely due to Pb oxidation).

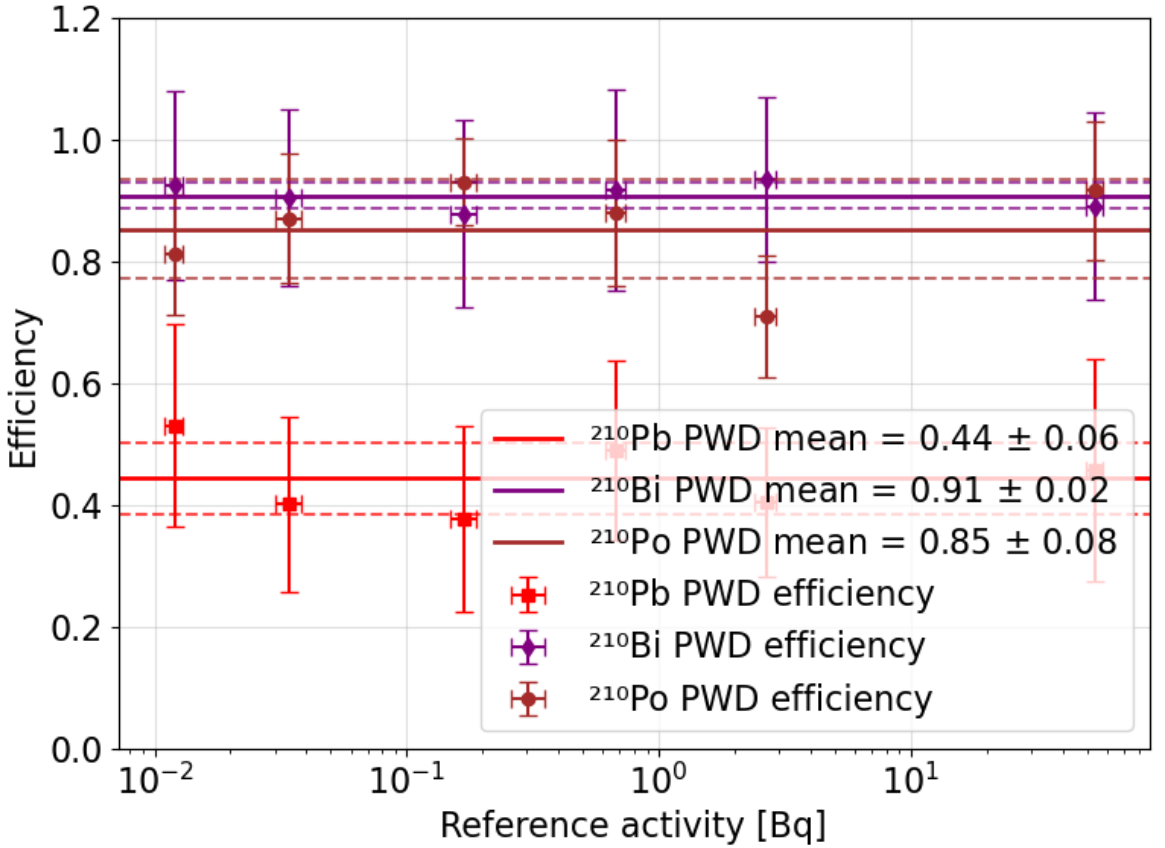
Different set of measurements were carried out to investigate and mitigate the issue.

Solution: lower Pb concentration in samples:
when 0.3 g of atomized Pb is dissolved, detection efficiency for ^{210}Bi and ^{210}Po become comparable to metallic Pb; only the energy quenching differs.
^{210}Pb efficiency remains significantly lower (44 ± 6 %).

New PSA set-up as necessary.
Samples degrade faster in time.

