

ジルコニウム96を用いた2つのニュートリノを 放出する二重ベータ崩壊事象の観測

2025年度宇宙線研究所共同利用研究

成果発表会

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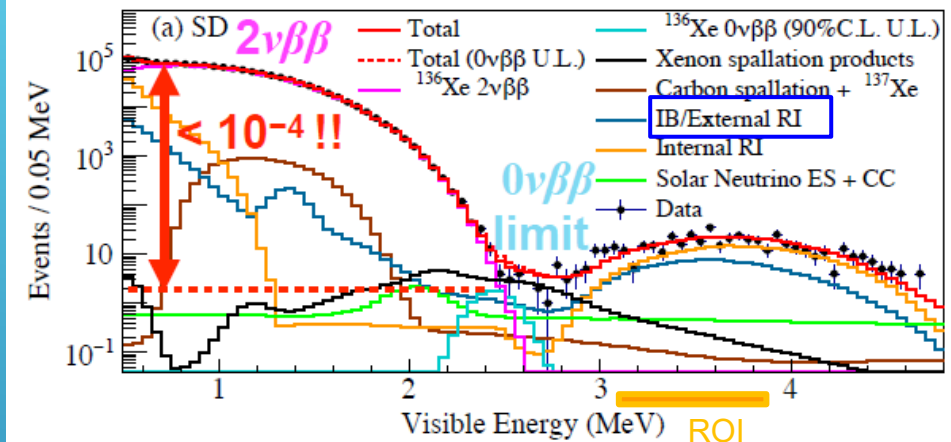
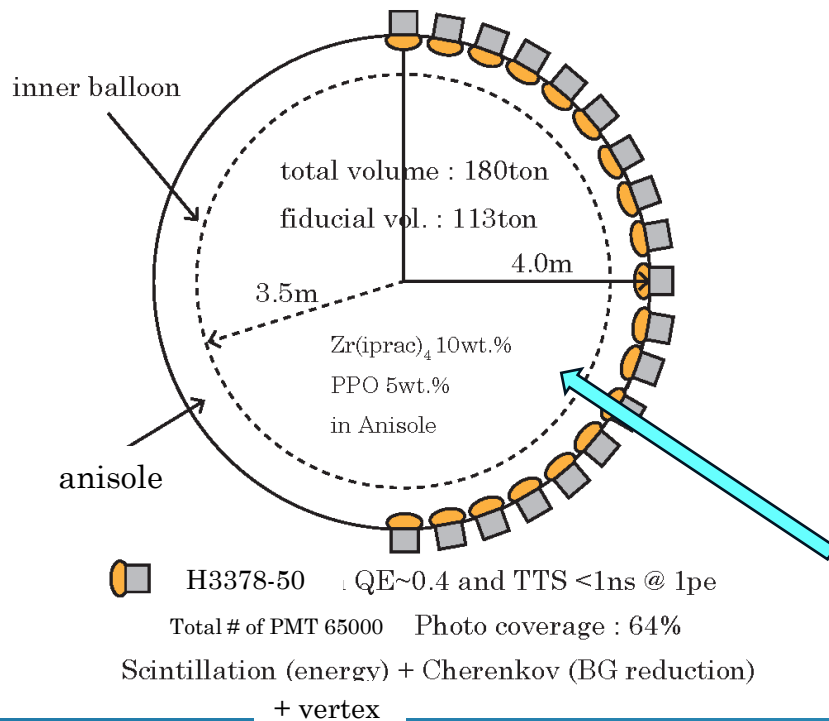
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ZICOS experiment for ^{96}Zr $0\nu\beta\beta$ observation

Conceptual design of ZICOS detector



I. Shimizu, Plenary talk at Neutrino2024 conference.

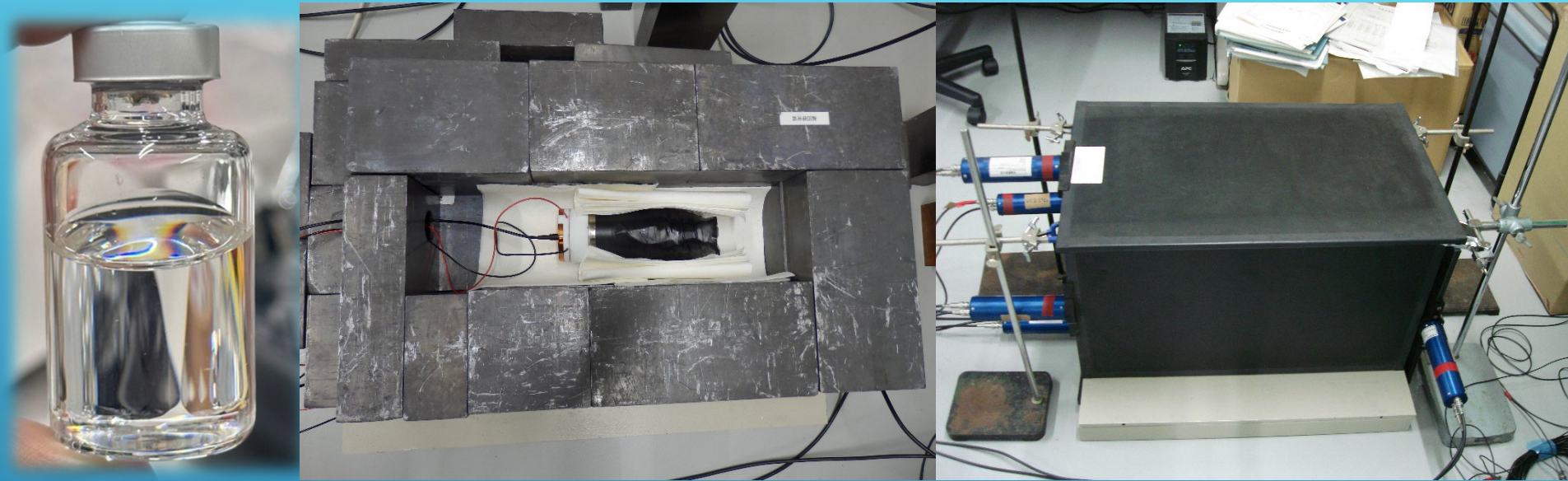
Liquid scintillator loaded ^{96}Zr

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

$$^{96}\text{Zr} : 45 \text{ kg (nat.)} \rightarrow 865 \text{ kg (50 \% enrich)} \rightarrow 1/20 \text{ BG}$$

$$T_{1/2}^{0\nu} > 4 \times 10^{25} \text{ yrs} \rightarrow 2 \times 10^{26} \text{ yrs} \rightarrow \sim 1 \times 10^{27} \text{ yrs}$$

BG measurement using 20mL quartz vial

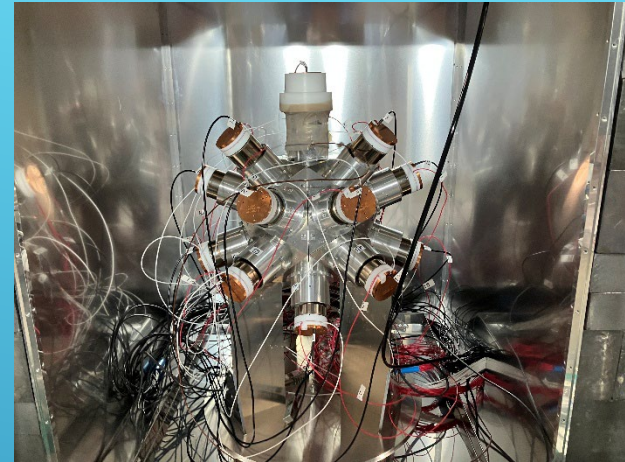
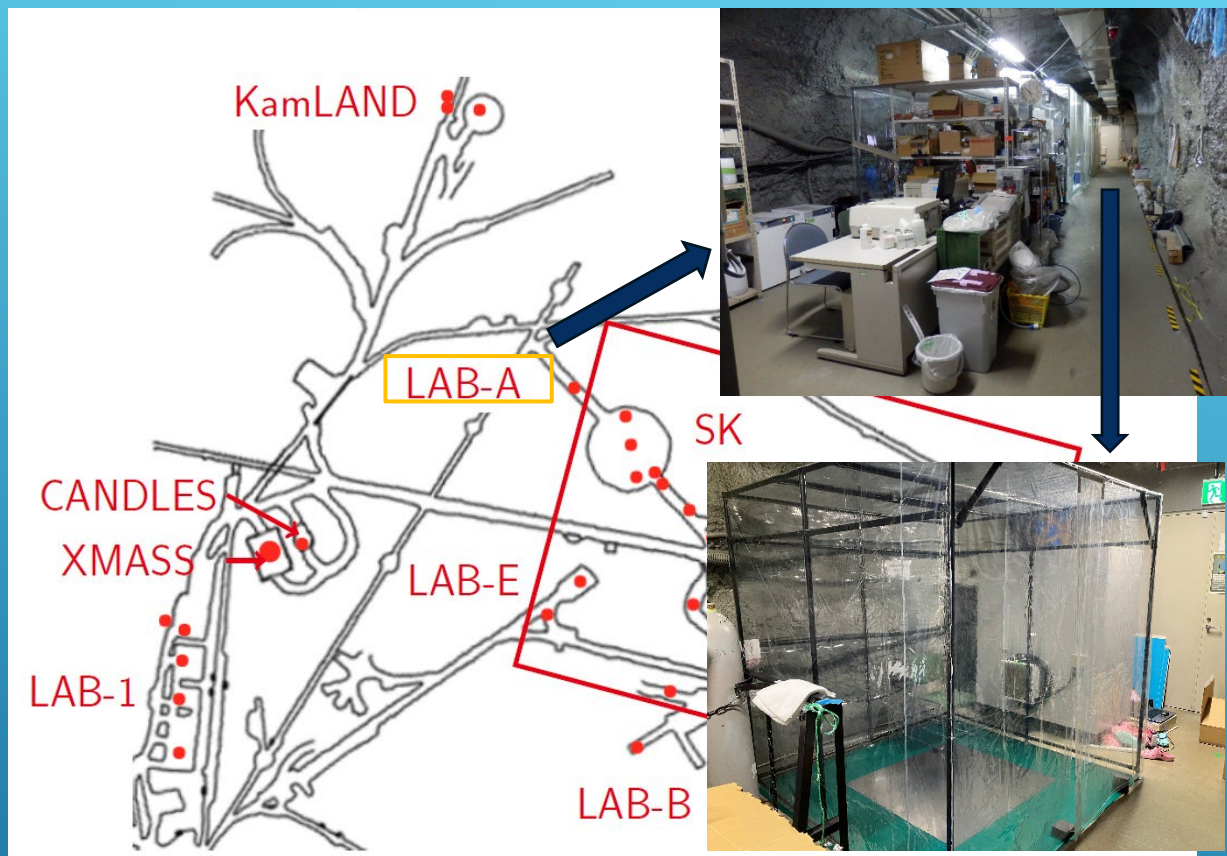


- Difficult to measure BG events for 2ν -ZICOS detector inside of Pb shield due to limit of number of Pb block.
- 17.5mL ZICOS LS filled in 20mL quartz vial.
- The vial contacted with XMASS PMT R10789 covered by Myra film.
- 10cm thickness of Pb radiation shield was prepared.
- In order to veto cosmic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

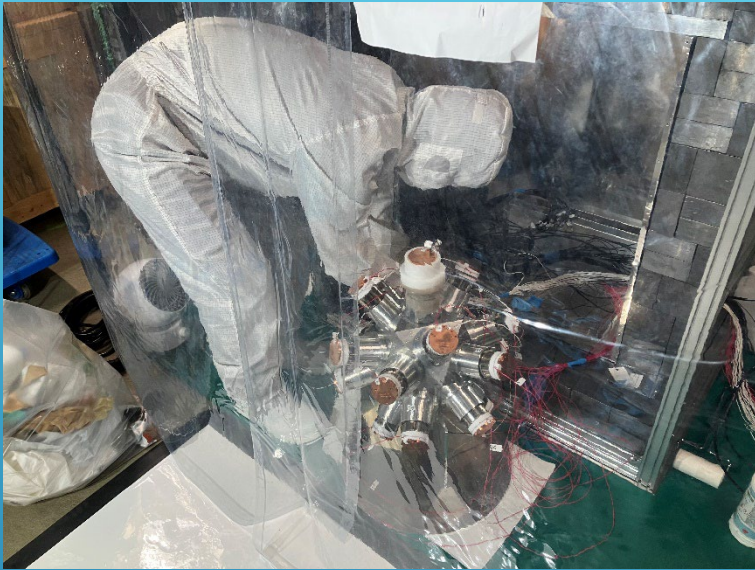
2ν-ZICOS for measurement of ^{96}Zr $2\nu\beta\beta$ events



- 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 etc were installed before starting experiment.

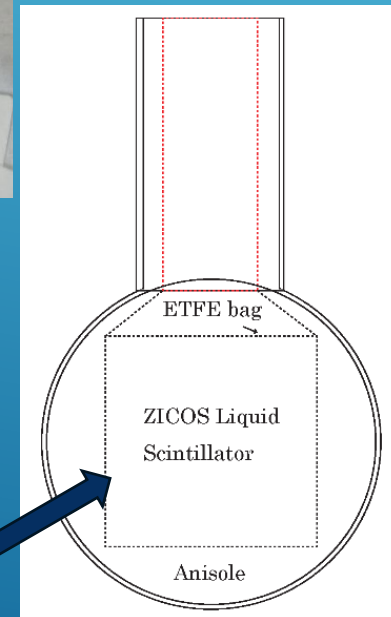
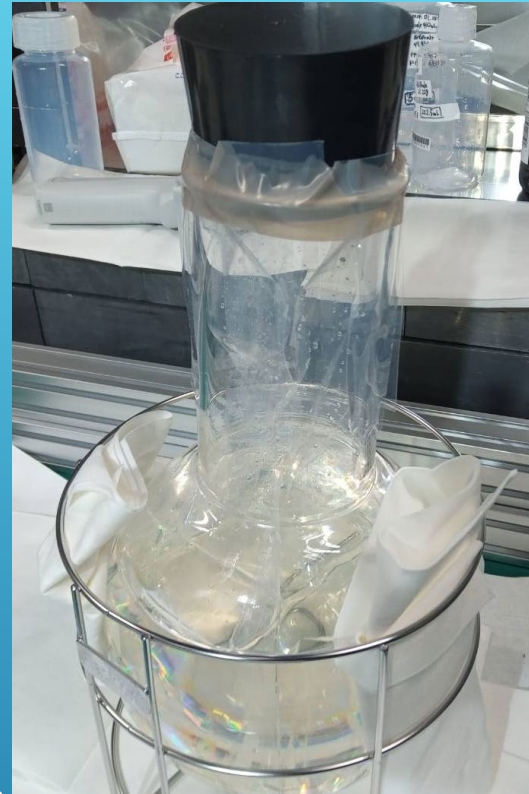
The first data taking implemented over 50 hours from November 22nd to 25th.

