Low background counting techniques - status and outlook

Dr. Matthias Laubenstein
Laboratori Nazionali del Gran Sasso
ITALY

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Activities of the Deep Underground Laboratories in ILIAS

Activity

Activity	
J1 : LBT-DUS Low Background Techniques for Deep Underground Science	Joint R&D for the improvement of the strengthening of the low background facilities and know-how of the UG labs
A1 : TARI-DUSL Transnational Access to the EU Deep Underground Science Laboratories	Support for the transnational access of research teams to the EU underground labs with priority to researchers from less favoured countries
N2 : EUNet-DUSL European Network of the Deep Underground Science Laboratories	Coordination and networking to support the management of common issues relevant in the operation of the UG Labs

J1 : low background techniques for deep underground science

Motivations :

Extremely low-level background techniques and instrumentation are an essential requirement for a number of topics in astroparticle physics, e.g.:

- ❖ search for ββ decay
- ❖ search for dark matter
- ❖ detection of low-energy neutrinos (solar, geo)

Fundamental topics common to most experiments are:
- selection of radiopure materials
- techniques for shielding against environmental backgrounds
- purification techniques

This is the main motivation to carry on a Joint Research Activity on Low Background Techniques coordinated by the UG Labs within ILIAS

Key R&D topics

❖ development and strengthening of the ultra low background facilities and instrumentation in the UG labs

❖ measurement and monitoring of the background components in the underground Labs – Development of background simulation codes

❖ application of low background techniques to interdisciplinary fields

❖ R&D on radiopurity of materials and purification techniques.

EU Support:

❖ personnel
❖ travel money
❖ contribution to equipment and consumables for selected specific activities

Comparison of radioassay techniques

Ge-spectroscopy	γ emitting nuclides
Rn emanation assay	^{226}Ra, ^{228}Th
neutron activation analysis	primordial parents
liquid scintillation counting	α,β emitting nuclides
mass spectrometry (ICP-MS; AMS)	primordial parents
AES + AAS analysis	primordial parents
X-Ray Fluorescence	primordial parents
alpha spectroscopy	α emitting nuclides

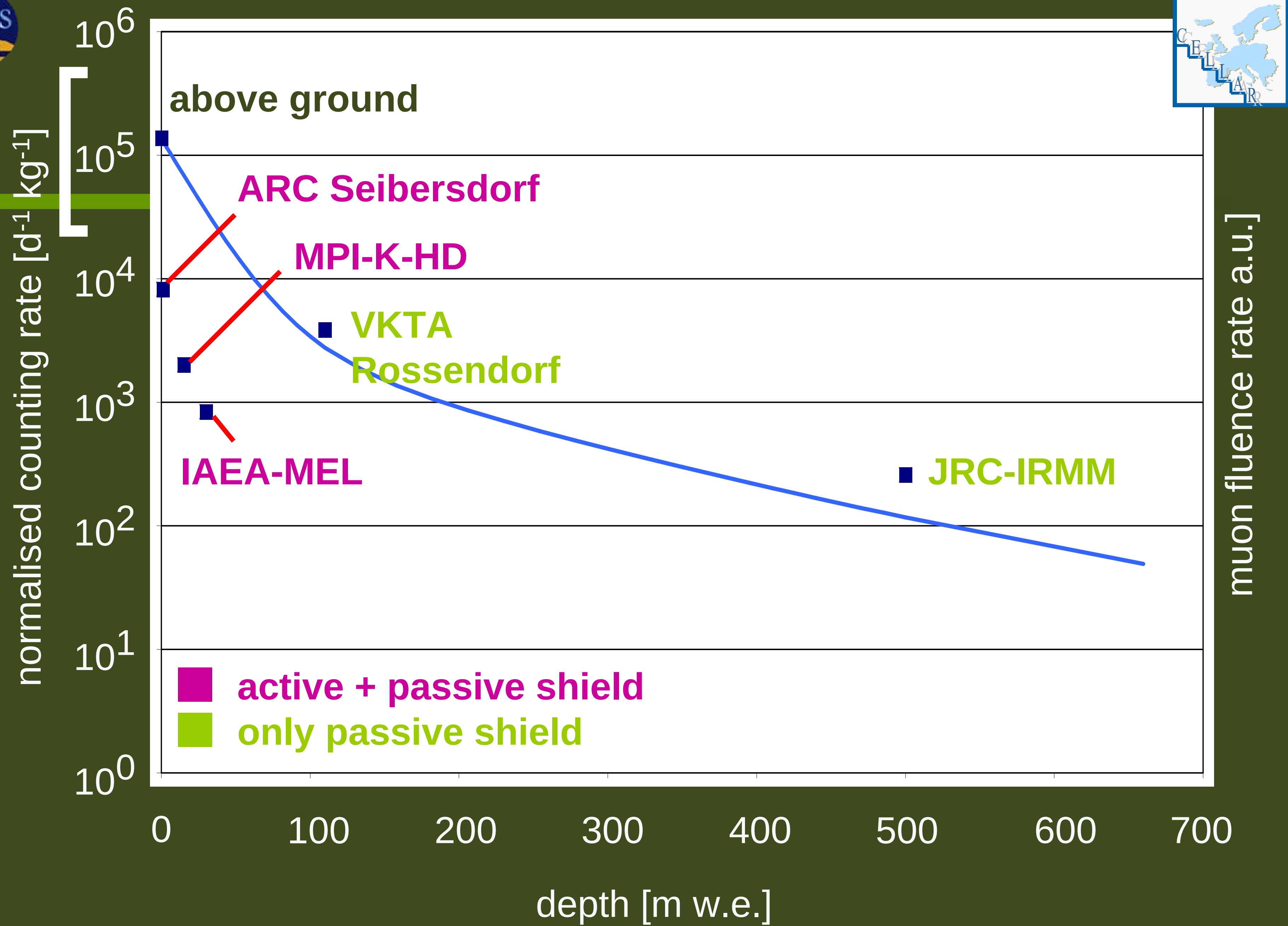
difficult to compare because each method has its special application

Sensitivities

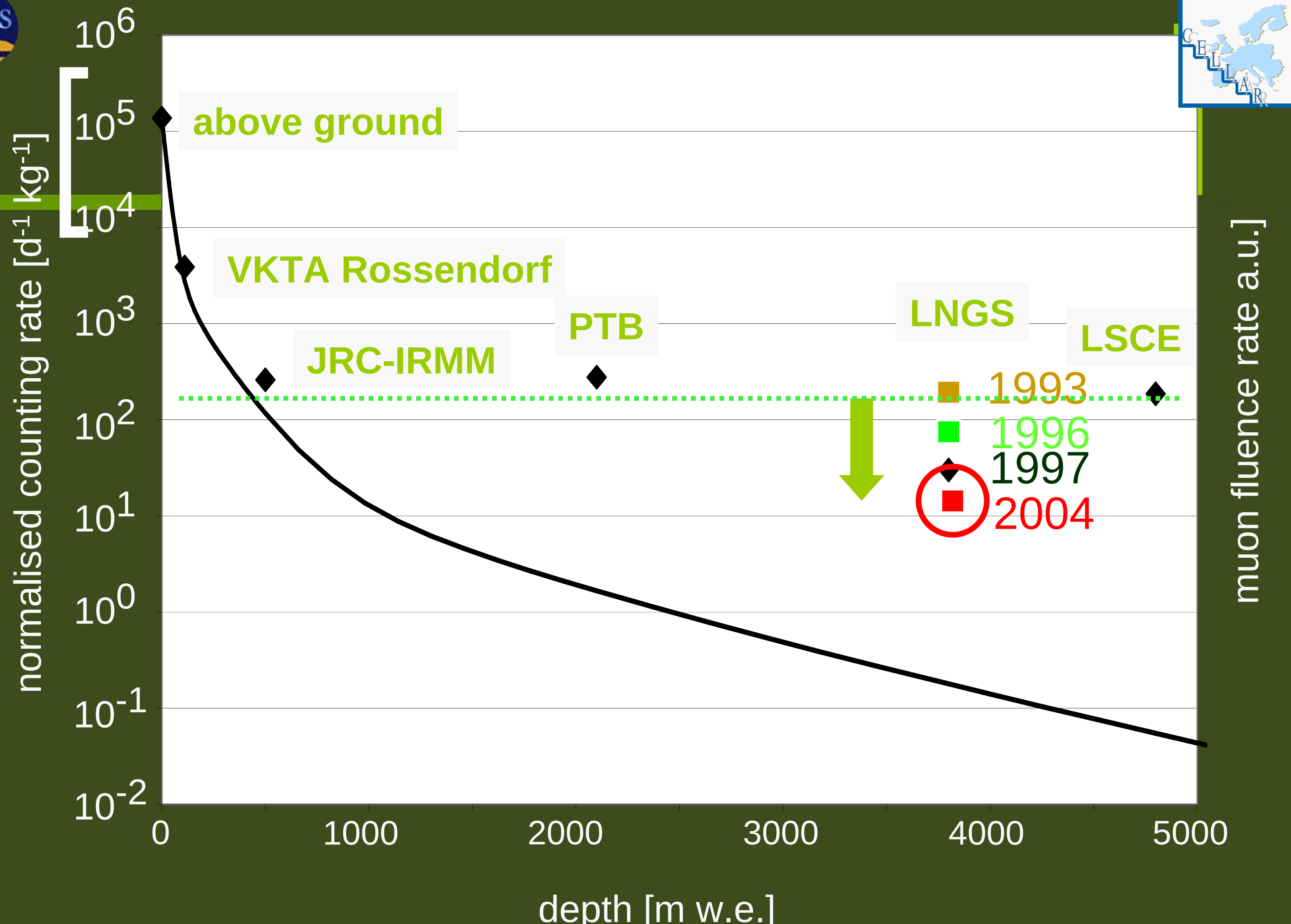
method	suited for	sensitivity for U/Th
Ge-spectroscopy*	γ emitting nuclides	10-100 μBq/kg
Rn emanation assay	^{226}Ra, ^{228}Th	0.1-10 μBq/kg
neutron activation analysis	primordial parents	0.01 μBq/kg
liquid scintillation counting	α,β emitting nuclides	1 mBq/kg
mass spectrometry (ICP-MS; AMS)	primordial parents	1-100 μBq/kg
AES and AAS analysis	primordial parents	1-1000 μBq/kg
X-Ray Fluorescence	primordial parents	10 mBq/kg
alpha spectroscopy	α emitting nuclides	1 mBq/kg

