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The iSERIES™ Radon Progeny Compensation Algorithm and its Application to Air Filters

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The iSERIES™ Radon Progeny Compensation Algorithm and its Application to Air Filters

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Abstract

Sandia has developed methods to perform a rapid count using Canberra's iSERIES™ Alpha/Beta Counting System, the iSOLO™. The purpose of this report is to show that by using the iSERIES™ built-in radon progeny compensation algorithm, the radon progeny-free gross alpha air concentration can be determined for a sample taken using a high volume air sampler or a lapel device within a few hours post-sampling. Also, a special form of the Currie[1] Minimum Detectable Activity estimation was derived for use with the radon progeny stripping algorithm after it was discovered that the expression used by the Canberra software did not take the counts due to radon progeny into consideration as part of the background. This rapid air sample analysis proves to be a vast improvement to the current method that has an average turn-around time between seven and ten days.

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Nomenclature

GPC	Gas-flow Proportional Counter
HiVol	High Volume
Lc	Critical Level
MDA	Minimum Detectable Activity
RCT	Radiological Control Technician
ROI	Region Of Interest
RPDP	Radiation Protection Dosimetry Program
RPSD	Radiation Protection Sample Diagnostics Program

Introduction

Some Sandia National Laboratories (SNL) experiments require quantification of alpha-emitting radionuclides suspended in the air in an area where there are likely elevated levels of environmental radon progeny ("radon progeny" is used here to describe the air-borne components of the Uranium and Thorium decay chains that can be collected on paper-type air filters). Personnel are required to wear lapel air samplers while performing work and high-volume air samplers are also used in these areas. These two different types of air samples are necessary to assess airborne activity levels. High volume air samples are taken to determine if an area exceeds safe working limits. When an area is deemed safe but there still exists a potential airborne contaminant concern, workers will wear breathing-zone air samplers that sample volumes of air at a typical breathing rate. The measured radioactivity of these lapel filters is used to assess the inhaled dose of the worker. In many situations at SNL, the speedy analysis of these samples is crucial to an experiment's success.

The current method used at Sandia for high-volume air samples and lapel filter analysis is to take a preliminary count of the filter on a Gas-flow Proportional Counter (GPC) when the filter is removed from the sampling device, then take another count seven days later to allow for the environmental radon progeny to decay. The GPC provides no energy information on the alpha particles it detects. Thus, the exact contaminants on the air filters are not known. The seven day wait time is used as a "rule-of-thumb" to ensure that the final result does not contain counts from progeny due to the Uranium or Thorium decay series. Since most operations are time-sensitive it is highly desirable to have an accurate measurement of the radon progeny-free gross alpha air concentration as soon as possible after sampling.

Hence, Sandia developed methods to perform a rapid count using Canberra's iSERIES™ Alpha/Beta Counting System, the iSOLO™. The purpose of this report is to show that by using the iSERIES™' built-in radon progeny compensation algorithm, the radon progeny-free gross alpha air concentration can be determined for a sample taken using a high volume air sampler or a lapel device within a few hours post-sampling. This rapid air sample analysis proves to be a vast improvement to the current method that has an average turn-around time between seven and ten days.

Process Development

Two lapel filters and two HiVol filters were used in typical air sampling devices (SKC breathing zone air sampler, and Staplex High Volume air sampler). The air samples were obtained from a space of known high radon progeny concentration to provide a worst-case scenario in terms of the ability of the iSERIES™ instruments to compensate for radon progeny. Each filter was counted on both an iSOLO™ and a GPC for five minutes. The filters were then spiked with approximately 300 dpm of a Pu-239 liquid standard diluted in ethanol to speed up the drying process. Table 1 includes the amount of spike and calculated activity for each filter. Following the spike, the filters were repeatedly counted on both instruments for five minutes per count over a number of hours to assess the behavior of both instruments as the natural radon progeny decay progressed.

The following activities were plotted over time:

- Uncompensated Gross Alpha Activity from the iSOLO™
- Radon progeny-Compensated Gross Alpha Activity from the iSOLO™
- Gross Alpha Activity from the GPC.

Alpha particles have a very small mean free path compared to less massive particles. Because of this, many alpha particles are attenuated by both the sample matrix and the air that exists between the sample and the detector. Both the iSOLO™ and the GPC unit are calibrated using electroplated, certified sources. The matrix effects of electroplated sources on the detection efficiency of these instruments are not comparable to those of the spiked air filters used in this study. Hence, one filter of each type (with no exposure to air flow) was spiked with nearly the same amount of Pu-239 standard. These matrix spiked filters were then counted a number of times on both instruments to establish an "expected" Pu-239 presence on the filter matrix. In this way, it can be shown that the gross alpha activity approaches this "expected" value as the radon progeny decays. Also, it can be shown that the compensated alpha activity calculated using the iSERIES™ algorithm is roughly equal to this matrix-corrected spike activity for all counts post-spike. Aliquots of the standard were slightly different for the sample spike and matrix spike. Thus, a correction factor for the matrix corrected spike activity was determined:

$$A_{aliquot\ corrected} = A_{measured}\left[\frac{M_{Sample}}{M_{Matrix\ Spike}}\right]$$

Where:

$A_{aliquot\ corrected}$ – *the aliquot corrected gross alpha activity due to spike (dpm)*

