ジルコニウム９６を用いたニュートリノを放出しない二重ベータ崩壊事象の探索XXVI
～ニュートリノを放出する二重ベータ崩壊事象観測実験の改良について～

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Miyagi University of Education　Y. Fukuda

Kamioka Observatory, ICRR, Univ. of Tokyo　S. Moriyama, K. Hiraide

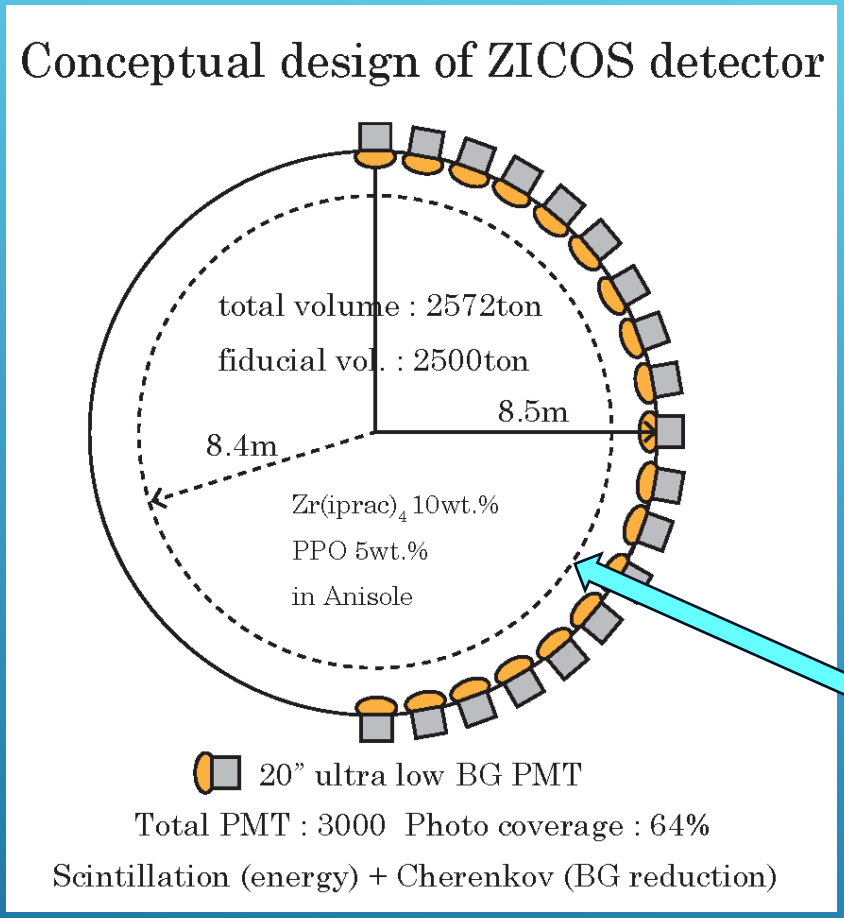
University of Fukui　I. Ogawa

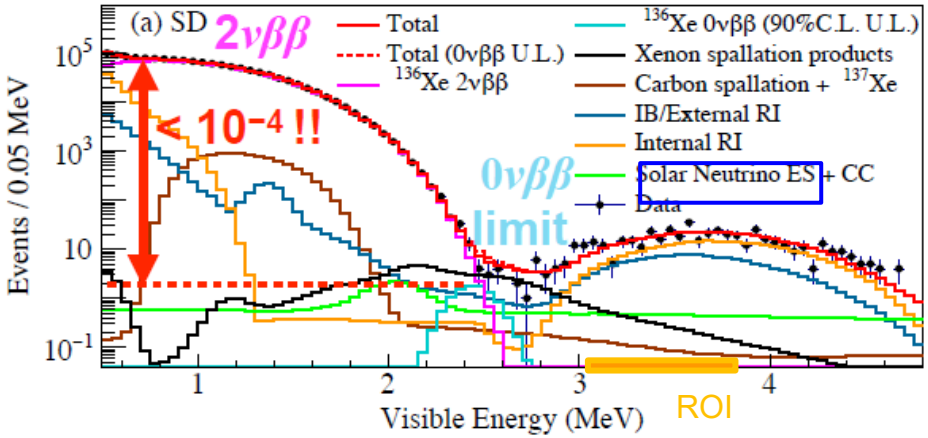
Tokyo University of Science　T. Gunji

University of Tokyo　S. Kurosawa

University of Toyama　Y. Nakano, S. Kato

ZICOS experiment for ^{96}Zr 0νββ observation





I.Shimizu, Plenary talk at Neutrino2024 conference.

Liquid scintillator loaded ^{96}Zr

NEMO3 : $T_{1/2}^{0\nu} > 9.1 \times 10^{21}$ yrs

^{96}Zr : 1ton (nat.) → 1/20 BG → 20 ton(50 % enrich)

$T_{1/2}^{0\nu} > 2 \times 10^{26}$ yrs → ~1×10^{27}yrs → ~1×10^{28}yrs

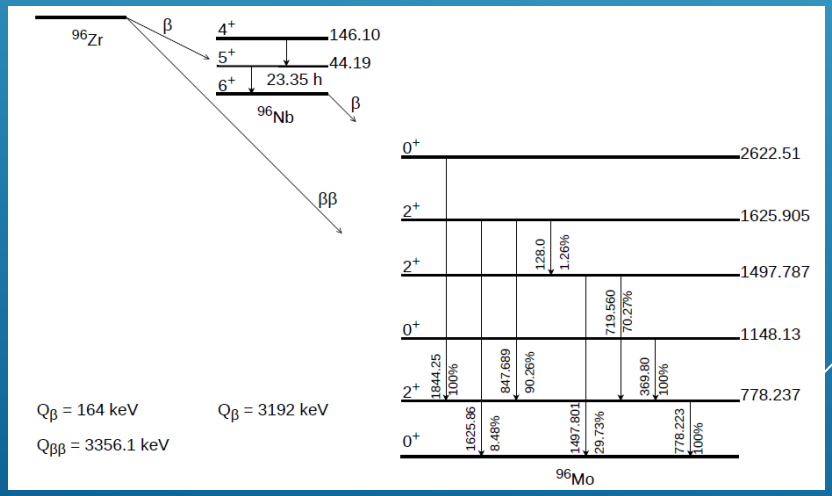
LS with In(iPrac)$_3$ could be used for solar CNO ν measurement.

Recent news from Russia for ^{96}Zr enrichment

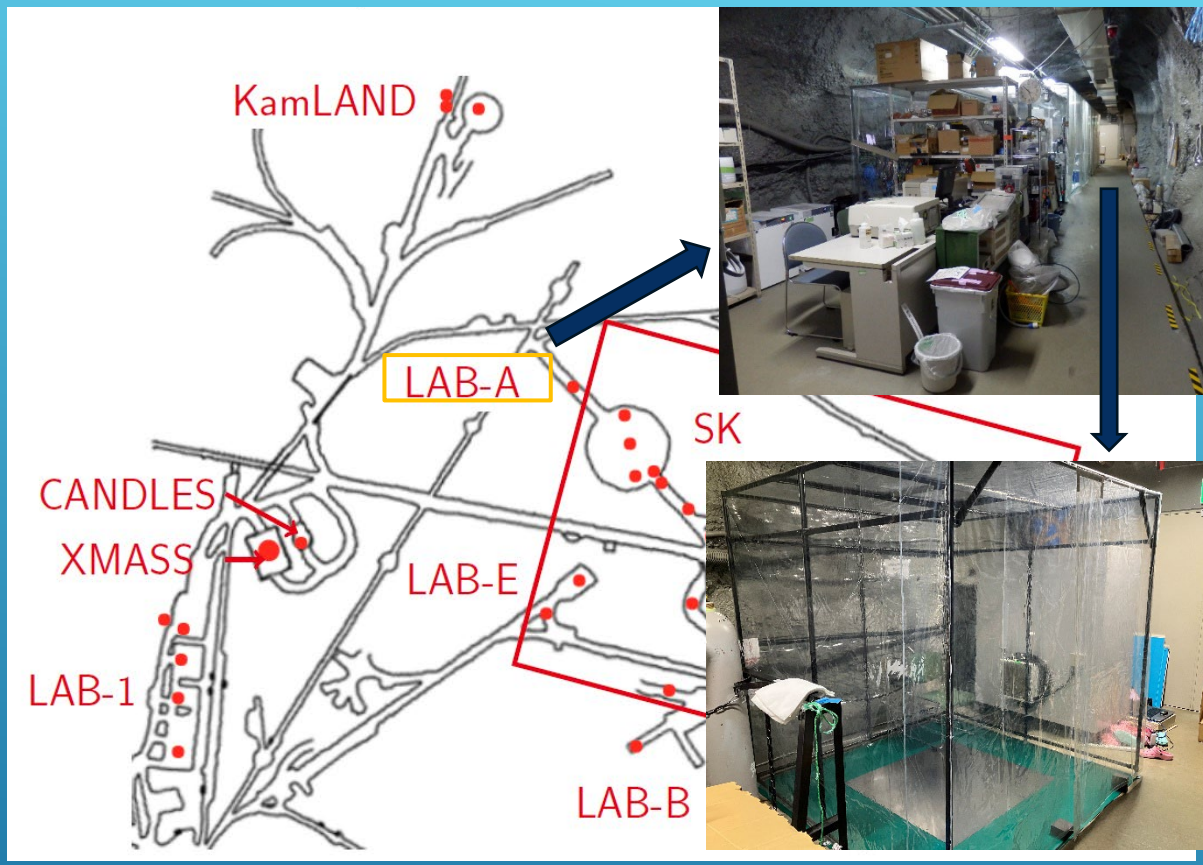
First investigation of ^{96}Zr samples enriched by the gas-centrifuge method for the use in rare-decay studies (arXiv:2605.13215v1)

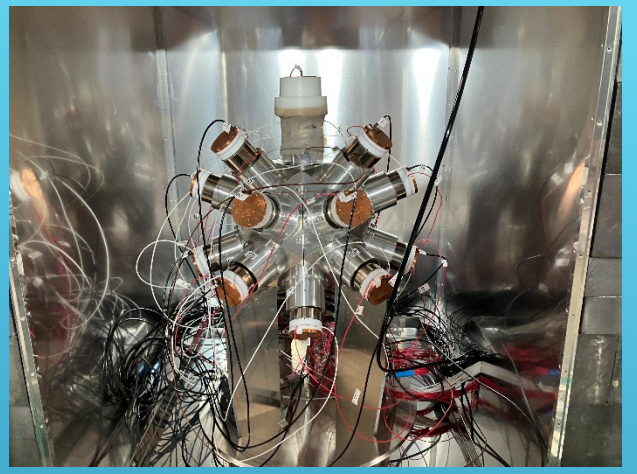
Abstract

For the first time, the isotope ^{96}Zr has been produced by the gas-centrifuge method (enrichment is 88.28% and full mass of ^{96}Zr is 179.816 g). The level of radionuclide impurities of the samples and thus their suitability for rare-event search have been investigated using three low-background HPGe detectors. The most stringent limit, obtained at sea level, on the half-life of the double beta decay of ^{96}Zr to the 0+1 excited state (1148 keV) of ^{96}Mo has been set $T_{1/2}(0\nu + 2\nu) > 3.9 \times 10^{19}$ yr (90% C.L.).



2ν-ZICOS for measurement of ^{96}Zr 2νββ events



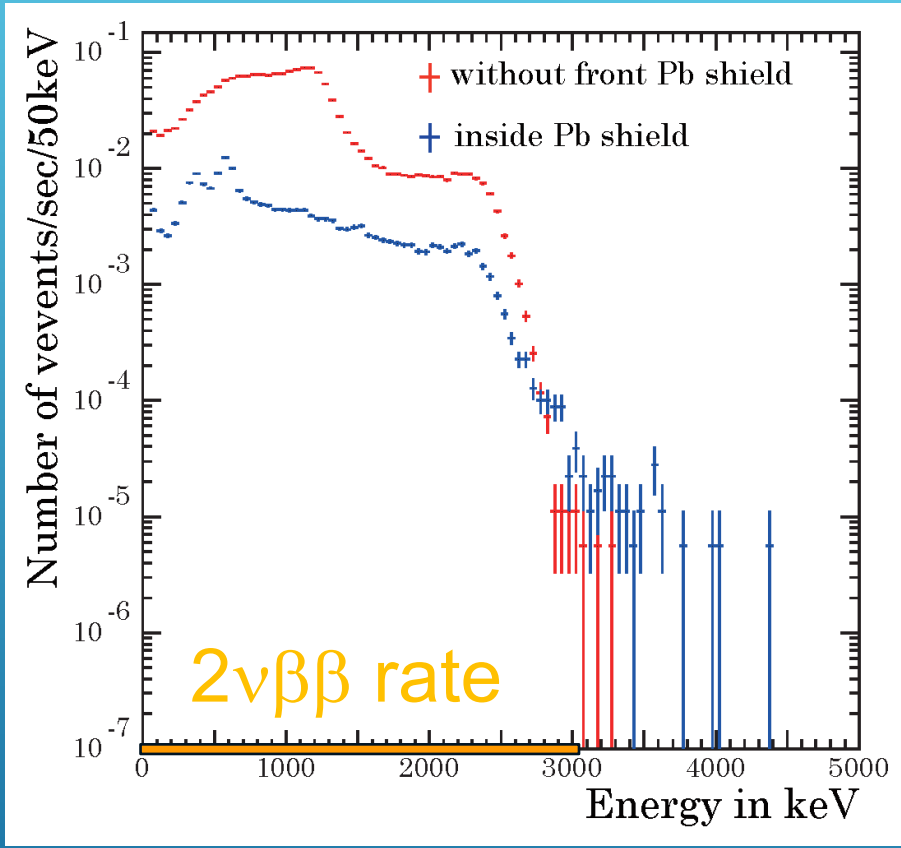




- 650 Pb blocks was used for radiation shield with 15cm thickness.
- N_2 gas was circulating within Al box.
- Background run without front Pb shield : 100800sec inside Pb shield : 180000 sec

The first data taking implemented over 50 hours from November 22nd to 25th.

Background events observed in Pb shield



- 2.615MeV γ events from ^{208}Tl were observed.
- Despite of expected 10^{-4} reduction for environmental γ due to 15cm Pb shield, BG rate was 3 order magnitude higher than expected rate.
- Shoulder structures were found around 2.6MeV

Measured event rate is over 4 order magnitude higher than expected 2νββ event rate (~10^{-7}event/s/50keV)

Background γ events from PMT mounting jig

https://doi.org/10.2116/bunsekikagaku.41.5_T83

A5052/A5056　U ~0.1ppm　Th ~0.5ppm

^{232}Th : ~2.0×10^{-4}Bq/g　^{238}U : ~1.2×10^{-3}Bq/g

Total mass of PMT mounting jig :4680g

Assuming radiative equilibrium

^{232}Th : ~1.0Bq　^{238}U : ~5.6Bq

Background γ events from Quartz GE214 flask

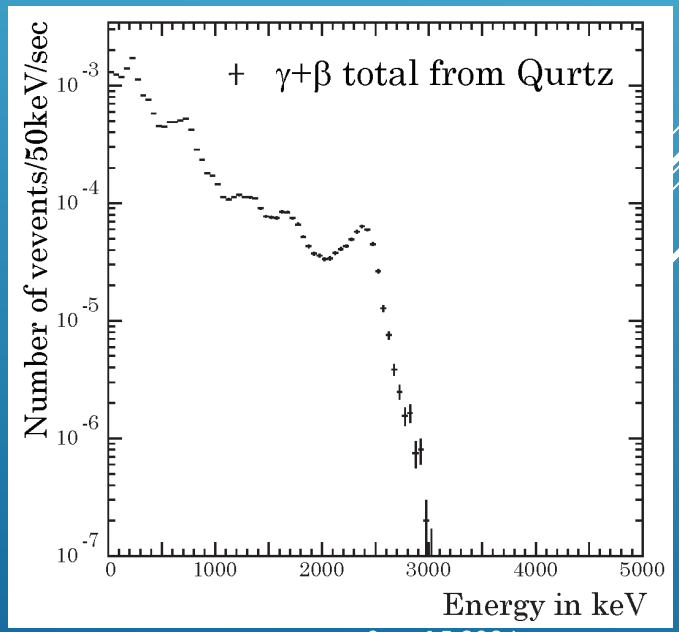
単位：ng/g

試料名	K	Th	U
RQ200	330	42	64
GE214	180	15	29
定量下限	5	1	1

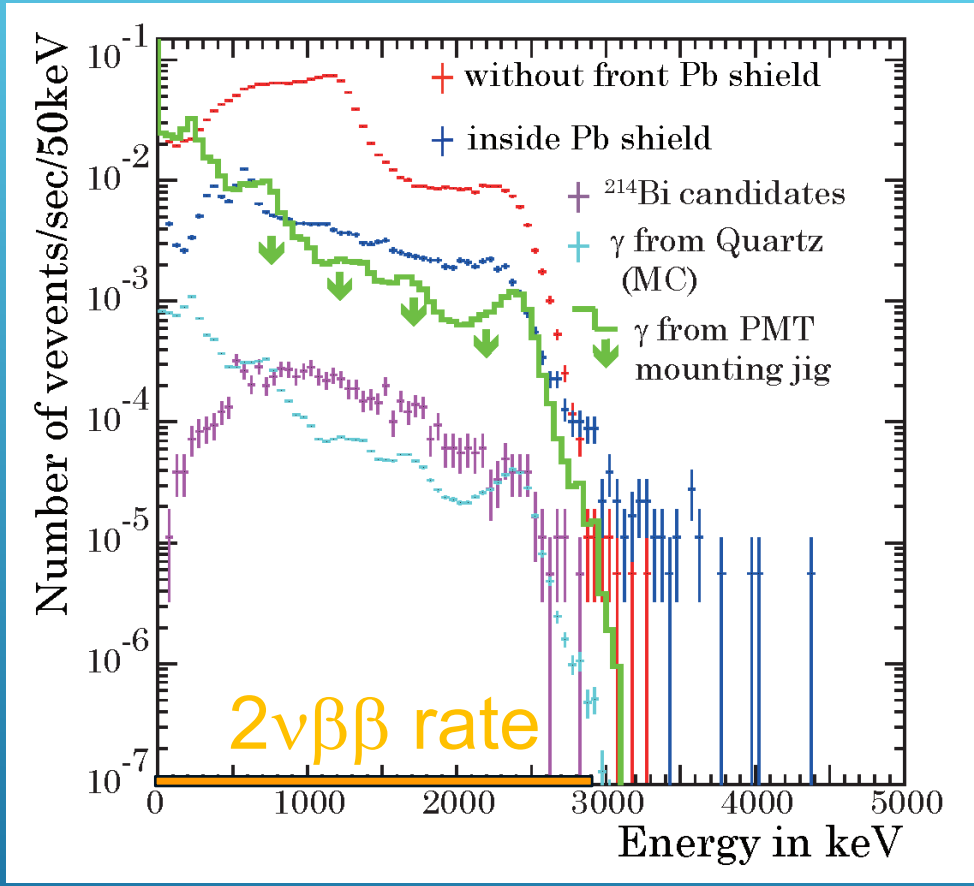
^{232}Th : 3.6×10^{-2}Bq

^{238}U : 2.1×10^{-1}Bq

^{40}K : 3.4×10^{-3}Bq



Background events observed in Pb shield



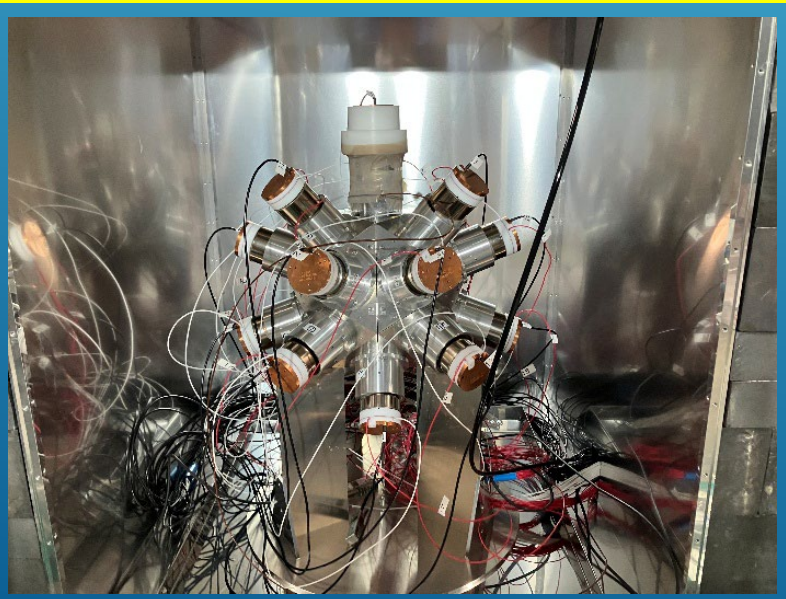
- About 10000 times larger events due to γ from PMT mounting jig.
- About 1000 times larger events due to γ from Quartz
- Same level of ^{214}Bi events exist in LS due to ^{226}Ra contamination and ^{222}Rn intrusion.

