

Purification of Liquid Scintillators for Low Radioactivity

Frank Calaprice
Princeton University
Borexino Experiment

Liquid Scintillator Radioactivity

- Extreme scintillator radio-purity is essential for solar neutrino measurements.
- Initial Borexino data on ^7Be , ^8B , and pep solar neutrino data relied on process equipment developed in the late 1990's.
 - Distillation
 - Water extraction
 - Nitrogen stripping
- Better accuracy on ^7Be , ^8B , and pep solar neutrinos and possible measurement of pp and CNO neutrinos motivate a second phase of Borexino with lower backgrounds.
 - A new campaign of re-purification to achieve lower backgrounds for a second phase of Borexino started in 2010.

The Borexino Detector

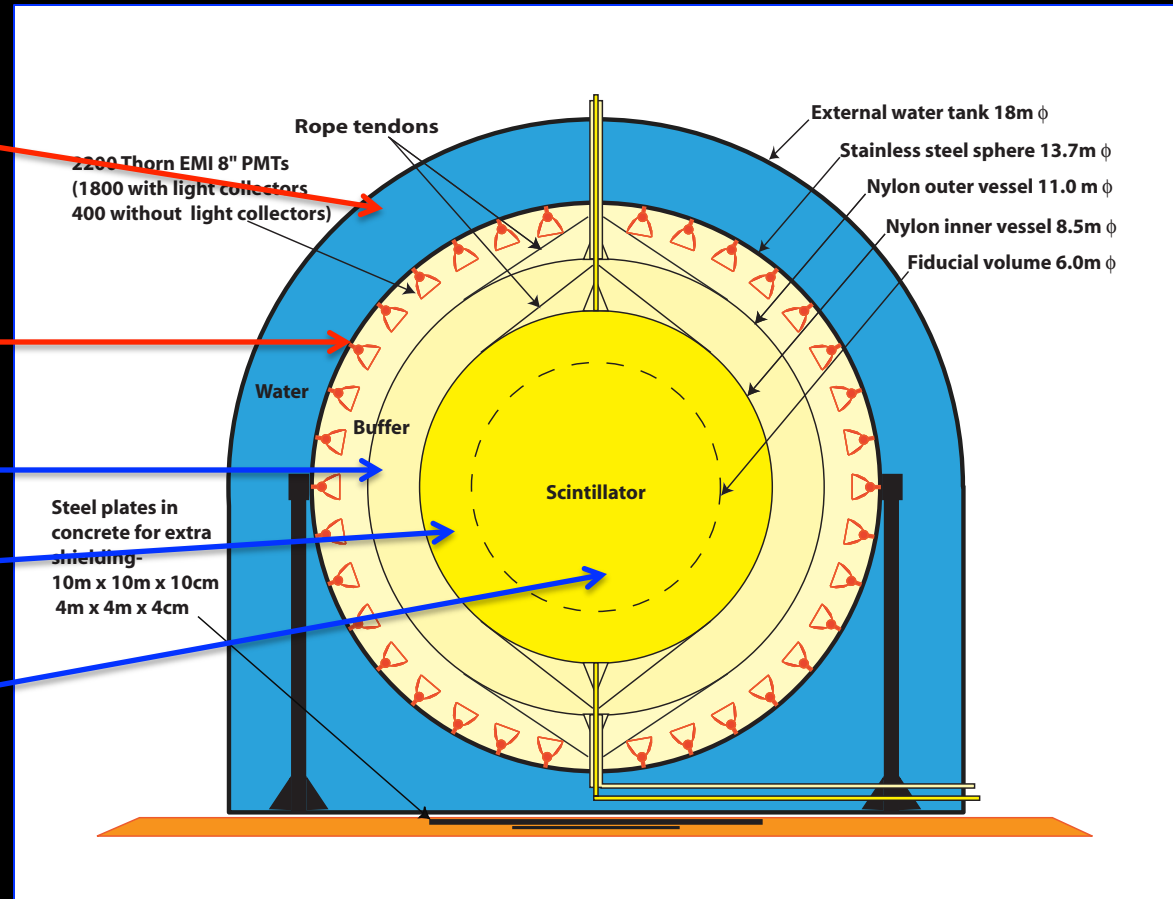
Outer Water
Detector

Inner Detector

Buffer

Scintillator

Fiducial Volume
(100 tons)



Borexino Filling Strategy

- Vessels inflated with high purity synthetic air.
- Vessels filled with de-ionized water.
- Scintillator filled with scintillator from top, while draining water from bottom.

