



# Ultralow background LS and related technologies

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# Outline



- Ultra-pure LS: BOREXINO
- Internal and external background: mitigation techniques
- LS purification
- Conclusions

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# BOREXINO at LNGS



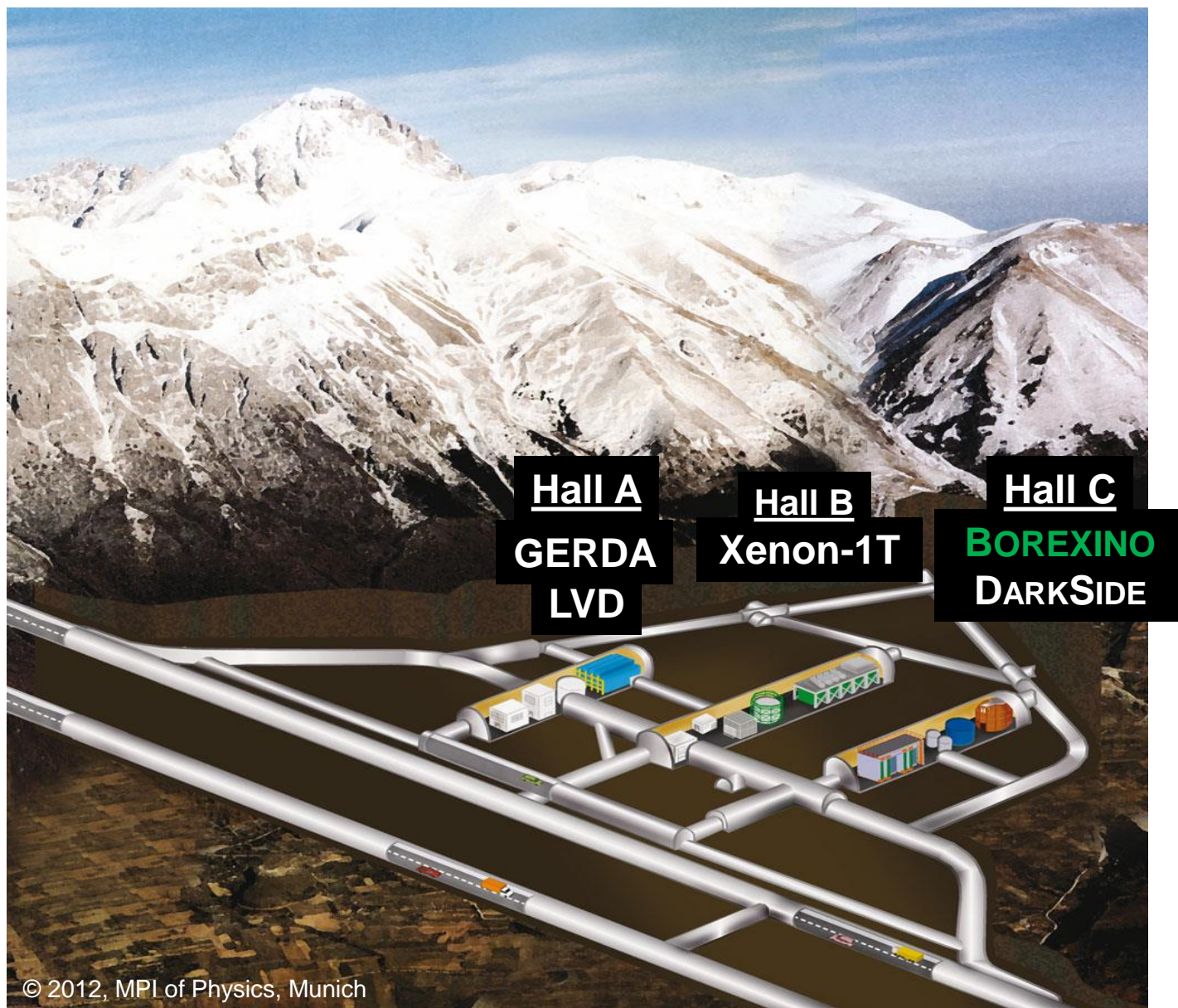
BOREXINO

Bcg mitigation

LS purification

Conclusions

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International Workshop On Next Generation Nucleon Decay And Neutrino Detectors, 03-05.11.2016 Beijing, China

# BOREXINO design



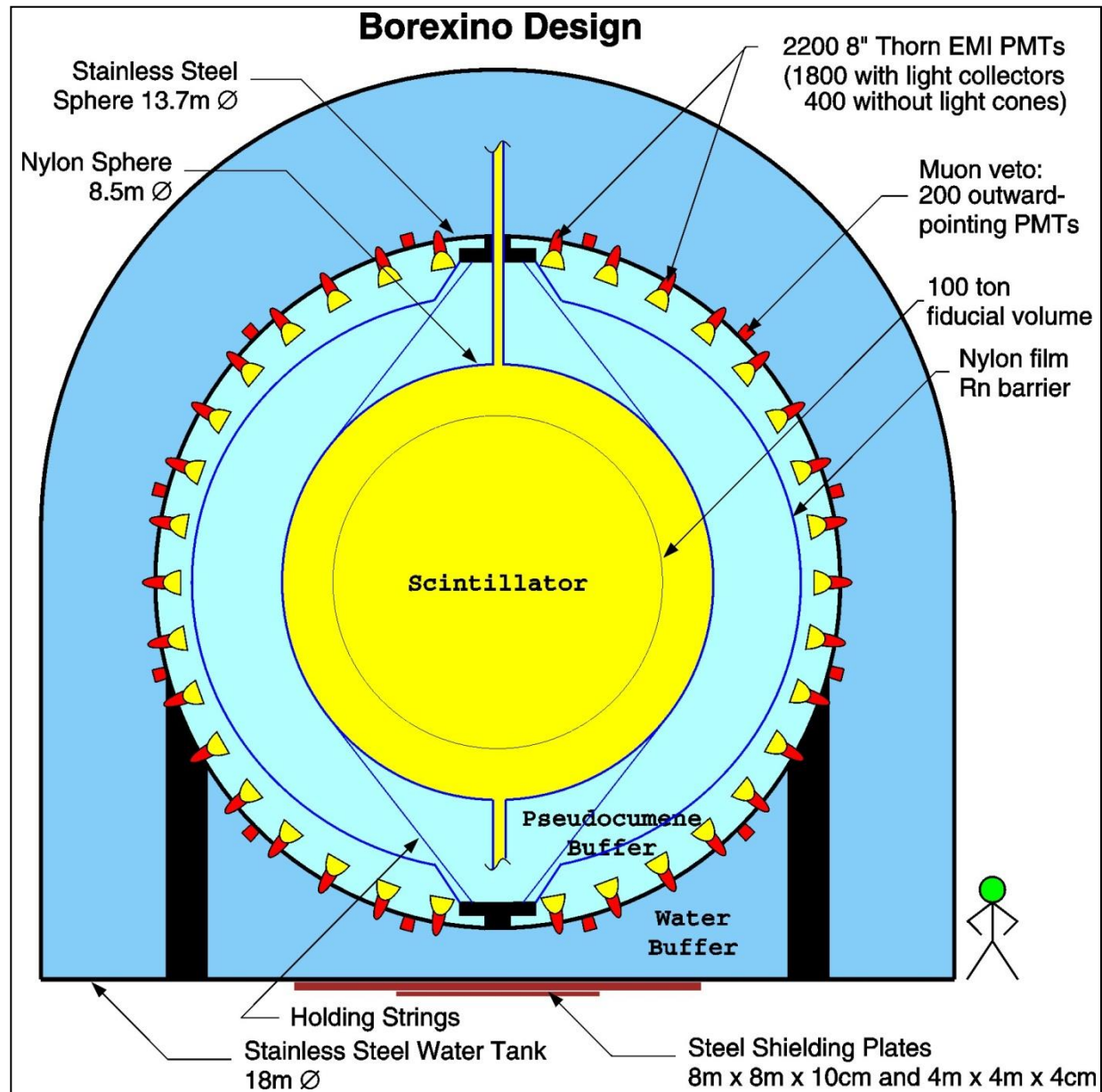
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# BOREXINO radio-purity

