Methods for Reduction of the Minimal Detectable Activity in Neutron Assay of High Mass/High Z Objects – 14504

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ABSTRACT

The Minimum Detectable Activity (MDA) for neutron counting techniques is usually governed by cosmic ray related background effects. In the case of assay of high mass objects where the constituent materials have high atomic number (Z>20) the neutron background will increase due to spallation events, which is often known as the “ship effect”, a term that was coined to describe the phenomenon of observed increases in background when neutron counters were taken on board large ships.

The MDA is a statistical concept that inherently reflects the variability in the background (i.e. between sample and background measurements). In the case of neutron background it is often found, over the course of a day, that backgrounds can vary by a considerably larger degree than would be predicted from Poisson statistics alone. The underlying cause of this variation is often not well known, but is likely to be driven by extraterrestrial phenomenon (e.g. solar activity), diurnal fluctuations, geomagnetic effects, weather and barometric pressure. Corrections for these effects could yield real-time methods for reducing MDA. For example, the barometric pressure is a direct measure of the mass of air overhead which is in turn related to the flux of cosmic rays at ground level.

Several methods for suppression of the ship effect are also available that can further reduce the MDA of any given technique. In particular the use of time correlation to identify pulses of neutrons associated with spallation may be employed with a variety of statistical filtering methods.

A study was performed in association with URS | CH2M Oak Ridge LLC (UCOR) on the potential improvements in MDA that can be achieved through the use of various methods for background compensation and reduction. A large background data set was evaluated in various applications.

The implications of this study indicate that the reduction in MDA for assay of large objects has significance for waste consignors who are seeking to categorize and classify waste according to its radioactivity and for the exemption of small quantities of fissile material. Particular benefits may be realized for decommissioning of gaseous diffusion plant waste arisings and in low-level / intermediate / transuranic waste segregation. Widespread adoption of this technology could yield cost benefits and improved operational efficiency for waste management professionals. Further to this, the principles of this method may also be applicable for improving the performance capability of neutron radiation portal monitors for the interdiction of illicit transportation of nuclear materials.

INTRODUCTION

Neutron counting systems are routinely used for screening and segregation of radioactive waste [1]. The Minimum Detectable Activity (MDA) of the equipment must be below the screening threshold and therefore optimization of the MDA is a key design requirement. MDA is governed by background rate, efficiency, and count time. The latter two parameters are often limited by cost and operational throughput requirements. Therefore, methods for reducing the background or changing its daily variability can yield significant advantages for waste management.

Cosmic ray related effects will often be a dominant source of background, particularly at locations that are isolated from stored waste or other neutron sources. In the case of assay of high mass objects where the constituent materials have high atomic number (Z>20 e.g. steel, copper, lead) the neutron background will increase due to spallation events.

For a given count time and efficiency, the MDA depends on the variability in the background (i.e. between sample and background measurements). The background is normally measured on a daily basis, but over the course of a day its magnitude can vary by a larger degree than would be predicted from Poisson statistics alone. Contributing effects include solar activity, diurnal and geomagnetic fluctuations as well as weather (barometric pressure). The uncertainty in the background could be reduced by increasing the frequency of its measurement (e.g. 3-4 times per operational day), but this strategy yields diminishing returns, due to the interruption of operations.

For assay systems that use coincidence counting, various statistical filtering methods have been deployed to reduce the cosmic background such as high order multiplicity rejection (relying on the fact that cosmic spallation tends to produce large bursts of neutrons) and the “local coincidence veto” method (cosmic interactions in the chamber walls tend to be detected in neighboring detector banks) [2]. However, in the case of total neutron counting where multiplicity / time stamped information may not available, such methods are of limited applicability.

A promising method has been identified to reduce a system’s MDA in real-time, by correcting for the background variation using various proxy measurements (e.g. pressure / solar activity). This method is applicable in a wide variety of neutron counting systems, but is particularly focused to total neutron instruments such as the slab counter.

NEUTRONS FROM COSMIC RAYS

Neutrons measured at ground level from cosmic ray sources are generated by various reactions. Cosmic rays enter the upper atmosphere as charged particles (e.g. protons, alpha particles) with energies >300 MeV [3]. A variety of cascade reactions in the atmosphere deliver a broad spectrum of particles to the Earth’s surface. Muons and secondary neutrons are thought to be largely responsible for reactions that lead to counts in ground based neutron detectors [4].

As the atmosphere acts as a shield, cosmic flux is highly dependent on the instrument’s elevation. Also, the Earth’s magnetosphere deflects incoming particles into a curved path. This results in different energy spectrum for different latitudes and longitudes, controlled by a geomagnetic cutoff, with minimum value at the magnetic poles. This is usually parameterized in terms of the effective vertical geomagnetic cutoff rigidity (momentum per unit charge) given in units of GV.

The cutoff rigidity contour plot for North America is shown in Figure 1. The value reaches a maximum at the magnetic equator. For a given type of particle, there is a cutoff energy (E_c) below which the particle will not be capable of entering the atmosphere as illustrated in Figure 2.