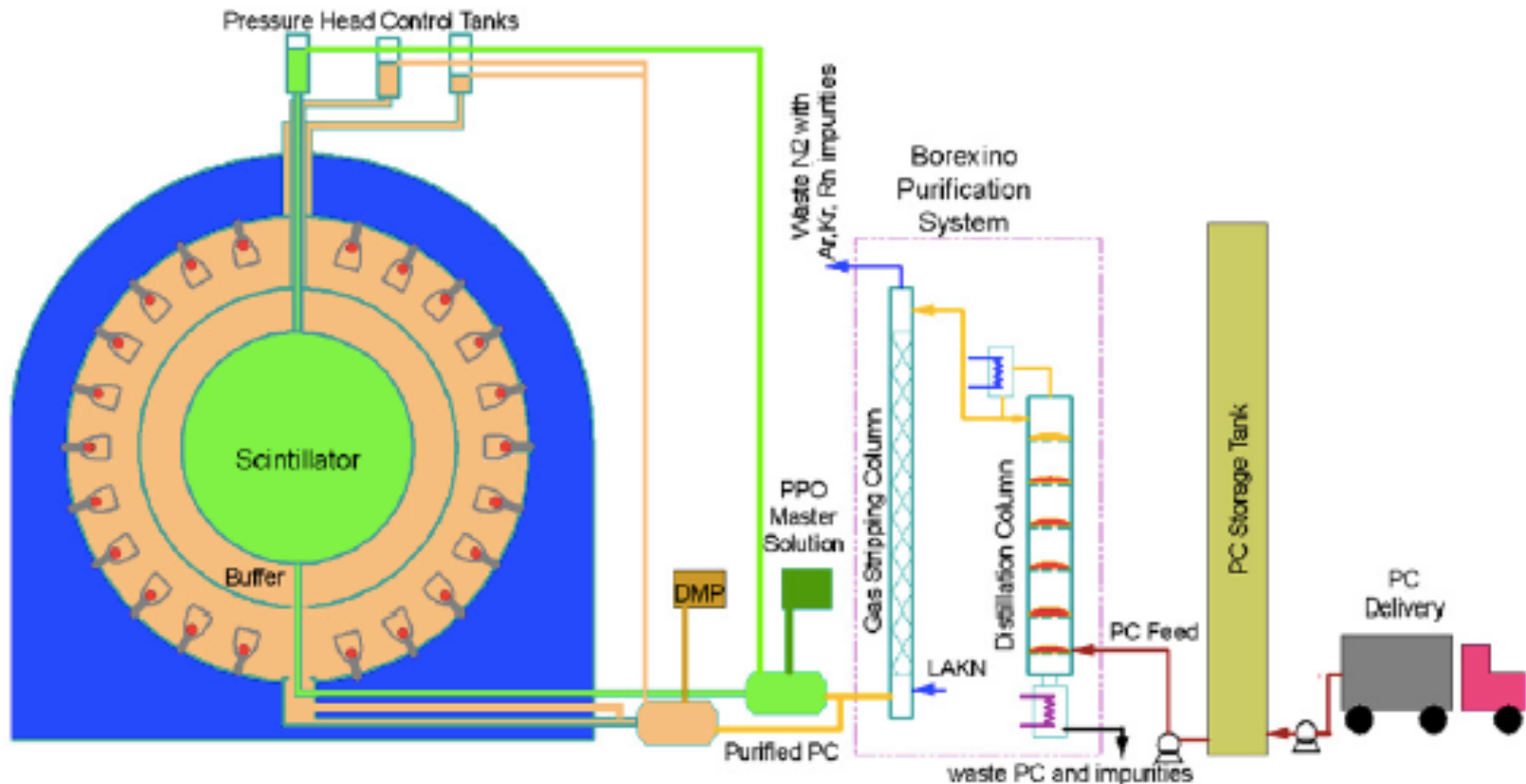
Purification Procedure for Borexino Scintillator (PC+1.5 g/l PPO)

- PPO wavelength shifter was purified by preparing a concentrated “master solution” of PPO dissolved in pseudocumene (PC).
 - The master solution was purified by a combination of water extraction followed by distillation.
- PC was shipped to temporary storage vessels, then distilled and mixed with PPO master solution while filling the vessels.

Borexino Purification System

Initial Filling detector

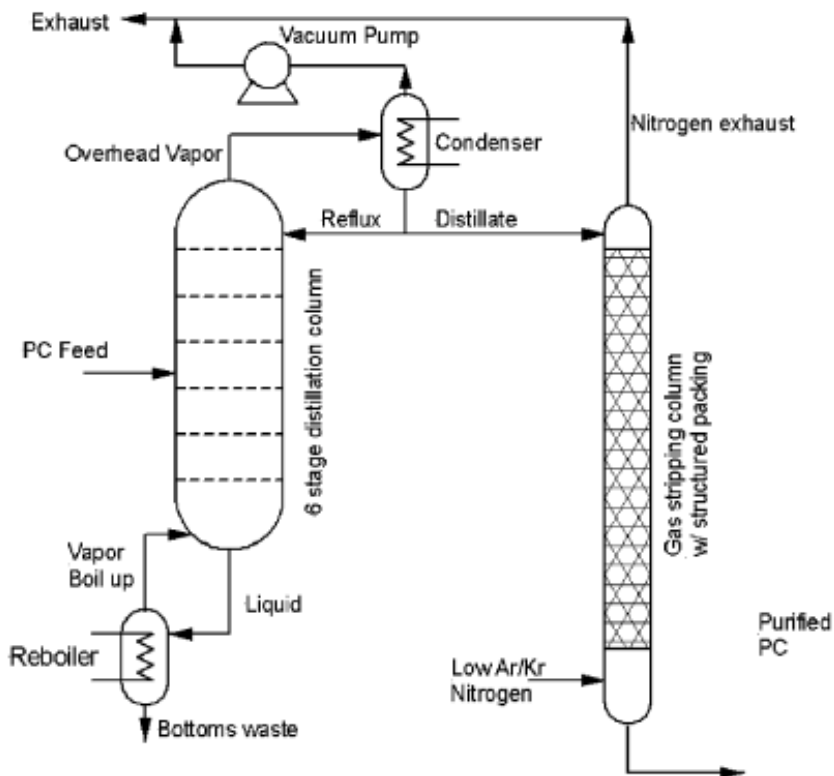
J. Benziger et al. / Nuclear Instruments and Methods in Physics Research A 587 (2008) 277–291