In a nutshell: the cleanest detector ever built



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Isotope	Specification for LS	Achieved after filling (2007 - 2010)
$^{238}\text{U}$	$\leq 10^{-16} \text{ g/g}$	$(5.3 \pm 0.5) \cdot 10^{-18} \text{ g/g}$
$^{232}\text{Th}$	$\leq 10^{-16} \text{ g/g}$	$(3.8 \pm 0.8) \cdot 10^{-18} \text{ g/g}$
$^{14}\text{C}/^{12}\text{C}$	$\leq 10^{-18}$	$(2.69 \pm 0.06) \cdot 10^{-18} \text{ g/g}$
$^{40}\text{K}$	$\leq 10^{-18} \text{ g/g}$	$\leq 0.4 \cdot 10^{-18} \text{ g/g}$
$^{85}\text{Kr}$	$\leq 1 \text{ cpd/100 t}$	$(30 \pm 5) \text{ cpd/100 t}$
$^{39}\text{Ar}$	$\leq 1 \text{ cpd/100 t}$	$\ll ^{85}\text{Kr}$
$^{210}\text{Po}$	not specified	$\sim (70) 1 \text{ dpd/100 t}$
$^{210}\text{Bi}$	not specified	$(20) 70 \text{ dpd/100 t}$

$$\left. \begin{aligned} A_{Bx} = \sum (bcg \text{ ev.}) &\sim 30 \frac{\text{cpd}}{100 \text{ t}} \sim 10^{-9} \frac{\text{Bq}}{\text{kg}} \\ A_{\text{water}} &\sim 10 \frac{\text{Bq}}{\text{kg}} \end{aligned} \right\} \rightarrow \text{activity reduction factor} \\ \textcolor{red}{f} \sim \textcolor{red}{10^{10}}$$



# Background mitigation techniques



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- 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 (PC buffer, water) suppression of external radiation
- Careful selection of construction materials and detector components with respect to content of radioactive isotopes,  $^{222}\text{Rn}$  emanation and permeability
- Preventing surface contamination
- Application of appropriate purification (liquids, gases) and cleaning techniques

# BOREXINO design



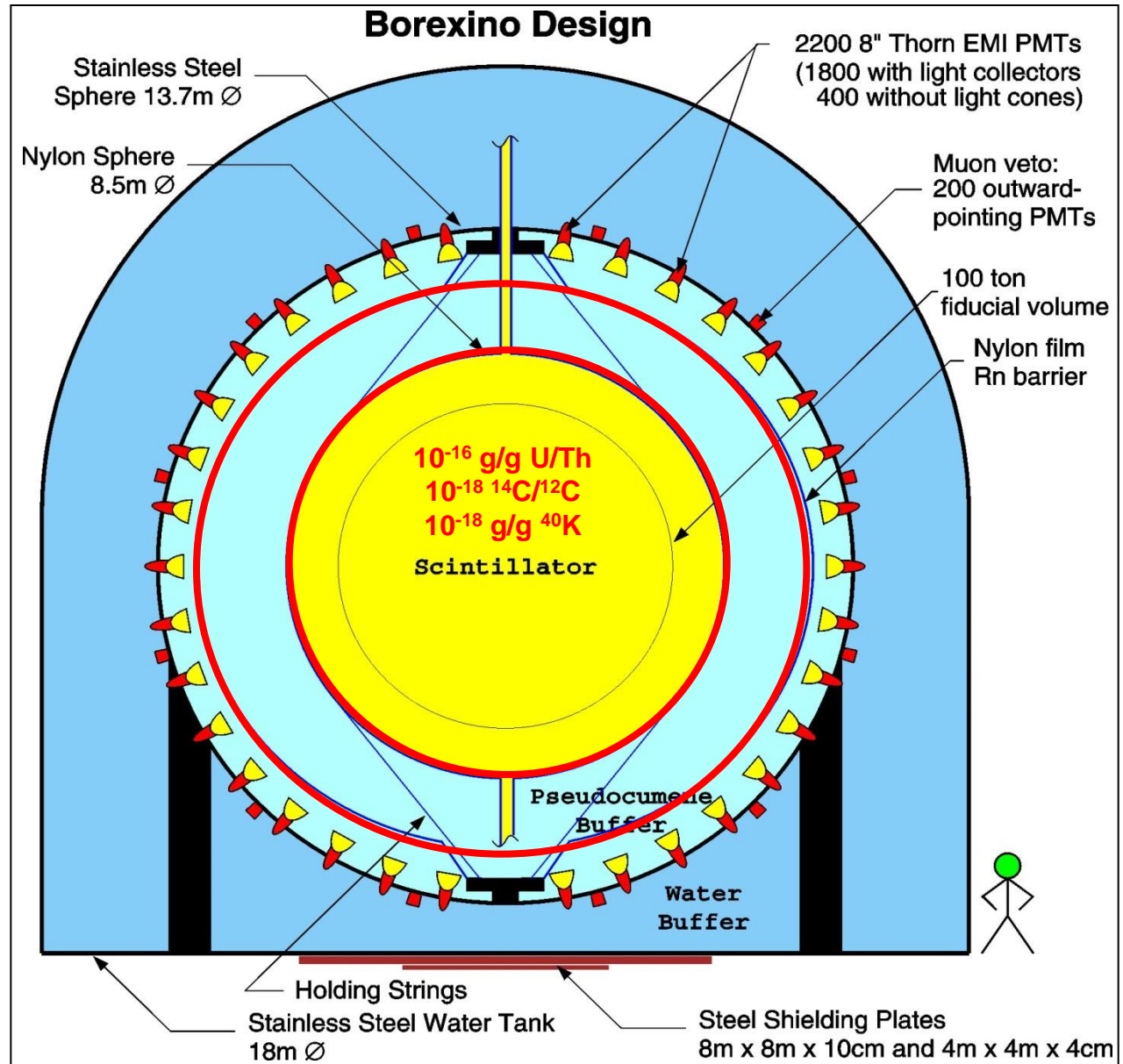
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# CTF – testing the scintillator



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CFT – 1: 1995  
CTF – 2: 2000  
CTF – 3: 2001-2003

- Phys. Lett. B, 422, 349 (1998)
- Astrop. Phys. 8(3), 141 (1998)
- NIM A406, 411 (1998)
- Physics Letters B 525, 29 (2002)
- Physics Letters B 563, 23 (2003)
- Physics Letters B 563, 37 (2003)
- JETP Lett. 78 No 5, 261 (2003)
- Eur. Phys. J. C 37, 421 (2004)
- Eur. Phys. J. C 47, 21 (2006)
- Phys. Rev. C 74, 045805 (2006)

$$\frac{^{14}\text{C}}{^{12}\text{C}} \sim 10^{-18}$$

$$C_{U/Th} \sim 4 \times 10^{-16} \text{ g/g}$$



# HPGe spectroscopy



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## GeMPIs at GS (3800 m w.e.)

