FNP Team

TIPP'17

Low Radiation Techniques

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Outline

- Introduction
- Low Radioactivity Assay Techniques
  - Gamma-ray spectroscopy
  - Detection of radon
  - Underground alpha spectroscopy
- Purification/Cleaning Techniques
  - Purification of gases
  - Surface cleaning techniques
- Summary

Introduction

Low-radiation (Low-level) techniques: experimental techniques, which allow to investigate very low activities of natural and/or artificially produced radio-isotopes.

- material screening (HPGe spectroscopy, ICP-MS, AMS, NAA)
- surface screening (α,β,γ spectroscopy)
- study of radioactive noble gases (emanation, diffusion)
- purification techniques (gases, liquids)
- background events rejection techniques (Active veto, PSD)
- modeling of background in experiments (Monte Carlo)

Low-level techniques are "naturally" coupled to the experiments looking for rare nuclear processes at low energies (detection of neutrinos, search for dark matter, search for 0ν2β decay, search for proton decay, ...), where the backgrounds identification and reduction plays a key role.

Low-level radioactivity

In BOREXINO we are detecting ~50 ν/d in 100 ton of LS

↓ **S/B_{int} ~1**

$A_{spec.}$ ~50 dpd/100 ton

↓

$A_{spec.} \sim 6 \times 10^{-9}$ Bq/kg (~0.5×10^{-15} g/g U = 0.5 ppq)

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$A_{spec.}$ (water) ~1 Bq/kg

↓ **Activity reduction factor needed**

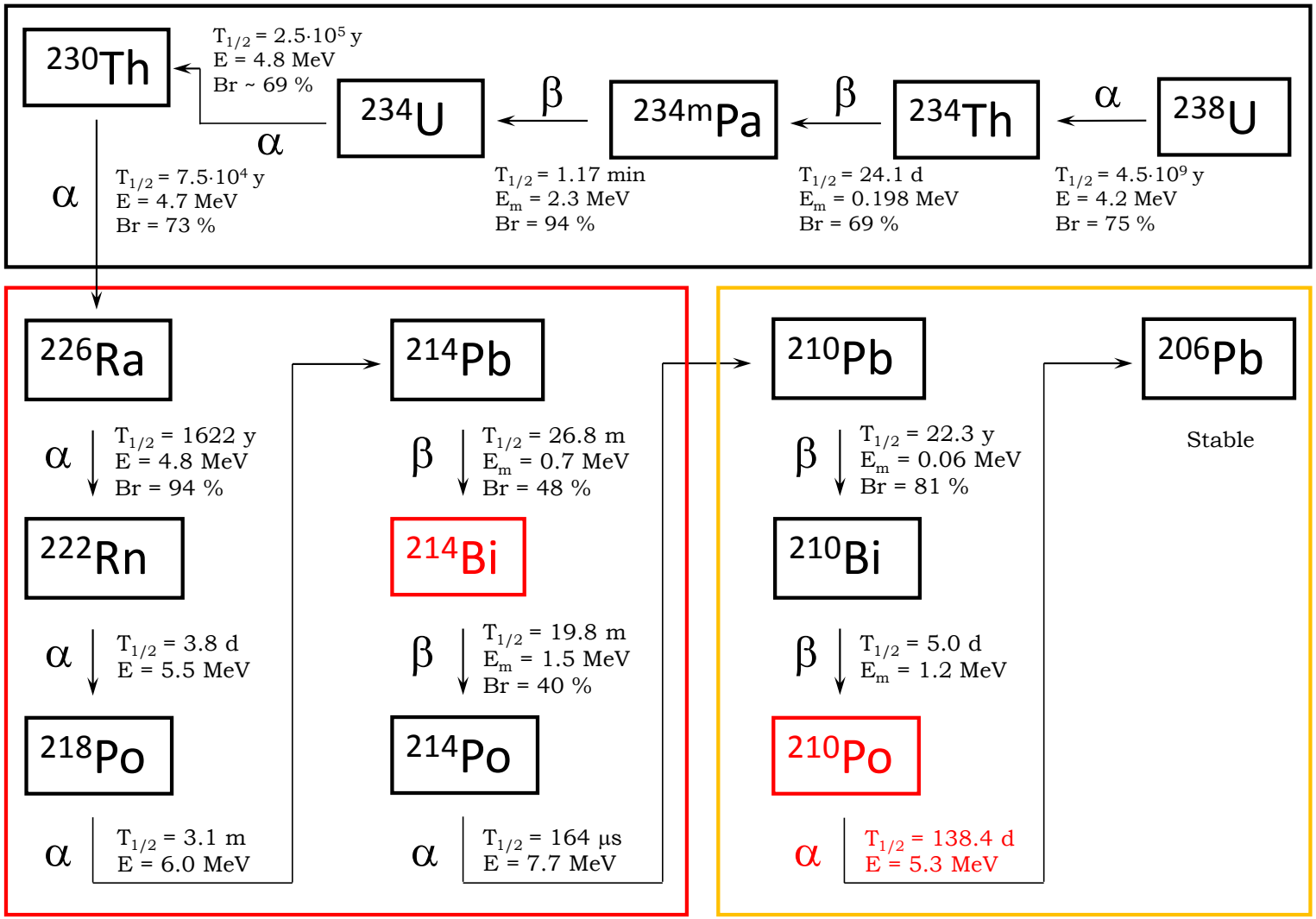
R ~ 10^{10} !!

Requirements for external components usually more relaxed (~ppt level)

Background mitigation techniques

- Graded shielding: traveling inward to the center, each component is protected from external radiation by the preceding one
- The radio-purity level is increasing towards the center
- Active (definition of FV, Čerenkov veto) and passive (buffer volume) suppression of external radiation
- Careful selection of construction materials and detector components with respect to content of radioactive isotopes, ^{222}Rn emanation and permeability
- Preventing surface contamination
- Application of appropriate purification (liquids, gases) and cleaning techniques

^{238}U decay chain



ICP-MS

- ICP-MS: Inductively Coupled Plasma Mass Spectrometry
- Presently very well established and very popular analytical technique
- ICP-MS supported by a proper sample preparation methodology allows for the analysis of various materials and specialty components important in ultralow background physics experiments
- Assay of materials, which can be put into liquid form (polymers, electronic components, wires/cables, metals, *etc.*)
- Extremely sensitive, fast (couple of days for a measurement), requires small amounts of sample (<1 g)
- Commercially available instruments can reach <0.1 ppt sensitivity for U/Th (<1 μBq/kg)

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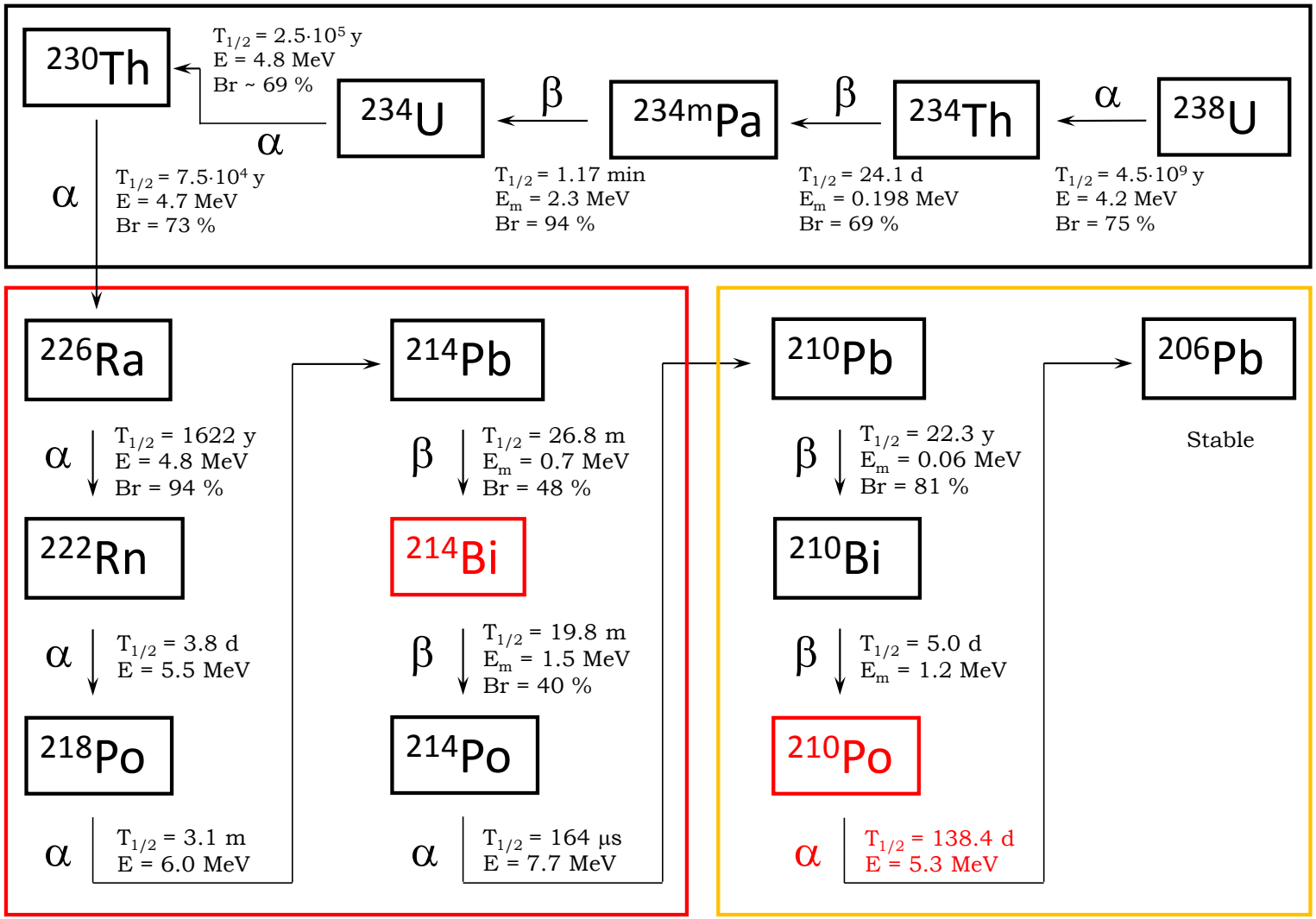
- ^{238}U may not be in secular equilibrium with ^{226}Ra !

^{238}U decay chain



Gamma-ray spectroscopy

Gamma-ray spectroscopy based on HPGe detectors is one of the most powerful techniques to identify γ-emmiters (Ra in the U/Th chain, ^{40}K, ^{60}Co,...).