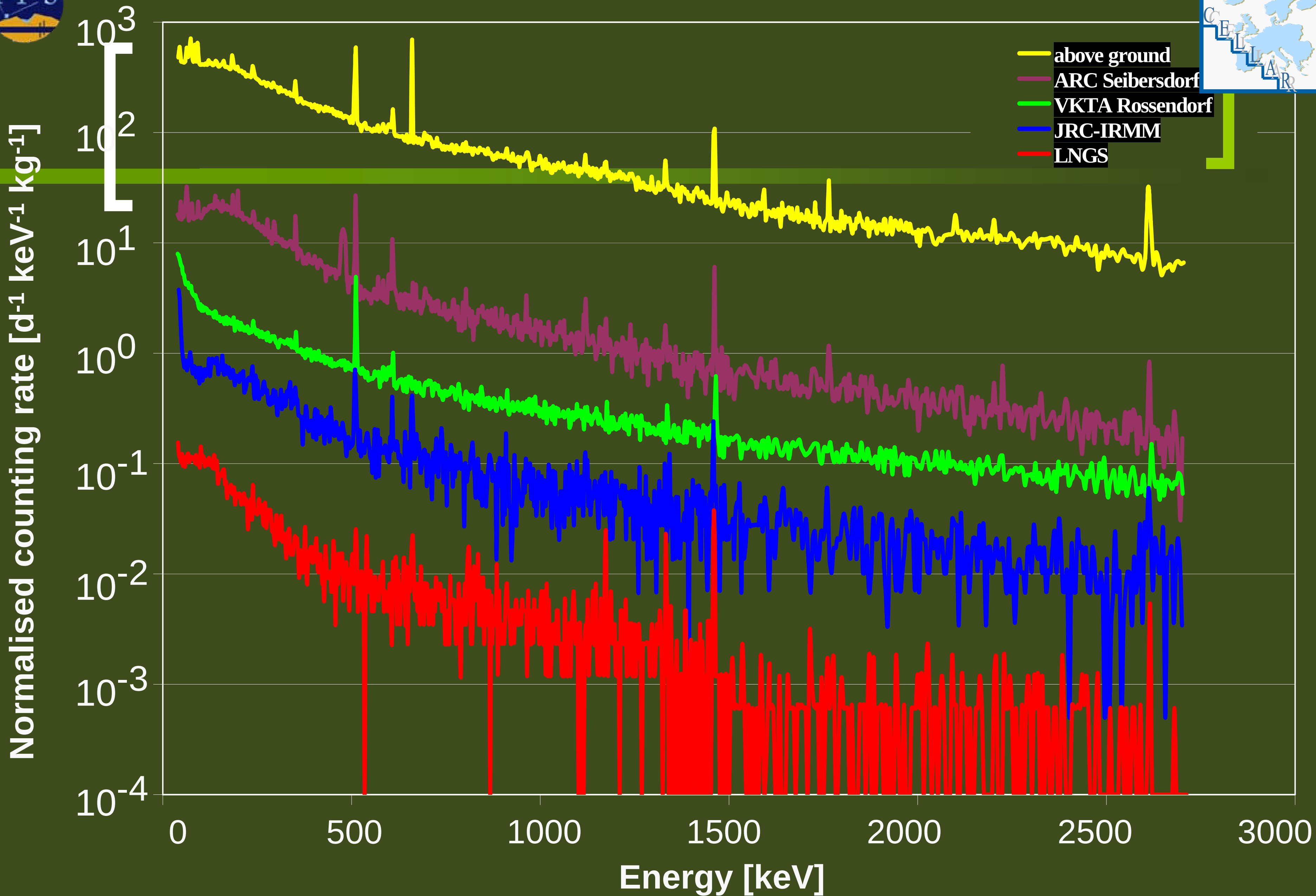
* Needs counting times from several weeks to several months

Germanium spectroscopy



above ground
ARC Seibersdorf
MPI-K-HD
VKTA
Rossendorf
IAEA-MEL
JRC-IRMM
active + passive shield
only passive shield
normalised counting rate [d-1 kg-1]
muon fluence rate a.u.]
10^0
10^1
10^2
10^3
10^4
10^5
10^6
0
100
200
300
400
500
600
700
depth [m w.e.]


above ground
VKTA Rossendorf
JRC-IRMM
PTB
LNGS
LSCE
1993
1996
1997
2004
normalised counting rate [d-1 kg-1]
muon fluence rate a.u.]
depth [m w.e.]
0
1000
2000
3000
4000
5000
10-2
10-1
100
101
102
103
104
105
106



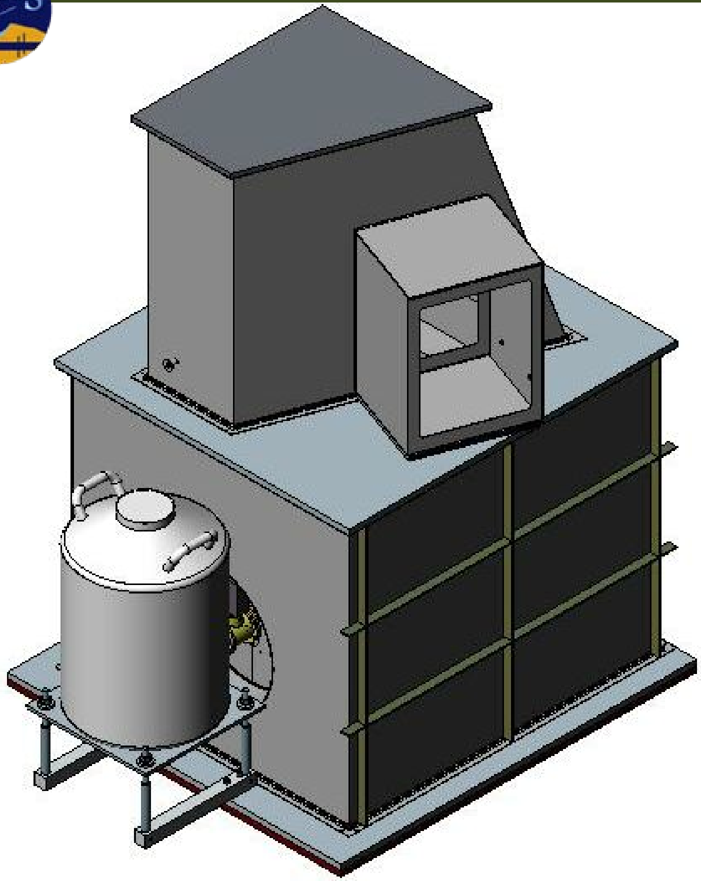
Normalised counting rate [d-1 keV-1 kg-1]
10^3
10^2
10^1
10^0
10^-1
10^-2
10^-3
10^-4
0
500
1000
1500
2000
2500
3000
Energy [keV]
above ground
ARC Seibersdorf
VKTA Rossendorf
JRC-IRMM
LNGS

