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高久雄一 環境科学技術研究所 ガドリニウム定量分析
公募研究 坂口綾 筑波大学 高マトリクス試料中の極微量放射性核種測定法確立

C01報告 超新星背景ニュートリノの高感度観測 でせまる宇宙星形成の歴史



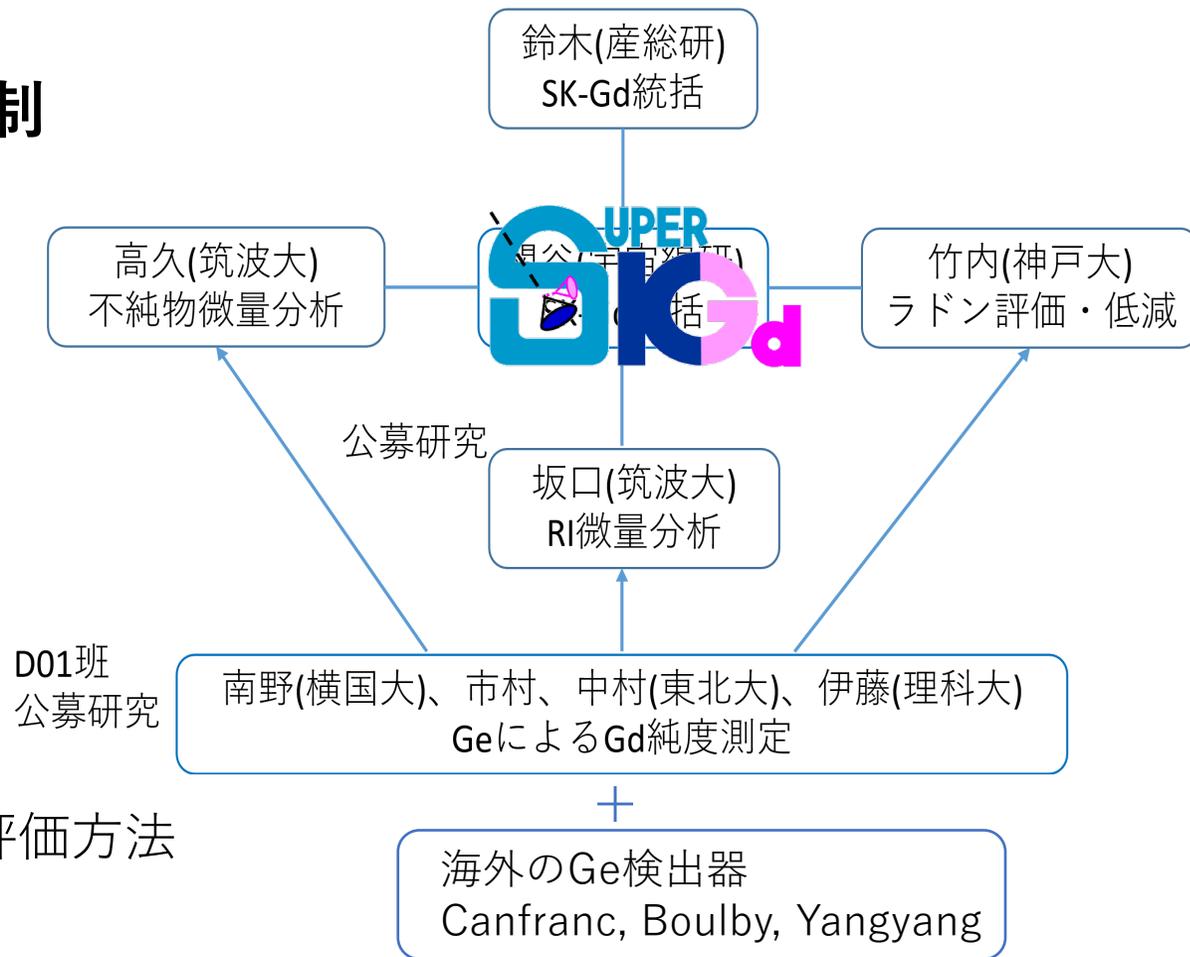
東京大学宇宙線研究所 関谷洋之

2021.5.21

新学術「地下宇宙」領域研究会

2021.5.21 ugap2021

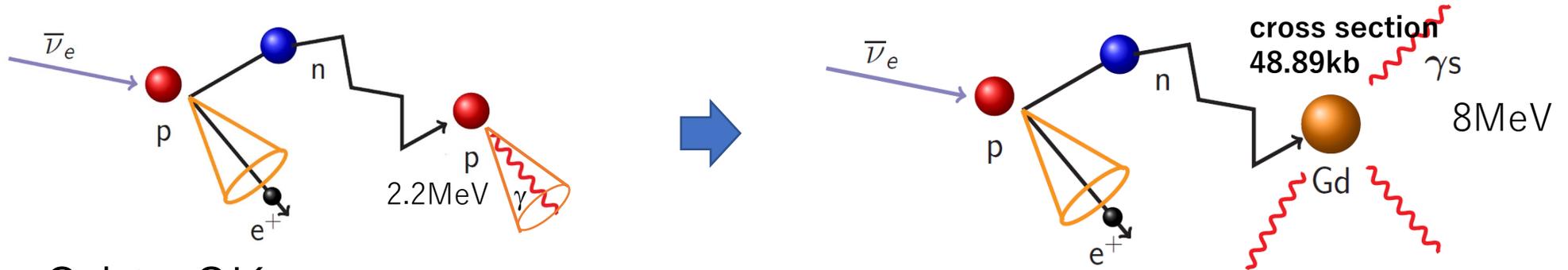
計画研究C01班の体制



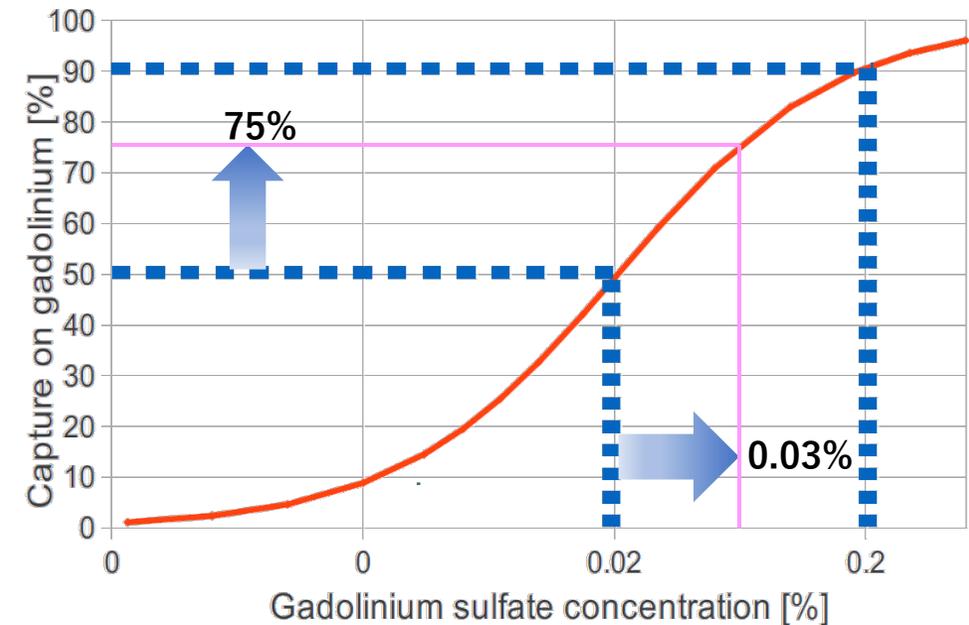
内容

- SK-Gdの目指すもの
- SKへ導入した低放射能硫酸ガドリニウムとその評価方法
- 2020年7月14日～8月17日に行ったGd導入の様子
- 導入したGd濃度の確認
- Gdによる中性子捕獲イベントの紹介
- 導入後の様子 (Dark noise、バクテリア?、LowE BG)
- Rn/Linac
- まとめ

SK-Gd project

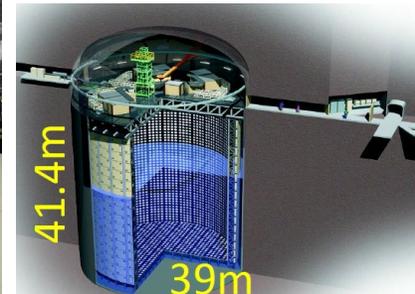
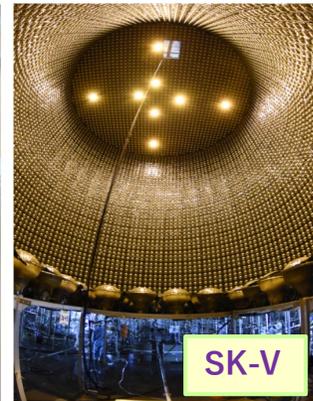
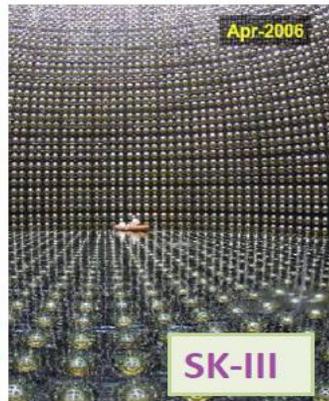
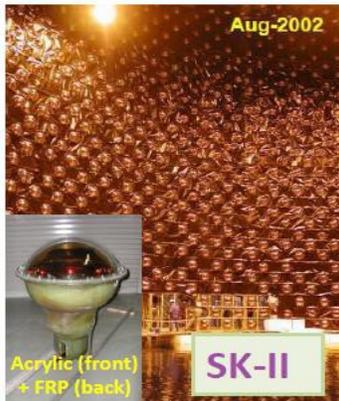
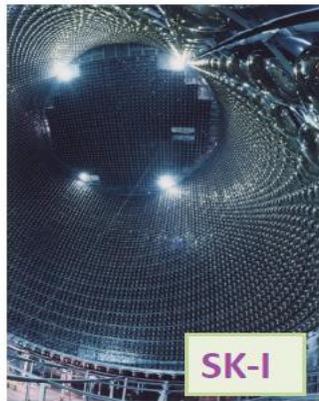
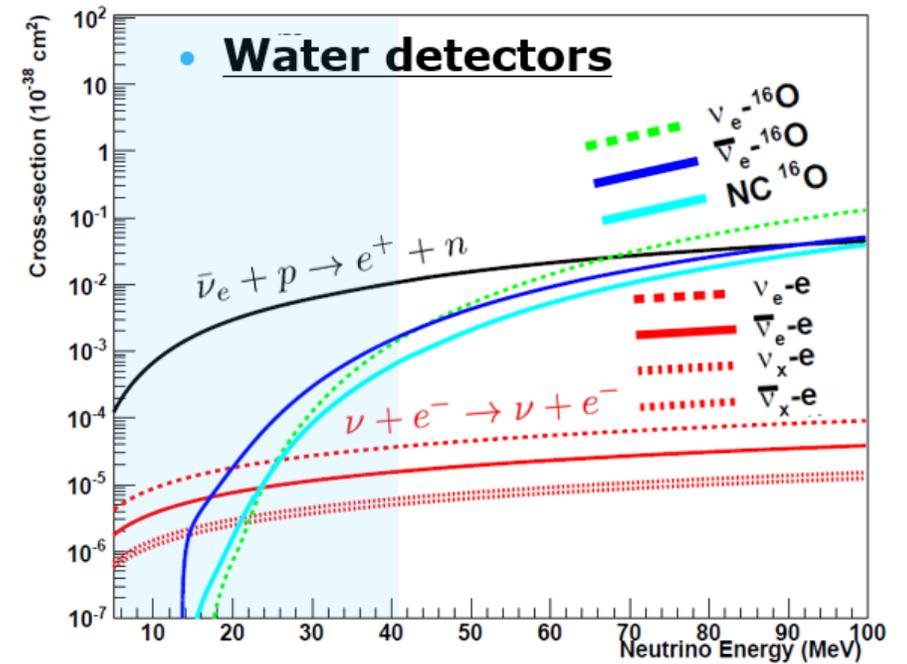


- Loading Gd to SK
 - To significantly enhance detection capability of neutrons from $\bar{\nu}$ interactions
 - 0.02% $\text{Gd}_2(\text{SO}_4)_3$ concentration for the 1st step
 - About 50% of neutron would be captured by Gd, enhancing neutron tagging efficiency by 2-3 times.
- Planned gradual increasement of Gd
 - Final target: 90% of neutron tagging
 - Aiming at 75% with this Kakenhi



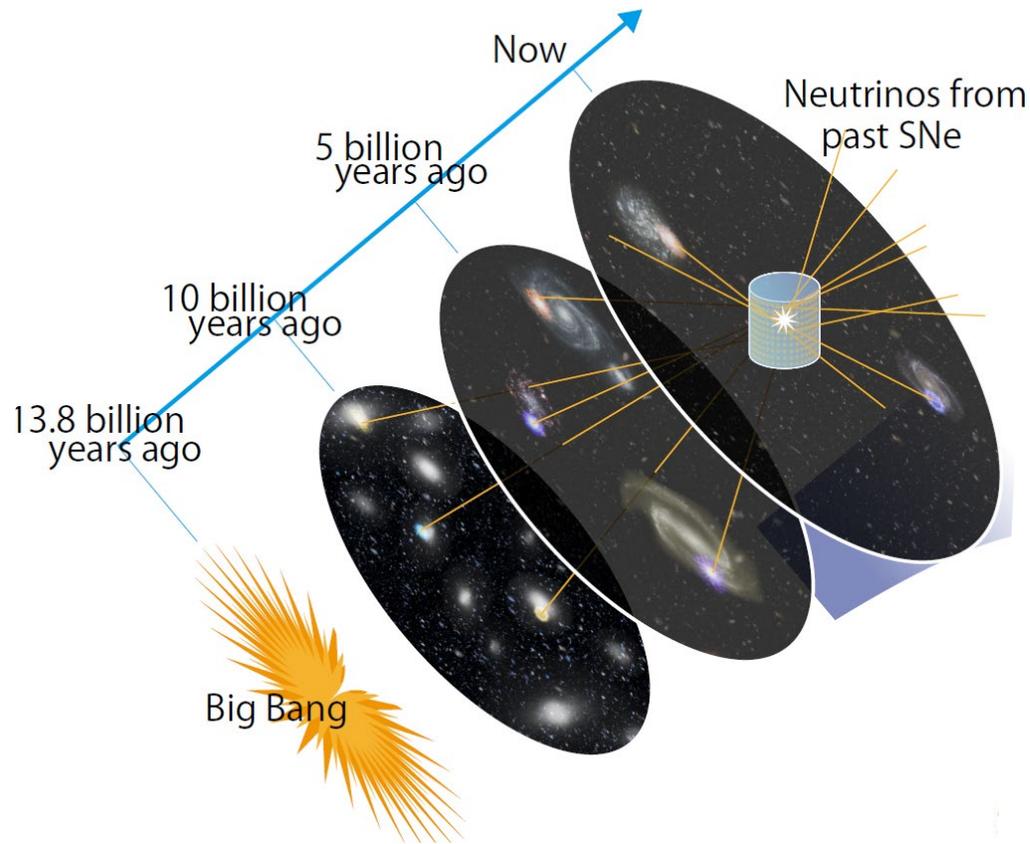
Super-Kamiokande VI

- Ring imaging Gd-doped water Cherenkov detector
 - **49468 tons of pure water with 5426kg of Gd**
 - 11129 50cm PMTs for Inner detector
- 1km (2700 mwe) underground in Kamioka
- Most sensitive to $\bar{\nu}_e$ through inverse beta decay, and the emitted neutron can be tagged with more than 50% efficiency.