2v-ZICOS detector

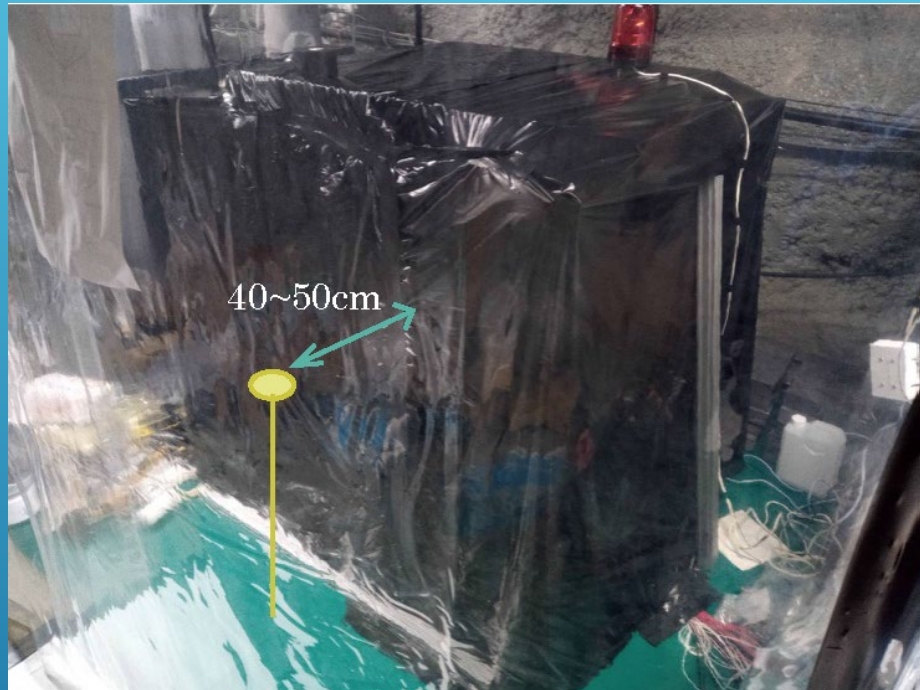


- 20 2" Ultra Low BG PMT Hamamatsu R10789.
- 16 cm diameter round bottom flask using pure Quartz.
- 0.724L of ZICOS LS loaded 69g of $\text{Zr}(\text{iPrac})_4$ including ^{96}Zr 0.27g.

9cm cubic ETFE bag for reducing external BG events

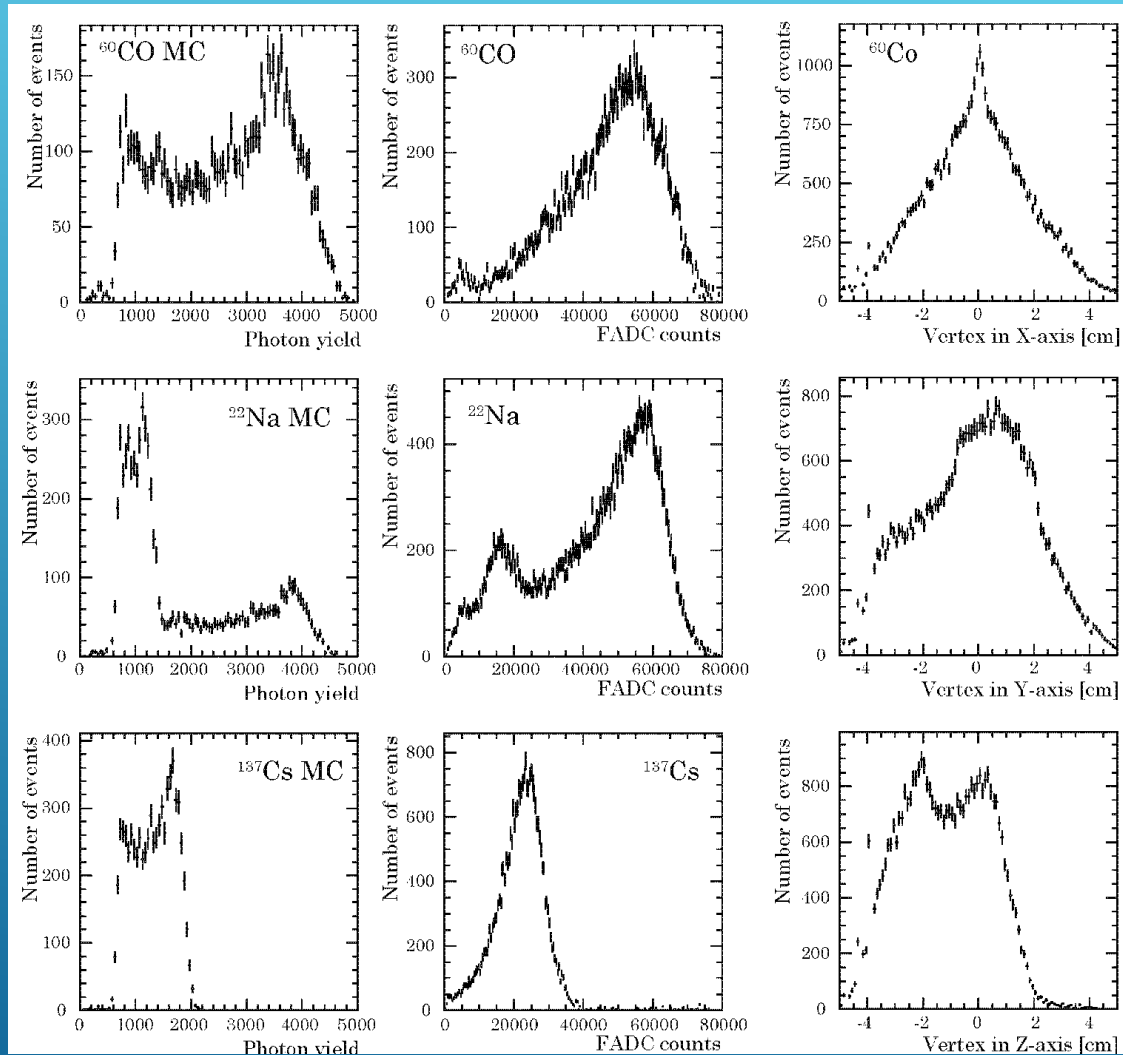


Energy calibration and DAQ for 2v-ZICOS detector



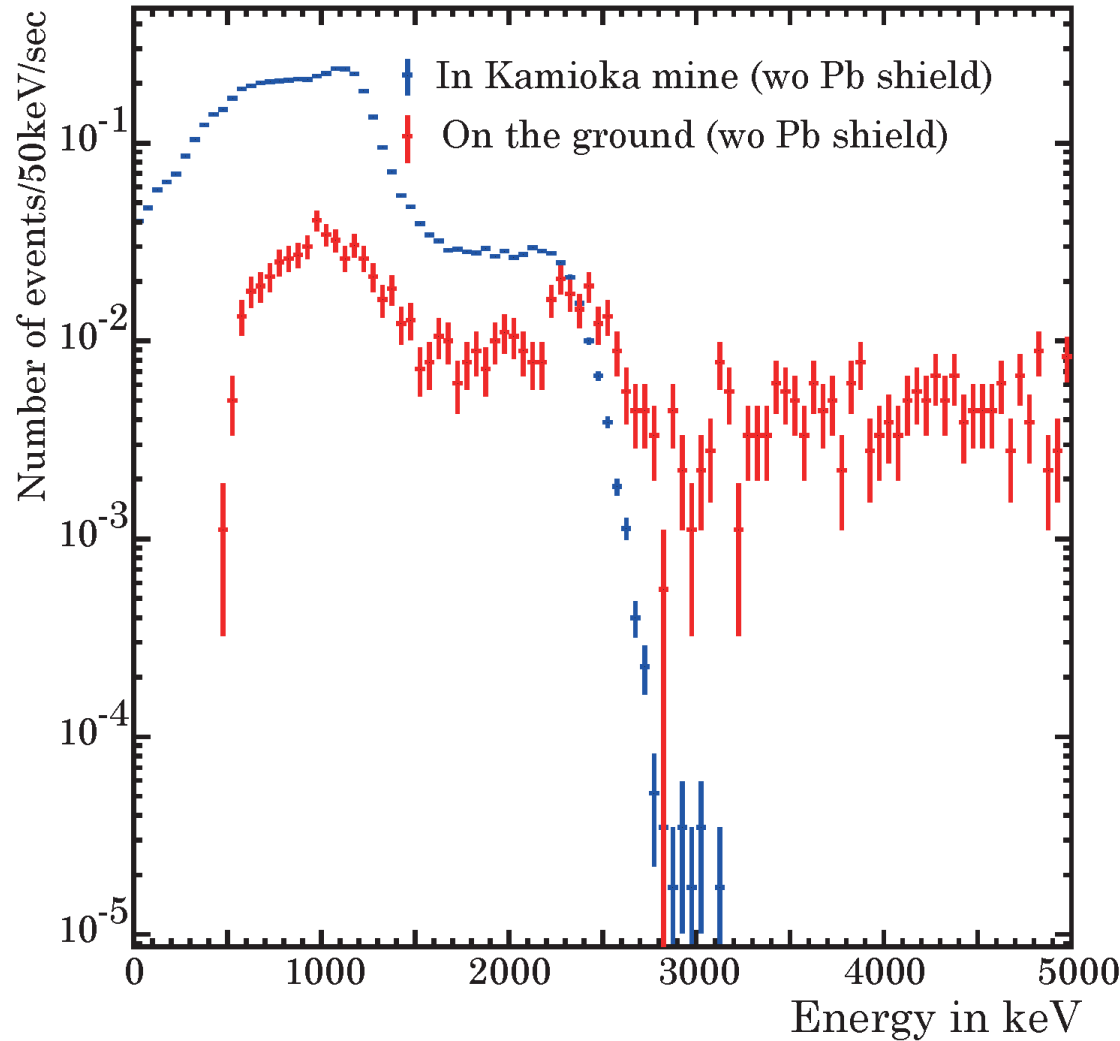
- Energy calibration was done by gamma sources with ^{137}Cs , ^{60}Co and ^{22}Na .
- Those sources were located at front of Al wall.
- All data were recorded by V1742 digitizer.

Energy calibration of 2ν-ZICOS detector



- Energy spectra seem to be reproduced by MC without trigger threshold.
- Energy scale looks have a good linearity.
- Reconstructed vertices look as expected.

Background events observed in the mine



- Only few muons were observed even in half day measurement .
- BG events from ^{40}K and ^{208}Tl were observed about over 10 times larger than the measurement on the ground, even only front shield was opened.

Present status and future plan

- Data analysis of background data measured in Pb radiation shield is now on going.
- First result will be reported in this Spring meeting.
- Detector upgrade in next fiscal year as planned.
 - Material of PMT mounting jig and inner shield wall will be replaced by Oxygen free copper.
 - Ultra pure quartz (<0.1 ppb for U/Th) will be used for round bottom flask with 30cm diameter.
 - ^{88}Y gamma source will be prepared in Kamioka mine for the energy calibration.
- Automated safety control is also necessary for long term stable measurement. (to be discussed in Kamioka safety committee)

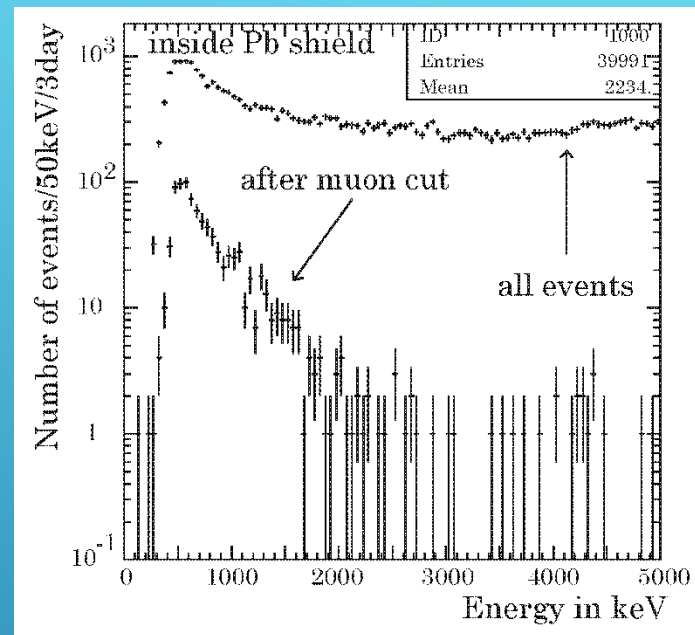
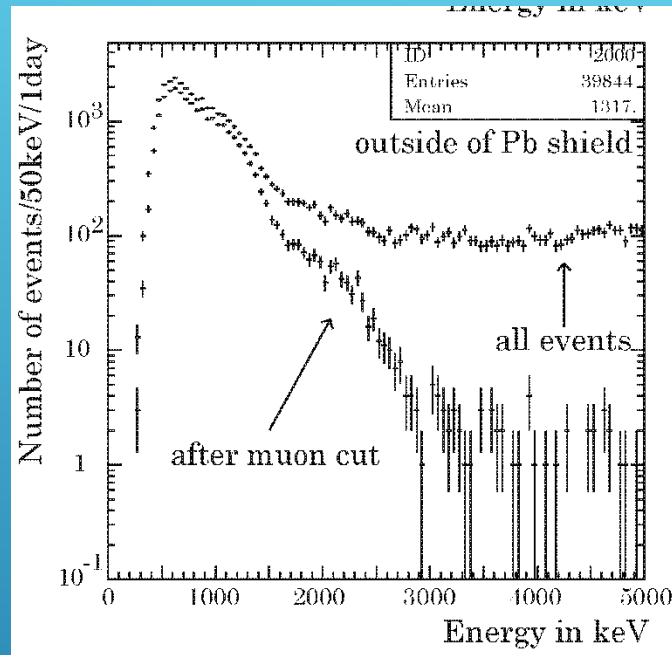
backup

