

The design, construction, and initial characterization of an ultra-low-background gas-proportional counting system

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Abstract Over the past several years, the Pacific Northwest National Laboratory (PNNL) has developed an ultra-low-background proportional counter (ULBPC) technology. The resulting detector is the product of an effort to produce a low-background, physically robust gas proportional counter for applications like radon emanation measurements, ground-water tritium, and ^{37}Ar . In order to fully take advantage of the inherent low-background properties designed into the ULBPC, a comparably low-background dedicated counting system is required. An ultra-low-background counting system (ULBCS) was recently built in the new shallow underground laboratory at PNNL. With a design depth of 30 m water-equivalent, the shallow underground laboratory provides approximately $100\times$ fewer fast neutrons and $6\times$ fewer muons than a surface location. The ULBCS itself provides additional shielding in the form of active anti-cosmic veto (via 2-in-thick plastic scintillator paddles) and passive borated poly (1 in.), lead (6 in.), and copper (~ 3 in.) shielding. This work will provide details on PNNL's new shallow underground laboratory, examine the motivation for the design of the counting system, and provide results from the characterization of the ULBCS, including initial detector background.

Keywords Ultra-low-background · Proportional counters · Shallow underground laboratory · Counting system

Introduction

Over the past several years, the Pacific Northwest National Laboratory (PNNL) has developed an ultra-low-background proportional counter (ULBPC) technology. The resulting detector is the product of an effort to produce a low-background, physically robust gas proportional counter for applications like radon emanation measurements, ground-water tritium, and ^{37}Ar [1]. The ULBPC was designed to handle a wider range of gas volumes, have higher efficiency, and be easier to assemble than current ultra-low background solutions. The materials chosen for the detector were specifically selected for their proven radiopurity. High-purity electroformed copper offers excellent radiopurity along with good electrical, thermal, and vacuum properties. The ULBPC body is an electroformed copper cylinder roughly 9 inches in length having a ~ 0.1 L internal volume. Instead of using a more traditional, less-radiopure material like glass or ceramic, the plastic polychlorotrifluoroethylene was chosen for the gas seal and high-voltage insulation for its very low gas permeability and reasonable measured background level. The ULBPC anode wire is 0.001-in.-diameter niobium, a metal that was proven in prior low-background work. The detector is held within an L-shaped oxygen-free high-conductivity (OFHC) copper holder that provides additional shielding from less-low-background parts, such as the brass valves on the gas fill capillary. An example of a ULBPC in an OFHC copper holder is shown in Fig. 1.

Recent work at PNNL demonstrated the measurement of a 1.50 ± 0.02 Bq sample of ^{37}Ar in a ULBPC in an above-ground modest lead shield [2]. This was a challenging measurement, with the signature of ^{37}Ar decay being a series of low-energy Auger electrons and X-rays summing to only 2.8 keV. To measure this very-low-energy signature, significant work was done to extend low-energy

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Fig. 1 The PNNL ultra-low background proportional counter. The cylindrical body of the detector is contained within a solid copper holder which provides additional shielding. A copper SHV enclosure (lower left region of figure), with a connector for applied bias, provides electrical shielding for the high-voltage penetration to the detector's inner volume. A double-valved assembly (extending vertically above the copper SHV enclosure) seals the detector's gas-fill capillary

background performance. This included cosmic-ray veto and pulse-shape discrimination to reject backgrounds and allow lower-threshold operation. While the anti-cosmic veto did serve to help reduce the observed backgrounds, the shielding geometry was far from ideal and the reported results do not represent the ultimate sensitivity attainable with this detector technology. In order to more fully take advantage of the inherent low-background properties designed into the ULBPC, a comparably low-background dedicated counting system is required.

Fully minimizing the ambient radiometric backgrounds will result in improved sensitivity from the ULBPC. The three key methods used to minimize backgrounds for traditional low-background experiments are depth, shielding, and radiopure materials. All three of these elements have been incorporated into the ultra-low-background counting system (ULBCS) recently deployed in the new Physical Sciences Facility at PNNL. The ULBCS was designed to accommodate 12 ULBPC detectors operating concurrently within the shielded inner measurement chamber, with supporting data acquisition electronics to handle coincident measurement of both sample and anti-cosmic veto data. The use of radiopure materials has already been described for the construction of the proportional counter; the following sections will describe how these three background mitigation techniques have been utilized in the design and construction of the ULBCS and will provide initial test results from ULBPC measurements performed within the system.