- GeMPI I operational since 1997 (MPIK)
- GeMPI II built in 2004 (MCavern)
- GeMPI III/IV constructed in 2007-2012 (MPIK/LNGS)
- World's most sensitive spectrometers

### GeMPI I:

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

**Sensitivity for U/Th:**  
 **$\sim 10 \mu\text{Bq/kg}$**

Appl. Rad. Isot., 53 (2000) 191  
Astrop. Phys. 18 (2002) 1



# Survey different materials



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

Astroparticle Physics 18 (2002) 1

# $^{222}\text{Rn}$ emanation



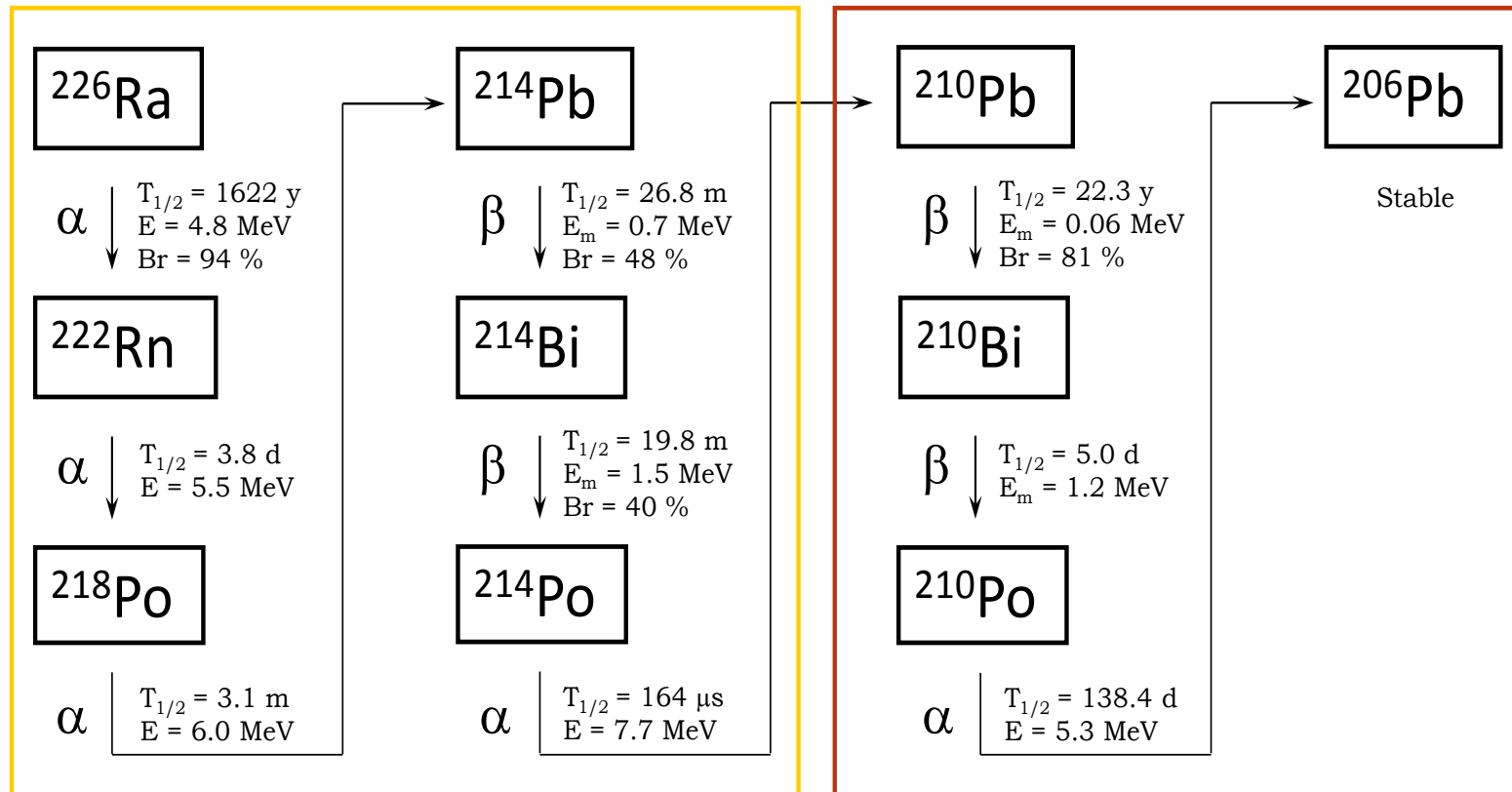
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# $^{222}\text{Rn}$ emanation



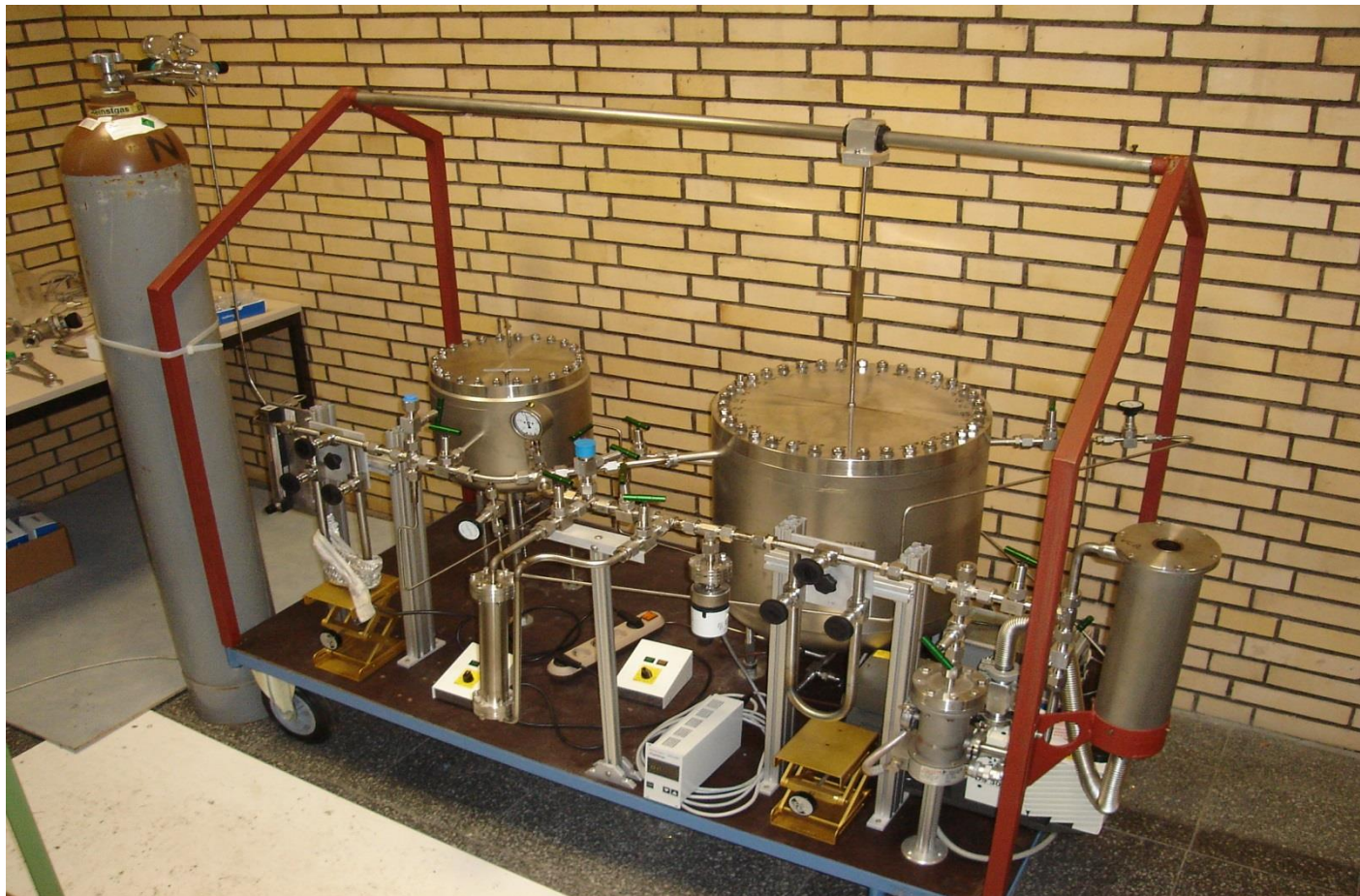
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**Blanks:**

