Identification of ^{210}Pb and ^{210}Po in the bulk of copper with low background alpha counter

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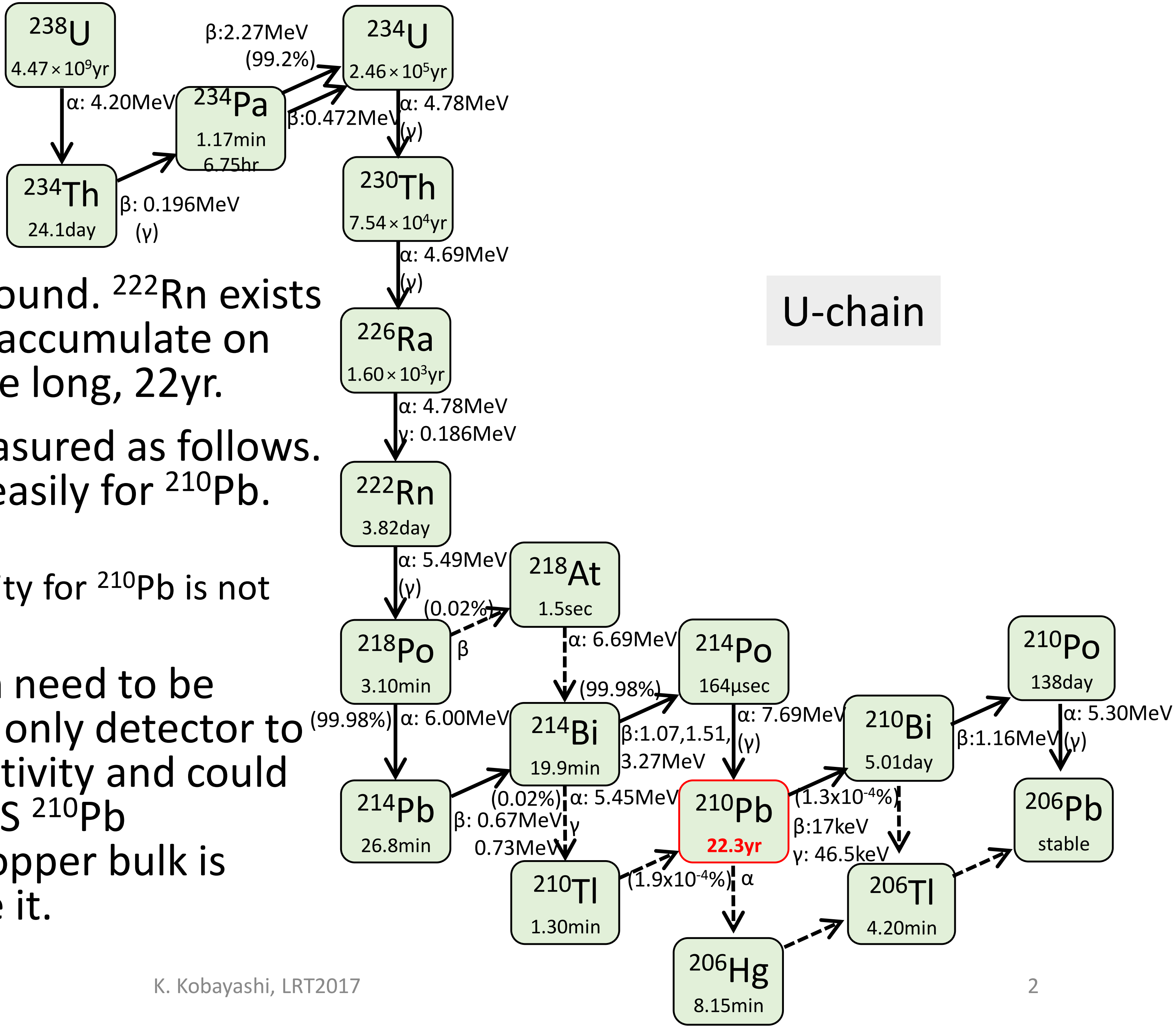
motivation

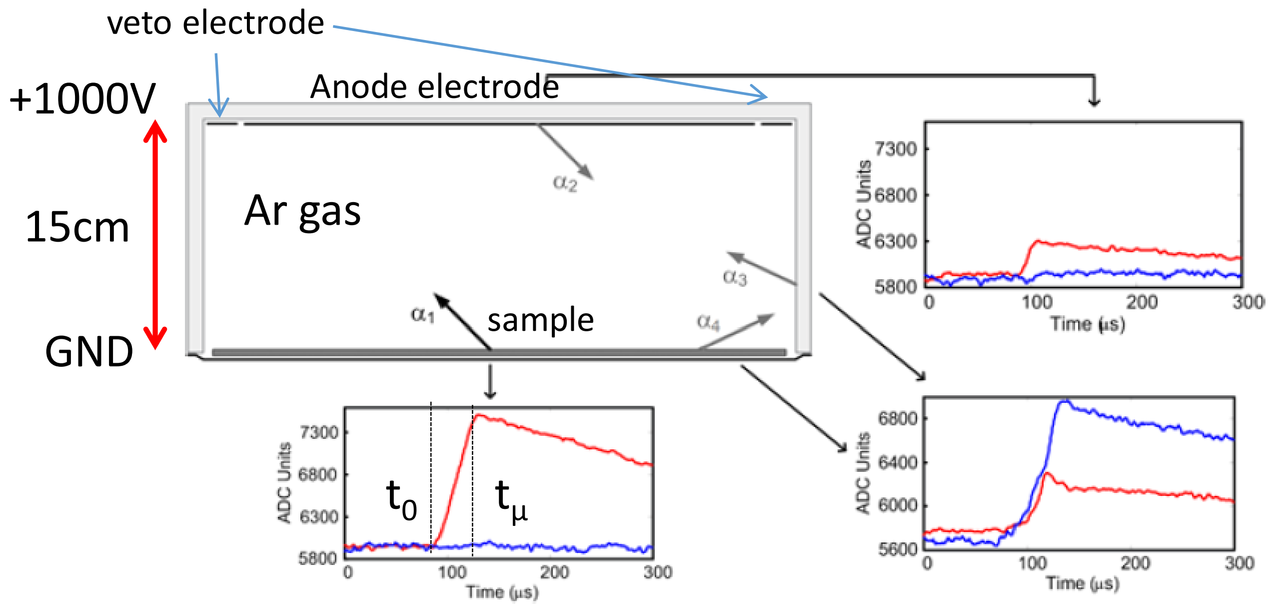


- ^{210}Pb could be the nasty background. ^{222}Rn exists in the air and its daughters can accumulate on materials. ^{210}Pb half-life is a little long, 22yr.
- U-chain background can be measured as follows. But equilibrium can be broken easily for ^{210}Pb.
  - ICP-MS/GD-MS: ^{238}U, ^{232}Th.
  - HPGe: ^{226}Ra (^{238}U, ^{210}Pb). Sensitivity for ^{210}Pb is not high (~100mBq/kg).
- Surface and bulk contamination need to be separated. Alpha counter is the only detector to distinguish bulk from surface activity and could have better sensitivity. In XMASS ^{210}Pb contamination in oxygen free copper bulk is important. We tried to measure it.

