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The Influence of Heterogeneity in Gamma Spectroscopy Analysis of Soil Contaminated with Weapons Grade Plutonium at the BOMARC Missile Accident Site, McGuire AFB NJ

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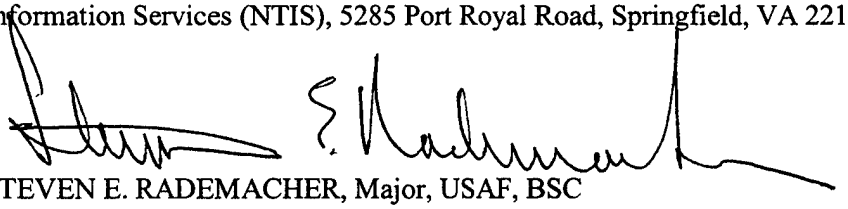

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The BOMARC Missile Site contains weapons grade plutonium (WGP) as the result of a nuclear weapon accident that occurred in 1960. Numerous environmental investigations have been performed on the site during the 40 years post accident. Many of the investigations have been focused on the location of the residual WGP and activity concentrations. WGP in the environment is notorious for its heterogeneous distribution in soils. Past characterization efforts have noted this characteristic for the WGP at the BOMARC site and the difficulties it presented in accurate description of site conditions. Direct measurement of WGP in soils can be accomplished through gamma- and alpha-spectroscopy. Due to the low-energy and low-frequency of the photons emitted from Pu-239/240, gamma spectroscopy is difficult and error prone. Direct measurement by the expensive and time-consuming process of dissolution, chemical extraction, and alpha-spectroscopy analysis is also error prone for heterogeneously distributed plutonium. A less costly and considerably more timely laboratory analysis consists of evaluation of the 60 keV gamma-ray emitted by Am-241, a decay product of WGP. This process relies on a known relationship between the Am-241 and the Pu-239/240. This report describes the errors inherent in this method for WGP from the BOMARC site and the reduction of errors from a conjugate counting methodology. The findings demonstrated that many of the soils were laden with heterogeneity that affected measurement accuracy. A sizeable fraction, however, had little observable heterogeneity. In addition, vertical distributions and correlation coefficients to a portable in-situ gamma-radiation measurement device are provided.

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List of Acronyms

AFIERA - Air Force Institute for Environment, Safety and Occupational Health Risk Analysis

BOMARC - Boeing Michigan Aeronautical Research Center

CI - confidence interval

EPA - Environmental Protection Agency

FIDLER - field instrument for detecting low energy radiation

HpGe - hyperpure germanium

HQ AFSC - Headquarters, Air Force Safety Center

HQ AMC - Headquarters, Air Mobility Command

NaI(Tl) - thallium-drifted sodium iodide

LANL - Los Alamos National Laboratory

LLL - Lawrence Livermore Laboratory

MDC - minimal detectable concentration

NAEG - Nevada Applied Ecology Group

NIST - National Institute for Standards and Technology

NTS - Nevada Test Site

ORNL - Oak Ridge National Laboratory

% CV - percent coefficient of variation

REECo - Reynolds Electrical and Engineering Company

RPD - relative percent difference

WGP - weapons grade plutonium

Abbreviations

Am - americium

cm - centimeter

^{o}C - degree Centigrade

g - gram

h - hour

keV - kilo electron volt

μm - micrometer

mm - millimeter

MeV - million electron volt

μ - linear attenuation coefficient

n - number

pCi - picocurie

Pu - plutonium

ρ - density

R^2 - squared correlation coefficient

σ - standard deviation

s - second

y – year

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Introduction

The Boeing Michigan Aeronautical Research Center (BOMARC) Missile Site is an inactive Air Force installation located in Plumstead Township, New Jersey. The site was an active nuclear missile defense site from 1958 – 1971. On June 7, 1960, a fire occurred in one of the shelters in which the shelter, missile, and warhead were partially consumed by the fire. The high explosive materials in the weapon ignited but did not detonate. The most intense period of the fire lasted about one hour. Water was applied to the shelter and weapons during the fire by the installation fire department. The fire melted the weapons grade plutonium (WGP) that was contained in the device. Turbulent local atmospheric conditions and the water applied during the fire contributed to scattering of WGP to the environment. WGP is the primary radiological concern at the site. The amount of WGP originally contained in the weapons remains classified. Los Alamos National Laboratory estimated that approximately 100 g of WGP remained on-site (Farley 1996). A recent characterization survey supports an estimate closer to 300 g (OHM 1998).

Quantification of WGP in soils is a technically challenging endeavor compared to many naturally-occurring or other man-introduced radiological contaminants. A large source of the difficulty with sampling and analysis of soil samples is related to the discrete particulate nature of plutonium contamination (referred to as "heterogeneity" in this report) (Bernhardt 1976). In the 1960's and early 1970's, the preferred analytical method for soil samples containing WGP was chemical dissolution and α-spectroscopy analysis. Because of practical limitations in aliquot masses prepared for dissolution (i.e., less than 50 g), the method was plagued with large uncertainties if plutonium contaminants were in discrete particle form. The uncertainties from this method were largely due to the inability to collect samples representative of the contaminated area rather than from systemic or random laboratory errors.

With improvements in detector technologies, alternate methodologies were developed. The Field Instrument for Detecting Low Energy Radiation (FIDLER) is a specialized NaI(Tl) detector designed for in-situ measurements of WGP (Tinney 1969). Later, high-resolution γ-spectroscopy systems were commonly used for laboratory assessment of plutonium activity concentrations from ^{241}Am quantification and knowledge of the relationship of the ^{241}Am and plutonium. This method, while dependent on accurate assessment or knowledge of the plutonium to ^{241}Am activity concentrations, can be used to evaluate samples with mass up to a kilogram. The ability to evaluate sample masses of this magnitude offers significant advantage over chemical dissolution and α-spectroscopy analysis where sample mass is highly limited. First, the γ-spectroscopy method does not require chemical procedures that are labor intensive and time consuming. Second, the larger sample mass ensures that the sample is more representative of the environmental area being evaluated. On a negative note, however, little research has been conducted to investigate the effect of heterogeneity on uncertainties on laboratory γ-spectroscopy analysis assessments.
This report summarizes the results of a sampling and laboratory analysis effort on BOMARC site soils. The primary purpose of the study was to better understand the effects of heterogeneity on sample collection and analysis through γ-spectroscopy of a WGP contaminant. The study demonstrated that the particulate nature of the WGP contaminant created significant levels of uncertainty in quantified activity concentrations for some samples evaluated. The impact of the uncertainties is largely dependent on the purpose of the assessment. For a site characterization

effort, a high degree of accuracy in analytical results may not be necessary. In this case, the impact is negligible. For a research study, however, uncertainties from a heterogeneously distributed contaminant may greatly limit the purpose of a study. For site closure measurements, where post remediation residual WGP contamination may be very low, uncertainties may be overwhelming and greatly limit a conclusion on the effectiveness of the effort. In addition, for low activity concentration samples, overall uncertainty will be exacerbated by that introduced by counting uncertainties alone. A method to reduce the effects of heterogeneity on sample analysis is provided.

Background

1. Weapons Grade Plutonium.

WGP is comprised primarily of ^{239}Pu, with lesser mass amounts of ^{238}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu. For many nuclear weapons, individual WGP isotopic assay information exists (classified). However, for the warhead on the BOMARC missile and other nuclear weapons produced during the same time, specific assay information is not available (Taschner 1998). Table 1 provides an estimate of the isotopic composition of the BOMARC missile based on information from Los Alamos National Laboratory (Taschner 1998) and soil analyses performed in 1997 (Rademacher 1999).

Table 1. Isotopic Composition (by mass) of WGP in BOMARC Weapon Based on Los Alamos National Laboratory Estimates and Soil Analyses (Rademacher 1999).

Isotope	Mass Fraction*	Radiological Half-life (y) **
Pu-238	0.0099	87.74
Pu-239	0.937	24,110
Pu-240	0.056	6,560
Pu-241	0.0047	14.35
Pu-242	Negligible	376,000

* Fractions in 1958 ** Walker *et al* 1984

There was variability in the isotopic composition of WGP among weapons produced during this period because of variables involved in WGP production. According to Los Alamos National Laboratory records (Taschner 1998), the plutonium in the BOMARC missile (involved in the fire at McGuire AFB) may have been separated in 1957/1958. The relative isotopic composition of WGP constituents changes over time due to radioactive decay. Shortly after chemical separation during production, the most significant change is due to the radioactive decay of ^{241}Pu:

$$^{241}\mathrm{Pu} \rightarrow {}^{241}\mathrm{Am} + {}^{0}_{-1}\beta .$$

The daughter product, ^{241}Am, is an α-particle emitter with a radiological half-life of 432 y (Walker *et al* 1984).

Table 2 lists the major radiation(s) emitted by the primary constituents of WGP. For ^{239}Pu and ^{240}Pu, only infrequent low-energy photons are emitted. Direct assessment of either of these isotopes in samples can be accomplished through high-resolution gamma spectroscopy if sample activities are reasonably high. Measurement accuracy can be severely affected if the density and elemental

composition of the sample are not well characterized. Better measurement accuracy can be obtained through chemical dissolution, separation, and alpha or mass spectroscopy. However, this measurement approach has shortcomings. To ensure measurement accuracy, the sample must be completely dissolved by strong acids. The requirement for complete dissolution is time consuming and limits sample mass. Many laboratories limit sample mass to about 10 g, with many commercial environmental laboratories using less than 2 g. Further, application of acid leaching methods on BOMARC soils was effective in solubilizing only about 50 % of the contaminant (Refosco 2001). Small sample mass can introduce variability into the activity assessment if WGP is heterogeneously distributed. Most importantly, the method is expensive because it involves wet chemistry and can be cost-prohibitive for a large number of samples. Because the α-particle energies of the ^{239}Pu and ^{240}Pu are very close, α-spectroscopy is incapable of resolving the two isotopes. Practically, for radiation protection purposes this does not present a problem because each isotope has the same activity to dose conversion factor (Eckerman 1999).

Table 2. Major Radiation Emissions of WGP Constituents (Scheien 1992).

Radionuclide	α-Particle Energies (MeV) & Frequency	β-Particle Energies (MeV) & Frequency	Photon Energies (MeV) & Frequency
Pu-239	5.155 (0.733) 5.143 (0.151) 5.105 (0.115)	None	0.113 (0.0005) 0.014 (0.044)
Pu-240	5.168 (0.735) 5.123 (0.264)	None	0.054 (0.0005) 0.014 (0.11)
Pu-241	None	0.021 (1.00)	None
Am-241	5.486 (0.852) 5.443 (0.128) 5.388 (0.014)	None	0.014 (0.427) 0.0595 (0.359) 0.026 (0.024)

Of the radionuclides in WGP, ^{241}Am has the most favorable photon emission characteristics with a 0.0595 MeV γ-ray at an emission frequency of 36 %. This photon is of sufficient energy to afford reasonable measurement sensitivity in high-resolution gamma spectrometry counting systems. Additionally, the attenuation is not as severe as that of the 17 keV x-ray emitted by ^{239}Pu or ^{240}Pu. In laboratory and in-situ environmental measurements of WGP, measurement of the 0.0595 MeV γ-ray from ^{241}Am is often used as a surrogate for $^{239/240}$Pu, provided the ratio of activities between the two elements is well known.

2. Plutonium Characteristics in the Environment.

Plutonium is a silvery-white metal that readily oxidizes in air. The compound can combine with oxygen to form many different compounds to include binary oxides, peroxides, hydroxides, and oxides of higher order. Plutonium dioxide, PuO_2, is the most stable of the oxides found in the environment and is formed under most conditions, especially when plutonium metal is ignited in air (Burley 1990). PuO_2 has a high melting point (2240 °C), has a high chemical stability, and is highly

insoluble in water (Burley 1990). The behavior of plutonium in soils can vary depending on the local soil characteristics and the form the plutonium is in at the time of introduction. Four sites have been extensively monitored in the U.S. up to 30 years after introduction of the plutonium into the environment: the Nevada Test Site (NTS), Oak Ridge National Laboratory (ORNL) in Tennessee, Mound Laboratory in Ohio, and Rocky Flats in Colorado (Burley 1990). The source of plutonium is different for each site. At NTS, the plutonium is dispersed as an oxide as the result of safety research studies. At Oak Ridge, plutonium in a holdup pond was released when a dike broke. At Rocky Flats, cutting oil contaminated with metallic plutonium was released from leaking storage drums; while at the Mound Facility, a low-pH solution of plutonium leaked from a waste transfer line. For the soils studied at NTS and Rocky Flats, extraction of plutonium from soils was very low (10 – 15 %) as compared to 60 – 85 % extraction from the soils of the Mound Facility and ORNL (Burley 1990). Thus, if the plutonium was introduced into the environment as an oxide or metallic form, it exhibited low solubility; whereas if introduction was in the form of a soluble compound, much greater mobility was exhibited at a later time. Furthermore, autoradiographic comparisons of Rocky Flats and Mound Facility soils have indicated that the soils of the former exhibited discrete particles of plutonium, while the latter had a more homogeneous dispersion (Burley 1990). Homogeneous dispersion of plutonium requires the material to be available in an ionic form. For plutonium at the Mound Facility, the release was in the form of an acid solution, which allowed a more uniform dispersal of the plutonium into the soil matrix. Because of the nature of the release, the plutonium at the BOMARC site is expected to exhibit characteristics similar to that released at Rocky Flats and NTS.

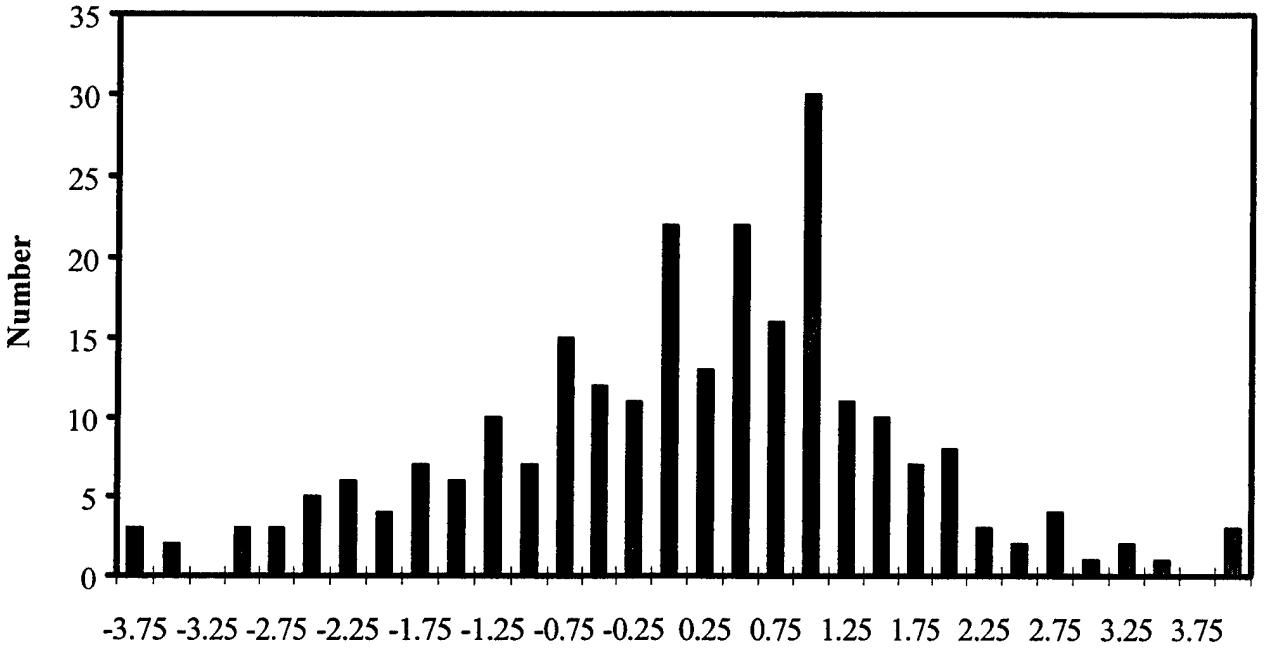
3. Studies of Plutonium at the BOMARC Site.

