The 4th J-PARC Symposium (J-PARC2024)

Radiomercury collected during an operation of the neutrino experimental facility, J-PARC

Eisuke Watanabe[1,2],
K. Takahashi[1,2], K. Saito[1,2], Y. Oyama[1,2], E. Lee[1,2], H. Matsumura[1],
G. Yoshida[1], K. Tsugane[1], S. Nagaguro[1,2], K. Bessho[1,2], and H. Yamazaki[1,2]
[1]KEK, [2]J-PARC

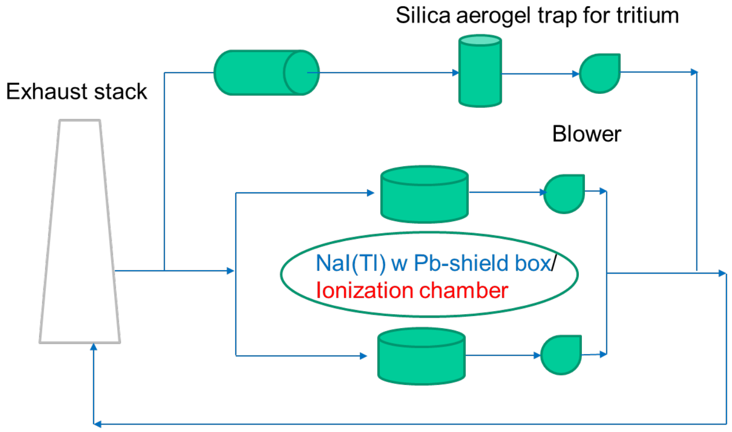
Radiation monitoring at J-PARC

- At J-PARC, radiation levels and the radioactivity in the exhaust air and drainage water are monitored.

- Airborne activity in exhaust
  - Radioactive gas

    Ionization chamber or Plastic scintillator / NaI(Tl) with Pb shield

    For H-3, silica aerogel traps and liquid scintillator



To measure activity without pause, gas monitors are duplicated

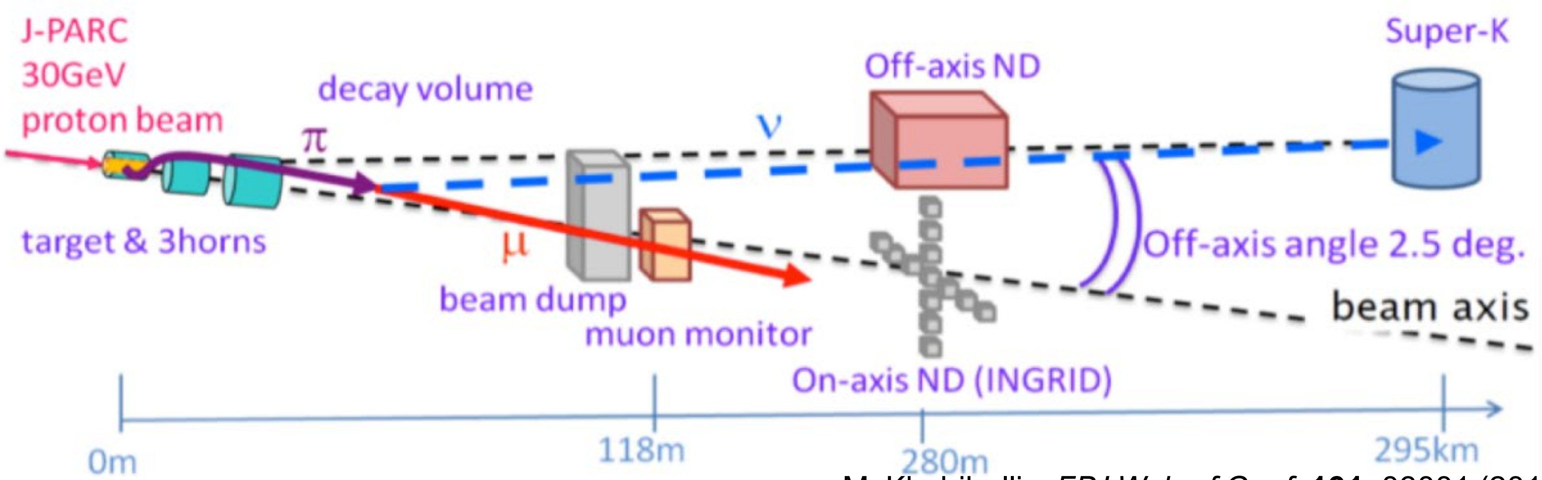
  - Radioactive dust

    Semi-conductor / GM or NaI(Tl) with filters

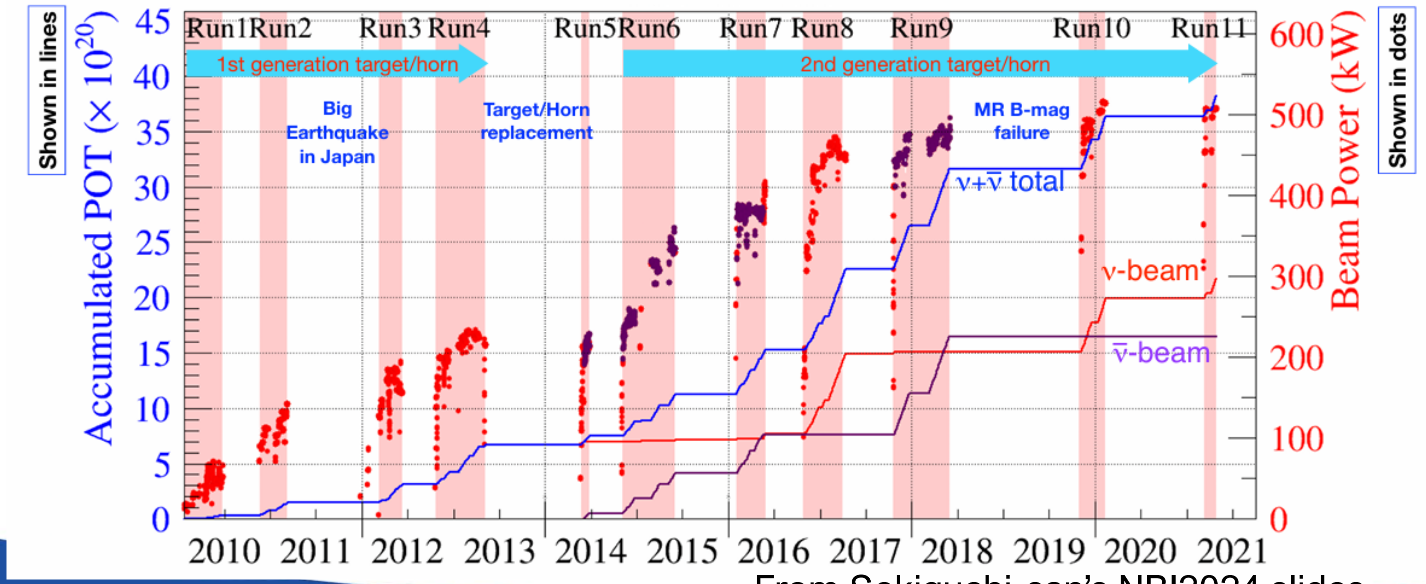
    Filters: A cellulose-glass fiber filter and a charcoal filter (for I, Hg etc.) replaced once a week and measured with Ge detector

J-PARC neutrino experimental facility

- ✓ For T2K experiment (Tokai to Kamioka Long Baseline Neutrino Oscillation Experiment)
- ✓ Beam power of 30-GeV proton achieved 800kW in 2024 and will 1.3MW in 2028.