20 l  $\rightarrow$  50  $\mu\text{Bq}$

80 l  $\rightarrow$  80  $\mu\text{Bq}$

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

Appl. Rad. Isot. 53 (2000) 371



# $^{222}\text{Rn}$ emanation: examples



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System	Sample	Description	$^{222}\text{Rn}$ em. rate
PC storage area	SS vessel TK1	114 m <sup>3</sup>	< 60 mBq
	SS vessel TK2	114 m <sup>3</sup>	(45 ± 8) mBq
	SS vessel TK3	114 m <sup>3</sup>	(24 ± 5) mBq
N <sub>2</sub> 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 <sup>2</sup>	< 0.12 mBq
	H <sub>2</sub> O extraction column + 24 SS packages	0.6 m <sup>3</sup> / 608 m <sup>2</sup>	(4.83 ± 0.70) mBq
	N <sub>2</sub> sparging column + 26 SS packages	0.2 m <sup>3</sup> / 280 m <sup>2</sup>	(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

# $^{222}\text{Rn}/^{226}\text{Ra}$ in water



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## STRAW: System for the $^{222}\text{Rn}$ and $^{226}\text{Ra}$ Assay of Water

- Placed at the BOREXINO water plant
- $^{222}\text{Rn}$  extraction from 350 liters
- $^{222}\text{Rn}$  and  $^{226}\text{Ra}$  measurements possible

$^{222}\text{Rn}$  detection limit:  $\sim 0.1 \text{ mBq/m}^3$

$^{226}\text{Ra}$  detection limit:  $\sim 0.8 \text{ mBq/m}^3$

Nucl. Instr. Meth. A 497 (2003) 407

$\text{H}_2\text{O}$ flow [ $\text{m}^3/\text{h}$ ]	$\text{HPN}_2$ flow [ $\text{kg}/\text{h}$ ]	$C_{\text{Rn}}$ [ $\text{mBq}/\text{m}^3$ ]	$C_{\text{Ra}}$ [ $\text{mBq}/\text{m}^3$ ]
2	30	$704 \pm 7$	$1.2 \pm 0.5$
1	20	$247 \pm 6$	$3.8 \pm 0.7$
1	30	$186 \pm 5$	$2.0 \pm 0.6$
Loop mode		$3.0 \pm 0.4$	$1.3 \pm 0.9$

# BOREXINO design



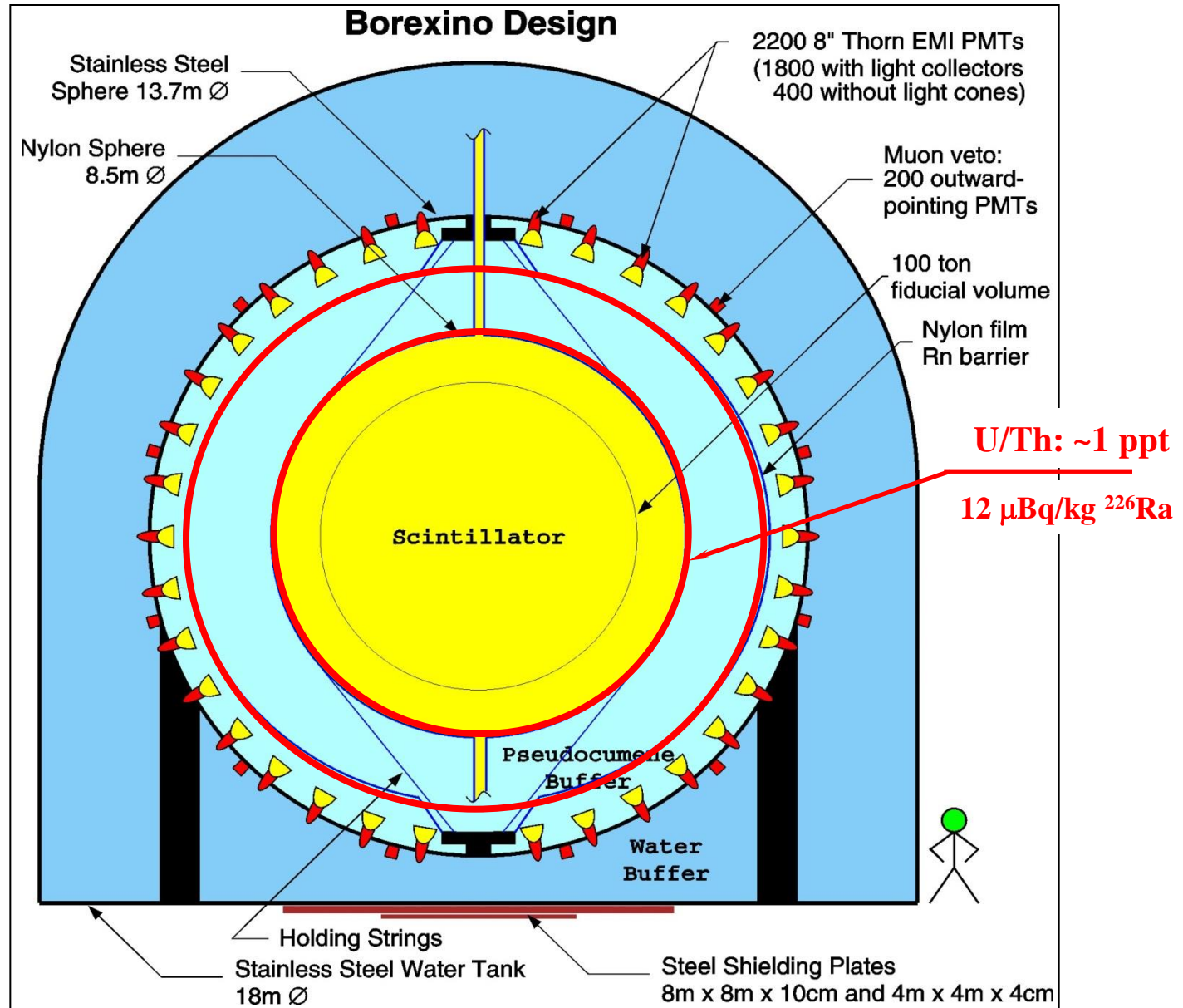
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# $^{222}\text{Rn}$ diffusion