There has been a considerable number of environmental surveillance activities on the BOMARC site over the past 40 years. AF personnel accomplished a majority of the surveillance, but in the last 10 years most of the activities have been accomplished by organizations under contract to the AF. Three recent reports on surveillance activities note problems associated with evaluation of soil concentrations because of heterogeneity effects (HQ AMC 1992, Watts and Collins 1992, Kennedy 1990). The first two references did not provide data to support problems associated with heterogeneity. The last reference provided actual site data. The data was comprised of in-situ γ-measurements with FIDLER and analysis for ^{241}Am using laboratory γ-spectroscopy with a hyperpure germanium (HpGe) detection system. The purpose of the study was to determine a relationship between the FIDLER and ^{241}Am activity concentrations. A regression of the two parameters provided a squared correlation coefficient of 0.171. The author concluded, "it appears that there is not a linear relationship between the data set." Likely, the lack of correlation is due to inability of the sampling and analysis methods to control the effects of heterogeneity.

In 1997, OHM Remediations Services Corp. performed a characterization effort to support the Remedial Investigation/Feasibility Study (RI/FS). The purpose of the characterization effort was to determine the extent of the contamination zone. Soil samples were analyzed by both α- and γ-spectroscopy. Many of the 249 samples had analyses for ^{241}Am accomplished through both methods. Figure 1 contains a histogram of the ratio of the ^{241}Am activity concentration for γ- to α-spectroscopy analyses. Due to the wide range of ratio values, the plot is of the logarithm of the ratios. The ratios ranged from 0.00049 to 2629, spanning more than six orders of magnitude. The

mean, median, and percent coefficient of variation (% CV) of the data are: 15.8, 1.17, and 1100 %, respectively. The evaluation of this data set was not provided in the characterization report. Headquarters, Air Force Safety Center (HQ AFSC) provided additional evaluations of the characterization study data (Rademacher 1999a). In general, the data evaluations from the HQ AFSC report noted high variability and the influence of heterogeneity.

Figure 1. Logarithm of Activity Concentration Ratio (γ- to α-Spectroscopy) for ^{241}Am (OHM 1998).



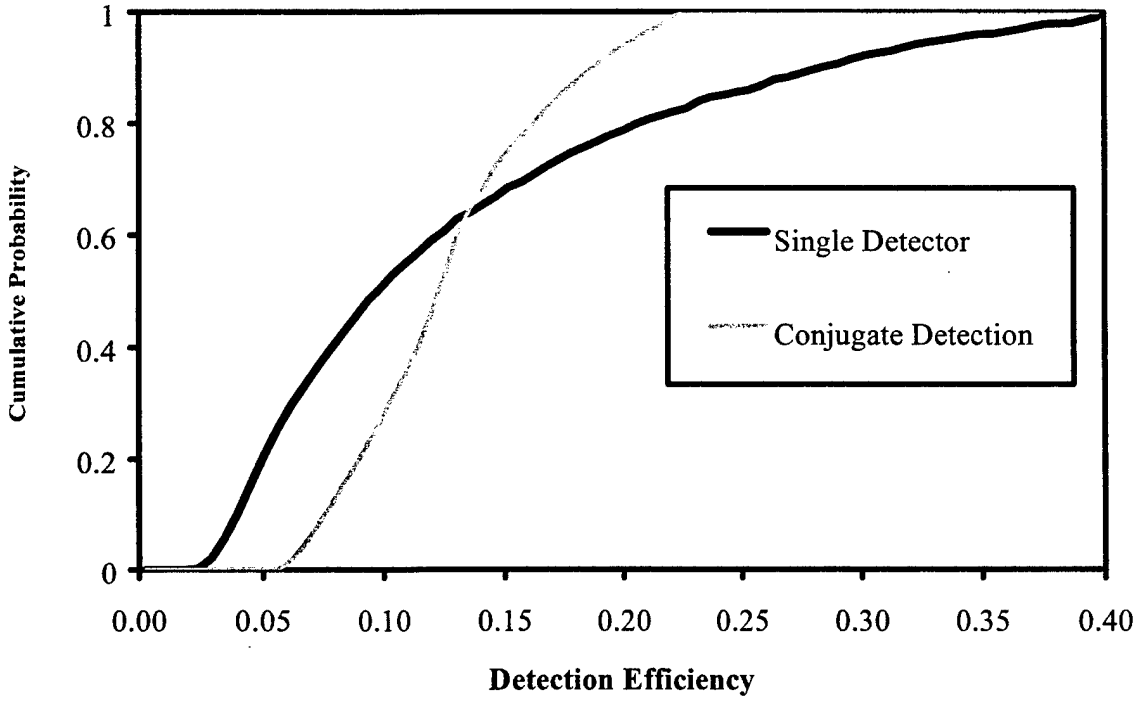
The high variability of the data in Figure 1 may be the result of many factors. First, HQ AFSC speculated that the α-spectroscopy evaluations were based on batch chemical recovery fractions in lieu of the preferred individual aliquot recovery method (Rademacher 2000a). The primary motivation for use of the former analytical method is reduced cost, because an ^{243}Am tracer is used in a fraction of the aliquots rather than all. Heterogeneity is the other factor that is believed to have a significant influence on the variability exhibited in the data of Figure 1. Due to the particulate nature of WGP, and the aliquot masses used for α-spectroscopy (~ 2 g) and γ-spectroscopy (~ 500 g), this factor is likely the most important.

In 2001, the Nuclear Engineering Department of Massachusetts Institute of Technology investigated the chemical nature of two BOMARC site samples (Plaue and Czerwinski 2001). In general, the soils were highly resistant to chemical attack, suggesting that the majority of the plutonium and americium contaminants were in oxide forms. For the BOMARC environment, it was predicted that

about 15 % of the contaminants are in soluble carbonate phase, indicating that the BOMARC plutonium has similar characteristics to that of NTS plutonium.

4. Theoretical Predictions of the Effects of Single Particle Heterogeneity on γ-Spectroscopy Analysis of WGP. HQ AFSC performed theoretical measurements to determine the effects of heterogeneity in WGP contamination in soils on γ-spectroscopy analyses (Rademacher 2000b). The analysis was performed for varied soil volumes, linear attenuation coefficients (μ), and sample containers. The calculations were made under the assumption that all of the WGP in a sample was contained in a single particle. This represents worst-case heterogeneity, while complete homogeneity represents the opposite. Figure 2 contains a plot of example detection efficiency calculations for a HpGe system and petri dish sample containers of similar dimension to those used in this study. The μ for this set of efficiency calculations was 0.38 cm^{-1}, while the referenced report contains calculations for additional values. The solid black line of the plot is the cumulative frequency distribution of detection efficiency for a single HpGe detection system. From the data, detection efficiency ranges from 0.025 to 0.4, had a mean of 0.13, and % CV of 76 %. The solid gray line of the plot is the cumulative frequency distribution of the mean detection efficiency for conjugate measurements (i.e., counted on both sides). For this distribution, mean detection efficiency for the paired calculations ranged from 0.06 to 0.23, with a distribution mean of 0.13, and a % CV of 30 %. Clearly, conjugate counting provides significant reduction in variability for measurement of soil samples with maximum heterogeneity. In this example, variability is reduced by almost four-fold.

Figure 2. Cumulative Frequency Distribution of Detection Efficiency for a Single Particle in a 126 cm^3 Volume Container (μ = 0.38 cm^{-1}).



5. Variability in α-Spectroscopy Analysis of Soils with WGP Contaminants.

a. General. While the primary purpose of this report is to better understand the effects of heterogeneity on γ-spectroscopy analysis of BOMARC soils, a summary of some observed effects on α-spectroscopy analyzed soils provides additional insight on the problems associated with heterogeneity. The analyses were of samples associated with investigation of WGP from NTS.

b. Nevada Applied Ecology Group (NAEG). From the NAEG, conflicting analytical data from investigations was observed on samples being analyzed for WGP contaminants (Bernhardt 1976). To further investigate the problem, one sample was split among three laboratories: Environmental Protection Agency (EPA), Reynolds Electrical and Engineering Company (REECo), and Lawrence Livermore Laboratory (LLL). Each laboratory further blended and homogenized the sub-sample, split the sub-samples into smaller aliquots of varying mass dependent on the laboratory, chemically dissolved the sample, and analyzed the plutonium extract by α-spectroscopy. The results of the comparison are in Table 3. From the data in the table, it is apparent that significant variability existed among the reported mean activity concentrations of the laboratories, with a range of 1.1 to 5.3 pCi g^{-1}. Also, the study demonstrated that within laboratory variability was high and was generally inversely related to aliquot mass. For the EPA data, the percent coefficient of variation (% CV) was 130 %. Even for the LLL data, where aliquot masses were high, relative percent difference (RPD) values were high among some paired analyses.

Table 3. Comparative Analysis of $^{239/240}$Pu in Soil from NTS (Adapted from Bernhardt 1976).

Lab	Aliquot Size (gram)	Number of Aliquots	$^{239/240}$Pu (pCi g^{-1})		% CV	$^{239/240}$Pu Range (pCi g^{-1})
			mean	σ		
EPA	1	14	1.1	1.4	130	0.23 – 5.3
REECo	10	10	2.3	1.5	66	0.66 – 5.2
LLL	25	2	3.3	NA	83*	1.9 – 4.6
	25	2	3.4	NA	6*	3.3 – 3.5
	100	2	4.8	NA	29*	4.1 – 5.5
	100	2	5.3	NA	48*	4.0 – 6.5

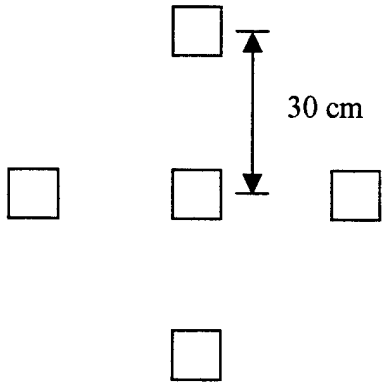
NA = Not Applicable * Relative Percent Difference

Methodology

1. Sample Collection. With a FIDLER instrument, the survey team scanned areas of the BOMARC site that did not contain concrete or asphalt overburdens. Areas with elevated instrument response were selected for sampling. Seven locations had five surface samples collected for a single FIDLER instrument measurement according to the spatial pattern of Figure 3. The surface samples were collected from the surface to a depth of 5 cm (2 in) with a stainless steel trowel. Between samples, the trowel was cleaned with distilled water. Samples were placed in plastic bags, with notation of sample number and depth. Sample chain-of-custody forms were prepared for the sample set. Prior to packaging for shipment, the samples were double-bagged. For the center locations in the sampling pattern, a sample was collected at a depth of 5 – 10 cm. For one center sample, a third sample at depth (10 – 15 cm) was collected. Twenty-three locations had only a surface soil sample collected in the center sub-sampling location. Ten locations had only a surface (0 – 5 cm) and a sample at depth (5 – 10 cm) that was collected in the center sub-sampling location. These 10 locations were sampled one year later than the other locations, with the intent of filling a data gap in low activity samples.

2. FIDLER Measurements. The FIDLER device used for the measurements had an energy window set to correspond to the 60 keV γ-ray from ^{241}Am. The detector was held in a stand, with a detector to ground surface separation distance of 30 cm. One-minute measurements were recorded.

Figure 3. Soil Sampling Pattern.



3. Sample Preparation. At the laboratory, the samples had mass determinations and filtration through a course mesh sieve to remove rocks. The remaining fraction was ashed in an oven at 500 °C for 24 h. The samples were blended and homogenized in a soil tumbler. The soil was incrementally placed into plastic petri dishes (9.5 cm in diameter x 2.54 cm in height) until completely filled, exhausting all of the soil. Due to variability in the mass of samples collected and rock fraction, aliquot number varied from 2 to 10 among each sample. Net sample mass was determined for each aliquot, with the typical being about 200 g. The petri dish lids were permanently sealed to the bases with a silicone sealant. The sample container exterior was cleaned and placed in a clean Zip-Lock™ container to prevent contamination of laboratory surfaces.

4. Analysis. Aliquot ^{241}Am activity concentration was assessed by laboratory γ-spectroscopy analysis on a 8.9 cm diameter HpGe detector encased in a 7.5 cm thick steel cave. The aliquot to detector face separation distance was about 4 mm. Each aliquot was assessed twice, with the sample aliquot containers being flipped between measurements. Measurement periods were set at 10,000 s (2.8 h). Absolute activity concentration was based on comparison to a prepared soil standard that was calibrated against a National Institute of Standards and Technology (NIST) traceable calibration source.

5. Data Evaluation. Estimated aliquot activity and standard counting errors were recorded for each measurement. For the paired petri dish measurements, the ratio and mean were calculated. Estimated aliquot activity concentrations were based on the mean. In the same way, the overall sample standard deviation and % CV were calculated. For locations with samples collected at multiple depths, ratios of the respective mean sample activity concentrations were calculated.

6. University of Pittsburgh Work. Capt Craig Refosco, an Air Force active duty health physicist, performed additional analysis of select samples in partial fulfillment of the requirements for the M.S. in Science Degree (Refosco 2001). Five samples were used for the analysis. Smaller aliquots were prepared by subdivision of 200 g aliquots into ten 20 g aliquots. The samples were placed in the lids of same type of petri dish that was used by AFIERA, enclosed by the petri dish bottom portion, and sealed with silicone. The aliquots were counted on a Bicron (12.7 cm diameter x 1.6 mm thick) NaI(Tl) and calibrated with an AFIERA prepared NIST traceable source.

Results

1. Sample Activity Concentrations. Table A in Appendix A contains the analytical results from the laboratory γ-spectroscopy. The reported $^{239/240}$Pu activity concentration for the individual aliquots is based on assessment of the ^{241}Am 60 keV γ-ray and an assumed $^{239/240}$Pu to ^{241}Am ratio of 5.4 (Rademacher 1999a). Uncertainty values are based on randomness due to counting statistics and are expressed at the 95 % confidence interval (CI). Aliquot $^{239/240}$Pu activity concentrations, estimated from individual aliquot measurements (one side alone), ranged from 1.1 to 68,500 pCi g^{-1}. For the mean of paired aliquot measurements, $^{239/240}$Pu activity concentrations ranged from 1.1 to 51,700 pCi g^{-1}.

2. Heterogeneity.

a. General. This study provides a description of heterogeneity at four levels. First, for seven sampling locations, surface soil samples within 30 cm of the central location describe variability within sampling regions. Second, the evaluation of heterogeneity at the sample level is described through multiple aliquot analysis of samples. Third, conjugate measurement of individual aliquots provides examination of heterogeneity at the aliquot level. Lastly, work performed at the University of Pittsburgh, describes heterogeneity at the sub-aliquot level of 20 g.

b. Heterogeneity at Sampling Level. Table 4 contains $^{239/240}$Pu activity concentrations for the surface soils of sample locations 1 through 7. For each location, results are provided for the center, west, east, north, and south sub-sample locations. The last column lists the variability of the measurements expressed in % CV. The variability among the locations ranges from 32 to 186 %. Sampling location 1 had the greatest level of variability, with the most extreme range of activity concentration being 3.2 to 2,792 pCi g^{-1}. The other three samples collected at this location had activity concentration clustered around 140 pCi g^{-1}. Sampling location 7 had the highest mean activity concentration among the sub-samples, being 9,647 pCi g^{-1}. For this sampling location, the ratio between the highest and the lowest activity concentration among sub-samples was about 4, while that of sample location 1 was almost 1,000.

