# Direct Counting of U, Th and K Levels in Candidate Support-Rope Materials for SNO

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## ABSTRACT

Using a unique three-crystal, low intrinsic background germanium detector (ULBT), we have directly determined the levels of U, Th and K in two candidate materials being considered for use in the SNO detector.

It is well understood that materials being considered for use in the SNO detector must meet the condition of having very low levels of intrinsic radioactivity. Maintaining low levels of radionuclides in the detector will help ensure that the background signals in the experiment are at a manageable level.

The support ropes, which will suspend the acrylic vessel in the cavity, are especially critical, as they will be attached at the equator of the vessel bearing the $D_2O$. If the radioactivity in these ropes is not kept at the lowest possible level, high levels of background events will result. Whatever radioactivity is present in these ropes also must be well characterized so that events generated by this radioactivity may be correctly represented in future Monte Carlo analyses.

In order to begin the selection process for the support ropes, two groups have been charged with making direct measurements (by $\gamma$-counting) of the content of U, Th and K in the candidate rope materials, Vectran and Kevlar. Our group has conducted its measurements using an ultralow intrinsic background, three-crystal germanium detector, originally designed for double-beta decay studies. This three-crystal detector has been placed in a research lab on the 4600 foot level of Creighton Mine. 10 inches of Pb and two inches of Cu have been used to surround the crystals and attenuate the gamma rays from the surrounding rock. Finally, a nominal flow of five liters per minute of clean $N_2$ was maintained into the shield. This purge gas flow was found sufficient to exclude airborne radon from the inside of the shield.

Previous study had demonstrated that a sufficient sensitivity to U, Th and K levels would be attained by using a 21 liter sample. Consequently, the Vectran and Kevlar filaments were wound onto spools of a Marinelli geometry of 21 liter volume. These spools were then placed into a holder within the shield so that the sample surrounds the detector crystals.

A simple noise suppression circuit using a piezoelectric microphone, an amplifier and a 555 timer is used to reject periods of excessive noise and vibration (such as those following blasts or during train movements). The timer is set to reject any counts which arrive after the circuit fires, for a period of about 25 s. Including the periods in which the circuit suppresses counting, the ratio of acquiring time to real time stands at about 85%.

Each sample was counted for a period which was dependent on the level of the radioactive nuclides in the sample, and on the background counts in the system. In practice, counting times in excess of one million seconds have been used. The position of the gamma ray peaks of interest have been fixed using calibrations involving either obtainable radioactive sources, or by allowing radon to enter the detector. Any radon admitted into the shield was found to have fallen to background levels within 24 hours.

Radioactivity which is counted by the detector system, but which is not present due to the

sample material, is a major concern. This radiation may be present due either to isotopes contained within the shield, detector and sample holders, or to radiation which "leaks" through the shield into the vicinity of the crystals (either as a residual radon level which is present despite the use of the nitrogen flush, or as gamma rays which penetrate the shield). Both sources of background may be nulled by counting the gamma rays present in an empty shield/sample holder. By determining the number of gamma rays present at relevant peak locations per a given number of seconds of counting with this empty shield, the number of gamma rays actually due to a sample is easily extracted.

The actual concentration of radioisotopes is determined via the use of a standard. Our standard is composed of vermiculite, loaded into the same 21 liter Marinelli geometry as the filament samples. The vermiculite itself is referenced back to a MnO standard. The vermiculite standard, with a much higher concentration of the radioisotopes of interest than the filament samples, was counted for an appropriate time period and the known concentrations of the standard were then related to the unknown concentrations of the samples.

Concern regarding the difference between the density of the samples and the vermiculite standard prompted the use of a sisal rope standard. The sisal rope would have confirmed the efficiency calibration of the vermiculite standard, and would have shown that the self-absorption of gamma rays in a material as dense as our sample filament was not a problem. However, the measurements involving the sisal rope standard did not agree with those involving the vermiculite standard. This is further discussed below. Potential problems in the characterization of the sisal standard are being explored. For the purposes of this work, we will exclusively use the results from the vermiculite standard.

Our planned procedure was to energy calibrate the detector, place a sample into the holder, count for the appropriate time, then replace the sample with the standard. Energy calibrations would be done as frequently as possible during the measurement, with the vermiculite standard serving as both an energy and an efficiency calibration. Then another sample could be loaded and the process repeated. Finally, all samples would be removed and the background counted to determine the number of gammas due to intrinsic background in the detector system.

In the actual counting done with this system, several problems arose. The first, and most serious, was a failure of several components in the preamplifier for the number one crystal of the detector (D1). Spectra taken from D1 initially contained a very high background compared to D2 and D3, especially at high energies. By replacing two high-voltage capacitors and a load resistor, the pre-amplifier was brought back into service, and D1 now produces spectra consistent with those produced by D2 and D3.

All the crystals, but most especially D1, also became unusually sensitive to external vibration at one point, manifesting this sensitivity as microphonic noise in the gamma ray spectra. It became necessary to open the shield and to realign the detector in the sample holder, in order to assure that there was no physical coupling between the shield and the detector.

Finally, a drifting of the energy calibration of D2 has been a recurring problem. This drift was first seen during the measurement of the Vectran filament sample, and was thought to be corrected by the replacement of an amplifier module. The problem was not present during the low-vibration shut-down period of Creighton Mine during January and February of 1994. This drift recently recurred in the background measurement during March of 1994, and became a serious enough problem that the pre-amplifier contacts were cleaned and the bias supply, amplifier and coaxial cables were all changed. At present, all energy calibrations are stable.

1) Vermiculite Standard:

The standard used in this work is a 21 liter container of vermiculite in a Marinelli geometry, with a mass of 2277 g. The radioactivity has been well characterized, and is shown in Table 1.

Table 1 - Concentration of radionuclides of interest in the vermiculite standard (all error bars are 2σ).

| Isotope | Concentration |
|---|---|
| Th | 2141 (94) ppb |
| U | 77 (14) ppb |
| K | 4.74 (20) % |

The standard was first counted for a total of 136,800 s. Each saved interval of data was analyzed for the appearance of an anomalous number of counts at the positions where radon would be expected. All data where no anomalous levels were found were accepted and included in a summed gamma ray spectrum. This spectrum was then analyzed, and the total number of counts for each gamma ray energy of interest from the three crystals was determined.

A second standard run, using the same sample and procedures as the first, was made at a later date. The total counting time for this second run was 323,674 s. It was found that the count rates for the regions of interest in the second run agreed extremely well with those from the first. For purposes of this work, the second (longer) standard run will be used in data analysis. The data from the second standard run are listed in Table 2. Figure 1 shows the 2614 keV region from the second vermiculite standard run.

As the samples all have extremely low activities, and as there was no evidence for energy calibration drift during either standard run, the three crystals could be counted on a single multichannel analyzer through a multiplexer (mux), with the three crystals offset from one another. The offset was nominally 50 channels at 1300 keV (determined by using a $^{60}$Co source).