Design and construction

Background mitigation

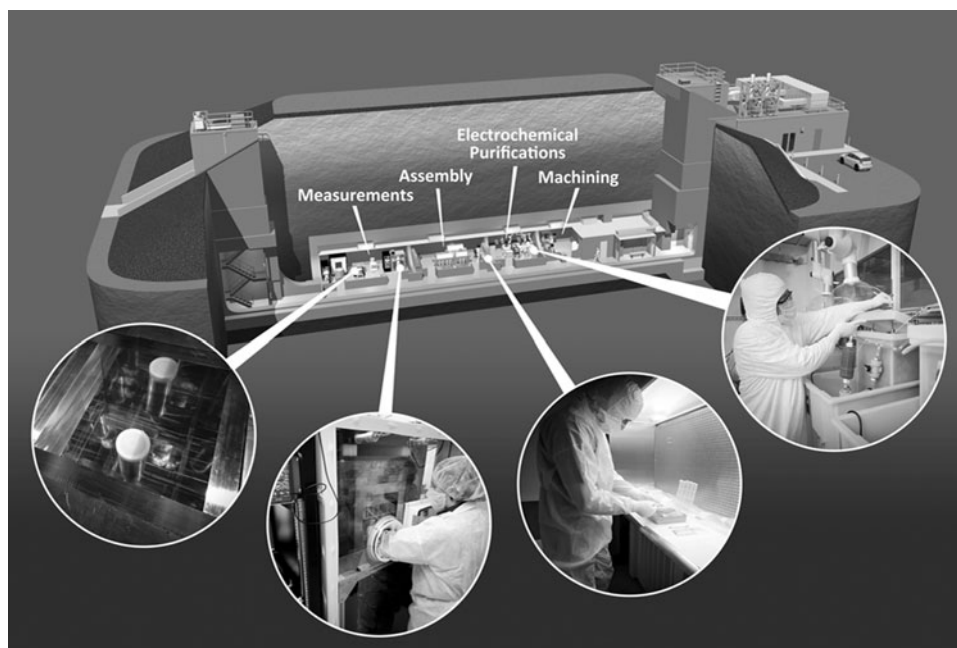
Depth

A new shallow underground laboratory was recently built on the PNNL Richland, WA, campus to house capabilities for clean fabrication and machining, electroformed copper production and cleaning, clean detector assembly, underground materials storage, and low-background radiometric measurements. The laboratories located within this facility are cleanrooms rated to either class 10 k (for the measurements hall and the machine shop) or class 1 k (for the electroforming lab and detector assembly hall). As is represented in Fig. 2, essentially every step in the process of the manufacture, construction, and testing of an ultra-low-background detector can occur in a clean, low-background environment within this underground facility.

The design of the underground laboratory was driven by specific goals for background reduction. Cosmic-ray backgrounds are relatively well understood and methods for determining their impact to a low-background system are well documented [3]. Backgrounds from neutrons are among the most difficult to suppress and thus were a primary concern. At the earth's surface they are produced by both protons and muons interacting with material around a low-background lab, e.g. the concrete floor; after several meters water-equivalent (mwe), the proton contribution to backgrounds is negligible. The impact of these induced fast neutrons to a low-background experiment would be observed in both (a) direct interactions in detectors and, most importantly for proportional counters, (b) activation of the detector materials resulting in events not rejected by traditional anti-cosmic methods. Increasing the depth of a facility is essentially the only way to reduce the incident cosmic ray rate. Given that practical considerations mandated the lab be of shallow depth, the desired effective depth chosen was 30 mwe. The 30 mwe target was expected to yield about 100 times fewer fast neutrons from cosmic-ray interactions in materials. For cosmic-ray backgrounds affecting measurements, the 30 mwe target was expected to provide about a sixfold reduction. No special steps were planned to mitigate gamma-ray backgrounds, as each instrument was envisioned to supply adequate gamma shielding. Building ventilation was designed to have several full air exchanges per hour, so no special steps to mitigate radon for lab air were taken.

