Shielding concepts for low-background proportional counter arrays in surface laboratories

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Abstract

Development of ultra low background gas proportional counters has made the contribution from naturally occurring radioactive isotopes – primarily α and β activity in the uranium and thorium decay chains – inconsequential to instrumental sensitivity levels when measurements are performed in above ground surface laboratories. Simple lead shielding is enough to mitigate against gamma rays as gas proportional counters are already relatively insensitive to naturally occurring gamma radiation. The dominant background in these surface laboratory measurements using ultra low background gas proportional counters is due to cosmic ray generated muons, neutrons, and protons. Studies of measurements with ultra low background gas proportional counters in surface and underground laboratories as well as radiation transport Monte Carlo simulations suggest a preferred conceptual design to achieve the highest possible sensitivity from an array of low background gas proportional counters when operated in a surface laboratory. The basis for a low background gas proportional counter array and the preferred shielding

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configuration is reported, especially in relation to measurements of radioactive gases having low energy decays such as ^{37}Ar .

Keywords:

Gas proportional counter system, Cosmic ray shielding, Low background radiation detection, ^{37}Ar

1. Introduction

Low background radiometric measurement systems are typically located in underground laboratories to shield against cosmic-ray products (e.g., protons, neutrons, and muons) that create background event rates and ultimately limit a system's sensitivity reach. The effort to further improve the sensitivity of such underground systems, in one case, has led to the development of ultra low background gas proportional counters made from ultra-pure electroformed copper. These underground proportional counters are further operated in shields to exclude naturally occurring radiation in the laboratory from contributing signals to the detector system. As a consequence of these development efforts the proportional counters now produced have a contribution from naturally occurring radioactive isotopes – primarily α and β activity in the uranium and thorium decay chains – that is entirely inconsequential to instrumental sensitivity levels when the measurements performed in *above-ground* surface laboratories. This allows for a single-minded focus on re-addressing the challenge of the cosmic-ray product induced background contribution in an above-ground proportional counter system. This article presents the rational and conceptual design for a proportional counter array designed for above-ground operation, based upon an assessment of the

background contributions from both cosmic-ray products and naturally occurring radioactivity. As a concrete example, measurement of environmental naturally occurring levels of ^{37}Ar is chosen as an application to explore.

2. Proportional counter backgrounds

Recent reported measurements and simulations presented below provide the basis for bracketing the expected performance of a ultra-low-background counter in a surface laboratory. This section describes the prior measurements and presents simulations used as a basis for inferring the detection system's expected sensitivity. To provide a concrete quantitative estimate, ^{37}Ar detection sensitivity is evaluated. The 2.8 keV emission after ^{37}Ar decay (Barsanov et al. (2007)) provides an uncomplicated nearly single-point energy deposition within the proportional count gas (Xiang et al. (2007)). Thus understanding the performance of the radiation detection is particularly simple allowing a straight forward discussion of background contributions. Furthermore the discussion of ^{37}Ar measurements is motivated by a desire to understand the ultimate reach of surface laboratory systems (Xiang et al. (2008), Xiang et al. (2009)) that ideally would have sensitivity to the naturally occurring levels of in the environment (Cao et al. (2009), Riedmann and Purtschert (2011)).

2.1. Surface cosmic-ray background

A previous gas proportional measurement of ^{37}Ar in a surface laboratory (Aalseth et al. (2011)) provides a point of reference for understanding the backgrounds of a single detector, surface laboratory gas proportional counter system. The single proportional counter used in that measurement was an

44 ultra-low background proportional counter (ULBPC) fabricated from ultra-
 45 pure electroformed copper (Aalseth et al. (2009)). The ULBPC is roughly
 46 9" in length and 1" in diameter with an internal volume of ~ 0.1 L and was
 47 operated in a lead shield consisting of 2", 4", and 8" thick walls (top, sides,
 48 and bottom, respectively). Two 2" thick polyvinyl toluene (PVT) cosmic-
 49 ray veto panels were placed above and below the shield, having 33" $\times$ 14" and
 50 56" $\times$ 19" footprints, respectively. This shielding arrangement is *not* ideal due
 51 to the absence of cosmic-ray veto panel coverage on four sides. Nevertheless,
 52 as reported (Aalseth et al. (2011)), the cosmic veto shield was able to reduce
 53 the background in the 1-20 keV measurement region by a factor of ~ 16.5 .
 54 These results are reproduced in Figure 1 and show that while the cosmic-ray
 55 veto panels reduced the background from cosmic rays, the fact that the *shape*
 56 of the background spectrum is unchanged implies the dominant background
 57 remains due to untagged (un-vetoed) cosmic ray products.