Table 2 - Analysis of standard run number two (323,674 s). All errors are 2σ. Detectors indicates the number of the crystals producing useful data at each energy (without contamination by neighbouring peaks in the sum spectrum).

| Isotope | Peak Energy (keV) | Detectors | Gross Counts | Net Counts |
|---|---|---|---|---|
| Th | 2614 | D1,D2,D3 | 5792 | 5532(156) |
| | 911 | D1,D2,D3 | 76395 | 7680(762) |
| | 583 | D2 | 25309 | 4380(430) |
| U | 1764 | D1,D2,D3 | 2044 | 872(114) |
| | 1001 | - | - | - |
| | 609 | D2,D3 | 41794 | 2466(570) |
| K | 1461 | D1,D2,D3 | 315217 | 299405(1150) |

It was deemed useful to test whether or not the gamma rays of interest to us are free of sample self-absorption problems, as compared to the standard. In other words, do gamma rays emitted from the samples have the same chance of passing through the remainder of the sample and being counted in the crystals as the gamma rays emitted from the standard. The density of vermiculite is quite low when compared to the filament samples, so this is not an idle concern.

To answer this question, a sample composed of 21 liters of sisal rope was manufactured, as

were all other samples used, by P. Jagam of the University of Guelph. The mass of this 21 l of sisal rope in a Marinelli geometry was 13.454 kg, and the radioisotopic content of the sisal was measured to be:

U: 43 ppb
K: 3697 ppm
Th ($^{228}$Ac): 322 ppb
Th ($^{208}$Tl): 231 ppb

Table 3 - Analysis of sisal rope run (68,400 s). All errors are 2σ. Detectors indicates the number of the crystals producing useful data at each energy (without contamination by neighbouring peaks in the sum spectrum).

| Isotope | Peak Energy (keV) | Detectors | Gross Counts | Net Counts |
|---|---|---|---|---|
| Th | 2614 | D1,D2,D3 | 339 | 317(38) |
| | 911 | D1,D2,D3 | n/a | 0 |
| | 583 | D2 | 4496 | 650(182) |
| U | 1764 | D1,D2,D3 | 180 | 122(30) |
| | 1001 | - | - | - |
| | 609 | D2,D3 | 4669 | 380(190) |
| K | 1461 | D1,D2,D3 | 25186 | 23950(326) |

The above data may be compared to the vermiculite standard, given the known concentrations and masses of the two standard samples. In all cases, the count rate from the sisal standard was lower than that of the vermiculite standard, and we may express this as a ratio between the count rate in the sisal and the count rate in the vermiculite. These ratios are (all errors are 2σ):

2614 keV: 0.42 (6)
583 keV: 0.43 (24)
1764 keV: 0.20 (8)
609 keV: 0.22 (16)
1461 keV 0.82 (2)

The ratios of each isotope appear to be equal, regardless of the energy which is used to determine the ratio. This would indicate that; (1) the concentrations as listed for the sisal rope sample are incorrect, since concentrations at different energies would not be equal unless the sample actually contained that concentration of the isotope, and (2) the problem of self-absorption may be discounted.

It is impossible for the sisal standard to have been contaminated and thus cause this problem, as the concentrations are all too low compared to the vermiculite. It is also extremely unlikely that the sealed vermiculite standard has been so seriously contaminated (factor of 2x in Th, factor of 5x in U).

Since the other group doing direct gamma-ray counting (LBL) is using a vermiculite standard,

we will ignore this discrepancy for now and assign fault to the sisal standard. Our final conclusions are based on relative levels in the two filament samples, regardless, so the standard will factor out. For the remainder of this work, the vermiculite standard will be the sole standard.

2) Background:

In order to determine the empty-shield background, the shield and sample holder were counted. The detector was calibrated using either a $^{60}Co$ and $^{137}Cs$ source, or a 2% thoriated welding rod. Further calibrations were accomplished using either of the two possible energy calibrators (depending on the precision desired) or, at one point, by allowing radon to enter the system. The radon entry was conducted primarily to determine the lengths of time that nitrogen flushing might be discontinued without contamination, and also to determine how long after flushing is restarted that the system still remains contaminated.

Drifting of the D2 detector began after about 800,000 s of data had been taken with the empty shield. This entire portion of data was discarded. The hardware was modified so that data from detectors D1 and D3 continued to be gathered on a single MCA through the mux, while data from D2 was gathered on a separate MCA. This would allow a severe drift of D2 to take place without compromising the data. About 1.2 Ms of data was acquired using this configuration. At this time, a third MCA was added and all three crystals were now being acquired on separate MCA cards. A further 1.9 Ms of data were acquired. Spectra were analyzed and the result combined. Figure 2 shows the 2614 keV region (with arrows indicating gamma energies) for the last 1.9 Ms of data.

The background values used in this work are shown in Table 4. Note that only the crystals listed under "Detectors" in Table 2 are analyzed.

Table 4 - Background counts for each useful crystal at each energy of interest (3,135,369 s). All errors are $2\sigma$. Count rate is given as number of net counts in the region of interest per million seconds of acquired data.

| Isotope | Peak Energy (keV) | Detectors | Net Counts | Rate per Ms |
|---|---|---|---|---|
| Th | 2614 | D1,D2,D3 | 37.5(164) | 12.0(52) |
| | 911 | D1,D2,D3 | 59.3(376) | 18.9(120) |
| | 583 | D2 | 30.0(404) | 9.6(129) |
| U | 1764 | D1,D2,D3 | 73.8(224) | 23.5(71) |
| | 1001 | - | - | - |
| | 609 | D2,D3 | 240.0(566) | 76.5(181) |
| K | 1461 | D1,D2,D3 | 201.2(310) | 64.2(99) |

3) Kevlar Sample:

The Kevlar 21 liter sample has a mass of 18.858 kg. The stability of the energy calibrations for the three crystals was good throughout the run, so clear and well-defined peaks are present. For example, the peaks at 1461 keV resulting from K are shown in Figure 3, while the peaks at 2614 keV resulting from Th are shown in Figure 4.

Each saved interval of data was examined for deviations in both the gross count rate, and for anomalous levels of counts under the prominent peaks due to radon (242, 295 and 352 keV). While the most sensitive peaks for radon detection would seem to be 352 and 609 keV, and these will be used in the future, spot checks on the reliability of using the above three energies as opposed to 352

and 609 keV show no difference in the ability to determine whether data has been compromised by the presence of radon. Figure 5 shows the gross count rate as a function of time, while Figure 6 shows the total net counts under the three energy regions corresponding to radon gamma rays, as a function of time. Only the first 70 hours of data are shown, to demonstrate the rapid decrease in radon levels within the shield once the nitrogen flush is begun.

