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HIGH RESOLUTION GAMMA-RAY SPECTROMETRY OF CULVERTS CONTAINING TRANSURANIC WASTE AT THE SAVANNAH RIVER SITE (U)

by

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ABSTRACT

A number of concrete culverts used to retrievably store drummed, dry, radioactive waste at the Savannah River Site (SRS), were suspected of containing ambiguous quantities of transuranic (TRU) nuclides. These culverts were assayed in place for Pu-239 content using thermal and fast neutron counting techniques [1]. High resolution gamma-ray spectrometry on 17 culverts, having neutron emission rates several times higher than expected, showed characteristic gamma-ray signatures of neutron emitters other than Pu-239 (e.g., Pu-238, Pu/Be, or Am/Be neutron sources). This study confirmed the Pu-239 content of the culverts with anomalous neutron rates [2] and established limits on the Pu-239 mass in each of the 17 suspect culverts by in-field, non-intrusive gamma-ray measurements.

INTRODUCTION

The TRU waste culverts are concrete cylinders approximately 2 m in diameter and 2 m tall with 15 cm thick walls and a 15 cm thick removable lid. Each can accommodate up to 14 stainless 200-liter (55-gallon) Type-A drums containing dry TRU contaminated waste. Each waste drum contains a PVC drum liner and is sealed before storage. The culverts are stored above ground on a concrete drainage pad in the SRS burial ground in a close-packed configuration blocking side access to most of the culverts. Neutron measurements on 118 culverts confirmed the Pu-239 loadings and determined they were critically safe. Higher than anticipated neutron emission rates were observed from 17 culverts which were then studied by high resolution gamma-ray spectroscopy.

EXPERIMENTAL METHODS

A high purity intrinsic germanium (HPGe) detector mounted in a down-looking cryostat was placed on top of the culverts for gamma-ray spectroscopic measurements. The HPGe detector had a 1.84 keV FWHM energy resolution at 1332 keV and a 26.0 % relative efficiency[3]. Energy and efficiency calibrations were performed using NBS-traceable gamma-ray sources. A two-point digital stabilizer compensated for gain shifts using the 88 keV line from a Cd-109 source fixed to the detector along with the 2614 keV gamma ray from Tl-208 natural background. The Cd-109 source also provided an internal check of live time/dead time corrections. Spectra were acquired with a computer-based 16K-channel multichannel analyzer and the data stored on removable disk. A small trailer equipped with a motor generator and an air conditioner housed the electronics and data acquisition system for the burial ground measurements. The detector signals were fed to the trailer through ≈35 meters of cable. A battery operated uninterruptible power supply maintained a constant voltage on the equipment.

A specially designed, graded lead shield, 5 cm thick, minimized the detection of radiations from any neighboring culvert. Cadmium covered the detector end-cap to reduce the thermal neutron flux. Borated polyethylene was interposed between the culvert and the detector to thermalize any fast neutrons and further reduce the neutron background. Measurements were made at two different elevations on each culvert, one with the detector sitting directly on the culvert lid and one sitting on a table 70 cm above the lid. An overhead crane was used to position the detector/shield assembly. The measurements were conducted at the point of highest dose rate on each culvert.

EXPERIMENTAL RESULTS

Gamma-ray spectra were accumulated and recorded on disk. Typical counting times were 4 and 16 hours for the detector sitting on the culvert lid and on the elevated table, respectively. Table I lists the principal radiations observed in the spectra and notes their corresponding origins. Many of the radiations arise from radiative capture gamma rays in the surrounding waste matrix, from inelastic scattering of the alpha particles on low-Z materials, and from (α,n) reactions. Some of the artifact radiations are highly dependent on the chemical form of the plutonium.