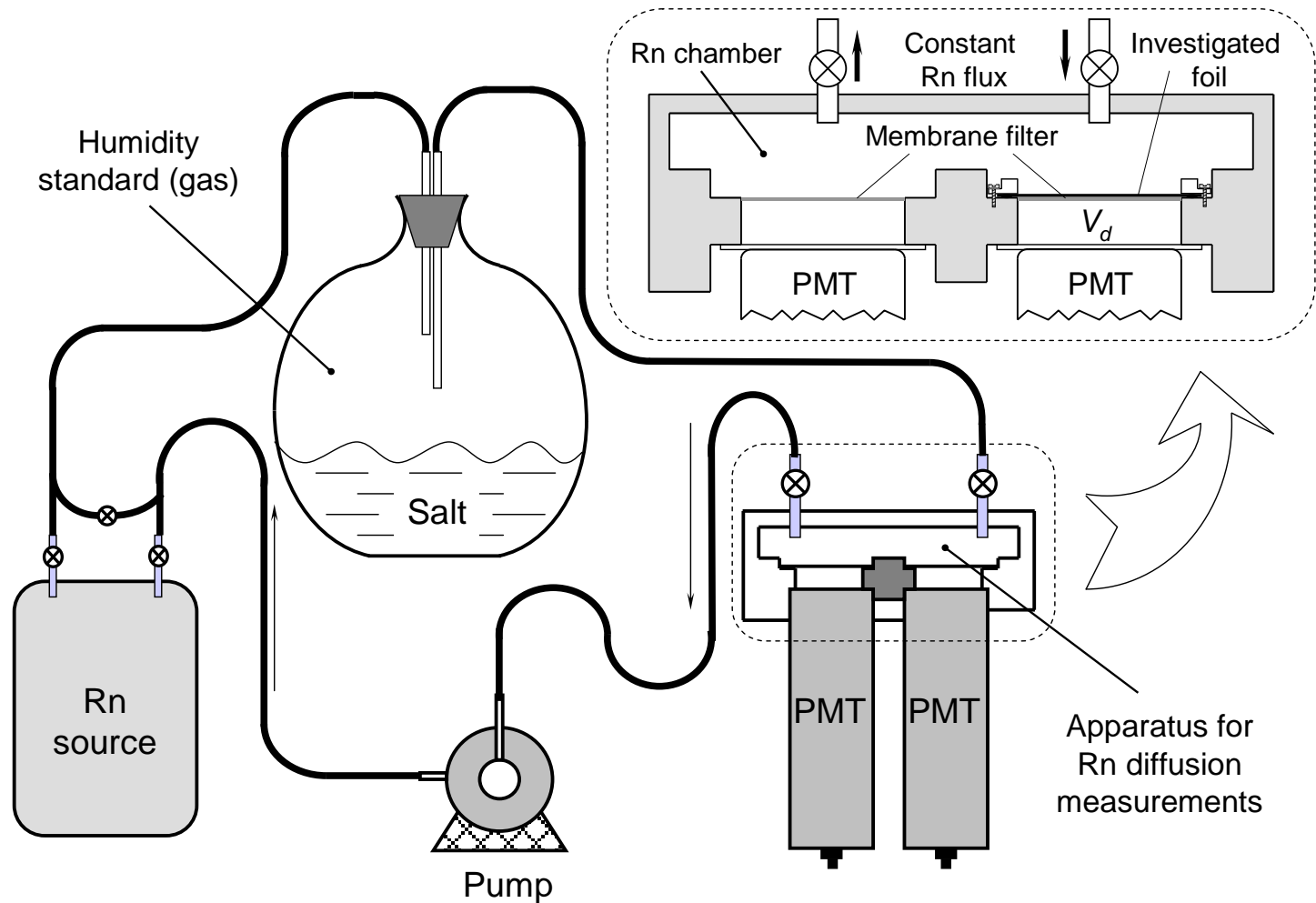
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Sensitivity:  $D \sim 10^{-13} \text{ cm}^2/\text{s}$   
 $d_e \sim 2 \mu\text{m}$



# $^{222}\text{Rn}$ diffusion



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

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

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

# $^{226}\text{Ra}$ in/on BOREXINO nylon



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

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

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

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

$$A_{\text{wet}} = A_{\text{sf}} + A_{\text{bulk}}$$

**Separation of the bulk  
and surface  $^{226}\text{Ra}$  conc.  
was possible through  
 $^{222}\text{Rn}$  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



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

A. Pocar, PhD Thesis (2003)



# Inflation of vessels in SSS



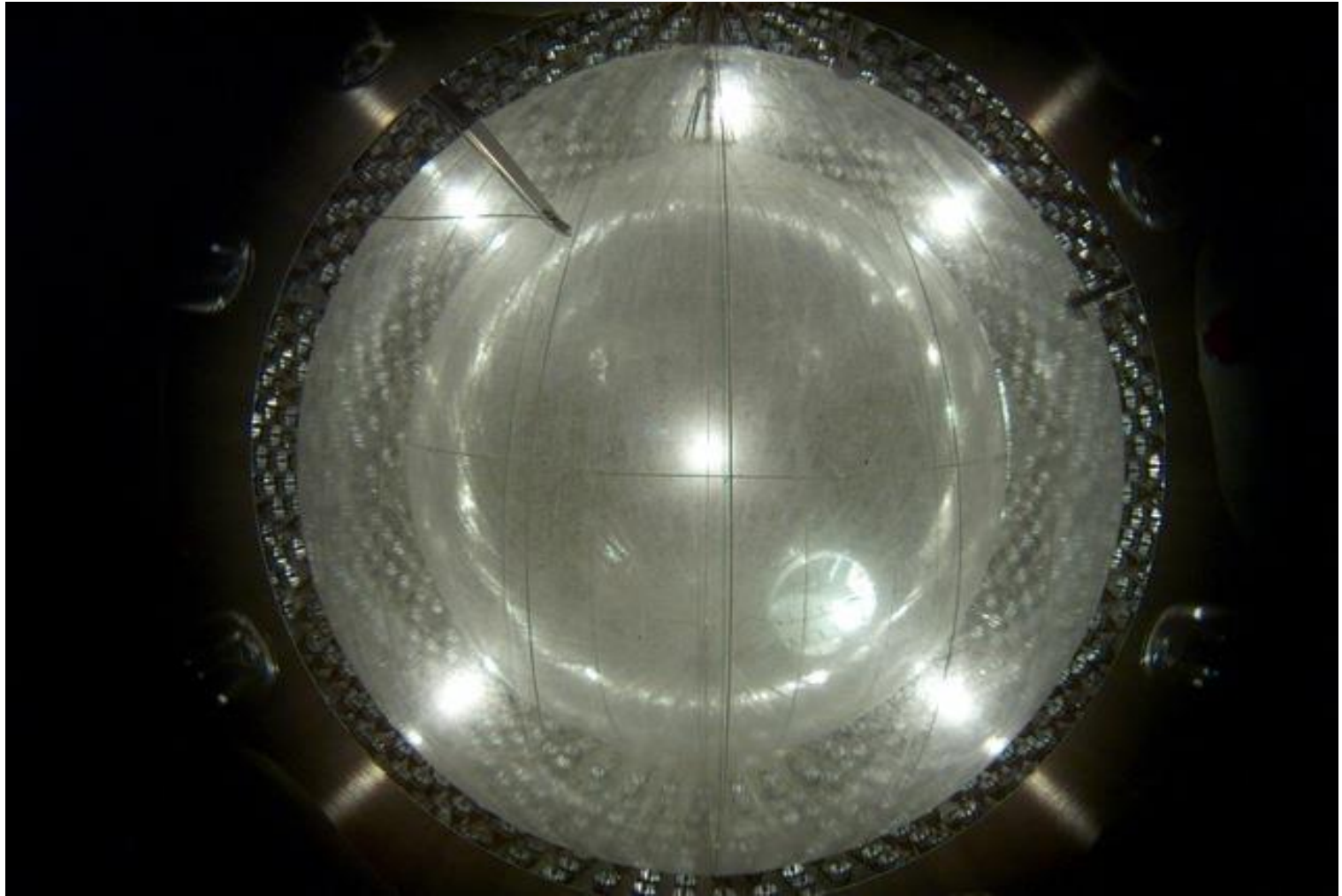
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The nylon vessels were inflated in the sphere  
with synthetic air:  $C_{Rn} < 100 \mu\text{Bq}/\text{m}^3$

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



# LS purification



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## Effective scintillator purification processes

- Efficient removal of radioactive elements
  - distillation
  - water extraction
  - nitrogen stripping
- Development of cleaning methods to remove radioactivity from equipment surfaces
- Purification of scintillator before and after filling detector

# LS purification during filling



- Vessels inflated with synthetic air
- Vessels filled with high-purity deionized water
- Scintillator distilled and stripped with LAK nitrogen during filling
- Detector filled with scintillator from top, while draining water from bottom

*J. Benziger et al. / Nuclear Instruments and Methods in Physics Research A 587 (2008) 277–291*