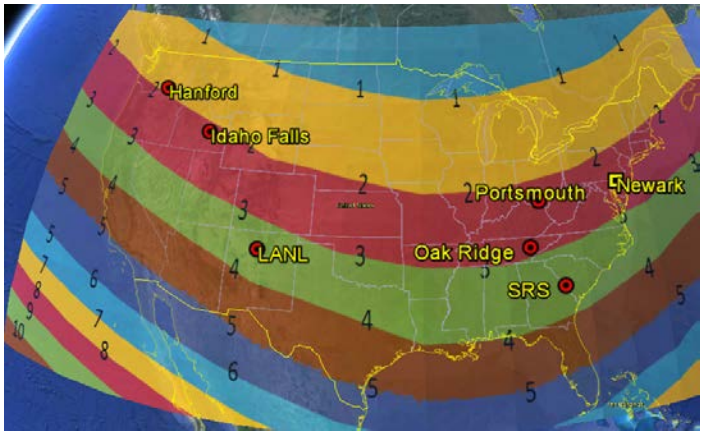

Fig. 1. Effective Geomagnetic Rigidity Cutoff (GV) for North America

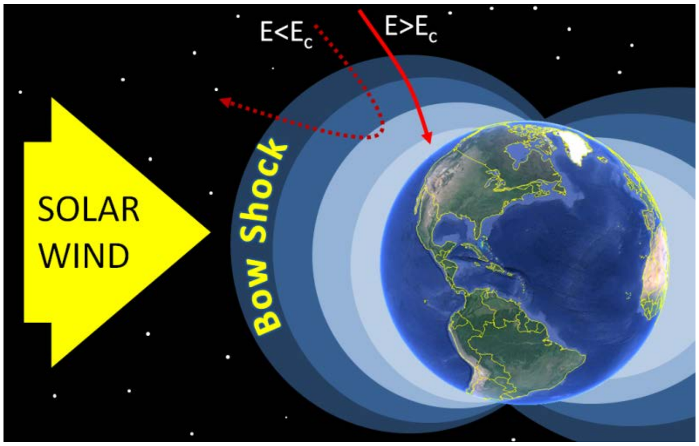

Fig. 2. Illustration of Cut-Off Energy (E_c) for Cosmic Protons

The rigidity cutoff at various worldwide sites is given in Table I. The final column gives the neutron flux (relative to Newark, Delaware, USA) - a function of both cutoff rigidity and elevation [5].

TABLE I. Effective Geomagnetic Cutoff Rigidity and Relative Neutron Flux at Various Sites

Location	**Effective Geomagnetic Cut-Off Rigidity (GV)**	**Altitude (m)**	**Relative Neutron Flux (/Newark)**
Chiang Mai, Thailand	16.8	2560	2.88
Rokassho, Japan	8.8	0	0.67
Beijing, China	8.7	446	0.97
Rome, Italy	6.3	60	0.81
Los Alamos, NM, USA	3.5	2182	4.87
Oak Ridge, TN, USA	3.0	280	1.20
Portsmouth, OH, USA	2.2	205	1.15
Newark, DE, USA	2.1	50	*1.00 (Ref)*
Richland, WA, USA	2.0	118	1.07
Idaho Falls, ID, USA	2.0	1500	3.41

The solar wind shapes and modifies the geomagnetic field. Consequently, as solar activity varies with time, the flux and energy spectrum of cosmic rays will also vary. The impact of coronal mass ejections, often leads to a decrease in observed neutron count rates at various monitoring stations around the world, an effect known as the Forbush decrease [6]. This reduction is due to the interaction of the charged particles (in the solar wind) with the Earth's geomagnetic field which often results in a net reduction in cosmic ray neutron flux at ground level. Count rates are typically reduced by 10-20% and gradually return to normal after 8-12 days. The onset of the event occurs approximately 18-20 hours after the solar flare is detected. This delay is due to the transit time of the charged particles between the Earth and Sun.

A Forbush event that occurred in October 2003 is illustrated in Figure 3. Data points are plotted from the Bartol Institute's Neutron Monitor in Newark, Delaware. This event was preceded by a solar fare of magnitude X17 (X-class flares are the highest classification).

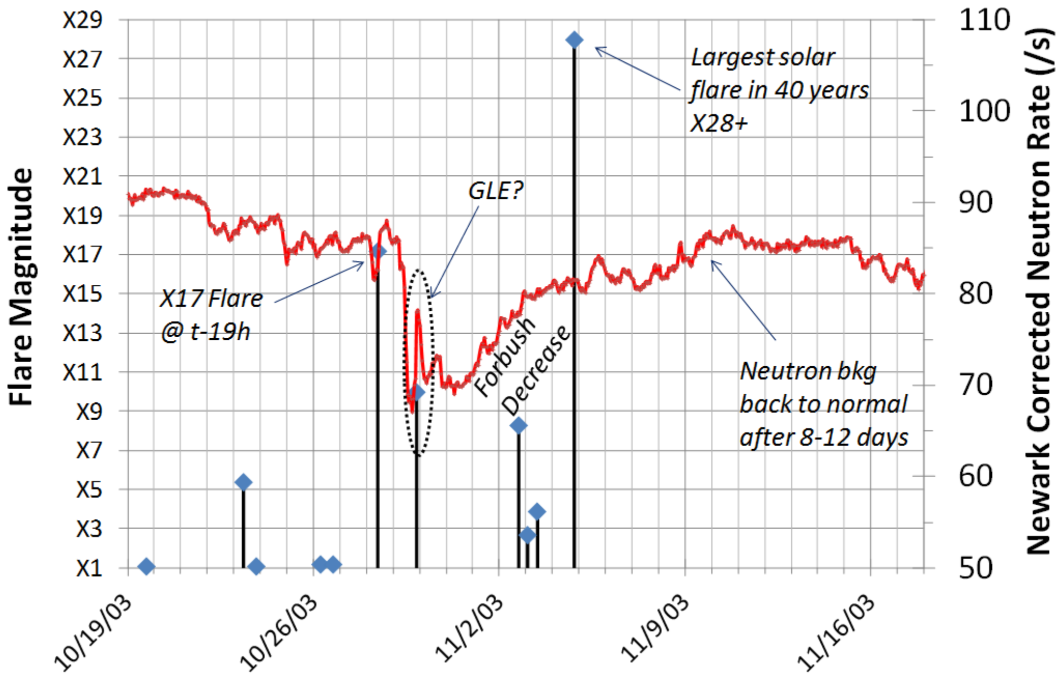

Fig. 3. Newark Count Rate During Major Solar Flare (October 2003)

There are several interesting features of this event that merit discussion. Firstly, a short lived peak occurs just after the decrease minimum and appears to be associated with a magnitude X10 flare. This type of increase is known as a Ground Level Event (GLE) which is associated with the arrival of particles from the Sun travelling at relativistic speeds. GLEs are much rarer than Forbush decreases. Secondly, a very high magnitude flare occurred on November 4th 2003 (X28 or higher) which has no observable impact on the Newark monitor count rate. In fact many X-class flares have no effect on neutron background rates and, conversely, many short-term temporal fluctuations are not associated with observed flares at all.