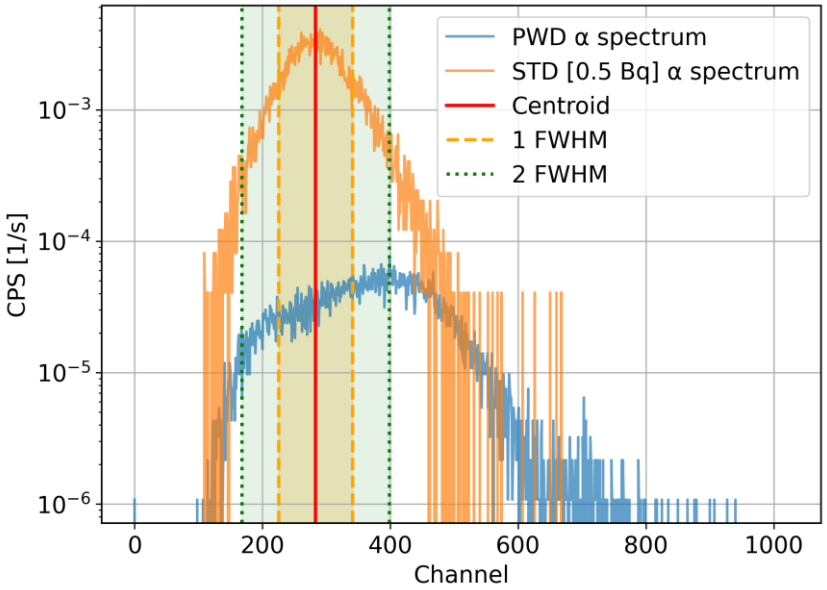




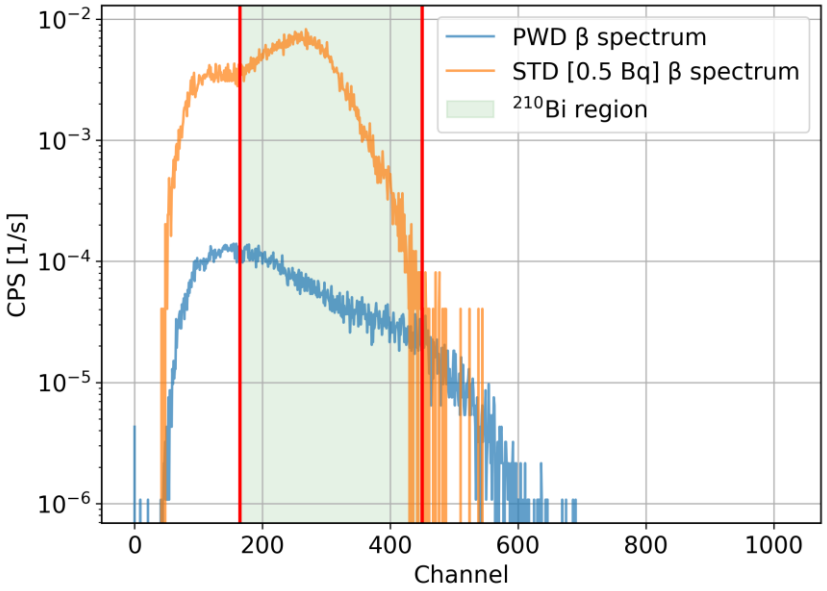


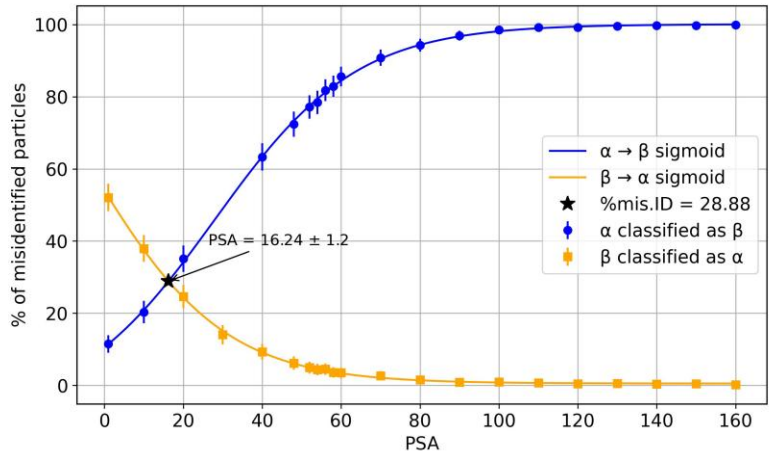
Results on atomized Pb

α spectra



β-spectra



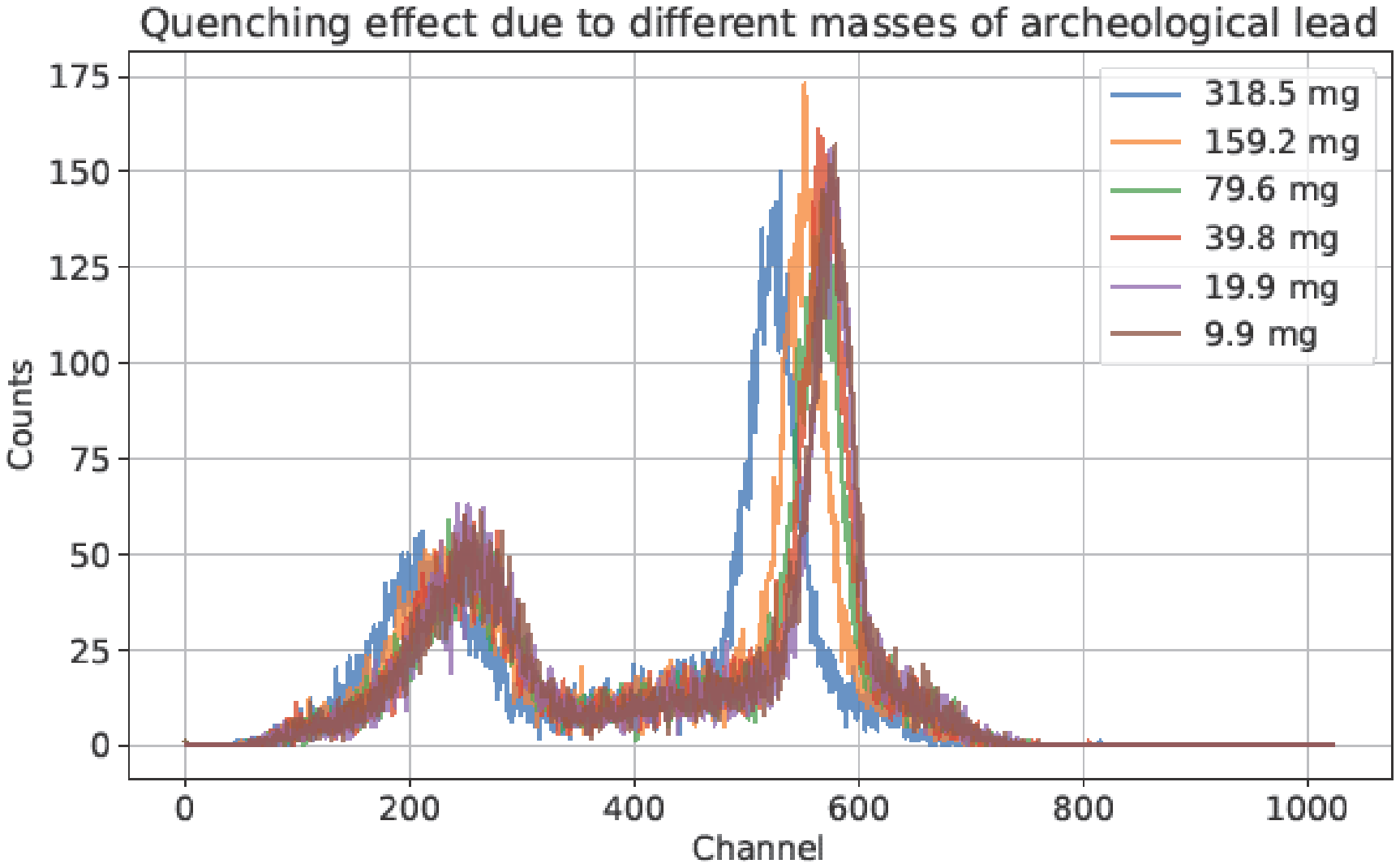


Expected ^{210}Po sensitivity after a 45 d measure < 282 mBq/kg.

m_{powder} = 0.3 g
$m_{metallic}$ = 0.79 g

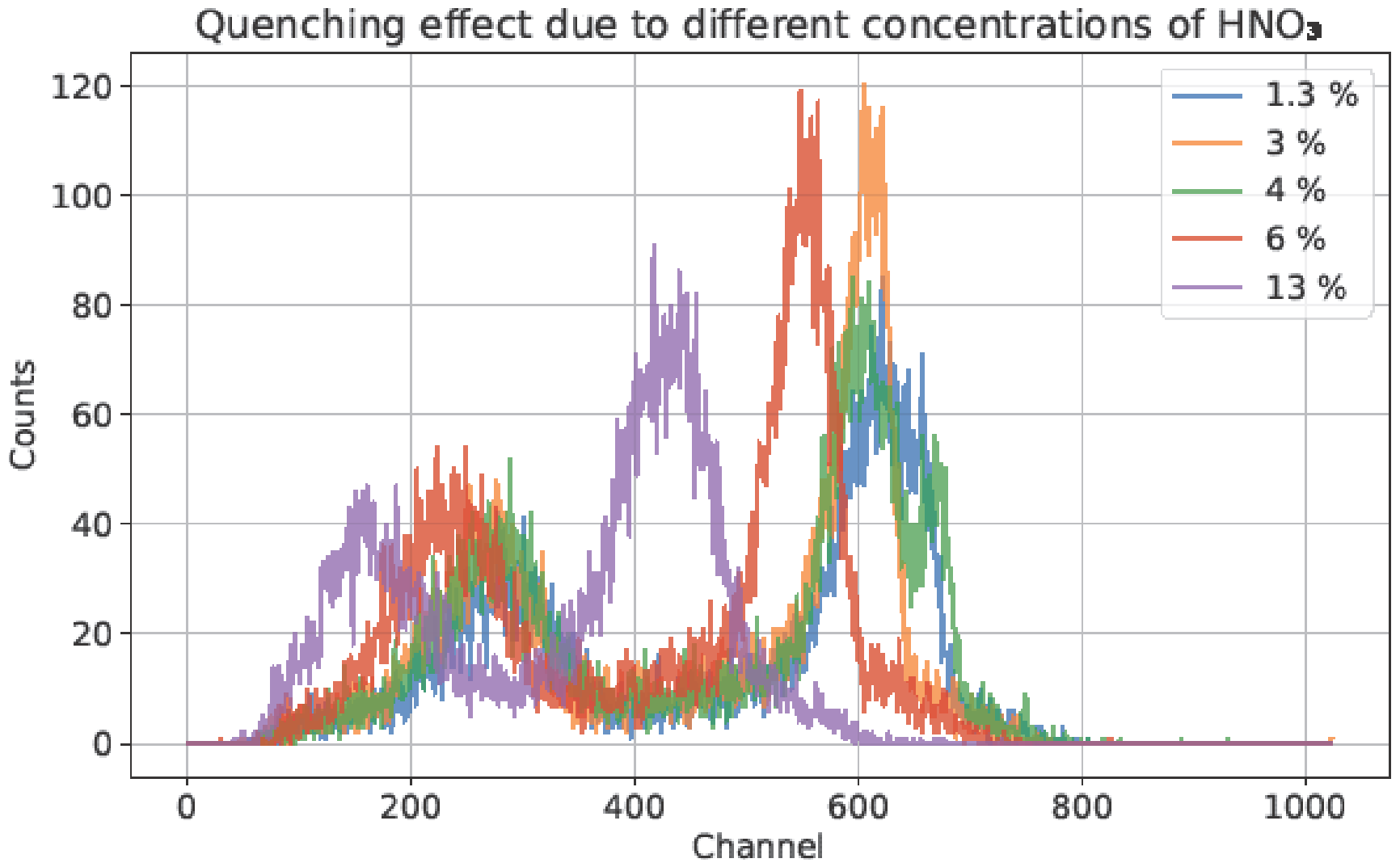
Sample	^{210}Pb [mBq/kg]	^{210}Bi [mBq/kg]	^{210}Po [mBq/kg]	Time [d]
Atomized Pb	< 2100	< 1150	< 563	11
Archaeolead 2	< 286	< 167	< 93	44

Quenching: Pb mass



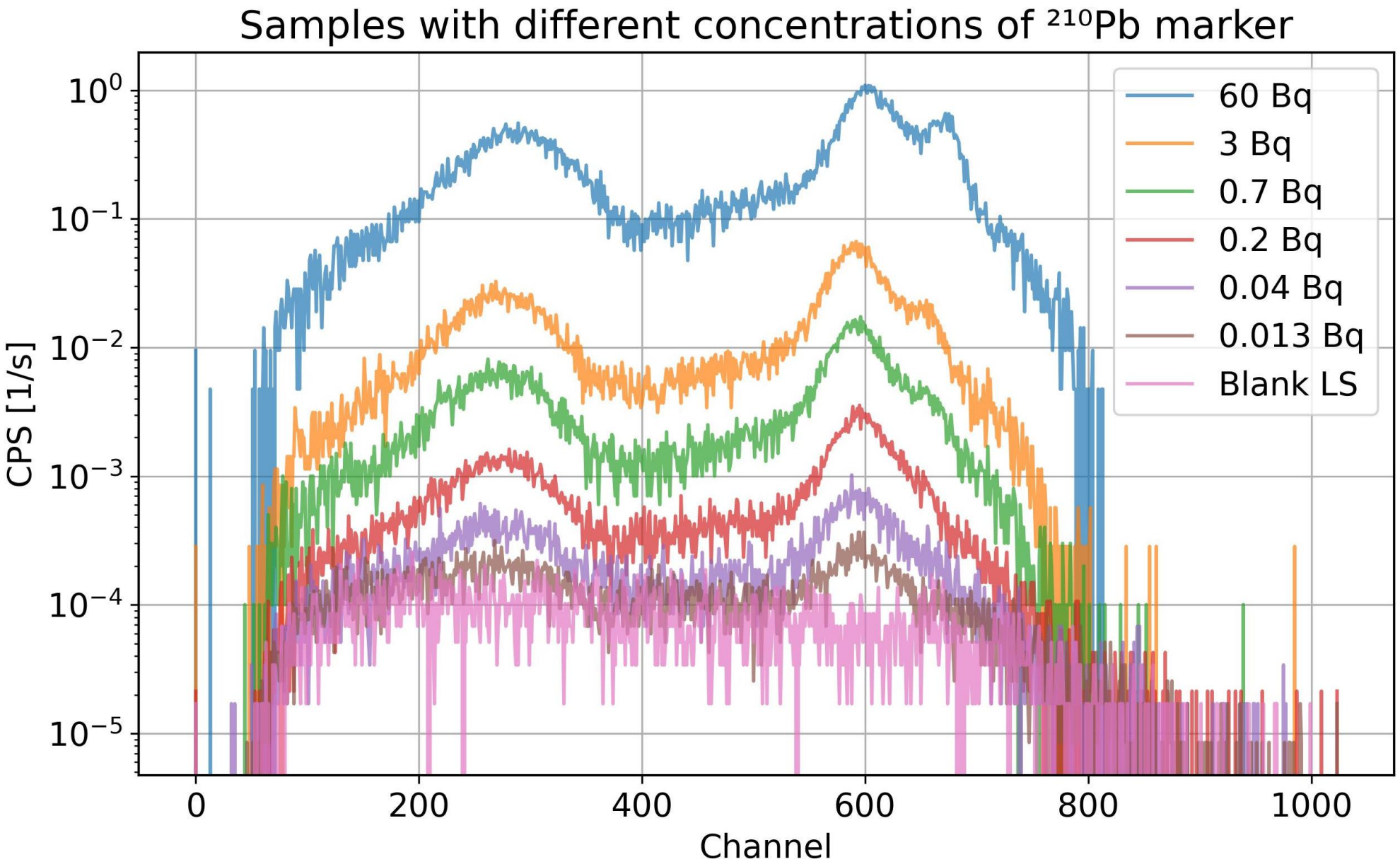
- No α/β discrimination
- Low measure time (15 min)
- 8/20 set
- Quenching effects visible but not excessively impactful.