Low background alpha-counter (XIA Ultra-Lo-1800)

We installed it at clean room in Kamioka underground to understand the surface background in Jan. 2015. We have been studied surface/bulk ^{210}Po contents.

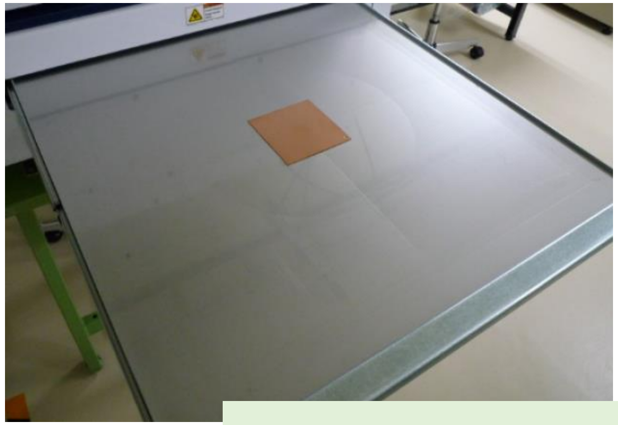
	specification
efficiency	>90% of 2pi
energy resolution	<9%FWHM at 4.6MeV
energy range	1-10MeV
sample size	707cm^2 (ϕ30cm disk), 1800cm^2 (42cm*42cm)
maximum sample weight	9kg
maximum sample thickness	6.3mm
achievable background level	10^{-4}alpha/cm^2/hr



signal
veto signal

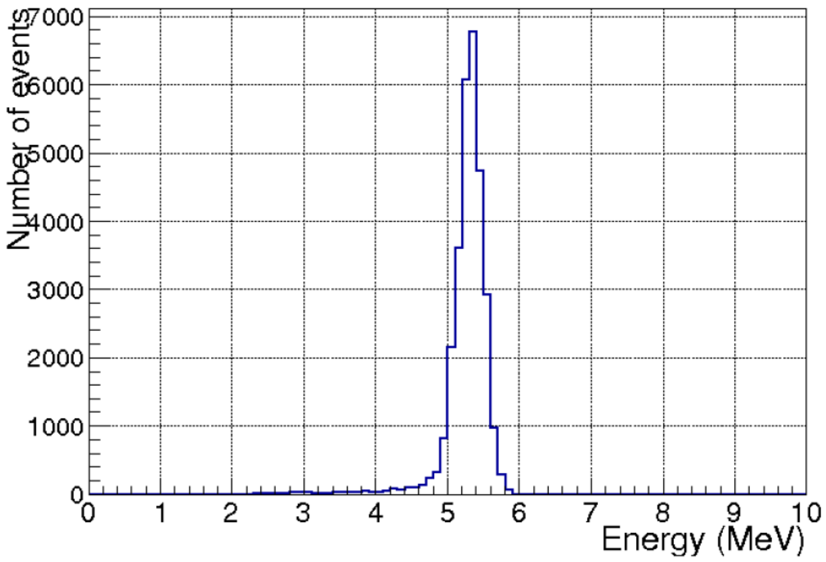
By measuring rise time (t_μ-t_0), surface event can be distinguished from event in Ar gas or ceiling.

Stability of alpha counter

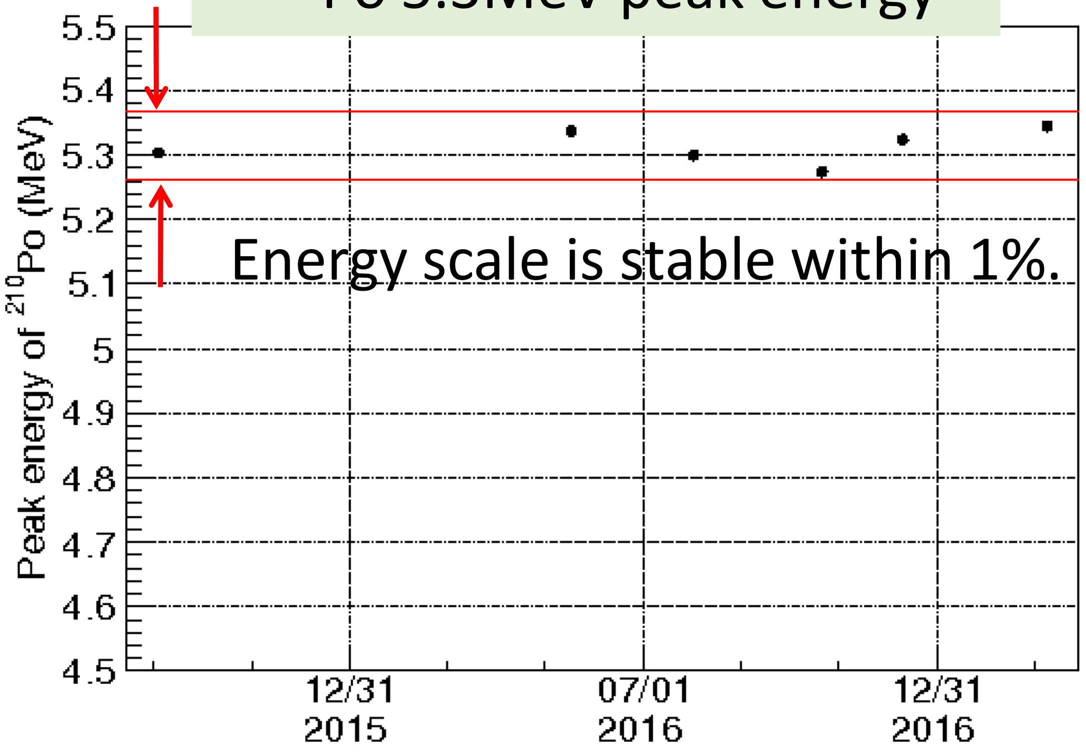


Using surface ^{210}Po 5.3MeV, we are constantly monitoring the energy scale.

Energy distribution of ^{210}Po 5.3MeV in one example



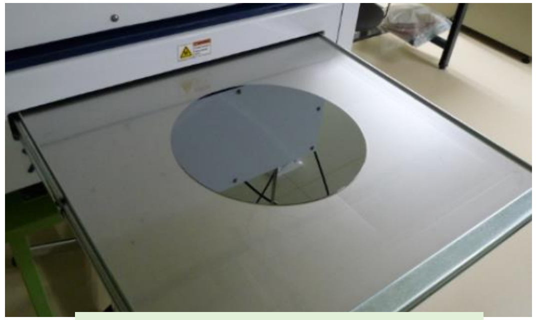
Time variation of ^{210}Po 5.3MeV peak energy



