

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XXV
～ニュートリノを放出する二重ベータ崩壊事象
観測実験の初期結果報告～

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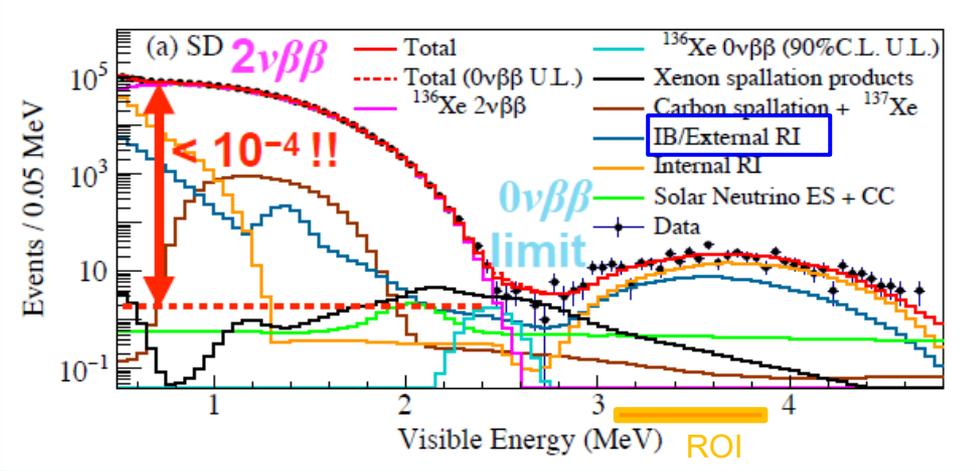
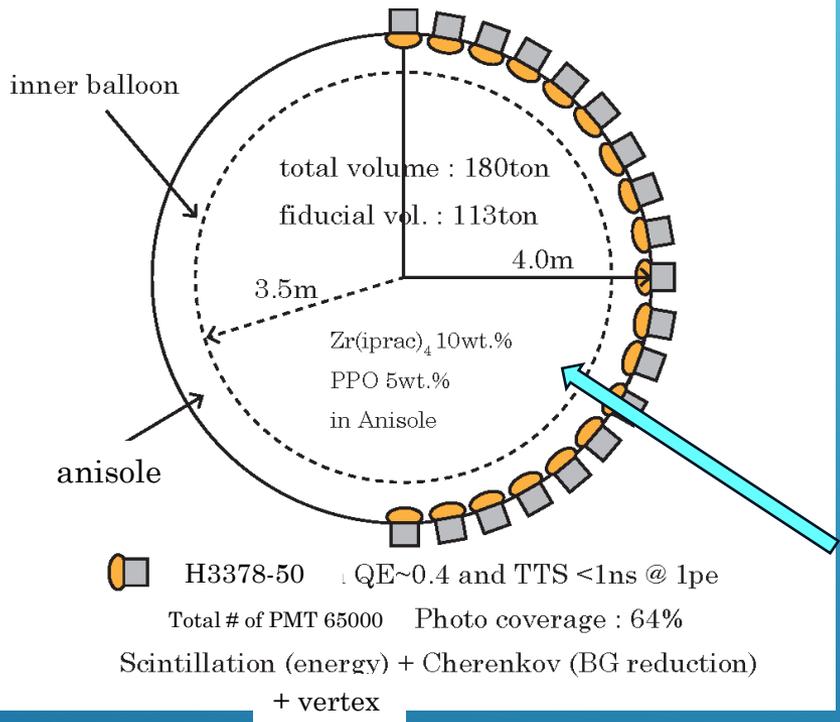
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ZICOS experiment for ^{96}Zr $0\nu\beta\beta$ observation

Conceptual design of ZICOS detector



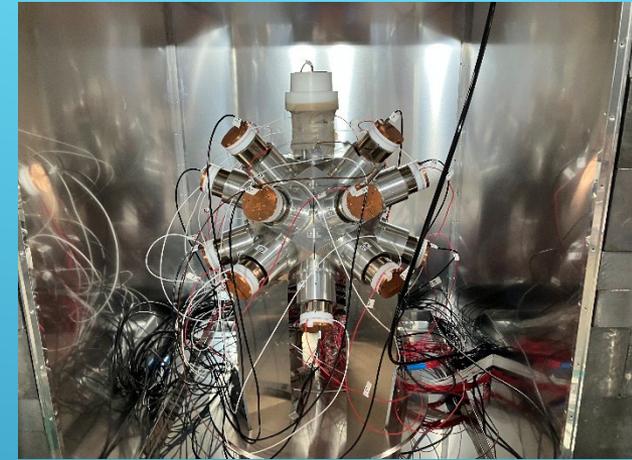
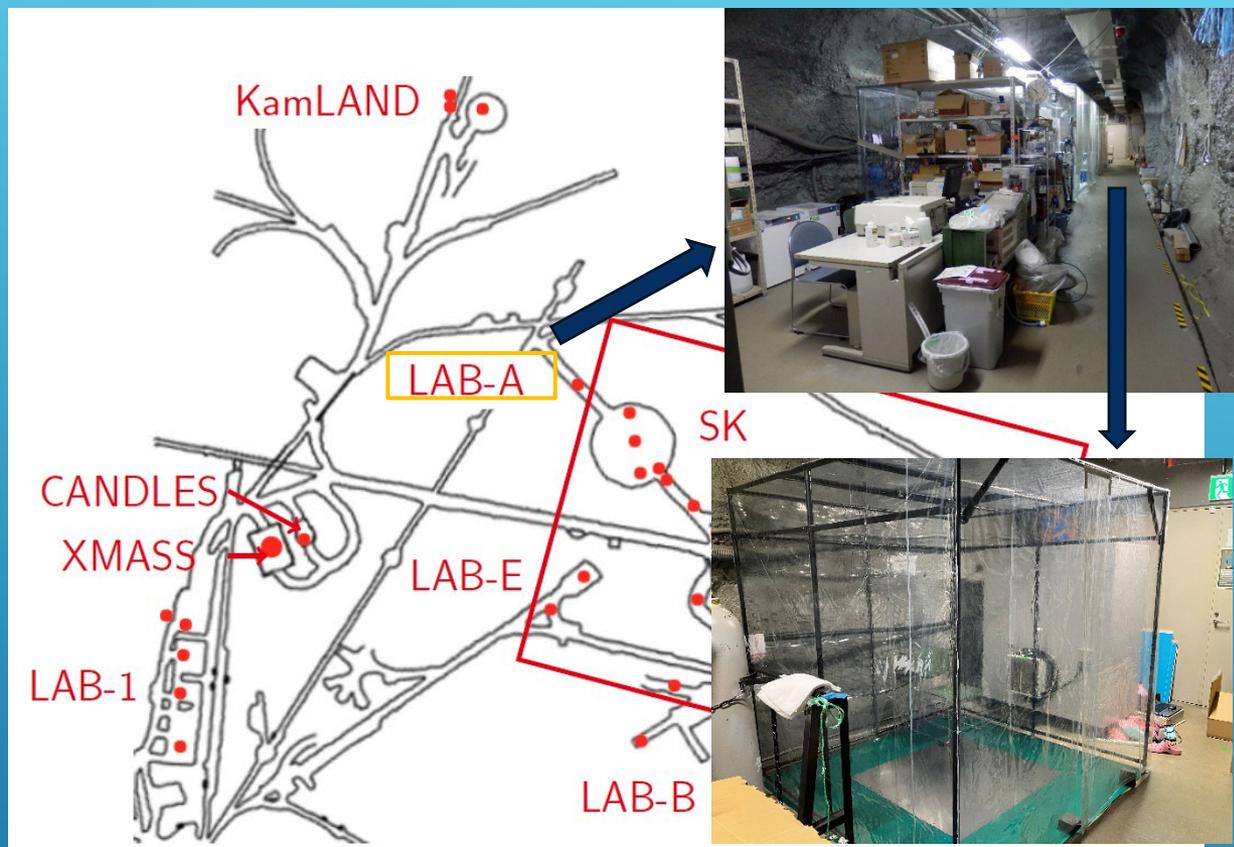
I. Shimizu, Plenary talk at Neutrino2024 conference.

Liquid scintillator loaded ^{96}Zr

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

^{96}Zr : 45 kg (nat.) \rightarrow 865 kg (50 % enrich) \rightarrow 1/20 BG
 $T_{1/2}^{0\nu} > 4 \times 10^{25}$ yrs $\rightarrow 2 \times 10^{26}$ yrs $\rightarrow \sim 1 \times 10^{27}$ yrs

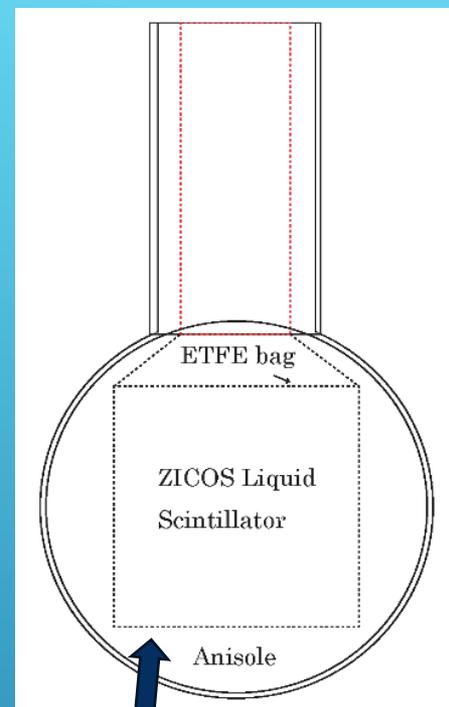
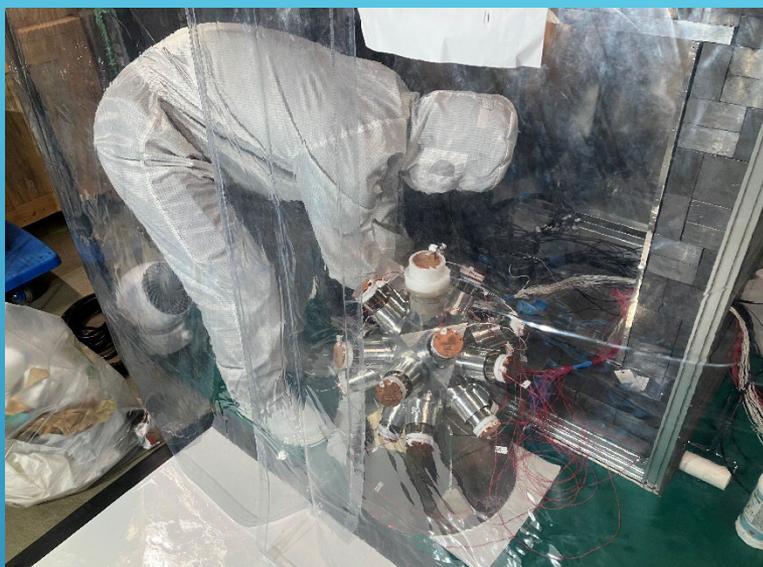
2ν -ZICOS for measurement of ^{96}Zr $2\nu\beta\beta$ events



- Clean booth was built in Lab-A at Kamioka mine.
- 650 Pb blocks was used for radiation shield with 15cm thickness.
- All safety issues such as fire alarm, leak sensor etc were installed before starting experiment.

The first data taking implemented over 50 hours from November 22nd to 25th.
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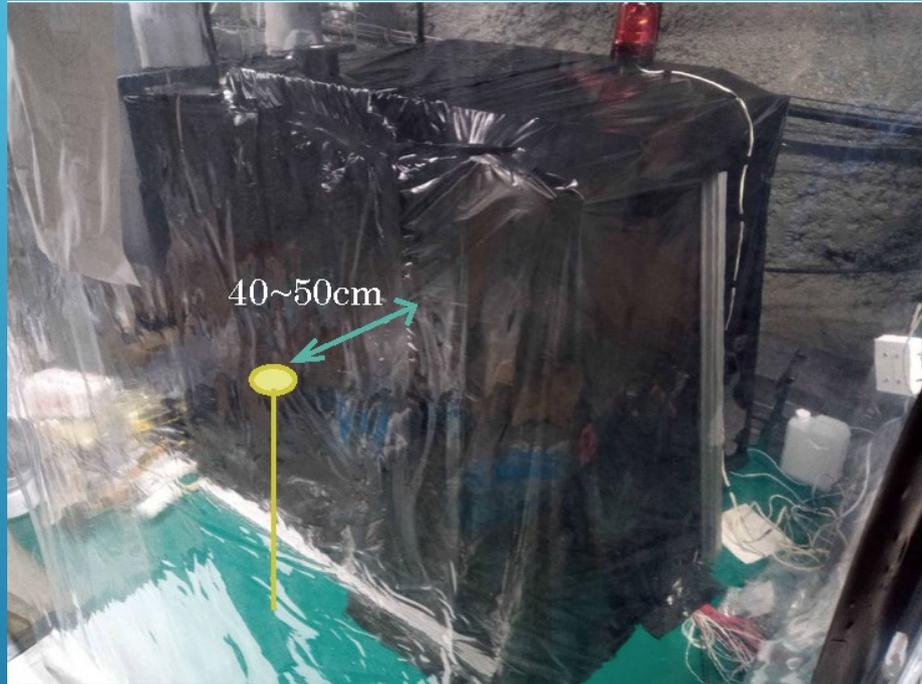
2ν-ZICOS detector



- 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 $\sim 70/yr$

9cm cubic ETFE bag for reducing external BG events

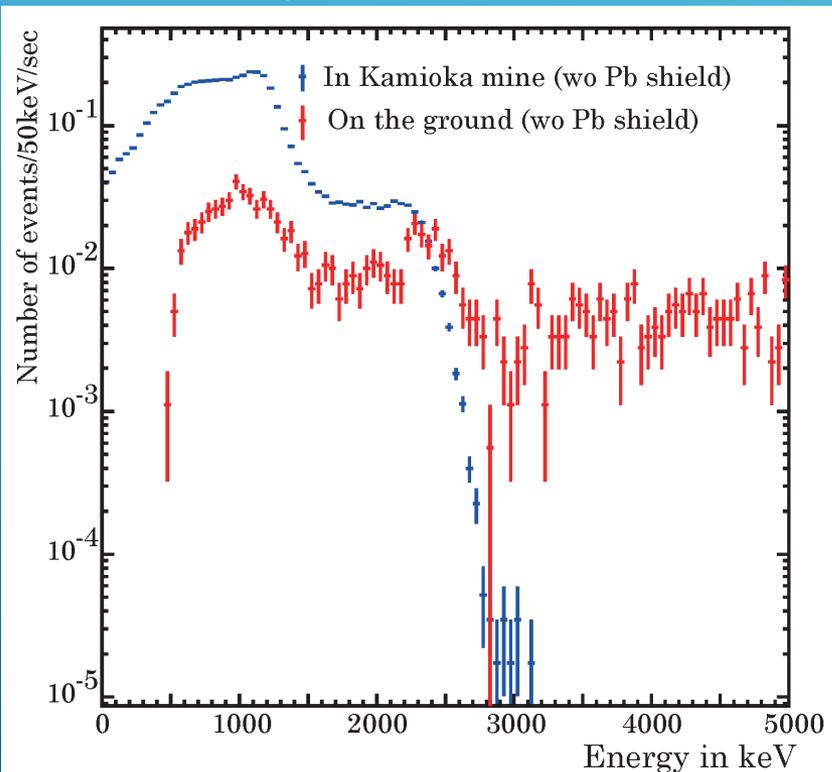
Energy calibration and DAQ for 2v-ZICOS detector



- ^{137}Cs , ^{60}Co and ^{22}Na source were used for energy calibration. (please look at **Kato-san's talk**)
- Those sources were located at front of Al wall without front Pb shield.
- All data were recorded by V1742 digitizer.

Data taking in 2025

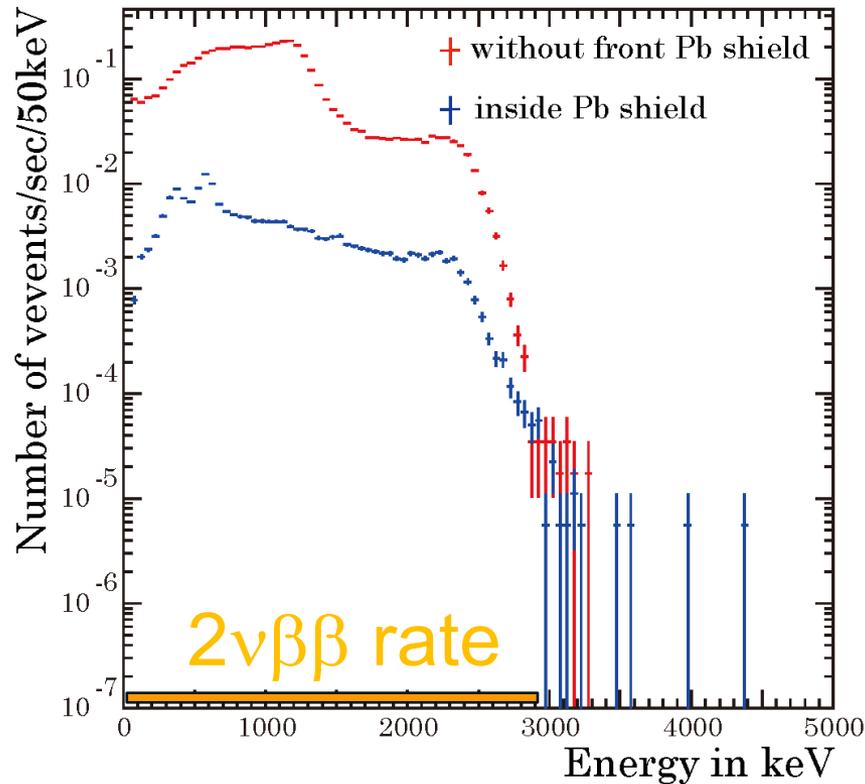
- Period : 2025.Nov.22nd – 26th
- Background run without front Pb shield : 100800sec
- Background run inside Pb shield : 180000 sec



- A few muons were observed even half day measurement.
- Environmental γ events from ^{40}K and ^{208}Tl were clearly observed in case of without front Pb shield.
- Those amount was about 10 times higher than events measured on the ground.

