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STATISTICAL DESIGN AND ANALYSIS OF ENVIRONMENTAL STUDIES FOR PLUTONIUM AND OTHER TRANSURANICS AT NAEG "SAFETY-SHOT" SITES

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ABSTRACT

This paper is centered around the use of stratified random sampling for estimating the total amount (inventory) of $^{239-240}Pu$ and uranium in surface soil at ten "safety-shot" sites on the Nevada Test Site (NTS) and Tonopah Test Range (TTR) that are currently being studied by the Nevada Applied Ecology Group (NAEG). The use of stratified random sampling has resulted in estimates of inventory at these desert study sites that have smaller standard errors than would have been the case had simple random sampling (no stratification) been used.

Estimates of inventory are given for ^{235}U, ^{238}U, and $^{239-240}Pu$ in soil at A Site of Area 11 on the NTS. Other results presented include average concentrations of one or more of these isotopes in soil and vegetation and in soil profile samples at depths to 25 cm. The regression relationship between soil and vegetation concentrations of ^{235}U and ^{238}U at adjacent sampling locations is also examined using three different models.

The applicability of stratified random sampling to the estimation of concentration contours of $^{239-240}Pu$ in surface soil using computer algorithms is also investigated. Estimates of such contours are obtained using several different methods. The planning of field sampling plans for estimating inventory and distribution is discussed.

1. INTRODUCTION

The Nevada Applied Ecology Group (NAEG), established by the Atomic Energy Commission (Nevada Operations Office) in July 1970, is currently engaged in an environmental sampling program for plutonium and other transuranics at 10 "safety-shot" sites on the Nevada Test Site (NTS) and the Tonopah Test Range (TTR) in the State of Nevada, U.S.A. At most of these sites a chemical explosive was detonated in close proximity to arrangements or assemblies of plutonium and/or uranium resulting in contamination of the surrounding soil and vegetation. The tests were conducted between 1954 and 1963 so that the contamination has been exposed to the effects of weathering in a desert environment for up to 21 years. The sites being studied on the NTS are Project 57 site in Area 13, GMX site in Area 5, and A, B, C, and D sites in Area 11 (Plutonium Valley). Those on the TTR are the 4 Roller Coaster sites known as Double Track and Clean Slates 1, 2, and 3.

Objectives of the current NAEG program include estimating the total amount (inventory) and geographical distribution of $^{239-240}Pu$ and/or ^{235}U and ^{238}U in surface (0-5 cm) soil. More than 2000 soil and vegetation samples have been collected during 1973-74 using stratified random sampling, wherein each study site was first divided into subregions (strata) on the basis of general activity levels in the soil, and within which soil samples were collected at random locations. Statistical analyses of resulting data have been reported in [1], [2] and [3]. Some of the soils data have also been examined in [4] relative to the design of more efficient field sampling designs using "double sampling".

The purpose of the present paper is twofold: (i) to present some analyses of soil and vegetation data (primarily ^{234}U, ^{235}U, ^{236}U, ^{238}U, and $^{239-240}Pu$) that have recently become available from A Site in Area 11, and (ii) to describe and discuss the field sampling design we have used for estimating inventory and geographical distribution. The expense of plutonium analysis is such that careful attention should be given to the use of efficient field sampling designs that hold down total costs, but which still yield estimates of sufficient accuracy and precision to meet the objectives of the study. Our latest efforts at estimating concentration contours using polynomial regression and moving average (nearest neighbor) estimation routines are also given here.

2. METHODS

2.1 Estimating Inventory

The geographical distribution of plutonium at the safety-shot sites is characterized by high activity levels in the immediate vicinity of ground zero (GZ) with concentrations decreasing rapidly with distance. Hence, if n samples are collected at randomly chosen locations over the entire area, the resulting standard error (S.E.) of the mean ($\bar{x}$) concentration per unit area will be large, as will that of the estimate of inventory.

A sampling design we have found to be generally effective in obtaining estimates of inventory with smaller S.E.'s than possible with the above approach, is known as stratified random sampling wherein the study area is divided into L strata (subregions) based on general activity levels in the soil, and where within each stratum, soil samples are collected at random locations (see [3], Appendix A; [5], Chapters 5 and 5A for computational details). In general, the area immediately surrounding GZ is designated as a separate stratum as is the low activity level region farthest removed from GZ, with several strata demarcated at intermediate activity levels between these two extremes (methods used to define stratum boundries are discussed below). When the radionuclides are distributed about GZ as described above for the safety-shot sites, we have found that the mean concentrations for the various strata ($\bar{x}_h$, h = 1, 2, ..., L) will vary widely depending on the general activity level of the soil within the stratum, and that the stratum S.E.'s ($s_h/\sqrt{n_h}$, h = 1, 2, ..., L) will be smaller than that for the estimate $\bar{x}$. As discussed in [5] (page 100), this is a situation where we might expect stratified random sampling to result in a more precise estimate of inventory than would be obtained without stratification.