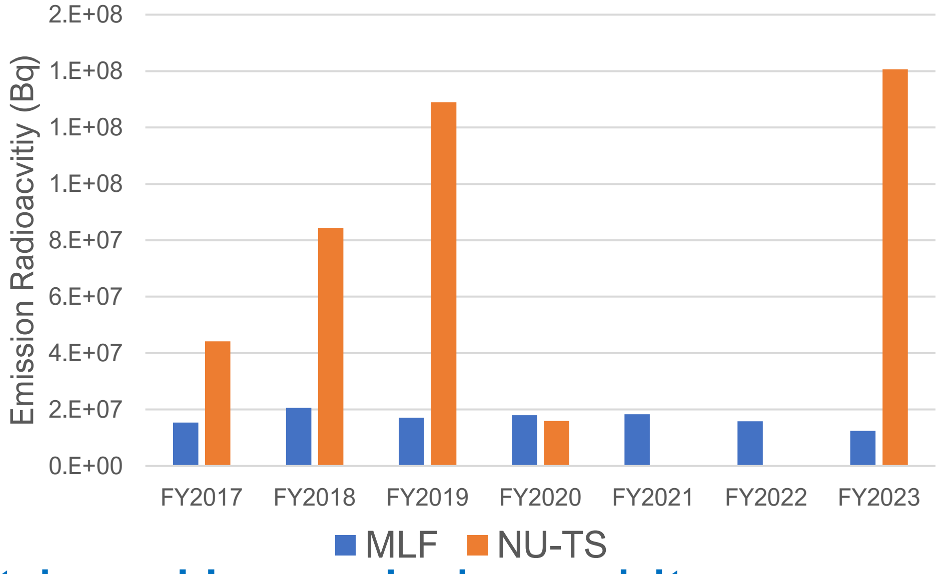
M. Khabibullin, *EPJ Web of Conf*. **191**, 03001 (2018).



From Sekiguchi-san's NBI2024 slides

Radioisotope emission

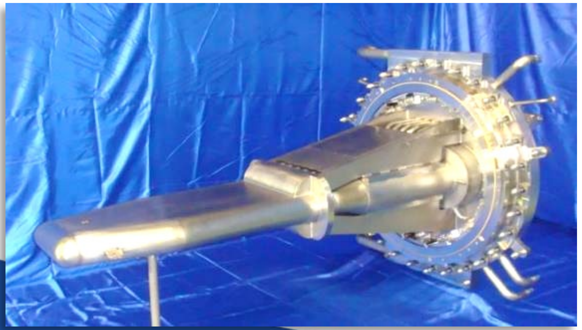
Emission radioactivity of ^{197}Hg

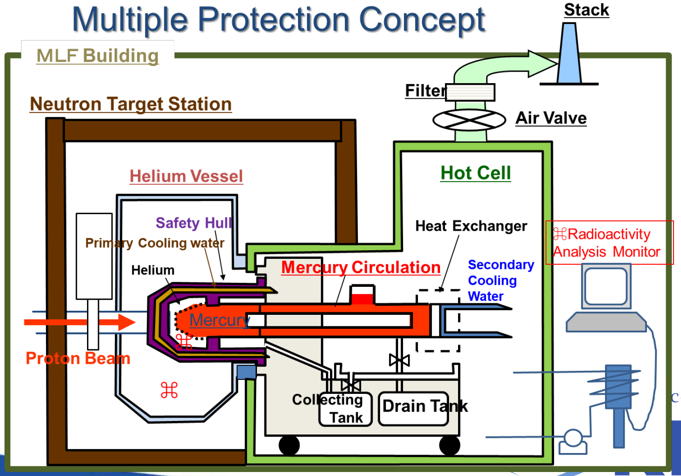


Radiomercury (rad-Hg) is observed in exhaust air.

- MLFs have huge radioisotopes within the Hg target but do not emit externally.
- Neutrino buildings emit more rad-Hg than MLF.

✓ MLF neutron source: Liquid Mercury





✓ Neutrino : He-cooled graphite

Where is rad-Hg from…?

Radiomercury in J-PARC

Rad-Hg in the Neutrino exp. facility

- Specific activities in exhaust air are much lower than the regulatory limits.
- It cannot be a barrier to Neutrino physics.

- However, its origin is unclear.

Study on rad-Hg

- To deduce source and production mechanism of rad-Hg in the Neu. TS buildings.

- To show that the rad-Hg at the Neu. exp. facility is not related to MLF.

- To understand behavior of rad-Hg to establish measuring and controlling method.
  - Contributing for whole of safe management of J-PARC.

Outline

Deducing the source and production mechanism of rad-Hg

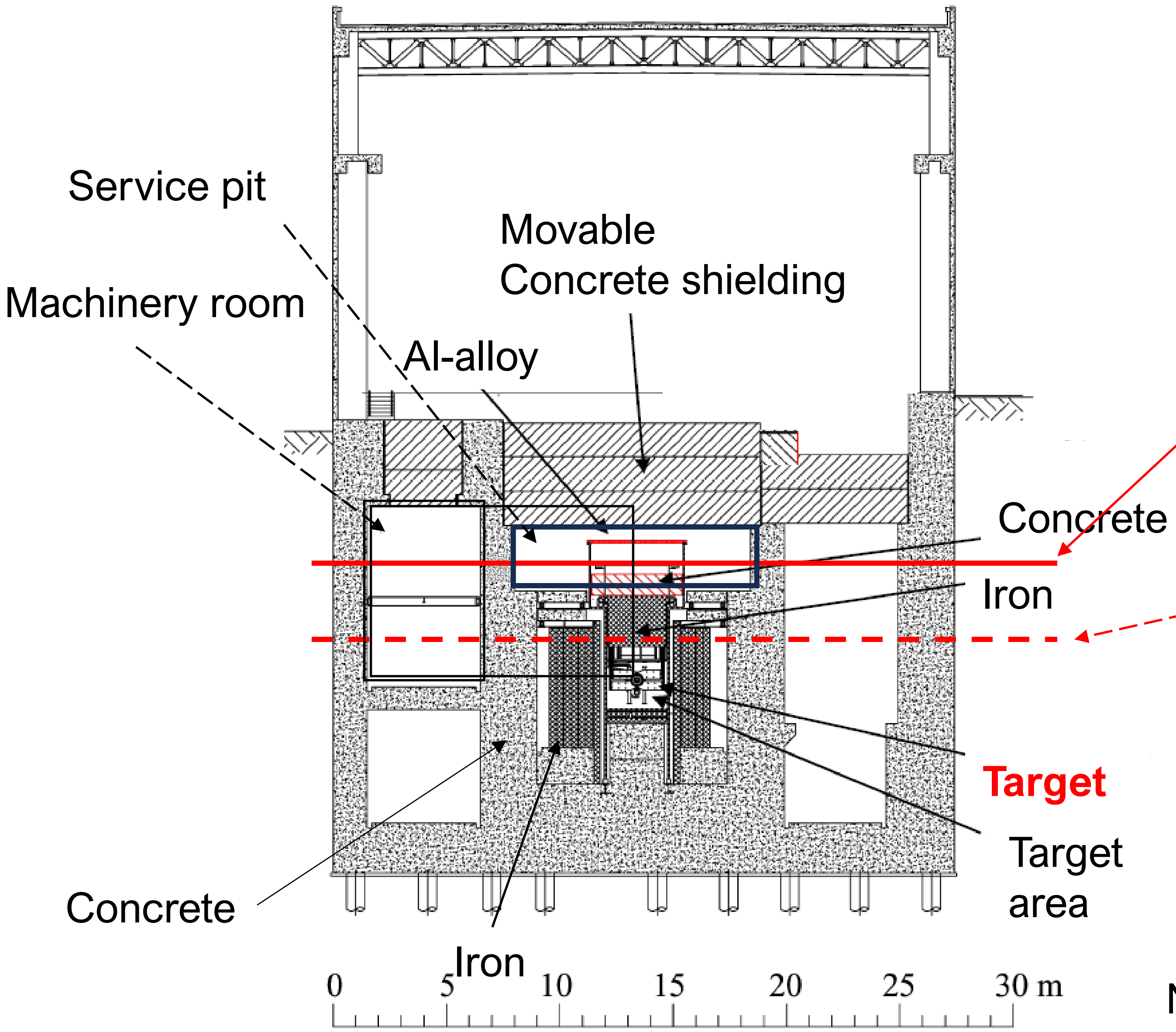
✓Experimentally collecting radiomercury with activated carbon (AC)
✓Radiation environment calculation with PHITS code
✓Model radioisotope yield calculation with PHITS code

