Gran Sasso high sensitivity instrumentation for trace radioactivity measurements the LNGS

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Plenary ECFA Meeting 2016

Laboratori Nazionali del Gran Sasso, Italy

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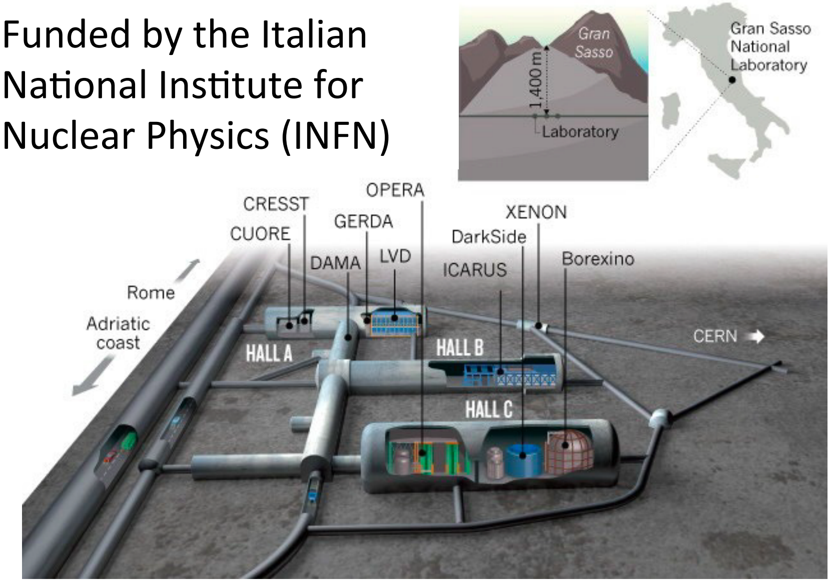
- Many thanks to:

Maria Laura Di Vacri and Stefano Nisi
for the part on ICP-MS

Gran Sasso National Laboratory (LNGS)

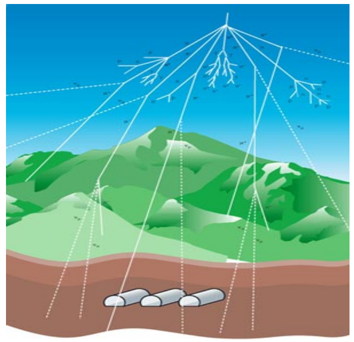
Funded by the Italian National Institute for Nuclear Physics (INFN)



- The largest and most advanced underground facility in the world devoted to neutrino and astroparticle physics research

- The 1400 m of rock (3600 mwe) above the laboratory provides:
• cosmic ray flux reduction: 10^6
• neutron flux reduction: 10^4

An ideal low-background environment to operate experiments searching for rare events



- Environmental radioactivity around the detector

use of suitable shielding to suppress the environmental radioactivity around the detector (i.e. lead, copper, PE)

- Natural radioactivity in the materials used for the detector construction (K, Pb, Th and U)

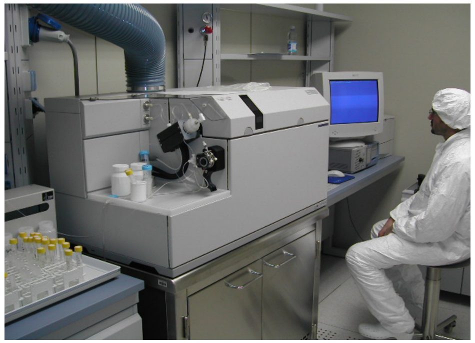
- strict selection of radiopure materials
- very high levels of radiopurity are required
- extremely sensitive analytical techniques

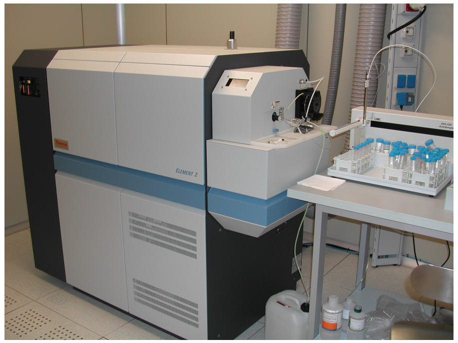
Ultra-low level radioactivity counting facilities at LNGS

STELLA (SubTErranean Low Level Assay)

- γ spectrometry (High-Purity Ge Detectors, HPGE)
- α spectrometry (Silicon PIPS detectors)
- liquid scintillation counters

Inductively coupled plasma mass spectrometry (ICP MS)



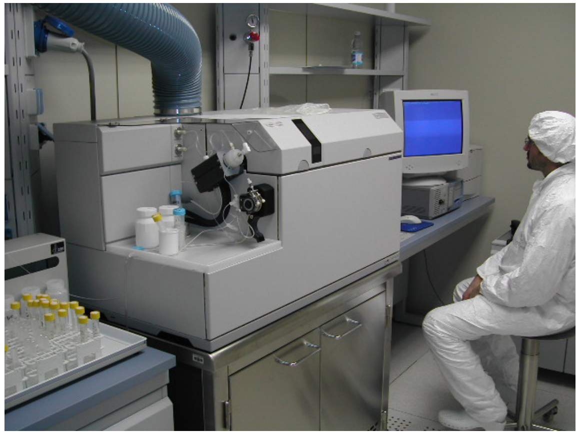


- 7500a Agilent quadrupole ICP MS
- Element2 Thermo Double Focusing High Resolution ICP MS
- Class 1000 clean room
- Sub boiling distillation system for reagents purification

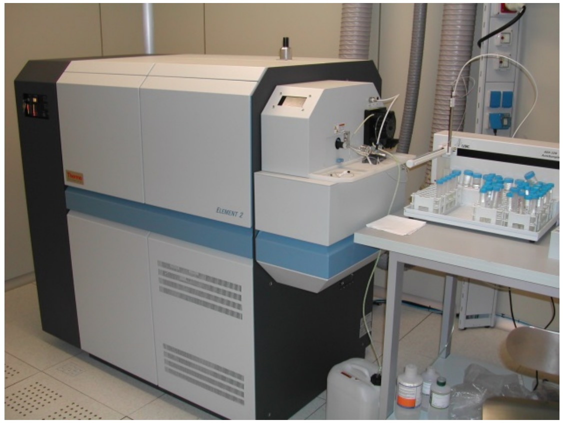
ICP MS measurements of low-level radioactivity in solid materials to be used in experiments

Mass spectrometry at LNGS

ICP MS



Agilent 7500a
quadrupole mass analyzer

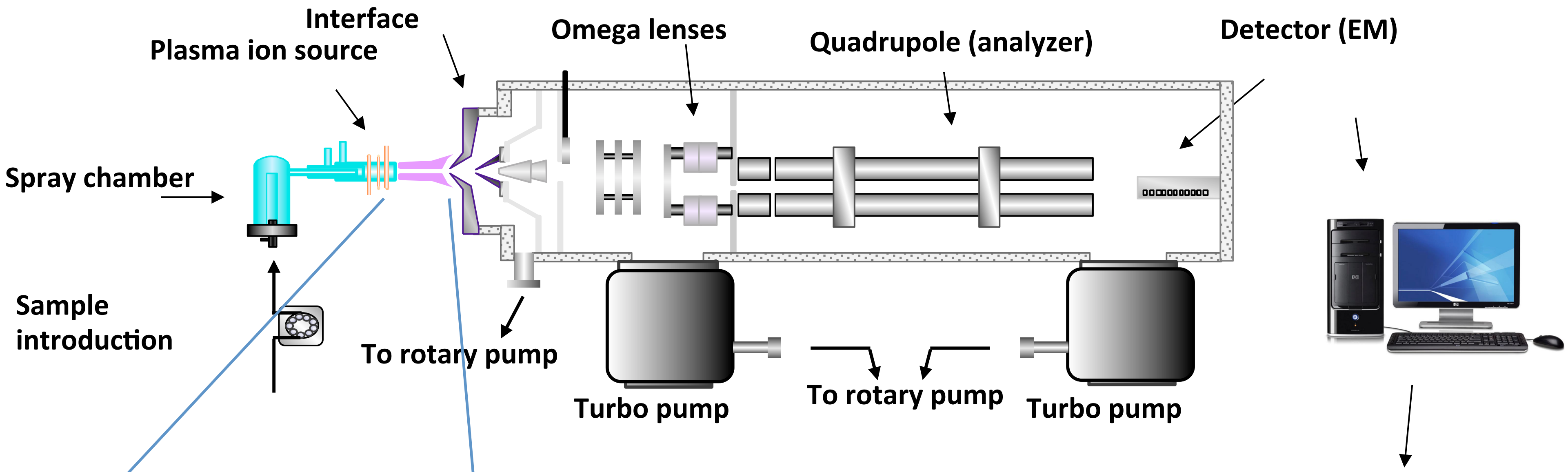


Thermo Element2 double focusing high resolution mass analyzer equipped with ESI APEX-Q desolvator

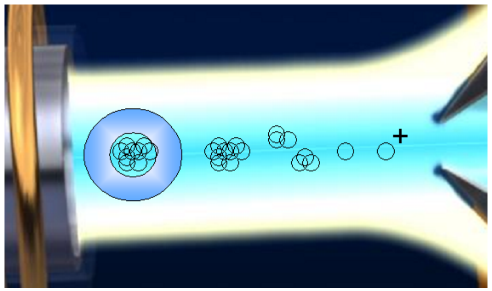
TIMS

Finnigam MAT 261/262
Thermal Ionization MS
Multicollector detector
(precise Isotope analysis)

ICP MS: trace element analysis

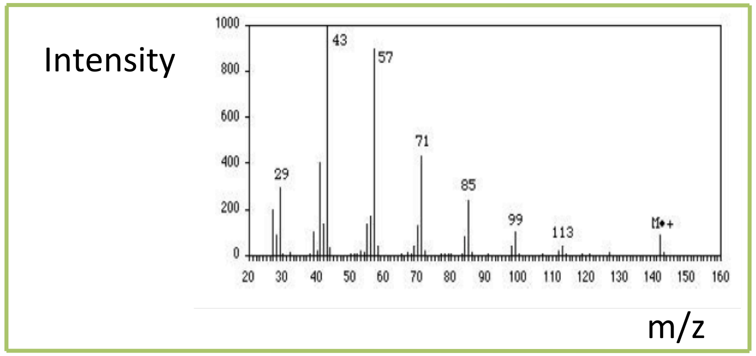


Plasma source (10000 K)



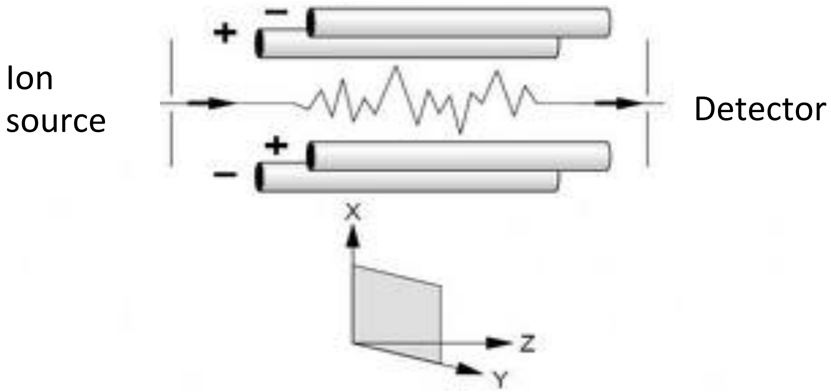
1. The aerosol is dried
2. Particles are decomposed to atoms
3. Atoms are ionized