58 *2.2. Shallow underground cosmic-ray background*

59 Pacific Northwest National Laboratory (PNNL) operates a shallow un-
 60 derground laboratory in Richland, WA having an overburden of ~ 30 meters
 61 water equivalent overburden shielding against cosmic rays (Aalseth et al.
 62 (2012)). The overburden of the underground laboratory acts as a shield
 63 against cosmic-ray products, stopping protons and reducing neutrons, both
 64 produced in the atmosphere by spallation processes. The cosmic-ray muon
 65 flux – a more penetrating cosmic-ray product – is reduced by $\sim 85\%$ in the un-
 66 derground laboratory and is used as the final quantitative metric for evaluat-
 67 ing the effective depth of the underground laboratory (Aalseth et al. (2012)).

68 The relevance of the underground laboratory to a surface laboratory mea-

Figure 1: The background from cosmic rays passing through an ULBPC, before and after application of the PVT-based cosmic-ray veto. Data was collected in a small shield located in a surface laboratory.

69 surement system is as a point of comparison for an ideal measurement sensi-
 70 tivity level. Detectors of the same ULBPC design described in the previous
 71 section 2.1 were deployed in the shallow underground laboratory. As noted in
 72 the previous section, the dominant background for these proportional coun-
 73 ters systems, when operated on the surface, is the residual cosmic-ray (direct
 74 or induced) backgrounds despite the use of scintillator-based cosmic-ray veto
 75 detectors. Thus the measurement sensitivity of a ULBPC measurement made
 76 in the shallow underground facility sets a practical lower bound on the poten-
 77 tial sensitivity one could hope to achieve in a surface laboratory measurement
 78 system.

79 Figure 2 presents results from 19.0 live-days of background data collected
 80 from an ULBPC operated in the shallow underground laboratory (Aalseth
 81 et al. (2013)). The proportional counter was located within a low background
 82 shield consisting of: (1) a gas-tight enclosure, (2) an active anti-cosmic veto
 83 detector consisting of 5-cm-thick scintillating plastic panels covering five sides
 84 of the shield, (3) a layer of 30% borated polyethylene with a thickness (2.5
 85 cm) selected to provide opacity for thermal neutrons, (4) a passive lead layer
 86 (15 cm) to limit external gamma-ray induced interactions, and (5) a pas-
 87 sive high-purity copper inner layer with a thickness (7.6 cm) to limit in-
 88 duced interactions from bremsstrahlung gamma-ray production due to ^{210}Pb
 89 (~ 30 Bq/kg) in the passive lead layer. The proportional counter referred to
 90 here was operated at 7 atmospheres of pressure of P-10 counting gas.

91 The upper histogram of Fig. 2 is the gross detector counts while the
 92 lower histogram is the data remaining after the cosmic-ray veto system is
 93 employed to reject events (“vetoed data”). The post-cosmic ray veto data

Figure 2: The background in an ULBPC, before and after application of the scintillating plastic cosmic-ray veto. Data was collected in a proportional counter shield located in the PNNL shallow underground laboratory.

94 appears as a continuum background which is believed to be due to internal
 95 backgrounds (e.g. β , α , and potentially neutrons) and the P-10 counting
 96 gas containing ^{39}Ar (β -emitter: $E_{\beta}^{\text{end}} = 565$ keV; $t_{1/2} = 269$ years). For
 97 comparison to the ^{37}Ar sensitivity level achieved in the previous above group
 98 measurement (~ 45 mBq/SCM), the shallow underground laboratory ULBPC
 99 ^{37}Ar measurement has an initial reported sensitivity of ~ 2 mBq/SCM. The
 100 details of this measurement and the precise sensitivity analysis is reported
 101 in a separate publication (Aalseth et al. (2013)).