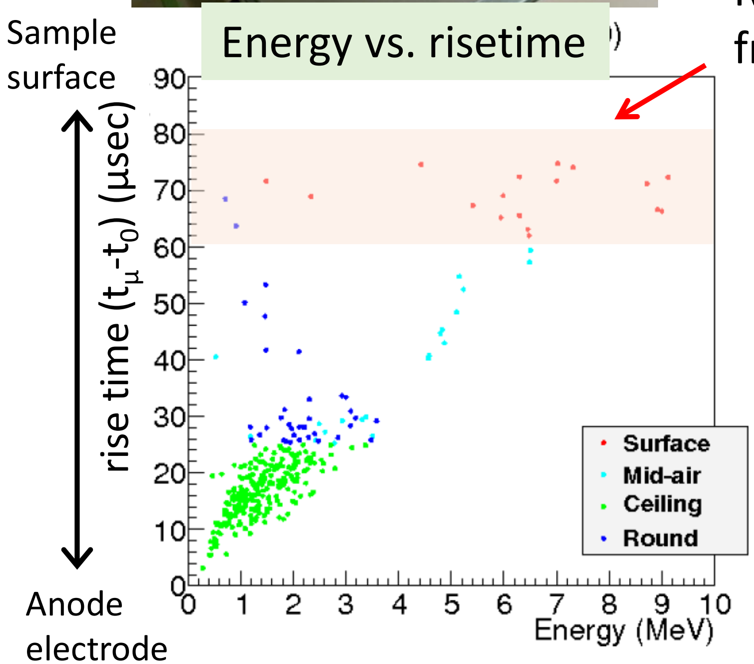
Energy scale is stable within 1%.

Background estimate
by Silicon wafer measurement

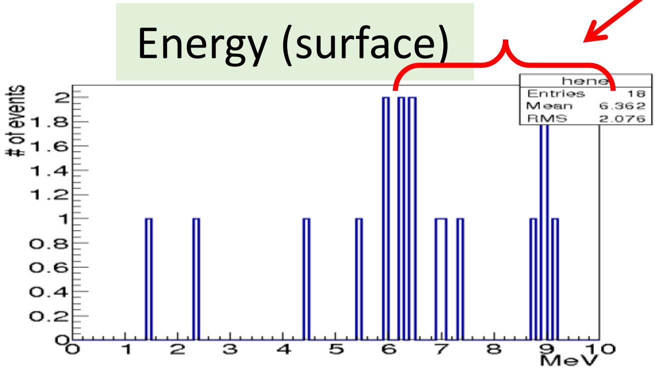
run#	218 (707cm^2 active region)
purge	0min (no sample change)
duration	11.69days (Nov. 17th, 2016 - Nov. 30th, 2016)
sample	Silicon wafer (polished silicon wafer made by Shin-Etsu chemical. Φ300mm and 775um in thick. Resistivity is 1~100Ωcm. Boron doped.) First 1 day data cut.



Red: alpha events from sample surface

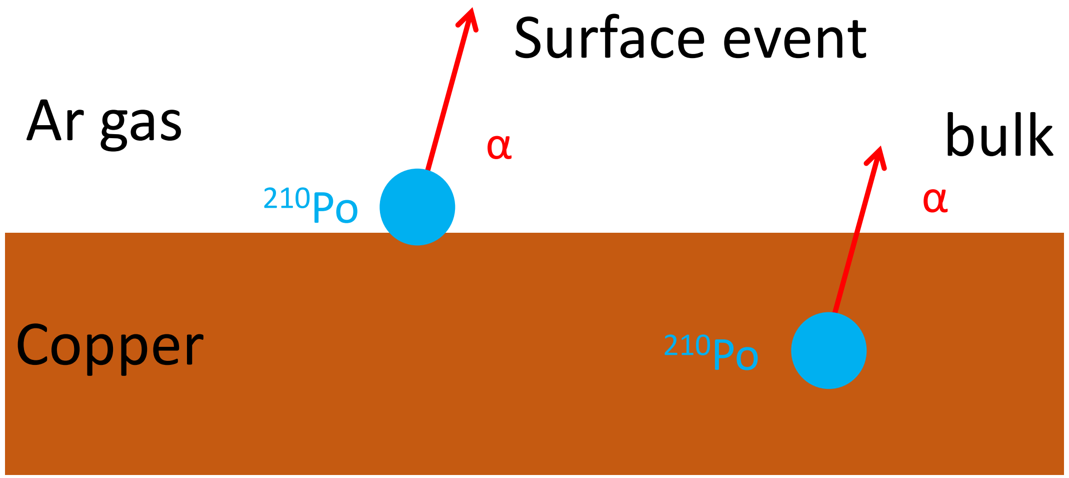


Background of daughter nuclei from ^{222}Rn and ^{220}Rn in Ar gas

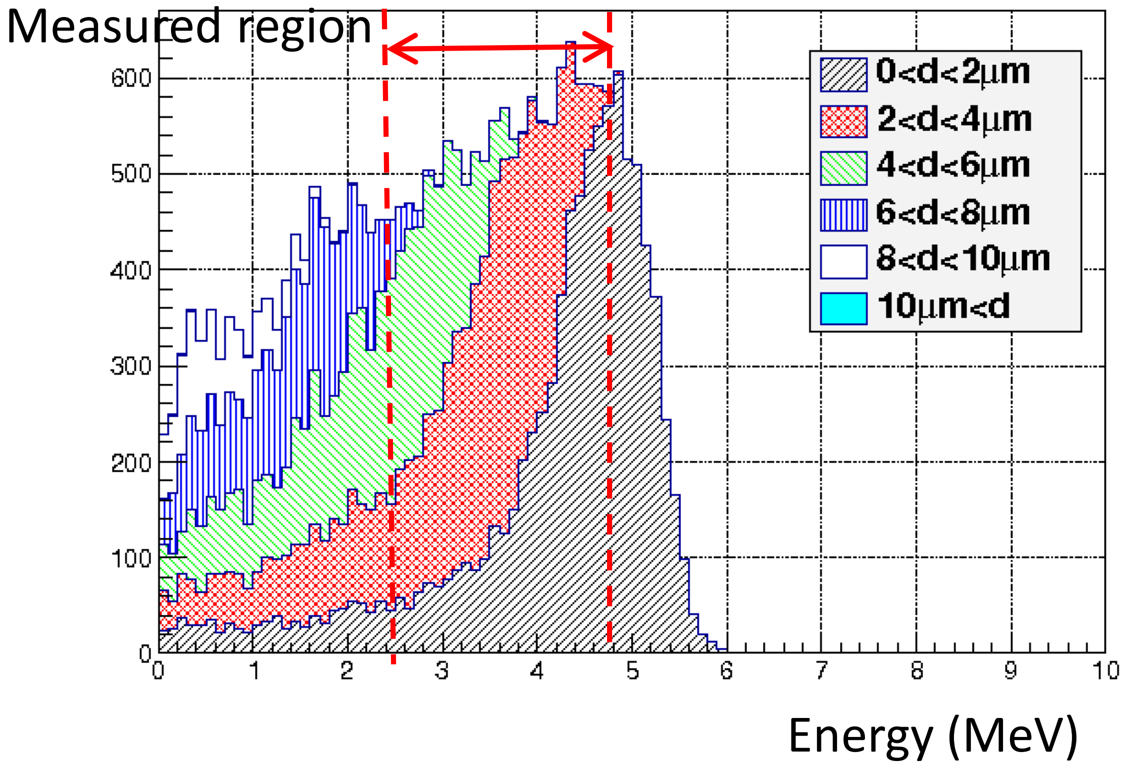


Background emissivity is (1.1±0.3)x10^{-4}α/cm^2/hr. They are mainly from ^{222}Rn and ^{220}Rn and their energy is higher. In 2.5<E<4.8MeV, background emissivity is (5.6±5.6)x10^{-6}α/cm^2/hr.