Quenching: HNO_3



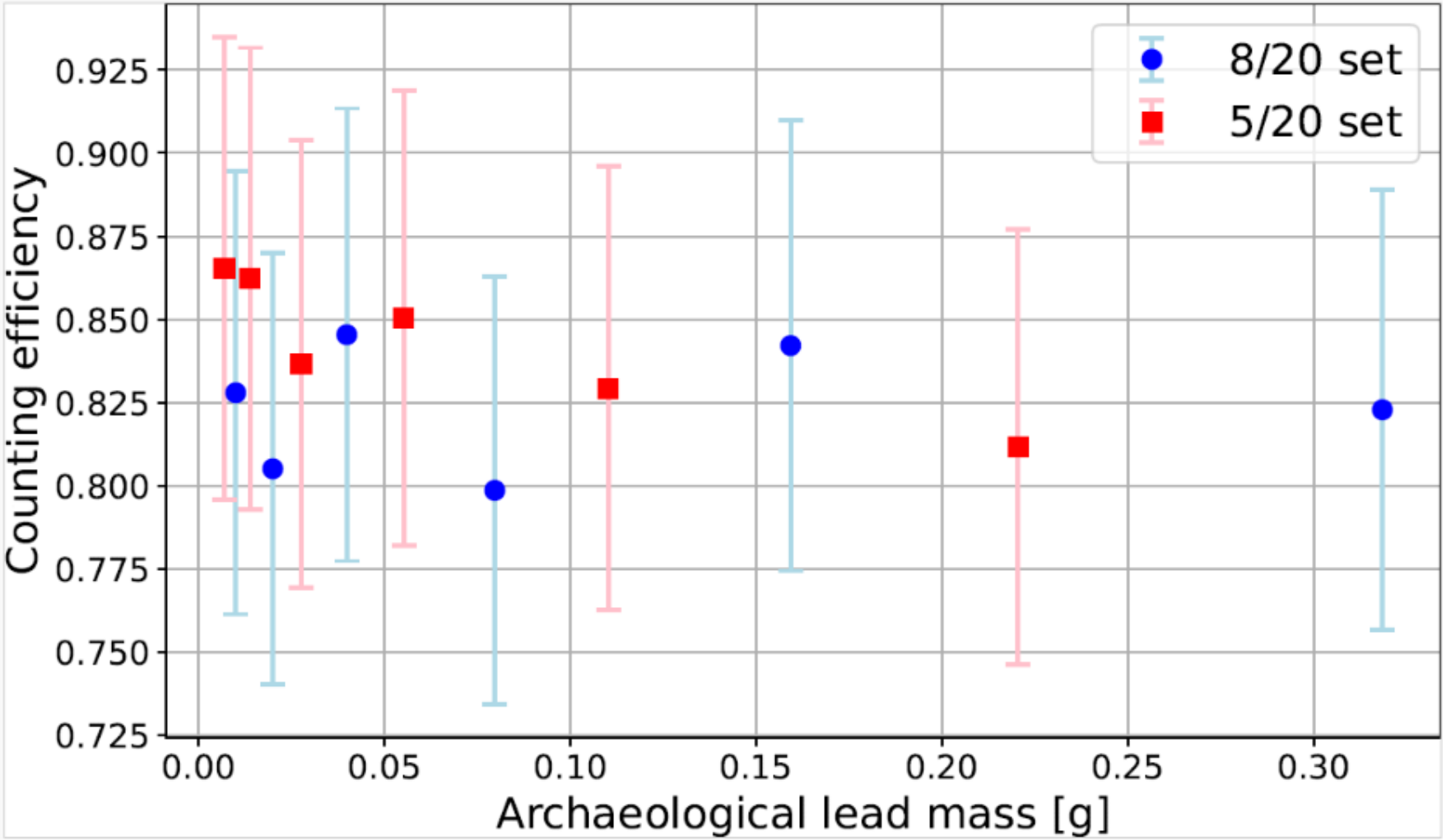
- No α/β discrimination
- Low measure time (15 min)
- 8/20 set
- Highly impactful quenching effects