The gamma-ray signatures of each culvert helped resolve many of the ambiguities observed in the neutron emission rates. For instance, several culverts were found to contain isotopic neutron sources of Am/Be or Pu/Be. In these cases, the characteristic prompt gamma rays from the (α ,n) reaction on Be at 4425 keV and the single and double escape peaks were obvious in the spectra. This unique feature in a spectrum obtained from a culvert containing an Am/Be source is shown in Figure 1. Many of the culverts contained Pu-238 which correlated well with the 766 keV gamma ray and with the 2614 keV gamma ray from Tl-208. While the latter is not in the 238 decay chain, the production process introduces U-232 as an impurity. The Cd capture gamma rays were used as an internal check of the neutron emission rates and correlated well with the field measurements [1].

To determine the sensitivity for the detection of Pu-239 in an unknown waste matrix, measurements were performed using small sources of Pu-239 and Pu-238 in several chemical forms (metal, oxide, and fluoride) in an empty culvert. Absorption coefficients were determined for known components (e.g., culvert lid, waste drum liners and lids, polyethylene, etc.) by placing the Pu sources and other radioisotopic standards at well defined geometrical locations within the test culvert. Calibration curves were developed for each configuration and the results compared to the predictions of a gamma-ray transport computer code [4]. The results of the calibration runs on the simulated waste helped establish the detection sensitivity and limits for the assays performed on the actual waste culverts in the field. It was determined that a 10 gm source of Pu-239 could be detected on the bottom of the culvert surrounded by a matrix of drums packed with polyethylene in an overnight count.

The analysis of all spectra was performed using SPECTRAN-AT [5] and also with MicroSAMPO [5]. Any discrepancies were resolved by hand analysis. Figure 2 shows a typical spectrum obtained in the SRS burial ground from one of the suspect TRU waste culverts. This Figure displays the prominent Pu-239 gamma rays used in this study. The peak intensities were first adjusted for the intrinsic efficiency of the HpGe detector. The data were then corrected for known absorbers. These included the borated polyethylene surrounding the detector (density 0.92 gm/cc), the concrete lid on the culvert (density 2.35 gm/cc), one thickness of PVC drum liner (density 1.2 gm/cc) and one drum lid. These attenuation factors were generated with the MICROSHIELD computer code after they had been experimentally verified during the calibration runs.

The rate of gamma rays striking the detector at each energy, uncorrected for the waste matrix, was then determined. This rate, was then correlated with the reported Pu-239 content. It was found that five of the gamma rays associated with the decay of Pu-239 correlated well with the reported Pu-239 content. The other Pu-239 gamma rays were either too weak for useful measurements or had considerable interference from other radiations in that spectral region. A summary of the correlations with the detector sitting on the lid of each culvert is given in Table II. The reported Pu-239 loading as compared to the calculated loading from the 414 keV gamma ray rate is shown in Figure 3. The result of multiple regression analyses for the five gamma rays confirms the Pu-239 loadings for the 17 suspect culverts to a precision of ± 101 gms.

To assess the accuracy of the reported Pu-239 content in the waste, a number of models were generated with the Pu-239 in a variety of distributions and configurations. First the Pu-239 was uniformly distributed throughout the barrels of waste and the count rates for the five gamma rays calculated at the two detector locations. The density of the waste matrix was varied from the maximum of 0.2 gm/cc to no self absorption. (The maximum had been experimentally determined by packing a drum with polyethylene.) The distribution was then changed to a point source at various radial and axial locations and the count rates compared to the experimental rates at various matrix densities. Other models generated were multiple point sources and disk sources at various locations within the culvert. A total of 43 different models were generated with the MICROSHIELD computer code. As an example of the comparison of the calculated count rates at the different gamma-ray energies, Figure 4 shows a comparison of the experimental count rates with the calculated count rates for the reported Pu-239 content as a point source lying in the bottom of a barrel under the detector in a uniform waste matrix of 0.2 gm/cc of polyethylene. The figure shows the ratio for all culverts at the two measurement locations. The average ratio is 2.1±1.3 for all the culverts. If the density were reduced by 30%, the ratio would be unity. The point source configuration and the uniformly distributed source represent the count rate extremes and differ by a factor of ≈200. The energy of the Pu-239 gamma rays are too close to determine any self-absorption differences caused by large lumps of plutonium.