Diffused Supernova Neutrino Backgrounds

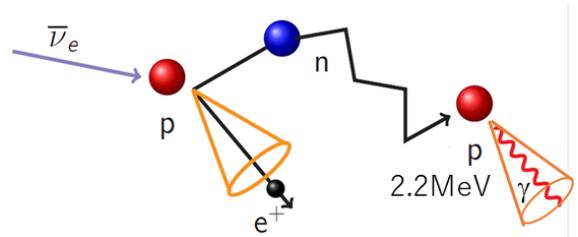
Supernova Relic Neutrino



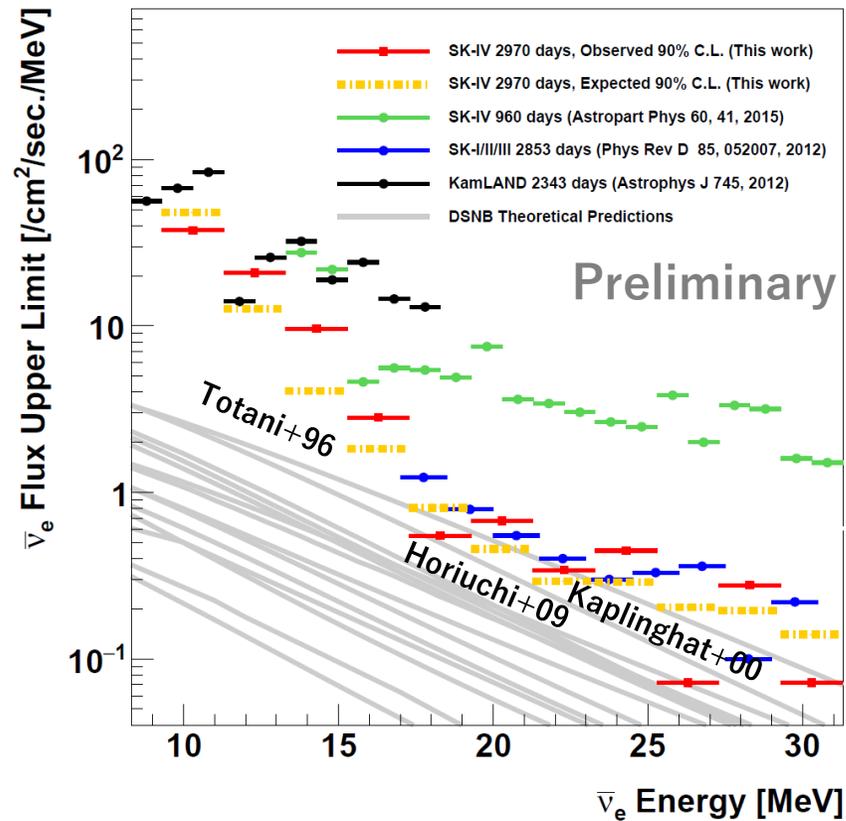
- Neutrinos produced from the past SN bursts and diffused in the current universe.
 - ~ a few SN explosions every second $\rightarrow O(10^{18})$ SNe so far in this universe
 - Can study history of SN bursts with neutrinos

$$\frac{dF_\nu}{dE_\nu} = c \int_0^{z_{\max}} R_{\text{SN}}(z) \frac{dN_\nu(E'_\nu)}{dE'_\nu} (1+z) \frac{dt}{dz} dz$$

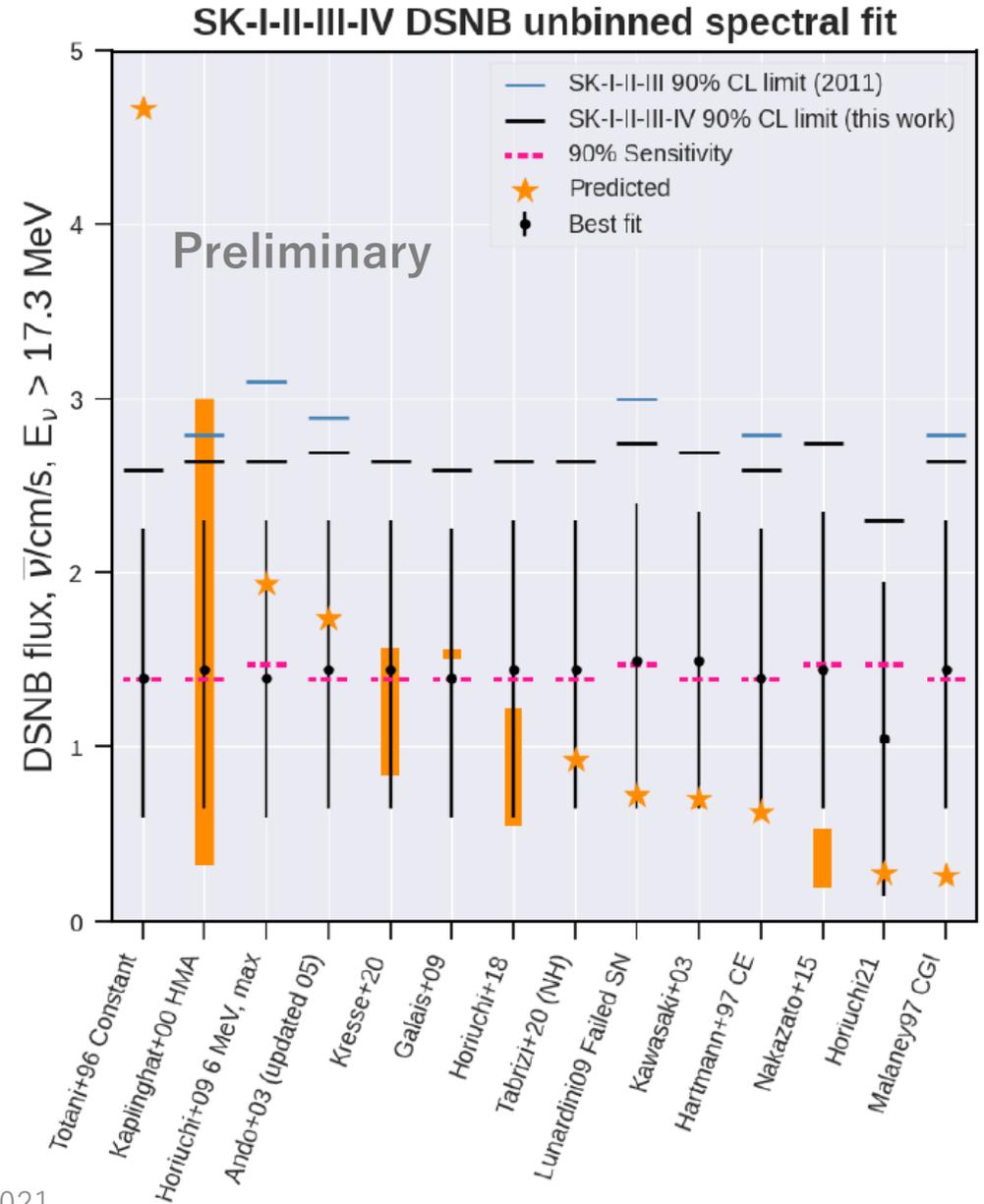
DSNB search in SK-IV



Pure water, with neutron tagging

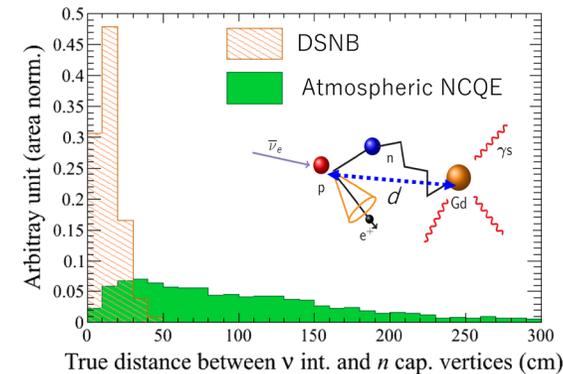
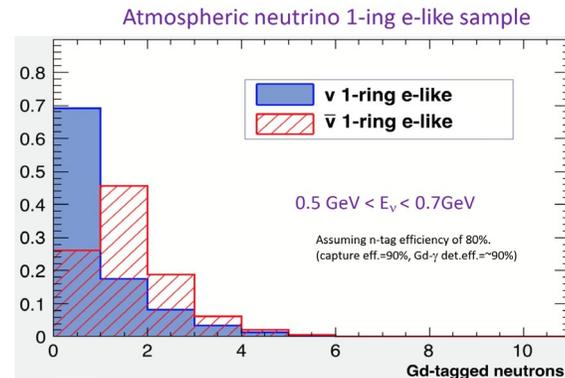
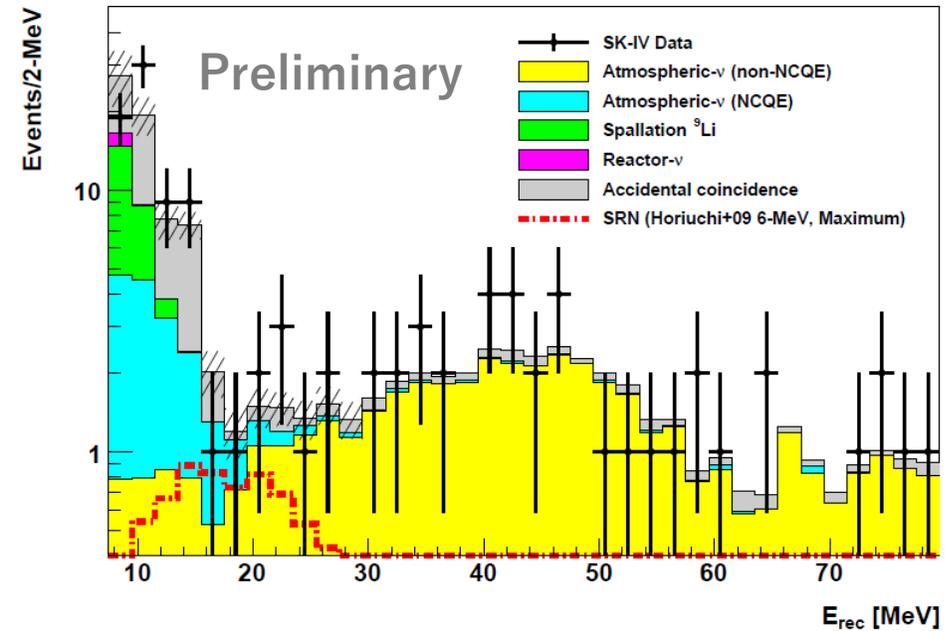
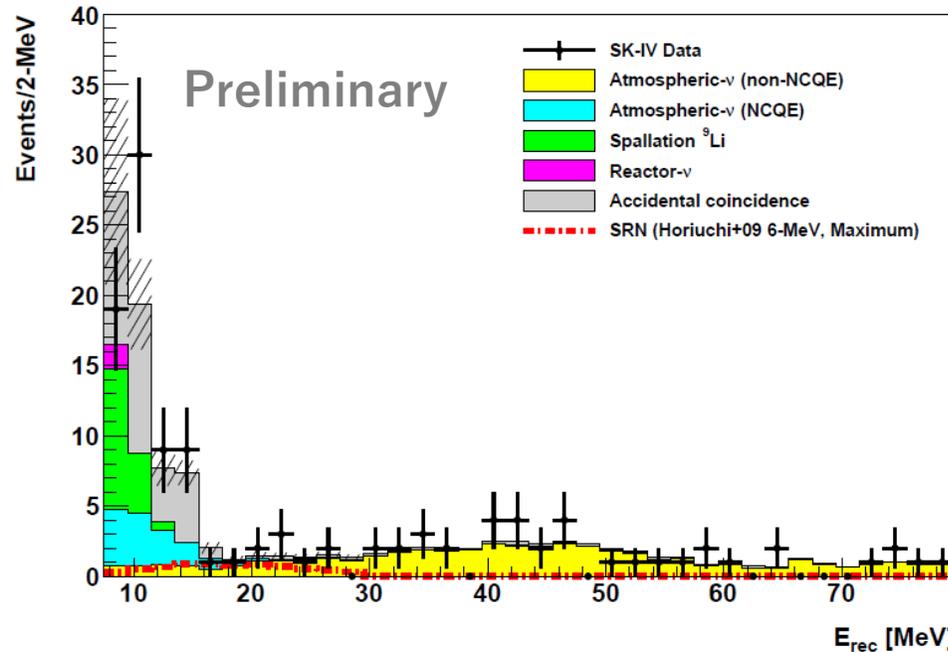


Already touched the predicted region!



DSNB search in SK-IV

Pure water, with neutron tagging



With Gd n-tag,
Neutron multiplicity cuts and
topology cuts will reduce these BG

SK-Gd sensitivity will cover many predictions !

Technologies for Low RI $Gd_2(SO_4) \cdot 8H_2O$

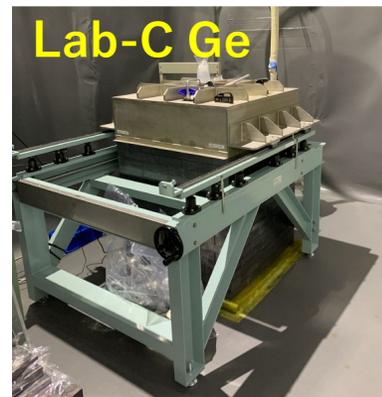
ICP-MS

- Established the analysis of ^{238}U , ^{232}Th , and ^{226}Ra at ppt level

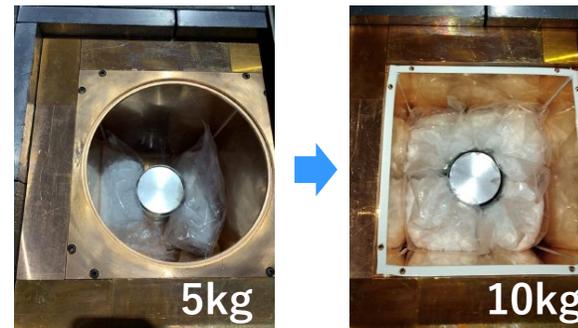
D班、公募研究 坂口さん
S.Ito, H.Ito, K. Ichimura et al.

Ge

- Sensitivity of ~ 0.2 mBq/kg was achieved by increasing the sample weight
- Sensitivity of ~ 0.5 mBq/kg was achieved by applying the Ra disk analysis method.



Enlarged sample room



Ra-disk method



Ra captured resin disk after Gd sulfate solution passed

Another new Ge is under commissioning!

Requirement for 0.1%Gd-loading

Radioactive chain	Part of the chain	SRN (mBq/kg)	Solar-v(mBq/kg)
^{238}U	^{238}U	< 5	-
	^{226}Ra	-	< 0.5
^{232}Th	^{228}Ra	-	< 0.05
	^{228}Th	-	< 0.05
^{235}U	^{235}U	-	< 30
	$^{227}Ac/^{227}Th$	-	< 30

$^{238}U < 0.5$ mBq/kg → 400 ppt
 $^{232}Th < 0.05$ mBq/kg → 13 ppt

N.B. We don't have methods to measure 0.05mBq/kg of ^{228}Ra

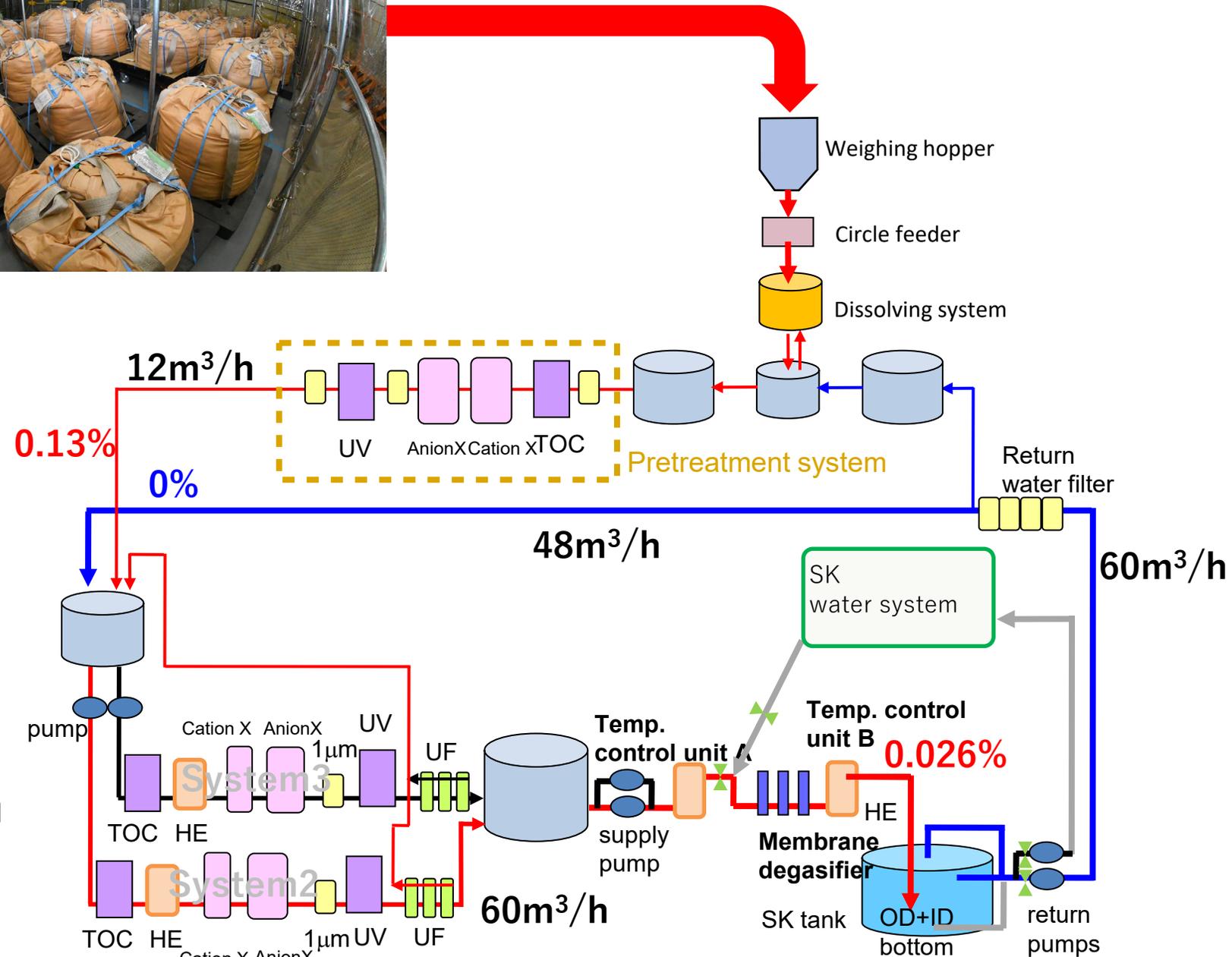
13tons of $Gd_2(SO_4) \cdot 8H_2O$



- Ge detected Th-chain RI $\sim 1\text{mBq/kg}$ level in the latter half of the products.
- After Ra extraction measurement, it turned out they are ^{228}Ra i.e. ^{232}Th itself was removed during the production process, but ^{228}Ra remains.
- The quality of the feedstock fluctuated greatly and could not be controlled.
- For the next Gd-loading, extra processes will be applied to be independent from the quality of feedstocks