No PSA tests



- No α/β discrimination
- Different measure times (5 min up to 2 d)
- 8/20 set

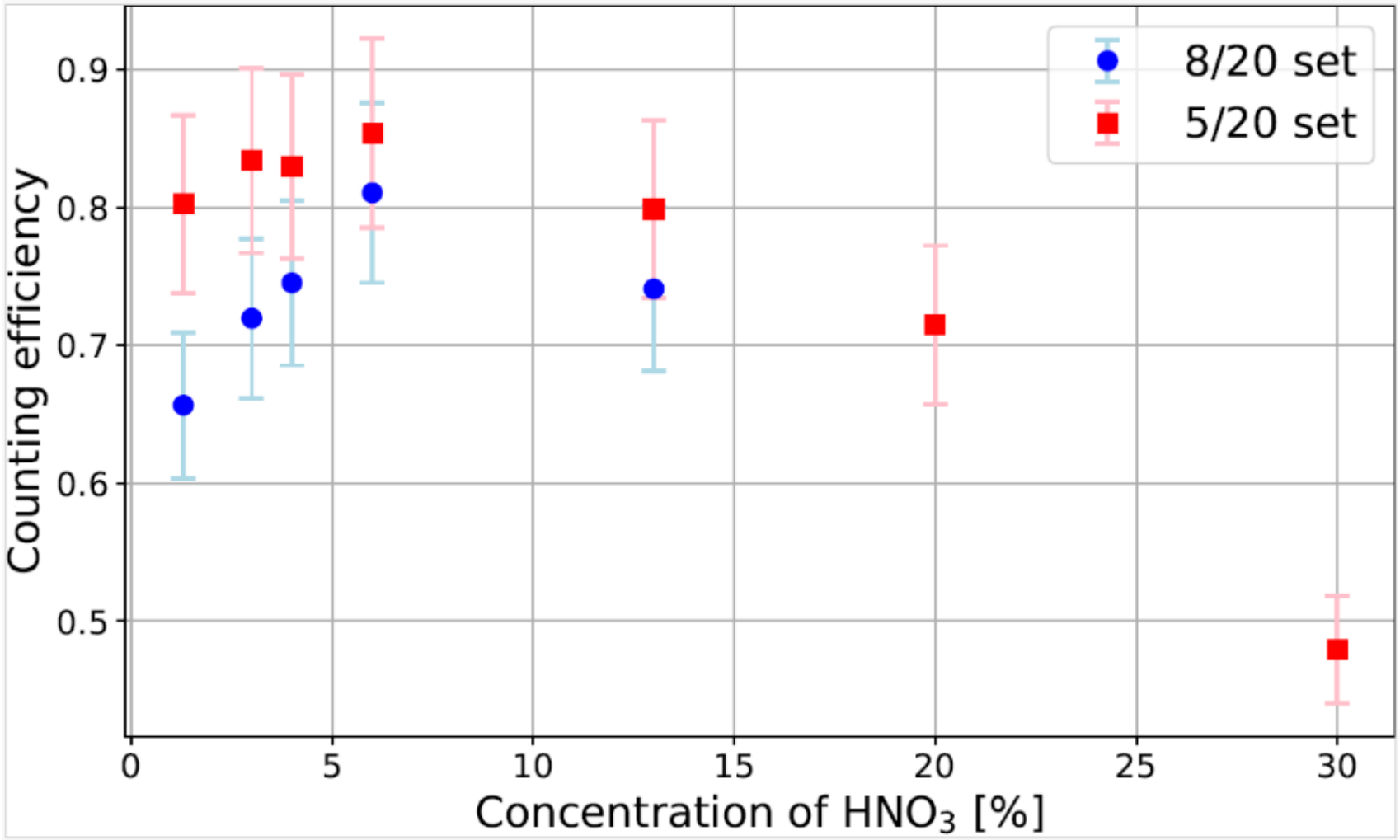
Efficency



2 set of samples:
8/20 = 8 ml sample + 12 ml LS
5/20 = 5 ml sample + 15 ml LS

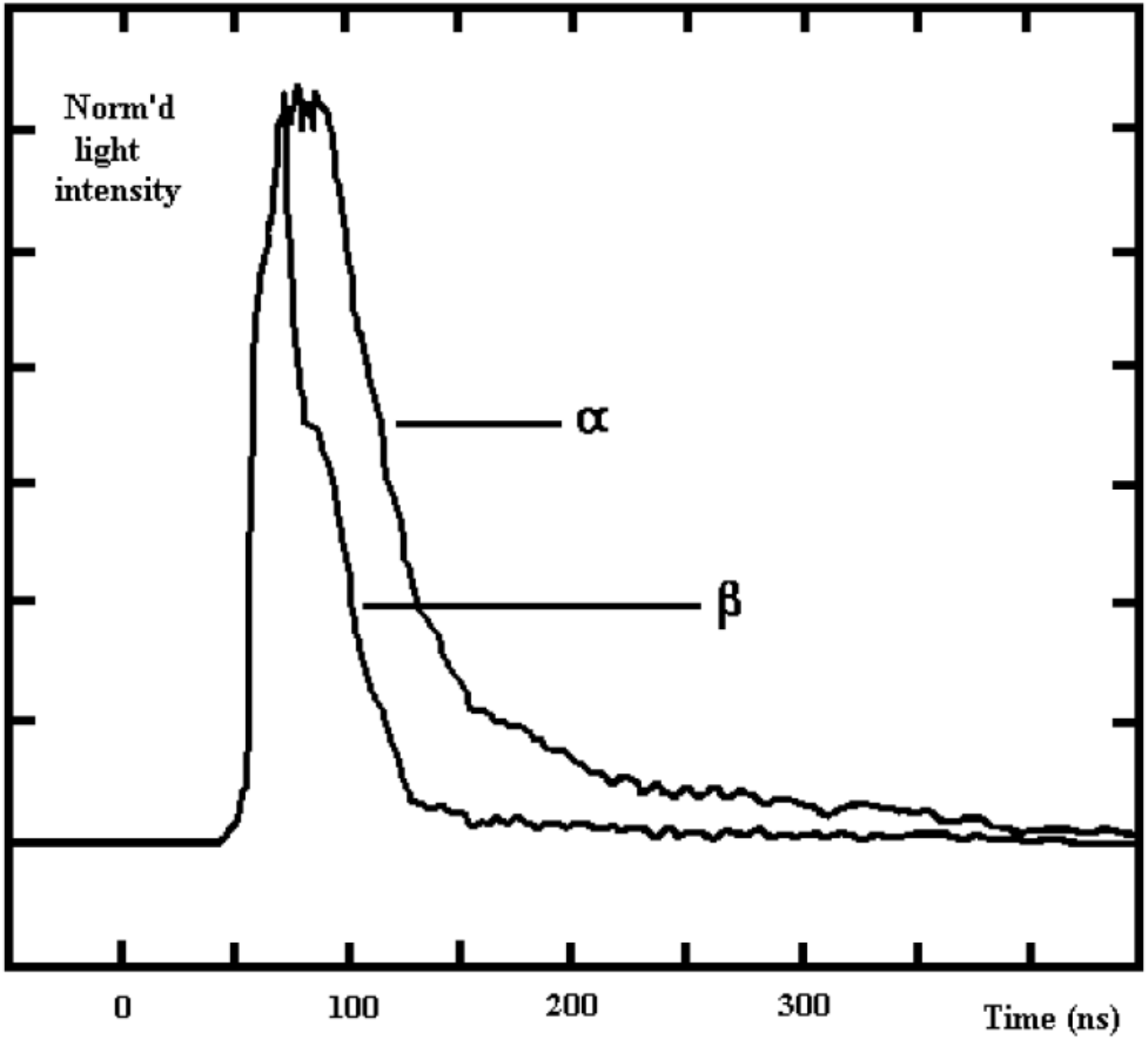
$\varepsilon_{8/20} = 82 \pm 2\ \%$
$\varepsilon_{5/20} = 85 \pm 2\ \%$

Efficency



Quenching and thereore counting efficiency strongly depends on HNO_3 concentration

Pulse Shape Analysis



Quantulus allows α/β discrimination analyzing the signal lenght.

PSA-dedicated samples are required to determine the optimal PSA value. PSA samples should closely resemble actual samples to ensure accuracy.

PSA is an arbitrary integer value from 1 to 200. Since it is not possible to save pulse shape, to operate a posterior pulse-shape discrimination, it is necessary to scan all possible PSA values with pure α/β emitters

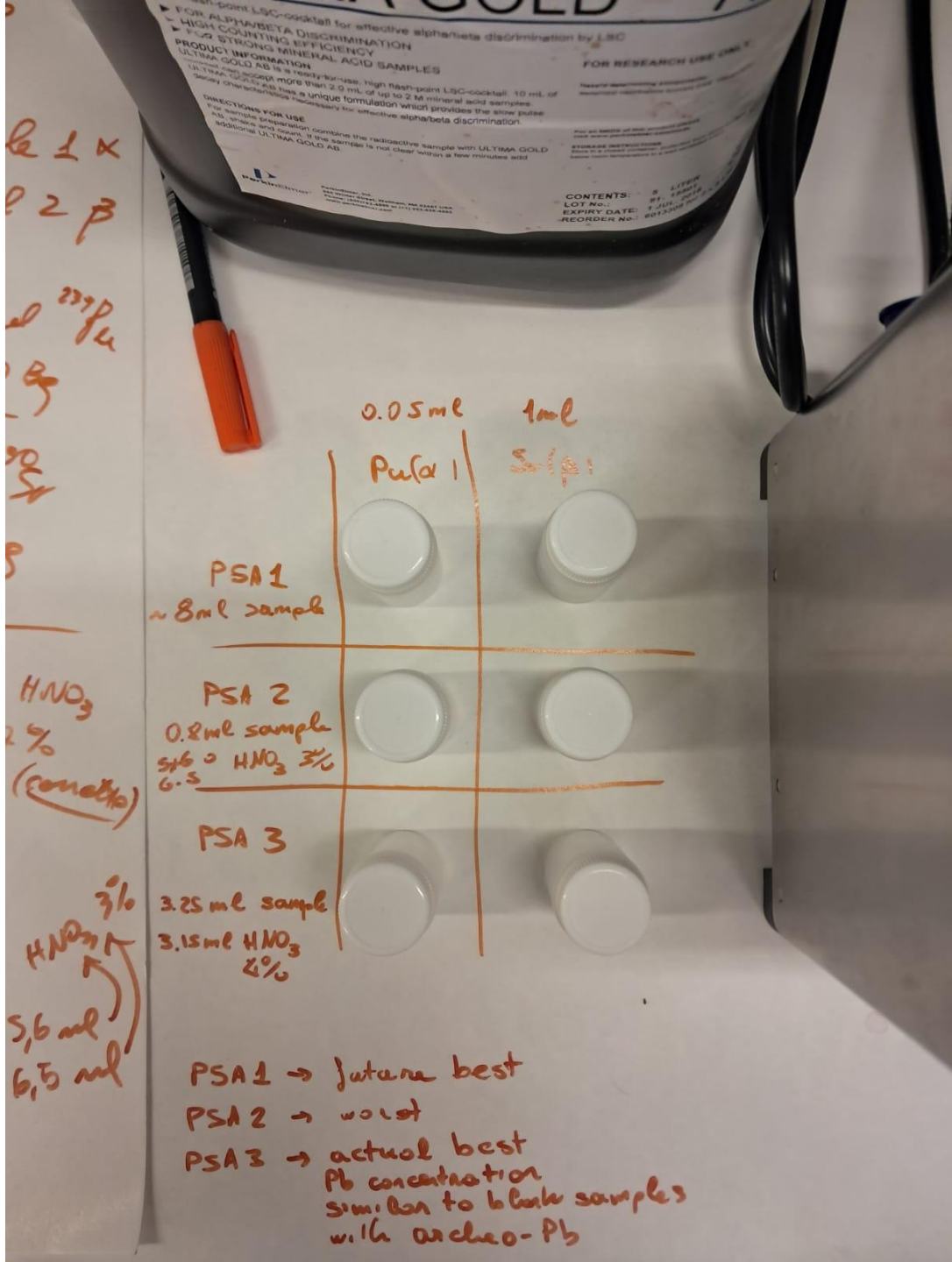
PSA samples

Preparation of 3 sets of 8/20 samples:

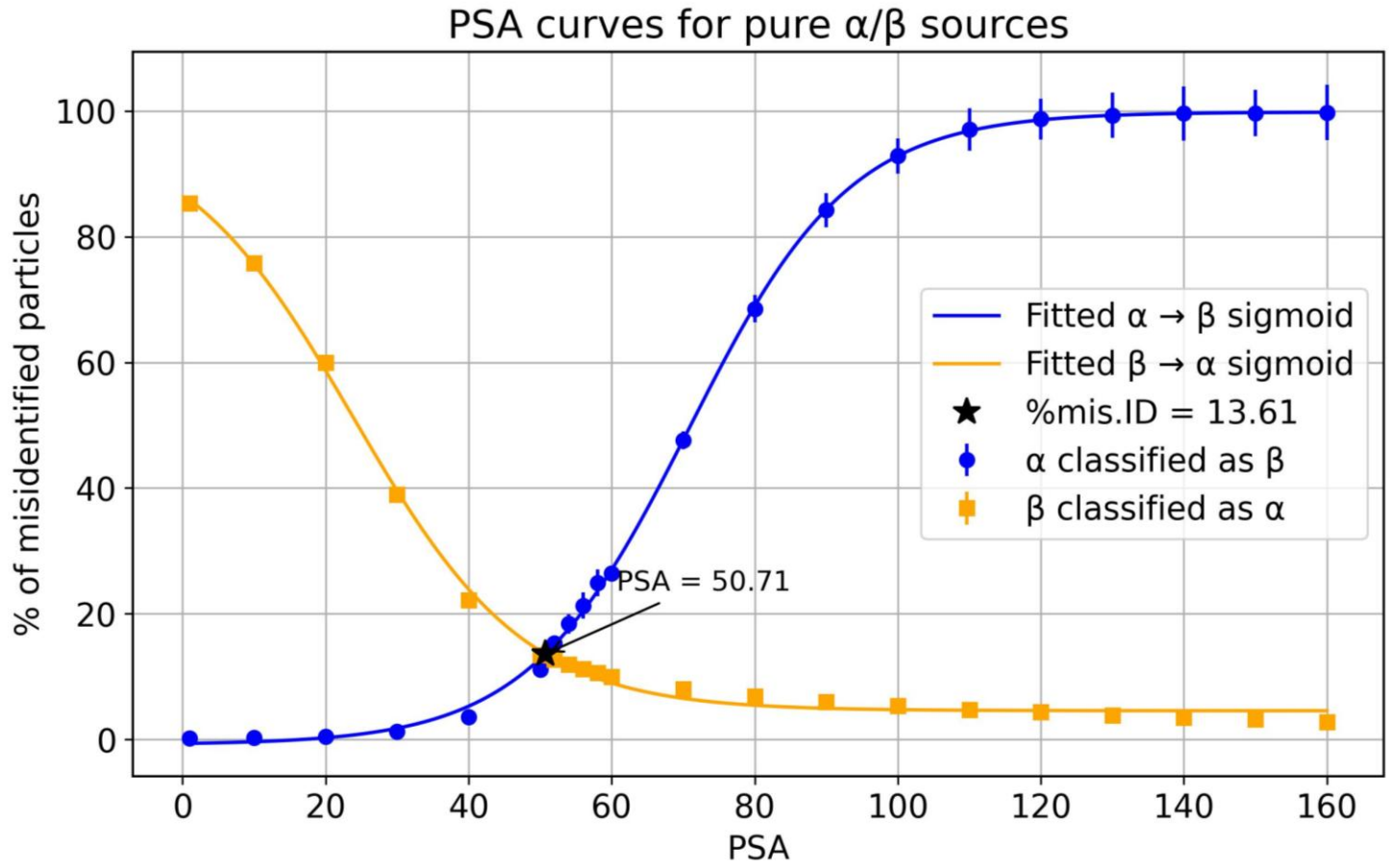
1. Sample with 0.704 g of dissolved archeological lead: optimal setup to maximize Pb sensitivity
2. Sample with 0.062 g of dissolved archeological lead: least favorable setup to maximize Pb sensitivity
3. Sample with 0.286 g of dissolved archeological lead: midpoint for the possible mass of dissolvable lead.

For each condition, 2 samples are prepared and subsequently spiked with a pure α/β emitter

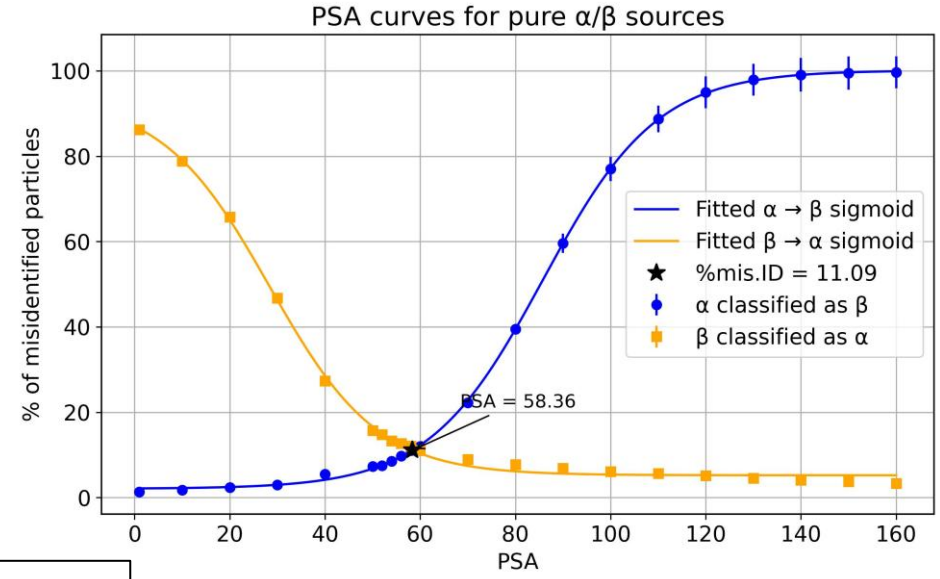
Pure α source: ^{238}Pu
Pure β source: ^{90}Sr



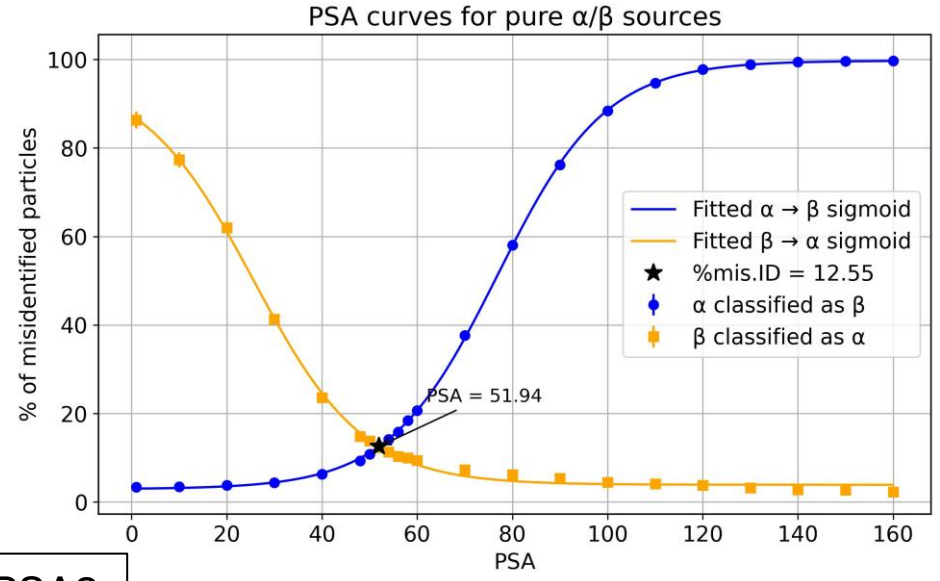
PSA Tests



PSA1, for best sensitivity samples!