Condensing the reported Pu-239 contents into point sources located in the bottom of each layer of drums yielded gamma-ray count rates consistent with the measured values at the expected waste matrix densities. While a unique value of the Pu-239 content is not possible from such measurements, the predicted values were within a factor of five of the reported values when the maximum waste density was used in the point source model. Reductions in the density reduced the overall deviations from the experimental values.

CONCLUSIONS

Pu-239 masses were estimated using the peak intensities from each of the five principle gamma rays. When appropriate attenuation coefficients are used, each peak should give the same mass result within a certain statistical variance. After correcting for known attenuations, arriving at the "best" attenuation coefficients is then an itterative process. Comparing the itterative result with known matrix effects, one can estimate the consistency of the method. Comparing the observed rates with the detector in two locations with the predictions using known or expected absorption coefficients yielded confirming evidence of the accuracy of the method. However, due to the large number of variables in the waste matrix, accurate inventories of Pu-239 in the waste was impossible by this technique. True far-field measurements were not possible due to the close proximity of other culverts.

The higher than expected neutron count rates for many of the culverts are attributed to neutron sources other than Pu-239. While the gamma-ray assay of the TRU waste cannot be used to uniquely determine the quantity of Pu-239 in the TRU waste, it demonstrated consistency with the estimates made prior to loading the waste drums contained in the culverts. Applying known attenuation coefficients to the data, the precision was determined to be ±101 gms. Using internally consistent attenuation coefficients with a point source model, the measured masses agree with the loading data within a factor of three for the 17 suspect culverts. This study confirms that there is little likelihood of an inadvertent criticality in the entire group of TRU waste culverts.

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Table 1. The Principal Gamma Rays Observed in the Spectra Recorded during Assay of the TRU Waste Culverts and their Origin.

Energy* keV	Origin
345.0	Pu-239
375.0	Pu-239
393.1	Pu-239
413.7	Pu-239
451.5	Pu-239
558.0	Cd capture
609.3	Bi-214
646.0	Pu-239
651.0	Cd capture
727.0	Bi-212
742.8	Pu-238,Pa-234
766.4	Pu-238,Pa-234
769.4	Pu-239
786.0	Pu-238,Bi-212
890.0	F (α,α')
911.2	Ac-228
969.0	Ac-228
1001.0	Pa-234m
1274.0	F (α,α')
1620.6	Bi-212
1764.5	Bi-214
1943.0	Ca capture
2223.0	H capture
2614.4	Tl-208
3538.0	Si capture
4425.0	Be capture
4933.0	Si capture
7117.0	Fe capture
7630.0	Fe capture
7645.0	Fe capture

* Gamma-ray energies from radioactive decay taken from Reference 6, capture gamma-ray energies taken from References 7 and 8, and inelastic scattering gamma-ray energies taken from Reference 9.

Table 2. Summary of the results of the correlation of the gamma-ray count rates with the reported Pu-239 contents of the 17 suspect culverts.

Energy (keV)	Corr. Coef. (R^2)	Std. Err. of Y-est.(gms)
345.0	0.803	99.5
375.0	0.772	107
393.1	0.818	95.7
413.7	0.749	112
451.5	0.593	143