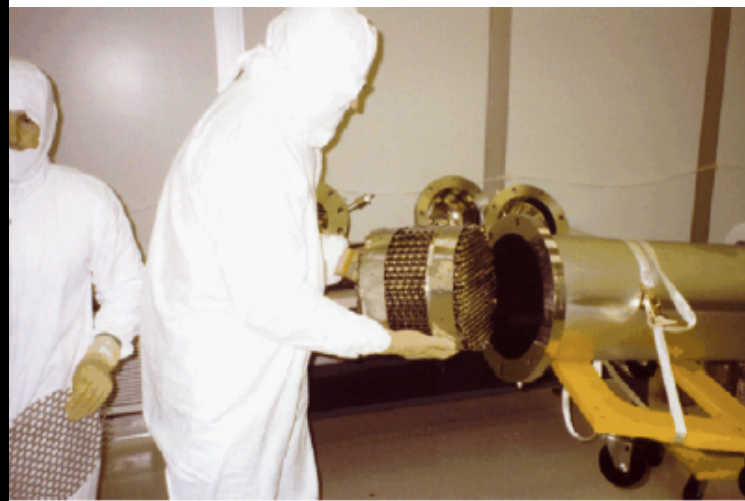
Scintillator Purification Plants

- Distillation, water extraction, and nitrogen stripping of PC at 1 m³/hr
- Distillation of concentrated PPO+PC in CTF purification plant at 20 liters/hr
- Low-Argon-Krypton (LAK) N₂ gas for stripping.

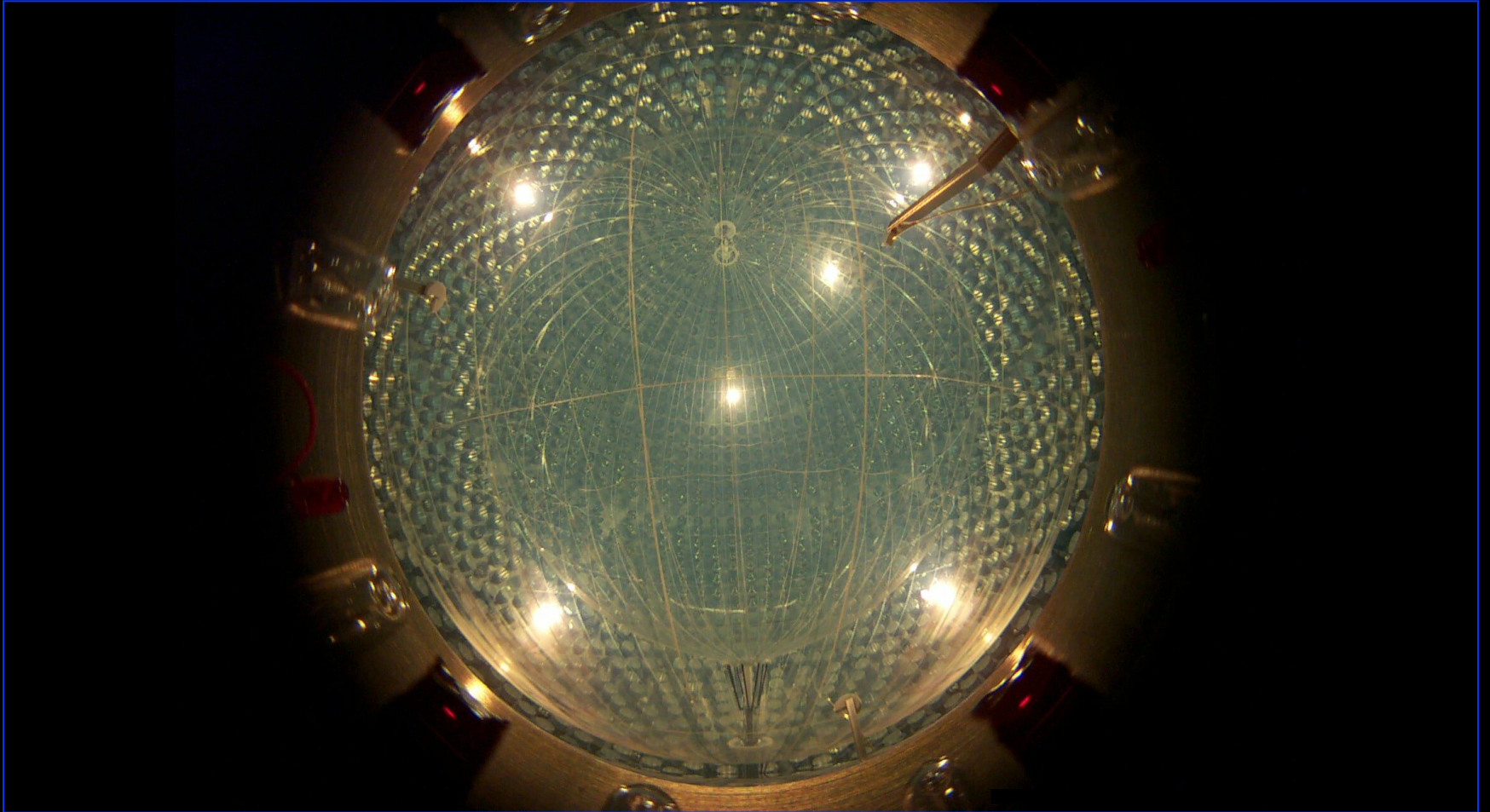
6-tray distillation column and gas stripping column with packing.