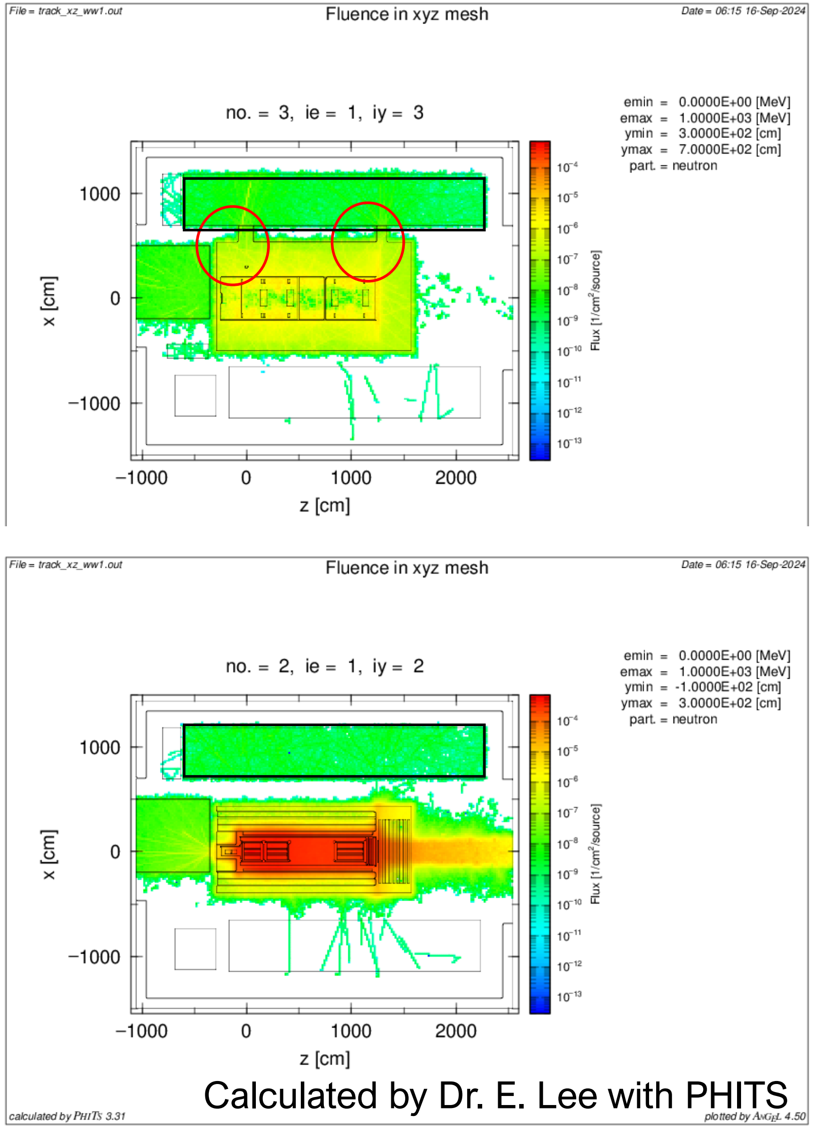
Establishing measurement method of rad-Hg

✓Preliminary experiment using chemical-modified AC

Radiation environment of the neutrino exp. facility

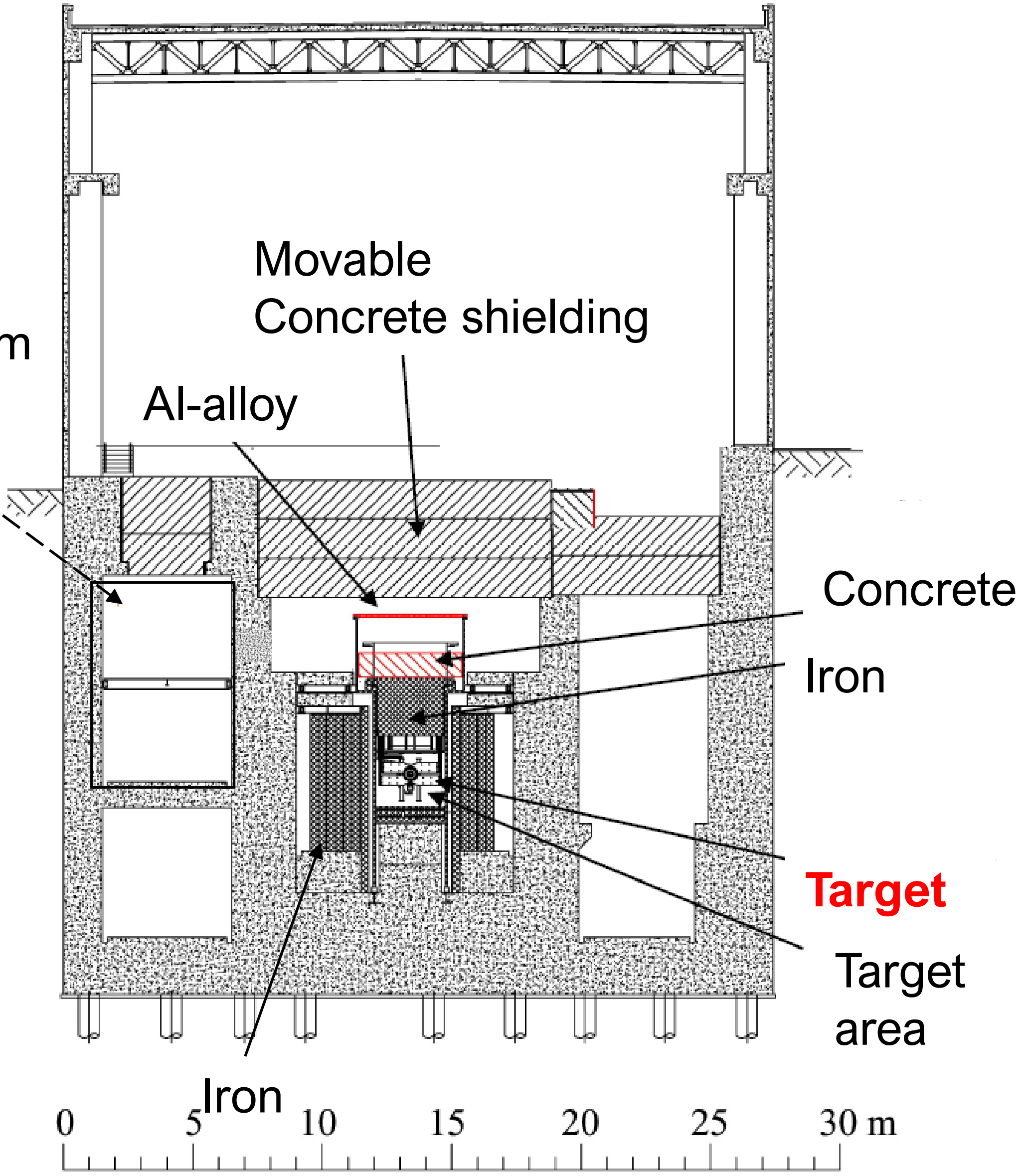
Target Station Building, Neutrino Exp. Facility



