Low-Radioactivity Techniques

Cracow, Oct 1-4, 2024

The BOREXINO low-background techniques

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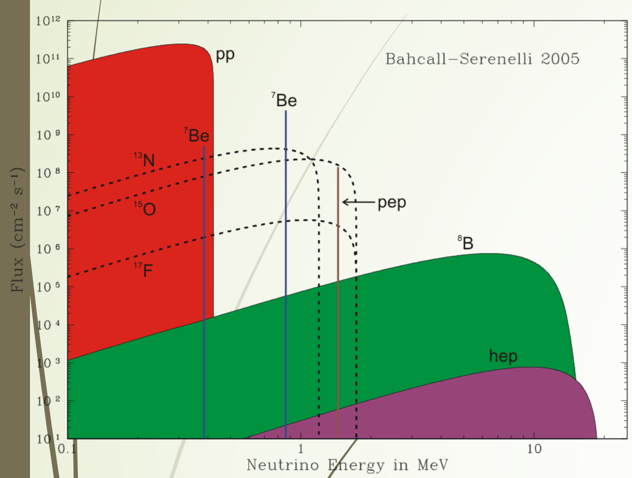
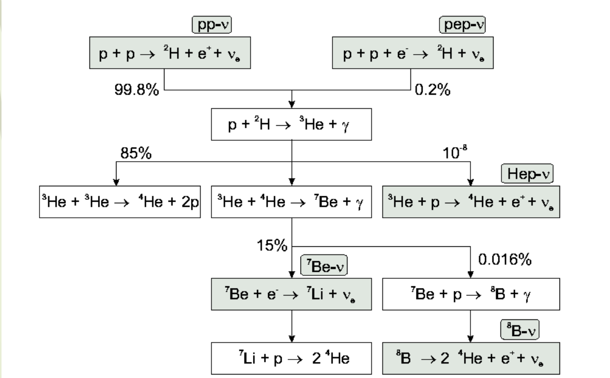
BOREXINO



Bahcall–Serenelli 2005
pp
7Be
7Be
13N
15O
17F
pep
8B
hep
Flux (cm⁻² s⁻¹)
Neutrino Energy in MeV
0.1
1
10



pp-ν
p + p → 2H + e+ + νe
pep-ν
p + p + e- → 2H + νe
99.8%
0.2%
p + 2H → 3He + γ
85%
10-8
Hep-ν
3He + 3He → 4He + 2p
3He + 4He → 7Be + γ
3He + p → 4He + e+ + νe
15%
0.016%
7Be-ν
7Be + e- → 7Li + νe
7Be + p → 8B + γ
8B-ν
7Li + p → 2 4He
8B → 2 4He + e+ + νe

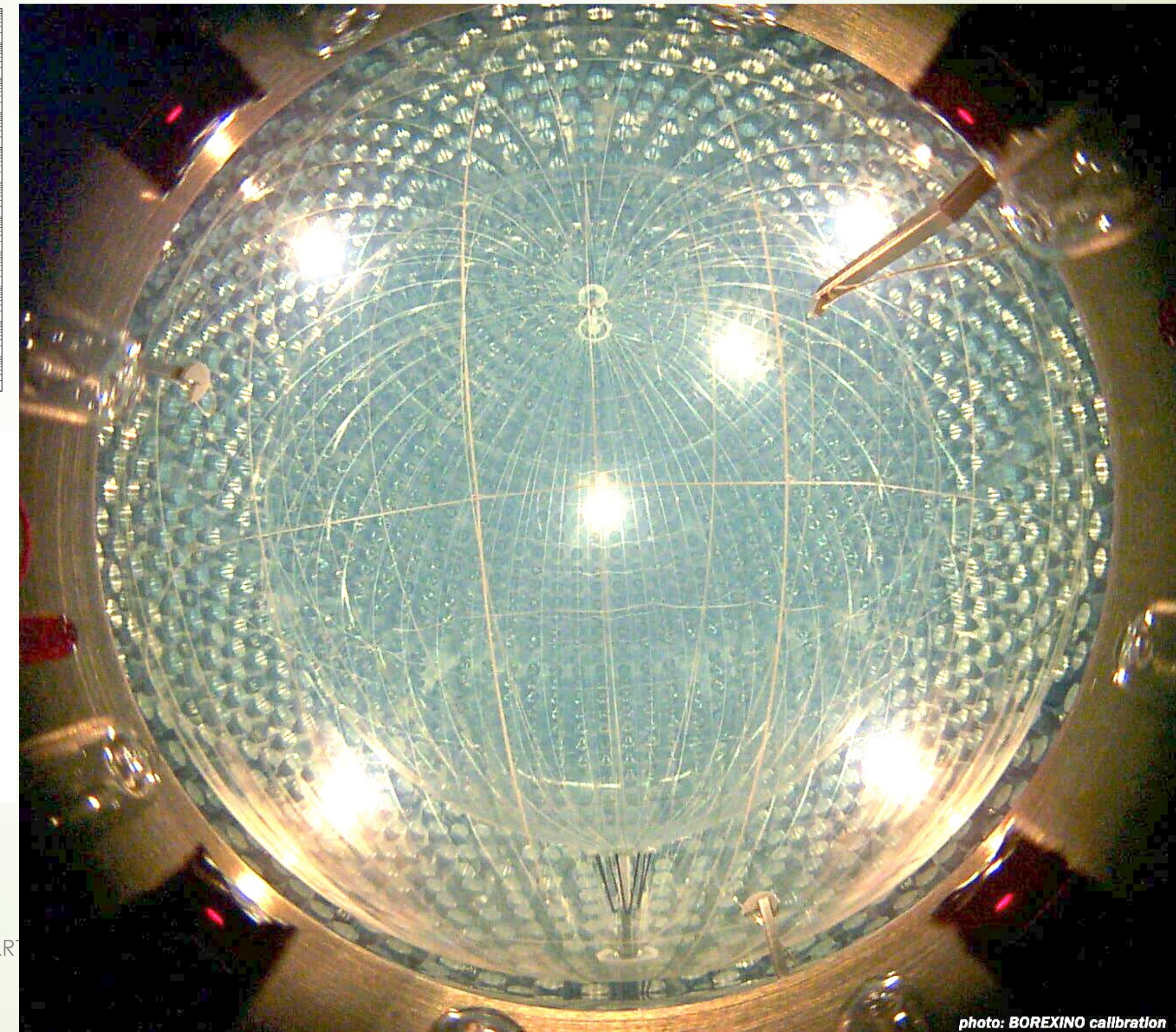
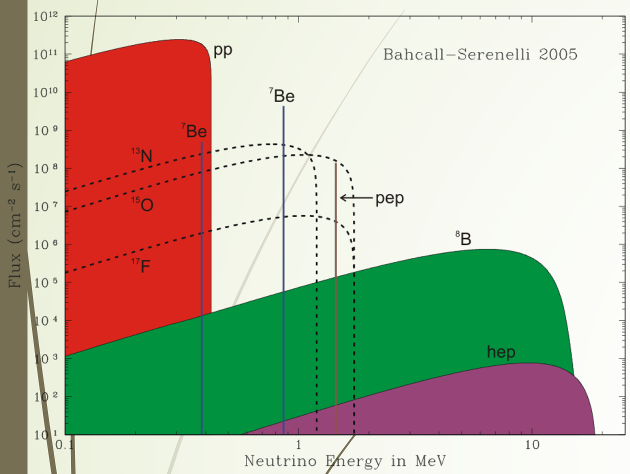
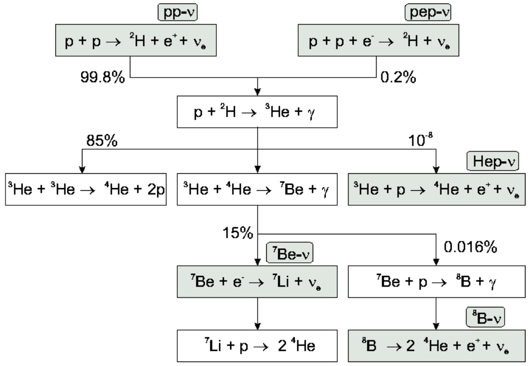
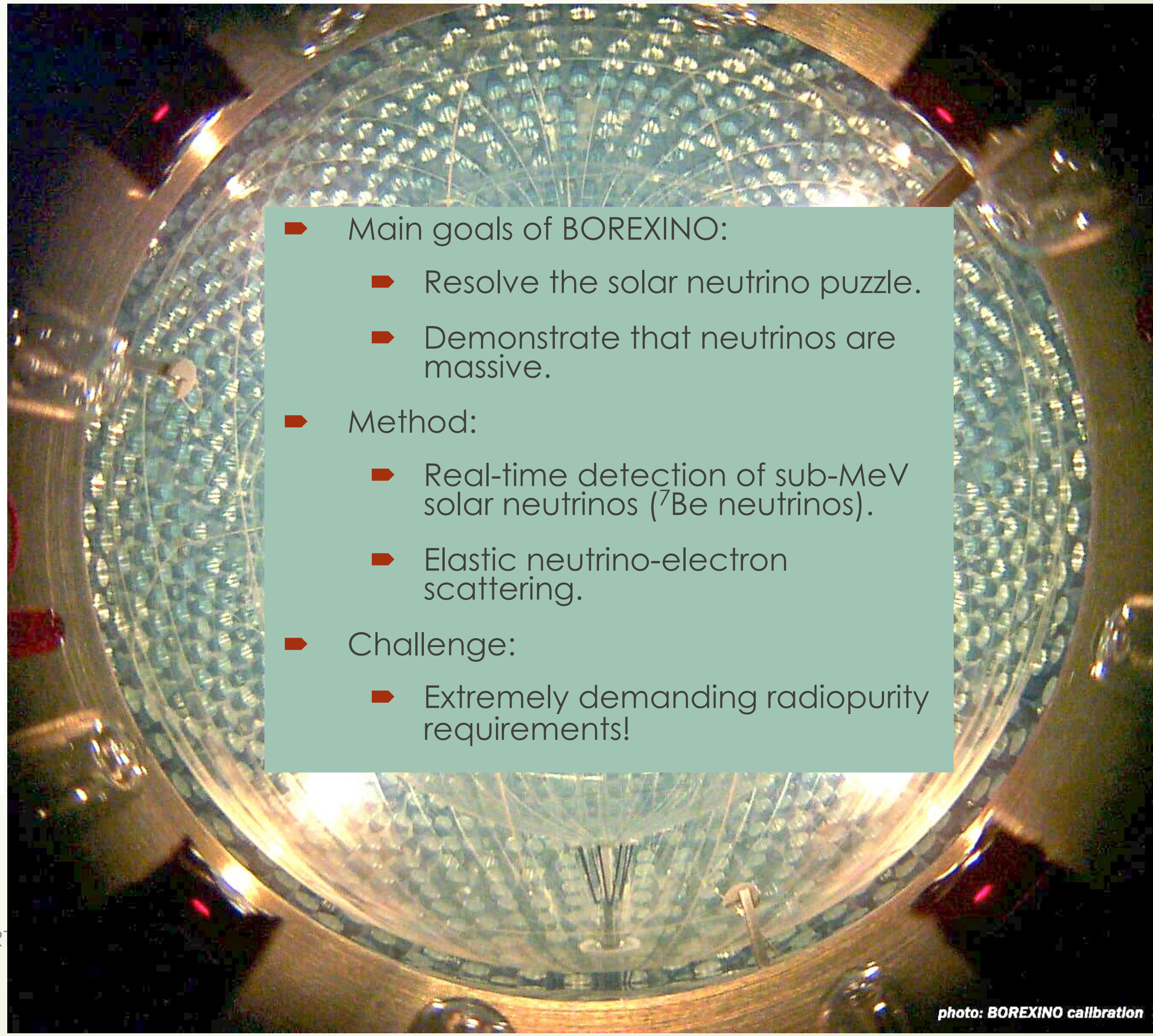