- excellent energy resolution (~ 2 keV around 1.33 MeV)
- high purity (HPGe) detectors (low intrinsic background)

In order to reach high sensitivity it is necessary:

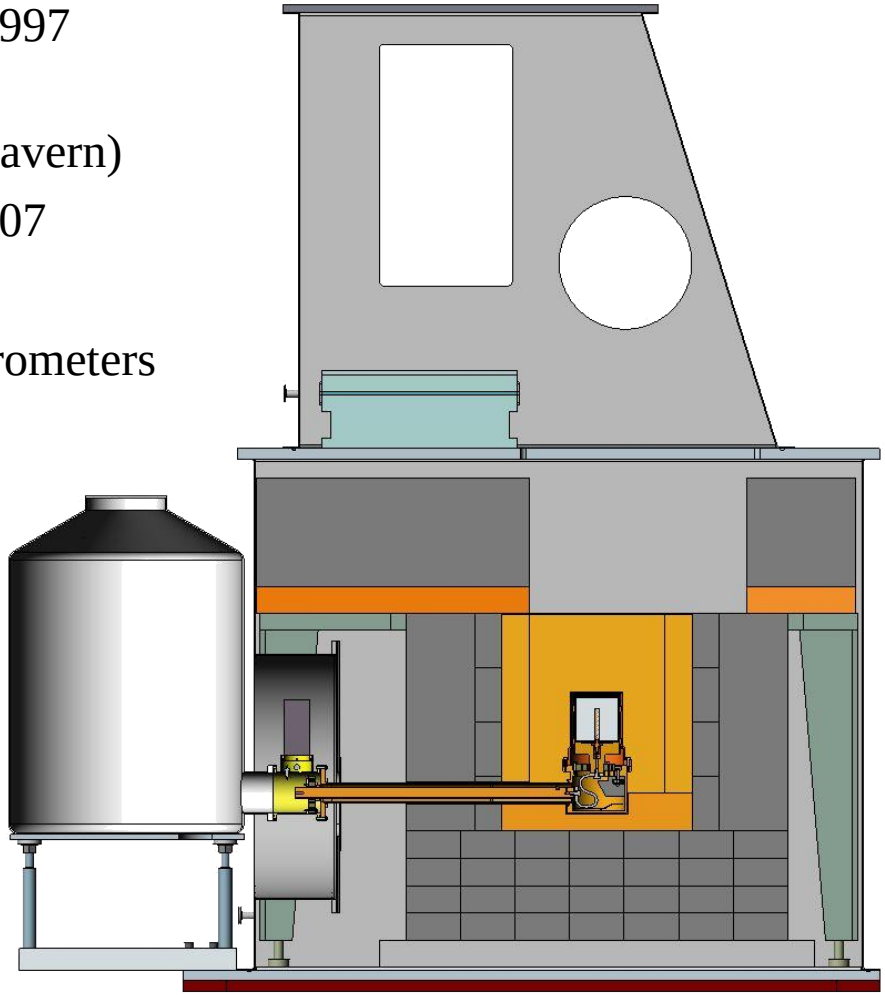
- reduce backgrounds originating from external sources
  - active/passive shielding (underground locations)
  - reduction of radon in the sample chamber
- assure (reasonably) large volumes of samples
- assure precise calculations/measurements of detection efficiencies

Highly sensitive Ge spectroscopy is a perfect tool for material screening

Gamma-ray spectroscopy

GeMPIs at GS (3800 m w.e.)

- GeMPI I operational since 1997 (MPIK-HD)
- GeMPI II built in 2004 (MCavern)
- GeMPI III constructed in 2007 (MPIK-HD/LNGS)
- Worlds most sensitive spectrometers



GeMPI I:

- Crystall: 2.2 kg, ε_r = 102 %
- Bcg. Index (0.1-2.7 MeV): 6840 cts/kg/year
- Sample chamber: 15 l

Sensitivity:
~10 μBq/kg

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Astrop. Phys. 18 (2002) 1

Survey of different materials

Sample	Description	^{226}Ra [mBq/kg]	^{228}Th [mBq/kg]	^{40}K [mBq/kg]
Stainless Steel	AISI304L: SSS	4.6 ± 0.9	11.4 ± 1.1	< 14
	SS for pipes	< 14	< 10	< 34
	SS for flanges	6.2 ± 1.2	6.5 ± 1.6	< 13
PMTs inner parts	Dynodes	< 280	450 ± 163	< 240
	Ceramic plates	170 ± 50	310 ± 60	960 ± 450
	Al for dynodes	1190 ± 100	980 ± 80	2800 ± 600
PMTs ancillary parts	Mu metal	57 ± 20	< 27	< 180
	Volt. div. board	170 ± 60	80 ± 40	770 ± 360
	Voltage divider	680 ± 30	320 ± 20	3200 ± 320
Glass	Sand for glass	40 ± 3	< 3.1	< 25
	ETL LB glass	820 ± 230	130 ± 12	500 ± 120
	Base glass	520 ± 90	410 ± 90	(2.2 ± 0.6) ·10^5

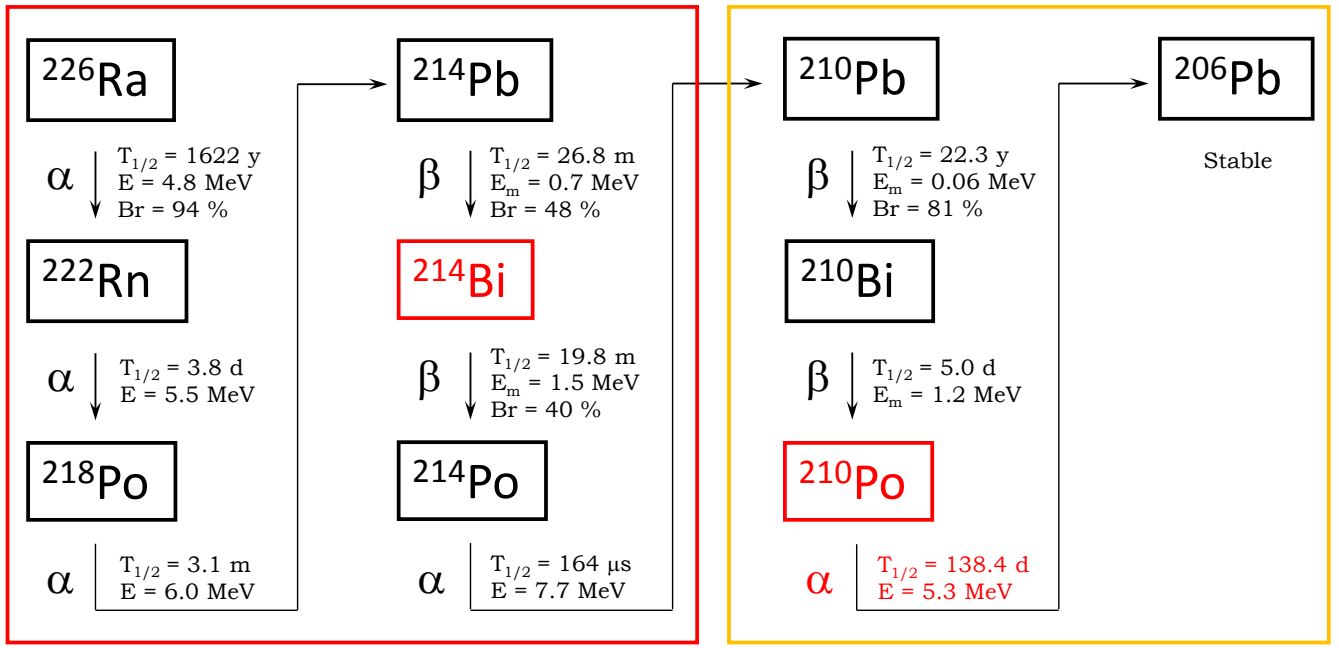
Astroparticle Physics 18 (2002) 1

Survey of stainless steel for GERDA

No.	Specific activity [mBq/kg]			
	^{228}Th	^{226}Ra	^{40}K	^{60}Co
1 D	5.1 ± 1.0	2.9 ± 1.0	< 3.9	6.5 ± 0.5
2 G	< 0.27	< 0.35	< 1.1	13.0 ± 0.6
3 D	1.1 ± 0.4	< 0.84	< 3.3	15.1 ± 0.5
4 D	< 2.6	< 2.2	< 6.2	14.4 ± 1.0
5 D	< 1.1	< 1.2	< 2.8	11.6 ± 0.5
6 D	< 0.8	< 0.6	< 1.7	16.7 ± 0.4
7 G	< 0.20	< 1.3	< 2.8	45.5 ± 2.1
8 G	< 0.11	< 0.24	< 0.93	14.0 ± 0.1
9 G	< 0.41	< 0.74	< 1.1	13.8 ± 0.7
10 G	< 1.0	< 1.3	< 6.8	17.1 ± 0.7
11 G	1.5 ± 0.2	1.0 ± 0.6	< 0.81	18.3 ± 0.7

NIM A 593 (2008) 448

^{222}Rn detection

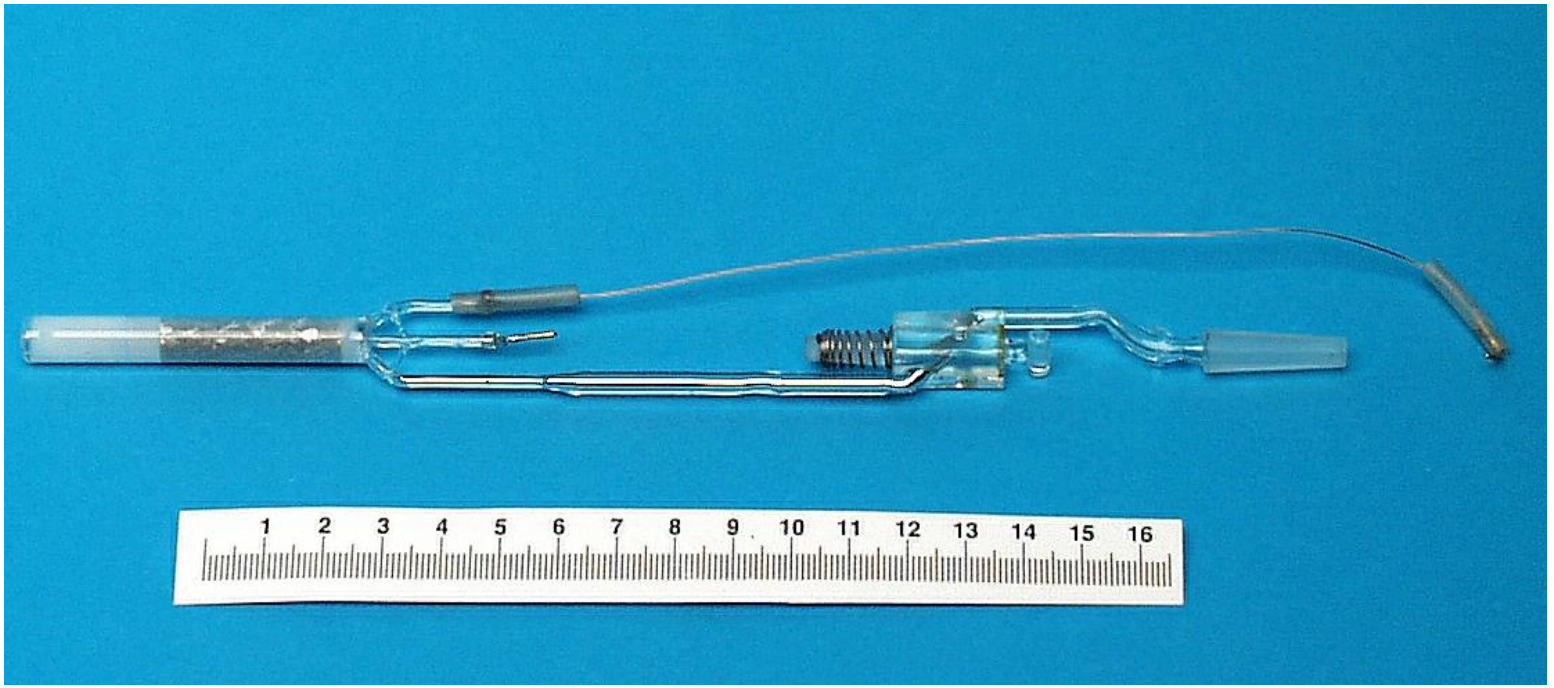


Radon ^{222}Rn and its daughters form one of the most dangerous source of background in many experiments