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)

Energy spectra using Quartz vial detector

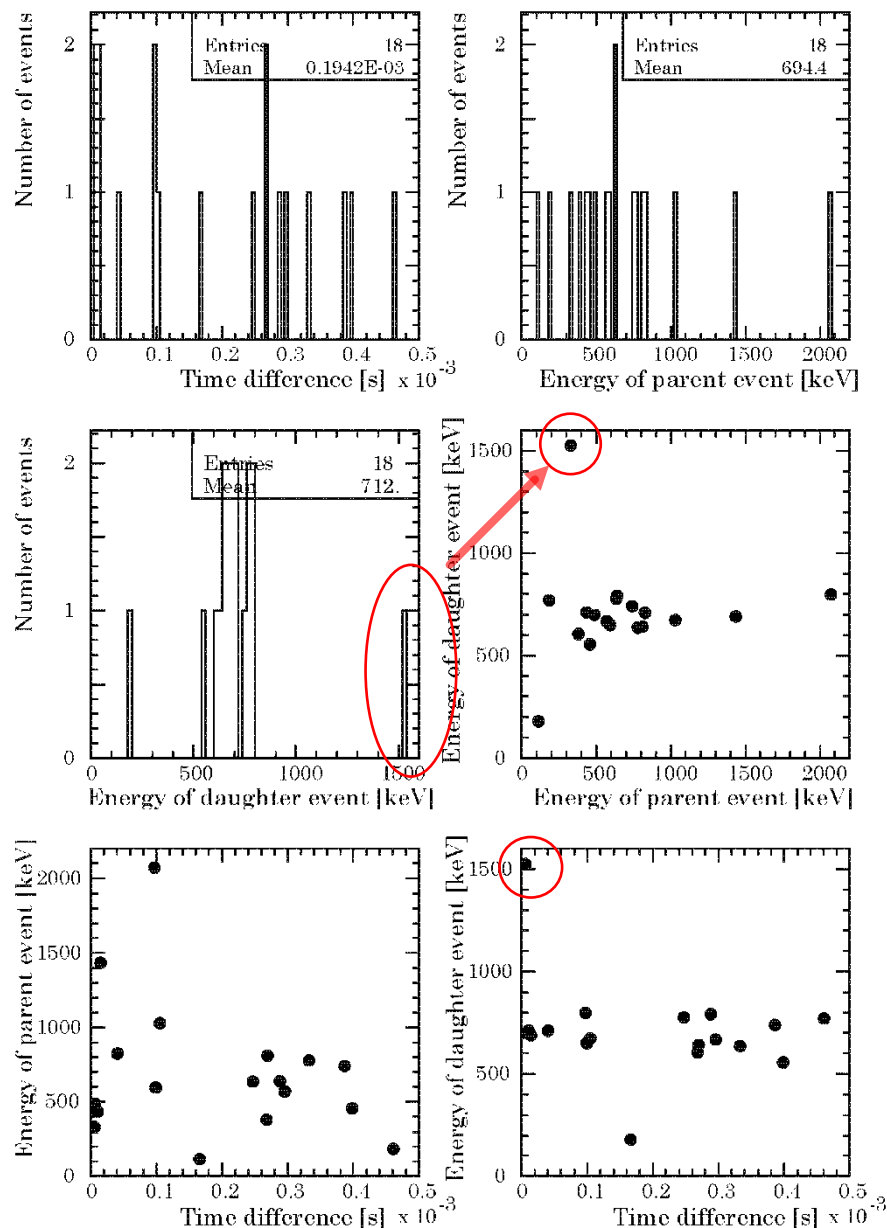


Outside of shield

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

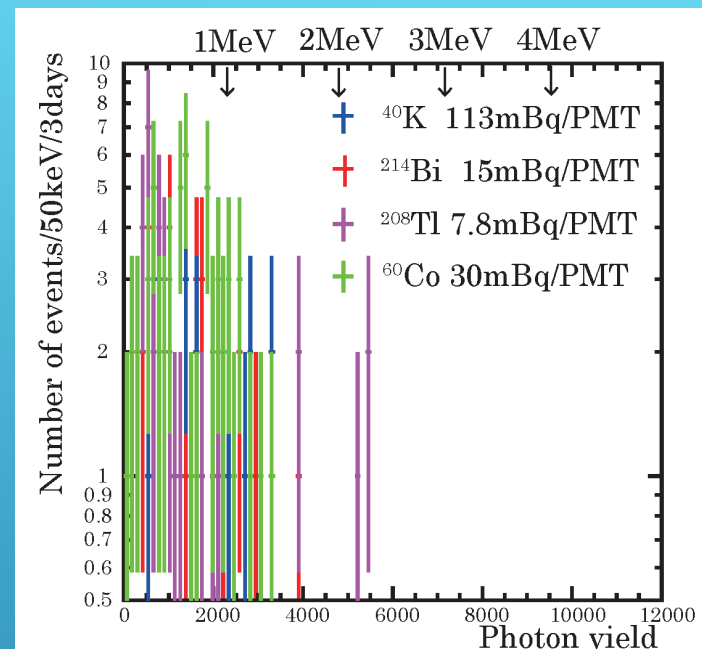
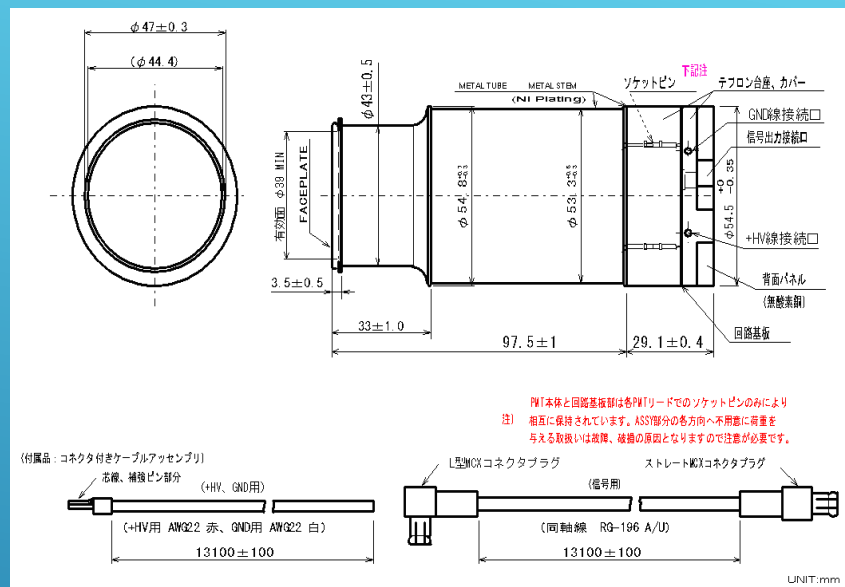
Search for ^{214}Bi - ^{214}Po candidates



- 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)

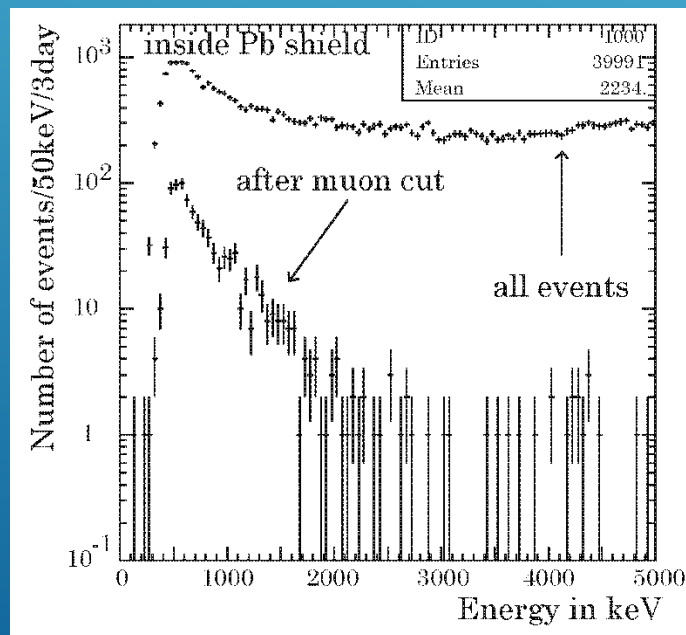
17 events look like ^{214}Bi - ^{214}Po candidates. But they don't contribute residual events.

BG events from PMT R10789

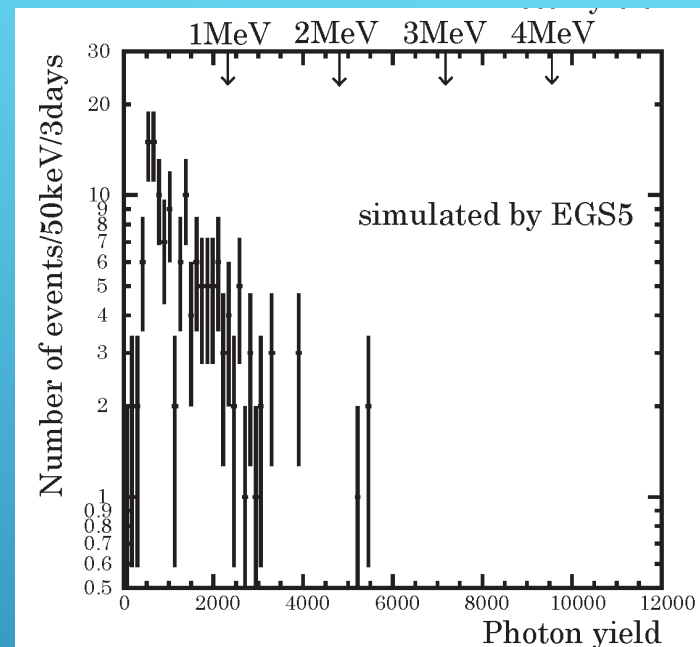
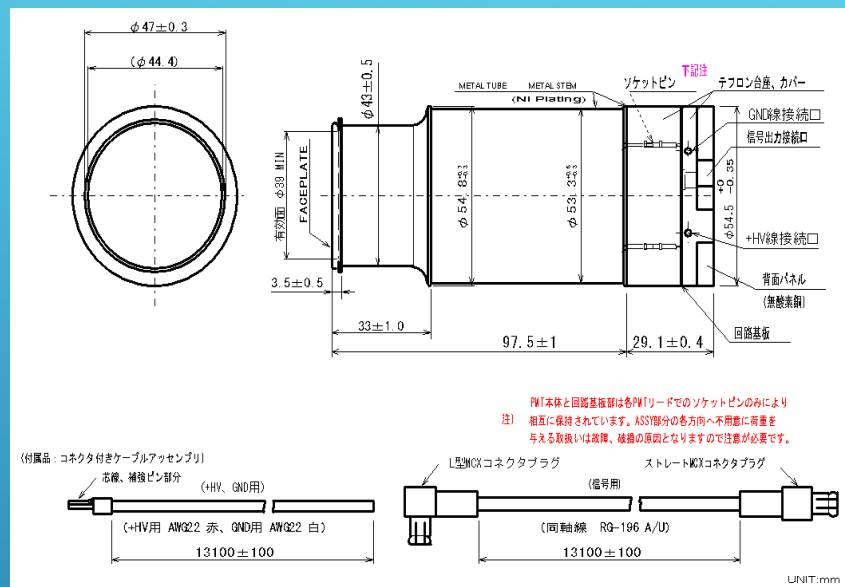


Ref: NIMA 922(2019)171-176

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

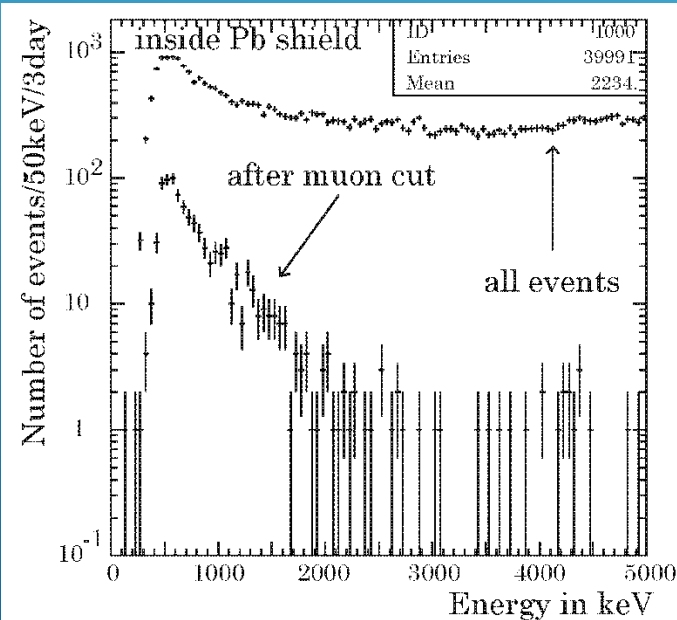
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< 0.78 mBq/PMT of ^{228}Ra