➡ The price of $Gd_2(SO_4) \cdot 8H_2O$ gets increased

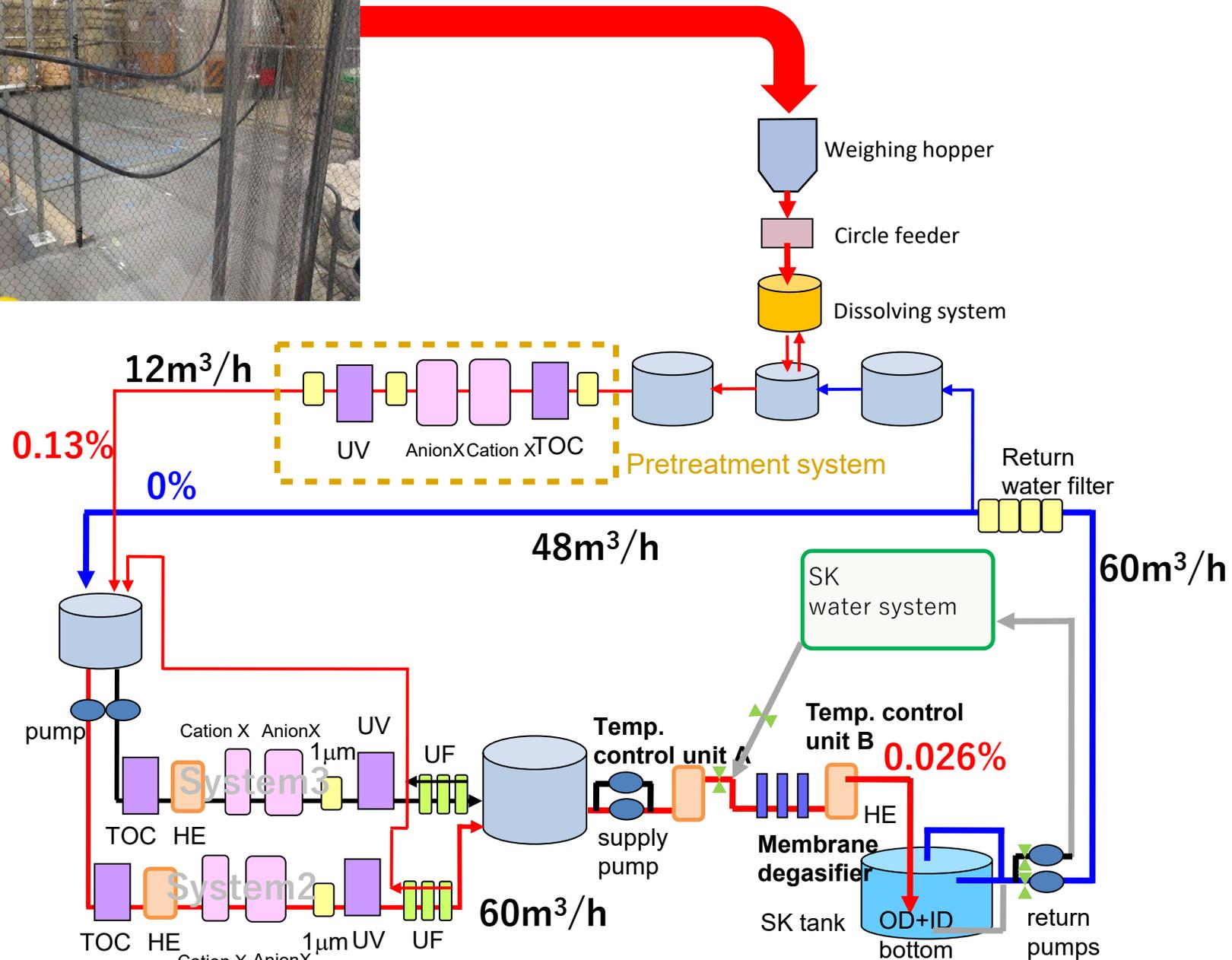
Gd-loading procedure & systems



Started on July 14

- 0.026% loading with 60 m³/h
- Takes 35 days

Gd-loading procedure & systems

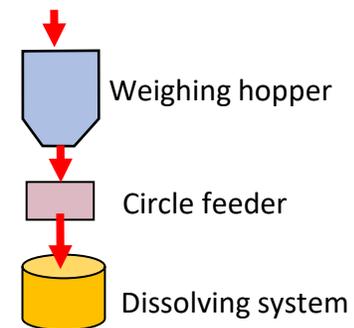


Finished on Aug. 17

- 0.026% loading with 60 m³/h
- Takes 35 days



Direct supply with shovels and buckets



Just after adding 8.2kg of $Gd_2(SO_4)_3 \cdot 8H_2O$

10 minutes later

Pictures

One sequence:
8.2kg(→8.7kg) of powder in 768L
30minues/cycle



The record

On Aug 17
finished loading 13212 kg of
“ $\text{Gd}_2(\text{SO}_4)_3 \cdot 8\text{H}_2\text{O} + 2.5\% \text{ water}$ ”
= 12884 kg of $\text{Gd}_2(\text{SO}_4)_3 \cdot 8\text{H}_2\text{O}$

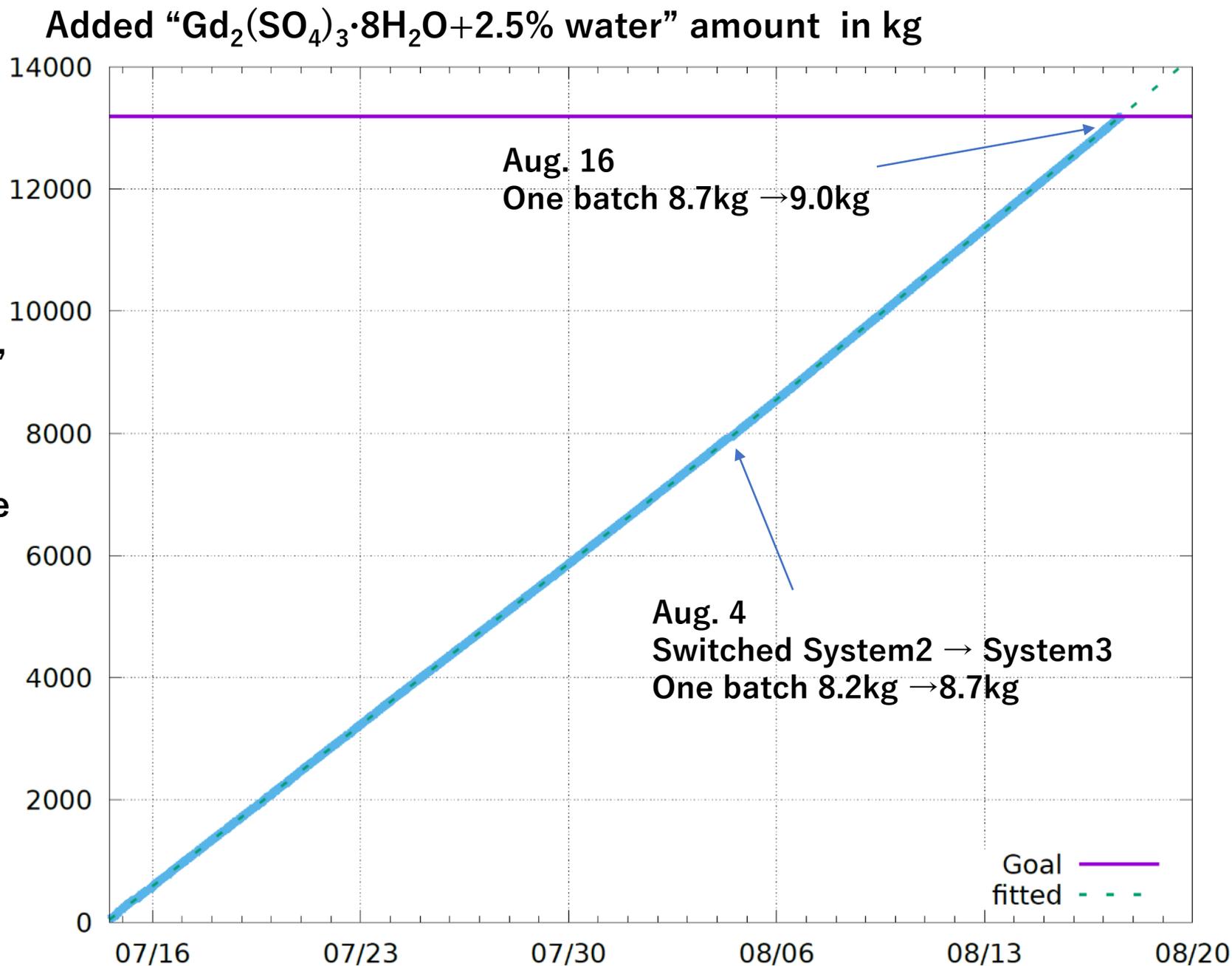
The water in the tank and the
water recirculation system
= 49468000 kg

I mean the pure water amount in the system, so
49468000m3. Anyway both 12884./49468000 and
12884./(49468000+12884) give you 0.000260.

= 0.0260% $\text{Gd}_2(\text{SO}_4)_3 \cdot 8\text{H}_2\text{O}$

= 0.0210% $\text{Gd}_2(\text{SO}_4)_3$

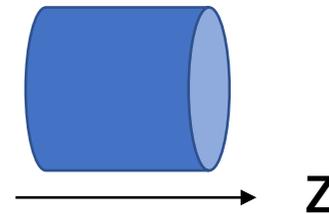
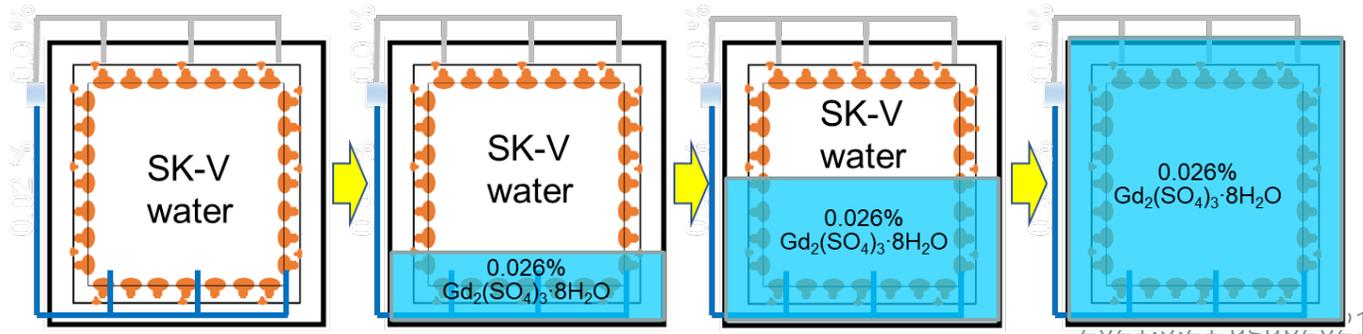
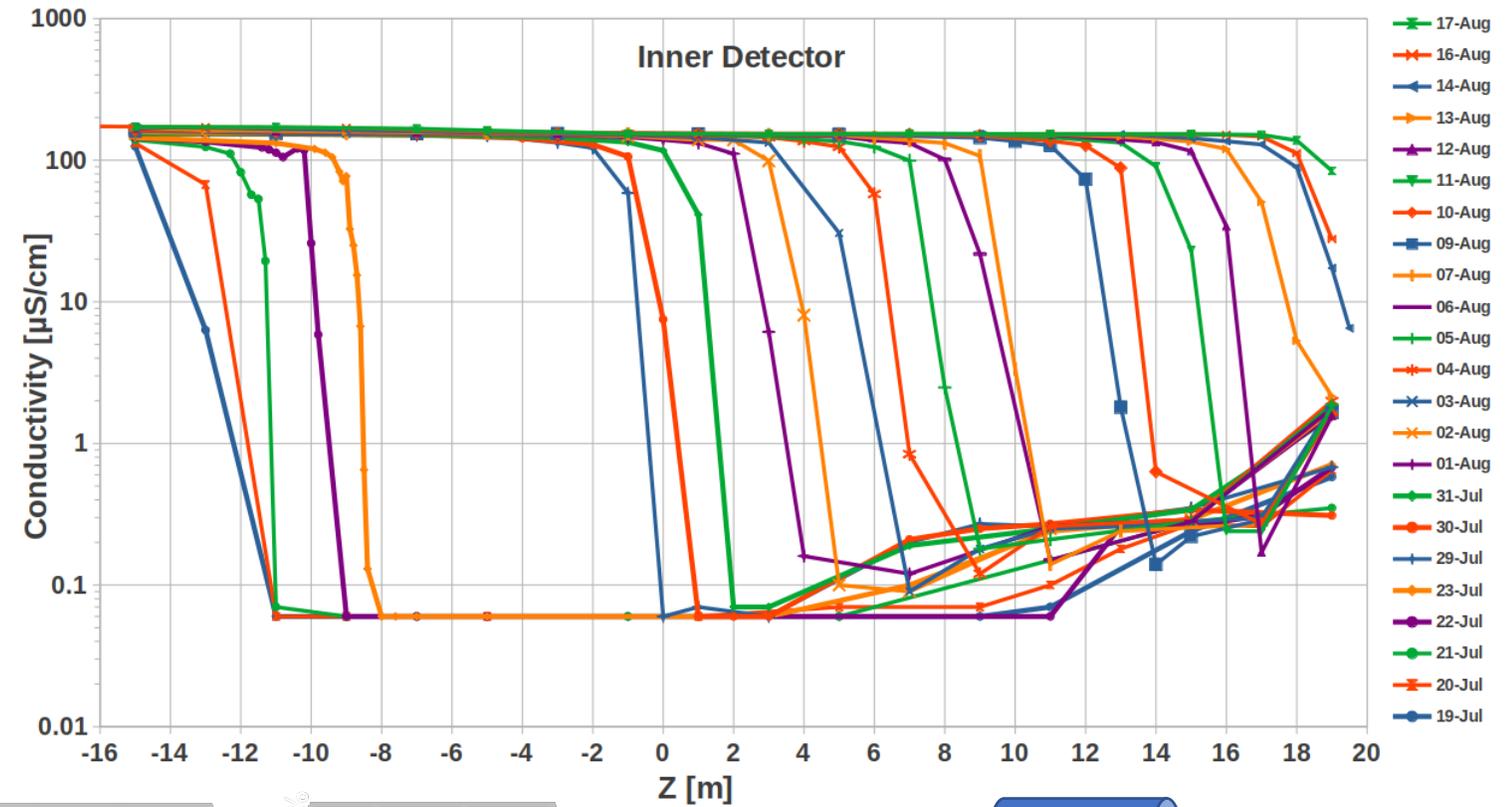
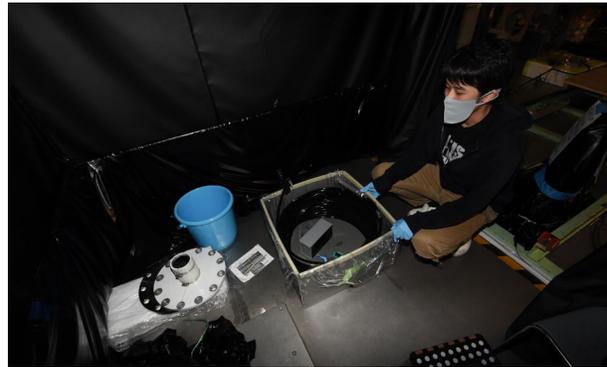
= 0.0110% Gd



Gd concentration checked by Conductivity

Water sampled directly from various positions in the tank by insertion of tube

LI.Marti

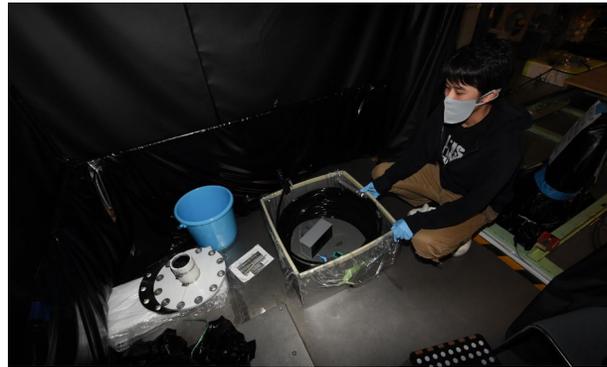


Observed the bottom-up accumulation

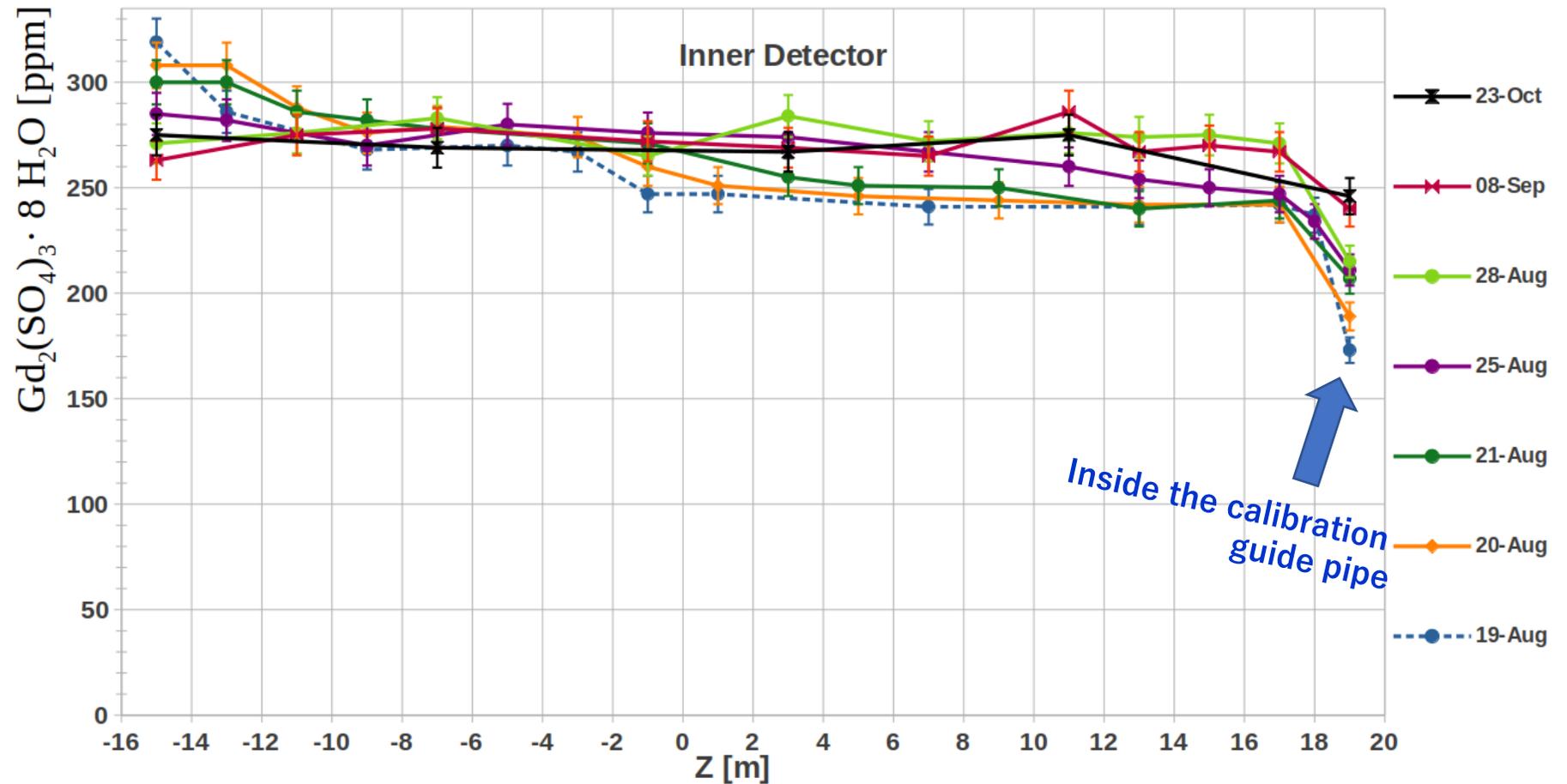
Direct concentration measurement after loading