- inert noble gas
- belongs to the ^{238}U chain (present in any material)
- high diffusion and permeability
- wide range of energy of emitted radiation (with the daughters)
- surface contaminations with radon daughters (heavy metals)
- broken equilibrium in the chain at ^{210}Pb level

^{222}Rn detection

Proportional counters (MPIK-HD)



- Developed for the GALLEX/GNO experiment
- Hand-made at MPIK-HD (~1 cm^3 active volume)
- In case of ^{222}Rn only α-decays are detected
- 50 keV threshold
  - bcg: 0.1 – 2 cpd
  - total detection efficiency of ~ 1.5
- Absolute detection limit ~ 30 μBq (15 atoms)

^{222}Rn detection

Cryogenic radon detector

- Cryogenic adsorption of Rn with subsequent counting of the alpha decays of Rn and the daughters
- Simultaneous and real-time detection of emanated ^{222}Rn **and ^{220}Rn** (under vacuum)
- **Emanation tests of small samples**
- **Attachable to vacuum vessels for direct emanation measurements**
- Volume of ~2 L

- Detection efficiency : 25 %
- Resolution (5.5 MeV) : ~40 keV

- **Background (^{222}Rn) : ~0.8 cpd**
- **Detection limit : ~20 µBq (10 ^{222}Rn atoms)**

Tests of the DARKSIDE-50 cryostat

Empty cryostat

A_{sat} = (0.14 ± 0.04) mBq

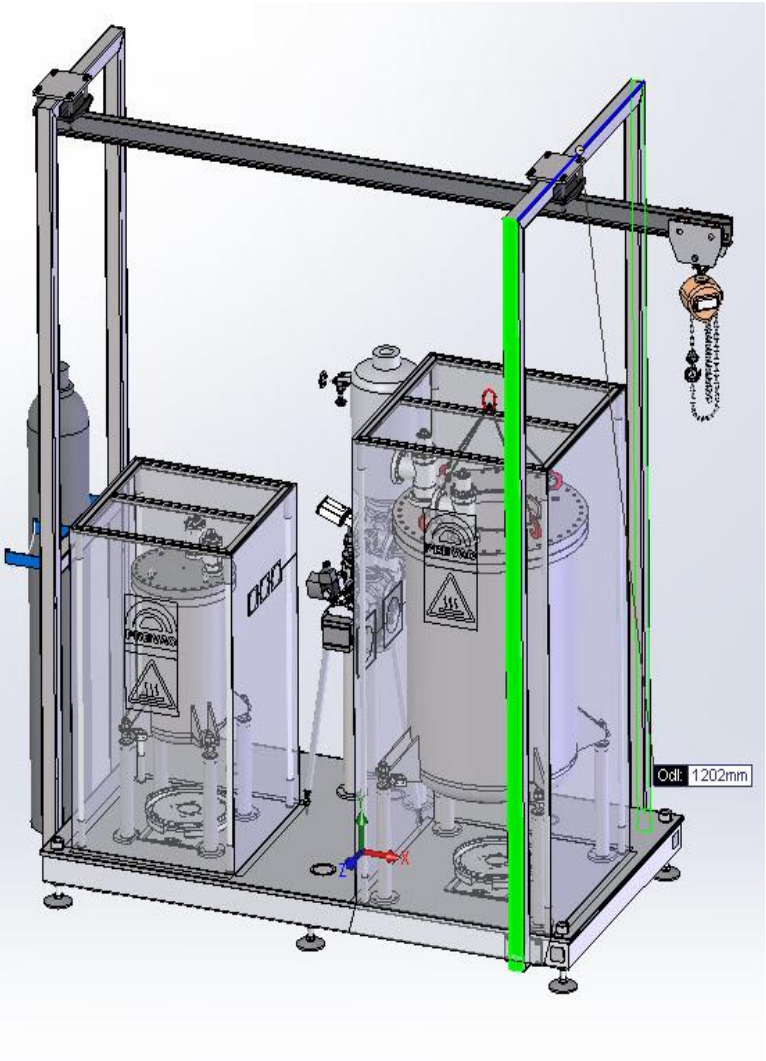
(typical rate for SS vessel of this size)

Cryostat with TPC

A_{sat} = (13.5 ± 4.0) mBq

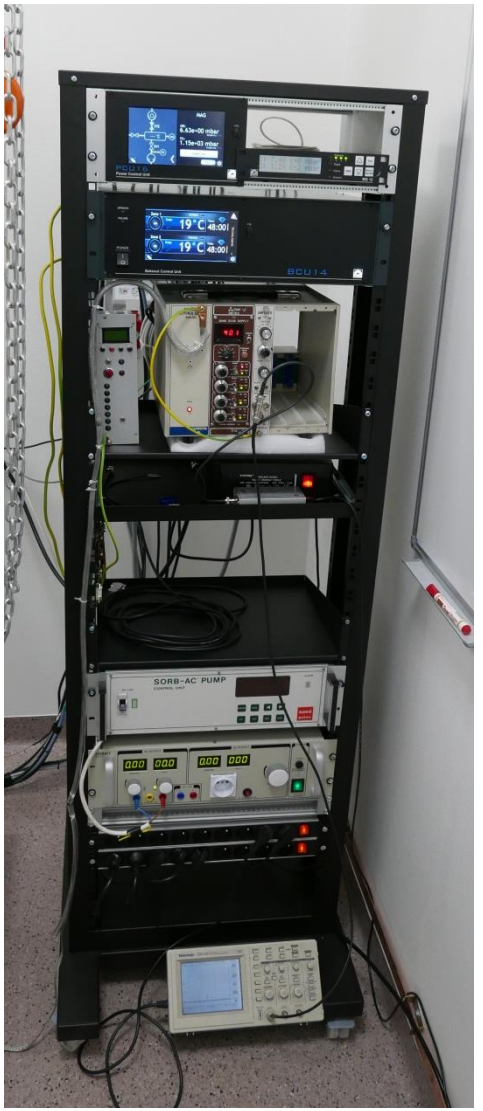
$DS\text{-}50_{data}$ < 0.3 mBq

System of emanation chambers



- Three UHV chambers, fully electro-polished and metal-sealed, 12 L, 50 L and 250 L, available
- Chambers coupled to the cryogenic Rn detector
- Integrated automatic pumping system
- Integrated automatic heating system (emanation tests up to 150 ^{o}C possible)
- Simultaneous real-time detection of emanated ^{220}Rn and ^{222}Rn
- Detection limit of ~100 μBq

System of emanation chambers



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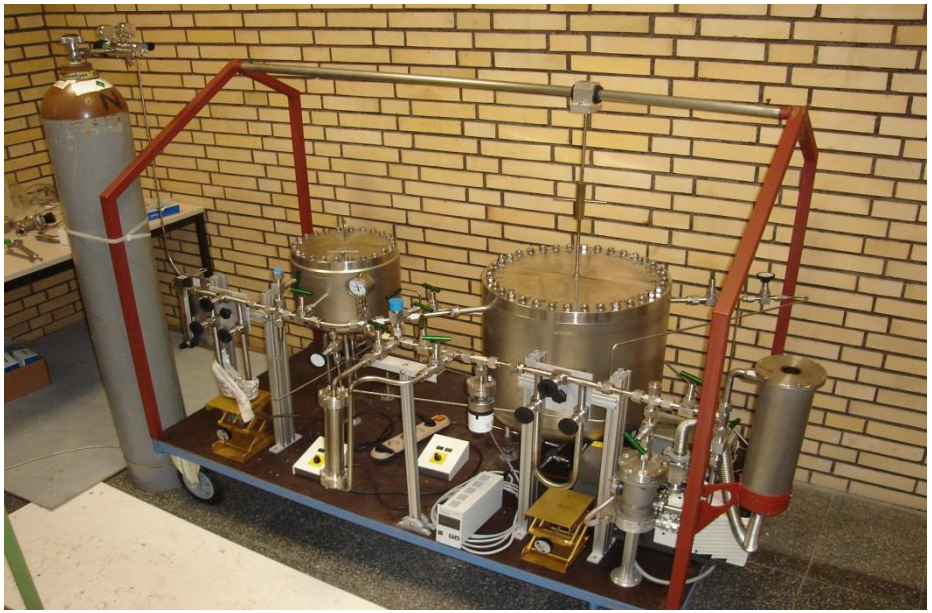

MPIK-HD chambers

Absolute sensitivity
~100 μBq [50 atoms]