The counter at Newark uses 9 large BF_3 detectors surrounded by a lead collar inside a polyethylene box [7]. It has performed continuous neutron counting since 1964 as part of a worldwide array of similar counters. The locations are shown in Figure 4, with more than 30 monitors still operating in 2013.

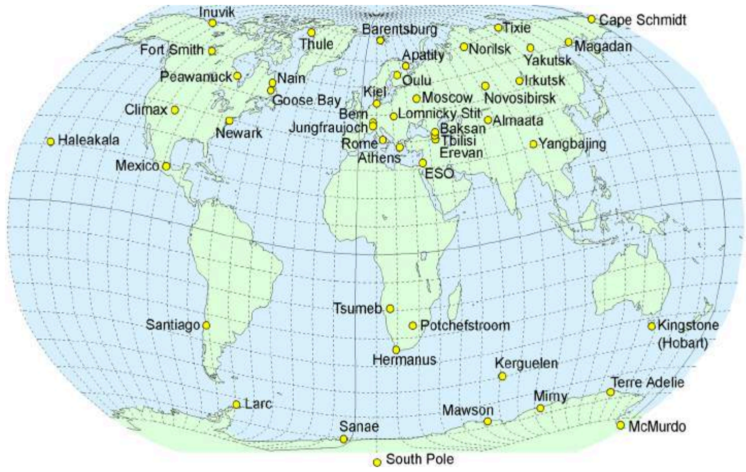

Fig. 4. Locations of various cosmic ray neutron monitor

Data from Newark (see Figure 5) provides an excellent demonstration of the long term relationship between ground level neutron flux and solar activity over 4 solar cycles (>44 years). This is co-plotted with the incidence of individual x-class flares and the annualized sum flare rate. Also shown (from 1997 onwards) is the annualized solar x-ray flux as measured with the Geostationary Operational Environmental Satellite system (GOES) operated by the US National Oceanic and Atmospheric Administration [8].

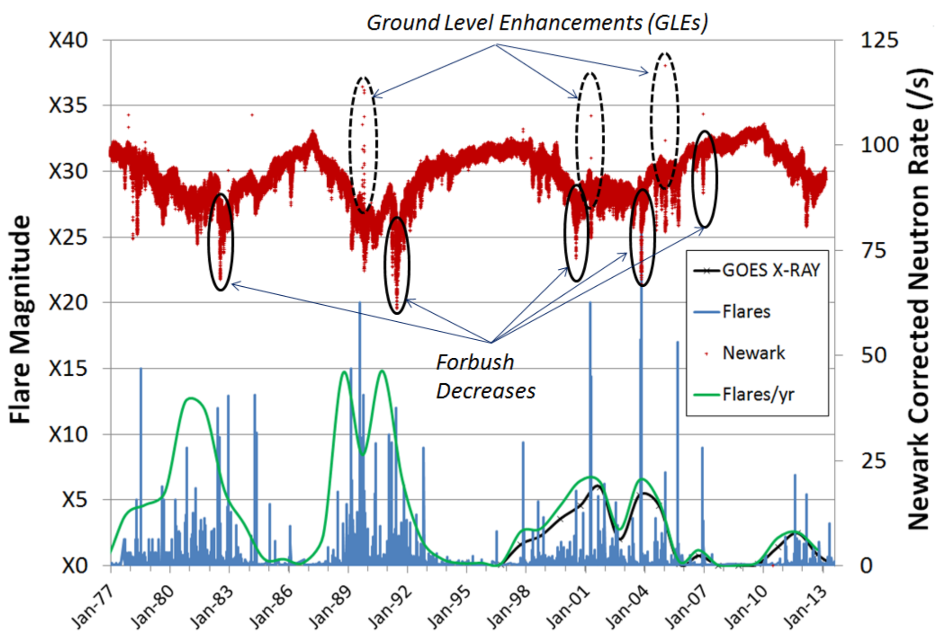

Fig. 5. Solar Flares / GOES X-ray Flux with Newark Count Rate (1977-2013)

Figure 6 (based on Newark data) indicates a clear relationship between solar flare magnitude and the Forbush reduction in count rate at minimum value. It can therefore be seen that the effect will be less than 5% decrease for a magnitude X2 flare or lower.

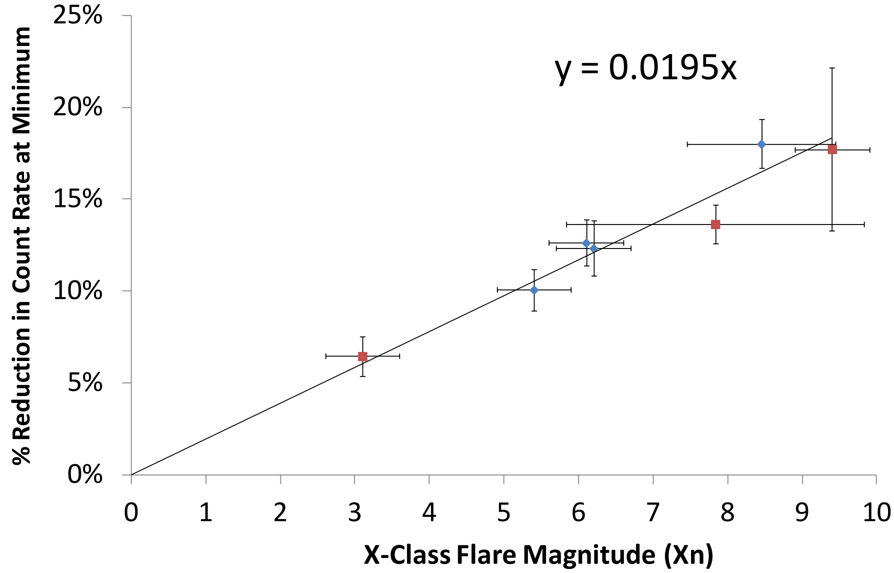

Fig 6. Correlation between Forbush Decrease in Count Rate and X-Class Flare Magnitude

Clearly, the solar activity is partly responsible for short and long term variations in neutron background rates. Real-time data (e.g. from satellite x-ray observations) could be used as an indicator of (possible) short term variation in background rate over a 24 hour period, but the user must take care to not place over-reliance on this method. For example, it may be appropriate to instigate special treatment on assays acquired during a 48 hour period after flares of magnitude X3 or above; this may require re-assay of containers where the result is close to the MDA.