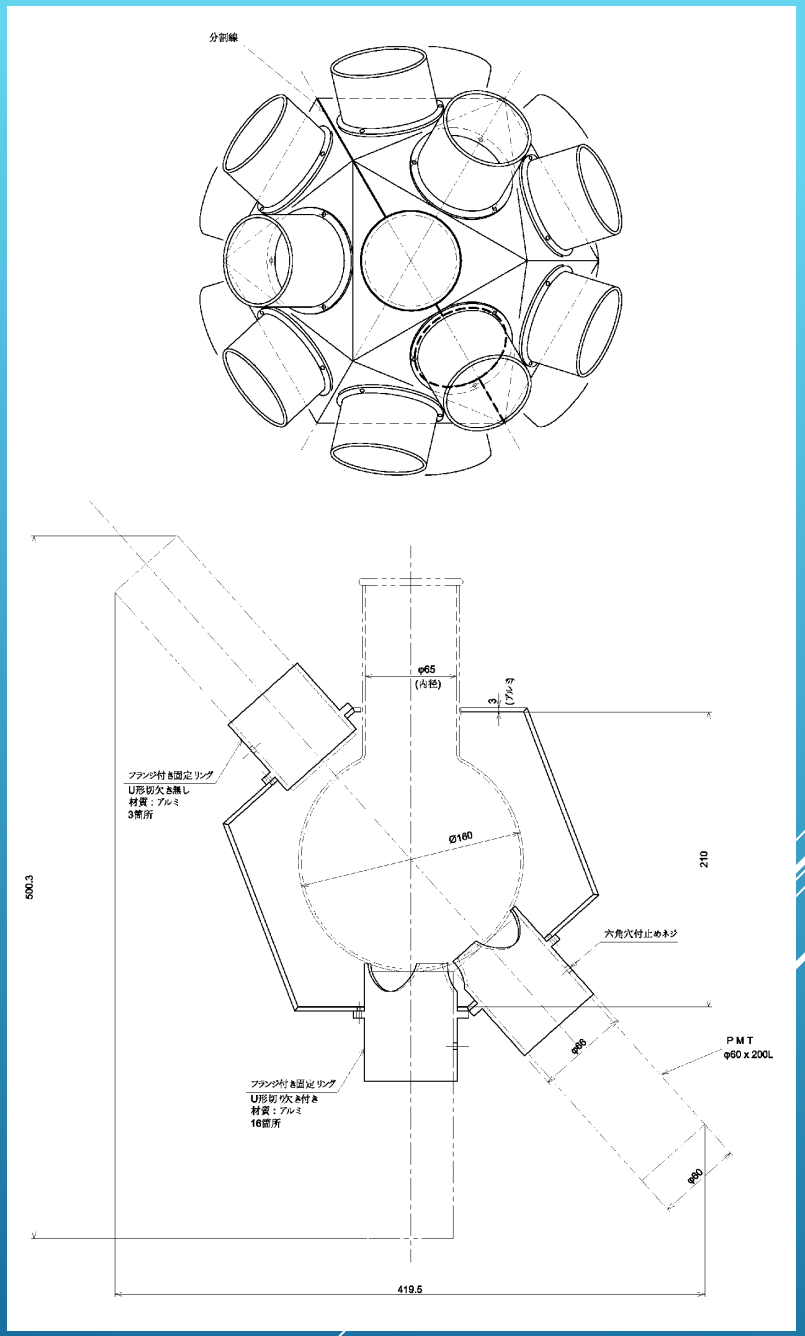
Those backgrounds will be removed by replacing clean materials.

Plan to improvement ①

Using oxygen-free copper for R10789-11 PMT mounting jig instead of ordinal aluminum.

Ge detector and quick BG measurement will be done right after purchase and deliver new mounting jig.





Plan to improvement ②

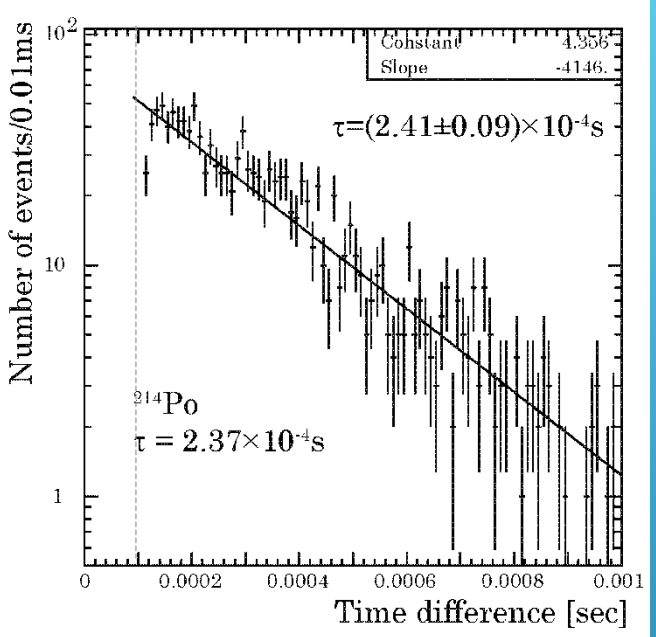
Using Synthetic quartz glass SH110 from Shin-Etsu Quartz Products Co., Ltd. instead of GE214

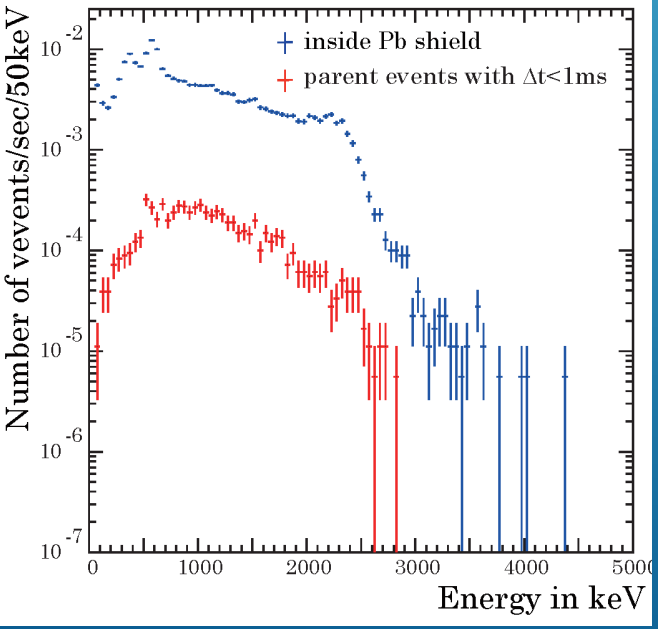
品種名	金属不純物(ppm)								OH基
	Na	K	Li	Mg	Cu	Fe	Ca	Al	
TSC-3	0.3	0.2	0.2	<0.05	<0.05	0.05	0.4	15	170
TSC-4	0.2	0.08	<0.05	<0.05	<0.05	0.1	<0.05	8	170
HERALUX-E	0.3	0.4	0.6	0.05	<0.05	0.1	0.5	15	20
HERALUX-E-LA	0.05	0.1	0.05	0.05	<0.05	0.1	0.5	15	20
HSQ330	0.1	0.3	0.6	<0.05	<0.05	0.1	0.5	15	20
HSQ130	0.1	0.3	0.6	<0.05	<0.05	0.1	0.5	15	<10
HSQ135	0.9	0.7	0.6	<0.05	<0.05	0.3	0.5	15	<10
SO-210	0.4	0.4	0.4	0.05	<0.05	0.2	0.2	15	—
OM-100	0.2	0.4	0.6	0.05	<0.05	0.2	1.2	15	—
SH110	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	250
SH100	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<1
SH200	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	800
SH120	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<1

Al	As	B	Ca	Cd	Cr	Cu	Fe	K	Li	Mg	Mn	Na	Ni	P	Sb	Ti	Zr	OH	Type
14	<0.002	<0.2	0.4	<0.01	<0.05	<0.05	0.2	0.6	0.6	0.1	<0.05	0.7	<0.1	<0.2	<0.003	1.1	0.8	<5	

Measure BG using Ge detector right after purchase of quartz (it takes some months to make a flask).

Plan to improvement ③





- 0.1ms< Δt<1ms 1250events (+~650events). No event Δt<0.1ms due to dead time of V1742 digitizer.
- This will be solved by using V1751 digitizer (no dead time acquisition)

Not only ^{214}Bi -^{214}Po events but ^{212}Bi-^{212}Po events will be removed by time diff. cut due to $T_{1/2}$~3 ×10^{-7}sec if exist.

Schedule

- U/Th in PMT mounting jig should be measured by Ge detector and BG γ also should be simulated.
- New PMT mounting jig and quartz round flask will be prepared until the end of this year and next February, respectively. Ge detector measurement will be done right after purchase of those materials.
- Quick BG measurement using plastic scintillator will be done just after deliver them.
- Next DAQ with a few days same as last November will be conducted at next March.

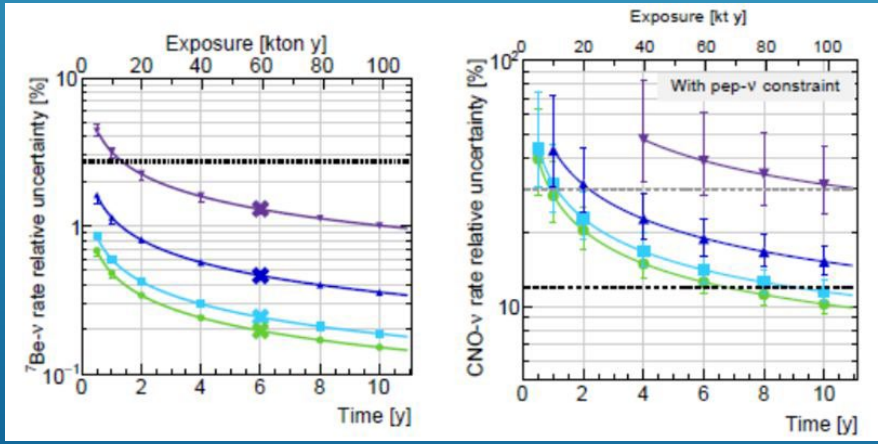


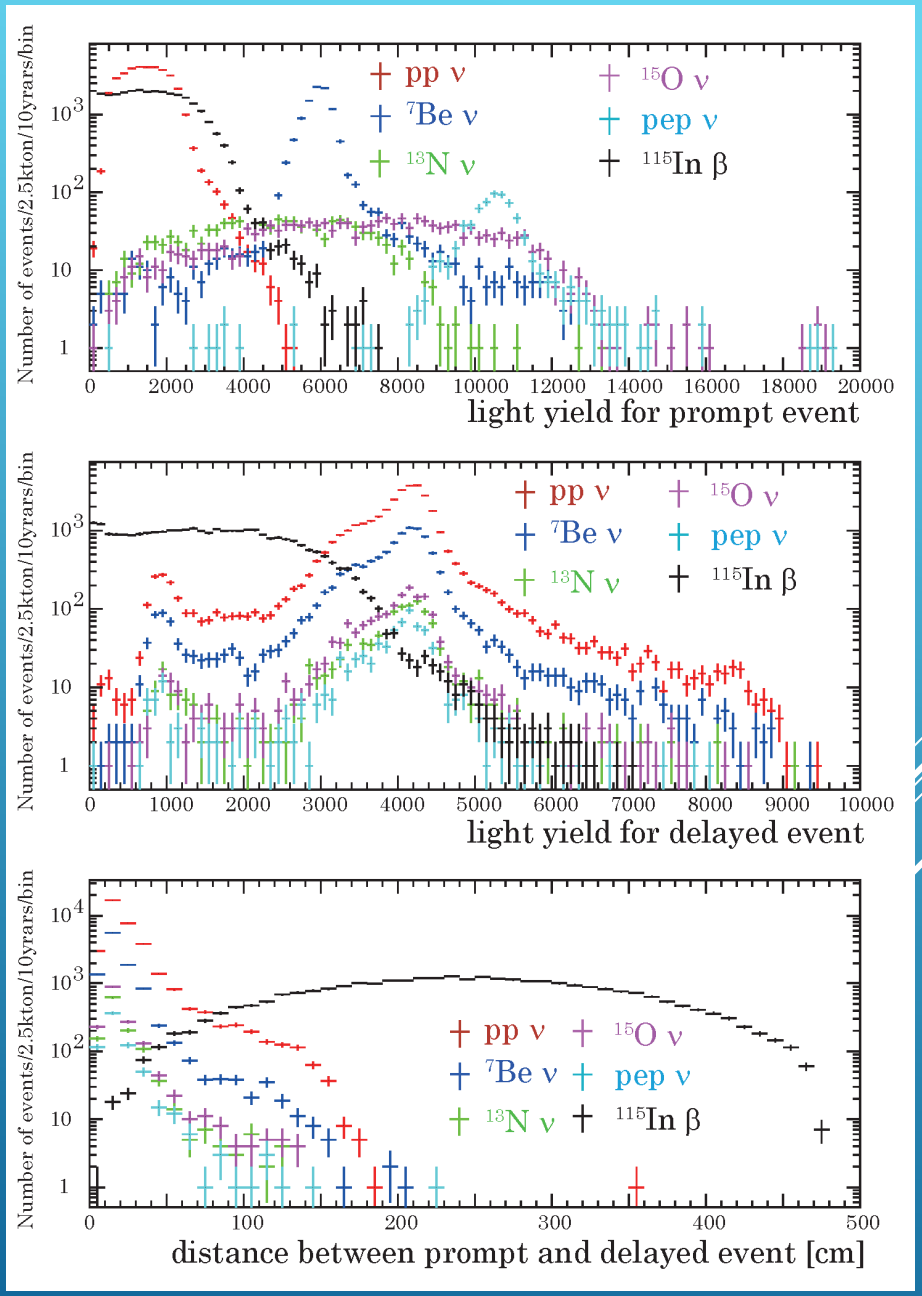
For the enrichment of ^{96}Zr, Japan Nuclear Fuel Ltd has already been interested in the gas-centrifuge and continue to discuss…

backup

Solar CNO ν observation using LS with In(iPrac)$_3$

- Assuming 2.5kton LS detector with R=8.4m, including ^{115}In 50 ton.
- ^{7}Be ν 1% (JUNO 0.4%)
- CNO ν 1.8% (JUNO 10~15%)





Background γ events from Quartz GE214

単位：ng/g

試料名	K	Th	U
RQ200	330	42	64
GE214	180	15	29
定量下限	5	1	1

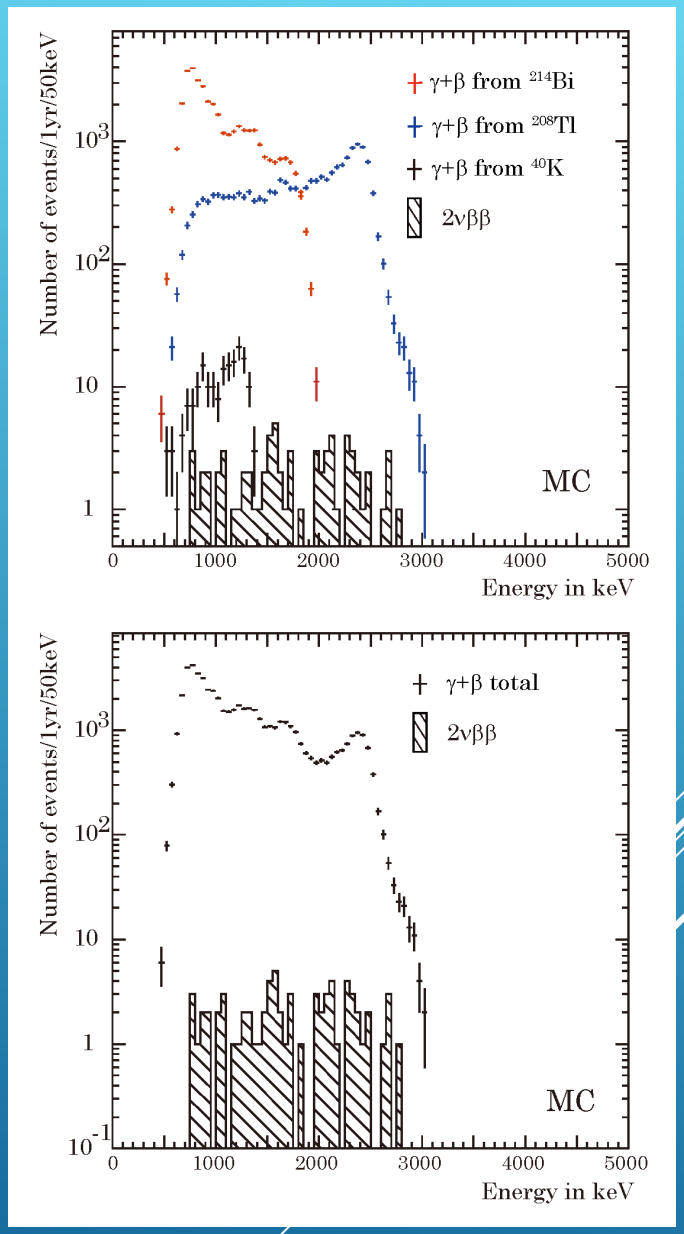
^{232}Th : 6.09 × 10^{-5}Bq/g

^{238}U : 3.58 × 10^{-4}Bq/g

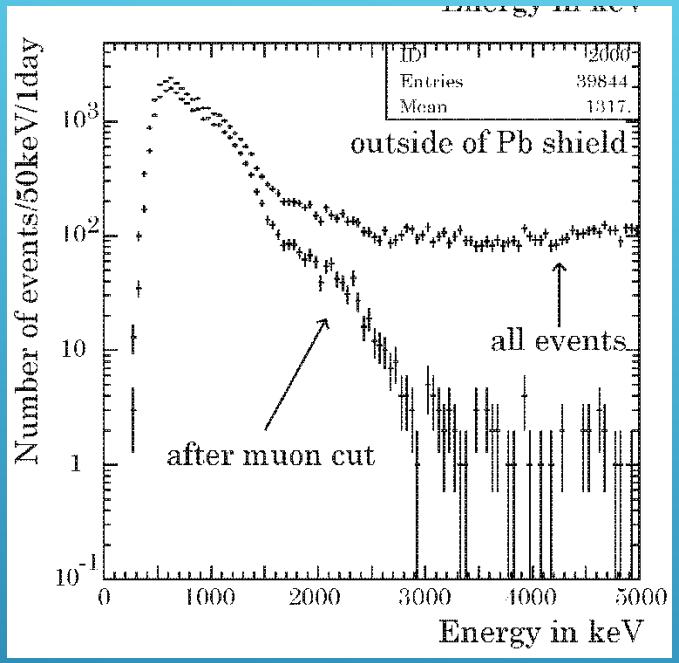
^{40}K : 5.59 × 10^{-6}Bq/g

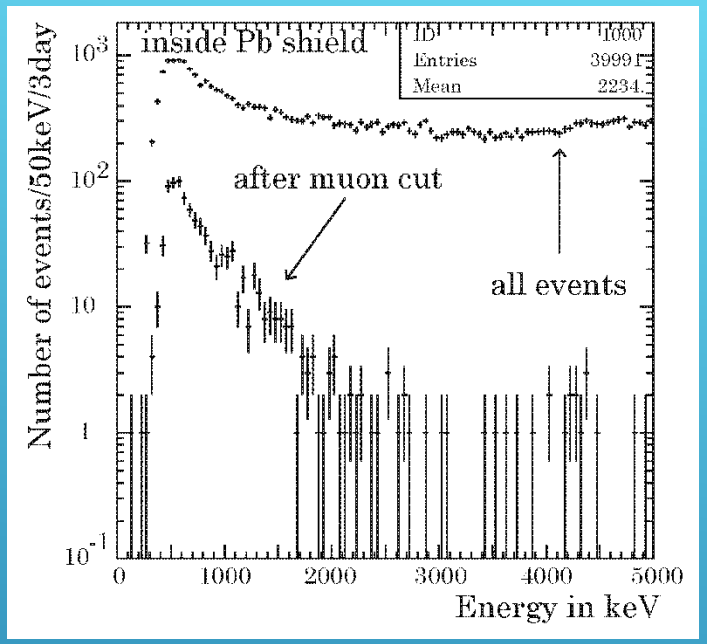
3 order magnitude higher level

Amount of observed BG events might not be explained by Quartz only.