Table 4. Sub-Sample $^{239/240}$Pu Activity Concentrations.

Sample Location	Pu-239/240 Activity Concentration (pCi g^{-1})						Variability (% CV)
	Center	W	E	N	S	Mean	
1	2,792.2	165.4	127.3	3.2	138.6	645	186
2	594.1	268.8	184.2	354.9	553.5	391	46
3	232.8	180.9	251.1	98.6	173.5	187	32
4	1,004.8	1,227.3	437.9	774.6	1,049.6	899	34
5	577.5	278.8	572.3	543.6	199.8	434	42
6	2,207.1	75.9	885.3	2,560.7	4,681.5	2,082	85
7	5,273.2	12,888.2	19,889.6	3,866.5	6,319.3	9,647	69

c. **Heterogeneity at Sample and Aliquot Level.**

Figure 4 contains a histogram of the individual aliquot data from Table A-1 for sample 14 (abbreviated notation based on the last digits of the base sample number). Apparent is the vast discrepancy among individual aliquot activity concentrations. The mean aliquot activity concentrations are: 544, 7,742, and 90 pCi g^{-1}, respectively for increasing aliquot number. The ratio between mean activity concentration of aliquots 2 and 3 is 86. With the extensive blending and homogenizing procedures applied to the samples during preparation, heterogeneity of this magnitude was not anticipated. While the composition of an individual aliquot cannot be inferred from the data presented here, it is clear that in the case of aliquot 2, that the majority of sample activity is concentrated in a single or possibly a few particles. If the activity is primarily concentrated in a single particle, the minimum particle diameter is 162 μm, under the assumption that the plutonium is in a dioxide form (ρ = 9.8 g cm^{-3}). Regardless of the particle(s) size(s) and composition, the ratio of the estimated activity concentration from the down to up measurements is 4.3, and this discrepancy may introduce significant errors if the sample was measured in one configuration only.

Figure 4. Sample 14 $^{239/240}$Pu Activity Concentration for Individual Aliquots (Mean = 2792 pCi g^{-1}, % CV = 176 %).

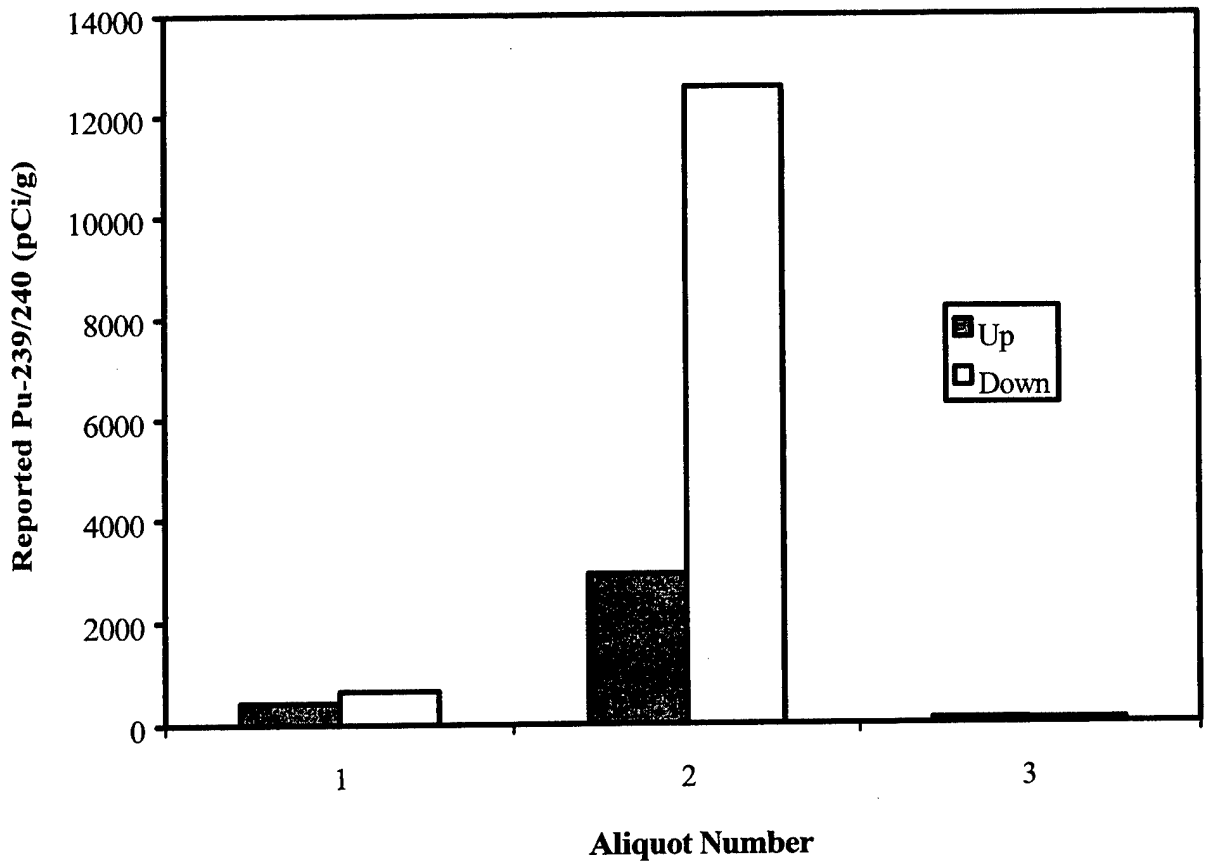


Figure B-1 of Appendix B contains a histogram of the individual aliquot data from Table A for sample 67. Apparent from the histogram, aliquots 1, 3, and 5 have relatively consistent activity concentration, while 2 and 4 are considerably higher. The mean aliquot activity concentrations are: 92, 607, 101, 500, and 55 pCi g^{-1}, respectively for increasing aliquot number. For aliquots 2 and 4, the ratio of down to up is 1.7 and 1.4, respectively. Because of the high sample activity concentration of these aliquots, the high conjugate counting ratios are indicative of heterogeneity effects rather than uncertainties of counting statistics. For these aliquots, if the activity is largely associated by single particles, the minimum particle diameters would be 69 and 65 μm, respectively (dioxide form of plutonium). For aliquots 1, 3, and 5, the plutonium activity appears to be relatively homogenous in distribution, based on agreement between respective conjugate measurements.

Figure B-2 contains a histogram of the individual aliquot data from Table A for sample 15. Like sample 14, one aliquot (2) contains significantly higher activity concentration than the others. The disparity between the up and down measured activity concentration, 4.6, is indicative of a single particle dominating the aliquot activity. Figure B-3 contains a histogram of the individual aliquot data for sample 53. Like samples 14 and 15, this sample has one aliquot that contains a significantly higher mean activity concentration than the others. Sample 53 is unique to this data set in having one of the highest individual aliquot mean activity concentrations, 33,200 pCi g^{-1}. Like many of the other high activity concentration aliquots, the ratio of the conjugate measurements is high with a value of 2.6. For this aliquot, if the activity concentration is concentrated in an individual particle, the minimum particle diameter is 260 μm (dioxide form of plutonium).

The samples discussed above provide a unique description of heterogeneity of WGP and represent the greatest degree of heterogeneity among those evaluated in this study. Many of the samples had less apparent heterogeneity. Figure B-4 contains a histogram of the individual aliquot data for sample 75. From the plot, it is apparent that close agreement exists among the conjugate aliquot measurements and reasonably well among all measurements. The % CV for the individual measurements was 72 %. Figure B-5 contains a histogram of the individual aliquot data for sample 60. This sample has a mean activity concentration of 128 pC g^{-1} and a % CV of only 13 %. The variability among aliquots and conjugate measurements is low, suggesting a homogenous sample.

Figure 5 contains a scatterplot of % CV of individual aliquot measured $^{239/240}$Pu activity concentration versus the mean sample activity concentration. The plot describes the heterogeneity that exists among individual aliquots of a sample. Samples with the highest variability among the measurements are those with the highest mean activity concentration. For samples with mean activity concentration between 100 and 1,000 pCi g^{-1}, the % CV values range from very low values to about 160 %. Thirteen samples have mean activity concentrations between 10 - 100 pCi g^{-1}, and have % CV values ranging from 18 to 128 %. Seven samples have mean activity concentrations less than 10 pCi g^{-1}, and have % CV values ranging from 25 to 137 %. The samples in this range of activity concentration have particular significance to the BOMARC site because the remediation criterion for the site is 8 pCi g^{-1}.

Figure 6 contains a scatterplot of the ratio of the conjugate measurements of $^{239/240}$Pu activity concentrations for individual aliquots (reciprocal plotted for ratios less than 1). The plot describes the heterogeneity that exists within individual aliquots. In general, samples with the highest ratios are those with the highest mean $^{239/240}$Pu activity concentration. The ratios ranged from 1.0 to 4.6,

Figure 5. Percent Coefficient of Variation (% CV) of Individual Aliquot Measured $^{239/240}$Pu vs. Mean Sample $^{239/240}$Pu.

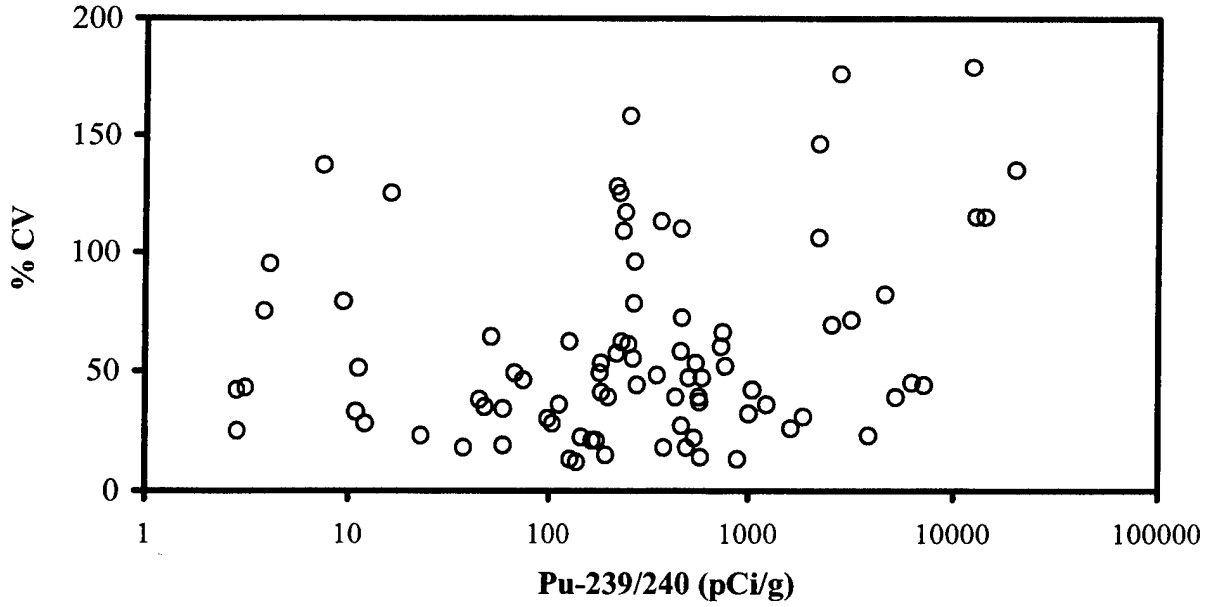
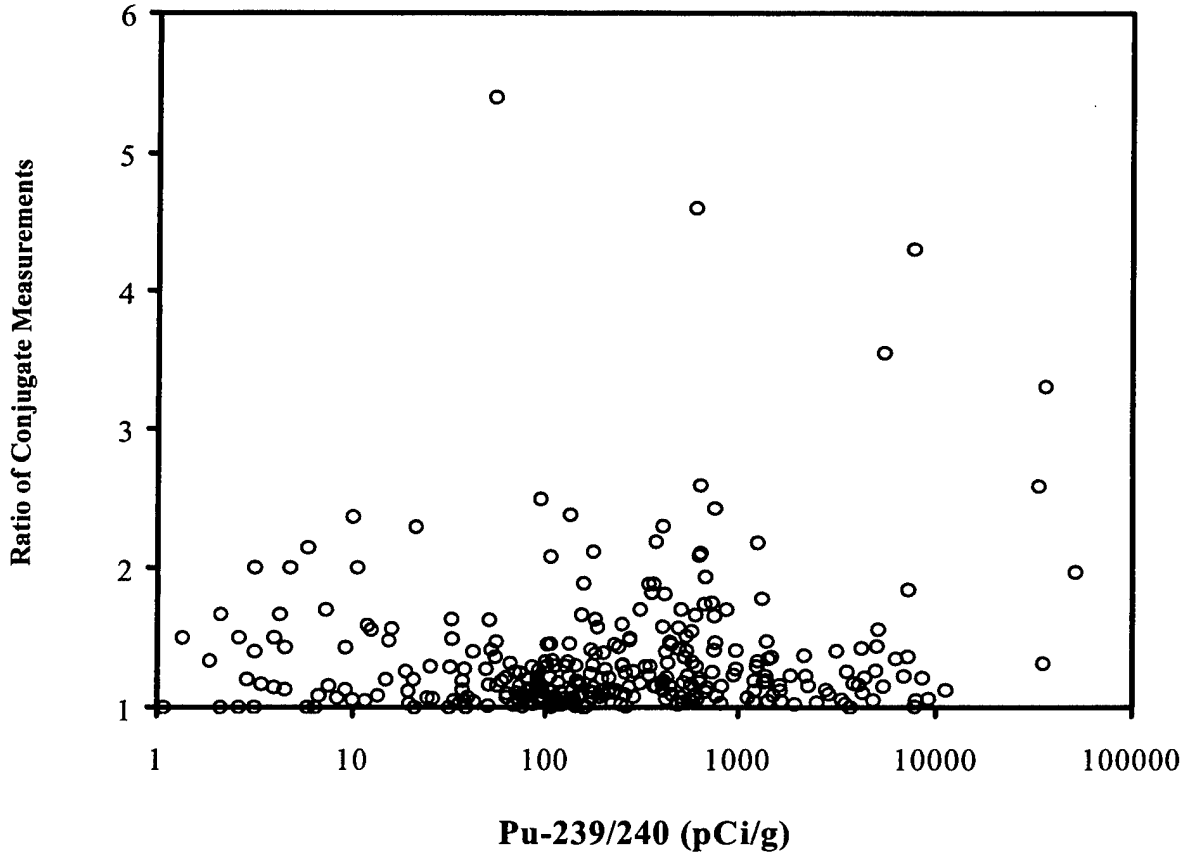


Figure 6. Ratio of Conjugate Measurements of $^{239/240}$Pu for Individual Aliquots.



with the majority being below 3. In general, higher ratios are indicative of a greater degree of heterogeneity, indicating that the highest heterogeneity exists among samples of the highest activity concentration. Many of the samples have ratios near one. It could be argued in general that these samples have a low degree of heterogeneity. On an individual sample basis, however, conclusions cannot be made, due to the potential for single or multiple particle samples having the particle(s) on the mid-line of the sample. The effect of heterogeneity was exhibited for samples across entire range of activity concentration.