Only data which passes the above test was included in the final sum spectrum. Signals from all three detectors were accumulated in a single MCA spectrum, the three detectors being offset in energy. Gross and net counts for each region of interest, as well as final calculated concentrations, are given in Table 5.

Table 5 - Final results for the Kevlar filament sample, background corrected. Mass of the sample was 18.858 kg. Counting time was 1,929,915 s. All errors are 2σ. Where a peak was not present above background, a 2σ limit is shown. Only crystals for which an efficiency calibration exists are shown.

| Isotope | Peak Energy (keV) | Gross Counts | Net Counts | Concentration |
|---|---|---|---|---|
| Th | 2614 | 61 | 23.8(134) | 186(153) ppt |
| | 911 | 1164 | 121.6(960) | 686(550) ppt |
| | 583 | n/a | 0 | <6710 ppt |
| U | 1764 | 166 | 29.6(349) | 53(71) ppt |
| | 1001 | - | - | - |
| | 609 | 1277 | 93.4(1023) | 59(75) ppt |
| K | 1461 | 3493 | 2801(130) | 8.98(80) ppm |

Averaging the concentrations obtained from the 2614 keV and 911 keV gamma rays of Th yields a Th concentration of 222(147) ppt. Averaging the concentrations from the 1764 keV and 609 keV gamma rays of U yields a U concentration of 56(52) ppt. All error bars are 2σ.

4) Vectran Sample:

The measurement of the 21 liter Vectran sample, mass of 14.124 kg, presents us with the same difficulty as the background run, in that a drift in the D2 energy calibration was occurring. However, where the background run contains no large peaks which could aid us in determining how severe the drift has been, the Vectran filament contains sufficient K that the peak at 1461 keV may be used to determine where the D2 peak is in relation to D1 and D3 at other energies.

In this run, the 1461 keV gamma ray allowed sufficient counts for the calibration to be adjusted using the amplifier gain while the run was going on. Data were again accumulated using a single MCA and the mux, with the peaks from the three crystals offset in energy.

The region near 1461 keV in the sum spectrum is shown in Figure 7, while the region near 2614 keV is shown in Figure 8. The accumulated data were binned into intervals and examined for the intrusion of radon using both gross count rate and net counts under radon peaks (Figures 9 and 10). Once again, only data which were shown to be radon free by virtue of these tests were included in the final sum spectrum. The final results are given in Table 6.

Table 6 - Final results for the Vectran filament sample, background corrected. Mass of the sample was 14.124 kg. Counting time was 1,454,976 s. All errors are 2σ. Where a peak was not present above background, a 2σ limit is shown. Only crystals for which an efficiency calibration exists are shown.

| Isotope | Peak Energy (keV) | Gross Counts | Net Counts | Concentration |
|---|---|---|---|---|
| Th | 2614 | 47 | -1.5(193) | <271 ppt |
| | 911 | n/a | 0 | <7764 ppt |
| | 583 | n/a | 0 | <5238 ppt |
| U | 1764 | 145 | 26.8(320) | 116(159) ppt |
| | 1001 | - | - | - |
| | 609 | n/a | 0 | <776 ppt |
| K | 1461 | 16227 | 12009(230) | 68.2(42) ppm |

5) Conclusions:

Two samples, Vectran and Kevlar filaments, have had their concentrations of the radioisotopes Th, U and K measured using direct counting techniques. Each of the samples has had its absolute radioisotope concentrations established using a 21 liter vermiculite standard and a background subtraction. The final values are:

Table 7 - Final values for concentrations of radionuclides in the two samples, background corrected. All errors are 2σ.

| Nuclide | Kevlar | Vectran | Δ(Kevlar-Vectran) |
|---|---|---|---|
| Th | 222(147) ppt | <271 ppt | 222(308) ppt |
| U | 56(52) ppt | 116(159) ppt | -60(167) ppt |
| K | 8.98(80) ppm | 68.2(42) ppm | -59.2(43) ppm |

There is a clear difference between the two filaments only in their K concentrations. There is no detectable difference in the Th or U concentrations at the 2σ level.

It is, of course, possible that the levels of radioactivity seen in the samples are due to contamination. For example, if we examine the Kevlar values, an upper limit of about 100 ppt U in an 18.858 kg sample corresponds to less than 1.9 μg of U. Assuming that the rock dust in the area contains 10 ppm Th and 2 ppm U, the addition of 0.95 g of rock dust. This contamination would then result in a Th concentration of 504 ppt. However, 950 mg of rock dust is a sizeable quantity. Based on the cleanliness of the shield as evidenced from the background runs, and from the vermiculite standard run, we would conservatively estimate that there is less than 50 mg of rock dust within the shield. The presence of this dust is removed by background correction. We would further estimate that there is likely less than 50 mg of rock dust on a sample. These values yield upper limits of 0.5 μg of Th and 0.1 μg of U. These levels of Th and U, in turn, would yield concentrations of 27 ppt Th and 5 ppt U for a 18.858 kg sample.

Each sample is wrapped in several layers of saran. When the sample is about to be loaded, the outer (presumably dirty) layer is removed and discarded. It is extremely unlikely, in our

estimation, that contamination at the levels necessary to duplicate our measurements was introduced into the sample through handling. If contamination was introduced onto the filament during manufacture through faulty handling, then we would likely view the material and supplier as unsatisfactory, in any case.

ACKNOWLEDGEMENTS

Sample preparation, as well as discussion on problems and techniques, by P. Jagam of Guelph is gratefully acknowledged. The continuing loan of the ULBT detector by J.J. Simpson of Guelph is also recognized.