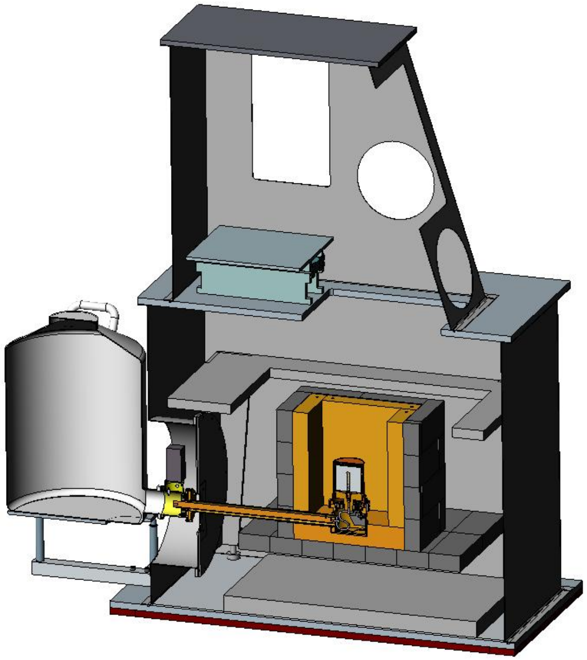
GeMPI

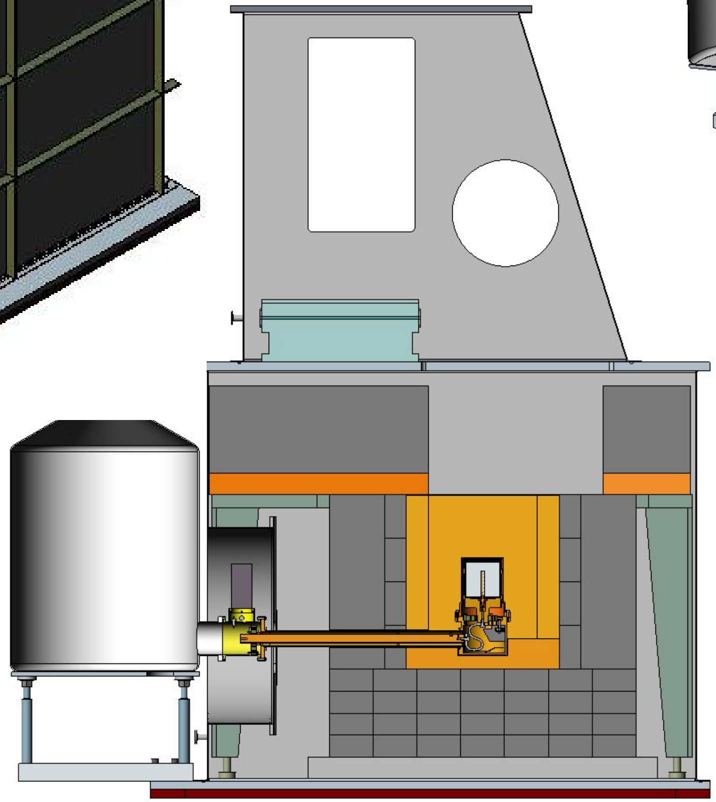
Operated at

LNGS

(3800 m w.e.)





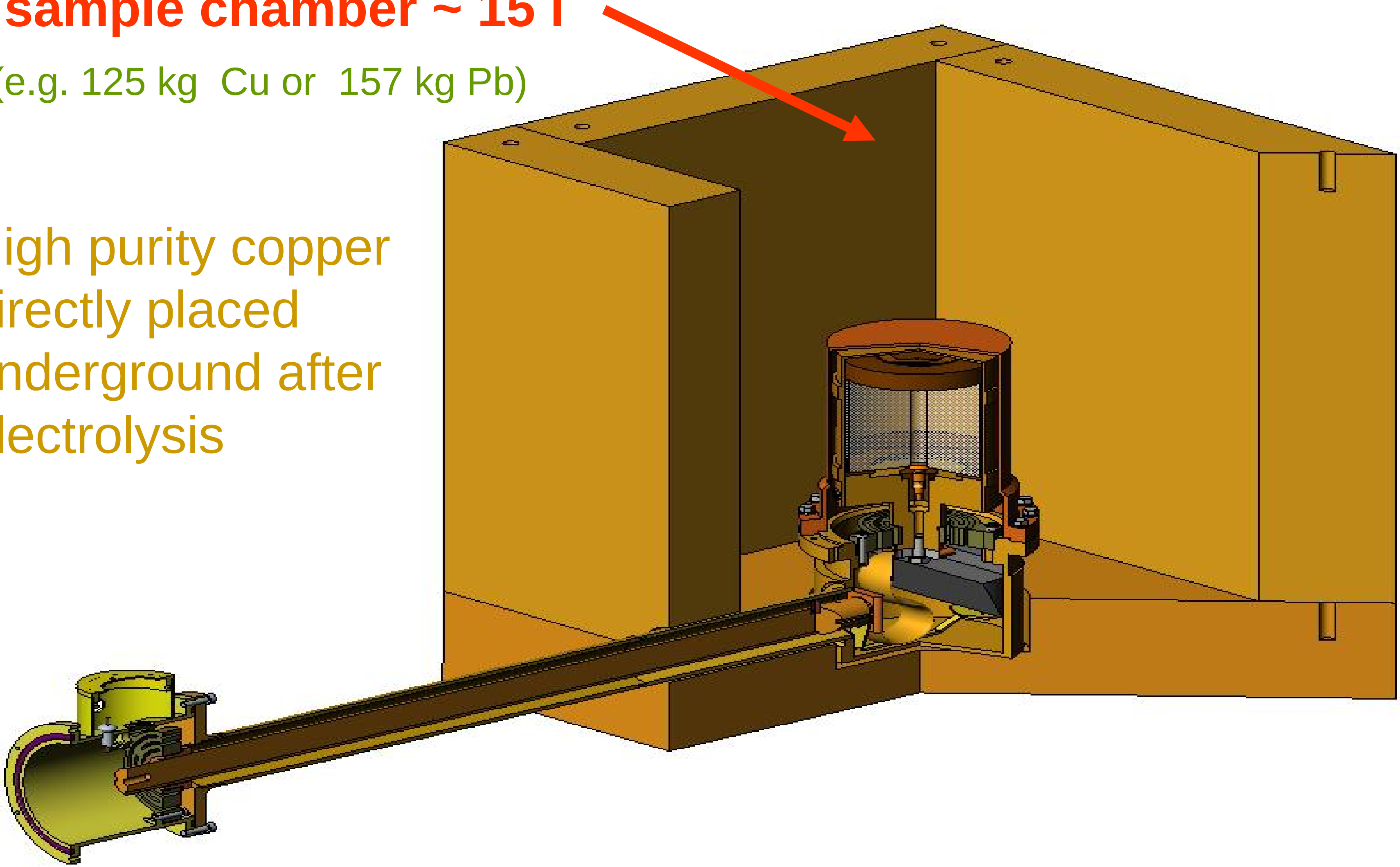


G. Heusser

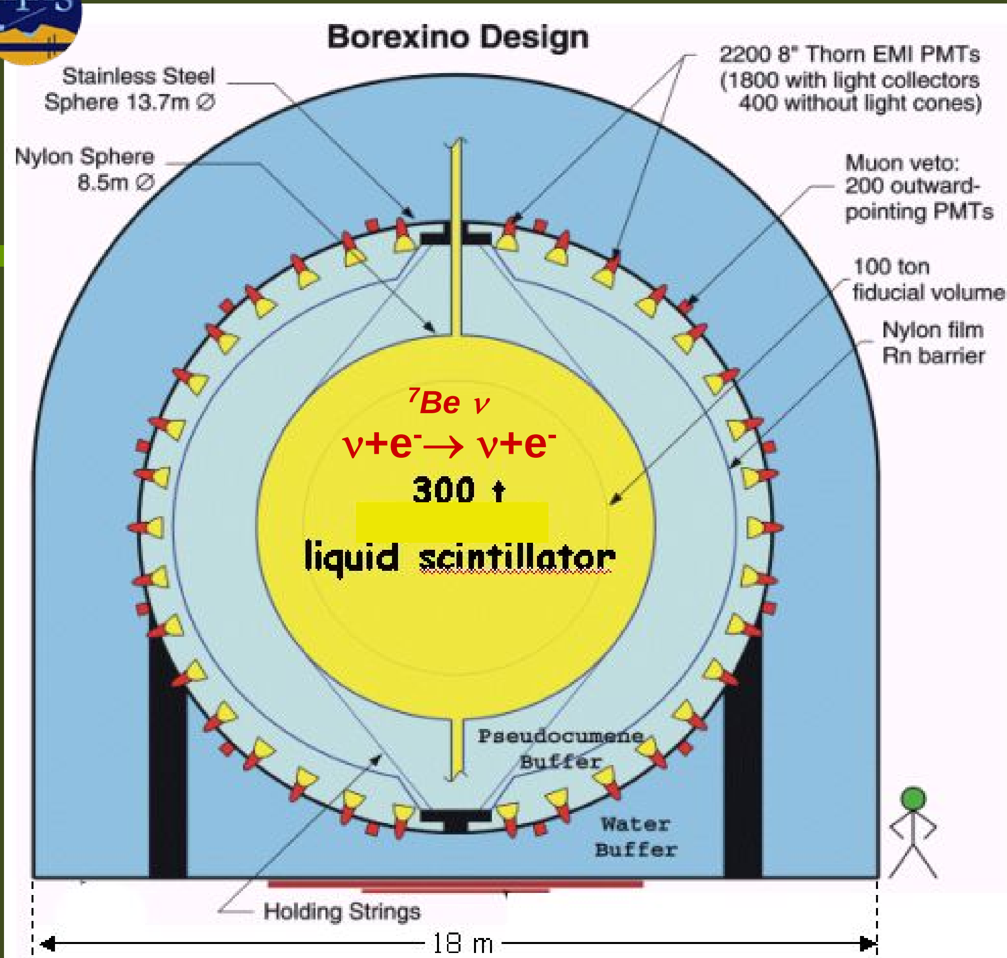
effective volume of sample chamber ~ 15 l

