Low background technologies for astroparticle experiments

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UGAP2024

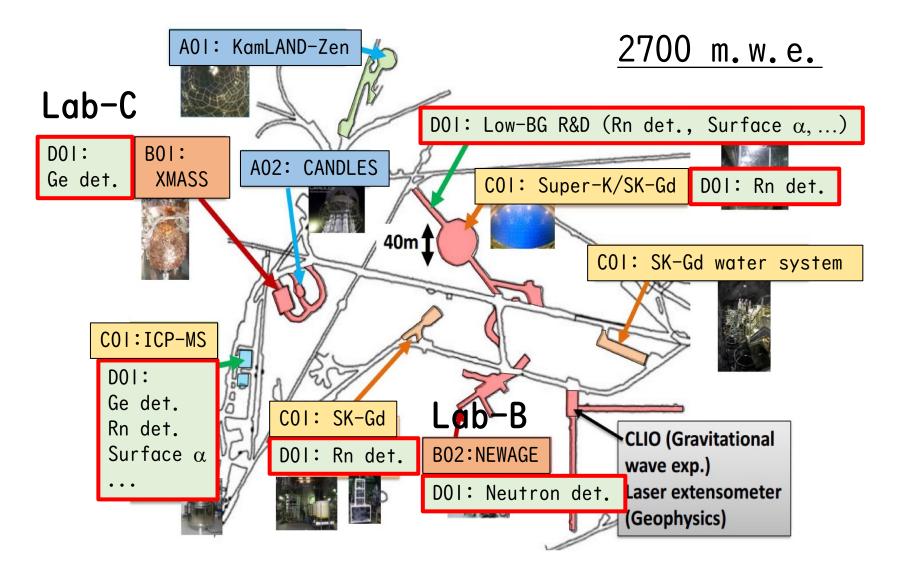
Mar. 6th, 2024

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- Summary

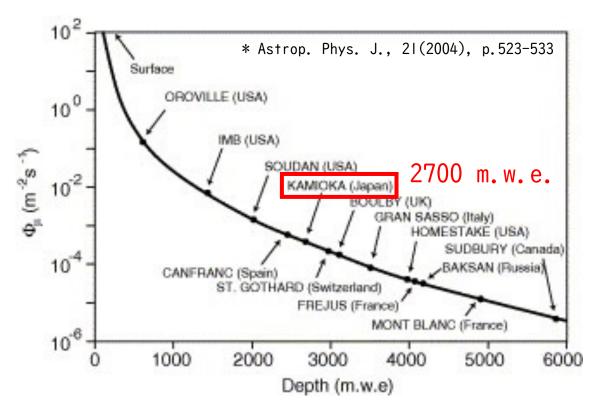
Kamioka underground laboratory

Kamioka Underground Laboratory

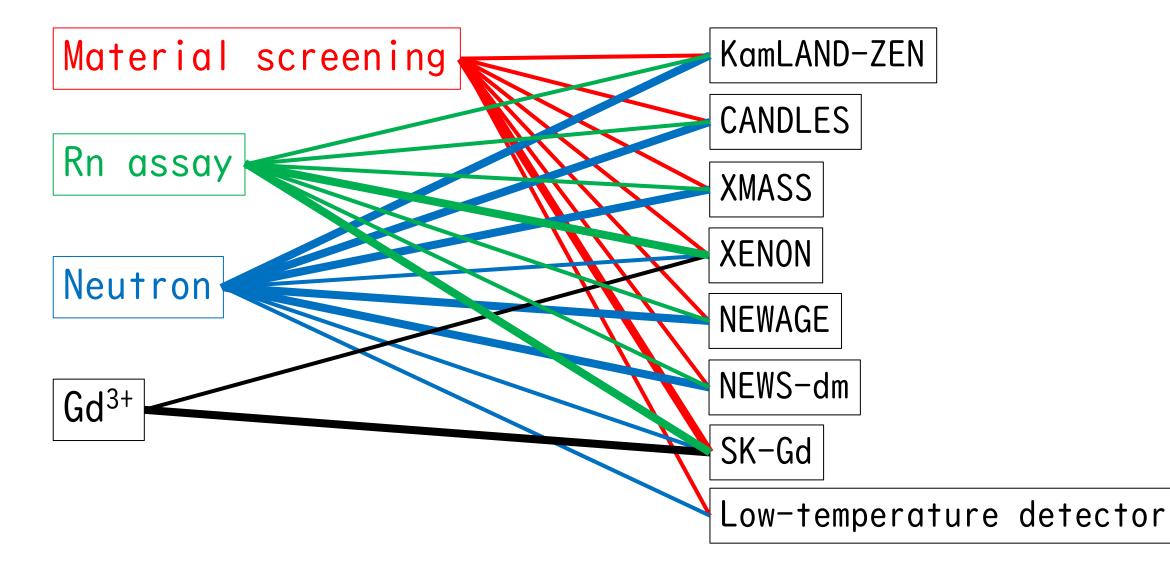


Kamioka Underground Laboratory

- Cosmic ray μ is 10⁻⁵ times less than on the ground.
 - Cosmogenic radioactive nuclei & Neutrons originating from nuclear spallation due to cosmic ray μ are suppressed.