Assembly was done in Princeton radon-suppressed cleanroom.



Borexino Filled 2007



^7Be neutrino & background rates in BX1 (2007) data

- ^7Be neutrino rate: $47 \pm 7 \pm 12$ cpd/100 t (25%)
- ^{85}Kr beta decay: $27 \pm 7 \pm 5$ cpd/100t
 - Introduced in air leak during filling operation
- ^{210}Bi beta decay : $15 \pm 4 \pm 5$ cpd/100t
 - Daughter of 22-yr ^{210}Pb
- ^{210}Po alpha decay: ~ 8000 cpd/100t !!!
 - 5.3 MeV (quenched to ~ 400 keV)
 - Seriously out of secular equilibrium with ^{210}Pb - ^{210}Bi
 - ~ 800 times higher than ^{210}Bi .
 - Major background with unknown origin.
 - New results may offer partial explanation.

^7Be neutrino and background rates in BX2 (2008)

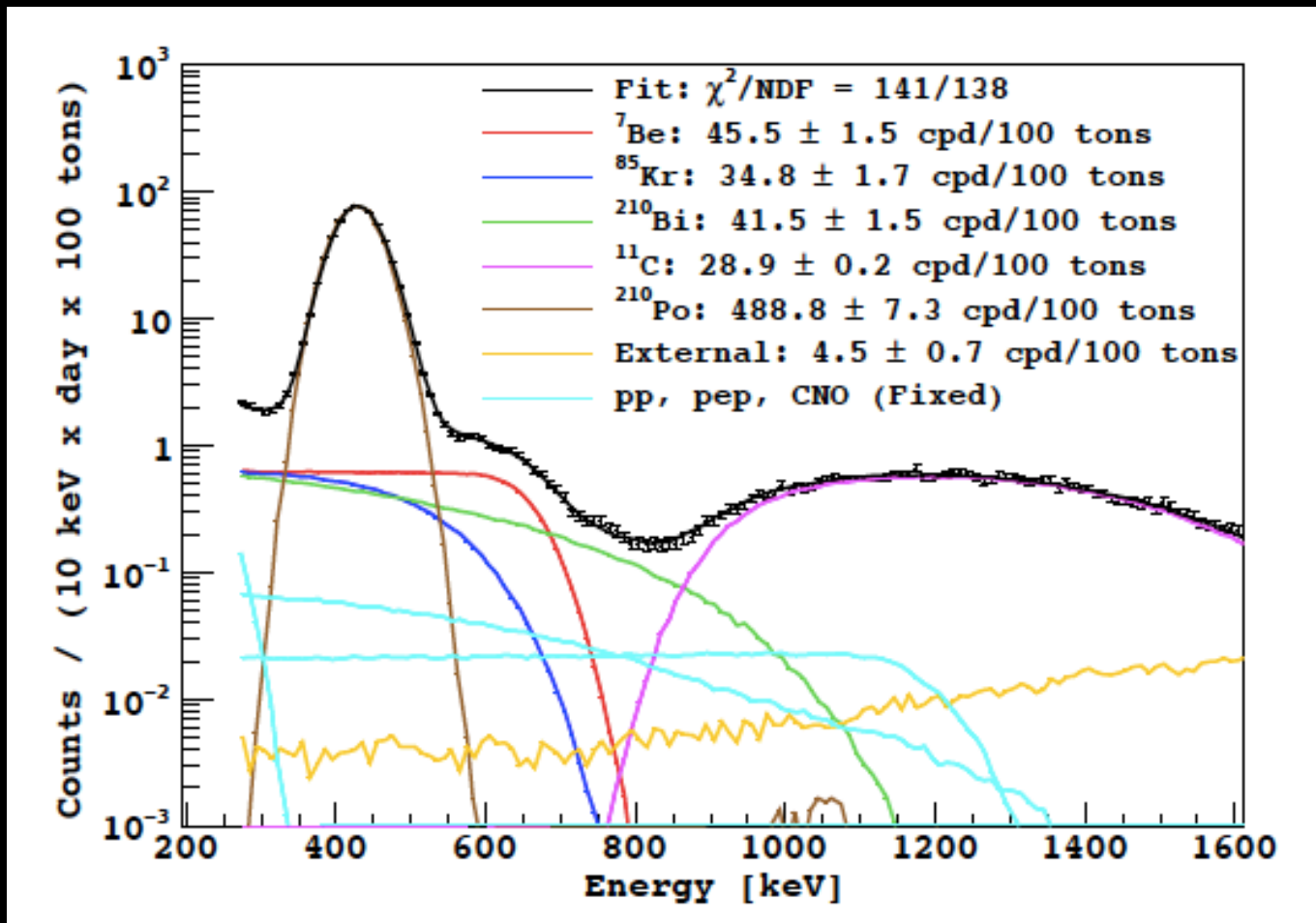
- ^7Be neutrino rate: $49 \pm 3 \pm 4$ cpd/100 t (10%)
- ^{85}Kr beta decay: 25 ± 3 cpd/100t
- ^{238}U chain (Bi-Po): $1.6 \pm 0.1 \times 10^{-17}$ g/g
- ^{232}Th chain (Bi-Po) $6.8 \pm 1.5 \times 10^{-18}$ g/g
- ^{210}Bi beta decay : 23 ± 2 cpd/100t
 - Rate has increased; reasons not completely clear.
- ^{210}Po alpha decay:
 - Rate decreasing with half-life of 138 d.