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Copper foil

Not treated: (1.7 ± 0.2) μBq/m^2
Water cleaning: (1.2 ± 0.2) μBq/m^2

Steel foil untreated: (10 ± 1) μBq/m^2
After water cleaning: (4.6 ± 0.6) μBq/m^2

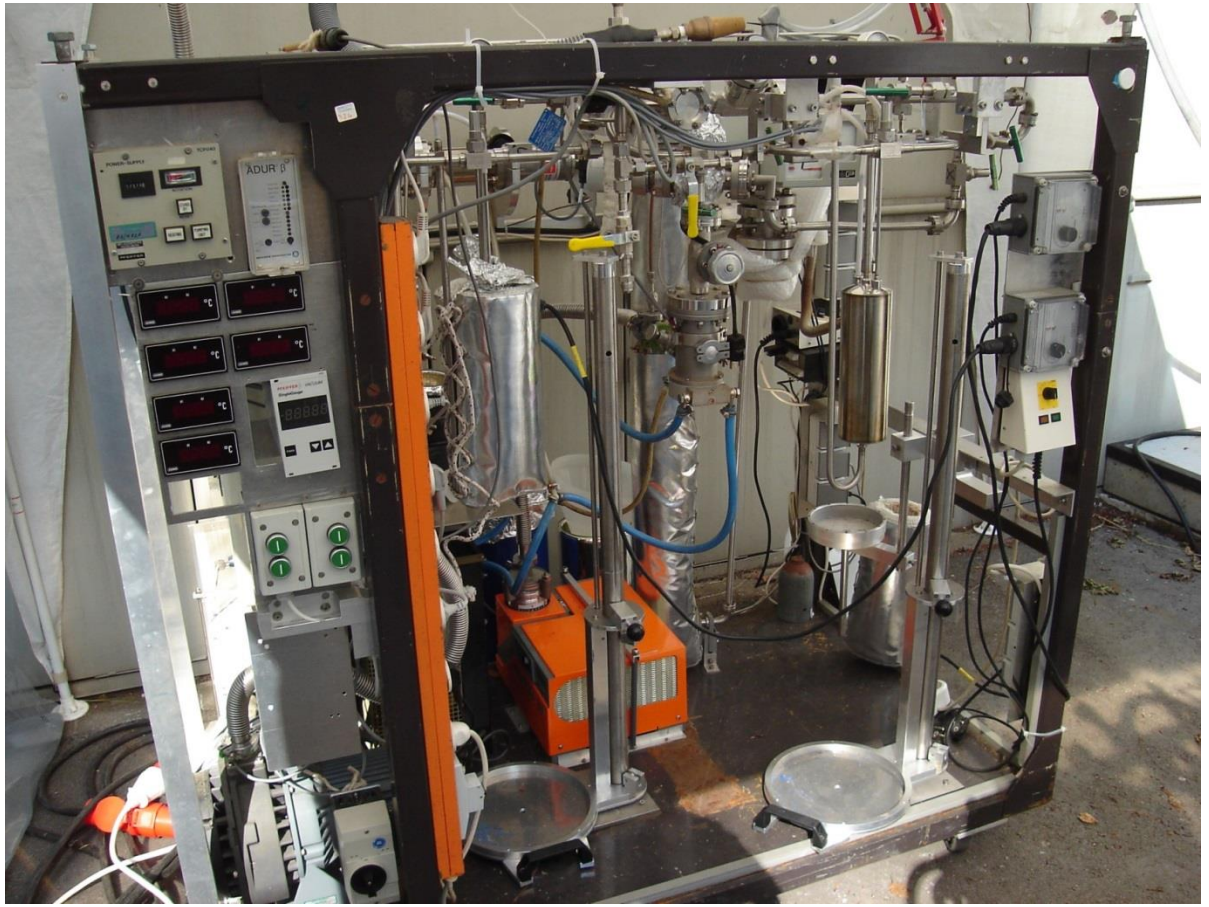
Teflon foil: < 9 μBq/m^2 (< 13 μBq/kg)
BOREXINO IV foil: < 0.8 μBq/m^2

AIP Conf. Proc., Vol. 785, p. 142

^{222}Rn in gases (MPIK-HD)

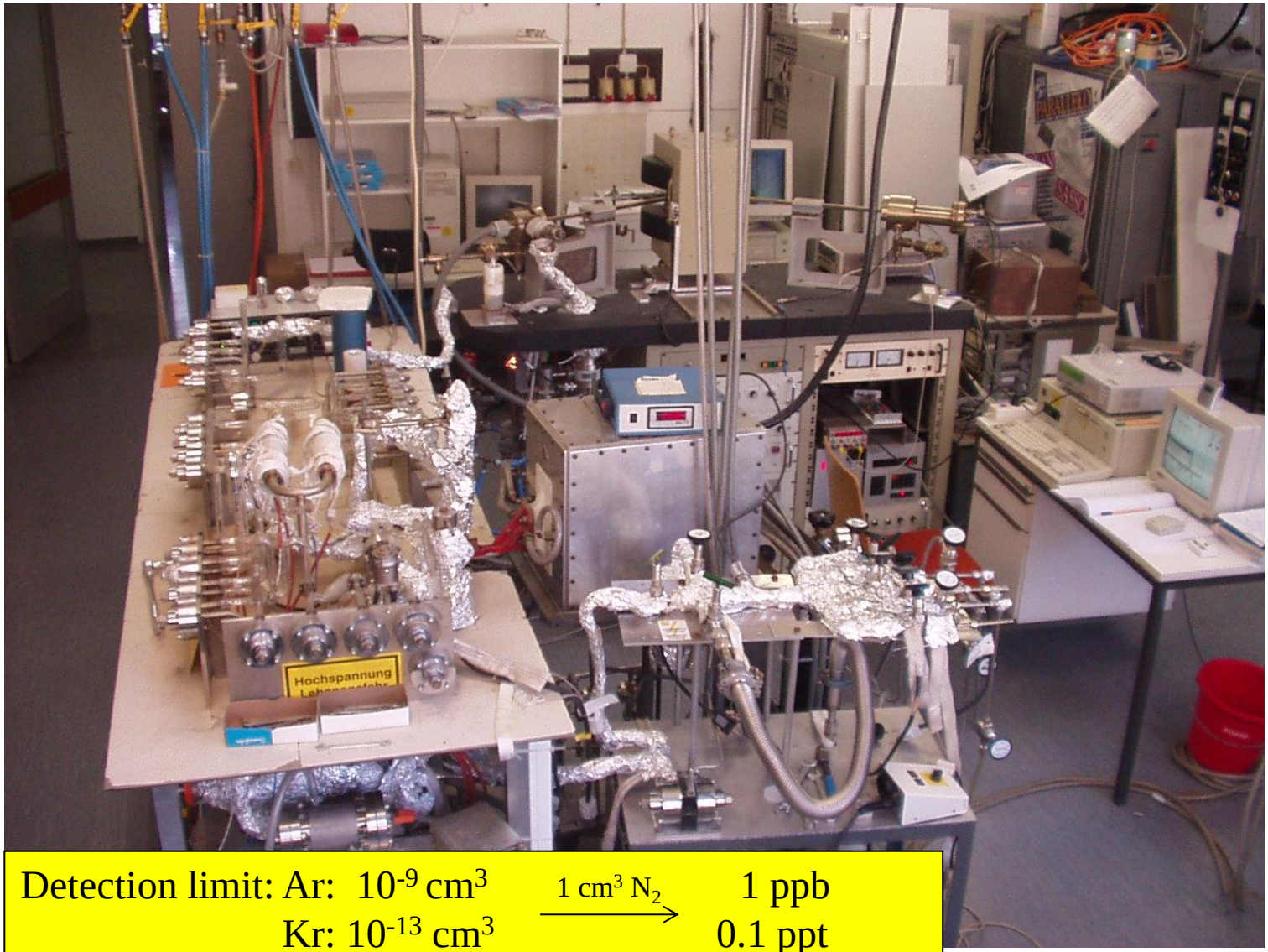
- ^{222}Rn adsorption on activated carbon
- Several AC traps available (MoREx/MoRExino)
- Pre-concentration from 100 – 200 m^3

^{222}Rn detection limit:
~0.5 μBq/m^3 (STP)
[1 atom in 4 m^3]



Appl. Rad. Isot. 52 (2000) 691

Ar and Kr in nitrogen (MPIK-HD)



Detection limit: Ar: 10^{-9} cm^3 $\xrightarrow{1\ \text{cm}^3\ N_2}$ 1 ppb
Kr: 10^{-13} cm^3 0.1 ppt

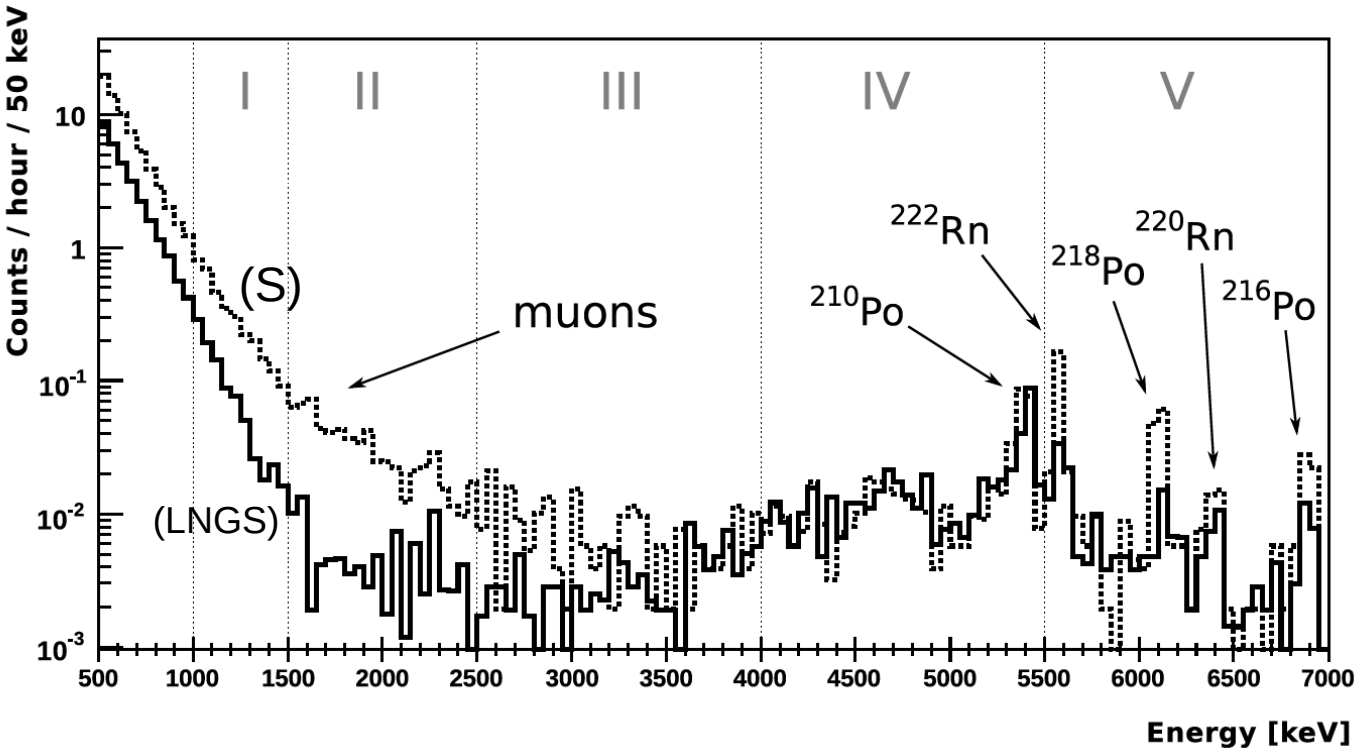
^{222}Rn in Argon (GERDA)

Date	Quality	Sample size	^{222}Rn activity [mBq/m^3] when measured	after production
29.09.04	Ar 4.6	117 m^3	2.9 ± 0.2	> 8
04.11.04	Ar 4.6	141 m^3	0.20 ± 0.02	—
08.06.05	Ar 5.0	200 m^3	6.0 ± 0.1	8.4 ± 0.2
21.11.05	Ar 5.0	85 m^3	0.048 ± 0.004	—
28.11.06	Ar 5.0 (GS)	4 m^3	< 0.020	—
13.06.05	Ar 6.0	104 m^3	0.11 ± 0.01	0.38 ± 0.02

