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Hanford Tank Farms Vadose Zone Monitoring Project

Correlation of Spectral Gamma Log Response and Sr-90 Concentrations for a Steel-Cased Borehole

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by
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Executive Summary

In passive gamma-ray logging, the presence of anomalous gamma activity without detectable spectral lines associated with specific radionuclides may indicate the presence of a high-energy beta-emitting radionuclide such as strontium-90 (^{90}Sr). Brodzinski and Nielson (1980) described a means of estimating ^{90}Sr concentrations by measurement of bremsstrahlung radiation in the 60- to 236-keV range. Baseline spectral gamma logging in the 241-B Tank Farm detected anomalous incoherent gamma activity with no identifiable gamma lines in several boreholes north and east of tank B-110. Interpretations presented in the B Tank Farm Report (DOE 1999) suggested that this activity represented a probable subsurface ^{90}Sr plume. Laboratory analysis of soil samples from a new borehole (299-E33-46 / C3360) drilled in this region subsequently confirmed the presence of ^{90}Sr. Concentrations as high as 11,000 picocuries per gram (pCi/g) were reported. This report presents a comparison of spectral gamma log data from this borehole to laboratory ^{90}Sr values. Shape factor analysis (Wilson 1997, 1998) is shown to be useful in identifying zones of probable ^{90}Sr and a correlation between net counts in the 60- to 350-keV range, and ^{90}Sr concentration is established for the specific casing configuration. Recommendations are made for additional investigations to more fully investigate the nature of the bremsstrahlung phenomena with respect to cased boreholes, and to determine the effect of casing thickness on bremsstrahlung radiation inside the casing.

Background

During characterization logging in the 241-B Tank Farm, the SGLS detected anomalous incoherent gamma activity in boreholes northeast of tank B-110. Specifically, boreholes 20-10-02, 20-08-07, and 20-07-11 exhibited intervals of anomalous gamma activity with no evidence of well-defined energy peaks that would be diagnostic of specific radionuclides. Figure 1 (attached) shows a combination plot for borehole 20-10-02. Note the anomalous total gamma activity between approximately 69 and 85 feet (ft) in both the spectral gamma logging system (SGLS) total gamma log (fifth plot) and the tank farms gross gamma log (sixth plot). This anomaly does not appear to be related to either man-made radionuclides or variations in natural radionuclides. Figure 2 illustrates two typical spectra from borehole 20-10-02. Spectrum 3A2A1076 (60 ft) is typical of an uncontaminated portion of the borehole. Spectrum 3A2A1036 (80 ft) is an example of anomalous gamma activity. Note that no clearly defined gamma energy peaks are present other than those associated with natural radionuclides. Both represent approximately the same concentrations of natural radionuclides, and yet 3A2A1036 has a gross count rate approximately three times that of 3A2A1076. Note that the bulk of the difference in counts occurs as incoherent gamma counts below approximately 600 keV. Brodzinski and Nielson (1980) and Wilson (1997) suggested that these incoherent counts may result from bremsstrahlung associated with the interaction between high-energy beta particles from strontium-90/yttrium-90 (^{90}Sr/^{90}Y) decay and the steel casing. In the B Tank Farm Report (DOE 1999) it was postulated that a subsurface zone of ^{90}Sr contamination had been encountered in this borehole and others in the vicinity.

Borehole C3360 (299-E33-46) was drilled in May 2001 to investigate this region and to collect soil samples for laboratory analysis. ^{90}Sr concentrations have been determined in samples at selected depths. The ^{90}Sr concentration data were provided by Dr. Jeff Serne of Pacific Northwest National Laboratory. Only the concentrations were provided; no details about the lab assay methods or assay standards were given. For the purposes of the investigation described in this report, the concentrations were assumed to be accurate.

The depth interval from 50 to 120 ft in borehole 299-E33-46 was logged with the SGLS, and man-made gamma-ray-emitting nuclides were determined to be absent, or present in negligible concentrations. Thus, emissions from such nuclides did not introduce significant extraneous spectral background. The photons in the borehole not due to natural radioactive sources were bremsstrahlung created by collisions and accelerations of the beta emissions from ^{90}Sr/^{90}Y decay. The borehole was therefore viewed as a facility to test theories about the bremsstrahlung contributions to passive gamma-ray spectra and the correlations of such bremsstrahlung signals to the ^{90}Sr concentrations.

The guide to the investigation was spectral shape factor analysis work performed by R.D.Wilson, most of which was reported in *Spectrum Shape-Analysis Techniques Applied to the Hanford Tank Farms Spectral Gamma Logs* (Wilson 1997). This work presented results of model studies using the MCNP radiation transport code and model experiments. Unfortunately, in relation to Wilson's work, borehole 299-E33-46 is an imperfect facility for this investigation, for at least two reasons:

- the borehole casing in Wilson's MCNP model was 0.313-inch (in.)-thick steel, whereas the steel casing in borehole 299-E33-46 is 0.514 in. thick. The effect of casing thickness on generation and transmission of bremsstrahlung gamma rays is unknown.

- the beta source in Wilson's MCNP model was "distributed uniformly 2 centimeters (cm) radially into the formation and extending ± 15.24 cm (± 6 in.) axially" (with respect to the center of the gamma-ray detector), whereas the ^{90}Sr distribution along borehole 299-E33-46 appears to be somewhat non-uniform.

Shape Factor Analysis

The technique of shape factor analysis is described in Wilson (1997, 1999). This technique was developed as a means to assess the distribution of cesium-137 (^{137}Cs) and cobalt-60 (^{60}Co) with respect to the borehole axis. For both ^{137}Cs and ^{60}Co, a shape factor, SF1, was defined as the ratio between the energy peaks and the increased spectral noise due to Compton scattering. SF1 is sensitive to spectral differences for a borehole-confined source, a source uniformly distributed in the formation, and a remote source. However, it is only valid when the respective contaminant is present and has no bearing on identification of ^{90}Sr. SF1 will not be further discussed in this document. Another shape factor was defined to assess the distribution of counts in the low-energy continuum:

$$SF2 = \frac{counts(60-350keV)}{counts(350-650keV)} \qquad \text{(Wilson 1997)}$$

This factor is sensitive to differences in the scattered portions of the spectrum from gamma emitters and from bremsstrahlung sources. In particular, SF2 is capable of identifying the presence of low-energy bremsstrahlung radiation from the decay of $^{90}Sr/^{90}Y$ and is able to distinguish this spectral effect from the enhanced low-energy response obtained from remotely located ^{137}Cs and ^{60}Co. Wilson's model studies indicated that "*for virtually all gamma-emitting contaminants and for all possible source distributions, SF2 never exceeds a value of about 5. An SF2 value greater than 5 is evidence for the presence of a bremsstrahlung-producing energetic beta emitter, such as $^{90}Sr/^{90}Y$.*" (Wilson 1997). Other reports (Wilson 1997, 1999) have suggested that SF2 values may be as high as 20 in zones with significant ^{90}Sr concentration.