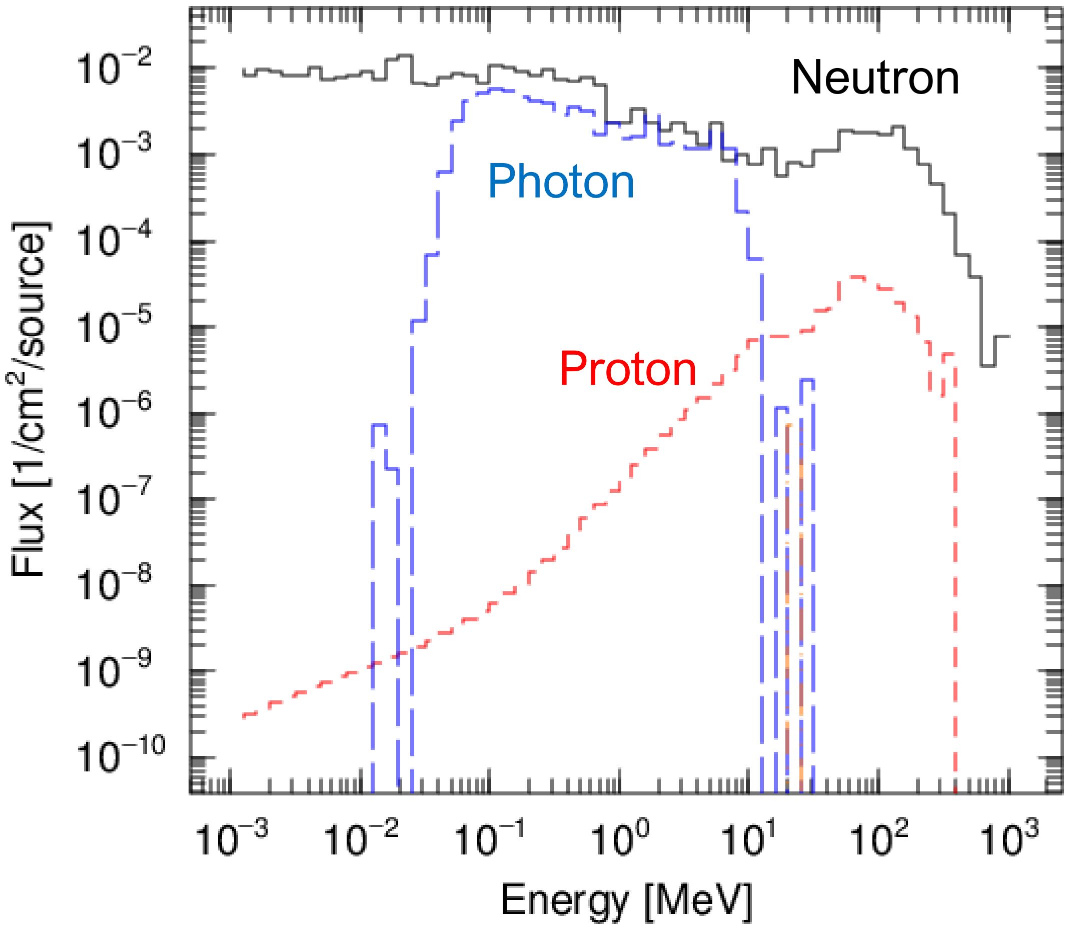


Neutrons fill the machinery room through two doors.

Target Station Building, Neutrino Exp. Facility



Average spectrum of target-side wall
in the machinery room



Much protons and neutrons with 100-200 MeV

Collecting radiomercury with activated carbon cartridge

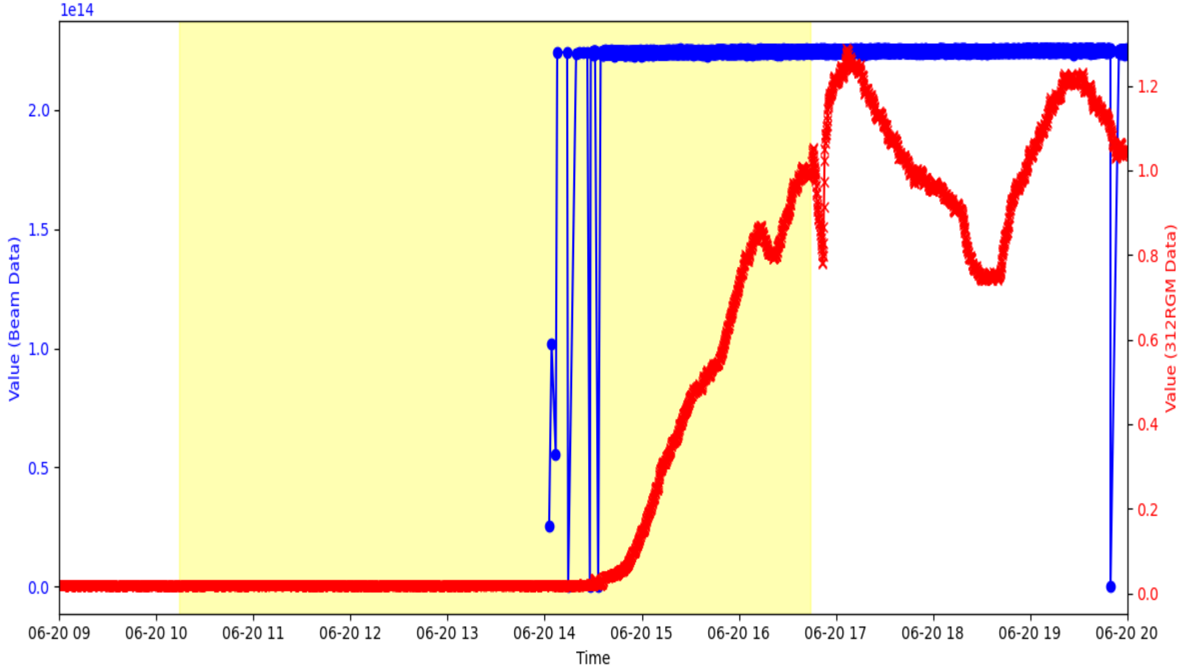
Air sampling

Method

- Air sampling experiment was conducted in June 2024 operation.
- The air in the machinery room was collected for 134 min at 100 L/min flow rate.
- Gas monitors (ionization chamber) in the outside of interlocked area were used.
- Activated carbon cartridges “CHC” (ADVANTEC), filter paper HE-40T (ADVANTEC) for dust filters were used.