9.1 ± 2.2 mBq/PMT of ^{40}K

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



BG events from Quartz

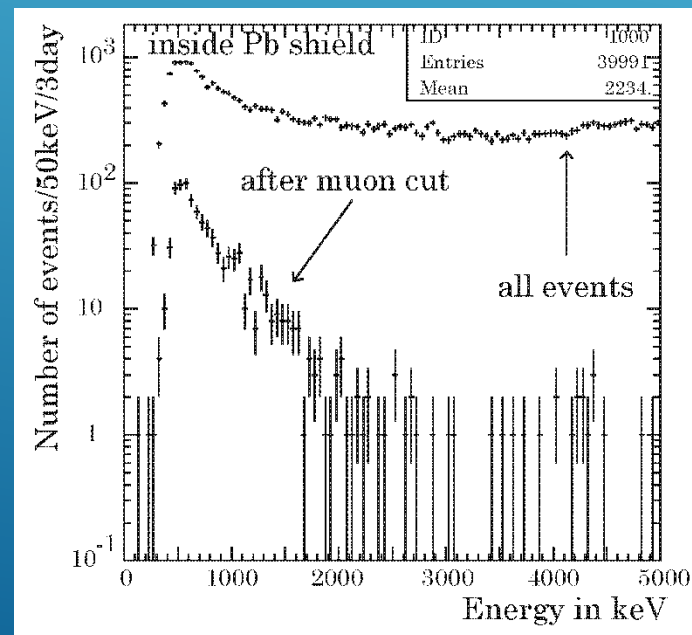
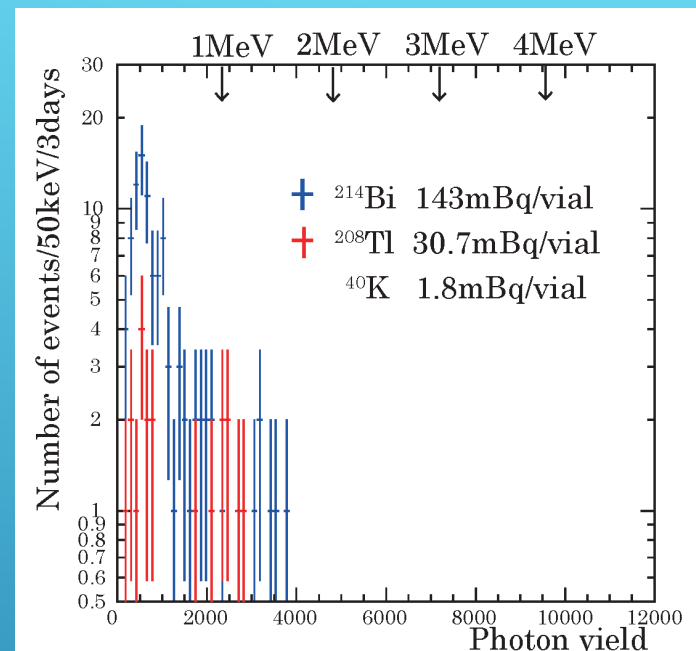


表 分析結果

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



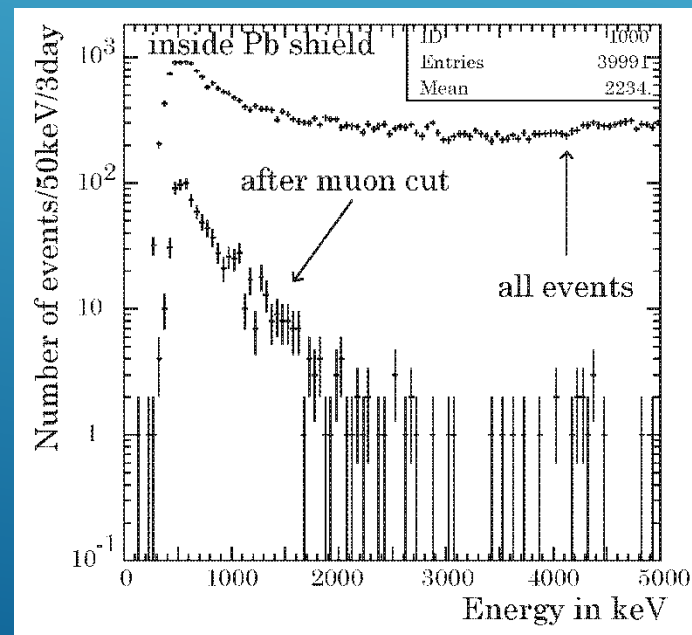
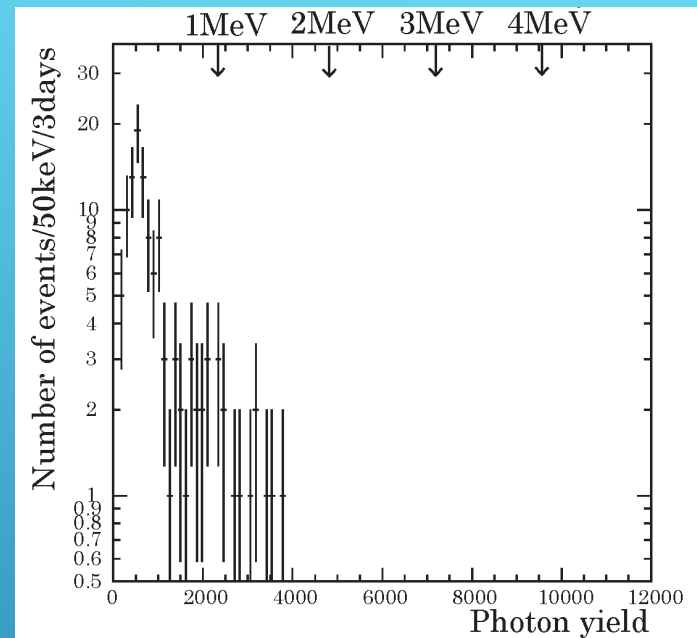
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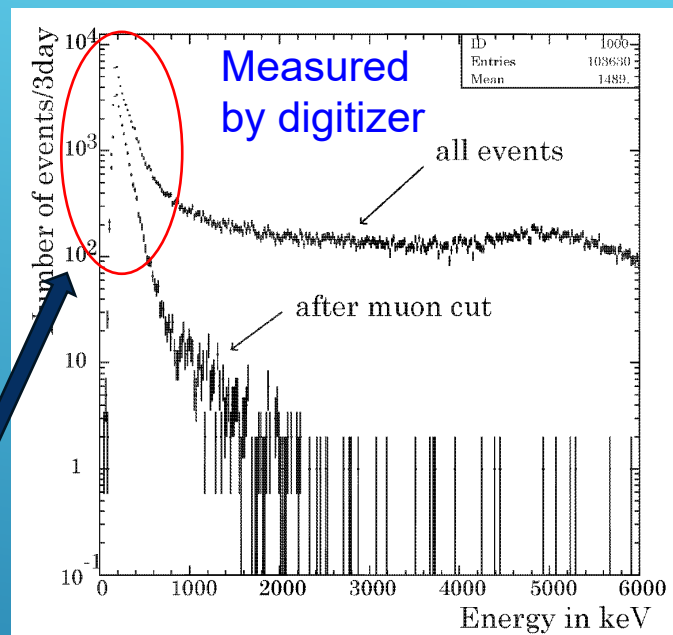
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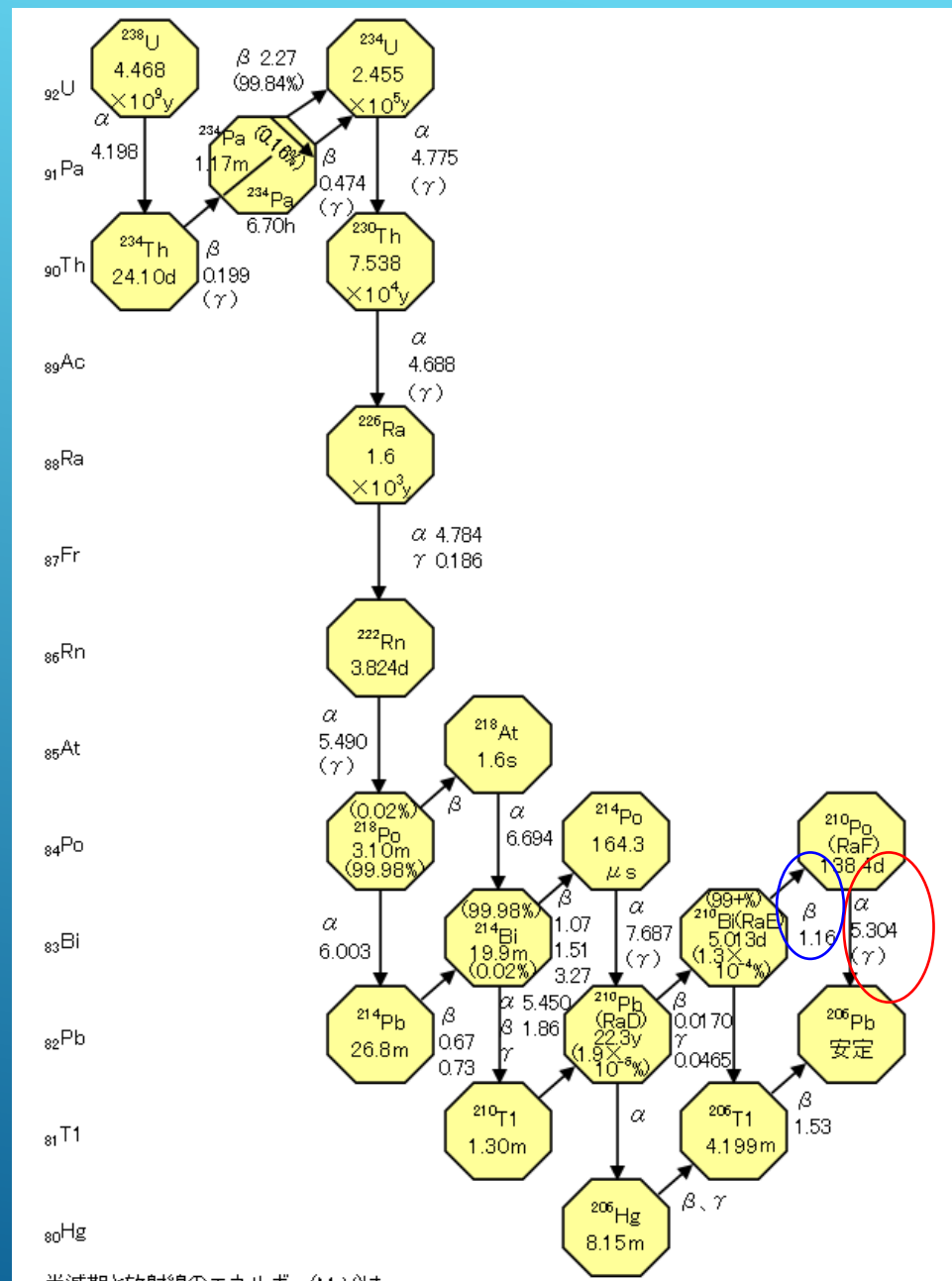
Even more 50 times larger
BG events could not explain
the residual events.



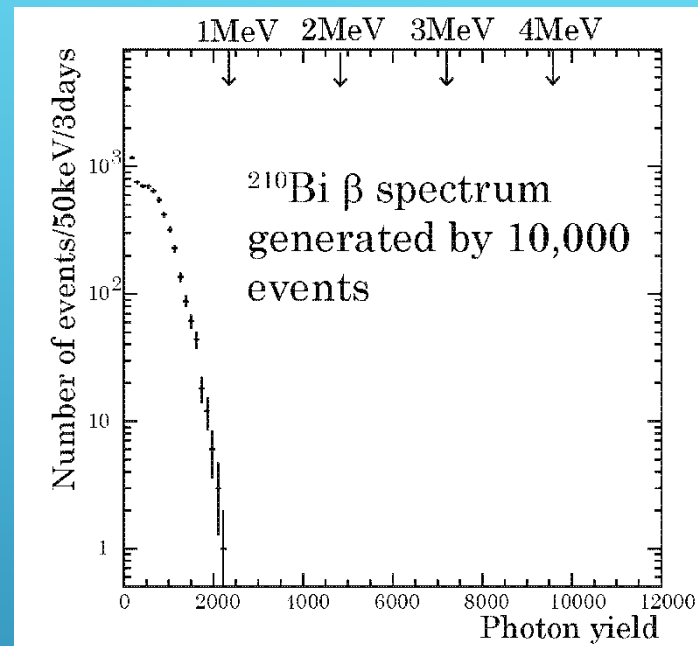
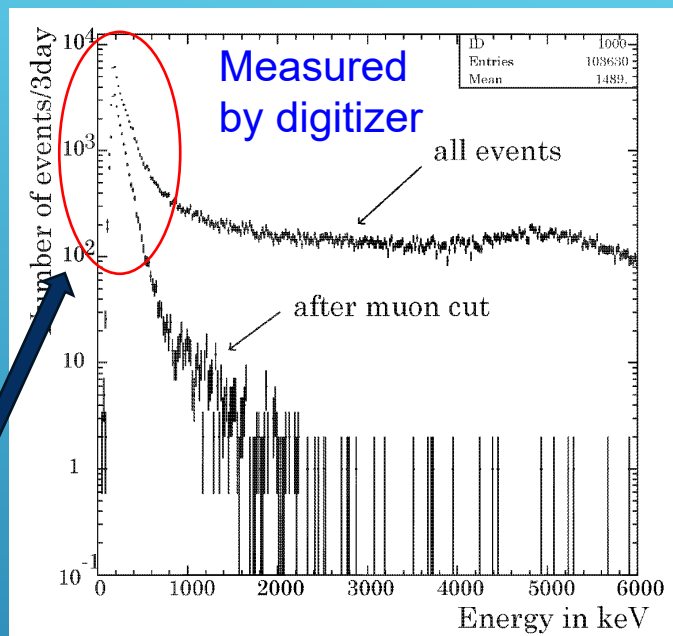
BG events from surface of Quartz