Figure 3 shows the shape factor log for borehole 20-10-02. Note that within the region of anomalous total gamma activity between 69 and 85 ft, SF2 attains a value of approximately 8. In this log, SF2 is calculated from counts in the two spectral windows after the contribution from naturally occurring radionuclides has been removed, leading to the erratic behavior of SF2 in uncontaminated intervals.

Results from Borehole 299-E33-46

Figure 4 (plot A) shows a plot of total gamma count rate and count rate in the 60- to 350-keV range for the 50- to 120-ft depth interval in borehole 299-E33-46. Figure 4 (plot B) shows laboratory ^{90}Sr concentrations plotted at the same depth scale. Figure 4 (plot C) shows SF2 and SF2* calculated as the ratio between counts in the 60- to 350-keV range and counts in the 350- to 650-keV range. SF2 is calculated using count rates corrected for natural radionuclides, while SF2* is calculated using gross (uncorrected) counts in the two energy windows. A correlation appears to exist between laboratory ^{90}Sr concentrations and either total gamma count rate or count rate for 60 to 350 keV. Furthermore, SF2* has a value of about 3.3 to 3.6 in uncontaminated areas, increasing to greater than 6 in intervals of high ^{90}Sr concentrations.

Figures 5 (plot A) and 5 (plot B) show two HPGE spectra from borehole 299-E33-46. Both plots show the same two spectra; counts (vertical axis) are plotted on a log scale in Figure 5 (plot A), and on a linear scale in Figure 5 (plot B). The upper spectrum (FOCA1024.S0) was recorded at a depth of 62.0 ft, where the ^{90}Sr concentration was 11245 ±363 pCi/g. The lower spectrum (FOCA1100.S0) was recorded at a depth of 100.0 ft, where the ^{90}Sr concentration was zero, or close to zero. The full energy peaks are all associated with natural background; the peak for the 1460.8-keV gamma ray of ^{40}K, and the peak for the 2614.5-keV gamma ray of ^{208}Tl are labeled. Because spectrum FOCA1024.S0 contains no evidence of man-made gamma-ray emitters, the offset relative to spectrum FOCA1100.S0 is presumably due to bremsstrahlung associated with ^{90}Sr beta emissions.

Spectrum FOCA1024.S0 seems generally consistent with Wilson's MCNP simulation, which indicated that most of the bremsstrahlung contribution would appear in the part of the spectrum below 500 keV (see Figure 8 in Wilson 1997).

Although the shape of spectrum FOCA1024.S0 more or less agrees with Wilson's model, the gross count rate apparently does not. Spectrum FOCA1024.S0 has a gross count rate equal to 2713.12 counts per second (cps), and spectrum FOCA1100.S0 has a gross count rate of 162.82 cps. Because FOCA1024.S0 has a ^{90}Sr contribution but FOCA1100.S0 does not, the difference of 2550.3 cps would seem to be attributable to the ^{90}Sr contribution (assuming the potassium-uranium-thorium background is more or less uniform). The ^{90}Sr concentration corresponding to FOCA1024.S0 was 11,245 picocuries per gram (pCi/g), meaning that the measurement sensitivity to ^{90}Sr was about 0.23 cps per pCi/g. This is almost an order of magnitude higher than the value of 0.028 cps per pCi/g estimated by Wilson.

Because the sensitivity was in substantial disagreement with Wilson's estimate, the sensitivity was recalculated using 22 spectra from ^{90}Sr-contaminated depths. A background gross count rate of 159.3 ± 13.4 cps (uncertainty = ± 2σ) was determined by calculating the average gross count rate of the spectra from depths below 90 ft, where the ^{90}Sr concentrations were zero, or close to zero. The background was subtracted from each of the 22 spectra, then each gross count rate was divided by the associated ^{90}Sr concentration. The average of the sensitivity values was 0.26 ± 0.18 cps per pCi/g, which agreed with the initial finding.

SF2 for FOCA1024.S0 was calculated by subtracting counts from FOCA1100.S0 in each channel, computing the sums of the remainder over 60 to 350 keV and 350 to 650 keV, and dividing the two numbers. SF2* was calculated by dividing the total counts in the same channel ranges. SF2 was calculated to be 6.61, and SF2* was calculated to be 6.34. These values are somewhat lower than those encountered in borehole 20-10-02, and considerably lower than the value of 20 suggested by Wilson's model studies. This discrepancy may be due at least in part to the greater casing thickness. The maximum range of a 2.28-million electron volts (MeV) beta particle in iron is estimated to be on the order of 0.06 in., suggesting that incident beta radiation only interacts with a relatively thin outer layer of the casing and that the remaining casing material simply attenuates the bremsstrahlung gamma activity. Because lower energy gamma rays are attenuated to a greater degree, a lower value of the SF2 ratio in thicker casing would result.

In spite of the sensitivity discrepancy, and differences between Wilson's MCNP model and the casing thickness and ^{90}Sr distribution presented by borehole 299-E33-46, values for several parameters were calculated from the SGLS spectra, and correlations between these parameter values and the ^{90}Sr concentrations were investigated.

Spectra from various depths, mostly where ^{90}Sr was present, were analyzed as follows. Count rates for two spectral windows were calculated, 60 keV to 350 keV (window 1) and 350 keV to 650 keV (window 2). Both windows are corrected for background using stripping factors based on potassium-40 (^{40}K), uranium-238 (^{238}U), and thorium-232 (^{232}Th) peaks. The ratio of the corrected rate for window 1 to the corrected rate for window 2 is Wilson's shape factor 2 (SF2). SF2 is plotted in relation to ^{90}Sr concentration in Figure 6. Also plotted in Figure 6 is a "modified" SF2, designated SF2*, which is the ratio of total counts in window 1 to total counts in window 2. Note that both SF2 and SF2* increase with increasing ^{90}Sr. Both are greater than 5 when ^{90}Sr concentrations are greater than 1,000 pCi/g, and both seem to reach a maximum value between 6 and 7. At low ^{90}Sr concentrations, however, SF2 varies widely, while SF2* seems to have a relatively stable value between 3.3 and 3.6. From a mathematical perspective, this behavior should be expected; in intervals with no contamination, the corrected values for windows 1 and 2 should be close to zero, or even slightly negative. Division of numbers close to zero can result in unpredictable results. At high ^{90}Sr concentrations, the counts due to bremsstrahlung dominate the spectra, and subtraction of background has little effect. In this borehole, one could infer that values of SF2 (or SF2*) greater than 5 indicate the presence of ^{90}Sr, while values less than 4 indicate that ^{90}Sr concentrations are less than 500 to 1,000 pCi/g. For uncontaminated spectra, SF2* appears to be preferable because it approaches a relatively stable value. Figure 4 (plot C) shows both SF2 and SF2* plotted as a function of depth. Note that SF2 is only stable where ^{90}Sr is present, while SF2* achieves stable values in both the contaminated interval and the uncontaminated interval.