Mass Spectrum



Mass analyzers

Quadrupole mass filter

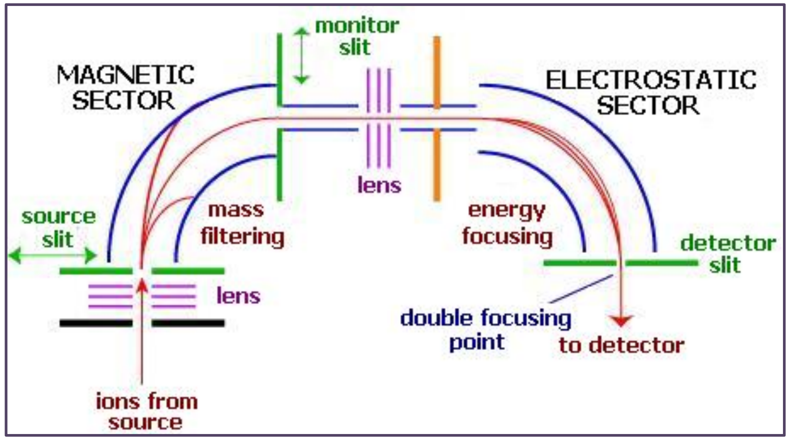


RESOLUTION POWER

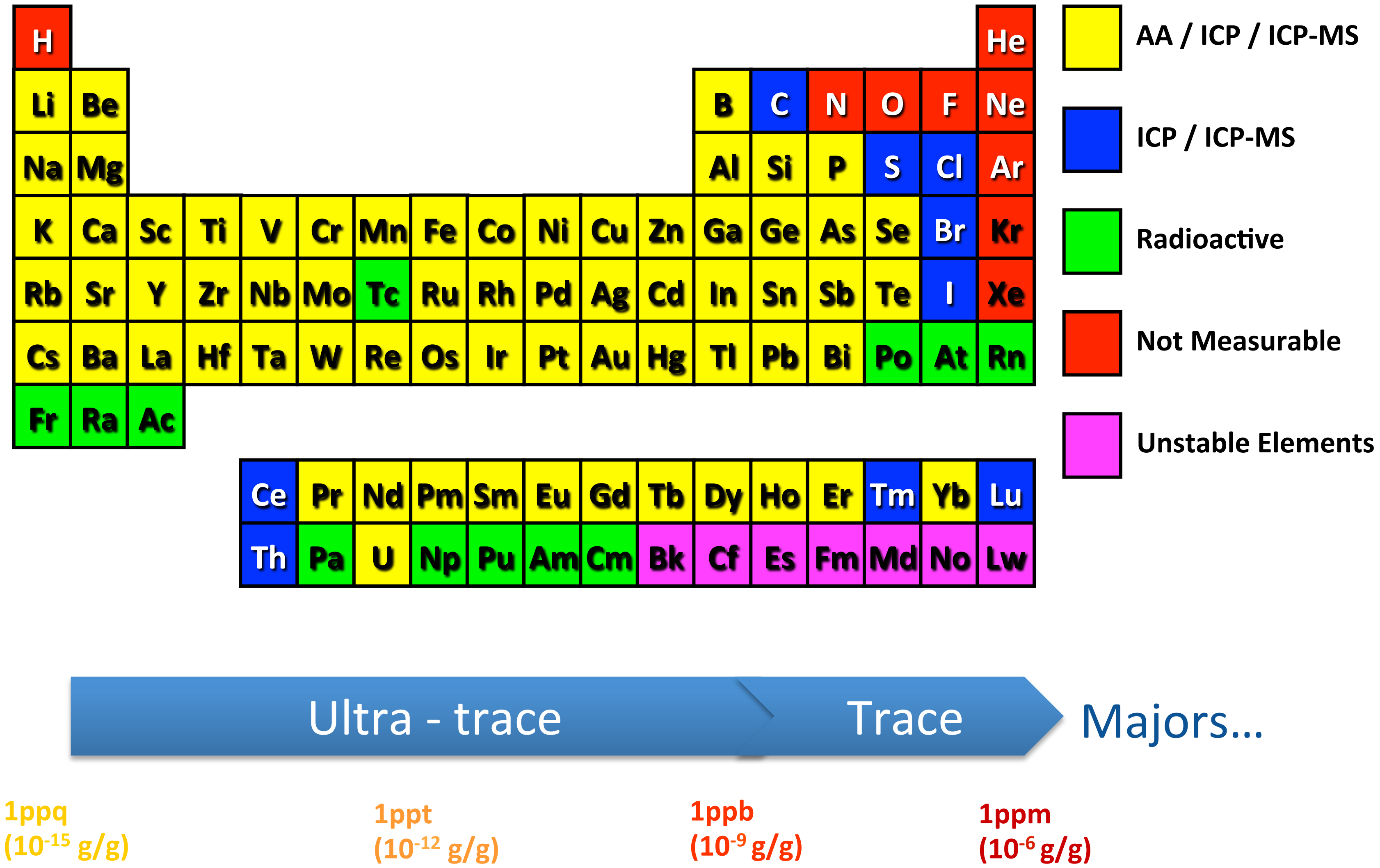
$$R = \Delta M/M$$

R ≈ 300 low resolution
R ≈ 4000 medium resolution
R ≈ 10000 high resolution

Double focusing mass analyzer (MSA + ESA)
High Resolution Mass Spectrometry

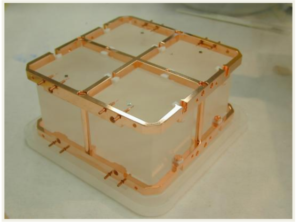


Measurable elements

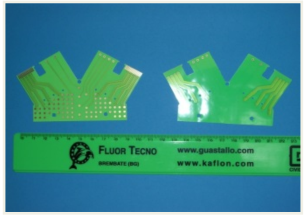


ICP MS main activity at LNGS: material screening for low background physics applications (K, Pb, Th U)

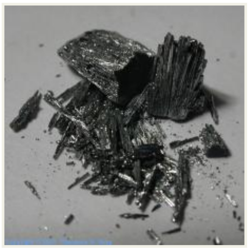
- ≈ 200 samples/year (complex matrices)
- few hundreds samples/year (reagents and water)




Cu, TeO2 and reagents
-CUORE-



Printed Circuit Board (PCB)
-GERDA-



Metals and alloys
-GERDA, XENON, DARK SIDE-

Al-Mylar insulating foils
-XENON, DARK SIDE-

Issues in ICP MS ultra-trace analysis for ultra-low background applications

- **Lack of validated methods and reference materials** (unusual matrices or analytes)

- High **risk of contamination** during sample preparation (we are looking for very very low concentrations!!!)

- **Background**

- **Isobaric interferences**

- **Sensitivity**, especially for solid samples (the instrument does not tolerate high matrix content, dilution is necessary) and **matrix effect**

Limiting factors to the achievement of strict detection limits required by neutrino experiments!!!

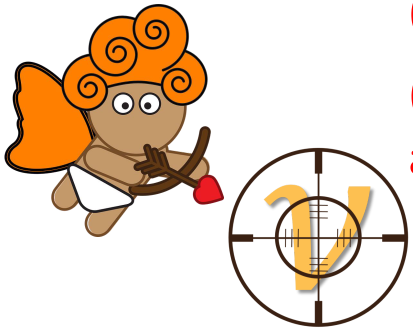
Our goal

Develop analytical procedures involving:
- matrix separation
- analyte pre-concentration
- background and contamination reduction
- increase of sensitivity

Improve detection limits in ICP MS radiopurity assay to meet low background experiment demanding requirements

Our current DL:

Sample	Liquid	Metals (solid)		Plastic (solid)
		Dissolution and dilution	Analyte separation	
Amount	0.5-5.0 mL	0.05-5.0 g	0.5-5.0 g	0.5-5.0 g
Element	ppt	ppt	ppt	ppt
K	500	50000	-	50000
Pb	1	500	-	100
Th	0.01	100	0.5	10
U	0.01	100	0.5	10



CUPID

(CUORE Upgrade with Particle Identification)
arXiv:1504.03612v1

A proposed future ton-scale bolometric 0νββ experiment, that will be built based on the experience gained in CUORE

CUPID aim: reaching a sensitivity for the effective Majorana mass of the order of 10 meV

↓

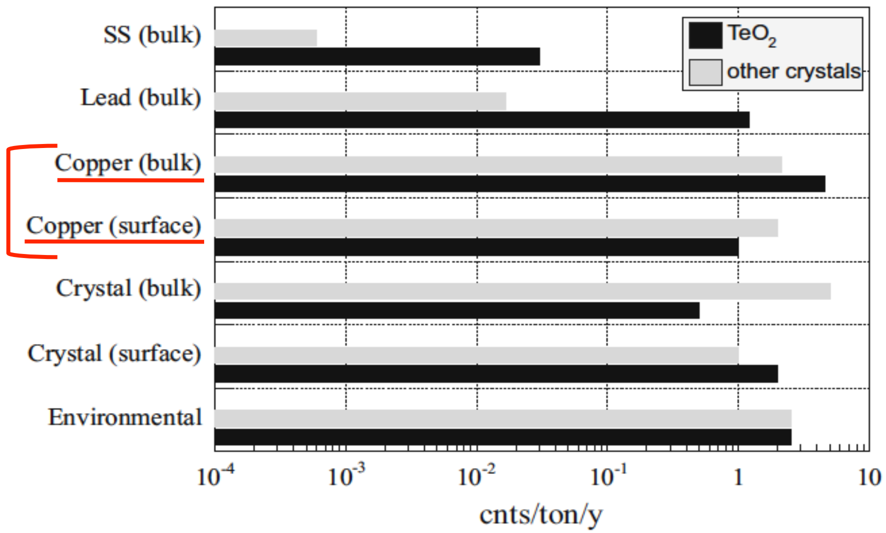
Ambitious target!

↓

Strict reduction of the background!