Atomic Absorption Spectrometer

LI.Marti



After the Gd-loading, the concentration was getting uniform



Gd concentration and Neutron capture time

Number of captures in Δt

$$\frac{dN_n(t)}{dt} \Delta t \propto - (n_{Gd} \sigma_{Gd} + n_p \sigma_p) v_n \Delta t N_n(t)$$

$N_n(t)$: number of neutron

v_n : neutron velocity

$n_{Gd} n_p$: number of nuclei in unit volume

$\sigma_{Gd} \sigma_p$: capture cross section of Gd

Once neutrons are thermalized, v_n becomes \sim constant

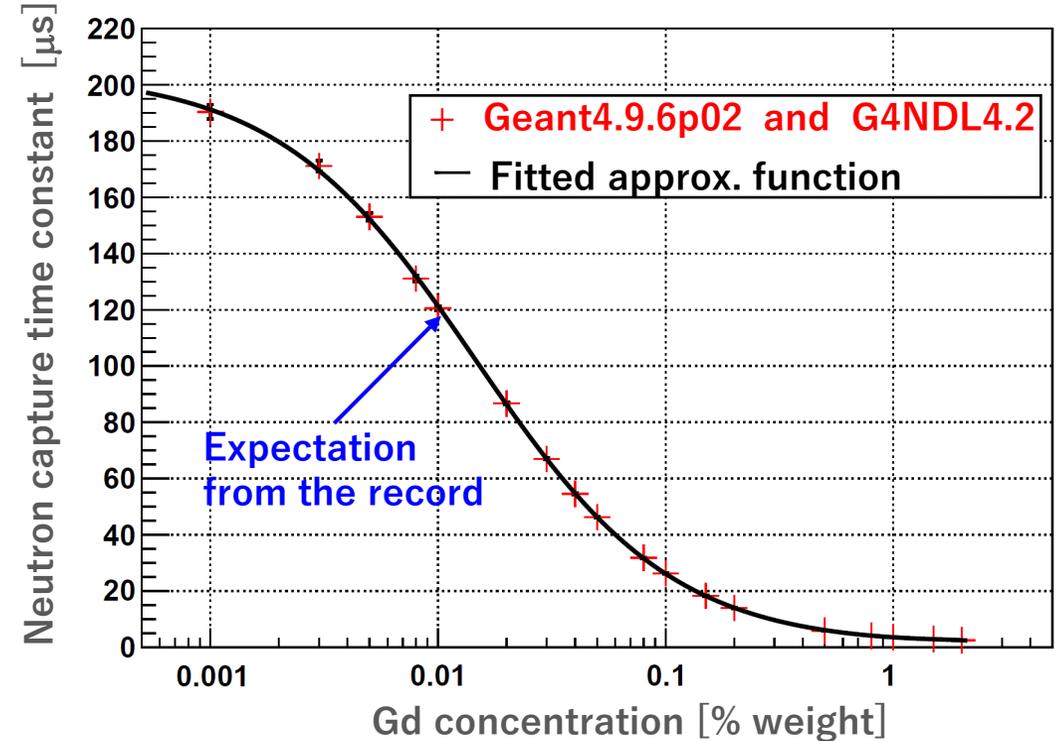
$$N_n(t) \propto \exp(- (n_{Gd} \sigma_{Gd} + n_p \sigma_p) t)$$

$$\tau \propto \frac{1}{n_{Gd} \sigma_{Gd} + n_p \sigma_p}$$

Gd concentration



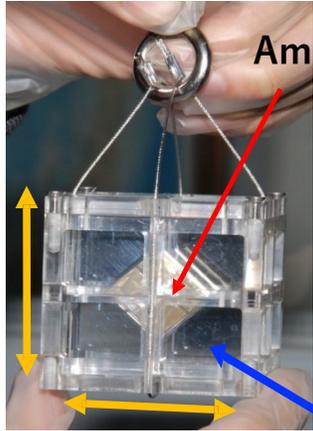
T.Yano



Approx. function

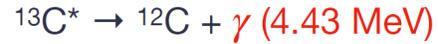
$$c_{Gd} \approx \left(\frac{8.19}{\tau [\mu s]} - 0.0371 \right) \times \frac{1}{310} \times 100 [\%]$$

Am/Be + BGO neutron source



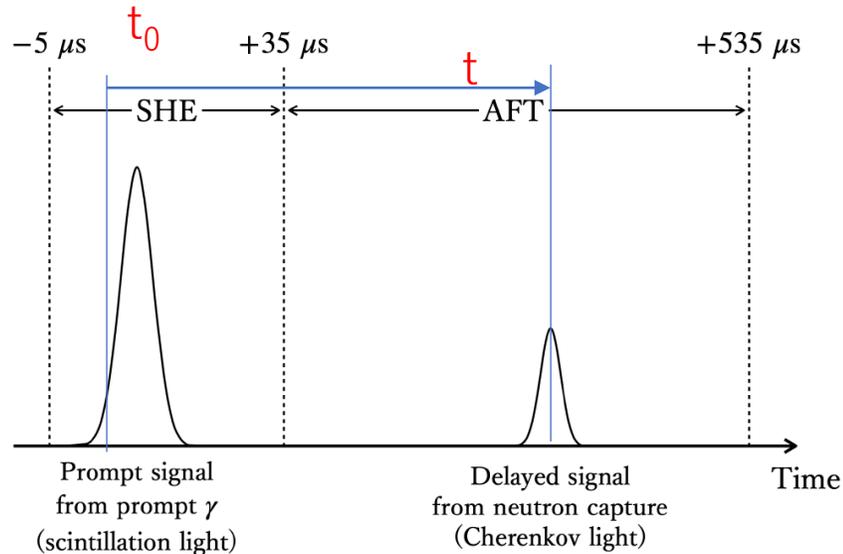
Am/Be source

100~200 neutrons/s



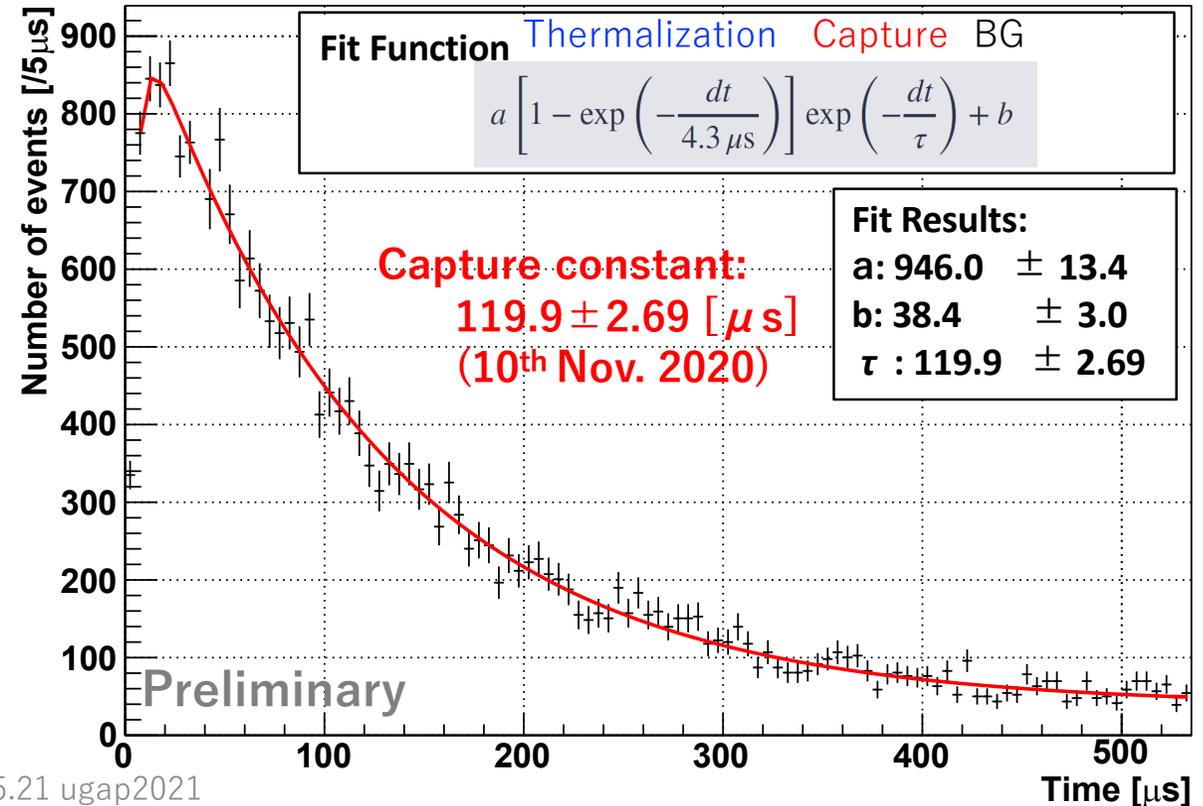
5cm

8 BGO Crystals



- The trigger is the scintillation of 4.4 MeV γ emitted from the Am/Be source simultaneously with the neutrons in the BGO crystal. (SHE trigger threshold 64 hits in 200ns).
- All the PMT hits from -5 to 535 μs before and after the trigger are stored and searched for neutron signals. (sub trigger threshold 30 hits)

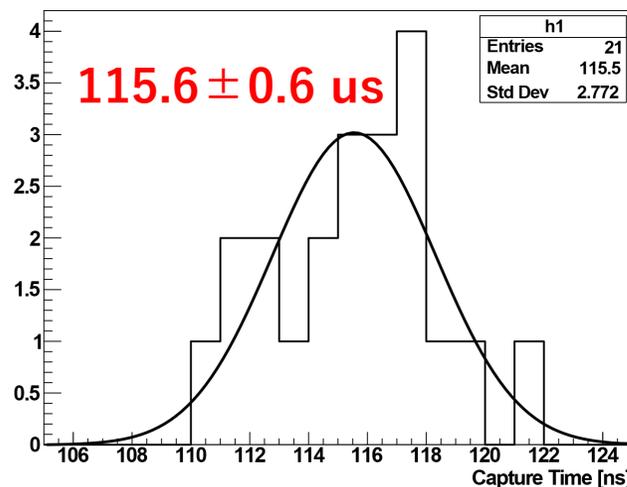
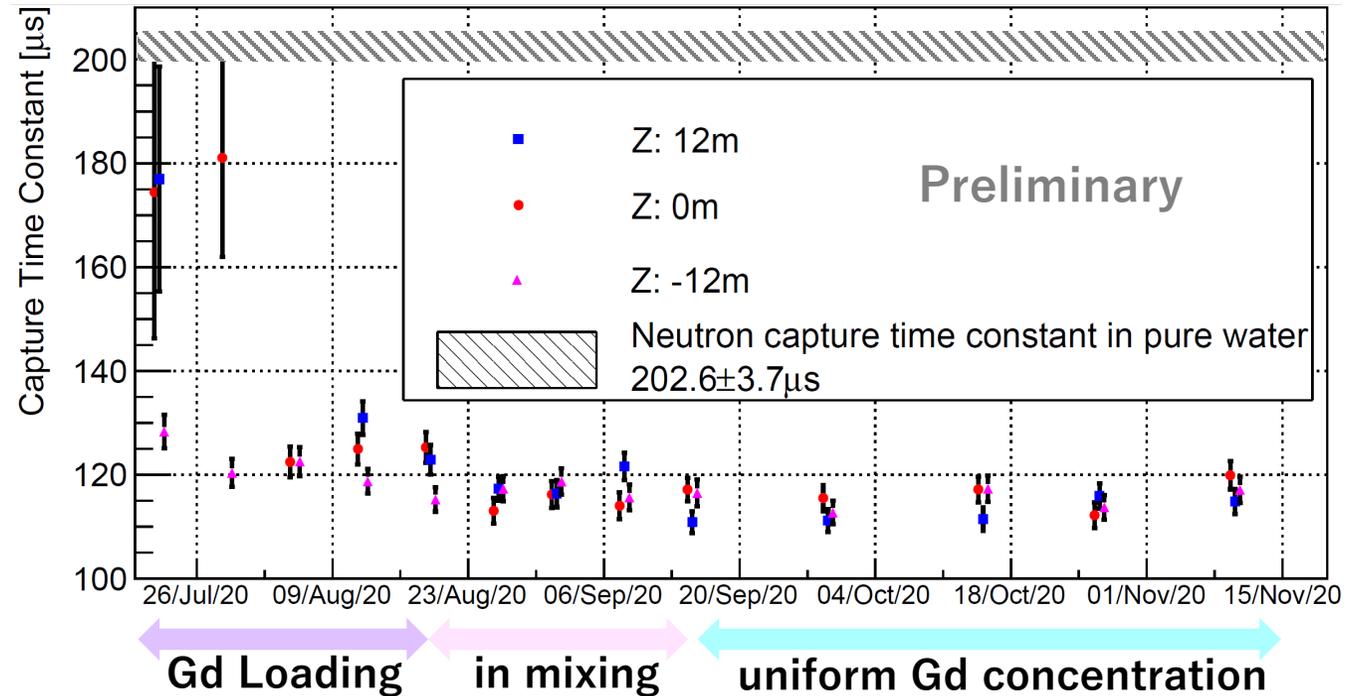
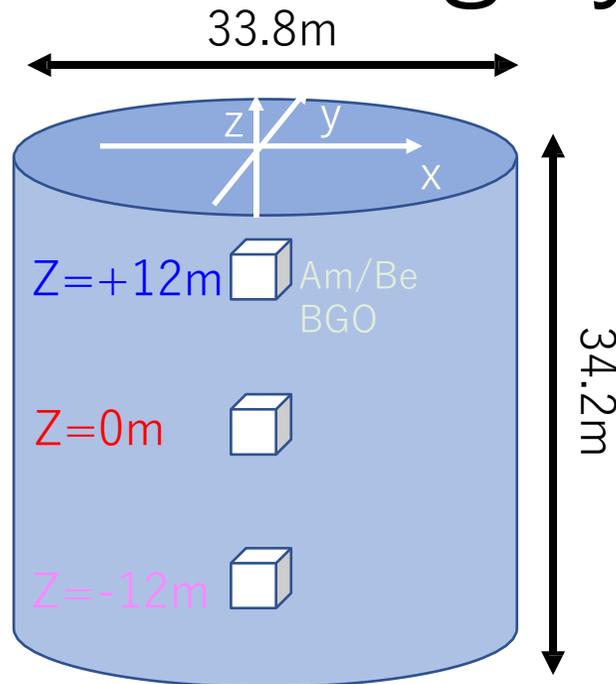
T.Yano



Gd monitoring by Am/Be

every 2 weeks

T.Yano



After September
all Z positions

Derived Gd concentration
 $109.1 \pm 1.2 \text{ ppm}$

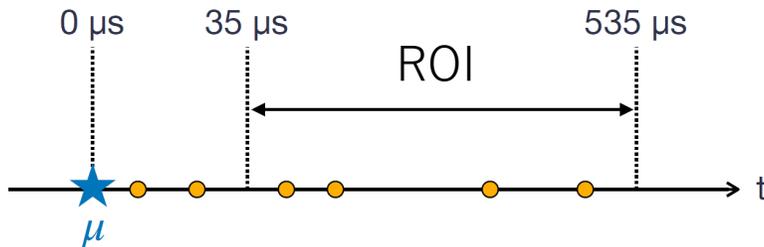
consistent with the record

Spallation neutron by muon

• Event selection

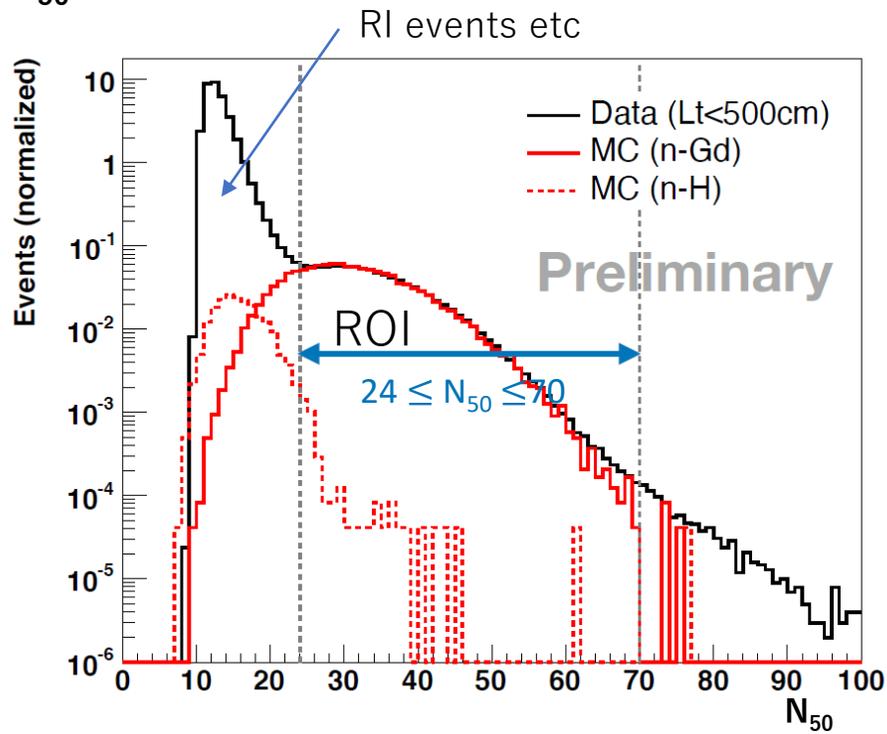
M.Shinoki

Timing selection



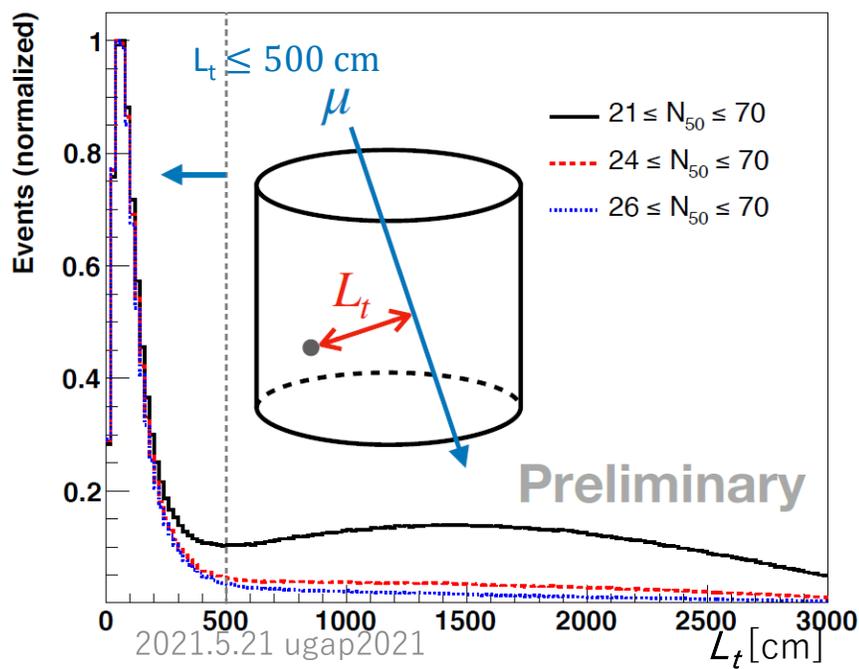
- Michel decay-e $\sim 2.2\mu\text{s}$
- Neutron thermalization $\sim 4.3\mu\text{s}$
- PMT after pulses $10\sim 20\mu\text{s}$