The frequency of various classes of flare is shown in Figure 7. It can be seen that during active solar periods there are an average of 6 flares per annum of magnitude X3+. During solar low activity periods this type of flare occurs approximately once per annum. Periods of high and low activity last for roughly 5.5 years during the 11 year solar cycle. The next low period is predicted to start in 2016.

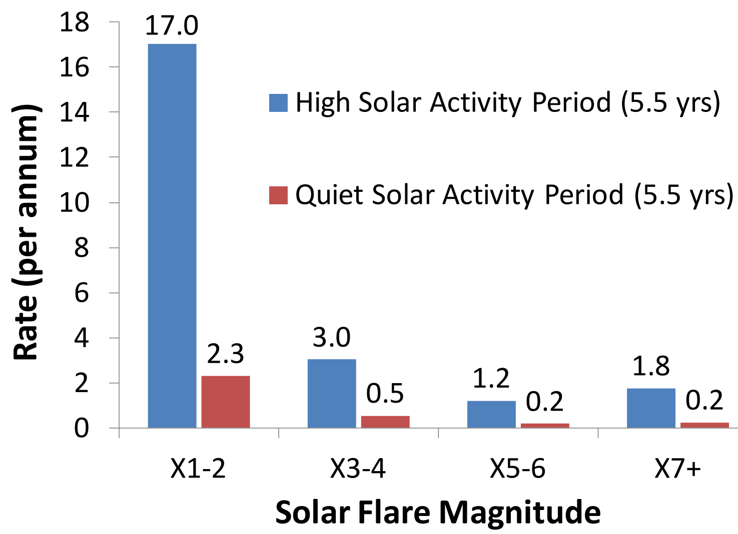

Fig. 7. Annual Rate of Occurrence of X-Class Solar Flares

BAROMETRIC PRESSURE CORRECTION

Weather can have a significant effect on cosmic neutron background. A dependency is known to exist between surface neutron count rate and the local barometric pressure (a measure of the overhead shielding air mass). A simple correction function (Equation 1) can be used to derive the background rate (B^*) at a reference pressure value (P_0) based on the measured pressure P:

$$B^* = Be^{\beta(P-P_0)} \quad \text{(Eq. 1)}$$

P_0 is usually set to the pressure at the start of the day (when the background is measured). β can be determined at the measurement location by taking a series of background and pressure measurements. Continuous measurements over a 48 hour period (e.g. on a weekend) is usually sufficient to provide a good estimate of β. The procedure simply involves plotting the natural logarithm of background rate against pressure and determining the slope by linear regression.

To refine the estimate of β, additional data may be accumulated over the representative period of operation, however some care must be taken to ensure that the cosmic ray term has remained reasonably constant during the accumulation period. For example, this requirement may not be satisfied during periods of very high solar activity.

The assumption of invariant cosmic flux can be checked by cross-reference of the data to neutron backgrounds acquired over the same period with a similar counter. The reference counters should be of a similar type, elevation and rigidity cutoff to each other to ensure that the flux and energy spectra of the primary and secondary particles are similar. For example, by inspection of Table I it can be seen that the Newark monitor represents a good proxy for many assay systems located at United States DOE sites including Hanford, Portsmouth and Oak Ridge.

It may be possible to use the proxy data to correct the background observed in assay equipment. Clearly the data from cosmic ray monitors such as Newark will not necessarily be available on a real-time basis, but one application could be to derive a more accurate estimate of β by performing a proxy monitor correction over the period used to establish the pressure correction.

DIURNAL BACKGROUND VARIATION

It has long been known that a small variation of approximately +/- 0.3% occurs in the cosmic neutron rate over a 24 hour period (the diurnal effect) due to Earth's motion through the interplanetary magnetic field [3].

Figure 8 shows the change in daily count rate Δ (normalized to the respective daily mean) averaged for each hour of the day over the period 2001-2012 for Newark. The data can be fit to local time of day (t) using the function in Equation 2 with a major diurnal (24 hour period) sinusoidal harmonic and a minor semi-diurnal (12 hour period) harmonic.

$$\Delta = A_1 Cos\left(\frac{2\pi(t-\phi_1)}{24}\right) + A_2 Cos\left(\frac{2\pi(t-\phi_2)}{12}\right) \quad \text{(Eq. 2)}$$

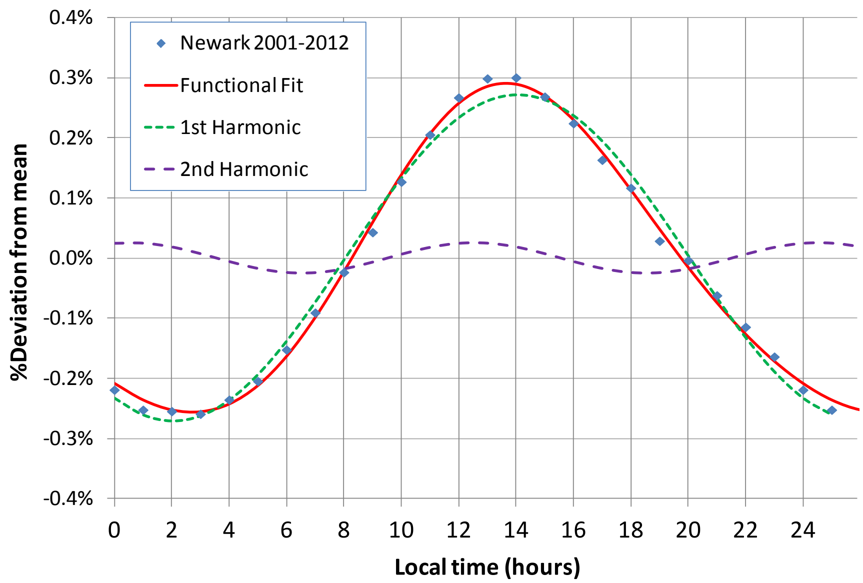

Fig. 8. Diurnal Variation in Newark Count Rate

The peak occurs approximately 1 hour after local noon. The functional fit parameters are given in Table II calculated over 3 solar cycles. The data from Newark also exhibits seasonal variation (+/- 0.6% peaking in February) and solar cycle variation (+/- 6% with the peak occurring during the 11-year solar cycle minimum).