Energy spectra using Quartz vial detector





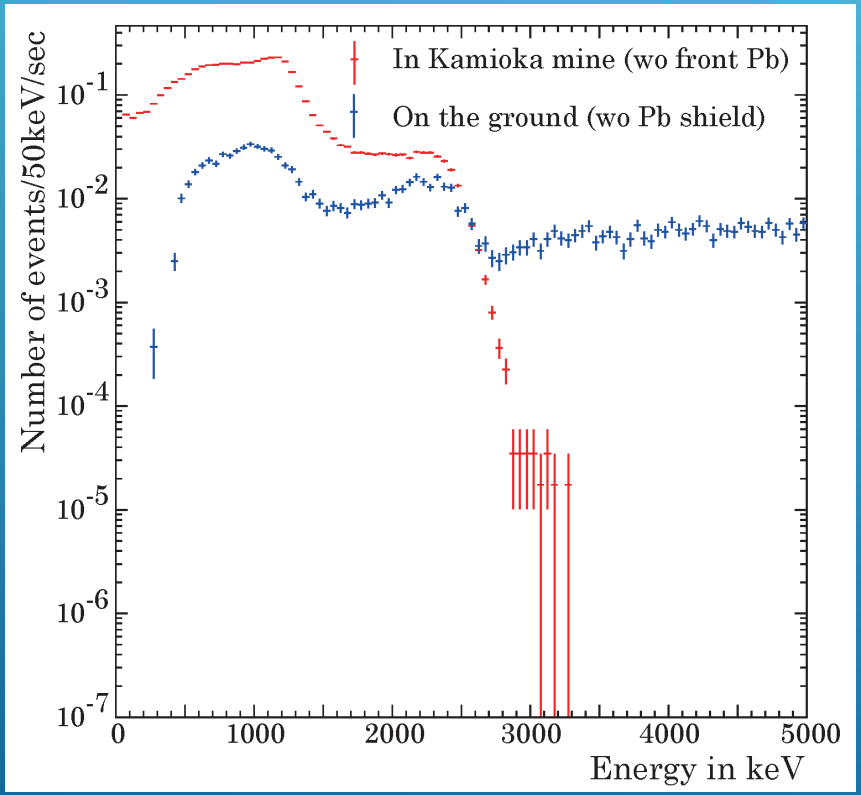
Outside of shield

Inside of shield

- Compton scattering gammas from environmental ^{40}K and ^{208}Tl decay were observed for outside of Pb shield.
- Some background events were found inside of Pb shield.
- Assuming these events from U/Th in LS, we should search for ^{214}Bi -^{214}Po candidates ($\tau=1.643\times10^{-4}$sec)

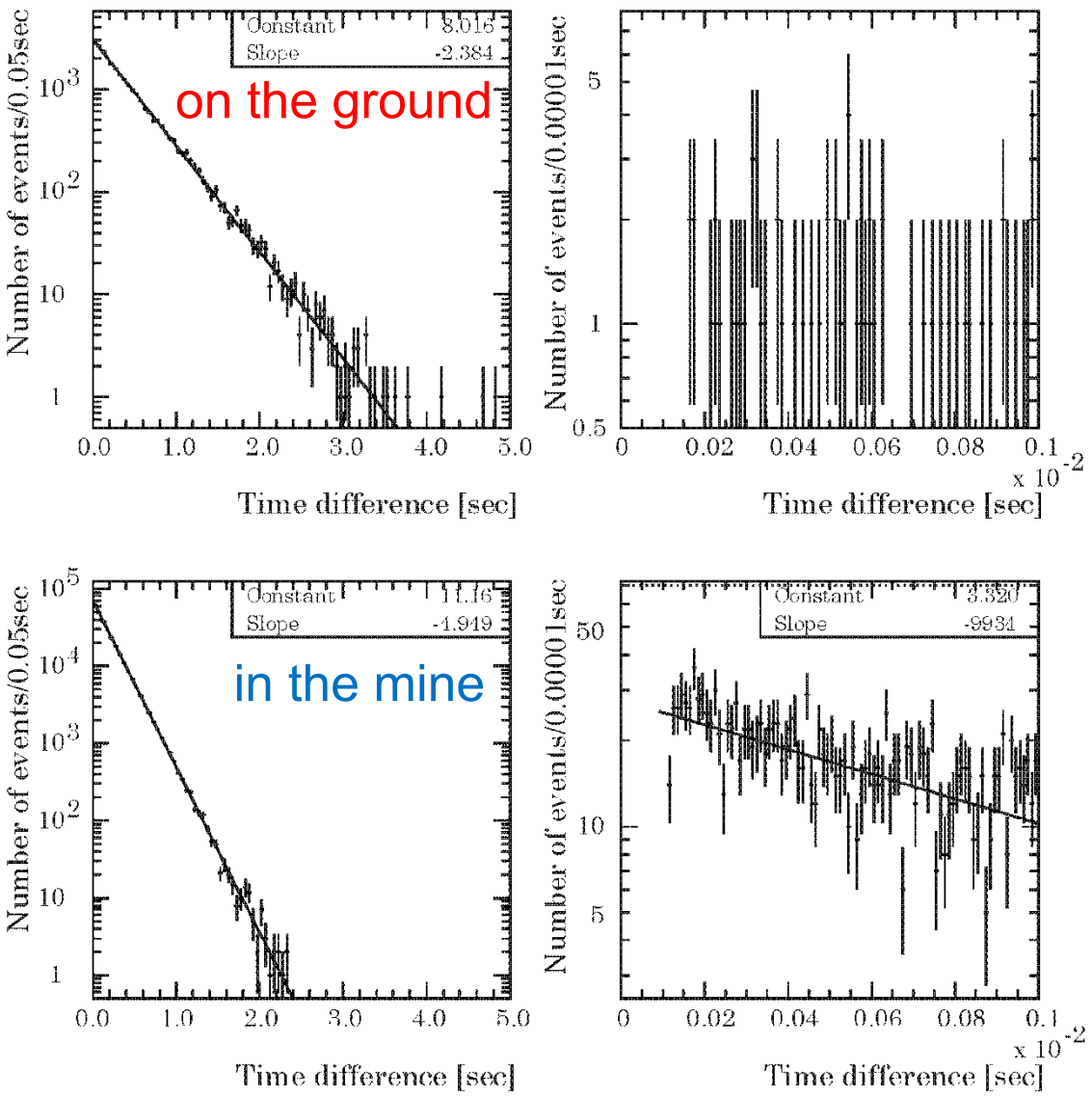
Data taking in 2025

- Period : November 22nd – 26th 2025
- Background run without front Pb shield : 100800sec
- Background run inside Pb shield : 180000 sec



- Most of muons observed on the ground was disappeared.
- Environmental γ events from ^{40}K and ^{208}Tl were clearly observed in case of without front Pb shield.
- Amount of U/Th was almost same but ^{40}K was about 10 times higher than the ground. This is due to the difference of trigger threshold.

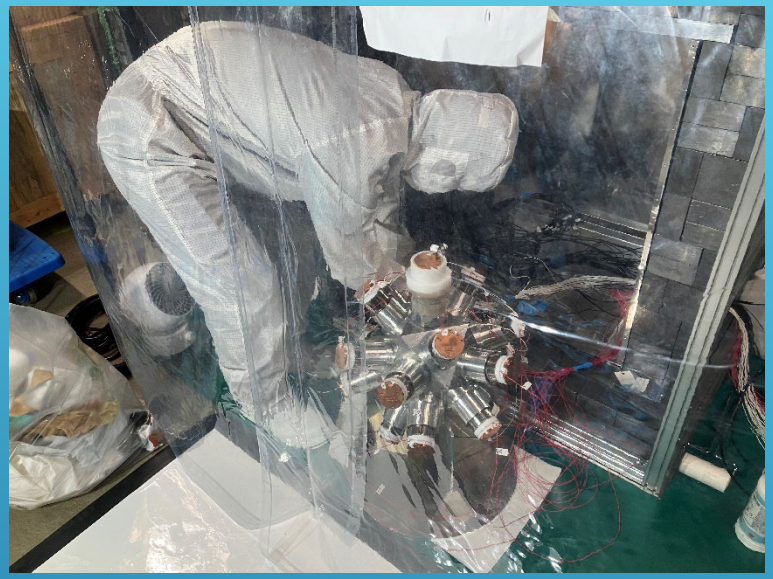
Data quality check

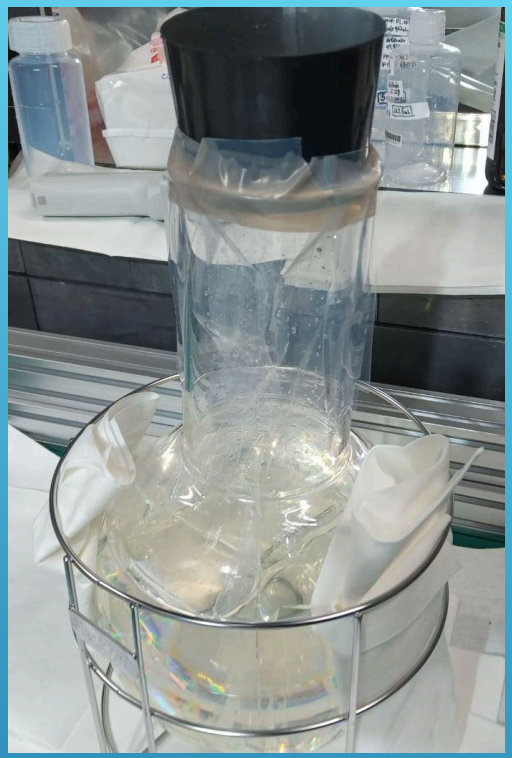


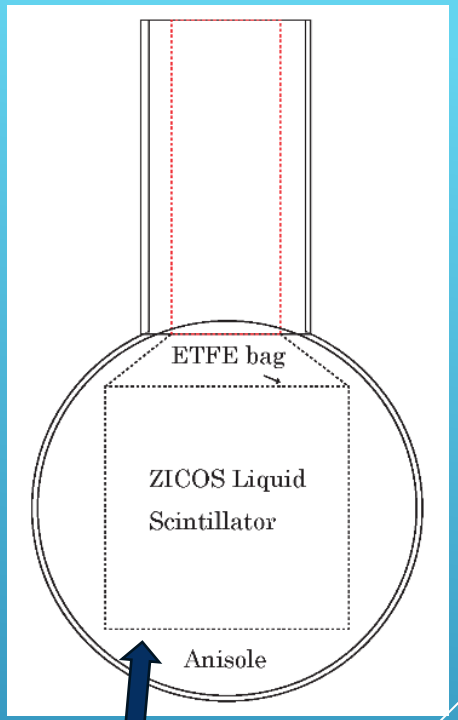
- Trigger rate on the ground was ~2Hz due to cosmic μ.
- No dead time for BG events

- Trigger rate in the mine was ~5Hz due to BG events.
- Nor no dead time for those events.

2ν–ZICOS detector



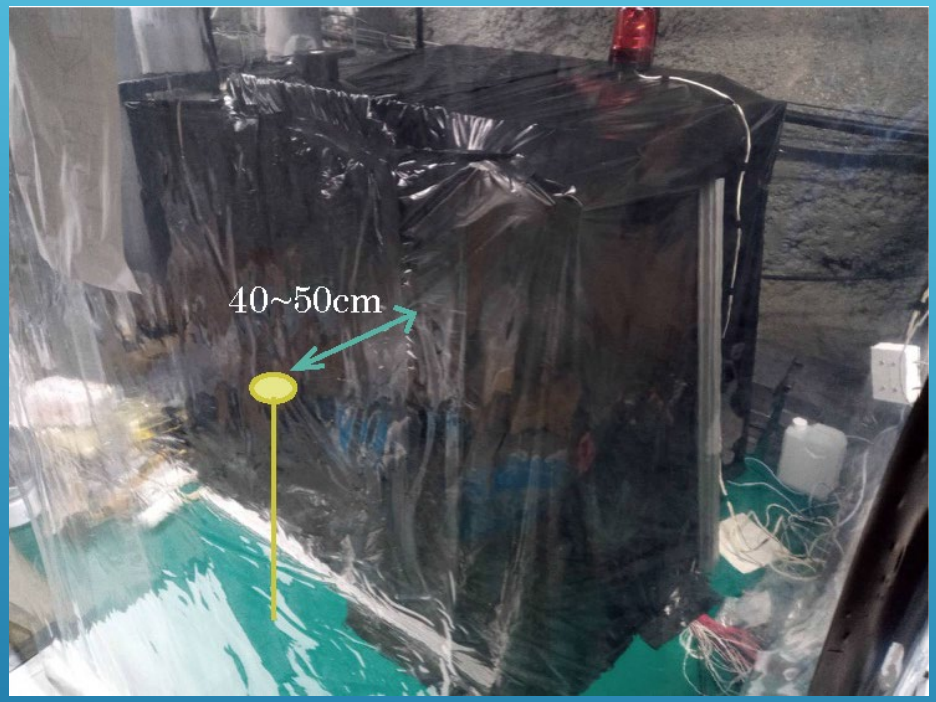




- 20 2” Ultra Low BG PMT Hamamatsu R10789.
- 16 cm diameter round bottom flask using pure Quartz GE214.
- 0.724L of ZICOS LS loaded 69g of $Zr(iPrac)_4$ including ^{96}Zr 0.27g.
- Expected $2\nu\beta\beta$ events ~70/yr

9cm cubic ETFE bag for reducing external BG events

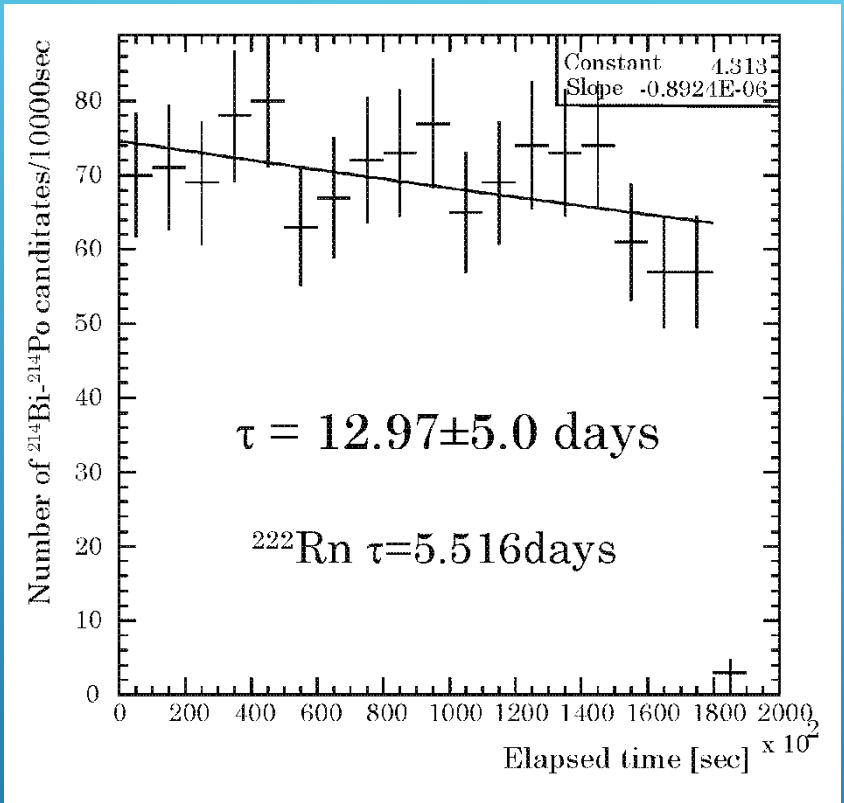
Energy calibration and DAQ for 2ν-ZICOS detector

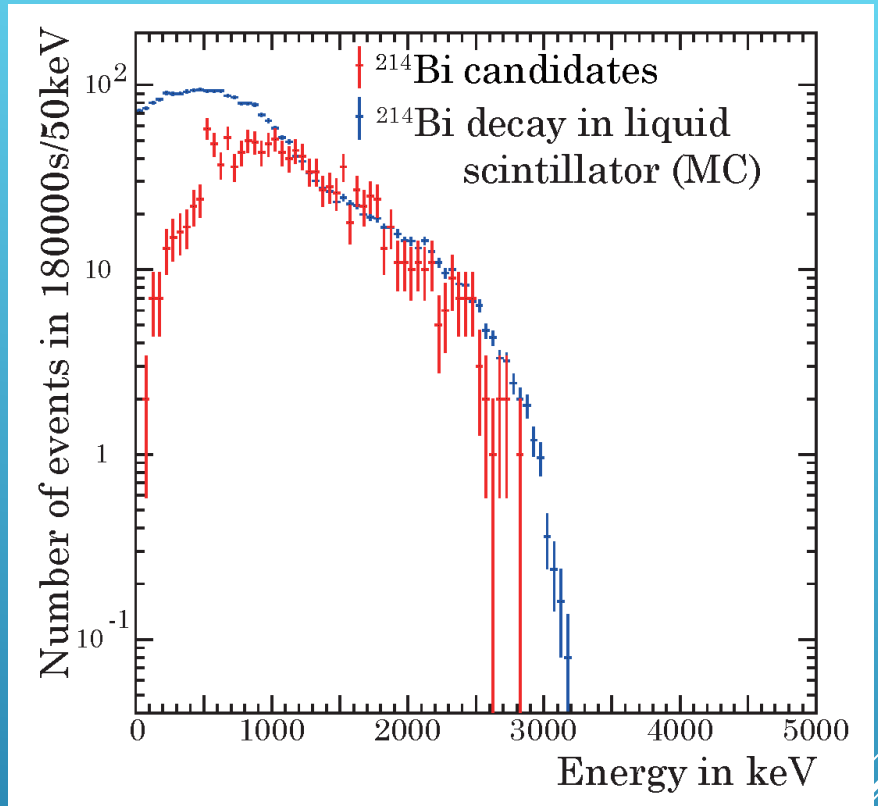




- ^{137}Cs, ^{60}Co and ^{22}Na source were used for energy calibration.
- Those sources were located at front of Al wall without front Pb shield.
- All data were recorded by V1742 digitizer.

^{222}Rn contaminated in liquid scintillator?

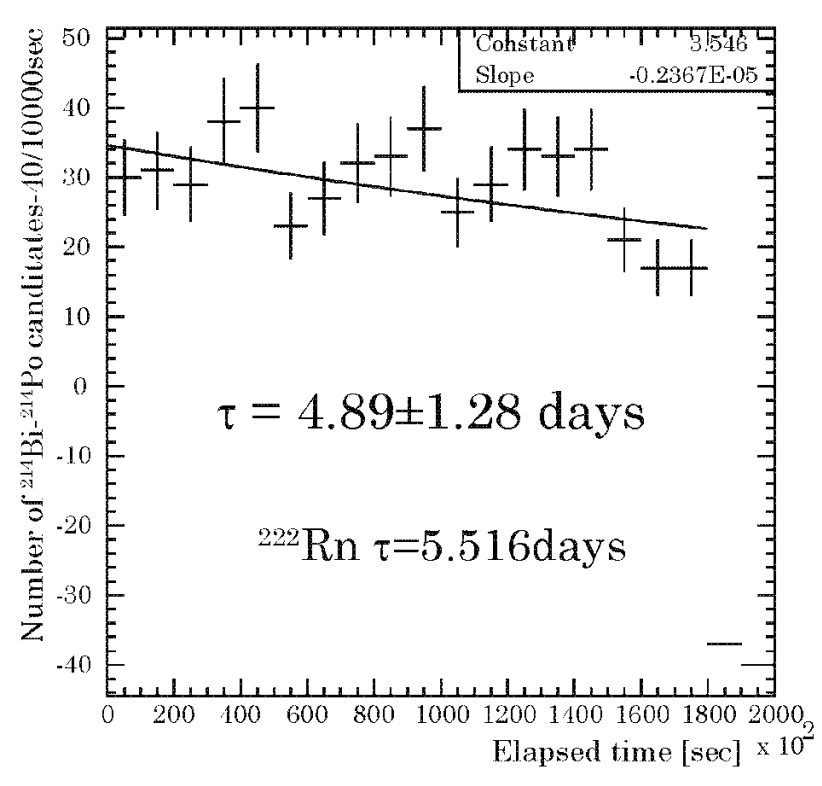


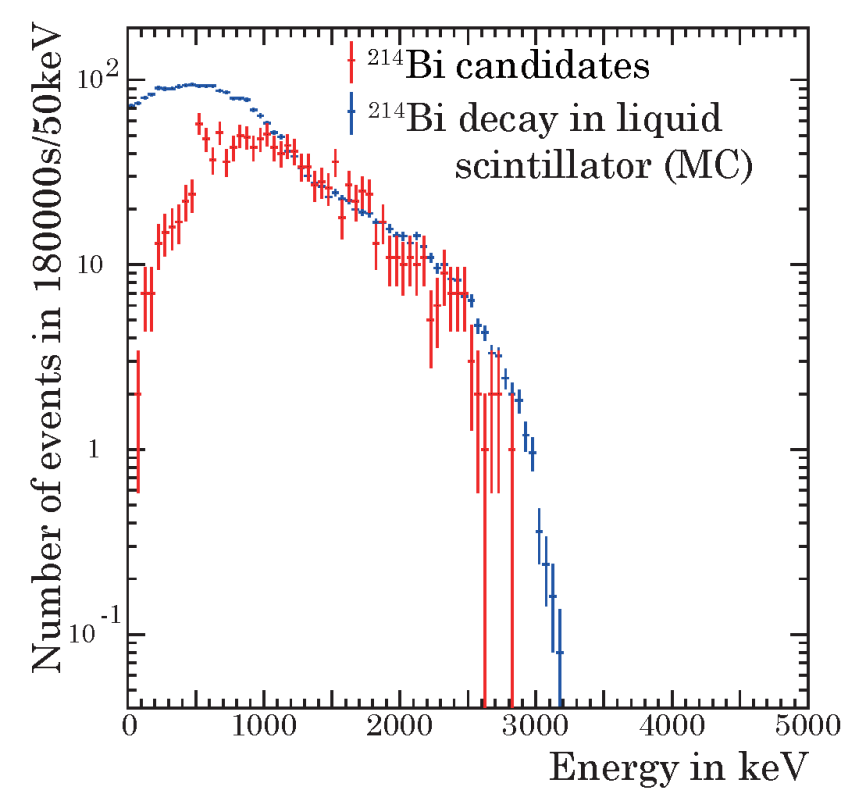


Time slope is inconsistent with lifetime of ^{222}Rn decay.($T_{1/2}$=3.8235days)

Spectral shape is almost consistent with ^{214}Bi β decay in LS obtained by simulation.

^{222}Rn contaminated in liquid scintillator?





Time slope is consistent with lifetime of ^{222}Rn decay, if 4mBq comes from ^{226}Ra decay.

Spectral shape is almost consistent with ^{214}Bi β decay in LS obtained by simulation.