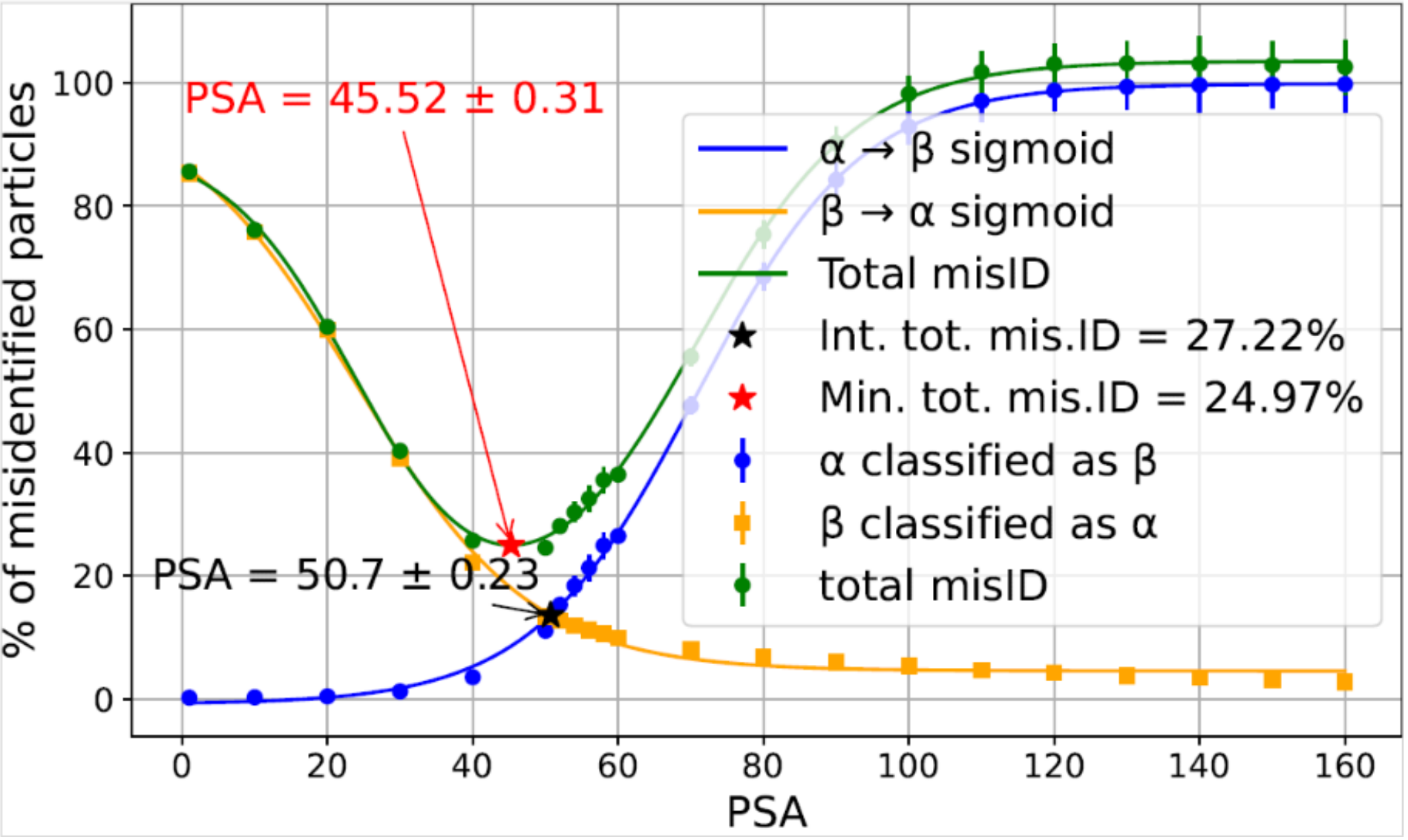


PSA2



PSA3

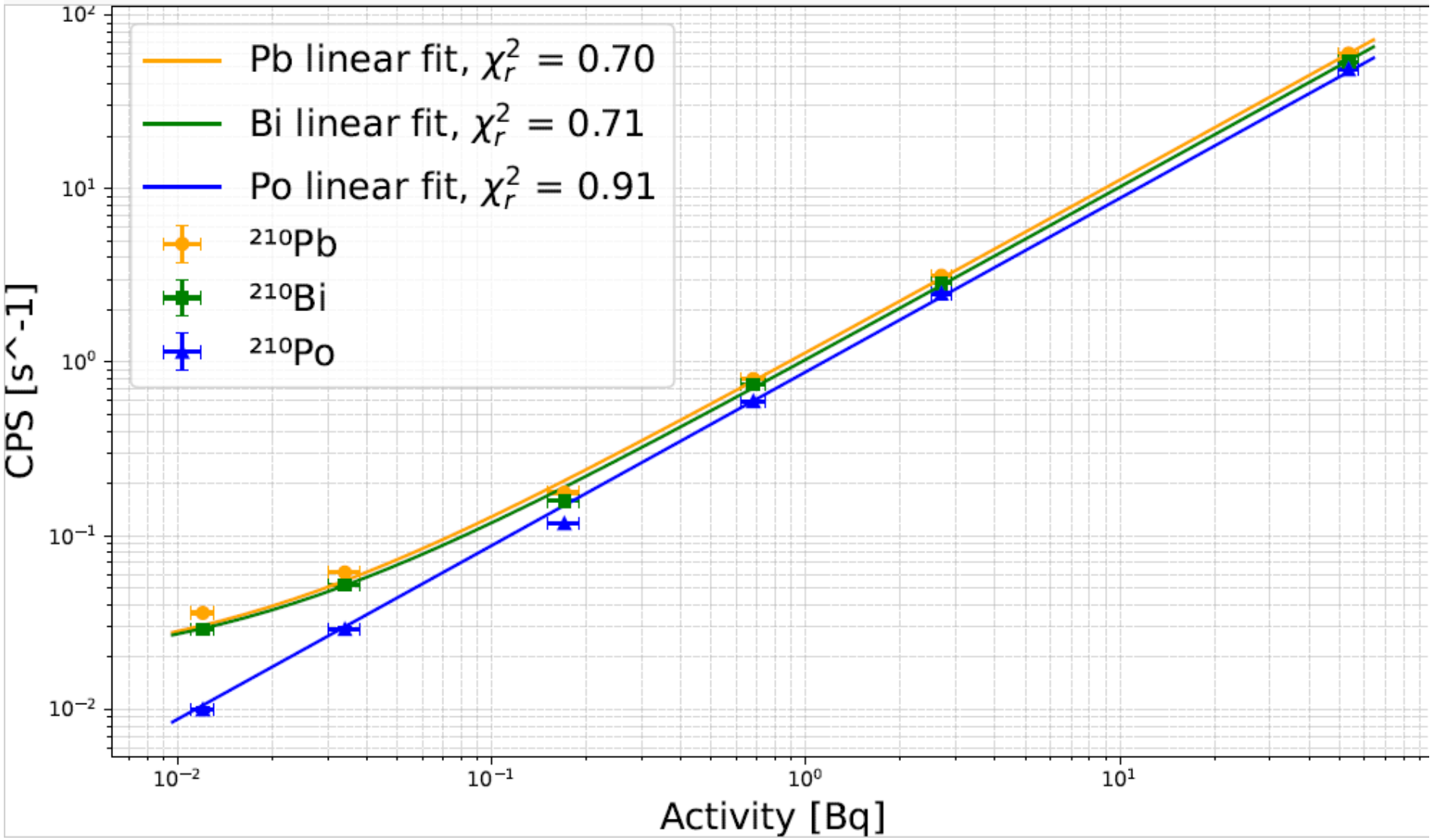
PSA optimum



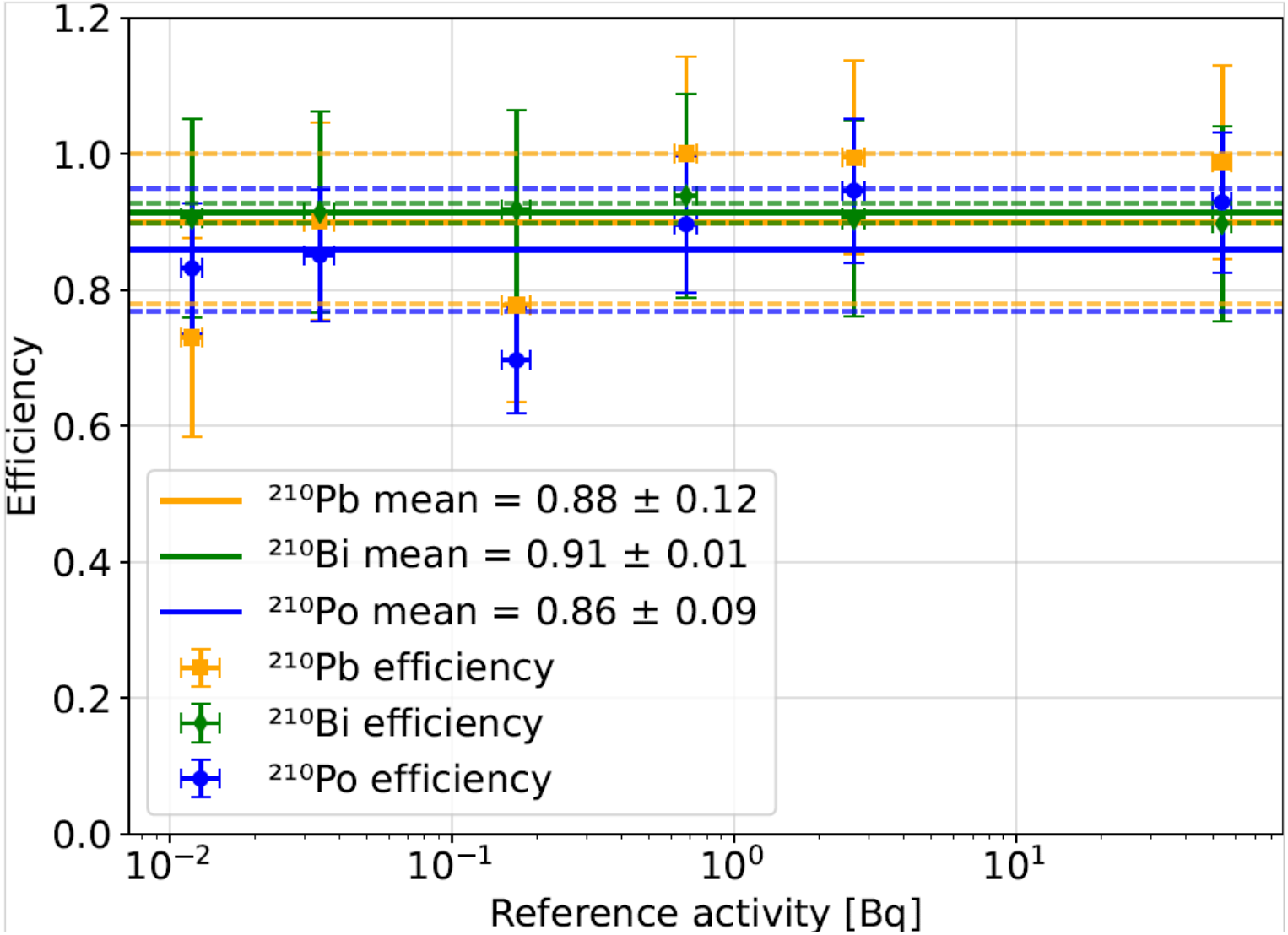
RED STAR:
Minimization of total particle misID, resulting in higher leakage of β→α