photo: BOREXINO calibration

BOREXINO

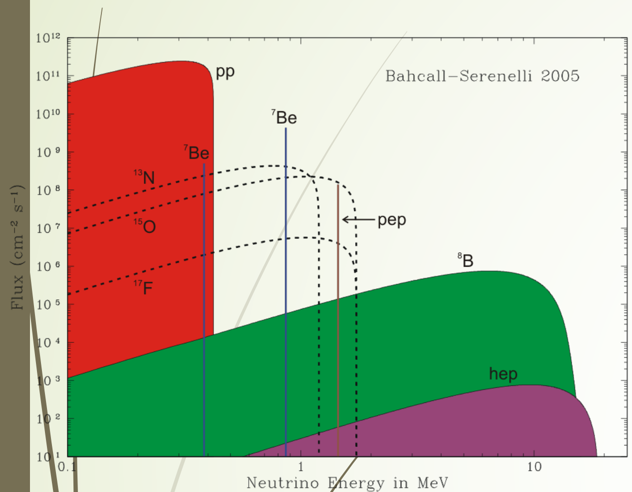


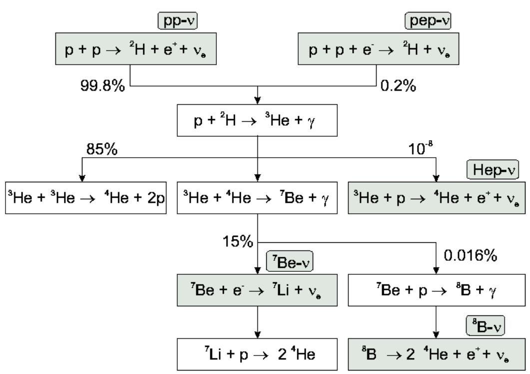


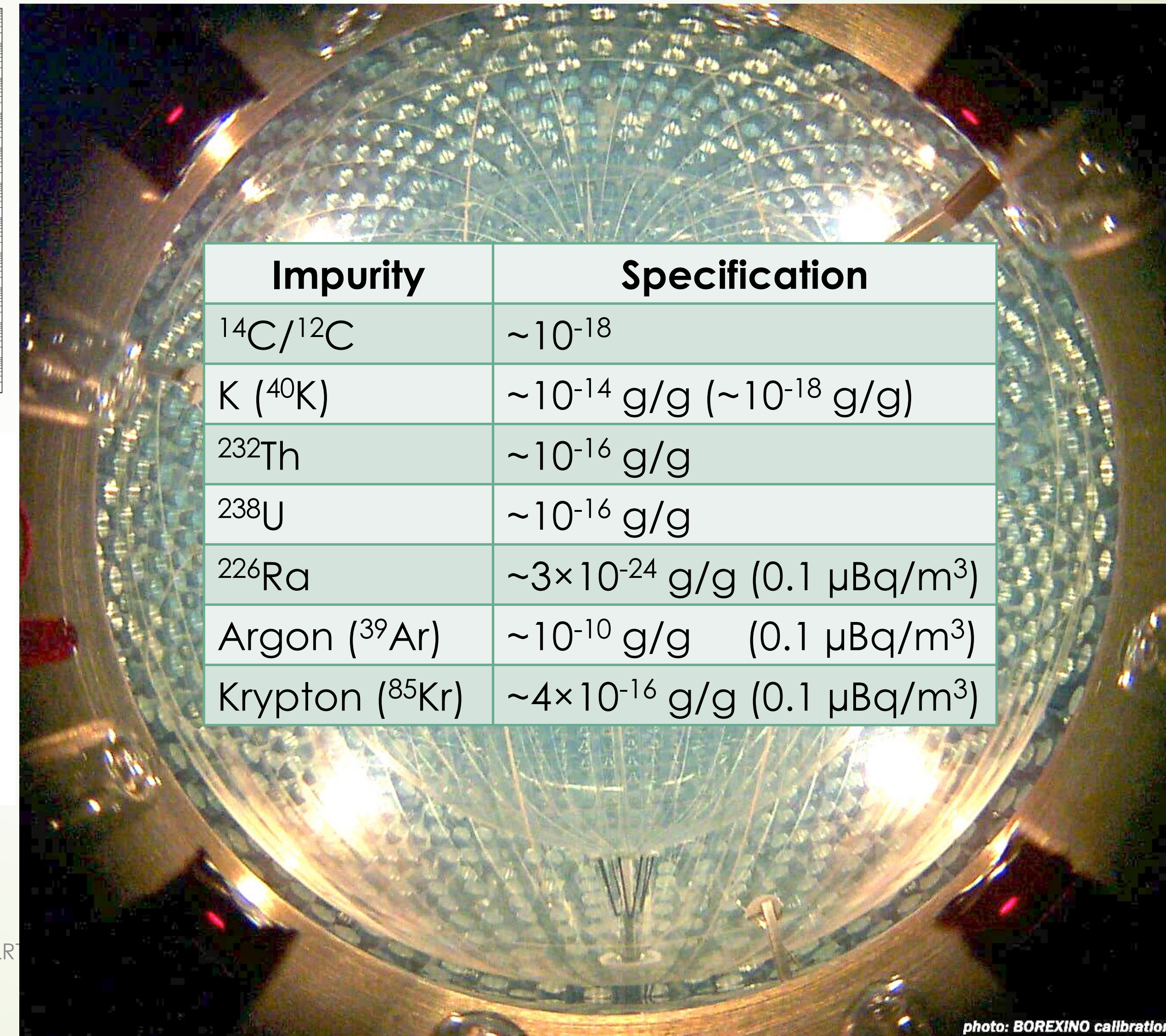


- Main goals of BOREXINO:
  - Resolve the solar neutrino puzzle.
  - Demonstrate that neutrinos are massive.
- Method:
  - Real-time detection of sub-MeV solar neutrinos (^{7}Be neutrinos).
  - Elastic neutrino-electron scattering.
- Challenge:
  - Extremely demanding radiopurity requirements!

BOREXINO







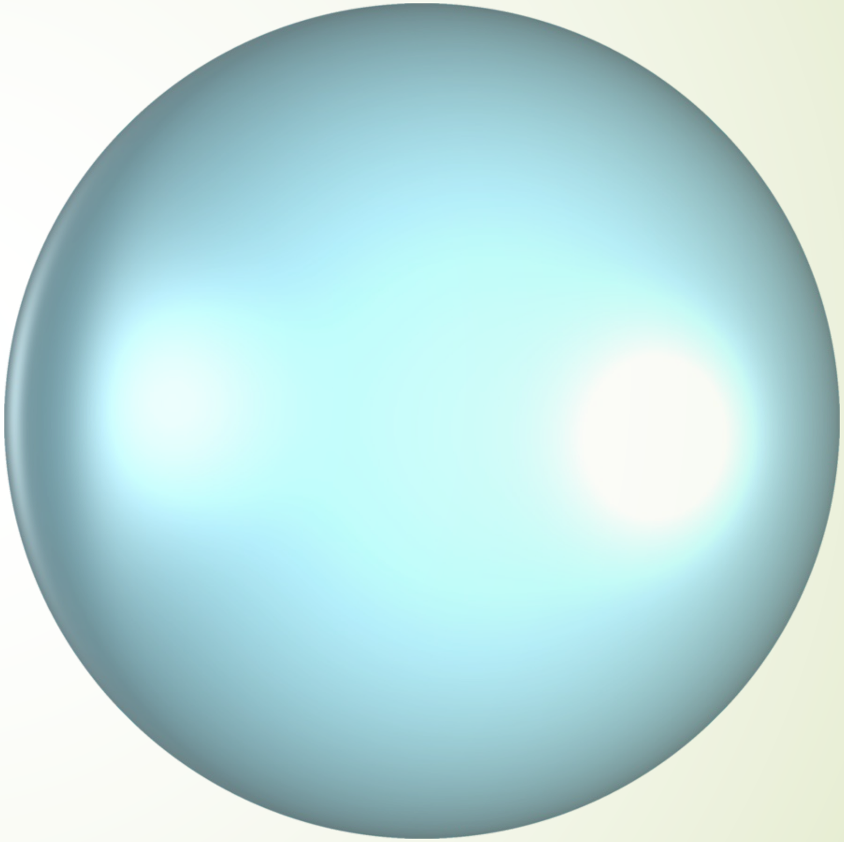
Impurity	Specification
$^{14}C/^{12}C$	~10^{-18}
K (^{40}K)	~10^{-14} g/g (~10^{-18} g/g)
^{232}Th	~10^{-16} g/g
^{238}U	~10^{-16} g/g
^{226}Ra	~3×10^{-24} g/g (0.1 µBq/m^3)
Argon (^{39}Ar)	~10^{-10} g/g (0.1 µBq/m^3)
Krypton (^{85}Kr)	~4×10^{-16} g/g (0.1 µBq/m^3)

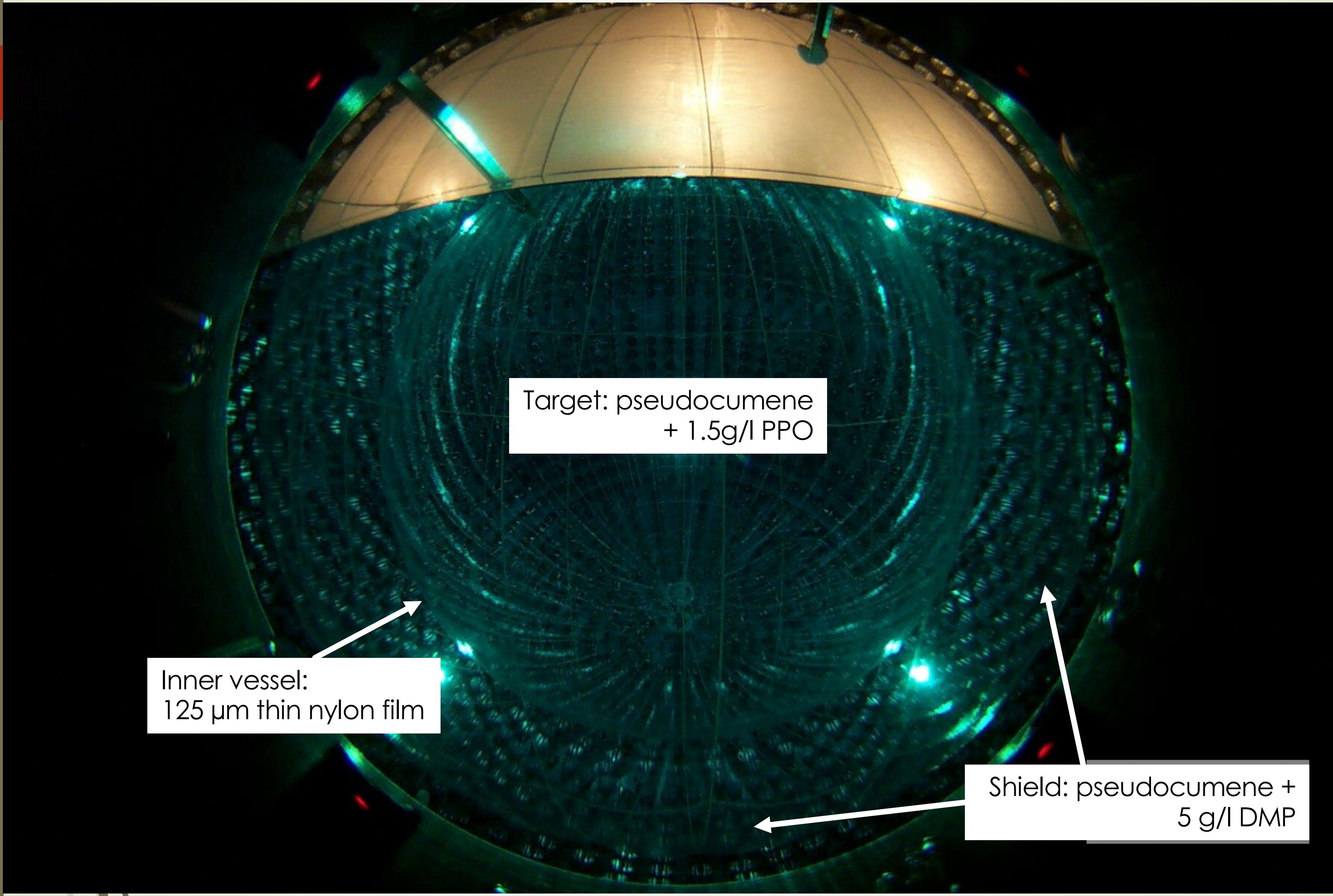
Target selection

- Target must be:
  - Large mass (~100 tons).
  - Extremely high purity.
  - High signal response.
- Solids (monocrystals) can be very pure, but small and cannot be re-purified.
- Liquid target:
  - Large mass possible.
  - High purity achievable.
  - Re-purification „easily“ possible.
- Organic liquid scintillator with high light yield: Pseudocumene with wavelength shifter PPO.

Detector design

- Ultralow-level maxim: All materials are radioactive!
- → Avoid materials as much as possible.
- Minimize surface-to-volume ratio: Sphere!
- Use low mass container: Nylon foil.
- Use same liquid as target and shield.




Target: pseudocumene
+ 1.5g/l PPO
Inner vessel:
125 µm thin nylon film
Shield: pseudocumene +
5 g/l DMP

Radioactivity in materials:

- The radioactivity activity A of all detector materials has to be measured (material screening campaign).

$$A = \lambda \times N$$

- If λ small → N is large:
  - Inductively Coupled Plasma - mass spectrometry (ICP-MS).
  - Neutron Activation Analysis (NAA).
    - Neutron capture makes λ large.