At maximum, an environment γ event rate might be 100 times higher than event rate on the ground.

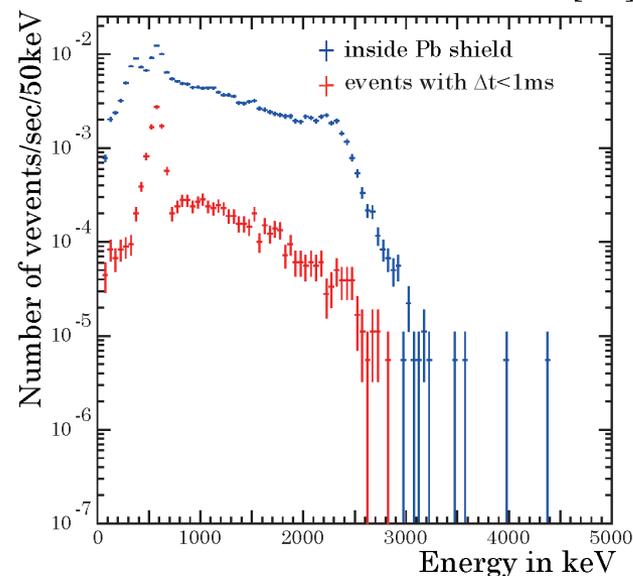
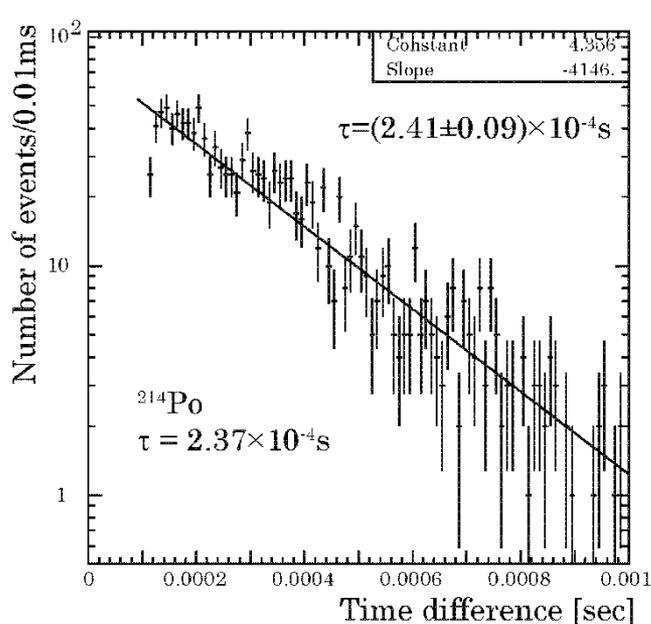
Background events observed in Pb shield



- 2.615MeV γ events from ^{208}Tl were observed.
- Despite of 10^{-4} reduction for environmental γ with 15cm Pb shield, BG rate was 3 order magnitude higher than expected rate.
- Peaks were found around 600keV and 400keV.
- Shoulder events were found above 2.6MeV

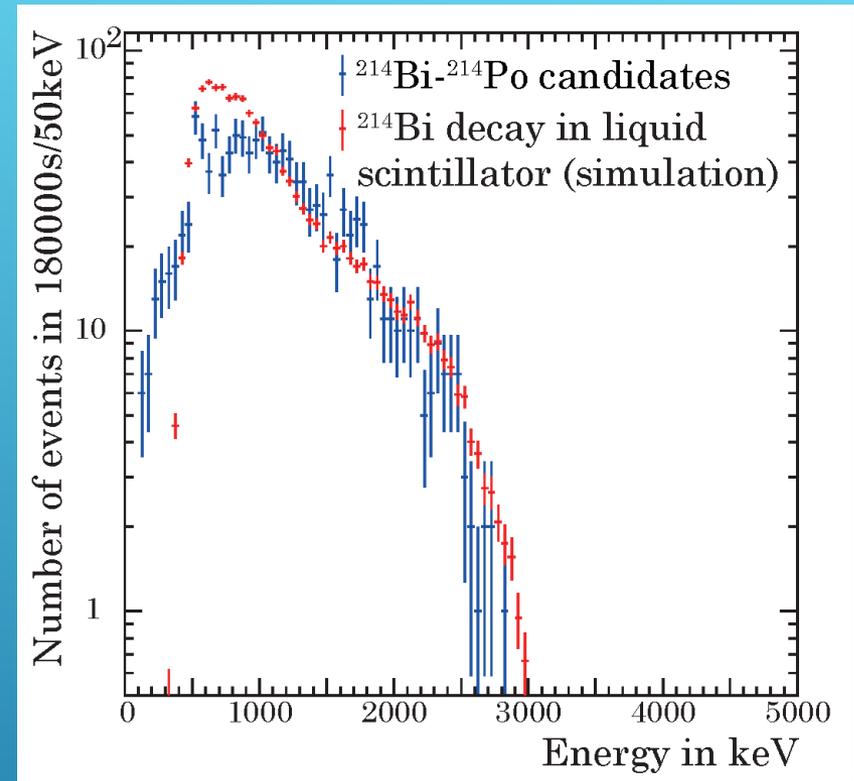
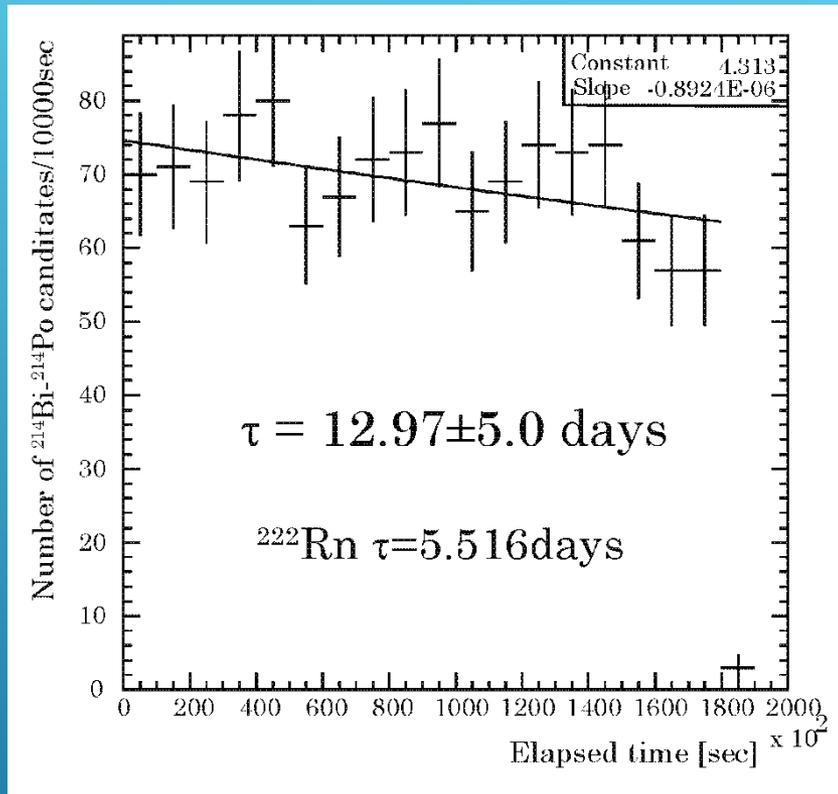
Measured event rate is over 4 order magnitude higher than expected $2\nu\beta\beta$ event rate ($\sim 10^{-7}$ event/s/50keV)

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



- $0.1\text{ms} < \Delta t < 1\text{ms}$ 1250 events (+650 events). No event $\Delta t < 0.1\text{ms}$ due to dead time.
- Time slop is consistent with ^{214}Po lifetime.
- Parent events look have a β spectrum of ^{214}Bi decay.
- Peak at $\sim 600\text{keV}$ should be caused by α s of ^{214}Po decay.
- Amount of ^{214}Bi β decay events are **3 order magnitude** higher than expected $2\nu\beta\beta$ event rate.

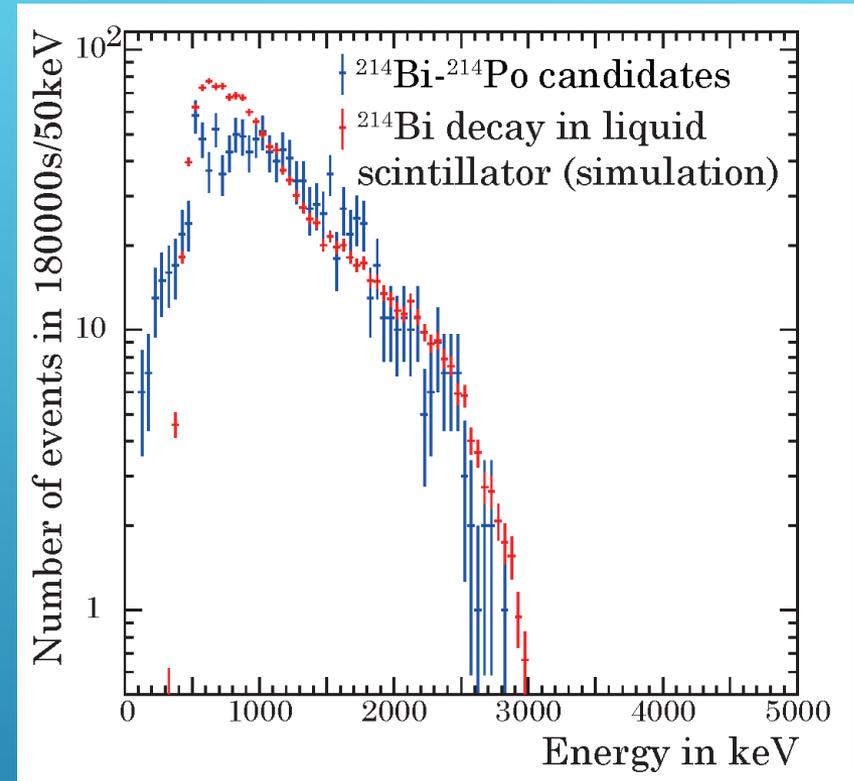
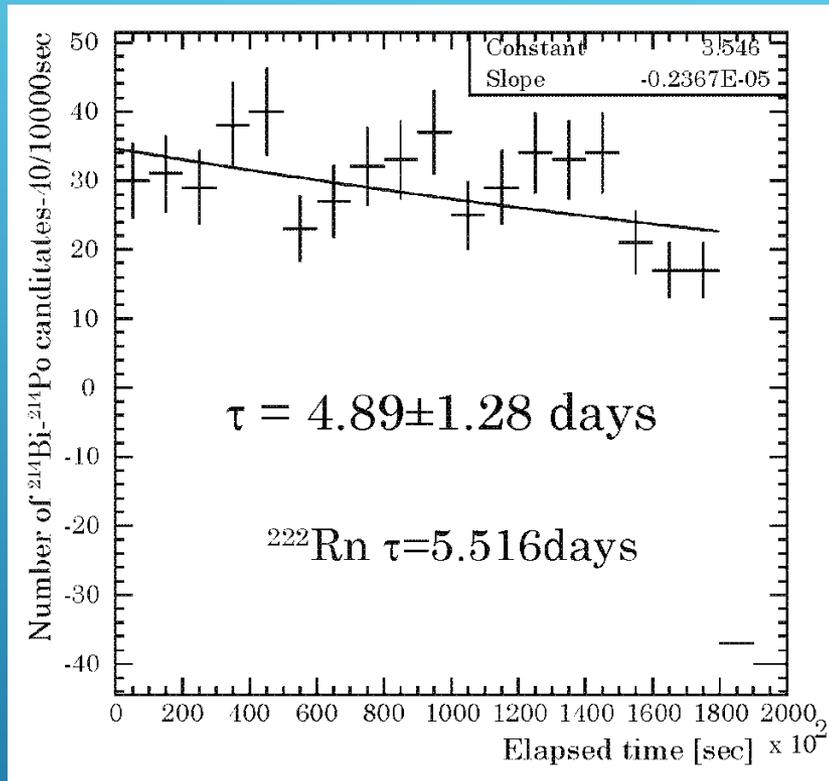
^{222}Rn contaminated in liquid scintillator?



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

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.

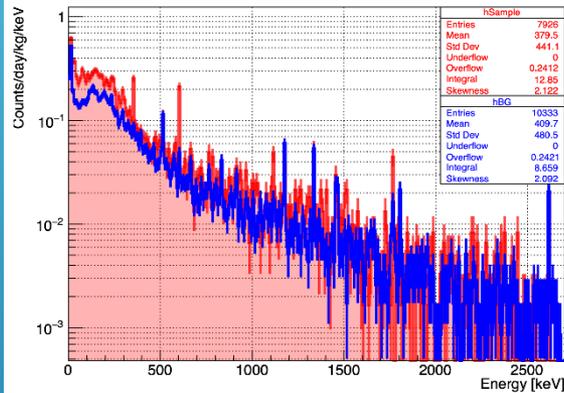
Background measurement using Ge detector

2024/11/07 LabC02

ZICOS LS

sample 224.6g, 35.2days

Unit: mBq/kg



summary table

unit : mBq/kg

	value	error	limit
U			
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

40K	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

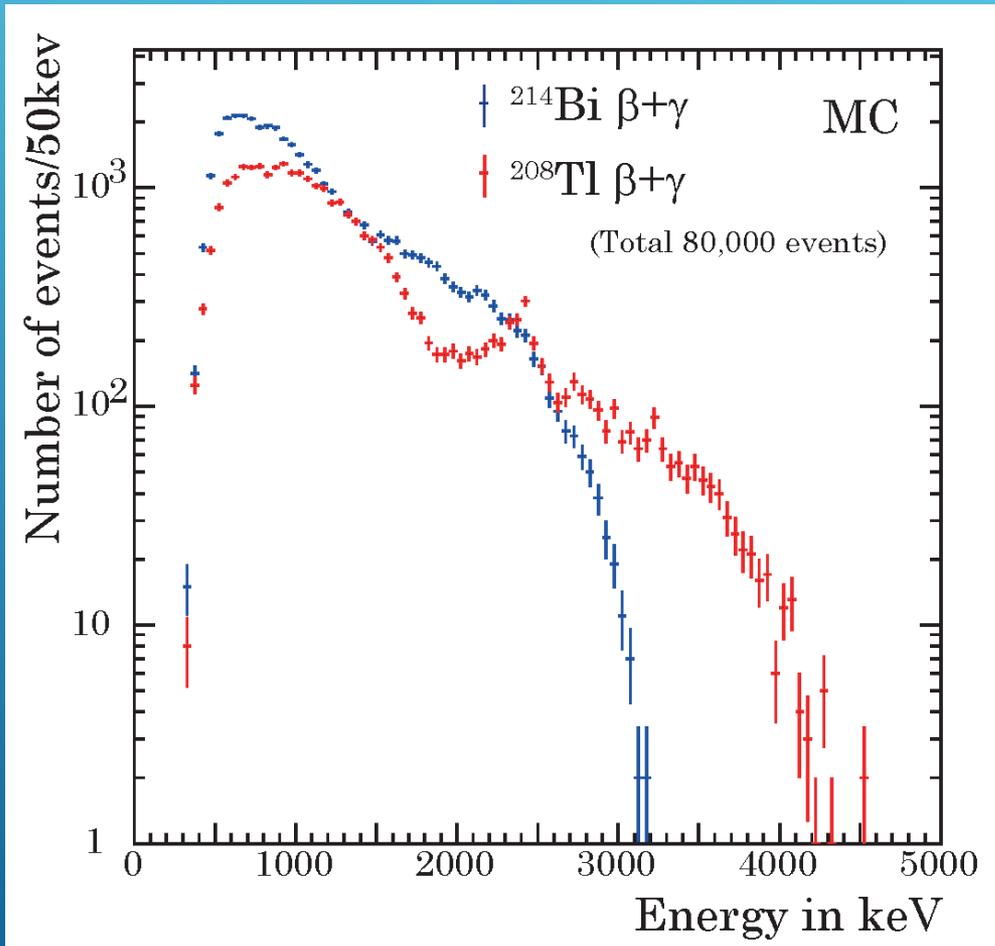
²³⁸ U upper (²³⁸ U)	²³⁸ U middle (²²⁶ Ra)	Th-chain (²²⁸ Ra)	Th-chain (²²⁸ Th)	U-235 (²³⁵ U)	U-235 (below ²²³ Ra)	K-40	Co-60	Cs-137	Lu-176
<12.85	2.72 ± 1.33	<4.36	<1.80	<6.97	<3.28	<15.50	<0.777	<0.97	<0.66

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

Almost consistent with measured value by Ge

How much amount of Th-chain in LS?