102 *2.3. GEANT simulations on cosmic ray backgrounds*

103 The above results from the surface measurement (Aalseth et al. (2011))
 104 and the shallow underground sensitivity estimate (Aalseth et al. (2013)) lead
 105 to a desire to understand the impact cosmic rays have on the background
 106 in the proportional counters in the ^{37}Ar measurement region. The relevant
 107 characteristics are the counter's length, radius, operating pressure, and the
 108 orientation of the cylindrical axis with respect to the vertical direction. For
 109 the moment, the orientation is fixed so the proportional counter axis is par-
 110 allel the Earth's surface (i.e., the tube is horizontal) as this is how the initial
 111 above ground measurement was performed (Aalseth et al. (2011)). A sim-
 112 ple GEANT model of an array of ULBPCs was prepared and cosmic-ray
 113 neutrons, protons, and muons were passed through the counter array. The
 114 Cosmic-ray Shower Library (CRY 1.5) (Hagmann et al. (2007) and Hagmann
 115 et al. (2008)) was used as input to GEANT to generate the cosmic ray neu-
 116 trons, protons, and muons with approximately the correct relative proportion
 117 and angular distribution of incoming direction.

118 For each of the simulations performed, the integral number of counts in

Figure 3: Proportional counter optimization study. Cosmic rays generated from the CRY package are simulated in GEANT. The geometry and pressure of the proportional counter is varied. Shown here is the impact of radius and pressure on the totally energy deposited in the proportional counter gas. For an ^{37}Ar measurement the sensitive energy region is 2–5 keV, thus designs that *increase* the total energy deposited by cosmic rays assist in *reducing* the background in the ^{37}Ar measurement region. Note: 1 inch = 25.4 mm and 1 atm = 101.3 kpa.

119 the energy range from 2 to 5 keV was determined and used as representative of
 120 the *relative* importance of a particular proportional counter's length, radius,
 121 and operating pressure. In this way it is possible to ascertain the preferred
 122 proportional counter design for minimizing the cosmic ray background in an
 123 above ground system, in this example targeting measurement of the 2.8 keV
 124 signature of ^{37}Ar decay. The results are presented in Tables 1, 2, and 3.
 125 Figure 3 presents a sub-set of the simulations that illustrate the impact of
 126 counter radius and pressure on the measured background energy spectrum.
 127 The broad peak seen in each energy spectrum is essentially due to the amount
 128 of energy deposited along the ionizing particle's path length through the
 129 counter (i.e., "minimum ionizing particle" dE/dx). Thus, larger radius and
 130 higher pressure tubes show a broader spectral peak pushed to higher energy.

131 Putting aside other aspects of making a feasible, operational proportional
 132 counter, the summary conclusion from these simulation results is that the
 133 ideal counter is short in length, large in radius, and high in pressure. Note
 134 that large radius and high pressure creates a proportional counter configura-
 135 tion requiring very high potential bias for operation (i.e., high V/cm in the
 136 gas). In comparing the relative effect of pressure and radius, pressure has a
 137 greater impact. Thus a tube construction design driven by ability to achieve
 138 a desired high pressure, effectively selects the proportional counter radius by
 139 being required to keep the high voltage bias below 5000 V, a typical commer-
 140 cial high voltage bias supply module. Having fixed the operating pressure
 141 and radius to obtain proportional charge gain with a reasonable high voltage
 142 bias power supply, the final requirement is obtaining the desired sample-size
 143 target by selecting the proportional counter length.

Table 1: *Length series*. Integral counts for the energy range 2–5 keV per 10^5 simulated cosmic rays. Pressure = 2 atm and radius = 1 inch for all simulations in this table. Note the counts do not simply double as the length doubles due to an "edge effect" in the simulation since the cosmic rays were generated from a 1000 mm by 1000 mm surface centered above the proportional counter.

Length (mm)	100	200	300	400	500
Counts (2–5 keV)	596	1017	1334	1622	1776

Table 2: *Pressure series*. Integral counts for the energy range 2–5 keV per 10^5 simulated cosmic rays. Length = 200 mm and radius = 1 inch for all simulations in this table.

Pressure (atm)	2	5	10	15
Counts (2–5 keV)	1017	167	59	37

Table 3: *Radius series*. Integral counts for the energy range 2–5 keV per 10^5 simulated cosmic rays. Length = 200 mm for all simulations in this table.