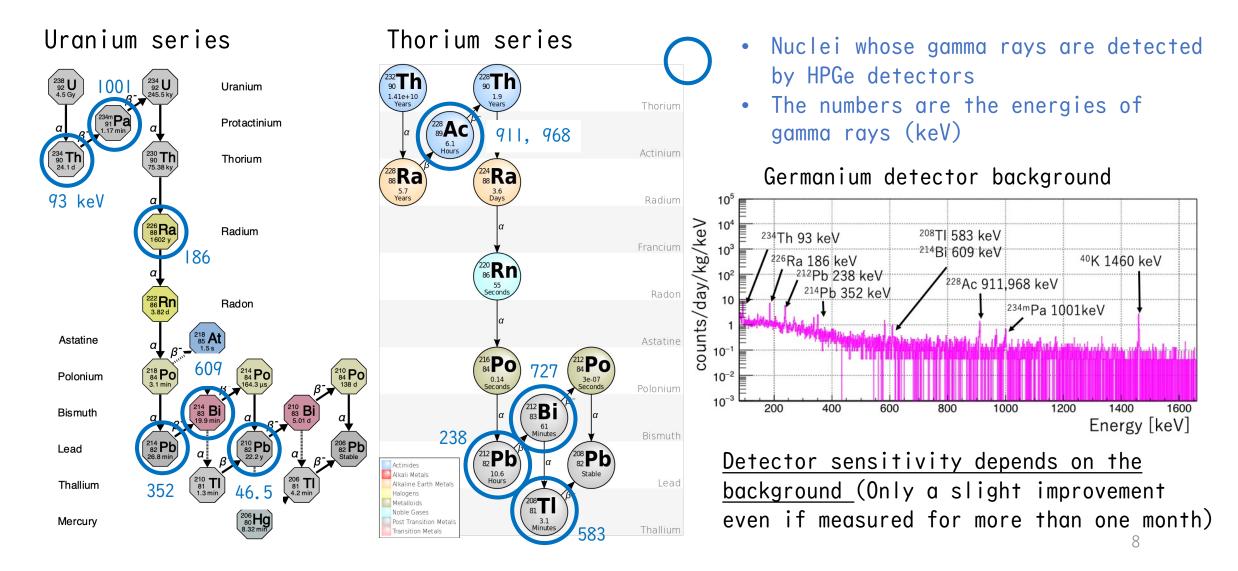
Cooperation overview



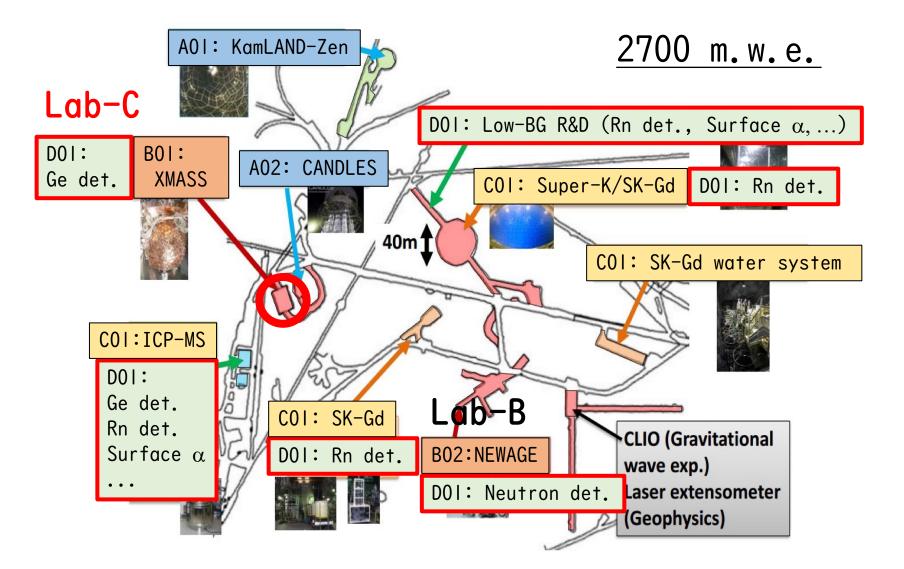
*HPGe: High-Purity Germanium

Material screening with HPGe* detectors

Measurement using HPGe detectors

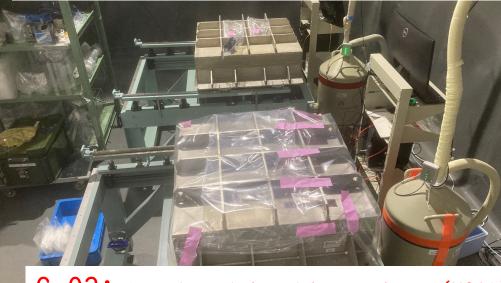


Two HPGe detectors in Lab-C



Two HPGe detectors in Lab-C

GeOl: Developed in XMASS collaboration Delivered in 2016



GeO2: Developed in this project (UGAP) Delivered in 2021

- P-type Coaxial HPGe detectors
 - Relative efficiency*: >80%
 - Low background Aluminum endcap
 - Developed in collaboration with Mirion Technologies (T2FA series)
 - Transported from France to Japan by sea to prevent cosmogenic activation

* Relative efficiency to a NaI(TI) detector with a diameter & length of 3 inches for 1332.5 keV gamma total absorption peak from a ⁶⁰Co point source placed 25 cm on the detector.

GeO2: Photos at the time of delivery

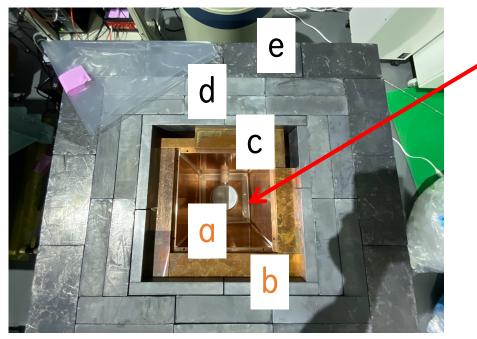


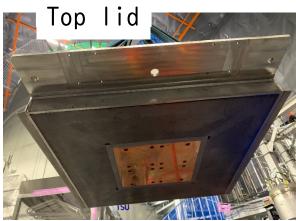


Performance

- Relative efficiency: 82.5%
- Energy resolution (FWHM)
 - 0.81 keV for 122 keV gammas (⁵⁷Co)
 - 1.74 keV for 1332 keV gammas (⁶⁰Co)

GeO2: Radiation shield



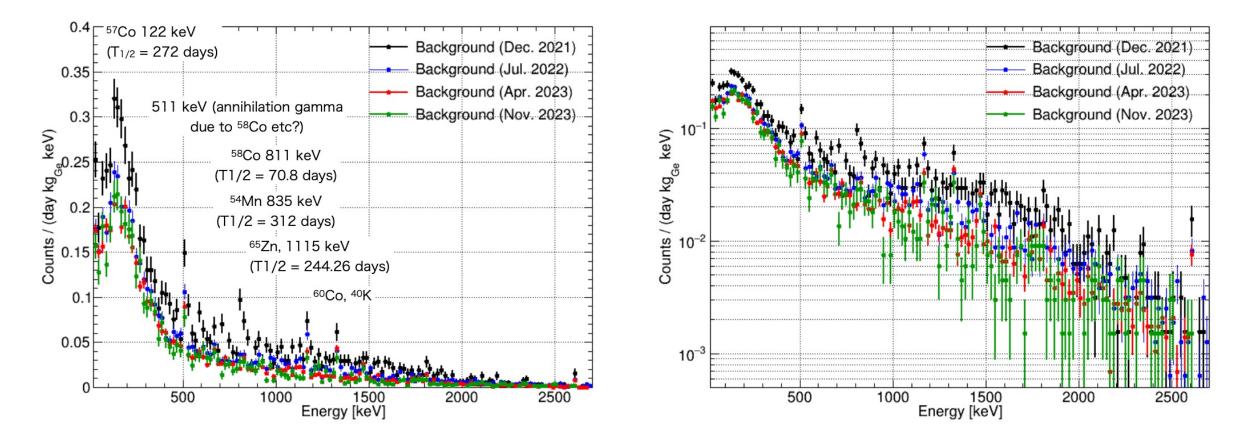


Sample space with Rn free air: capable of measuring 0 (10 kg) of gadolinium sulfate

a: I cm 6N grade CU
 (surface etching with HNO₃*)
b: 5 cm OFHC Cu
c: 2.5 cm Pb (²¹⁰Pb: 5±3 Bq/kg)
 (surface etching with HNO₃*)
d: I0 cm Pb (²¹⁰Pb: ~35 Bq/kg)
 except for the top lid
e: I0 cm Pb (²¹⁰Pb: ~180 Bq/kg)

* Surface etching with HNO_3 : Soak in 4% HNO_3 solution for 20 min. 12

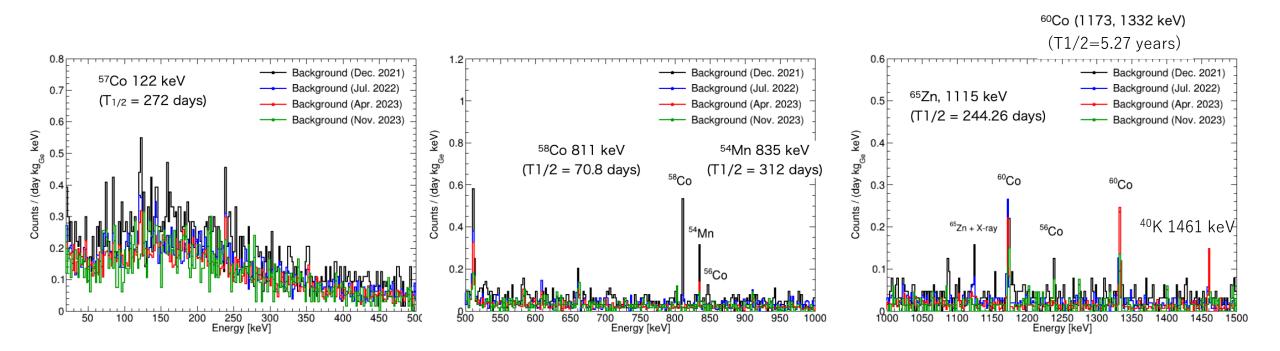
Ge02: Background spectrum



 $\begin{array}{c|cccc} \underline{\text{Count rate in } 40-2700 \ \text{keV:}} \\ \hline 140.3\pm2.1 \ \text{cpd/kg}_{\text{Ge}} \rightarrow 100.0\pm1.1 \ \text{cpd/kg}_{\text{Ge}} \rightarrow 84.3\pm0.8 \ \text{cpd/kg}_{\text{Ge}} \rightarrow 80.0\pm1.5 \ \text{cpd/kg}_{\text{Ge}} \\ \hline (\text{Dec. 2021}) & (\text{Jul. 2022}) & (\text{Apr. 2023}) & (\text{Nov. 2023}) \end{array}$

13

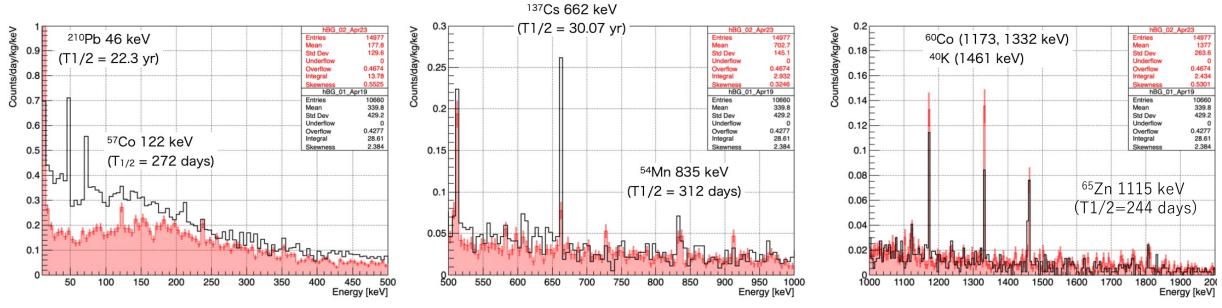
GeO2: Background spectrum



Nucleus with relatively short half-lives are steadily decreasing.