The PNNL Shallow Underground Laboratory was constructed with 2,856 ft² of underground class 1 k or 10 k cleanroom space, of which 2,021 ft² is dedicated to low-background detector system production/R&D. The 4-ft-thick concrete floor of the ~650 ft² measurements hall

Fig. 2 Cut-away rendering of the PNNL Shallow Underground Laboratory, Physical Sciences Facility Building 3425, featuring the capabilities of each of the laboratories within



where the ULBCS is housed is located ~ 38 ft below the ground surface. To help achieve the 30 mwe effective depth, a total thickness of ~ 42 ft of compacted backfill (berm height of ~ 21 ft above ground level) was layered on top of the 4-ft-thick concrete ceiling. The application of a berm on top of the underground facility was required in order to achieve the desired mwe depth due to the relative shallowness of the water table at the site. Data from an experiment to calculate effective depth using the anti-cosmic veto system of the ULBCS to measure the reduction in the observed cosmic-ray rate is presented in the “Testing” section below.

Shielding

When planning to reduce the radiometric backgrounds that would be observed by the ULBPC detectors operating in the ULBCS, significant emphasis was placed on shielding. Although having the system at a shallow depth underground will help to reduce the backgrounds due to incident cosmic particles, these backgrounds will not be completely eliminated. Likewise, backgrounds from the surround building materials must also be addressed. The approach taken was that of a “graded shield”, with multiple layers of shielding materials. The material and thickness of each layer of shielding were selected to target a specific background.

As is typical for these types of applications, lead was chosen as the primary material used for shielding the detectors from ambient room gamma-ray backgrounds. The thickness of the lead was chosen to achieve a target background limit of only 0.1 counts/day from surrounding materials. A series of radiation transport simulations were

performed using the GEANT4 code [4] to determine this thickness. The simulations used room background as measured by a high-purity germanium (HPGe) detector as the basis for an incident flux. For a limited number of pre-selected lead shielding thicknesses the expected number of counts/day to be observed by the detector were calculated, the results of which can be seen in Fig. 3. The GEANT4 model of the ULBPC was benchmarked against a measurement using the 59-keV gamma from a ^{241}Am calibration source. A correction factor of 1.2 determined from this benchmarking was applied to the GEANT4 results. In order to achieve the desired 0.1 counts/day limit in the detector from room backgrounds, a thickness of 6 inches of lead was chosen for the shield.

Similar calculations were performed to determine the thickness of copper necessary for the inner measurement chamber. While lead is an excellent material for primary shielding, it can represent a source of background itself. Of principle concern are the lead X-rays and the bremsstrahlung radiation from the decay of ^{210}Pb introduced during refining. In order to minimize the contributions from the ^{210}Pb to the observed background in the detector, one must either use ancient lead or provide an additional layer of high-radiopurity, low-background shielding material between the lead and the detector. Of the readily available moderate- to low-Z metals, copper is among the most radiopure. The thickness of copper for the walls of the inner measurement chamber was determined using a GEANT4 model. For this set of calculations, methods established for simulating low-level backgrounds observed in HPGe were used; a source term of ^{210}Pb was located in the model on the inside surface of the lead surrounding the

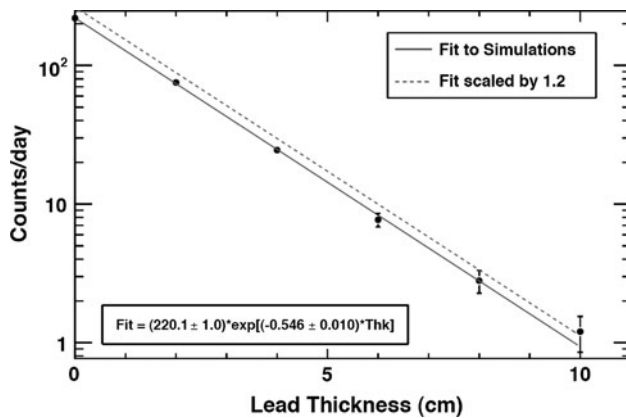


Fig. 3 GEANT4 calculation of expected counts/day in an ULBPC with varying thickness of lead used to shield room backgrounds