N_{50} selection



Distance to μ track selection

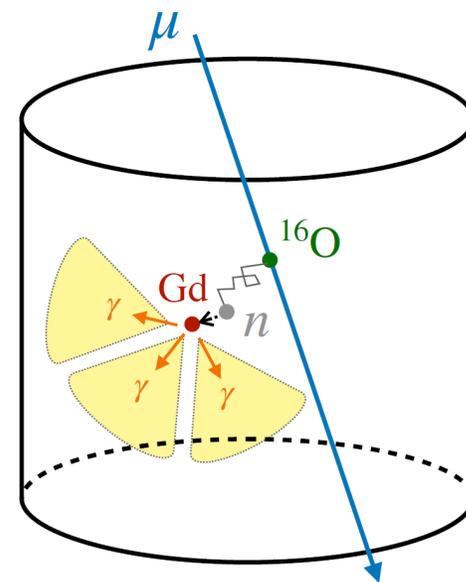
Neutron capture occurs near the muon track



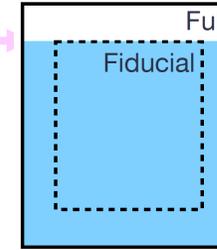
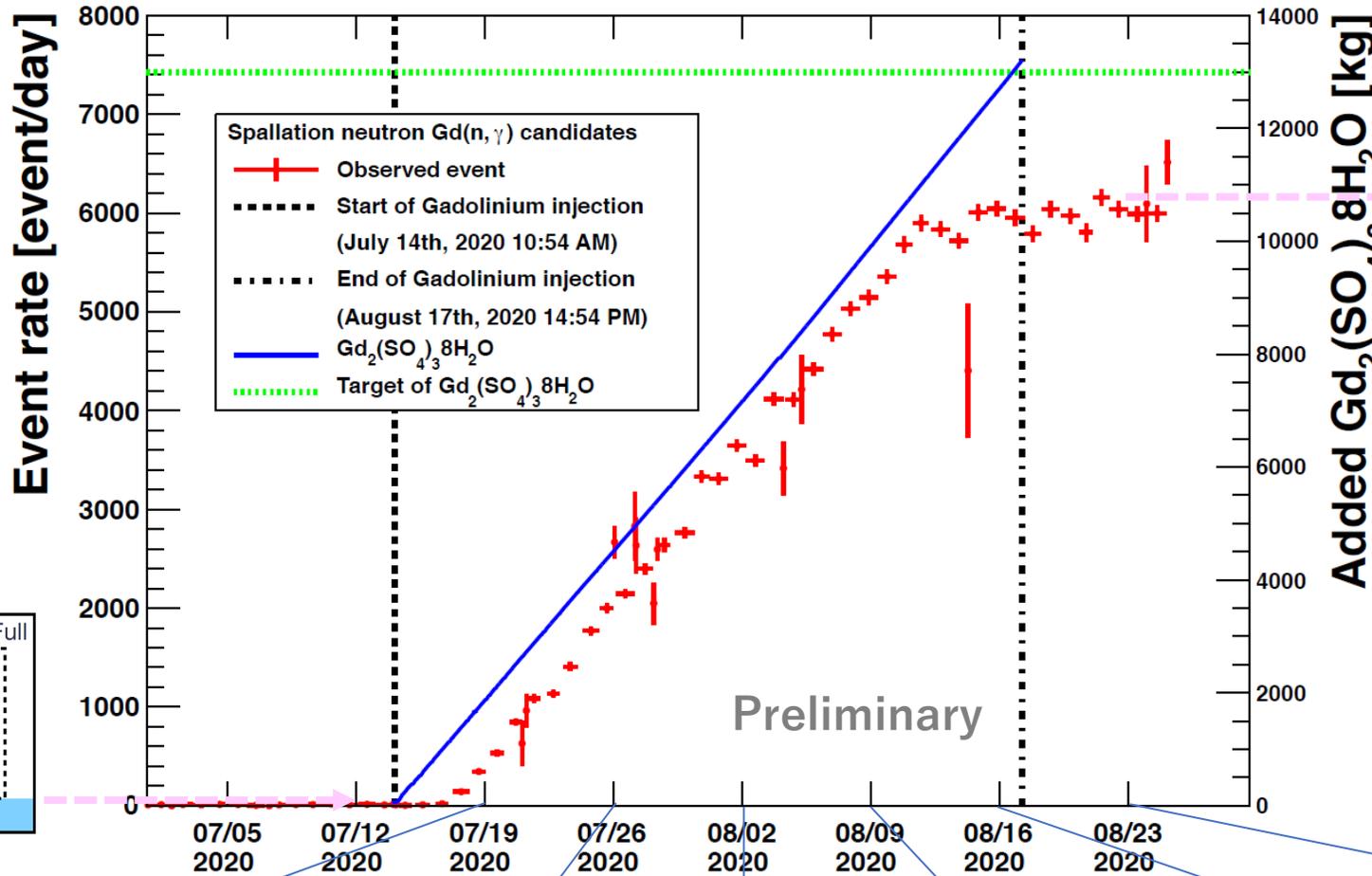
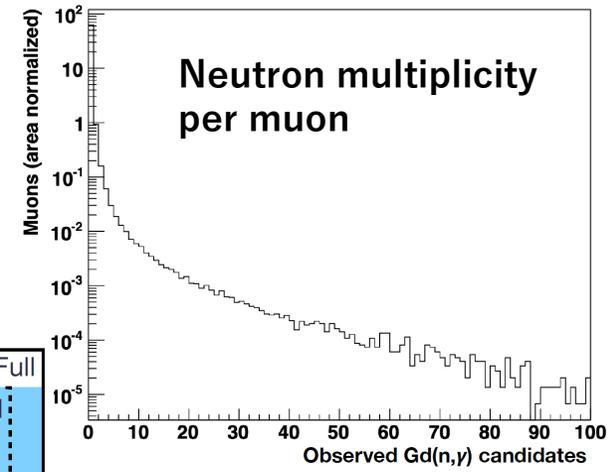
List of spallation products

Isotope	Half-life (s)	Decay mode	Yield (total) ($\times 10^{-7} \mu^{-1} \text{g}^{-1} \text{cm}^{-2}$)	Yield ($E > 3.5 \text{ MeV}$) ($\times 10^{-7} \mu^{-1} \text{g}^{-1} \text{cm}^{-2}$)	Primary process
n			2030		
¹⁸ N	0.624	β^-	0.02	0.01	¹⁸ O(n,p)
¹⁷ N	4.173	β^-n	0.59	0.02	¹⁸ O(n,n+p)
¹⁶ N	7.13	$\beta^- \gamma$ (66%), β^- (28%)	18	18	(n,p)
¹⁶ C	0.747	β^-n	0.02	0.003	(π^- ,n+p)
¹⁵ C	2.449	$\beta^- \gamma$ (63%), β^- (37%)	0.82	0.28	(n,2p)
¹⁴ B	0.0138	$\beta^- \gamma$	0.02	0.02	(n,3p)
¹³ O	0.0086	β^+	0.26	0.24	(μ^- ,p+2n+ μ^- + π^-)
¹³ B	0.0174	β^-	1.9	1.6	(π^- ,2p+n)
¹² N	0.0110	β^+	1.3	1.1	(π^+ ,2p+2n)
¹² B	0.0202	β^-	12	9.8	(n, α +p)
¹² Be	0.0236	β^-	0.10	0.08	(π^- , α +p+n)
¹¹ Be	13.8	β^- (55%), $\beta^- \gamma$ (31%)	0.81	0.54	(n, α +2p)
¹¹ Li	0.0085	β^-n	0.01	0.01	(π^+ ,5p+ π^+ + π^0)
⁹ C	0.127	β^+	0.89	0.69	(n, α +4n)
⁹ Li	0.178	β^-n (51%), β^- (49%)	1.9	1.5	(π^- , α +2p+n)
⁸ B	0.77	β^+	5.8	5.0	(π^+ , α +2p+2n)
⁸ Li	0.838	β^-	13	11	(π^- , α +2H+p+n)
⁸ He	0.119	$\beta^- \gamma$ (84%), β^-n (16%)	0.23	0.16	(π^- , ³ H+4p+n)
¹⁶ O			351		(γ ,n)
¹⁵ N			773		(γ ,p)
¹⁴ O			13		(n,3n)
¹⁴ N			295		(γ ,n+p)
¹⁴ C			64		(n,n+2p)
¹³ N			19		(γ , ³ H)
¹³ C			225		(n, ² H+p+n)
¹² C			792		(γ , α)
¹¹ C			105		(n, α +2n)
¹¹ B			174		(n, α +p+n)
¹⁰ C			7.6		(n, α +3n)
¹⁰ B			77		(n, α +p+2n)
¹⁰ Be			24		(n, α +2p+n)
⁹ Be			38		(n,2 α)
sum			3015	50	

S.Li and J.Beacom, Phys. Rev. C 89, 045801 (2014)



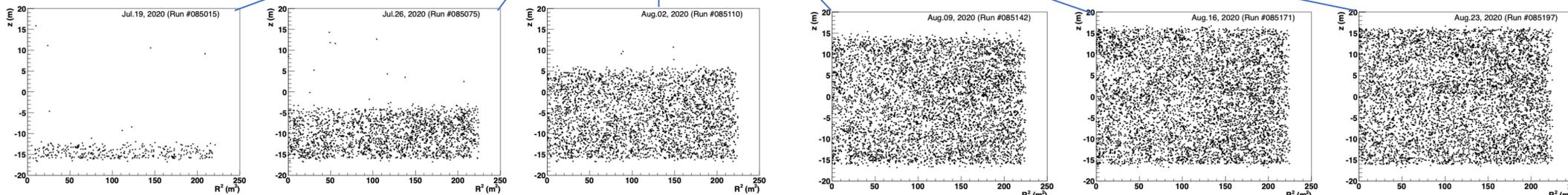
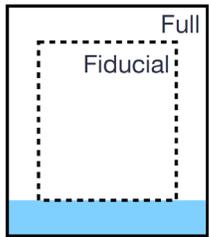
Neutron event rates vs. Gd-loading



Neutron yields in water is under calculation

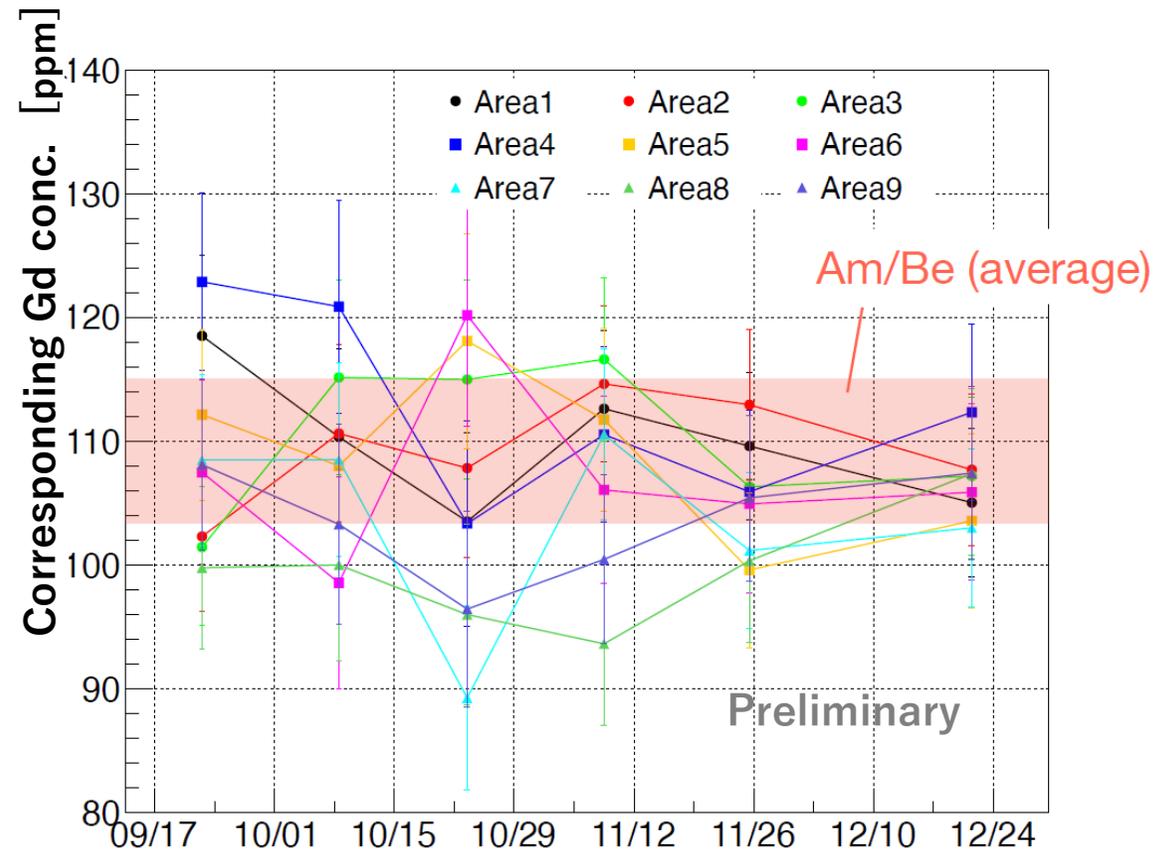
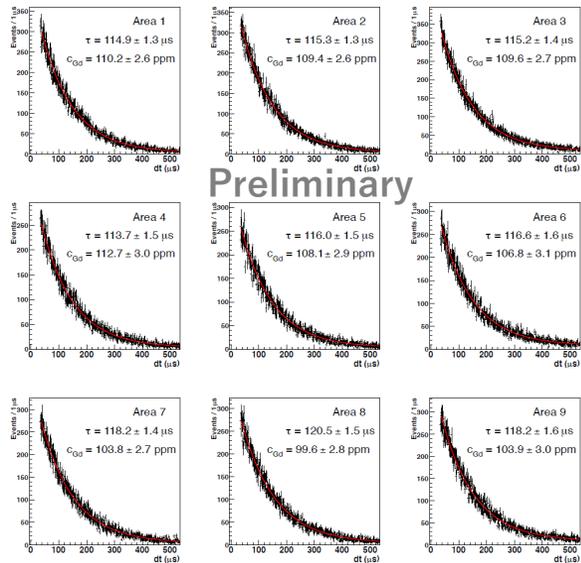
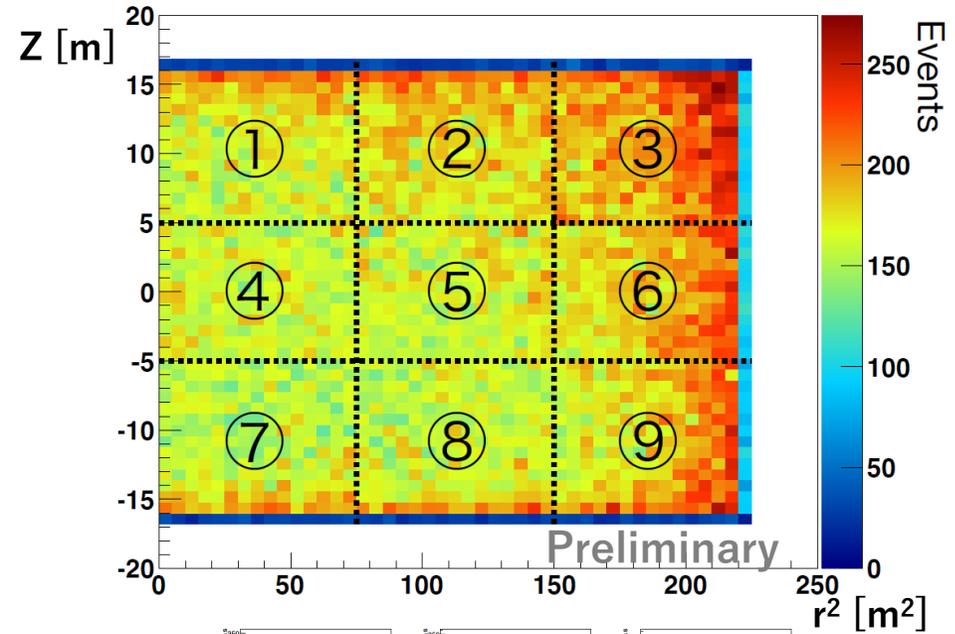
$$Y_n = \frac{N_n}{N_\mu L_\mu \rho V_{corr}}$$

- N_n : The number of neutrons produced by cosmic-ray muons
- N_μ : The number of selected muons
- L_μ : The average path length of muons
- ρ : The density of Gd water
- V_{corr} : Fiducial volume / Full volume



Gd monitoring by spallation neutron

M.Shinoki



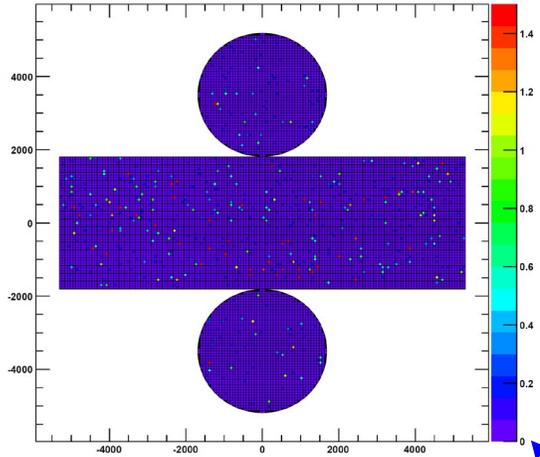
The position dependence has been decreased.