TABLE II. Functional Fit Parameters for Diurnal Cosmic Neutron Variation (Newark)

Solar Cycle #	**21-22**	**22-23**	**23-24**
Start	9/19/1979	10/24/1990	11/27/2001
Mid-point	4/6/1985	5/10/1996	6/15/2007
Finish	10/23/1990	11/26/2001	12/31/2012
Diurnal Parameters			
A1	0.300%	0.263%	0.271%
$\phi 1$	14:01	12:42	14:02
Semi-Diurnal Parameters			
A2	0.036%	0.034%	0.026%
$\phi 2$	13:16	12:24	12:31
Functional Fit Parameters			
Amplitude	0.303%	0.265%	0.274%
Peak Time	13:46	12:32	13:38
Data %RSD	0.218%	0.191%	0.197%

ASSAY SYSTEM LONG TERM BACKGROUND STUDY

A long term neutron background study was performed using the UCOR (URS | CH2M Oak Ridge LLC) Uranium Neutron Counting System (UNCS). This system consists of a large concrete enclosure with arrays of polyethylene-moderated He-3 neutron detection tubes mounted on each interior facet. The UNCS is effectively an array of slab counters. The left and right walls and the floor and ceiling each have sixteen detectors attached. The back wall and the door of the system each have eight attached. The signals from all of the detectors are combined together to provide a single total neutron count which is then used to determine the uranium content of waste items using assumed enrichment and chemical composition. A standard background count is 1800 seconds long. The UNCS is designed to measure large metallic items contaminated with uranium compounds by measurement of neutrons emitted from (alpha,n) reactions. The large measurement chamber is capable of assay of items as large as intermodal containers.

The UNCS software was configured to gather long term background data that is typical of a total neutron counter. When the system was not performing operational measurements, a rolling series of automated 1800 second background counts were acquired and saved to a database. Backgrounds were continually acquired in this manner for a period of 8 months in 2011-2012.

The pressure correction function (Equation 1 with β=0.0075) was applied to the UNCS data. The result is shown in Figure 9 (corrected and uncorrected data). A Forbush event occurred following an X4 flare (March 7th 2012) and can be observed in this data set. A few (high) outliers are observed which are believed to be due to movement of waste containers near to the assay system

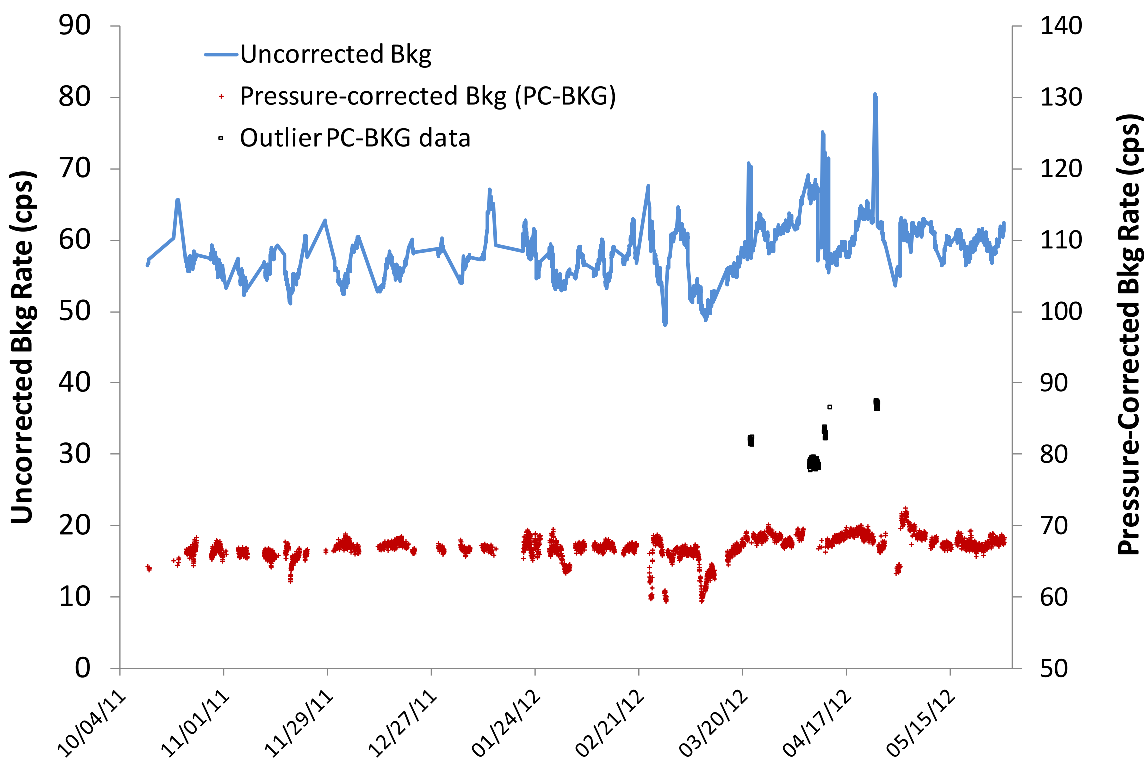

Fig. 9. Effect of Pressure Correction on UNCS Backgrounds

The UNCS backgrounds are plotted in Figure 10 over the course of the 24 hour day. The plot shows the background rate relative to the daily mean. Each point represents the hourly average over the 8 month study period. Also plotted are the equivalent average barometric pressure points relative to the daily average pressure. Clearly, it can be seen that pressure and uncorrected background are in anti-phase to each other. A variation of approximately +/-0.13% in pressure over the day (or +/-13 mbar) leads to a change in background of +/-1.0%. The effect of pressure correction is illustrated here (squares in Figure 10) such that daily variation in background is smoothed out.

