



Experience of gas purification and radon control in BOREXINO

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on behalf of the Borexino collaboration

Outline



- BOREXINO
- ^{222}Rn control
- Purification of nitrogen
- Summary

BOREXINO at LNGS

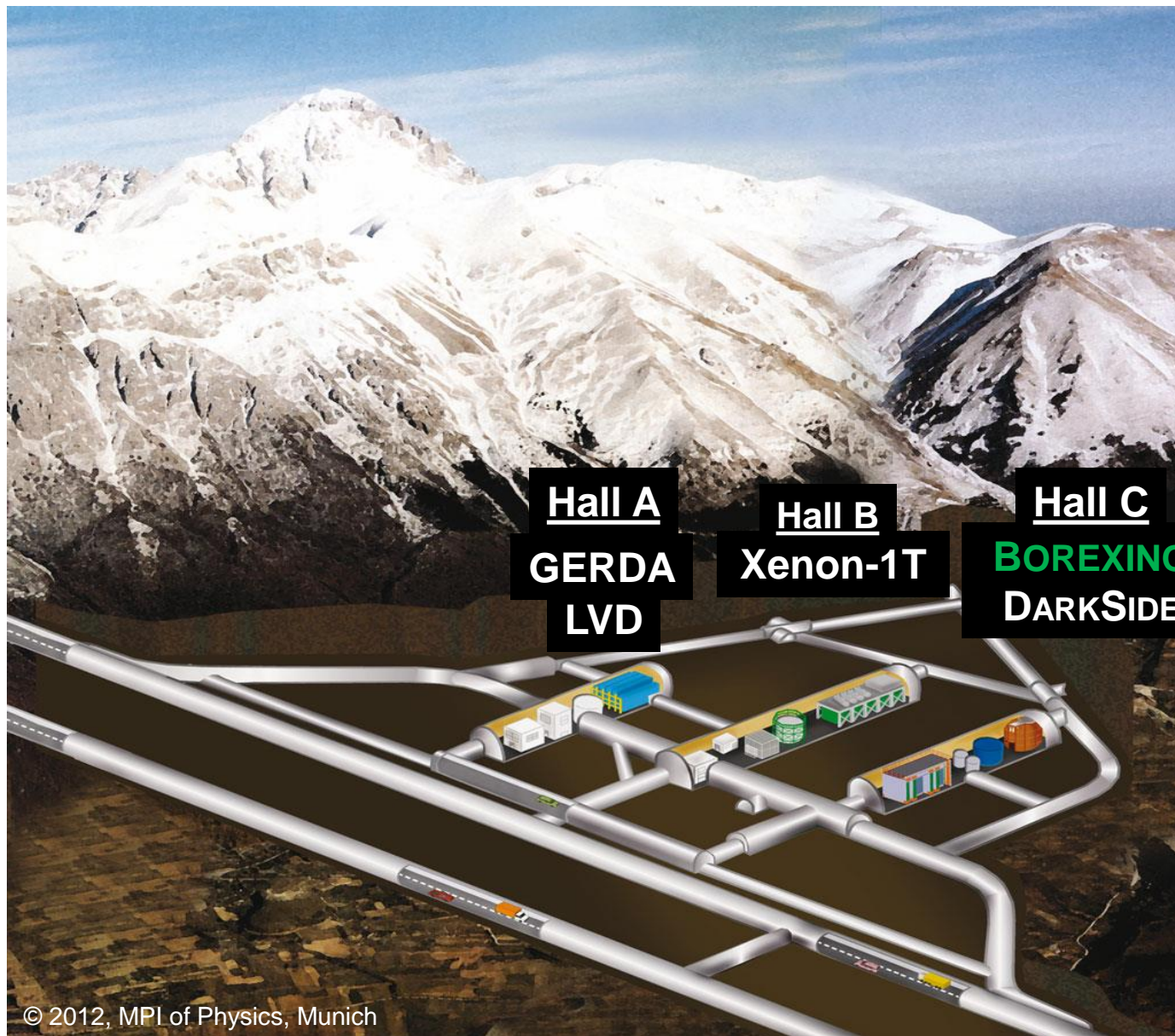


BOREXINO

^{222}Rn control

N_2 purification

Summary



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Low Radioactivity Techniques 2017, 23-27 May 2017, Seoul, Korea