Activated carbon cartridge (ADVANTEC)

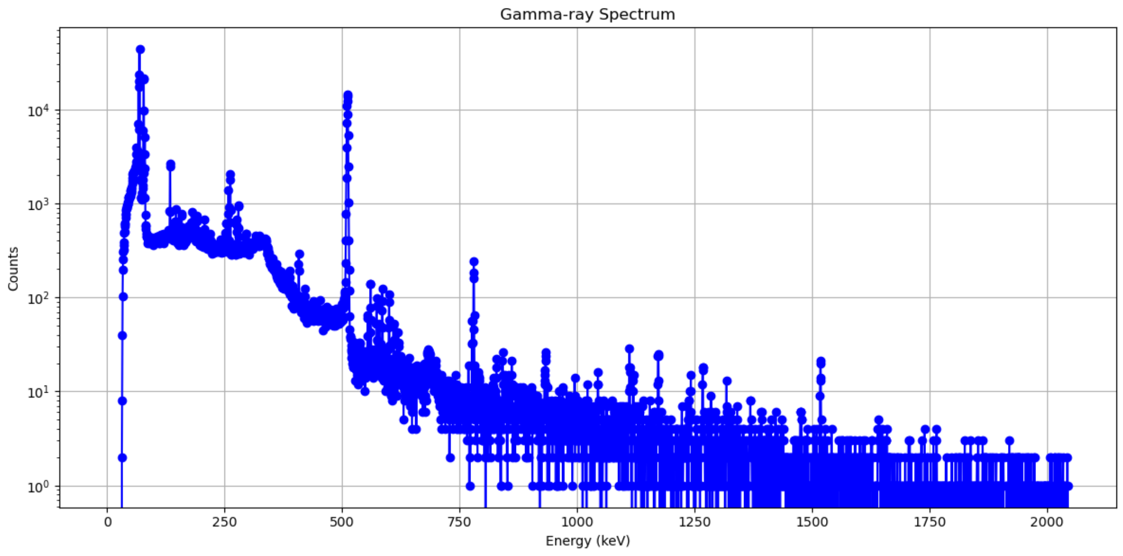
Neutrino TS operation and radioactivity in RGM

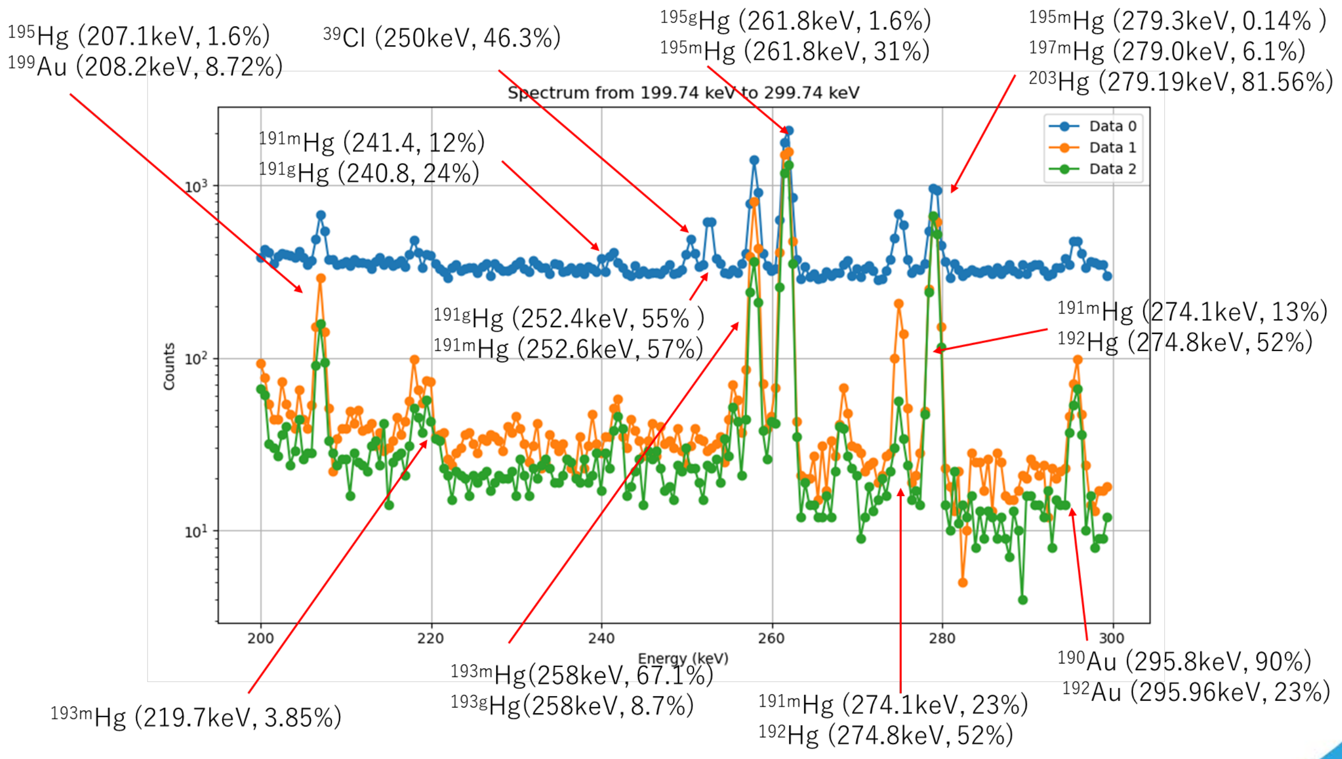


Gamma-ray analysis

- ✓ Sample was transported from the neu. TS to the rad. measurement bldg.
- ✓ Gamma-ray measurement with Ge semiconductor detector.

Gamma-ray spectrum of CHC with rad-Hg





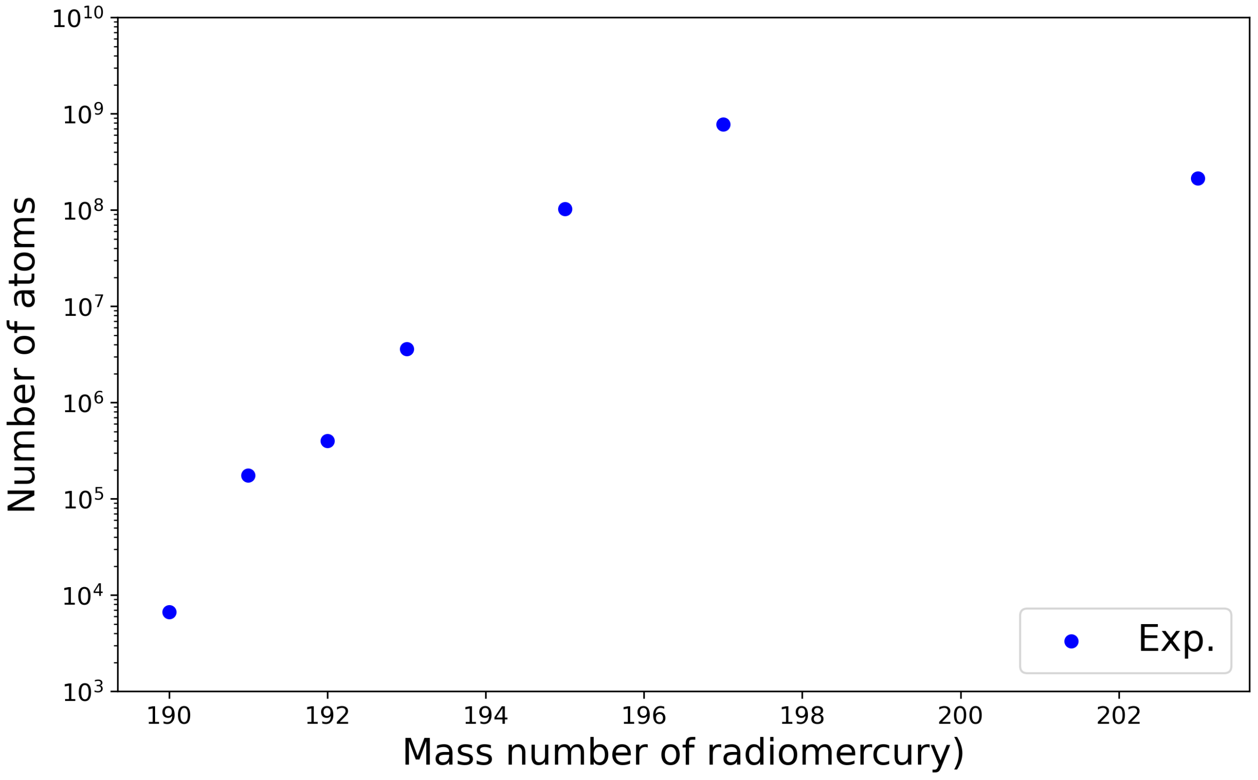
(Cooling Time: 2023 s, 19602s, 59786s)

Collection radiomercury in the machinery room

✓Observed various rad-Hg isotopes on an activated carbon filter.