$A_{measured}$ – *the measured gross alpha activity (dpm)*

M_{Sample} – *the mass of the Pu-239 standard deposited on the sample (g)*

$M_{Matrix\ Spike}$ – *the mass of the Pu-239 standard deposited on the matrix spike (g)*

Table 1: Experimental Sample Information

Air Filter	Spike Quantity (mL)	Spike Activity (dpm)	Air Exposure (L)
HiVol #1	0.243	292.0	10200
HiVol #2	0.254	305.3	10200
Lapel #1	0.2534	304.5	1804
Lapel #2	0.2498	300.2	1814
HiVol Matrix Spike	0.2541	305.4	0
Lapel Matrix Spike	0.2546	306.0	0

Results and Discussion

High Volume Air Samples

Two HiVol (approx. 10,000 L) air samples were taken from a non-ventilated space next to a low background vault. Figures 1 and 2 show the measured activity for each sample (solid lines) compared with the aliquot-corrected, matrix spike value (dotted lines). Initially, the samples were saturated with radon progeny which makes the gross alpha activity of the sample nearly an order of magnitude larger than the spike value. With the use of the Canberra radon progeny compensation algorithm, the "Compensated Gross Alpha" activity reported by the iSOLO™ much more accurately represents the synthetic alpha activity on the filter. However, the compensated value does not reach the average matrix spike value until a few hours have passed. The compensated gross alpha activity seems to be an over-estimate of the non-radon progeny activity within the first few hours. This may be due to the small presence of the short-lived radon progeny that are not compensated for in the algorithm. Despite this discrepancy, the algorithm proves to be effective in providing, within a few hours, an accurate estimate of the non-radon progeny gross alpha air concentration.

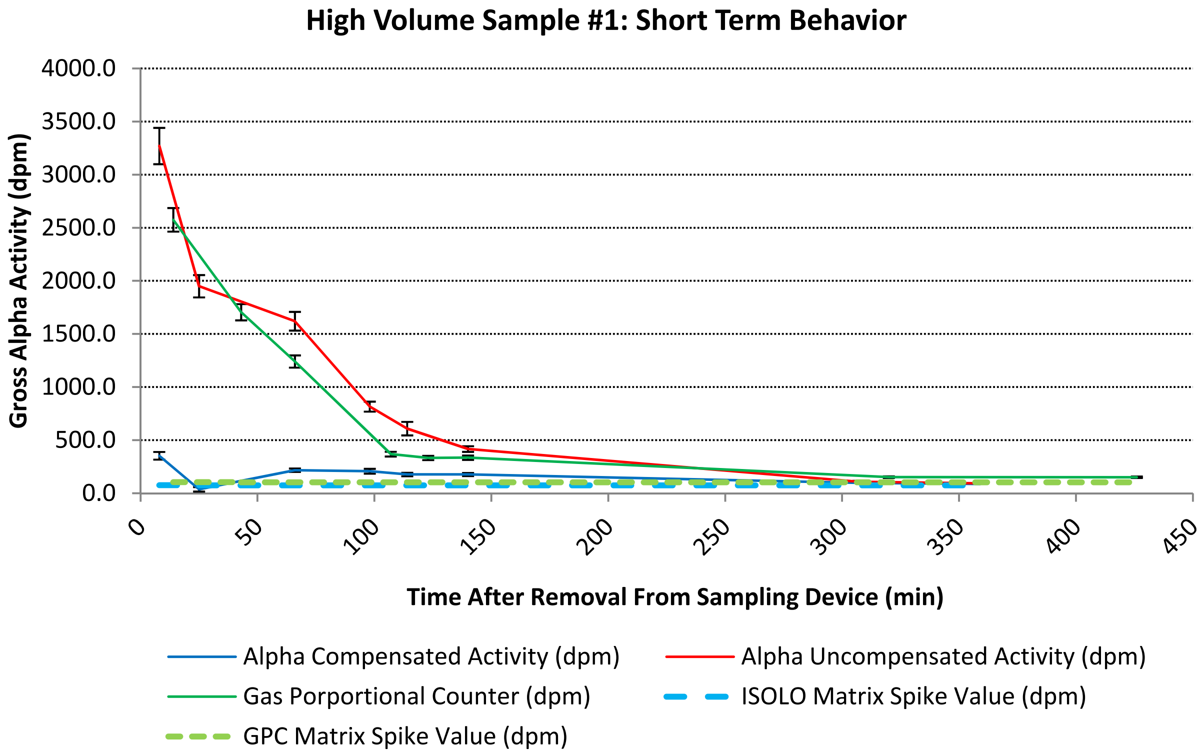

Figure 1: *HiVol #1 Gross Alpha Activity (dpm) as a function of time for counts made on the day of sampling. The solid green line represents measurements made on a GPC. The solid red line represents uncompensated measurements on the iSOLO™. The solid blue line represents compensated measurements on the iSOLO™. Error bars represent 1-sigma uncertainties.*