BOREXINO design

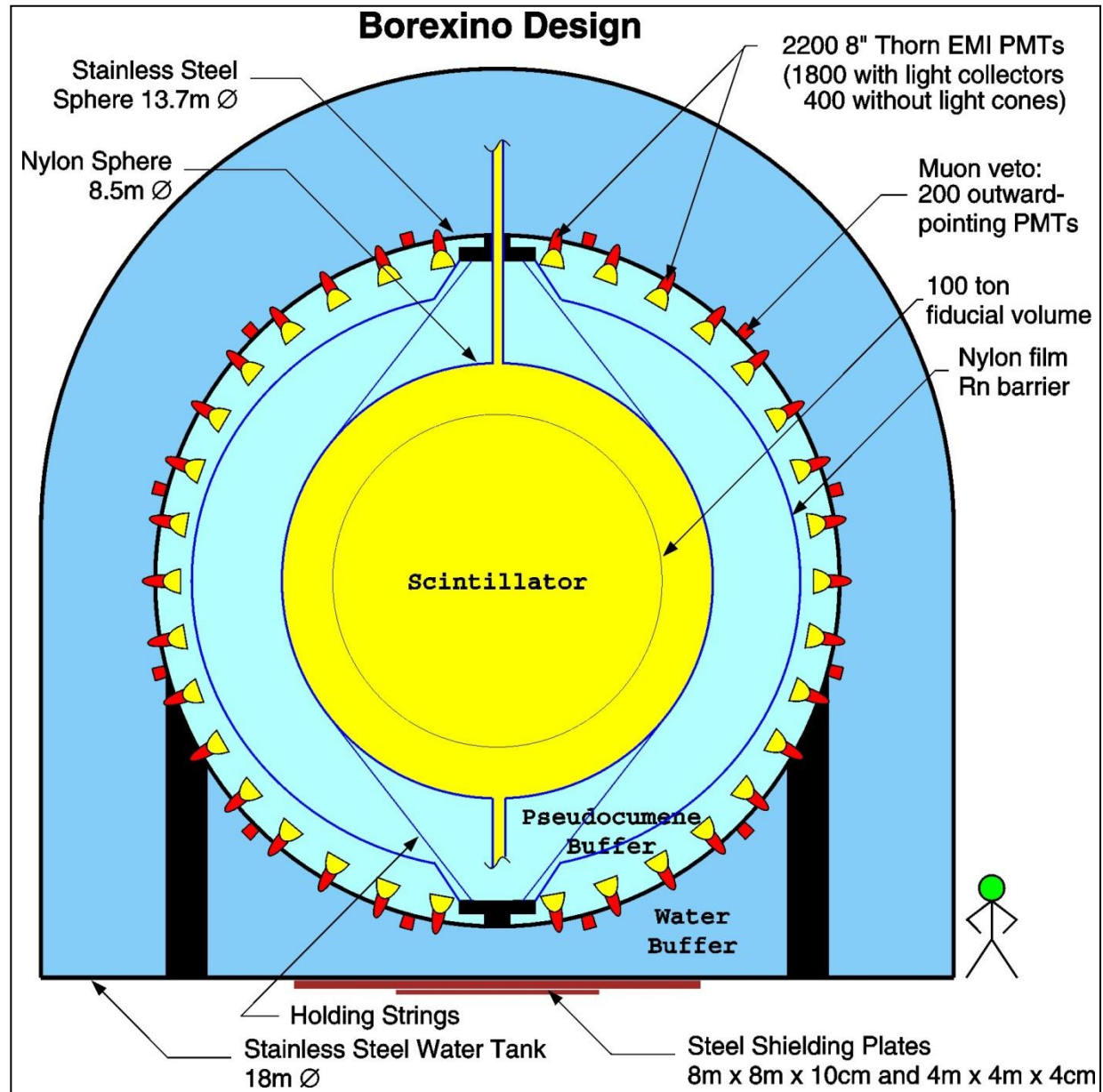


BOREXINO

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Low Radioactivity Techniques 2017, 23-27 May 2017, Seoul, Korea

BOREXINO radio-purity



In a nutshell: the radio-purest detector ever built

BOREXINO

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Summary

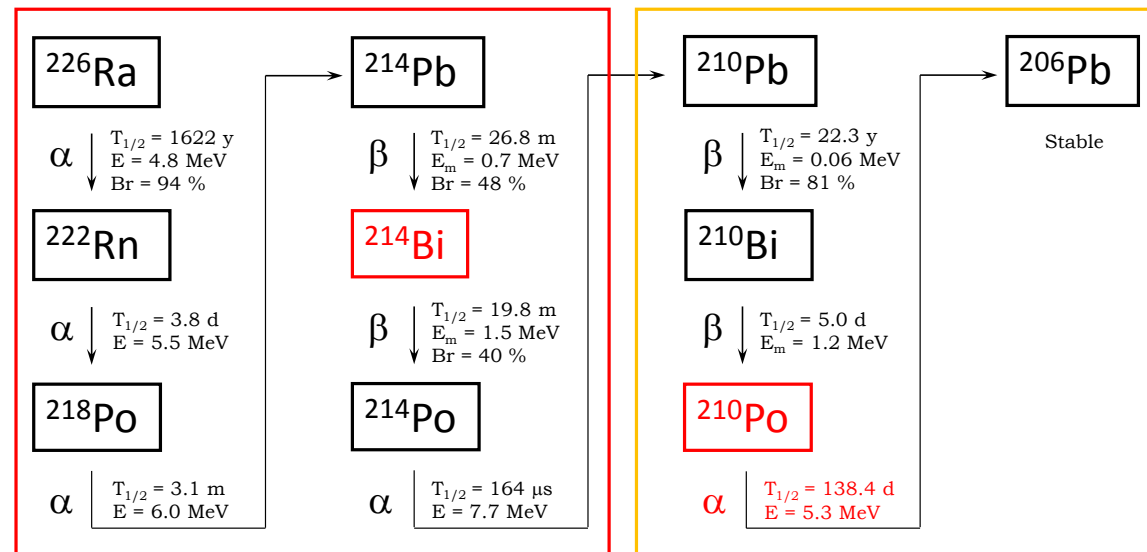
Isotope	Specification for LS	Before purification	After purification
^{238}U	$\leq 10^{-16} \text{ g/g}$	$(5.3 \pm 0.5) \cdot 10^{-18} \text{ g/g}$	$< 0.8 \cdot 10^{-19} \text{ g/g}$
^{232}Th	$\leq 10^{-16} \text{ g/g}$	$(3.8 \pm 0.8) \cdot 10^{-18} \text{ g/g}$	$< 1.0 \cdot 10^{-18} \text{ g/g}$
$^{14}\text{C}/^{12}\text{C}$	$\leq 10^{-18}$	$(2.69 \pm 0.06) \cdot 10^{-18} \text{ g/g}$	unchanged
^{40}K	$\leq 10^{-18} \text{ g/g}$	$\leq 0.4 \cdot 10^{-18} \text{ g/g}$	unchanged
^{222}Rn	$\leq 1 \text{ cpd/100 t}$	see ^{238}U	see ^{238}U
^{85}Kr	$\leq 1 \text{ cpd/100 t}$	$(30 \pm 5) \text{ cpd/100 t}$	$\leq 5 \text{ cpd/100 t}$
^{39}Ar	$\leq 1 \text{ cpd/100 t}$	$\ll ^{85}\text{Kr}$	$\ll ^{85}\text{Kr}$
^{210}Po	not specified	$\sim (70) 1 \text{ dpd/100 t}$	unchanged
^{210}Bi	not specified	$(20) 70 \text{ dpd/100 t}$	$(20 \pm 5) \text{ cpd/100 t}$

Sources of ^{222}Rn in BOREXINO



- All external systems wetted by LS and nitrogen (LS storage tanks, LS purification systems: water extraction column, stripping column, nitrogen lines, *etc.*)
- Emanation from the Stainless Steel Sphere and from the PMTs
- Emanation from the nylon scintillator vessel
- Nitrogen

Decays of ^{222}Rn and its short-lived daughters can directly contribute to the detector's background, but even more important is the accumulation of the long-lived ^{210}Pb (followed by ^{210}Bi and ^{210}Po) in the scintillator.