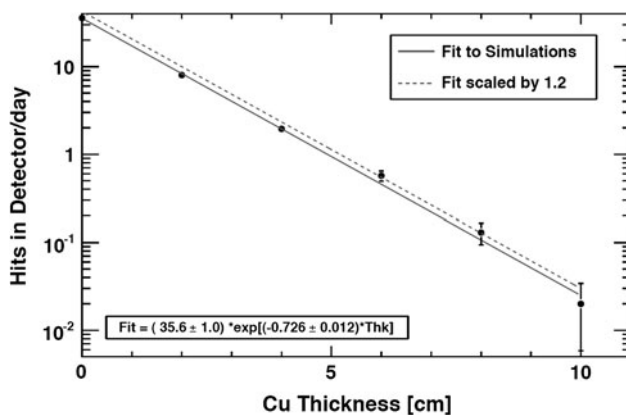


Fig. 4 GEANT4 calculation of expected counts/day in an ULBPC with varying thickness of copper used to shield a ^{210}Pb bremsstrahlung source term

copper [5]. For a limited number of copper shielding thicknesses, the expected number of counts/day to be observed by the detector was calculated. The results can be seen in Fig. 4. By applying the experimental correction to the GEANT4 result and determining an exponential fit to the simulated data, a thickness of ~ 3 in. of copper was chosen for the inner shield liner.

Although the ULBPC is not especially sensitive to neutrons, some neutron shielding was included in the ULBCS. As noted above, the 30 mwe target depth is expected to yield about 100 times fewer fast neutrons from cosmic-ray interactions in materials. However, the room backgrounds of thermal neutrons in the underground lab were expected to be reduced by a much smaller factor. Neutron scattering on hydrogen in the P-10 counting gas (90 % argon, 10 % methane) is a recognized proportional counter background source. Thus, a 1-in.-thick layer of 30 % (by weight) borated polyethylene surrounds the lead shield. With a macroscopic thermal neutron cross section

of 14.5 cm^{-1} , this thickness of absorber should stop essentially all incident thermal neutrons.

Cosmic muon backgrounds

Cosmic-ray-induced backgrounds are the dominant source of background in the ULBPC during above-ground testing, even in a well-shielded enclosure. The PNNL ULBPC design will only reach its background potential if operated with some overburden to attenuate cosmic-rays and, at shallow underground lab depths, a high-efficiency anti-cosmic veto system. The ULBCS was designed with two-inch-thick polyvinyl toluene (PVT) scintillator panels (BC408) located on five of the six sides of the enclosure (no panel on the door) to provide active anti-cosmic veto of muon backgrounds. Even with the one side absent, all vectors through a detector in the inner chamber cross at least one of the veto paddles. Each panel has two 1.5-in. photo-multiplier tubes along one of the (shorter) narrow sides for light collection. The signals from all five of the veto panels are summed into a single preamp, with the resulting summed and amplified signal passed to the PXI-based DGF Pixie-4 digitizer (from XIA, LLC) [6]. The electronic readout system identifies event triggers for both ULBPC and PVT signals. Coincidences are identified and labeled to allow offline cosmic-ray veto and all events involving a ULBPC are digitized to allow offline pulse-shape discrimination.

Radon

Ambient radon in the room air is another potential source for ULBPC backgrounds. Thus, efforts were made in the design of the system to mitigate radon backgrounds. First, the design of the ULBPC holders and ULBCS inner chamber minimize air volume around the detector. Then, polycarbonate panels surround the ULBCS structure, creating an air-tight enclosure. Access to the inner volume is provided through an air-lock. Manipulation of detectors and electrical connections occur through gloveports. Both the full-shield enclosure and the air-lock are plumbed with copper lines providing a continuous purge of dry nitrogen gas from liquid nitrogen boil-off. Independent flow controllers can be used to regulate the purge rate to the two volumes; bleed valves prevent over-pressurization. The dry nitrogen purge for the main air volume is plumbed directly into the inner measurement chamber. This is intended to sweep ambient radon from the measurement space outward.

Surface contamination

During the construction of the ULBPC detectors, great care is taken to minimize any surface contamination on detector

parts in order to reduce backgrounds. Detector assembly is performed in a class 1 k cleanroom and all parts are cleaned prior to assembly to remove possible U and Th dust and radon daughters that may reside on component surfaces from exposure during manufacture and handling. Similar care was taken during the construction of the ULBCS to minimize potential low-level contamination. Prior to assembly, all components of the structure underwent a cleaning procedure specific to the type of material and its proximity to the inner measurement chamber.