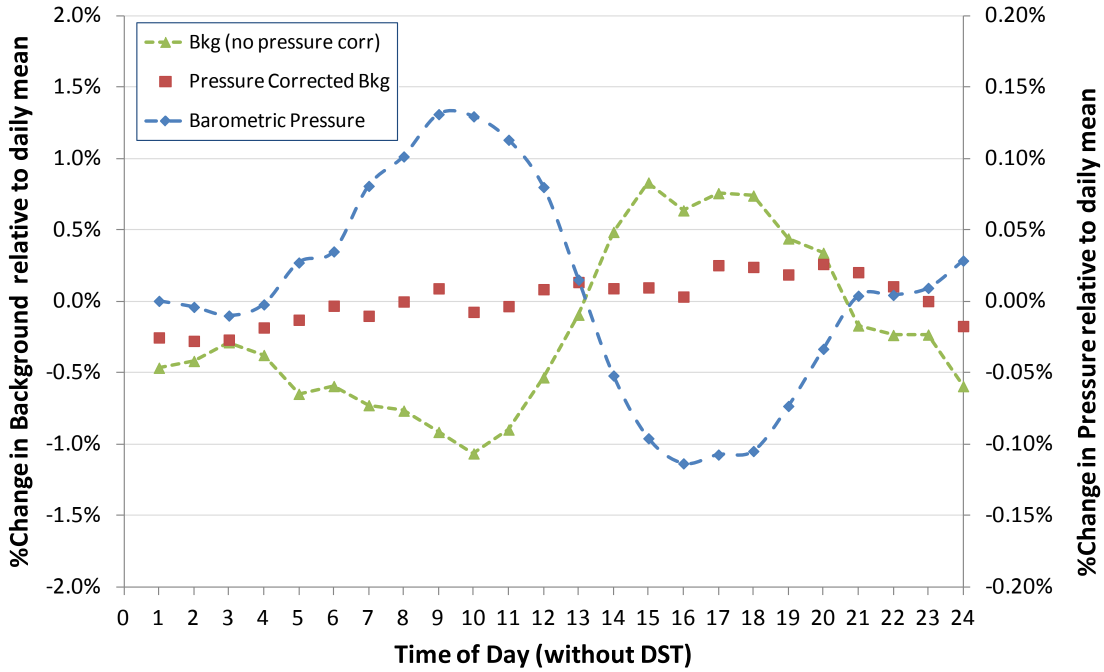

Fig.10. Daily Background Variation for UNCS (Oct 2011 - May 2012)

More detail is provided in Figure 11 which shows daily variation of the pressure corrected points (squares) co-plotted with the Newark diurnal function (solid line) from Equation 2, and the actual measured rate from Newark over the equivalent study period (dotted line). The UNCS data are roughly in agreement with the Newark diurnal fluctuations albeit with some minor discrepancies which are likely due to the effect of local operations (e.g. movements of waste containers at certain times of day).

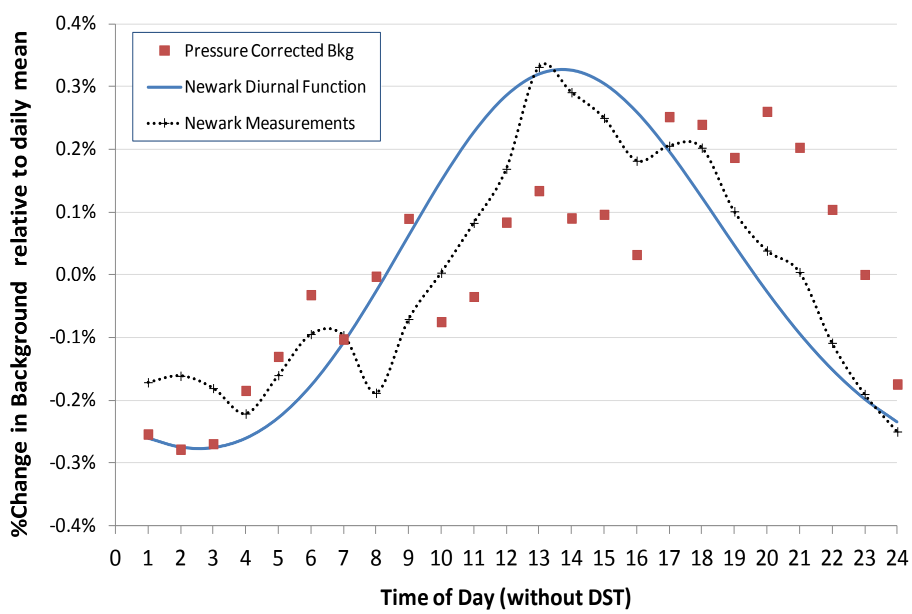

Fig. 11. UNCS and Newark Daily Backgrounds compared to Diurnal Function (Oct 2011- May 2012)

Over a given short-term period (a few days), the correlation between pressure corrected UNCS data and Newark data is good. See for example, Figure 12 that presents data for the end of October 2011. The Pearson product-moment correlation coefficient [9] between the data sets is 0.717 for this time period (a value of 1.0 indicates perfect linear correlation). The plot shows the expected Poisson variation (horizontal lines) and the Newark diurnal (sinusoidal) function (Equation 2) for comparison.