How the bulk signal looks like in alpha counter?



Energy distribution (5.3MeV copper bulk alpha MC simulation)



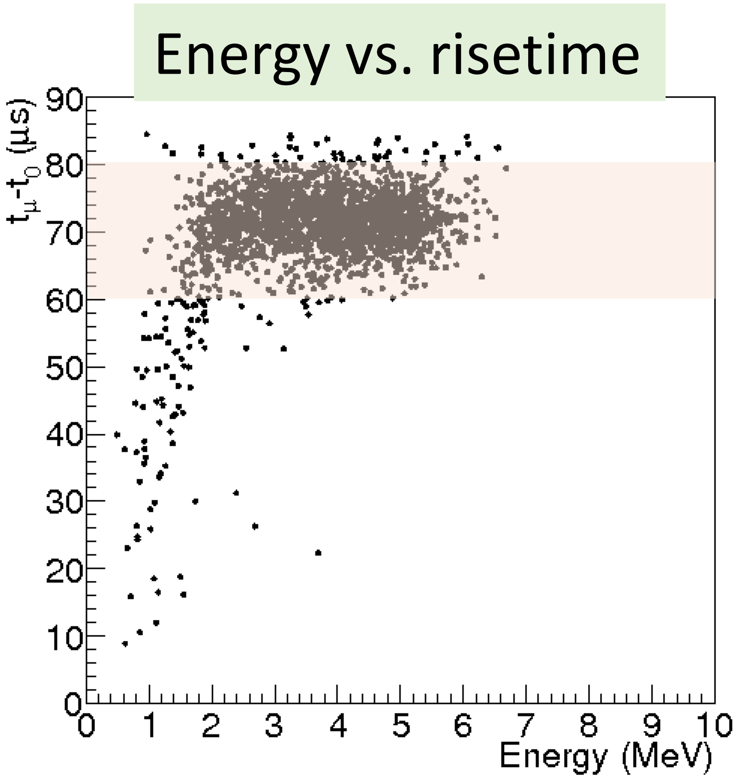
- Alpha range in copper is ~10μm.
- Energy distribution is continuous.
- According to the MC estimate, events in 2.5<E<4.8MeV come from 2<d<6um mainly (d: distance from alpha generation point to the copper surface.).
- Conversion factor from emissivity to bulk contamination is 249 (Bq/kg)/(alpha/cm2/hr) in 2.5<E<4.8MeV

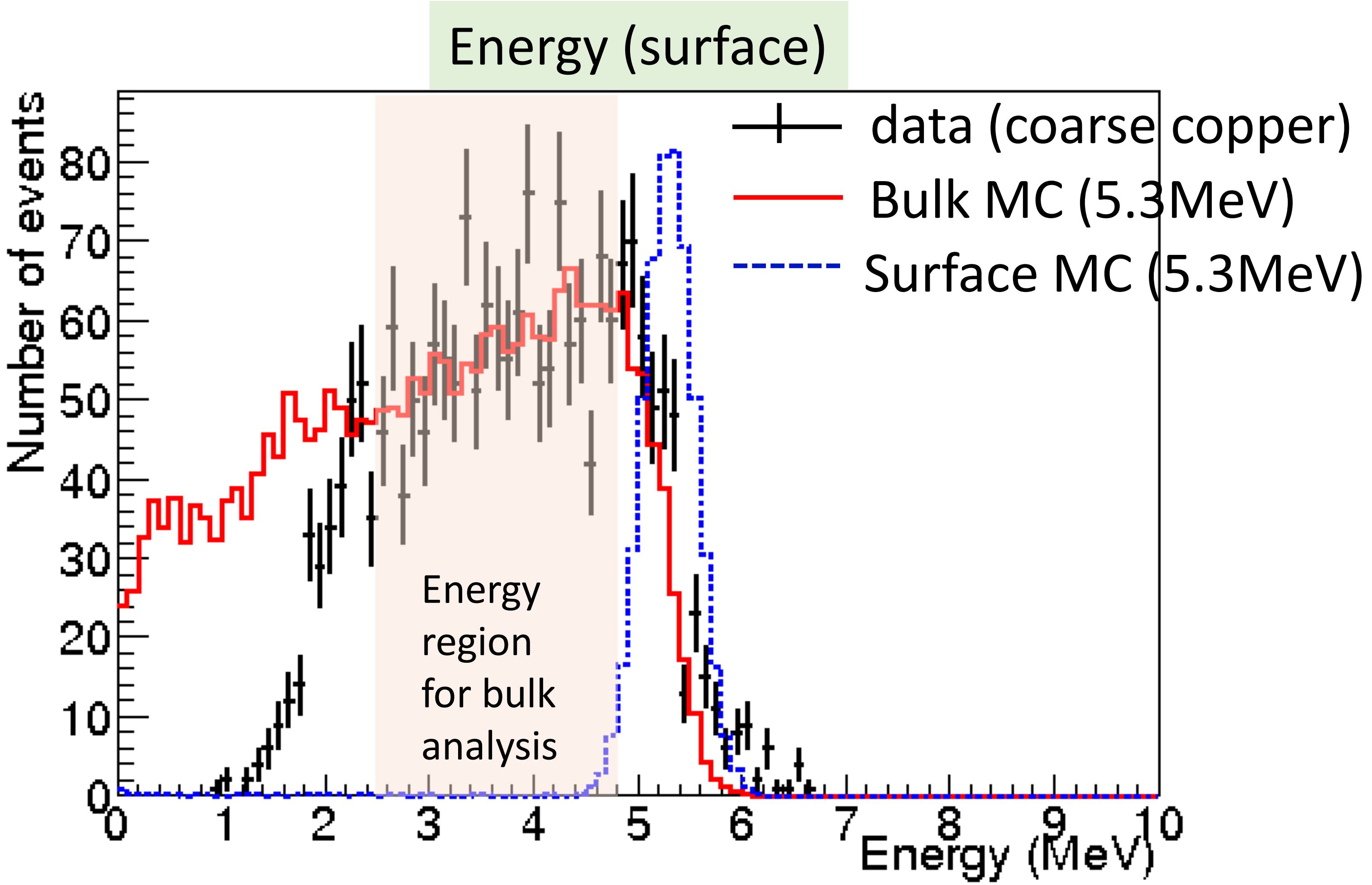
→ Background level corresponds to 1.4mBq/kg

To measure ^{210}Pb/^{210}Po contamination in copper bulk:

1. Background from the alpha particle counter itself should be small.
   - UltraLo-1800 was installed underground. Background level in 2.5<E<4.8MeV is (5.6±5.6)x10^{-6}α/cm^2/hr.
2. Radioactive contamination on the sample surface should be minimized.
   - We apply electro-polish to remove surface RI. Also we confirmed the surface activity before EP is not large.
   - Sample exchange is done in a clean room.
   - We kept the samples in the EVOH (ethylene-vinylalcohol copolymer) bag during storage.
   - We don't use the surface energy region (around 5.3MeV). In 2.5<E<4.8MeV, contribution from surface events is <5%.
3. The sample surface roughness should be much smaller than track length of alpha-rays (~10μm).
   - After the EP, surface roughness is <<1μm by laser microscope check.
4. Alpha-rays from radioactive nuclei other than ^{210}Po should be negligible.
   - By GD-MS, we confirmed <100ppt in ^{238}U and ^{232}Th → <1.2mBq/kg in ^{238}U and <0.4mBq/kg in ^{232}Th.