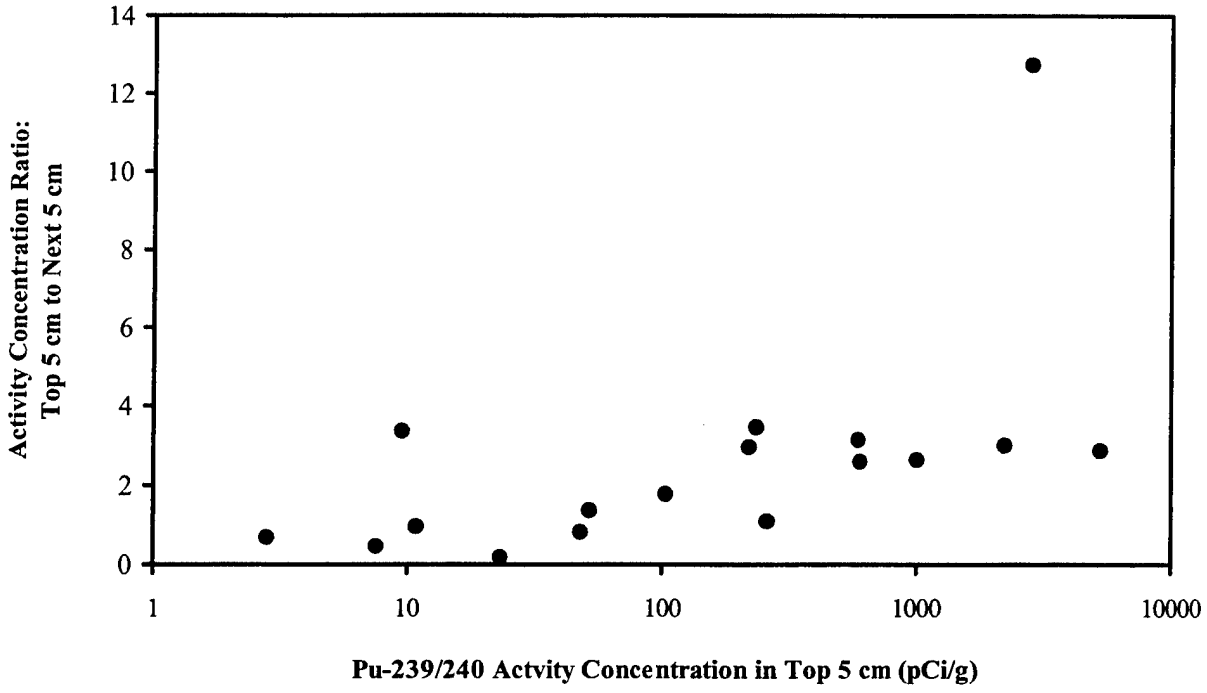
3. University of Pittsburgh Sub-Aliquot Data.

Samples: 50, 51, 52, 57, and 79 had 10 sub-aliquots analyzed from the first aliquot. The results of the analysis are listed in Table B-3 with the results from the AFIERA analysis. For each set of sub-aliquots, the mean, % CV, and relative percent difference (RPD) between the mean and AFIERA result. For four of the sub-aliquot groups, the degree of heterogeneity observed among the sub-aliquots was low, with the maximum to minimum sub-aliquot ratios ranging from 2.9 to 4.3 and range of % CV values ranging from 32 to 50. For these samples, it was initially assumed that the contaminant was uniform with the ratio of the AFIERA conjugate measurements having ratios less than 1.17. Further, for samples 50, 51, and 52, the entire samples overall had a low degree of heterogeneity, with combined aliquot % CV values of 39, 31, and 14. For sample 57, the combined aliquot % CV was 71, but the greatest degree of heterogeneity within this sample was expected in aliquot 2, with a conjugate measurement ratio of 1.36, as compared to 1.15 in aliquot 1 (the aliquot that was sub-divided).

The aliquot from sample 79 exhibited the greatest degree of heterogeneity among the five samples with a % CV among the sub-aliquots at 306 %. The first sub-aliquot contained over 97 % of the total aliquot activity and had an activity concentration 800 times the lowest activity sub-aliquot. If all the activity for this sub-aliquot is comprised of a single particle, the minimum diameter is 250 μm. Another interesting observation with this aliquot is the existence of a sub-aliquot with an intermediate activity concentration: 12.7 times that of the lowest, but 63 times lower than the highest. For the remaining sub-aliquots, the activity concentration was fairly uniform.

4. Depth Distribution. Seventeen locations had samples collected at the 5 – 10 cm depth. For one location, another sample was collected at a depth of 10 – 15 cm. The ratio of the $^{239/240}$Pu activity concentration in the top 5 cm to the next 5 cm is plotted in Figure 7. The ratios range from 0.2 to 12.7. While the ratio among paired samples has a significant degree of variability, among the higher activity concentration samples, better agreement existed, with the median being about 3. For all paired samples, the median ratio is 2.6. The sample with the highest ratio is comprised of samples 14 and 15 (14 being the top 5 cm). Sample 14 (see Figure 4) has a % CV of 176 % among its aliquots. This value is second highest among the sample evaluated. As well, this sample was in the group of samples that exhibited a high degree of spatial heterogeneity (see Table 4, sampling location #1). The unusually high heterogeneity exhibited in this sample may in part explain non-agreement with the depth distribution of the other high activity concentration samples.

Figure 7. Activity Concentration Ratio: Top 5 cm to Next 5 cm vs. $^{239/240}$Pu Activity Concentration in Top 5 cm.



5. FIDLER Correlation Coefficients.

The correlation between concentration of WGP in soils and the response of portable γ-radiation instruments is important in assessing instrument minimum detectable concentrations (MDCs). The FIDLER instrument is the most common for in-situ γ- radiation measurements of WGP contaminated soils. Figure 8 contains a scatterplot of FIDLER response versus the mean $^{239/240}$Pu activity concentration in the top 5 cm of soil of those from the first sampling effort. Apparent from the plot is a high degree of variability in the data set, based on the low squared correlation coefficient (R^2) of 0.35. The intercept of the linear regression was set at 1,000 cpm, the background count rate of the FIDLER in an area of the site where WGP is known not to have been deposited.

The regression analysis is affected to a significant degree by the two highest activity concentration data points. Another regression of the data is provided in Figure 9, with the two highest activity concentration data points truncated from the plot. Visually, the data are in better agreement with the regression line and is reflected in a significantly higher R^2 of 0.84. The slope of the regression analysis was increased by a factor of 2.3 over that of the complete data set. Table 5 provides a list of estimated correlation coefficients of FIDLER response and $^{239/240}$Pu activity concentration for the BOMARC site, based on an assumed activity concentration ratio between 5 cm successive lifts of 2.6.

Figure 8. Scatterplot of FIDLER Response vs. $^{239/240}$Pu Activity Concentration (Full Data Set, n = 30)

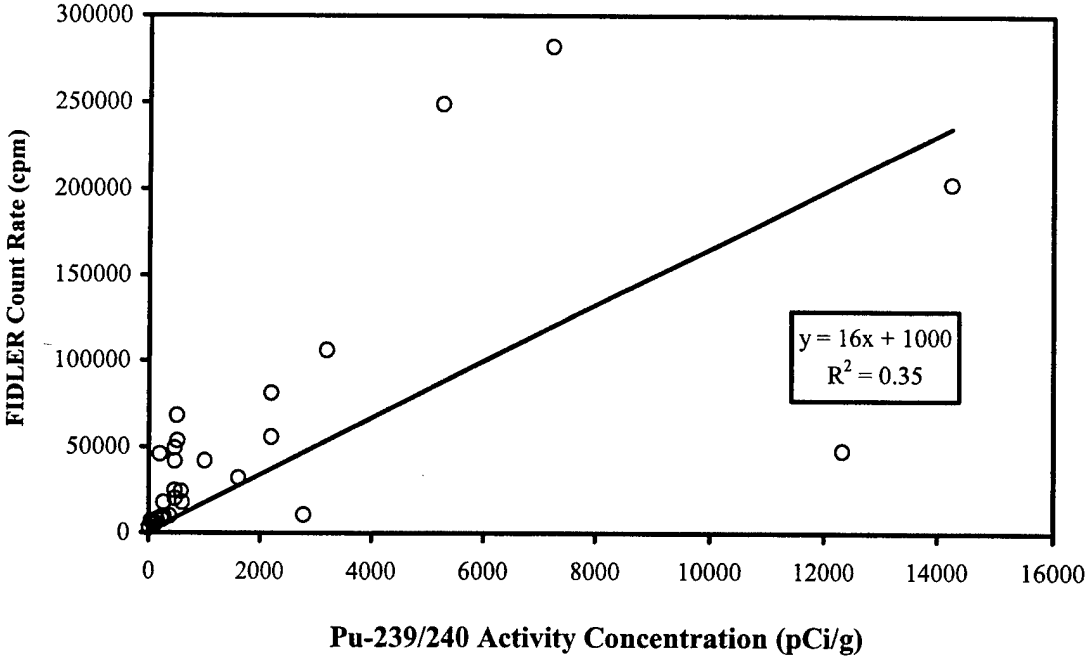


Figure 9. Scatterplot of FIDLER Response vs. $^{239/240}$Pu Activity Concentration (Truncated Data Set, n = 28)

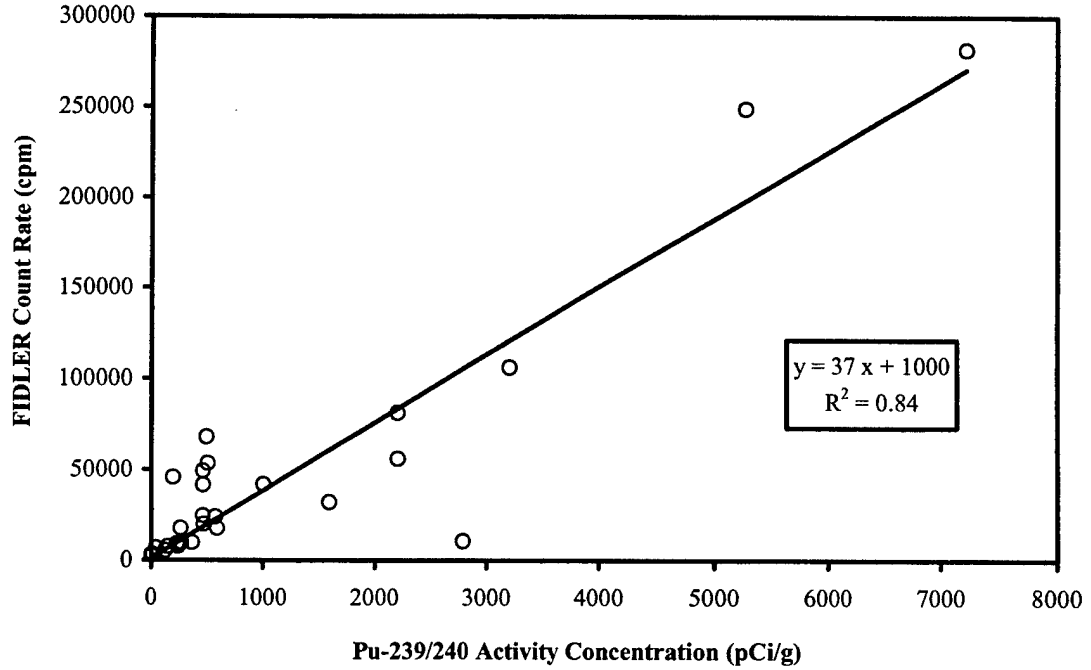


Table 5. Estimated Correlation Coefficients of FIDLER Response and $^{239/240}$Pu Activity Concentrations.

Averaging Depth* (cm)	Correlation Coefficient (cpm g pCi^{-1})
5	37
10	53
15	72

* Assummed ratio of $^{239/240}$Pu between successive 5 cm lifts is 2.6.

Discussion and Conclusions

Heterogeneity impacts laboratory evaluation of soil samples when γ-radiation evaluation of the 60 keV photon from ^{241}Am is used with an assumed constant ratio between ^{241}Am and $^{239/240}$Pu. Heterogeneity was observed at every level of evaluation: sub-aliquot, aliquot, sample, and sampling location. The conjugate counting methodology used aided in assessing the degree of heterogeneity. For many aliquots evaluated, the conjugate counting methodology improved measurement accuracy. For WGP samples, the conjugate counting methodology should be implemented unless homogeneity can be verified.

Due to the heterogeneity among aliquots from the same sample, the conjugate method should be modified to accommodate larger samples. This requirement necessitates larger diameter detectors. HpGe detectors are not commonly manufactured with diameters in excess of 10 cm. NaI(Tl) detectors are a reasonable alternative. AFIERA has purchased a pair of 20 cm diameter x 2 mm thick NaI(Tl) detectors. Over 2,000 g of soil can be held in sample containers of the same diameter and 4.5 cm in height.

Heterogeneity impacts the accuracy of the estimated correlation coefficient between the FIDLER response and $^{239/240}$Pu activity concentrations. Clear from the evaluation provided here, careful review of the data and accurate soil measurement techniques should be used to estimate this parameter. Evaluation of vertical distribution of the contaminant is critical to in-situ γ-measurements if they are being used to meet remediation or screening criteria. For the evaluation provided here, there was good agreement in 10 of the 17 soil samples evaluated for vertical distribution of the contaminant.

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Mr. William Walker – Laboratory Sample Analysis

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Appendix A

Laboratory $^{239/240}$Pu Analysis Results

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Table A-1. Individual Aliquot $^{239/240}$Pu Activity Concentrations.
(1999 Sampling)

Base Sample Number	Sampling Location		Aliquot #1					Aliquot #2				
			Pu-239/240 Activity Concentration (pCi/g)					Pu-239/240 Activity Concentration (pCi/g)				
	#	Sub-sample	Up		Down		HIGH / LOW	Up		Down		HIGH / LOW
			Value	Uncer.	Value	Uncer.		Value	Uncer.	Value	Uncer.	
GS0000014	1	0-2 C	655	152	434	101	1.51	12,570	2,920	2920	678	4.31
GS0000015		2-4 C	226	52	203	47	1.11	217	50	1,000	232	4.60
GS0000016		0-2 W	146	34	230	54	1.57	160	37	179	42	1.12
GS0000017		0-2 E	73	17	80	18	1.10	115	27	242	56	2.11
GS0000018		0-2 N	3.8	1.1	2.7	0.5	1.40	2.2	0.5	1.6	0.5	1.33
GS0000019		0-2 S	138	32	162	38	1.17	123	29	131	30	1.07
GS0000020	2	0-2 C	629	146	884	205	1.41	535	124	935	217	1.75
GS0000021		2-4 C	126	29	87	20	1.45	1,080	250	444	103	2.42
GS0000022		0-2 W	192	45	167	39	1.15	381	89	648	150	1.70
GS0000023		0-2 E	312	72	196	45	1.59	164	38	165	38	1.01
GS0000024		0-2 N	617	143	453	105	1.36	233	54	395	92	1.70
GS0000025		0-2 S	945	219	571	133	1.65	243	56	456	106	1.88
GS0000026	3	0-2 C	90	21	95	22	1.05	248	58	221	51	1.12
GS0000027		2-4 C	46	11	63	15	1.36	69	16	144	34	2.08
GS0000028		0-2 W	115	27	103	24	1.12	220	51	286	66	1.30
GS0000029		0-2 E	136	32	145	34	1.07	257	59	467	109	1.82
GS0000030		0-2 N	99	23	83	19	1.19	113	27	150	35	1.33
GS0000031		0-2 S	169	39	235	55	1.39	170	39	189	38	1.11
GS0000032	4	0-2 C	1,080	252	1,400	325	1.29	911	212	623	145	1.46
GS0000033		2-4 C	500	116	379	88	1.32	315	73	387	90	1.23
GS0000034		0-2 W	1,250	290	1,700	395	1.36	1,570	365	1,450	336	1.09
GS0000035		0-2 E	536	125	366	845	1.47	581	135	633	1467	1.09
GS0000036		0-2 N	1,430	333	1,080	251	1.33	578	135	698	162	1.21
GS0000037		0-2 S	748	174	807	187	1.08	1,690	393	952	221	1.78