FIGURE 1 - 2614 keV REGION FROM VERMICULITE STANDARD RUN

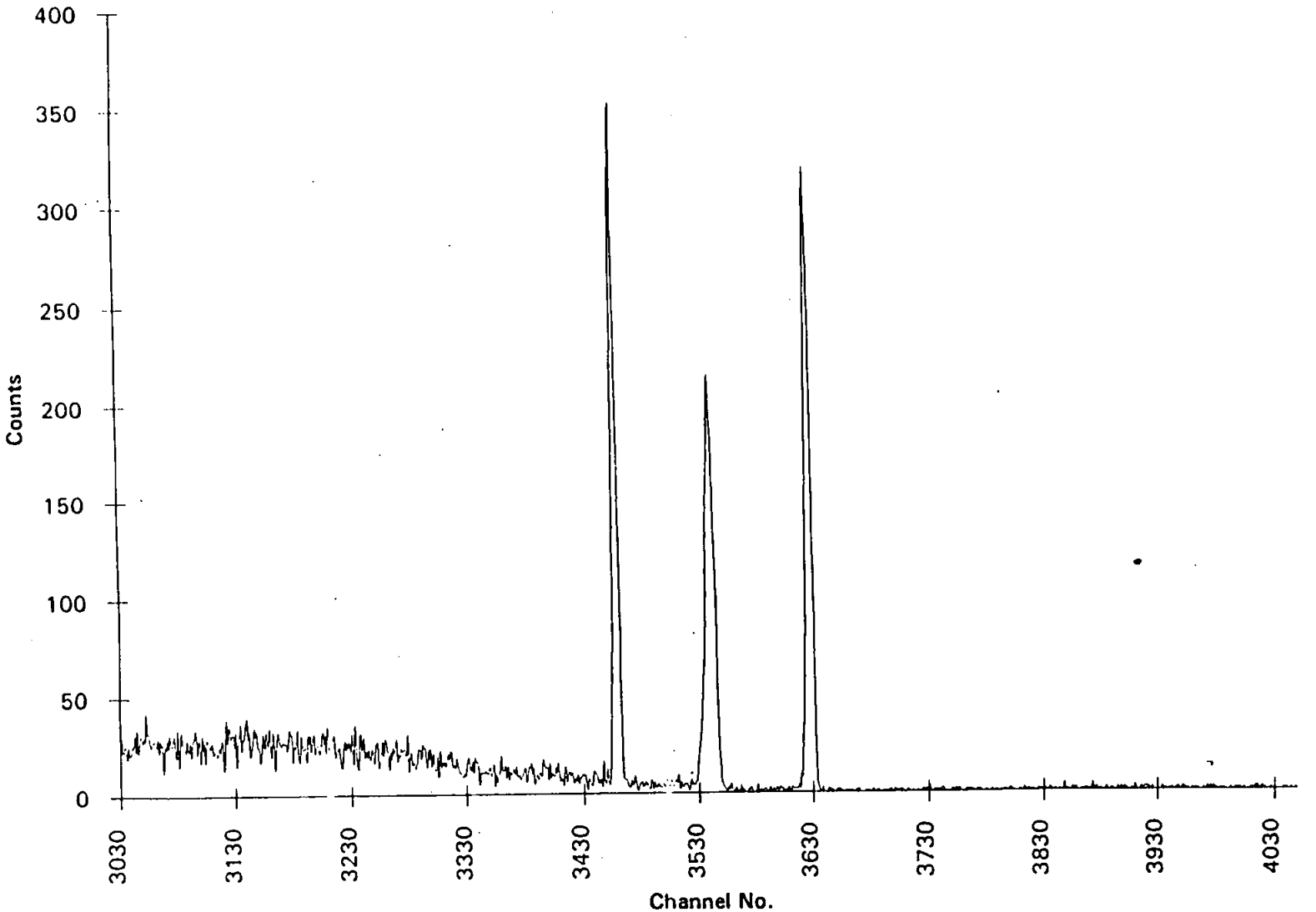

FIGURE 2 - 2614 keV REGION FROM FINAL 1.9 Ms OF BACKGROUND DATA