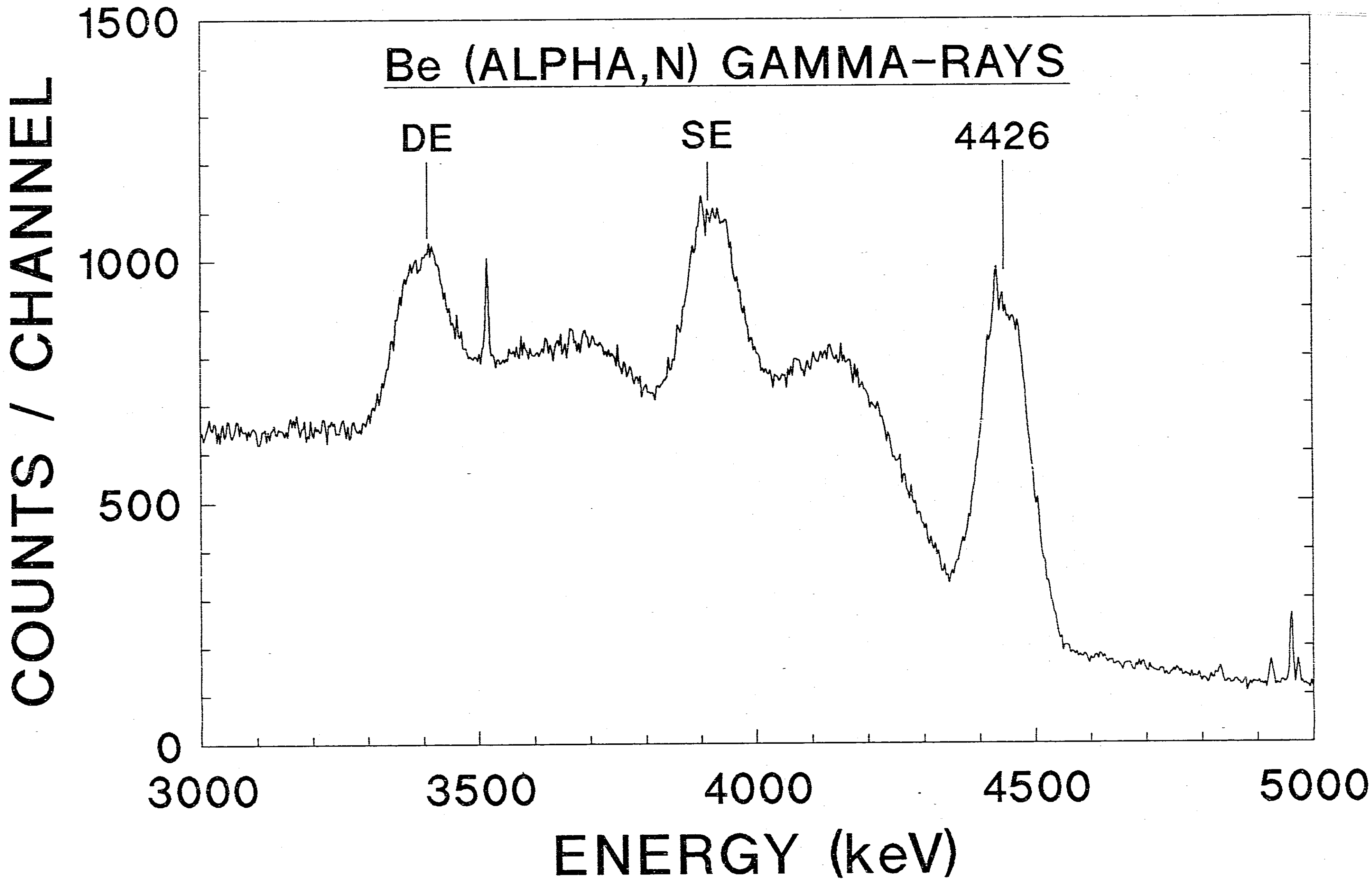
Figure 1. A portion of the gamma-ray spectrum taken on a culvert containing an isotopic Am/Be neutron source. The region of the spectrum shows the severely Doppler broadened prompt gamma ray at 4425 keV along with the single and double escape peaks.