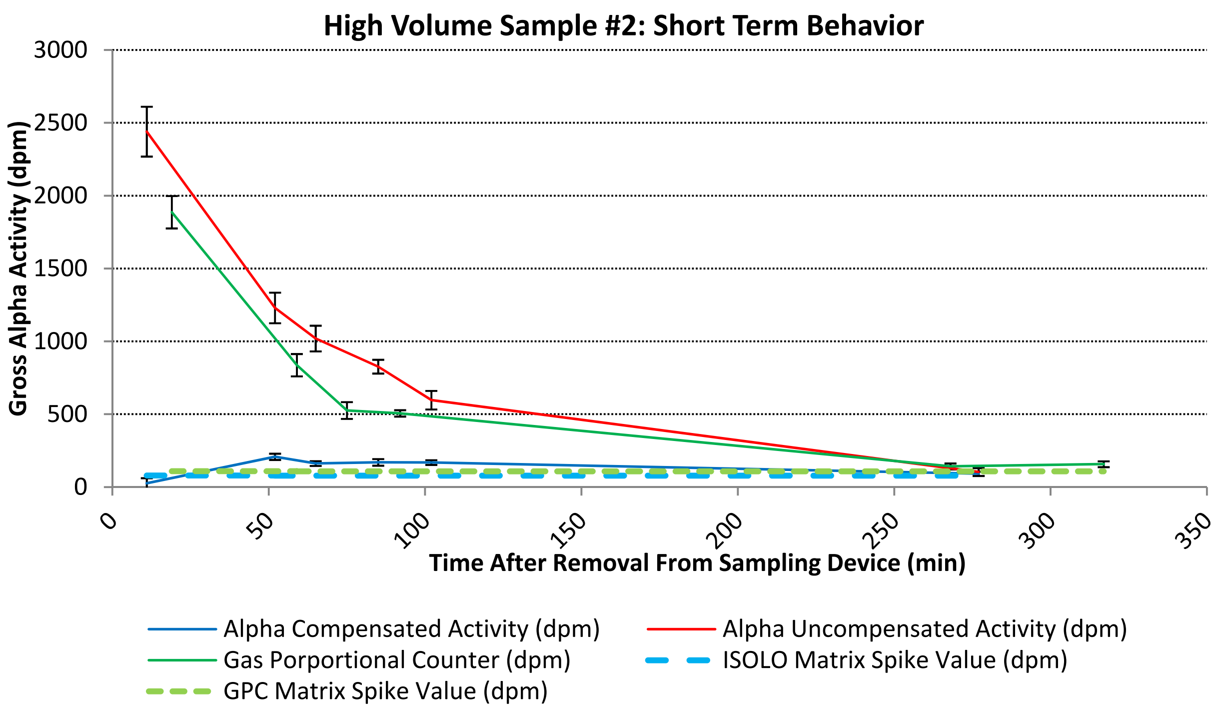

Figure 2: *HiVol #2 Gross Alpha Activity (dpm) as a function of time for counts made on the day of sampling. The solid green line represents measurements made on a GPC. The solid red line represents uncompensated measurements on the iSOLO™. The solid blue line represents compensated measurements on the iSOLO™. Error bars represent 1-sigma uncertainties.*

<u>Lapel Air Filter Samples</u>

Two duplicate air samples were taken in the same location using breathing zone air samplers. Figures 3 and 4 show the measured activity (solid lines) as well as the average matrix spike value (dotted lines) as a function of time over the day of sampling.

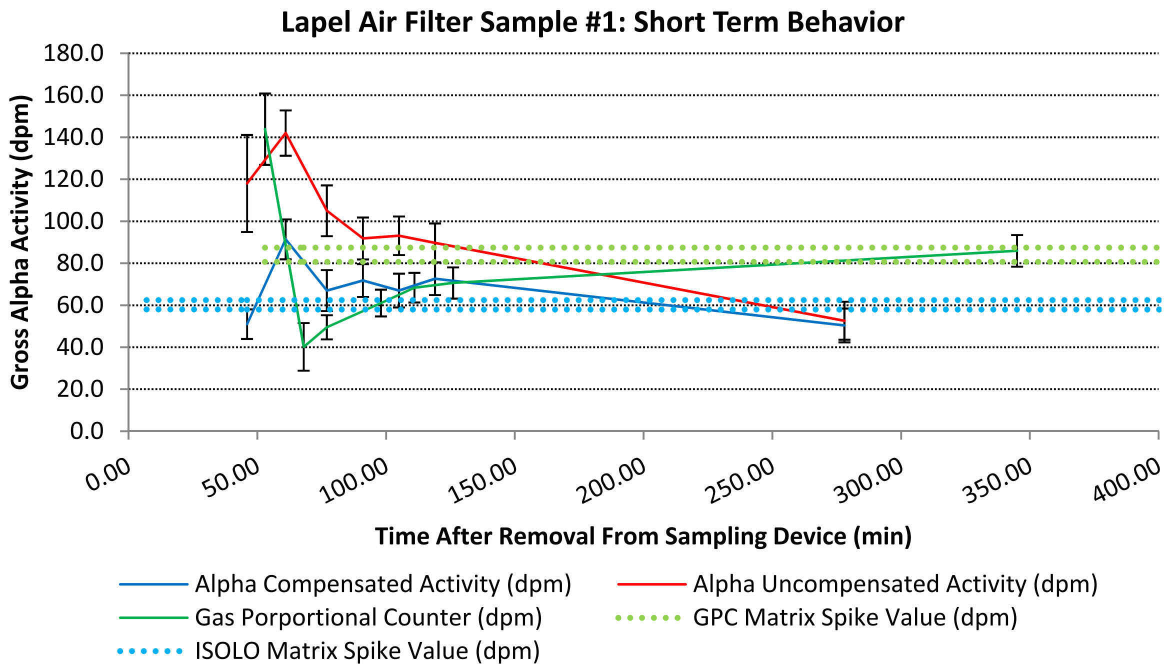

Figure 3: *Lapel #1 Gross Alpha Activity (dpm) as a function of time for counts made on the day of sampling. The solid green line represents measurements made on a GPC. The solid red line represents uncompensated measurements on the iSOLO™. The solid blue line represents compensated measurements on the iSOLO™. Error bars represent 1-sigma uncertainties.*