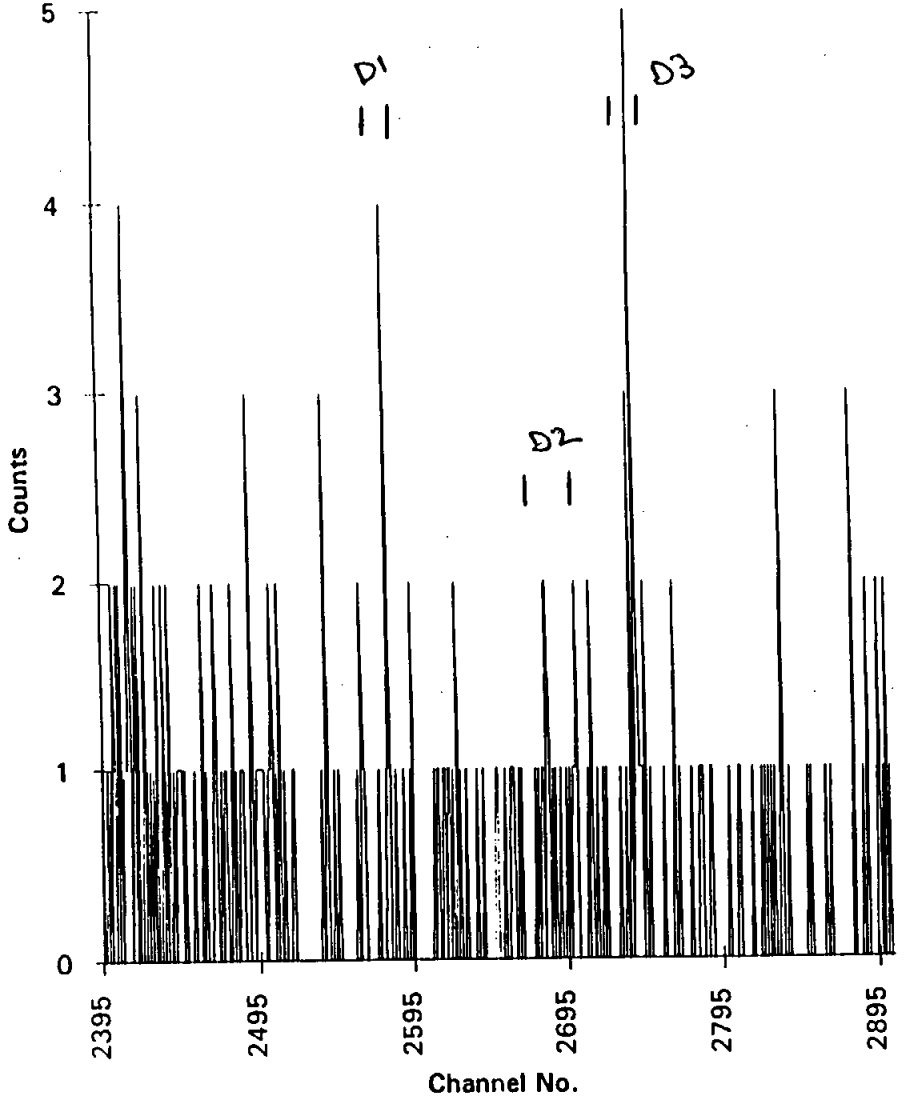

FIGURE 3 - 1461 keV REGION IN KEVLAR

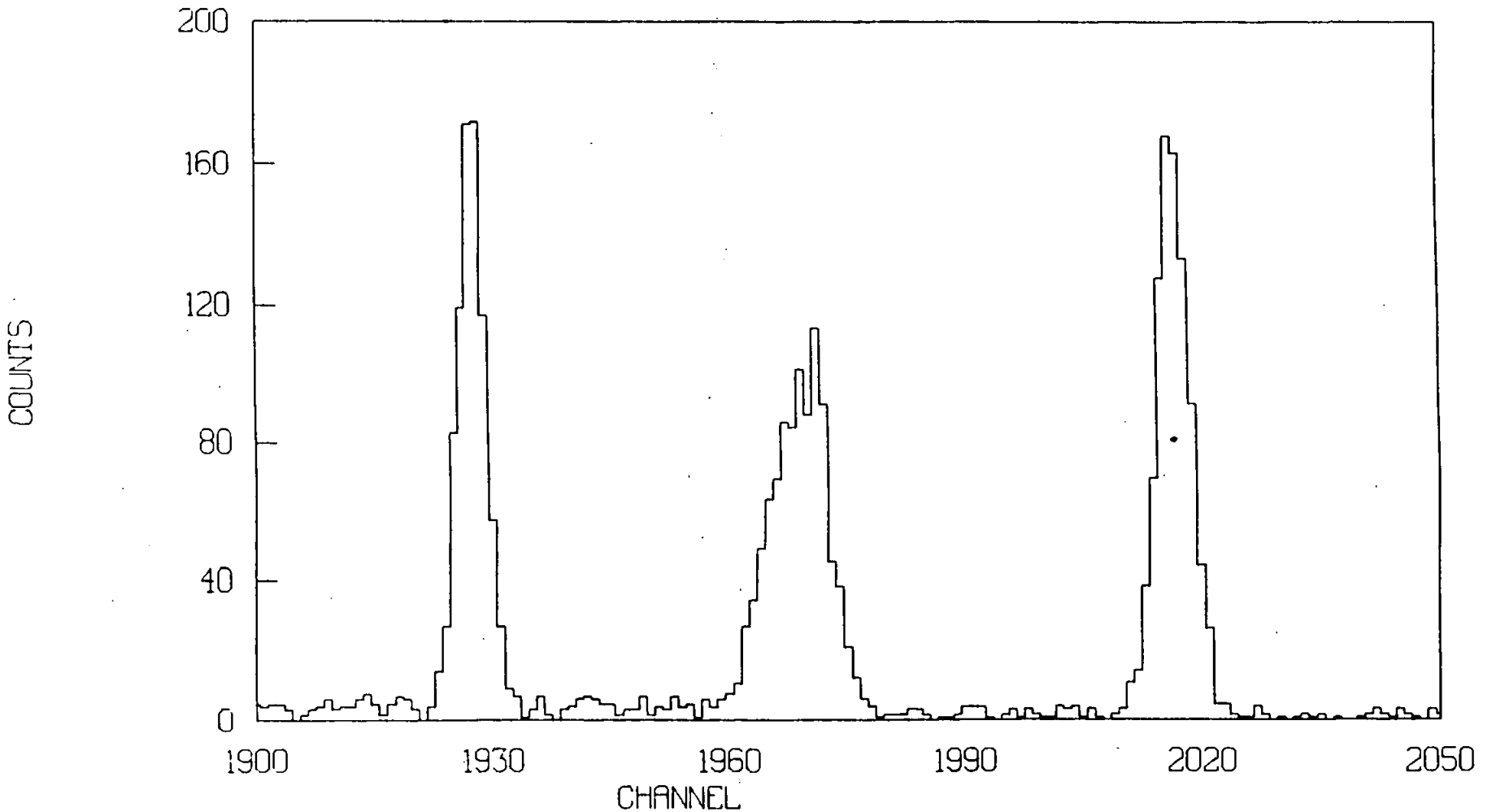

FIGURE 4 - 2614 keV REGION IN KEVLAR

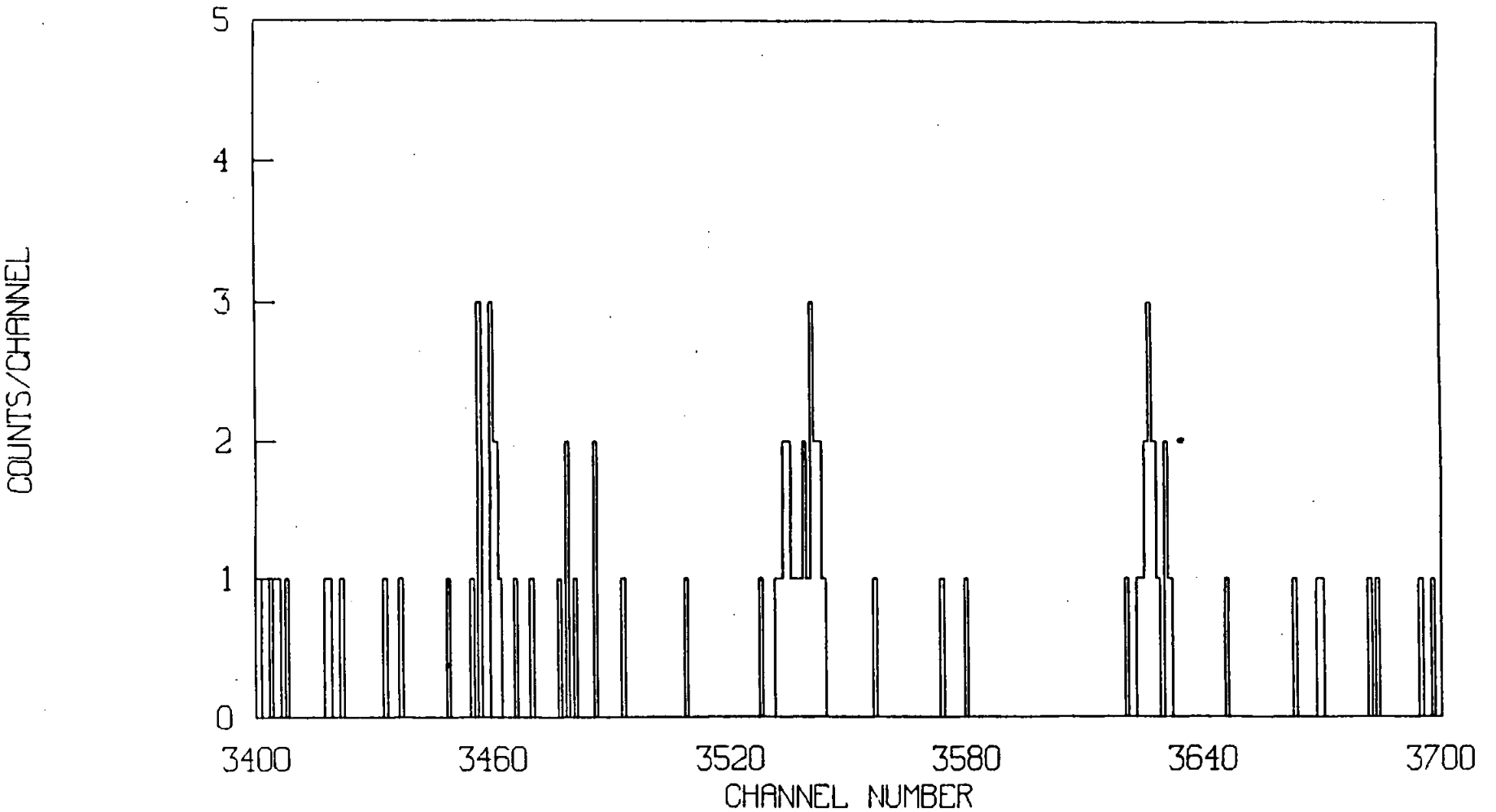