Examination of Figure 6 indicates that neither SF2 or SF2* appears to be useful as a quantitative indicator of ^{90}Sr concentration. Over the range of about 500 to 5,000 pCi/g, both shape factors appear to increase with increasing ^{90}Sr content, but above about 5,000 pCi/g, SF2 and SF2* values appear to remain relatively constant. This behavior can be explained by the fact that both numbers are ratios. Below about 500 pCi/g, the contribution to the gamma spectrum from bremsstrahlung associated with ^{90}Sr/^{90}Y decay is relatively minor. The behavior of SF2 is erratic because counts due to background have been removed and only "noise" is left to calculate the ratio. SF2* assumes a stable value, which represents the ratio based on typical levels of natural radionuclides. Between about 500 and 5,000 pCi/g, the bremsstrahlung contribution becomes increasingly more important and the ratio changes. Above about 5,000 pCi/g, the bremsstrahlung contribution dominates the spectra; counts in both windows increase proportionately, and both SF2 and SF2* exhibit little or no change with increasing concentration.

Count rates are more likely to exhibit a correlation with ^{90}Sr concentration over a wider range. Figures 7 and 8 show total gamma count rate and count rate in the 60- to 350-keV energy range plotted as a function of ^{90}Sr concentration. Each figure shows both the total or gross count rate, as well as the net count rate after subtraction of background. The values plotted against ^{90}Sr concentration are based on a 3-point average of the SGLS data, centered on the midpoint depth of the sample. Also shown on Figure 7 is a line corresponding to the sensitivity of 0.26 cps per pCi/g determined above. This shows reasonable agreement with the net count values. However, the total count rate is subject to variation associated with the presence of any man-made radionuclides or with variations in natural radionuclide concentrations.

The results of model studies (Wilson 1997) indicated that the bulk of the gamma activity associated with bremsstrahlung occurs in the 60- to 350-keV window. Following Wilson's method, the net counts in this window can be determined by subtracting the background associated with the natural radionuclides ^{40}K, ^{238}U, and ^{232}Th, using stripping ratios developed by Koizumi and reported by Wilson (1997). The "net cps (60-350 keV)" plotted in Figure 8 are determined in this manner. With one exception, this plot shows a strong linear trend. A least-squares regression was used to estimate the sensitivity of SGLS net counts in the 60- to 350-keV energy window to ^{90}Sr concentration. It was determined that:

$$N_{(60-350KeV)} = 0.19 \times C_{^{90}Sr} \qquad (R^2 = 0.874)$$

Where N is the net count rate and C is the ^{90}Sr concentration in picocuries per gram. This relationship is plotted as a line on Figure 8. It can be re-arranged to:

$$C_{^{90}Sr} = 5.24 \times N_{(60-350KeV)}$$

This relationship was developed for a borehole casing thickness of 0.514 in. Because the nature of bremsstrahlung generation associated with the interaction between high-energy beta particles and steel casing is poorly understood, the equation above should not be used where the casing thickness is significantly different from 0.514 in.

Conclusions

Analysis of borehole spectral gamma measurements and laboratory determination of ^{90}Sr concentration in samples from borehole 299-E33-46 (C3360) have shown that relationships exist between SF2 and ^{90}Sr concentration and between net counts in the 60- to 350-keV energy window and ^{90}Sr concentration. Comparison of the borehole and laboratory data resulted in the following findings and observations:

- The borehole casing in Wilson's MCNP model was 0.313-in.-thick steel, whereas the steel casing in borehole 299-E33-46 is 0.514 in. thick. The effect of casing thickness on generation and transmission of bremsstrahlung gamma rays is unknown.

- The beta source in Wilson's MCNP model was "distributed uniformly 2 cm radially into the formation and extending ± 15.24 cm (± 6 in.) axially" (with respect to the center of the gamma-ray detector), whereas the ^{90}Sr distribution along borehole 299-E33-46 appears to be non-uniform.

- Only the borehole interval from 50 to 120 ft is used for the comparison studies. The entire borehole was logged, but other intervals were logged with slightly different equipment and procedures. Variations in logging system response would complicate the comparison of log data and laboratory values.

- Comparison of gamma spectra from intervals of high ^{90}Sr concentration with spectra from low concentration intervals indicated that bremsstrahlung associated with ^{90}Sr decay resulted in greatly increased low energy counts, with the bulk of the activity less than 350 keV.

- After subtracting background, total gamma counts were compared to ^{90}Sr concentration. The average sensitivity was calculated to be approximately 0.26 cps per pCi/g, almost an order of magnitude higher than the value of 0.028 cps per pCi/g predicted by Wilson (1997).

- SF2 values on the order of 6 to 8 indicate the presence ^{90}Sr concentrations greater than 1,000 pCi/g. Wilson (1997) predicted the existence of a correlation between ^{90}Sr concentration and SF2, but the observed correlation does not fully conform to Wilson's expectations. Wilson's MCNP model predicted SF2 values greater than 20 in the presence of ^{90}Sr.

- A modified shape factor, SF2*, is defined as the ratio between total counts in the 60 to 350 keV and 350- to 650-keV windows. Because background is not subtracted, SF2* tends to remain stable in the absence of contamination. SF2* assumes a value between 3.3 and 3.7 in uncontaminated intervals and increases to greater than 6 in intervals with high ^{90}Sr concentration. For ^{90}Sr concentrations on the order of 500 to 1,000 pCi/g, SF2* values are transitional between 3.7 and 6.

- Cross-plots of SGLS total gamma versus ^{90}Sr concentration show a linear trend, particularly when total counts are corrected for background. However, total counts are affected by the presence of man-made radionuclides, as well as by variations in natural radionuclides.

- Cross-plots of SGLS total counts and net counts in the 60- to 350-keV energy range versus ^{90}Sr concentration also show a strong linear trend. For this energy window, background counts can be estimated from the 1461-, 1764-, and 2615-keV peaks, using stripping ratios developed by Koizumi. The net counts show a good correlation with ^{90}Sr concentration. The sensitivity is about 0.19 cps per pCi/g. Similar corrections could possibly be made for limited amounts of ^{137}Cs and ^{60}Co, using stripping ratios estimated from modeling and shape factor experiments. This would allow ^{90}Sr concentrations to be estimated in the presence of other contaminants.

Recommendations

The results of this study and previous experience in the B Tank Farm and elsewhere indicate that, at least in the absence of other contaminants, ^{90}Sr can be detected by spectral gamma logging in steel-cased boreholes. Concentrations also appear to be estimable, at least to an order of magnitude. Unfortunately, a number of unknown factors must be investigated before a widely applicable relationship can be established. Specific recommendations for future work include the following:

- <u>Conduct experiments to investigate the nature of gamma-ray generation and transmission associated with bremsstrahlung</u>.