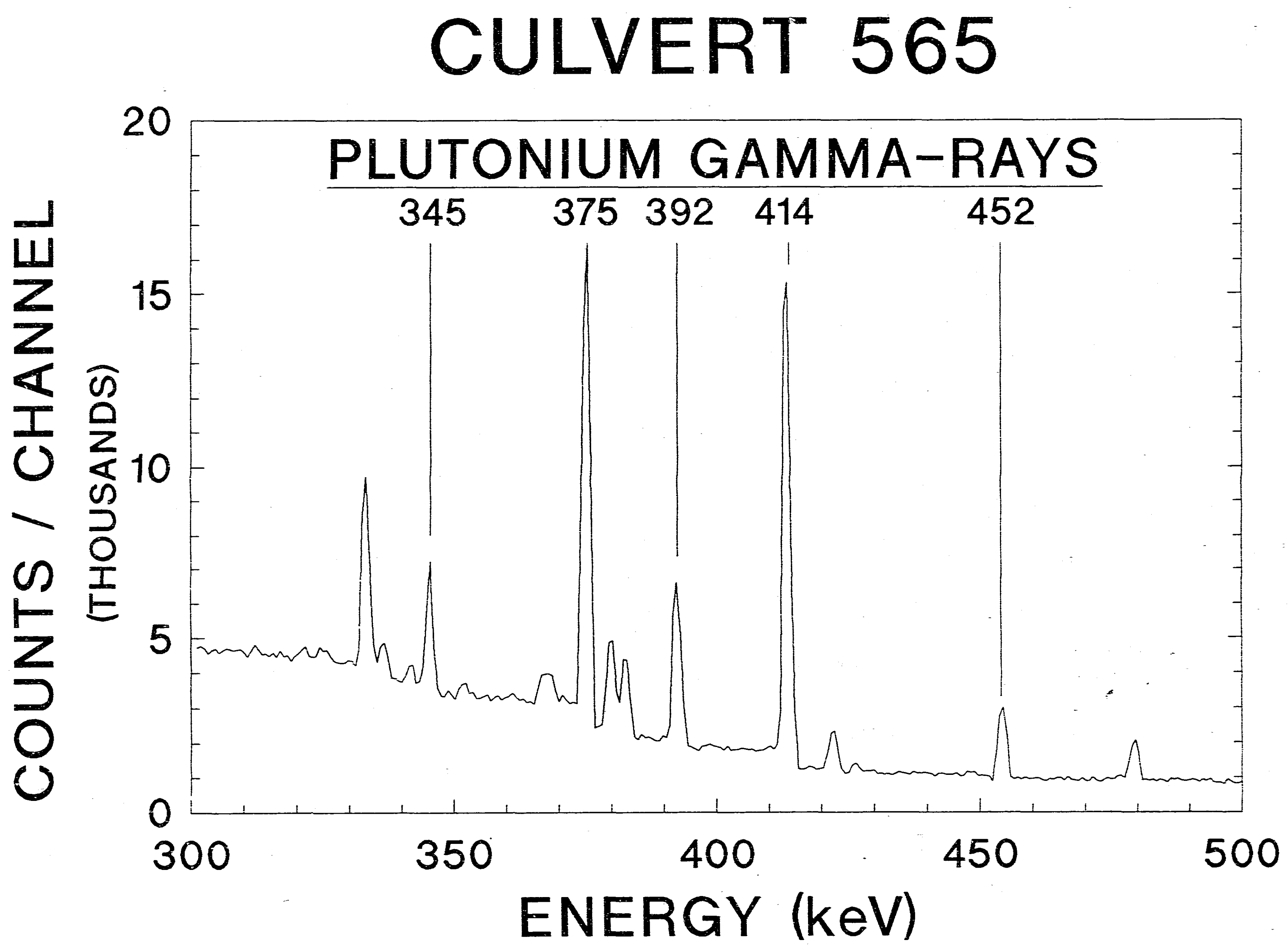
Figure 2. A portion of the gamma-ray spectrum showing the prominent peaks from Pu-239 used in this study.

Figure 3. Pu-239 contents of each culvert as reported and as calculated from the correlation with 414 keV gamma-ray intensity.