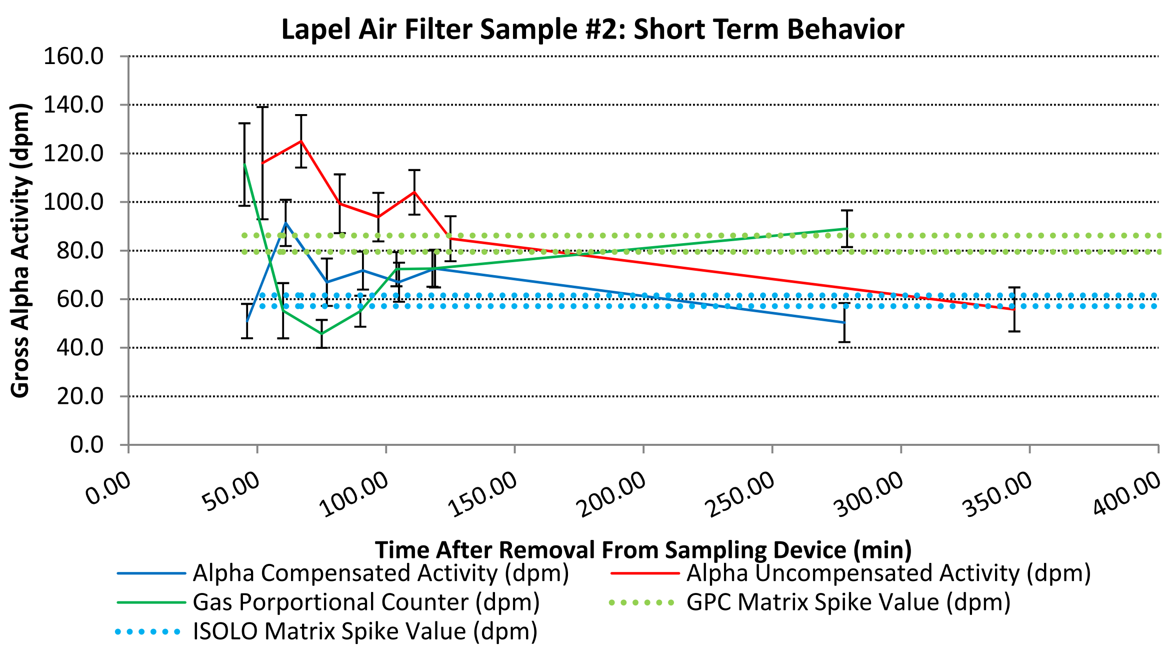

Figure 4: *Lapel #2 Gross Alpha Activity (dpm) as a function of time for counts made on the day of sampling. The solid green line represents measurements made on a GPC. The solid red line represents uncompensated measurements on the iSOLO™. The solid blue line represents compensated measurements on the iSOLO™. Error bars represent 1-sigma uncertainties.*

Note that the curves produced with these lapel filters are not quite as smooth as those produced from the high volume air samples. This may be explained by:

- The volume of air sampled on the lapel filters were on the order of 2000 L over an eight hour period. There is less radon progeny trapped on the filter thus increasing the statistical uncertainty of the measurements.
- The filter was completely wetted by the 0.25 mL ethanol spike which could have washed much of the radon progeny into the filter, unnaturally decreasing the radon progeny counts. The HiVol filter is larger in diameter and was not completely wetted during the spiking process.
- The lapel air filters used were roughly twice as thick and more porous then the HiVol air filters. This increased porosity could allow the radioactive materials present on the filter to migrate, causing the inconsistent detection efficiency for each count.

Despite the strange behavior of the activity vs. time curves shown in Figures 3 and 4, the radon progeny-compensated gross alpha activity of the filter approaches the matrix spike value rapidly (within the first 5 hours). This shows that for a lower radon progeny background (more representative of actual conditions) the iSERIES™ instruments are effective in producing a radon progeny-free gross alpha activity within the first day upon removal from the sampling device.

Decrease of Radon Progeny Activity

The gross activity due to radon progeny (uncompensated iSOLO™ counts minus the compensated iSOLO™ counts) found on HiVol Filter #1 was plotted as a function of time (Figure 5). The radon progeny activity is a smooth exponential function of time. The R^2 value of the fit was 0.9951. The decay constant of this exponential function corresponds to a half life of 37.7 minutes.