  Discrepancies exist in the relationship between ^{90}Sr concentrations and gross gamma count rate and in the behavior of SF2 in the presence of ^{90}Sr. In both cases, the relationship observed in the field differs significantly from predictions based on radiation transport modeling. These discrepancies may be due to the difference in casing thickness, or to errors in the way bremsstrahlung is addressed in the model. A relatively simple experiment can be set up to investigate the effects of casing thickness on gamma activity from bremsstrahlung. Gamma spectra would be recorded from a detector placed a short distance from a ^{90}Sr source. Steel plates with various thickness values between about 0.25 and 1.0 in. would be placed between the source and the detector, and the data would be evaluated to determine the effect of casing thickness on gamma response. This experiment would also be modeled with the radiation transport code and model results compared to measurement data.

- <u>Perform additional modeling to investigate the effects of casing thickness, ^{90}Sr concentrations, and the presence of other radionuclides on spectral gamma response</u>.

  The borehole geometry with a distributed source is much more difficult to construct in a physical model, but numerical modeling can be performed to estimate response.

- <u>Collect additional sample data where possible and log all boreholes in which samples containing elevated ^{90}Sr concentrations are encountered.</u>

  Comparison of borehole log data and lab data in multiple boreholes will help validate the model studies.

References

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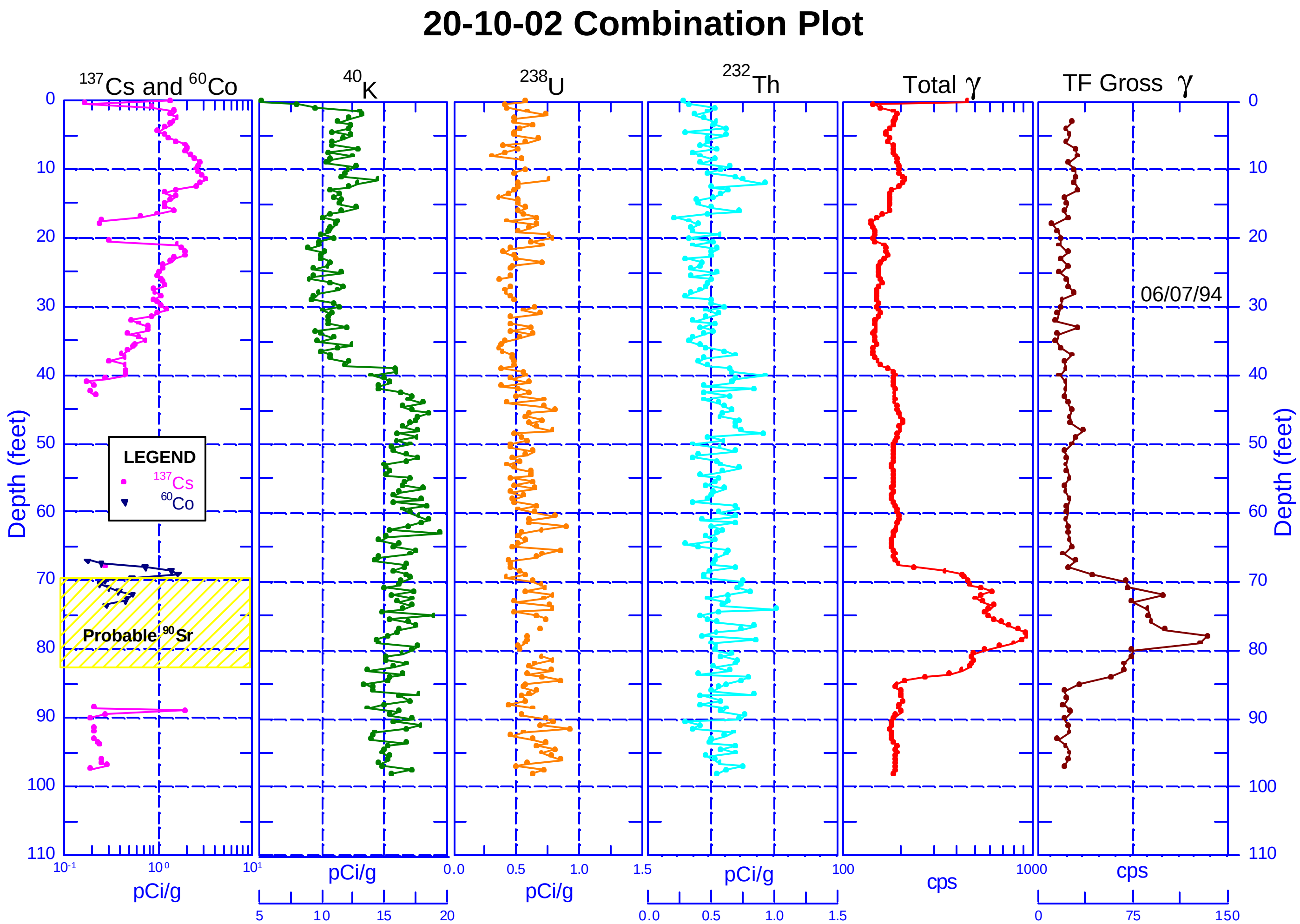

20-10-02 Combination Plot
137Cs and 60Co
40K
238U
232Th
Total γ
TF Gross γ
Depth (feet)
LEGEND
137Cs
60Co
Probable 90Sr
06/07/94
pCi/g
cps