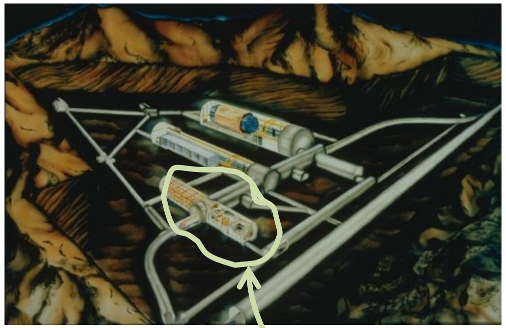
The CUPID experimental set up

Copper is envisaged to be one of the CUPID detector materials

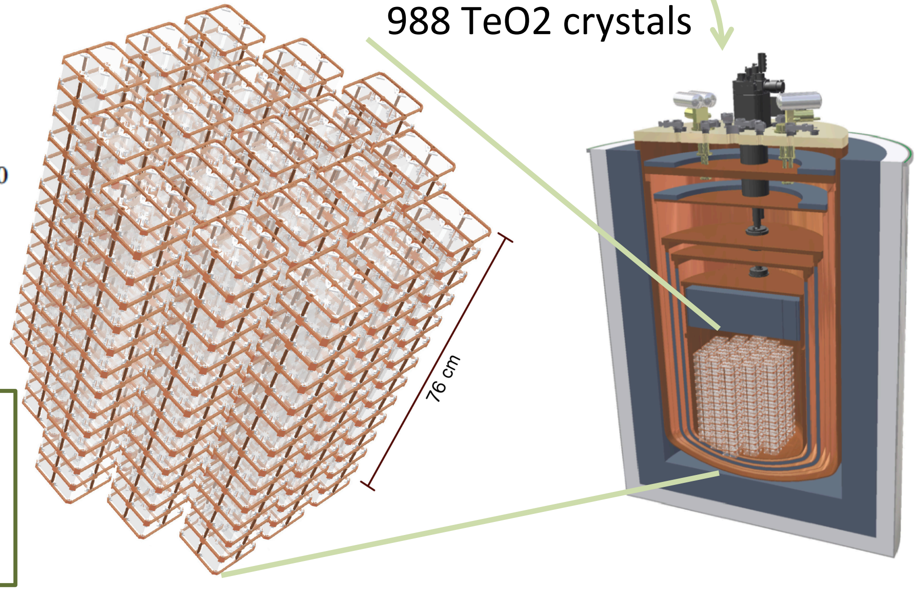


Background budget (upper limits)

Eur. Phys. J. C (2014) 74:3096



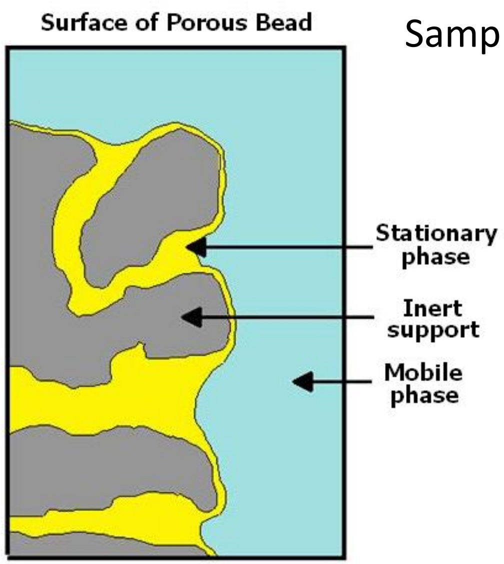
CUORE
T=10 mK
988 TeO2 crystals

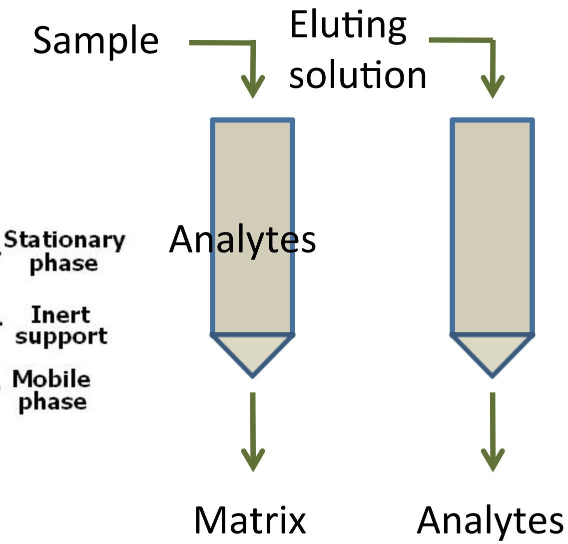


This translates in contamination level of the order of **ppt** or **fraction of ppt** of **Th and U in copper**

Development of an analytical procedure for the improvement of ICP MS detection limits for Th and U in copper

Extraction chromathography





Capacity factor k': $k' = D \frac{V_s}{V_m}$

Advantages:

- Matrix removal
- Analyte pre-concentration

Disadvantages:

- Time consuming
- Reagents
- Risk of contamination
- Higher amount of sample

Conclusions

- The developed procedure for the extraction and pre-concentration of Th and U allowed to improve ICP MS DL in copper of a factor ≈ 35 for Th and ≈100 for U

- Two resins have been tested, one of them gave very good results for both Th and U, the other one gave excellent results for U but not acceptable for Th

Future upgrade for DL improvement

- **Enhance Th recovery %** with UTEVA resin (which is ≈90% with other matrices)

- Improve the column rinsing efficiency to further **reduce background**

- Increase the sample amount to **reduce the dilution factor** (saturation tests)

- Further reduce DL in ICPMS measurement of Th and U ultra-traces in copper

- Study and develop similar procedures for the measurement of Pb and K

The Low Background Facility STELLA at the LNGS


EG&G ORTEC

Low background counting laboratory

some of the HPGe detectors working

type	volume [cm^3]	rel. efficiency	FWHM [keV]
GeBer n-type	235	56%	2.0
GeMi p-type	403	86%	1.9
GePV p-type	363	91%	1.8
GsOr p-type	414	96%	1.9
GePaolo p-type	518	113%	2.0
GeCris p-type	465	120%	2.0
GeMulti p-type	4×225	4×96%	2.0

Activities

- **material screening (e.g. GerDA, Borexino, CUORE, Xenon)**
- CELLAR
- environmental radioactivity
- small fundamental physics research projects
- meteorite measurements

What is ULGS?

Ultra Low-level Gamma-ray Spectrometry

i.e. low-level g-spectrometry with additional background reduction by using active shields, material selection and/or underground laboratories

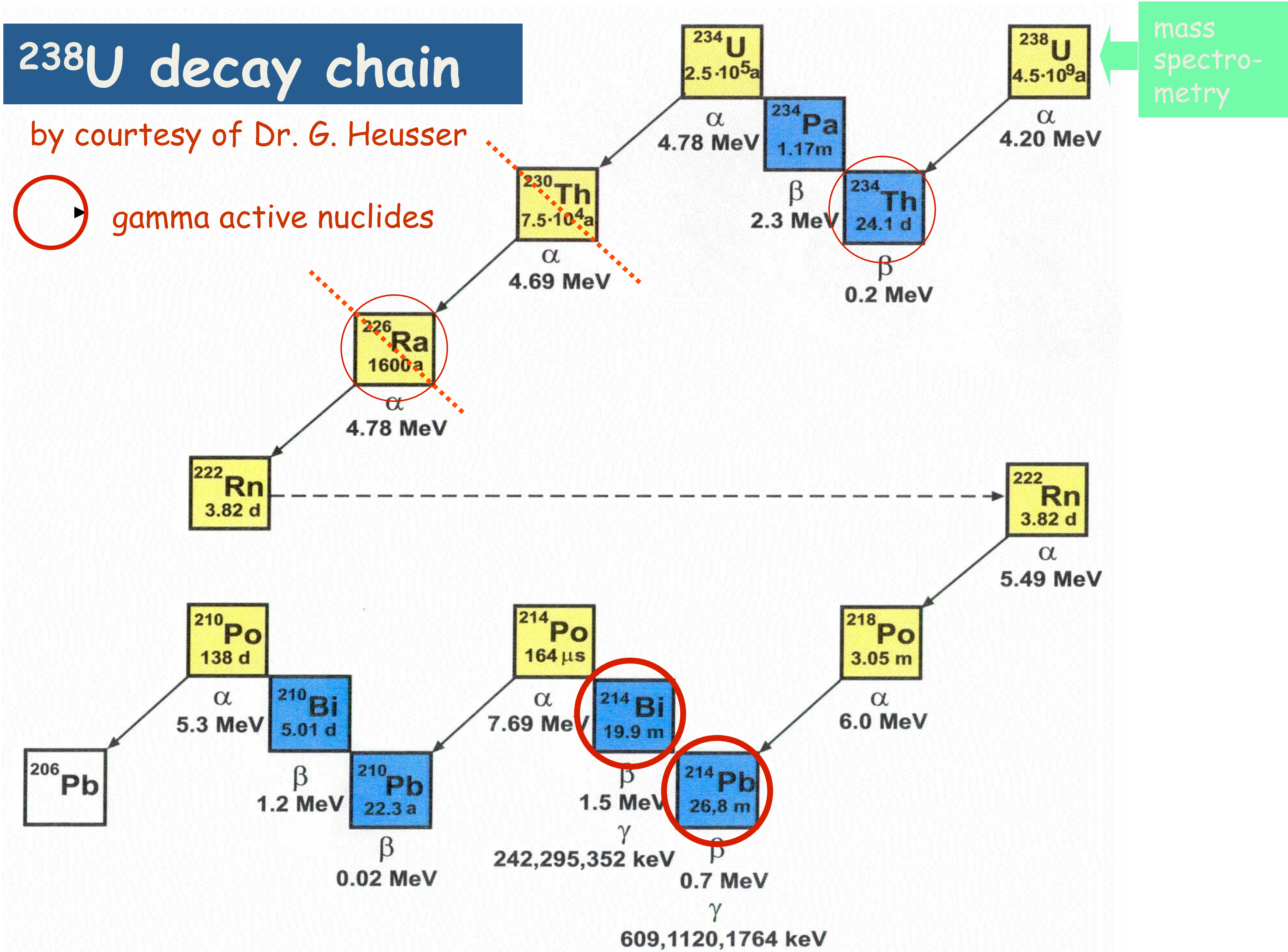
Sensitivity

Method	Detection limit for U and Th [Bq/kg]
ULGS (non-destructive) γ emitters	10^{-5} - 10^{-4}
ICP-MS (destructive) primordial parents	10^{-6} - 10^{-5}
ULGS + NAA primordial parents	10^{-7}