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

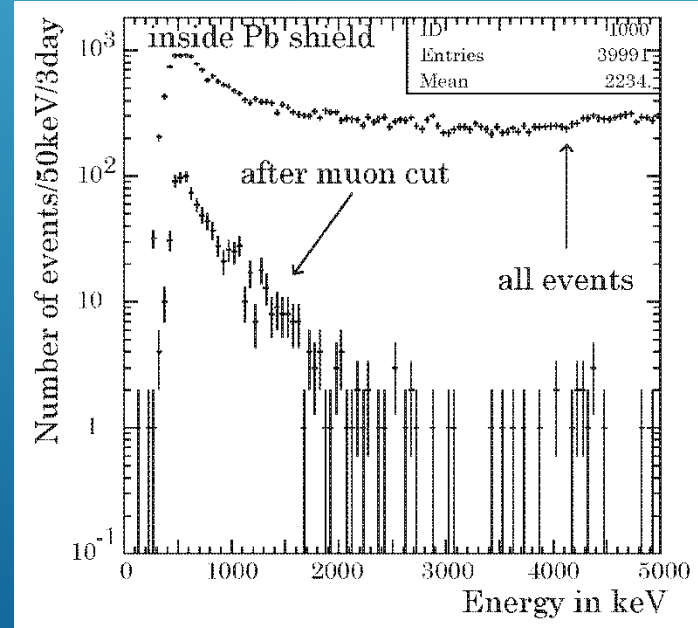


BG events from surface of Quartz

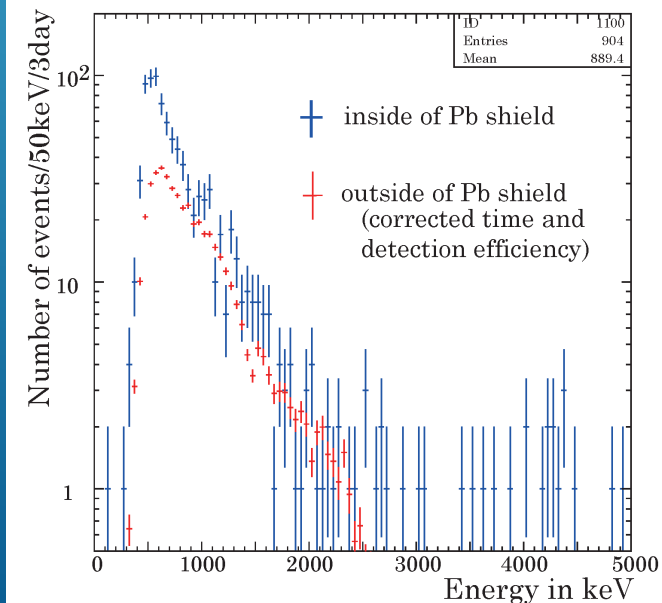
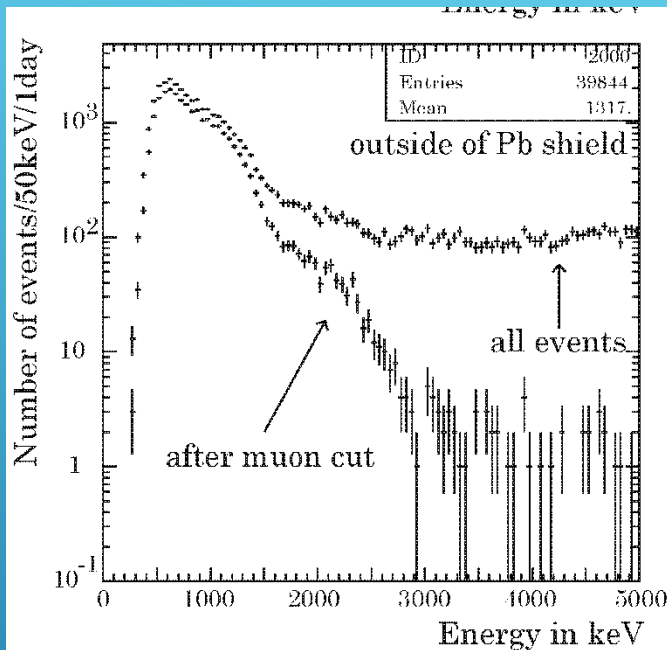


α from ^{210}Po ? if so, β from ^{210}Bi should be observed by same order of events.

Observed rate correspond to $2\text{mBq}/\text{cm}^2$, but XMASS estimated as $1\mu\text{Bq}/\text{cm}^2$.



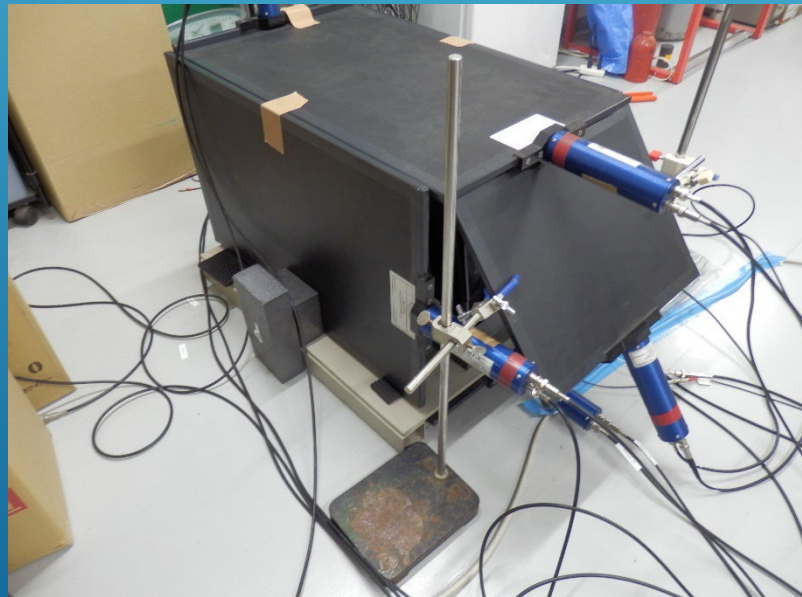
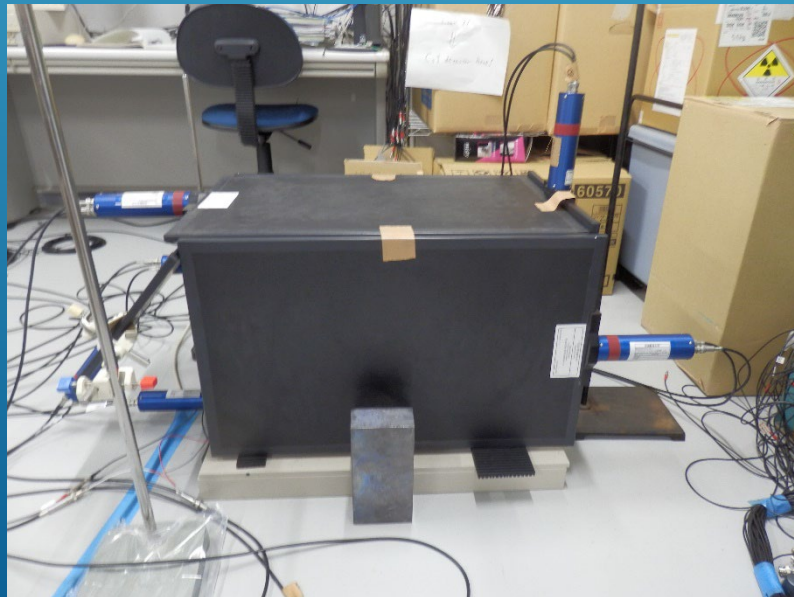
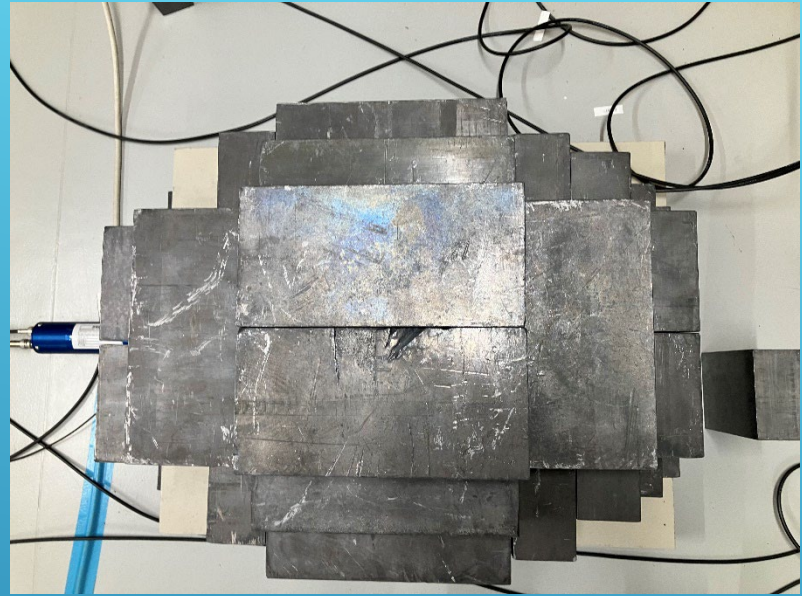
Comparing distributions using env. gammas



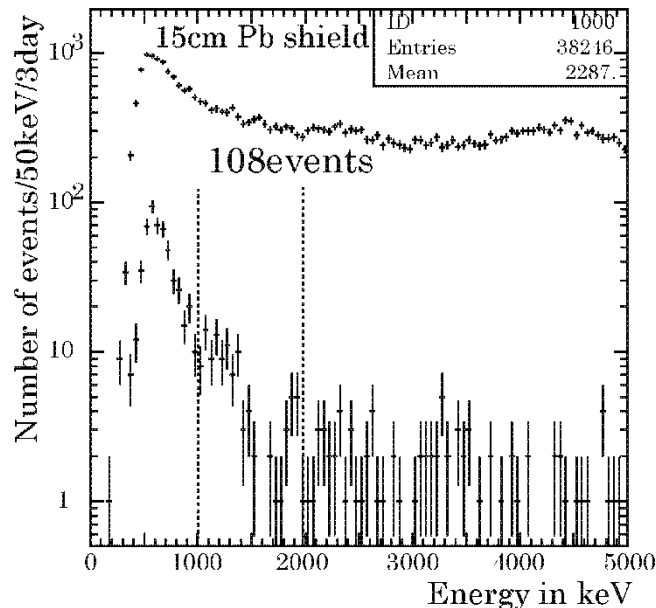
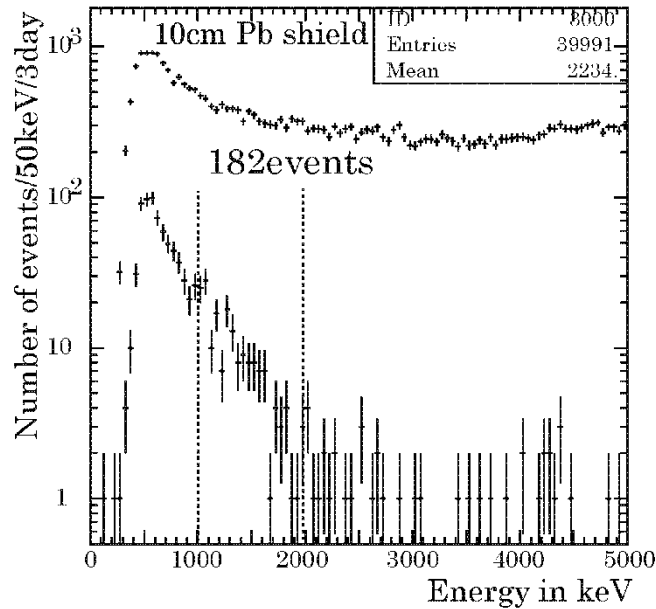
Almost the energy spectrum of residual events is consistent with environmental gammas observed at outside of shield.

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



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

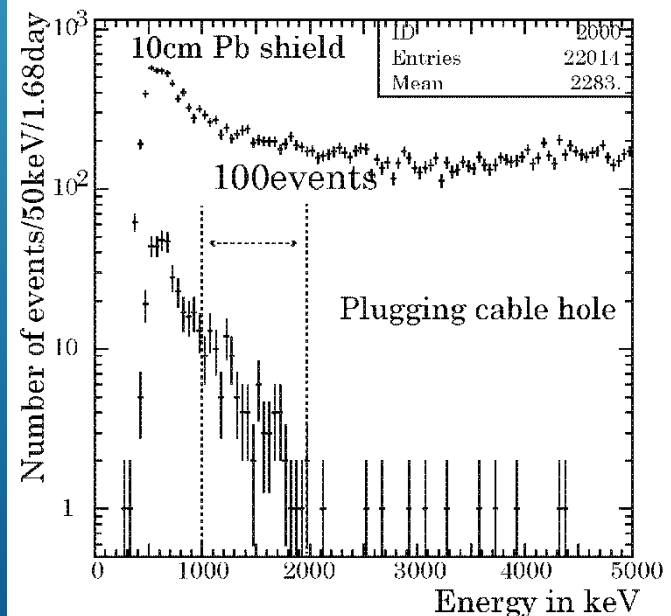
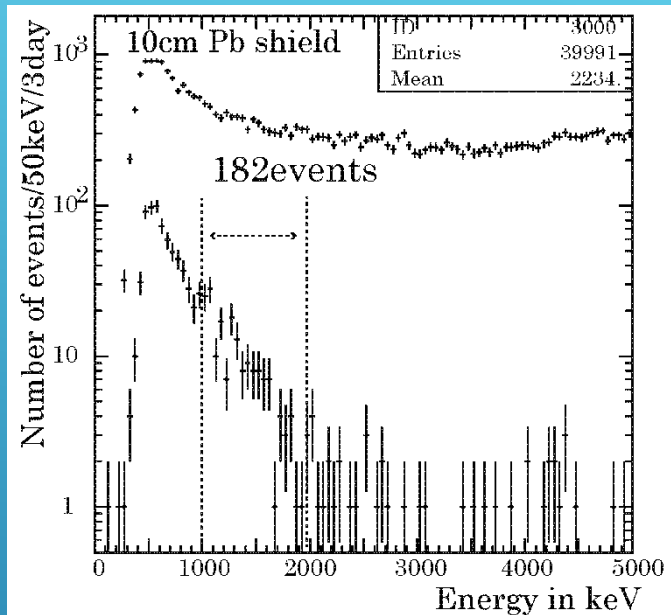
結論： 外部ガンマ線が若干見えていた可能性