Figure 1

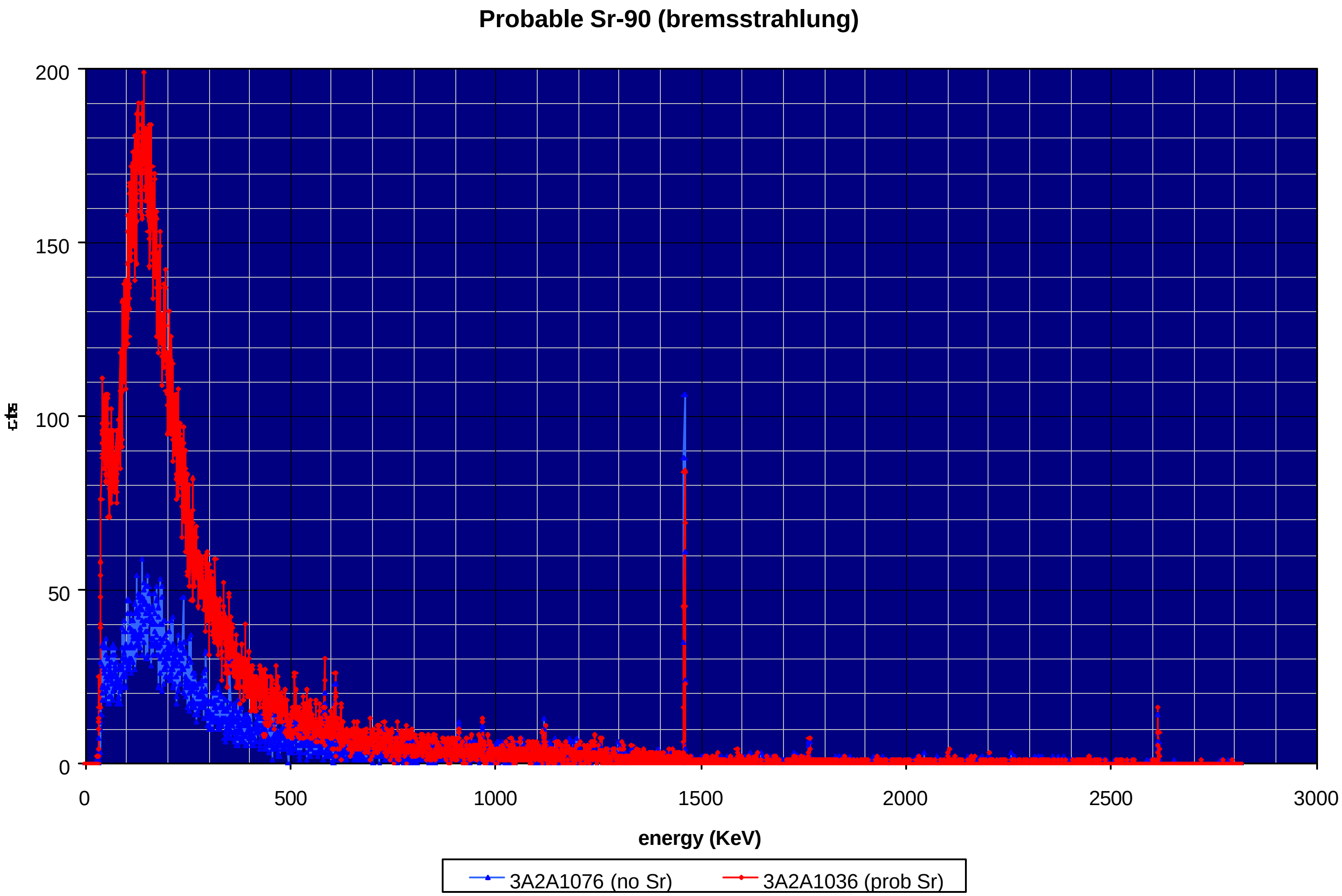

Probable Sr-90 (bremsstrahlung)
cts
0
50
100
150
200
0
500
1000
1500
2000
2500
3000
energy (KeV)
3A2A1076 (no Sr)
3A2A1036 (prob Sr)

Figure 2

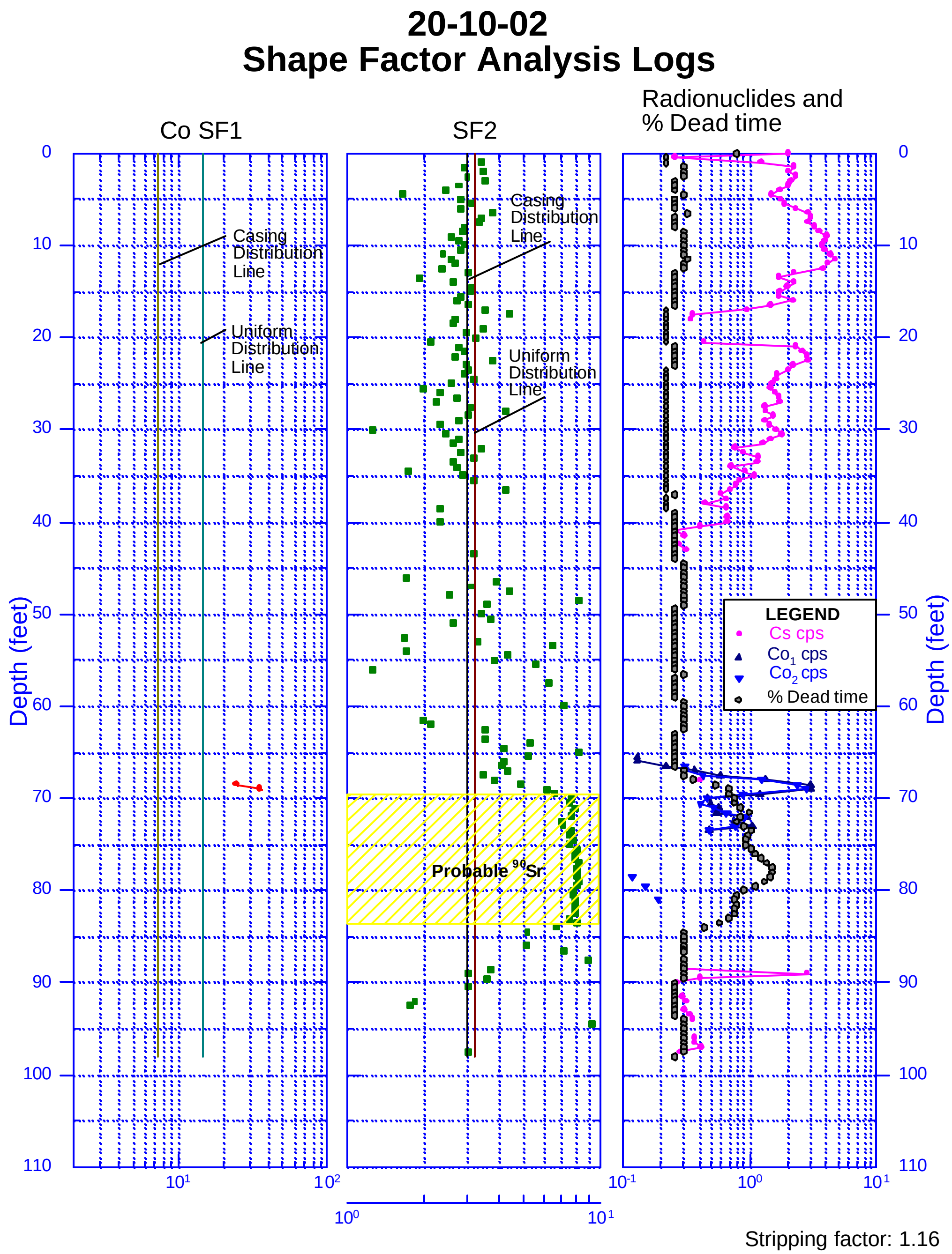
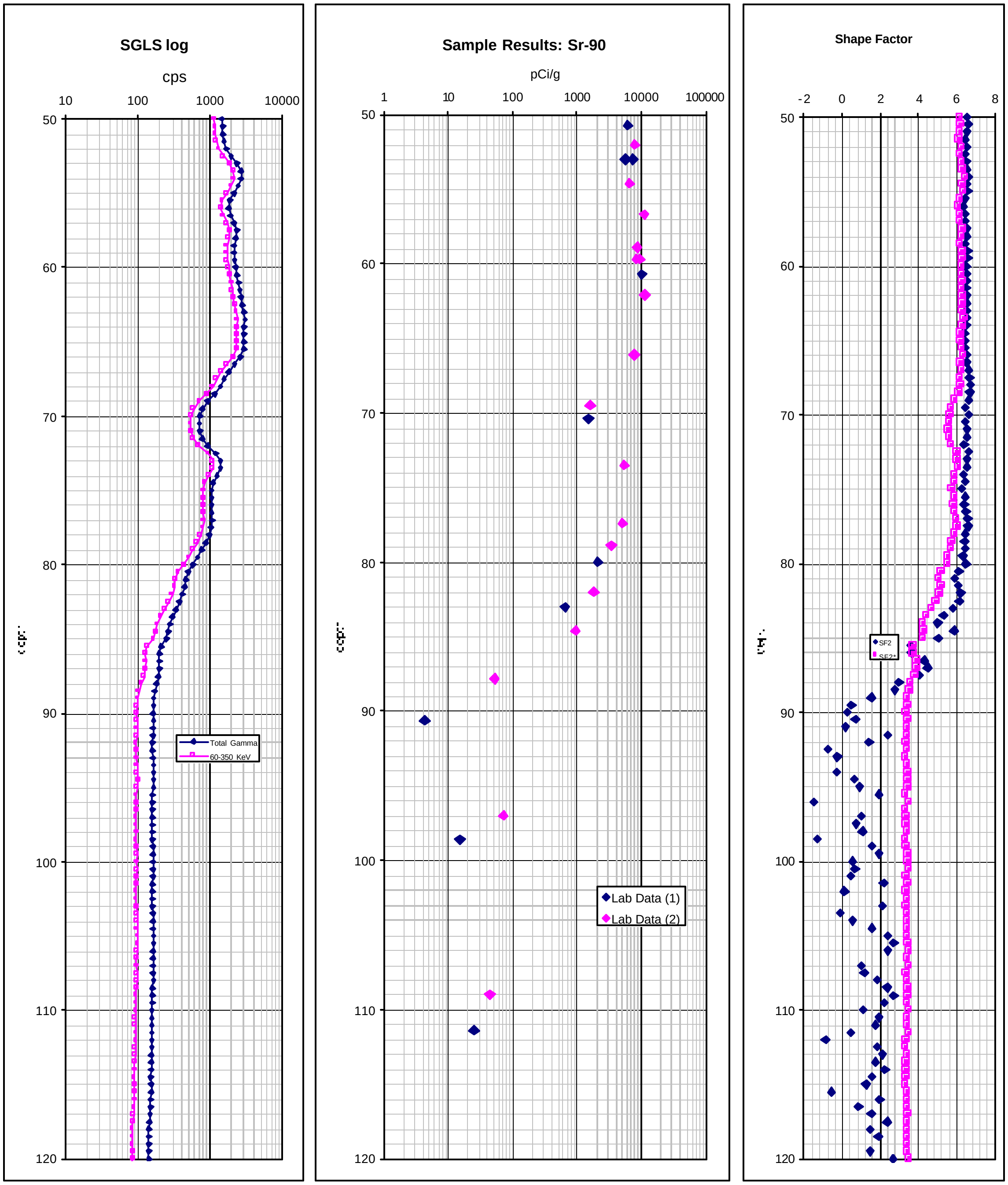