Table A-1. Individual Aliquot $^{239/240}$Pu Activity Concentrations (Continued)
(1999 Sampling)

Base Sample Number	Sampling Location		Aliquot #3					Aliquot #4				
			Pu-239/240 Activity Concentration (pCi/g)					Pu-239/240 Activity Concentration (pCi/g)				
	#	Sub-sample	Up		Down		HIGH / LOW	Up		Down		HIGH / LOW
			Value	Uncer.	Value	Uncer.		Value	Uncer.	Value	Uncer.	
GS0000014	1	0-2 C	84	19	96	22	1.13					
GS0000015		2-4 C	82	19	100	23	1.22	82	19	65	15	1.25
GS0000016		0-2 W	143	33	136	32	1.05					
GS0000017		0-2 E										
GS0000018		0-2 N	5.4	1.1	3.2	0.5	1.67					
GS0000019		0-2 S										
GS0000020	2	0-2 C	258	60	324	75	1.26					
GS0000021		2-4 C	102	24	92	22	1.11	86	20	101	23	1.17
GS0000022		0-2 W	115	27	111	26	1.04					
GS0000023		0-2 E	190	44	80	1.1	2.38					
GS0000024		0-2 N	232	54	182	42	1.27					
GS0000025		0-2 S										
GS0000026	3	0-2 C	485	113	258	60	1.88					
GS0000027		2-4 C	55	13	47	11	1.16	36	8.1	37	8.6	1.03
GS0000028		0-2 W										
GS0000029		0-2 E										
GS0000030		0-2 N	68	16	78	18	1.15					
GS0000031		0-2 S	131	31	147	35	1.12					
GS0000032	4	0-2 C										
GS0000033		2-4 C	309	72	399	93	1.29					
GS0000034		0-2 W	653	152	744	173	1.14					
GS0000035		0-2 E	222	52	289	68	1.30					
GS0000036		0-2 N	359	83	501	116	1.40					
GS0000037		0-2 S										

Table A-1. Individual Aliquot $^{239/240}$Pu Activity Concentrations (Continued)
(1999 Sampling)

Base Sample Number	Sampling Location		Aliquot #5					Aliquot #6				
			Pu-239/240 Activity Concentration (pCi/g)					Pu-239/240 Activity Concentration (pCi/g)				
	#	Sub-sample	Up		Down		HIGH / LOW	Up		Down		HIGH / LOW
			Value	Uncer.	Value	Uncer.		Value	Uncer.	Value	Uncer.	
GS0000014	1	0-2 C										
GS0000015		2-4 C	106	25	112	26	1.06					
GS0000016		0-2 W										
GS0000017		0-2 E										
GS0000018		0-2 N										
GS0000019		0-2 S										
GS0000020	2	0-2 C										
GS0000021		2-4 C	140	32	163	38	1.16	207	48	110	25	1.89
GS0000022		0-2 W										
GS0000023		0-2 E										
GS0000024		0-2 N										
GS0000025		0-2 S										
GS0000026	3	0-2 C										
GS0000027		2-4 C	84	19	97	23	1.15					
GS0000028		0-2 W										
GS0000029		0-2 E										
GS0000030		0-2 N										
GS0000031		0-2 S										
GS0000032	4	0-2 C										
GS0000033		2-4 C										
GS0000034		0-2 W										
GS0000035		0-2 E										
GS0000036		0-2 N										
GS0000037		0-2 S										

Table A-1. Individual Aliquot $^{239/240}$Pu Activity Concentrations (Continued)
(1999 Sampling)

Base Sample Number	Sampling Location		Combined Aliquot Statistical Parameters	
	#	Sub-sample	Mean Activity Concentration (pCi/g)	Percent Coefficient Of Variation
GS0000014	1	0-2 C	2,790	176
GS0000015		2-4 C	219	128
GS0000016		0-2 W	165	21
GS0000017		0-2 E	127	62
GS0000018		0-2 N	3.2	43
GS0000019		0-2 S	139	12
GS0000020	2	0-2 C	594	47
GS0000021		2-4 C	228	125
GS0000022		0-2 W	269	78
GS0000023		0-2 E	184	41
GS0000024		0-2 N	352	48
GS0000025		0-2 S	554	53
GS0000026	3	0-2 C	233	62
GS0000027		2-4 C	68	49
GS0000028		0-2 W	181	49
GS0000029		0-2 E	251	61
GS0000030		0-2 N	99	30
GS0000031		0-2 S	174	21
GS0000032	4	0-2 C	1,000	32
GS0000033		2-4 C	381	18
GS0000034		0-2 W	1,230	36
GS0000035		0-2 E	438	39
GS0000036		0-2 N	775	52
GS0000037		0-2 S	1,050	42

Table A-1. Individual Aliquot $^{239/240}$Pu Activity Concentrations (Continued)
(1999 Sampling)

Base Sample Number	Sampling Location		Aliquot #1					Aliquot #2				
			Pu-239/240 Activity Concentration (pCi/g)					Pu-239/240 Activity Concentration (pCi/g)				
	#	Sub-sample	Up		Down		HIGH LOW	Up		Down		HIGH LOW
			Value	Uncer.	Value	Uncer.		Value	Uncer.	Value	Uncer.	
GS0000038	5	0-2 C	659	153	825	192	1.25	321	75	505	117	1.58
GS0000039		2-4 C	234	55	293	68	1.25	84	19	122	28	1.45
GS0000040		0-2 W	274	64	251	58	1.09	239	56	523	122	2.19
GS0000041		0-2 E	609	142	388	90	1.57	416	97	875	203	2.10
GS0000042		0-2 N	481	112	417	97	1.15	586	136	690	160	1.18
GS0000043		0-2 S	268	62	266	62	1.01	135	31	130	30	1.04
GS0000044	6	0-2 C	2,420	562	8,590	1,990	3.55	492	114	5023	117	1.02
GS0000045		2-4 C	393	91	476	111	1.21	1,630	379	1,210	281	1.35
GS0000046		0-2 W	1,720	399	789	183	2.18	393	91	441	103	1.12
GS0000047		0-2 E	849	197	1,040	243	1.23	881	205	766	178	1.15
GS0000048		0-2 N	1,260	293	1,500	349	1.19	2,480	576	2,570	596	1.03
GS0000049		0-2 S	1,240	288	1,520	353	1.23	8,180	1,900	7,790	1,810	1.05
GS0000050	7	0-2 C	4,800	1,110	3,960	920	1.21	3,980	923	3,170	736	1.25
GS0000051		2-4 C	2,640	612	2,960	687	1.12	1,970	457	1,930	447	1.02
GS0000052		4-6 C	534	124	623	145	1.17	648	149	720	167	1.11
GS0000053		0-2 W	5,370	1,250	7,240	1,680	1.35	6,240	1,450	7,630	1,770	1.22
GS0000054		0-2 E	4,500	1,040	4,080	946	1.10	34,800	8,090	68,470	15,900	1.96
GS0000055		0-2 N	4,160	966	3,560	826	1.17	2,760	642	3,020	702	1.09
GS0000056		0-2 S	7,780	1,810	9,400	2,180	1.21	3,280	760	3,460	803	1.06
GS0000057	8	0-2	2,440	567	2,120	493	1.15	8,360	1,940	6,150	1,430	1.36
GS0000058	9	0-2	8.6	2.2	9.7	2.2	1.13	11	2.7	12	2.7	1.05
GS0000059	10	0-2	3.8	1.1	2.7	0.5	1.40	2.2	0.5	1.6	0.5	1.33
GS0000060	11	0-2	133	31	157	37	1.18	126	29	114	27	1.11
GS0000061	12	0-2	2.2	0.5	1.6	0.5	1.33	3.8	1.1	8.1	1.6	2.14

Table A-1. Individual Aliquot $^{239/240}$Pu Activity Concentrations (Continued)
(1999 Sampling)

Base Sample Number	Sampling Location		Aliquot #3					Aliquot #4				
			Pu-239/240 Activity Concentration (pCi/g)					Pu-239/240 Activity Concentration (pCi/g)				
	#	Sub-sample	Up		Down		HIGH / LOW	Up		Down		HIGH / LOW
			Value	Uncer.	Value	Uncer.		Value	Uncer.	Value	Uncer.	
GS0000038	5	0-2 C										
GS0000039		2-4 C										
GS0000040		0-2 W	200	46	186	43	1.07					
GS0000041		0-2 E										
GS0000042		0-2 N										
GS0000043		0-2 S										
GS0000044	6	0-2 C	604	140	639	148	1.06					
GS0000045		2-4 C	551	128	582	135	1.05	535	124	515	110	1.04
GS0000046		0-2 W	504	117	666	155	1.32					
GS0000047		0-2 E										
GS0000048		0-2 N	6,240	1,453	4,010	932	1.56	1,310	305	1,100	256	1.19
GS0000049		0-2 S										
GS0000050	7	0-2 C	7,880	1,830	7,850	1,820	1.00					
GS0000051		2-4 C	1,230	285	1,180	274	1.04	2,510	583	1,830	426	1.37
GS0000052		4-6 C	496	115	532	124	1.07	692	161	536	124	1.29
GS0000053		0-2 W	47,900	11,100	18,600	4,320	2.58	5,970	1,390	4,160	967	1.43
GS0000054		0-2 E	3,720	864	3,720	865	1.00					
GS0000055		0-2 N	4,730	1,200	4,970	537	1.05					
GS0000056		0-2 S	9,480	2,200	8,960	2,080	1.06	4,400	1,020	3,790	881	1.16
GS0000057	8	0-2	3,680	854	2,630	610	1.40	1,550	359	1,730	402	1.12
GS0000058	9	0-2	12.4	3.2	18.4	4.3	1.48					
GS0000059	10	0-2	5.4	1.1	3.2	0.5	1.67					
GS0000060	11	0-2	120	25	146	34	1.22	109	25	117	27	1.08
GS0000061	12	0-2										

Table A-1. Individual Aliquot $^{239/240}$Pu Activity Concentrations (Continued)
(1999 Sampling)

Base Sample Number	Sampling Location		Aliquot #5					Aliquot #6				
			Pu-239/240 Activity Concentration (pCi/g)					Pu-239/240 Activity Concentration (pCi/g)				
	#	Sub-sample	Up		Down		HIGH / LOW	Up		Down		HIGH / LOW
			Value	Uncer.	Value	Uncer.		Value	Uncer.	Value	Uncer.	
GS0000038	5	0-2 C										
GS0000039		2-4 C										
GS0000040		0-2 W										
GS0000041		0-2 E										
GS0000042		0-2 N										
GS0000043		0-2 S										
GS0000044	6	0-2 C										
GS0000045		2-4 C										
GS0000046		0-2 W										
GS0000047		0-2 E										
GS0000048		0-2 N										
GS0000049		0-2 S										
GS0000050	7	0-2 C										
GS0000051		2-4 C	1,570	363	1,600	371	1.02	1,390	323	1,450	337	1.04
GS0000052		4-6 C	578	134	549	127	1.05	455	106	639	149	1.40
GS0000053		0-2 W										
GS0000054		0-2 E										
GS0000055		0-2 N										
GS0000056		0-2 S										
GS0000057	8	0-2	1,750	406	1,650	382	1.06					
GS0000058	9	0-2										
GS0000059	10	0-2										
GS0000060	11	0-2										
GS0000061	12	0-2										

Table A-1. Individual Aliquot $^{239/240}$Pu Activity Concentrations (Continued)
(1999 Sampling)

Base Sample Number	Sampling Location		Combined Aliquot Statistical Parameters	
	#	Sub-sample	Mean Activity Concentration (pCi/g)	Percent Coefficient Of Variation
GS0000038	5	0-2 C	578	37
GS0000039		2-4 C	183	53
GS0000040		0-2 W	279	44
GS0000041		0-2 E	572	39
GS0000042		0-2 N	544	22
GS0000043		0-2 S	200	39
GS0000044	6	0-2 C	2,220	146
GS0000045		2-4 C	737	60
GS0000046		0-2 W	752	66
GS0000047		0-2 E	885	13
GS0000048		0-2 N	2,560	69
GS0000049		0-2 S	4,680	82
GS0000050	7	0-2 C	5,270	39
GS0000051		2-4 C	1,850	31
GS0000052		4-6 C	584	14
GS0000053		0-2 W	12,900	115
GS0000054		0-2 E	19,900	135
GS0000055		0-2 N	3,870	23
GS0000056		0-2 S	6,320	45
GS0000057	8	0-2	3,210	71
GS0000058	9	0-2	12	28
GS0000059	10	0-2	3.2	43
GS0000060	11	0-2	128	13
GS0000061	12	0-2	3.9	75

Table A-1. Individual Aliquot $^{239/240}$Pu Activity Concentrations (Continued)
(1999 Sampling)

Base Sample Number	Sampling Location		Aliquot #1					Aliquot #2				
			Pu-239/240 Activity Concentration (pCi/g)					Pu-239/240 Activity Concentration (pCi/g)				
	#	Sub-sample	Up		Down		HIGH / LOW	Up		Down		HIGH / LOW
			Value	Uncer.	Value	Uncer.		Value	Uncer.	Value	Uncer.	
GS0000062	13	0-2	856	199	493	115	1.74	82	19	103	24	1.26
GS0000063	14	0-2	64	15	54	12	1.19	39	9.2	63	15	1.63
GS0000064	15	0-2	153	36	212	49	1.38	107	25	109	25	1.02
GS0000065	16	0-2	1,150	268	1,080	251	1.07	1,630	378	1,700	394	1.04
GS0000066	17	0-2	1,750	406	1,510	351	1.16	1,280	139	1,520	247	1.18
GS0000067	18	0-2	97	23	86	20	1.13	757	176	456	106	1.66
GS0000068	19	0-2	706	164	457	106	1.54	486	113	572	132	1.18
GS0000069	20	0-2	39,800	9,230	30,300	7,030	1.31	3,490	810	4,960	1,150	1.42
GS0000070	21	0-2	5,070	1,180	5,830	1,350	1.15	4,400	1,020	5,550	1,290	1.26
GS0000071	22	0-2	106	25	105	24	1.01	645	150	1,100	254	1.70
GS0000072	23	0-2	257	60	294	69	1.14	198	46	191	45	1.04
GS0000073	24	0-2	832	193	809	188	1.03	408	95	355	83	1.15
GS0000074	25	0-2	302	70	281	65	1.07	860	200	412	96	2.09
GS0000075	26	0-2	205	48	231	54	1.13	854	199	1,090	253	1.28
GS0000076	27	0-2	611	142	498	116	1.23	419	97	443	103	1.06
GS0000077	28	0-2	170	39	181	42	1.07	301	70	545	127	1.81
GS0000078	29	0-2	196	45	237	55	1.21	213	50	196	45	1.09
GS0000079	30	0-2	16,800	3,900	55,500	12,900	3.30	399	93	346	81	1.15

Table A-1. Individual Aliquot $^{239/240}$Pu Activity Concentrations (Continued)
(1999 Sampling)