Radius (mm)	9.79	16.04	22.29	28.54	34.79
<i>Pressure = 2 atm</i>					
Counts (2–5 keV)	2134	1017	661	498	424
<i>Pressure = 10 atm</i>					
Counts (2–5 keV)	81	59	66	73	77

144 2.4. *External gamma-ray backgrounds*

145 Above ground proportional counter measurements using a OFHC copper
146 proportional copper were performed (Mace et al. (2013)) to determine the
147 affects of lead shielding on non-cosmic ray backgrounds. The shield used in
148 this study was composed of 2" thick lead in a cave of dimensions 8"×8"×16"
149 (W×H×L). Above and below the cave were 1" thick sheets of cadmium (neu-
150 tron absorber) and 1" thick PVT scintillator panels (cosmic-ray veto). This
151 shield design was then rebuilt increasing the lead shielding thickness to 4".
152 The OFHC copper proportional counter was also operated outside the shield
153 in a typical laboratory bench-top environment. Figure 4 shows the results
154 of these comparative shielding studies. The average background rate was
155 calculated over an energy range of 3-400 keV.

156 These results are consistent with the results previously discussed from an
157 above-ground, shielded ULBPC (Sec. 2.1) (Aalseth et al. (2011)). The results
158 presented in Fig. 4 clearly demonstrate the need for lead shielding, though
159 thicknesses of greater than 2" do not appear to provide a large additional im-
160 provement in background. It seems reasonable to conclude from these results
161 2" of lead shielding is more than adequate to shield against external ubiqui-
162 tous primordial gamma-ray emitting radioactivity sources, though perhaps
163 less lead could be employed (perhaps 1") if the system's weight became a
164 design consideration.

Figure 4: Results from a shielding study using an above ground OFHC copper proportional counter operated at 3 atm of P10 counting gas.

165 2.5. Proportional counter construction material backgrounds

166 To compare the backgrounds rates in an ULBPC¹ and an OFHC copper
 167 proportional counter², a test was performed in the PNNL shallow under-
 168 ground laboratory using the apparatus described in Aalseth et al. (2013)
 169 composed of cosmic ray and lead shielding arrangement. Spectroscopic his-
 170 tograms of the data from each of the two counters (both operated at ~ 1 atm)
 171 show a generic shape indicative of leakage of un-vetoed cosmic ray induced
 172 backgrounds, both before and after the application of a cut from the cosmic
 173 ray veto system. The residual rates in each counter after application of the
 174 cosmic ray veto system were 187.3 ± 1.8 cpd and 335.8 ± 3.1 cpd in the
 175 ULBPC and OFHC copper counters, respectively. Although it would not
 176 be considered a “conservative” assumption, one could presume the ULBPC
 177 has essentially zero background counts from the copper material and there-
 178 by conclude the difference in the two rates is representative of the absolute
 179 background rate of a proportional counter constructed from OFHC copper
 180 at 148.5 ± 3.6 cpd. Alternatively, the conservative assumption is of course
 181 to assume *all* the residual background events are due to internal radioac-
 182 tive backgrounds in the construction materials, that is, a background rate
 183 of 335.8 ± 3.1 cpd. These ranges of internal backgrounds from the OFHC
 184 copper proportional counter construction materials figure in estimating the
 185 lowest minimum detectable concentration (MDC) that can be achieve from
 186 a OFHC copper proportional counter operated in a surface laboratory.

¹Specifically, PNNL counter number 2U03.

²Specifically, PNNL counter number OFHC2.

187 3. Absolute gas counting methods

188 In the case of a single proportional counter instrument, determining the
189 absolute activity concentration of an unknown radioactive gas sample re-
190 quires a calibration of the counter’s efficiency. This efficiency is often driven
191 by the details of the proportional counter design at the ends of the tube
192 where electric fields lines may not produce collectible charge transport with
193 the same proportional gain characteristics as the central portions of the tube.
194 It is possible to use two (or more) separate but “identical” proportional
195 counters to effectively remove any uncertainty related to the ends of the
196 proportional tubes (Unterweger (2007)). In summary explanation, assuming
197 the end effects of the compared proportional counters are the same (hence
198 the term “identical”), subtraction of the shorter counter’s results from the
199 longer counter’s results effectively creates a virtual proportional counter vol-
200 ume equal to the difference in the two counters’ volumes but absent of any
201 end effects.