Background spectrum comparison between GOI and GO2

GeOl: Oct. 2018 & Apr. 2019 (LT = 44 days), GeO2: Apr.-Jul. 2023 (LT = 86 days)



Count rate in 40-2700 keV:

GeOI: I40.3 \pm 2.1 cpd/kg_{Ge} \rightarrow GeO2: 84.3 \pm 0.8 cpd/kg_{Ge} (30% improvement)

- ¹³⁷Cs and ²¹⁰Pb peaks have been dramatically improved.
- Continuous components below 600 keV have also been improved.

GeOl & GeO2: Background spectrum

https://doi.org/10.1093/ptep/ptad136

Detector	Ge01	Ge02			
Date	Dec. 2019	Dec. 2021	Jul. 2022	Apr. 2023	Nov. 2023
Measurement time (d)	23.0	19.0	47.2	86.2	19.9
Count rate $(kg_{Ge}^{-1} day^{-1})$					
Integral $40 - 2700$ keV	112.6	140.2	100.0	84.3	80.0
208 Tl, 2614 keV	$0.08 {\pm} 0.04$	$0.25 {\pm} 0.09$	$0.16 {\pm} 0.05$	$0.13 {\pm} 0.03$	$0.03 {\pm} 0.03$
214 Bi, 609 keV	$0.39{\pm}0.10$	$0.25 {\pm} 0.09$	$0.38{\pm}0.07$	$0.23 {\pm} 0.04$	$0.24{\pm}0.08$
60 Co, 1333 keV	$0.41 {\pm} 0.10$	$0.66 {\pm} 0.14$	$0.48 {\pm} 0.08$	$0.68{\pm}0.07$	$0.48 {\pm} 0.12$
40 K, 1461 keV	$0.44{\pm}0.11$	$0.31 {\pm} 0.10$	$0.44 {\pm} 0.07$	$0.42 {\pm} 0.05$	$0.18{\pm}0.07$
$^{137}Cs, 662 \mathrm{keV}$	$1.29{\pm}0.18$	$0.53 {\pm} 0.13$	$0.38{\pm}0.07$	$0.32{\pm}0.05$	$0.42{\pm}0.11$
210 Pb, 46.5 keV	$3.24{\pm}0.29$	$0.69 {\pm} 0.14$	$0.64{\pm}0.09$	$0.59{\pm}0.06$	$0.27 {\pm} 0.09$

Nuclei have relatively short half-lives are steadily decreasing.

GO2:Comparison with HPGe detectors around the world

https://doi.org/10.1093/ptep/ptad136

	Site	Detector	Crystal mass [kg]	Relative efficiency [%]	FWHM at 1333 keV [keV]	BG rate 60 - 2700 keV $[\text{kg}_{Ge}^{-1} \text{ d}^{-1})]$
Japan	Kamioka	Ge02 (This work) Ge01 [<mark>2</mark>]	1.68 1.68	80 80	1.82 2.39	81.3±0.7 104.5
Italy	LNGS	Gator [16] GeMPI [16]	2.2 2.2	100.5 98.7	1.98 2.20	89.0 ± 0.7 24 ± 1
UK	BUGS	Belmont [2] Merrybent [2]	3.2 2.0	160 100	1.92 1.87	90.0 145.0
Spain	LSC	GeOroel [2] Asterix [2] GeAnayet [2]	2.31 2.13 2.26	109 95.1 109	2.22 1.92 1.99	128.7 171.3 461.2
US	BHUC	Maeve [17]	2.0	85	3.19	956.1
Swiss	LVdA	GeMSE [16,18]	2.0	107.7	1.96	88±1

We have developed an ultra-low BG HPGe detector with the world's highest level of sensitivity.

GO2:Comparison with HPGe detectors around the world

*STELLA at LNGS, TAUP2023

	detector	total and peak background count rate [d ⁻¹ kg ⁻¹ Ge]			
		40-2700 keV	352 keV	583 keV	1461 keV
	GeMi	555 ± 7	4.1 ± 1.0	1.4 ± 0.5	6.1 ± 0.8
	GePV	498 ± 5	2.6 ± 0.7	1.8 ± 0.4	3.2 ± 0.4
	GsOr	442 ± 5	2.0 ± 0.5	0.76 ± 0.35	4.2 ± 0.5
	GePaolo	222 ± 2	1.1 ± 0.3	0.31 ± 0.16	1.8 ± 0.2
	GeCris	115 ± 2	0.29 ± 0.22	< 0.13	0.88 ± 0.22
	GeMPI	71 ± 2	< 0.07	< 0.06	0.24 ± 0.03
	Ge02	84 <u>+</u> 1	0.44±0.05	0.25 <u>+</u> 0.04	0.42 <u>+</u> 0.05
We have developed an ultra-low BG HPGe detector with the world's highest level of sensitivity.					

$Gd_2(SO_4)_3$ screening for SK-Gd

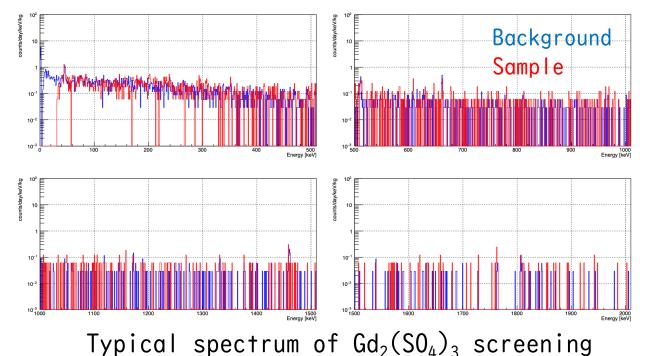
- $Gd_2(SO_4)_3$ dissolved in Super-Kamiokande
 - 13 tons in 2020
 - 26 tons in 2022
- Requirements for RI in $Gd_2(SO_4)_3$ for SK-Gd

Chain	Isotope	Criterion [mBq/kg]	Physics target
23811	$^{238}\mathrm{U}$	< 5	SRN
1000	226 Ra	< 0.5	Solar
²³² Th	232 Th	< 0.05	Solar
	228 Ra	< 0.05	Solar

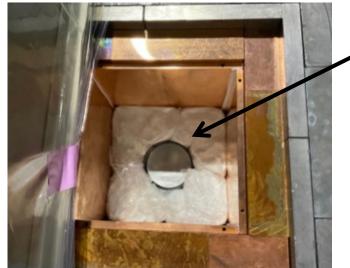
- Select an appropriate method for each nucleus
 - ²³⁸U, ²³²Th: ICP-MS
 - Other nuclei: HPGe detector

$Gd_2(SO_4)_3$ screening with HPGe

- In 2022, 26 tons of $Gd_2(SO_4)_3$ dissolved in SK-Gd was delivered in 37 lots.
 - HPGe detector measurements were performed on 34 lots using GeOl & GO2. The other lots were measured by European collaborators.