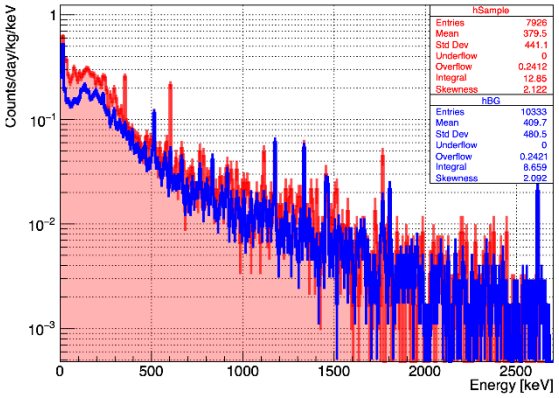
Background measurement using Ge detector

2024/11/07 LabC02

ZICOS LS

sample 224.6g, 35.2days

Unit: mBq/kg



summary table

unit : mBq/kg

U	value	error	limit
234Th	−8.75	10.21	13.07
234Pa	49.10	55.15	119.69
226Ra	−13.43	90.67	116.06
214Pb	0.26	3.30	4.48
214Bi	2.72	1.33	4.42
210Pb	98.94	55.91	170.51
Mid.U-chain	2.72	1.33	4.42
UpperU-ch	−6.83	10.04	12.85

Th	value	error	limit
228Ac	1.85	1.96	4.36
212Pb	2.38	1.66	4.51
212Bi	0.69	7.94	10.85
208Tl	−2.20	1.43	1.83
Th-chain	−2.11	1.41	1.80

	value	error	limit
60Co	−0.063	0.607	0.777

	value	error	limit
40K	6.03	7.40	15.50

	value	error	limit
137Cs	−0.14	0.76	0.97
134Cs	0.04	0.53	0.72

	value	error	limit
235U	0.56	5.01	6.97
231Th	7.46	10.10	20.38
231Pa	−26.28	39.40	50.44
227Th	3.94	9.07	15.55
below223Ra	−3.86	2.57	3.28

	value	error	limit
54Mn	−0.63	0.58	0.75
58Co	0.19	0.50	0.83
176Lu	−0.57	0.52	0.66

	value	error	limit
138La	0.41	1.40	2.20

BG：BG_20240909.dat

eff：20241107_ZICOS_LS.eff

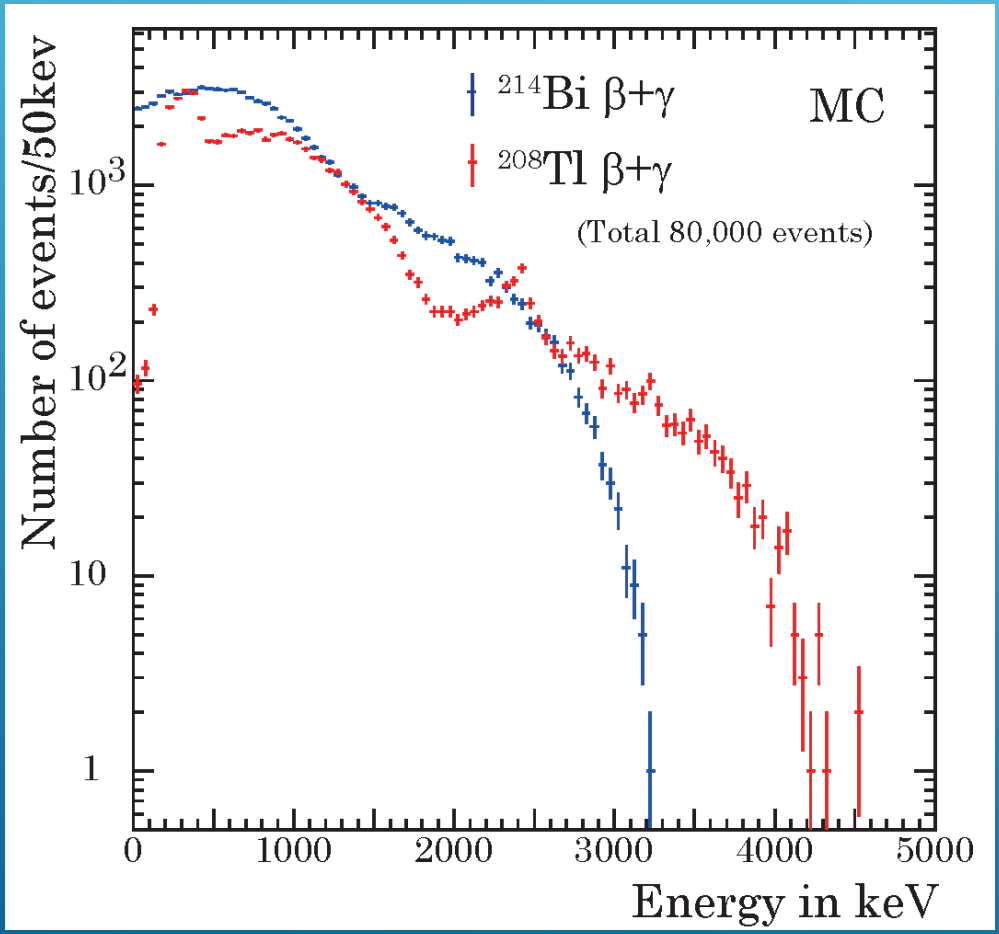
^{238}U upper (^{238}U)	^{238}U middle (^{226}Ra)	Th-chain (^{228}Ra)	Th-chain (^{228}Th)	U-235 (^{235}U)	U-235 (below^{223}Ra)	K-40	Co-60	Cs-137	Lu-176
<12.85	2.72±1.33	<4.36	<1.80	<6.97	<3.28	<15.50	<0.777	<0.97	<0.66

2.72±1.33mBq/kg ⟺ 5.7mBq/kg (constant)

Almost consistent with measured value by Ge

How much amount of Th-chain in LS?

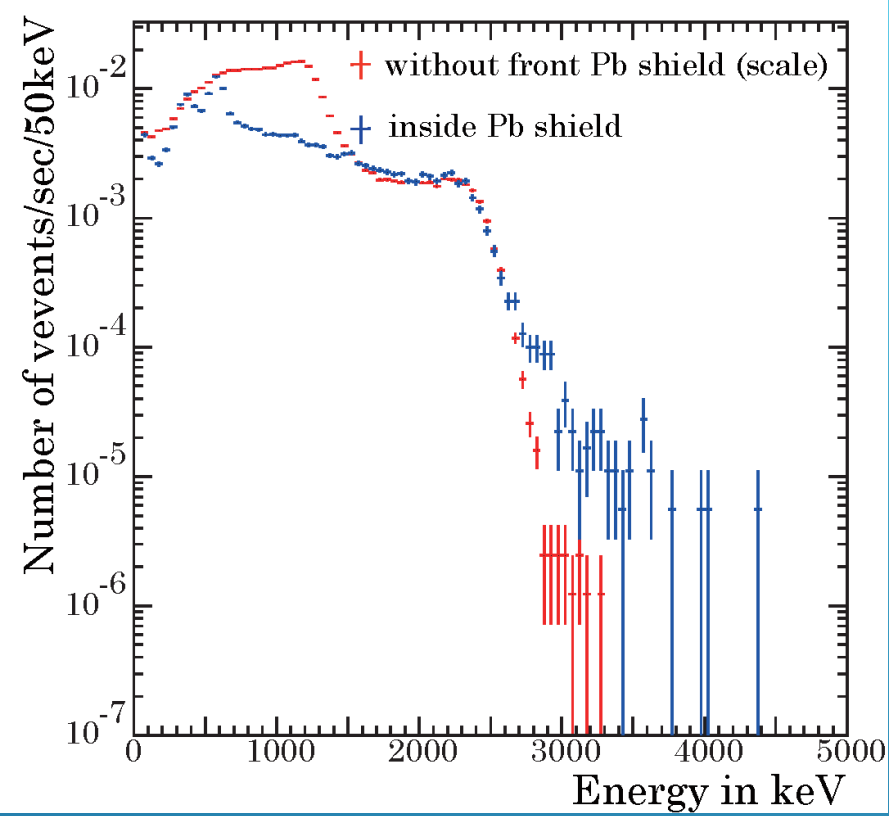
No ^{212}Bi-^{212}Po candidates ($T_{1/2}=2.99\times10^{-7}$sec) was observed due to dead time of V1742 digitizer.

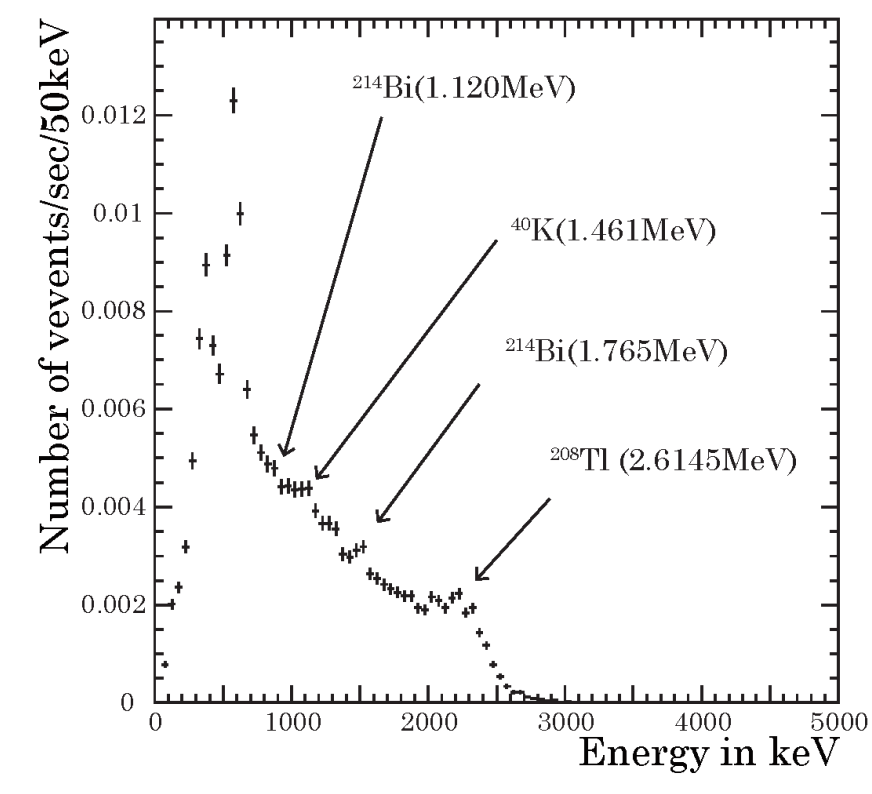


Energy spectrum of ^{208}Tl β decay should have a higher energy tail than that of ^{214}Bi beta decay.

Not so much Th-chain backgrounds exist in liquid scintillator.

What is major source of measured events ?

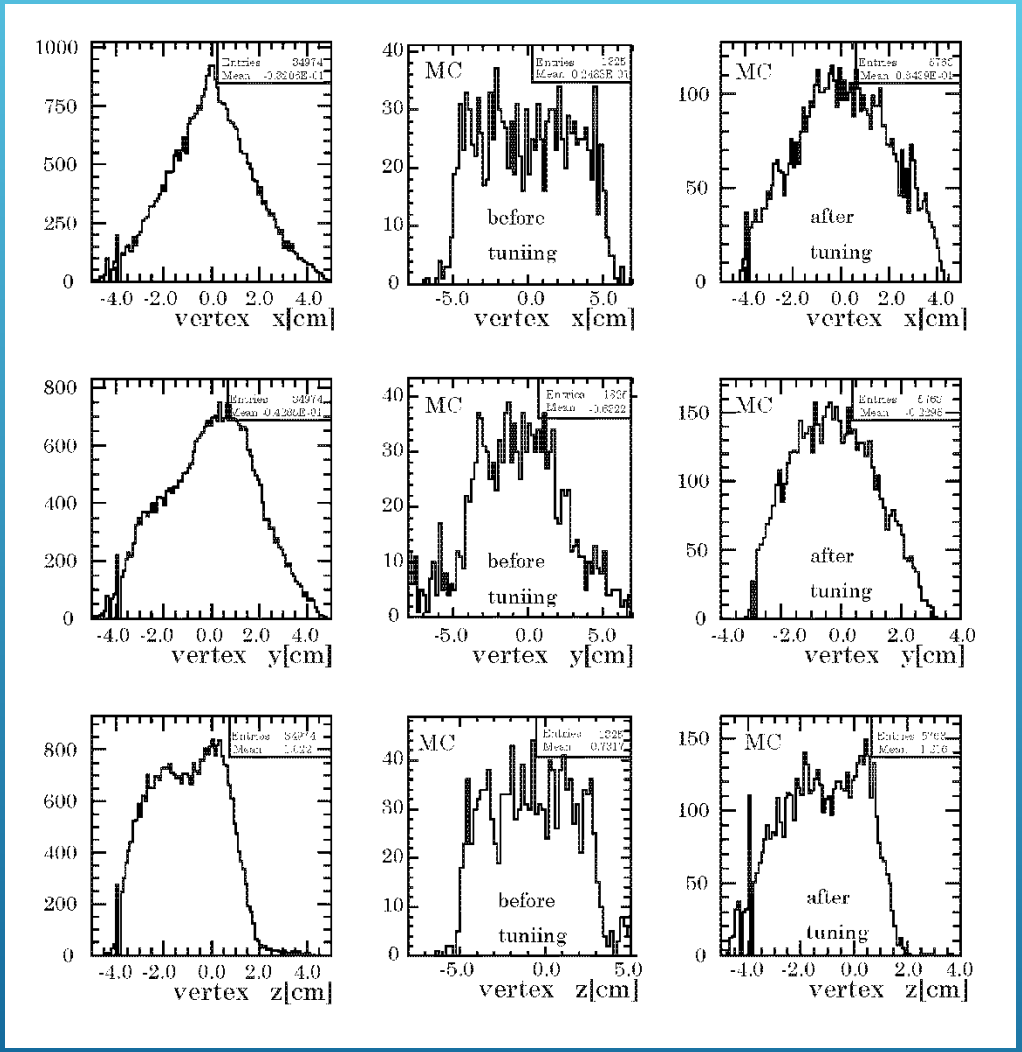




Spectral shape around 2.4MeV is consistent with 2.6145MeV γ from ^{208}Tl.

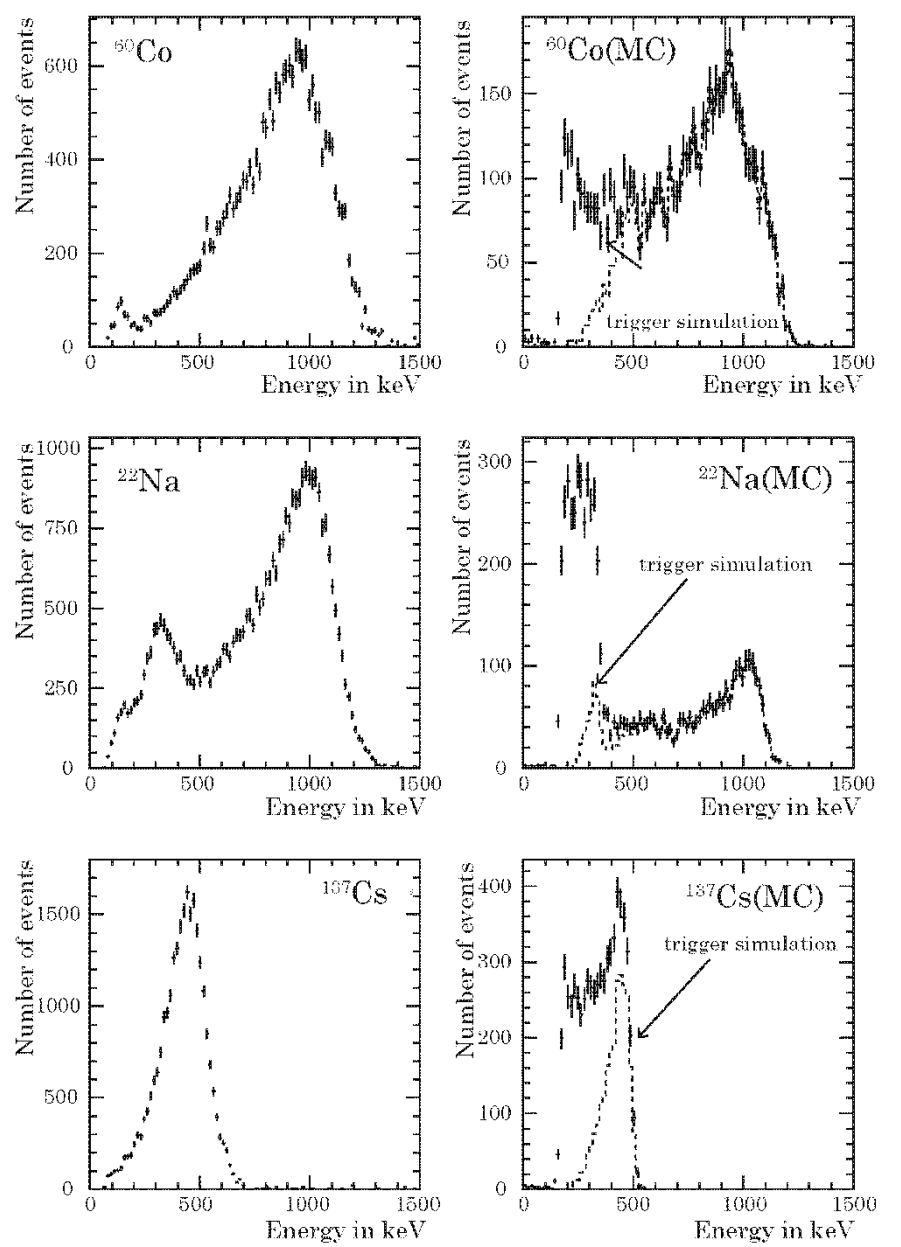
Major gamma spectra from U/Th series and ^{40}K were observed.

Monte Carlo tuning using calibration source



- MC tuning was used for ^{60}Co data.
- Tuning parameters are refractive index of LS for reflection + refraction of light between liquid and air and liquid scintillator level.
- Vertex position of data are almost reproduced by MC after tuning.

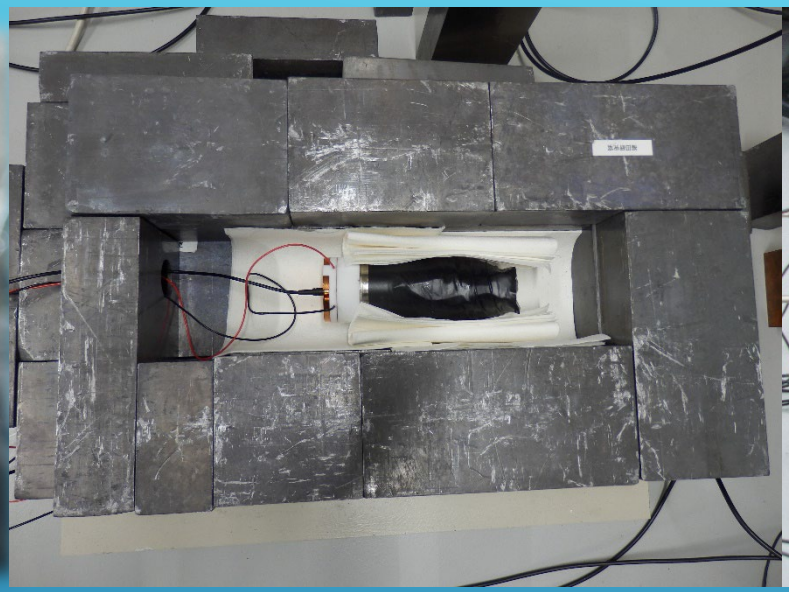
Trigger simulation using calibration source

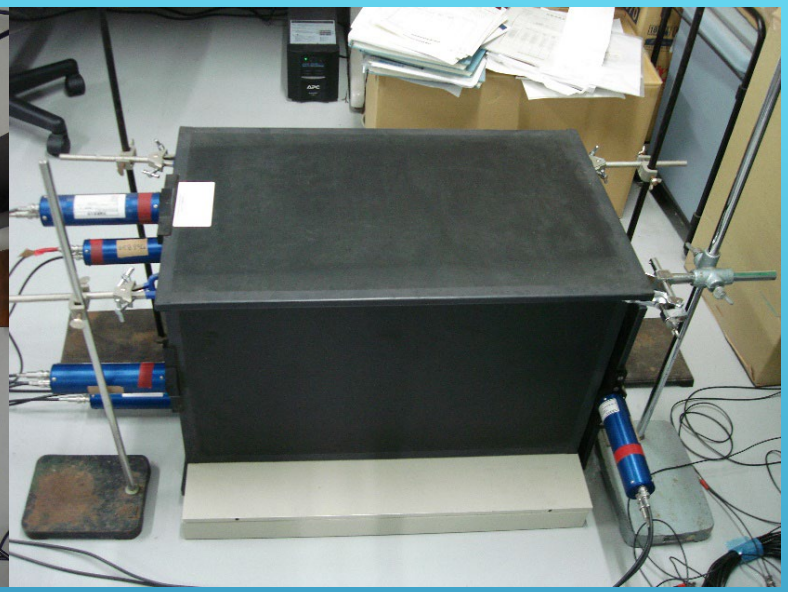


- Trigger simulation was used for number of photon received by PMT#1 and PMT#20.
- Typical trigger efficiency around 600keV is 70%.
- Therefore ^{214}Bi-^{214}Po candidates could be lost about 30% in DAQ.
- Calibration constant for MC was also obtained by same method as data.

BG measurement using 20mL quartz vial







- Difficult to measure BG events for 2ν-ZICOS detector inside of Pb shield due to limit of number of Pb block.
- 17.5mL ZICOS LS filled in 20mL quartz vial.
- The vial contacted with XMASS PMT R10789 covered by Myra film.
- 10cm thickness of Pb radiation shield was prepared.
- In order to veto cosmic muons, plastic scintillators were located around Pb shield.