Control of ^{222}Rn in BOREXINO



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^{222}Rn control

N_2 purification

Summary

- Check of all subsystems for ^{222}Rn emanation (extensive screening campaign)
- Minimization of ^{222}Rn diffusion through the Inner Vessel (identification of material with low diffusion coefficient) + installation of the Rn barrier
- Identification of the IV material with sufficiently low ^{226}Ra content ($12 \mu\text{Bq/kg}$)
- Nitrogen purification

Detection of ^{222}Rn – counters

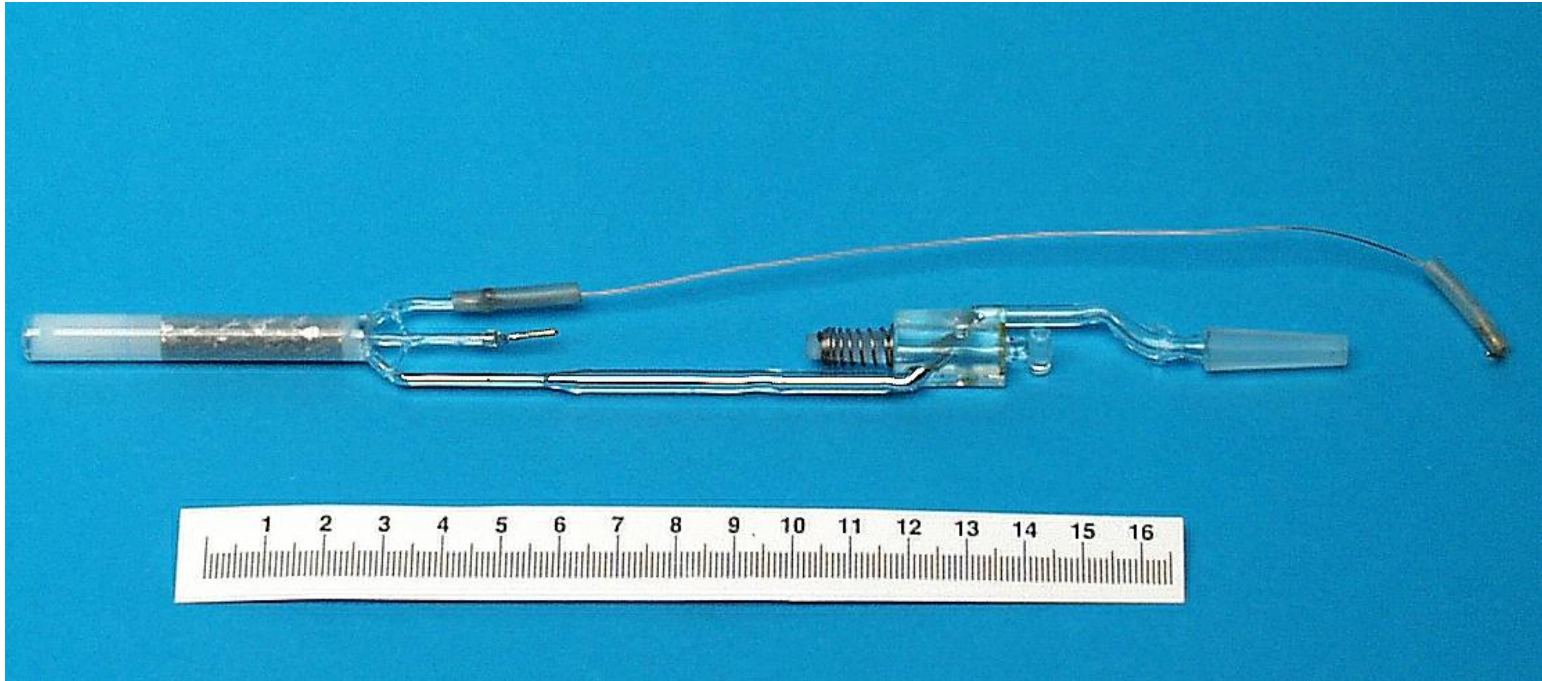


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^{222}Rn control

N_2 purification

Summary



- Developed for the GALLEX/GNO experiment
- Hand-made at MPI-K ($\sim 1 \text{ cm}^3$ active volume)
- In case of ^{222}Rn only α -decays are detected
- 50 keV threshold
 - bkg: 0.1 – 2 cpd
 - total detection efficiency of ~ 1.5
- **Absolute detection limit $\sim 30 \text{ } \mu\text{Bq}$ (15 atoms)**

^{222}Rn emanation (MPI-K)



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^{222}Rn control

N_2 purification

Summary



Blanks:

20 l \rightarrow 50 μBq

80 l \rightarrow 80 μBq

Absolute sensitivity
 $\sim 100 \mu\text{Bq}$ [50 atoms]

Appl. Rad. Isot. 53 (2000) 371

^{222}Rn emanation: examples



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^{222}Rn control

N_2 purification

Summary

System	Sample	Description	^{222}Rn em. rate
LS storage area	SS vessel TK1	114 m ³	< 60 mBq
	SS vessel TK2	114 m ³	(45 ± 8) mBq
	SS vessel TK3	114 m ³	(24 ± 5) mBq
N_2 distribution line	Electrical heater		(0.92 ± 0.29) mBq
	Particle Filter		(0.34 ± 0.13) mBq
	1.5" distrib. line	~ 100 m long	(0.47 ± 0.13) mBq
LS purification plant	SS package	25 m ²	< 0.12 mBq
	H ₂ O extraction column + 24 SS packages	0.6 m ³ / 608 m ²	(4.83 ± 0.70) mBq
	N_2 sparging column + 26 SS packages	0.2 m ³ / 280 m ²	(1.78 ± 0.21) mBq