Base Sample Number	Sampling Location		Aliquot #3					Aliquot #4				
			Pu-239/240 Activity Concentration (pCi/g)					Pu-239/240 Activity Concentration (pCi/g)				
	#	Sub-sample	Up		Down		HIGH / LOW	Up		Down		HIGH / LOW
			Value	Uncer.	Value	Uncer.		Value	Uncer.	Value	Uncer.	
GS0000062	13	0-2	116	27	96	22	1.21	89	21	115	27	1.29
GS0000063	14	0-2	25	5.9	27	6.5	1.06					
GS0000064	15	0-2	155	36	143	33	1.08	146	34	142	32.9	1.03
GS0000065	16	0-2	809	188	1,140	264	1.41	1,660	385	2,040	473	1.23
GS0000066	17	0-2	1,250	289	1,200	279	1.04	2,430	564	1990	461	1.22
GS0000067	18	0-2	86	20	115	27	1.33	587	136	414	96	1.42
GS0000068	19	0-2	292	68	377	88	1.29	454	105	382	89	1.19
GS0000069	20	0-2	3,560	826	3,470	807	1.02					
GS0000070	21	0-2	11,900	2,760	10,600	2,460	1.12					
GS0000071	22	0-2	112	26	1,450	34	1.29					
GS0000072	23	0-2	253	59	247	57	1.02	216	50	179	42	1.20
GS0000073	24	0-2	617	143	572	133	1.08	218	51	241	56	1.10
GS0000074	25	0-2										
GS0000075	26	0-2	373	87	542	126	1.45	274	64	189	44	1.45
GS0000076	27	0-2	380	88	443	103	1.17	577	134	599	139	1.04
GS0000077	28	0-2	209	49	187	43	1.12					
GS0000078	29	0-2	160	37	160	37	1.00					
GS0000079	30	0-2	503	117	486	113	1.04					

Table A-1. Individual Aliquot $^{239/240}$Pu Activity Concentrations (Continued)
(1999 Sampling)

Base Sample Number	Sampling Location		Aliquot #5					Aliquot #6				
			Pu-239/240 Activity Concentration (pCi/g)					Pu-239/240 Activity Concentration (pCi/g)				
	#	Sub-sample	Up		Down		HIGH / LOW	Up		Down		HIGH / LOW
			Value	Uncer.	Value	Uncer.		Value	Uncer.	Value	Uncer.	
GS0000062	13	0-2										
GS0000063	14	0-2										
GS0000064	15	0-2										
GS0000065	16	0-2	9,430	2,190	5,130	1,190	1.84	1,240	287	1,460	339	1.18
GS0000066	17	0-2										
GS0000067	18	0-2	45	10	66	15	1.47					
GS0000068	19	0-2										
GS0000069	20	0-2										
GS0000070	21	0-2										
GS0000071	22	0-2										
GS0000072	23	0-2	1,660	3867	1,130	262	1.47					
GS0000073	24	0-2										
GS0000074	25	0-2										
GS0000075	26	0-2										
GS0000076	27	0-2										
GS0000077	28	0-2										
GS0000078	29	0-2										
GS0000079	30	0-2										

Table A-1. Individual Aliquot $^{239/240}$Pu Activity Concentrations (Continued)
(1999 Sampling)

Base Sample Number	Sampling Location		Aliquot #7 Pu-239/240 Activity Concentration (pCi/g)					Combined Aliquot Statistical Parameters	
	#	Sub-sample	Up Value	Up Uncer.	Down Value	Down Uncer.	HIGH / LOW	Mean Activity Concentration (pCi/g)	Percent Coefficient Of Variation
GS0000062	13	0-2						244	117
GS0000063	14	0-2						46	38
GS0000064	15	0-2						146	22.3
GS0000065	16	0-2	1,280	297	1,140	266	1.12	2,210	106
GS0000066	17	0-2						1,610	26
GS0000067	18	0-2						271	96
GS0000068	19	0-2						466	27
GS0000069	20	0-2						14,300	115
GS0000070	21	0-2						7,230	44
GS0000071	22	0-2						368	113
GS0000072	23	0-2						463	110
GS0000073	24	0-2						507	47
GS0000074	25	0-2						464	58
GS0000075	26	0-2						470	72
GS0000076	27	0-2						496	18
GS0000077	28	0-2						266	55
GS0000078	29	0-2						194	15
GS0000079	30	0-2						12,300	179

Uncertainty Values at the 95 % Confidence Level Based on Uncertainties from Counting Uncertainties Only

Blank Cells Represent Samples Where an Aliquot Did Not Exist (Aliquot Number Varied from 2 to 10 per Sample)

Table A-2. Individual Aliquot $^{239/240}$Pu Activity Concentrations.
(2000 Sampling)

Base Sample Number	Sampling Location		Aliquot #1					Aliquot #2				
			Pu-239/240 Activity Concentration (pCi/g)					Pu-239/240 Activity Concentration (pCi/g)				
	#	Sub-sample	Up		Down		HIGH / LOW	Up		Down		HIGH / LOW
			Value	Uncer.	Value	Uncer.		Value	Uncer.	Value	Uncer.	
GS0000669	31	2-4	49	11	35	8.1	1.40	39	9.2	39	9.2	1.00
GS0000670	32	2-4	154	36	191	44	1.24	93	22	125	29	1.34
GS0000671	33	2-4	61	14	65	15	1.07	75	17	96	22	1.29
GS0000672	34	2-4	41	9.7	38	8.6	1.07	117	27	111	26	1.05
GS0000673	35	2-4	3.8	1.1	2.7	0.5	1.40	2.7	0.5	2.7	0.5	1.00
GS0000674	36	2-4	14	3.2	16	3.8	1.20	21	4.9	21	4.9	1.00
GS0000675	37	2-4	50	12	51	12	1.01	2.7	0.5	3.8	1.1	1.40
GS0000676	38	2-4	2.7	0.5	1.6	0.5	1.67	1.1	0.4	1.1	0.5	1.00
GS0000677	39	2-4	66	15	60	14	1.10	57	13	75	17	1.31
GS0000678	40	2-4	465	108	899	208	1.93	150	35	161	37	1.08
GS0000679	31	0-2	55	13	43	10	1.28	135	31	54	12	2.49
GS0000680	32	0-2	32	7.6	32	7.6	1.00	21	4.9	18	4.3	1.12
GS0000681	33	0-2	69	16	68	16	1.02	35	39	39	9.2	1.12
GS0000682	34	0-2	200	46	286	66	1.43	108	25	109	25	1.01
GS0000683	35	0-2	5.4	1.1	9.2	2.2	1.70	3.8	1.1	5.4	1.1	1.43
GS0000684	36	0-2	9.7	2.2	10	2.7	1.06	13	3.2	14	3.2	1.08
GS0000685	37	0-2	2.2	0.5	2.2	0.5	1.00	3.8	1.1	4.3	1.1	1.14
GS0000686	38	0-2	3.2	1.1	3.2	1.1	1.00	1.6	0.5	2.2	0.5	1.33
GS0000687	39	0-2	109	25	159	37	1.46	77	18	82	19	1.07
GS0000688	40	0-2	223	52	333	77	1.50	145	34	144	34	1.01

Uncertainty Values at the 95 % Confidence Level Based on Uncertainties from Counting Uncertainties Only

Table A-2. Individual Aliquot $^{239/240}$Pu Activity Concentrations (Continued)
(2000 Sampling)

Base Sample Number	Sampling Location		Aliquot #3					Aliquot #4				
			Pu-239/240 Activity Concentration (pCi/g)					Pu-239/240 Activity Concentration (pCi/g)				
	#	Sub-sample	Up		Down		HIGH / LOW	Up		Down		HIGH / LOW
			Value	Uncer.	Value	Uncer.		Value	Uncer.	Value	Uncer.	
GS0000669	31	2-4	36	8.6	28	6.5	1.29					
GS0000670	32	2-4	76	18	77	18	1.01	90	21	96	22	1.07
GS0000671	33	2-4	61	14.0	43	10	1.41	34	8.1	41	9.2	1.19
GS0000672	34	2-4	62	15	77	18	1.25					
GS0000673	35	2-4	3.2	0.5	4.9	1.1	1.50	1.1	0.4	1.6	0.5	1.50
GS0000674	36	2-4	7.0	1.6	6.5	1.6	1.08	6.5	1.6	6.5	1.6	1.00
GS0000675	37	2-4	1.6	0.5	2.2	0.5	1.33	13	3.2	30	7.0	2.29
GS0000676	38	2-4	2.2	0.5	3.2	0.5	1.50	3.2	1.1	6.5	1.6	2.00
GS0000677	39	2-4	42	9.7	43	10	1.04	52	12	60	14	1.16
GS0000678	40	2-4	98	23	127	30	1.30	117	27	194	45	1.66
GS0000679	31	0-2	25	5.9	41	9.7	1.63	56	13	68	16	1.21
GS0000680	32	0-2	19	4.3	20	4.9	1.03	24	5.4	25	5.9	1.07
GS0000681	33	0-2	33	7.6	35	8.1	1.05	39	9.2	37	8.6	1.04
GS0000682	34	0-2	164	38	126	29	1.30	440	102	481	112	1.09
GS0000683	35	0-2	3.2	1.1	3.8	1.1	1.17	9.2	2.2	15	3.2	1.59
GS0000684	36	0-2	19	4.9	12	2.7	1.57	11	2.7	7.6	1.6	1.43
GS0000685	37	0-2	27	5.9	39	9.2	1.49	7.0	1.6	6.5	1.6	1.08
GS0000686	38	0-2	2.7	0.5	2.7	0.5	1.00	3.8	1.1	3.2	0.5	1.17
GS0000687	39	0-2	87	20	77	18	1.13	126	29	108	25	1.18
GS0000688	40	0-2	340	79	290	68	1.17	144	34	203	47	1.41

Uncertainty Values at the 95 % Confidence Level Based on Uncertainties from Counting Uncertainties Only

Table A-2. Individual Aliquot $^{239/240}$Pu Activity Concentrations (Continued)
(2000 Sampling)

Base Sample Number	Sampling Location		Aliquot #5					Aliquot #6				
			Pu-239/240 Activity Concentration (pCi/g)					Pu-239/240 Activity Concentration (pCi/g)				
	#	Sub-sample	Up		Down		HIGH / LOW	Up		Down		HIGH / LOW
			Value	Uncer.	Value	Uncer.		Value	Uncer.	Value	Uncer.	
GS0000669	31	2-4										
GS0000670	32	2-4										
GS0000671	33	2-4										
GS0000672	34	2-4										
GS0000673	35	2-4										
GS0000674	36	2-4	7.0	1.6	14	3.2	2.00	7.0	1.6	8.1	2.2	1.15
GS0000675	37	2-4	3.2	1.1	3.2	1.1	1.00					
GS0000676	38	2-4	5.9	1.6	14	3.2	2.36					
GS0000677	39	2-4	74	17	67	16	1.10					
GS0000678	40	2-4	73	17	88	21	1.20					
GS0000679	31	0-2	19	4.3	23	5.4	1.20					
GS0000680	32	0-2	21	4.9	17	3.8	1.26					
GS0000681	33	0-2	75	17	71	17	1.05	34	7.6	43	9.7	1.27
GS0000682	34	0-2	139	32	226	52	1.63	201	46	154	36	1.31
GS0000683	35	0-2	9.7	2.2	8.6	2.2	1.13	22	5.4	29	6.5	1.29
GS0000684	36	0-2	8.6	2.2	8.1	2.2	1.07	7.0	1.6	6.5	1.6	1.08
GS0000685	37	0-2	2.2	0.5	4.3	1.1	2.00	3.2	0.5	2.7	0.5	1.20
GS0000686	38	0-2	1.6	0.4	2.2	0.5	1.33	3.2	1.1	3.8	1.1	1.17
GS0000687	39	0-2	85	20	88	21	1.03	159	37	133	31	1.19
GS0000688	40	0-2	121	28	123	29	1.02	575	133	251	58	2.30

Uncertainty Values at the 95 % Confidence Level Based on Uncertainties from Counting Uncertainties Only

Table A-2. Individual Aliquot $^{239/240}$Pu Activity Concentrations (Continued)
(2000 Sampling)

Base Sample Number	Sampling Location		Aliquot #7					Aliquot #8				
			Pu-239/240 Activity Concentration (pCi/g)					Pu-239/240 Activity Concentration (pCi/g)				
	#	Sub-sample	Up		Down		HIGH / LOW	Up		Down		HIGH / LOW
			Value	Uncer.	Value	Uncer.		Value	Uncer.	Value	Uncer.	
GS0000669	31	2-4										
GS0000670	32	2-4										
GS0000671	33	2-4										
GS0000672	34	2-4										
GS0000673	35	2-4										
GS0000674	36	2-4										
GS0000675	37	2-4										
GS0000676	38	2-4										
GS0000677	39	2-4										
GS0000678	40	2-4										
GS0000679	31	0-2										
GS0000680	32	0-2										
GS0000681	33	0-2										
GS0000682	34	0-2										
GS0000683	35	0-2	4.3	1.1	4.9	1.1	1.13					
GS0000684	36	0-2	9.7	2.2	15	3.2	1.56					
GS0000685	37	0-2	2.2	0.5	2.2	0.5	1.00	5.9	1.6	5.9	1.6	1.00
GS0000686	38	0-2	3.2	0.5	3.2	1.1	1.00					
GS0000687	39	0-2	70	16	74	17	1.05	117	27	119	28	1.02
GS0000688	40	0-2	331	77	223	52	1.48	17	4.0	92	22	5.40

Uncertainty Values at the 95 % Confidence Level Based on Uncertainties from Counting Uncertainties Only

Table A-2. Individual Aliquot $^{239/240}$Pu Activity Concentrations (Continued)
(2000 Sampling)

Base Sample Number	Sampling Location		Aliquot #9					Aliquot #10				
			Pu-239/240 Activity Concentration (pCi/g)					Pu-239/240 Activity Concentration (pCi/g)				
	#	Sub-sample	Up		Down		HIGH / LOW	Up		Down		HIGH / LOW
			Value	Uncer.	Value	Uncer.		Value	Uncer.	Value	Uncer.	
GS0000669	31	2-4										
GS0000670	32	2-4										
GS0000671	33	2-4										
GS0000672	34	2-4										
GS0000673	35	2-4										
GS0000674	36	2-4										
GS0000675	37	2-4										
GS0000676	38	2-4										
GS0000677	39	2-4										
GS0000678	40	2-4										
GS0000679	31	0-2										
GS0000680	32	0-2										
GS0000681	33	0-2										
GS0000682	34	0-2										
GS0000683	35	0-2										
GS0000684	36	0-2										
GS0000685	37	0-2										
GS0000686	38	0-2										
GS0000687	39	0-2										
GS0000688	40	0-2	1,160	269	3,000	696	2.58	240	56	254	59	1.06

Uncertainty Values at the 95 % Confidence Level Based on Uncertainties from Counting Uncertainties Only

Table A-2. Individual Aliquot $^{239/240}$Pu Activity Concentrations (Continued)
(2000 Sampling)

Base Sample Number	Sampling Location		Combined Aliquot Statistical Parameters	
	#	Sub-sample	Mean Activity Concentration (pCi/g)	Percent Coefficient Of Variation
GS0000669	31	2-4	38	18
GS0000670	32	2-4	113	36
GS0000671	33	2-4	59	34
GS0000672	34	2-4	74	46
GS0000673	35	2-4	2.8	42
GS0000674	36	2-4	11	51
GS0000675	37	2-4	16	125
GS0000676	38	2-4	4.2	95
GS0000677	39	2-4	60	19
GS0000078	40	2-4	237	109
GS0000679	31	0-2	52	64
GS0000680	32	0-2	23	23
GS0000681	33	0-2	48	35
GS0000682	34	0-2	219	57
GS0000683	35	0-2	9.5	79
GS0000684	36	0-2	11	33
GS0000685	37	0-2	7.5	137
GS0000686	38	0-2	2.9	25
GS0000687	39	0-2	104	28
GS0000688	40	0-2	255	158