Before starting 2ν-ZICOS in the mine

- We measured BG events on the ground using quartz vial inside Pb shield with cosmic muon veto.
- BG events from U/Th/K in both PMT R10789 and Quartz vial should be quite few.
- Observed ^{214}Bi-^{214}Po candidate was consistent with the result of ^{226}Ra 2.72 ± 1.33 mBq/kg measured by Ge detector in Kamioka mine.
- Surface event rate is inconsistent with XMASS estimation.
- Residual events seemed to not be internal BG events. In fact, 40% of residual events are caused by external γ.
- We moved all equipment to Kamioka and constructed the radiation shield last October.
- Safety equipment such as fire alarm, automatic telephone, liquid leak sensor were prepared before November.
- The first data taking implemented over 50 hours from November 22nd to 25th

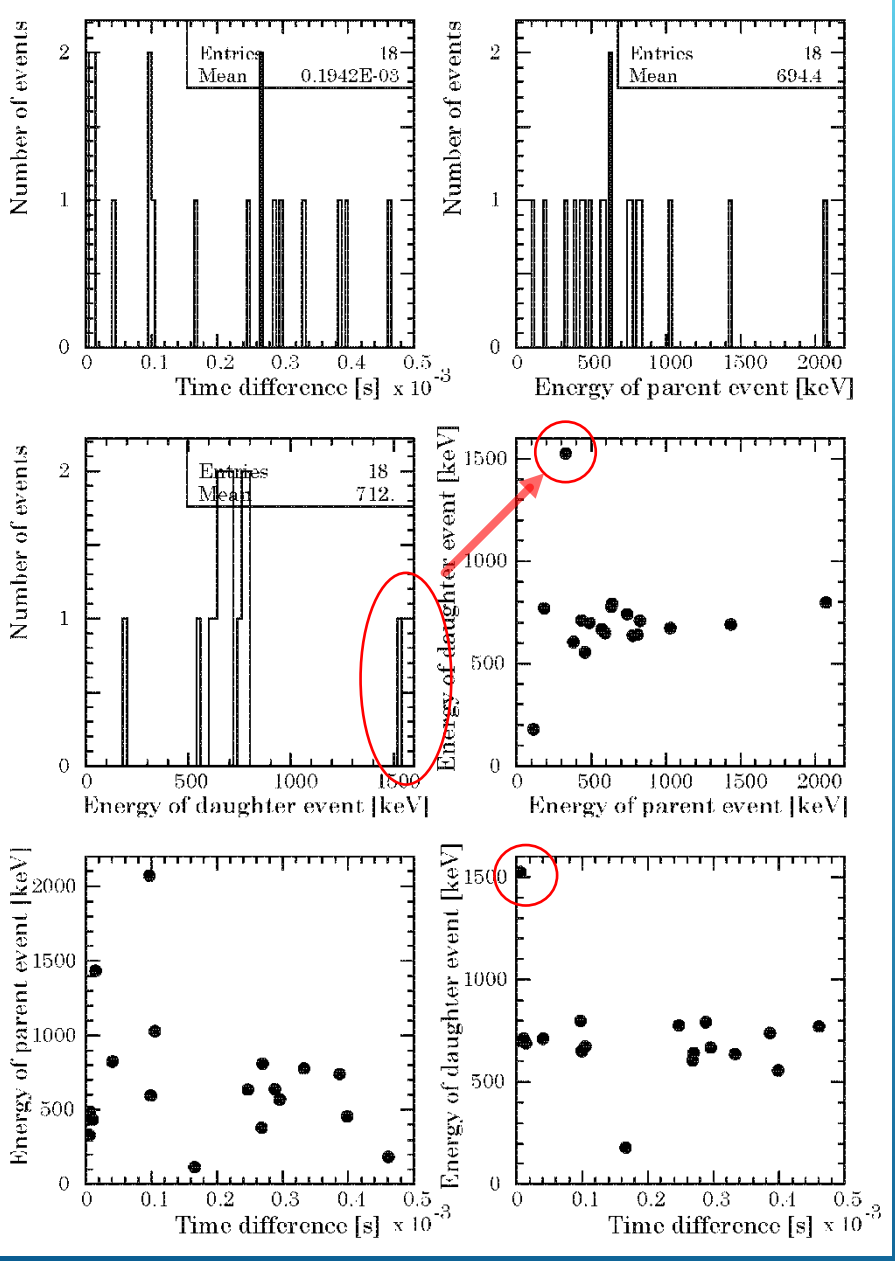
Sep 15,2026

Why 2ν-ZICOS should do in Kamioka mine?

There is huge backgrounds for the observation of $2\nu\beta\beta$ events on the ground due to cosmic muons.

- Cosmic muon rate is ~2Hz on the ground.
- Most significant spallation product is ^{11}C which decays in beta plus and emits total energy ~2MeV.
- Maximum 270 events per day will be observed on ground.
- The half-life of ^{11}C is 1200sec and the detector should have no live-time if veto for all muons.
- In the mine, the cosmic muon rate will be 2×10^{-5} Hz, so dead-time is about 2.4% even if veto for all muons.
- Expected event rate due to ^{11}C, which is caused by neutron produced by muon spallation, is ~20 events per year using measured neutron flux. (same expected number was obtained by KamLAND data. Ref:Phys.RevC.107.054612)

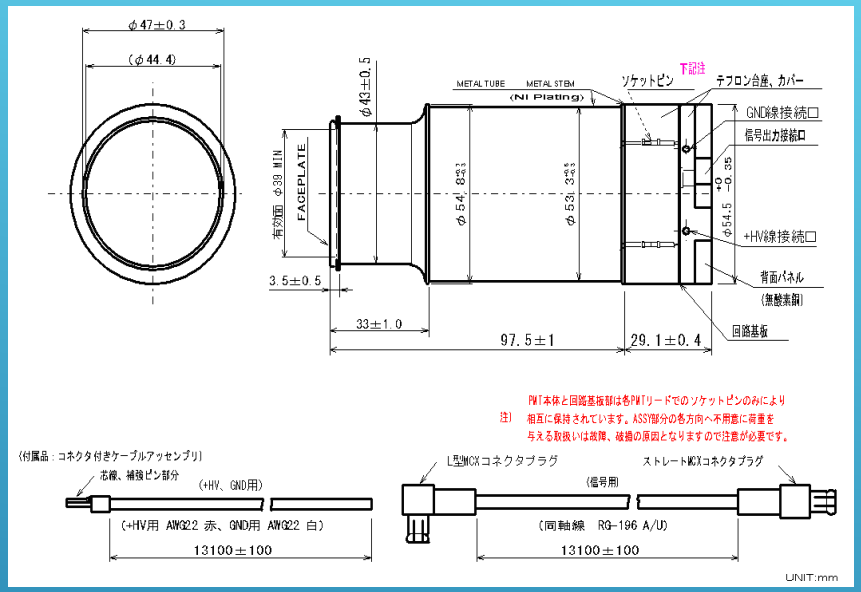
Search for ^{214}Bi-^{214}Po candidates

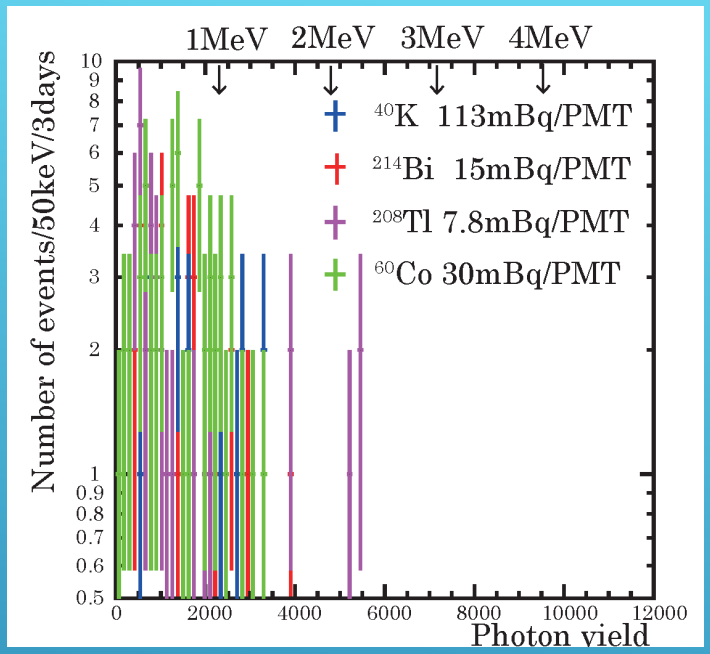


- 18events have time difference < 5×10^{-4} s except parent muon
- Parent events have wider energy distribution than daughter events
- Daughter events have narrow distribution (like α quenching)

17events looks ^{214}Bi-^{214}Po candidates. But they don't contribute residual events.

BG events from PMT R10789





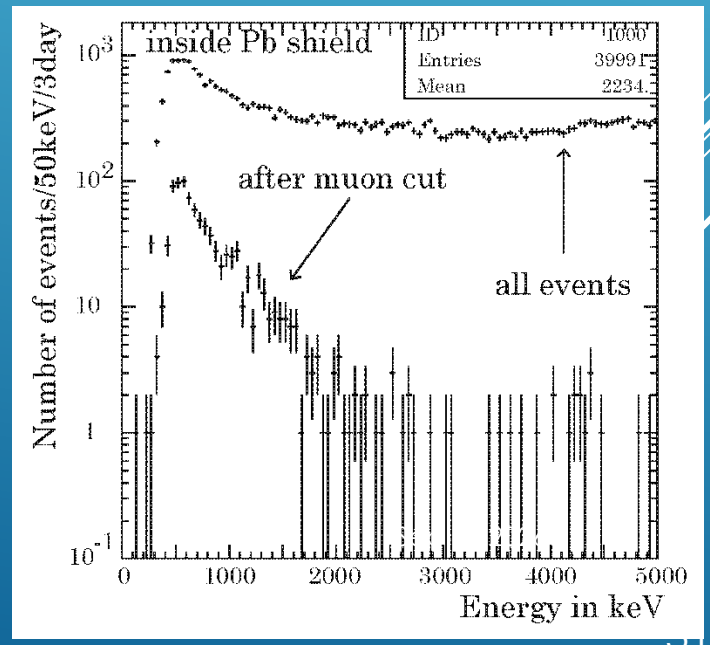
Ref: NIMA 922(2019)171-176

1.2 ± 0.3 mBq/PMT of ^{226}Ra

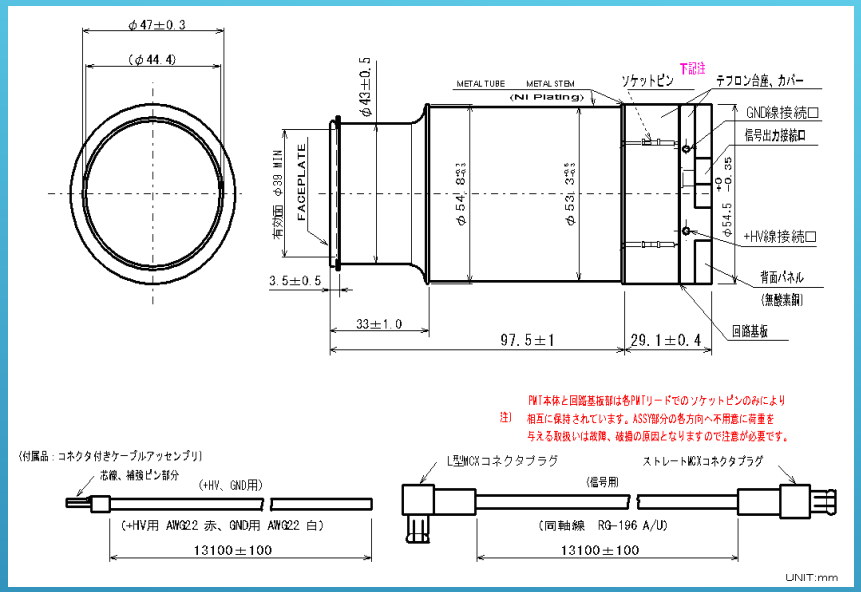
< 0.78 mBq/PMT of ^{228}Ra

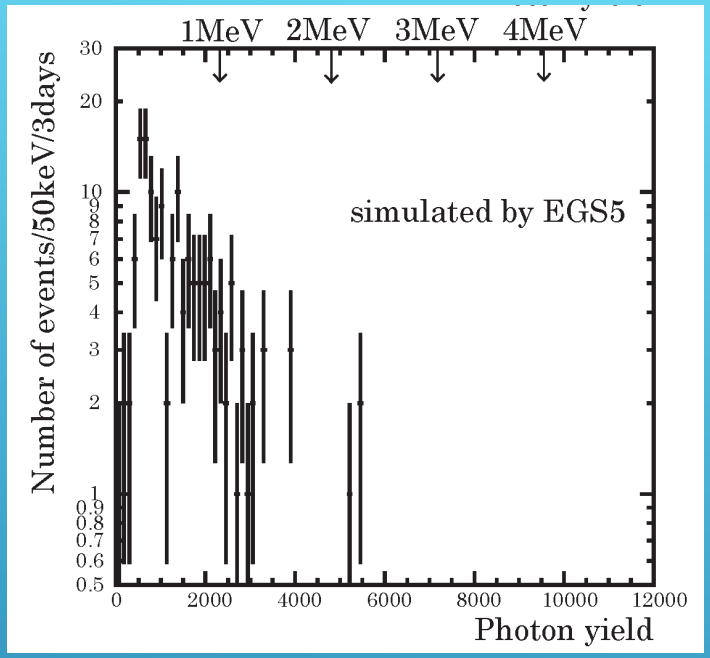
9.1 ± 2.2 mBq/PMT of ^{40}K

2.8 ± 0.2 mBq/PMT of ^{60}Co.



BG events from PMT R10789





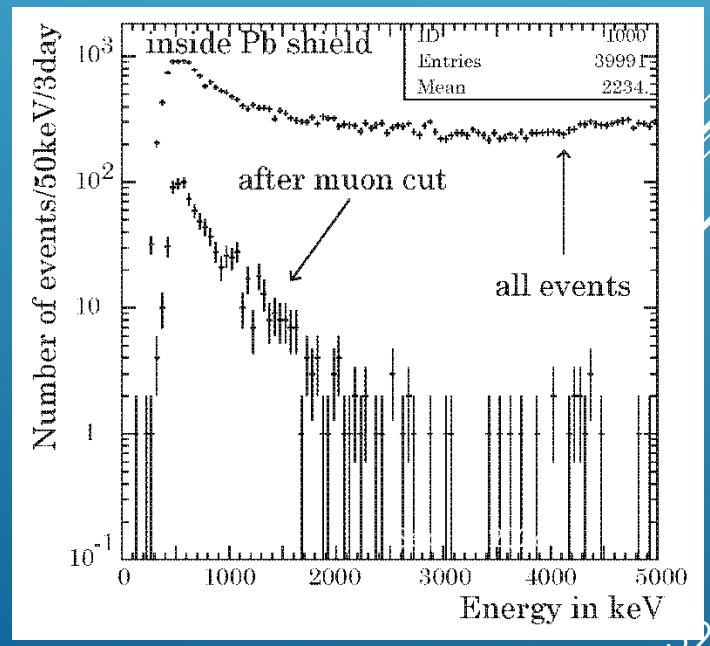
Ref: NIMA 922(2019)171-176

1.2 ± 0.3 mBq/PMT of ^{226}Ra

< 0.78 mBq/PMT of ^{228}Ra

9.1 ± 2.2 mBq/PMT of ^{40}K

2.8 ± 0.2 mBq/PMT of ^{60}Co.



Need ~40 times larger BG events to explain the residual events.

BG events from Quartz

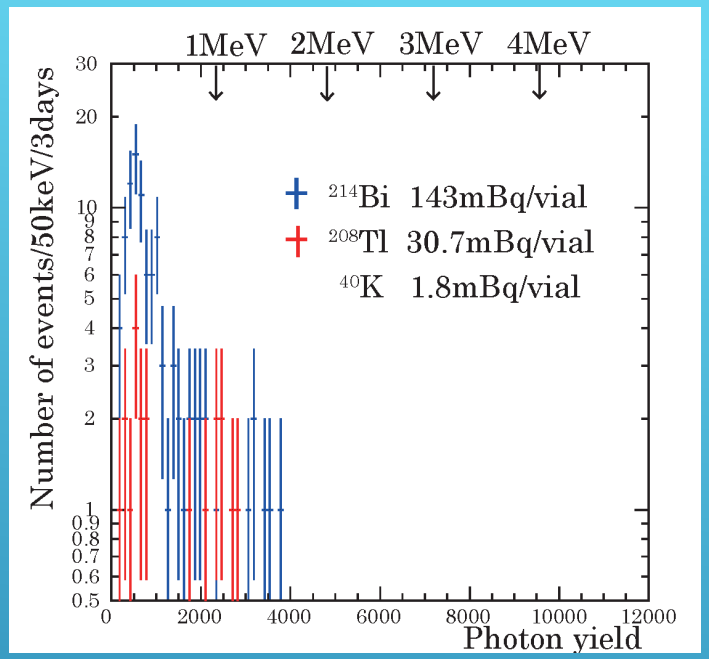


表　分析結果

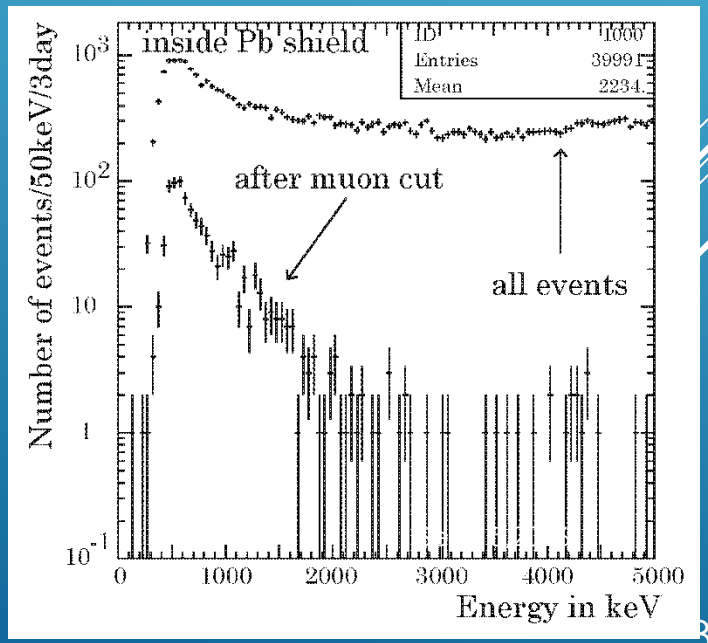
単位：ng/g

試料名	K	Th	U
RQ200	330	42	64
GE214	180	15	29
定量下限	5	1	1

Measured by Toshiba analysis using IPC mass spectrometer



14.3mBq/vial of ^{214}Bi
3.07mBq/vial of ^{208}Tl
0.18mBq/vial of ^{40}K



BG events from Quartz

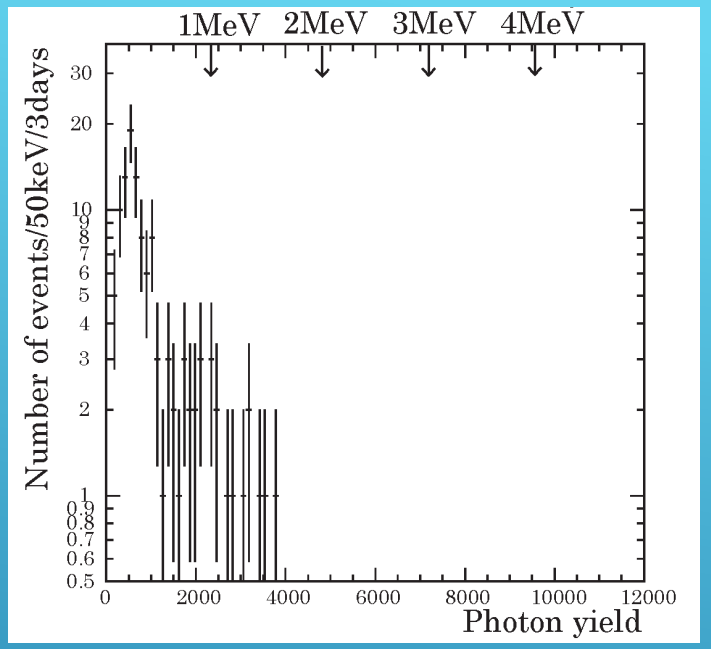


表 分析結果

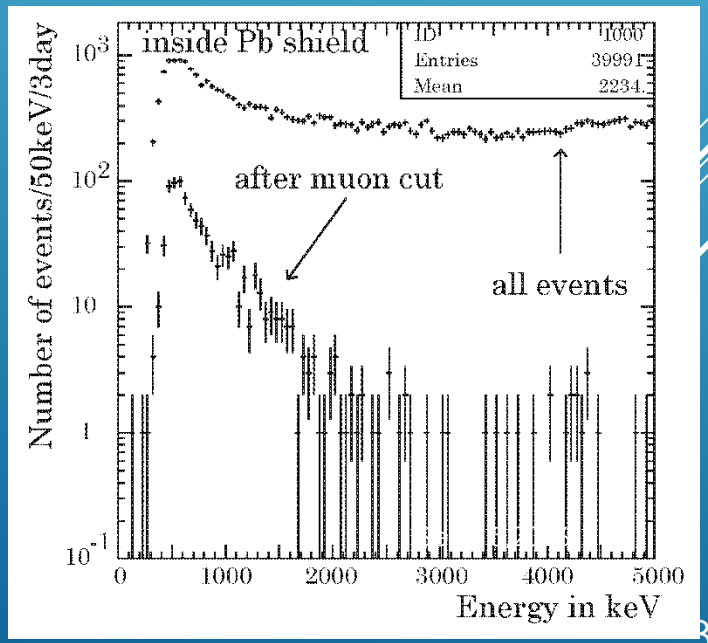
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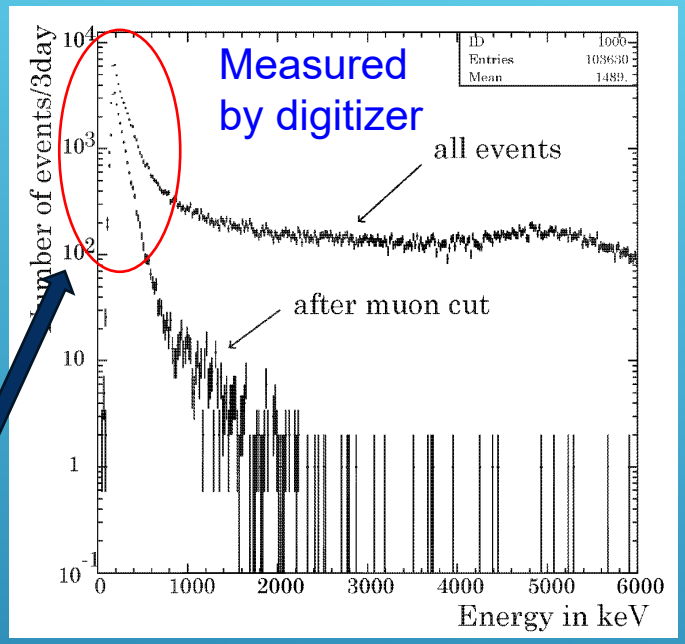