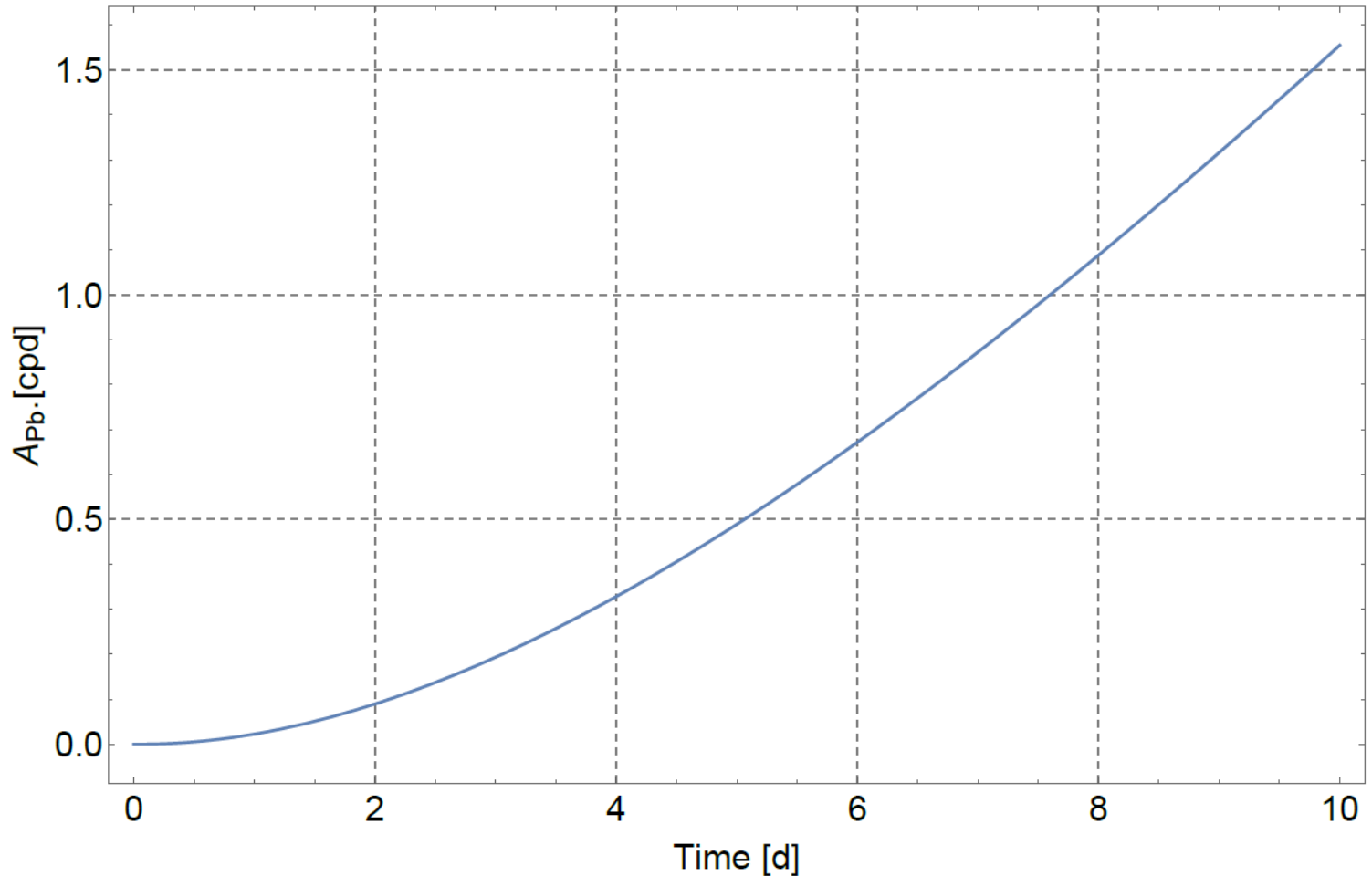
Over 1000 entries in the DB!

Astroparticle Physics 18 (2002) 1
 LRT 2004 proceedings, p. 141 – 149
 Int. J. Mod. Phys. A29 (2014) 1442009

^{222}Rn emanation: examples



Expected ^{210}Pb activity in 100 ton of the BOREXINO scintillator as a function of the storage time in a tank with the ^{222}Rn emanation rate of 40 mBq. 1 cpd of ^{210}Pb is reached already after 7.5 d.