Nuclide	Half-life (sec)	Activity (Bq)	err
Hg-190	1200	1.24.E+01	3.10.E+00
Hg-191g	2940	3.04.E+01	8.00.E+00
Hg-191m	3000	2.23.E+01	9.60.E+00
Hg-192	17460	3.90.E+01	3.40.E+00
Hg-193g	13680	2.80.E+01	1.23.E+01
Hg-193m	42480	5.23.E+01	5.60.E+00
Hg-195g	35640	8.05.E+02	7.36.E+01
Hg-195m	149760	2.92.E+02	2.38.E+01
Hg-197g	230904	2.28.E+03	2.90.E+02
Hg-197m	78480	2.08.E+02	4.80.E+00
Hg-203	4025376	4.54.E+01	2.00.E-01

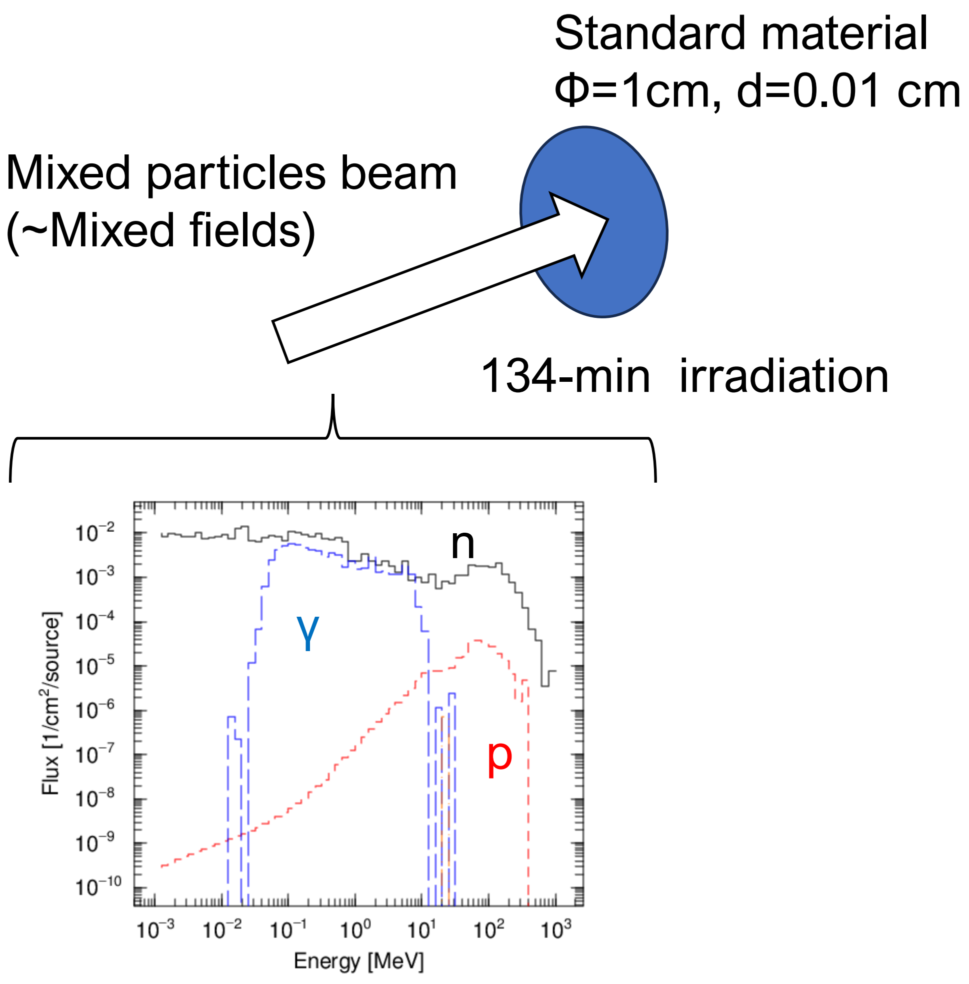
(At the end of collection)