- If λ large → N is (too) small.
  - Direct radioactive counting.
  - Requires very sensitive ultralow background screening stations.

NAA and ICP-MS

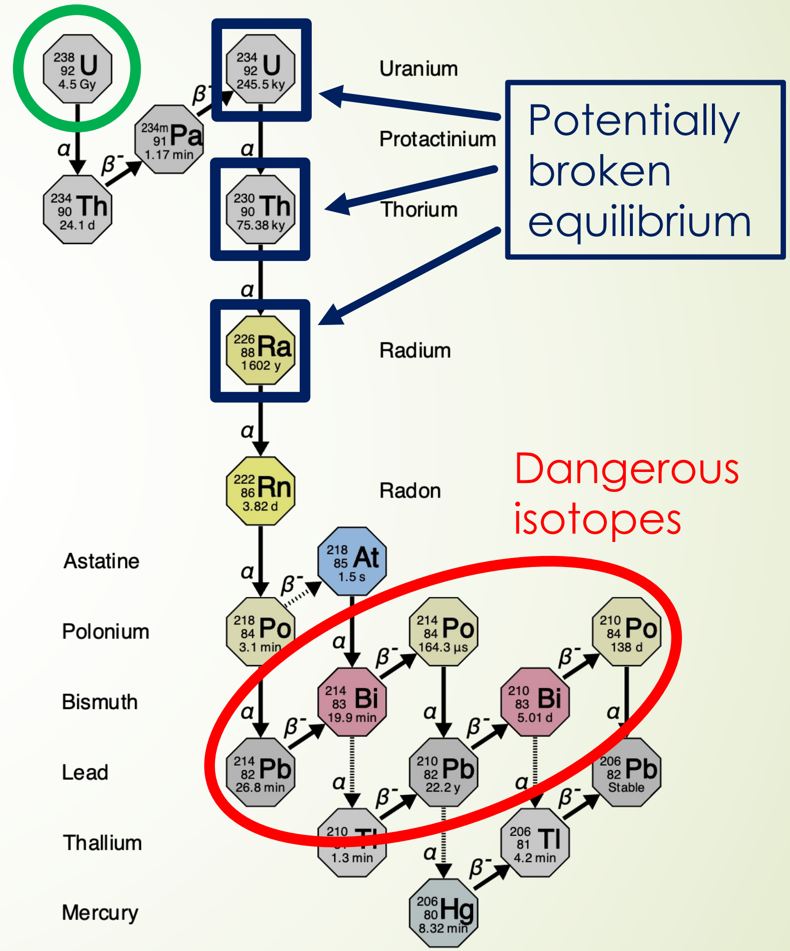
- Mostly used for screening of organic liquids, shielding water and nylon.
- Some results from *Astrop. Phys. 18 (2002) 1.*

Sample	Technique	^{238}U [g/g]	^{232}Th [g/g]	K [g/g]
Lab water	NAA	4×10^{-15}	2×10^{-15}	$< 8\times10^{-13}$
Shielding water	ICP-MS	3×10^{-14}	3×10^{-14}	1×10^{-14}
Nylon pellets	ICP-MS	5×10^{-13}	1×10^{-12}	---
Scintillator	NAA	$< 2\times10^{-16}$	$< 2\times10^{-15}$	$< 4\times10^{-12}$

- Very high sensitivity.
- Further improvement possible by chemical separation and high resolution devices.

Radioactive decay chain

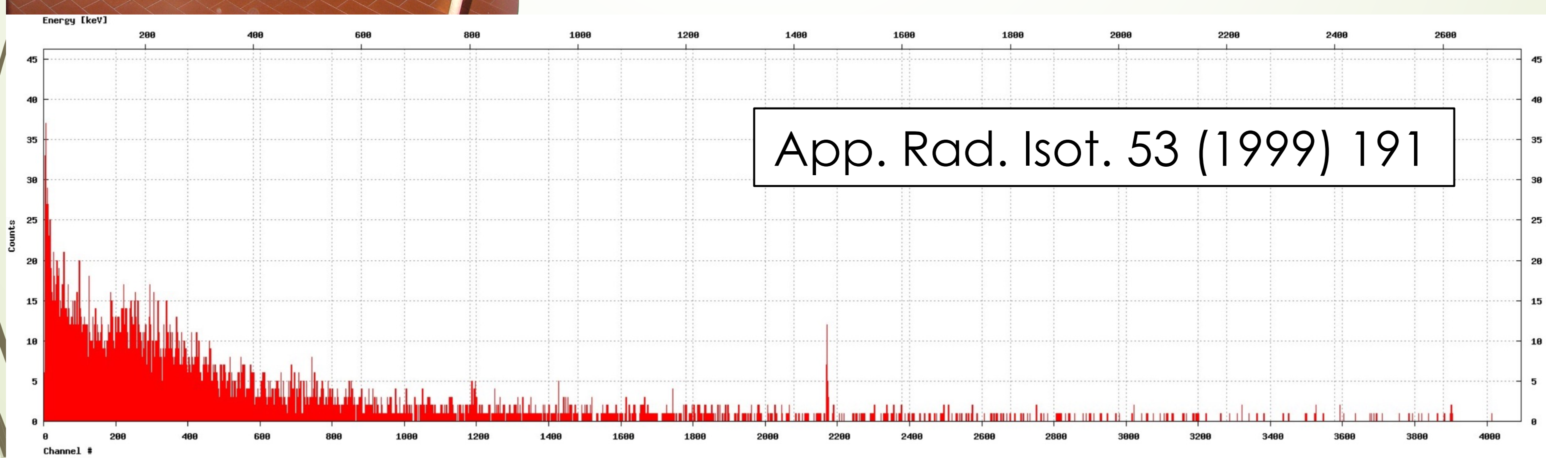
- ICP-MS and NAA measures long-lived mother isotopes.
- Materials undergo chemical processes during production.
- Secular equilibrium may be broken.
- Direct counting is crucial, even if ICP-MS / NAA suggests, that contamination level is okay.



By http://commons.wikimedia.org/wiki/User:BatesIsBack

GeMPI (1997): Germanium from MPI

- The first ultralow background HPGe spectrometer for material screening.
  - Located at LNGS.
  - Build from screened materials only.
  - Large sample chamber.
  - Air-lock, N_2-flushing.
  - Cosmic-ray exposure minimized during construction.
- Sensitivity: O(10 µBq/kg) for $^{226}Ra/^{228}Th$.

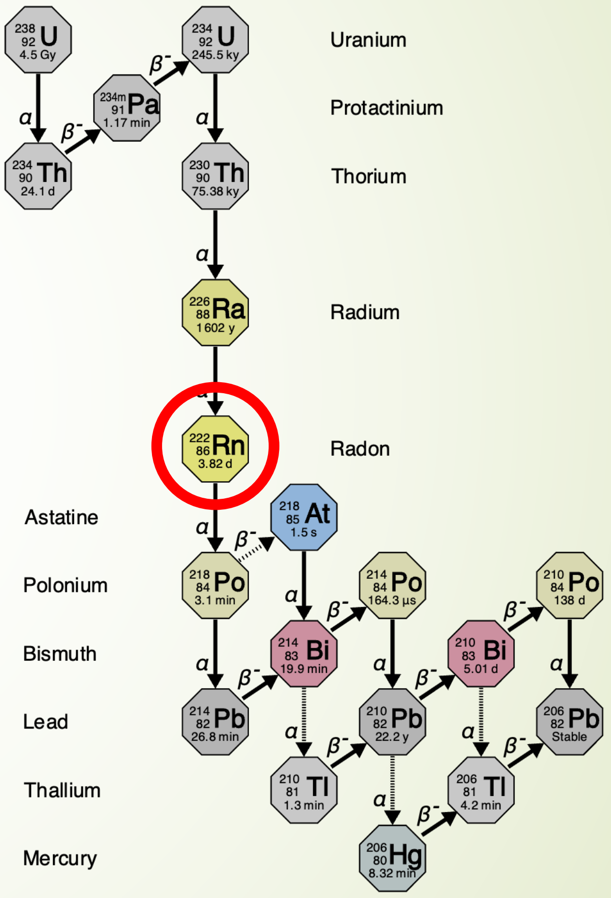


App. Rad. Isot. 53 (1999) 191

Radon: A special case!

- The only gaseous element in natural decay chains.
- Mother of all "dangerous" isotopes.
- May diffuse out of materials.
- Most dangerous is ^{222}Rn (t_H = 3.8 d).
- ^{222}Rn emanation rate NOT directly correlated to ^{226}Ra concentration.
- Complementary material screening program needed:

^{222}Rn emanation measurements with few atom sensitivity.



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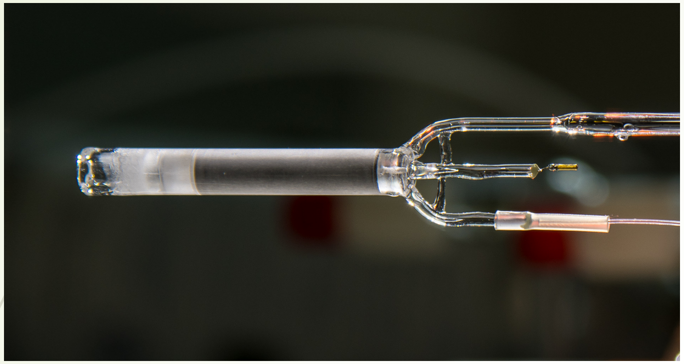
^{222}Rn screening station at MPIK



Low background proportional counter

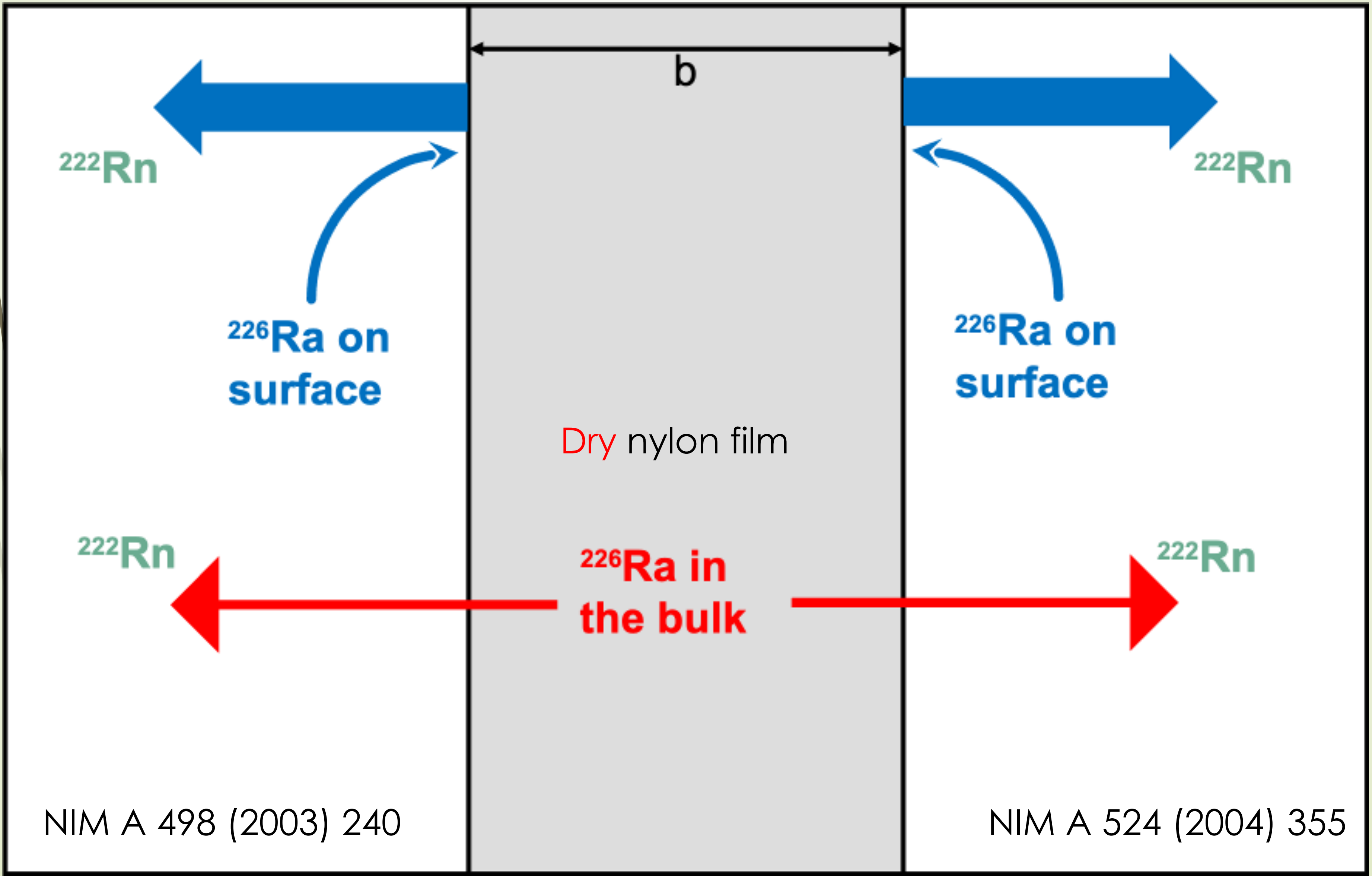
Sample purification and counter filling

Detection limit:
~20 µBq (10 ^{222}Rn atoms)