The estimate of inventory $\hat{I}_s$ using stratified random sampling is the sum of the stratum estimates of inventory ($A_h \bar{x}_h$). The S.E.$_2$of $\hat{I}_s$ is the square root of the sum of the stratum inventory variances ($A_h^2 s_h^2/n_h$), where A_h is the area in square meters of the h^{th} stratum.

Methods used to define stratum boundries and determine the number of samples to collect within each stratum have been discussed in detail in [3] and [6]. Briefly, strata were determined by taking ^{241}Am count per minute (CPM) readings on a grid system using the portable FIDLER (Field Instrument for the Determination of Low Energy Radiation) with a 5 in. NaI crystal. These counts approximate the plutonium contamination in surface soil. At A Site, where the FIDLER could not be used due to background interference, soil samples were collected at grid intersections and counted in the laboratory on a Ge(Li) counter for ^{241}Am.

The number of soil samples n_h collected in the h^{th} stratum was determined using as a guide the optimum allocation formula given in [5] (Eq. 5.20). The allocation depends on the relative sizes of strata and on the variability in concentrations from sample to sample within strata. Estimates of these stratum variances are required for the allocation formula. Ideally, these should be available from pilot studies conducted earlier at the sites to be studied, but often such information does not exist. As discussed in [6], we lacked such information in Area 13 and instead assumed that the coefficient of variation $c(= s_h/\bar{x}_h)$ was equal to 1 in all strata and that $\bar{x}_h$ was proportional to the median FIDLER reading in the h^{th} stratum.

The usual 95% confidence limits $\hat{I}_s \pm 2[\text{S.E.}(\hat{I}_s)]$ are appropriate if $\hat{I}_s$ is normally distributed. The data from Area 13 were examined in [1], and for most strata, they fit the lognormal distribution [7] better than the normal. If data are lognormally distributed, a method in [8] may be used to compute exact confidence intervals for the inventory I_h of each stratum, but not for I_s.

2.2 Estimating Concentration Contours

Soil and Vegetation plutonium data available from the stratified random sampling for inventory were used in [3] to experiment with estimating concentration contours. A computer program[1] entitled SURFACE II was used in this effort [9]. Briefly, the contour lines were estimated using each plutonium concentration datum z_i ($i = 1, 2, \ldots, N$; N = total observations over all strata) each with its north-south, east-west coordinates x and y, respectively. An estimated plutonium concentration $\hat{z}_g$ is obtained at regular intervals (grid nodes) on a grid laid over the study site using a local weighted average of trends or slopes in the neighborhood around each grid node. This is a two-phase procedure described in Appendix C of [9] resulting in a grid matrix of values $\hat{z}_g$.

Several search methods are available on SURFACE II for selecting "nearby" data points. "NEAR" selects the k nearest data points without regard to direction or distance and was used in [3] with k = 8. Estimated grid matrices and contour (isopleth) lines (obtained by linear interpolation between the $\hat{z}_g$ values of the grid matrix) are given in [3] for soil and vegetation for the Project 57 and GMX study sties. Other search methods available include "QUAD" and "OCTANT", where the area about each grid node is divided into 4 or 8 equal

[1] Undergoing revision at the present time.

sections, respectively, with specified numbers of data points required in each section before z_g will be estimated at the grid node. A trend analysis ("TREND"), which fits a least squares n^{th} degree ($n \leq 6$) polynomial regression equation ([15], Chapter 6; [16], Chapter 4) to the data in terms of the geographical coordinates x and y, can also be used on SURFACE II.

Finally, a moving average method known as "universal Kriging" is discussed in [9], but will not be operational until the fall of 1975 when a new manual will be issued.[2] Kriging is based upon regionalized variable theory developed by Matheron ([10], [12], [13]). The method requires a structural analysis to estimate the correlation structure between points in the field at various distances apart. One advantage of Kriging over the other moving average methods discussed here is that an estimate of the variance of $\hat{z}_g$ can be computed, although in practice it may only be an approximation [13]. Also, the structural analysis required to use Kriging gives an objective means of estimating the optimum search radius for data.