^7Be neutrino and background rates in BX3 (2011)

- ^7Be neutrino rate: $46 \pm 1.5 \pm 1.6$ cpd/100t (5%)
- ^{85}Kr beta decay: $28.0 \pm 2.1 \pm 4.7$ cpd/100t
- ^{210}Bi beta decay : $40.3 \pm 1.5 \pm 2.3$ cpd/100t
 - Rate increased due to scintillator operations, and other (?) effects.
- ^{210}Po alpha decay: ~ 500 cpd/100t
 - Decreasing with expected half-life.

Borexino Energy Spectrum (BX3)

Phys. Rev. Letts. 107 141302 (2011)



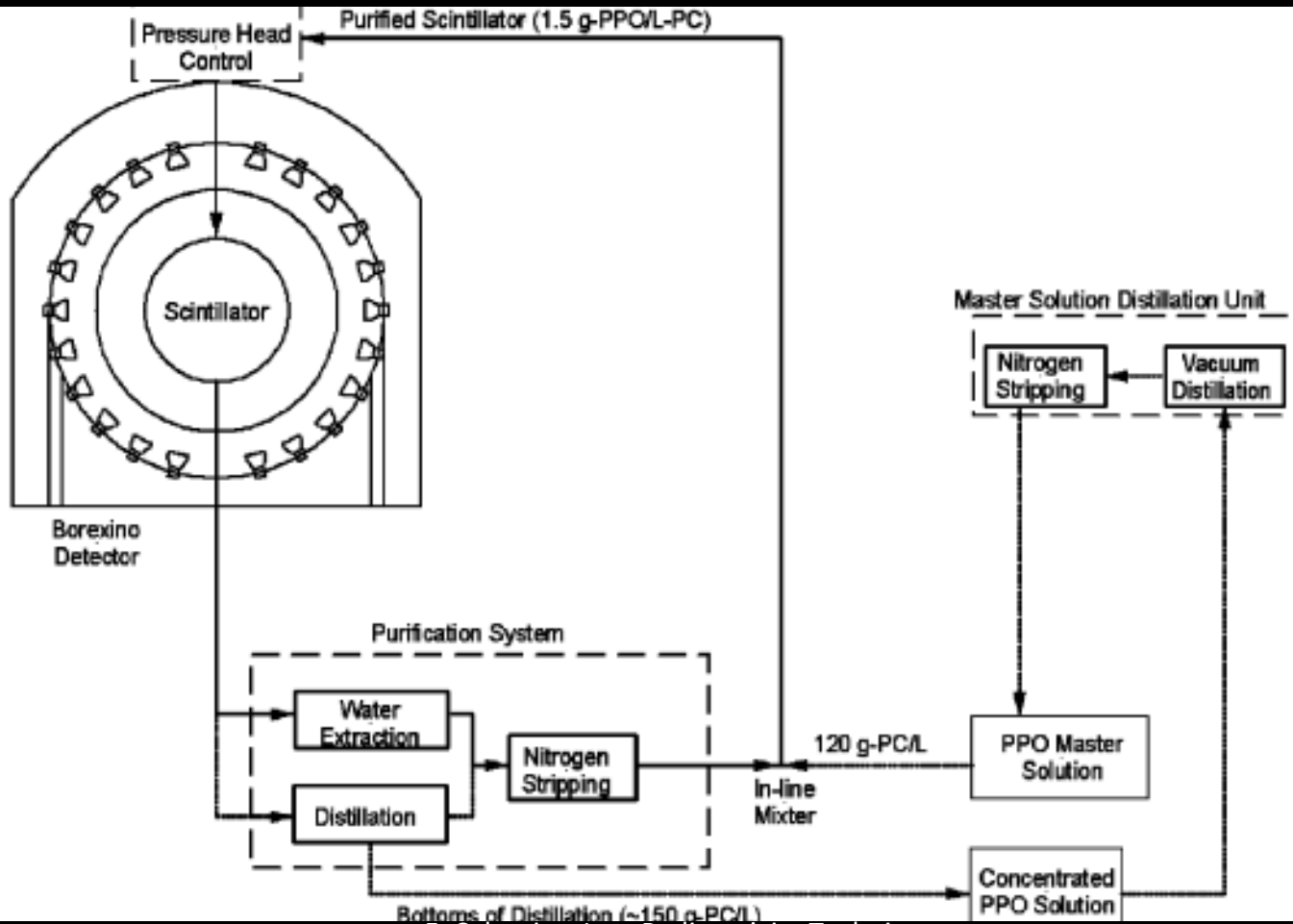
Re-Purification of BX Scintillator

2010-2011

- From 2007 to 2010 the ^{210}Bi background increased
 - Causes: Scintillator refill operations to cope with vessel leak. Other?
 - ^{210}Bi rate: 15 \rightarrow 70 cpd/100t
 - ^{85}Kr rate: 30 cpd/100t (constant)
- To reduce background, scintillator was re-purified using two processes:
 - Water extraction to remove ^{210}Pb (^{210}Bi)
 - Nitrogen stripping to remove ^{85}Kr and other volatiles.