Large ^{210}Pb contaminated sample (coarse copper)





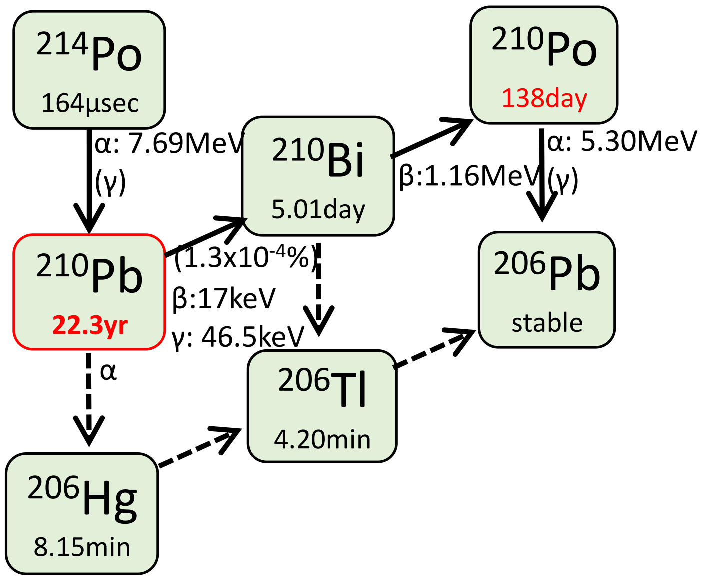
- In coarse copper sample MMC provided, large ^{210}Po activity in bulk region is seen.
- Energy selection for bulk analysis is determined to 2.5<E<4.8MeV through this comparison
- Shape of energy distribution in data agrees with that of MC.

Calculation method to derive ^{210}Pb/^{210}Po contamination

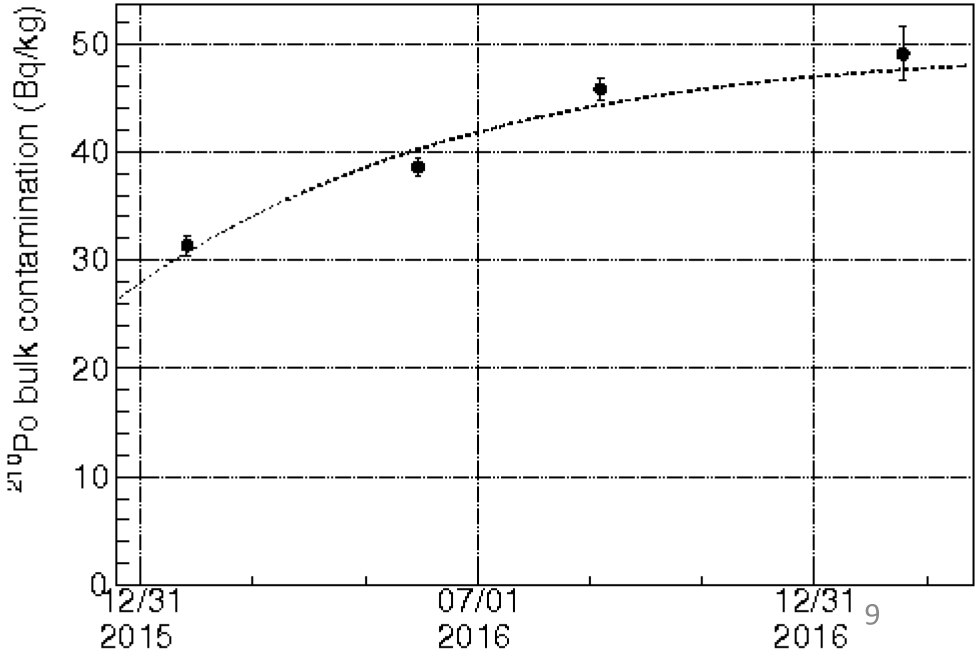
$$N_0(t) = N_0(0)e^{-\frac{t}{\tau_0}}$$

$$N_1(t) = N_0(0)\frac{\frac{1}{\tau_0}}{\frac{1}{\tau_1}-\frac{1}{\tau_0}}\left(e^{-\frac{t}{\tau_0}} - e^{-\frac{t}{\tau_1}}\right) + N_1(0)e^{-\frac{t}{\tau_1}}$$

τ_0 =32.17yr (lifetime of ^{210}Pb)
τ_1 =0.55yr (lifetime of ^{210}Po)
Ns are numbers measured by alpha counter (after background subtraction. Background is measured by Si wafer sample).
N_0 (t): amount of ^{210}Pb in 1kg at t=t
N_1 (t): amount of ^{210}Po in 1kg at t=t
t=0: the time when the samples are delivered.