Uncertainty Values at the 95 % Confidence Level Based on Uncertainties from Counting Uncertainties Only

Blank Cells Represent Samples Where an Aliquot Did Not Exist (Aliquot Number Varied from 2 to 10 per Sample)

Table A-3. University of Pittsburgh - Individual Sub-Aliquot Estimated $^{239/240}$Pu Activity Concentrations

Base Sample Number	Sub-Aliquot Number	Estimated Pu-239/240 Activity Concentration (pCi/g)*		Relative Percent Difference
		U. of Pittsburgh**	AFIERA	
GS0000050	1	6,979 ± 350	4,381	0.34
	2	5,884 ± 295		
	3	3,633 ± 182		
	4	5,392 ± 270		
	5	6,963 ± 349		
	6	4,276 ± 214		
	7	3,013 ± 151		
	8	2,001 ± 100		
	9	2,417 ± 121		
	10	3,098 ± 156		
	Mean	4,366		
	% CV	42		
GS0000051	1	2,611 ± 131	2,799	2.8
	2	2,055 ± 103		
	3	5,279 ± 265		
	4	4,878 ± 245		
	5	1,943 ± 97		
	6	3,238 ± 163		
	7	2,077 ± 104		
	8	4,078 ± 204		
	9	1,223 ± 62		
	10	1,398 ± 70		
	Mean	2,878		
	% CV	50		
GS0000052	1	687 ± 131	579	- 3.3
	2	585 ± 103		
	3	465 ± 265		
	4	442 ± 245		
	5	503 ± 97		
	6	626 ± 163		
	7	747 ± 104		
	8	415 ± 204		
	9	872 ± 62		
	10	259 ± 70		
	Mean	560		
	% CV	32		

Table A-3. University of Pittsburgh - Individual Sub-Aliquot Estimated $^{239/240}$Pu Activity Concentrations (Continued)

Base Sample Number	Sub-Aliquot Number	Estimated Pu-239/240 Activity Concentration (pCi/g)*		Relative Percent Difference
		U. of Pittsburgh**	AFIERA	
GS0000057	1	2,655 ± 133	2,281	5.5
	2	3,751 ± 188		
	3	2,566 ± 129		
	4	1,152 ± 58		
	5	2,615 ± 131		
	6	3,579 ± 179		
	7	2,453 ± 123		
	8	1,522 ± 76		
	9	2,156 ± 108		
	10	1,650 ± 83		
	Mean	2,410		
	% CV	35		
GS0000079	1	272,436 ± 13,646***	36,123	25
	2	455 ± 23		
	3	447 ± 23		
	4	4,321 ± 22		
	5	442 ± 22		
	6	534 ± 27		
	7	554 ± 28		
	8	490 ± 25		
	9	697 ± 30		
	10	342 ± 17		
	Mean	28,062		
	% CV	306		

* $^{239/240}$Pu to ^{241}Am Ratio of 5.4 Assumed

** 95 % Confidence Level

*** Single Particle PuO_2 Estimated Diameter = 250 μm

Appendix B

Select Plots of $^{239/240}$Pu Activity Concentration For Individual Aliquots

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Figure B-1. $^{239/240}$Pu Activity Concentrations for Individual Aliquots (Sample 67) [Mean $^{239/240}$Pu = 271 pCi g^{-1}, % CV = 96%].

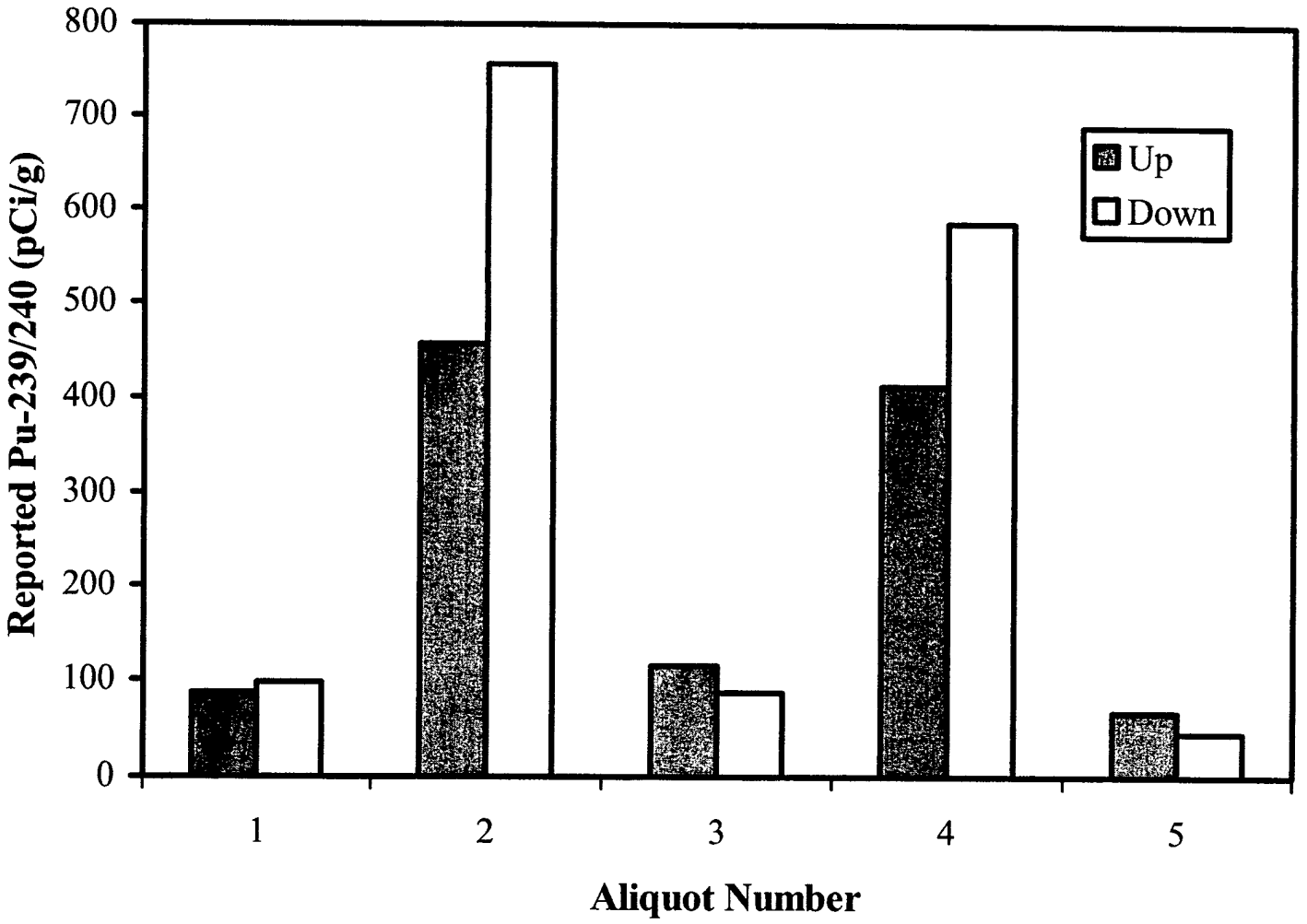

Figure B-2. $^{239/240}$Pu Activity Concentrations for Individual Aliquots (Sample 15) [Mean $^{239/240}$Pu = 219 pCi g^{-1}, % CV = 128 %].

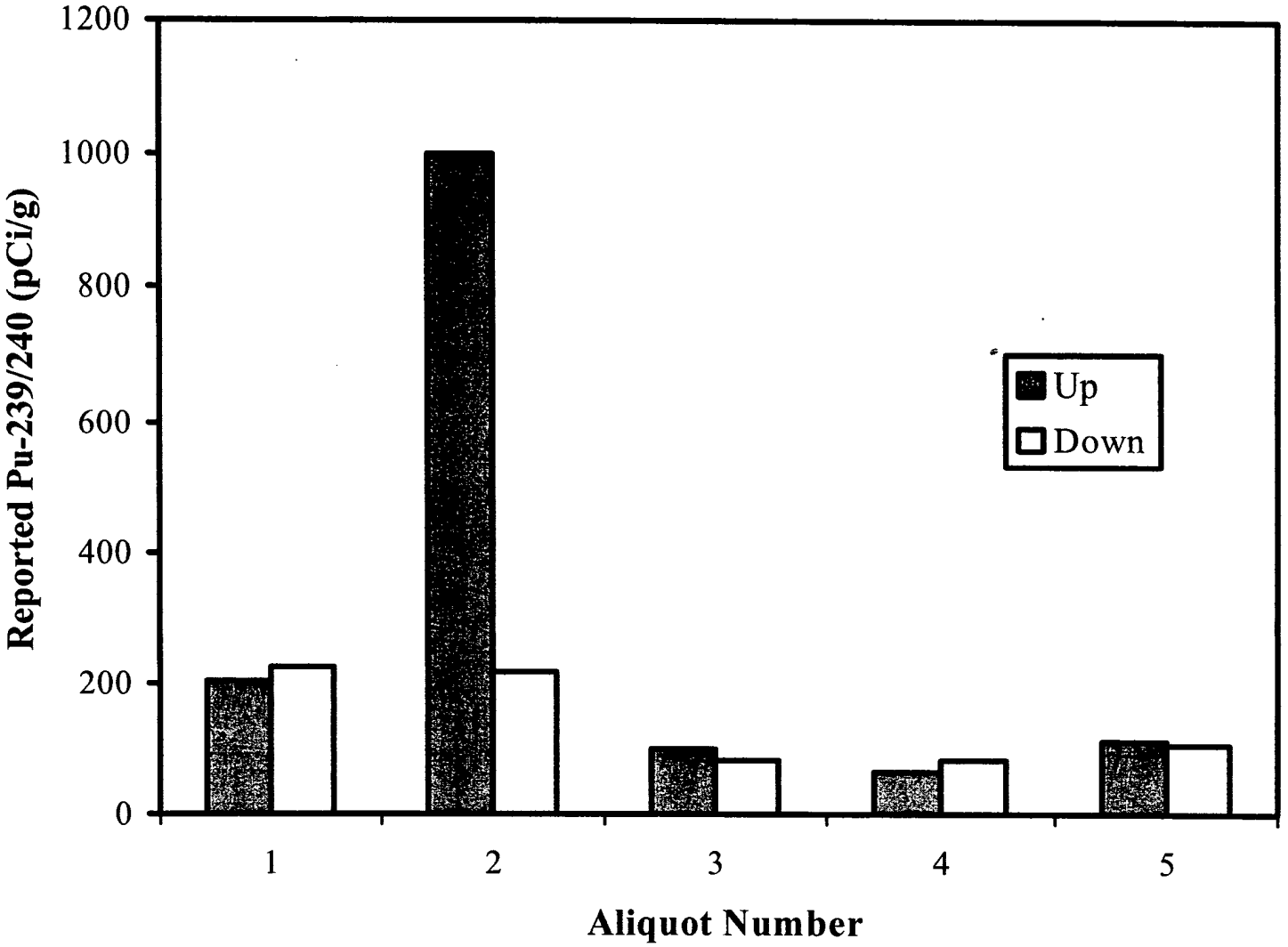

Figure B-3. $^{239/240}$Pu Activity Concentrations for Individual Aliquots (Sample 53) [Mean $^{239/240}$Pu = 12,900 pCi g^{-1}, % CV = 115 %].

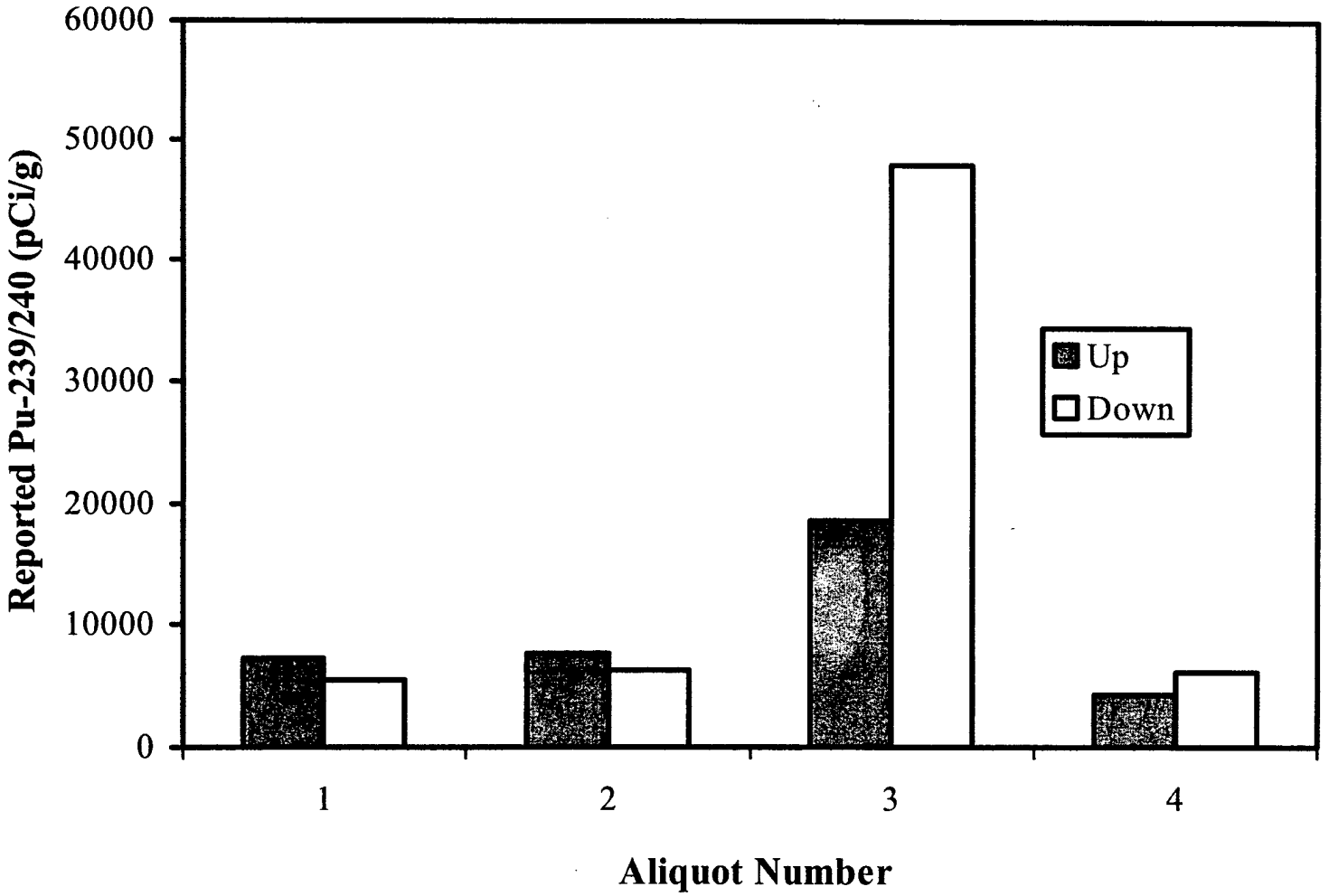

Figure B-4. $^{239/240}$**Pu Activity Concentrations for Individual Aliquots (Sample 685) [Mean** $^{239/240}$**Pu = 7.5 pCi g^{-1}, % CV = 137 %].**