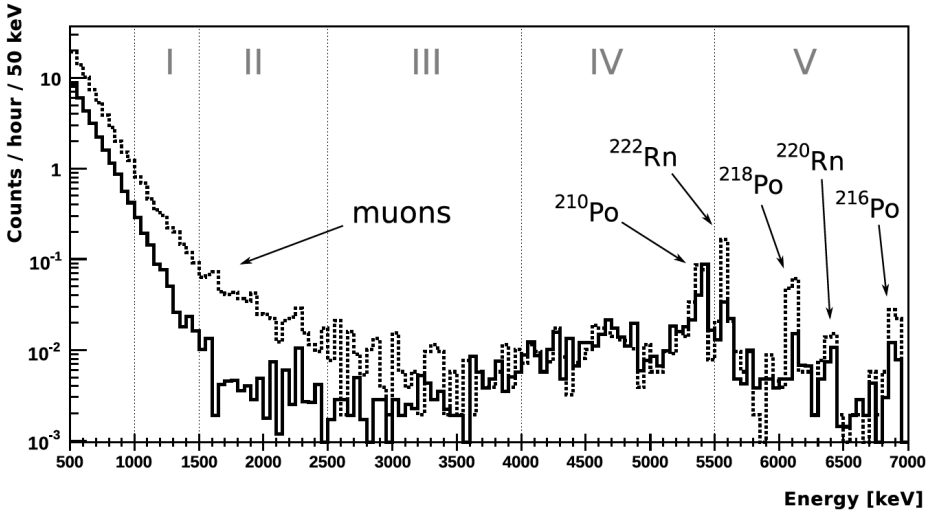
^{222}Rn content in Ar 100-1000 higher compared to N_2

Underground alpha spectroscopy

- Alpha spectroscopy of thick sources is sensitive to cosmic rays
- Long-term background measurements were performed in different configurations and locations:
  - spectrometer on the surface (S),
  - spectrometer on the surface with an active veto detector (SA),
  - spectrometer in the underground lab of LNGS (Italy)



Underground alpha spectroscopy



Energy region	Energy range [keV]	S [cts h^{-1}]	SA [cts h^{-1}]	LNGS [cts h^{-1}]
I	1000-1500	3.486 ± 0.082	2.225 ± 0.092	0.955 ± 0.030
II	1500-2500	0.666 ± 0.036	0.404 ± 0.039	0.095 ± 0.009
III	2500-4000	0.219 ± 0.020	0.145 ± 0.023	0.098 ± 0.010
IV	4000-5500	0.478 ± 0.030	0.389 ± 0.038	0.477 ± 0.021
V	5500-7000	0.478 ± 0.030	0.419 ± 0.040	0.207 ± 0.014

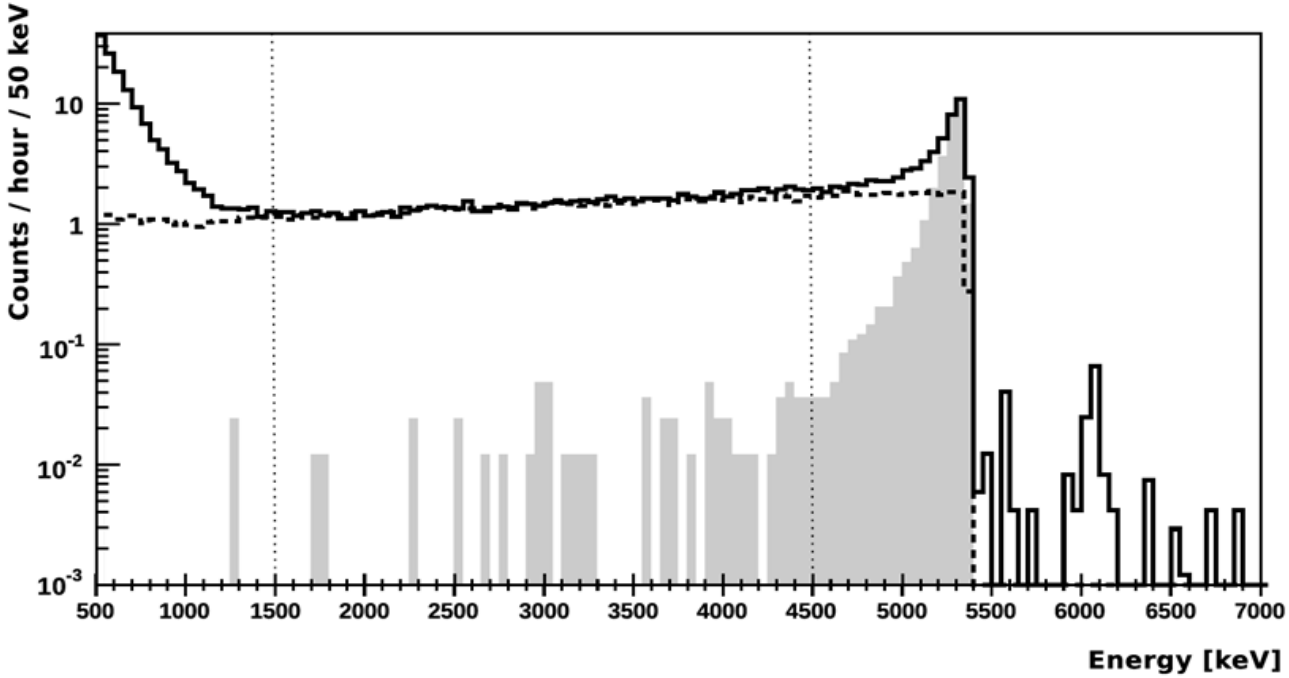
Background measurement	δ	ROI [MeV]	ε [cts h^{-1}/ (Bq kg^{-1})]	A_B [cts h^{-1}]	Δt [h]	A_{0min} [Bq kg^{-1}]
S	0.3	2.2 – 5.4	0.12	0.715 ± 0.037	72	3.73
SA	0.3	2.2 – 5.4	0.12	0.494 ± 0.043	72	3.37
LNGS	**0.3**	**1.5 – 5.4**	**0.14**	**0.566 ± 0.023**	**72**	**2.83**

^{210}Po diffusion problem

The observed peak at 5.3 MeV comes from the diffusion-enhanced ^{210}Po surface activity.

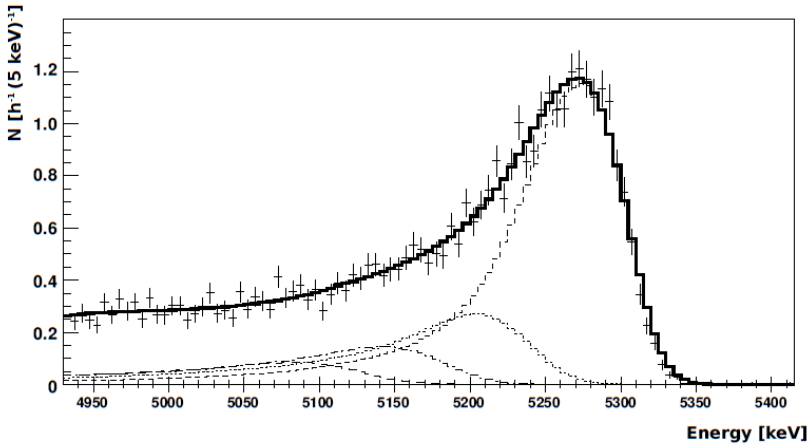
Analysis of ^{210}Po specific activity for a thick sample should be performed for the energy region below the ^{210}Po peak and above the tail from the cosmogenic background. Recommended window: 1.5 – 4.5 MeV.

Such procedure avoids systematic mistakes in determination if specific activities when the calibration standard and the sample have different ^{210}Po distributions near the surface.

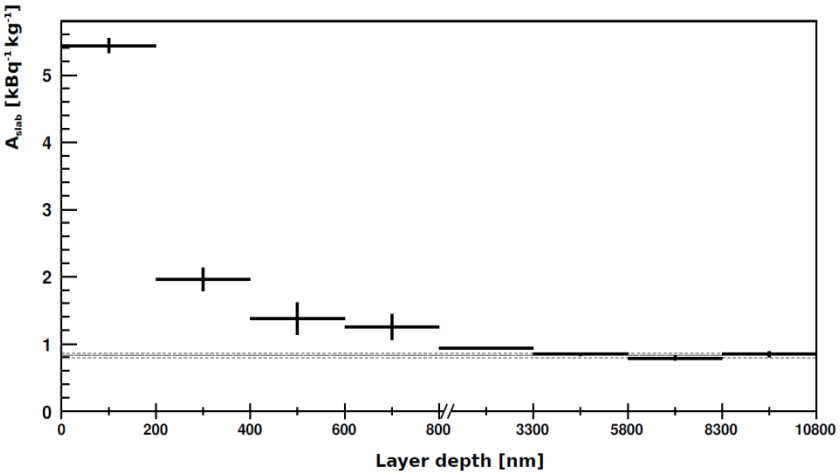


^{210}Po diffusion problem

Background measurement	δ	ROI [MeV]	ε [cts h^{-1}/ (Bq kg^{-1})]	A_B [cts h^{-1}]	Δt [h]	A_{0min} [Bq kg^{-1}]
S	0.3	2.2 – 4.5	0.08	0.408 ± 0.028	72	4.27
SA	0.3	2.2 – 4.5	0.08	0.240 ± 0.030	72	3.69
LNGS	0.3	1.5 – 4.5	0.10	0.292 ± 0.017	72	2.99



In the model the disc was divided into 200 nm slabs with their specific activities treated as free parameters in the global fit to the measured spectrum (solid line). Individual spectra from the first four layers are displayed.



Specific activities of individual slabs reproduced from the global fit are shown on the left. The values start to agree with nominal specific activity of the source ((826 ± 33) Bq kg^{-1}) for the depth of about 3 μm below the surface – effective range of the polonium diffusion in lead.