 - Distillation was deferred to develop a new distillation system for PPO.
- Six purification operations were done, each of ~ 1 month duration.
 - Each operation processed all the scintillator in the detector once (320 m^3).
 - Data were acquired to evaluate backgrounds after each operation.
 - Not all six operations were successful.

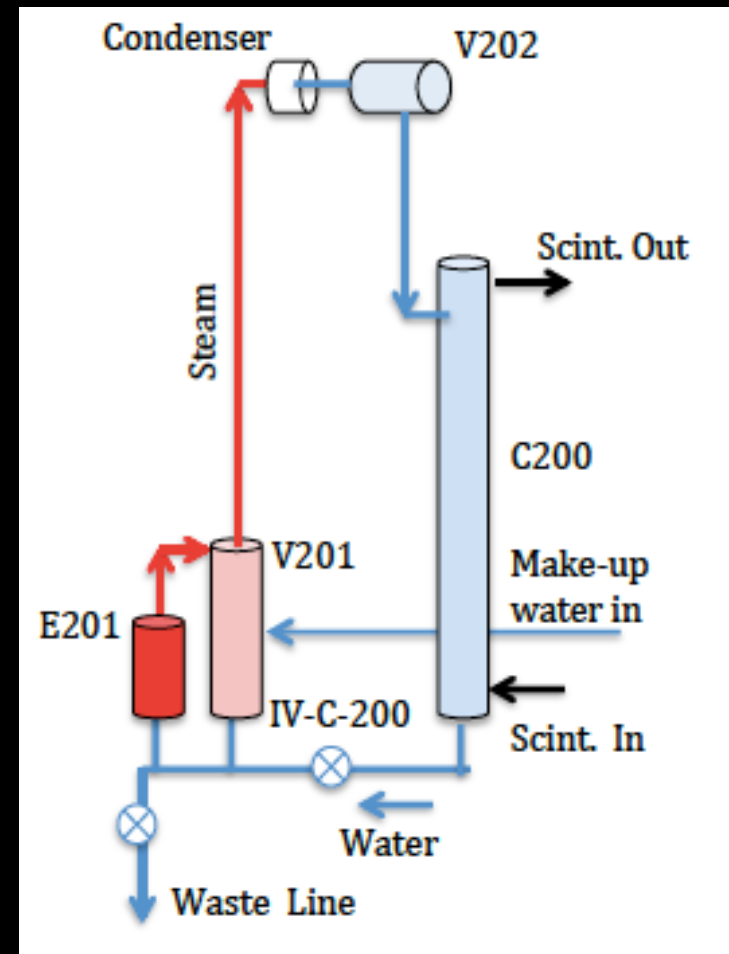
Borexino Re-Purification Systems

Options for Water Extraction or Distillation followed by Nitrogen Stripping



Water Extraction System

- Water is supplied by BX Water Plant (see M. Giammarchi talk).
- Single stage evaporator (E201-V201) produces steam that is condensed into V202.
- Water from V202 flows into top of packed column C200 and returns to V201.
- Scintillator enters bottom of C200, flows upwards against water and exits at top.
- 10% of water is wasted out bottom of E201-V201 at same rate as make-up water is added.
- Waste design is flawed.