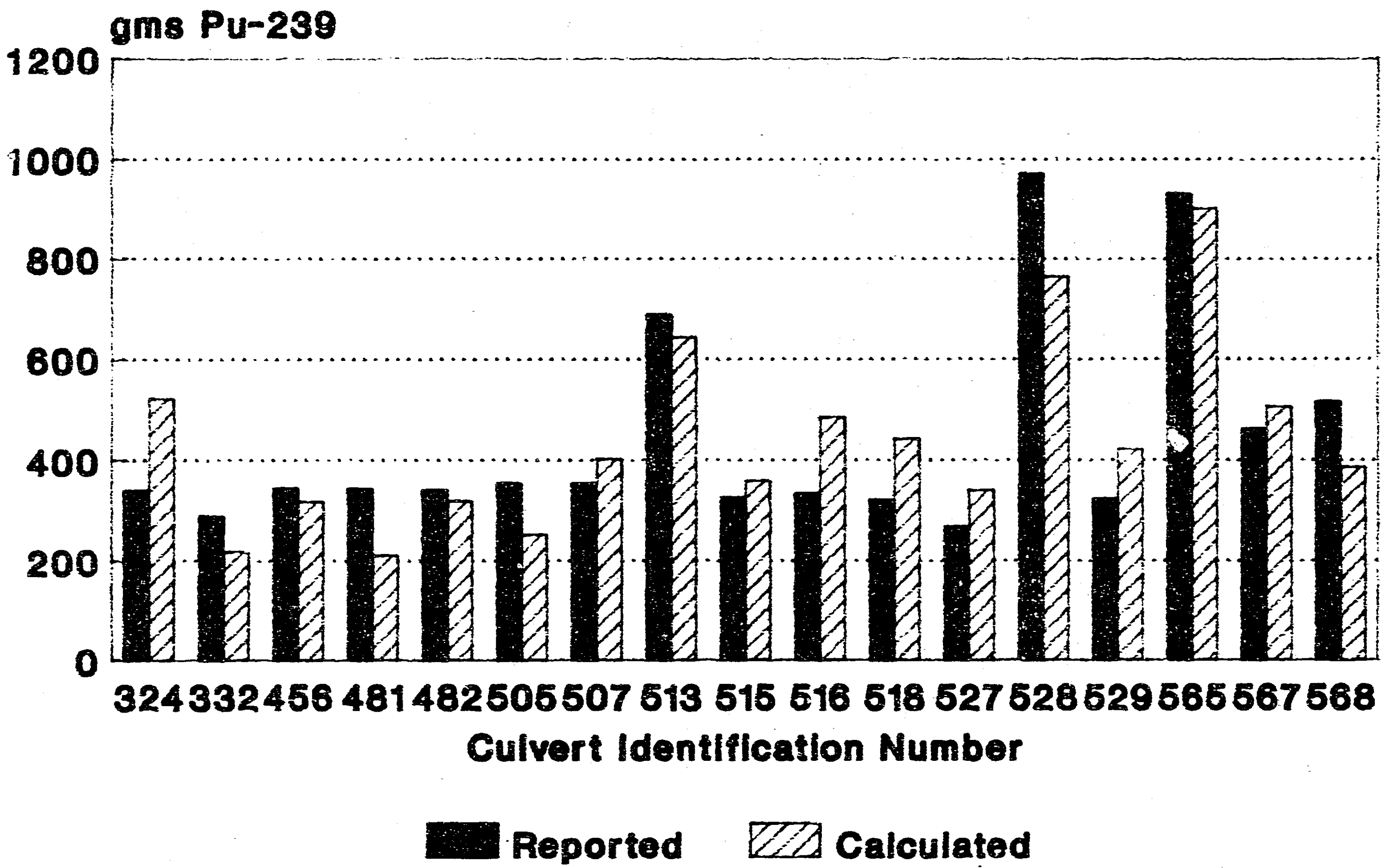
Figure 4. The ratio of experimental to calculated count rates for the 414 keV gamma ray from Pu-239 from each TRU waste culvert. See text for model details.