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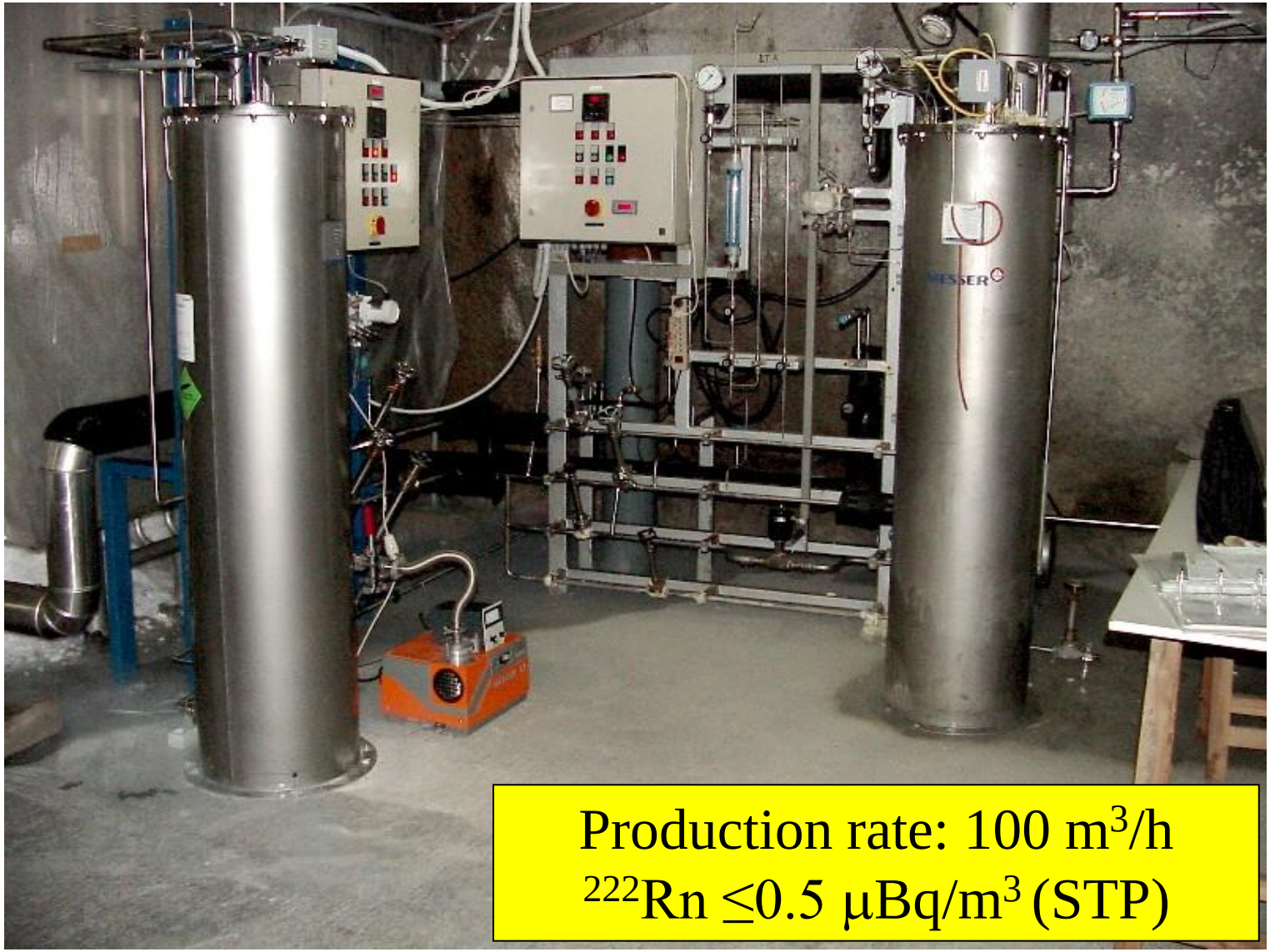
Purification of gases

- Radio-pure gases are widely used in low-background detection systems and experiments
  - drying
  - cleaning/purging volumes
  - sparging liquids (last purification step)
  - blanketing
- Liquid gases are widely applied as target material in the detectors used e.g. to search for neutrino-less double beta decay or dark matter interactions
- Removal of ^{222}Rn and other radioactive isotopes (^{39}Ar, ^{85}Kr) crucial (for ^{222}Rn, due to its emanation, often in-situ purification is needed)

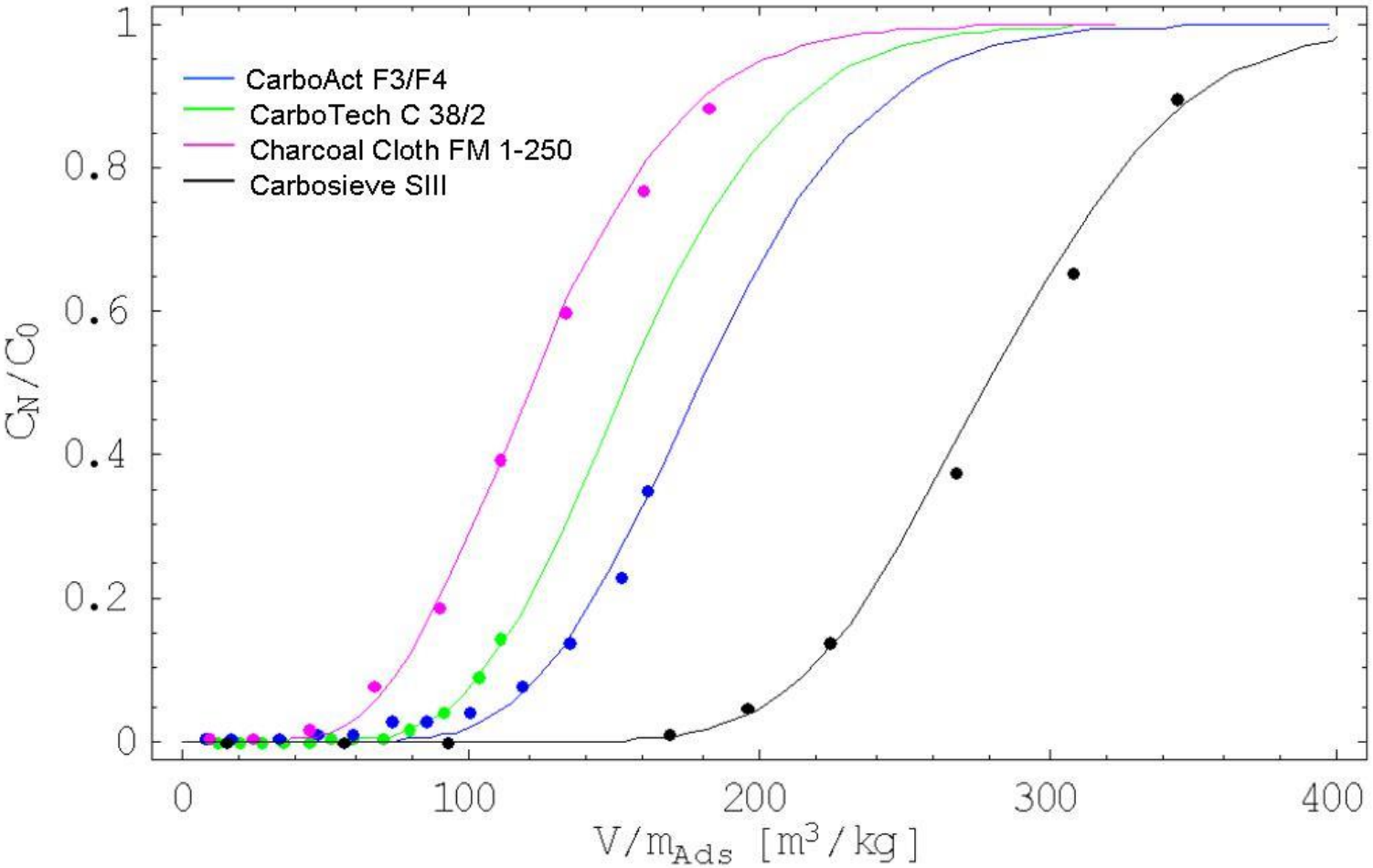
Purification of nitrogen from ^{222}Rn

BX N_2 purification plant: LTA – Low Temperature Adsorber

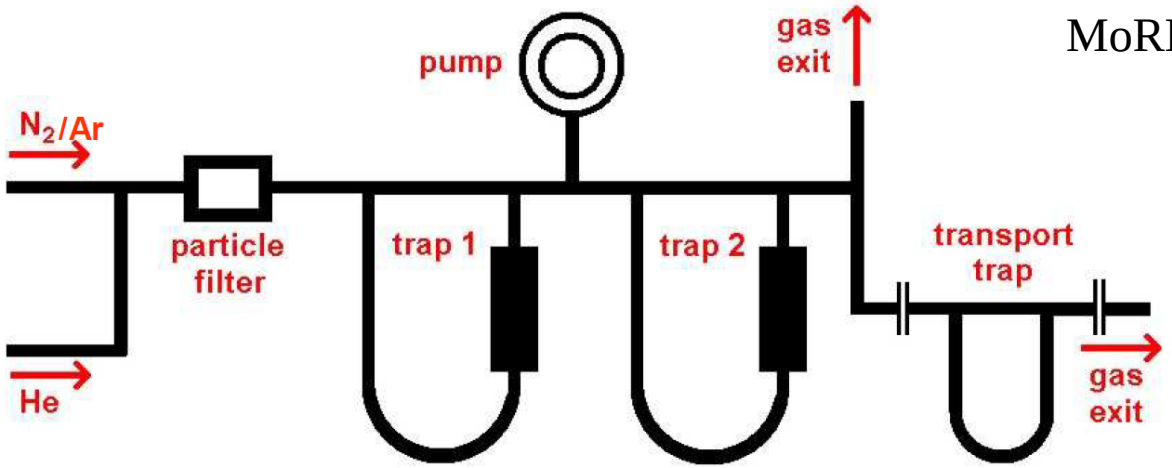


Production rate: 100 m^3/h
^{222}Rn ≤0.5 μBq/m^3 (STP)

Purification of nitrogen from Kr



Purification of argon from ^{222}Rn



MoREx (MPI-K HD)

Argon quality	Sample size [m^3]	^{222}Rn activity [mBq/m^3] before purif.	after purif.	Red. factor [1/kg]	Remarks
4.6	141	0.20 ± 0.02	≤0.0005	≥ 2700	gas phase
5.0	200	6.0 ± 0.1	0.60 ± 0.02	167	liquid phase
6.0	104	0.11 ± 0.01	0.006 ± 0.001	305	liquid phase