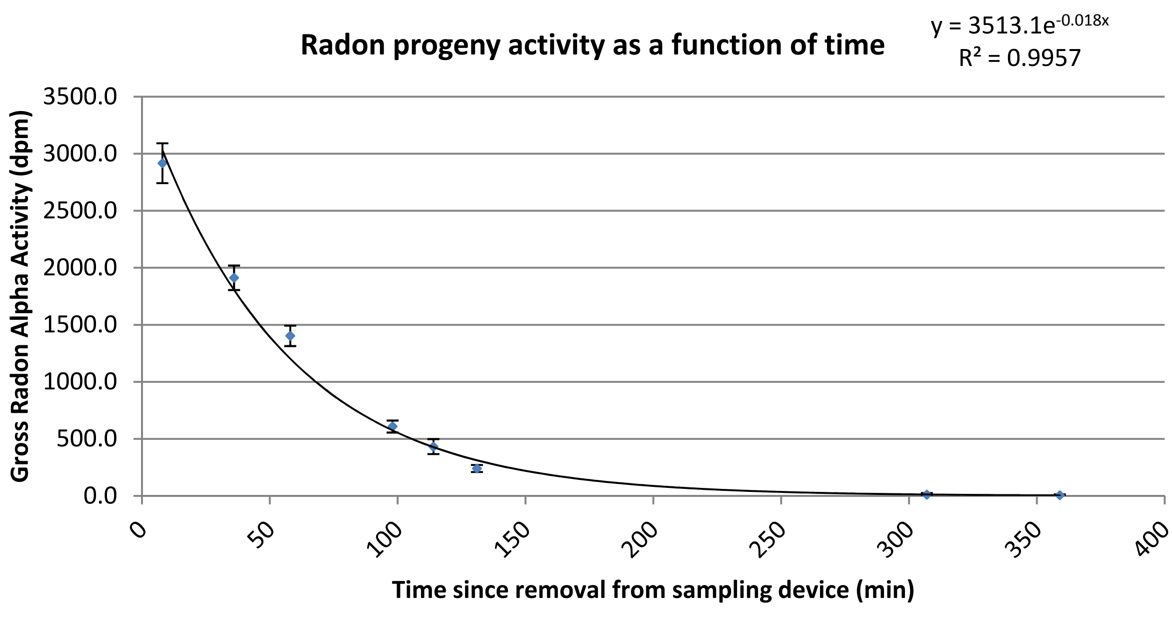

Figure 5: *Gross radon progeny activity as a function of time since removal from the sampling device*

To determine the cause of this observation, an energy calibration was needed for the iSOLO™ to provide an energy vs. channel relationship. The calibration was performed using a mixed alpha quality control source that contains U-233, Pu-239, Am-241, and Cm-244. As expected, the results show that energy is a linear function of channel number. Using this calibration, the energy of the unknown component of the radon progeny peak corresponds to an energy of ~7.6 MeV (Figure 6). A secondary peak was also observed (only on the preliminary count of HiVol#1) with an energy around 6 MeV. This peak was not seen in later counts and was assumed to have decayed below background levels.