No ^{212}Bi - ^{212}Po candidates ($T_{1/2}=2.99 \times 10^{-7}\text{sec}$) was observed due to dead time of V1742 digitizer.

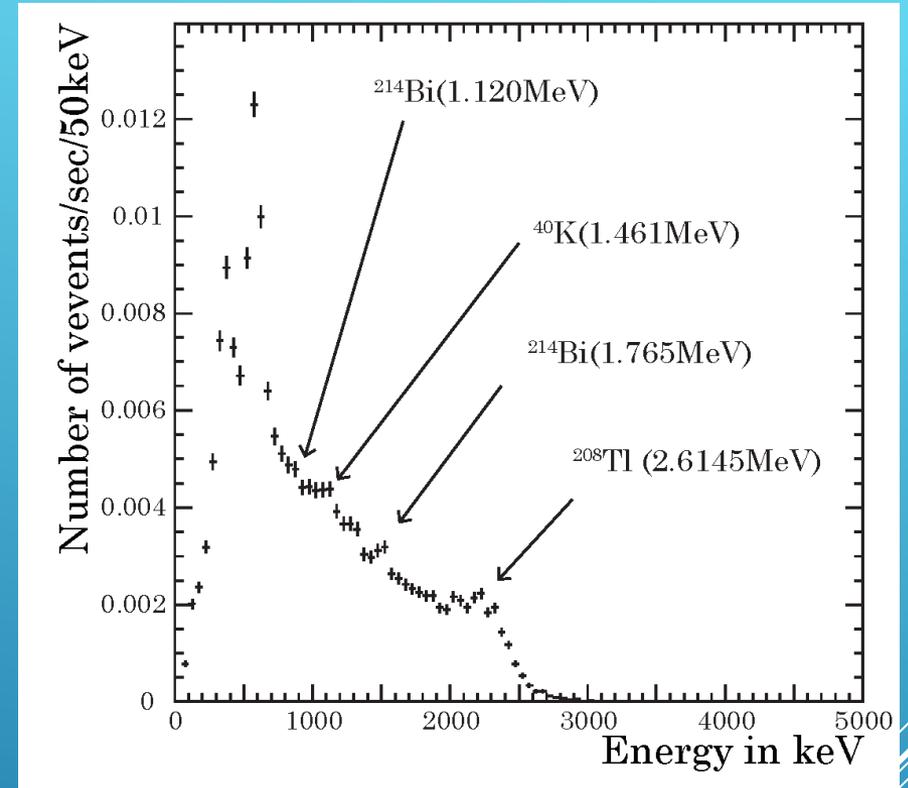
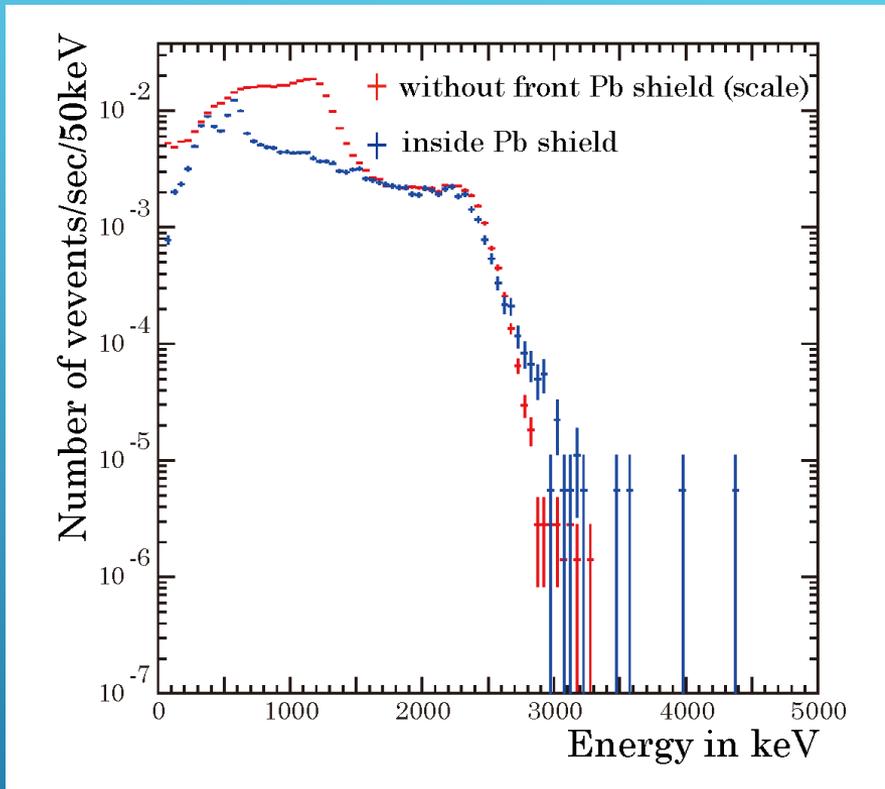


Energy spectrum of ^{208}Tl β decay should have a higher energy tail than that of ^{214}Bi beta decay.



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

What is major source of measured events ?



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

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

Background γ events from Quartz

単位 : ng/g

試料名	K	Th	U
テトラキス (アセト酢酸イソプロピル) ジルコニウム	-	-	-
定量下限	400	50	50

※表中の「-」表記は、定量下限以下であることを示します。

単位 : ng/g

試料名	K	Th	U
四塩化ジルコニウム	-	-	-
定量下限	200	50	50

※表中の「-」表記は、定量下限以下であることを示します。

単位 : ng/g

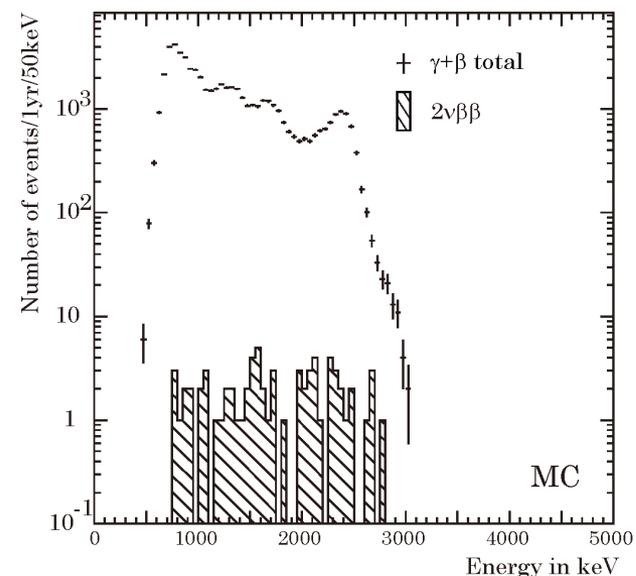
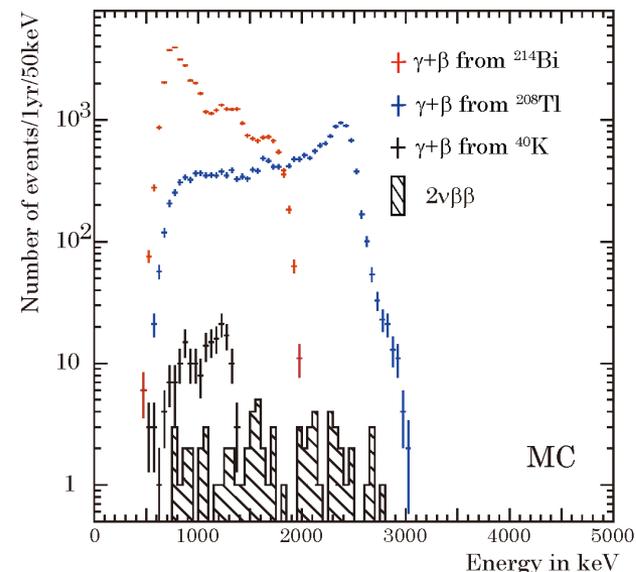
試料名	K	Th	U
RQ200	330	42	64
GE214	180	15	29
定量下限	5	1	1

^{232}Th : $6.09 \times 10^{-5} \text{Bq/g}$

^{238}U : $3.58 \times 10^{-4} \text{Bq/g}$

^{40}K : $5.59 \times 10^{-6} \text{Bq/g}$

3 order magnitude higher level



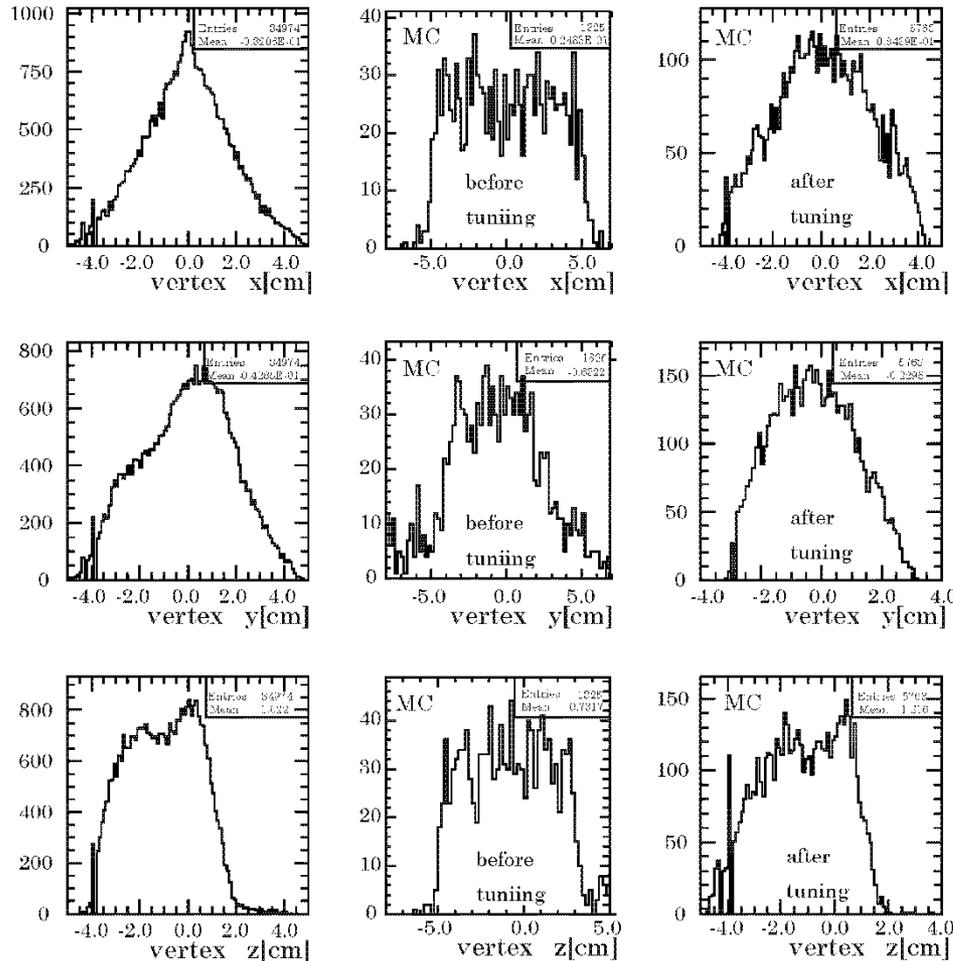
Present status and future plan

- Measured background event rate is over **4 order magnitude higher** than expected $2\nu\beta\beta$ event rate.
- Clear 1250 events of ^{214}Bi - ^{214}Po candidates were observed. It is almost **3 order magnitude higher**, so we need distillation for Anisole and review for synthesis process of $\text{Zr}(\text{iPrac})_4$ to reduce ^{226}Ra contamination.
- However external γ events from Quartz should be **3 order magnitude higher**, major γ s might come from Al structure of PMT mounting jig. Need clean material such as **ultra-pure synthetic quartz** and **oxygen free copper** as scheduled KAKENHI project.
- Need **20cm Pb shield (15+5cm thickness)** to reduce sufficiently events from environmental γ s.

backup

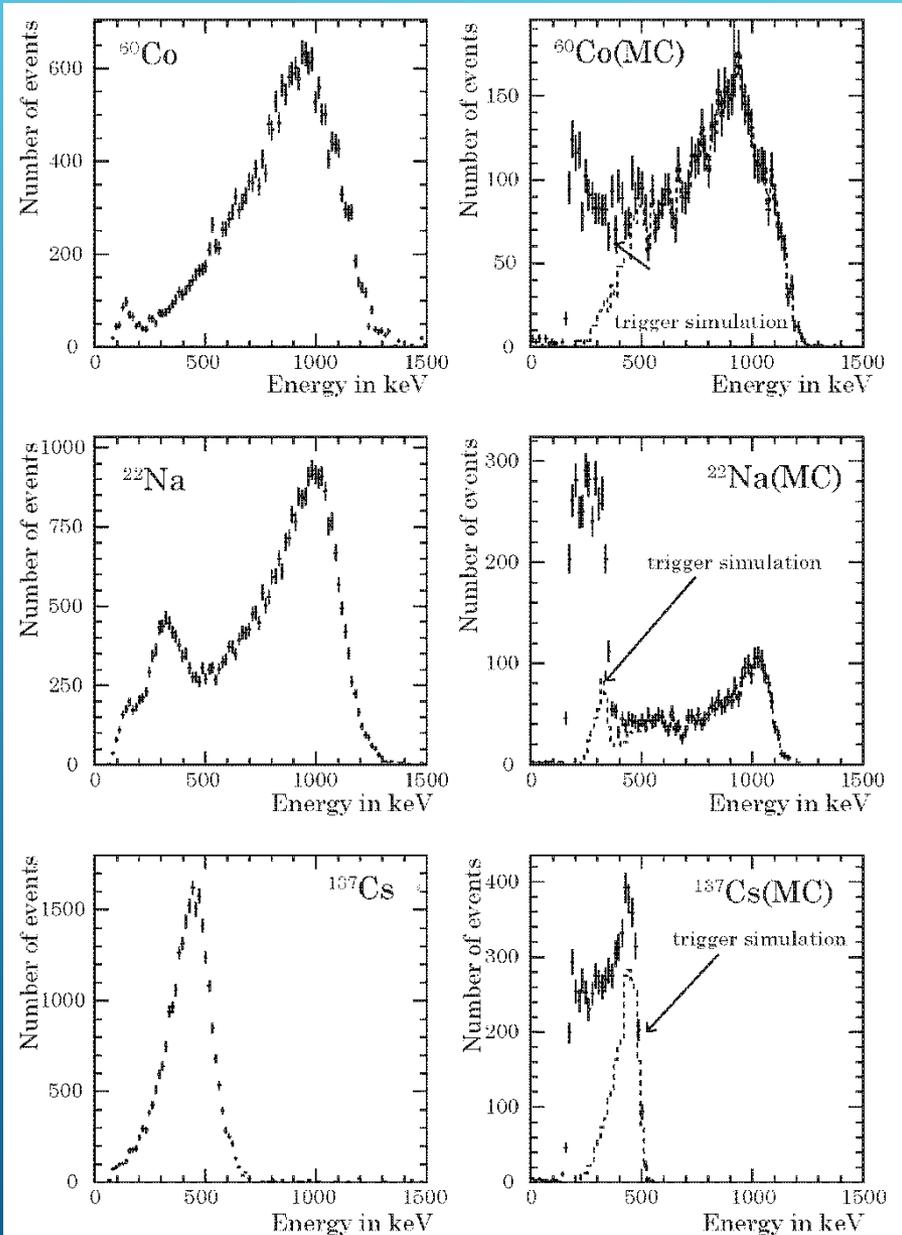
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Monte Carlo tuning using calibration source



- 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.