20-10-02
Shape Factor Analysis Logs
Co SF1
SF2
Radionuclides and
% Dead time
Depth (feet)
Casing
Distribution
Line
Uniform
Distribution
Line
Probable 90Sr
LEGEND
Cs cps
Co1 cps
Co2 cps
% Dead time
Stripping factor: 1.16

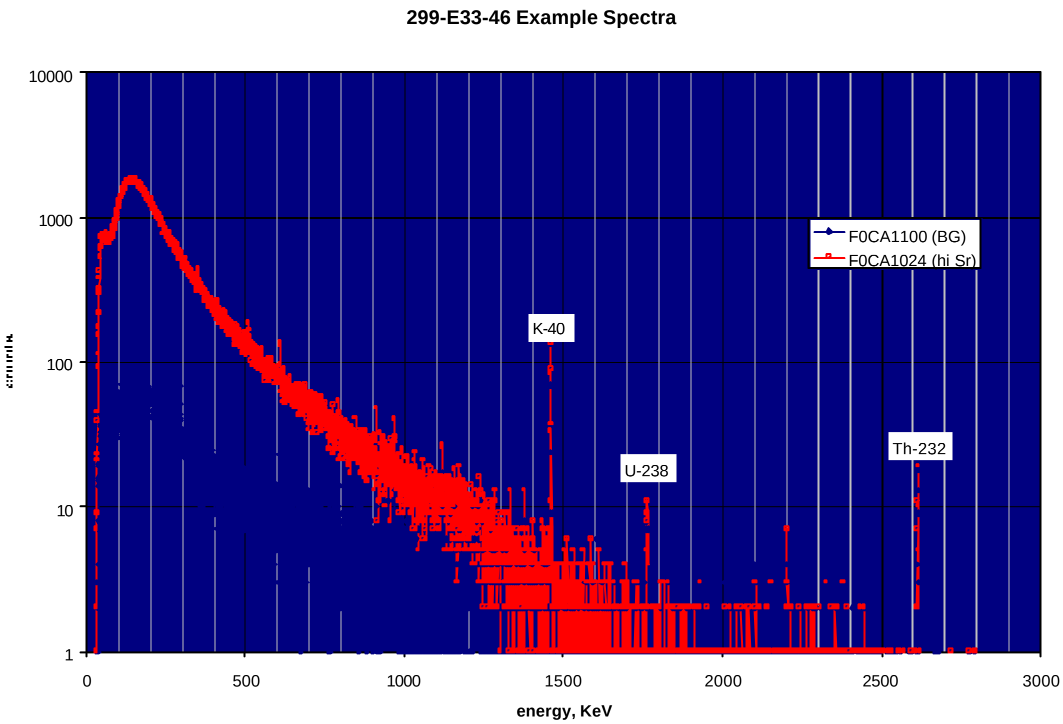
Figure 3

299-E33-46 (C3360)