The copper bricks forming the inner measurement chamber received the most stringent cleaning protocol. After machining, each brick was scrubbed with laboratory detergent (Micro 90) and rinsed in de-ionized water to remove any oils. The copper bricks were then cleaned using the standard PNNL method for etching and passivating copper [7]. The copper pieces were then dried in a laminar flow hood and double bagged in clean polyethylene for transport to the measurement hall.

Other structural materials also underwent a rigorous cleaning process. For parts small enough to fit in an ultrasonic bath, sonication for 30 min in an ethanol bath was used for cleaning. This process would be repeated up to three times depending on the initial cleanliness of the parts. The parts were then allowed to dry in a laminar flow hood and double bagged in clean polyethylene for transport to the measurement hall. Larger structural materials, such as extruded aluminum struts, received an initial cleaning via a mild detergent followed by a de-ionized water rinse to remove oils from handling. These parts were allowed to dry in a class 10 k cleanroom or a laminar flow hood, then double bagged in clean polyethylene. The lead bricks and borated polyethylene received a wipe-down with ethanol-soaked, lint-free cleanroom wipes.

Mechanical aspects

Once the radiometric properties of the system had been defined the other mechanical aspects of the ULBCS (shown in Fig. 5a) were considered. The number of ULBPC detectors and the size and shape of the ULBPC defined the form-factor of the inner counting chamber of the counting system. Since the required thicknesses of copper and lead build outward from the inner chamber, the capacity of the inner chamber largely drives the overall system weight. A configuration that allowed 12 detectors to be operated simultaneously (6 oriented “up”, 6 oriented “down”) provided a compromise between maximizing detection capabilities and minimizing required shielding size and weight. A close-up view of the inner measurement chamber with an ULBPC in an “up” position is presented in Fig. 5b. By tailoring the contours of the inner chamber to the

L-shape of the ULBPC holder, the total mass of the shield is reduced. In order to simplify application of the borated polyethylene and anti-cosmic veto panels to the outside of the shield, the void spaces created by contouring the shielding material were filled with nylon blocks. The total mass of the lead and copper shield is just over 5.5 tons (3/4 ton of copper, the remainder lead).

Due to the limited amount of floor space available in the measurements hall, minimizing overall system footprint was important. Thus, a design with a vertically translating door was chosen. The cave was constructed on an elevated table surface with the ~1.75-ton door lowering toward the floor to provide access to the inner chamber. This design provided the added benefit of having the top surface of the door act as a work surface while accessing the inner chamber. Door motion is provided by a 0.5 HP motor (1,700–1,750 rpm) with integral brake and two 5,000-lb-rated screw jacks. Jack life is predicted at 1,300,000 in. of travel, based on the door loading. At a conservative estimate of 5 cycles per day and 5 days per week, the jacks should last 20 years.

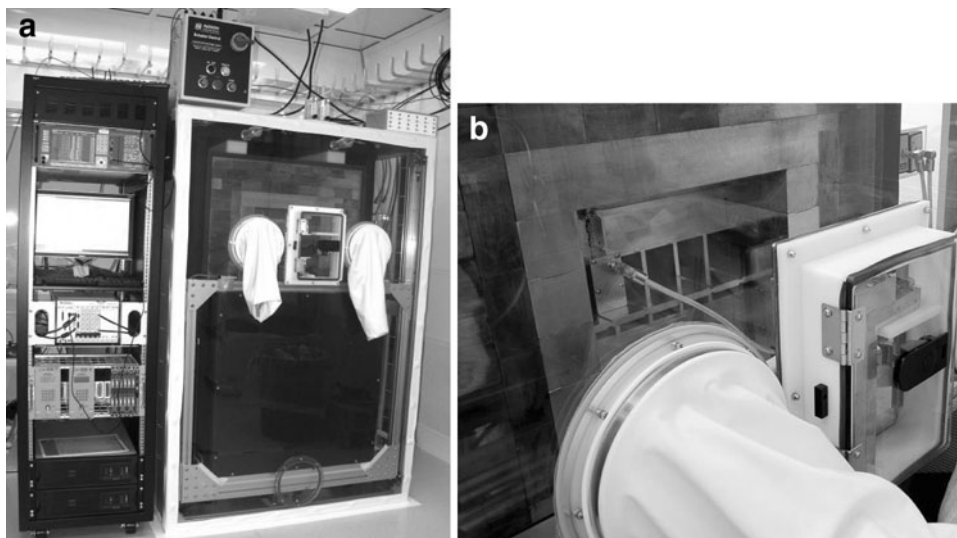
Additional safety features have been incorporated into the mechanical operation of the ULBCS. Limit switches restrict door travel for both upward and downward motion. A pair of optical break-beams located on the top surface of the door prevents operator injury during door travel. An additional emergency stop button on the control panel may be used to halt door movement. Failure of any of the safety devices prevents movement of the door. Loss of power results in the door locking in place.