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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**

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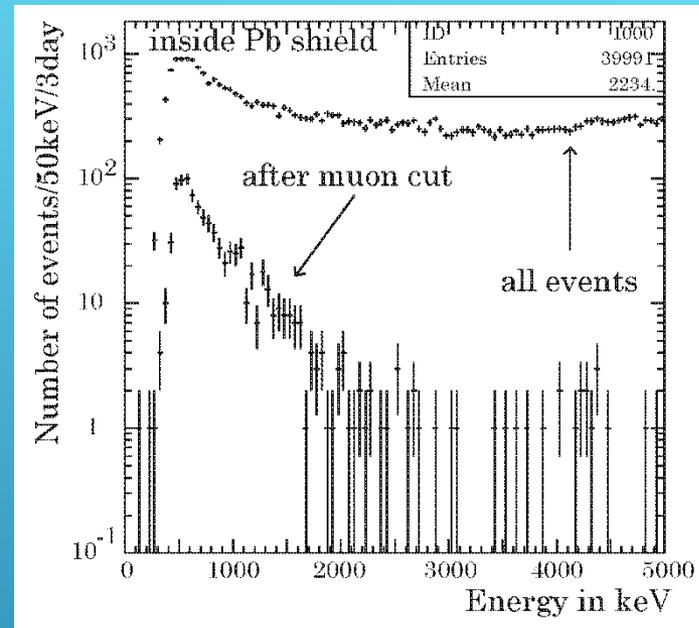
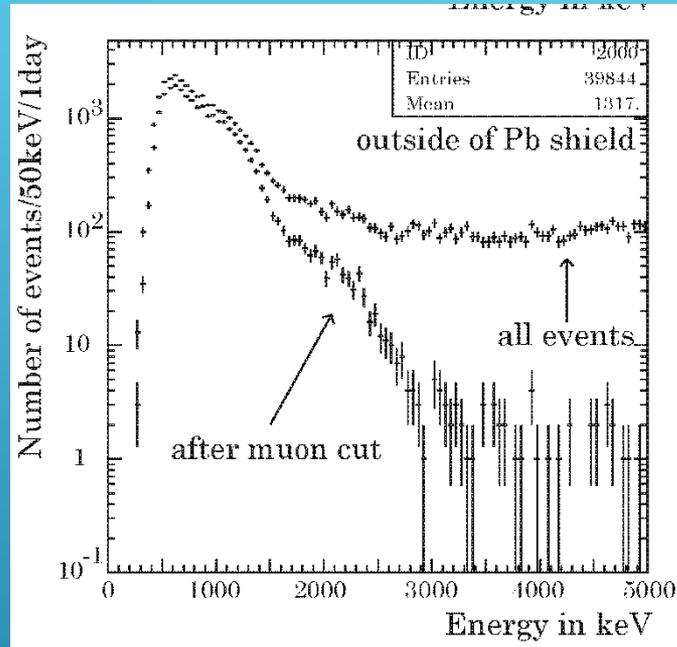
Why 2ν -ZICOS should do in Kamioka mine?

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

- Cosmic muon rate is $\sim 2\text{Hz}$ on the ground.
- Most significant spallation product is ^{11}C which decays in beta plus and emits total energy $\sim 2\text{MeV}$.
- 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 $2 \times 10^{-5} \text{ Hz}$, 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)

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Energy spectra using Quartz vial detector



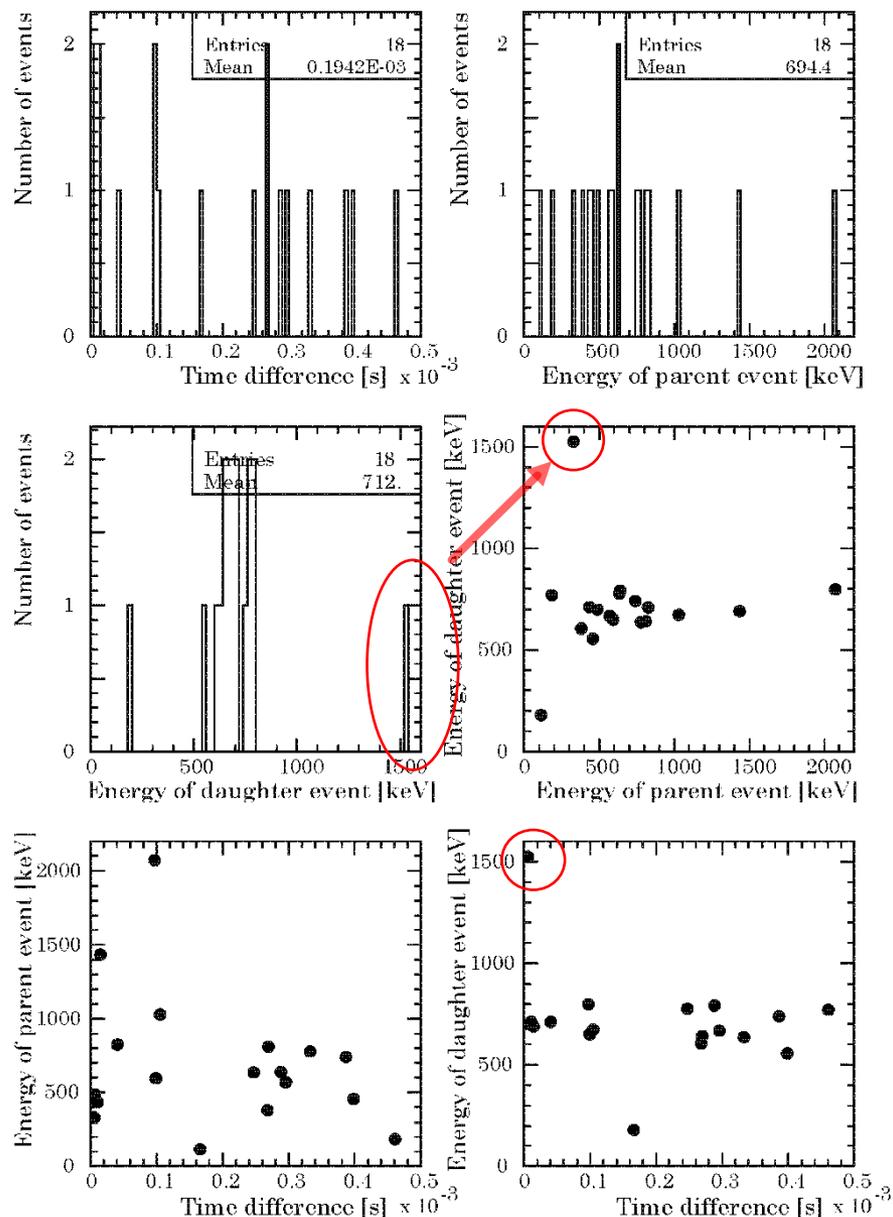
Outside of shield

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 ($\tau = 1.643 \times 10^{-4} \text{sec}$)

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

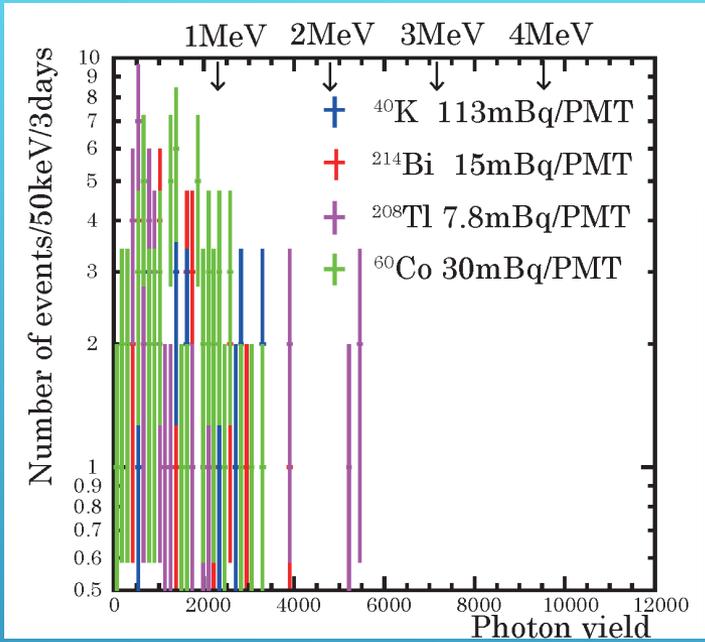
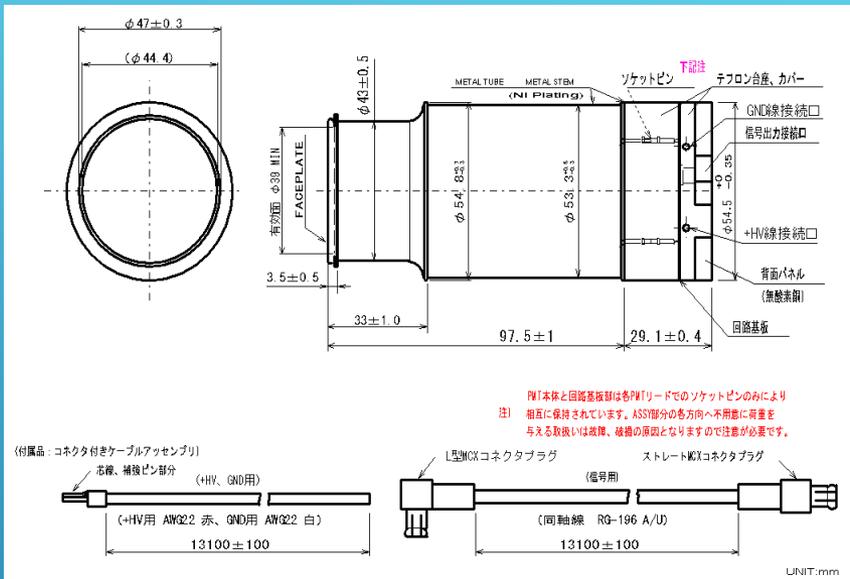


- 18 events have time difference $< 5 \times 10^{-4}$ s except parent muon
- Parent events have wider energy distribution than daughter events
- Daughter events have narrow distribution (like α quenching)

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

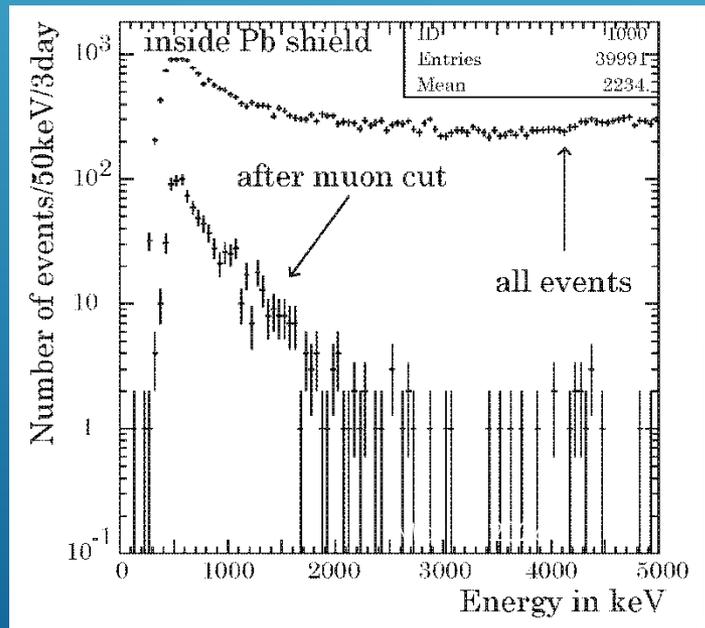
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BG events from PMT R10789

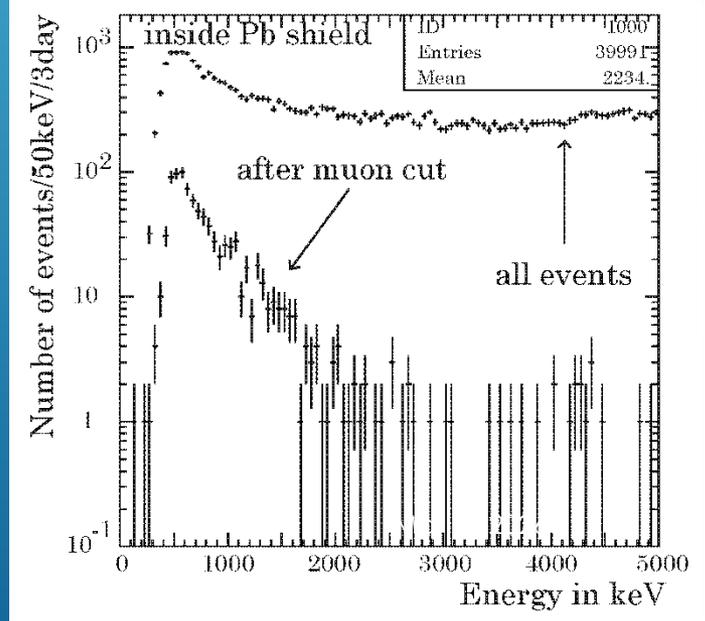
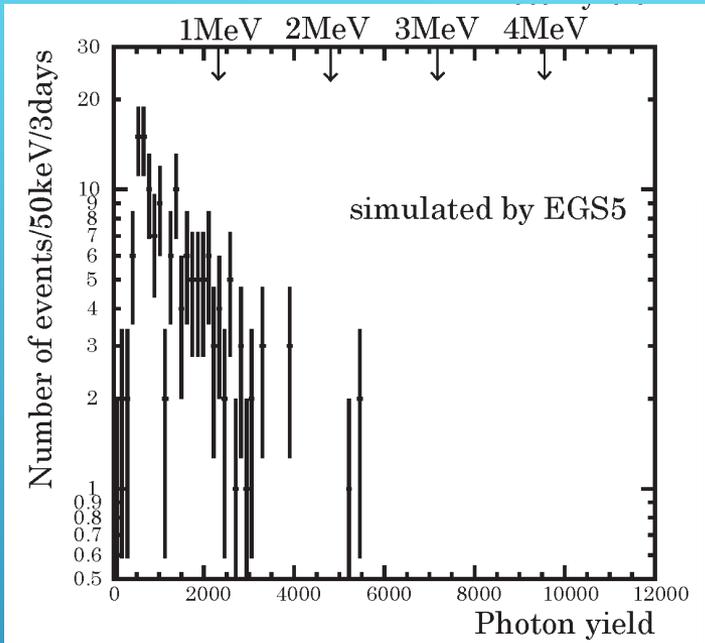
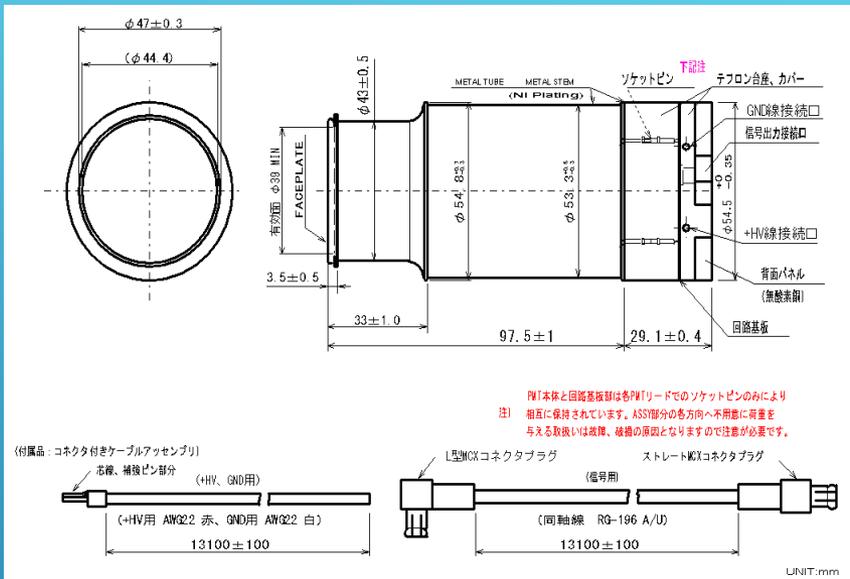


Ref: NIMA 922(2019)171-176

- 1.2 ± 0.3 mBq/PMT of ²²⁶Ra
- < 0.78 mBq/PMT of ²²⁸Ra
- 9.1 ± 2.2 mBq/PMT of ⁴⁰K
- 2.8 ± 0.2 mBq/PMT of ⁶⁰Co.