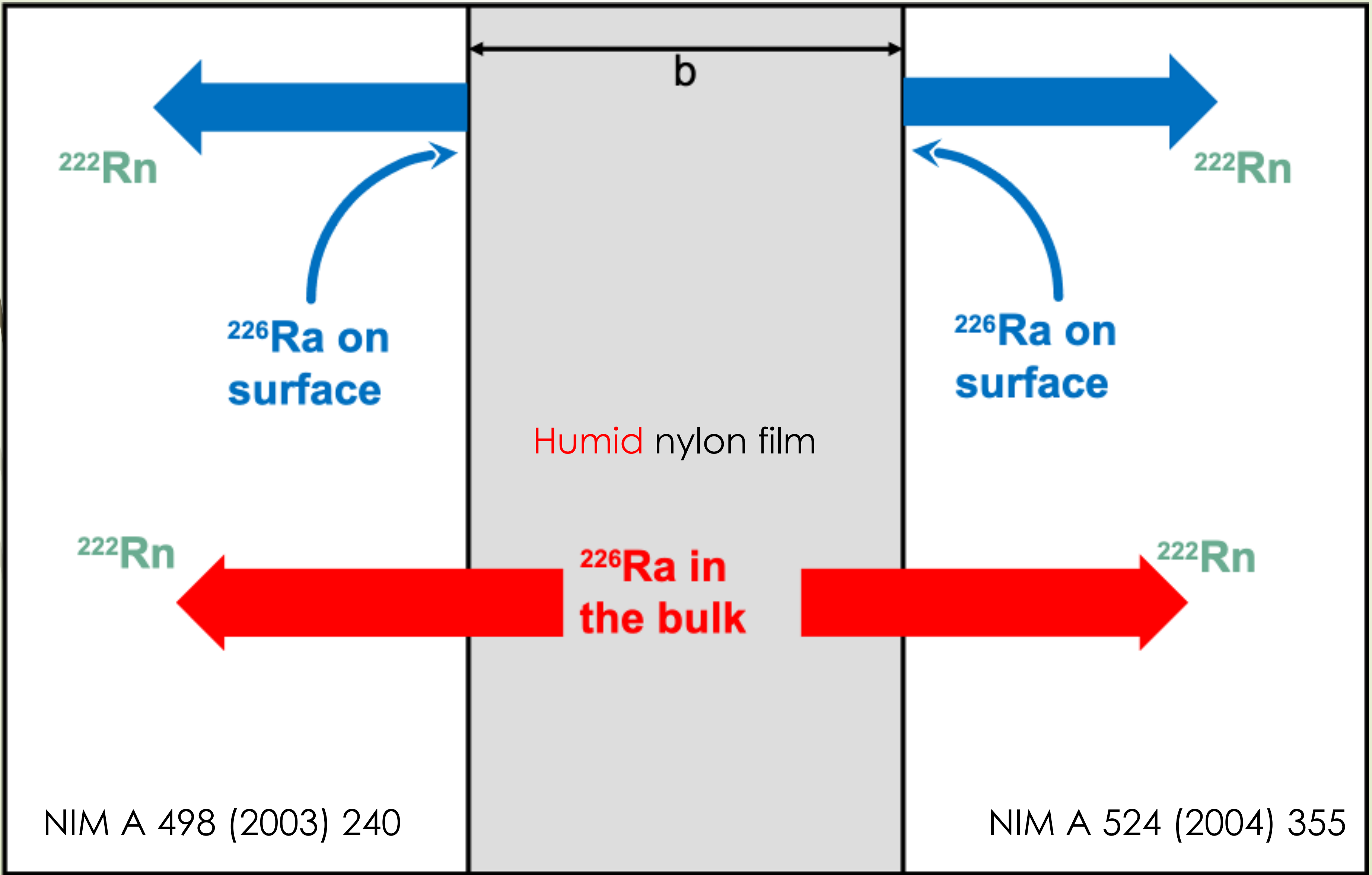
Small emanation vessel (with sample)

Large emanation vessels (up to 80 liters)

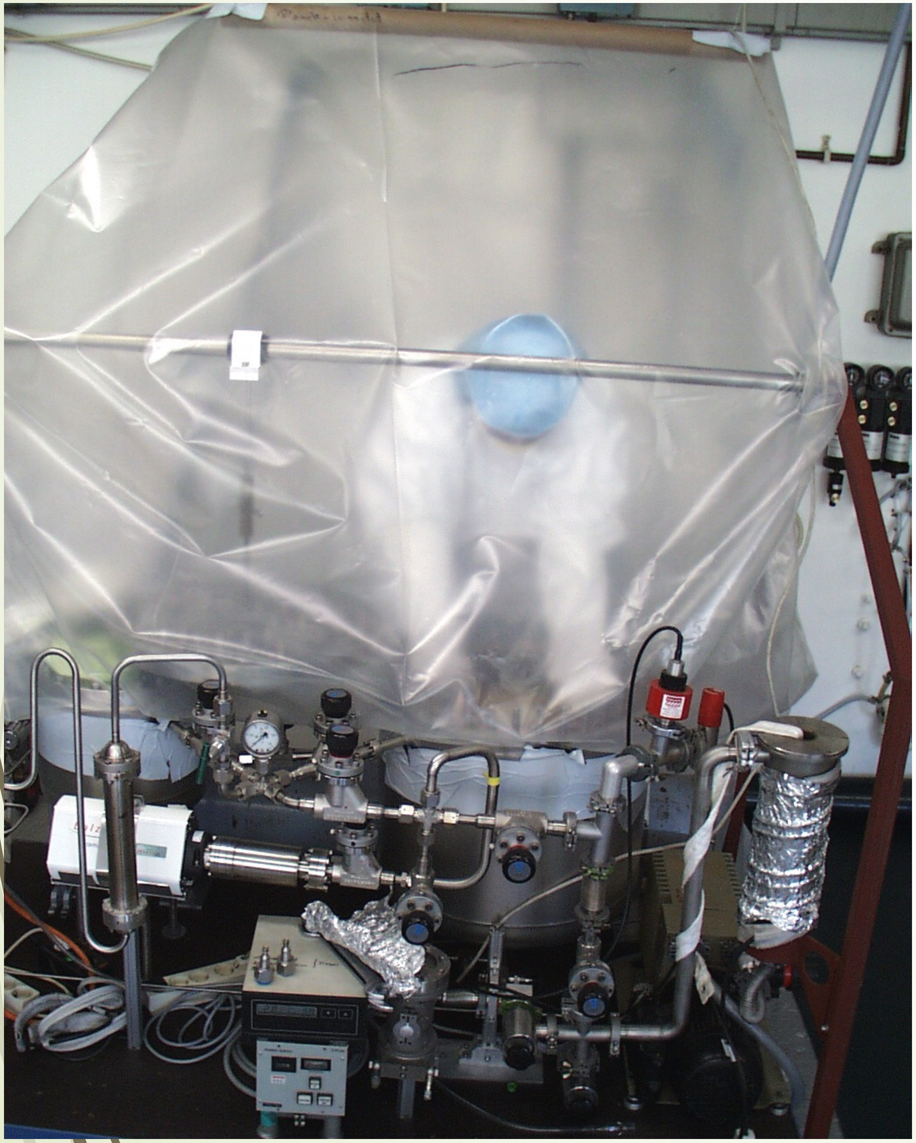
^{222}Rn emanation from nylon film



^{222}Rn emanation from nylon film



^{222}Rn emanation from nylon film



Dry and humid measurement allows to disentangle bulk and surface contamination!

A_{bulk}: <21 µBq/kg

$A_{surface}$: <0.8 µBq/m^2

NIM A 524 (2004) 355

How to demonstrate the BOREXINO feasibility?

- Expected ^{7}Be solar neutrino reaction rate:

  ~ 2×10^{-4} cts / (keV × kg × year)

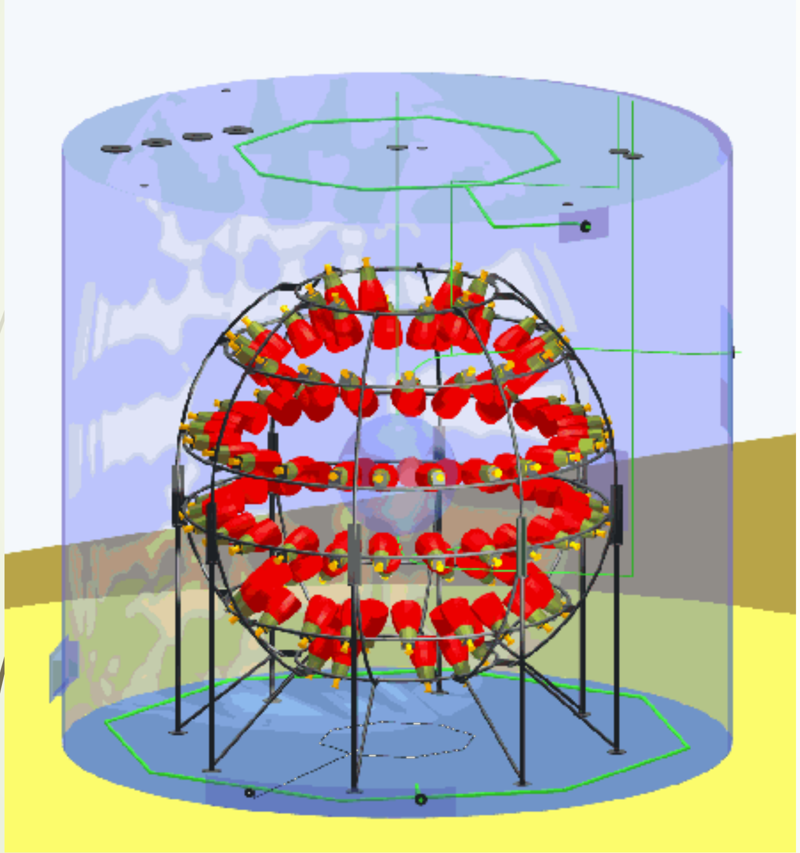
- For comparison: Background index of LEGEND-200 (Neutrino 2024 conference):

  $(5.3 \pm 2.2) \times10^{-4}$ cts / (keV × kg × year)