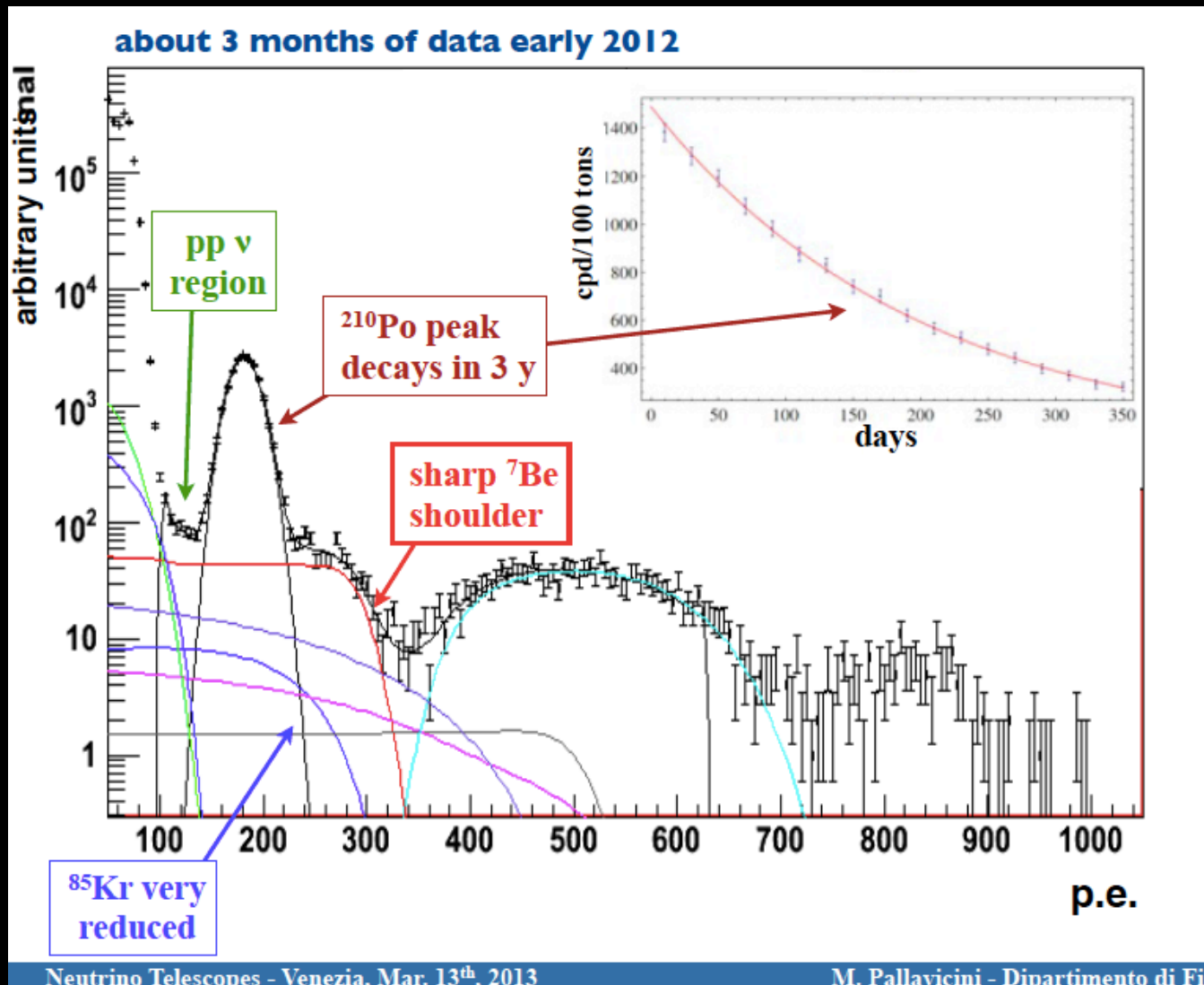
Operational Details of Borexino Water Extraction System

- Flow rates of scintillator and water
 - Scintillator: 800 kg/hr
 - Water: 200 kg/hr
- Recycling water
 - Water is distilled and re-used to reduce cost of disposing large quantities of water contaminated with liquid scintillator.
 - Water from bottom of evaporator E201 is wasted at rate of 20 kg/hr to eliminate non-volatile impurities.
 - Fresh water is added to the evaporator at 20 kg/hr to make up for water that is wasted.
 - Make-up water is supplied from the Borexino water purification system (see M. Giammarchi talk).