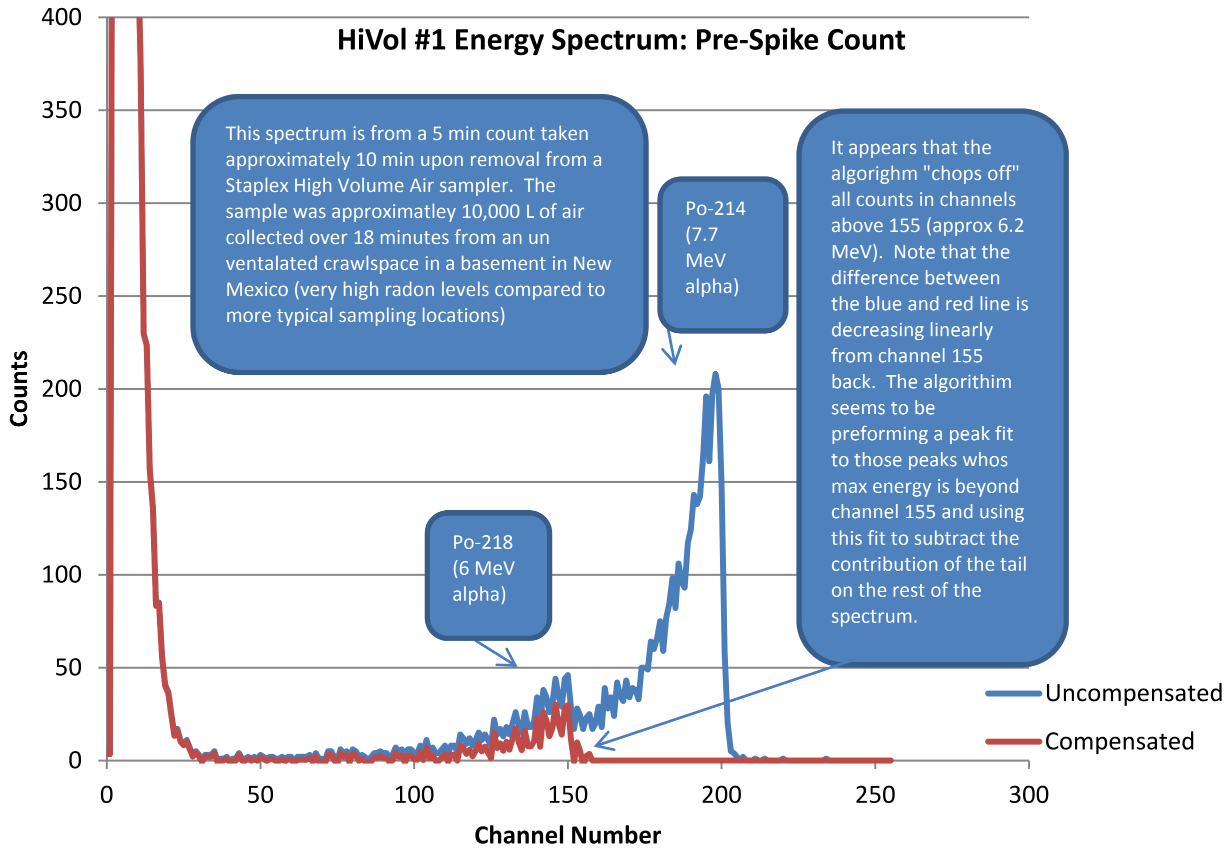

Figure 6: *iSOLO™ Energy spectrum taken over a 5 minute count roughly ten minutes upon removal from the Staplex High Volume Air Sampler*

Po-214 is one of the daughters in the U-238 decay series. It is formed by the beta-minus decay of Bi-214 with a half-life of 19.9 minutes. Po-214 has a half-life on the order of microseconds. Hence Bi-214 and Po-214 are in secular equilibrium, meaning that every time a Bi-214 beta decays, its daughter, Po-214 "immediately" decays by emitting a 7.6 MeV alpha particle. The detected alpha radiation observed from this process would appear to have a half-life corresponding to that of Bi-214.

The much smaller peak (with a maximum energy of around 6 MeV) is due to the decay of Po-218, the great-grandparent of Po-214. Po-218 has a 3 minute half-life which explains the rapid decrease in activity between the 1st and 2nd count of the air filter and its relatively small magnitude on the first count. The peak due to Po-218 was not compensated by the algorithm and was counted as unnatural alpha activity. The failure of the algorithm to compensate for this naturally occurring alpha emitter is not detrimental to the proposed use of this instrument. As long as air filters are given enough time for the

Po-218 to decay (about 30 min) the compensation algorithm seems to be effective in subtracting the contributions of the other longer-lived naturally-occurring radionuclides from the gross count.

Detection Levels: Minimum Detectable Activity (MDAs)

The sample-specific MDA calculated by the Canberra iSOLO™ software does not consider the counts due to radon progeny as part of the system background**,** which leads to artificially low minimum detectable activities. RPSD typically bases MDA estimations on the work of Lloyd A. Currie[1]. L.A. Currie defines the Critical Level (L_c) as:

Where:
L_C - The non-negative point on a normal distribution centered about zero where the percentage of the area under that distribution is equal to a desired confidence level
k – the number of standard deviations from zero (positive) to obtain a desired confidence level
- the expected uncertainty of a zero activity sample in counts per minute

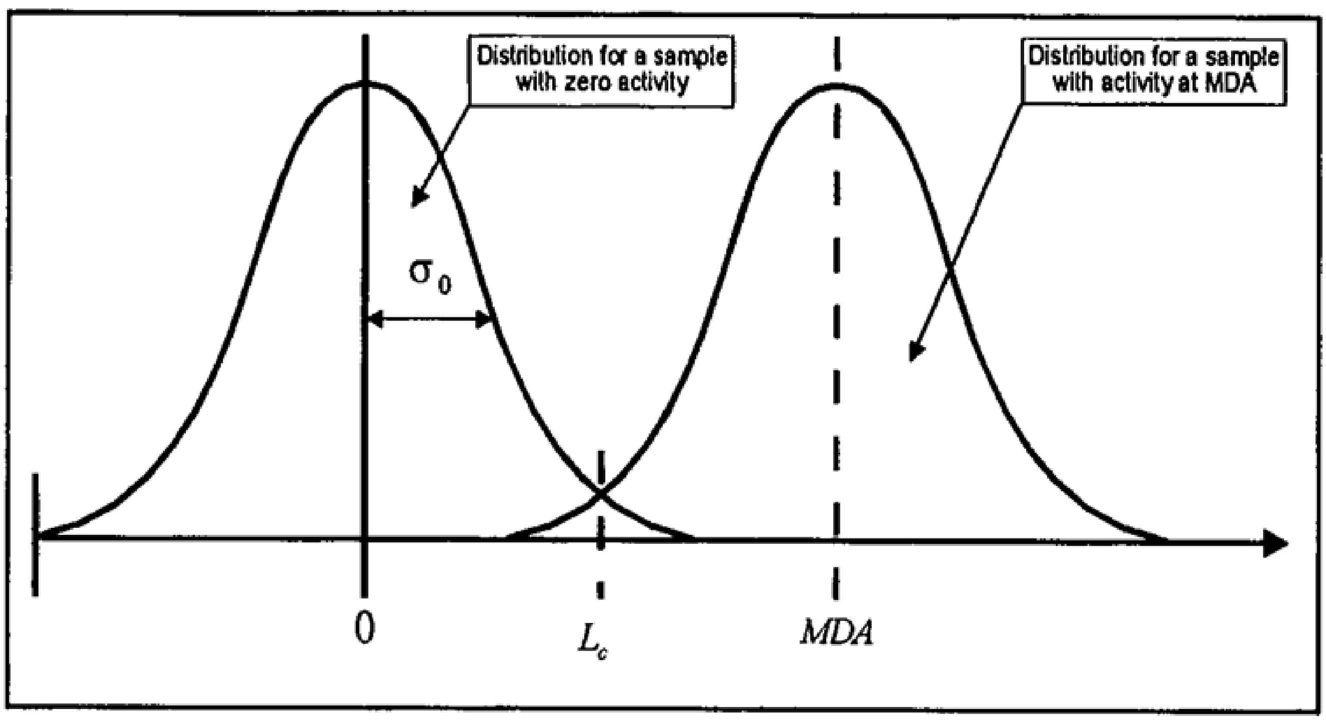

Figure 7: *A graphical representation of the concepts of Critical Level and Minimum Detectable Activity as defined by Lloyd A. Currie.*