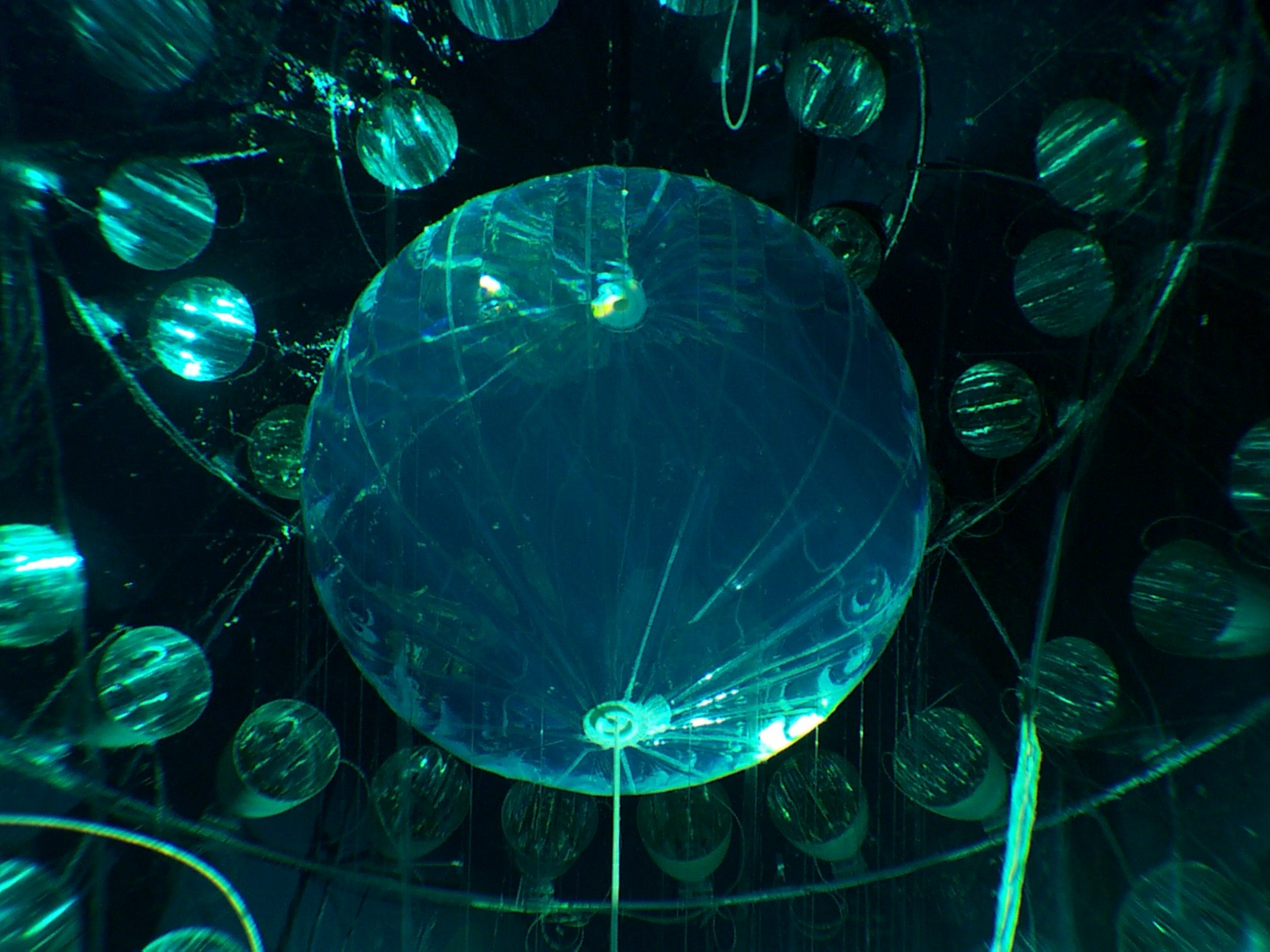
- Can U/Th/Ra/K be reduced to that level?
- Organic scintillator: Is ^{14}C sufficiently low?

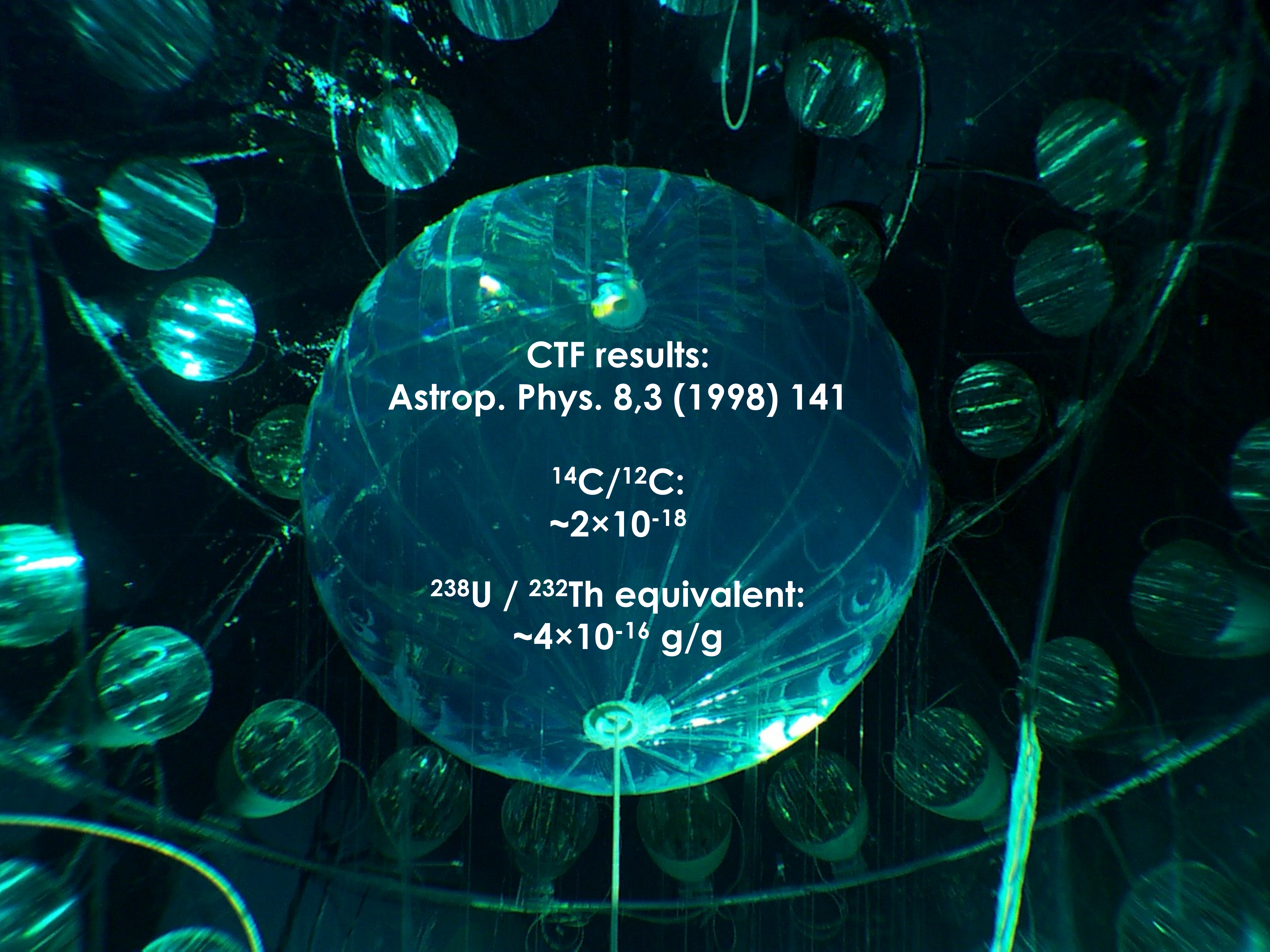
→ A BOREXINO demonstrator was needed.

CTF: Counting Test Facility

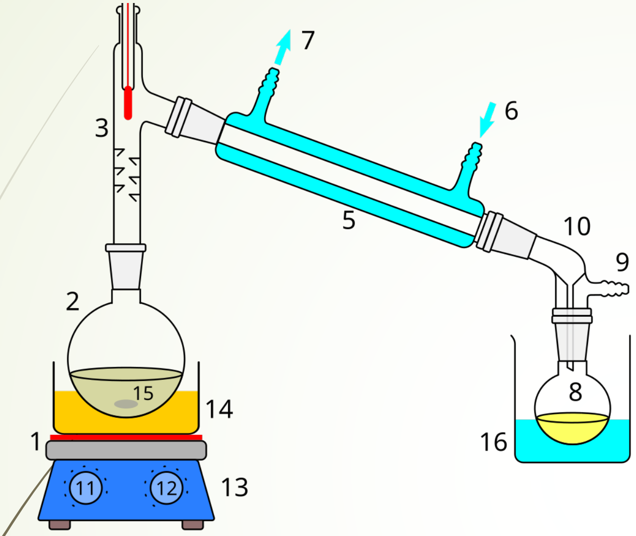


- A “small-scale” version of BOREXINO.
- ~5 tons of liquid scintillator in a nylon ballon.
- Shielded by 1000 tons of ultrapure water.
- Viewed by 100 PMTs.
- An ultralow background detector on its own.




CTF results:
Astrop. Phys. 8,3 (1998) 141
$^{14}C/^{12}C$:
$\sim 2\times 10^{-18}$
^{238}U / ^{232}Th equivalent:
$\sim 4\times 10^{-16}$ g/g

Scintillator purification



Distillation:

- Less volatile impurities remain in liquid phase.

Water extraction:

- Water (high polarity) doesn't mix with scintillator (non-polar).
- But ionic impurities are transferred into aqueous phase.

Scintillator purification

- Purification columns installed next to detector in underground laboratory.
- Filled with large surface stainless steel packing materials.
- Precision cleaned, assembled in clean-room.

- Water extraction column:
  - ^{222}Rn screening result : <0.12 mBq/packing.
  - 608 m^2 of stainless steel surface inside.
  - ^{222}Rn activity: (4.8 ± 0.7) mBq

    (8 ± 1) $\mu Bq/m^2$

J. Mod. Phys. A 29,16 (2014) 1442009

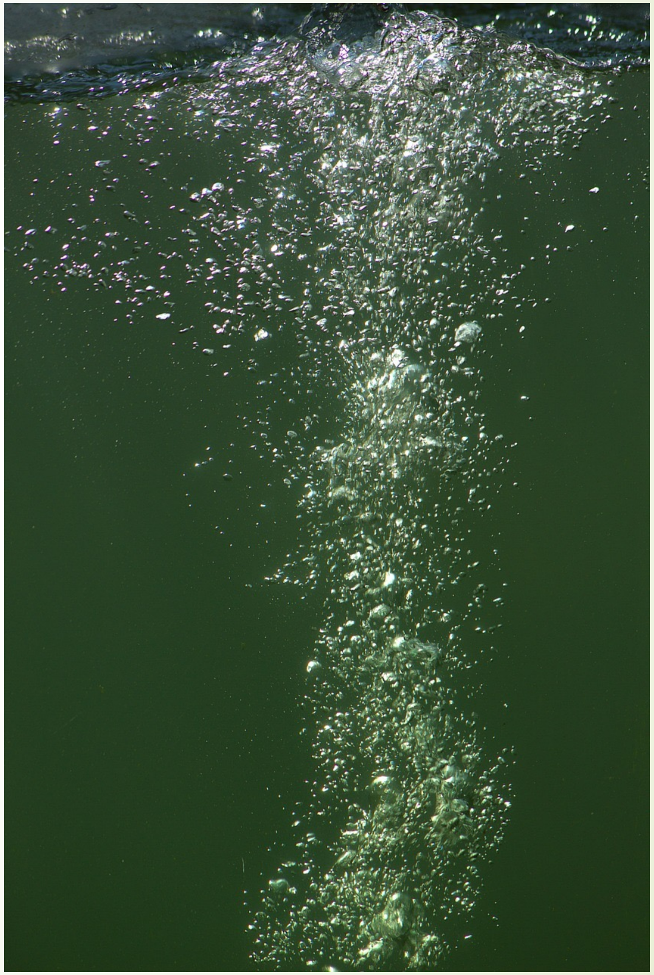
Radioactive noble gases:

- ^{222}Rn contamination was still very challenging, due to:
  - Continuous ^{222}Rn emanation from all materials.
  - Surface contamination (dust = ^{222}Rn source).
  - Good solubility of radon in organic liquid.

- There are other radioactive noble gases:
  - Cosmogenic ^{39}Ar: 14 mBq/m^3 in air.
  - Anthropogenic ^{85}Kr: ~1.5 Bq/m^3 in air.

Dissolved radioactive noble gases were the remaining challenge for BOREXINO.

Removal of gaseous impurities from liquids



- Sparging with pure nitrogen carries away dissolved impurities.
- But relatively high solubility of noble gases in organic liquid.
- Large amount ($O(100 m^2/h)$) of ultrapure N_2 needed:
  - ^{222}Rn: <7 µBq/m^3
  - Kr: <140 ppq

^{222}Rn-free High purity N_2 (HPN_2)

- Purification by cryo-trapping of radon on high-purity (!) activated carbon.
- Direct liquid nitrogen purification is convenient (although less efficient).
- Measurement of ^{222}Rn concentration by collection from N_2 and counting with proportional counters:

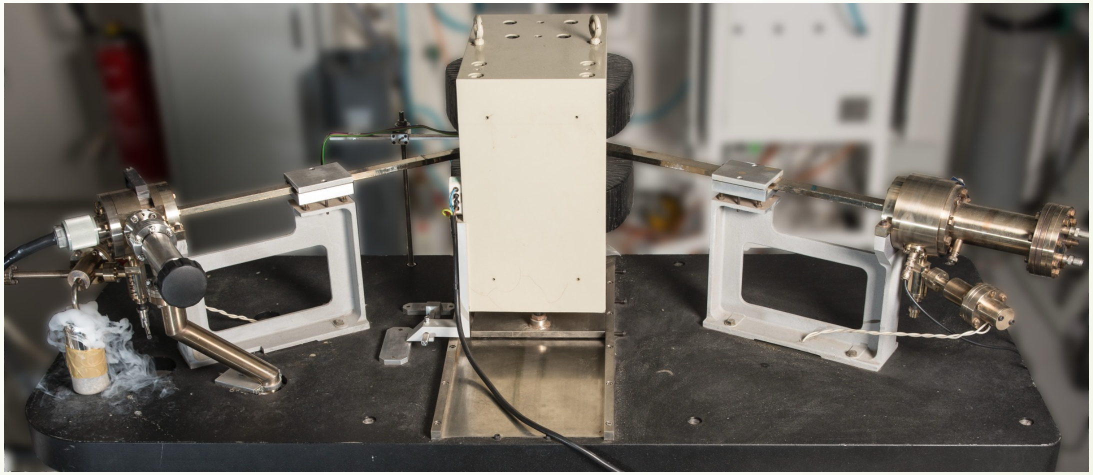
  <0.5 μBq/m^3 (STP)

- Production plant for continuous supply of HPN_2 at 100 m^3/h flow-rate.
- HPN_2 is particularly useful for continuous sparging of stored scintillator:
  - No ^{210}Pb introduction

App. Rad. Isot. 52 (2000) 691

Low Ar-Kr-Nitrogen (LAKN$_2$): How to tackle the Kr problem?