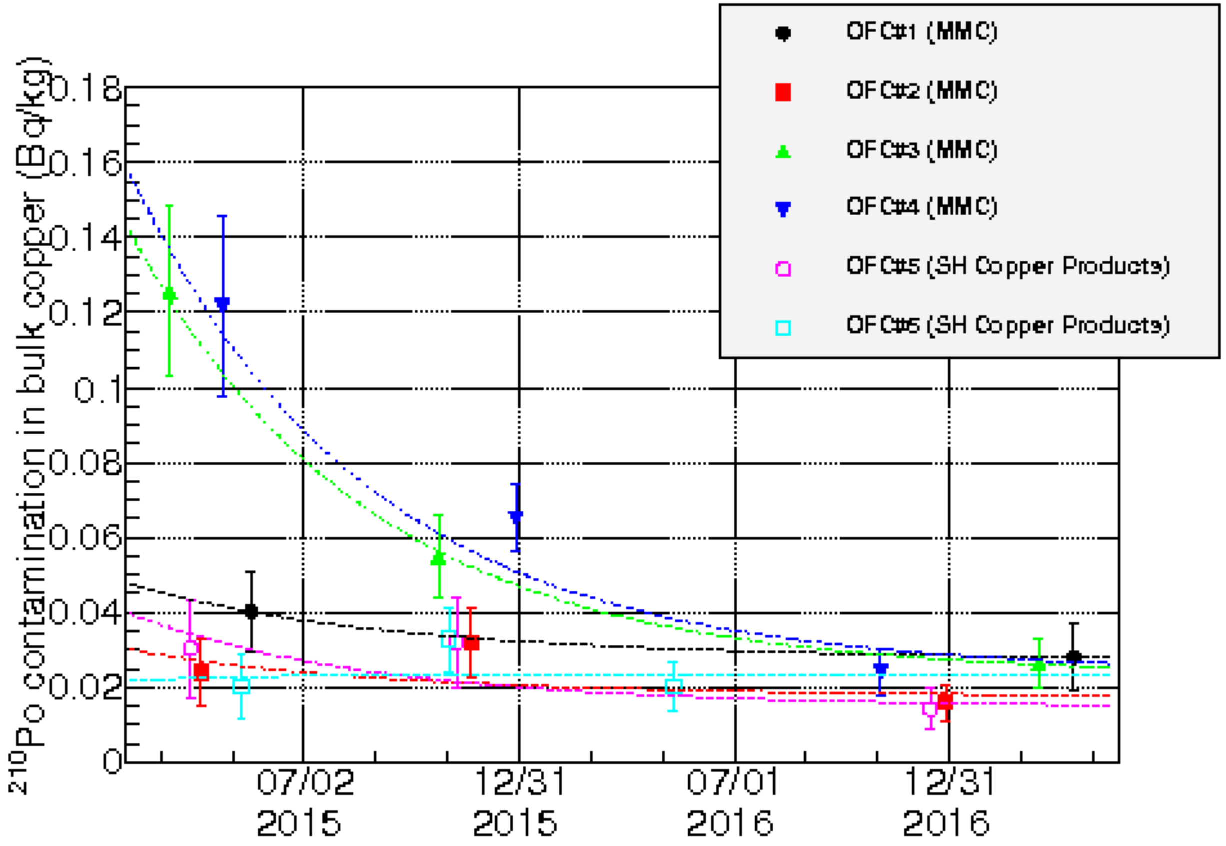


Time evolution (coarse copper)



Equilibrium is largely broken.
^{210}Pb: 52.1±1.2Bq/kg, ^{210}Po: 14.8±2.1Bq/kg
The result is consistent with that of HPEGe:
55.6±1.5(stat.)+16.7-5.6(syst.)Bg/kg
→ We confirmed that this method work well.

OFC (oxygen free copper, C1020) samples (rolled)

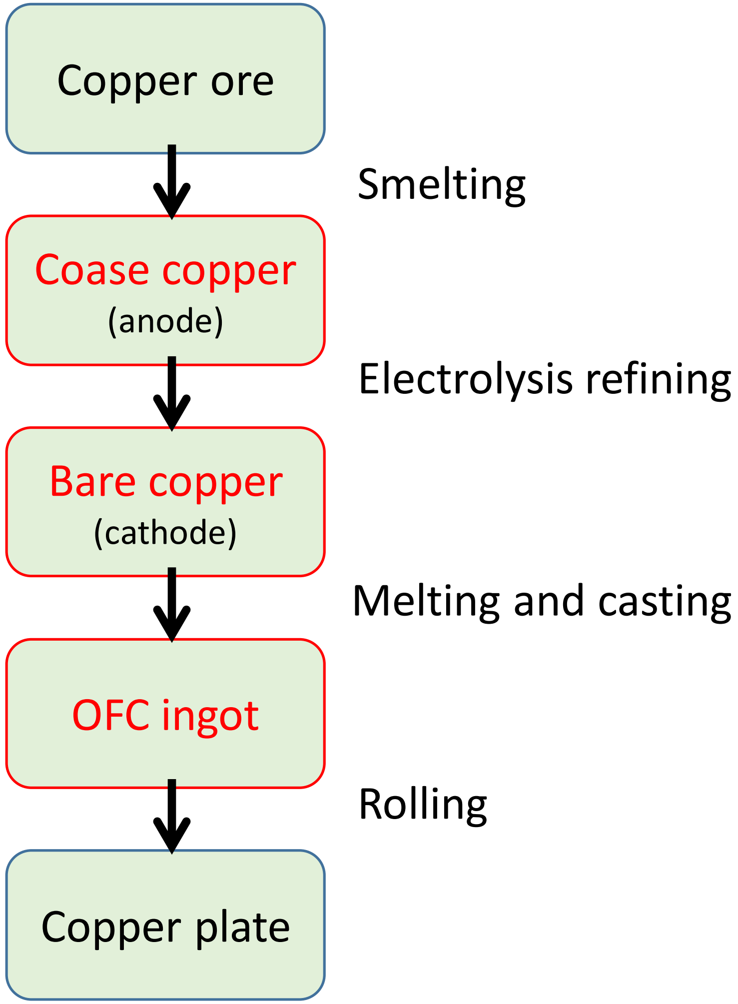


	^{210}Pb (mBq/kg)	^{210}Po (mBq/kg)
OFC#2 (MMC)	18.6±5.1	30.3±12.5
OFC#3 (MMC)	24.8±6.8	144±23
OFC#4 (MMC)	25.7±5.9	160±14
OFC#5 (SH copper products)	15.6±5.7	40.2±16.7
OFC#6 (SH copper products)	24.5±7.4	21.8±15.2

MMC: Mitsubishi Material Corporations

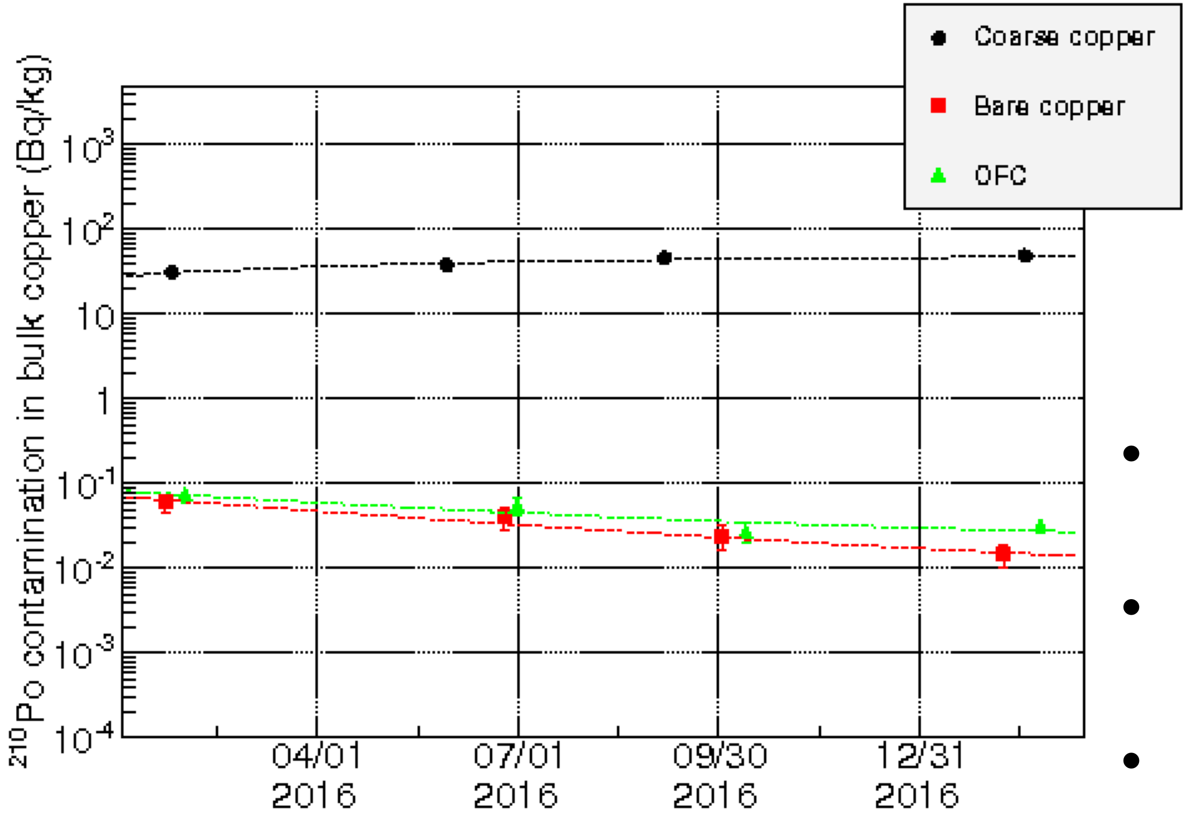
- Equilibrium is largely broken. Though ^{210}Po is widely ranged (21.8~160mBq/kg), ^{210}Pb are within 15.6~25.7mBq/kg. This is the first measurement of ^{210}Pb in OFC bulk.
- OFC Class 1 was also investigated. ^{210}Pb contamination is same level as OFC C1020.