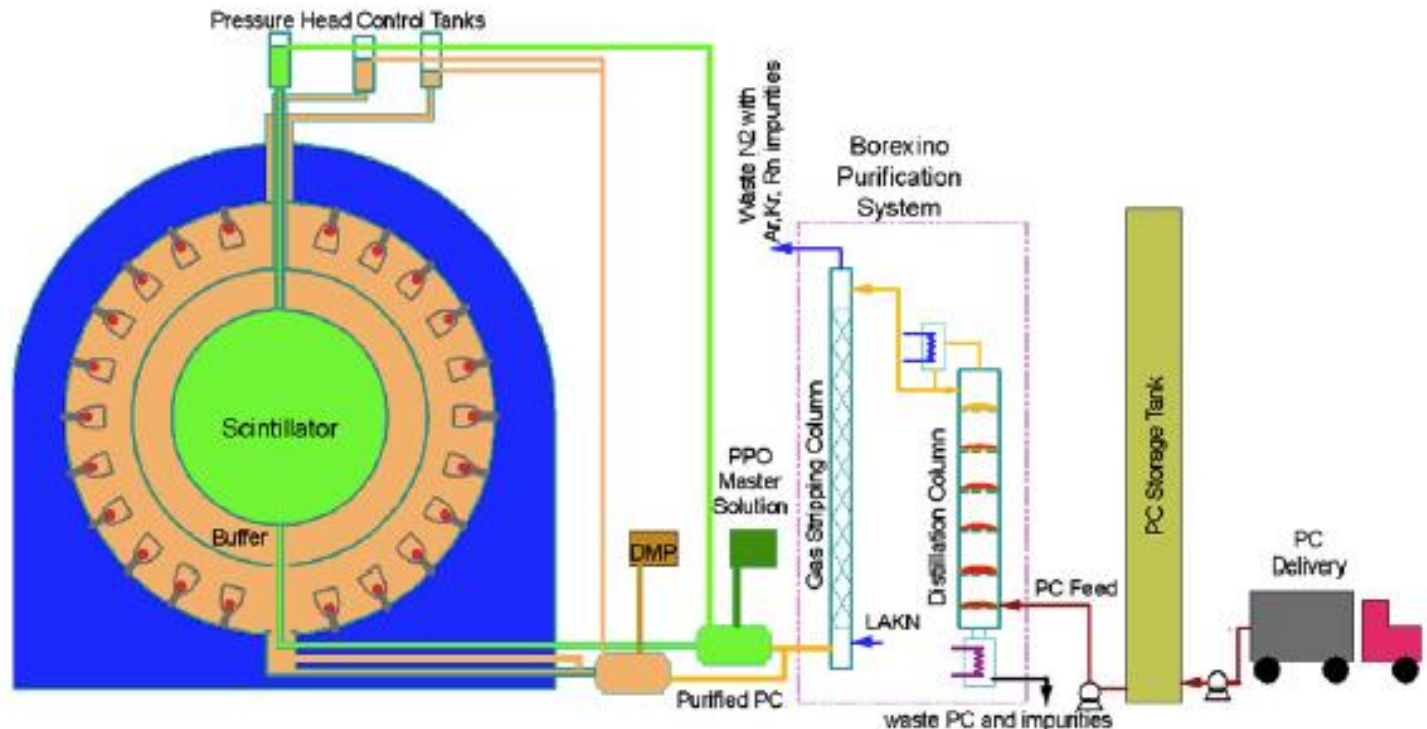
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# BOREXINO design



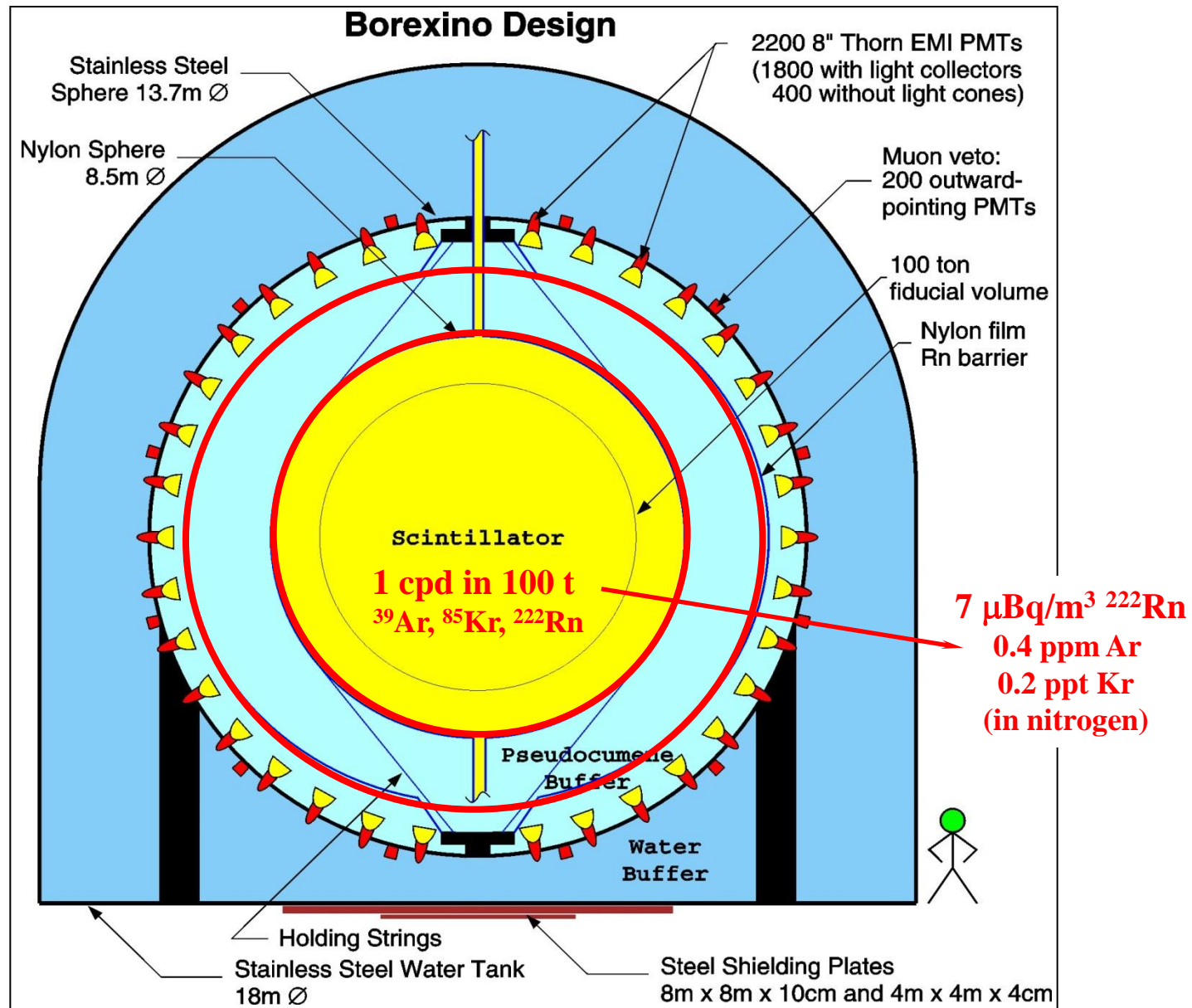
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# $^{222}\text{Rn}$ in gases: MoREx