BOREXINO

^{222}Rn control

N_2 purification

Summary

BOREXINO design

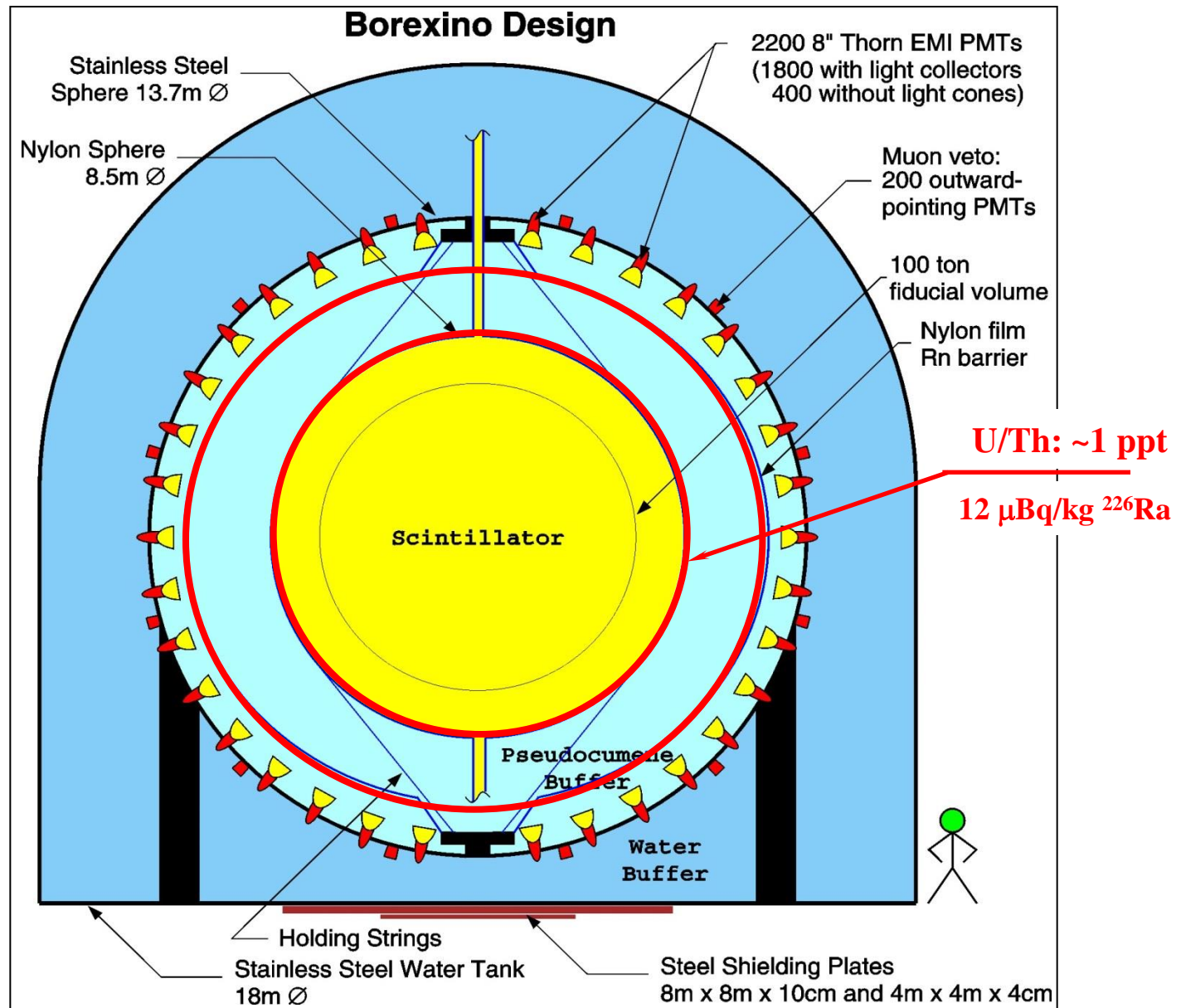


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^{222}Rn control

N_2 purification

Summary



^{222}Rn diffusion

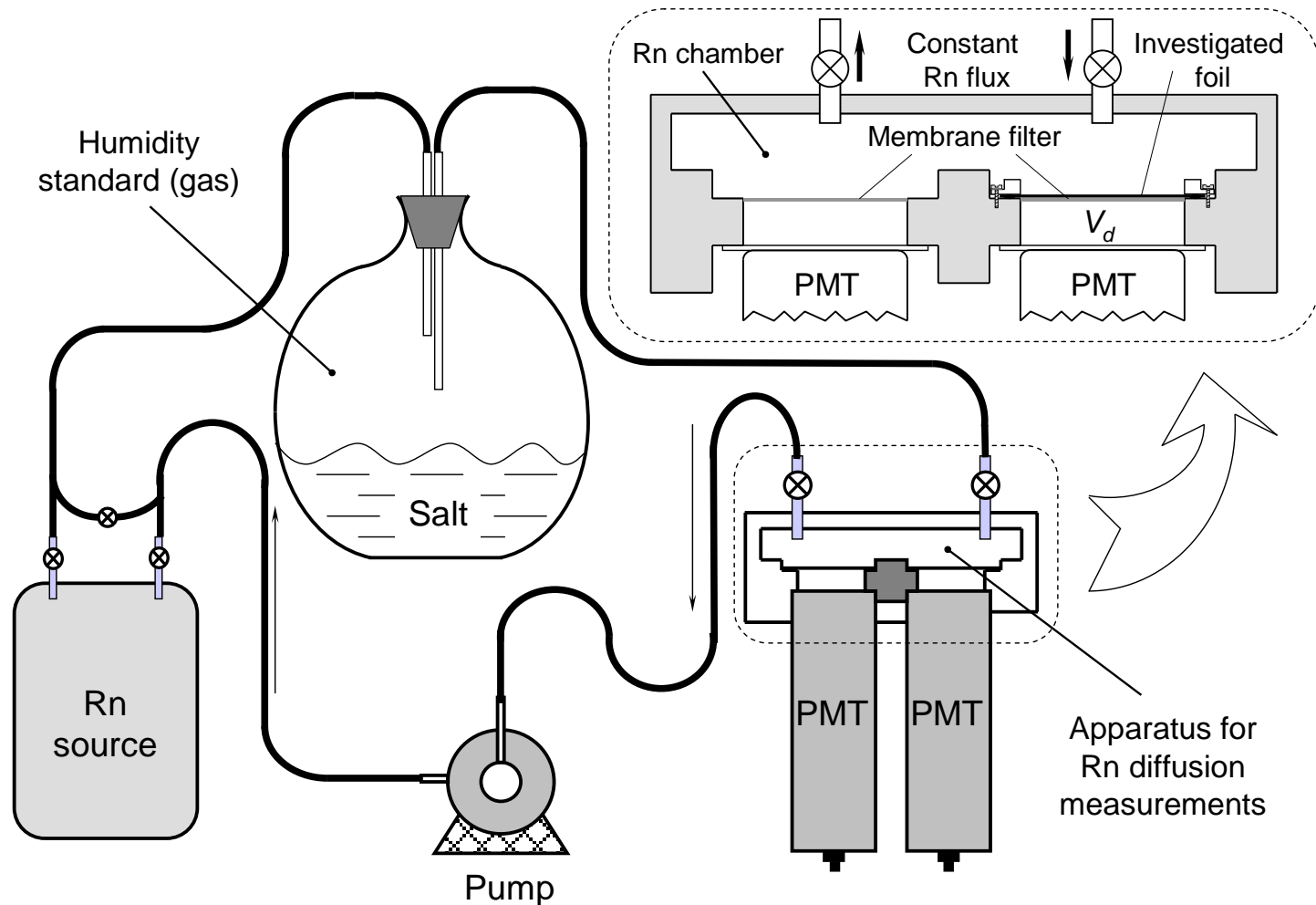


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^{222}Rn control

N_2 purification

Summary



Sensitivity: $D \sim 10^{-13} \text{ cm}^2/\text{s}$
 $d_e \sim 2 \mu\text{m}$

^{222}Rn diffusion



Results obtained for the 0.018 mm thick C38F film (BOREXINO)