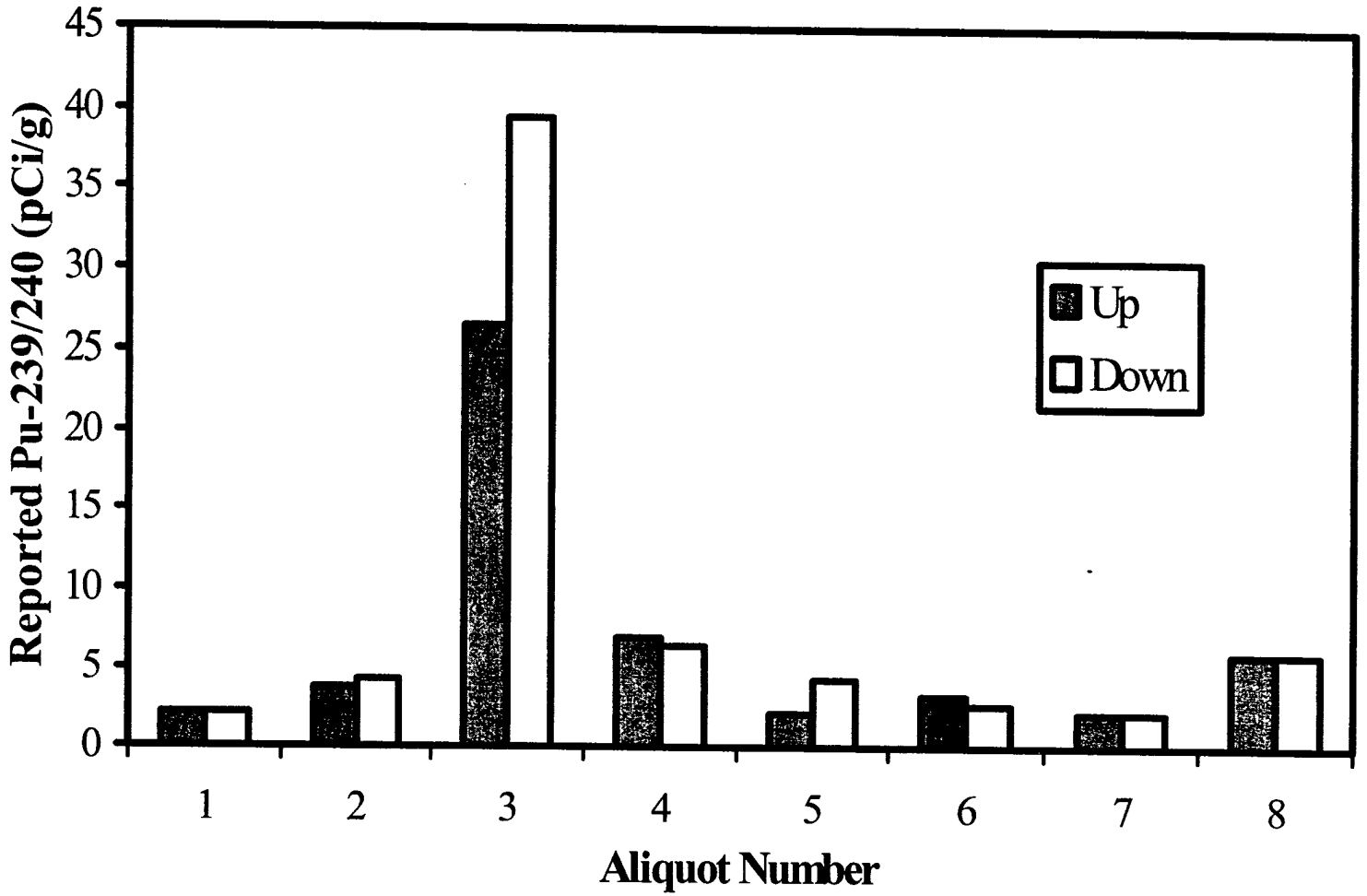

Figure B-5. $^{239/240}$Pu Activity Concentrations for Individual Aliquots (Sample 675) [Mean $^{239/240}$Pu = 16 pCi g^{-1}, % CV = 125 %].

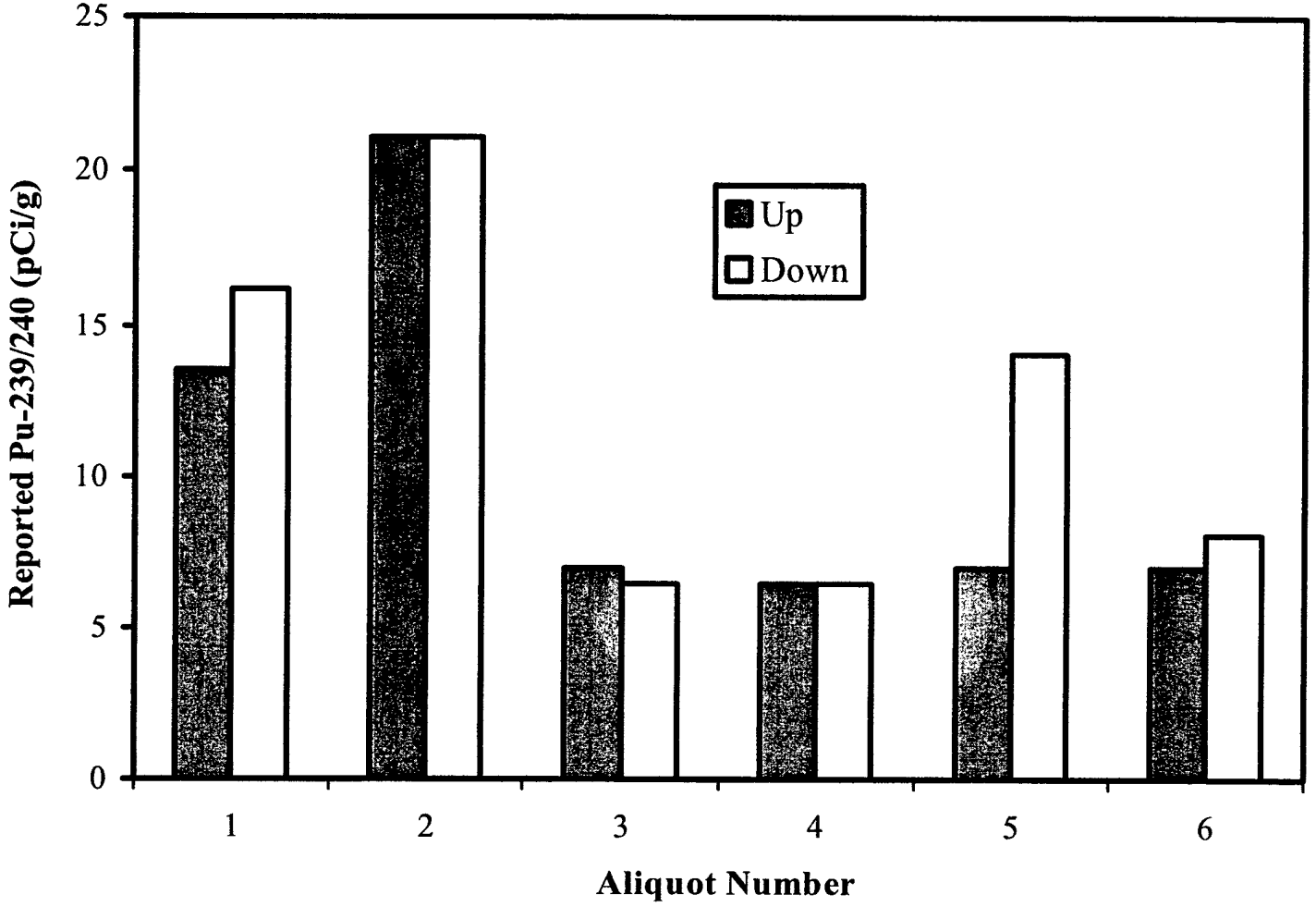

Figure B-6. $^{239/240}$Pu Activity Concentrations for Individual Aliquots (Sample 75) [Mean $^{239/240}$Pu = 470 pCi g^{-1}, % CV = 72 %].

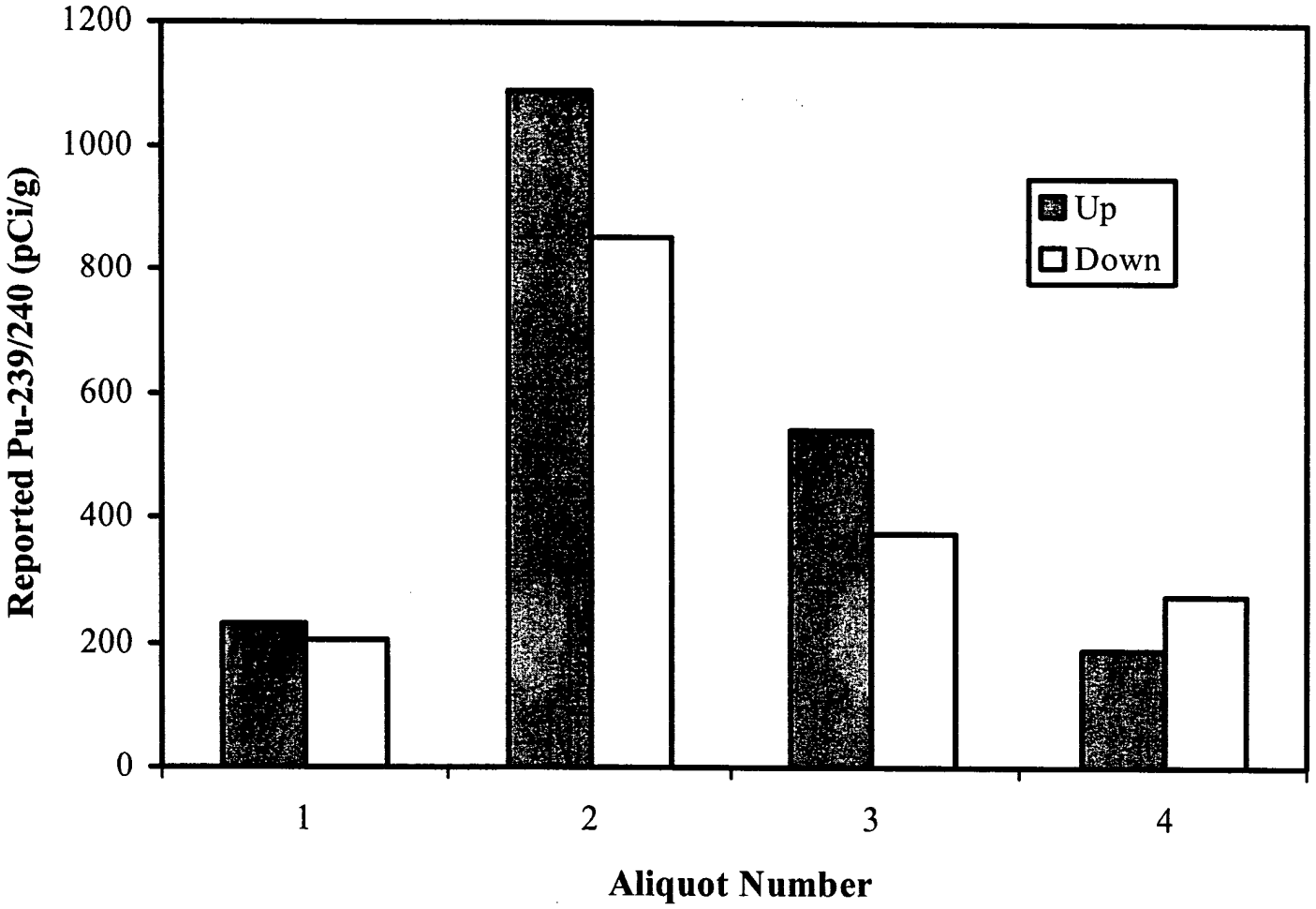

Figure B-7. $^{239/240}$Pu Activity Concentrations for Individual Aliquots (Sample 60) [Mean $^{239/240}$Pu = 128 pCi g^{-1}, % CV = 13 %].

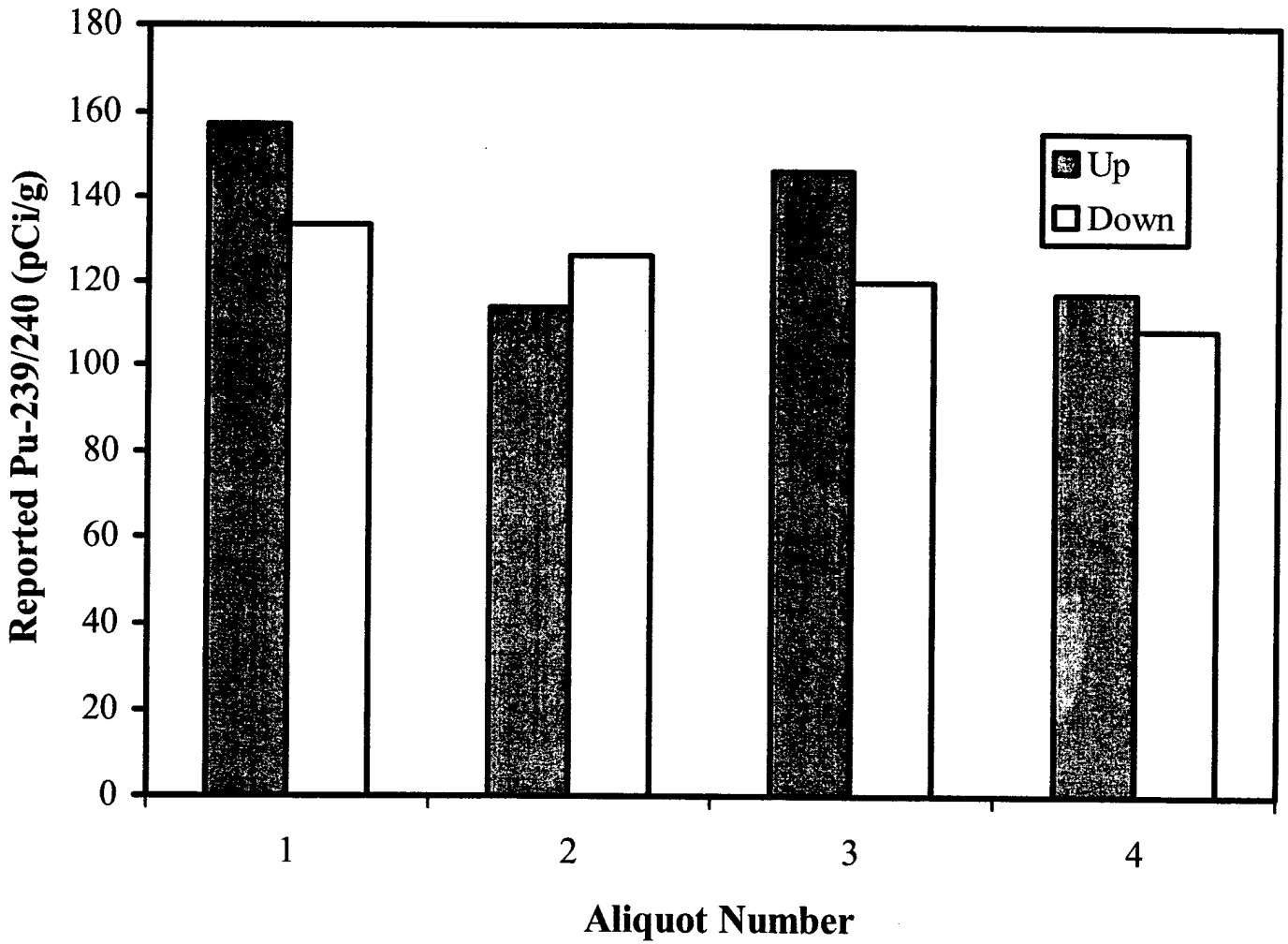

Figure B-8. $^{239/240}$Pu Activity Concentrations for Individual Aliquots (Sample 686) [Mean $^{239/240}$Pu = 2.9 pCi g^{-1}, % CV = 25 %].