202 3.1. Method of variable length proportional counters

203 The method of variable length proportional counters was explored pre-
204 viously using both OFHC copper proportional counters and ULBPC copper
205 tubes (Williams et al. (2013)). The focus of that work was to develop the
206 capability to directly prepare absolute activity concentration gas standards
207 in the range 0.05-10 Bq/cm³. In this report we consider the potential use
208 of the same method of variable length proportional counters as a compo-
209 nent of an quantitative assessment of sample gases. We perform this analysis
210 with the same context of measurements of naturally occurring levels of ³⁷Ar

211 (Riedmann and Purtschert (2011)). The reported concentration levels for
 212 soil gas are in the range of 1-100 Bq/SCM (SCM $\equiv$ standard cubic meter)
 213 of whole air. When 100% of the argon is extracted from whole air, this re-
 214 sults in a argon gas sample having an activity concentration in the range of
 215 approximately 1-100 $\mu\text{Bq}/\text{cm}^3$ – dramatically lower than considered in our
 216 prior work (Williams et al. (2013)). Furthermore, we take into considera-
 217 tion the prior described background rates of the above ground and below
 218 ground proportional counters described in Sec. 2. Recall the Figures 1 and
 219 2 report background rates of 42 mHz (over 0-20 keV) and 0.6 mHz (over
 220 1-15 keV) for the 100 cm^3 volume proportional counters. A rough estimate
 221 of the background rate *per keV* is then approximately 2 mHz/keV above
 222 ground to 0.04 mHz/keV for the shallow underground system. For the sake
 223 of providing a concrete sensitivity analysis, we presume we will be able to
 224 achieve a background rate in an above ground, well shielded proportional
 225 counter array of 0.5 mHz/keV/counter. This value is approximately a order
 226 of magnitude higher than achievable in a shallow underground laboratory
 227 (See Sec. 2.2), but a factor of 4 improvement in the background from the
 228 prior above ground system results (See Sec. 2.1).

229 3.2. Sensitivity and uncertainty analysis

230 As done previously (Williams et al. (2013)), using the approach outlined
 231 in the Guide to the Expression of Uncertainty (GUM) (ISO (1995)), an un-
 232 certainty model was constructed for sensitivity and uncertainty analysis using
 233 the GUM Workbench software tool (Metrodata-GmbH (2012)). The model
 234 represents the 1"-diameter ULBPC tubes described in Sec. 2.1 (Aalseth et al.
 235 (2009)). Two equations are employed, the first for two different length pro-

236 proportional counters ($V_S = 100 \text{ cm}^3$ and $V_L = 250 \text{ cm}^3$ volumes) and the second
 237 for the larger ($V_L = 250 \text{ cm}^3$ volume) tube alone. In this analysis, it is envi-
 238 sioned both proportional counters are loaded with an argon gas sample and
 239 10% methane by volume to make P-10, thus giving a mol fraction of $f_{\text{mol}} =$
 240 0.90. Data is then collected for 24 hours (86400 seconds). Equation 1 is used
 241 to analyze the results from the two tubes jointly while Equation 2 uses the
 242 same data but only considers the results from the longer of the two tubes.

$$A_{L-S} = \frac{(1 + k_t + k_w) \left(\frac{1}{f_{BR}} \right) \left[\left(\frac{S_L}{t} - \frac{B_L}{t} \right) - \left(\frac{S_S}{t} - \frac{B_S}{t} \right) \right]}{T_{STP} \left(\frac{P}{P_{STP}} \right) f_{\text{mol}} \left(\frac{1}{Z} \right) \left(\frac{V_L}{T} - \frac{V_S}{T} \right)} \quad (1)$$

243

$$A_L = \left(\frac{1}{\epsilon_{\text{fid}}} \right) \frac{(1 + k_t + k_w) \left(\frac{1}{f_{BR}} \right) \left[\left(\frac{S_L}{t} - \frac{B_L}{t} \right) \right]}{T_{STP} \left(\frac{P}{P_{STP}} \right) f_{\text{mol}} \left(\frac{1}{Z} \right) \left(\frac{V_L}{T} \right)} \quad (2)$$