1 Bq ^{238}U/kg ≅ 81 x 10^{-9} g/g
1 Bq ^{232}Th/kg ≅ 246 x 10^{-9} g/g
1 Bq ^{40}K/kg ≅ 32 x 10^{-6} g/g

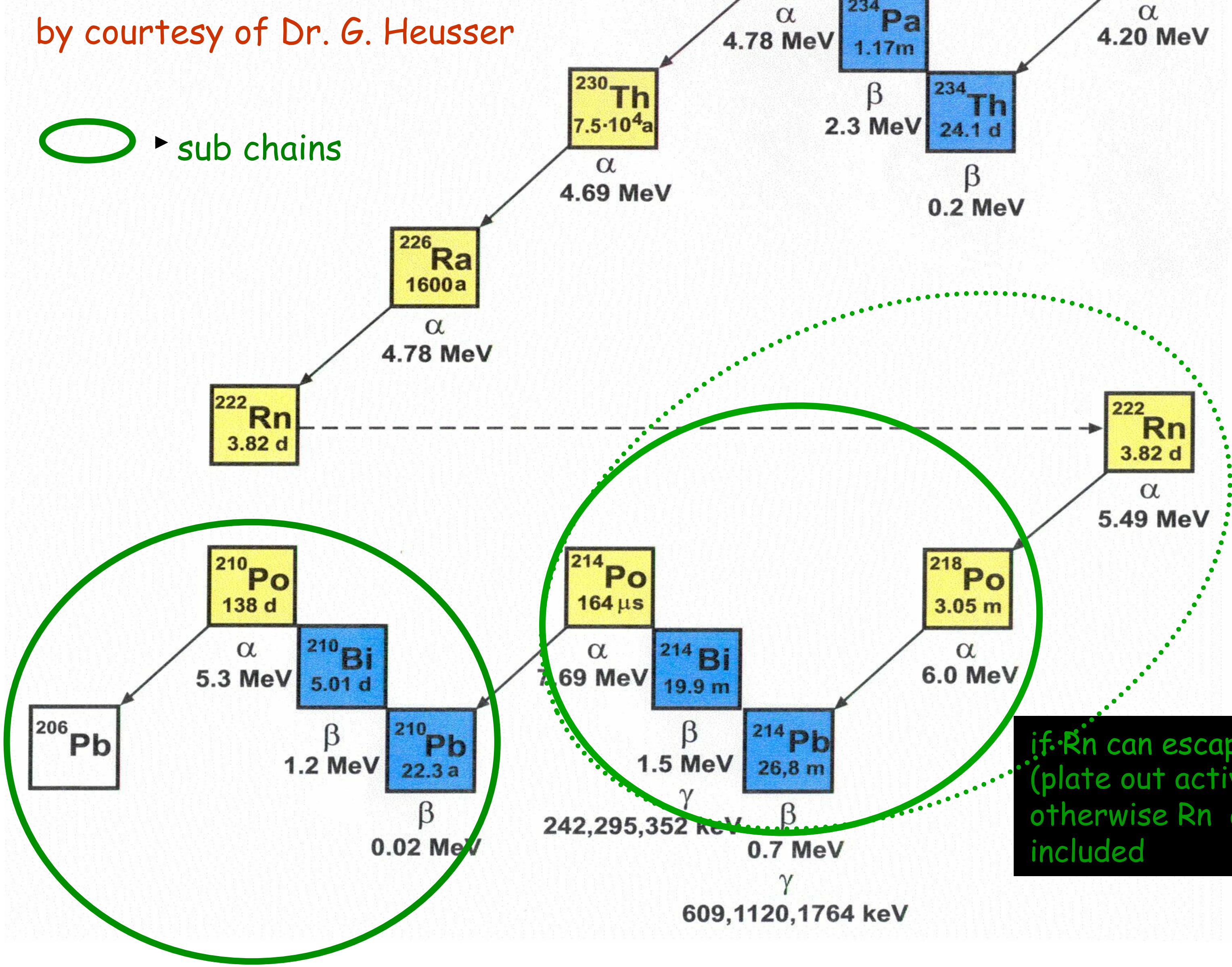
^{238}U decay chain

by courtesy of Dr. G. Heusser



^{238}U decay chain

by courtesy of Dr. G. Heusser



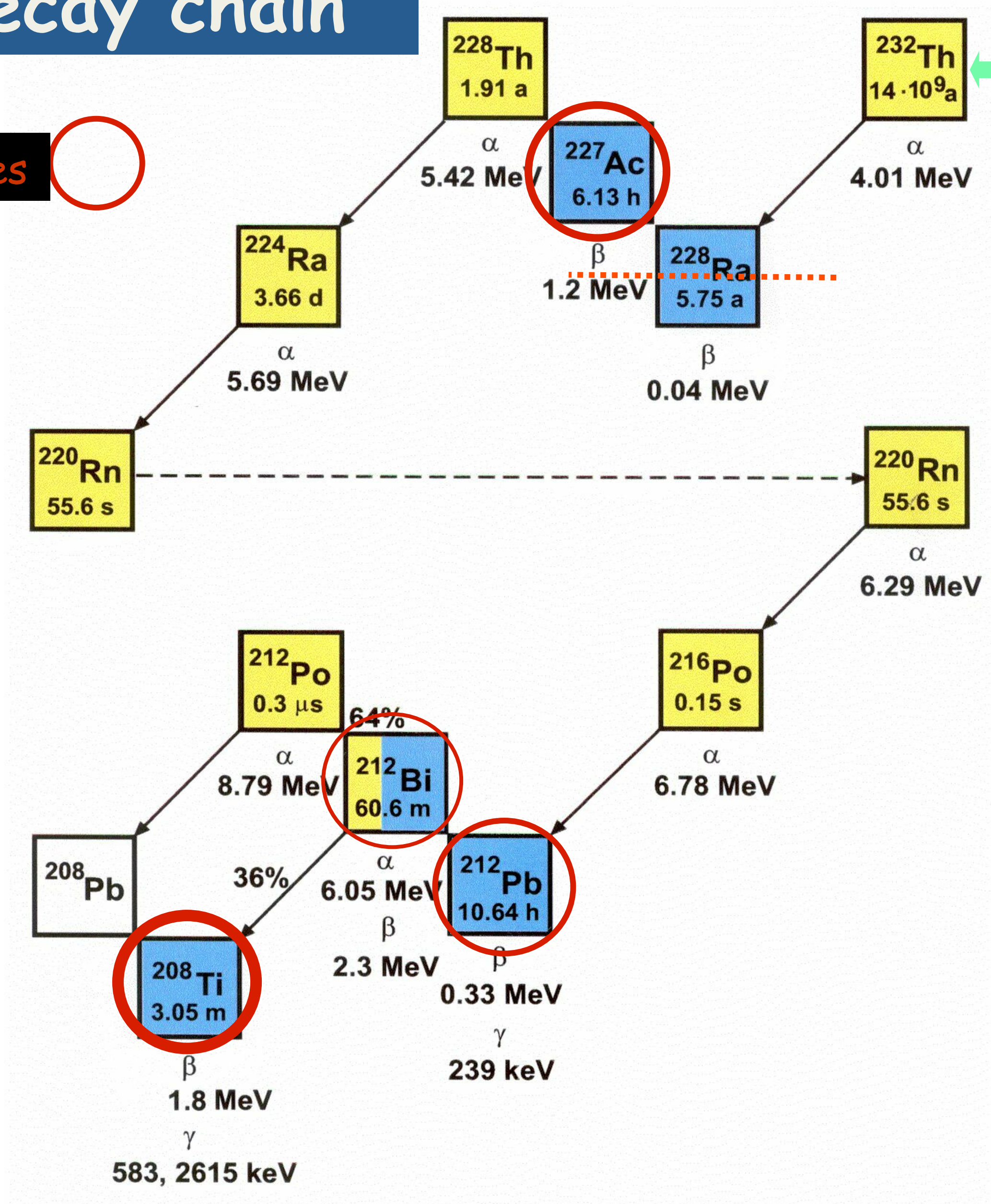
if Rn can escape, (plate out activity) otherwise Rn and Ra included

^{232}Th decay chain

by courtesy of Dr. G. Heusser

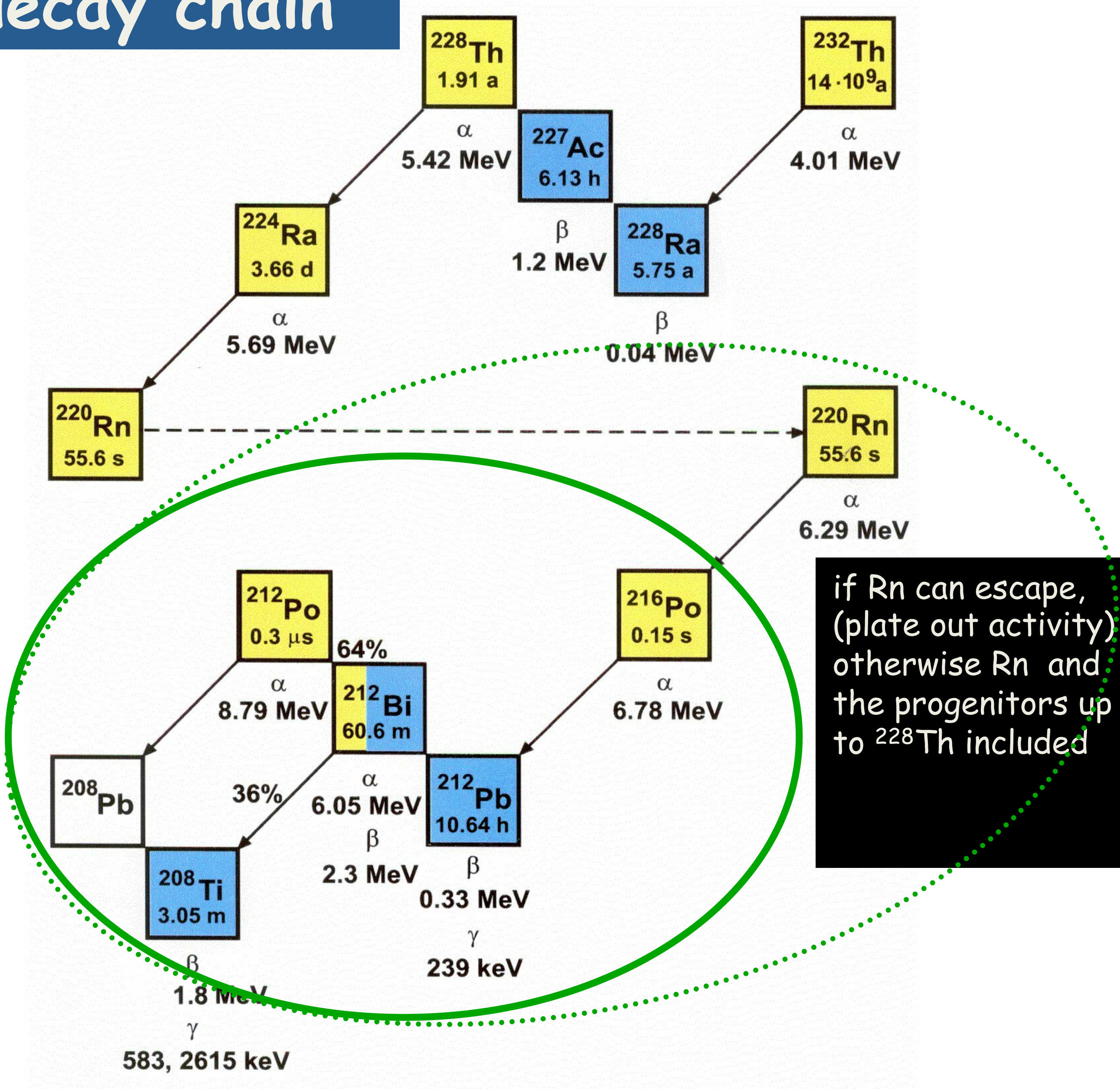
gamma active nuclides

mass spectrometry



^{232}Th decay chain

by courtesy of Dr. G. Heusser



if Rn can escape, (plate out activity) otherwise Rn and the progenitors up to ^{228}Th included

LNGS HPGe detectors

10 detectors installed, 8 p-type coaxial detectors, all LB or ULB configuration, one ULB well-type detector, one BEGe ULB detector

Sensitivity (U/Th):

commercial LB detectors O(mBq/kg)
commercial ULB detector O(0.5 mBq/kg)
custom ULB detector O(50 **μ**Bq/kg)