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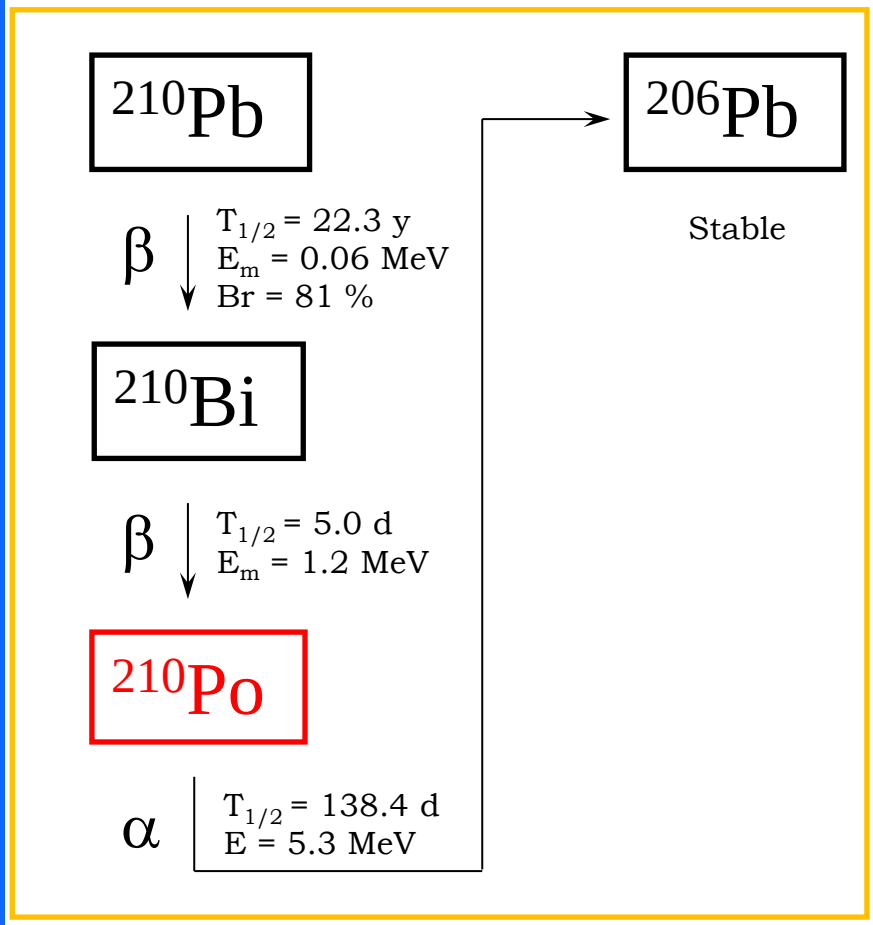
Cleaning of surfaces



- Disequilibrium in the chain: $A_{Rn} \neq A_{Pb}$
- In fresh material usually $A_{Pb} \neq A_{Po}$
- Pb may be plated out /implemented into the surface → ^{222}Rn-free atmosphere needed
- ^{210}Bi – pure β-emitter
- ^{210}Po:
  - degraded αs
  - diffusion
  - source of ns (α,n)
  - difficult to remove from surfaces (Cu)

High specific activities

- Samples in a form of discs with 50 mm diameter
- To increase the sensitivity samples were artificially loaded with ^{210}Pb, ^{210}Bi and ^{210}Po: placed in a strong ^{222}Rn source for several months (^{210}Po specific activities of ~100 Bq/m^2)
- Screening of ^{210}Po with an alpha spectrometer 50 mm Si-detector, bcg ~2 α/d (1-10 MeV) sensitivity ~20 mBq/m^2 (100 mBq/kg, ^{210}Po)
- Screening of ^{210}Bi with a beta spectrometer 2×50 mm Si(Li)-detectors, bcg ~0.18/0.40 cpm sensitivity ~10 Bq/kg (^{210}Bi)
- Screening of ^{210}Pb (46.6 keV line) with a gamma spectrometer 16 % - HPGe detector with an active and a passive shield

High specific activities

Isotope	Activity reduction factors after etching/electropolishing			
			Germanium	
	Copper	Stainless steel	NPGe	HPGe
^{210}Pb	50 / 300	100 / 400	100 / –	700 / –
^{210}Bi	50 / 300	100 / 800	400 / –	800 / –
^{210}Po	1 / 400	20 / 700	1000 / –	100 / –

Copper

- etching: 5 min in (1% H_2SO_4 + 3% H_2O_2) and 5 min in 1% citric acid
- electro-polishing (electrolyte): 85 % H_3PO_4 + 5 % 1-butanol ($C_4H_{10}O$)

Stainless steel:

- etching: (20 % HNO_3 + 1.7 % HF) and 15 % HNO_3
- electro-polishing (electrolyte): 40 % H_3PO_4 + 40 % H_2SO_4 + 3 % CrO_3

Germanium:

- etching: CP4 solution (45.45 ml HNO_3 + 27.27 ml HF + 27.27 ml CH_3COOH + 0.5 ml Br for 100 ml solvent) done by Canberra-France in Lingolsheim in cooperation with MPP Munich

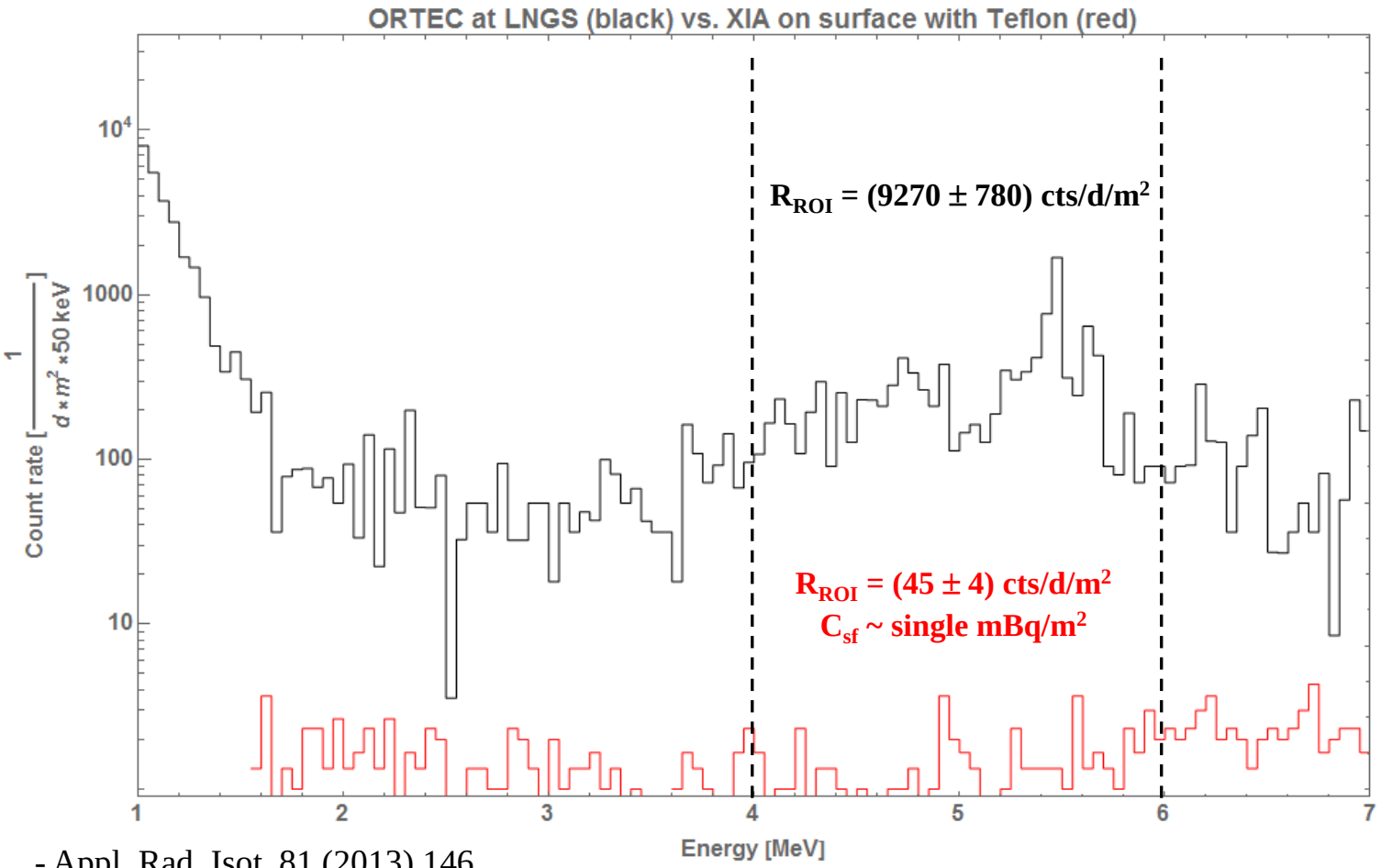
NIM A 676 (2012) 140
NIM A 676 (2012) 149

Low specific activities

- Only ^{210}Po studied
- Low background, large surface (LBS) alpha spectrometer
- Ar used as counting gas (3.5 l/min)
- Sample size: 43×43 cm / 30 cm diam. disc, a few mm thick
- PSD + veto guard (discrimination of background events)

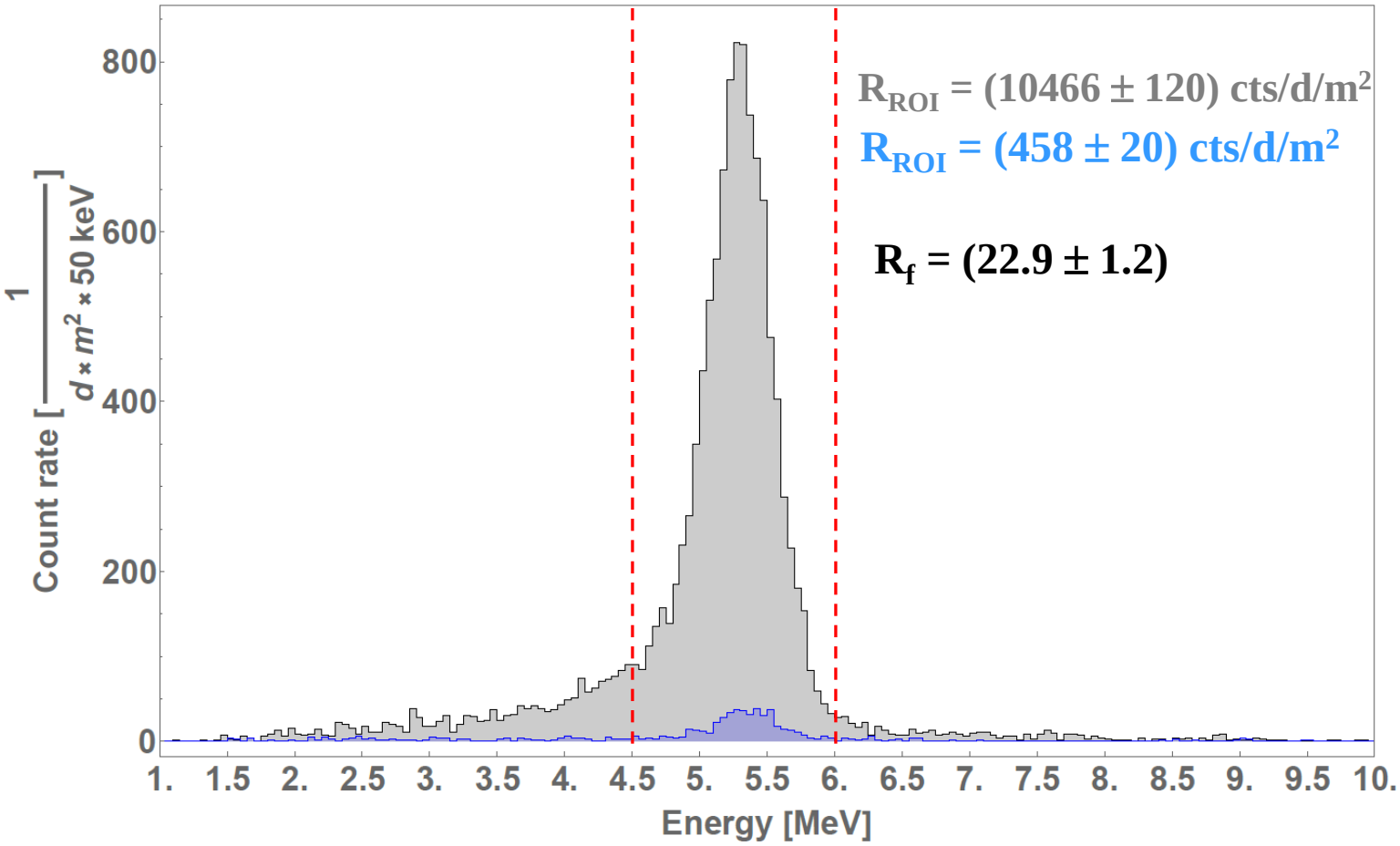
Background spectrum

Low background ORTEC α detector (40 mm diameter) at LNGS vs. LBS spectrometer: **factor ~200 improvement**.