The Canberra iSERIES™ instruments utilize adaptive spectrometry to correct for the presence of naturally occurring airborne radionuclides due to radon and thoron. The correction of gross counts that use this method involve not only system background subtraction but spectral "stripping" of unwanted natural counts. Thus, the expected uncertainty of a sample with "zero" activity (a sample with only system background and counts due to radon progeny that will be stripped from the gross activity) is sample-dependent. This value can be obtained by su mming in quadrature the uncertainty of the system background ***and*** the counts due to radon progeny, hence:

$$\sigma_0 = \sqrt{\sigma_{Bkg}^2 + \sigma_{RnTh}^2}$$

Where:

σ_{Bkg}^2 - *The predicted variance of the system background measurement*
σ_{RnTh}^2 – *The predicted variance of the naturally occurring radioactivity* measurement

And:

$$\sigma_{Bkg}^2 = \sigma_{R_S}^2 + \sigma_{R_B}^2$$

$\sigma_{R_S}^2 - Variance\ of\ the\ background\ count\ rate\ during\ the\ sample\ count$
$\sigma_{R_B}^2 - Variance\ of\ the\ background\ count\ rate\ during\ the\ system\ background\ count$

Or:

$$\sigma_{Bkg}^2 = \frac{R_B}{T_S} + \frac{R_B}{T_B}$$

$R_B - System\ background\ count\ rate\ [\frac{counts}{min}]$
$T_S - Sample\ count\ duration\ [min]$
$T_B - System\ background\ count\ duration\ [min]$

Since a count rate is simply the number of counts observed during a count divided by the duration of the count (in minutes) the expression becomes:

$$\sigma_{Bkg}^2 = \frac{N_B}{T_S T_B} + \frac{N_B}{T_B^2}$$

$N_B - The\ number\ of\ background\ counts\ observed\ in\ time\ T_B$
$T_B - The\ system\ background\ count\ time\ [min]$

$By\ factoring\ out$: $\frac{N_B}{T_S T_B}$, the expression becomes:

$$\sigma_{Bkg}^2 = \frac{N_B}{T_s T_B}\left(1 + \frac{T_S}{T_B}\right)$$

It can be shown that for cases in which the sample count time is equal to the background count time (which is the cas for σ^2_{RnTh}). The equation above reduces to:

$$\sigma^2_{RnTh} = \frac{2N_{RnTn}}{T_S^2}$$

Where N_{RnTn} is defined as the num ber of counts subtracted as interference in th e standard Region of Interest (ROI). Since the iSOLO™ and iMATIC™ does not report this value directly it must be computed using variables obtained from the iSERIES™' database,

$$\sigma^2_{RnTh} = \frac{2[\mathrm{N_{Raw}} - \mathrm{N_{Raw-RnTn}}]}{\mathrm{T}_S^2}$$

Where:

N_B – The sum of the system background counts obser ved in the alpha ROI during background count time, T_B

T_S – The sample count time

N_{Raw} – The sum of the sample counts observed in the alpha ROI during sample count time, T_S

$N_{Raw-RnTh}$ – The sum of the sam ple counts observed in the alpha ROI during sam ple count tim e T_S ***subtracted by*** the sum of the sample counts due to radon progeny interference in the standard ROI. This is equal to the total number of counts subtracted as interference in the standard ROI.

It follows that the MDA can be derived as a function of the confidence level fact or (k) and the critical level (L_C). The MDA is defined as the m ean of a nor mal distribution where a predeterm ined (by the k factor) probability exists such that sample can be said to have an activity above zero. The expression for MDA is as follows:

$$MDA = \frac{k^2}{\epsilon T_S} + \frac{2L_c}{\varepsilon}$$

Where:

ε - The detection efficiency (as a fraction)

The standard confidence level used by RPSD is 95% . To achieve such a confid ence level, the normal distribution is integrated from the mean (0 in this case) to kσ and set equal to 0.95. Then the equation is solved for k resulting in: k = 1.645. Substituting this value for k in the expression for L_C, one obtains the critical level and minimum detectable activity at 95% confidence (L_{C95} and MDA_{95} respectively):

$$L_{C_{95}} = \frac{1.645}{\epsilon}\sqrt{\frac{N_B}{T_sT_B}\left(1+\frac{T_S}{T_B}\right)+\frac{2[\mathrm{N_{Raw}}-\mathrm{N_{Raw-RnTn}}]}{\mathrm{T_s^2}}}$$

$$MDA_{95} = \frac{(1.645)^2}{\epsilon T_S} + \frac{2(1.645)}{\epsilon}\sqrt{\frac{N_B}{T_sT_B}\left(1+\frac{T_S}{T_B}\right)+\frac{2[\mathrm{N_{Raw}}-\mathrm{N_{Raw-RnTn}}]}{\mathrm{T_s^2}}}$$