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

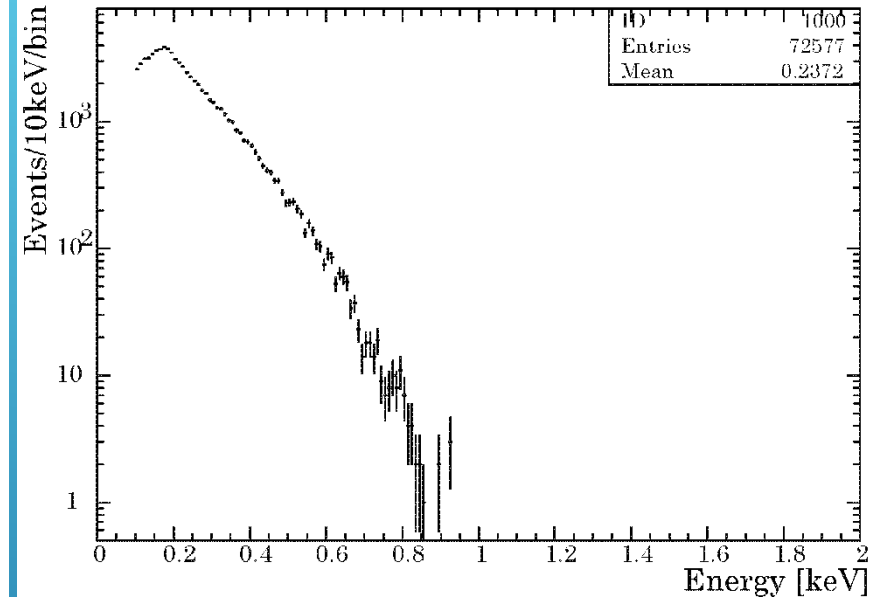
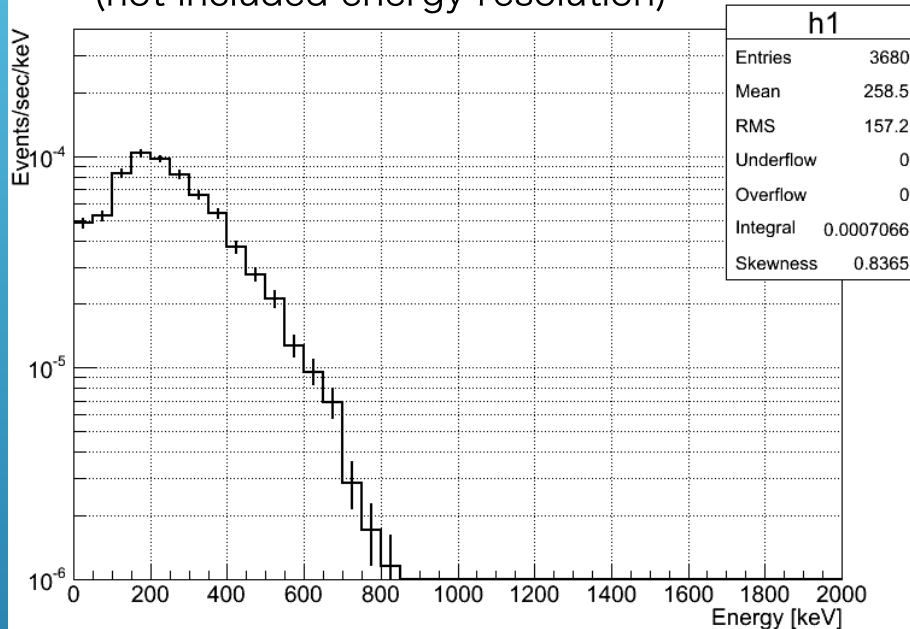
- ・穴を塞ぐ前 182事象/3日間
- ・穴を塞いだ後 100事象/1.68日間
= 179事象/3日間

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

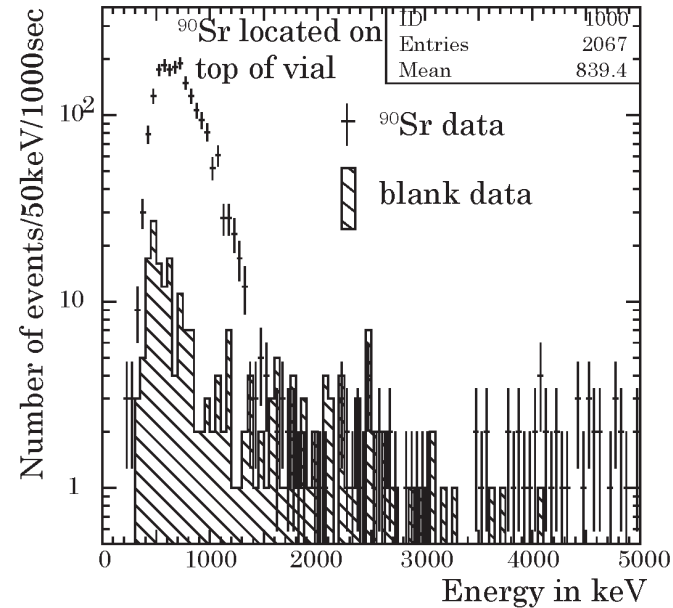
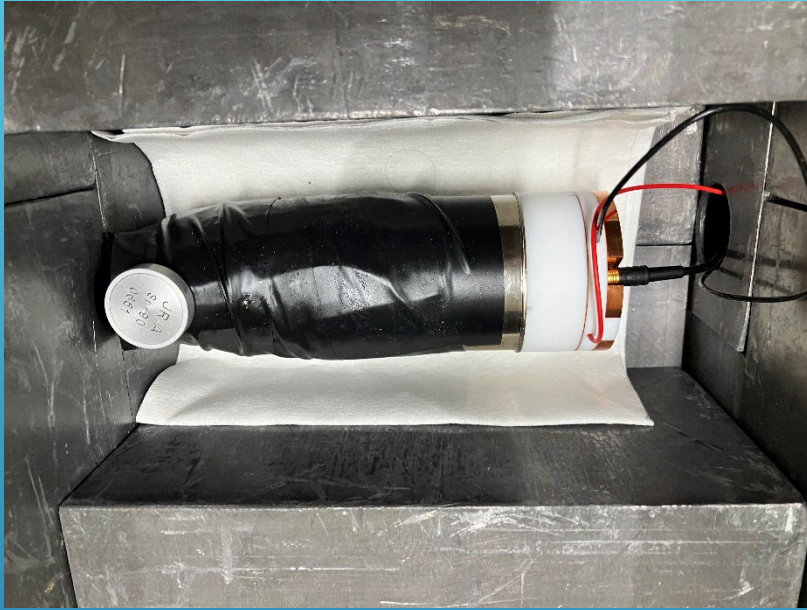
BG MC from ^{210}Bi Brems in Pb Shield
(not included energy resolution)



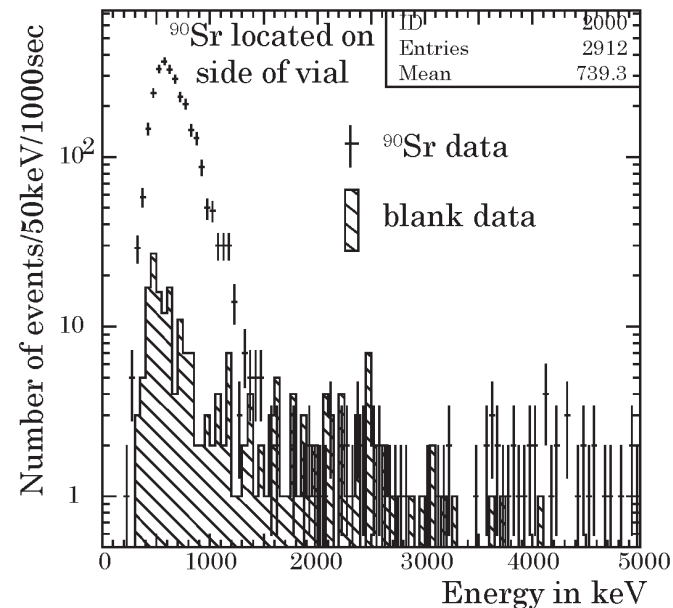
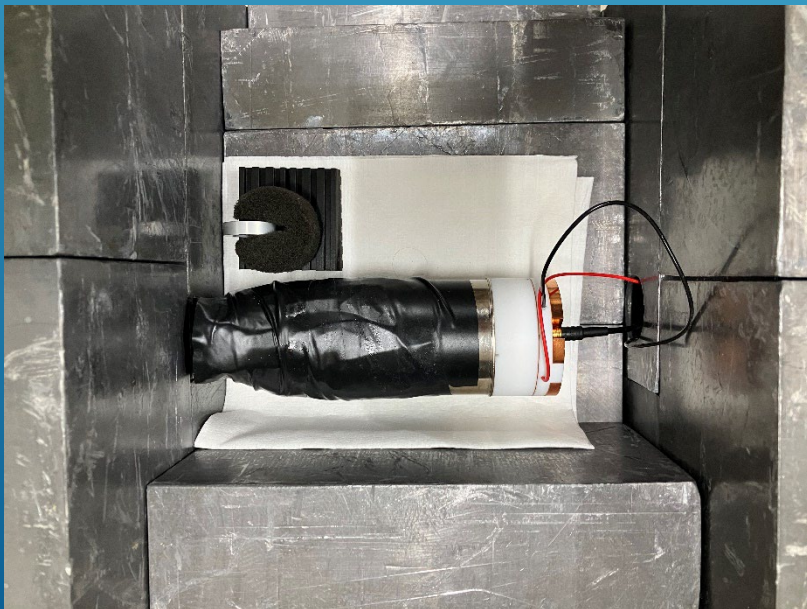
市村さんが振った鉛シールド内の ^{210}Bi のベータ事象からの制動放射事象のエネルギー分布と、EGS5で同様に振った事象のエネルギー分布

よく似ているが、EGS5の方が少しエネルギーが低い

鉛ブロックからの ^{210}Bi のベータ線が見える可能性

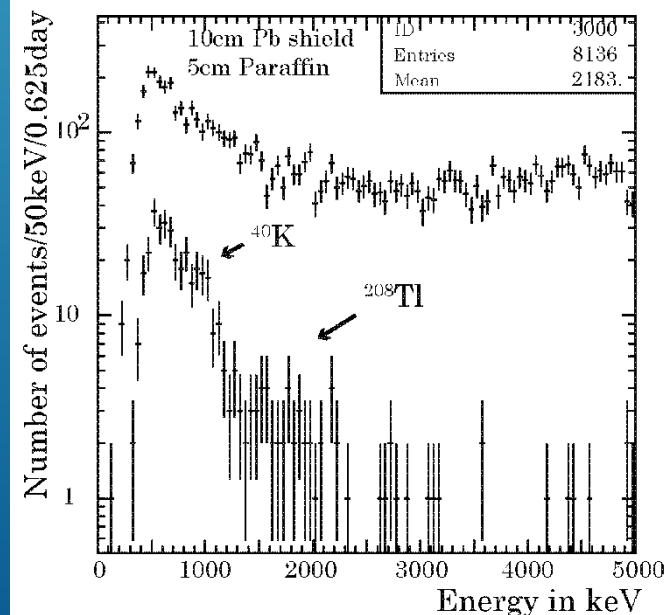
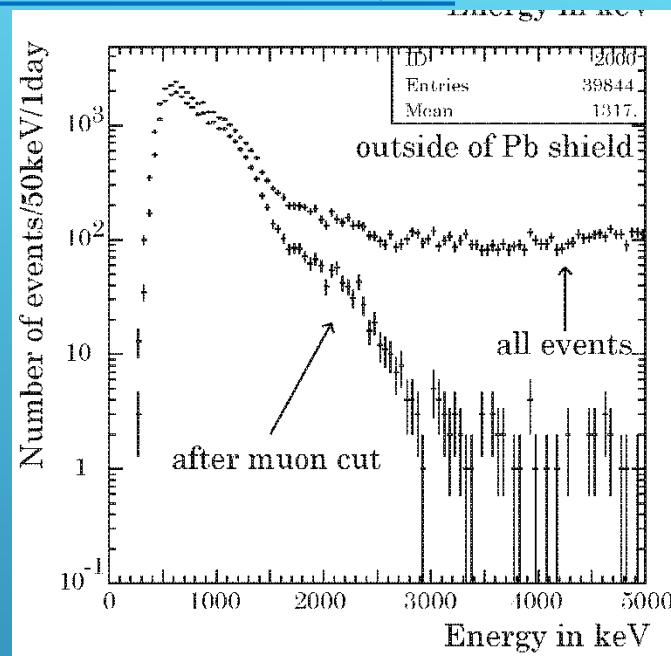
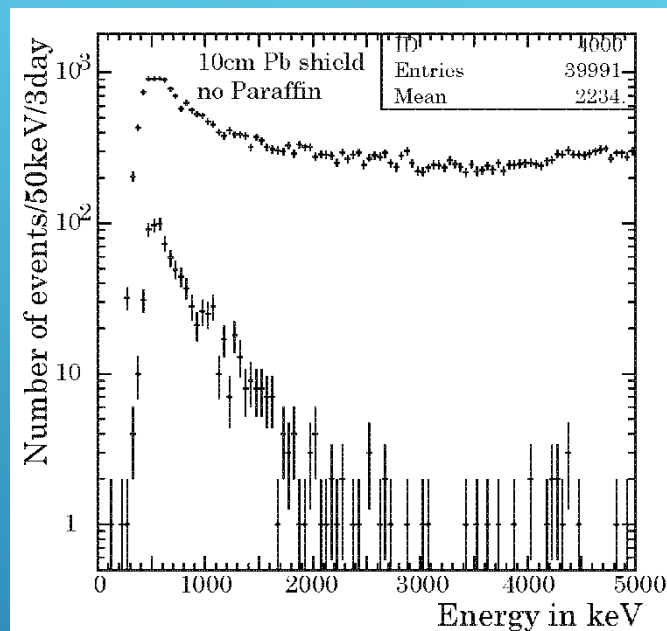


^{90}Sr のベータ線 ($E_{\text{max}} = 2.28\text{MeV}$) が見える。



鉛ブロックからの ^{210}Bi のベータが見える可能性がある

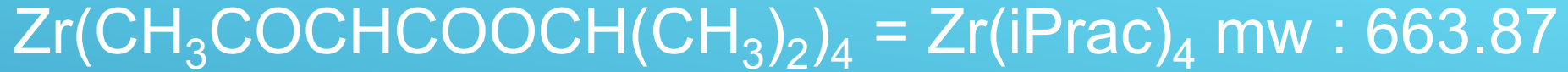
パラフィンブロックで囲んでBG測定



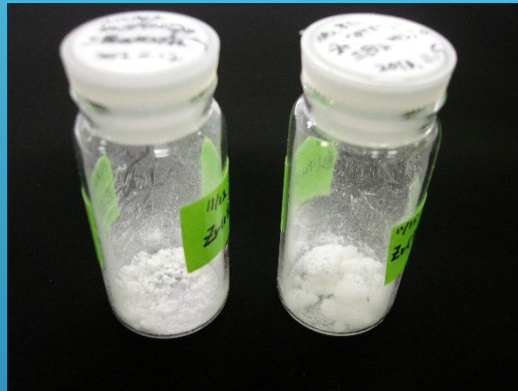
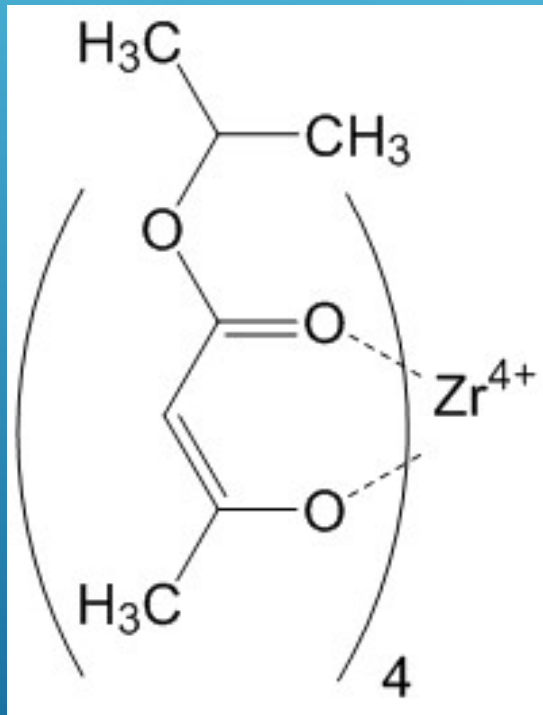
パラフィンで囲んだ場合の600keVの事象数が0.625時間で約30に対し、鉛だけの場合は72時間で約100なので、内部事象なら20事象存在するはずだが、予測される数しか観測されていない。