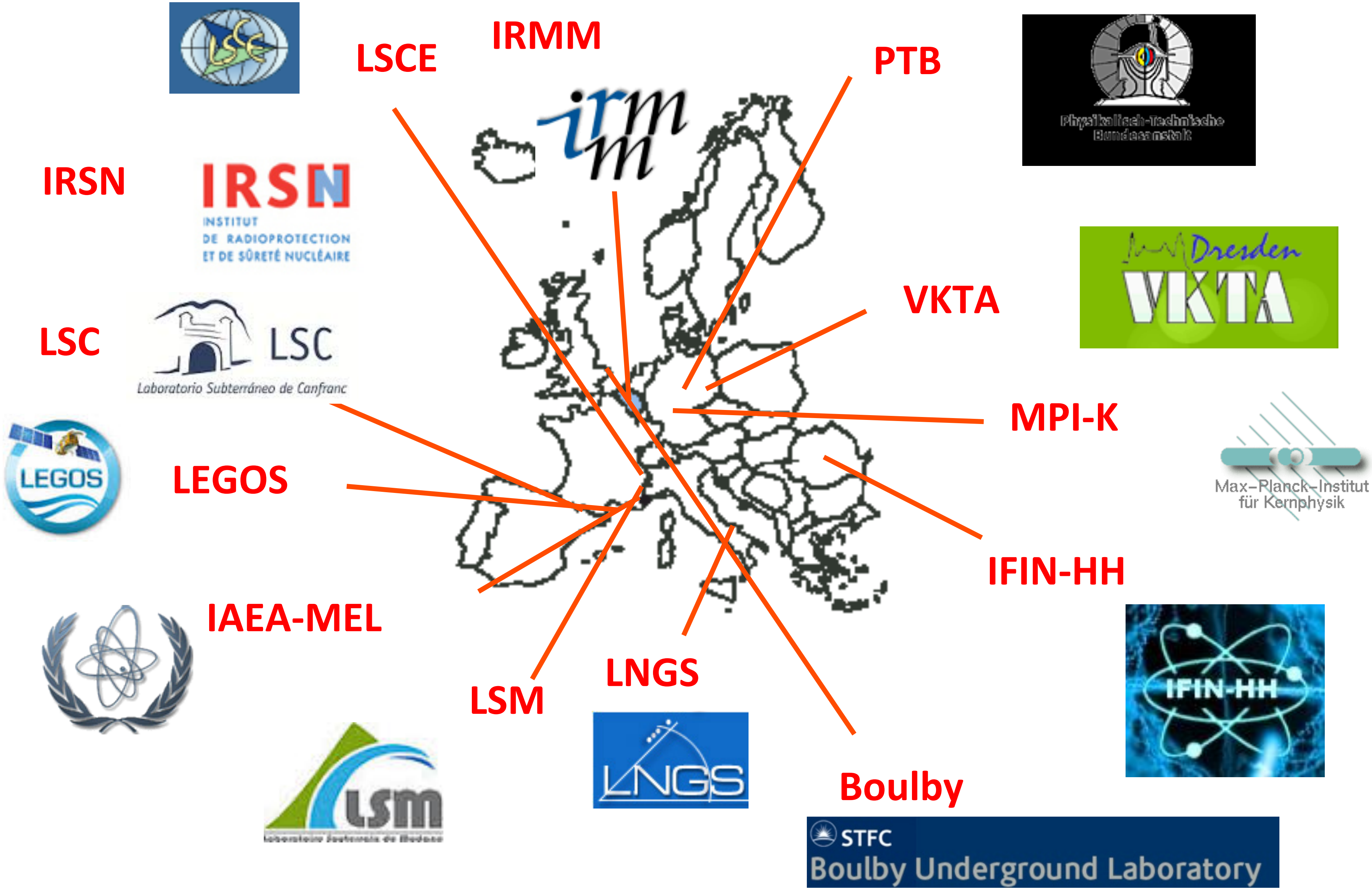
MPI-K (Prof. M. Lindner) GeMPI type detectors located at LNGS.

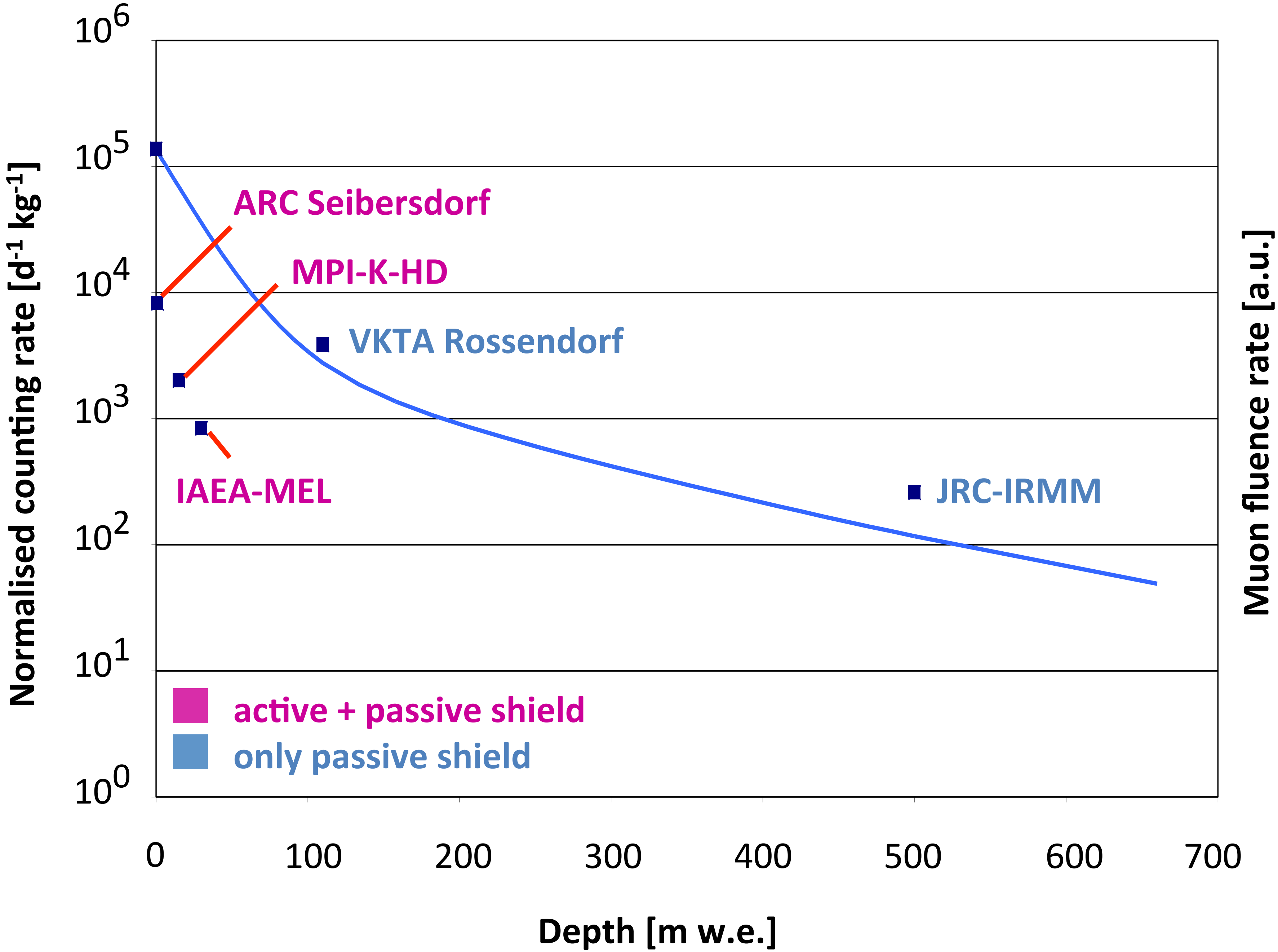
HPGe detectors

detector	total and peak background count rate [d^{-1} kg^{-1}_{Ge}]			
	40-2700 keV	352 keV	583 keV	1461 keV
GeBer	3686	3.3	1.5	4.6
GeMi	611	5.6	2.1	5.2
GePV	482	2.8	2.1	3.2
GsOr	469	2.4	0.76	4.3
GePaolo	226	0.83	0.38	1.4
GeCris	87	<0.39	<0.29	1.0
GeMPI	30	<0.20	<0.15	0.36