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

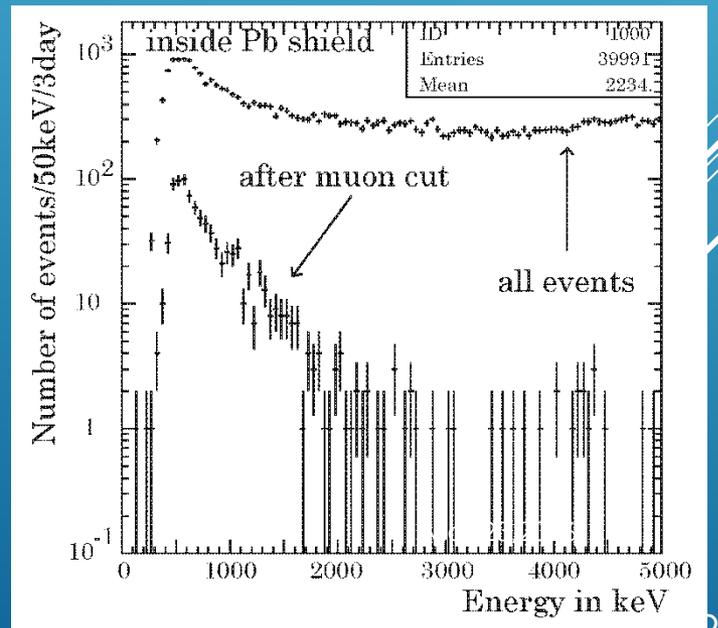
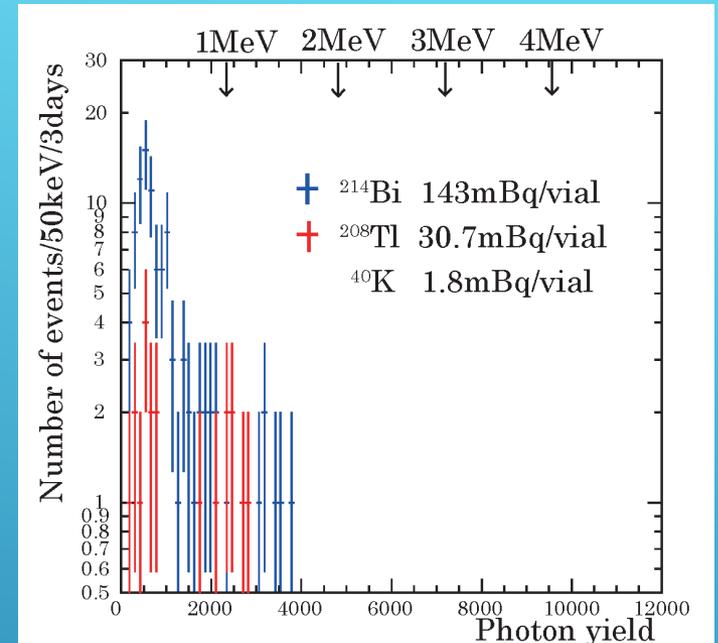


表 分析結果

試料名	単位 : ng/g		
	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



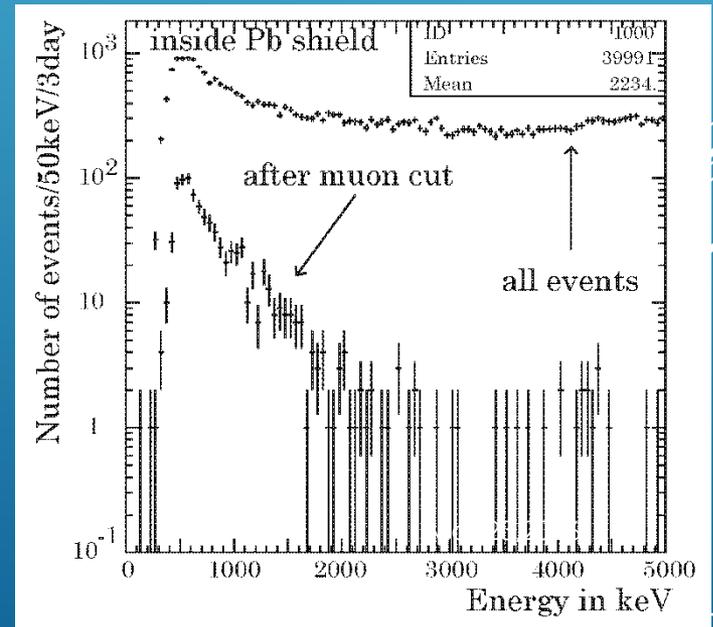
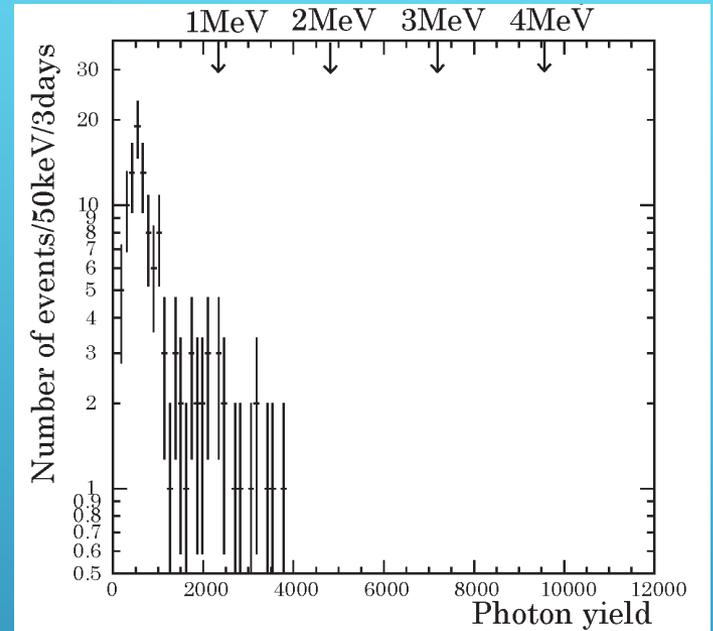
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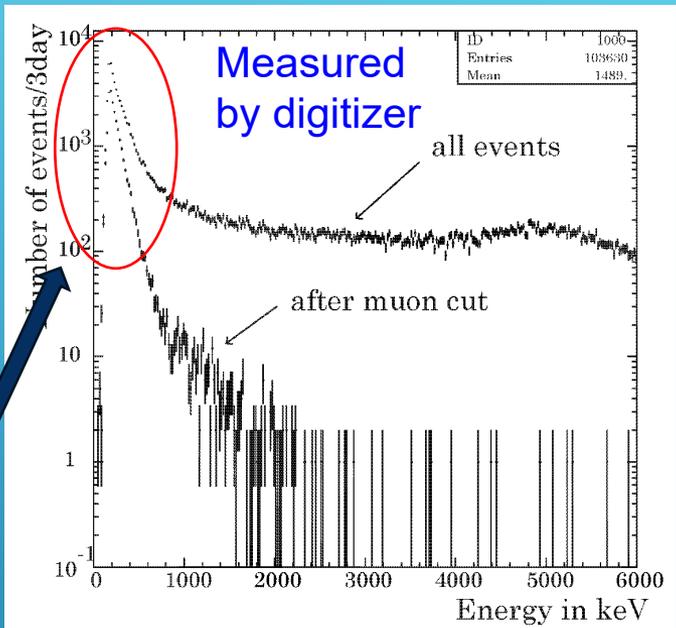
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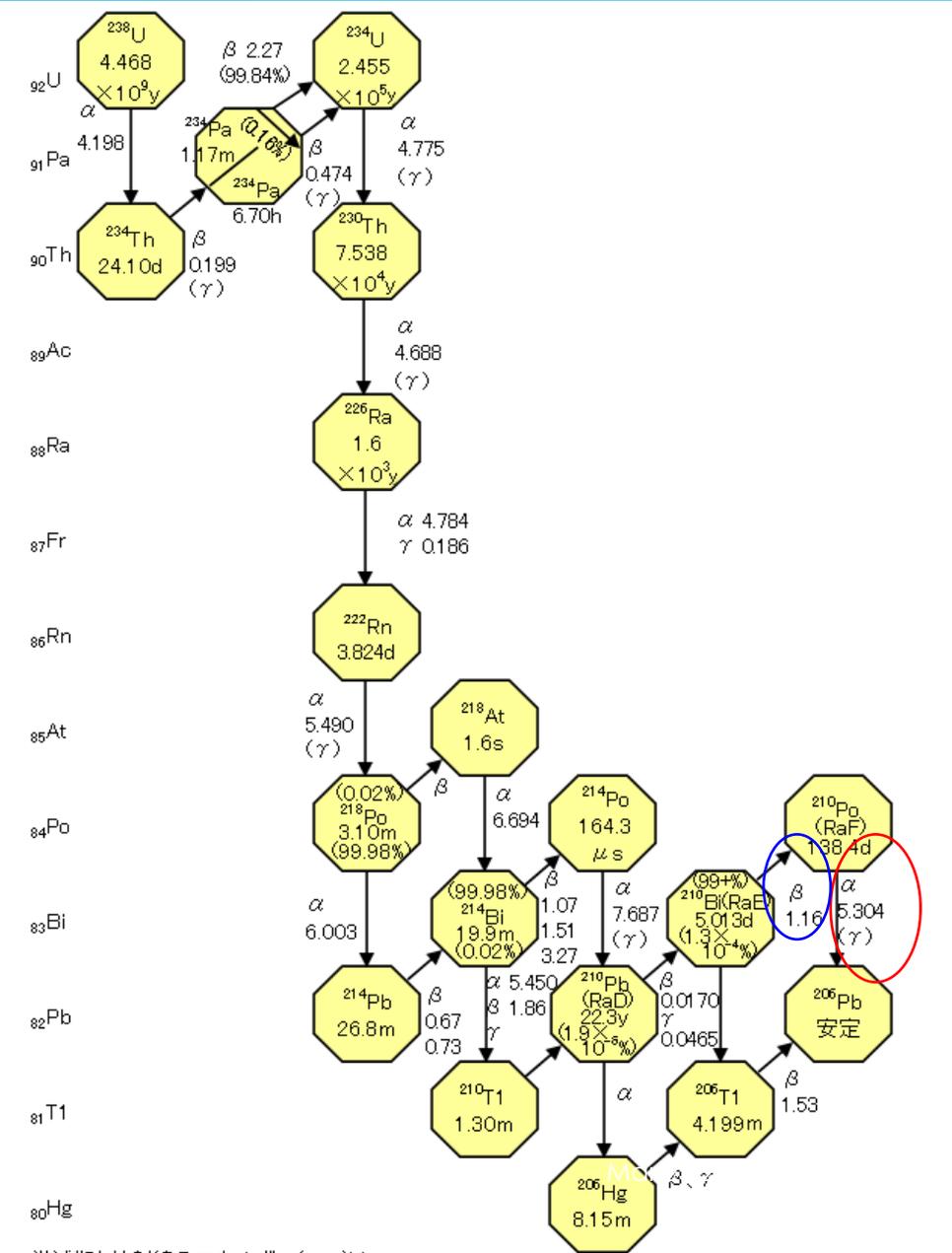
Even more 50 times larger BG events could not explain the residual events.



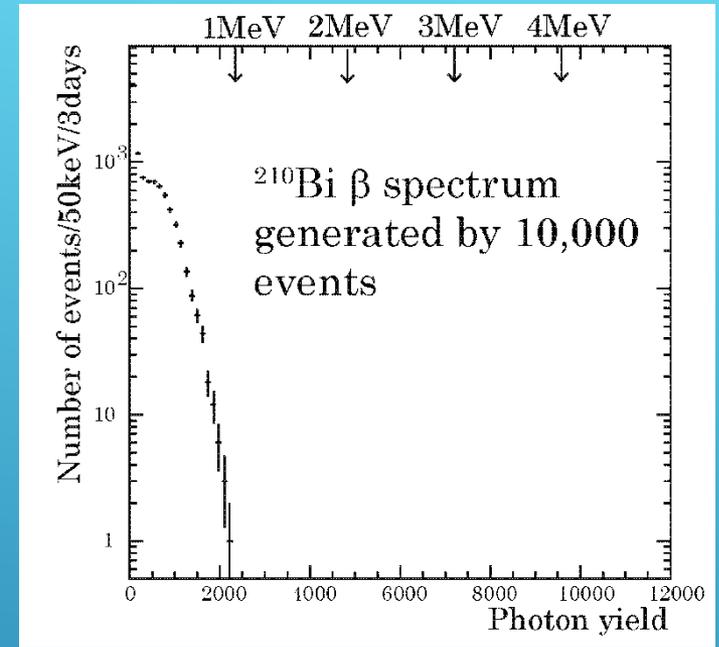
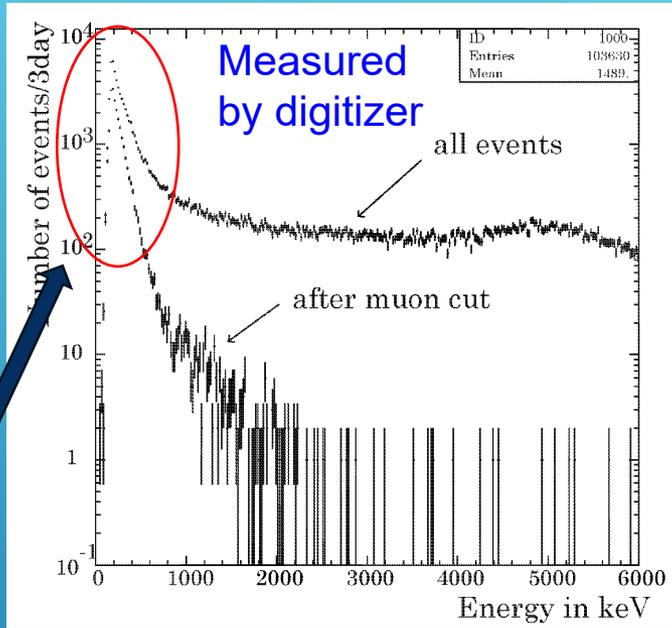
BG events from surface of Quartz



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

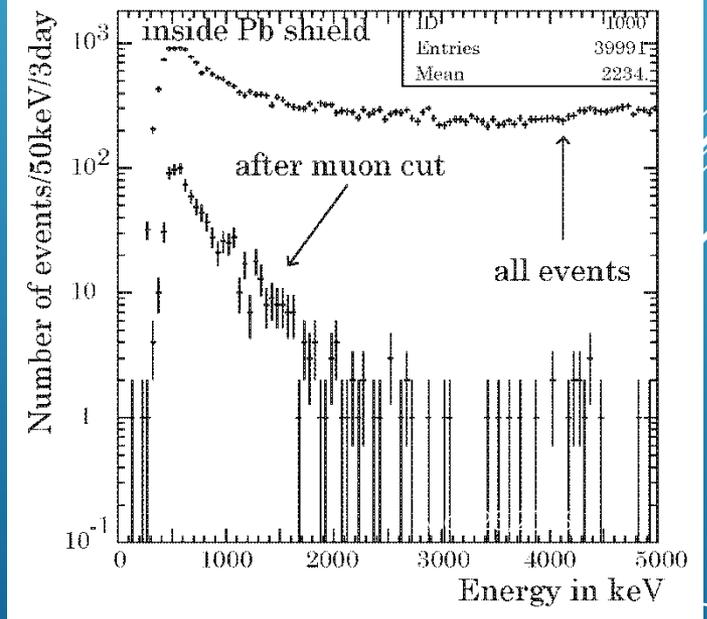


BG events from surface of Quartz

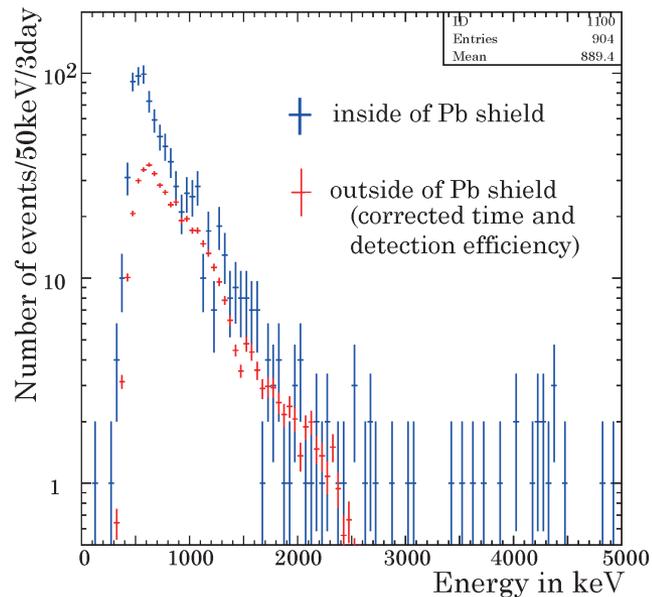
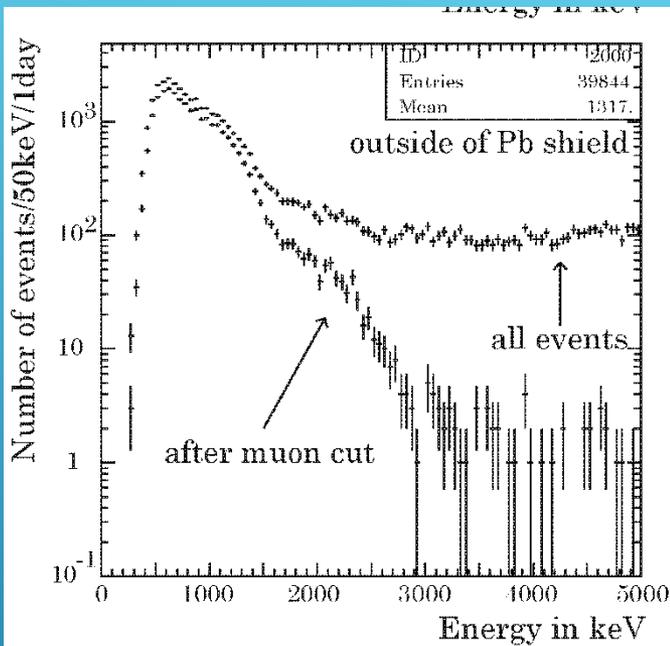


α from ²¹⁰Po? if so, β from ²¹⁰Bi should be observed by same order of events.

Observed rate correspond to 2mBq/cm², but XMASS estimated as 1 μ Bq/cm².