低エネルギー側は鉛からのベータか？

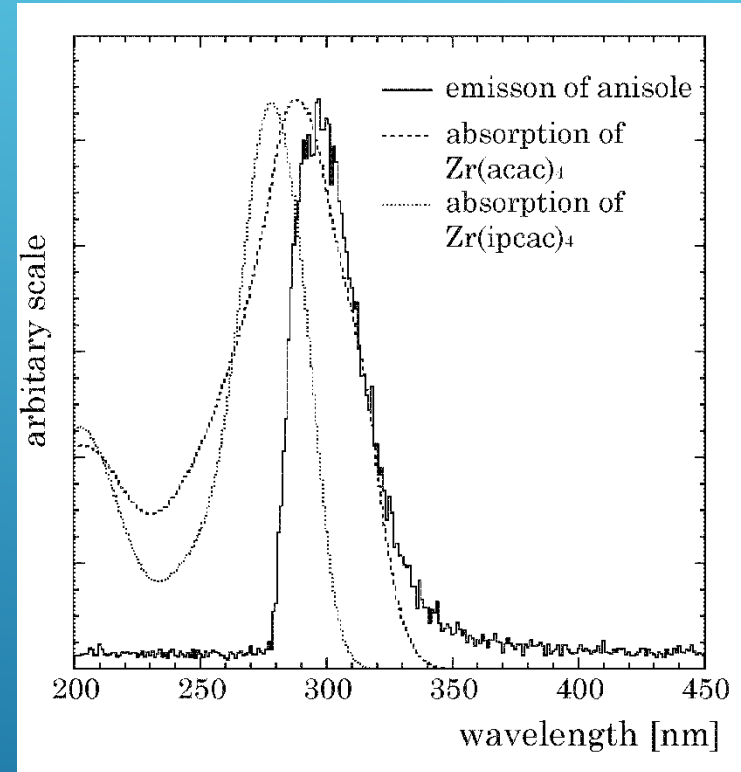
Zirconium β -keto ester complex



Tetrakis(isopropyl acetoacetate) Zirconium



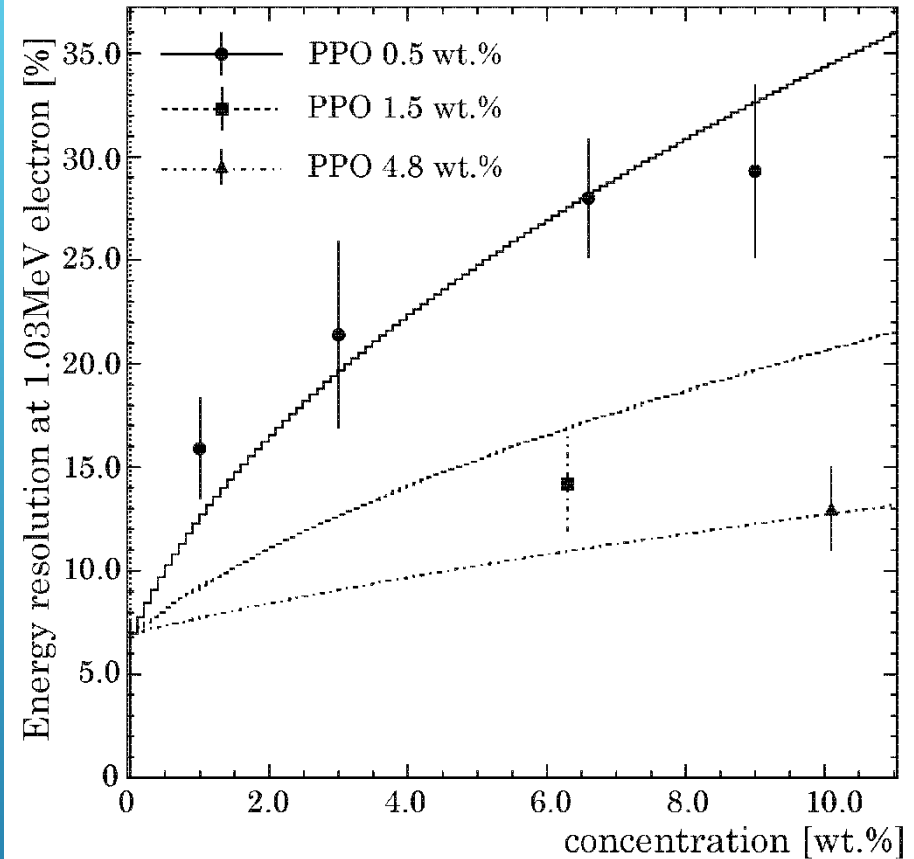
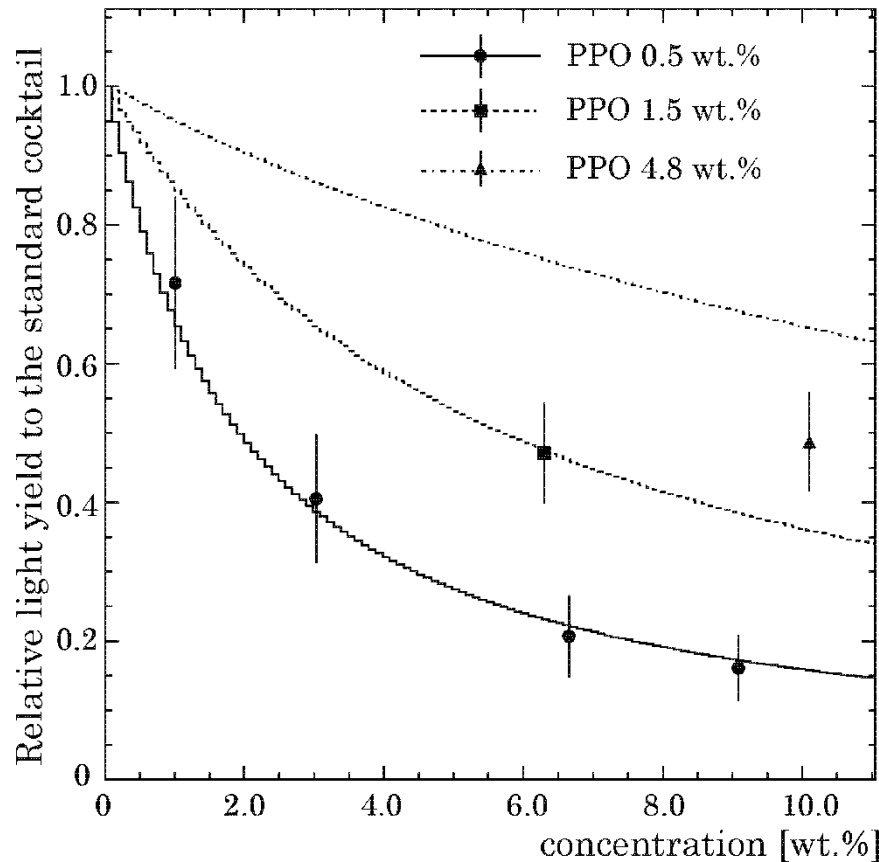
Synthesized by
Prof. T.Gunji
(Tokyo
University of
Science)



No commercial product

Shorter wavelength
for absorbance

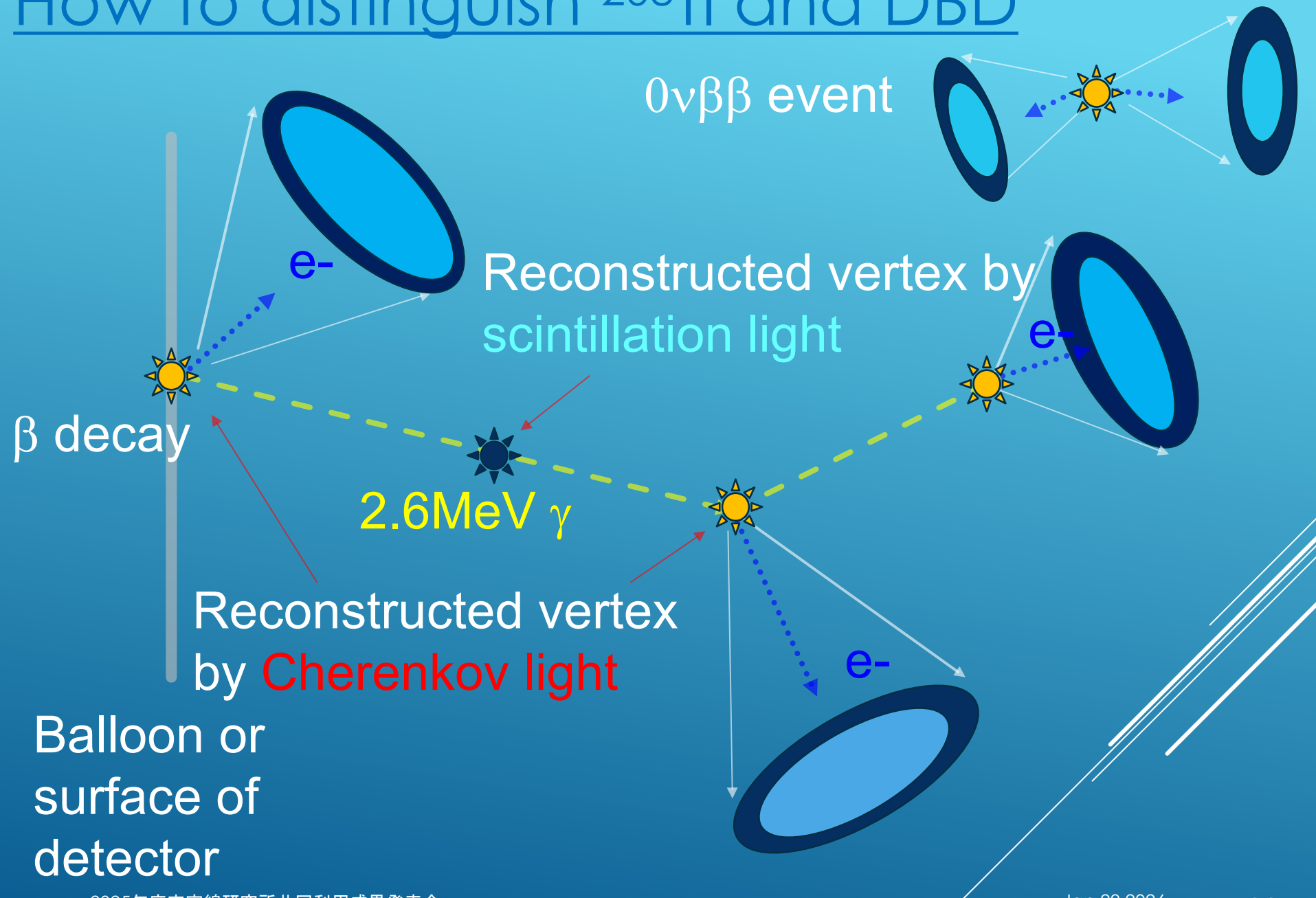
Improve of light yield and resolution



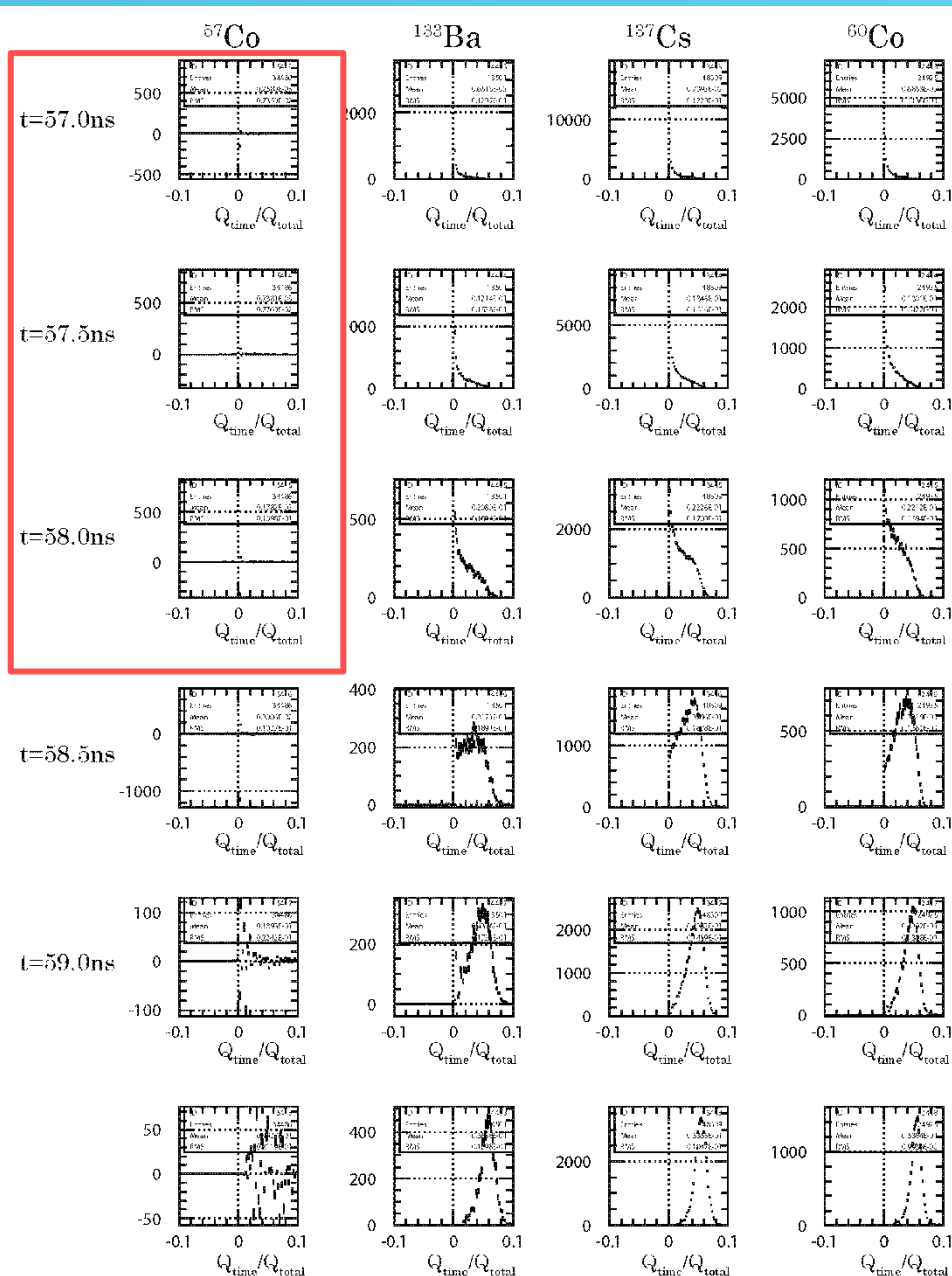
Zr(iPrac)₄ absorbs the scintillation photon and makes the energy resolution worth, however PPO could improve them.