CULVERT 481

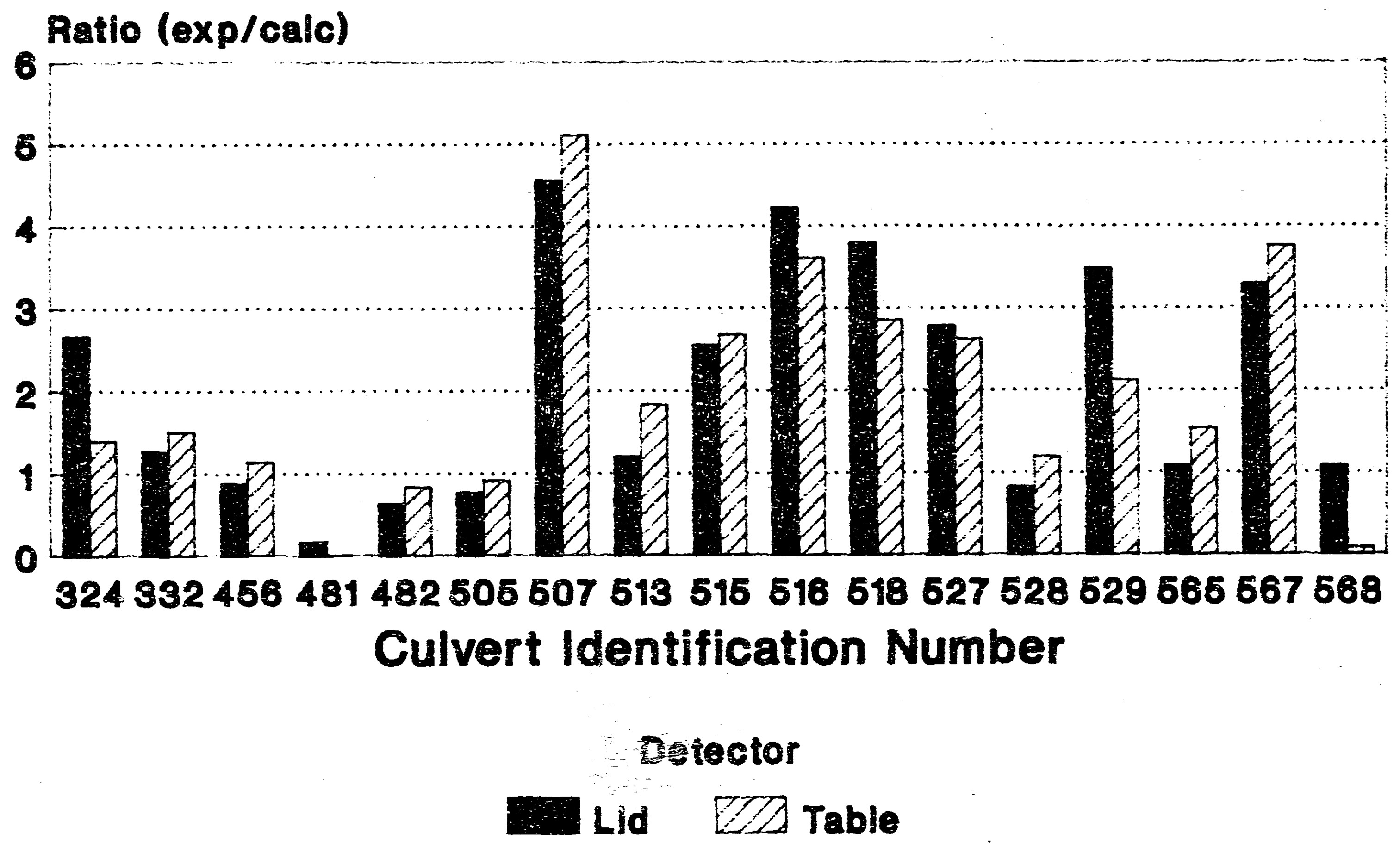

Be (ALPHA,N) GAMMA-RAYS
DE
SE
4426
1500
1000
500
0
COUNTS / CHANNEL
3000
3500
4000
4500
5000
ENERGY (keV)


CULVERT 565
PLUTONIUM GAMMA-RAYS
345
375
392
414
452
COUNTS / CHANNEL
(THOUSANDS)
0
5
10
15
20
300
350
400
450
500
ENERGY (keV)

Emperical Fit
to 414 keV Count Rate



Point Source Model (density = 0.2 gm/cc)



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