14.3mBq/vial of ^{214}Bi
3.07mBq/vial of ^{208}Tl
0.18mBq/vial of ^{40}K

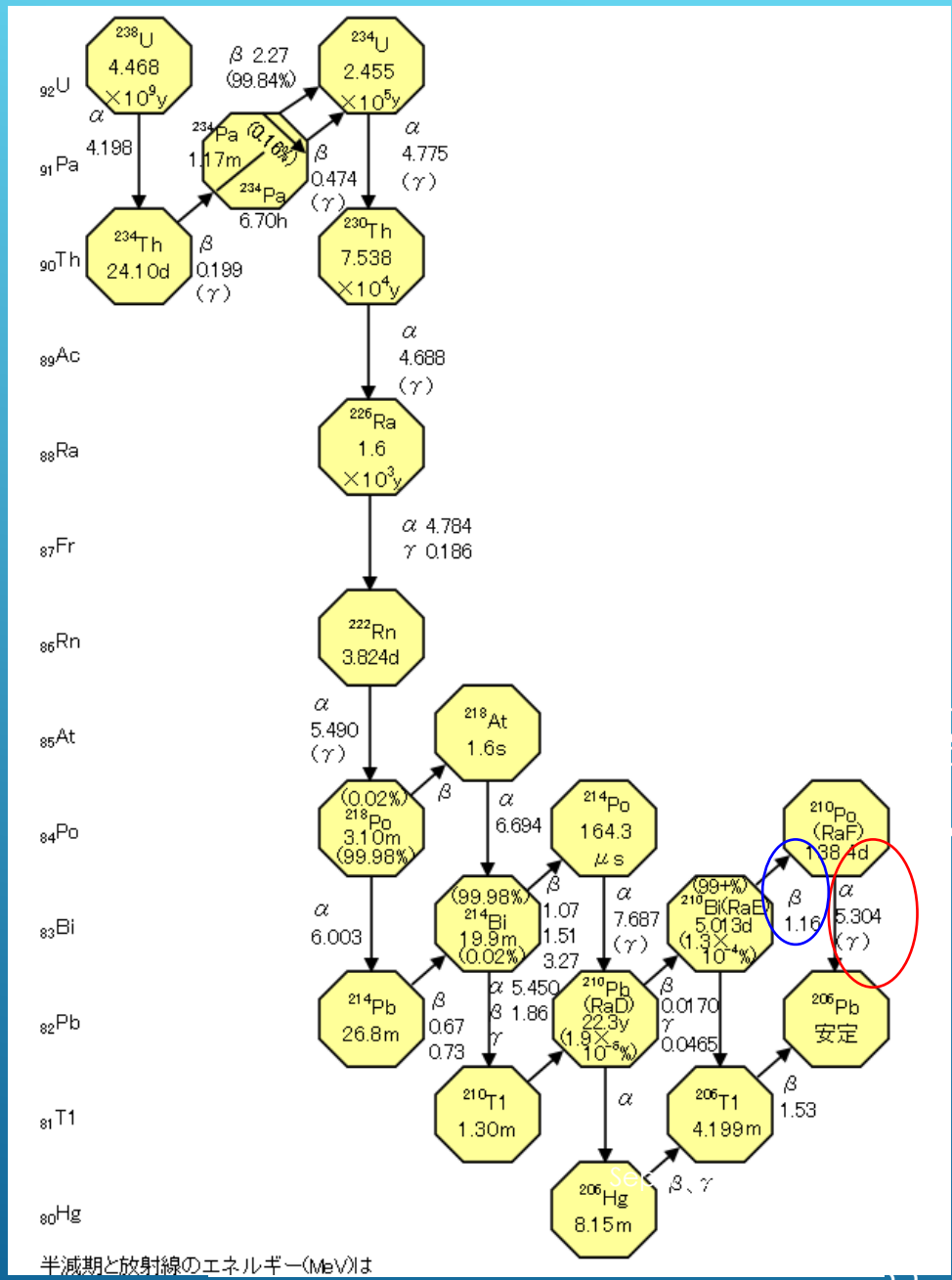


Even more 50 times larger BG events could not explain the residual events.

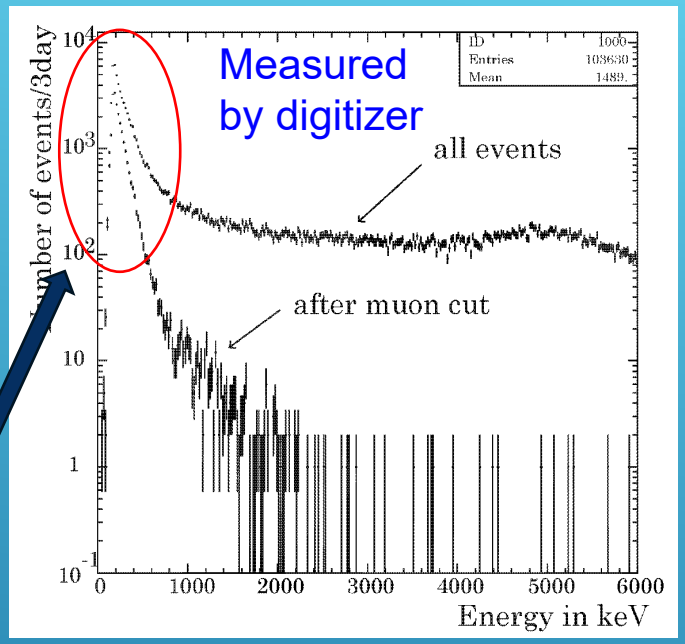
BG events from surface of Quartz

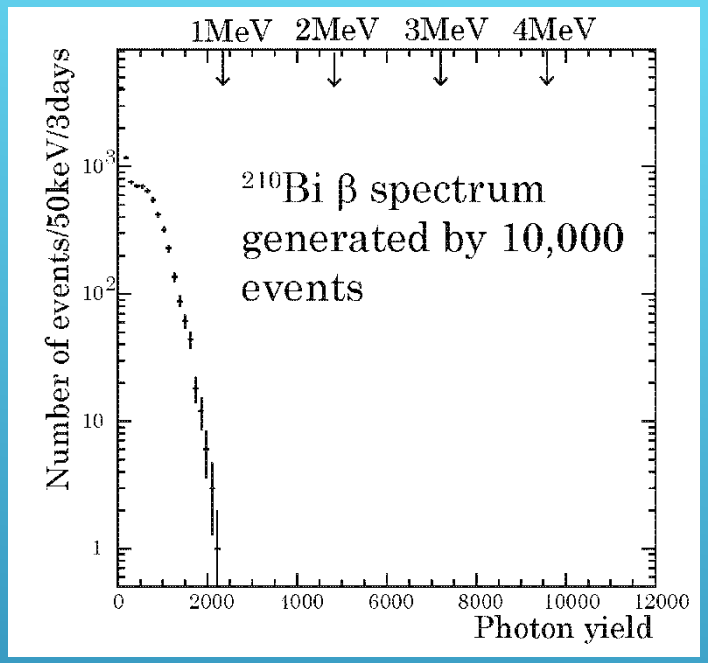


α from ^{210}Po? if so, β from ^{210}Bi should be observed by same order of events.

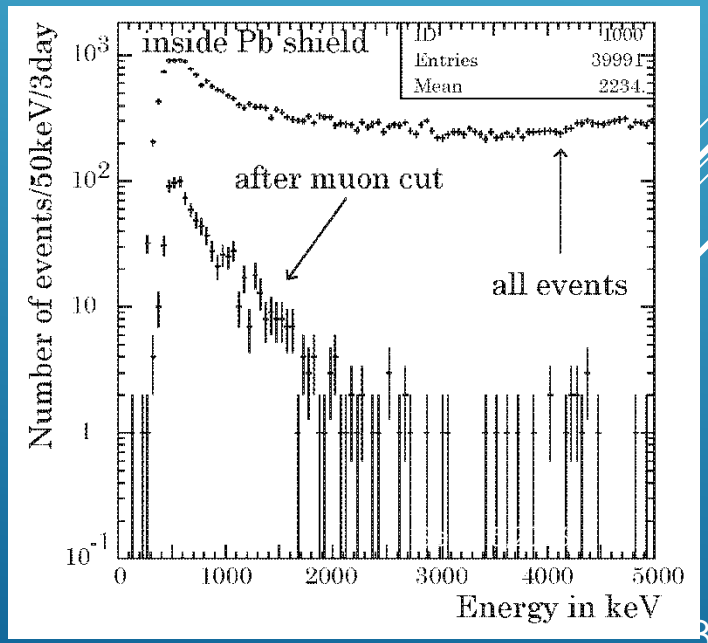


BG events from surface of Quartz



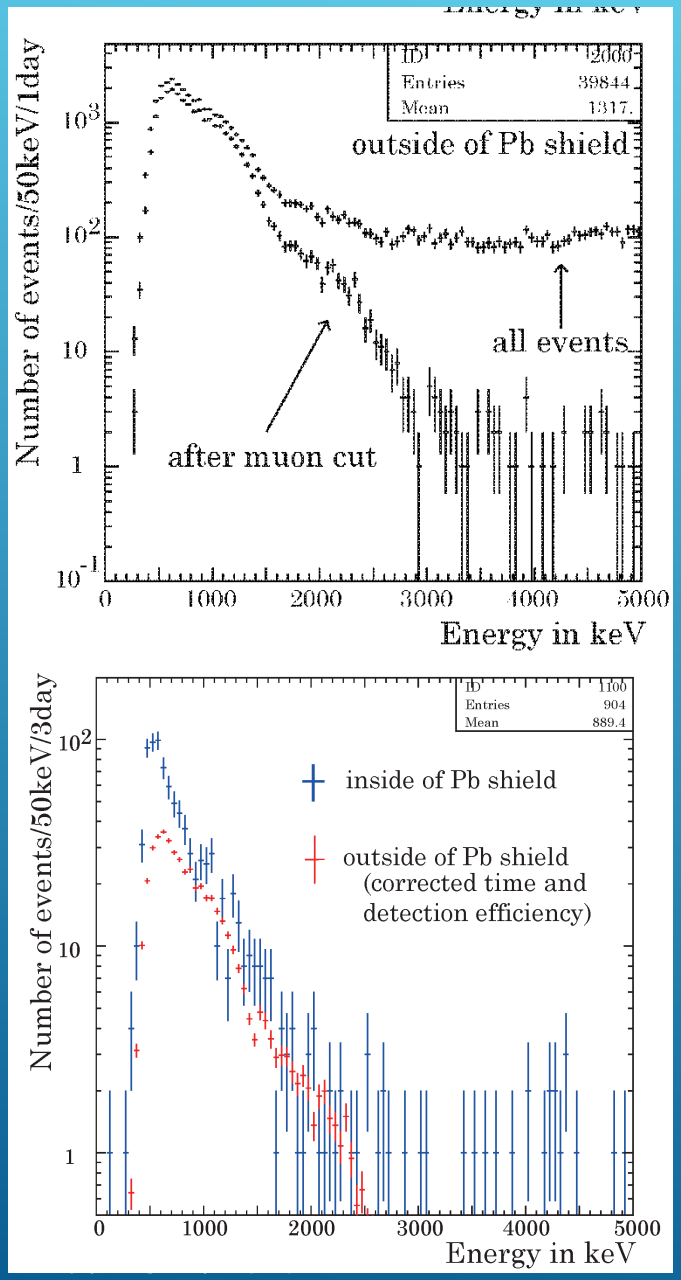


α from ^{210}Po? if so, β from ^{210}Bi should be observed by same order of events.



Observed rate correspond to 2mBq/cm^2, but XMASS estimated as 1μBq/cm^2.

Comparing distributions using env. gammas

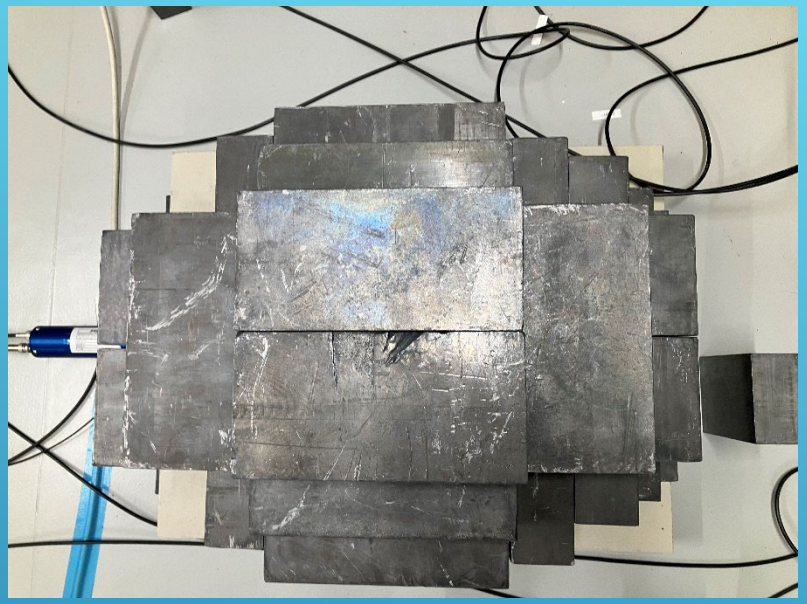


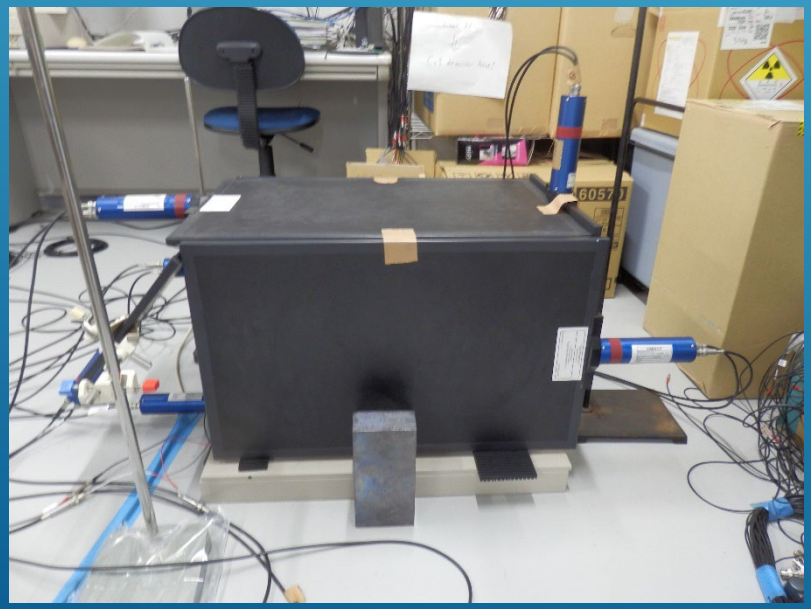
Almost the energy spectrum of residual events is consistent with environmental gammas observed at outside of shield.

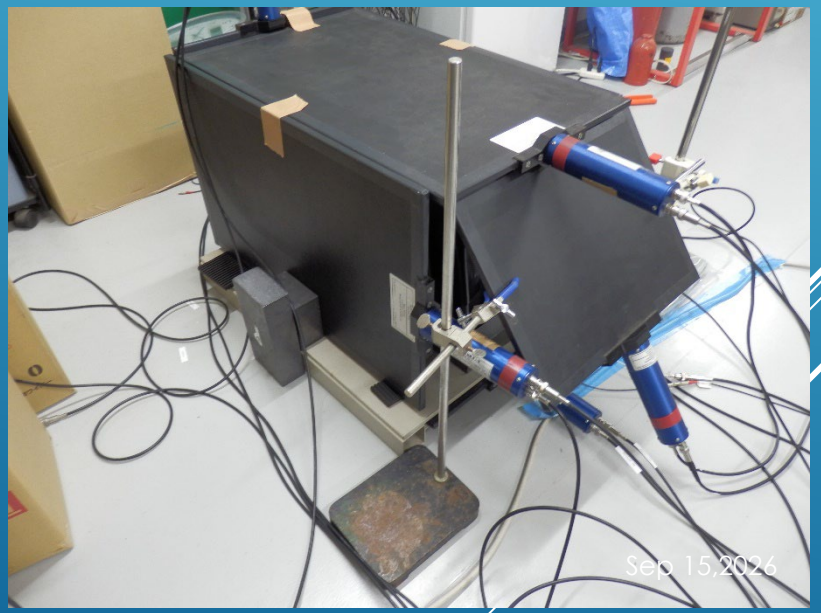
No obvious BG event exists except those above, we are still investigating the source of residual events.
(At least, it is not internal BG.)