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

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



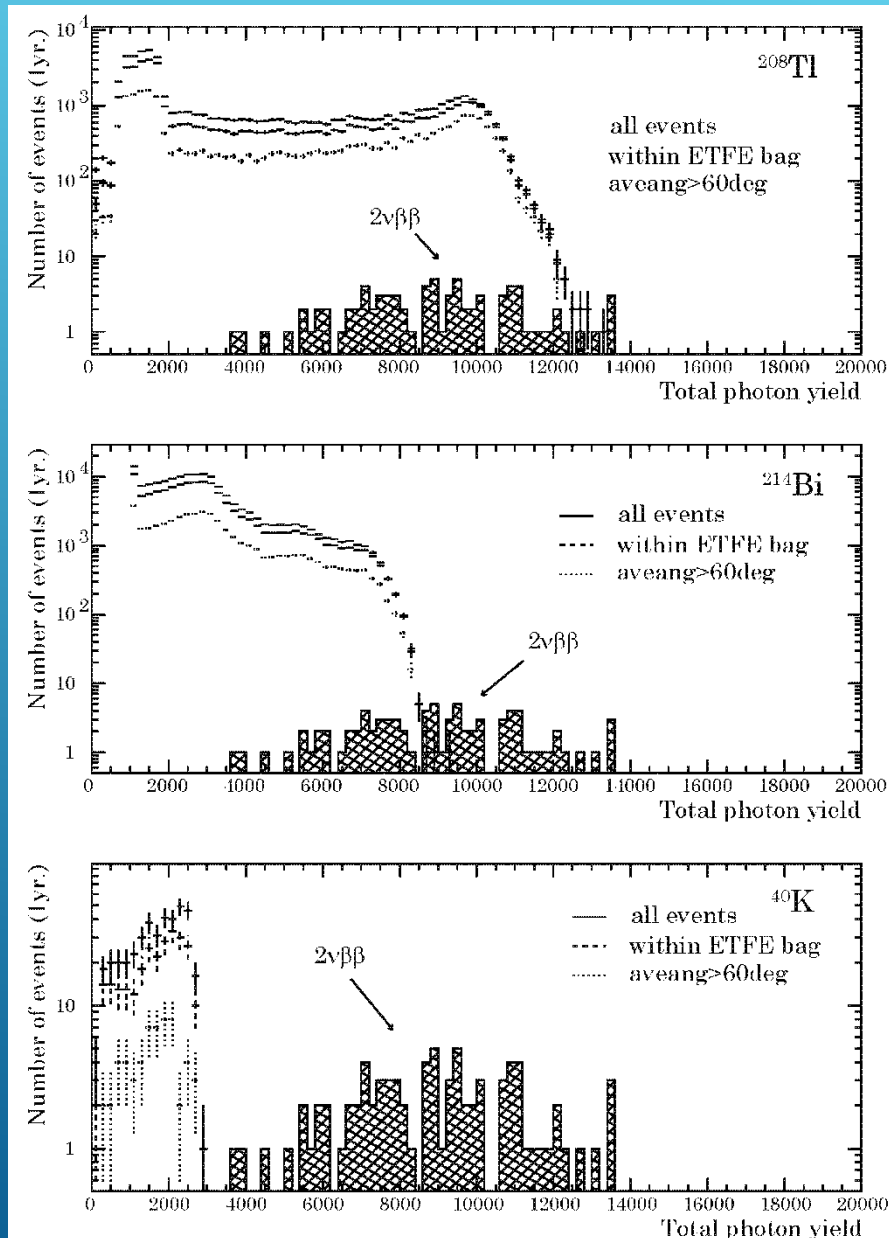
- $t=57\text{ns}\sim 58\text{ns}$ の時間帯で立ち上がり方に違いが見える
- エネルギーが大きいほど立ち上がり方も大きい
- $t>58.5\text{ns}$ の波形はエネルギーによらず一定

$t=57\text{ns}\sim 58\text{ns}$ 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

BG simulation in worst case

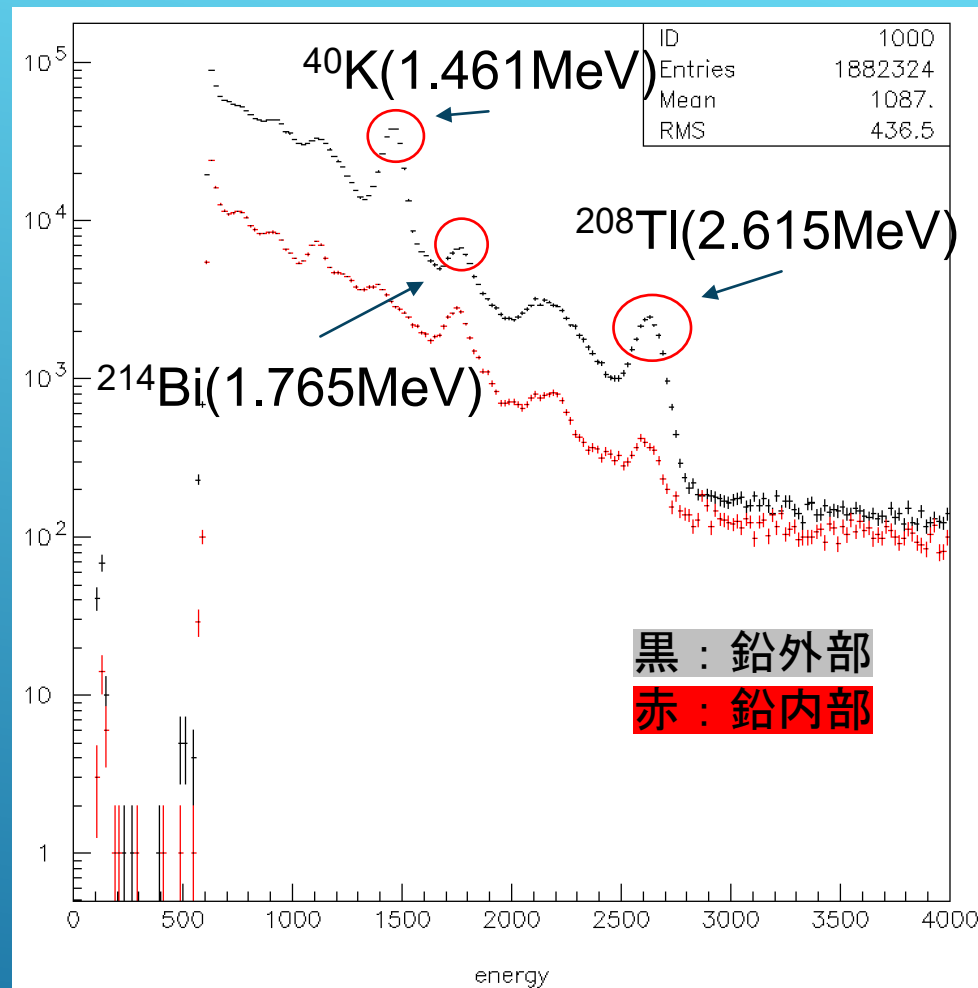


Assuming BGs from flask

- ^{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.

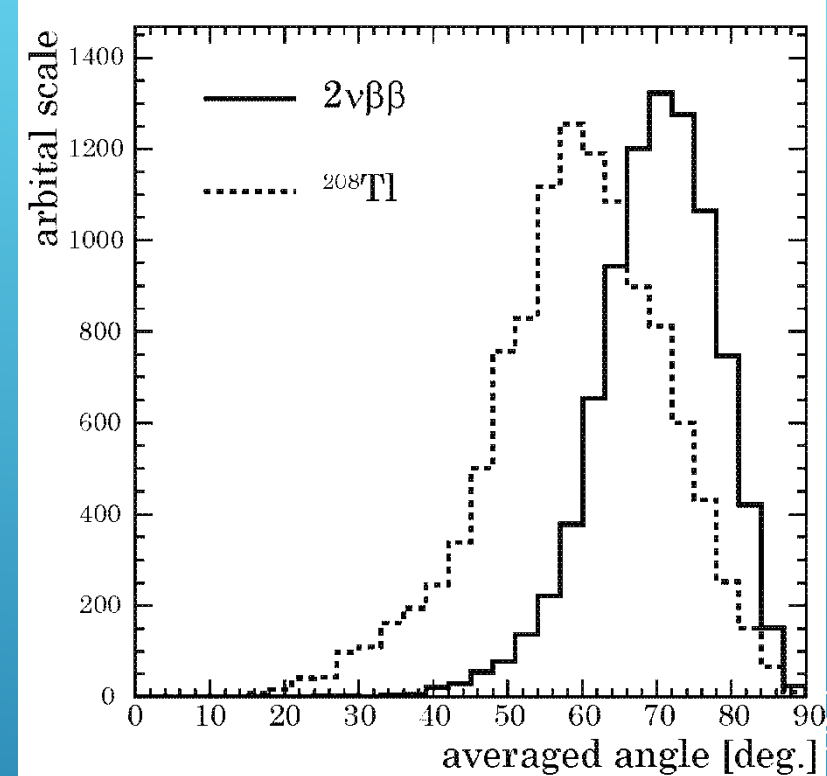
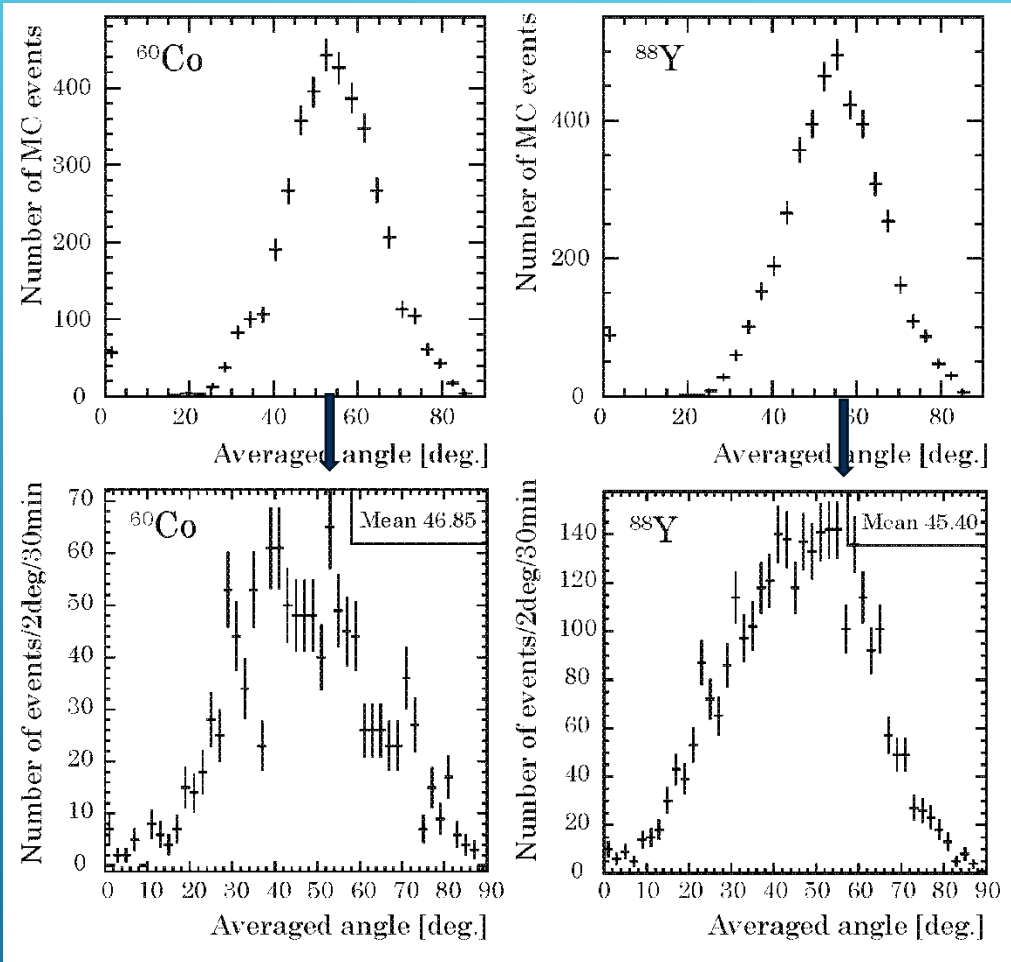
Need actual measurement for ^{96}Zr $2\nu\beta\beta$

10cm mini Pb shield and CsI detector



Due to limit of number of Pb block, we will use 15cm 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.