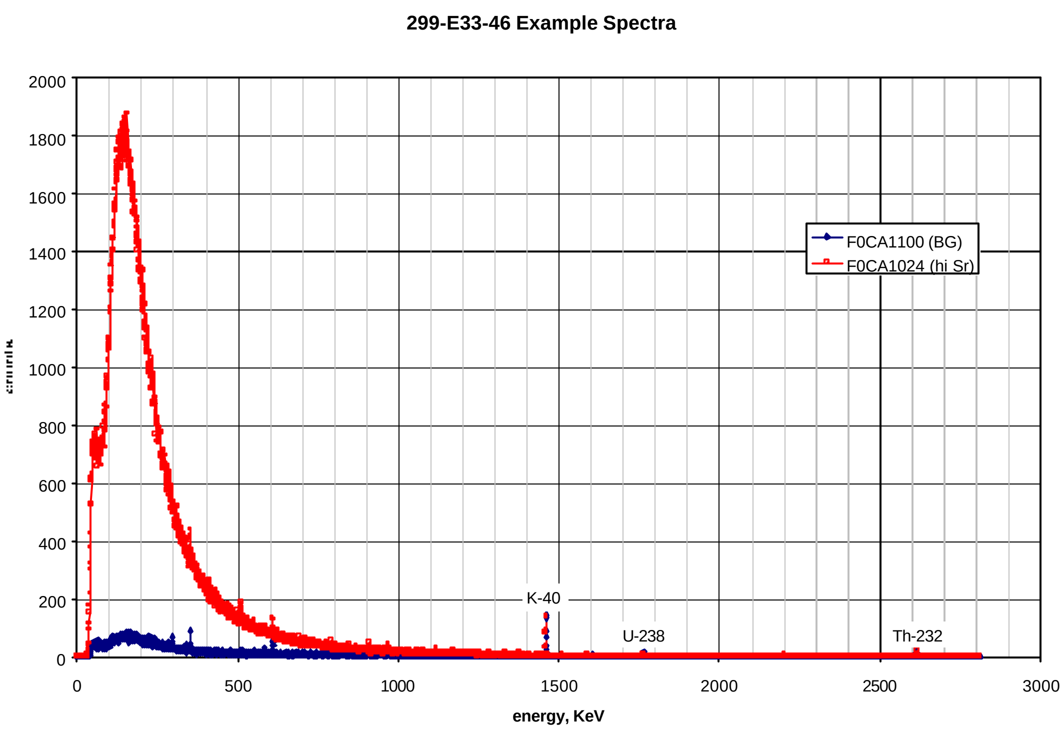


A B C

Figure 4



299-E33-46 Example Spectra
10000
1000
100
10
1
0
500
1000
1500
2000
2500
3000
energy, KeV
F0CA1100 (BG)
F0CA1024 (hi Sr)
K-40
U-238
Th-232



299-E33-46 Example Spectra
2000
1800
1600
1400
1200
1000
800
600
400
200
0
0
500
1000
1500
2000
2500
3000
energy, KeV
F0CA1100 (BG)
F0CA1024 (hi Sr)
K-40
U-238
Th-232

Figure 5

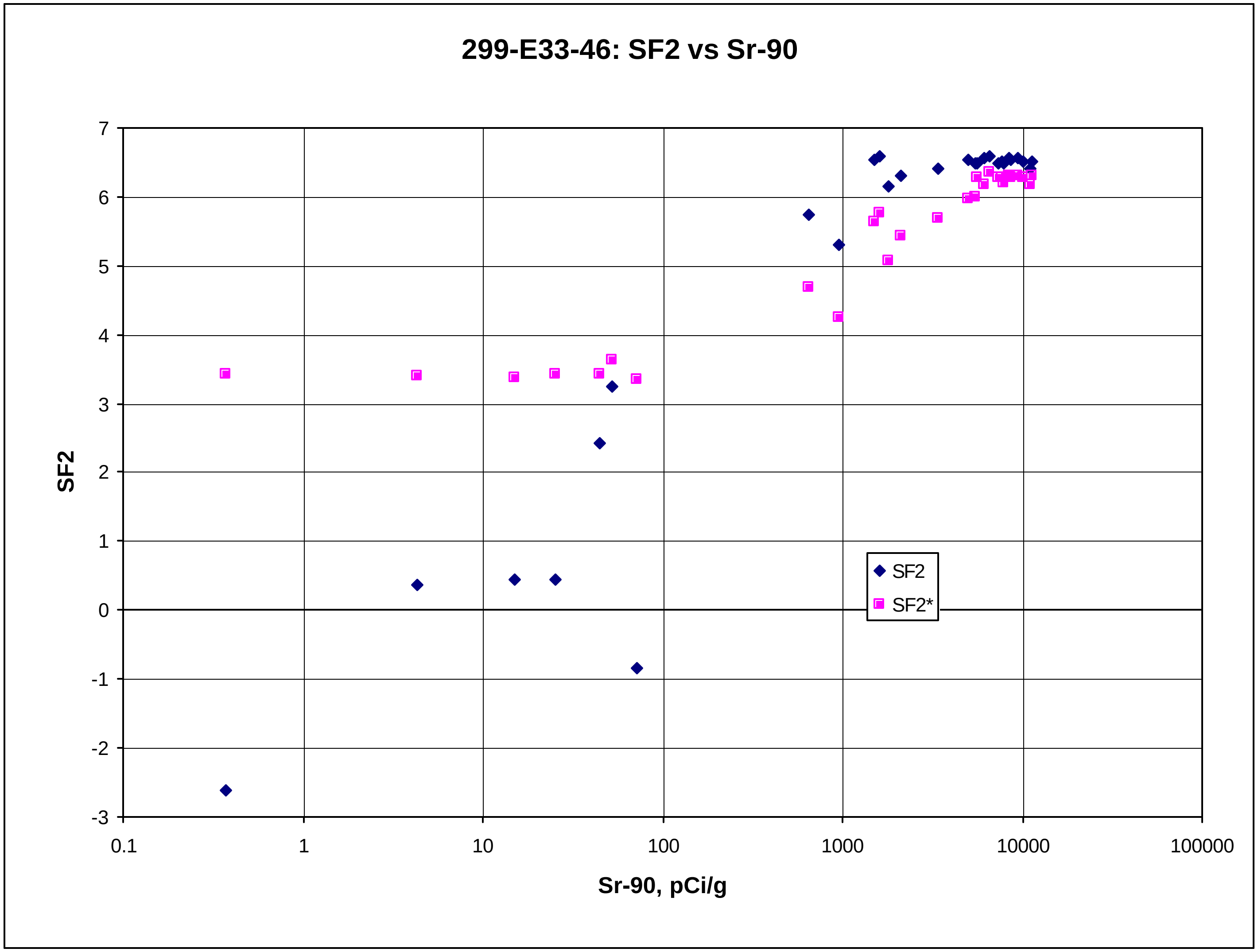

299-E33-46: SF2 vs Sr-90
SF2
7
6
5
4
3
2
1
0
-1
-2
-3
0.1
1
10
100
1000
10000
100000
Sr-90, pCi/g
SF2
SF2*