RH standard salt	RH in gas phase (%)	Water amount in nylon, M (%)	Diffusion coefficient, D (cm^2/s)	Solubility, S
$\text{Mg}(\text{ClO}_4)_2$	~ 0	~ 0	$(2.1 \pm 0.4) \times 10^{-12}$	4.5 ± 0.7
$\text{H}_3\text{PO}_4 \cdot \frac{1}{2}\text{H}_2\text{O}$	9 ± 1	0.72 ± 0.04	$(2.3 \pm 0.3) \times 10^{-12}$	2.5 ± 0.3
$\text{LiCl}_2 \cdot \text{H}_2\text{O}$	12 ± 1	0.87 ± 0.04	$(2.2 \pm 0.3) \times 10^{-12}$	2.2 ± 0.3
$\text{CaCl}_2 \cdot 6\text{H}_2\text{O}$	32 ± 2	2.09 ± 0.04	$(4.3 \pm 0.5) \times 10^{-12}$	1.8 ± 0.2
$\text{Na}_2\text{Cr}_2\text{O}_7 \cdot 2\text{H}_2\text{O}$	52 ± 2	3.74 ± 0.05	$(1.9 \pm 0.3) \times 10^{-11}$	1.4 ± 0.2
$\text{Na}_2\text{S}_2\text{O}_3 \cdot 5\text{H}_2\text{O}$	76 ± 2	6.35 ± 0.05	$(6.5 \pm 0.9) \times 10^{-11}$	1.5 ± 0.2
K_2CrO_4	88 ± 3	7.60 ± 0.05	$(1.3 \pm 0.2) \times 10^{-10}$	1.5 ± 0.2
$\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$	93 ± 3	9.12 ± 0.07	$(3.3 \pm 0.4) \times 10^{-10}$	1.0 ± 0.1
H_2O vapors	100 ± 3	10.14 ± 0.09	$(1.3 \pm 0.2) \times 10^{-9}$	0.7 ± 0.1

There is 3 orders of magnitude difference between the diffusion in the dry and in the foil saturated with water!

Nucl. Instr. Meth. A 449 (2000) 158
Nucl. Instr. Meth. A 524 (2004) 355

$$d_e = \sqrt{\frac{D}{\lambda}} \quad \begin{array}{l} d_e^d = 7 \mu\text{m} \\ d_e^w = 270 \mu\text{m} \end{array}$$

^{226}Ra in/on BOREXINO nylon



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^{222}Rn control

N_2 purification

Summary



1 ppt U required
($\sim 12 \mu\text{Bq/kg}$ for ^{226}Ra)

$$D_{\text{dry}} = 2 \times 10^{-12} \text{ cm}^2/\text{s} \quad (d_{\text{dry}} = 7 \mu\text{m})$$
$$D_{\text{wet}} = 1 \times 10^{-9} \text{ cm}^2/\text{s} \quad (d_{\text{wet}} = 270 \mu\text{m})$$

$$A_{\text{dry}} = A_{\text{sf}} + 0.14 \cdot A_{\text{bulk}}$$
$$A_{\text{wet}} = A_{\text{sf}} + A_{\text{bulk}}$$

Separation of the bulk
and surface ^{226}Ra conc.
was possible through
 ^{222}Rn emanation

Very sensitive technique:
($C_{\text{Ra}} \sim 10 \mu\text{Bq/kg}$)

Bx IV foil: bulk $\leq 15 \mu\text{Bq/kg}$
surface $\leq 0.8 \mu\text{Bq/m}^2$
total = $(16 \pm 4) \mu\text{Bq/kg}$ (1.2 ppt U equiv.)

NIM A 498 (2003) 240

Construction of nylon vessels



BOREXINO

^{222}Rn control

N_2 purification

Summary



Princeton clean room class 100 with ^{222}Rn -
reduced air (VSA filter): $C_{\text{Rn}} \sim 1 \text{ Bq/m}^3$

A. Pocar, PhD Thesis (2003)

Inflation of vessels in SSS

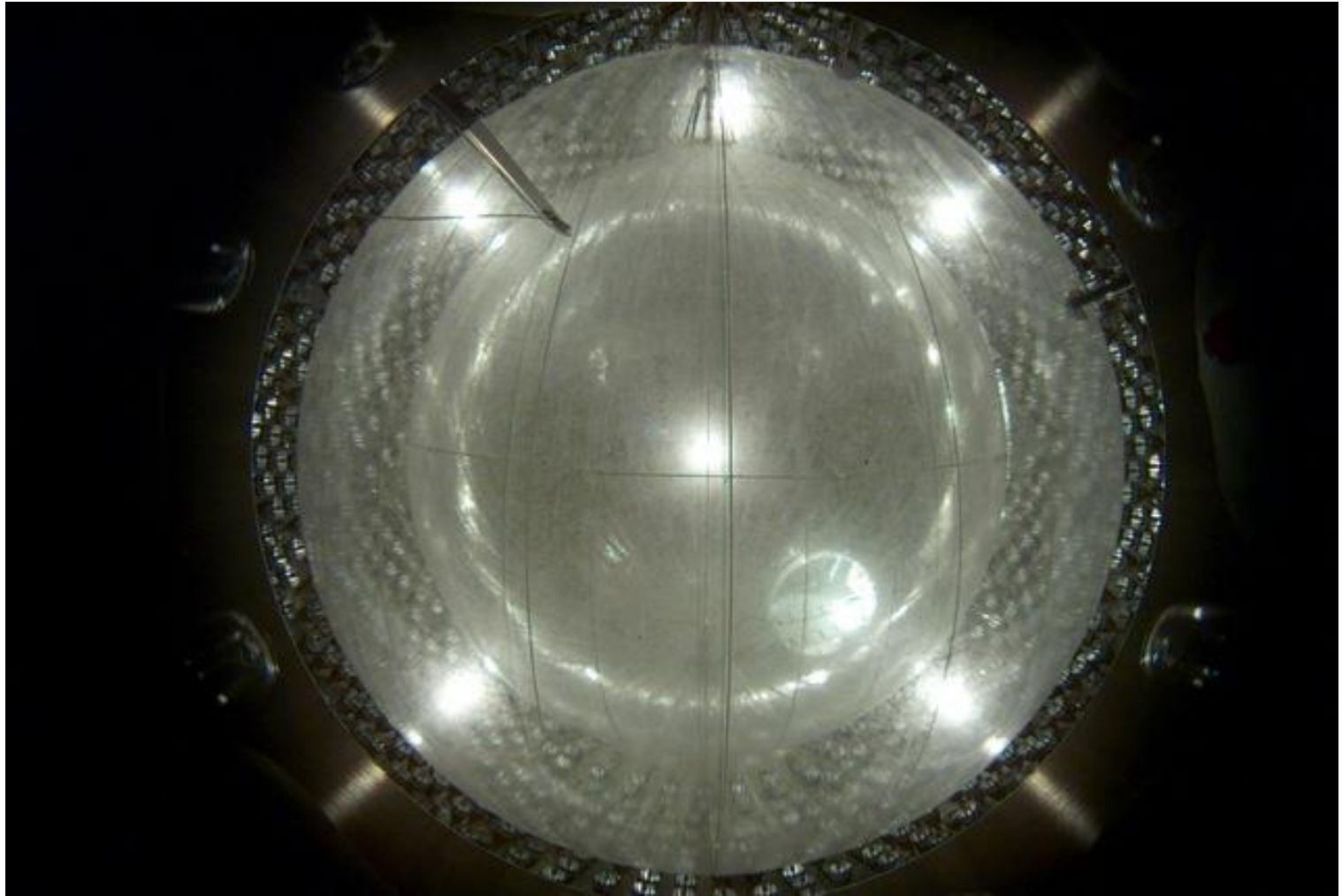


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^{222}Rn control

N_2 purification

Summary



The nylon vessels were inflated in the sphere
with synthetic air: $C_{\text{Rn}} < 0.1 \text{ mBq/m}^3$