20 days required to measure each lot



 $Gd_{2}(SO_{4})_{3}$

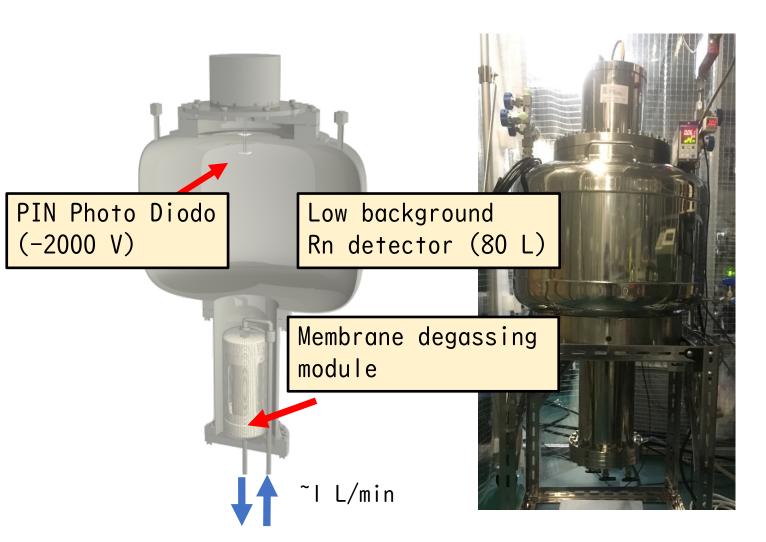
Rn assay in $Gd_2(SO_4)_3$ water

Continuous measurement of 222 Rn in Gd₂(SO₄)₃ water

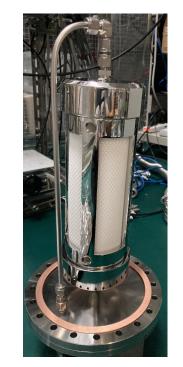
- Motivation
 - SK-Gd: β -decay of ²¹⁴Bi in the ²²²Rn daughter nucleus becomes a background in solar neutrino observation.
 - XENONnt: ²²²Rn affects the dead time of neutron veto water Cerenkov detector.
- Required sensitivity
 - The background is <1 mBq/m³.
- Development policy
 - Improve the existing water radon detector*.

* C. Mitsuda et al., NIMA 497 (2003) 414.

Continuous measurement of 222 Rn in Gd₂(SO₄)₃ water

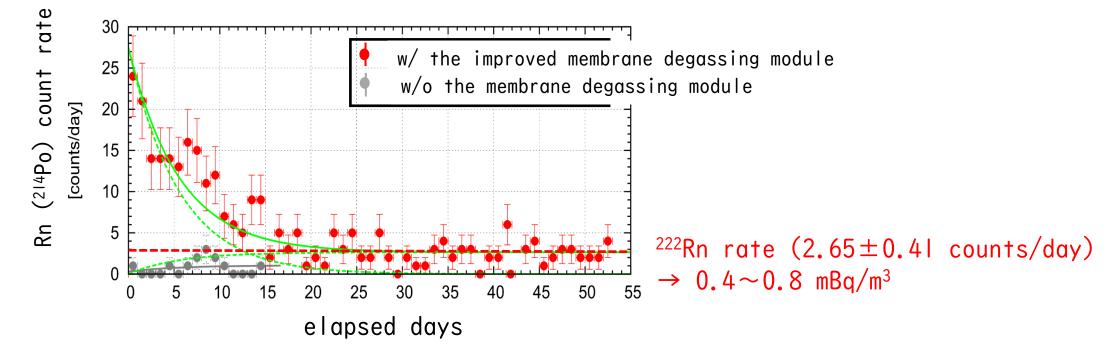


- The membrane degassing module is the main background source.
- Housing changed from resin to stainless steel.



Continuous measurement of 222 Rn in Gd₂(SO₄)₃ water

- We achieved the required background level of <1 mBq/m³.
 - IOx improvement from the past design.



• Four detectors are started operation in SK-Gd & XENONnT.

Automation of high-sensitivity 222 Rn measurement in Gd₂(SO₄)₃ water

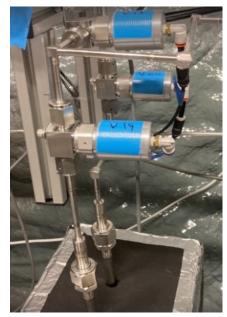
- High-sensitivity measurement method
 - After adsorbing ²²²Rn onto activated carbon cooled to -75°C, the activated carbon is heated to 150°C to release ²²²Rn.
 - Then, ²²²Rn is measured using a low-background radon detector.
- Problems with the conventional method
 - Complex valve operations had to be performed manually.
 - It was necessary to attach and detach the refrigerator and heater when cooling and heating the activated carbon.
- Goal
 - Automation of work with remote valves, temperature controller, and operation panel.

Automation of high-sensitivity $^{222}\mathrm{Rn}$ measurement in $\mathrm{Gd}_2(\mathrm{SO}_4)_3$ water

• Newly introduced equipment

Remote valves

for pure air



for Gd₂(SO₄)₃ water

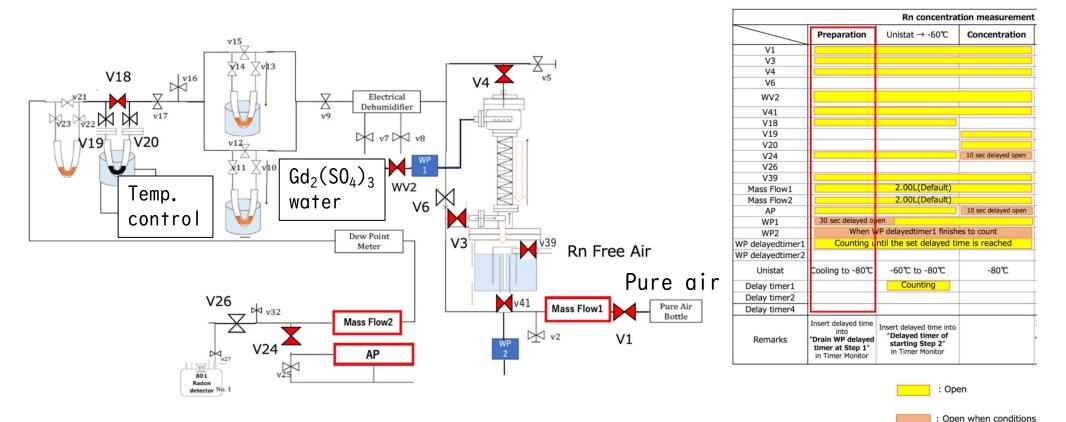


<u>Temp. controller</u> −75°C to 150°C

Operation panel



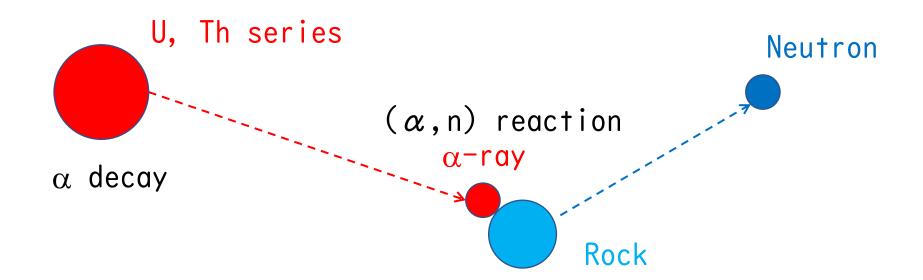
Automation of high-sensitivity 222 Rn measurement in Gd₂(SO₄)₃ water



We have successfully automated the work. The system will start operation in SK-Gd. Environmental neutron measurements in the underground

The primary sources of environmental neutrons in the underground

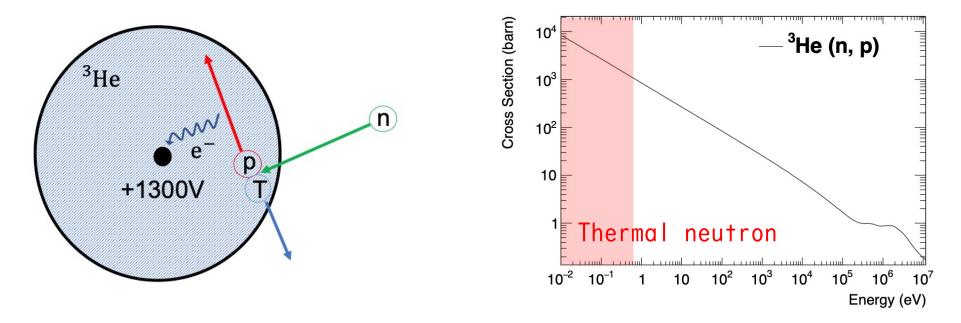
• (α, n) reaction between rock and α -rays generated by the decay of the U and Th series contained therein.