Results of 6 cycles of Re-purification

- ^{85}Kr : 30 cpd/100t \rightarrow < 5 cpd/100t
- ^{210}Bi : 70 cpd/100t \rightarrow 20 cpd/100t
- ^{210}Po : Essentially unchanged (?)
- ^{238}U (^{226}Ra): < 9.7×10^{-19} g/g
- ^{232}Th : < 2.9×10^{-18} g/g

Energy Spectrum after Re-Purification



Water Extraction and N₂ Stripping Requirements

- Large surface area between scintillator and water or N₂.
 - Promotes transfer of impurities from the scintillator to water or nitrogen.
 - Achieved with efficient structured packing in column (see photo).
- In general, concentration of impurities in nitrogen and water should be lower than in scintillator.
- Special nitrogen with low levels of ³⁹Ar and ⁸⁵Kr was used for stripping.
 - De-ionized water was used for water extraction, but may not be good enough.

General Comments

1. Nitrogen Stripping

- Nitrogen stripping removed ^{85}Kr very well.
 - The column was designed properly.
 - The upper limits of ^{39}Ar and ^{85}Kr in nitrogen was calculated in advance using standard chemical methods.
 - Methods to measure the argon and krypton in nitrogen were known.
 - Measurements were made to meet the requirements.

Low ^{39}Ar and ^{85}Kr (LAK) Nitrogen

H. Simgen, G. Zuzel LRT 2006

- Measurement techniques:
 - Noble gas mass spectrometry.
 - Low background gas proportional counter
- Borexino Requirements:
 - ^{85}Kr : $< 0.2 \mu\text{Bq m}^3$
 - ^{39}Ar : $< 0.6 \mu\text{Bq/m}^3$
- Tested European Suppliers and delivery system.

General Comments

2. Water Extraction

- ^{238}U reduced by factor >15 ($^{214}\text{Bi-Po}$ actually ^{226}Ra)
- ^{232}Th reduced by factor >3 ($^{212}\text{Bi-Po}$)
- ^{210}Pb reduced by factor of ~ 4
- ^{210}Po not reduced.

- The water extraction column was designed well, but...
- The system to distill and re-use water had a flaw that required a change in procedure to drain wastes properly. The change was implemented for last 2 operations, which gave the best performance.
- Studies of the water plant after the operations indicate high levels of ^{210}Pb , and especially ^{210}Po , in the water that contacts the scintillator.
- The ^{210}Pb and ^{210}Po are produced by ^{222}Rn in ground water.
 - De-ionization processes are not very effective for removing Pb and Po.
 - Distillation is needed, but may have to contend with volatile compounds of Po.

Water Plant

- Water source: De-ionized ground water from LNGS mountain.
- LNGS ground water has normal, but relatively high levels of radioactivity.
 - ^{222}Rn : $\sim 10,000 \text{ Bq/m}^3$ (measured 1996 & 2010).
 - ^{222}Rn daughters not known at time of operations.
 - ^{210}Pb , ^{210}Bi , ^{210}Po estimated to be $\sim 1 \text{ Bq/m}^3$
 - Off-line studies made after the operations roughly confirm the estimates.

Measurement Methods

- ^{208}Pb ICPMS used to infer reduction of ^{210}Pb
 - Show data.
 - Comment on solubility and compare to calcium, magnesium that are similar to radium.
- Auto-deposition of ^{210}Po on silver foils used for ^{210}Po measurements.
 - Show data.

Paper on ^{210}Pb and ^{210}Po in Water

REMOVAL OF LEAD-210 AND POLONIUM-210 FROM WATER BY DE-IONIZATION PROCESSES

B. RUSSELL, J. BENZIGER, F. CALAPRICE, A. FORMICOLA,
A. IANNI, M. LAUBENSTEIN, S. NISI

1. ABSTRACT

Efficient removal of radon daughters from experimental equipment and media is vital to optimize rare event detection sensitivity. The aim of this paper is to explore the extent to which ^{210}Pb and ^{210}Po are removed from water by standard de-ionization processes: reverse osmosis, continuous de-ionization, and ion exchange columns. In this work, we measured ^{210}Pb and ^{210}Po concentrations in water by ICPMS and alpha spectroscopy methods, respectively. De-ionization processes resulted in total reduction factors of approximately 850 and 7 for ^{210}Pb and ^{210}Po , respectively, which are insufficient for many low background direct detection experiments. Additional purification methods in conjunction with de-ionization are needed to produce de-contamination factors sufficient for sensitive detection of rare events.

Summary of Scintillator Purification

1. Successful Water Extraction & N₂ Stripping
 - ⁸⁵Kr, ²³⁸U (²²⁶Ra), ²³²Th, ²¹⁰Pb reduced.
2. Failure to reduce ²¹⁰Po significantly.
3. Discovery of radon daughters in de-ionized water at levels of 1-100 mBq/m³
 - New processes and equipment are being developed for more efficient removal of Pb and Po from water.
4. A productive second phase of Borexino looks promising.