244 In these equations, k_t is the ratio of those decays *not* measured to those
 245 decays that are measured. In this particular example that amounts to an as-
 246 sessment of the fraction of events contributing to the signal region of interest
 247 (ROI) for analysis. In a typical analysis, a Gaussian distribution is fit to the
 248 energy peak located at 2.8 keV. The central 82.6% of the peak distribution
 249 is used as the ROI thus giving a k_t value equal to $(100 - 82.6)/82.6 = 0.21$.
 250 A similar quantity, k_w is the ratio of those decay events that range out in the
 251 walls of the proportional coutner to those decay events that create a signal.
 252 In this particular case, it is assumed that nearly all (99%) of the ^{37}Ar de-
 253 cays produce event signals as the energy deposition is low energy and nearly
 254 point-like in the proportional count gas described here. Thus the k_w is $(100$
 255 $- 99)/99 = 0.01$. The uncertainty for these values is estimated relative to our
 256 expectations for how well we would be able to determine these values from

257 our current measurement experiences. The percentage of the fiducial volume
 258 of the longer proportional counter that is sensitive to ^{37}Ar decay is ϵ_{fid} . The
 259 branching fraction of the ^{37}Ar decay to the K-shell is f_{BR} and is considered
 260 a known constant for the purposes here. The count duration, t , is given in
 261 seconds. The proportional counters are assumed to operate at a pressure P
 262 $= 709.275$ kPa and at a temperature $T_{\text{L}} = T_{\text{S}} = 300$ K. The compressibility
 263 of P-10 gas is $Z = 0.998$ and is considered a known constant. All of these
 264 fixed values are summarized in Table 4.

265 The number of counts measured in the signal (S) region of interest (ROI)
 266 and the number of counts in the background (B) determined from side band
 267 regions away from the tails of the Gaussian distribution fit are represented in
 268 the equations by S_{L} , B_{L} , S_{S} , and B_{S} for the long (subscript L $\rightarrow 250$ cm³) and
 269 short (subscript S $\rightarrow 100$ cm³) proportional counters. The following com-
 270 parative analysis, the signal counts are estimated for three difference mea-
 271 surement cases of ^{37}Ar activity concentration in whole air: 1000 mBq/SCM,
 272 100 mBq/SCM, and 10 mBq/SCM. As described above, the background rate
 273 is assumed to 0.5 mHz/keV over an 0.8 keV width of energy region that is
 274 equivalent to the width of the 82.6% of the peak distribution is used as the
 275 signal ROI. The estimated counts in each energy region for each proportional
 276 counter are tabulated in Table 5 for the three ^{37}Ar activity concentration
 277 cases.

278 The GUM Workbench software tool (Metrodata-GmbH (2012)) output
 279 provides an *index* of the uncertainty contribution of a given input value and
 280 input value uncertainty. The higher the index percentage for a given input
 281 value and input value uncertainty, the more that input and its uncertainty

Table 4: Fixed inputs for the GUM uncertainty model. The volume values V_L and V_S are correlated via determination from the same calibrated reference.

Quantity	Unit	Value	Uncertainty
k_t	-	0.21	0.010
k_w	-	0.01	0.005
ϵ_{fid}	%	94.0	0.5
f_{mol}	mol/mol	0.90	0.01
t	s	86400	1
P	kPa	709.275	0.2
V_L	cm ³	250	0.625
V_S	cm ³	100	0.250
T	K	300	0.1
f_{BR}	%	90.2	constant
Z	-	0.998	constant
T_{STP}	K	273.15	constant
P_{STP}	kPa	101.325	constant

Table 5: Estimated signal region (S) and background region (B) counts for example 24-hour measurements described in the text for long (L) and short (S) proportional counters filled with argon extracted from whole air. Activity concentrations are given in mBq per standard cubic meter (SCM) of whole air.

Count Region	Activity (mBq/SCM)		
	10	100	1000
S_L	180	1008	9288
B_L	88	88	88
S_S	68	365	3335
B_S	35	35	35

282 *drives* the overall calculated uncertainty of the equation modeled. From
283 Table 5 one sees the “signal-to-noise” ratio ranges from ~ 1 (for 10 mBq/SCM
284 ^{37}Ar in the short counter) to ~ 100 (for 1000 mBq/SCM ^{37}Ar in the long
285 counter).