BLACK STAR:
Symmetrical misID rateo, lower β→α leakage

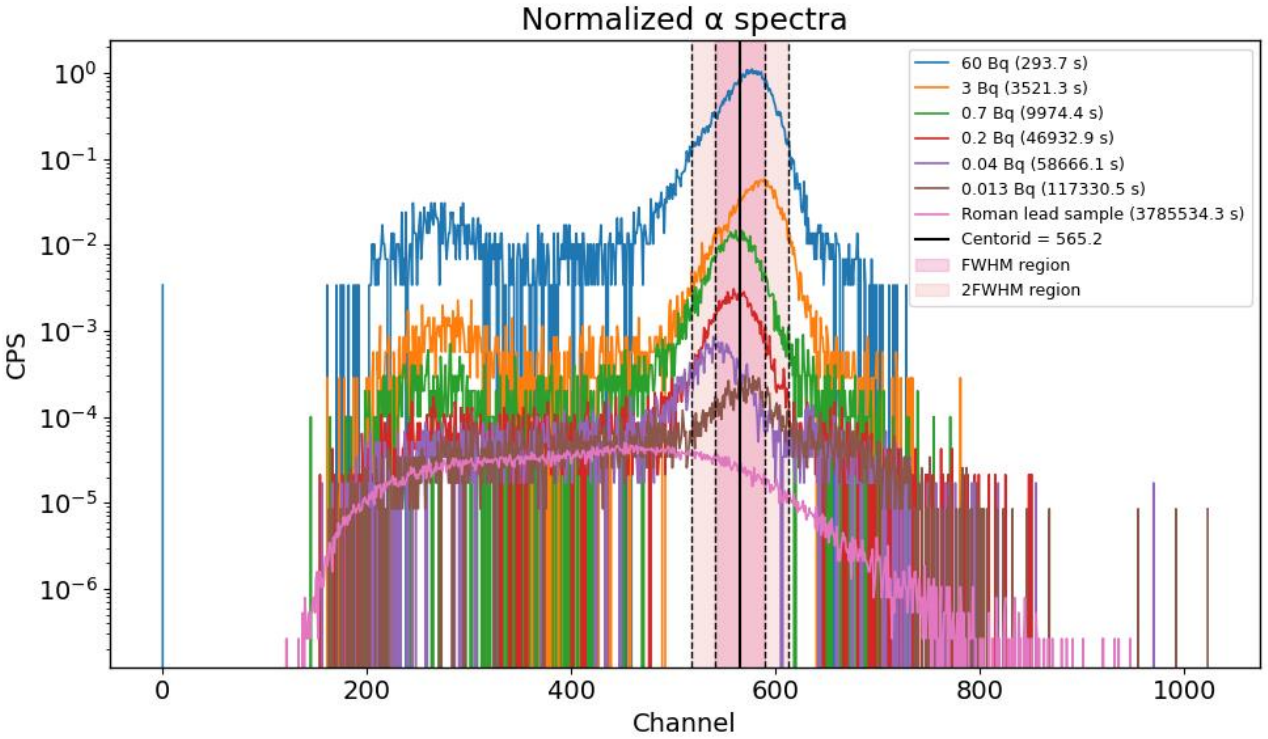
Linearity

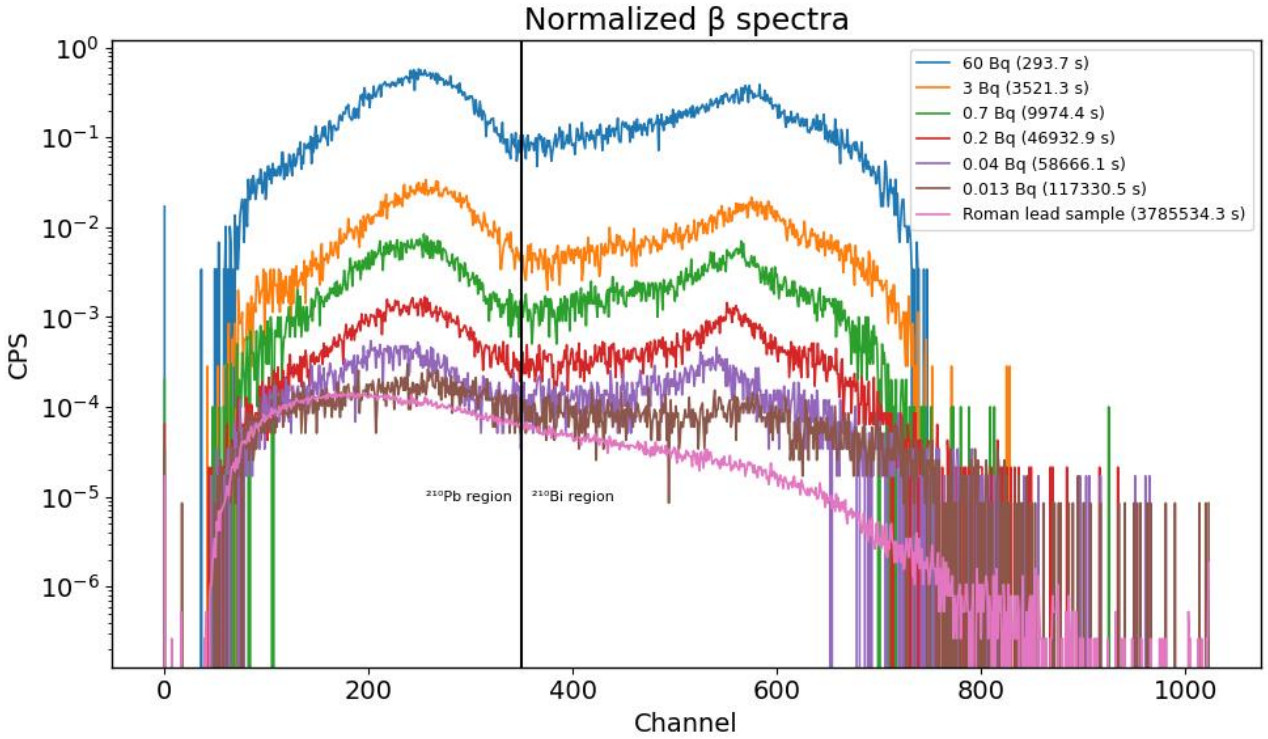

Pb linear fit, $\chi_r^2 = 0.70$
Bi linear fit, $\chi_r^2 = 0.71$
Po linear fit, $\chi_r^2 = 0.91$
^{210}Pb
^{210}Bi
^{210}Po
CPS [s^-1]
Activity [Bq]
10^{2}
10^{1}
10^{0}
10^{-1}
10^{-2}

Efficiency


1.2
1.0
0.8
0.6
0.4
0.2
0.0
Efficiency
10^{-2}
10^{-1}
10^{0}
10^{1}
Reference activity [Bq]
^{210}Pb mean = 0.88 ± 0.12
^{210}Bi mean = 0.91 ± 0.01
^{210}Po mean = 0.86 ± 0.09
^{210}Pb efficiency
^{210}Bi efficiency
^{210}Po efficiency

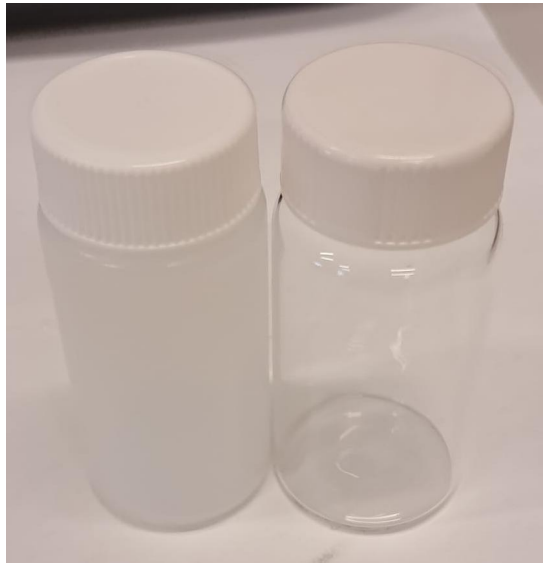
ROIs


Normalized α spectra
CPS
Channel
60 Bq (293.7 s)
3 Bq (3521.3 s)
0.7 Bq (9974.4 s)
0.2 Bq (46932.9 s)
0.04 Bq (58666.1 s)
0.013 Bq (117330.5 s)
Roman lead sample (3785534.3 s)
Centorid = 565.2
FWHM region
2FWHM region


Normalized β spectra
CPS
Channel
60 Bq (293.7 s)
3 Bq (3521.3 s)
0.7 Bq (9974.4 s)
0.2 Bq (46932.9 s)
0.04 Bq (58666.1 s)
0.013 Bq (117330.5 s)
Roman lead sample (3785534.3 s)
^{210}Pb region
^{210}Bi region

Set-up for a ^{210}Pb LSC campaign

WHAT IS A QUANTULUS SAMPLE?
20 ml vials containing a cocktail of mixed LS and liquid samples.
Usual LS to sample ratio are:
15 ml LS + 5 ml liquid sample
12 ml LS + 8 ml liquid sample (the one we used)



2 Quantulus vials made of PTFE (left) or glass (right)

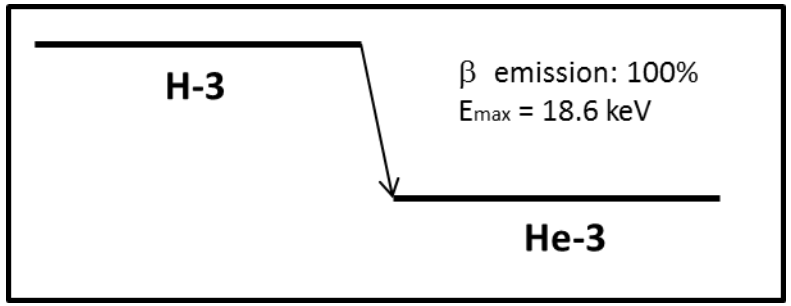
1. Finding the optimal detection set-up
   - Testing different ratio of LS vs liquid samples
   - Testing quenching effects due to sample acidity and concentration
2. Finding the optimal PSA set-up to allow correct α/β discrimination
   - Preparing 2 chemically identical samples to the optimized one
   - Spiking samples with pure α/β emitters
   - Scanning different PSA values (more on that later!)
3. Calibration curve with sample spiked with known activity of ^{210}Pb to identify ROIs and check detector linearity.
4. Samples measurements.