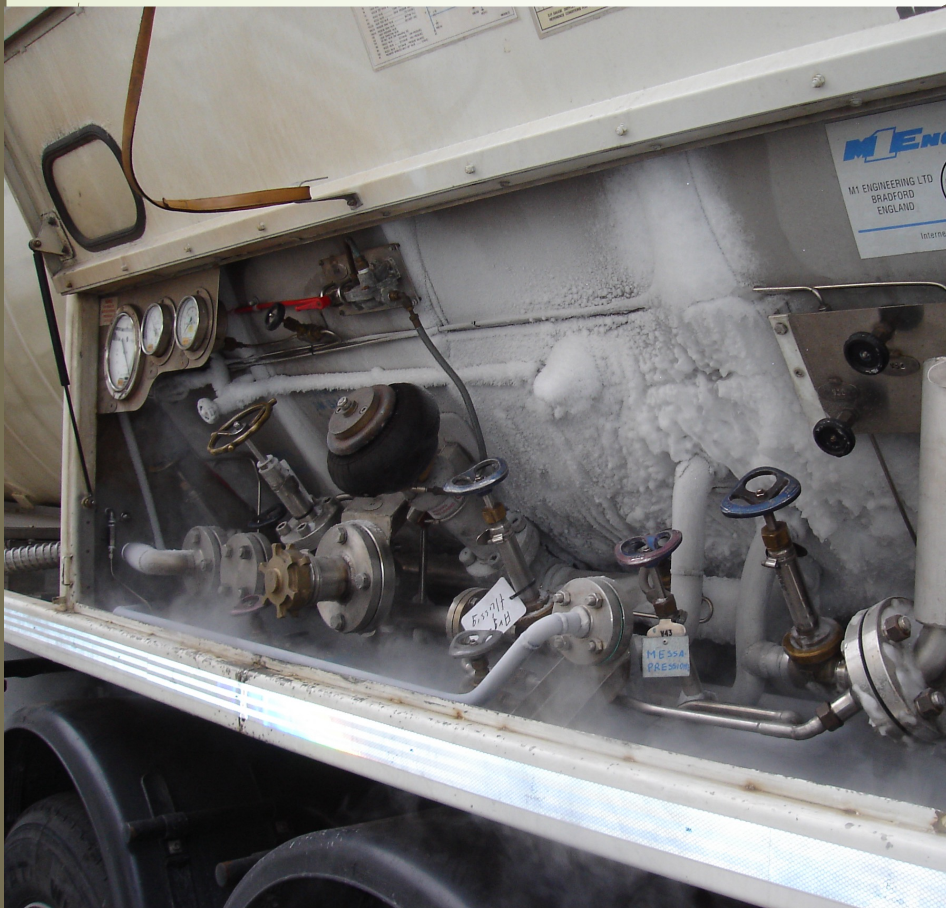
- Cryo-trapping? → Feasible, but difficult and expensive.
- Screening of N_2 from various European companies with RGMS: Rare Gas Mass Spectrometer (MPIK).



- Produced N_2 is pure enough (Kr <100 ppq), but re-contamination during transport and filling.

$LAKN_2$-tank from SOL company: Test of full delivery chain

LAKN$_2$-tank from SOL c… y: Test of full deliver…



Strict filling protocol to maintain ultra-high purity.

$LAKN_2$-tank from SOL company: Test of full delivery chain

$LAKN_2$-tank from SOL company: Test of full delivery chain

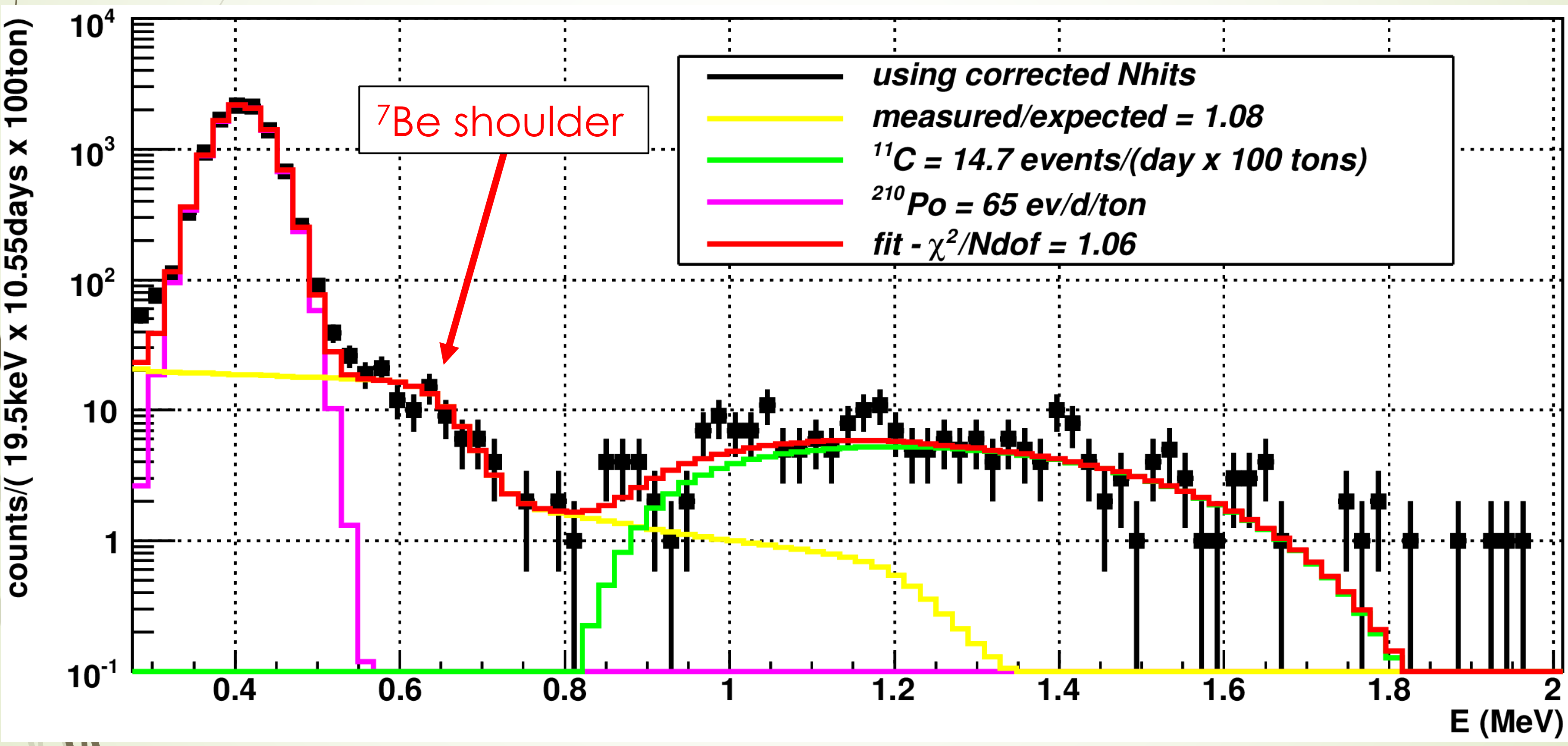
Results of delivery chain test:

Ar: (12.2 ± 1.7) ppb

Kr: (0.05 ± 0.01) ppt

Initial ^{222}Rn: (0.7 ± 0.2) $\mu Bq/m^3$

First spectrum (1 month of data)



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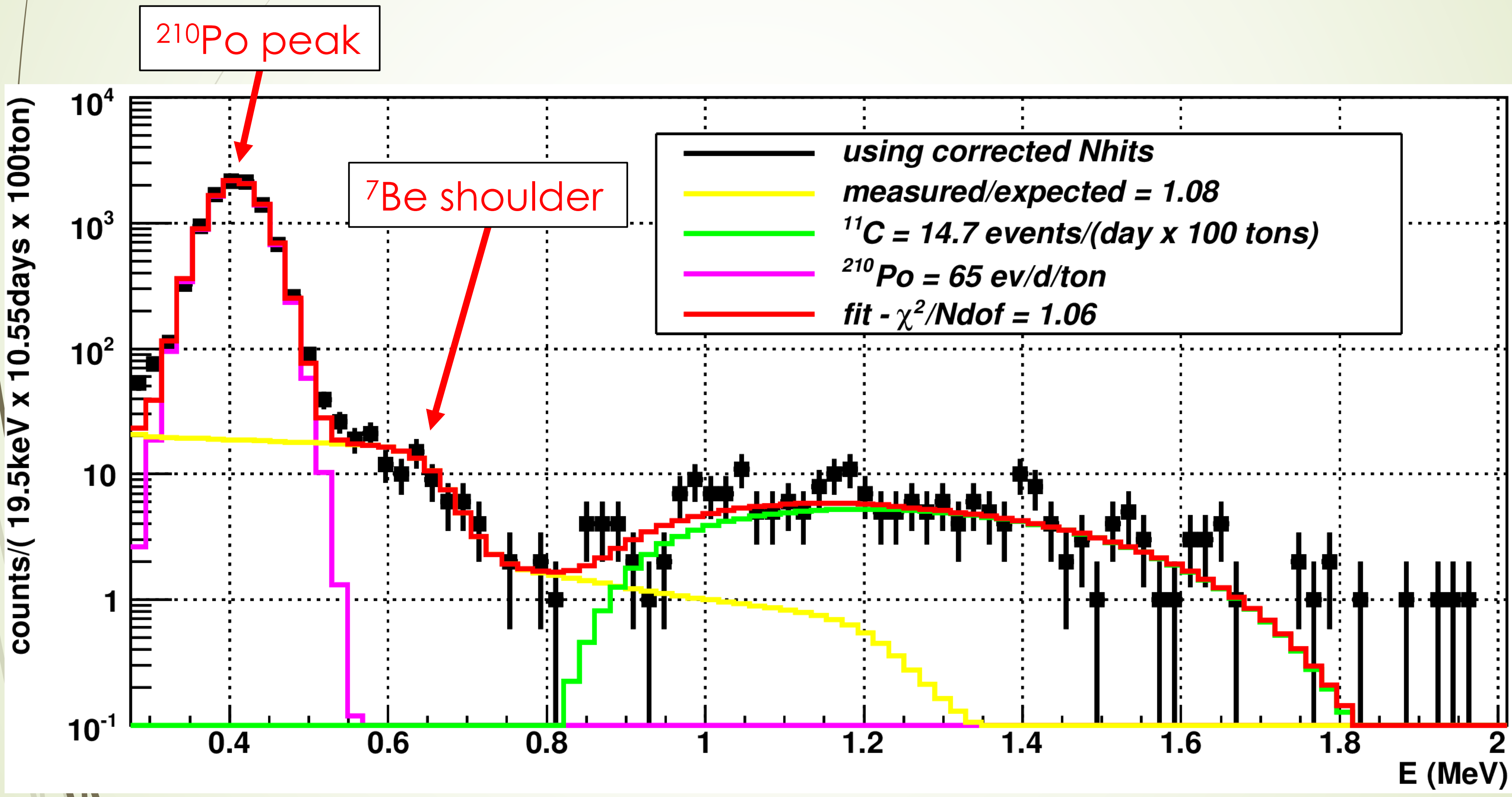
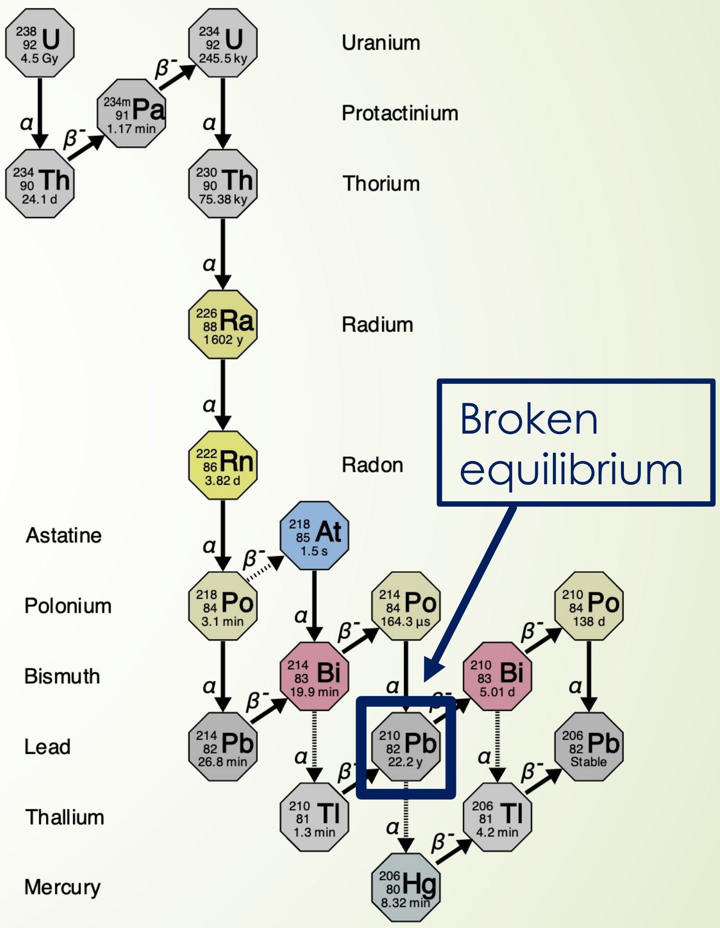