Int. J. Mod. Phys. A29 (2014) 1442009

^{222}Rn in gases



- ^{222}Rn adsorption on activated carbon
- Several AC traps available (MoREx, MPIK-HD)
- Pre-concentration from 100 – 200 m³ →

^{222}Rn detection limit:
~0.5 $\mu\text{Bq}/\text{m}^3$ (STP)
[1 atom in 4 m³]



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^{222}Rn control

N₂ purification

Summary

Appl. Rad. Isot. 52 (2000) 691

^{222}Rn removal from LN_2



BX N_2 purification plant: LTA – Low Temperature Adsorber



Production rate: $100 \text{ m}^3/\text{h}$
 $^{222}\text{Rn} \leq 0.5 \text{ } \mu\text{Bq}/\text{m}^3 \text{ (STP)}$

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^{222}Rn control

N_2 purification

Summary

Ar and Kr in nitrogen

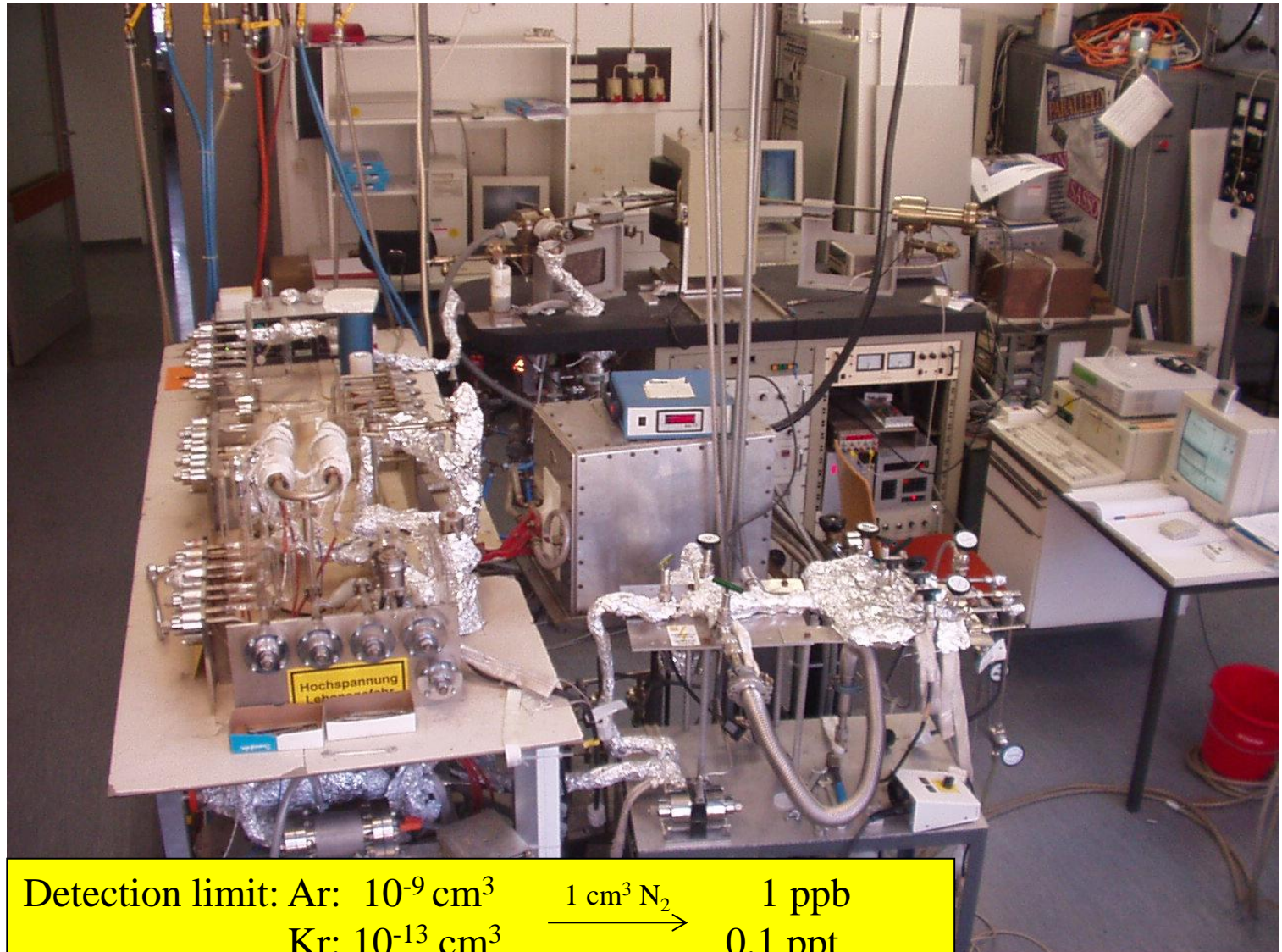


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^{222}Rn control

N_2 purification

Summary



Detection limit: Ar: 10^{-9} cm^3 $\xrightarrow{1 \text{ cm}^3 \text{ N}_2}$ 1 ppb
Kr: 10^{-13} cm^3 0.1 ppt

BOREXINO nitrogen



BOREXINO

^{222}Rn control

N_2 purification

Summary

Regular Purity Nitrogen:

- Technical 4.0 quality, not purified
- Production rate up to 100 m³/h (STP)
- ^{222}Rn (30 – 70) $\mu\text{Bq}/\text{m}^3$
- Ar ~ 10ppm, Kr ~ 30 ppt

High Purity Nitrogen:

- ^{222}Rn adsorption on charcoal (LTA)
- Achieved concentration (0.30 ± 0.09) $\mu\text{Bq}/\text{m}^3$
- Production rate up to 100 m³/h (STP)
- Ar and Kr not removed