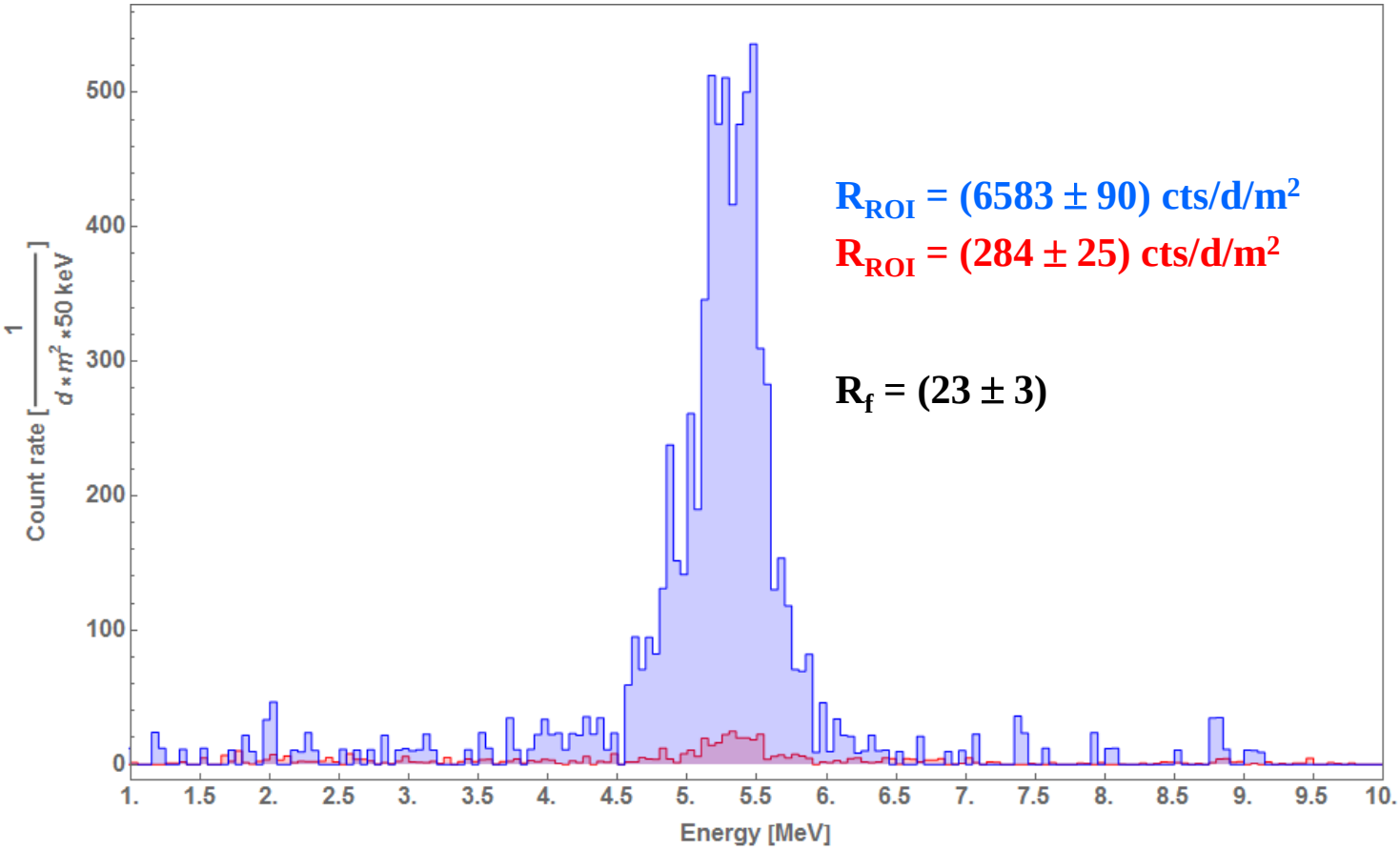
- Appl. Rad. Isot. 81 (2013) 146
- https://doi.org/10.1016/j.apradiso.2017.01.030

„Dynamic" etching



- Etching procedure: 5 x 1 min wash with a mixture of 1% H_2SO_4 + 3% H_2O_2
- Passivation with 1% citric acid at the end
- Washing in high-purity deionized water (18 MΩ×cm)

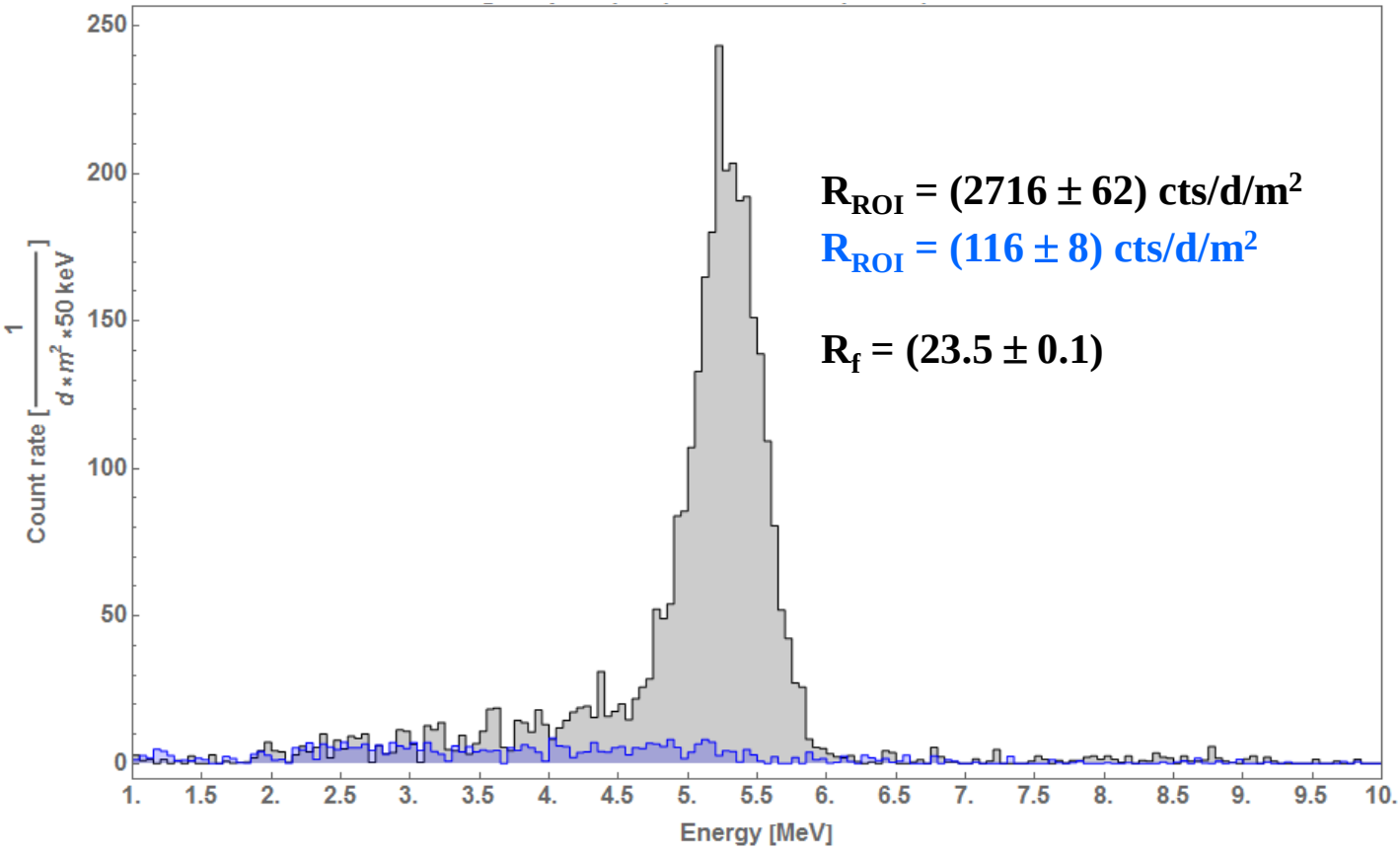
Electro-polishing of copper



- Polishing mixture: 95% H_3PO_4 + 1% 1-butanol
- Polishing conditions: 2.5 A/dm², 3 V, 20 min, distance between plates: 2 cm, room temperature

Electro-polishing of stainless steel

SS 1.4301 (304): sheet No. 2, 43 cm x 43 cm x 0.1 cm,



- Polishing mixture: 1:1 of 95% H_2SO_4 and 85% H_3PO_4
- Washing: 5 min in 15% HNO_3 and later in HP water
- Polishing conditions: 2.5 A/dm^2, 2 V, 25 min, distance between plates: 2 cm, T ~ 50 ^{o}C

DARKSIDE ^{222}Rn-free clean rooms

Avoiding deposition of long-lived ^{222}Rn daughters

- Class 10 - 100
- Radon daughters plating out on surfaces of the detector may cause dangerous alpha-induced nuclear recoils
- Dedicated scrubbing system reducing ^{222}Rn concentration in the air down to ~1 mBq/m^3 has been implemented
- DARKSIDE clean rooms are supplied with the ^{222}Rn-free air
- ^{222}Rn content in the clean rooms is monitored online by a dedicated detector

Typical radon in hall C air ~30 Bq/m^3

Cleanroom radon levels 5 – 50 mBq/m^3

Summary

- Low-radiation (low-level) techniques have "natural" application in experiments looking for rare nuclear processes at low energies.
- A lot of experience has been accumulated over the last years in this field resulting in background-free operation of some detectors (DS-50, GERDA II).
- Several detectors and experimental methods were developed, allowing measurements even at a single atom level.
- Effective purification and cleaning procedures have been established.
- However, for the next-generation, ton-scale experiments (DARKSIDE-20k, LEGEND, nEXO, XENONnT, *etc.*) achieved so far sensitivities of the screening facilities may not be sufficient – new developments are necessary.