FIGURE 5 - GROSS COUNT RATE VS TIME FOR KEVLAR

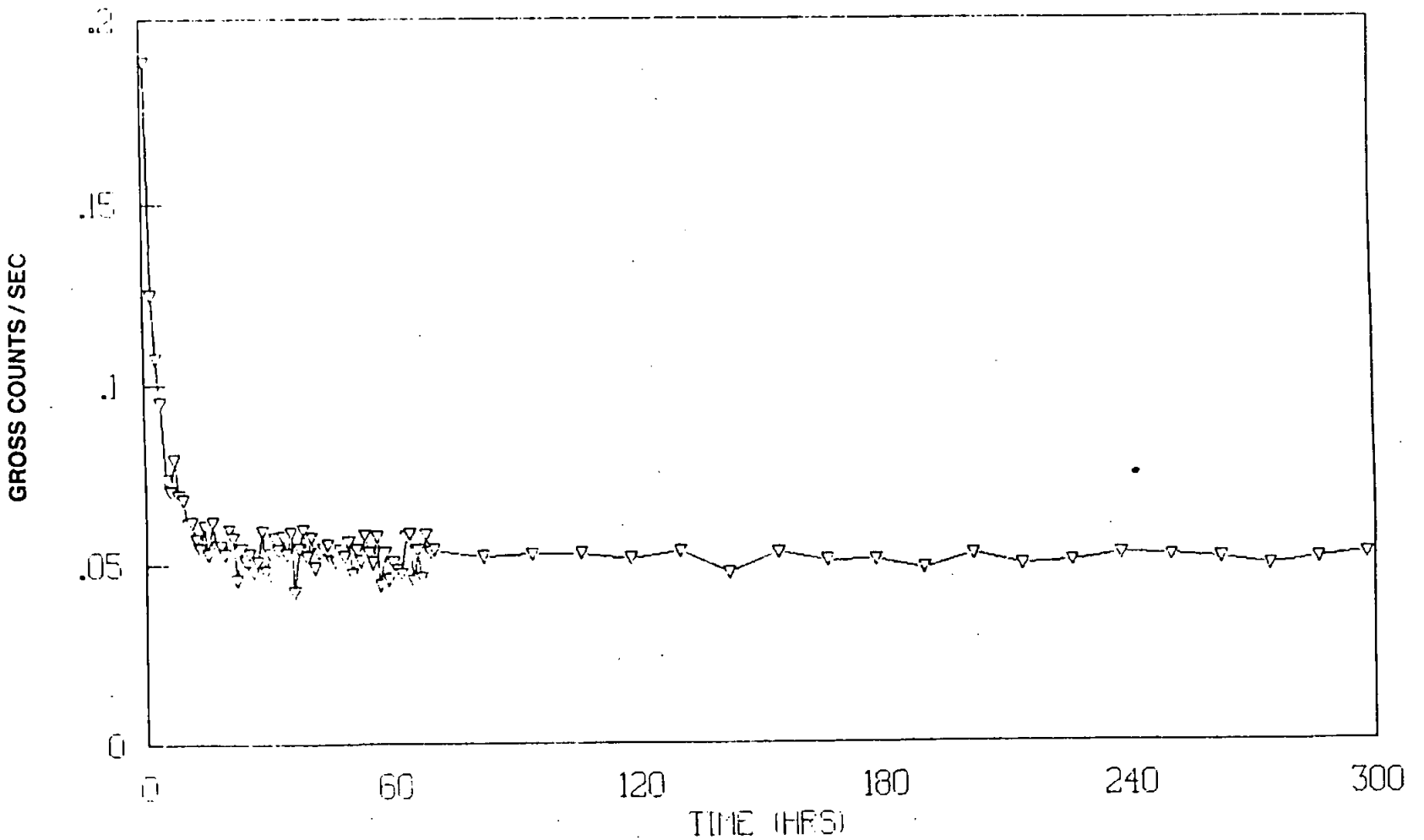

FIGURE 6 - NET COUNTS FOR PEAK REGIONS 242, 295, 352 keV IN KEVLAR