BG measurement inside 15cm Pb shield



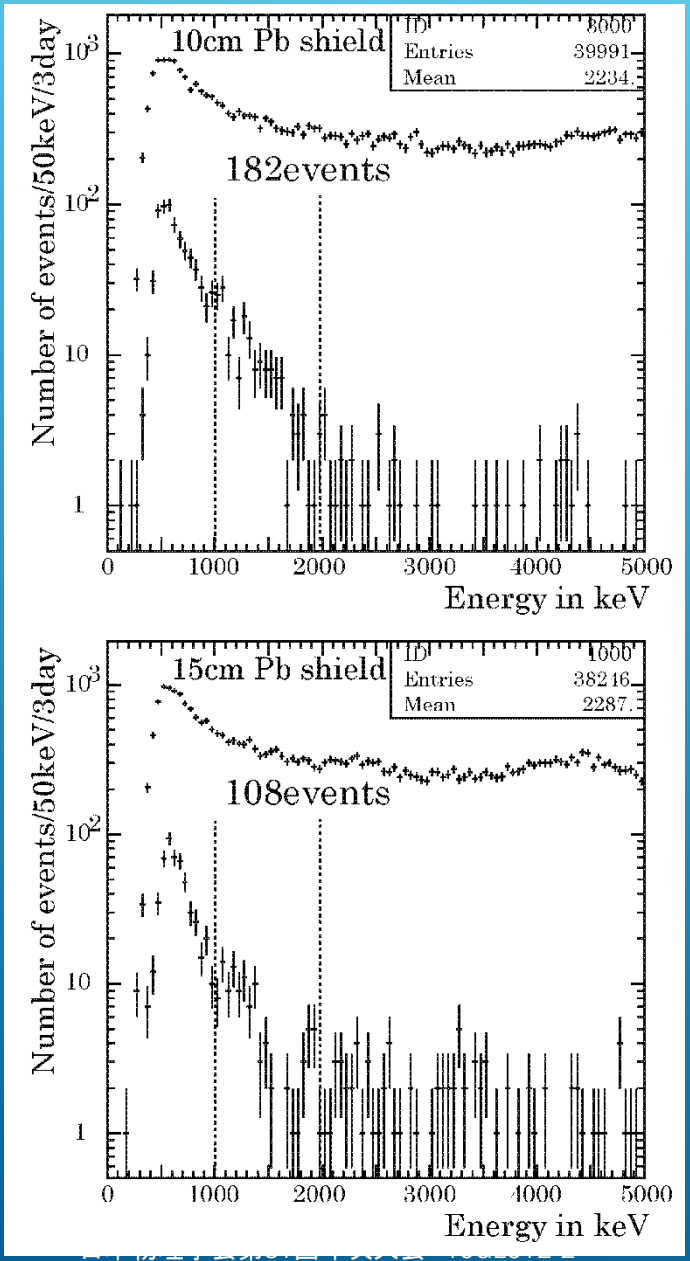






Sep 15,2026

鉛ブロックの厚さを15cmにしてBG観測



1MeV-2MeV間の事象数を比較

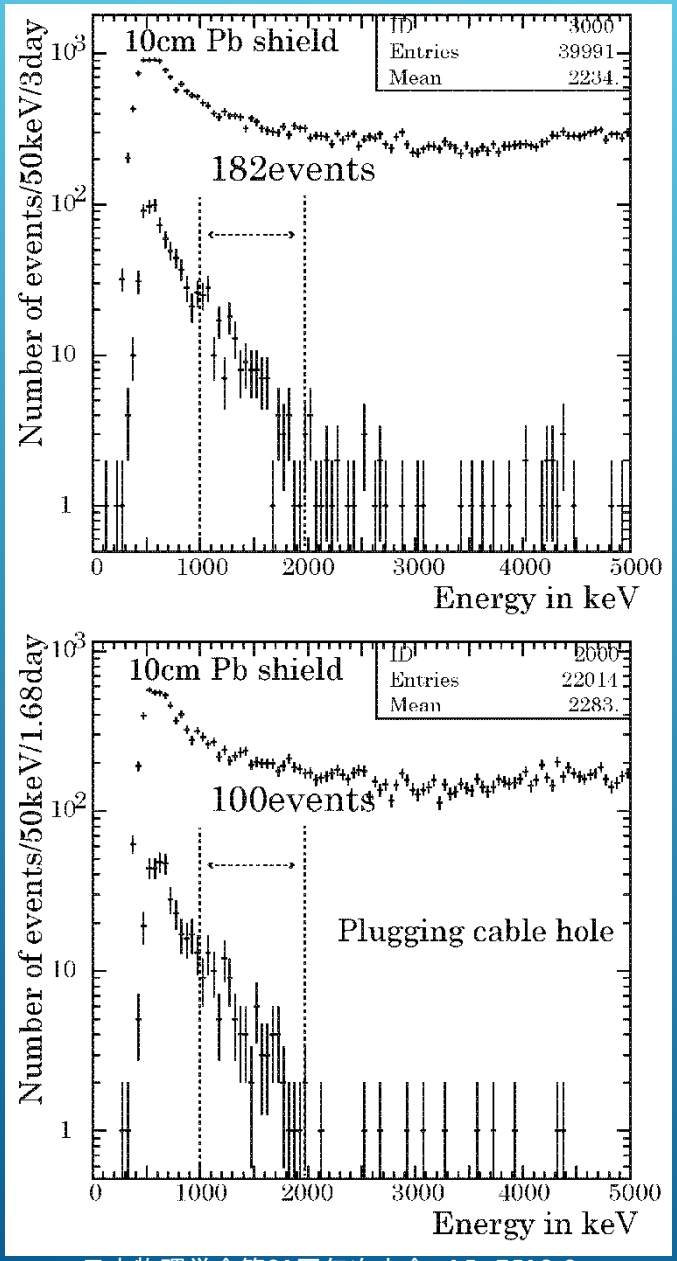
・10cmシールド　182事象/3日間

・15cmシールド　108事象/3日間

結果：　残存事象が30~40%減少

結論：　外部ガンマ線が若干見えていた可能性

Cable holeを鉛ブロックで塞いでBG観測



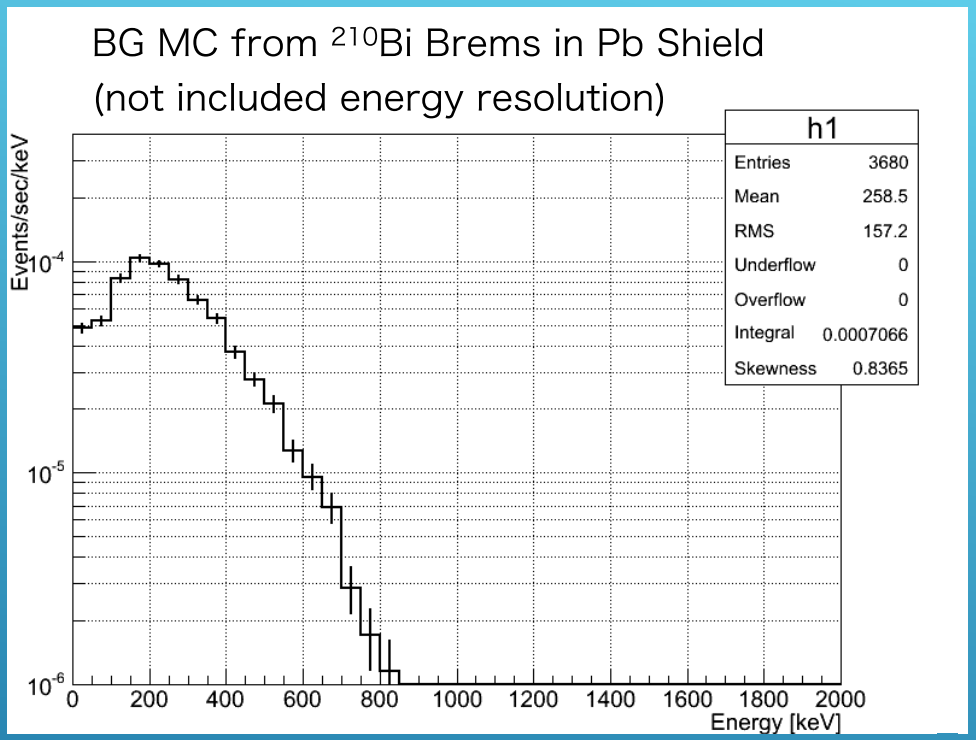
1MeV-2MeV間の事象数を比較

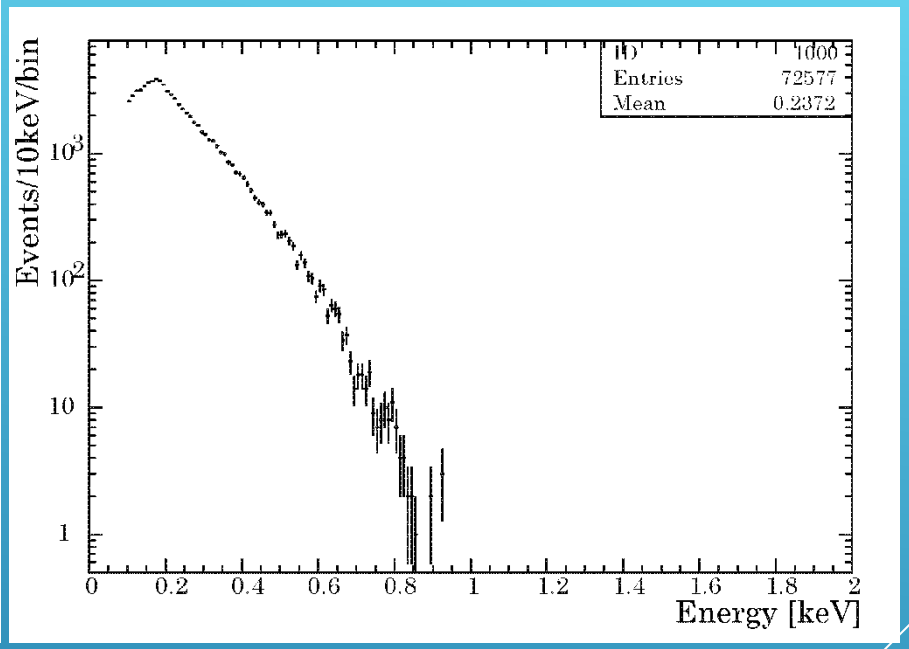
・穴を塞ぐ前　182事象/3日間

・穴を塞いだ後　100事象/1.68日間
＝　179事象/3日間

結論：　穴の効果は見られなかった

鉛ブロック内の^{210}Pbのベータ線からの制動放射

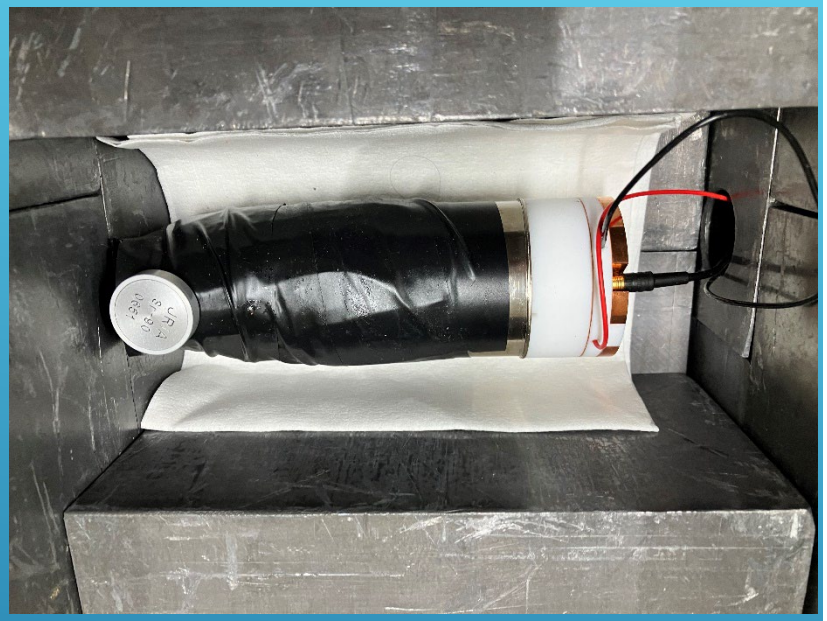




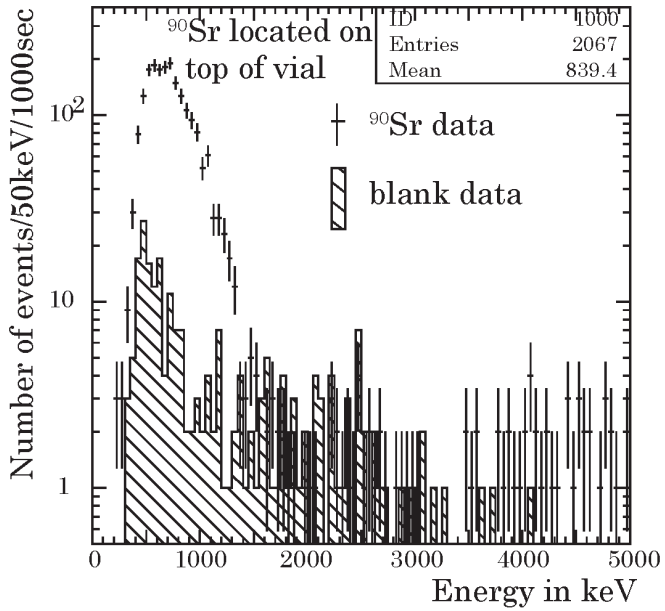
市村さんが振った鉛シールド内の^{210}Biのベータ事象からの制動放射事象のエネルギー分布と、EGS5で同様に振った事象のエネルギー分布

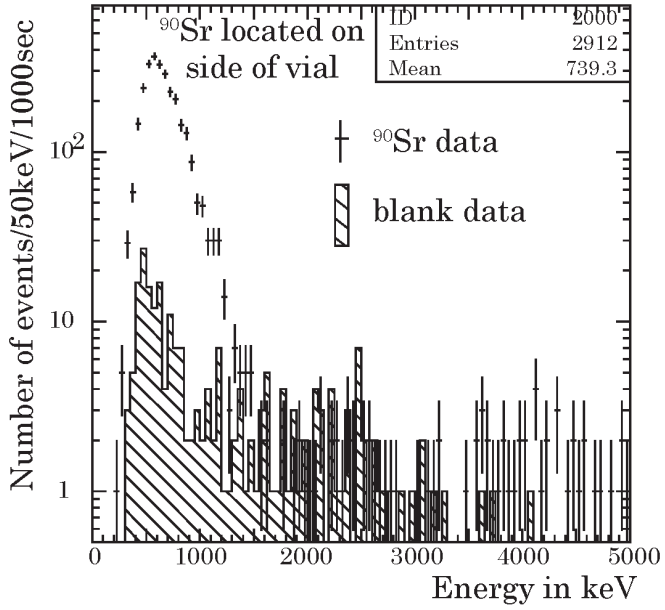
よく似ているが、EGS5の方が少しエネルギーが低い

鉛ブロックからの^{210}Biのベータ線が見える可能性





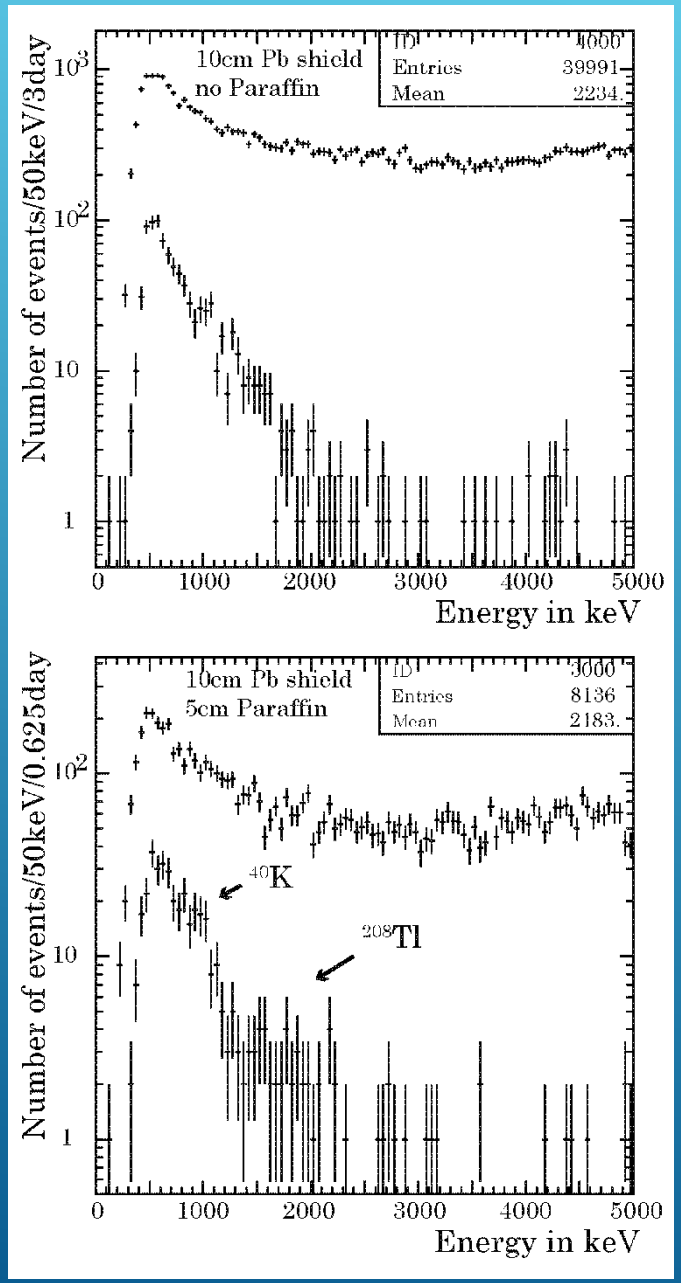


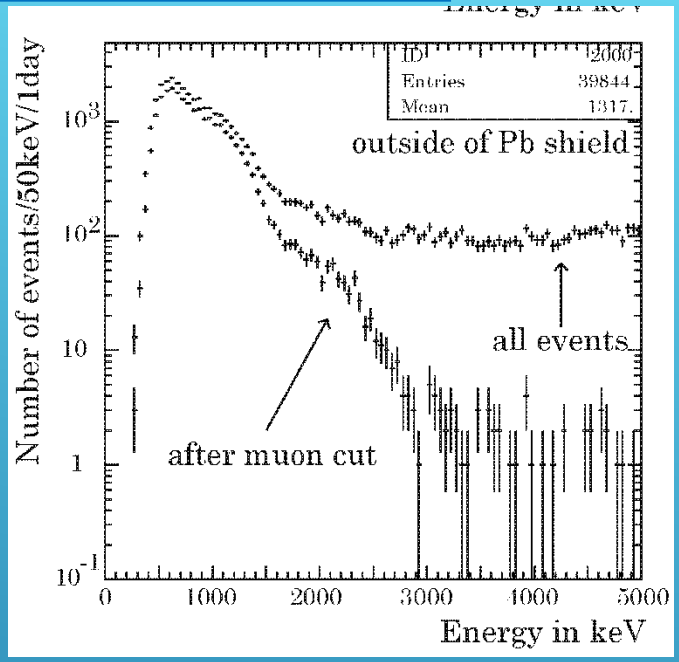


^{90}Srのベータ線（Emax=2.28MeV)が見える。

鉛ブロックからの^{210}Biのベータが見える可能性がある

パラフィンブロックで囲んでBG測定





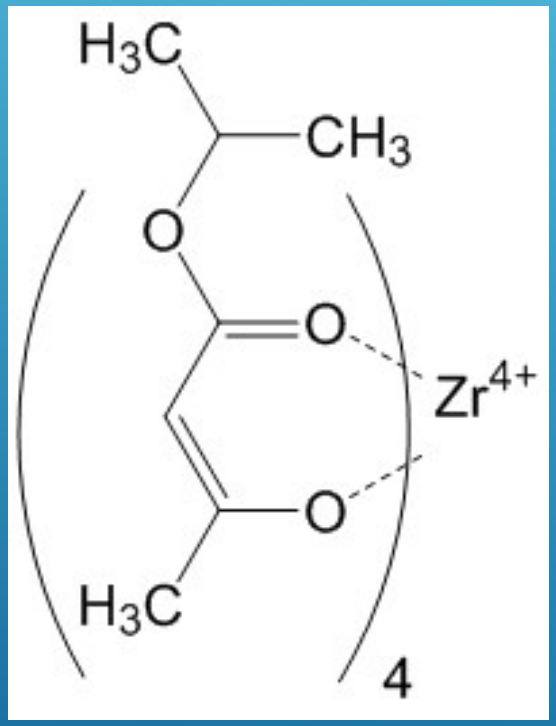
パラフィンで囲んだ場合の600keVの事象数が0.625時間で約30に対し、鉛だけの場合は72時間で約100なので、内部事象なら20事象存在するはずだが、予測される数しか観測されていない。

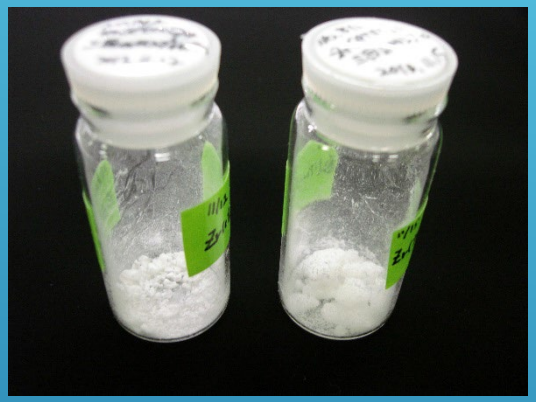
低エネルギー側は鉛からのベータか？

Zirconium β-keto ester complex

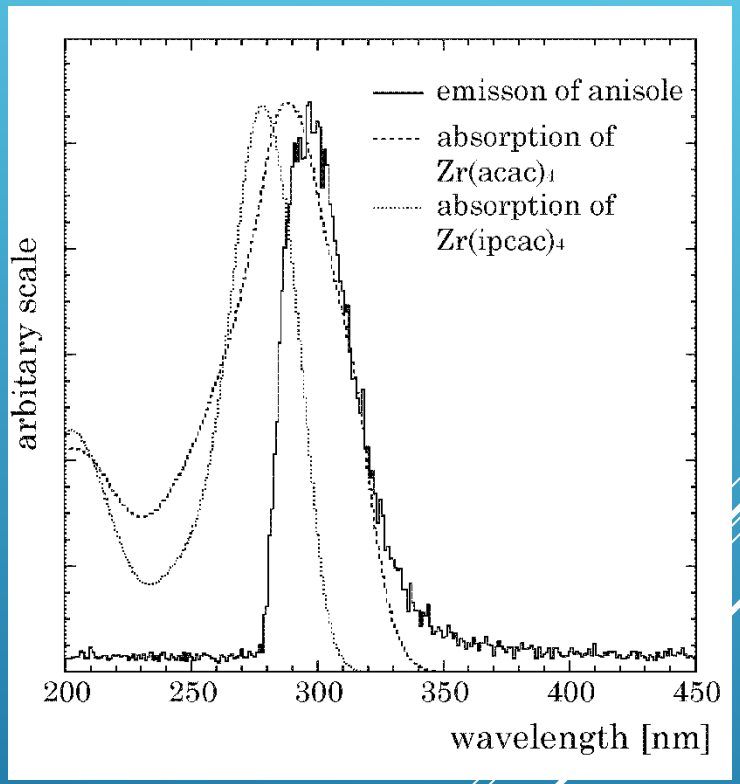
$Zr(CH_3COCHCOOCH(CH_3)_2)_4$ = $Zr(iPrac)_4$ mw : 663.87

Tetrakis(isopropyl acetoacetate) Zirconium





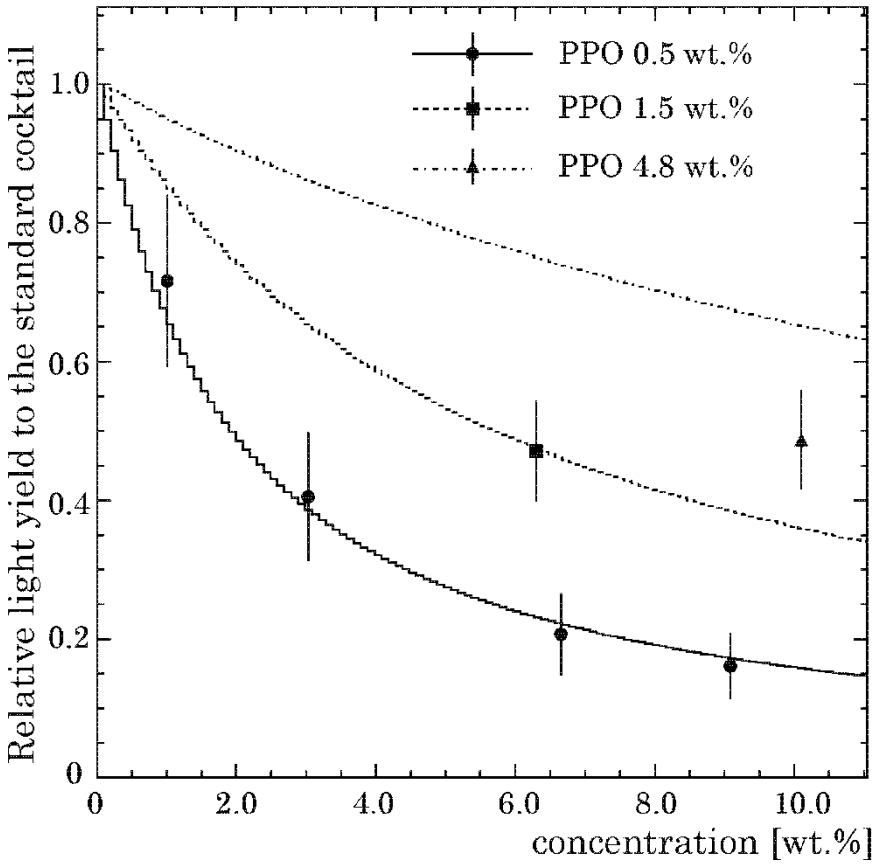
Synthesized by Prof. T.Gunji (Tokyo University of Science)