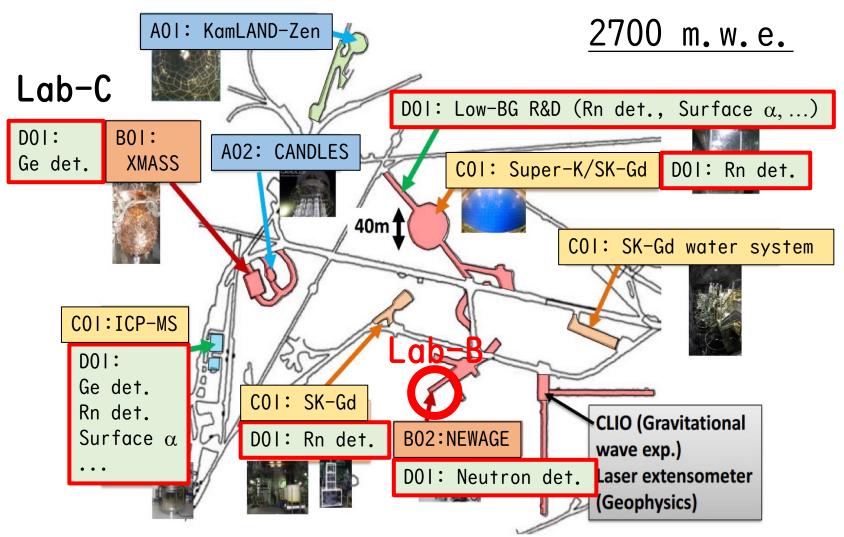
³He proportional counter

³He proportional counter

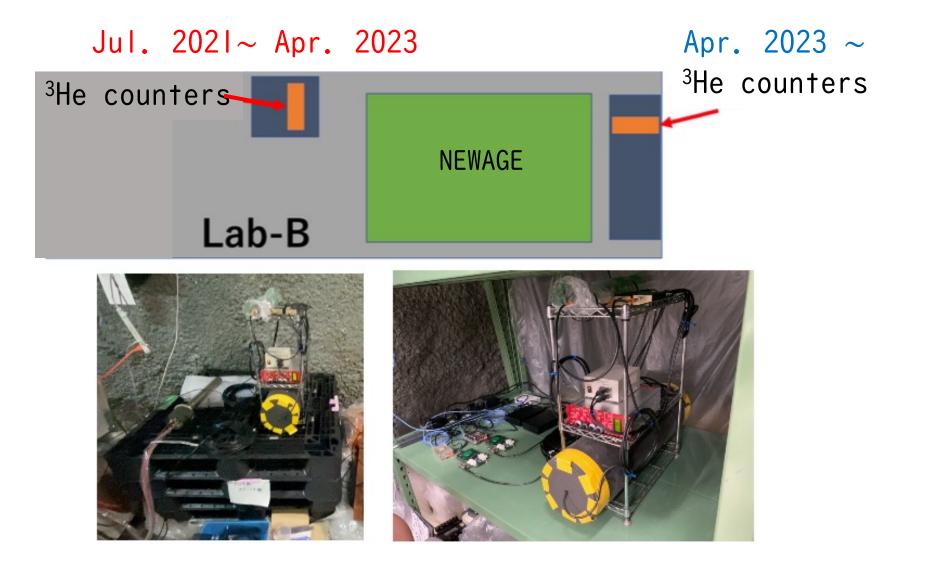
- 3 He + n \rightarrow p + T + 0.765 MeV
- High sensitivity to thermal neutrons
- Fast neutrons are measured after decelerating using a moderator (such as polyethylene).



Environmental neutron measurement in Lab-B

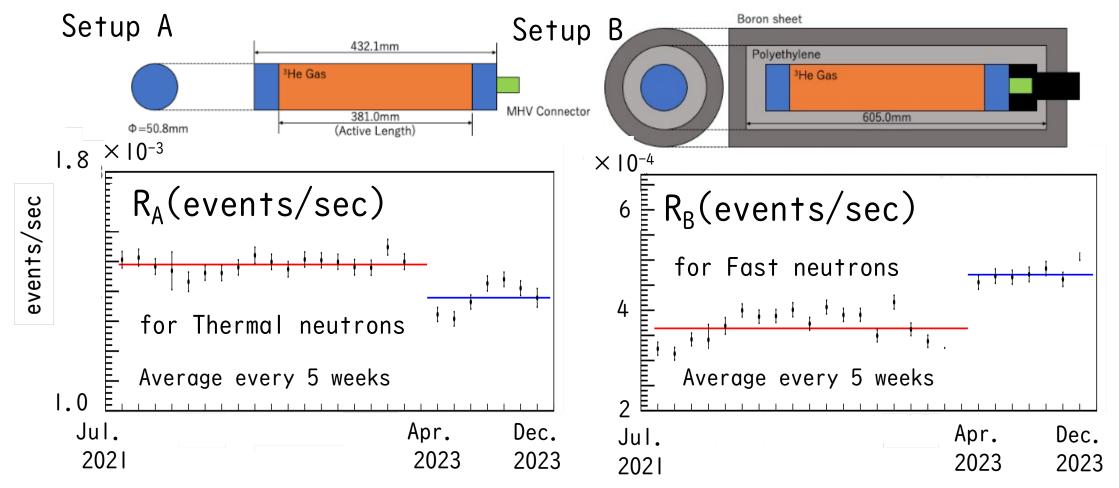


Two ³He proportional counters in Lab-B



Environmental neutron measurement in Lab-B with two ³He proportional counters

• Jul. 2021 ~ Apr. 2023, Apr. 2023 ~



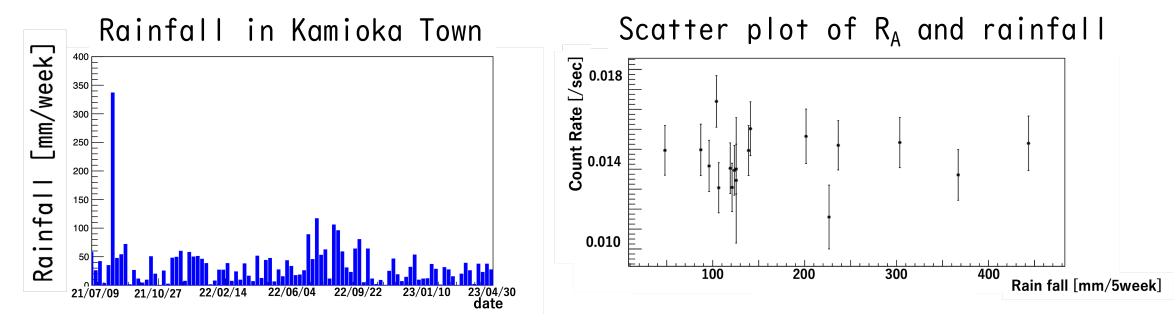
Correlation between Lab-B neutron flux and environmental parameters

We investigated the correlation between Lab-B neutron flux and environmental parameters from Jul. 2021 to Apr. 2023. I. Rainfall

- 2. Rn concentration in the mine-tunnel air
- 3. Humidity in the mine-tunnel air

Correlation with Rainfall

• Using rainfall data observed in Kamioka Town (~12 km from the Kamioka underground laboratory.)

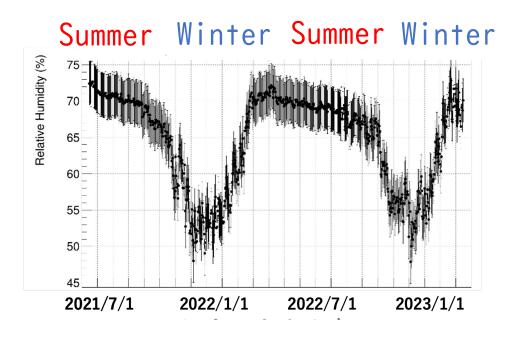


	R _A	R _B	R_A/R_B
correlation coefficient	0.17	0.12	-0.08

No correlation

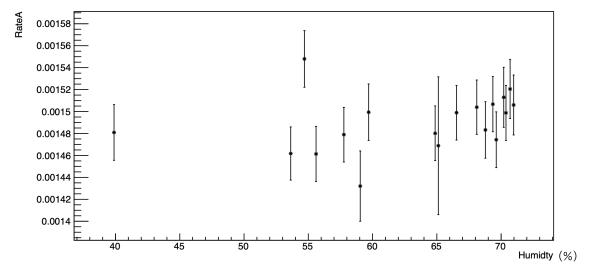
Correlation with Humidity in the mine-tunnel air