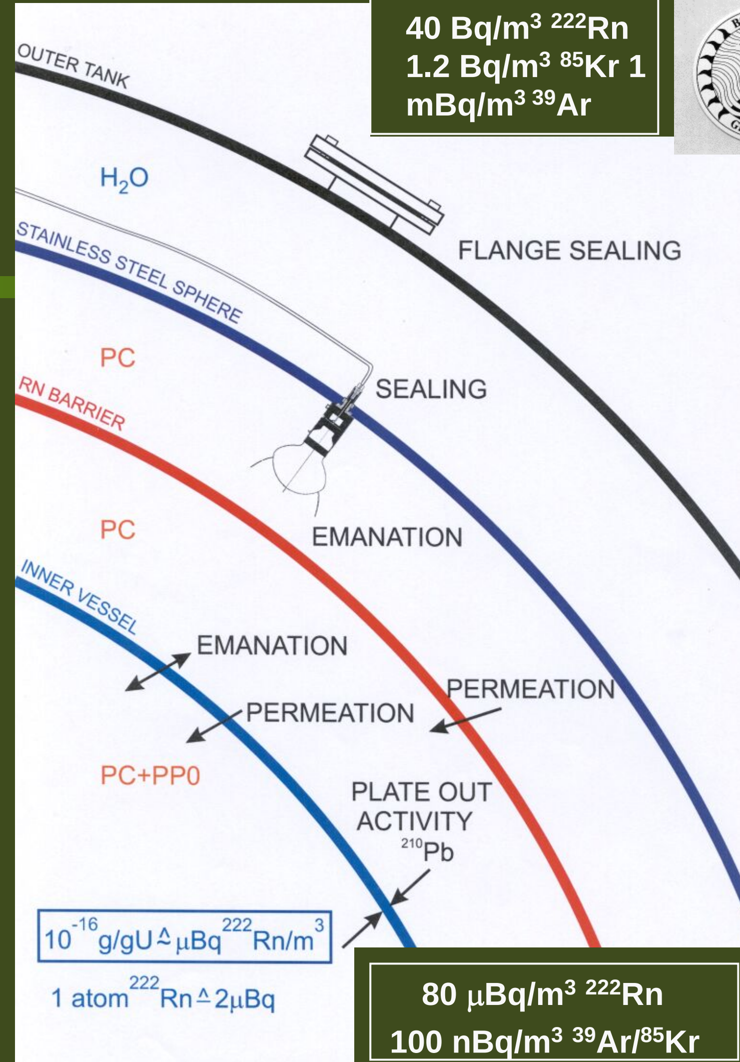
(e.g. 125 kg Cu or 157 kg Pb)

High purity copper directly placed underground after electrolysis



Rn emanation assay and gas proportional counting





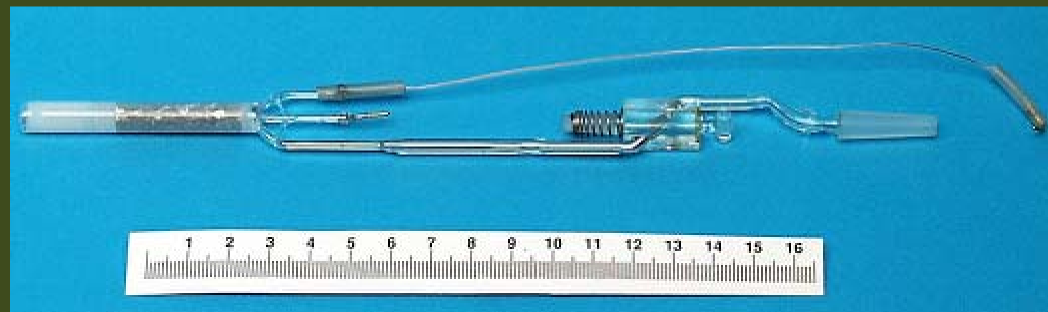
40 Bq/m^3 ^{222}Rn
1.2 Bq/m^3 ^{85}Kr 1 mBq/m^3 ^{39}Ar

80 μBq/m^3 ^{222}Rn
100 nBq/m^3 ^{39}Ar/^{85}Kr

- develop methods to detect noble gas radionuclides and ^{226}Ra (via ^{222}Rn) at the **μBq** level
- screen relevant materials and subsystems at that level
- provide nitrogen for scint. purification at the req. level

^{222}Rn (^{226}Ra) assay with proportional counting

^{71}Ga (ν_e, e$^-$)^{71}Ge



Ray Davis Jr. type miniature counter

efficiency for internal counting (> 15 keV): 148 %

background: (0.2 – 2) counts per day

$\Rightarrow$ about **30 µBq** ^{222}Rn easily detectable (monitoring)

Extract Rn from large quantities of water, nitrogen and as an emanation signal of subsystems of BOREXINO

H_2O: 1 mBq Ra/m^3
0.1 mBq/Rn/m^3

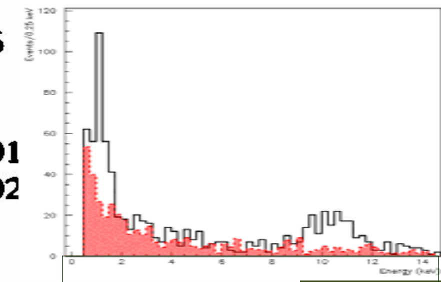
Reached sensitivities:

nitrogen: 0.5 µBq/m^3

surface emanation 0.5 µBq/m^2

composition of background for Fe cathode counters in Pb/Cu shield at LNGS

Source	Activity or flux at the position of the proportional counter	Count rate > 0.5 keV [cpd]
External sources		
Muons	3×10^{-8} cm^{-2} s^{-1}	0.005
Neutrons	$< 10^{-6}$ cm^{-2} s^{-1}	< 0.001
Gamma rays	$< 10^{-6}$ cm^{-2} s^{-1}	< 0.02
Rn + progenies	< 0.5 Bq m^{-3}	< 0.006
K, Th, U in copper of the shielding material	< 2, 1, 1 mBq/kg	< 0.02
Internal sources		
K in quartz	0.04 mBq/kg	0.0001
Th in quartz	< 0.01 mBq/kg	< 0.0002
U in quartz	< 1.2 mBq/kg	< 0.03
^{60}Co in iron cathode	< 7 mBq/kg	< 0.02
K in iron cathode	0.06 mBq/kg	0.001
^{226}Ra in iron cathode	< 3 mBq/kg	< 0.2
Th in iron cathode	< 0.3 mBq/kg	< 0.017
U in iron cathode	< 0.4 mBq/kg	< 0.03
Tritium in counting gas	6 TU	0.023
^{85}Kr in counting gas	< 0.12 Bq m^{-3}	< 0.01
Sum		< 0.39



All GNO runs recorded during the first 50 days

Background rates for counters used in GNO (0.5 keV ≤ E ≤ 15 keV) : 0.45

(8 with Fe cathode and 12 with Si cathode)

With pulse shape discrimination [counts/d]	
L-window fast	**K-window fast**
0.040	**0.025**

some contamination introduced during assembly (glassblowing)

NAA, ICP-MS, AMS

Neutron activation analysis

$$n + {}^{41}K \rightarrow {}^{42}K \xrightarrow[12.36\,h]{\beta^-} {}^{42}Ca\,, \quad \sigma_{th} = 1.2\,b\ (1.2\,b)$$

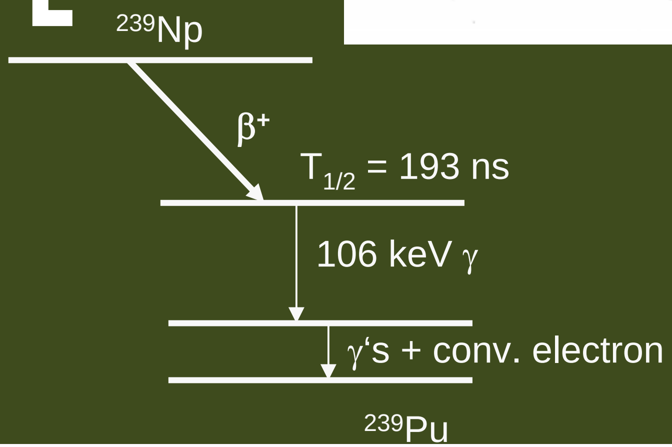
$$n + {}^{232}Th \rightarrow {}^{233}Th \xrightarrow[22.3\,m]{\beta^-} {}^{233}Pa \xrightarrow[27.0\,d]{\beta^-} {}^{233}U\,, \quad \sigma_{th} = 6.1\,b\ (7.8\,b)$$

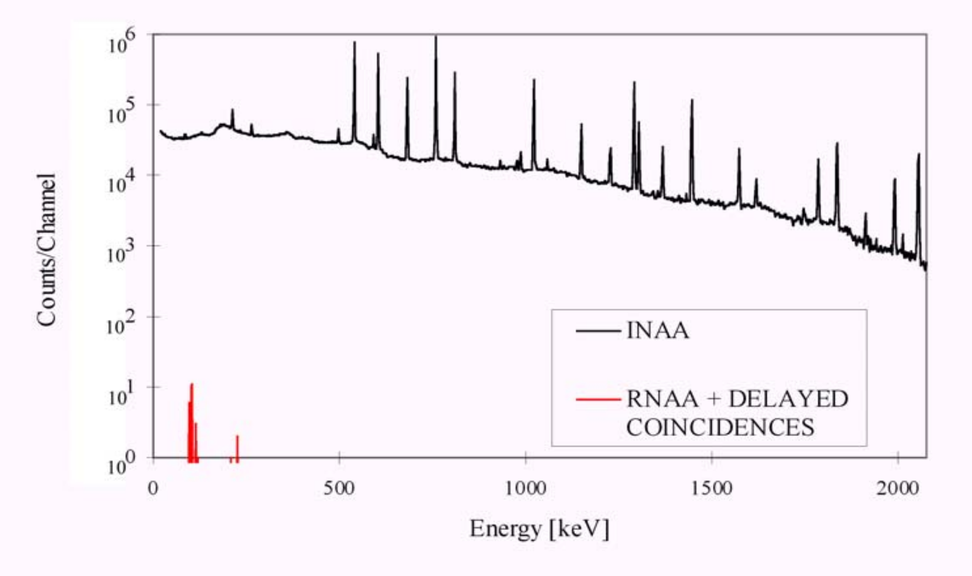
$$n + {}^{238}U \rightarrow {}^{239}U \xrightarrow[23.5\,m]{\beta^-} {}^{239}Np \xrightarrow[2.36\,d]{\beta^-} {}^{239}Pu\,, \quad \sigma_{th} = 2.3\,b\ (7.9\,b)$$