286 The results from Table 6 are informative because over the range of rele-
287 vant measurement activity concentrations, the contribution to uncertainty in
288 the determination from a given measurement shifts from a statistics limited
289 situation (for low activity concentration) to systematics limited situation (for
290 higher activity concentrations). Perhaps surprisingly the analysis selection of
291 the energy range over which the ^{37}Ar decay events are counted (the analysis
292 ROI) becomes an important consideration in the higher activity concentra-
293 tion measurements. Stated another way, the knowledge of the Gaussian

Table 6: The GUM Workbench index of contribution to uncertainty reported for various input parameters modeled in equations 1 and 2. The index of contribution to uncertainty is the relative contribution the input parameter and input parameter’s uncertainty contribution to the *overall* uncertainty reported on A_{L-S} or A_L . The index of contribution to uncertainty is reported on a scale from 0.0-1.0 where larger values indicate a *greater* contribution to the uncertainty of the modeled equation. The quantities A_{L-S} and A_L are reported in $\mu\text{Bq}/\text{cm}^3$. For conversion purposes, 10 mBq of ^{37}Ar per standard cubic meter (SCM) of whole air is equivalent to 1 $\mu\text{Bq}/\text{cm}^3$.

Relative uncertainty on reported activity concentration (%)						
	Length Compensated			Single Long Counter		
	Activity (mBq/SCM)			Activity (mBq/SCM)		
	10	100	1000	10	100	1000
A_{L-S}	65	13	4.4			
A_L				36	7.6	3.2

Index of contribution to concentration uncertainty						
S_L	0.485	0.656	0.551	0.669	0.828	0.433
B_L	0.237	0.057	0.005	0.327	0.072	0.004
S_S	0.183	0.237	0.198	-	-	-
B_S	0.094	0.023	0.002	-	-	-
k_t	0.000	0.015	0.139	0.002	0.047	0.265
ϵ_{fid}	-	-	-	0.000	0.017	0.099
f_{mol}	0.000	0.009	0.085	0.001	0.029	0.162

width of the ^{37}Ar decay peak – and the uncertainty associated with that knowledge – impacts how well known the fraction of the peak covered by the analysis ROI. An interesting aside is, for this example case, one can consider the choice between using the length compensated method or a single well calibrated counter as a choice between having the measurement uncertainty driven by *either* the statistics of the short counter in the length compensated method (i.e., S_S and B_L) *or* the systematic uncertainties associated with knowledge of the “sample size” (i.e., k_t , ϵ_{fid} , and f_{mol}).

4. Proportional counter array design

Figure 5 presents one conceptual realization of the cosmic ray background reduction and mitigation presented in this article. The individual proportional counters are oriented vertically as this will typically increase the path length through the gas, there-by pushing the energy measured from cosmic-ray products further away from the low energy ^{37}Ar signature at 2.8 keV. A closely packed array of proportional counters increases the ability to veto events based upon coincidence triggers between the tubes³. The inner shield shown in Fig. 5 is composed of 2"-thick standard lead bricks to eliminate external gamma-rays (See Sec. 2.4). The lead shield is surrounded by 1"-thick borated polyethylene to provide some thermal neutron absorption. Finally, and most importantly, the entire shield is enclosed in a active plastic scintillator veto system. The scintillator panels are 2"-thick to ensure the ability to separate muons from gamma rays. This shield as design is conceptual and

³In the low-rate configurations studied here, the accidental coincidence rate from the ^{37}Ar decays is inconsequential.

Figure 5: Concept for a shield intended for low background gas proportional counters located in surface laboratories.

weights approximately 1400 pounds (635 kg). The choice of variable proportional counter length is schematic and depends on the final measurement goal, sample volume, and activity concentration.

5. Gas preparation example

Figure 6: Flow diagram for the production of argon from an air sample.

In this article we have considered the measurement of ^{37}Ar as a concrete example to study shielding requirements for low-background gas proportional counters located in above ground (surface) laboratory settings where cosmic rays are expected to dominate the background of the system. As briefly mentioned earlier in this article, the choice of the proportional counter *length* is the least background-dependent handle available to adjust the system's capacity relative to a given sample preparation methodology. To further discuss

327 this sample-size aspect of system design considerations, we again reference
328 ^{37}Ar measurements in the context of determination of the naturally occurring
329 levels of ^{37}Ar in soils types and at depths within those soils (Riedmann and
330 Purtschert (2011)). We present a concept for processing a soil gas sample for
331 ^{37}Ar measurement as a specific example of a potential gas sample size to as-
332 sist in understanding the capacity requirements of gas proportional counters
333 as described in this article.