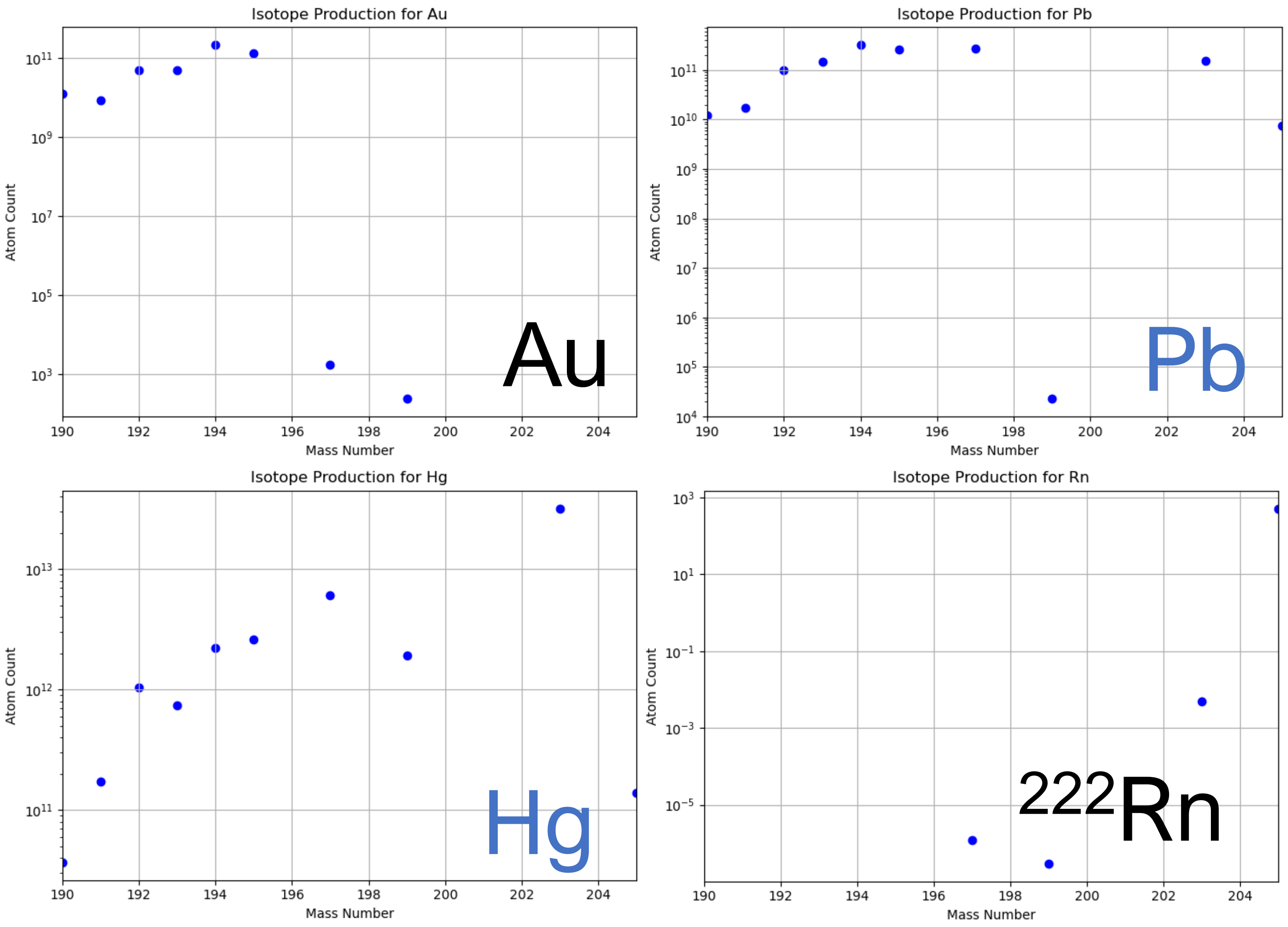


Comparison with model nuclear reaction calculation by PHITS

Model isotope production calculation

✓ Model calculations were conducted to deduce the “target material”





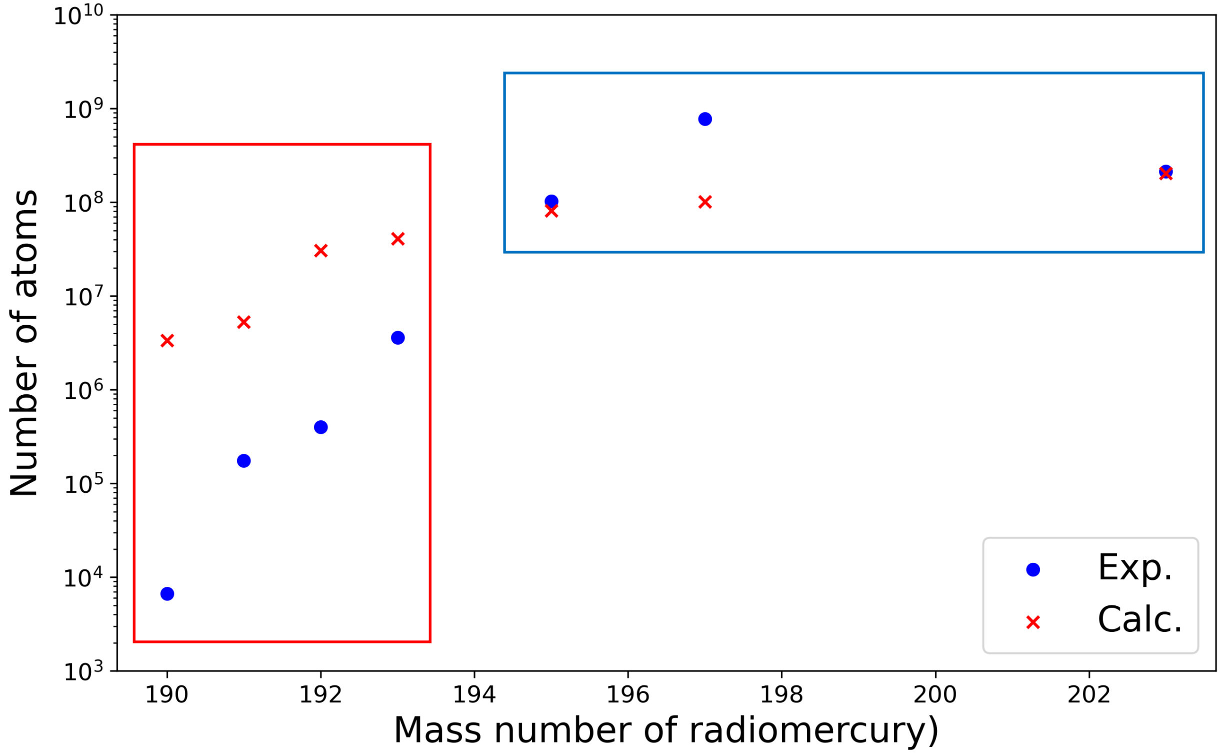
Hg and Pb are supposed to produce rad-Hg

Estimation of rad-Hg production

Source candidates;

- **Naturally present Hg (1.7 ng/m^3) and Pb (73 ng/m^3) in air**
- Building concrete (Pb)…?
- Some accelerator component…?

Collected and estimated amount of rad-Hg
in the air from the machinery room (V = 13.4 m^3)



Rad-Hg production can be roughly explained by the activation of heavy metals in the air.

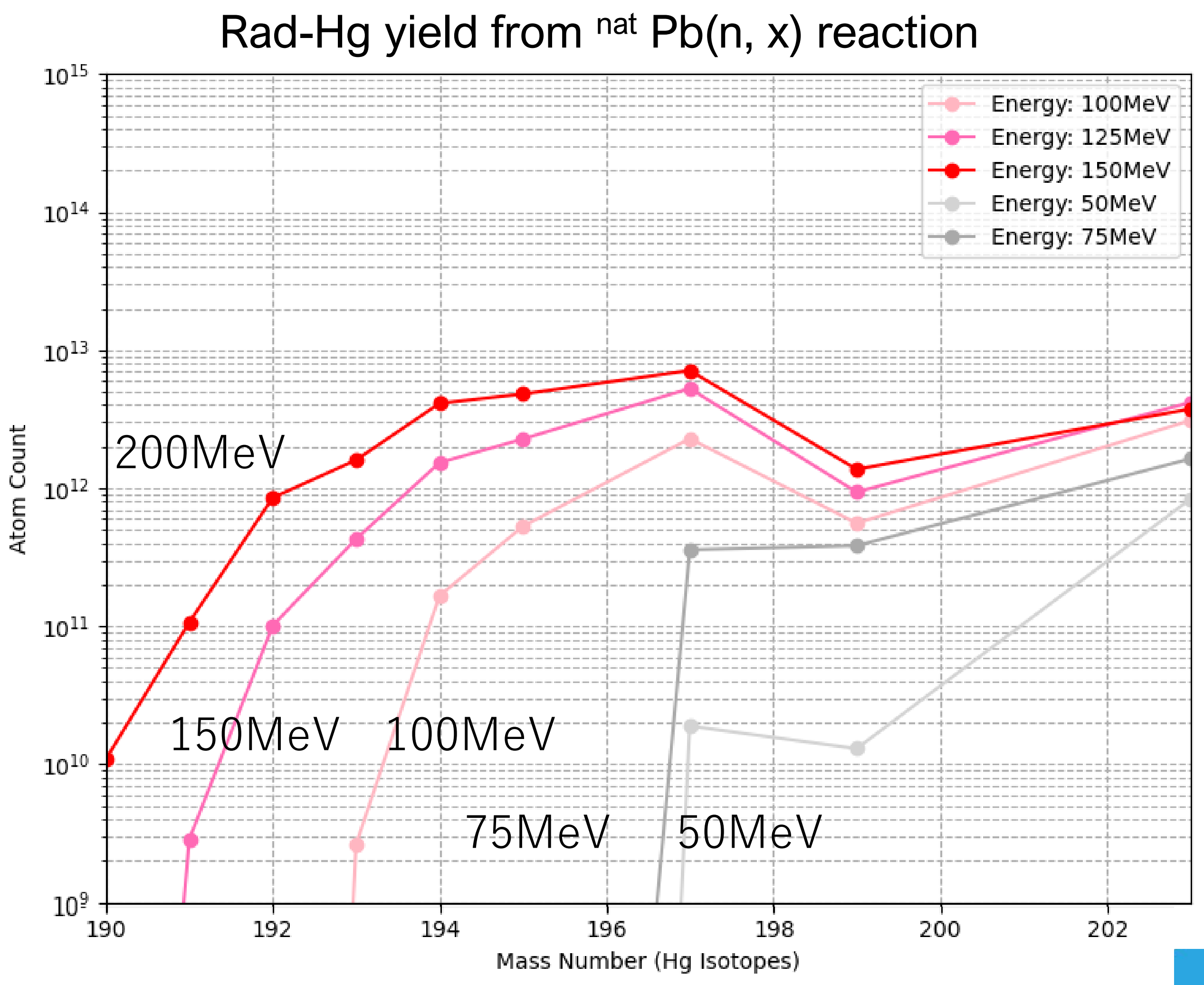
✓ Thermal neutron is not considered for calc.
  ➢ Hg-195, 197, 203 is underestimated