Long-lived ^{222}Rn daughters

- Any internal ^{226}Ra decay produces a ^{210}Pb atom (t_H = 22 years).
- More relevant: Accumulation of ^{210}Pb from ambient ^{222}Rn (particulates) during air exposure.
- ^{210}Po dynamics: Relatively long half-life and particular chemistry of polonium (different from Pb).



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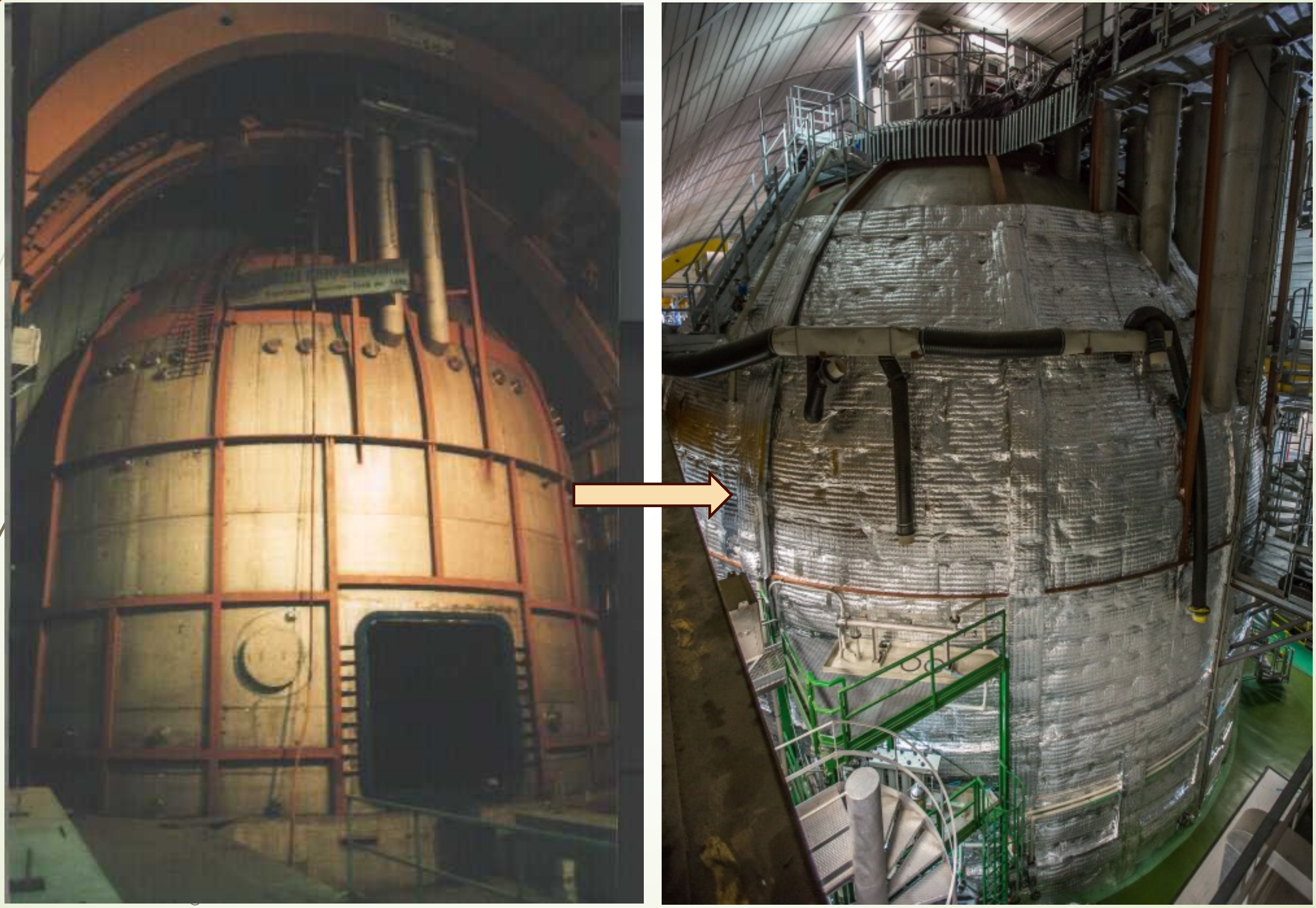
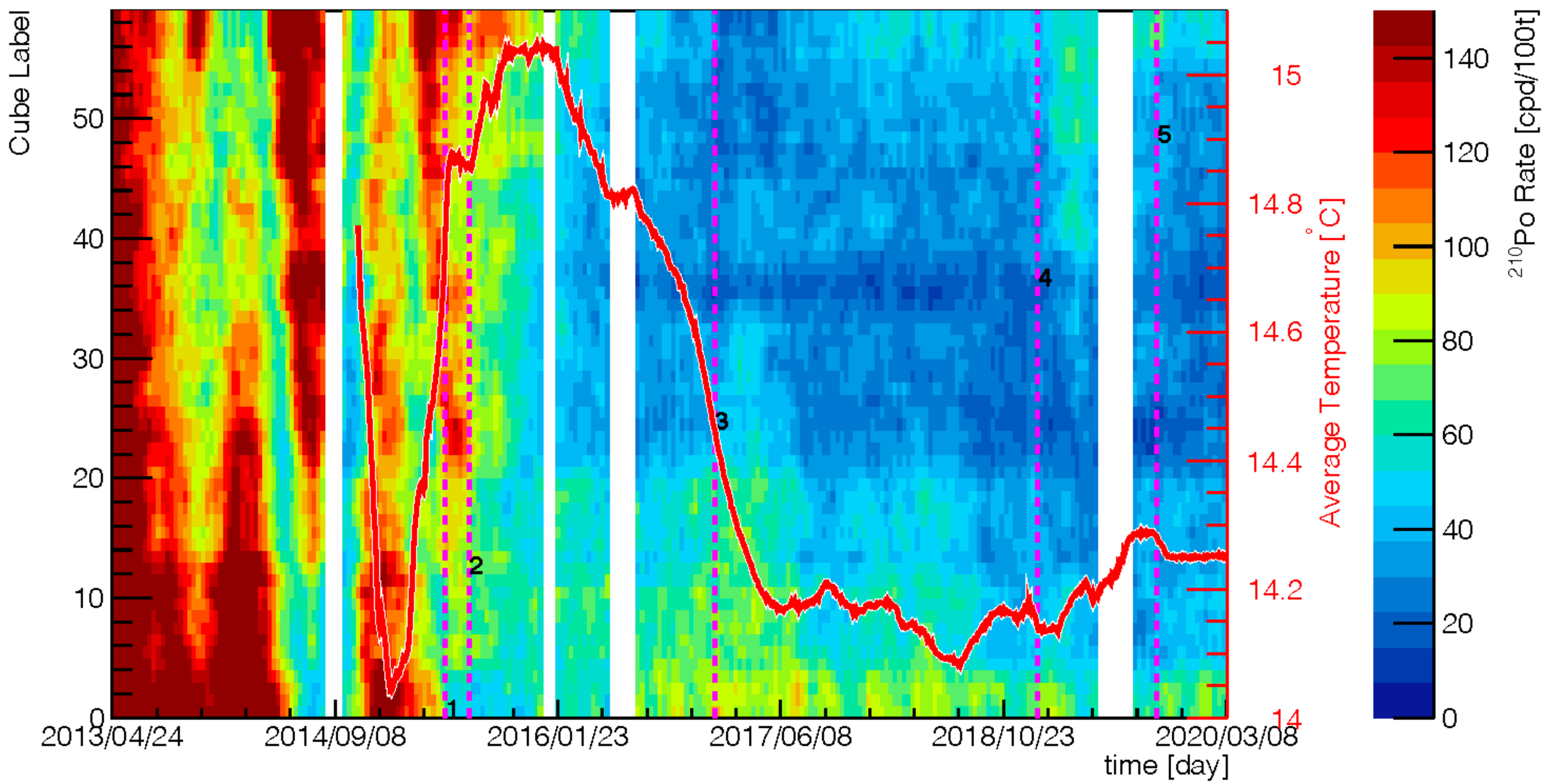
Thermal stabilization



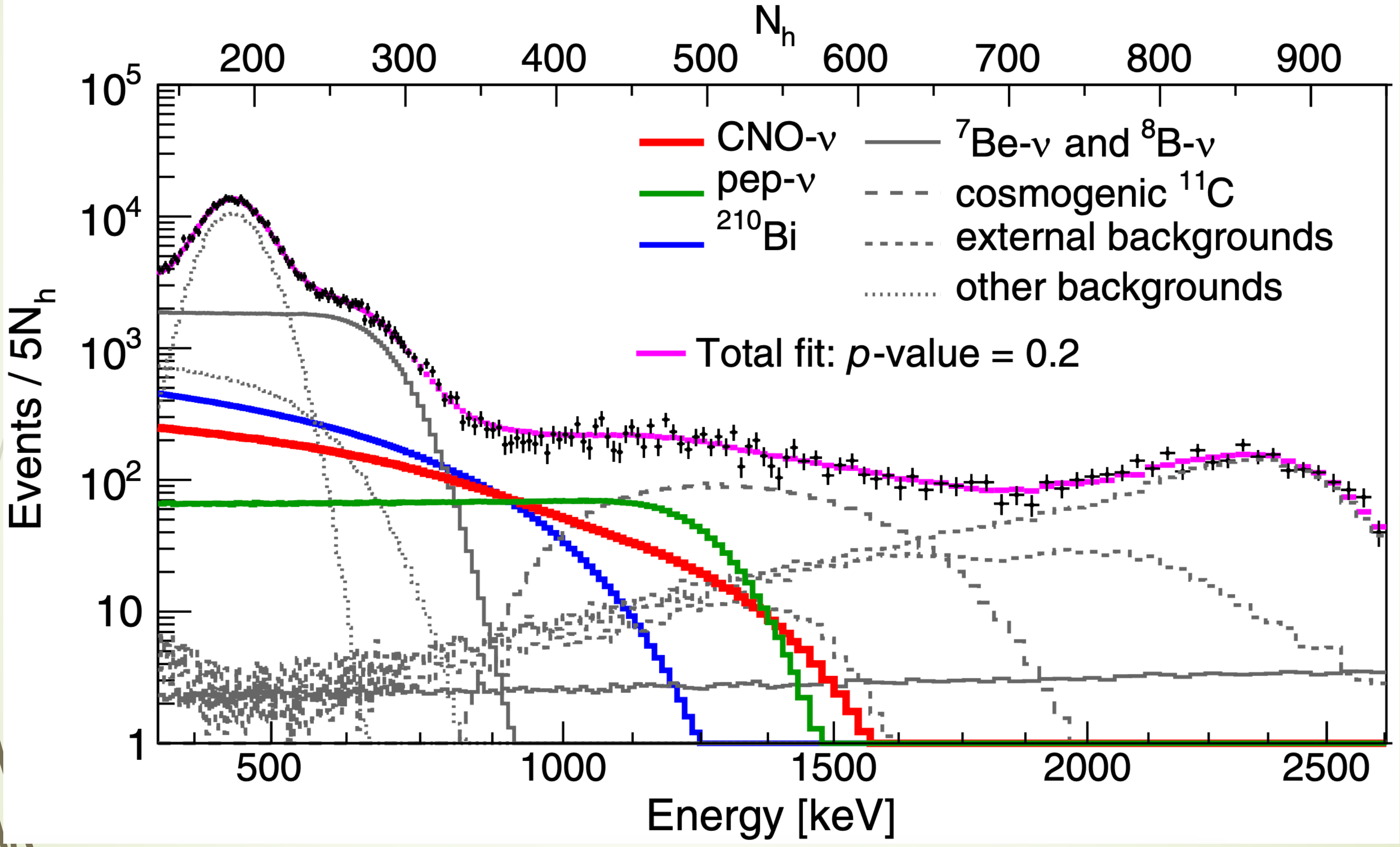
Thermal insulation and temperature monitoring + control: *NIM A 885 (2018) 38.*