Humidity in the mine-tunnel air



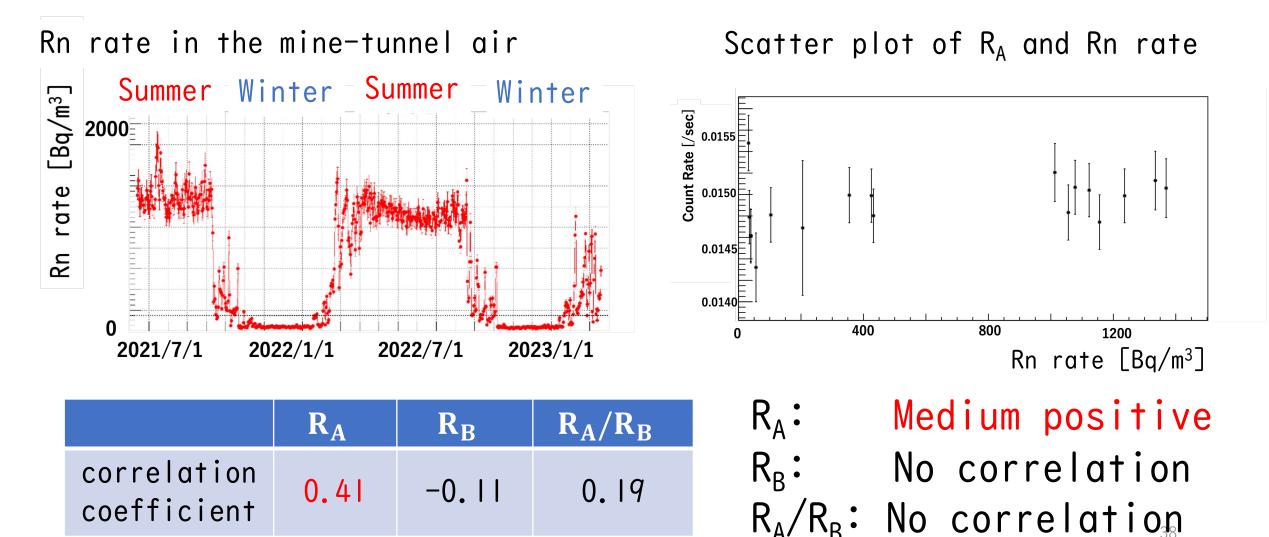
	R _A	R _B	R_A/R_B
correlation coefficient	0.28	-0.27	0.28

Scatter plot of R_A and Humidity



 R_A :Weak positive R_B :Weak negative R_A/R_B :Weak positive

Correlation with the Rn rate in the mine-tunnel air



Summary of long-term environmental neutron measurement in the underground

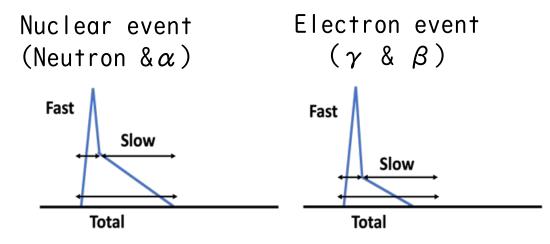
- Conclusion
 - No correlation with rainfall
 - <u>Weak correlation with humidity</u> in the mine-tunnel air
 - <u>Medium correlation with Rn rate</u> in the mine-tunnel air
- ToDo
 - Will verify the effects of humidity and Rn rate in the mine-tunnel through simulation

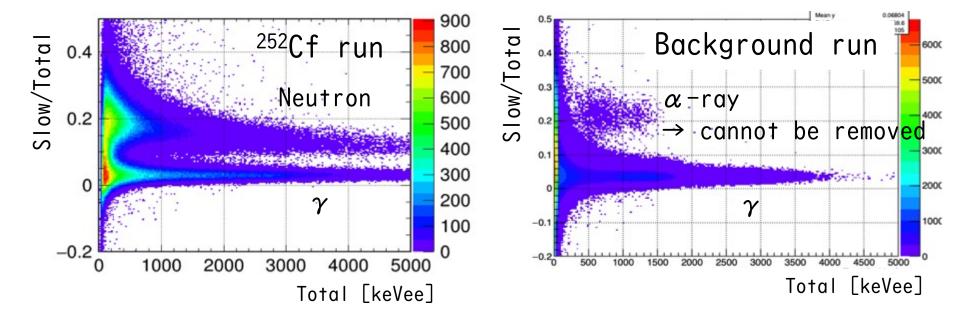
Liquid scintillator neutron detector

* Saint-Gobain BC501A

Liquid scintillator* neutron detector

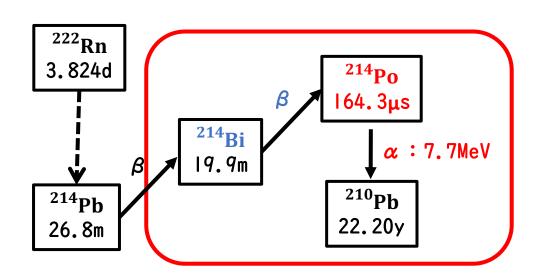
- Neutrons are detected through the scattered protons.
- $\gamma\text{-rays}$ and electrons can be removed by pulse-shape discrimination, but $\alpha\text{-}$ rays are difficult to remove.





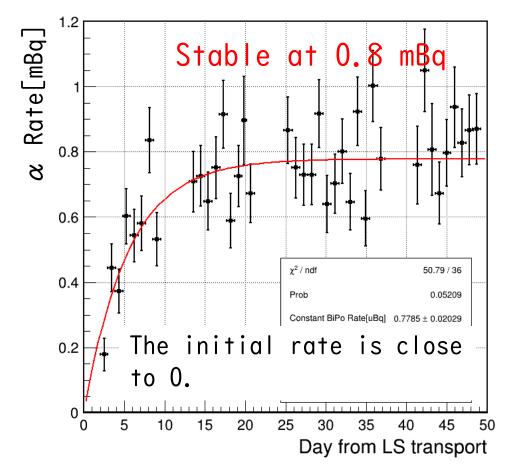
$\alpha\text{-ray}$ background of liquid scintillator detector

 Evaluate α-ray BG by delayedcoincidence of ²¹⁴Bi-²¹⁴Po

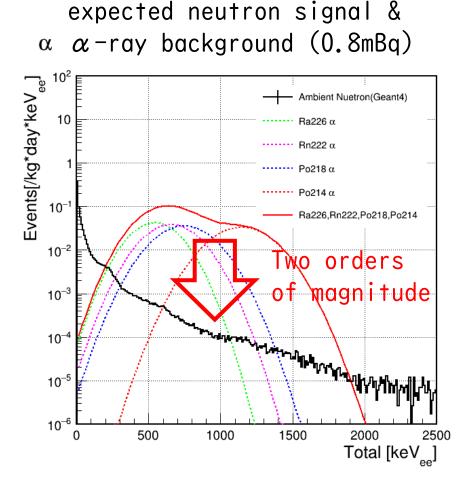


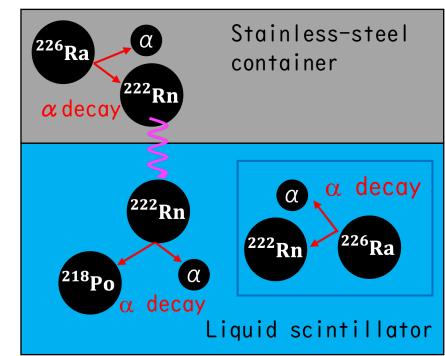
Since the half-life of ²¹⁴Po is short, $^{214}Bi-^{214}Po$ can be selected by Δt .

Bi-Po α -ray rate



$\alpha\text{-ray}$ background of liquid scintillator detector





- Reduction by detector improvement (x 1/10)
 - Additional purification of liquid scintillator
 - Surface treatment of stainless-steel containers
 - ^{222}Rn reduction by periodic N_2 bubbling

$\alpha\text{-ray}$ background of liquid scintillator detector

²²⁶Ra

1600y

²²²Rn

3.824d

²¹⁸Po

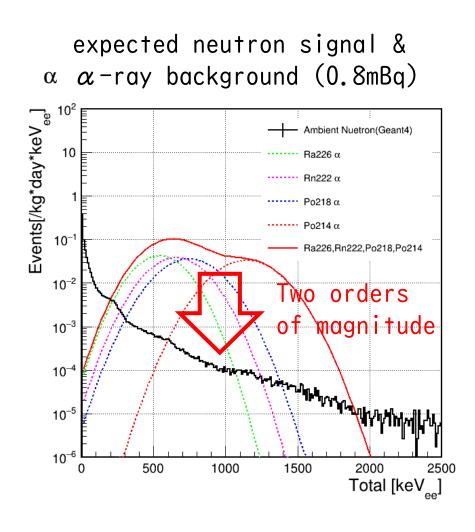
3.10m

²¹⁴Pb

26.8m

²¹⁴Bi 19.9m

α



Reduction through analysis (x 1/10)

²¹⁴Po

164.3µs

²¹⁰Pb

22.20v

α

• Tag the α -rays from ²²²Rn and ²¹⁸Po with delayed coincidence measurement.