Copper production



MMC (Mitsubishi Material Corporation) kindly cooperated for our study to understand the origin of ^{210}Pb in OFC. They provided us copper samples in each production step: coarse copper, bare copper, and OFC (ingot). All samples are machined (NOT rolled) and electro-polished (50um). We measured several times.

Coppers in each production step

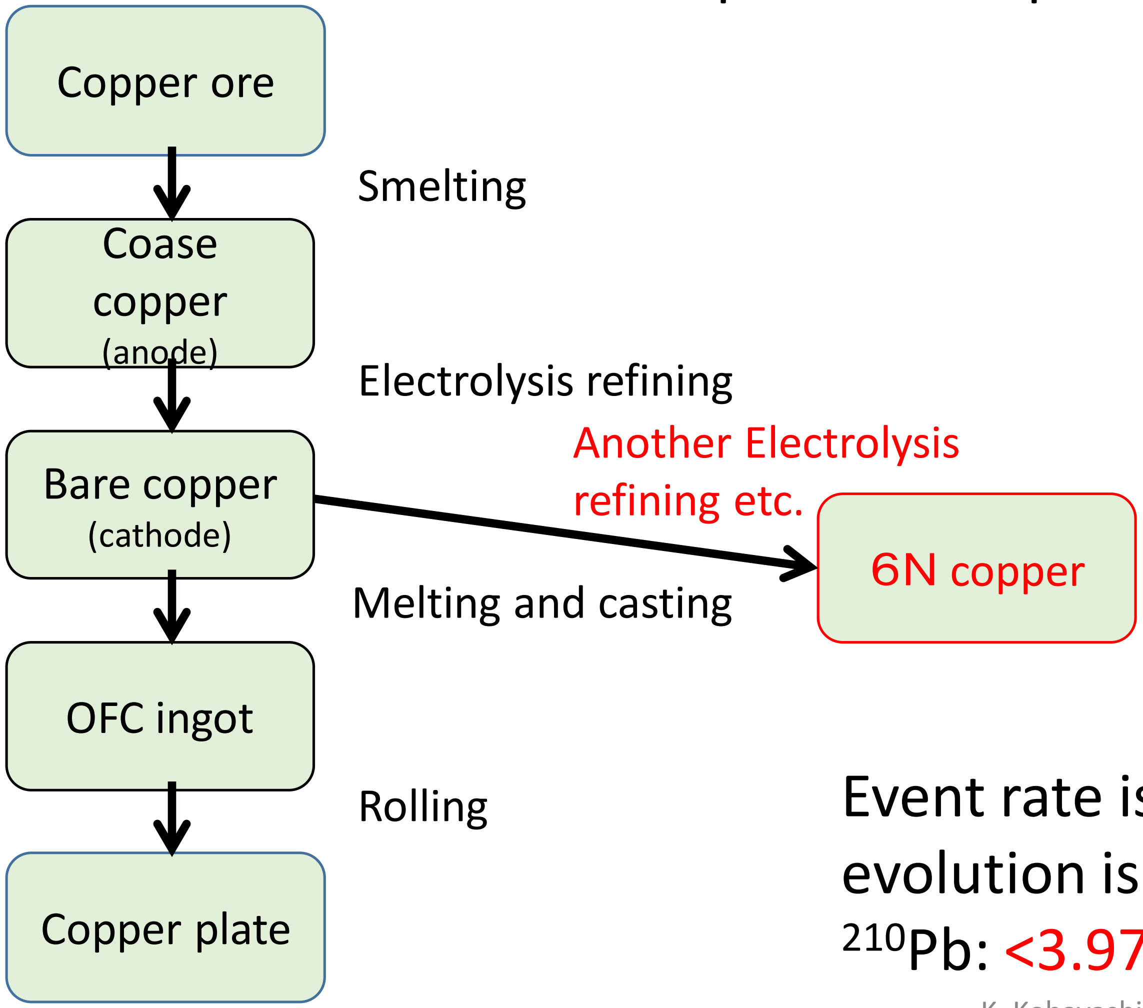


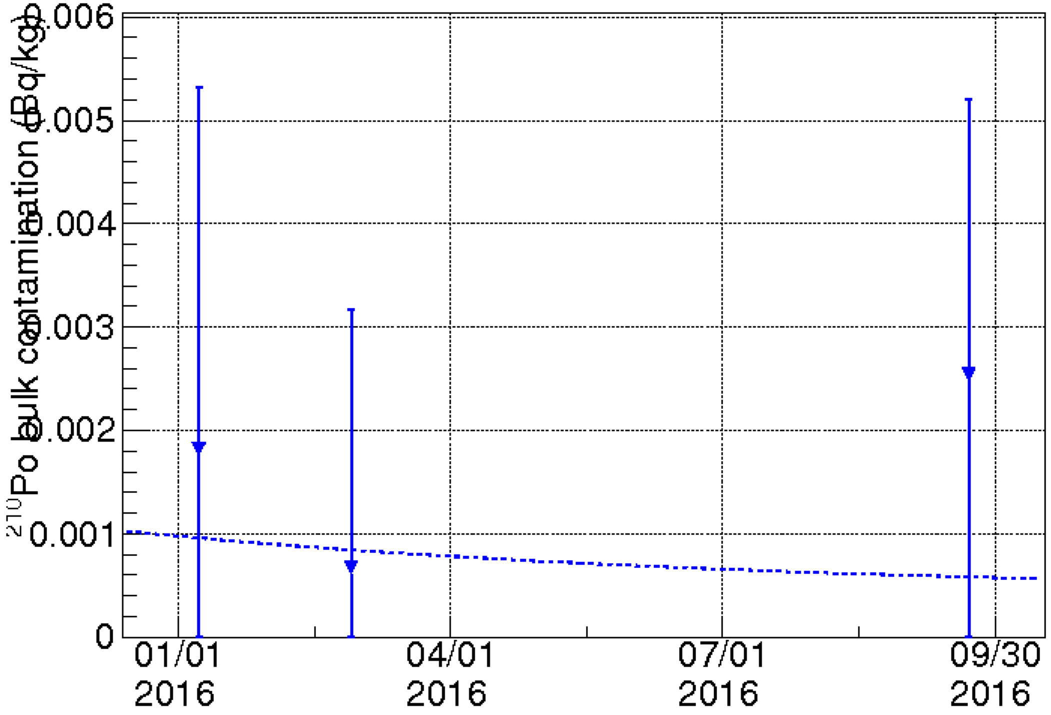
	^{210}Pb (mBq/kg)	^{210}Po (mBq/kg)
Coarse copper	52.1±1.2	14.8±2.1
Bare copper	7.0±5.8	107±24
OFC ingot	20.7±7.2	117±27

- ^{210}Pb contamination in Bare copper and OFC is ~1/1000 of coarse copper.
- OFC ingot is consistent with OFC samples (rolled) measurement (15.6-25.7mBq/kg).
- It indicates most of the ^{210}Pb is removed in the electrolysis process but the same does not seem to be true for the the daughter isotopes, ^{210}Bi and ^{210}Po.