Thermal stabilization



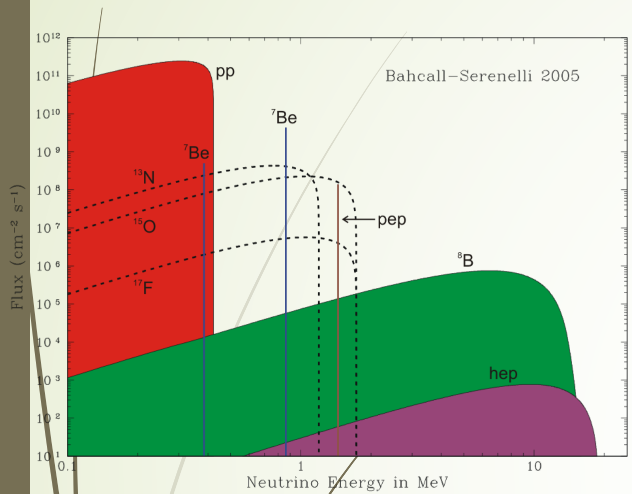
Nature 587 (2020) 577

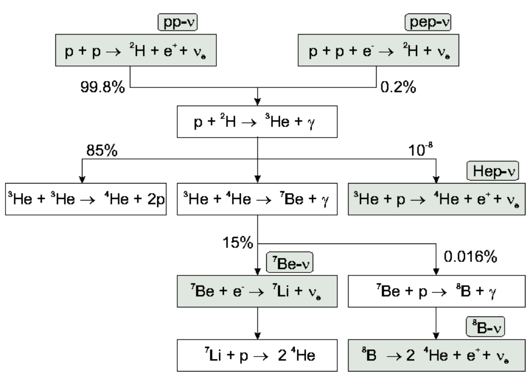
Detection of CNO neutrinos

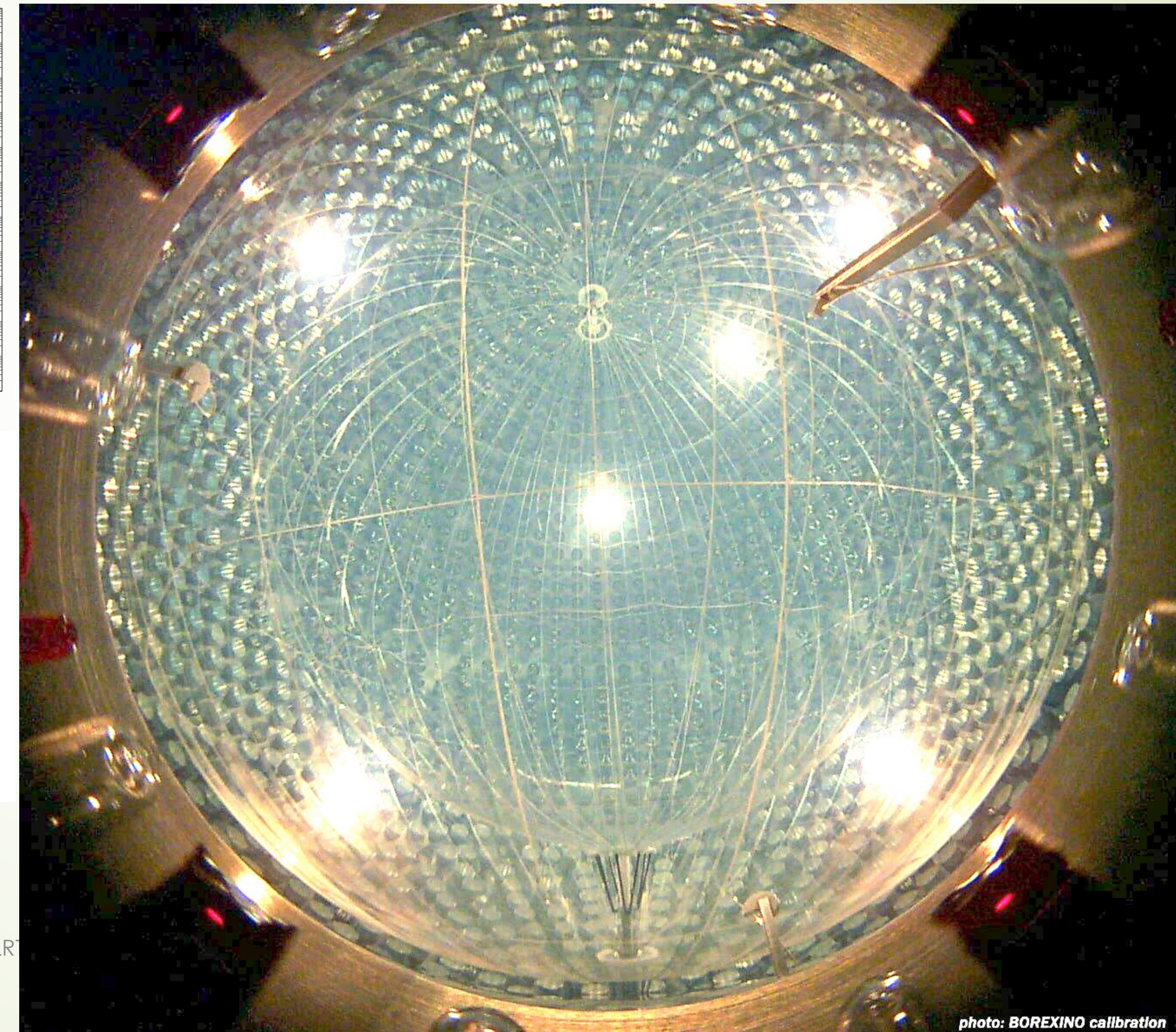


PRD 108, 102005 (2023)

BOREXINO purity







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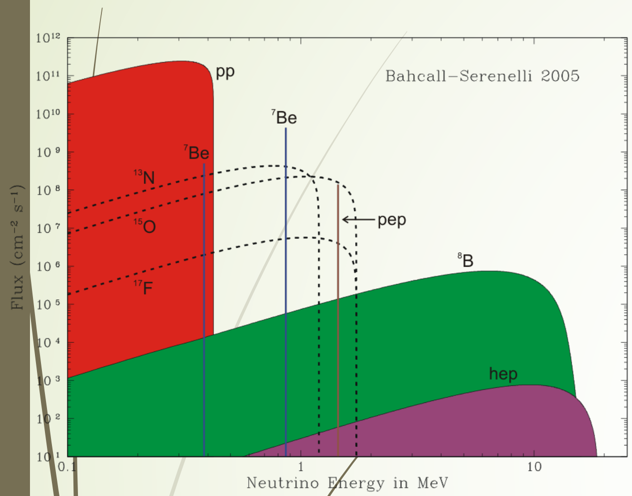

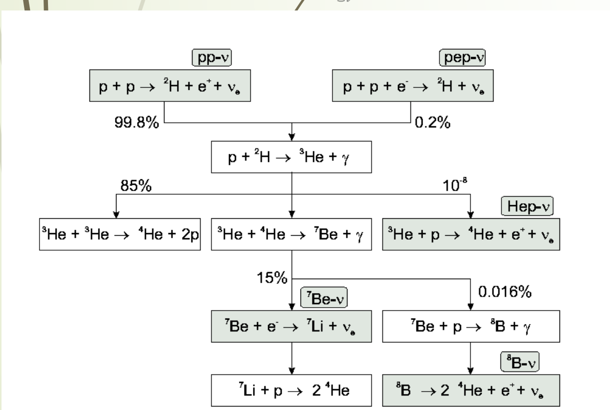

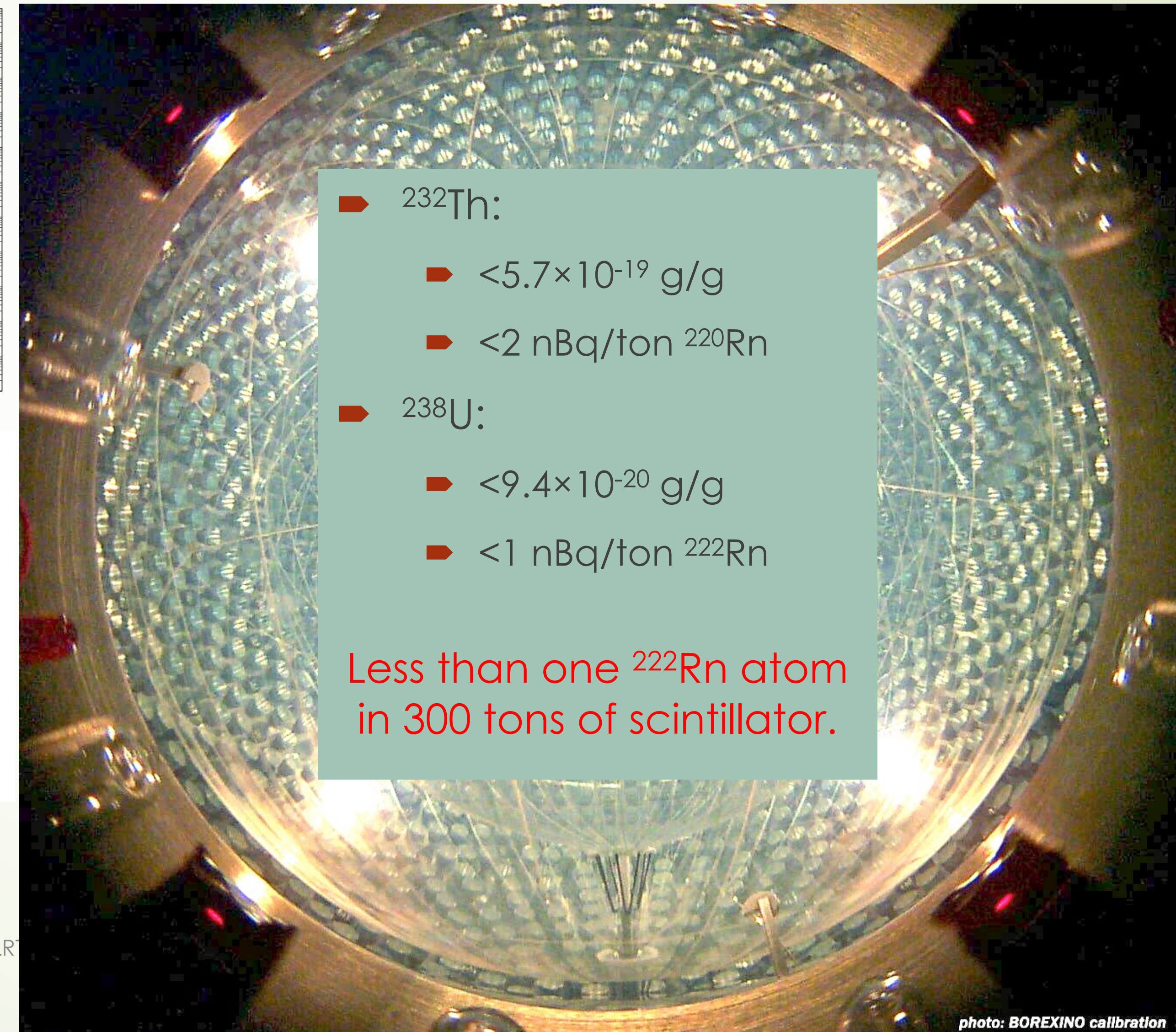

- ^{232}Th:
  - $<5.7\times10^{-19}$ g/g
  - <2 nBq/ton ^{220}Rn
- ^{238}U:
  - $<9.4\times10^{-20}$ g/g
  - <1 nBq/ton ^{222}Rn

Less than one ^{222}Rn atom in 300 tons of scintillator.

Summary

- BOREXINO was a pinoeering experiment with respect to ultralow background techniques.
- Developed methods are very useful for future experiments.
- Achieved radiopurity is still world leading.

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Let's beat BOREXINO and go beyond the limits!