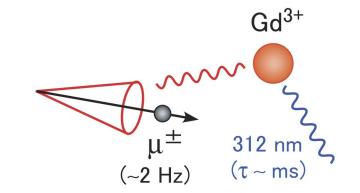
Assuming 222 Rn rate = 0.08 mBq

Time window	500 sec
Signal efficiency	41.2%
α -ray reduction	94.4%

Laser-induced luminescence spectroscopy for Gd³⁺

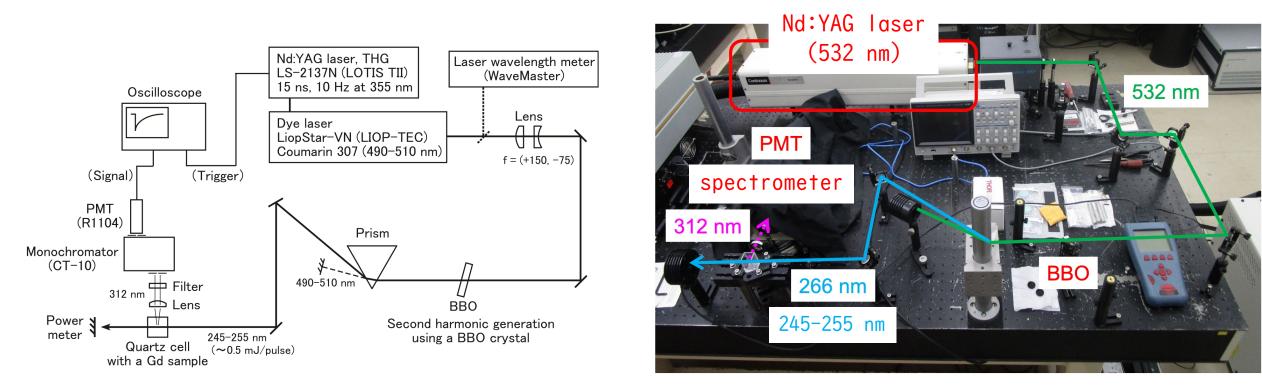
Laser-induced luminescence spectroscopy for Gd³⁺

- Gd³⁺ luminescence is a phenomenon in which Gd³⁺ is excited by a photon and emits a 312 nm photon after O(1) msec.
- In SK-Gd, Gd^{3+} luminescence due to a Cerenkov photon could become a background, but there was no detailed measurement of Gd^{3+} luminescence in $Gd_2(SO_4)_3$ water.
- We investigated the emission characteristics of Gd³⁺ with laser-induced luminescence spectroscopy.

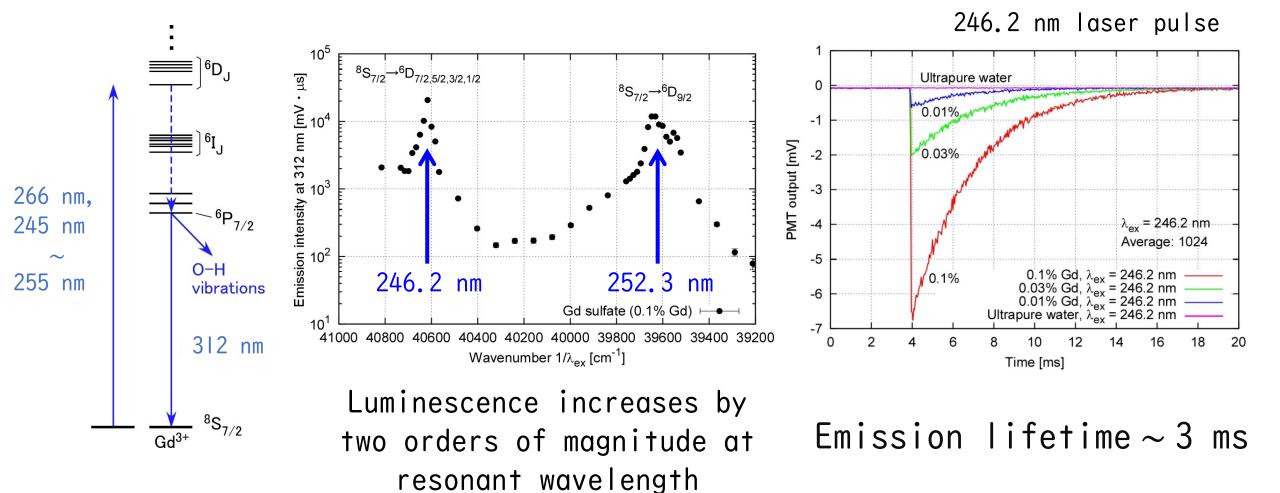


Laser-induced luminescence spectroscopy for Gd³⁺

266 & 245-255 nm laser is irradiated to the cell w/ $Gd_2(SO_4)_3$ water, and 312 nm photons emitted from the cell are observed with a PMT.



Laser-induced luminescence spectroscopy for Gd³⁺

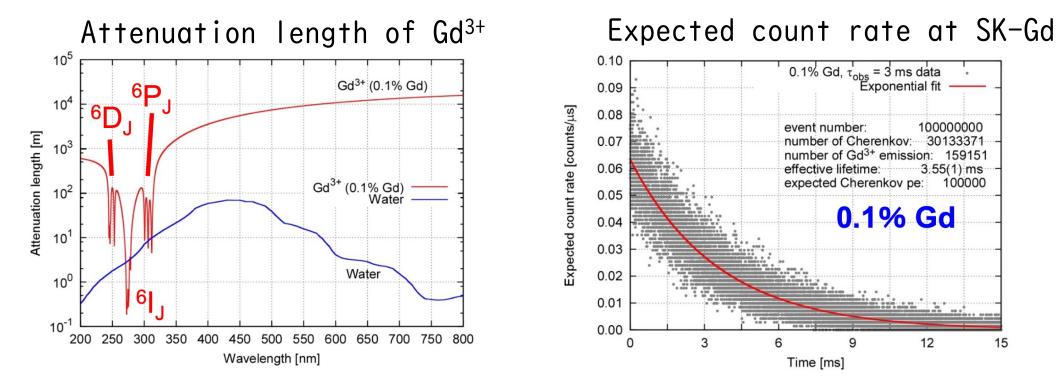


(246.2 nm & 252.3 nm)

48

Impact of Gd³⁺ luminescence on SK-Gd

A simulation study was performed with SK-Gd geometry.



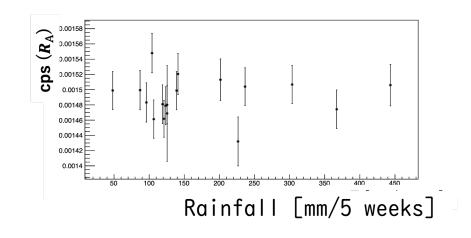
The expected count rate at SK-Gd is < 0.1 counts/ μ s; therefore, the impact on SK-Gd is small.

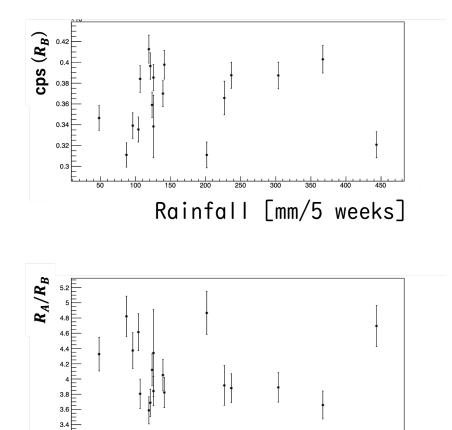
Summary

- Material screenings with HPGe detectors
 - An ultra-low BG HPGe detector with the world's highest level of sensitivity was developed.
- Rn assay in $Gd_2(SO_4)_3$ water
 - The required background level of <1 mBq/m³ is achieved.
- Environmental neutron measurements in the underground
 - Weak correlations with humidity in the mine-tunnel air.
 - Medium correlation with Rn rate in the mine-tunnel air.
- \bullet Laser-induced emission spectroscopy for Gd^{3+}
 - The effect on SK-Gd is small.