6N copper

6N (99.9999% pure copper) is made by another cycle of electrolysis in clean environment. -> Low ^{210}Pb contamination is expected. We purchased this 6N copper from MMC.



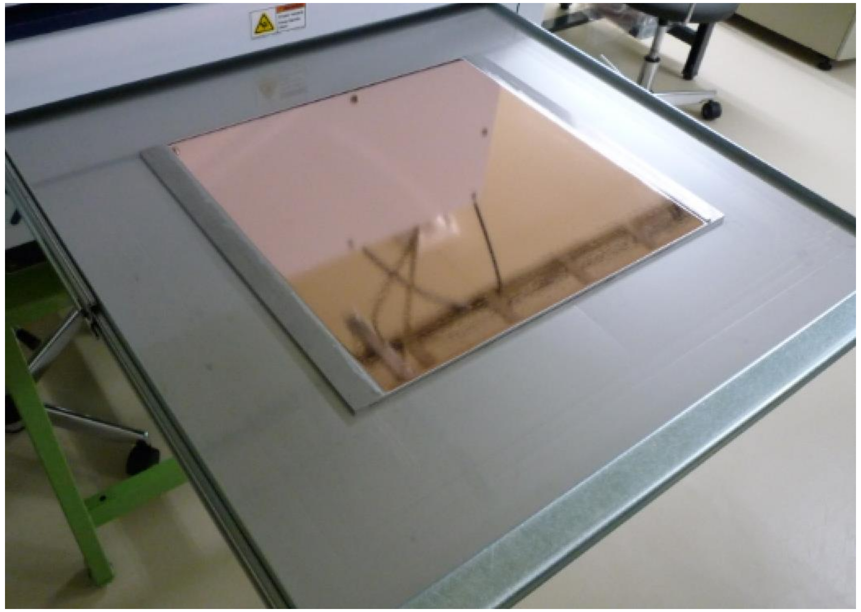


Event rate is consistent with background. No time evolution is observed.

^{210}Pb: <3.97mBq/kg, ^{210}Po: <5.06mBq/kg

Electro-formed copper

- Electro-formed copper plate is made by a different company from MMC. 0.5mm thickness copper is electro-formed (345mmx345mm) on SUS plate (35cmx40cmx4mm) and is electro-polished ~0.1mm .
- Event rate is consistent with background.
- ^{210}Pb: <2.8mBq/kg.



6N and electro-formed copper are the lowest ^{210}Pb contamination copper among our measured samples.

Application to other materials

- This method to measure ^{210}Pb and ^{210}Po contamination in copper bulk could apply to other materials. We generate MC simulations and estimate the sensitivity. We checked aluminum, stainless steel, titanium, tungsten, quartz glass, PTFE. The sensitivities are similar to copper, 0.75-2.29mBq/kg.
- Because the neutron from (α,n) reaction in material bulk is one of the expected background in the future dark matter experiment. It is important to measure not only upper U-chain contamination, but also ^{210}Pb contamination. This method is useful for the ^{210}Pb contamination understanding.

summary

- We established a new method to measure the ^{210}Pb and ^{210}Po contamination in copper bulk. The sensitivity to ^{210}Pb and ^{210}Po contamination is a few mBq/kg.
- Due to this high sensitivity, we succeeded in identifying and measuring the ^{210}Pb and ^{210}Po contamination in OFC bulk (^{210}Pb:15-26 mBq/kg). This ^{210}Pb is interpreted as a small residual from the electrolysis process based on the investigation of the coppers in each production step.
- The 6N and the electro-formed copper contaminate ^{210}Pb one order or more lower than OFC.
- The method we established here to measure the copper bulk contamination could apply to other materials. Our method can improve material screening and preparation for low background experiments.

Paper is now in preparation

backup

Laser microscope check (Ra)

	Before EP	After EP
Coarse copper	0.463um (2015.12.09)	0.684um (2016.01.27) 0.186um (2016.02.05)
Bare copper	0.113um (2015.12.09)	0.015um (2016.01.25)
OFC	0.143um (2015.12.09)	0.096um (2016.02.04)
6N copper	1.906um (2015.10.29)	0.073um (2016.01.12)
OFC (SH copper)	0.349um (2015.10.29)	
RFB copper		0.142um (2015.10.06)
6N copper (10cmx10cm)		0.017um (2016.05.02)
OFC EP+EP		0.017um (2016.01.18)
Shimosada copper	0.382um (2016.11.29)	

GDMS result

All the samples (coarse, bare, OFC, 6N copper) are investigated by GDMS by company.

	Coarse coper	Bare copper	OFC	6N copper
S	27	6.7	3.5	0.1
Cl	0.008	9.1	0.02	0.006
Ag	1100	8.6	16	0.15
^{208}Pb	2200	0.19	0.98	0.002
^{209}Bi	240	0.06	0.26	0.001
^{232}Th	<0.0001	<0.0001	<0.0001	<0.0001
^{238}U	<0.0001	<0.0001	<0.0001	<0.0001

- ^{238}U and ^{232}Th are all <0.1ppb.
- ^{208}Pb ratio (coarse, bare, OFC) is similar to ^{210}Pb of alpha counter measurement. -> ^{210}Pb in 6N copper is expected to be ~0.05mBq/kg

Summary of sensitivity study for bulk measurement by MC simulation

	Atomic mass (g/mol)	Density (g/cm^3)	Conversion factor ($1alpha/cm^2/hr(2.5<E<4.8MeV)=??Bq/kg$)	5.3MeV alpha-ray Range estimated by MC (μm)	Current sensitivity by alpha counter (mBq/kg)
Aluminum	26.98	2.70	361	22.0	2.02
Silicon	28.08	2.329	375	24.3	2.10
Titanium	47.87	4.506	306	15.7	1.72
Iron	55.85	7.874	282	9.9	1.58
SUS304		8.03	280	9.6	1.57
Kovar		8	272	10.2	1.52
copper	63.545	8.94	249	10.0	1.39
Zinc	65.38	7.14	248	12.6	1.39
Tin	118.7	7.365	198	15.3	1.11
Tungsten	183.84	19.25	134	8.5	0.75
Lead	207.2	11.34	139	15.1	0.78
Quartz glass		2.5	359	30.8	2.01
PTFE		2.14	410	24.3	2.29

All the Sensitivities (0.75-2.29mBq/kg) are similar to copper (1.39mBq/kg).