Comparing distributions using env. gammas

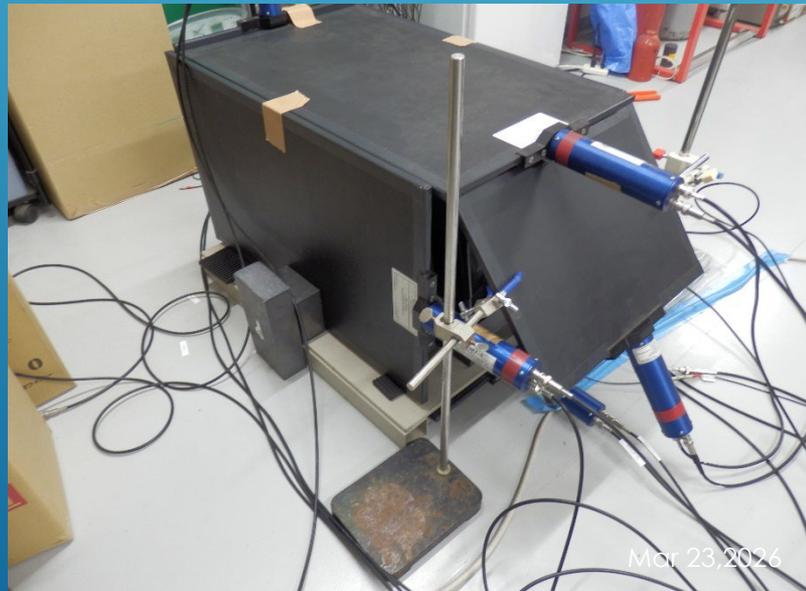
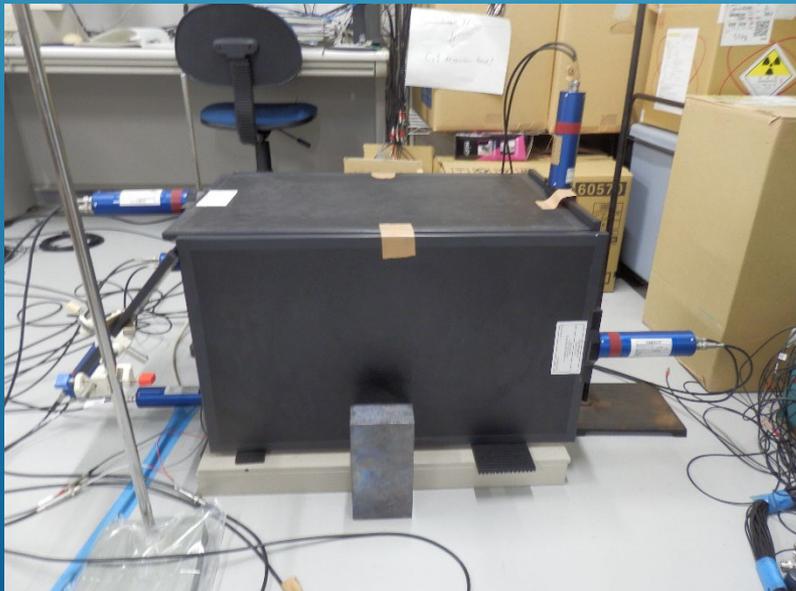
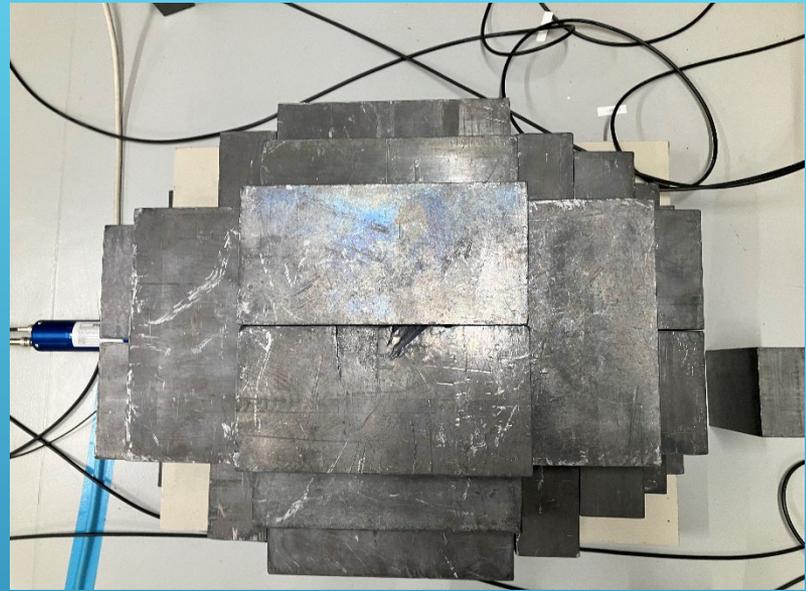


Almost the energy spectrum of residual events is consistent with environmental gammas observed at outside of shield.

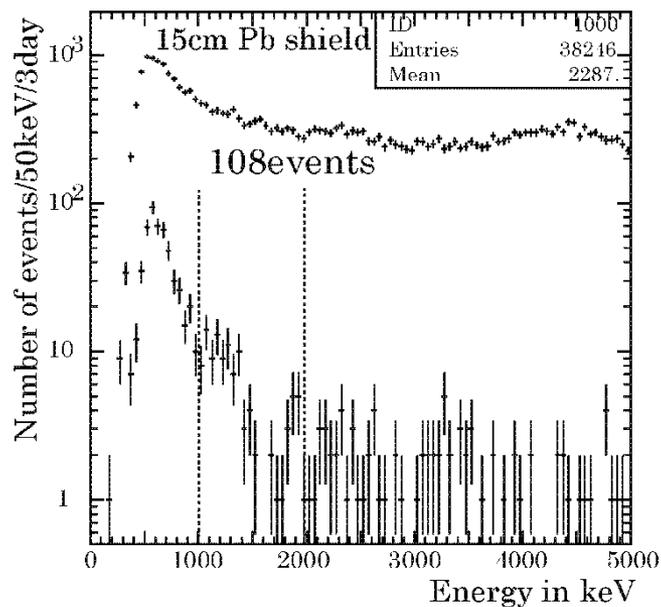
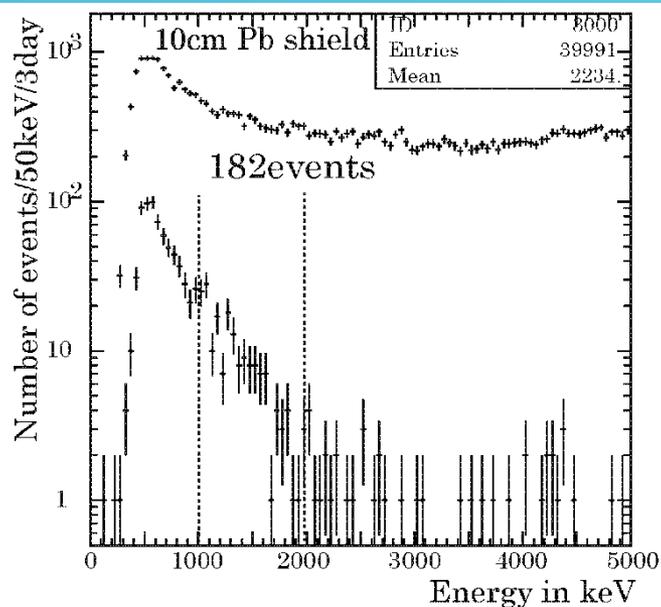
No obvious BG event exists except those above, we are still investigating the source of residual events.
(At least, it is not internal BG.)

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BG measurement inside 15cm Pb shield



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

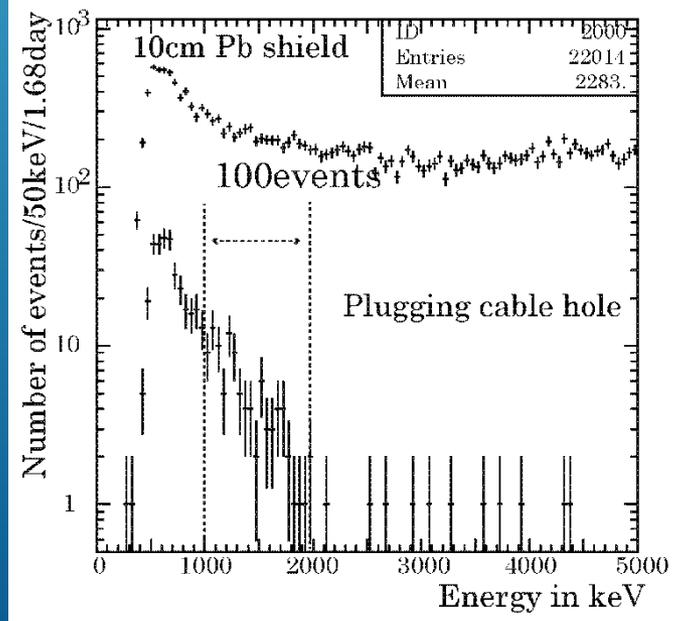
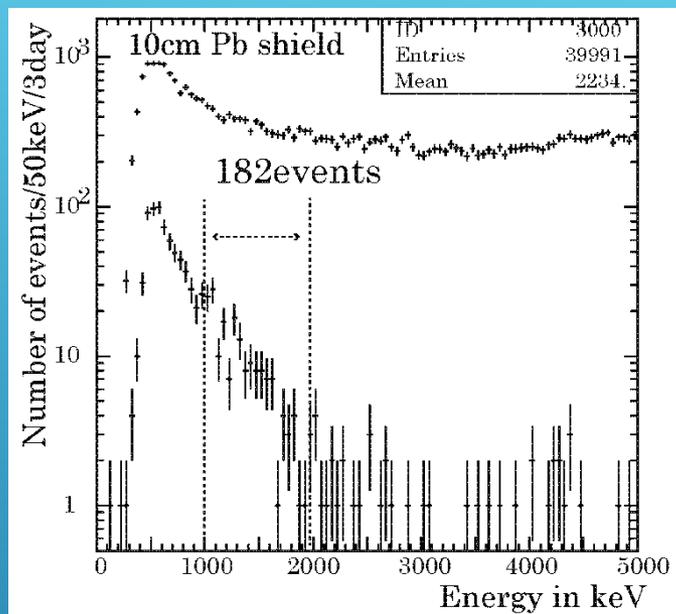
結論： 外部ガンマ線が若干見えていた可能性

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

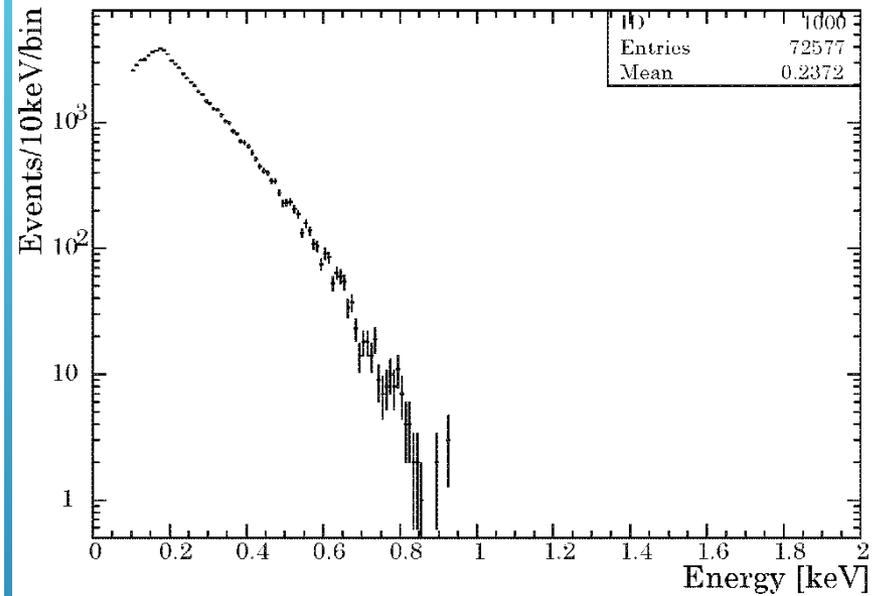
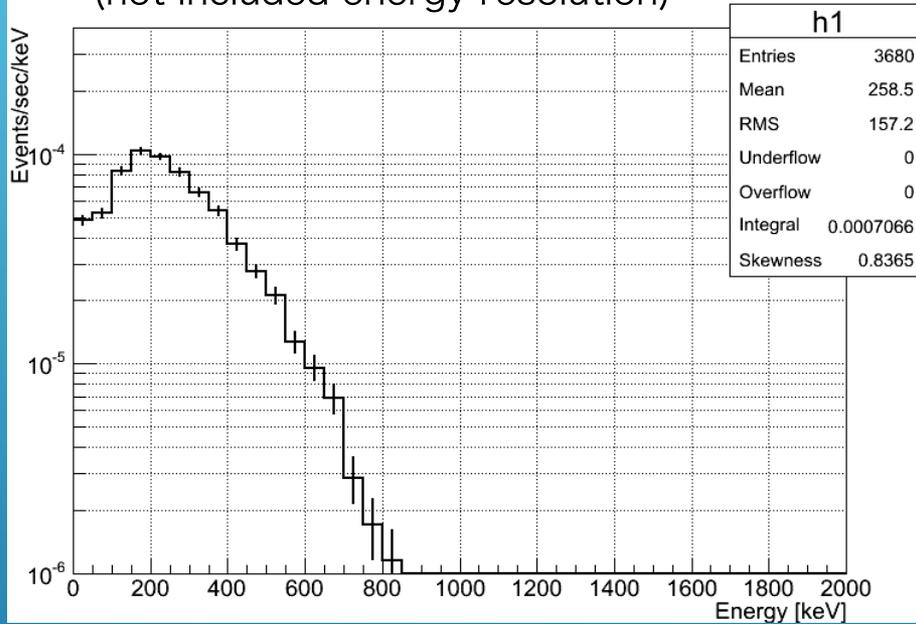
- ・穴を塞ぐ前 182事象/3日間
- ・穴を塞いだ後 100事象/1.68日間
= 179事象/3日間

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

BG MC from ^{210}Bi Brems in Pb Shield
(not included energy resolution)

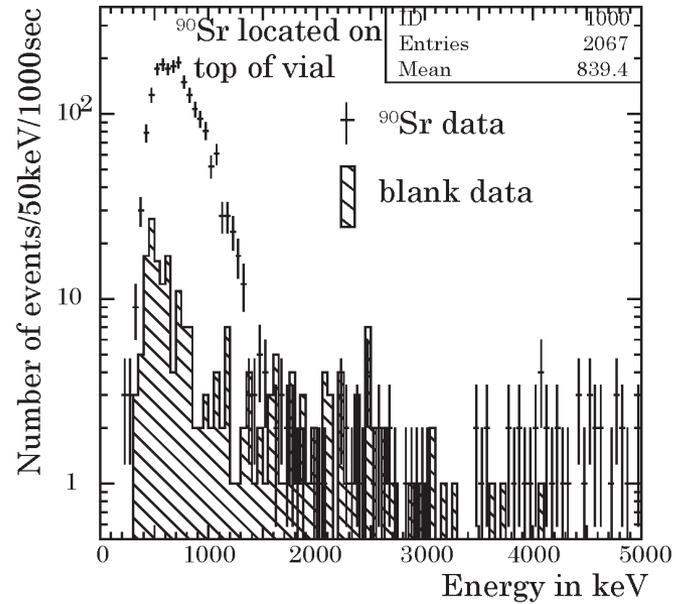
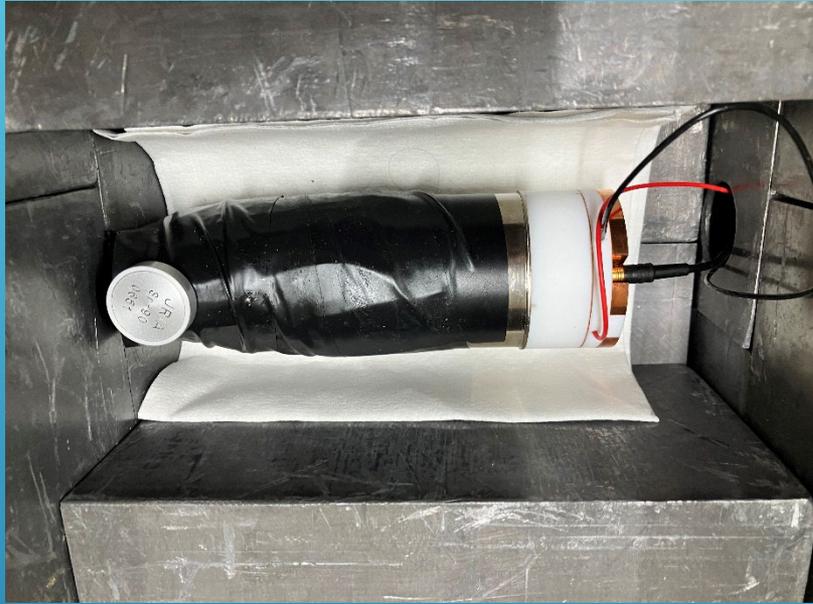