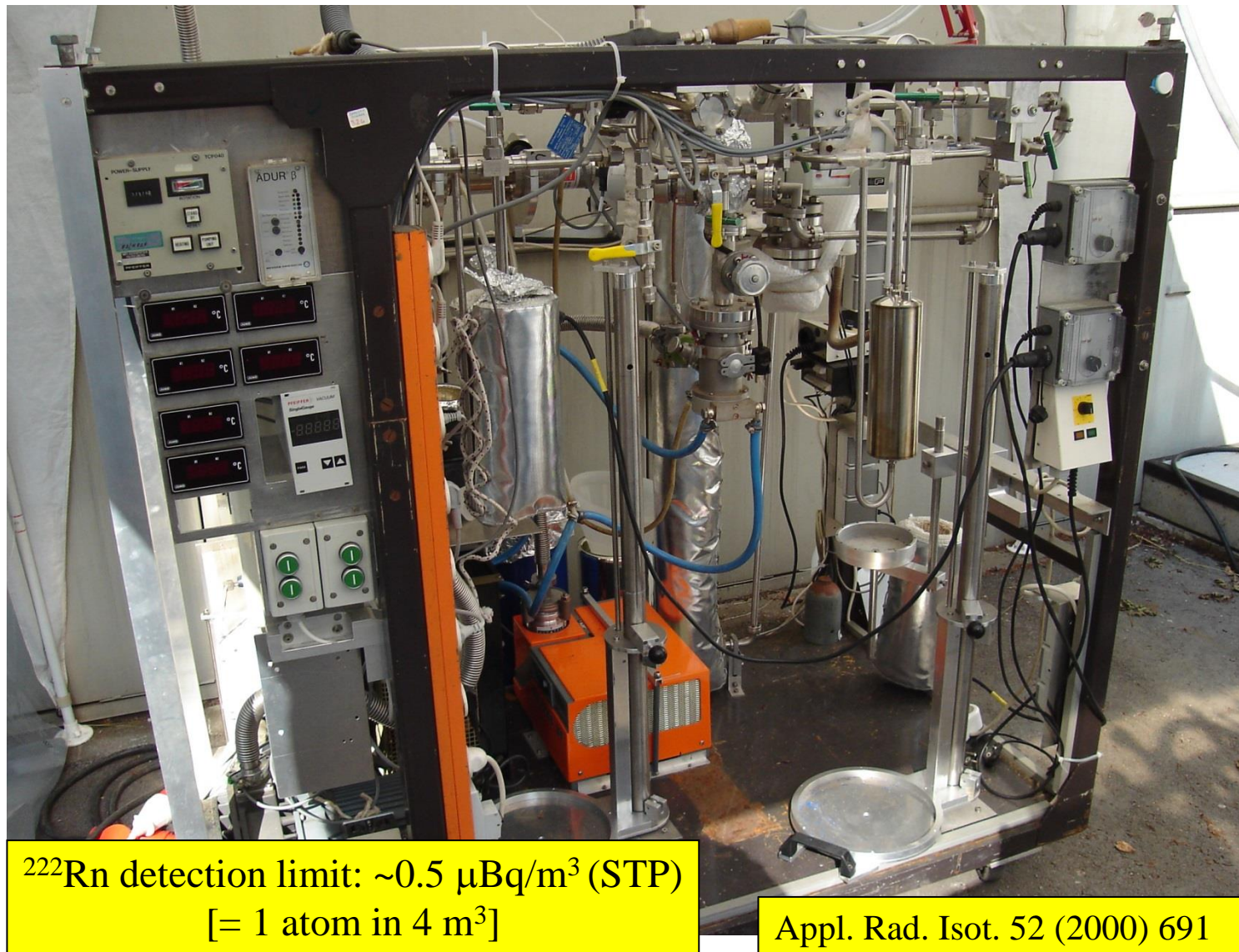
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$^{222}\text{Rn}$  detection limit:  $\sim 0.5 \mu\text{Bq}/\text{m}^3$  (STP)  
[= 1 atom in  $4 \text{ m}^3$ ]

Appl. Rad. Isot. 52 (2000) 691



# Ar and Kr in nitrogen



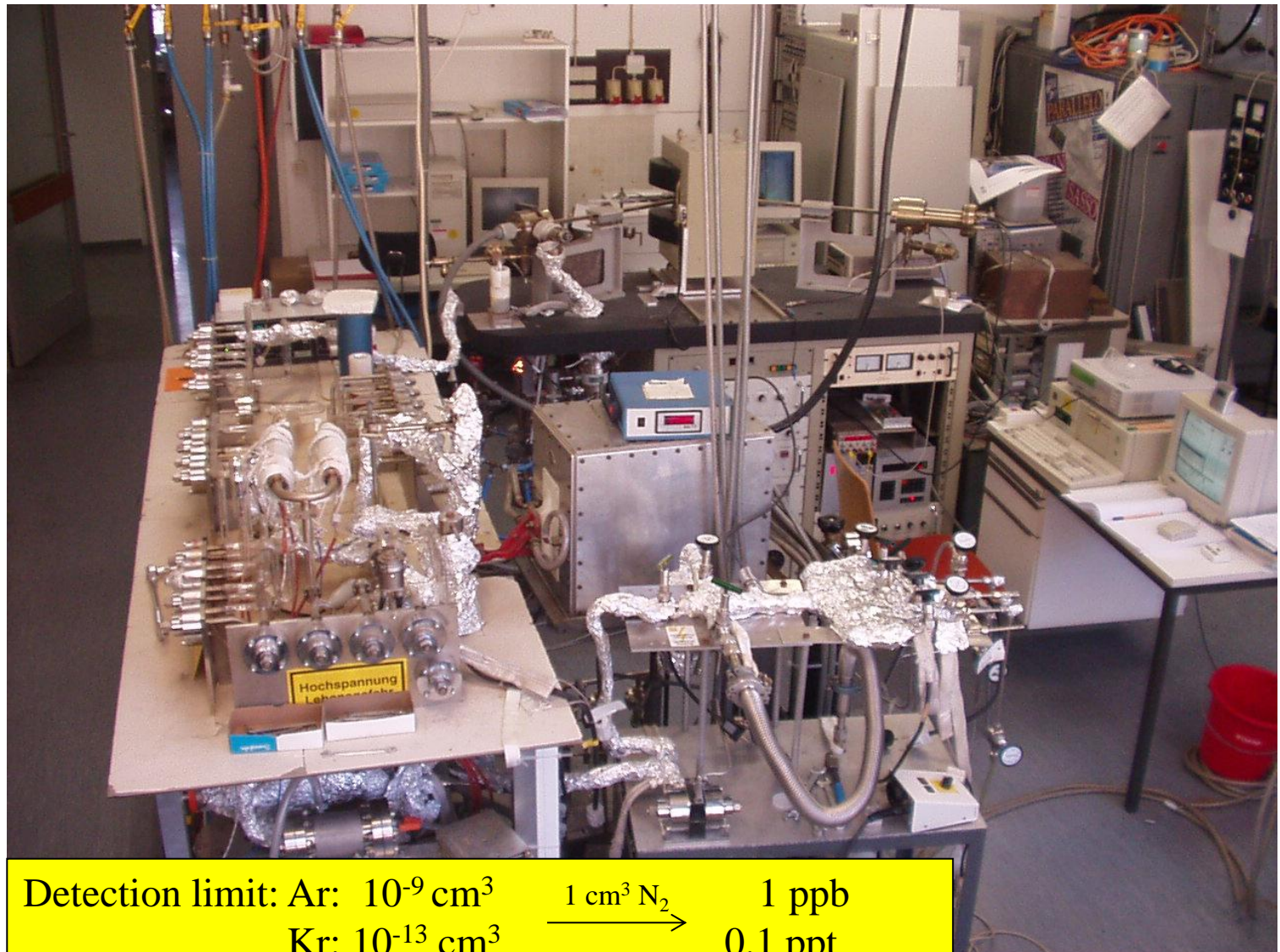
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Detection limit: Ar:  $10^{-9} \text{ cm}^3$   $\xrightarrow{1 \text{ cm}^3 \text{ N}_2}$  1 ppb  
Kr:  $10^{-13} \text{ cm}^3$  0.1 ppt

# BOREXINO nitrogen



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## Regular Purity Nitrogen:

- Technical 4.0 quality, not purified
- Production rate up to 100 m<sup>3</sup>/h (STP)
- <sup>222</sup>Rn (30 – 70) μBq/m<sup>3</sup>
- Ar ~ 10ppm, Kr ~ 30 ppt

## High Purity Nitrogen:

- <sup>222</sup>Rn adsorption on charcoal (LTA)
- Achieved concentration (0.30 ± 0.09) μBq/m<sup>3</sup>
- Production rate up to 100 m<sup>3</sup>/h (STP)
- Ar and Kr not removed