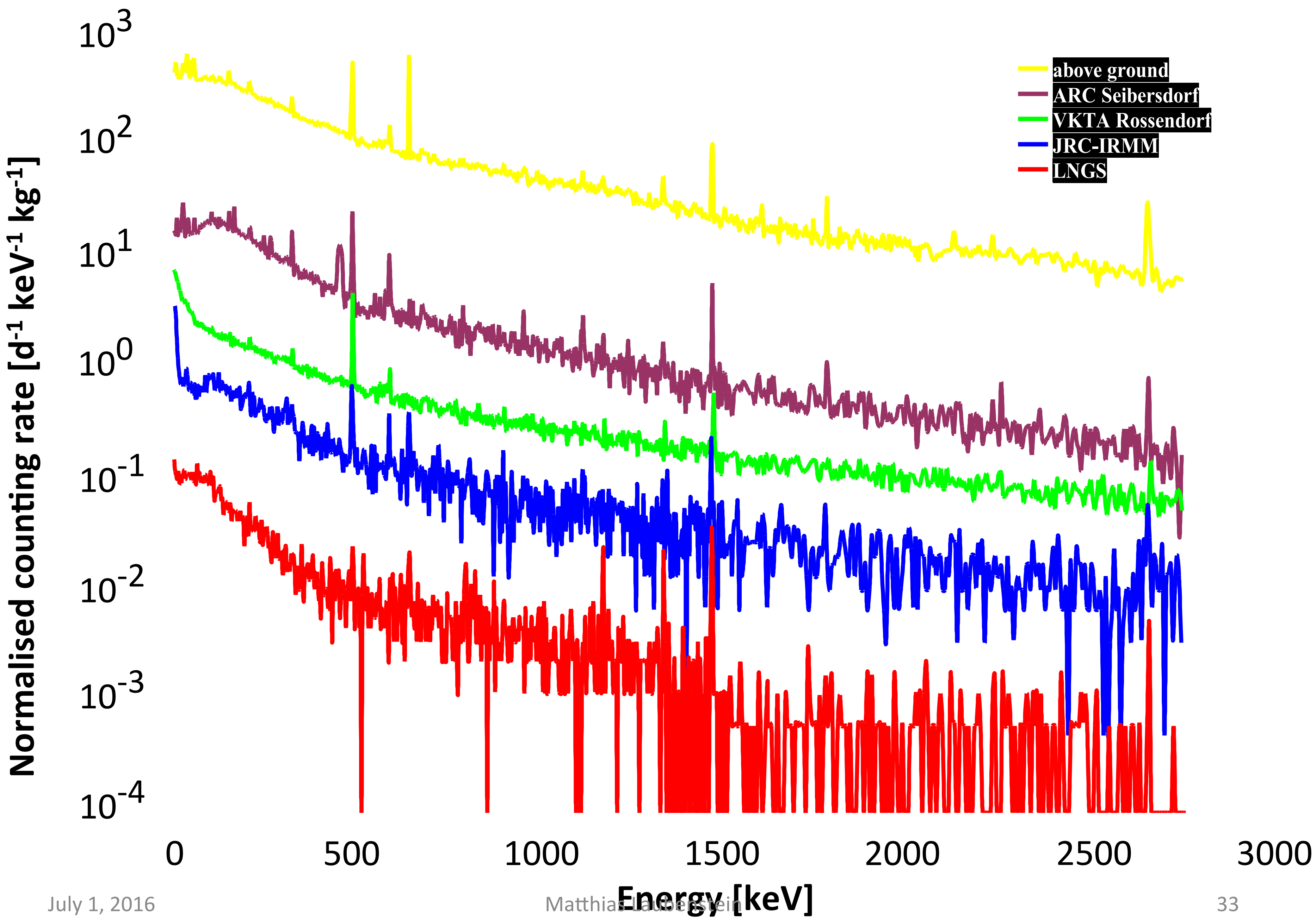
CELLAR

Collaboration of European Low-level underground LAboRatories

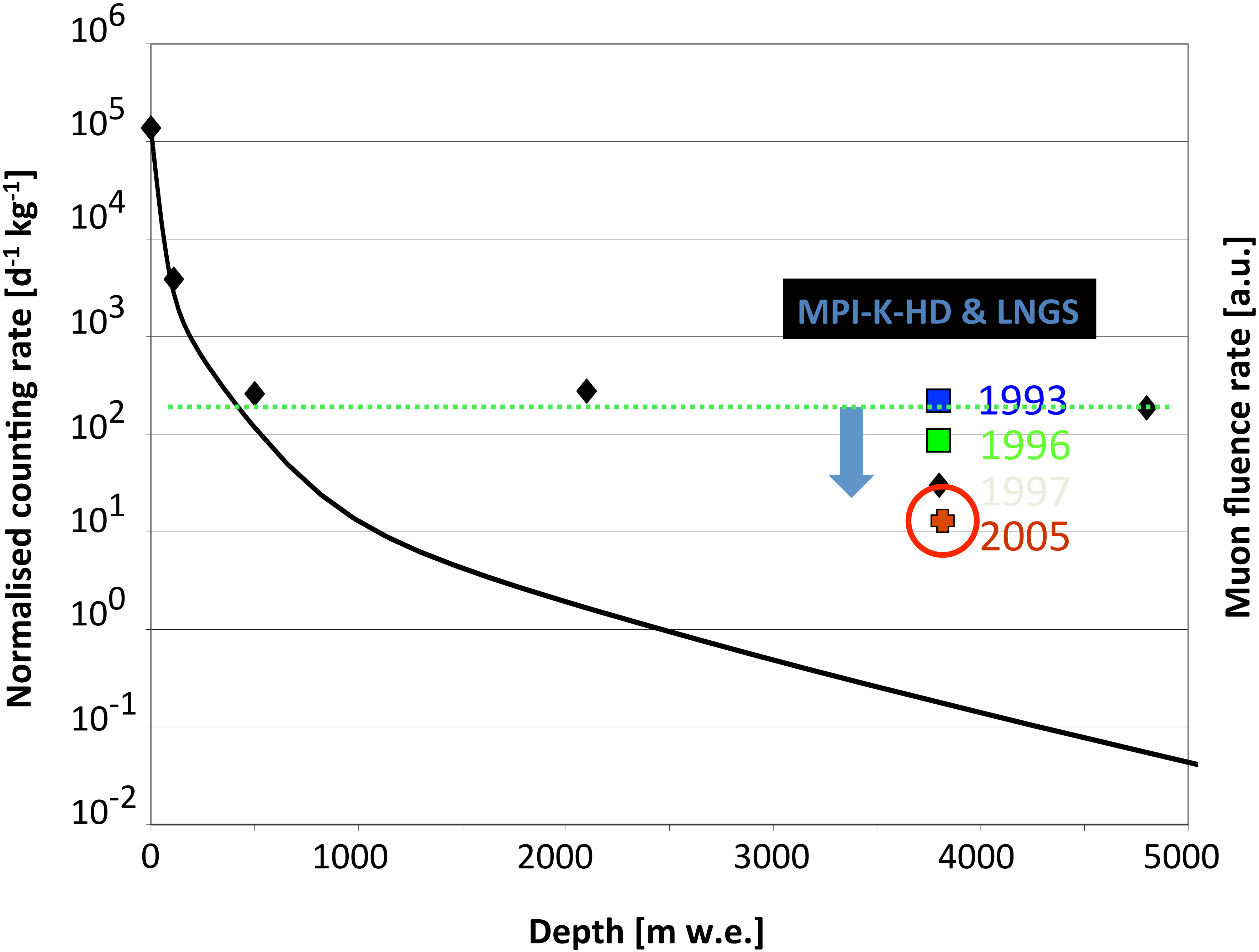

LSCE
IRMM
PTB
Physikalisch-Technische Bundesanstalt
IRSN
IRSN
INSTITUT DE RADIOPROTECTION ET DE SÛRETÉ NUCLÉAIRE
VKTA
Dresden
VKTA
LSC
LSC
Laboratorio Subterráneo de Canfranc
MPI-K
Max-Planck-Institut für Kernphysik
LEGOS
LEGOS
IFIN-HH
IAEA-MEL
IFIN-HH
LSM
LNGS
LNGS
lsm
Boulby
STFC
Boulby Underground Laboratory



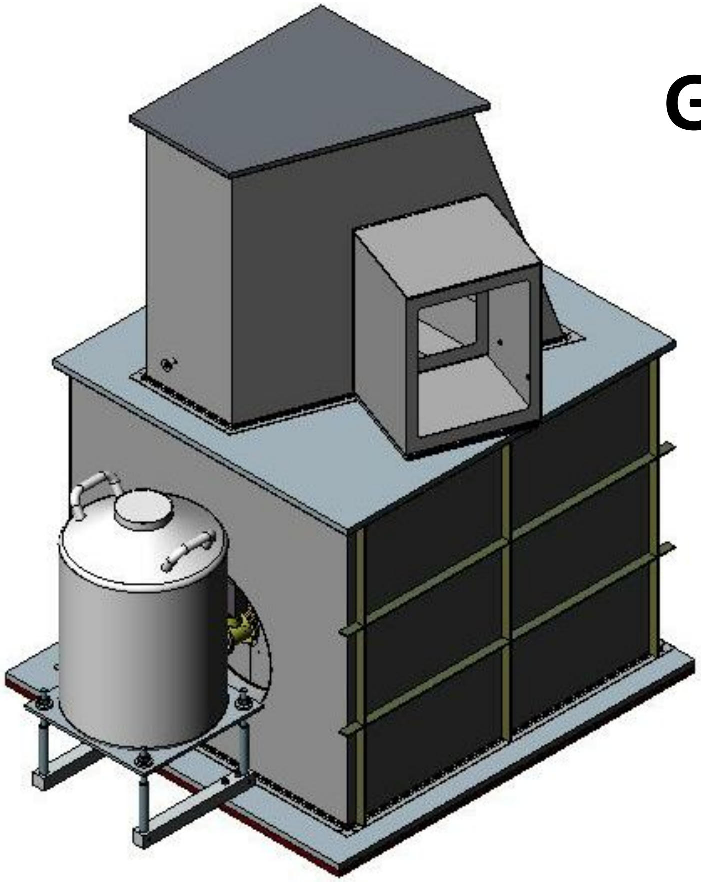
Normalised counting rate [d-1 kg-1]
Muon fluence rate [a.u.]
10^6
10^5
10^4
10^3
10^2
10^1
10^0
ARC Seibersdorf
MPI-K-HD
VKTA Rossendorf
IAEA-MEL
JRC-IRMM
active + passive shield
only passive shield
0
100
200
300
400
500
600
700
Depth [m w.e.]



above ground
ARC Seibersdorf
VKTA Rossendorf
JRC-IRMM
LNGS
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]



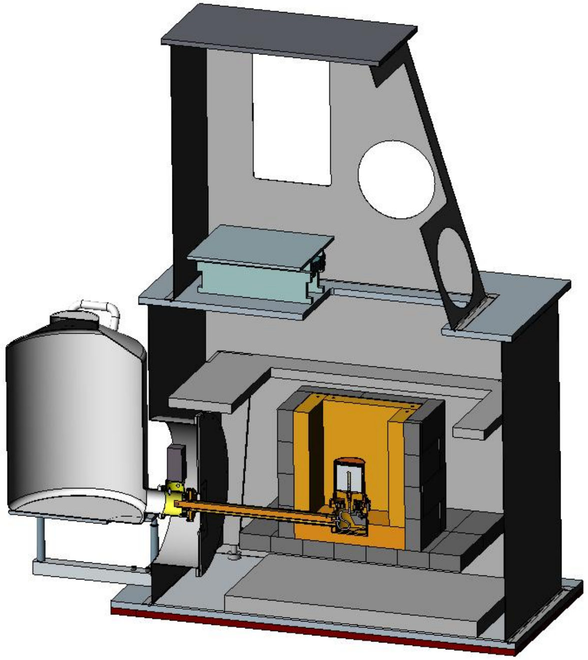
MPI-K-HD & LNGS
1993
1996
1997
2005
Normalised counting rate [d-1 kg-1]
Muon fluence rate [a.u.]
Depth [m w.e.]
10^6
10^5
10^4
10^3
10^2
10^1
10^0
10^-1
10^-2
0
1000
2000
3000
4000
5000