In terms of the iSERIES™ database variables:

For Alpha:

$$\boldsymbol{L_{C_{95}}} = \frac{1.645}{ALPHAEFFICIENCY}\sqrt{\frac{ALPHABACKGROUNDCOUNTS}{(COUNTTIME)(BKGCOUNTTIME)}\left(1+\frac{COUNTTIME}{BKGCOUNTTIME}\right)+\frac{2[\mathrm{ALPHASTDRAWCOUNTS}-\mathrm{ALPHASTDCOUNTSVALUE}]}{(\mathrm{COUNTTIME})^2}}$$

$$\boldsymbol{MDA_{95}} = \frac{(1.645)^2}{(ALPHAEFFICIENCY)(COUNTTIME)} + \frac{2(1.645)}{ALPHAEFFICIENCY}\sqrt{\frac{ALPHABACKGROUNDCOUNTS}{(COUNTTIME)(BKGCOUNTTIME)}\left(1+\frac{COUNTTIME}{BKGCOUNTTIME}\right)+\frac{2[\mathrm{ALPHASTDRAWCOUNTS}-\mathrm{ALPHASTDCOUNTSVALUE}]}{(\mathrm{COUNTTIME})^2}}$$

For Beta:

$$\boldsymbol{L_{C_{95}}} = \frac{1.645}{BETAEFFICIENCY}\sqrt{\frac{BETABACKGROUNDCOUNTS}{(COUNTTIME)(BKGCOUNTTIME)}\left(1+\frac{COUNTTIME}{BKGCOUNTTIME}\right)+\frac{2[\mathrm{BETARAWCOUNTS}-\mathrm{BETACOMPCOUNTSVALUE}]}{(\mathrm{COUNTTIME})^2}}$$

$$\boldsymbol{MDA_{95}} = \frac{(1.645)^2}{(BETAEFFICIENCY)(COUNTTIME)} + \frac{2(1.645)}{BETAEFFICIENCY}\sqrt{\frac{BETABACKGROUNDCOUNTS}{(COUNTTIME)(BKGCOUNTTIME)}\left(1+\frac{COUNTTIME}{BKGCOUNTTIME}\right)+\frac{2[\mathrm{BETARAWCOUNTS}-\mathrm{BETACOMPCOUNTSVALUE}]}{(\mathrm{COUNTTIME})^2}}$$

For the most common use of the iSERIES™ instruments, the ROI is the standard window that encompasses alpha particle energies between 3 – 6.4 MeV. As can be seen in Table 2, the true MDA calculated using the derived equation above is much larger than the MDA reported by the vendor software. These large MDA values are typically not acceptable for dosimetry-based decisions. Thus, for more statistically-viable results, in situations where there is are substantial radon progeny present in the sample, the user must wait up to 6 hours and perform a 30 minute count.

Table 2. Minimum Detectable Activity*					
Time Upon Removal (hr)	Count Time (min)	Radon Progeny Activity (dpm)	Compensated Alpha Activity (dpm)	Reported MDA (dpm)	Actual MDA (dpm)
1.1	5.00	1403	217.4	3.6	74.8
1.6	5.00	608	207.7	3.6	45.4
1.9	5.00	432	177.0	3.6	37.5
2.3	5.00	240	176.5	3.6	26.1
5.1	5.00	10.2	97.8	3.6	6.7
6.0	30.00	5.6	87.9	1.3	2.4
29.5	30.00	0.9	83.9	1.3	1.0

** Minimum Detectable Activity as a function of time removed from sampling device*

Conclusions

In summary, the iSERIES™ instruments can meet the requirements of a dosimetry-decision measurement of gross alpha concentration in air. This study also showed that the Canberra iSERIES™ instruments with the radon progeny compensation algorithm can be used to analyze air filters from an area with large amounts of naturally occurring environmental radioactivity and provide a relatively good[i] estimate of airborne contamination levels within 6 hours upon removal from the sampling device. However, the minimum detectable activity reported by the iSERIES™ instruments is incomplete in that it does not consider the subtracted radon progeny as background in the measurement. Due to this error, a different approach was taken where the counts due to progeny were considered as part of the background and the equation of minimum detectable activity was re-derived. Based on these findings and considerations, the best approach (and the one that will be used at RPSD) is as follows:

1. Obtain filter sample and count on iSOLO™ or iMATIC™ for 5 minutes for purposes of screening.
2. Observe spectrum, report any unusual peaks not due to natural background.
3. Wait 4-6 hours, count again on an iSOLO™ or iMATIC™ instrument for 30 minutes, observe spectrum, report values to customer.
4. Retain the filter if alpha/gamma spectrometry is subsequently required to obtain an even lower detection level.

i: The iSOLO™ yields a compensated MDA of 2.39 dpm and the GPC yields an MDA of 2.2 dpm for a 30-minute count 6 hours after removal from the sampling device.

References

[1] L. A. Currie "Limits for Qualitative Detection and Quantitative Determination: Application to Radiochemistry" Analytical Chemistry Vol. 40, No. 3, March 1968

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