LTA

LAK (Low Ar and Kr) Nitrogen:

- Spec. Ar < 0.4 ppm, Kr < 0.2 ppt
- ^{222}Rn < 7 $\mu\text{Bq}/\text{m}^3$
- Purification by adsorption on different materials extensively studied (successfully!)
- Cooperation with companies on the nitrogen survey
- Tests of the nitrogen delivery chain

Nitrogen survey

Nitrogen sample	C _{Ar} [ppm]	C _{Kr} [ppt]
MESSER (4.0)	200 ± 30	1680 ± 240
Air Liquide (4.0)	11.0 ± 1.3	40 ± 5
Linde AG, (7.0)	0.031 ± 0.004	2.9 ± 0.4
SOL (6.0)	0.0063 ± 0.0006	0.04 ± 0.01
Westfalen AG (6.0)	0.00050 ± 0.00008	0.06 ± 0.02
Goal (BOREXINO)	< 0.4	< 0.2

Tests of the delivery chains



Supplier/setup	C _{Rn} [$\mu\text{Bq}/\text{m}^3$]	C _{Ar} [ppm]	C _{Kr} [ppt]
Linde AG, 3-m ³ movable tank	1.2	0.018	0.06
SOL, 16-m ³ tank	8	0.012	0.02

BOREXINO LAK nitrogen



LAK Nitrogen tank installed at Gran Sasso



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^{222}Rn control

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Summary

$^{222}\text{Rn} \sim 8 \mu\text{Bq}/\text{m}^3$
 $\text{Ar} \sim 15 \text{ ppb}$
 $\text{Kr} \sim 0.1 \text{ ppt}$

Appl. Rad. Isot. 61 (2004) 197
AIP Conf. Proc. 897 (2007) 45
Int. J. Mod. Phys. A29 (2014) 1442009

Purification of nitrogen from Kr

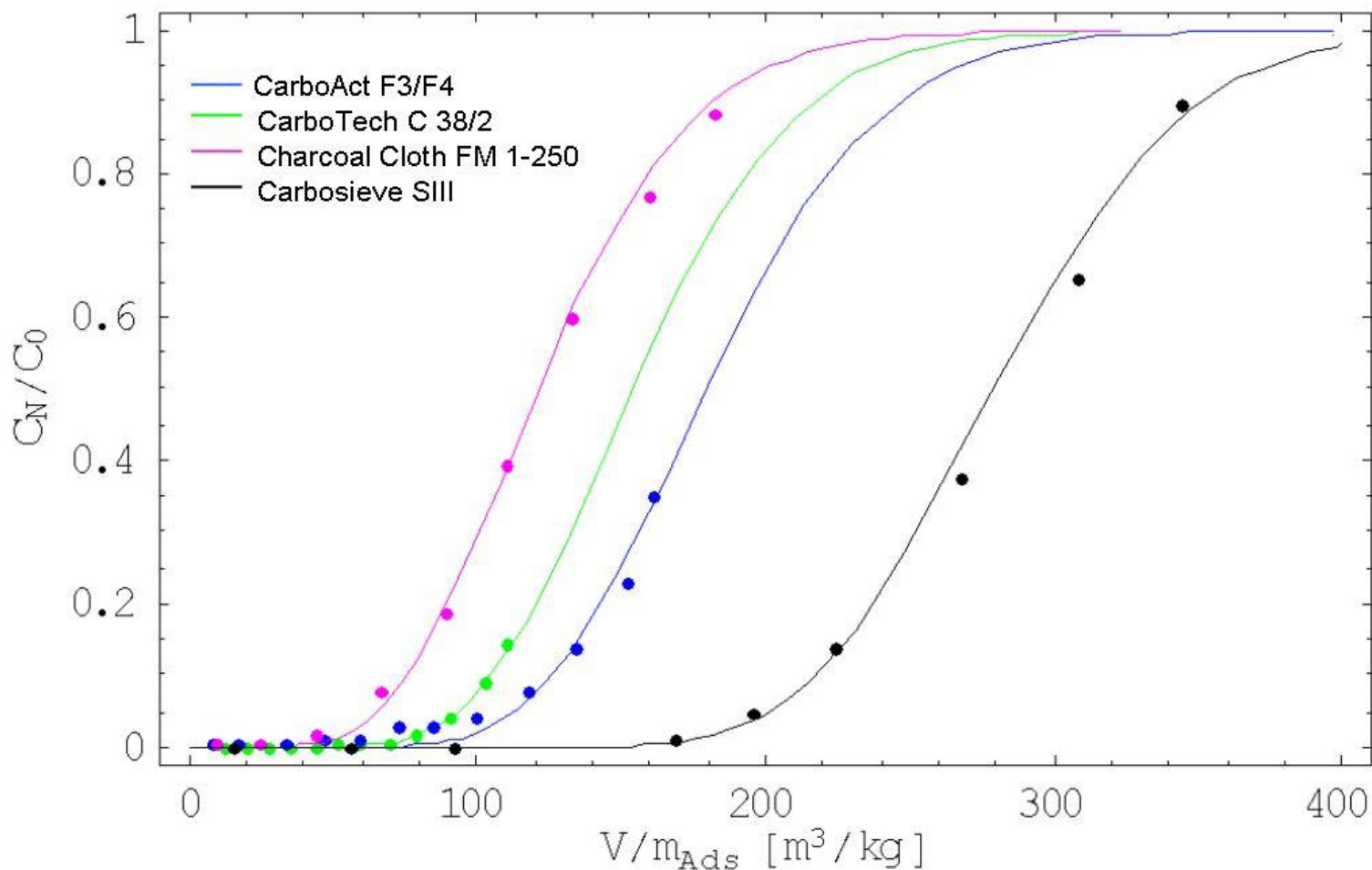


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Summary



Conclusions



BOREXINO

^{222}Rn control

N_2 purification

Summary

- BOREXINO has achieved an unprecedented background level in the liquid scintillator
- Strict quality control program including the assay of all components of the detector during its construction
- **+10 years of R&D, many people/institutions involved**
- Several detectors and experimental methods were developed allowing measurements even at a single atom level
- Most of the developed techniques are world-wide most sensitive (gamma-ray spectroscopy, ^{222}Rn detection, ^{222}Rn diffusion) and are applied in next-generation experiments (GERDA, XENON, DARKSIDE,...)