市村さんが振った鉛シールド内の ^{210}Bi のベータ事象からの制動放射事象のエネルギー分布と、EGS5で同様に振った事象のエネルギー分布

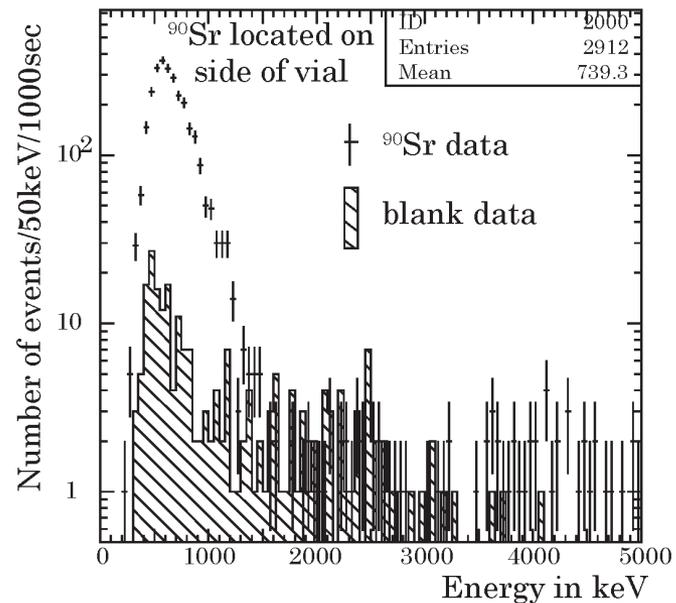
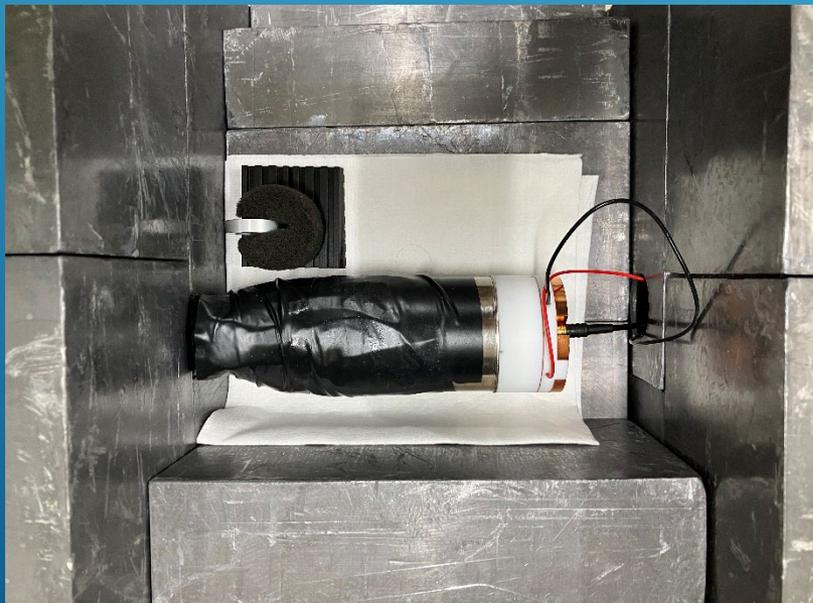
よく似ているが、EGS5の方が少しエネルギーが低い

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鉛ブロックからの ^{210}Bi のベータ線が見える可能性

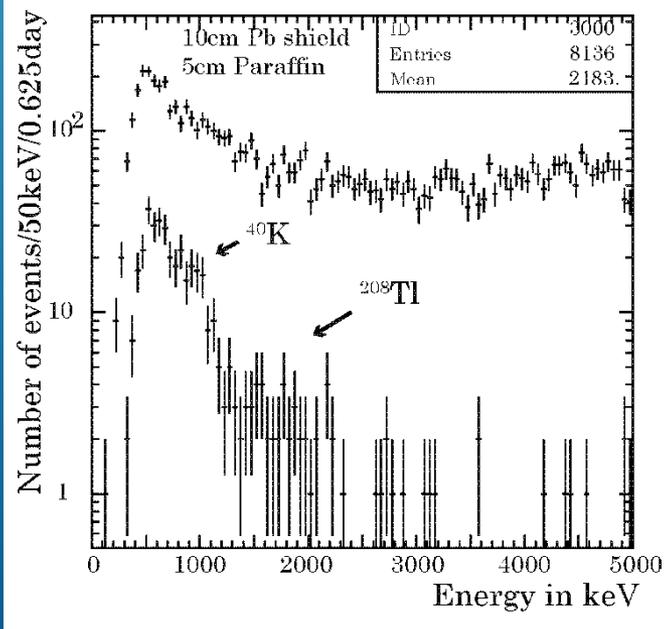
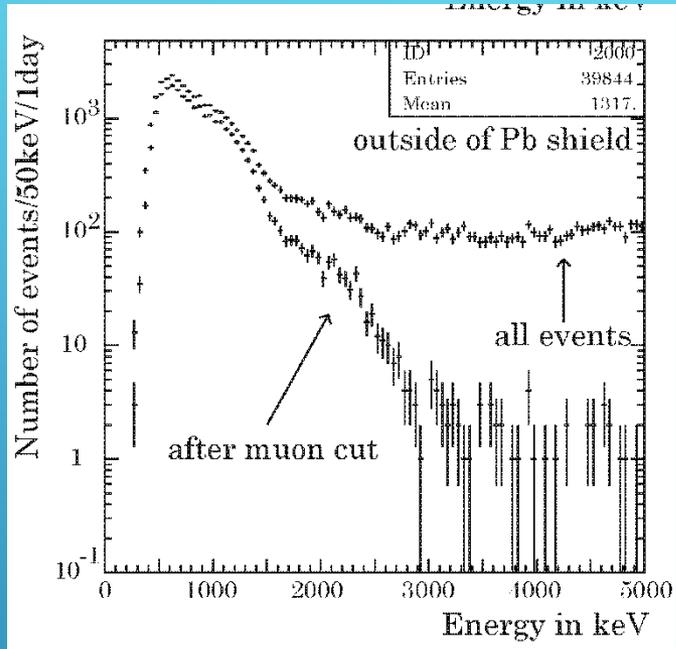
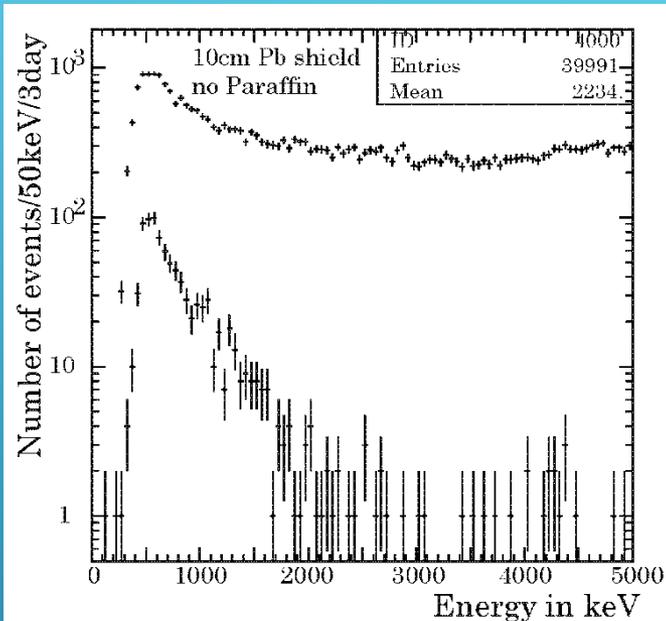


^{90}Sr のベータ線 ($E_{\text{max}} = 2.28\text{MeV}$)が見える。



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

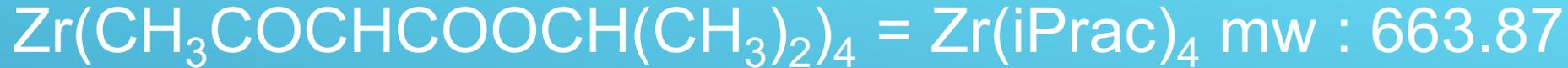


パラフィンで囲んだ場合の600keVの事象数が0.625時間で約30に対し、鉛だけの場合は72時間で約100なので、内部事象なら20事象存在するはずだが、予測される数しか観測されていない。

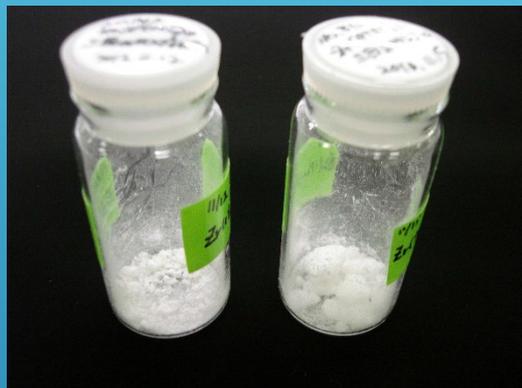
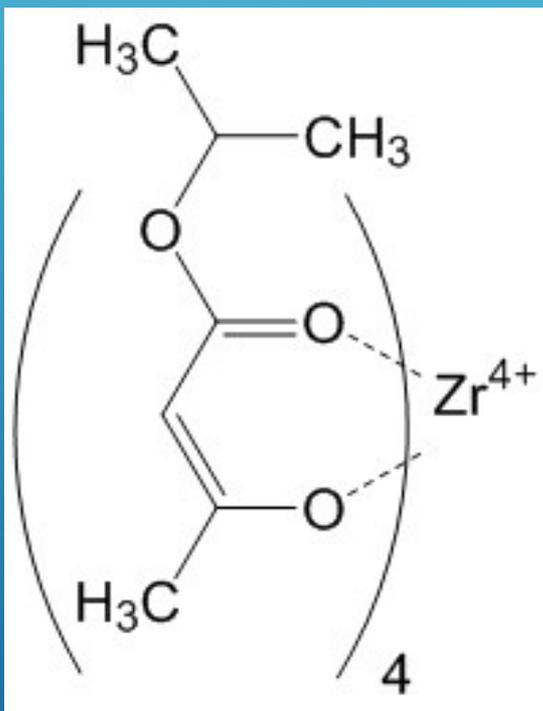
低エネルギー側は鉛からのベータか？

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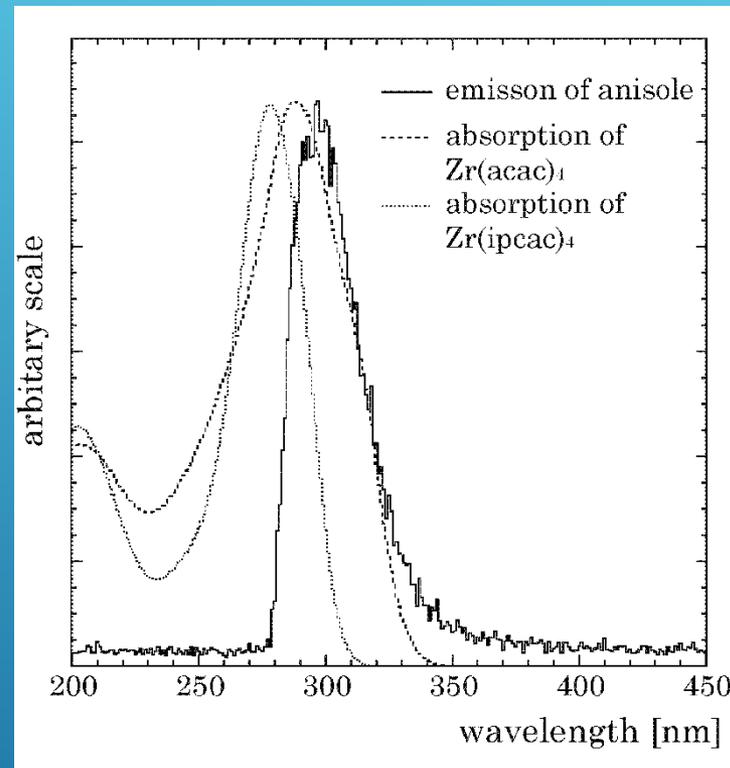
Zirconium β -keto ester complex



Tetrakis(isopropyl acetoacetate) Zirconium



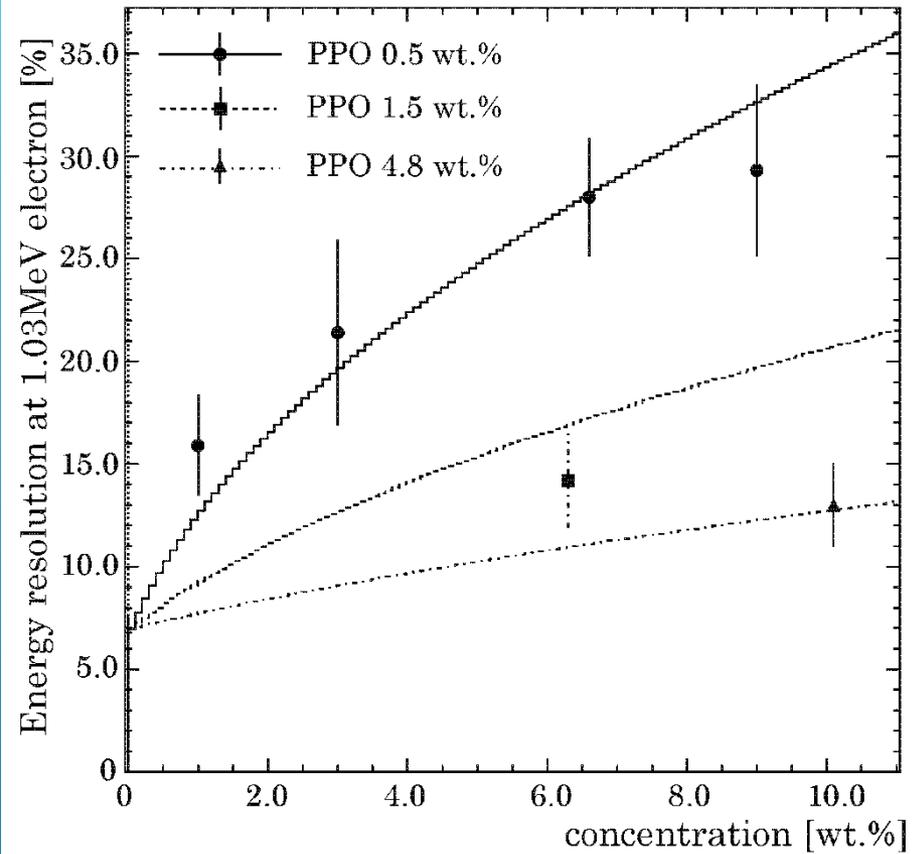
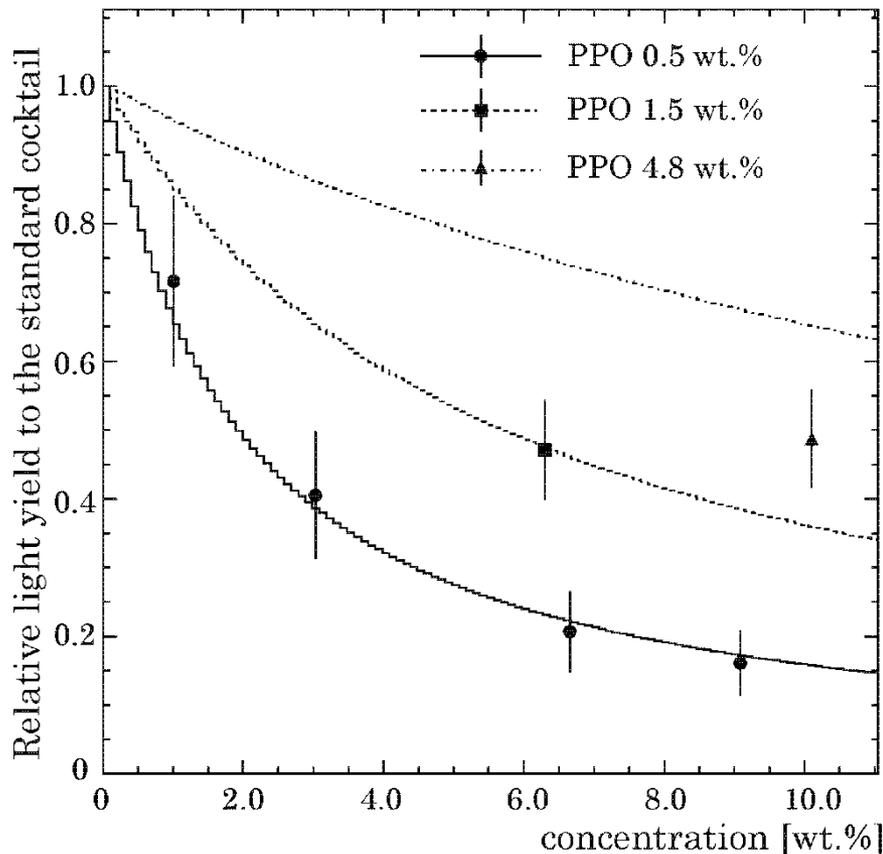
Synthesized by
Prof. T.Gunji
(Tokyo
University of
Science)