Backup

```
Neutron flux % rainfall
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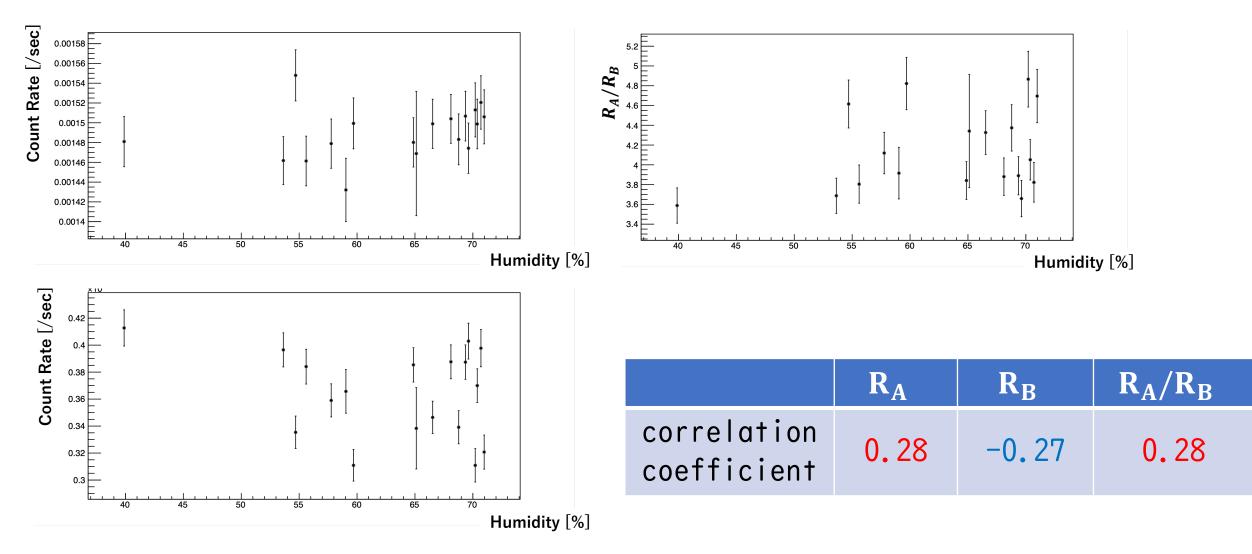


Rainfall [mm/5 weeks]

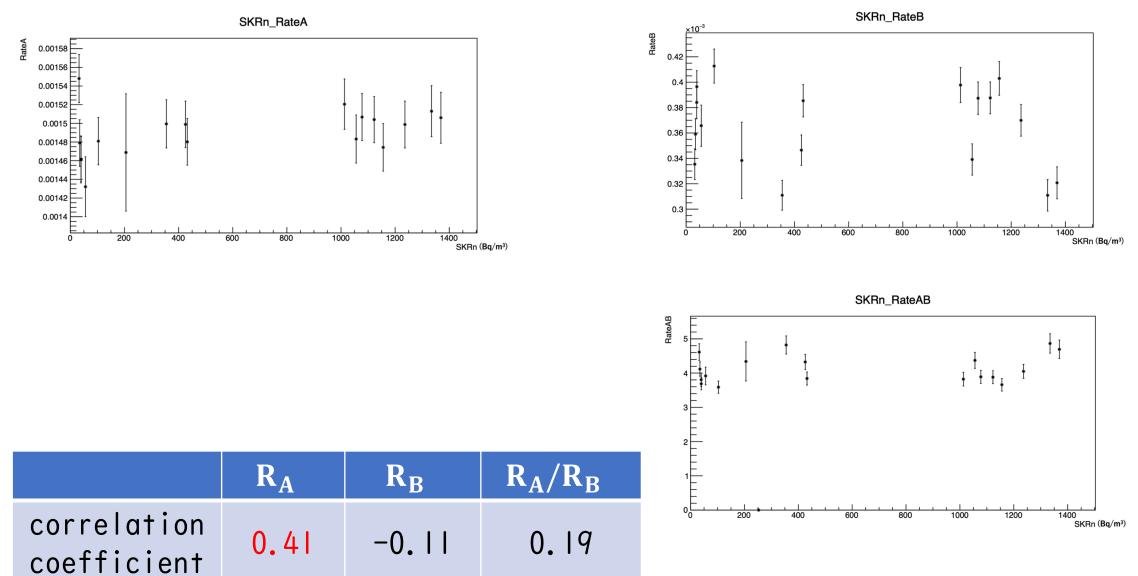
100

	R _A	R _B	R_A/R_B
correlation coefficient	0.17	0.12	-0.08

Neutron flux % Humidity



Neutron flux % Rn rate



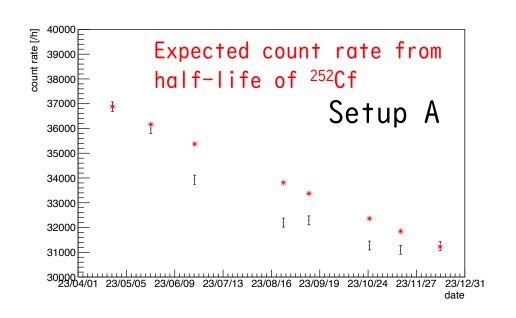
Calibration of ³He proportional counter

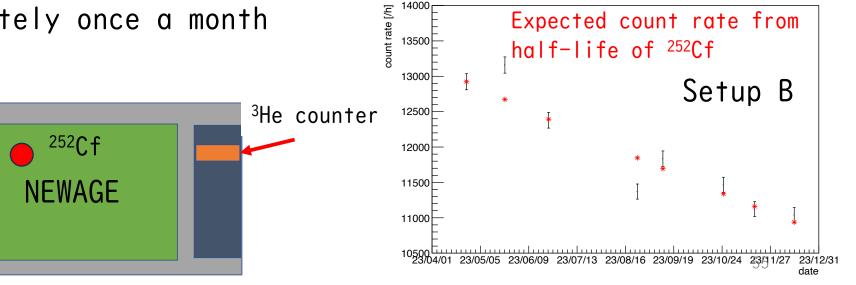
Check if the sensitivity of ³He proportional counter is stable.

• Started on Apr. 2023

Lab-B

- Irradiation with neutrons from ²⁵²Cf for I hour
- Conducted approximately once a month





Calibration of ³He proportional counter

The deviation from the expected rate is

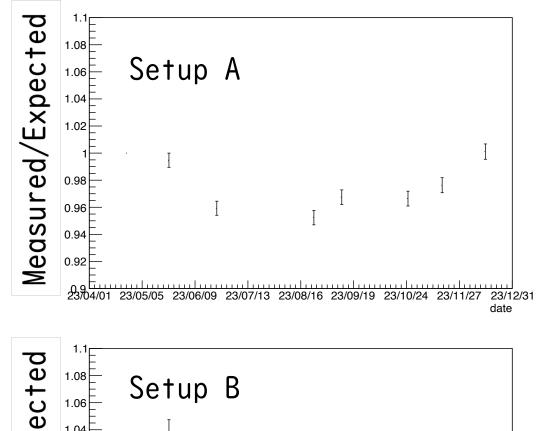
- Setup A: $-5\% \sim 0\%$
- Setup B: $-4\% \sim +4\%$

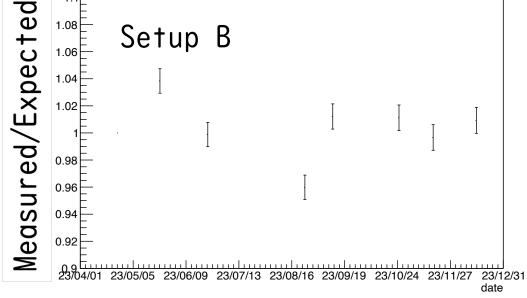
We are currently investigating the cause of the discrepancy from the expected value.

• It has been confirmed that the gain has stayed the same.

Large statistical error

 Considering increasing calibration time





Coefficient Interval Correlation 0.00 - 0.199Very Weak 0.20 - 0.399Weak 0.40 - 0.599Medium 0.60 - 0.799Strong Very Strong 0.80 - 1.000