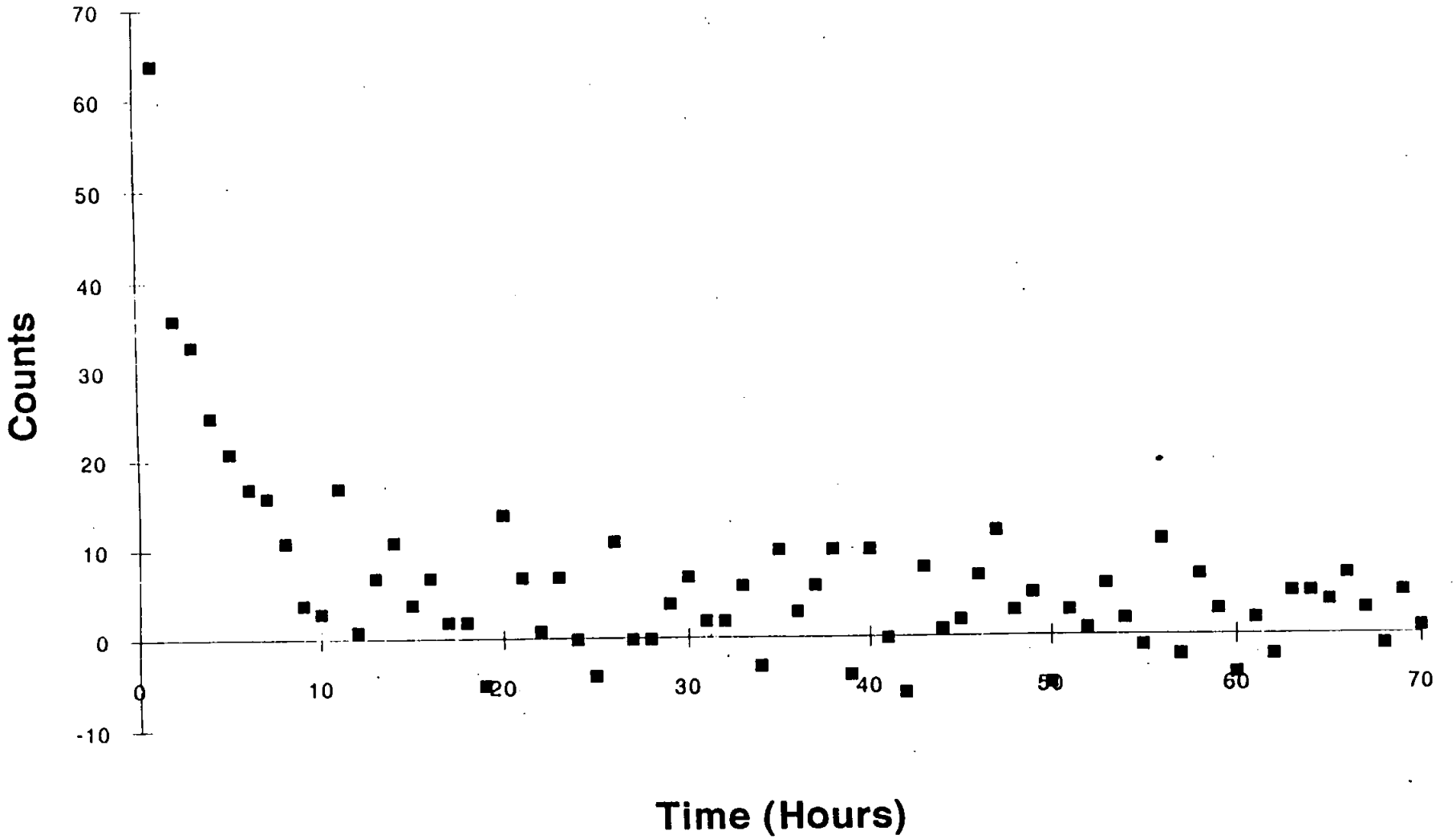

FIGURE 7 - 1461 keV REGION IN VECTRAN

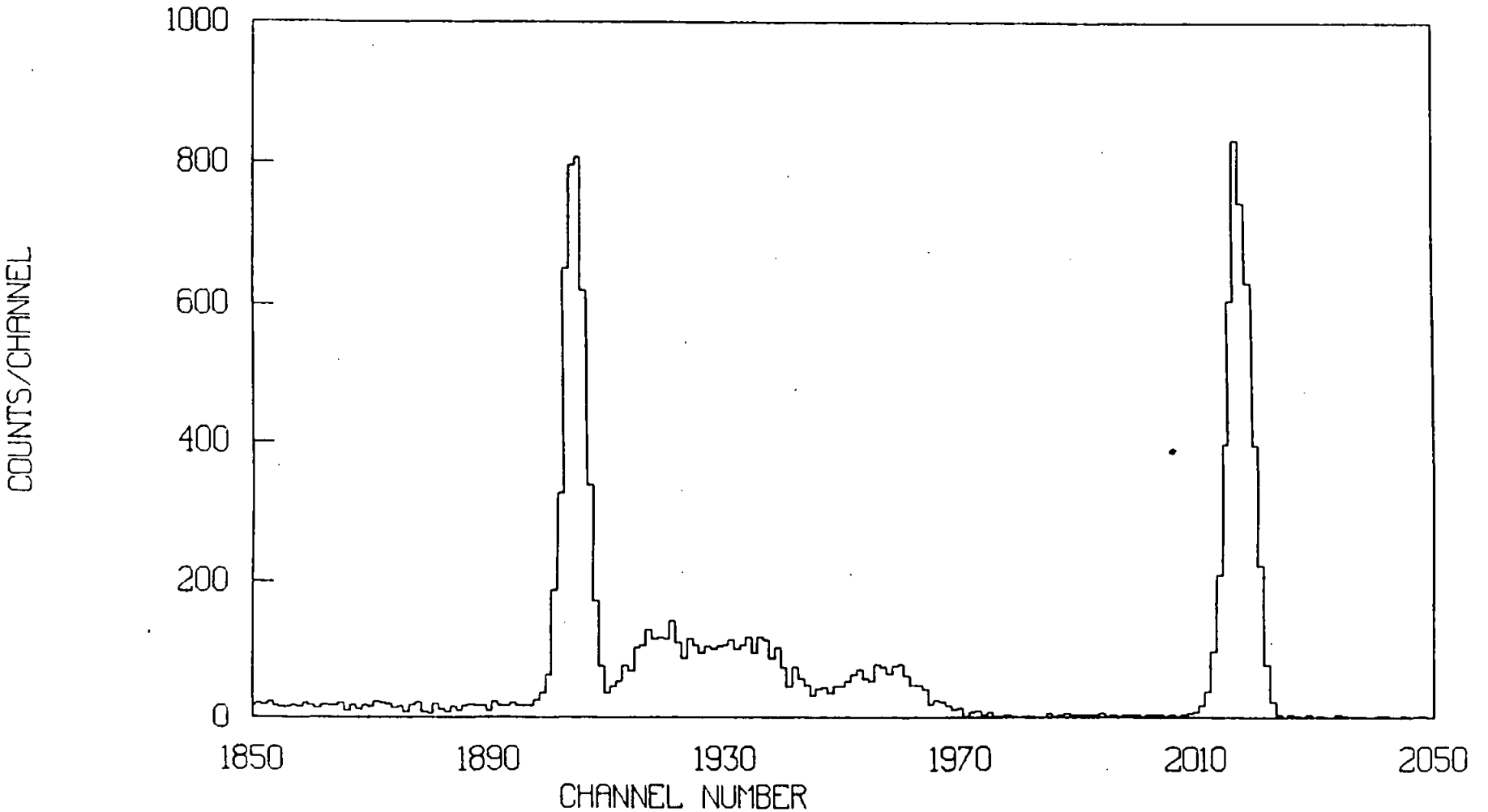

FIGURE 8 - 2614 keV REGION IN VECTRAN

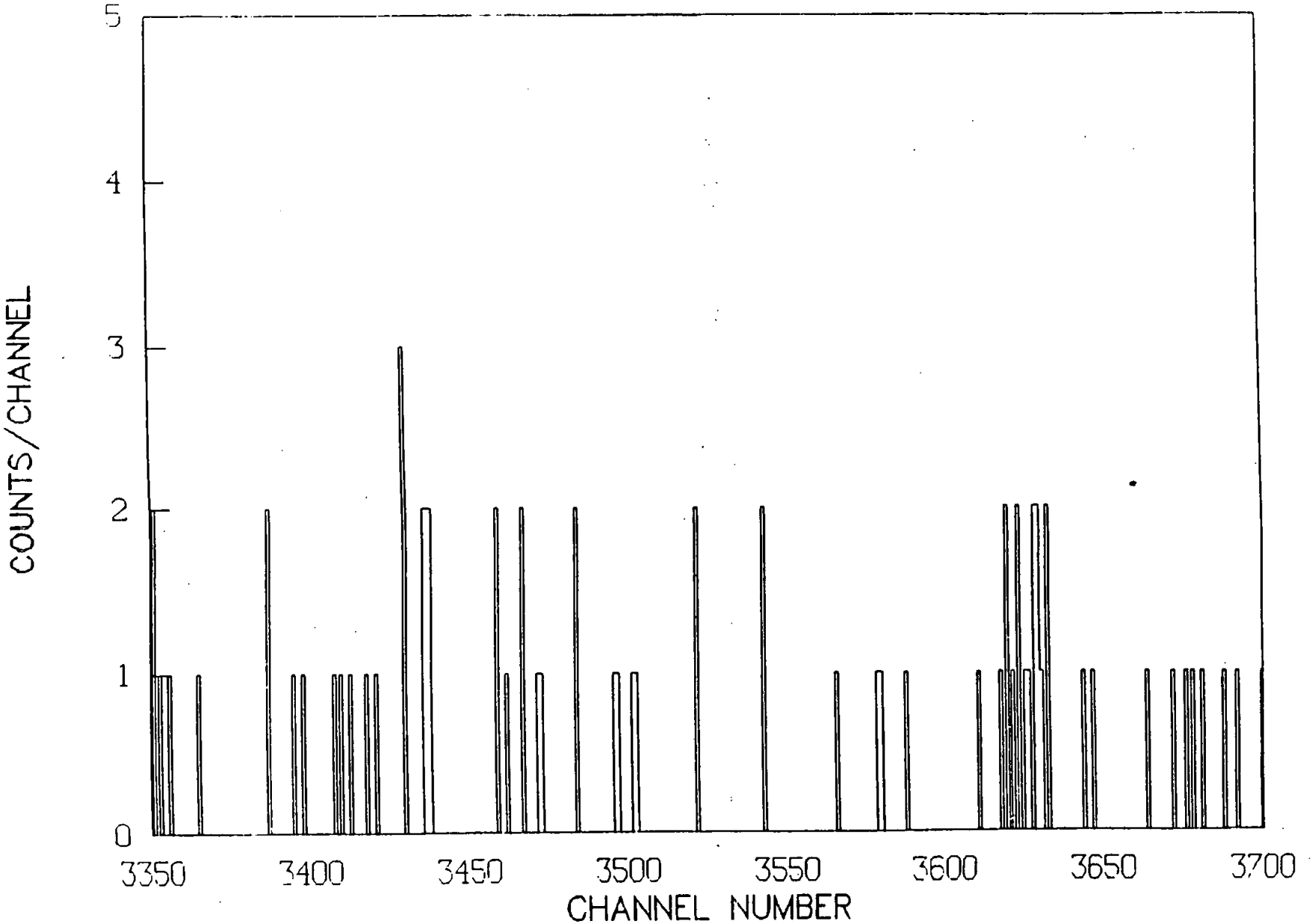