334 For the purposes of this article, we assume any dried soil-gas sample is
335 represented in rough volumetric composition by normal dry air: N_2 (78%),
336 O_2 (21%), Ar (0.9%), CO_2 (0.04%), and other minor constituents each at less
337 than 0.01%. The measurement goal is to determine the ^{37}Ar concentration in
338 $\sim\text{mBq/SCM}$ (Riedmann and Purtschert (2011)). Although a variety of gas
339 mixtures are possible for gas proportional counters methods, a natural choice
340 for an ^{37}Ar measurement is P-10 (90% argon and 10% methane), where the
341 90% argon fraction is supplied from the soil-gas sample.

342 As a general example, the ultra-low-background proportional counters
343 (ULBPCs) hold ~ 0.1 L of gas volume when filled at atmospheric pressure
344 and up to ~ 1 L of gas volume when filled at 10 times atmospheric pressure.
345 Although the ULBPC typically operates at fill pressures between these two
346 extremes, we first consider as a simple example the ~ 1 L of gas volume, 10
347 atmosphere fill case. In this case, $\sim 90\%$ of the fill gas is argon or ~ 0.9 L of
348 argon at atmospheric pressure. To obtain 0.9 L of argon from dried air (e.g.
349 the soil-gas sample), 100 L (or 0.1 SCM) of dried air is required as an initial
350 sample collection, assuming 100% collection efficiency when separating the
351 argon from the desiccated air. A 100% collection efficiency of argon gas from

dry air is unrealistic. Determining the actual representative argon collection efficiency is of primary interest as well as understanding the processing time.

To this end, an argon production test bench was developed to quantify the aspects of argon production efficiency as well as processing duration. A schematic flow diagram of the system is presented in Figure 6. The primary step in the separation of argon from dry air is a pressure swing absorption (PSA) oxygen (O_2) generator. The PSA O_2 generator eliminates the predominate nitrogen (N_2), producing an oxygen-argon mixture (O_2 -Ar mix), but at the loss of approximately 50% of the argon in the initial air sample. After the output of the oxygen (O_2) generator is collected, the O_2 -Ar mix is fed into a carrier gas of helium (He) and then mixed with hydrogen (H_2) for converting the hydrogen and oxygen into water vapor (H_2O). The water is collected and the gas stream composed of hydrogen, helium, argon, and trace oxygen and nitrogen is dried and passed through a reactive copper bed. The final gas stream is composed of argon and helium, that are finally separated via activated charcoal traps where a nominal 50% efficiency is assumed. In a typical process cycle, approximately 0.6 L of argon is produced from 150 L of initial air sample. This process cycle is composed of approximately 45 minutes of active processing. An entire cycle including regeneration time, requires approximately 2 hours. Thus, in this argon production test bench cycle approximately 0.6 L of argon sample is prepared every 2 hours. This sample size and production rate is on the scale relevant to the 24-hour count duration of ~ 1 L volumes described in Sec. 3.

375 **6. Conclusion**

376 The value of ultra low background proportional counters (and low back-
377 ground radiation detection systems in general) is typically achieved when
378 applied to measurements made in underground laboratories. In such under-
379 ground systems, where cosmic rays have been eliminated as a major contrib-
380 utor to the background of the system, the uranium, thorium, and potassium
381 content of the instrument components typically dominates. Conversely, de-
382 velopments of low background proportional counters have led to instruments
383 of such low internal background, that if they were to be operated on the sur-
384 face, the internal background contaminants effectively have zero contribution
385 to the system’s background. In such a case, the instrument and shielding de-
386 sign can focus exclusively on ways to mitigate the impact of cosmic rays prod-
387 ucts on the measurement. We have presented a conceptual thought process
388 on how to best reduce and mitigate against cosmic ray background contribu-
389 tions in a surface laboratory setting for gaseous proportional counters. We
390 analyzed the potential performance of such a system in the context of a sim-
391 ple radioactive gas, ^{37}Ar , at concentration levels relevant to environmental
392 measurements. We conclude it is potentially feasible to design and build an
393 actively and passively shielded, proportional counter array that is sensitive
394 to the range of naturally occurring ^{37}Ar soil gas activity concentration levels
395 in an above ground measurement system.

396 **Acknowledgments**

397 The research described in this paper was supported in part by the Ultra-
398 Sensitive Nuclear Measurements (USNM) Initiative, a Laboratory Directed

399 Research and Development Program at the Pacific Northwest National Labo-
 400 ratory, a multiprogram national laboratory operated by Battelle for the U.S.
 401 Department of Energy. Information Release PNNL-SA-106350.

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