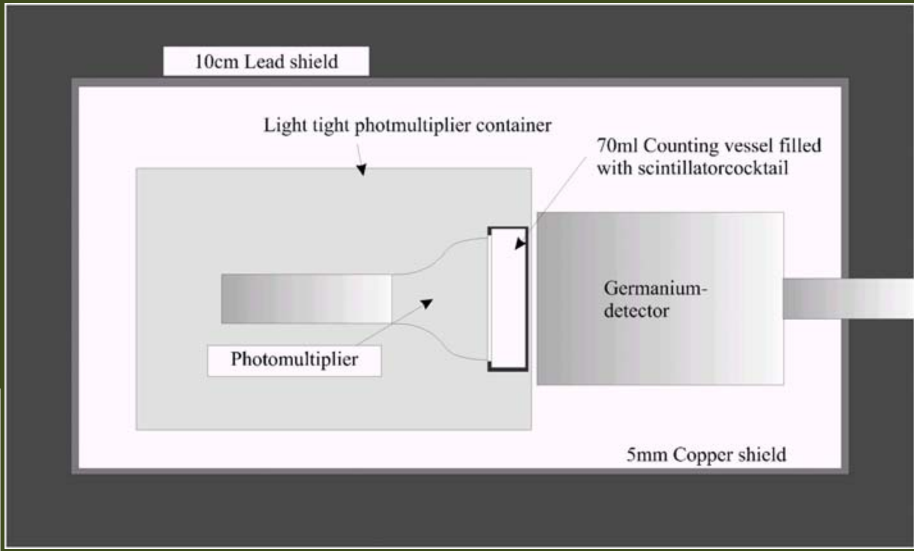
Sizeable cross sections and long enough half lives for delayed counting

RNAA (TU Munich)

$$n + {}^{238}U \rightarrow {}^{239}U \xrightarrow[23.5\,m]{\beta^-} {}^{239}Np \xrightarrow[2.36\,d]{\beta^-} {}^{239}Pu, \quad \sigma_{th} = 2.3\ b\ (7.9\ b)$$







PXE plus 1.5 g/l PPO

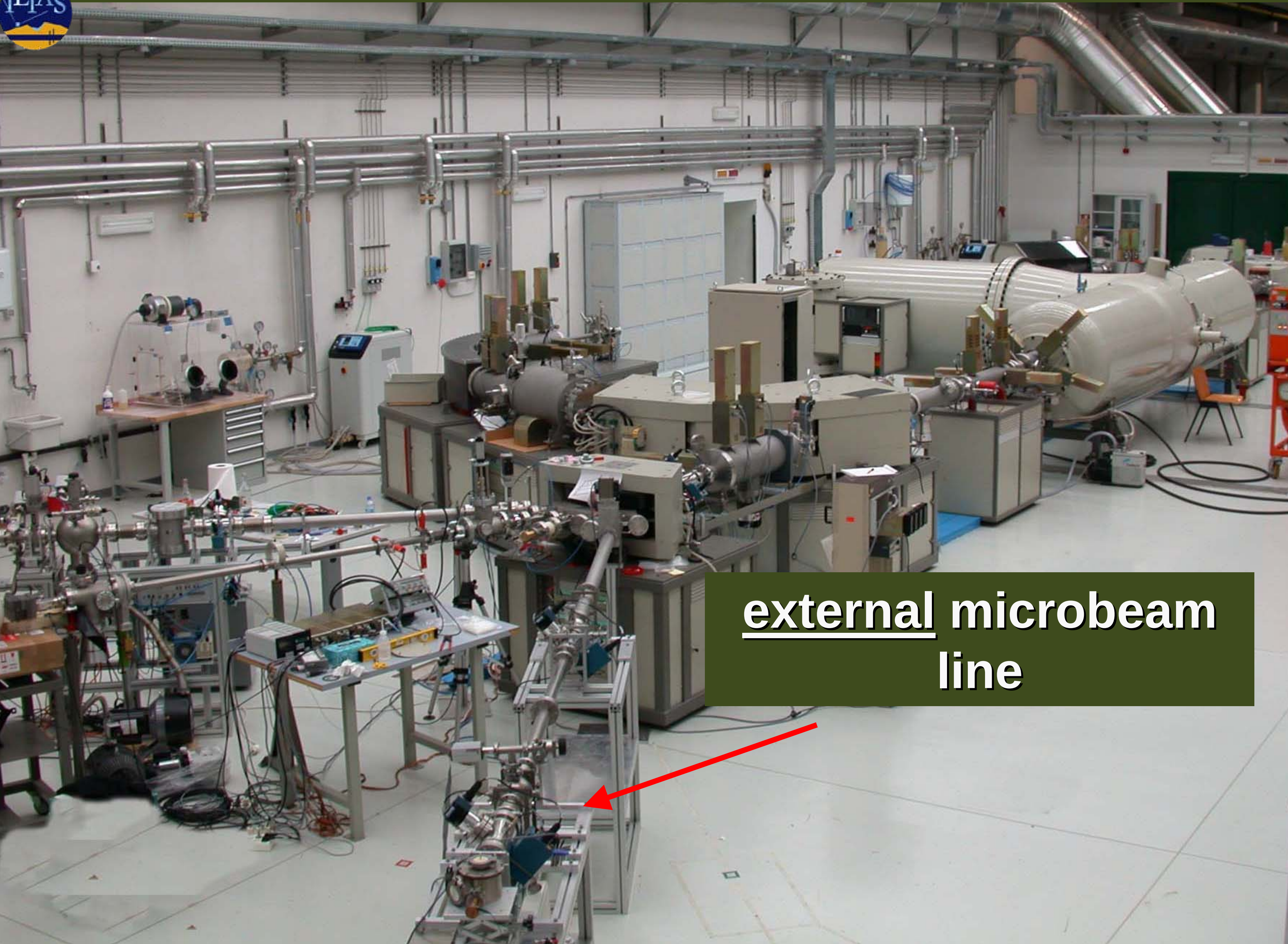
^{40}K $<7\cdot10^{-16}$ g/g	$\leq$ 0.19 µBq/kg
^{232}Th $<2\cdot10^{-16}$ g/g	$\leq 8\times10^{-4}$ µBq/kg
^{238}U $<1\cdot10^{-17}$ g/g	$\leq 1\times10^{-4}$ µBq/kg

Accelerator Mass Spectrometry (AMS)

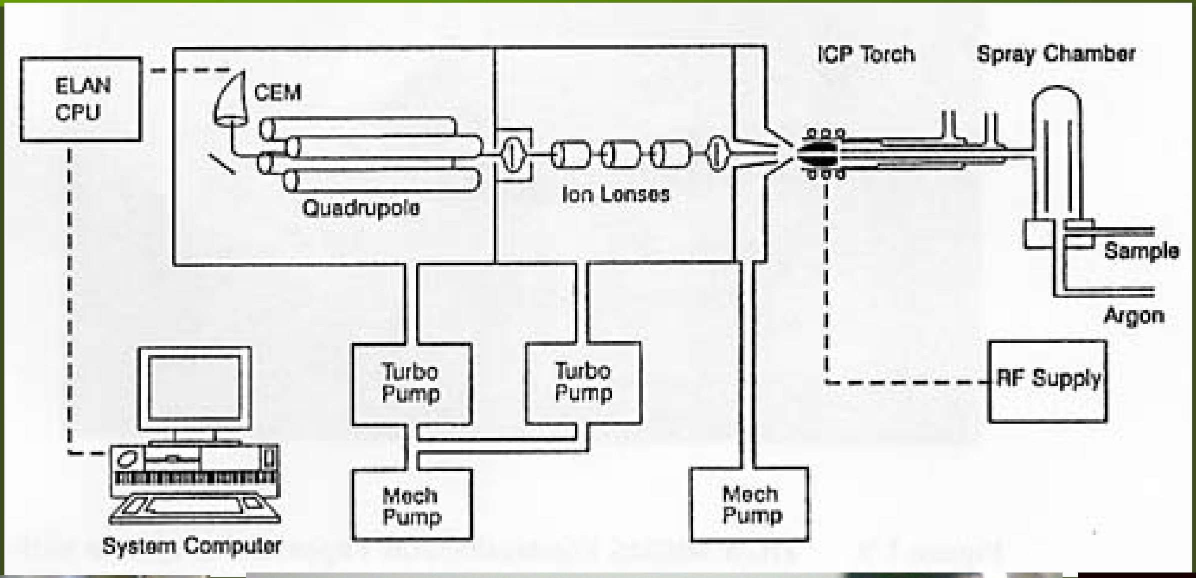
- a very sophisticated technique which detects rare isotopes