No commercial product

Shorter wavelength
for absorbance

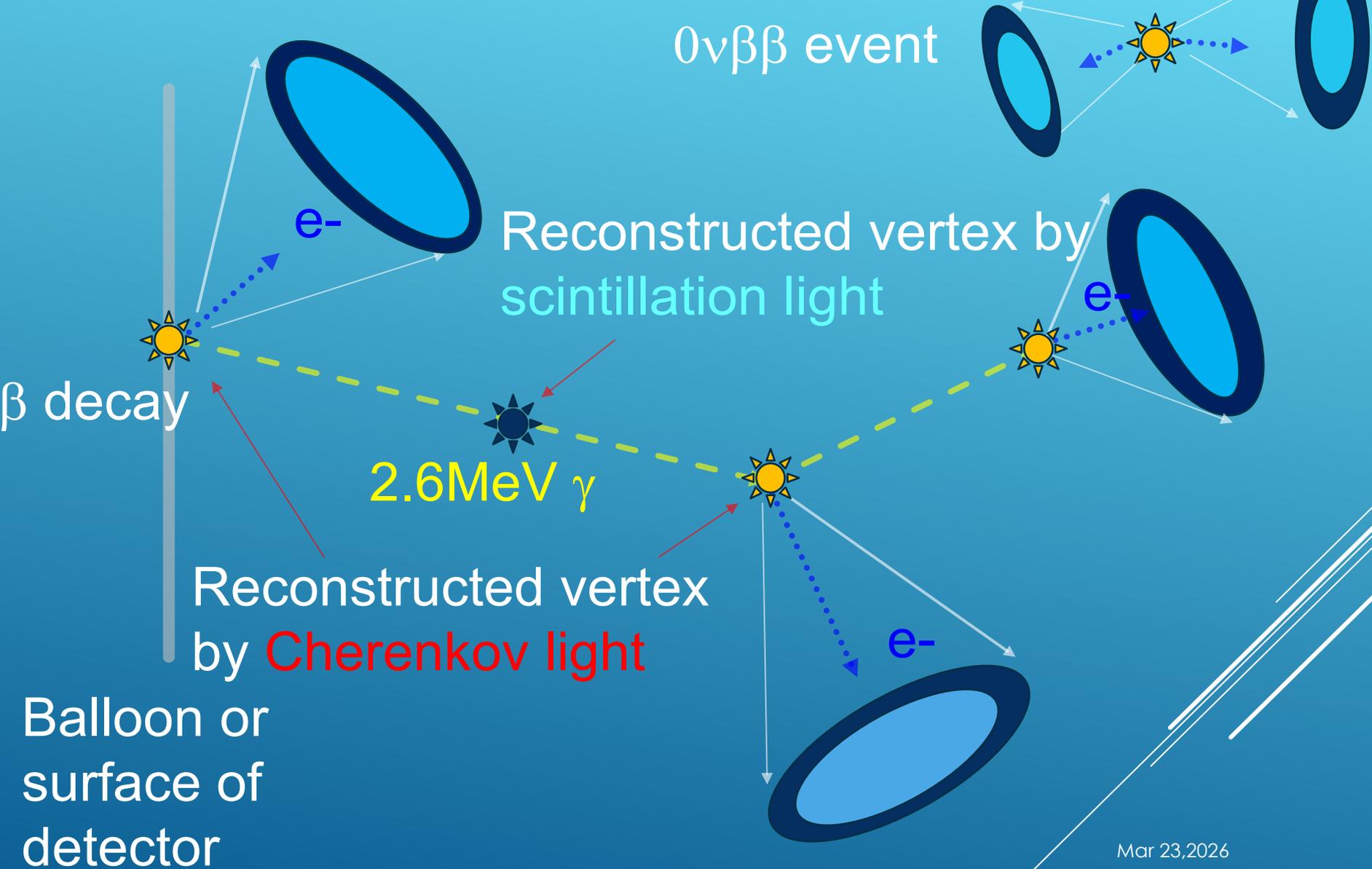
Improve of light yield and resolution



Zr(iPrac)₄ absorbs the scintillation photon and makes the energy resolution worth, however PPO could improve them.

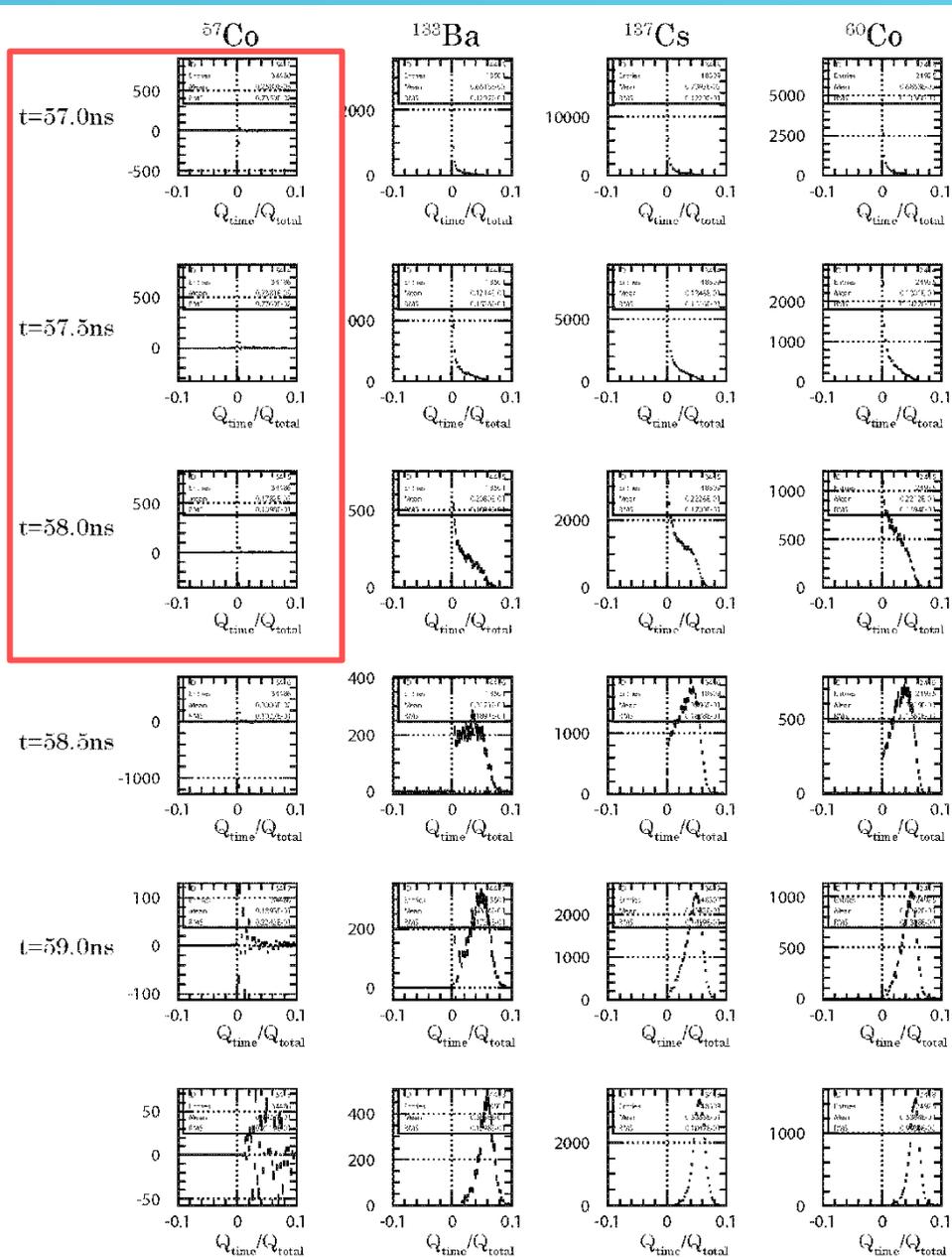
$$\begin{aligned}
 & 13.0 \pm 2.0\% \\
 & \frac{\sqrt{40\%/9\% \cdot 0.72 \cdot 3.35\text{MeV}/1.03\text{MeV}}}{} \\
 & = 4.1 \pm 0.6\% \text{ at } 3.35\text{MeV}
 \end{aligned}$$

How to distinguish ^{208}Tl and DBD



Balloon or surface of detector

ZICOS LSを用いたガンマ線事象の立ち上がり



- $t=57\text{ns}\sim 58\text{ns}$ 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
- $t>58.5\text{ns}$ の波形はエネルギーによらず一定

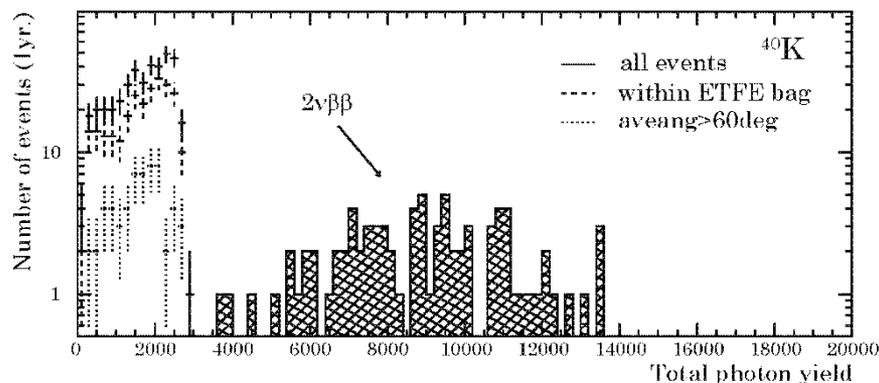
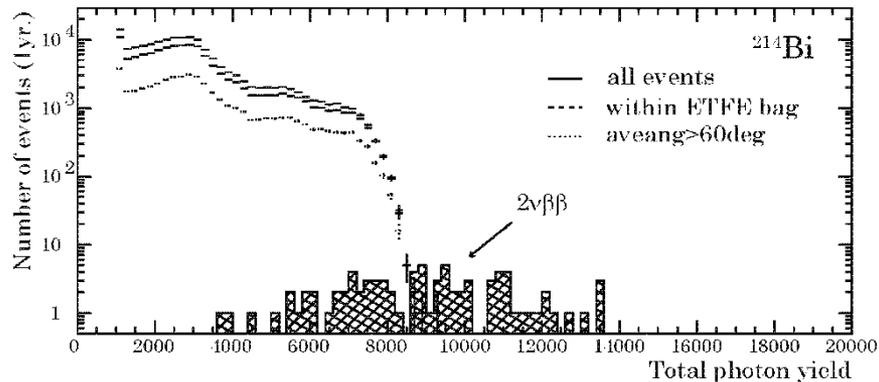
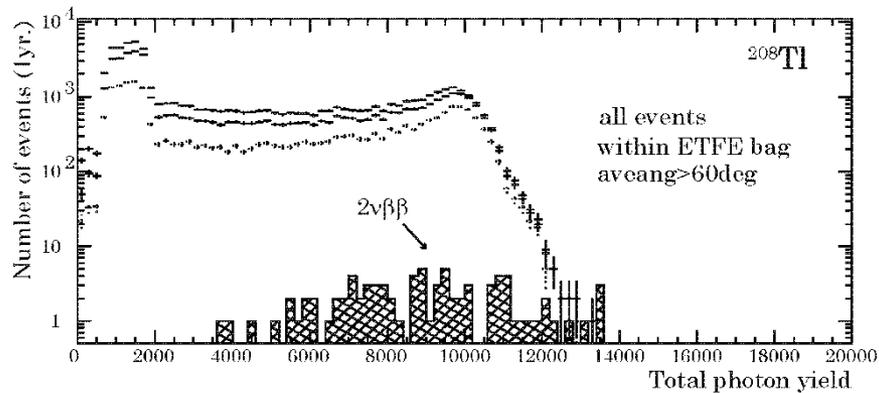
$t=57\text{ns}\sim 58\text{ns}$ の波形はチェレンコフ光が優勢と考えられる。



^{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

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BG simulation in worst case



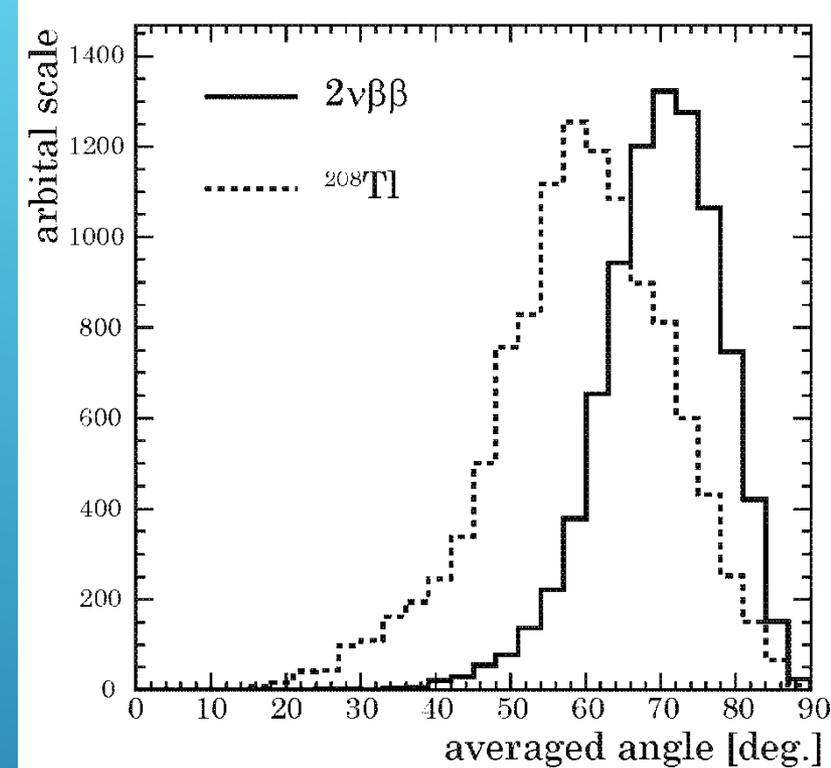
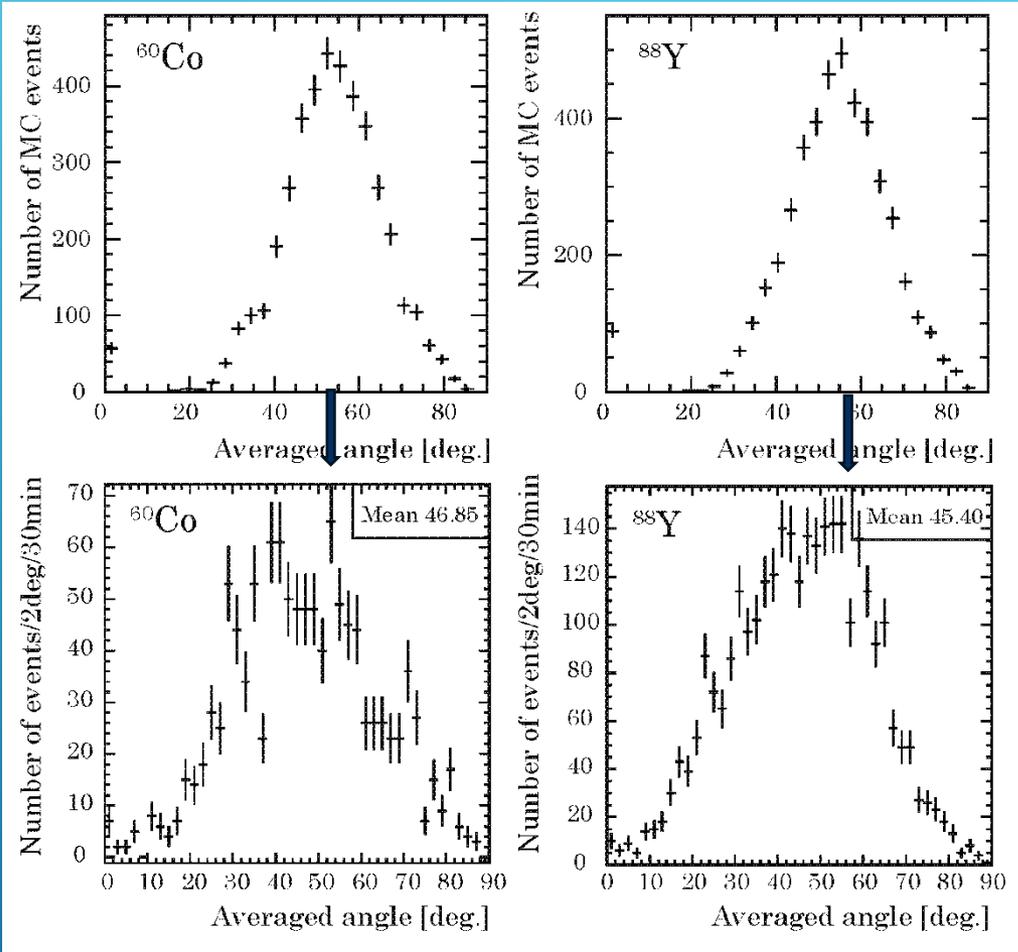
Assuming BGs from flask

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

Need actual measurement for ^{96}Zr $2\nu\beta\beta$

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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.