The Gd concentration has become uniform throughout the tank.

Dark noise issue

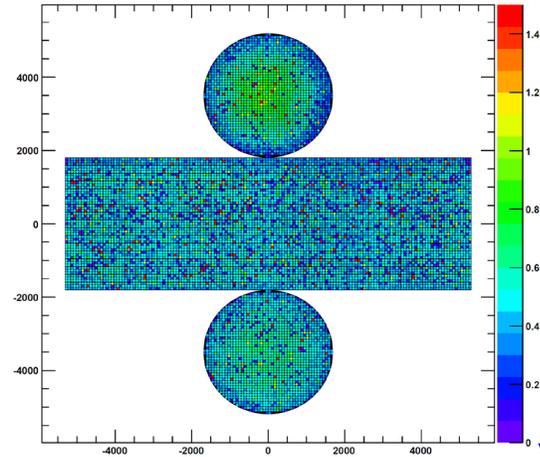
Jul.12.2020

Δ DR(Run82912-82911): Start Sun Jul 12 12:39:49 2020 2021-03-22 00:12:44



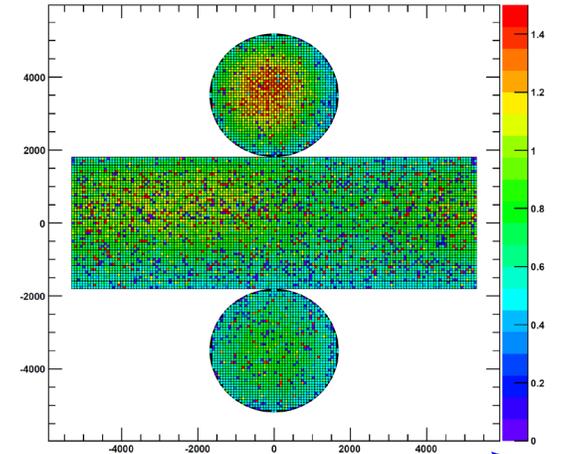
Sep.30.2020

Δ DR(Run85309-82911): Start Wed Sep 30 16:57:51 2020 2021-03-22 00:15:14

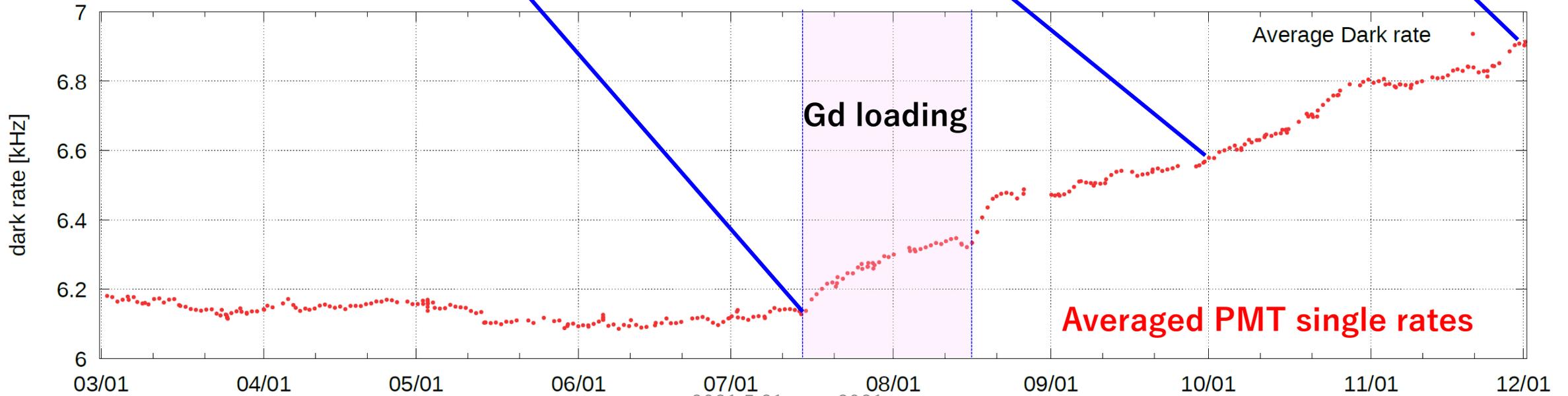


Nov.26.2020

Δ DR(Run85466-82911): Start Thu Nov 26 15:56:04 2020 2021-03-22 00:17:19



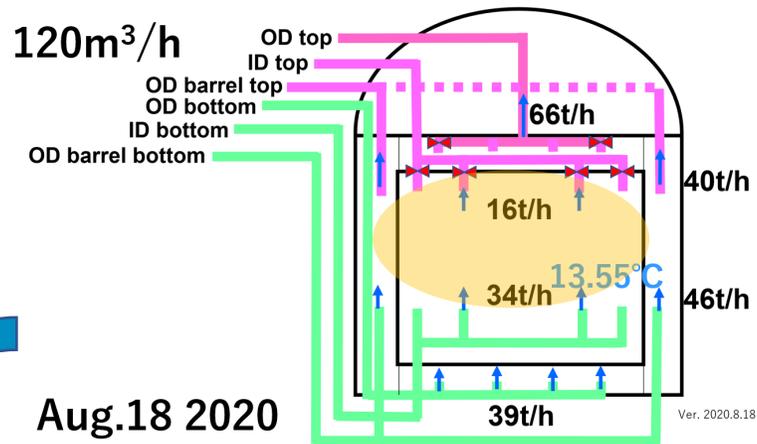
Increased noise rate [kHz]



Water stagnation and flow change

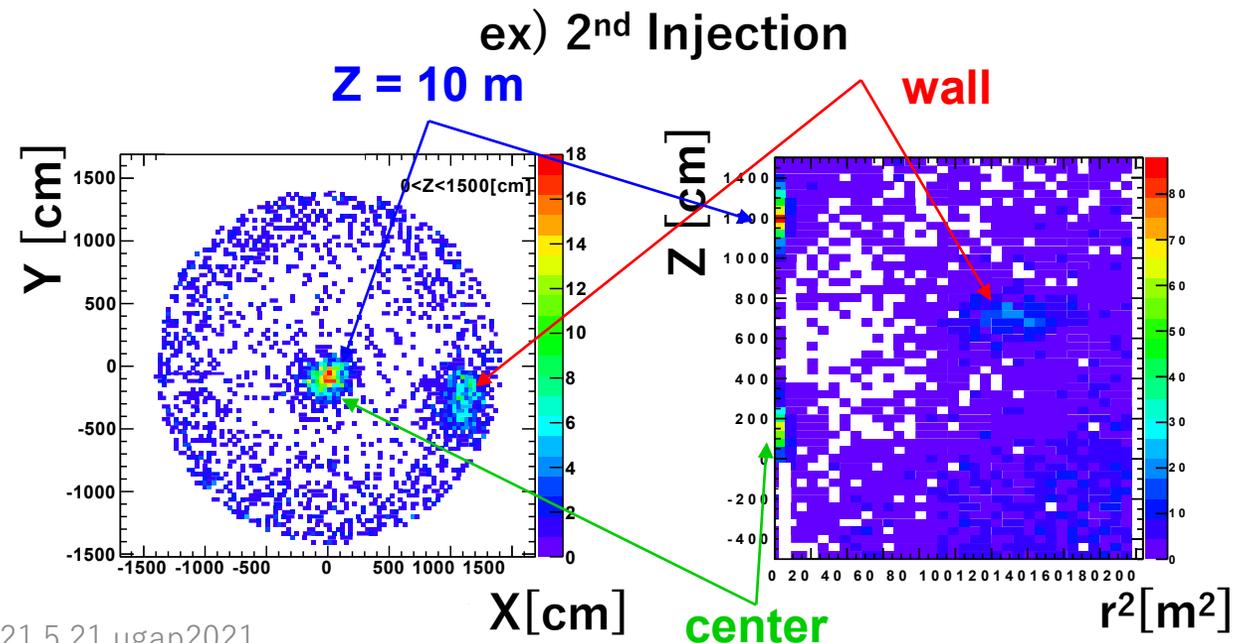
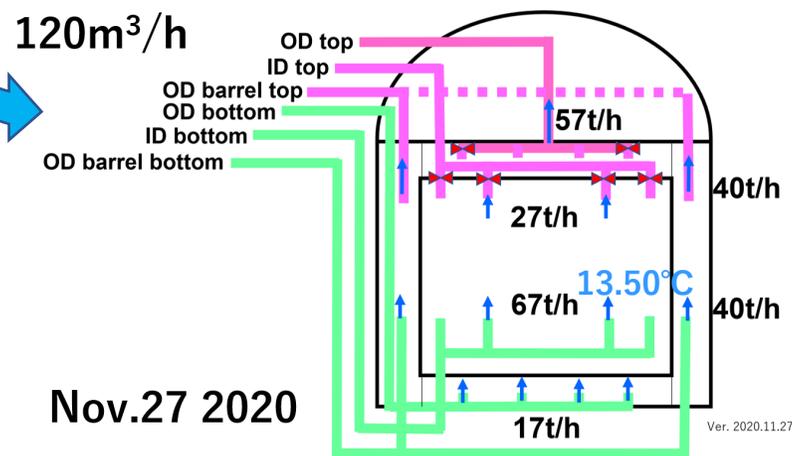
Y.Kanemura

- Water stagnation may cause the noise
- Forced water replacement by changing ID/OD flow balance and lowering supply water temperature



To see the effect ~10Bq Rn injected before and after the change

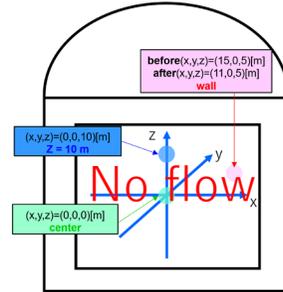
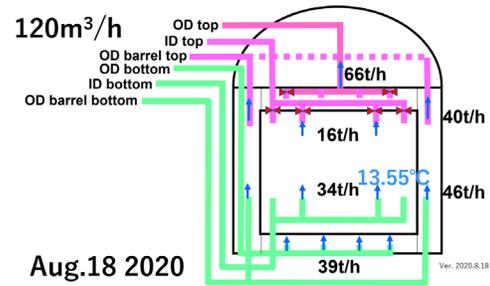
2020.11.7 : 1st Rn injection
2020.11.27: Change the water flow
2020.12.8: 2nd Rn injection



Flow in Z direction

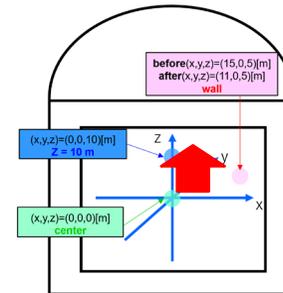
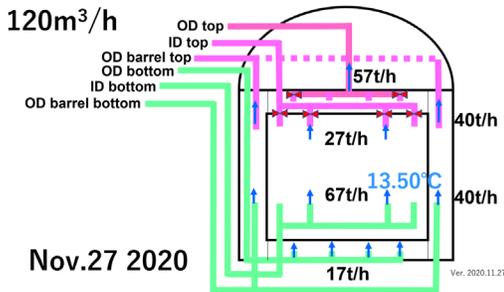
Y.Kanemura

Before

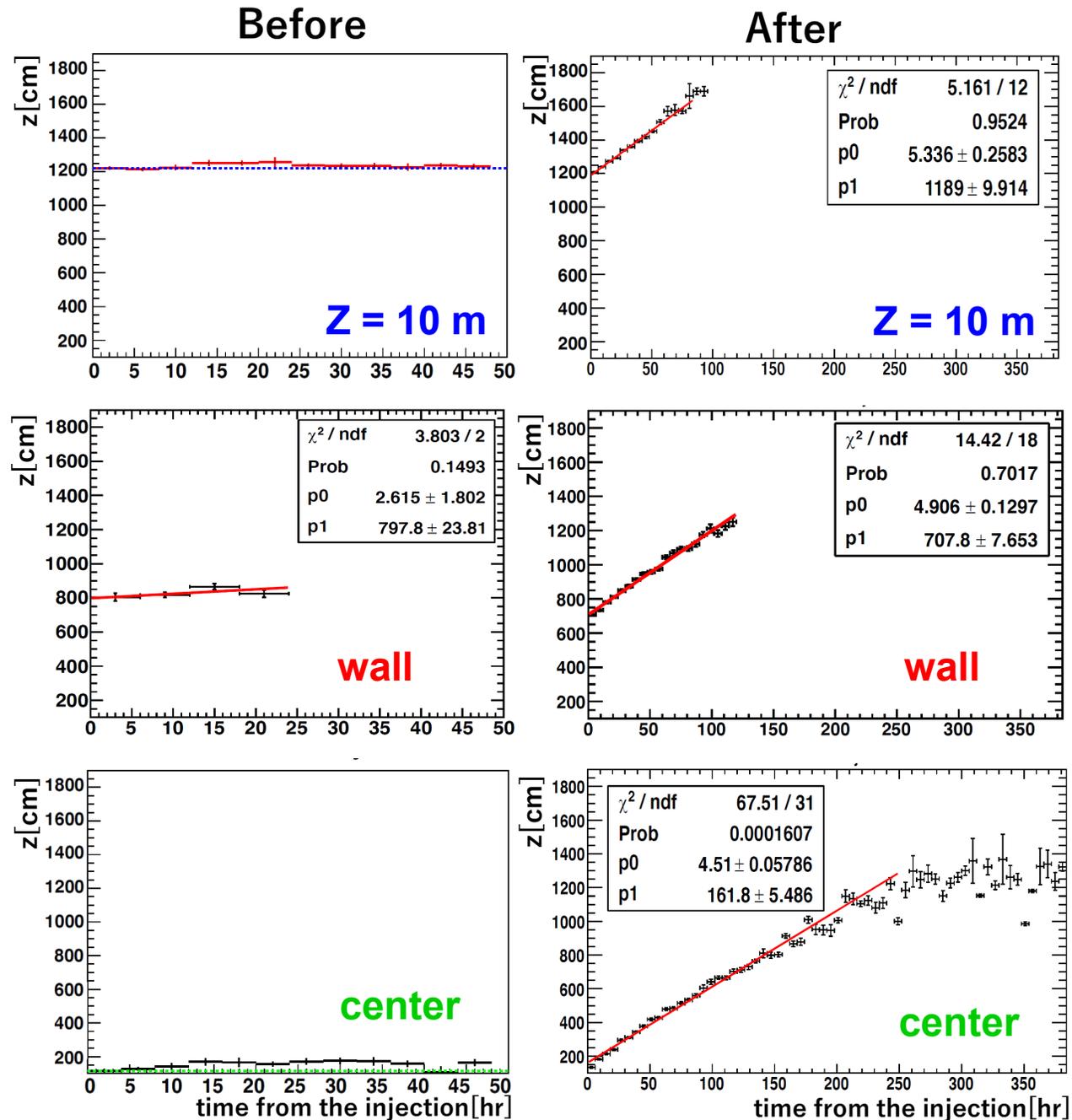


There was little water flowing → stagnation

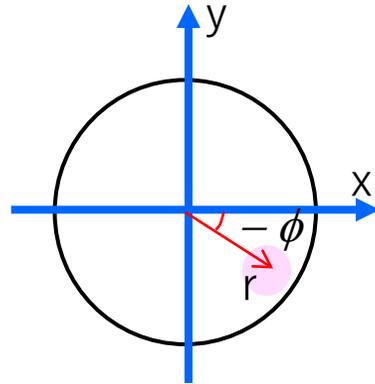
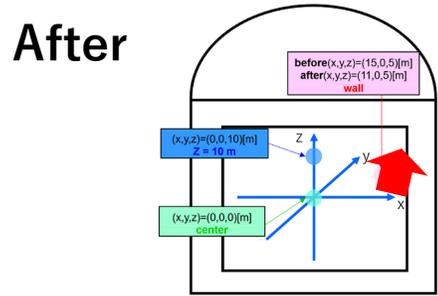
After



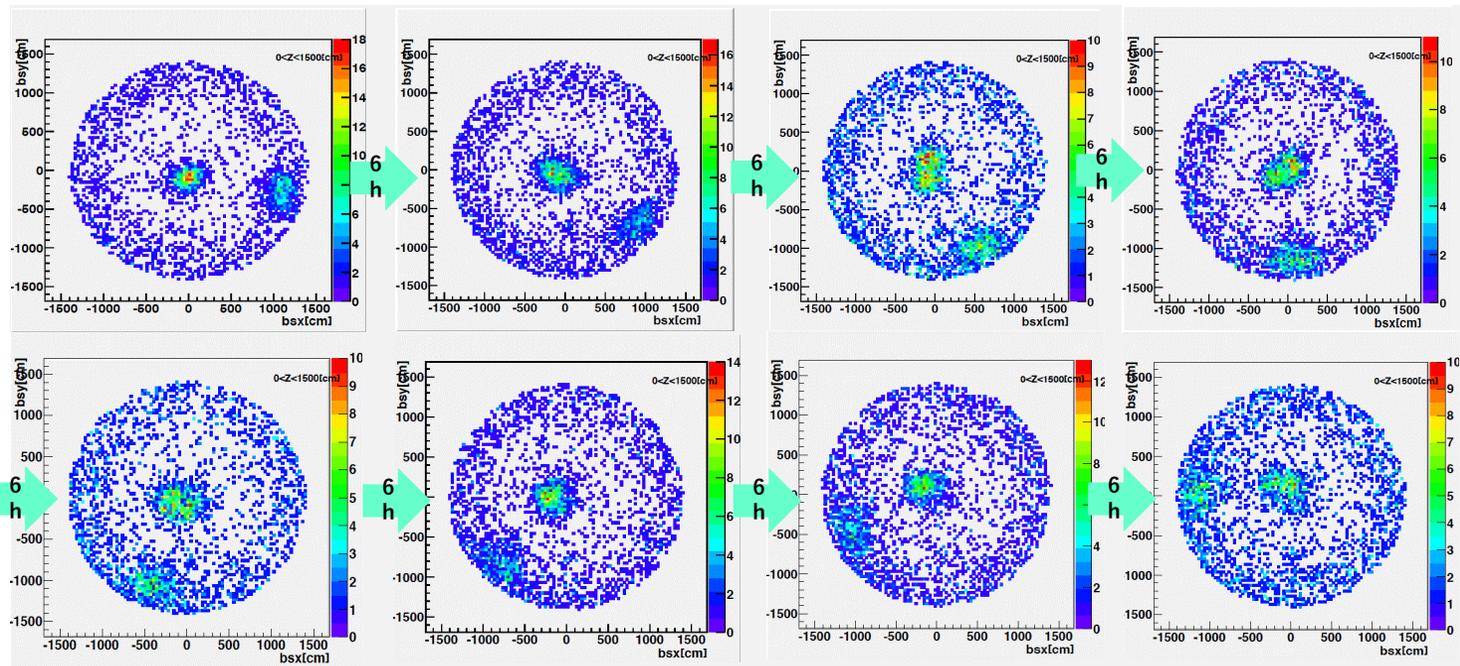
The water is rising at 5 cm/hr for all $Z > 0$
 After 250hr, it stagnates again as the temperature difference between the supply water and the tank water became small again.