LTA

## LAK (Low Ar and Kr) Nitrogen:

- Spec. Ar < 0.4 ppm, Kr < 0.2 ppt
- <sup>222</sup>Rn < 7 μBq/m<sup>3</sup>
- 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 <sub>Ar</sub> [ppm]	C <sub>Kr</sub> [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 <sub>Rn</sub> [μBq/m <sup>3</sup> ]	C <sub>Ar</sub> [ppm]	C <sub>Kr</sub> [ppt]
Linde AG, 3-m <sup>3</sup> movable tank	1.2	0.018	0.06
SOL, 16-m <sup>3</sup> tank	8	0.012	0.02



# BOREXINO LAK nitrogen

LAK Nitrogen tank installed at Gran Sasso



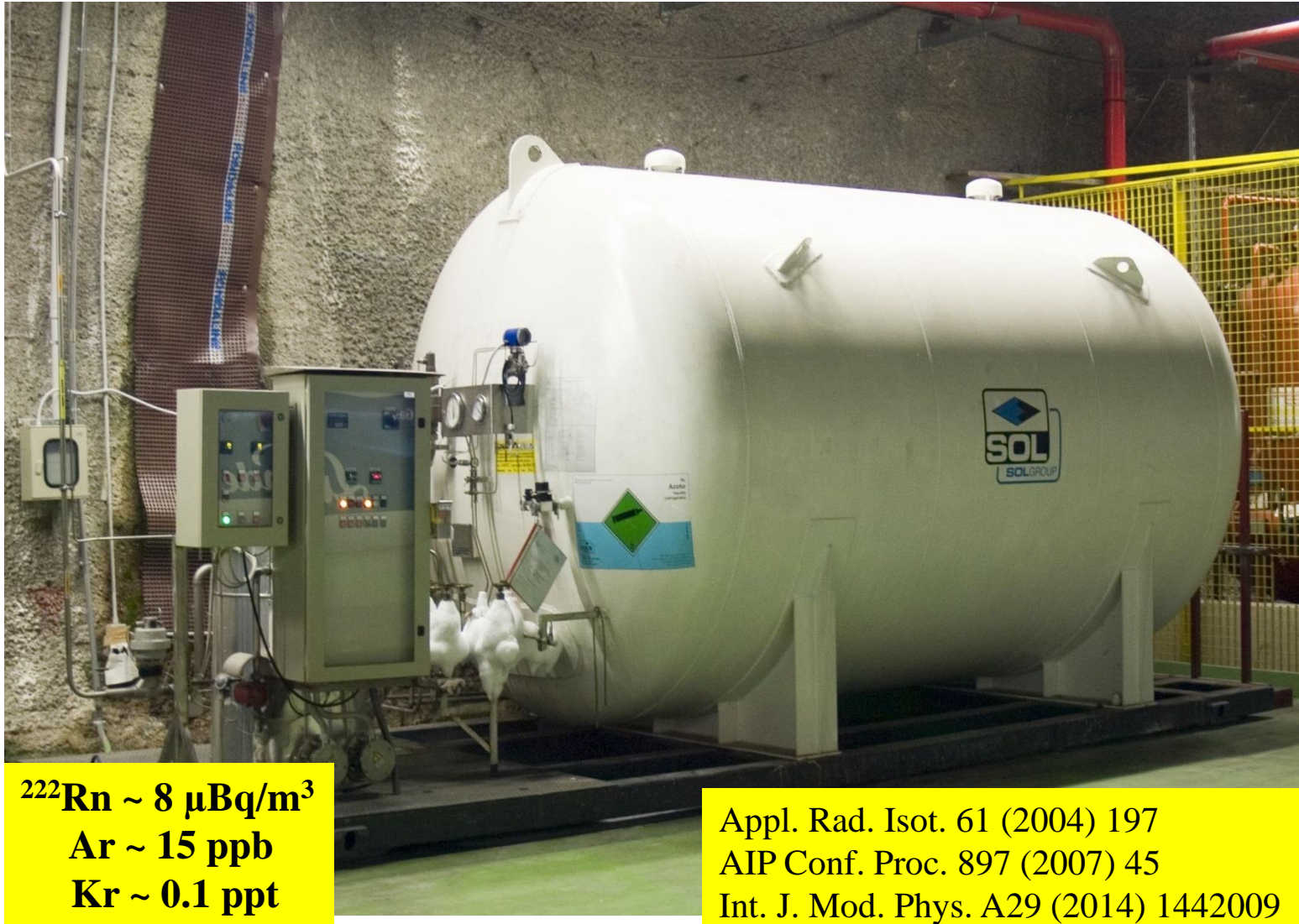
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$^{222}\text{Rn} \sim 8 \mu\text{Bq/m}^3$   
 $\text{Ar} \sim 15 \text{ ppb}$   
 $\text{Kr} \sim 0.1 \text{ ppt}$

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

# LS re-purification



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- From 2007 to 2010 the  $^{210}\text{Bi}$  background increased
  - Scintillator refills to compensate the vessel leak / others ?
  - $^{210}\text{Bi}$  rate: 20  $\rightarrow$  70 cpd/100 t
  - $^{85}\text{Kr}$  rate: 30 cpd/100 t (constant)
- To reduce background, scintillator was re-purified using two processes:
  - Water extraction to remove  $^{210}\text{Pb}$  ( $^{210}\text{Bi}$ )
  - Nitrogen stripping to remove  $^{85}\text{Kr}$  and other volatiles
- Six purification cycles were performed, each took  $\sim 1$  month
  - Each operation processed all the scintillator in the detector once ( $320 \text{ m}^3$ ).
  - Data were acquired to evaluate backgrounds after each operation.
  - Not all six operations were successful.



# BOREXINO radio-purity



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Isotope	Specification for LS	Achieved after filling (2007 - 2010)	After additional purification
$^{238}\text{U}$	$\leq 10^{-16}$ g/g	$(5.3 \pm 0.5) \cdot 10^{-18}$ g/g	$< 0.8 \cdot 10^{-19}$ g/g
$^{232}\text{Th}$	$\leq 10^{-16}$ g/g	$(3.8 \pm 0.8) \cdot 10^{-18}$ g/g	$< 1.2 \cdot 10^{-18}$ g/g
$^{14}\text{C}/^{12}\text{C}$	$\leq 10^{-18}$	$(2.69 \pm 0.06) \cdot 10^{-18}$ g/g	unchanged
$^{40}\text{K}$	$\leq 10^{-18}$ g/g	$\leq 0.4 \cdot 10^{-18}$ g/g	unchanged
$^{85}\text{Kr}$	$\leq 1$ cpd/100 t	$(30 \pm 5)$ cpd/100 t	$\leq 5$ cpd/100 t
$^{39}\text{Ar}$	$\leq 1$ cpd/100 t	$\ll ^{85}\text{Kr}$	$\ll ^{85}\text{Kr}$
$^{210}\text{Po}$	not specified	$\sim (70)$ 1 dpd/100 t	unchanged
$^{210}\text{Bi}$	not specified	$(20)$ 70 dpd/100 t	$(20 \pm 5)$ cpd/100 t

# Topics not discussed



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- $^{226}\text{Ra}$  adsorption on the nylon vessel (J. Rad. Nuc. Chem. 296 (2013) 639)
- $^{222}\text{Rn}$ -daughters deposition on the nylon vessel (E. Harding, Princeton)
- Tests of  $^{210}\text{Pb}$  removal from PC (J. Rad. Nuc. Chem. 296 (2013) 639)
- Online  $^{222}\text{Rn}$  monitoring with an electrostatic detector (NIM A 460 (2001) 272)
- Adsorption of noble gases on various porous materials (B. Freudiger, PhD Thesis (2003))
- NAA and ICP-MS measurements (Astroparticle Physics 18 (2002) 1)

# Conclusions



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- 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 (Ge spectroscopy,  $^{222}\text{Rn}$  detection,  $^{222}\text{Rn}$  diffusion) and are applied in next-generation experiments (GERDA, XENON, DARKSIDE,...)