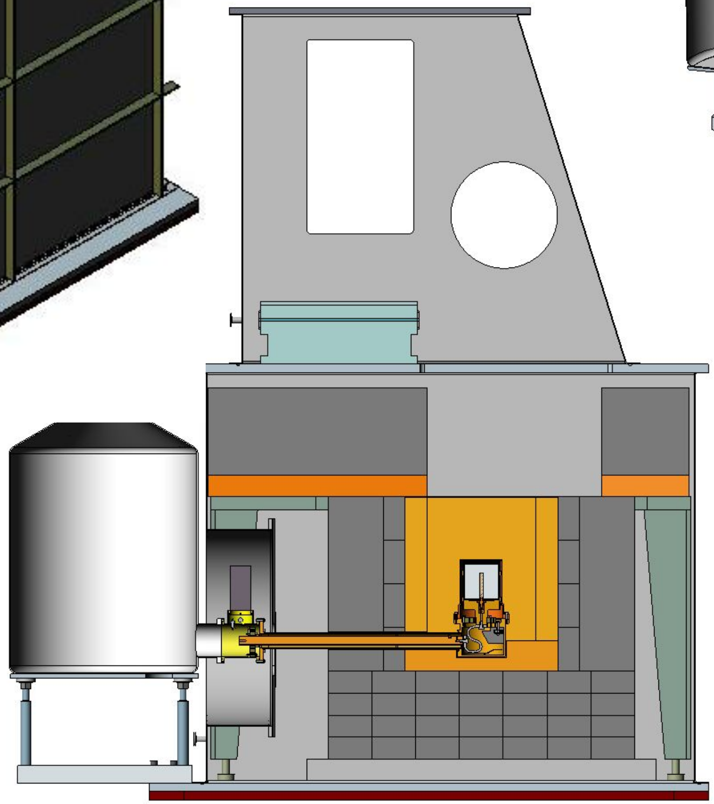


GeMPI type

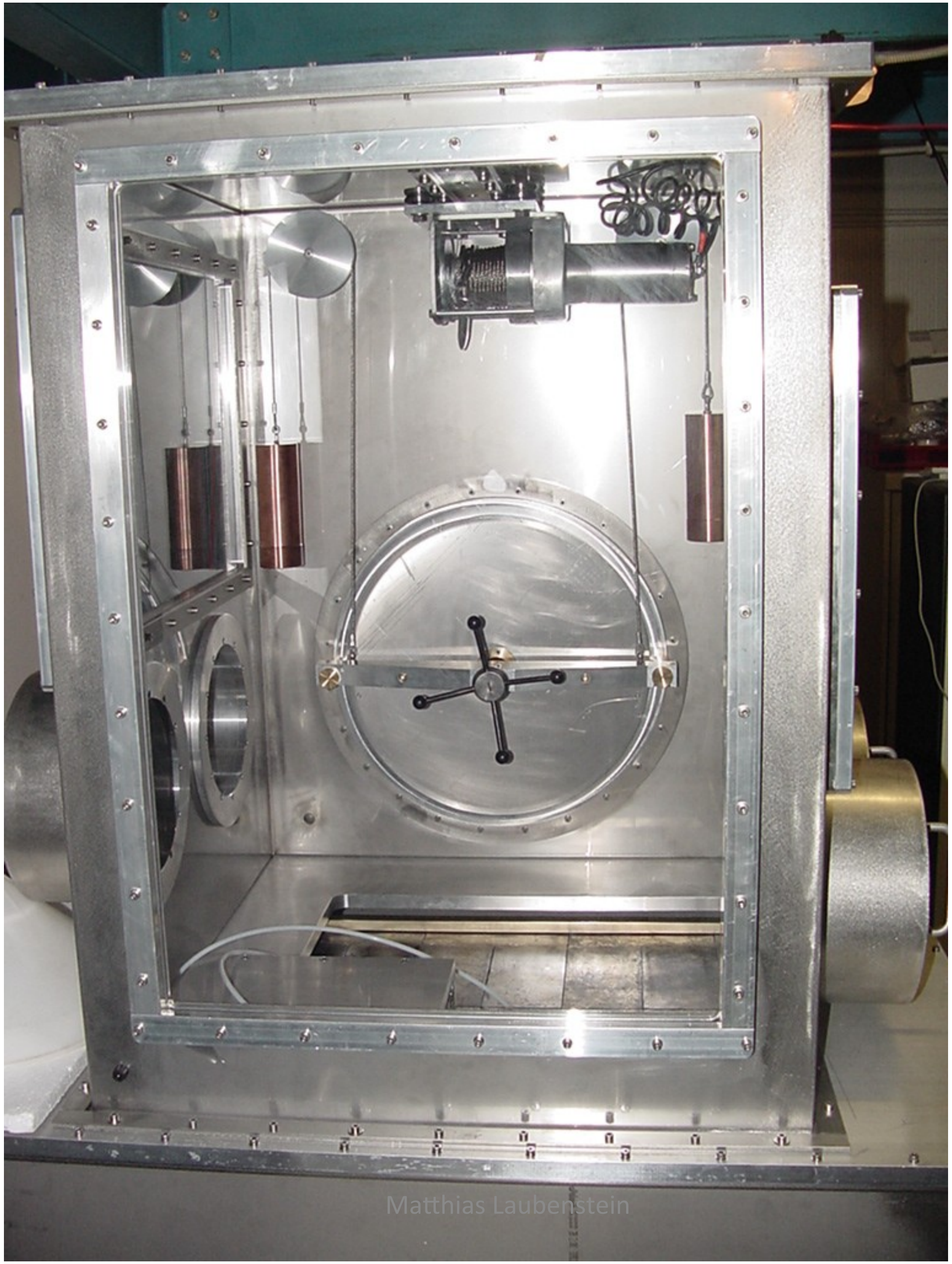
MPI-K detectors
operated at

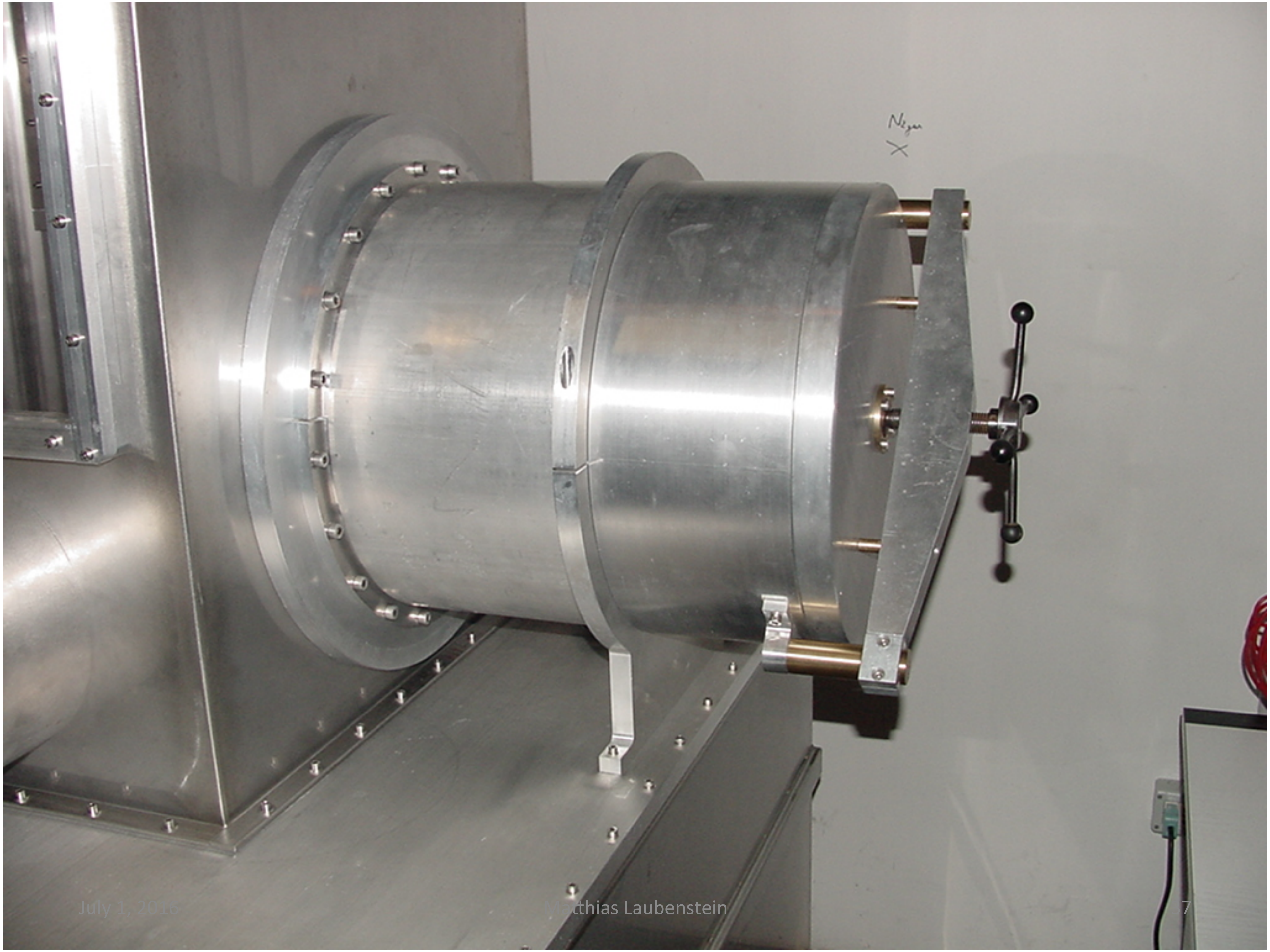
LNGS (3800 m w.e.)





G. Heusser
B. Prokosch
H. Neder
M. Laubenstein





GeMPI

material	activity [µBq/kg]				LN₂ shielding thickness against 2.6 MeV for E-4**
	^{226}Ra (U)	^{228}Th (Th)	^{40}K	various	intrinsic contamin. / (external)
lead (DowRun)	≤ 29	≤ 22	440	98 ^{207}Bi; 180 ^{60}Co 2.7x10^{7} ^{210}Pb	215 cm / (41.0 cm)
lead (Boliden)	≤ 27	≤ 26	460	≤ 13 ^{207}Bi; ≤ 11 ^{60}Co 2.3x10^{7} ^{210}Pb	
copper (Lens)	≤ 16	≤ 12	≤ 110	≤ 10 ^{60}Co	221 cm / (58.7 cm)
steel (foil)	600	200	1800	17000 ^{60}Co	290 cm / (66.3 cm)
steel (Lens)	1200	7100	≤ 3000	300 ^{60}Co	
water*	≤ 1	0.04 - 0.008	≤ 2		12cm / (464 cm)
liq. nitrogen	≤ 0.3 (^{222}Rn)			0.006 ^{39}Ar; 0.05 ^{85}Kr	(637 cm)
liq. argon	120 (^{222}Rn)			1.1x10^{6} ^{39}Ar; 30 ^{42}Ar	(396 cm)
concrete	8x10^{6}	1x10^{7}	9x10^{7}		

* BOREXINO

**backgroundindex at 2.039 MeV [counts/kg y keV]

Background (peak) count rates [c/kg y]

Energy [keV]	**GeMPI**	**HDM # 1-5**
352 (U/Ra)	**≤ 31**	**110 - 180**
609 (U/Ra)	**≤ 30**	**96 - 140**
583 (Th)	**≤ 23**	**18 - 42**
2615 (Th)	**17 ± 5**	**11 - 22**
1461 (K)	**90 ± 13**	**74 - 290**
100-2730 keV	**9760**	**12300**

Contamination of Cu [µBq/kg]