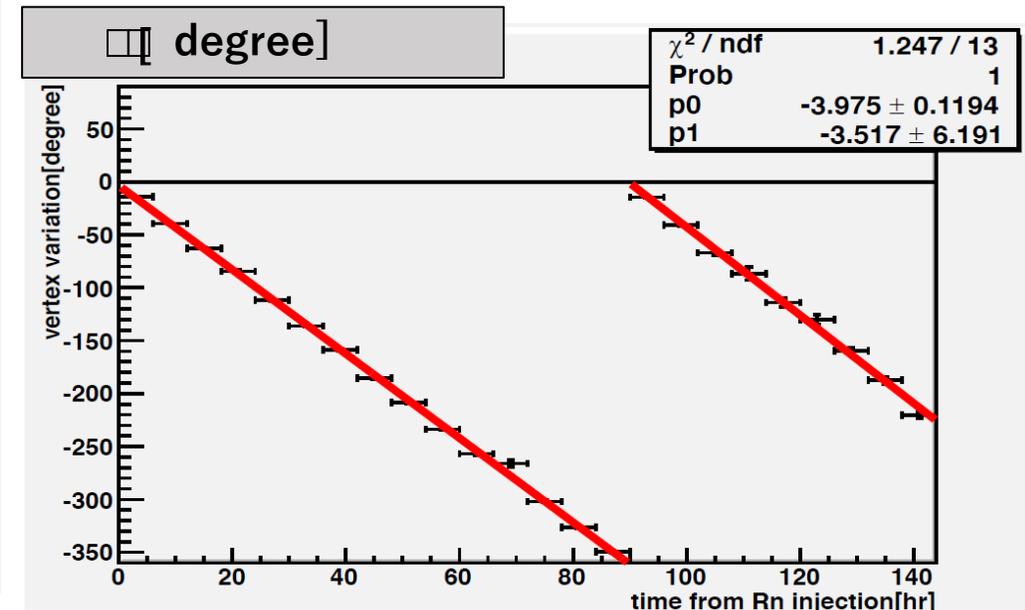
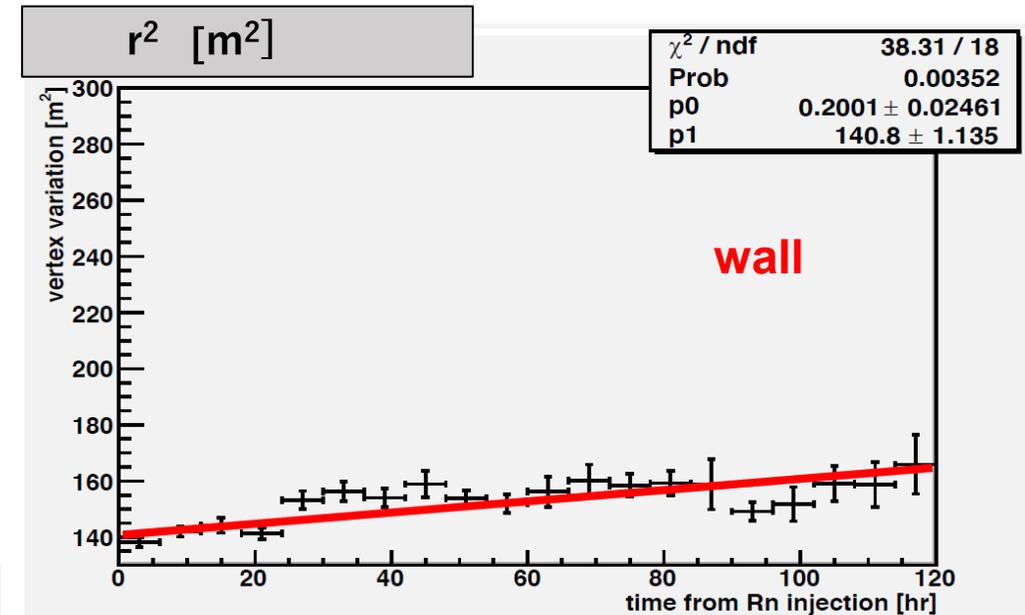
Flow in r and ϕ direction



Y.Kanemura



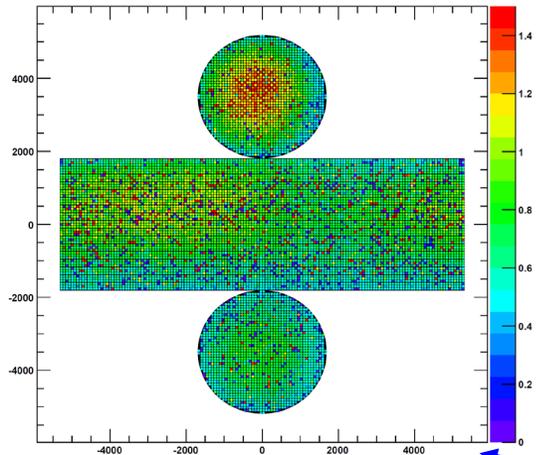
r : Moving toward the SK wall at a speed of 0.83 ± 0.10 [cm/hr]
 ϕ : Rotating clockwise at a speed of 4.0 [$^\circ$ /hr].



The results of the flow change

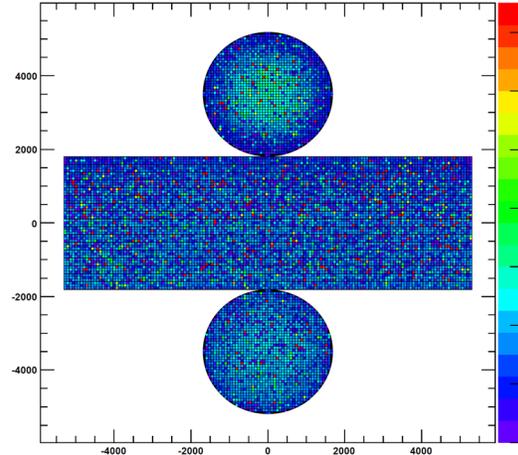
Nov.26.2020

Δ DR(Run85466-82911): Start Thu Nov 26 15:56:04 2020 2021-03-22 00:17:19



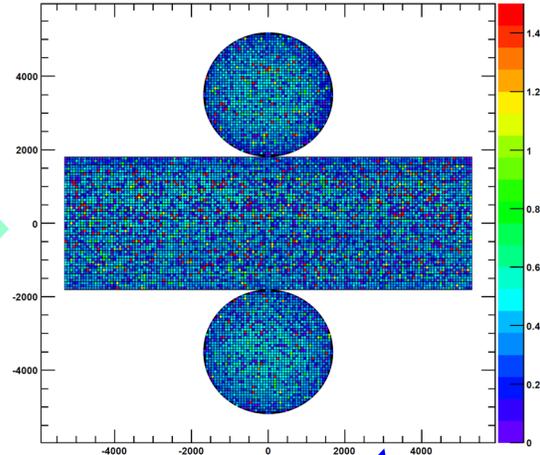
Dec.25.2020

Δ DR(Run85529-82911): Start Fri Dec 25 08:29:19 2020 2021-03-23 00:18:04

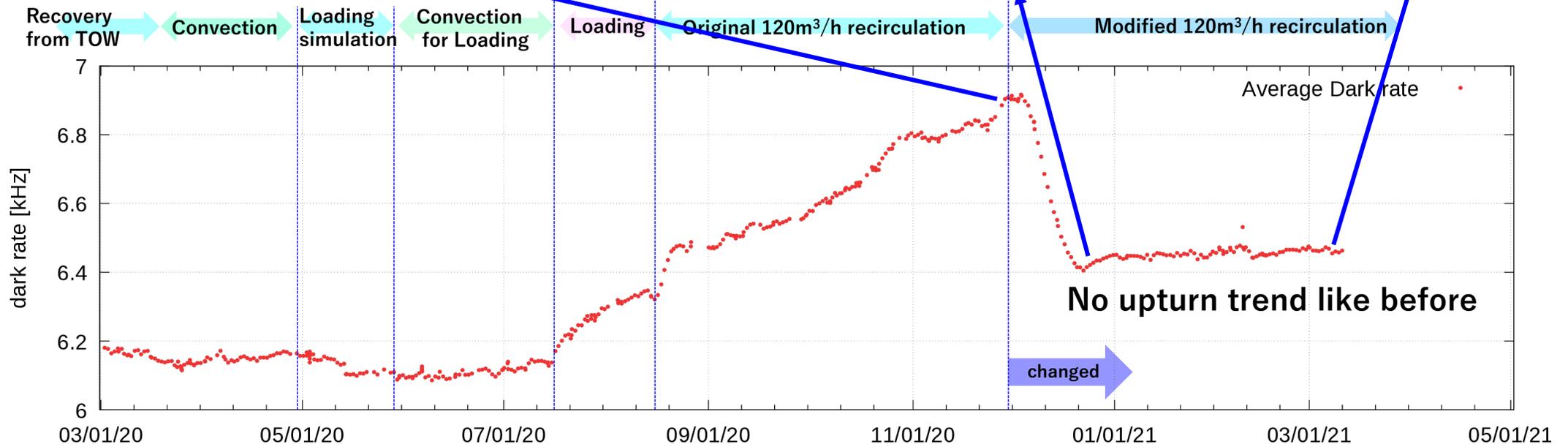


Mar.1.2021

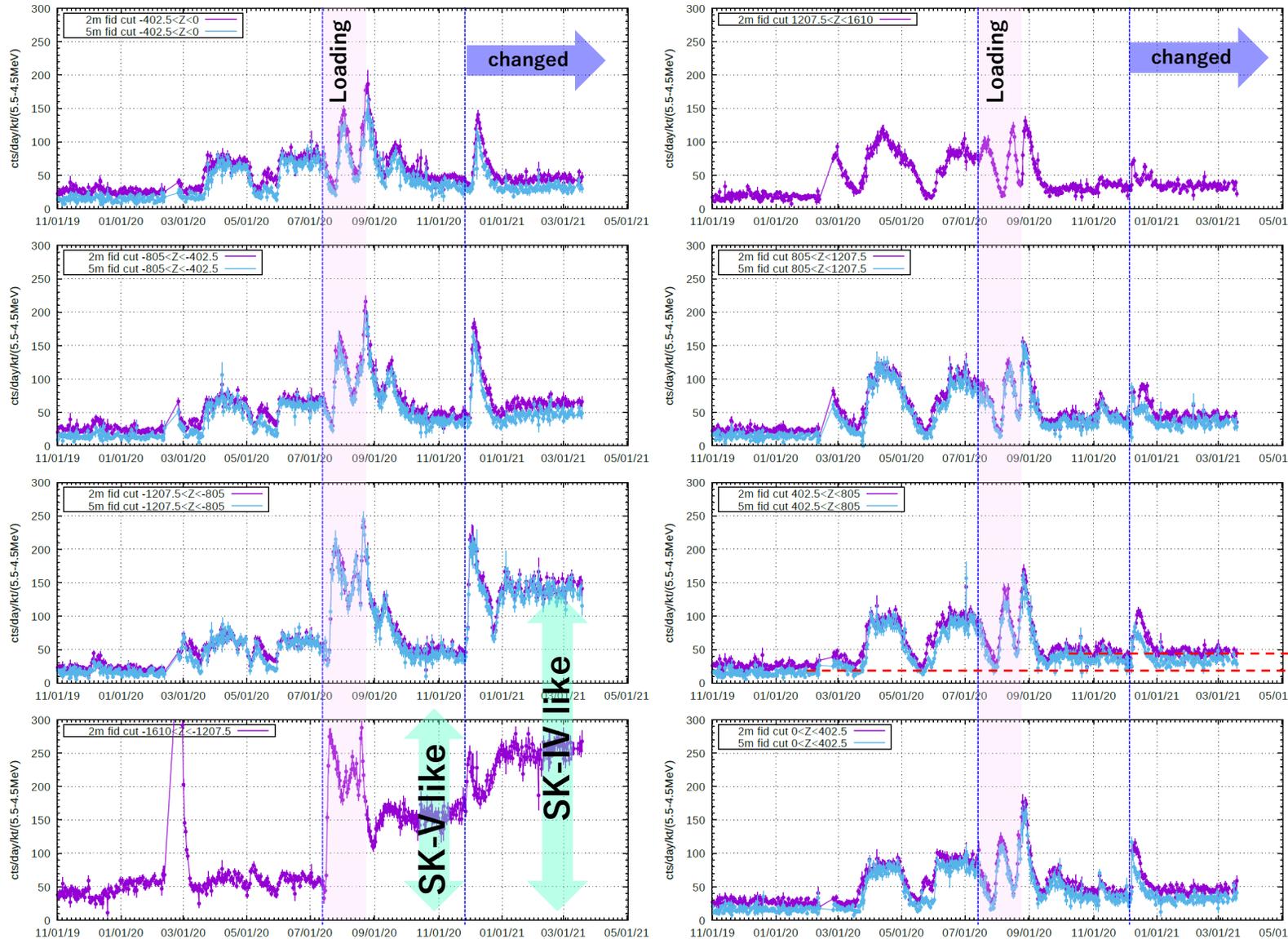
Δ DR(Run85763-82911): Start Mon Mar 1 09:04:57 2021 2021-03-23 00:20:07



Increased noise rate [kHz]



Flow change effect on the convection/Rn

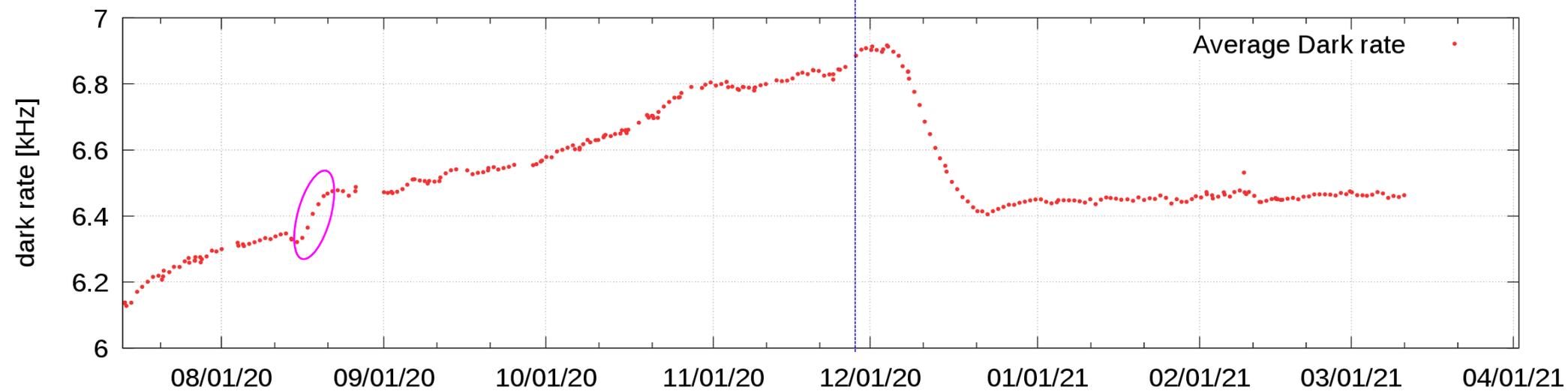
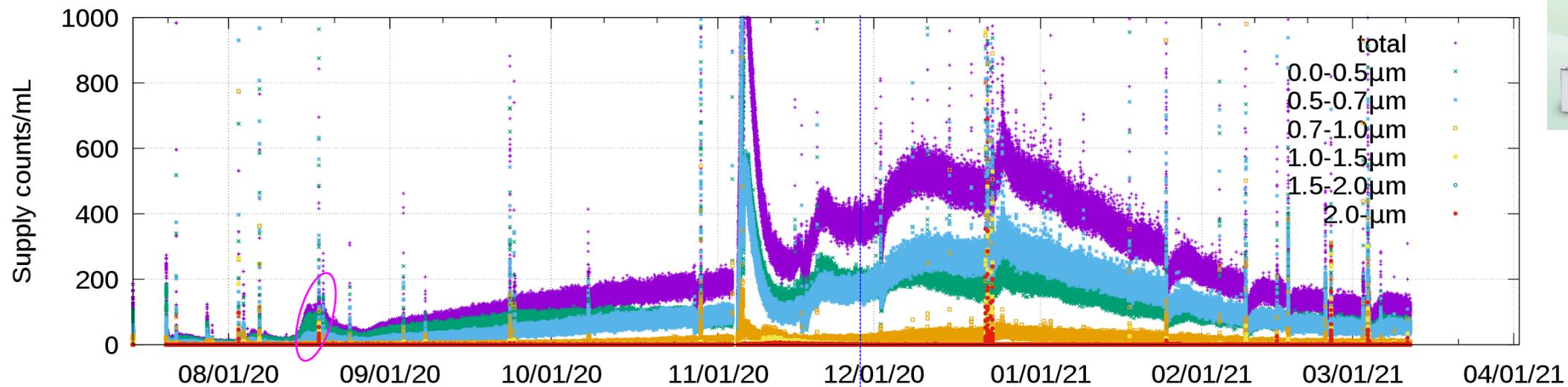


Even after the change,
convection region is kept $Z < -11\text{m}$
SK-IV like flow is realized

This is the issue!
 ^{222}Rn ? ^{228}Ra ? ^{226}Ra ? from Gd?