✓ 190 ≤ A ≤ 193 seems to be overestimated
  ➢ Much neutron lower than 100MeV?

Accurate understanding rad. env. of TS bldg.

- PHITS calculated much population of neutron with 100-200 MeV in the machinery room.
- By comparison between exp. and Calc,, TS machinery room has less neutron with > 100-200 MeV.

➢ We are planning actual measurements of the radiation field in the machinery room to verify “natural Hg and Pb hypothesis”



Towards a study of chemistry of rad-Hg: a preliminary result

Towards a chemistry of radiomercury

Amount of rad-Hg produced in the neutrino experimental facilities is sufficiently small.

- Even with 1.3 MW beam power, no barrier to neutrino physics.

On the other hand…

- Collection and measurement methods for airborne rad-Hg is not established enough.
- Important for the radiological control for all facility of J-PARC.

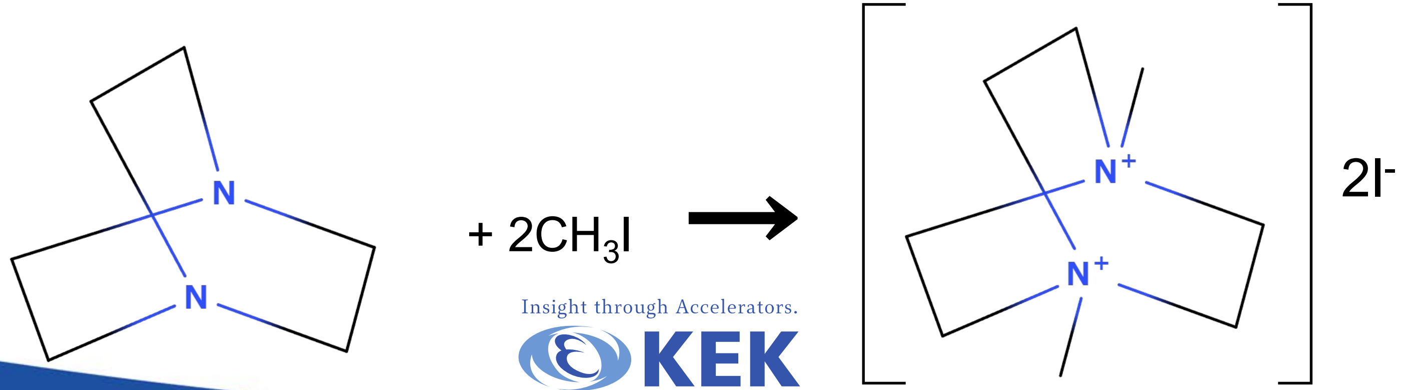
Chemical-modified Activated Carbon

- ✓ Few available data on mercury collection efficiency on AC.
  - ➢ Based on the study of radiomercury "production" in JAERI's reactor in 1970's.
- ✓ No data available on the efficiency of commercially available chemically modified AC.

In the 2024A neutrino operation…

Conducted rad-Hg collection using TEDA-containing activated carbon cartridges.

- Add 10% triethylenediamine to CHC to improve the adsorption rate of radioiodine.
- The adsorption rate for methyliodine is increased.

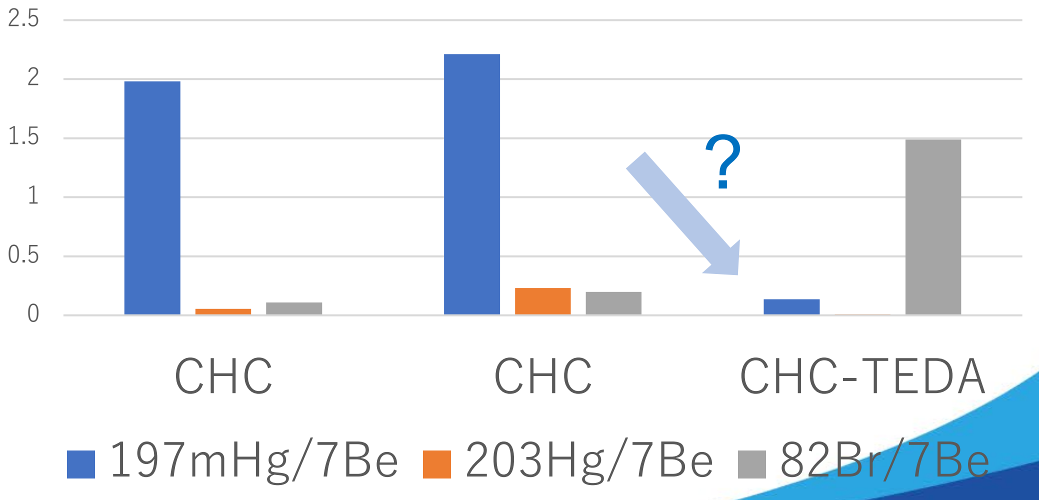


Adsorption experiment

		CHC				CHC-TEDA	
	Beam time	3d 16:22		4d 11:50		5d 00:43	
	POT	4.53E+19		6.05E+19		7.12E+19	
		Activity (Bq)	err	Activity (Bq)	err	Activity (Bq)	err
on CHC	^{197m}Hg	3.26.E+03	3.74.E+01	3.77.E+03	1.39.E+01	2.35.E+02	2.93.E+01
	^{203}Hg	9.03.E+01	6.41.E-01	3.95.E+02	1.19.E+00	1.45.E+01	2.76.E-01
	^{82}Br	1.78.E+02	9.02.E+00	3.40.E+02	4.60.E+00	2.58.E+03	2.24.E+01
on HE	^{7}Be	1.64.E+03	1.48.E+01	1.70.E+03	3.23.E+00	1.73.E+03	7.29.E+00
	^{24}Na	2.33.E+02	5.67.E+00	6.86.E+01	4.05.E-01	1.37.E+03	1.00.E+02

The reduction of rad-Hg adsorption

1) complex formation and volatilisation of Hg^0 and TEDA?
2) Decrease in the number of activated carbon adsorption sites due to TEDA impregnation?



Future plan: $FeCl_2$, $CuCl_2$-containing AC…

Summary

- J-PARC neutrino experimental facility emits much less rad-Hg than the regulation value.

- It is semi-quantitatively estimated that the source is probably naturally-present mercury and lead in the air.
- The accelerators upgrade has resulted in background observations of nuclides that were not previously observed.
- Rad-Hg in the neutrino facility would not be related to the MLF.

- We are trying to develop a method of collecting rad-Hg using chemically modified activated carbon in the future.