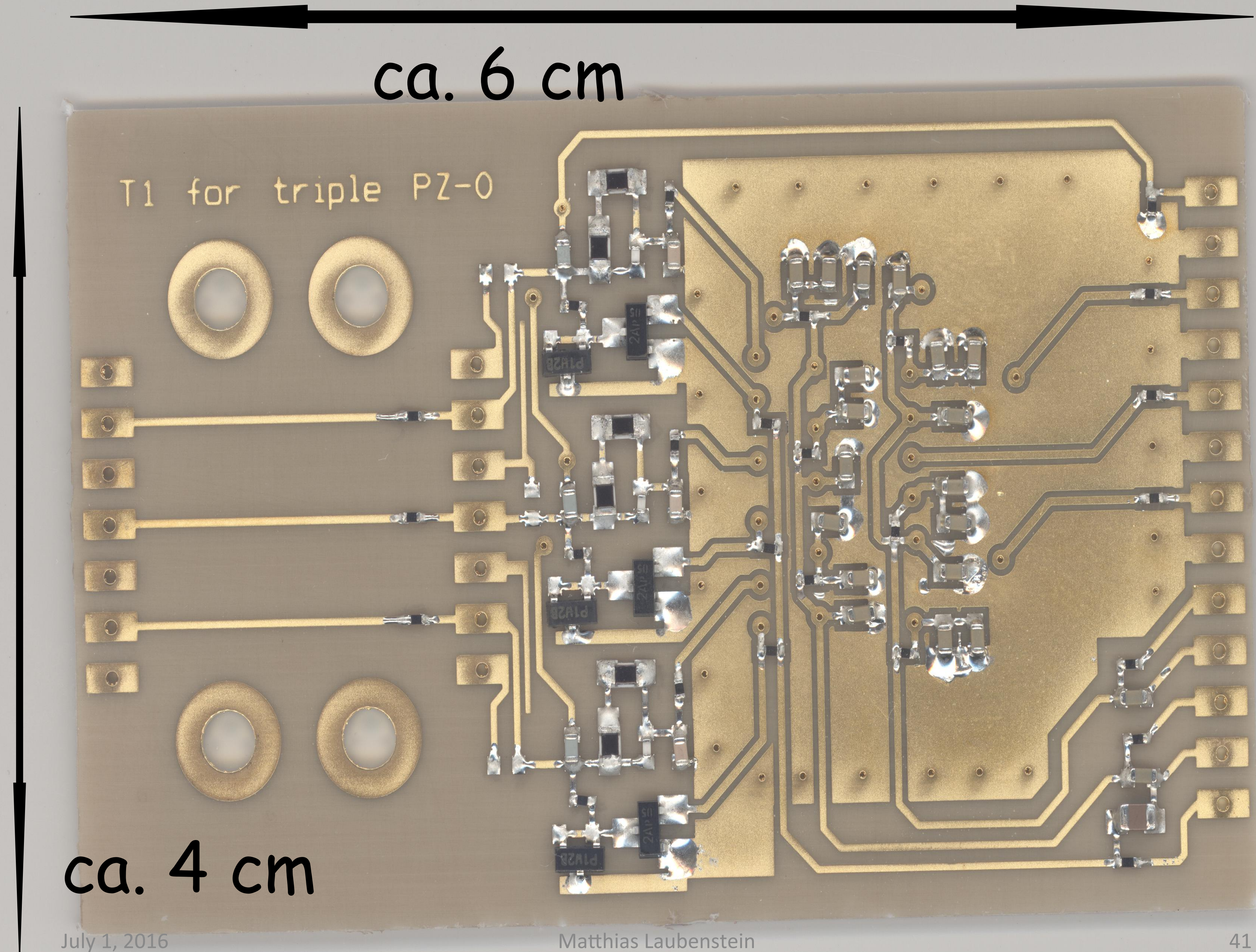
	^{226}Ra (U)	^{228}Th (Th)	^{40}K
Cryostat of ANG1	168 ± 8	84 ± 7	236 ± 61
Cryostat of ANG2	91 ± 4	10 ± 3	78 ± 22
Cryostat of ANG3	105 ± 5	84 ± 5	927 ± 46
Cryostat of ANG4	115 ± 3	87 ± 4	199 ± 4
Cryostat of ANG5	100 ± 4	26 ± 4	1632 ± 49
measured by **GeMPI***	≤ 16	≤ 12	≤ 110

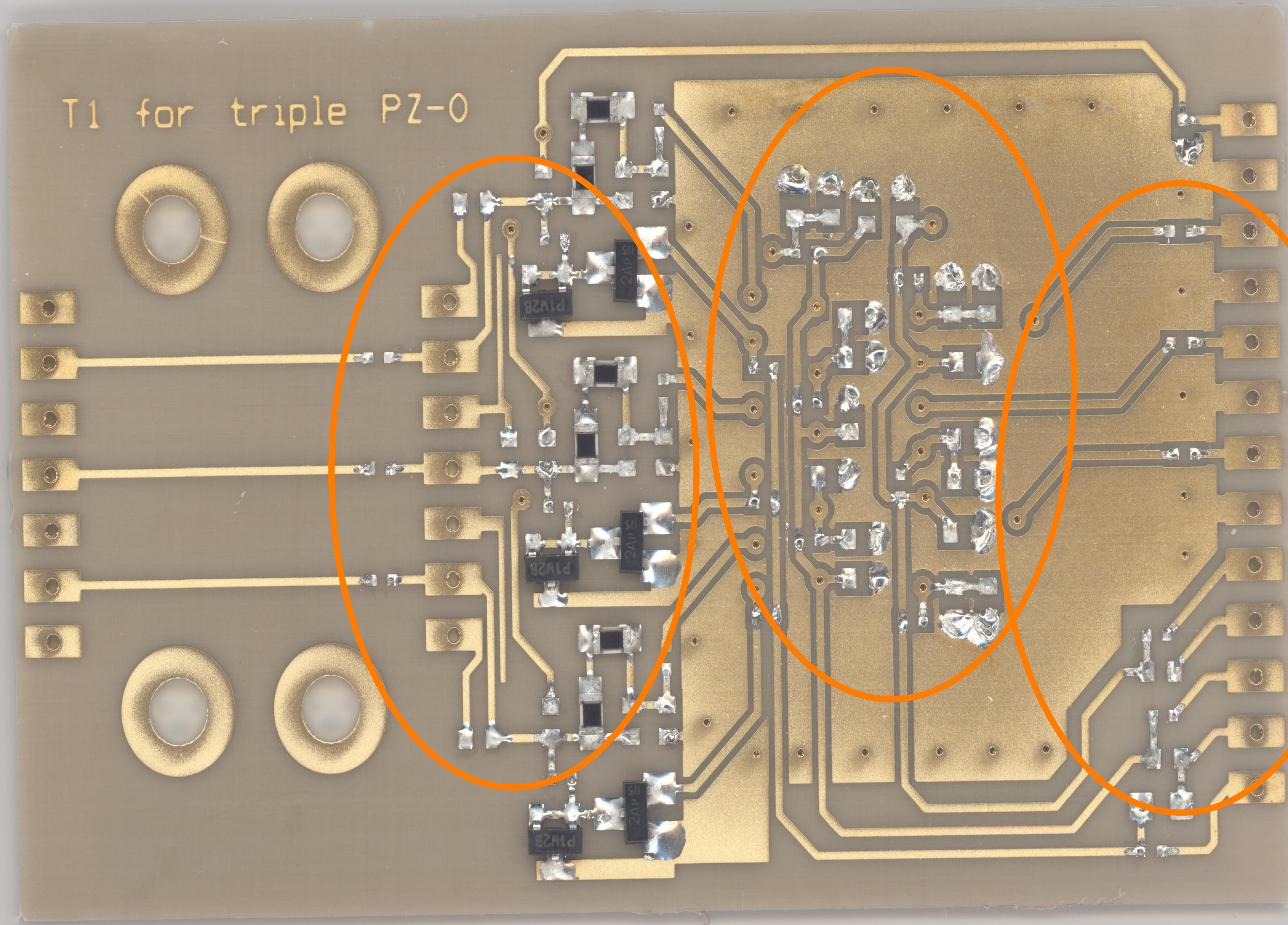
Monte Carlo simul. Ch. Doerr,Uni HD 2002

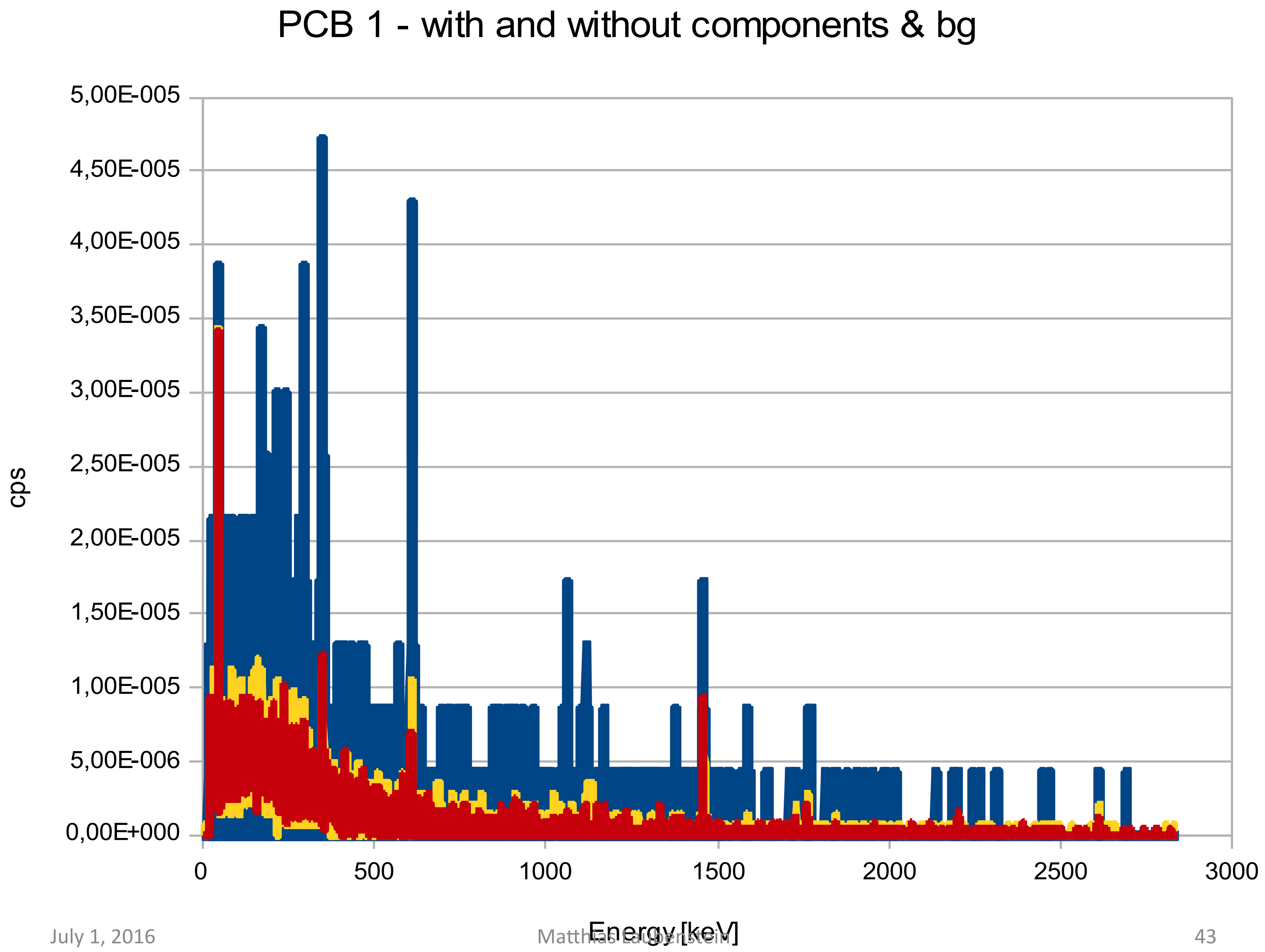
* 127 kg

→ surface contamination


GeDSG


ca. 6 cm
T1 for triple PZ-0
ca. 4 cm


T1 for triple PZ-0


PCB 1 - with and without components & bg
cps
5,00E-005
4,50E-005
4,00E-005
3,50E-005
3,00E-005
2,50E-005
2,00E-005
1,50E-005
1,00E-005
5,00E-006
0,00E+000
0
500
1000
1500
2000
2500
3000
Energy [keV]