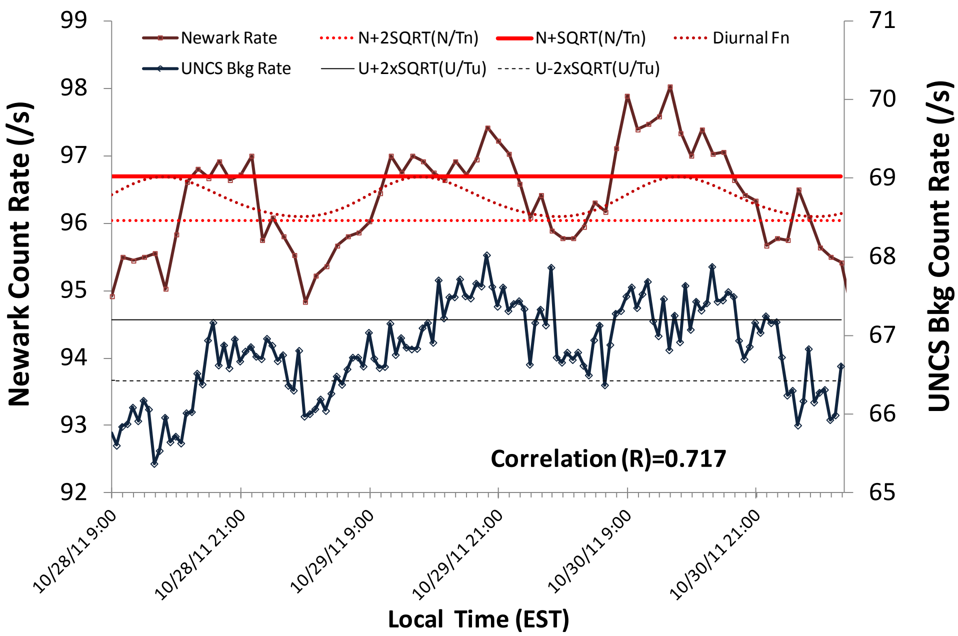

Fig. 12. Correlation of UNCS and Newark Data (Oct 28 – 30, 2011)

For a set of N background measurements of count time T, measured over a 24 hour period, the (non-Poisson) daily relative standard deviation (RSD) (F) in the background rate (B_i) is estimated in Equation 3. The $(\bar{B}T)^{-1}$ term is an estimate of the Poisson variance.

$$F = \sqrt{\frac{1}{N-1}\sum_{i=1}^{N}\left(\frac{B_i}{\bar{B}}-1\right)^2 - \frac{1}{\bar{B}T}} \qquad \text{(Eq. 3)}$$

F was calculated for the UNCS data set after applying various filtering / correction techniques including:

- Pressure Correction (PC) using a simple real-time barometric correction,
- Cosmic ray Neutron Monitor Correction (NMC) using data from Newark (non real-time),
- Active Day Rejection (ADR). This involves rejection of days where the RSD in the Newark data for that day exceeded a threshold set at its upper 95th percentile so that data from 5% of operational days are rejected.

A summary of the F term is illustrated in Figure 13. With no correction applied, F for UNCS was 1.60%. The simple PC method reduces F to 0.67%. The lowest value achieved was where all 3 methods were applied (F=0.41%). For comparison, the Newark data over the study period has F = 0.45%. The minimum value of F is governed by the diurnal (geomagnetic) term where F = 0.20%.

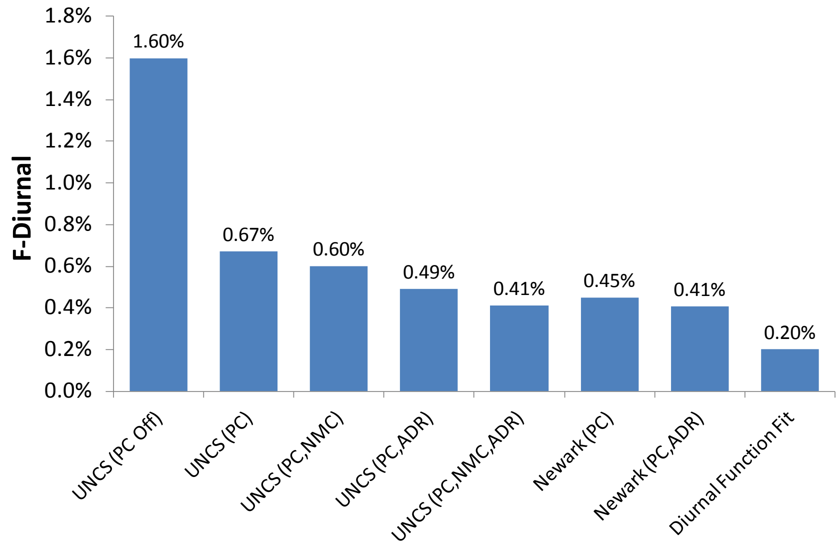

Fig. 13. Diurnal RSD (F) after Applying Various Correction Methods

MDA CALCULATIONS

In order to quantify the effect of background reduction and correction, the following scenario has been considered that is typical of an NDA measurement. A large steel object of mass M is assayed with 4 neutron slab counters (example shown in Figure 14).

Fig. 14. Neutron Slab Counter.

The object creates its own (unwanted) background due to spallation, so to reduce this effect, an assay is performed for count time T, in a shielded room depicted in Figure 15. The shielding reduces the local background in the slabs (measured outside as B_o) by a factor S_L and reduces the spallation rate in the assay object by S_G. The spallation neutron generation rate (measured outside) in the object per unit mass (of steel) is G_o. A typical value for G_o at sea level measured near to Richland, WA, USA is 0.038 n/s/kg-steel [4]. The absolute efficiency of the slab counters is ε_U (to neutrons from uranium emissions) and ε_G (to neutrons generated from cosmic ray spallation). If the mass ratio of U-234 to U-235 is R and the specific emission neutron rate per gram (U-234) (mostly from (alpha,n) emissions) is E_c then the system's "Currie-Method" MDA [10] is expressed in Equation 4.

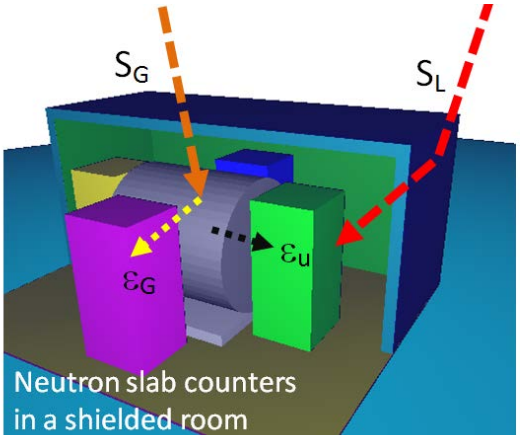

Fig. 15. Minimum Detectable Activity Scenario

$$MDA(g\,^{235}U) = \frac{2.71 + 4.65\sqrt{B_T T + B_T{}^2 T^2 F^2}}{R\varepsilon_u E_c^{^{234}U} T} \quad \text{(Eq. 4)}$$

The total background rate, B_T, is defined in Equation 5 by summation of two terms representing the local background and the background created by cosmic spallation in the assay object.

$$B_T = \frac{B_O}{S_L} + \frac{G_O M \varepsilon_G}{S_G} \quad \text{(Eq. 5)}$$

Figure 16 shows a contour plot of MDA versus count time and diurnal term (*F*) for a 1500 kg object contaminated with uranium where R = 0.01, assayed in a room where S_G= S_L= 1.5, B_o = *4* cps, ε_G = ε_U = 0.5% and E_c is 180 n/s/gU-234 for hydrated uranyl fluoride. This plot demonstrates that, for a given value of *F* (the hardest parameter to change), the MDA reduces to a point where increasing the count time will have very little impact on MDA.