Preparation of PWO samples

1. 0.2 g of PWO are dissolved in a 10 ml aqueous solution of NaOH 1:10.
2. The process is fairly rapid, no ultrasound heating needed.
3. 8 ml of dissolved sample are mixed with 12 ml of UltimaGoldAB LS.

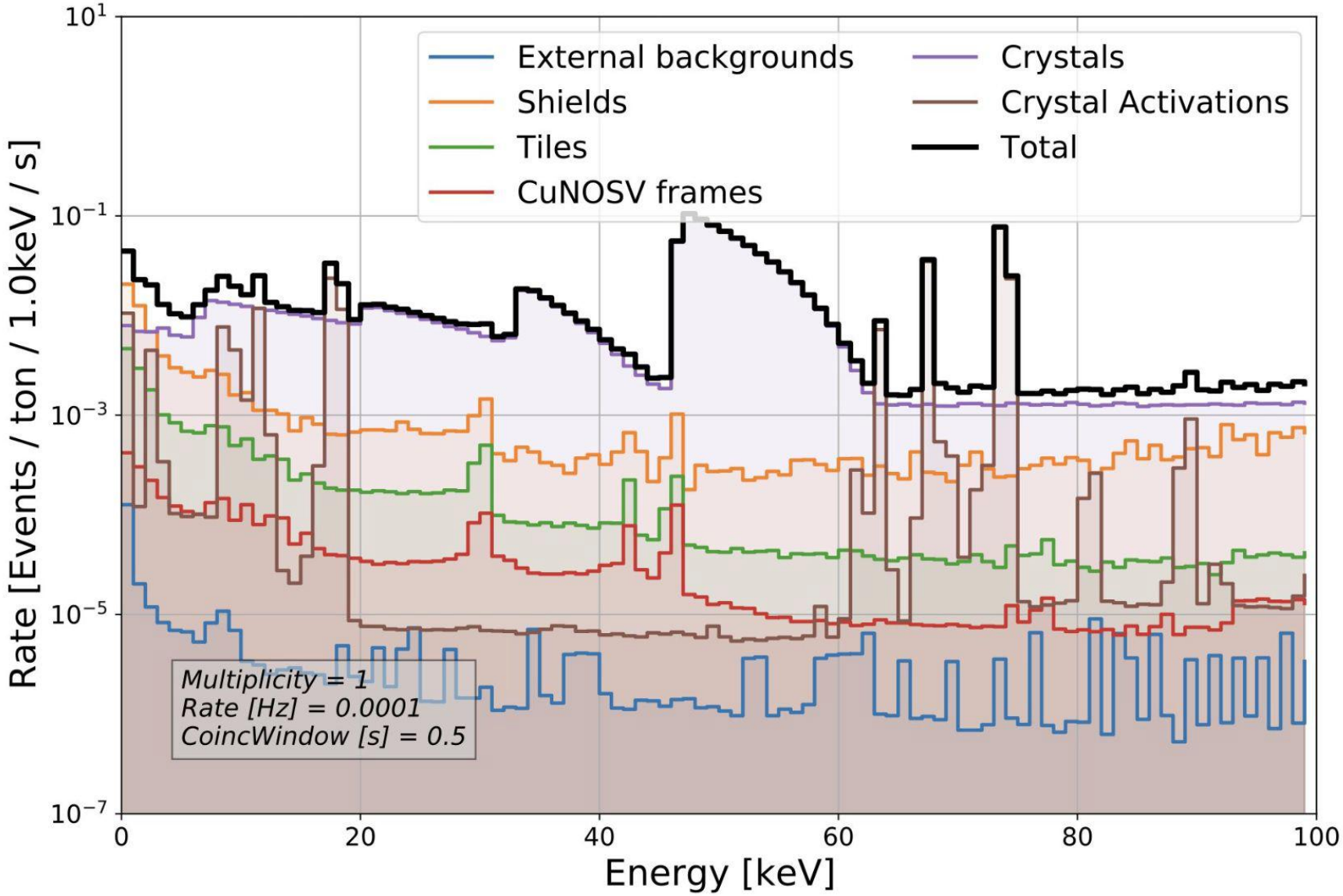
Blank samples prepared with a CMS PWO crystal that resulted slightly contaminated with ^{3}H (< 10-20 Bq/kg).

More calibration point needed since few ^{3}H standard were available.

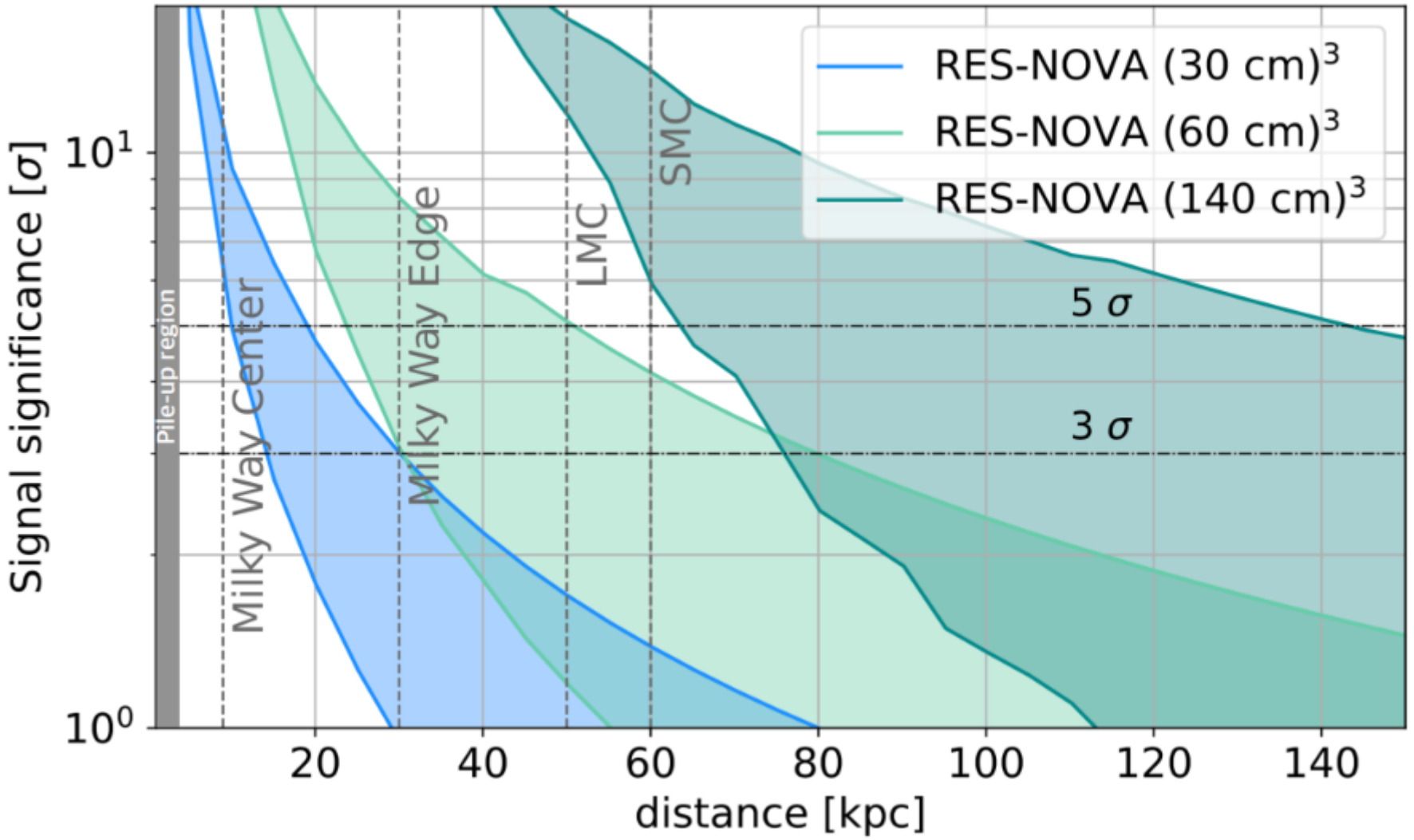


Dissolved PWO sample
(because apparently taking photos of the original crystal was too much to ask of me)

RES-NOVA full Background



RES-NOVA sensitivity



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