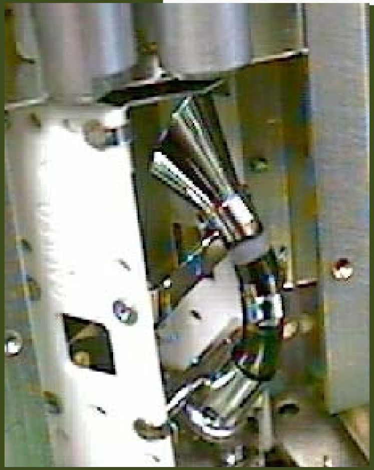
→ extraordinary sensitivity

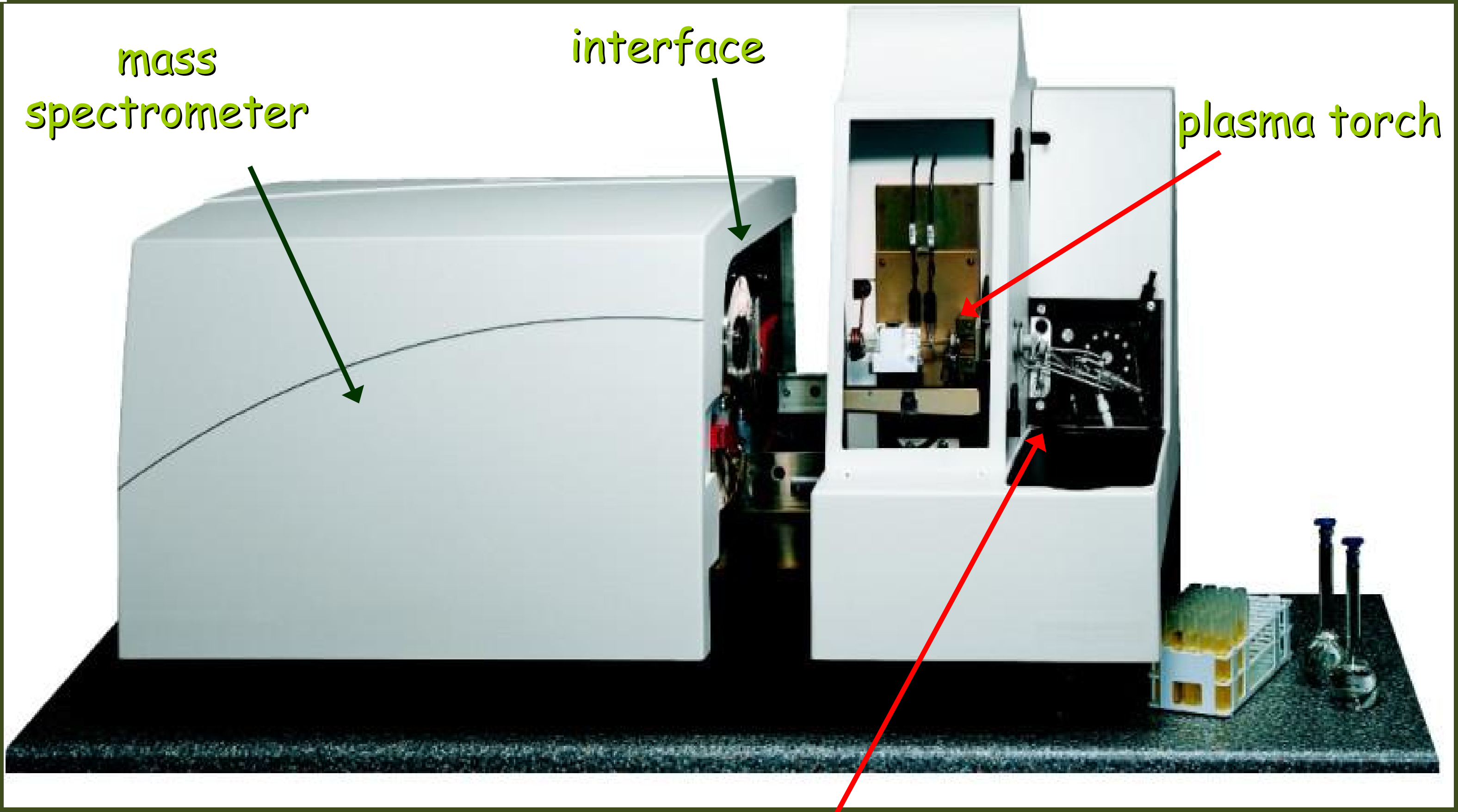
→ measurement of ^{10}Be, ^{14}C, ^{26}Al, ^{129}I and other radioisotopes of archaeological, geological, environmental interest


external microbeam line

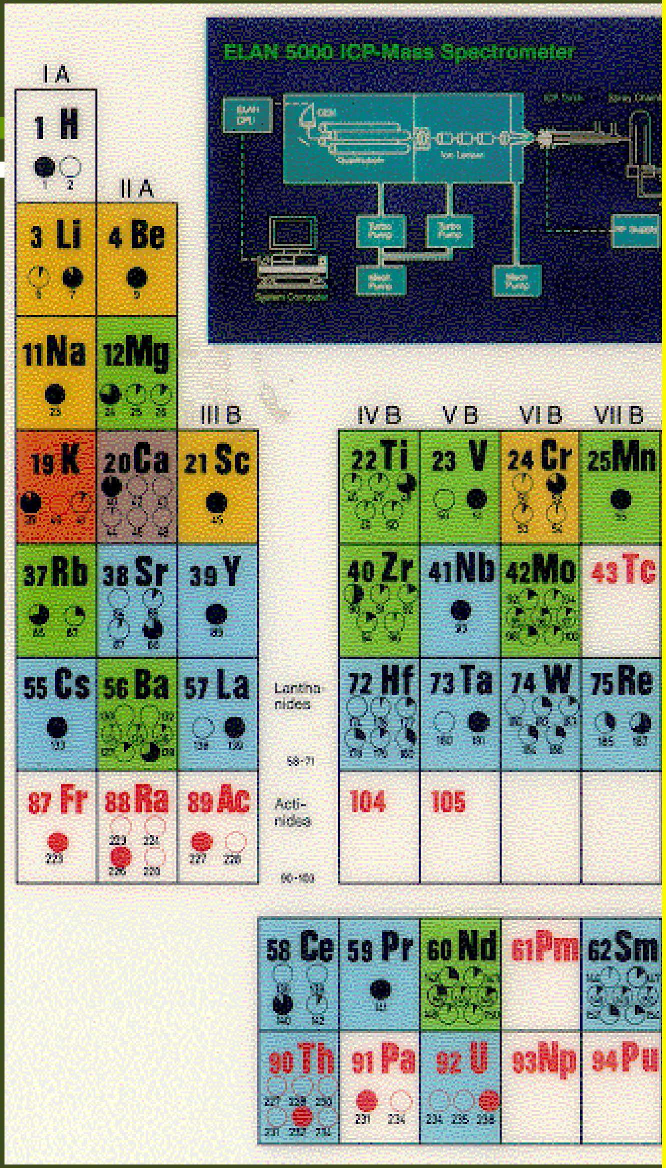
Inductively coupled plasma mass spectrometry (ICP-MS)






mass
spectrometer
interface
plasma torch
sample introduction

Detection limits of ICP-MS



^{238}U	~ 5×10^{-14} g	< 1 nBq
^{235}U	~ 5×10^{-15} g	< 1 nBq
^{234}U	~ 10×10^{-17} g	~ 20 nBq
^{236}U	~ 3×10^{-17} g	< 1 nBq
^{239}Pu	~ 3×10^{-17} g	< 100 nBq
^{240}Pu	~ 3×10^{-17} g	< 300 nBq
^{241}Pu	~ 3×10^{-17} g	~ 130 µBq
^{242}Pu	~ 3×10^{-17} g	< 1 µBq
^{241}Am	~ 3×10^{-17} g	~ 5 µBq
^{90}Sr	~ 1×10^{-15} g	~ 5 mBq
^{232}Th	~ 3×10^{-14} g	~ 1 nBq
^{230}Th	~ 3×10^{-17} g	~ 20 nBq

Comparison of detection limits

Table 3. Detection limit comparison (µg/L)

Element	ICP-MS	ICP-AES	Flame AAS	GFAAS
As	<0.050	<20	<500	<1
Al	<0.010	<3	<50	<0.5
Ba	<0.005	<0.2	<50	<1.5
Be	<0.050	<0.5	<5	<0.05
Bi	<0.005	<20	<100	<1
Cd	<0.010	<3	<5	<0.03
Ce	<0.005	<15	<200000	ND
Co	<0.005	<10	<10	<0.5
Cr	<0.005	<10	<10	<0.15
Cu	<0.010	<5	<5	<0.5
Gd	<0.005	<5	<4000	ND
Ho	<0.005	<1	<80	ND
In	<0.010	<30	<80	<0.5
La	<0.005	<0.05	<4000	ND
Li	<0.020	<1	<5	<0.5
Mn	<0.005	<0.5	<5	<0.06
Ni	<0.005	<10	<20	<0.5
Pb	<0.005	<20	<20	<0.5
Se	<0.10	<50	<1000	<1.0
Tl	<0.010	<30	<40	<1.5
U	<0.010	<30	<100000	ND
Y	<0.005	<0.5	<500	ND
Zn	<0.02	<1.0	<2	<0.01

ICP-MS, ICP-AES, Flame AAS: Detection limits (defined on the basis of 3 standard deviations of the blank)
GFAAS: Sensitivity (0.0044 absorbance) measured with 20 µL of sample
ND: Not determined

Special detectors (e.g. CTF)

Counting Test Facility (CTF)

- CTF is the prototype of Borexino. Its main goal was to verify the capability to reach the very low-levels of contamination needed for Borexino