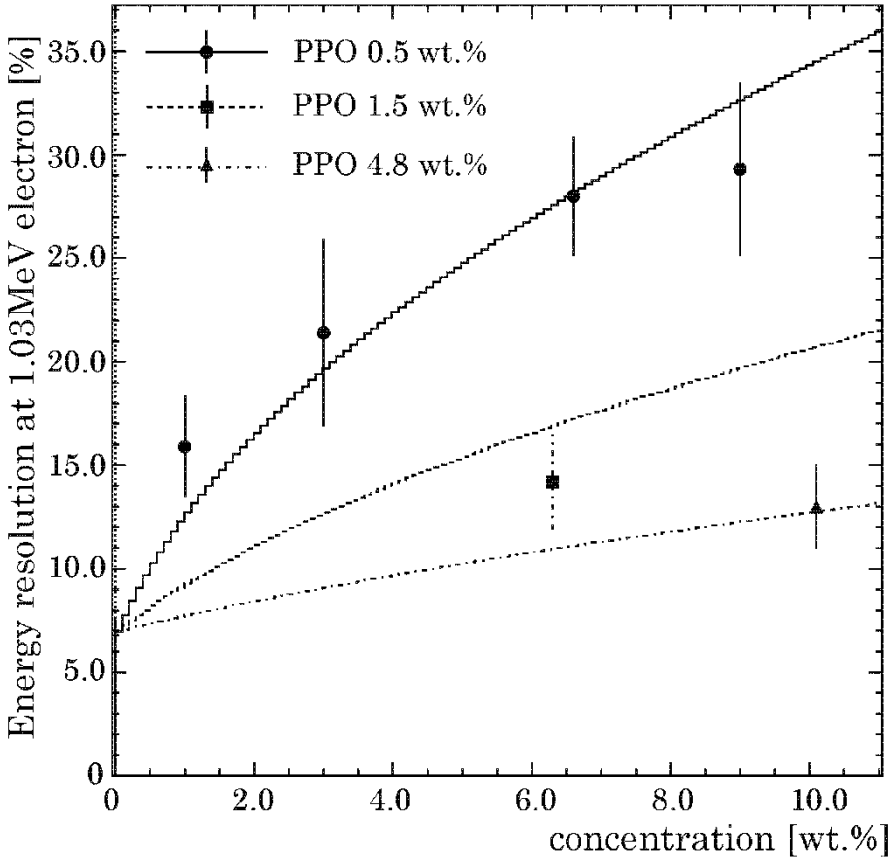


No commercial product

Shorter wavelength for absorbance

Improve of light yield and resolution





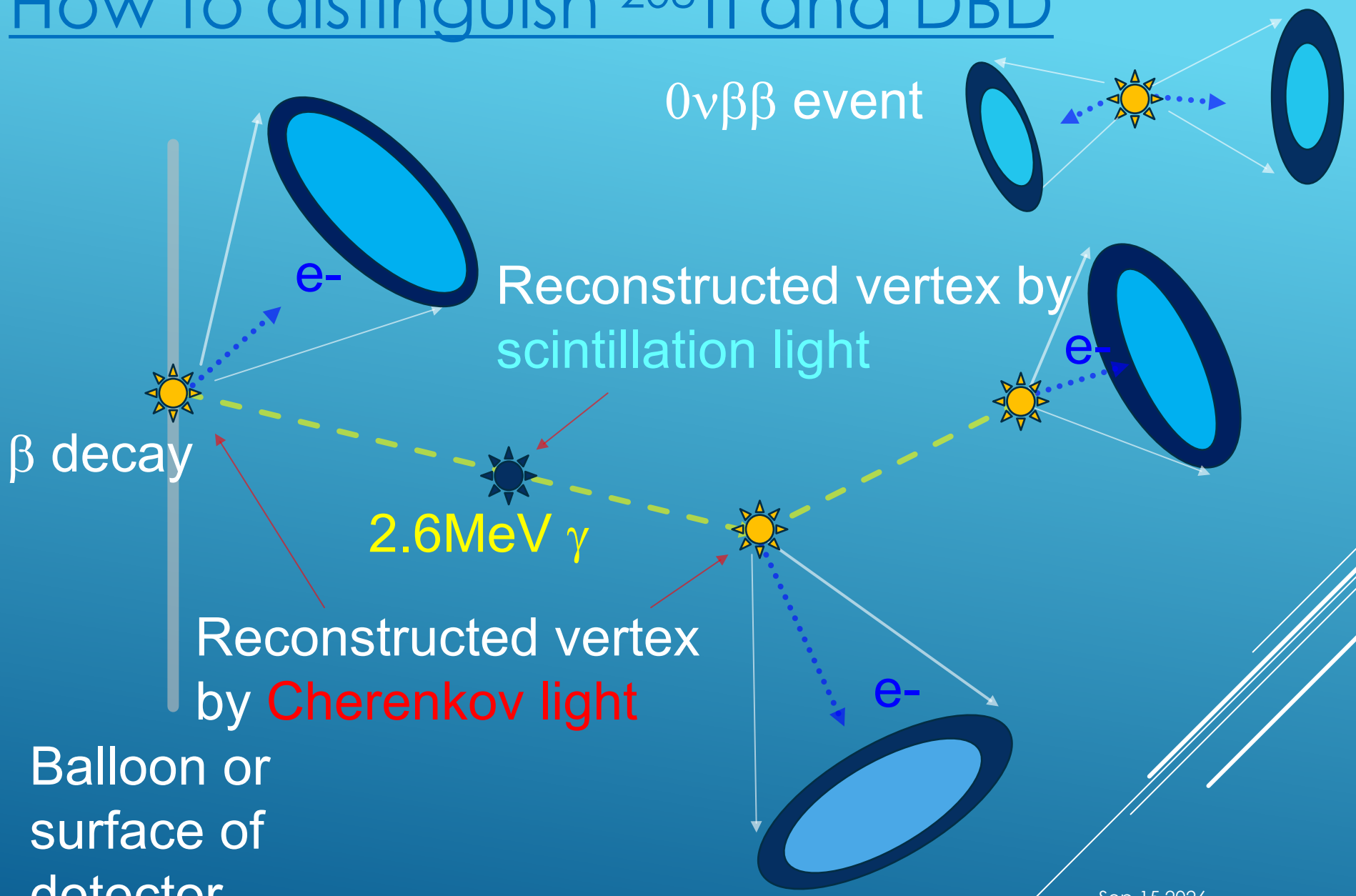
$Zr(iPrac)_4$ absorbs the scintillation photon and makes the energy resolution worth, however PPO could improve them.

$$\frac{13.0 \pm 2.0\%}{\sqrt{40\%/9\% * 0.72 * 3.35\text{MeV}/1.03\text{MeV}}}$$

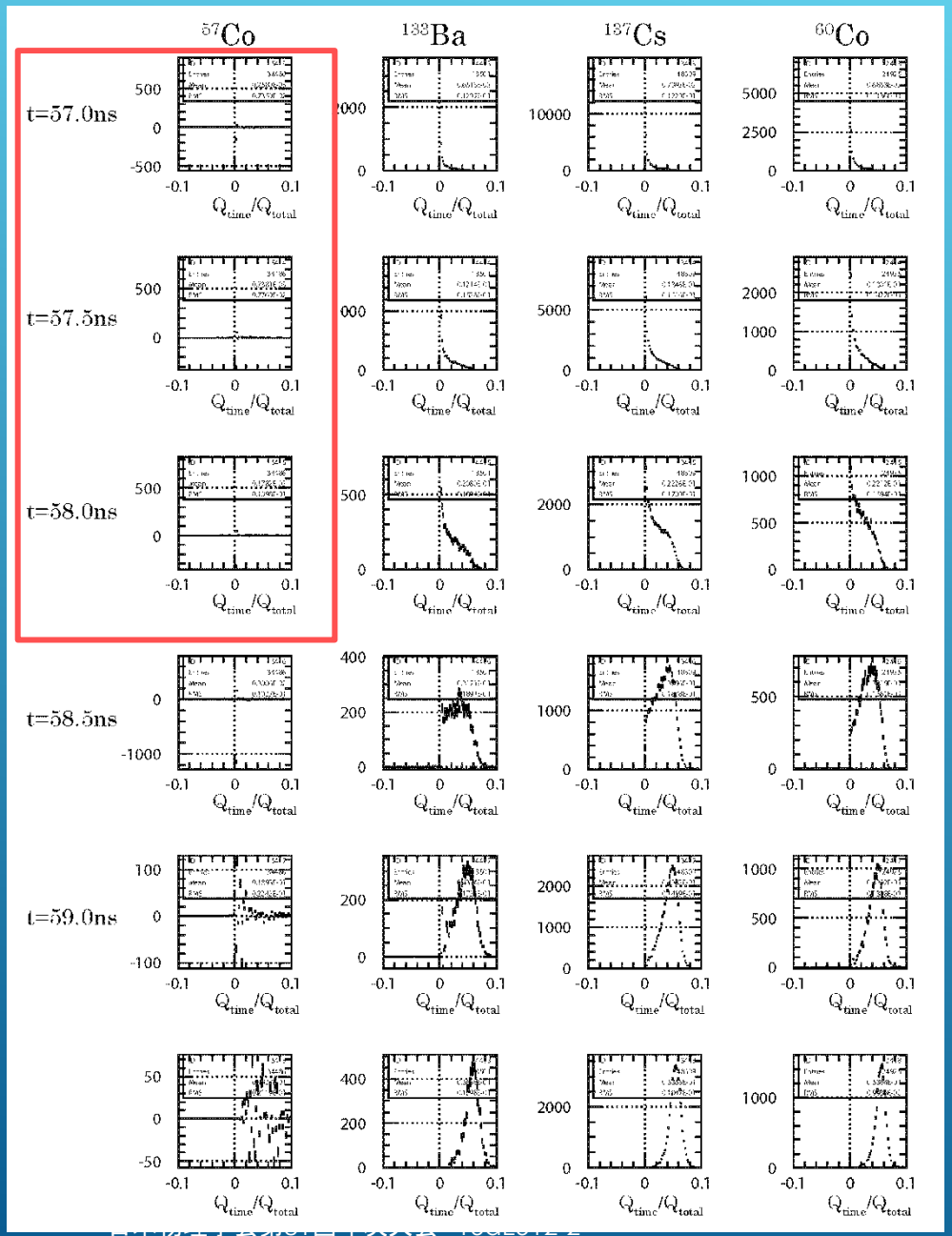
$$= 4.1 \pm 0.6\% \text{ at } 3.35\text{MeV}$$

日本物理学会第81回年次大会　15aE512-2

How to distinguish ^{208}Tl and DBD



ZICOS LSを用いたガンマ線事象の立ち上がり



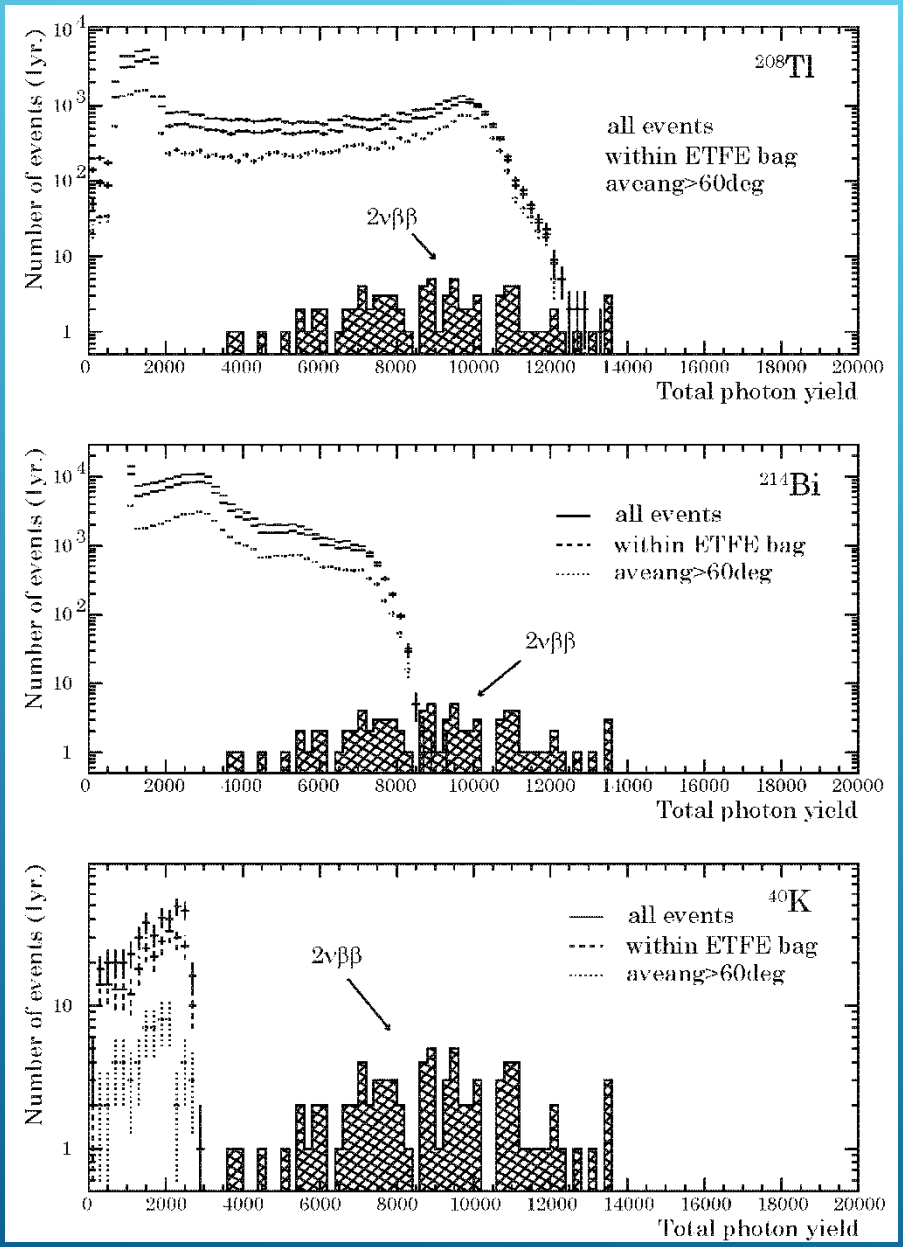
- t=57ns~58nsの時間帯で立ち上がり方に違いが見える
- エネルギーが大きいほど立ち上がり方も大きい
- t>58.5nsの波形はエネルギーによらず一定

t=57ns~58nsの波形はチェレンコフ光が優勢と考えられる。

^{57}Coの波形をシンチレーション光のテンプレートとして作成

Sep 15,2026

BG simulation in worst case



Assuming BGs from flask

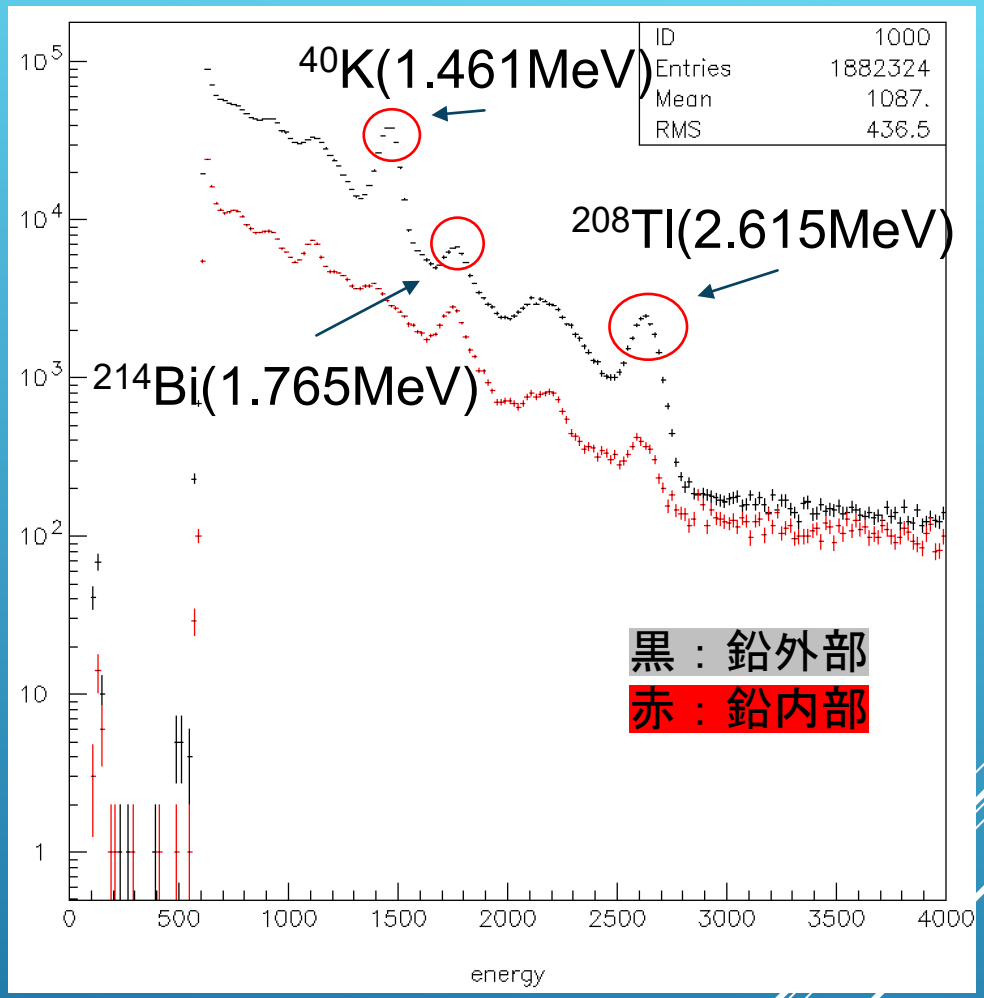
- ^{40}K affects only part of 2νββ observation.
- ^{214}Bi is significant BG, but small fraction of 2νββ events should be observed.
- ^{208}Tl is most serious BG for 2νββ. A few events might be observed.

Need actual measurement for ^{96}Zr 2νββ

10cm mini Pb shield and CsI detector

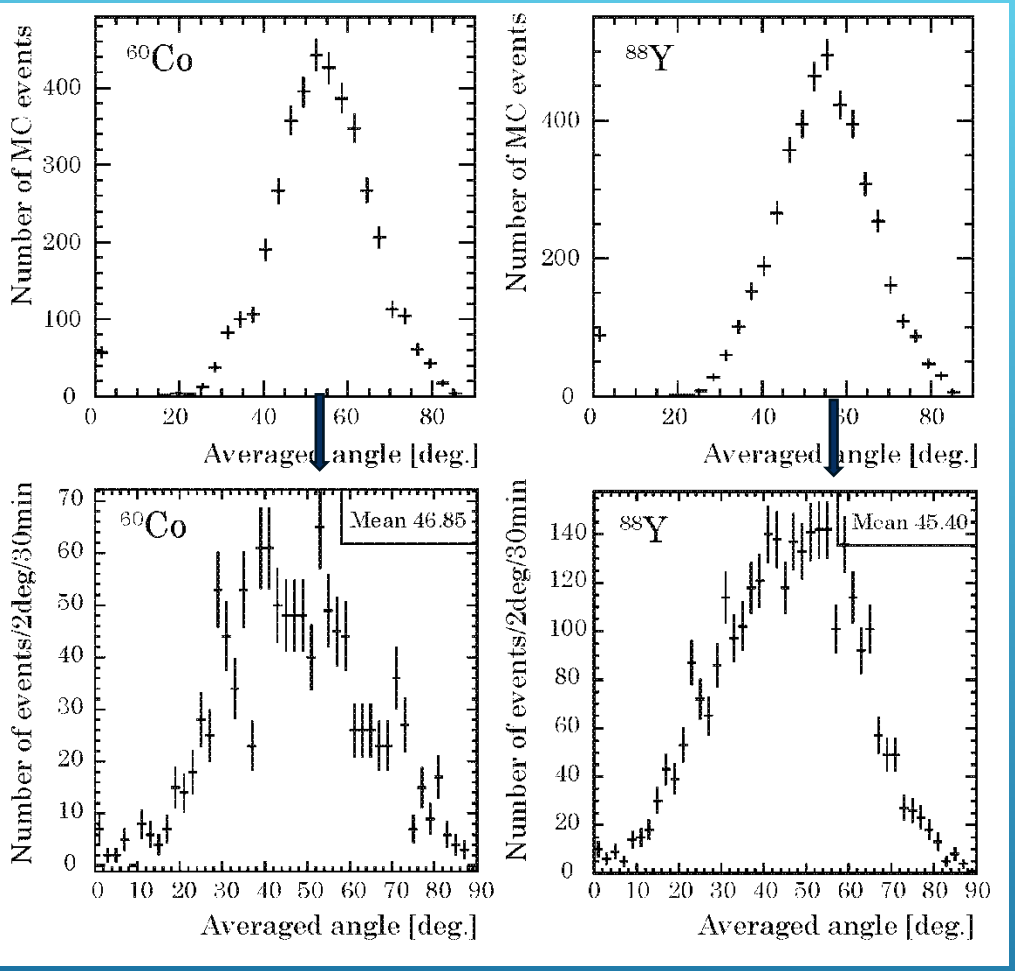


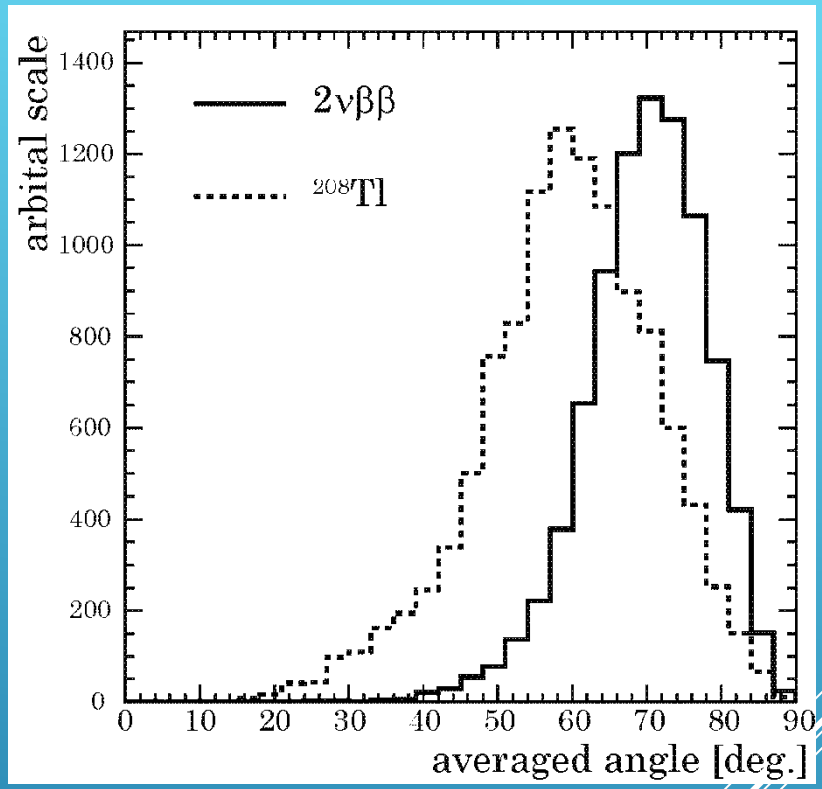




Due to limit of number of Pb block, we will use 15cmm thickness

Averaged angle using calibration data





The peak position were seen at 58 and 70 degree for $2\nu\beta\beta$ and ^{208}Tl β decay, respectively.

Averaged angle of simulation seems to roughly reproduce the calibration data (Selection of PMT which receives Cherenkov light might be OK), but vertex still does not reproduce well.