FIGURE 9 - GROSS COUNT RATE VS TIME FOR VECTRAN

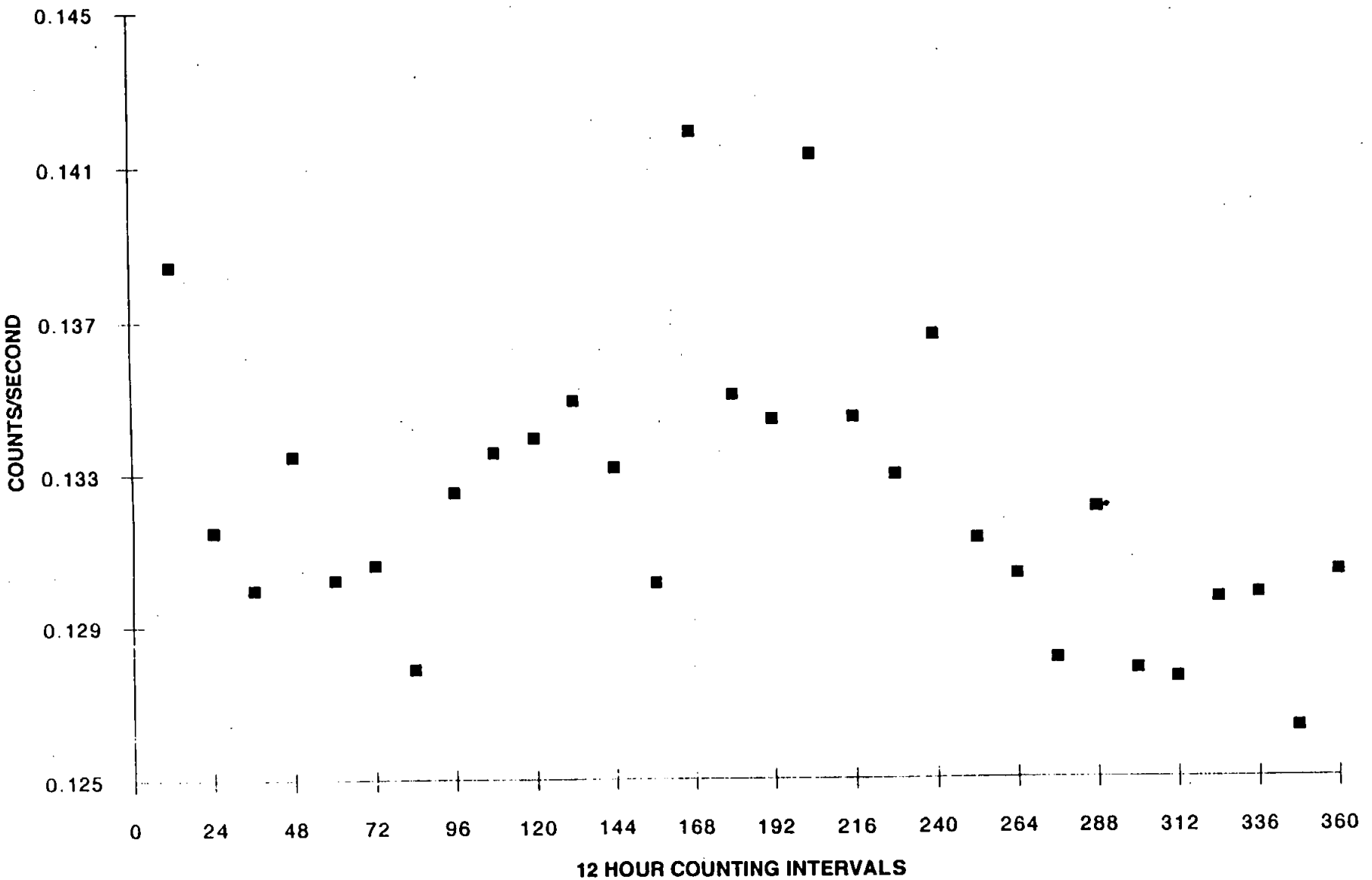

FIGURE 10 - NET COUNTS FOR PEAK REGIONS 242, 295, 352 keV IN VECTRAN