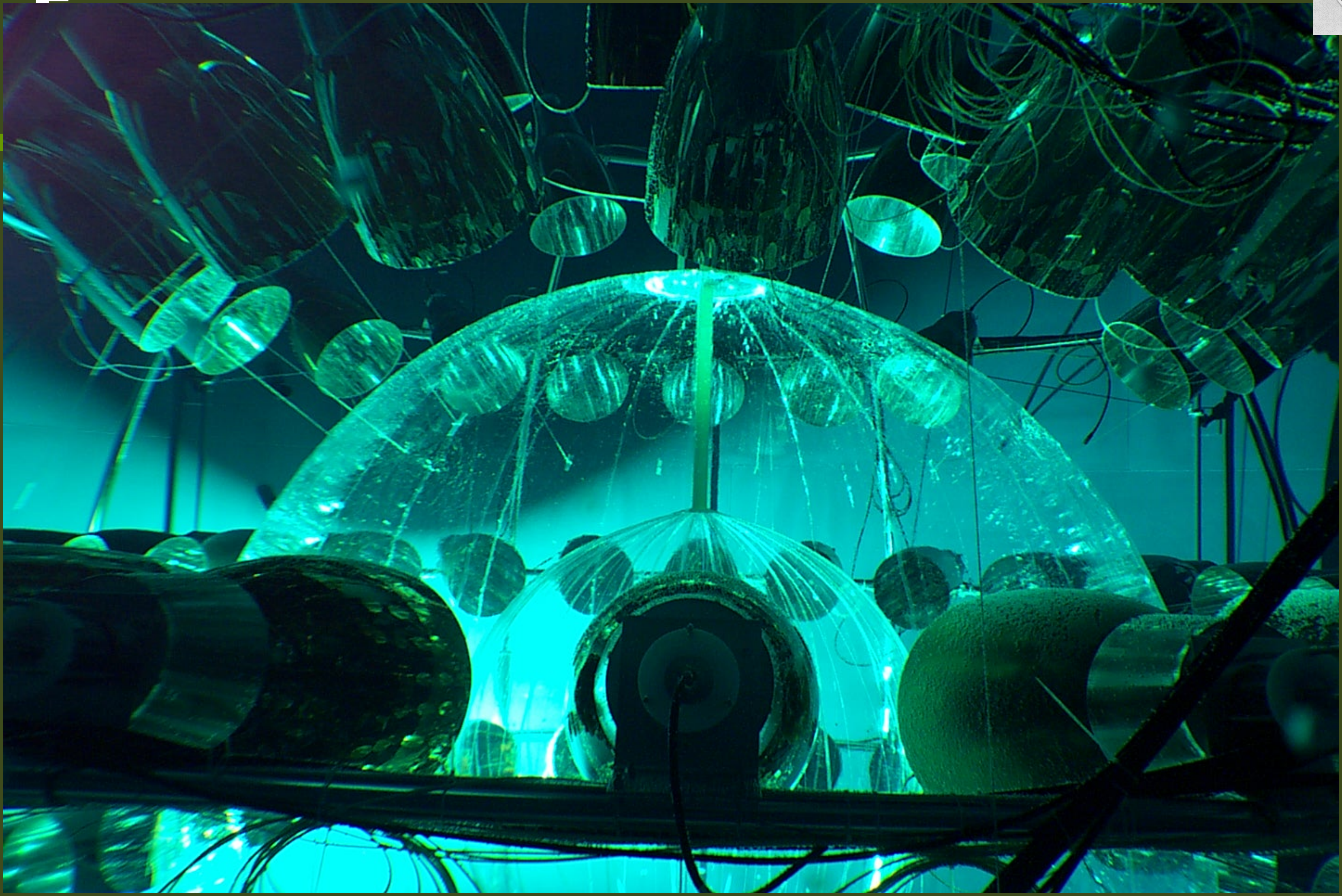
<u>CTF campaigns</u>

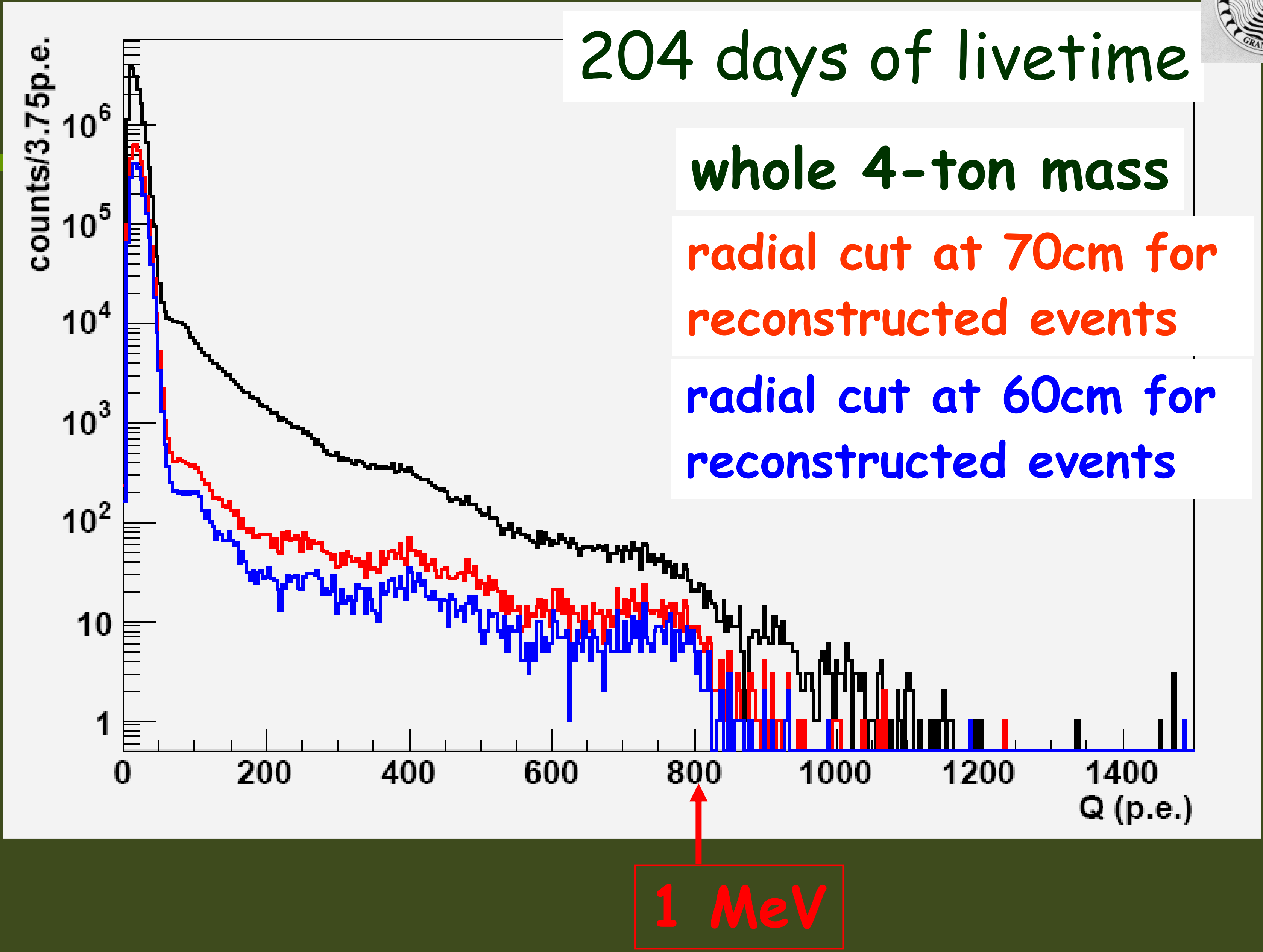
1. <u>CTF1</u>: 1995-1997
2. <u>CTF2</u>: 2000 (PXE)
3. <u>CTF3</u>: 2001 still ongoing

- 100 PMTs
- ~ 4 tons of scintillator
- 4.5 m thickness of water shield
- Muon-veto detector