Mis-reconstruction
by additional g from Gd?

Bacteria and dark rate in SK-VI

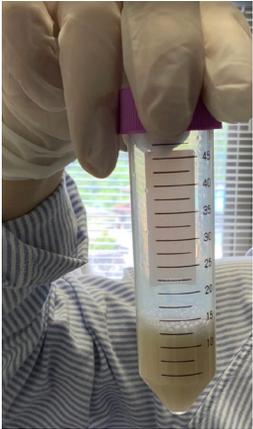


RION XL-10B

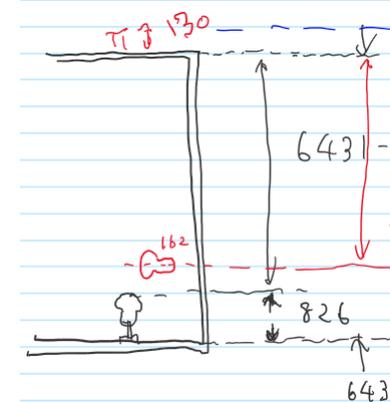
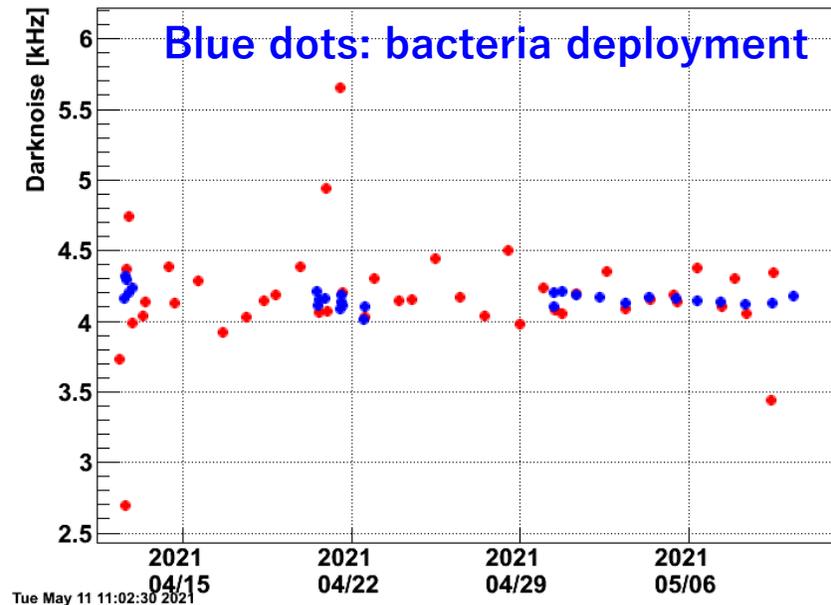
The bacteria: *Phyllobacterium myrsinacearum*

Prof. Yoshizawa AORI, UTokyo
Prof. Nomura Univ. of Tsukuba

- Single species dominates the Tank
 - *Phyllobacterium myrsinacearum* can live in pH 4 condition
- Tests in EGADS
 - SK rates: 1×10^{-6} photon/sell/sec
 - 10^{12} cells deployed in 200ton tank



Darknoise for ch. 162 (Last month)

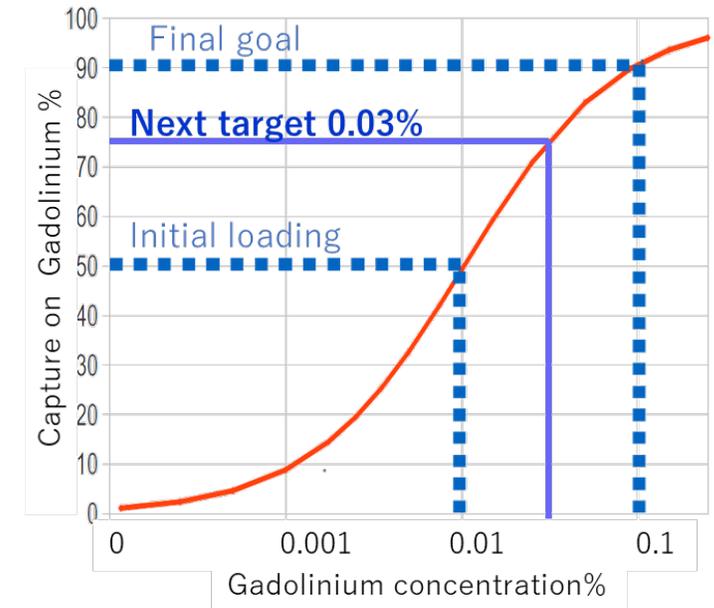


So far, no evidence of light emission
Too many parameters...
ex) Temperature/nutrition/growing phases etc

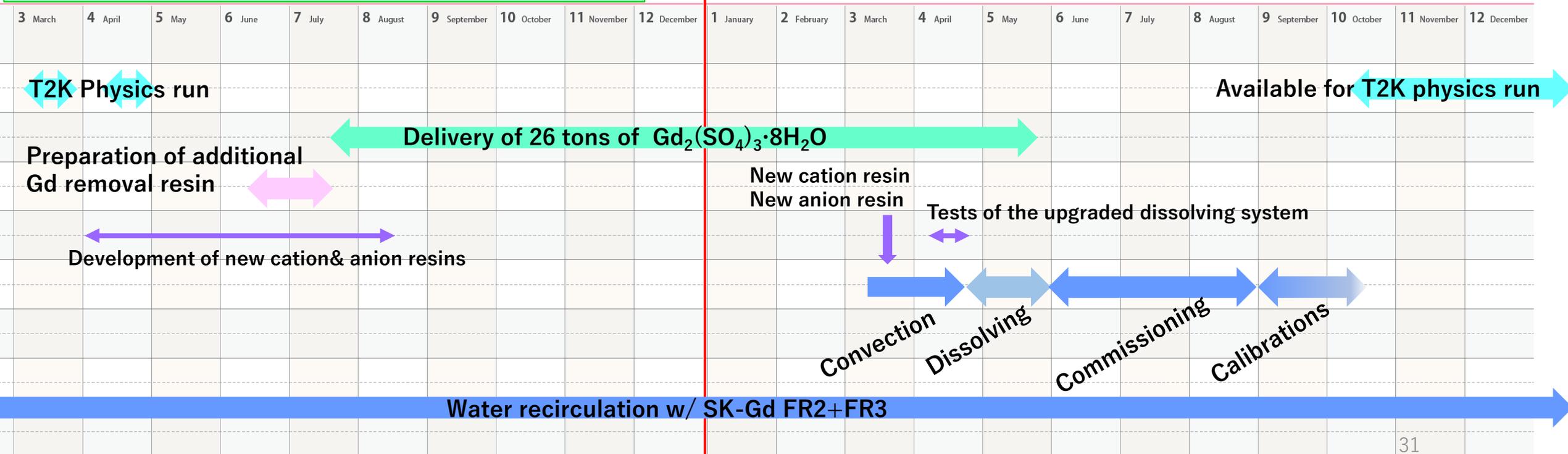
Next step

Planning to dissolve up to ~26 tons of additional $Gd_2(SO_4)_3 \cdot 8H_2O$ in 2021-2022

- Target Gd concentration: 0.03% (Currently 0.01%)
- Gd capture efficiency: 75% (Currently 50%)

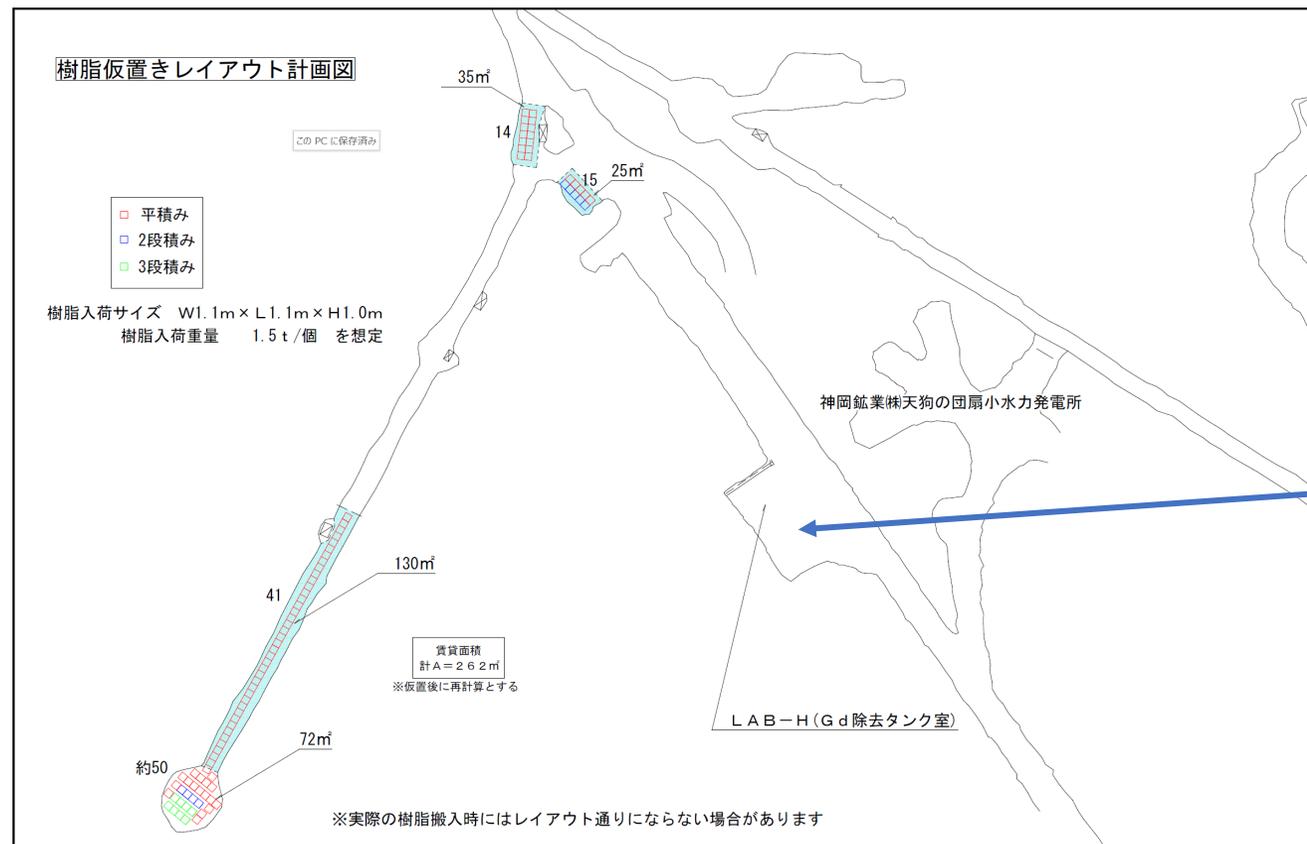


Possible plan for the next Gd-loading



Preparation for Gd-removal in Lab-H & I

- Lab-I(? 北20号) is under preparation for the storage of additional 153000L of cation exchange resin to collect Gd from SK-Gd T1.5



Lab-H Gd removal system
for SK-VI(0.01%Gd)
76000L of resin



Gd水溶液中でのRn分析高感度化

竹内康雄 神戸大学

脱気膜モジュール試作品

Gd水溶液中でのRn測定

- 目的: Gd水中 ^{222}Rn 測定を $\sim 0.5 \text{ mBq/m}^3$ の感度で行う装置を開発し、SK-Gd等で応用する。(D01+C01)
- 進捗: 脱気膜モジュールをステンレス化した脱気膜モジュールを試作し、D01にBGLレベル評価のために提供した。



ラドン検出器本体の低BG化

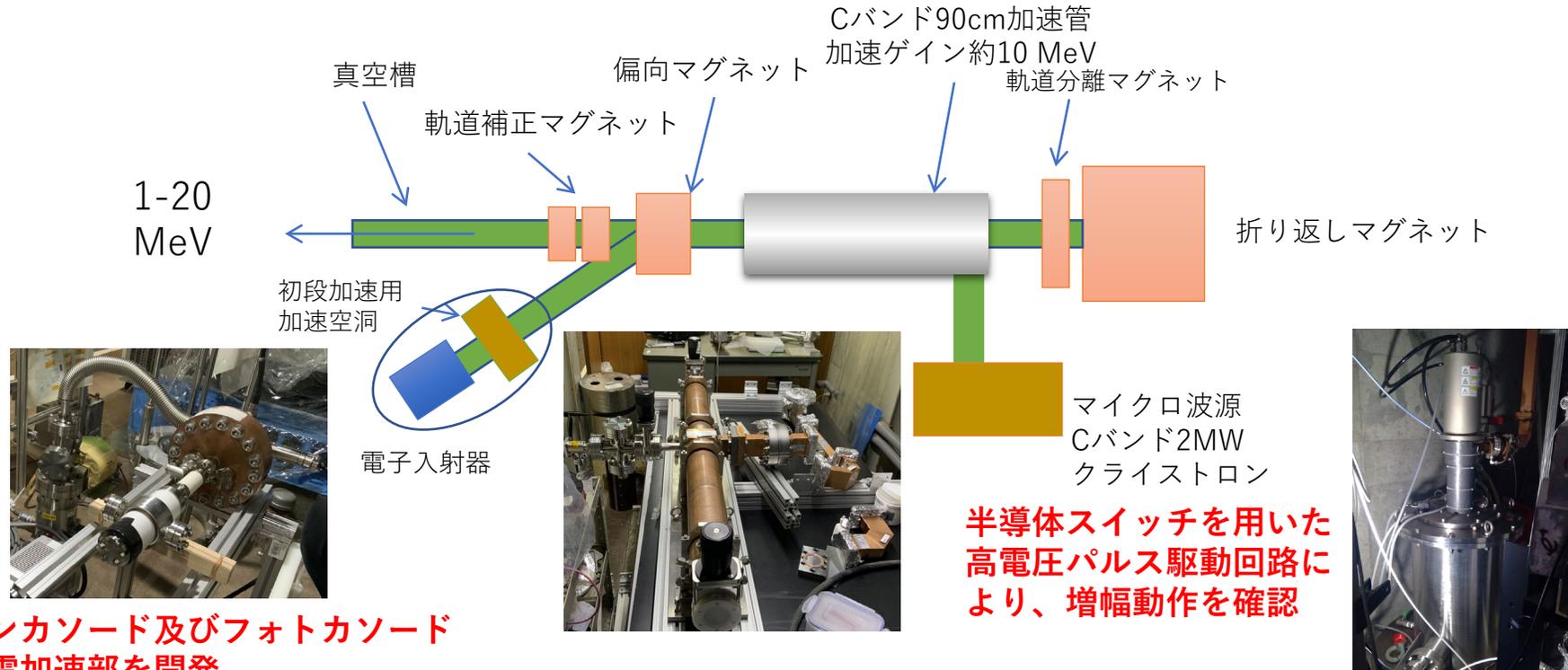
- 目的: D01で検討中の表面研磨技術をラドン検出器本体に適用して、低BG化を実現できるかどうか検証する。(昨年末頃からの新たな連携)
- 進捗: 業者さんと打ち合わせ中。今年度中に加工技術の検証(ラドン放出量に関する)と、その技術を適用したラドン検出器本体の試作を行いたい。

較正用電子加速器システム

鈴木良一

産業技術総合研究所

定在波型加速管を用いた往復加速により、小型加速管で20 MeV以上の電子加速を目指す



ナノカーボンカソード及びフォトカソード
電子銃と静電加速部を開発

今後

主加速管へ供給するクライストロンのマイクロ波出力を上げるとともに、電子入射器・前段加速部を主加速管に接続して、電子加速実験を行い、MeVオーダーの電子加速を実現する。

C01 1年のまとめ

- 2020年 0.011% Gdを導入し、ついにSK-Gdを開始した
 - DSNBを実際に探索できる感度を有しているはず
- 検出器全体でGdの濃度は一様になっている
 - 直接測定、Am/Be線源、Spallationによる中性子で確認
- 導入後、水が光っているように見える
 - 滞留と相関があり、水を一度”入れ替える”と低減した
 - 原因は調査中
- 低エネルギーBGが予想より多い
 - ^{228}Ra , ^{226}Ra , mis-reconstruction等の可能性を調査中
 - SK-IV程度の太陽ニュートリノ観測は可能
- 2022年 Gd 0.03%を目指している
 - 上記問題の対応
 - 原料の酸化ガドリニウムの放射性不純物レベルが高い、酸化Gd価格急上昇中
- Rn
 - $\sim 0.5 \text{ mBq/m}^3$ の感度を目指しD班との連携との連携がすすんでいる。
- Linac
 - 入射部およびクライストロンの準備が進む