Figure 6

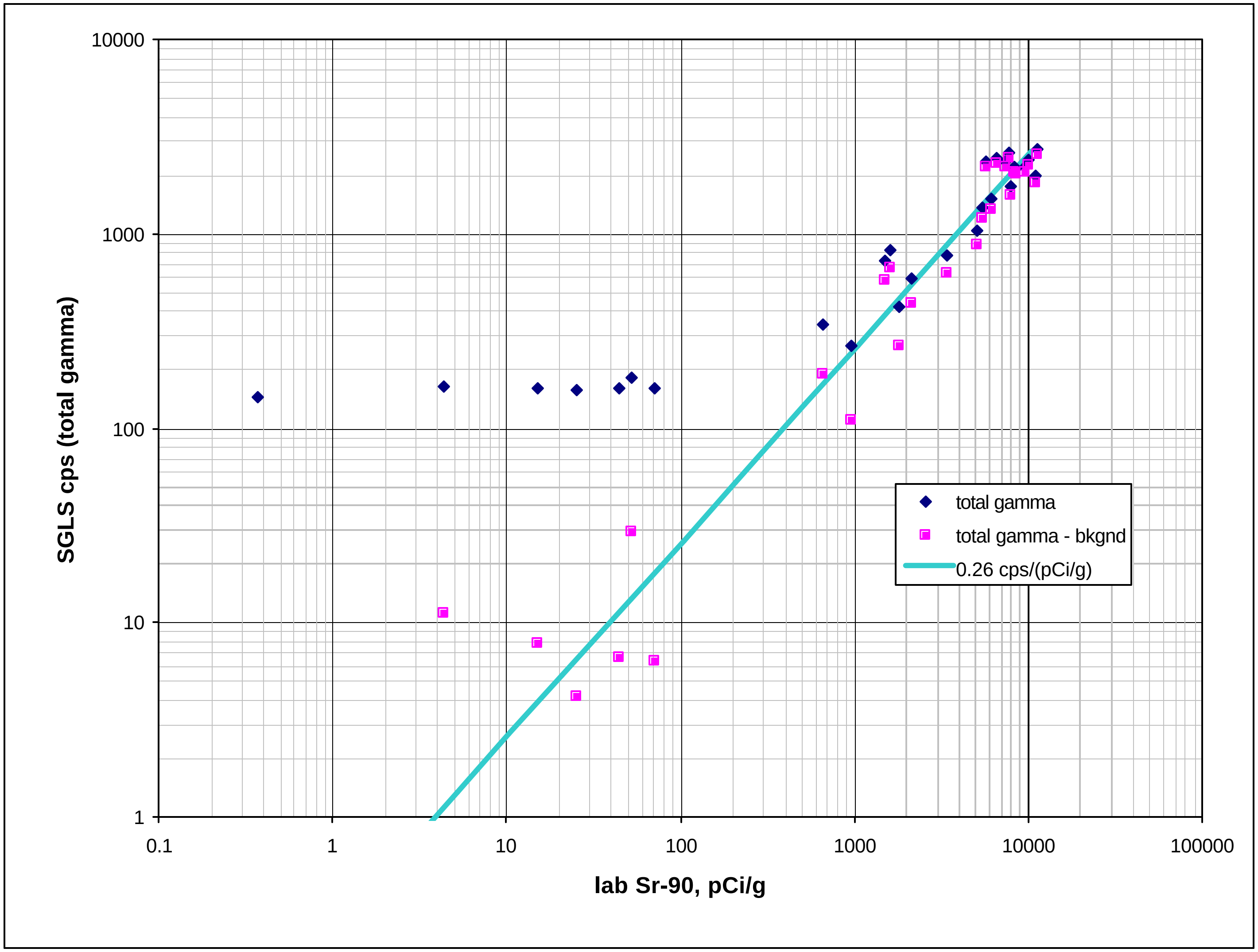

10000
1000
100
10
1
SGLS cps (total gamma)
0.1
1
10
100
1000
10000
100000
lab Sr-90, pCi/g
total gamma
total gamma - bkgnd
0.26 cps/(pCi/g)

Figure 7

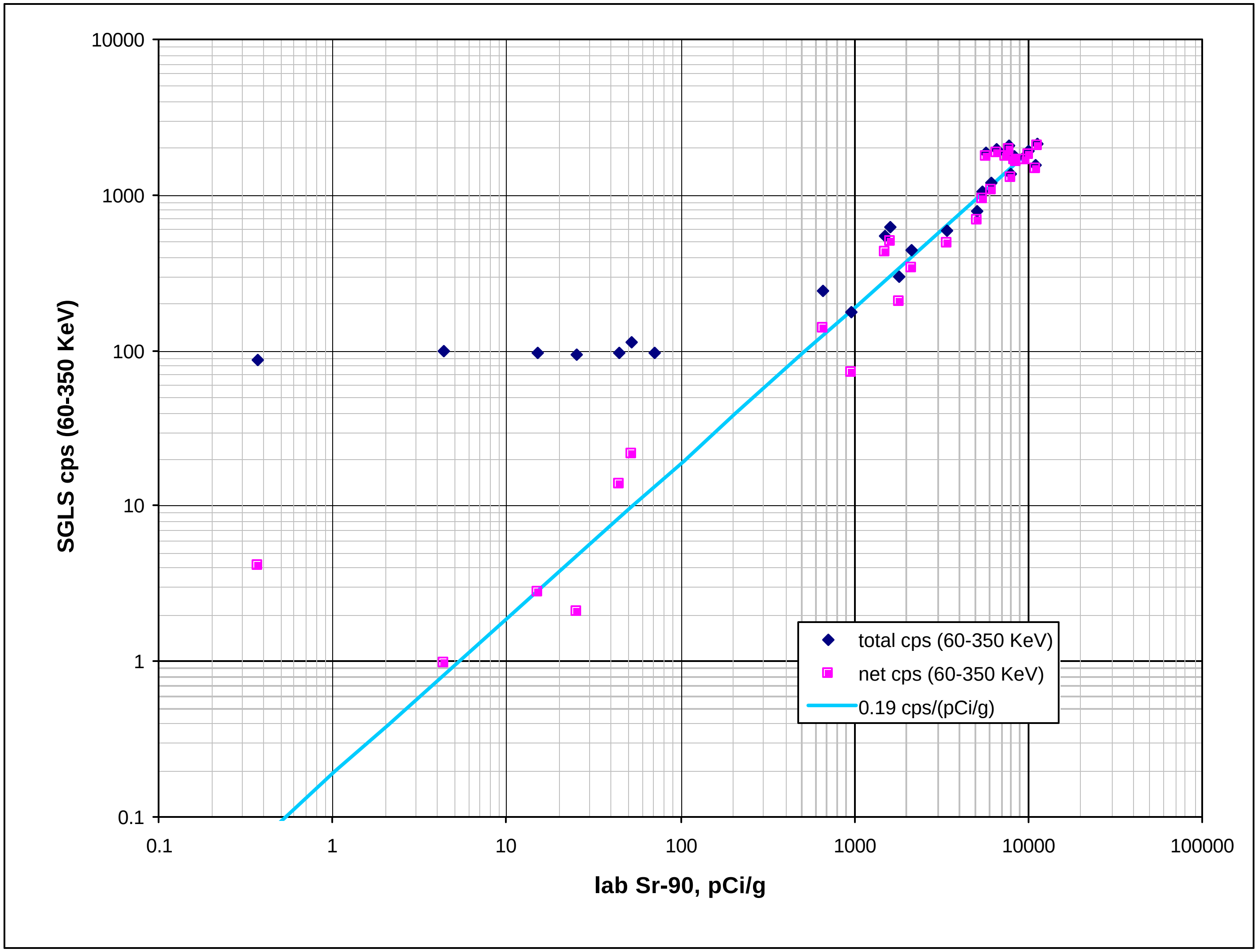

10000
1000
100
10
1
0.1
SGLS cps (60-350 KeV)
0.1
1
10
100
1000
10000
100000
lab Sr-90, pCi/g
total cps (60-350 KeV)
net cps (60-350 KeV)
0.19 cps/(pCi/g)

Figure 8