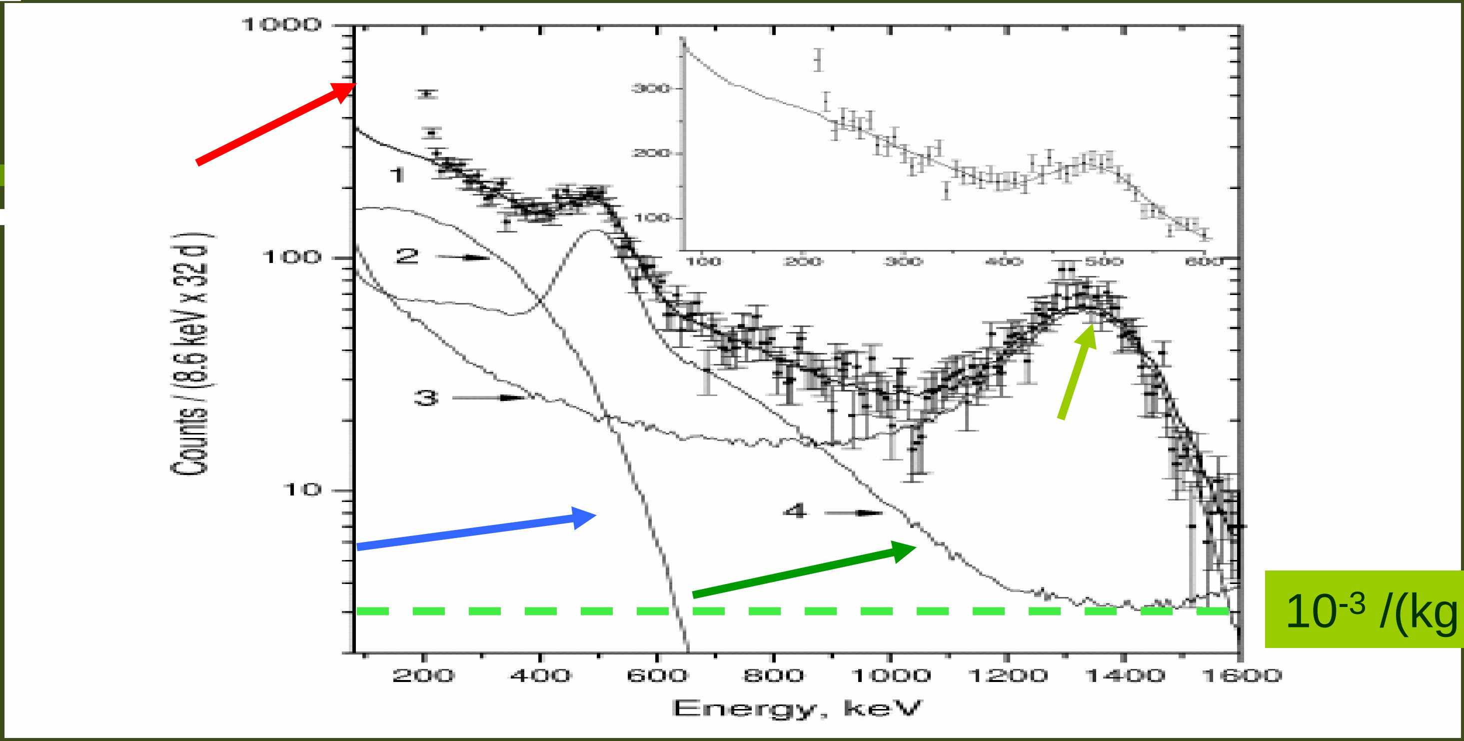
- $^{14}C/^{12}C \sim 10^{-18}$ (measured: $(1.94 \pm 0.09) \times 10^{-18}$))
- $^{238}U \sim 10^{-16}$ g/g (measured: $(3.5 \pm 1.3) \times 10^{-16}$) g/g, Rn daughters)
- $^{232}Th \sim 10^{-16}$ g/g (measured: $(4.4\ ^{+1.5}_{-1.0}) \times 10^{-16}$) g/g)

CTF high mass and very low levels of background contamination make it a unique detector to search for rare or forbidden processes with high sensitivity




204 days of livetime
whole 4-ton mass
radial cut at 70cm for reconstructed events
radial cut at 60cm for reconstructed events
counts/3.75p.e.
10^6
10^5
10^4
10^3
10^2
10
1
0
200
400
600
800
1000
1200
1400
Q (p.e.)
1 MeV

Background spectrum in CTF2



10^{-3} /(kg keV yr)

- The energy spectrum of the background counts in CTF is dominated by ^{14}C at low energy (<200 keV);
- Ar, Kr (up to ~700 keV);
- ^{238}U and ^{232}Th daughters (up to ~3 MeV);
- external ^{40}K (peak at 1.4 MeV + continuum);
- Muons mainly affect the very high energy region of the spectrum and can be effectively removed with the muon-veto detector

Outlook and conclusions

Well-known background reduction techniques

- muon veto
- anti-coincidence between detectors
- segmentation of readout electrodes
- pulse shape analysis (scintillators, semiconductors)
- coincidences in decay chain
- combination of complementary detection techniques (e.g. scintillation & ionisation)

Low-background electroformed copper - Soudan

Electroformed cups shown have wall thickness of only 250 mm!

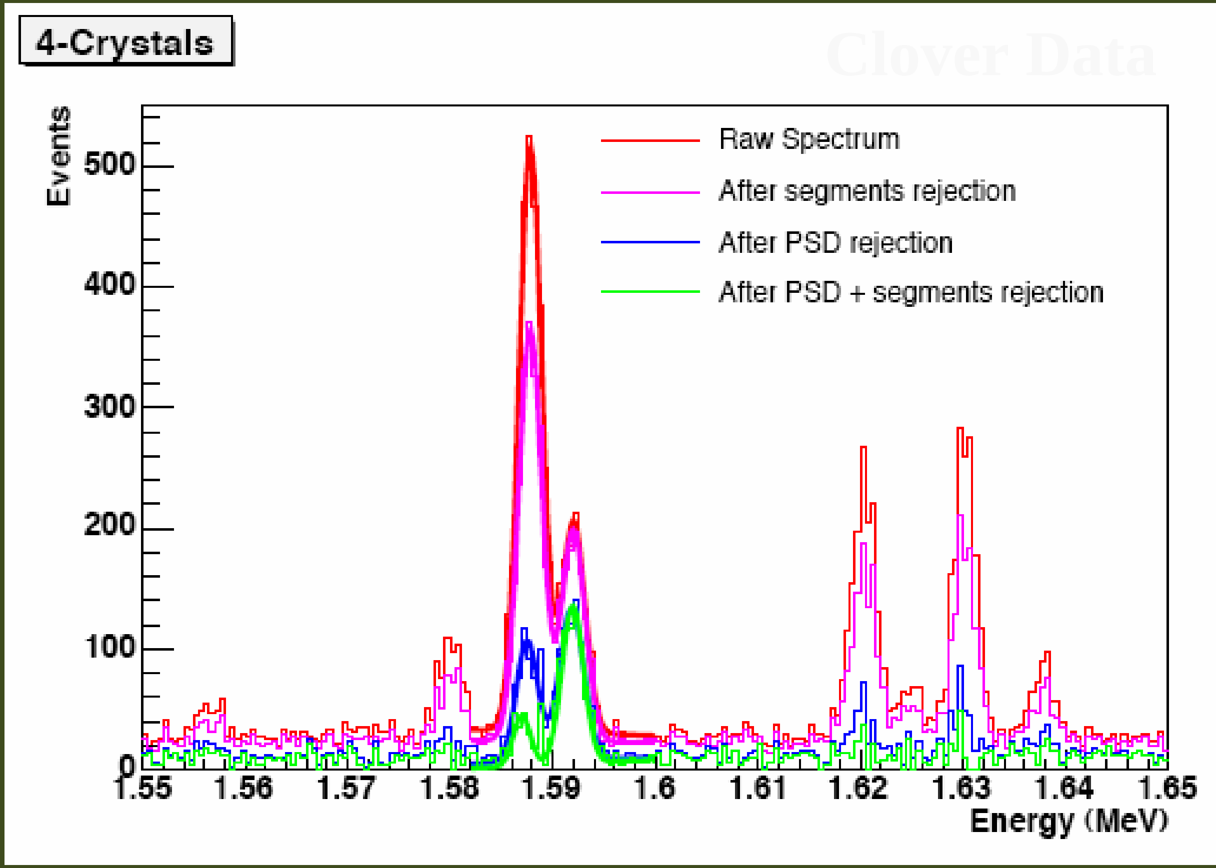
- can be easily formed into thin, low-mass parts
- UG electroforming can reduce cosmogenics
- pre-processing can reduce U-Th
- recent results suggest cleaner than thought

Low-background electroformed copper - Laboratorio Subterraneo Canfranc

- electroforming copper and lead melting facility started up

Segmentation R&D - Majorana collaboration

Data to confirm hypothesis that the PSD and segmentation cuts are orthogonal for Ge-diodes.



Ongoing improvement and R&D

- improved neutron shields (detector response, spectrum)
- improved material selection (more sensitive, better radiopurity e.g. $PbWO_4$ with archaeological lead)
- active shielding
- going deeper underground
- storage of freshly made construction materials underground
- multisegmented crystals or multiple crystals
- collaboration with producers (e.g. depleted Ge, crystal growing, Cu electroforming underground)
- the "ultimate" ultra-low background facility

Ongoing improvement and R&D

- future experiments need more sensitive screening techniques (< µBq/kg for ^{226}Ra) $\Rightarrow$ use of today's (e.g. CTF) or tomorrow's (e.g. GERDA) most sensitive detectors for screening

- future experiments need dedicated and highly sensitive screening and test techniques for measuring and monitoring *surface* contaminations (development and adaptation of existing techniques and methods to need, e.g. LA-ICP-MS)

- reorganisation and optimisation of existing screening facilities is necessary, because they are costly and measurement times can be rather lengthy

- harmonisation of how to report data and intercomparison programs for ultra low-level measurement techniques

New (and old) research applications

- ultra low-level chemistry
- particle astrophysics (material and techniques applicable to rare events experiments)
- space science (e.g. micro meteorites, Mars samples, cosmic activation products, comet tail samples)
- atmospheric samples ((very) short lived isotopes, radionuclide composition)
- ocean samples (deep ocean water - ^{60}Fe)
- in general application of low background techniques to interdisciplinary fields
  - low-level environmental radioactivity measurement and monitoring
  - radiodating (extension of determined ages towards the past)
  - geophysics (palaeoseismology, palaeogeology, sedimentation)

EU - FP7

- the ILIAS initiative in the EU FP6 should be pursued
- low background activities play an important role even outside the fundamental physics experiments
- proposal writing is ongoing