Testing

Cosmic-ray reduction

As construction on the ULBCS was only recently completed, initial characterization of the system is just now under way. The first test performed with the system was a measurement of the cosmic veto reduction provided by the shallow underground lab. During above-ground prototyping, functionality tests were performed with the veto panels, including a measurement of the cosmic backgrounds from the summed signals of all five panels. These tests were recently repeated with the fully-constructed ULBCS in the shallow underground facility. Figure 6 shows a comparison of backgrounds measured by the PVT panels in both above (329 Bldg.) and below ground (3425 Bldg.) labs. Thresholds for the PVT were set low enough to allow some room background gamma-rays to appear in the resulting spectra. Thus, analysis of this data for cosmic-ray response was limited to a region of interest.

Fig. 5 **a** The ULBCS, showing lead/copper shield in radon enclosure and 12-channel data acquisition system. **b** Close up of the copper inner chamber with capacity for 12 ULBPC detectors in holders



These data indicate an 83.7 % reduction in measured cosmic-ray flux. Using this reduction to estimate cosmic muon attenuation, one can calculate an effective depth of ~ 25 mwe for the system [3].

ULBPC background

Further characterization of the system is currently taking place. An initial measurement of the background observed in a ULBPC inside the system was recently completed. As already noted, cosmic-rays are the dominant source of background in the ULBPC during above-ground testing. For a bare ULBPC tested on a benchtop in an above ground lab at the standard operating parameters of 3 atm of P-10 and 2,150 V bias, a ~ 12 h measurement of the background showed a full-spectrum event rate of about

99,000 counts per day. Using those same operating parameters, a recent ~ 24 day measurement of detector backgrounds in the ULBCS in the shallow underground laboratory measured 234 counts per day for an energy region of 3–400 keV after all cuts. Significant gains were clearly made by operating in an ultra-low-background counting system in the shallow underground laboratory with an active anti-cosmic veto system.

Summary and outlook

The design and fabrication of the PNNL ULBCS for low-background proportional counters has been completed. GEANT4 modeling using HPGe measured room backgrounds and lead radiopurity as source terms was used to determine the amount of shielding necessary for the system. Additional efforts were made to reduce background contributions from neutrons, cosmic-ray muons, ambient room-air radon, and dust containing U and Th. Initial characterization of the system has recently begun. Measurements with the anti-cosmic veto system both above ground and then below ground have demonstrated a $\sim 6.1 \times$ reduction in measured cosmic-ray flux. A background of 234 counts per day was measured with an ULBPC located within the ULBCS over a ~ 24 day period, demonstrating successful reductions of both cosmic and room backgrounds. Additional measurements of the system background over a variety of operating pressures are planned to support a broad range of potential measurements.

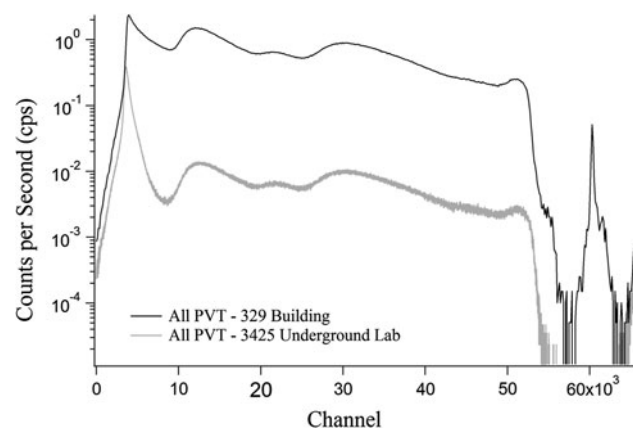


Fig. 6 Comparison of the summed backgrounds measured by all five PVT veto panels of the ULBCS both above ground (329 Bldg) and below ground (3425 Bldg). An 83.7 % ($\sim 6.1 \times$) reduction is seen in the cosmic signal, determined with a region of interest between channels 10 and 55 k

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