Table III illustrates the effect of pressure correction in this scenario. The MDA for a 20 minute count with pressure correction is equivalent to a 60 minute count without pressure correction.

TABLE III. MDA Values (g U-235) for various count times and diurnal terms

Count Time (minutes)	MDA (g U-235)			
	24h Solar Variation (F=0.2%)	Newark Corrected (F=0.4%)	Pressure Corrected (F=0.7%)	Without Correction (F=1.6%)
20	25.6	26.1	27.5	34.8
30	21.0	21.6	23.2	31.5
40	18.2	18.9	20.7	29.7
50	16.3	17.1	19.1	28.6
60	14.9	15.8	17.9	27.8
90	12.3	13.3	15.8	26.5

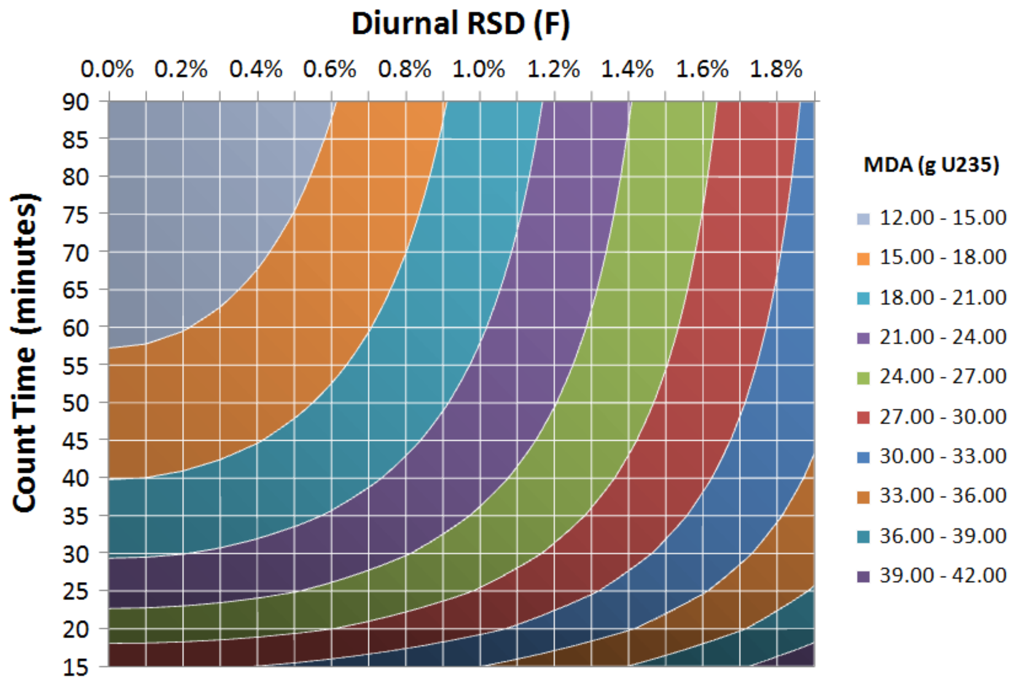

Fig. 16. MDA as a function of count time and diurnal term (F)

CONCLUSIONS

A study has been performed on the potential improvements in neutron assay MDA that can be achieved through the use of various methods for background compensation and reduction. A large background data set has been evaluated in various applications.

It has been demonstrated that a simple barometric pressure correction will significantly reduce a typical total neutron counting system MDA and allow much faster measurement times (a factor of 3 count time reduction has been demonstrated - from 60 minutes to 20 minutes).

There is a quantifiable short-term and long-term relationship between solar activity (x-ray flares) and neutron background rates which may be useful in identifying periods of high background variability. As a result, it is recommended that MDAs should be determined during periods of steady / low solar activity such that the diurnal cosmic neutron variation represents a typical value.

A proxy measurement for the cosmic background contribution can be made by reference to a neutron measurement station located at a region with similar geomagnetic cutoff rigidity and elevation to the assay equipment. Statistical correlation has been demonstrated over a 9 month period between a neutron monitor station in Newark, DE and backgrounds measured with a large neutron assay system (the UNCS) in Oak Ridge, TN.

Several other methods for suppression of cosmic background neutrons are available that can further reduce the MDA. In particular the use of time correlation to identify pulses of neutrons associated with spallation may be employed with a variety of statistical filtering methods.

The reduction in MDA for assay of large objects has significance for waste consignors who are seeking to categorize waste according to its radioactivity and for the exemption of small quantities of fissile material. Particular benefits may be realized for decommissioning of gaseous diffusion plant waste arisings and in low-level / intermediate / transuranic waste segregation. Widespread adoption of this method could yield cost benefits and improved operational efficiency for waste management professionals. Further to this, the principles of this method may also be applicable for improving the performance capability of neutron radiation portal monitors for the interdiction of illicit transportation of nuclear materials.

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ACKNOWLEDGEMENTS

The Bartol Research Institute neutron monitor program is supported by the United States National Science Foundation under grants ANT-0739620 and ANT-0838839, and by the University of Delaware Department of Physics and Astronomy and the Bartol Research Institute.

Data used to support this study was developed in conjunction with URS | CH2M Oak Ridge LLC (UCOR), the Department of Energy's (DOE's) cleanup contractor for the DOE Oak Ridge Reservation, primarily focused on cleanup of East Tennessee Technology Park (the former K-25 uranium enrichment facility).