3. "A SITE" DATA

The three strata established at A Site are shown in Fig. 11 of [3]. Both surface (0-5 cm) and profile (10 separate samples in 2.5 cm increments down to 25 cm) samples were collected within each stratum. A vegetation sample was collected near each surface or profile sample (usually within 5 ft) as described in [2] and are referred to here as paired observations. Using the optimum allocation formula in [5] and taking into account budget constraints, 20 surface soil samples were collected at random locations within strata 1 and 3, and 30 within stratum 2. Stratum 3 is the area immediately surrounding GZ. Most of the uranium in the device was ^{235}U.

3.1 Radioisotope Concentrations

The data for $^{239-240}Pu$, ^{235}U and ^{238}U are summarized in Table I. The average ^{235}U concentrations increase from stratum 1 to stratum 3, reflecting the preponderance of this isotope in the device. As expected, a more modest increase occurs for ^{238}U. The $^{239-240}Pu$ levels are low compared with the nearby B, C, or D sites in Plutonium Valley ([3], Table 16).

The ratio of vegetation mean to soil mean for ^{235}U and ^{238}U (Table I) decreases with decreasing distance from GZ. This was also reported in [2] for $^{239-240}Pu$ at the other safety-shot sites who attributed it to a differential particle size distribution occurring within the fallout patterns of the safety-shots. The average concentrations of ^{234}U and ^{236}U for vegetation (no results are available for soil) are about 100 times less than the vegetation means for ^{235}U and ^{238}U, and increase moderately for strata near GZ. The (mean, S.E., n) for vegetation ^{234}U in strata 1, 2, and 3 are (0.093, 0.027, 7), (0.55, 0.14, 20), and (0.82, 0.17, 6), respectively, in units of ng/g dry. These results for ^{236}U are (0.025, 0.0082, 7), (0.20, 0.05, 20), and (0.31, 0.065, 6).

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[2] Personal communication with J. C. Davis, Kansas Geological Survey, University of Kansas, Lawrence, Kansas.

3.2 Inventory

Table II gives the estimated inventories of ^{235}U, ^{238}U, and $^{239-240}Pu$ in surface soil at A Site. The sizes A_h of the 3 strata used to compute these inventories are $A_1 = 125,592$, $A_2 = 7,714$ and $A_3 = 475$ square meters. Sufficient data is not presently available to accurately estimate the inventory of ^{235}U or ^{238}U contributed by the device as opposed to that occurring naturally. The fact that most uranium in the device was ^{235}U is reflected in the results of Table II.

The efficiency of stratified random sampling in estimating I_s relative to that obtained by taking samples at random locations over the entire area can be measured by the ratio

$$R = \frac{\text{S.E. using simple random sampling (no strata)}}{\text{S.E. using stratified random sampling}},$$

where the numerator S.E. is approximated using Eq. 5A.27 in [5]. For ^{235}U the value of R was 8.6, i.e., if simple random sampling (no strata) had been used to estimate the inventory of ^{235}U, the S.E. of that estimate would have been about 8.6 times larger than the S.E. reported for ^{235}U in Table II. For ^{238}U the value of R was a more modest 1.3 resulting, basically, from smaller differences in stratum average concentrations. The ratio R for $^{239-240}Pu$ was only 0.82 since the strata boundries do not divide the area into different plutonium concentration zones. We note, however, from [3] that R for plutonium at the other study sites ranged from 1 to 6 with a median of 3.

3.3 Profile Samples and Soil Vegetation Comparisons

Four profile samples from A Site are currently available for study (Fig. 1). For two of these profiles the decrease in concentrations with depth can be described by the sum of two exponential terms, the parameter estimates being statistically significant at $\alpha = .05$ or less. The first term describes the rapid decrease in the first 5 cm, while the second term fits the much slower rate of decline at greater depth. There is a great deal of variability between profile samples at different locations ([3] gives $^{239\ 240}Pu$ concentrations for profiles at other safety-shot sites) arising perhaps from mechanical disturbance by rodents or man, or from the occurrance of different soil types at depth over the area. Figure 1 shows that except possibly for the profile 53 feet from GZ, ^{238}U surface concentrations are not greater than those at depth, due presumably to the fact that most ^{238}U present is natural uranium.

The relationship between uranium concentrations in paired soil and vegetation samples is examined in Fig. 2 for ^{235}U. The estimated linear correlation is 0.93 for ^{235}U and 0.35 for ^{238}U. The higher correlation for ^{235}U could be related to the preponderance of this isotope in the device. Three models have been fit to the data in Fig. 2. Two of these are weighted linear expressions through the origin, i.e., $y = \beta_1 x$ and $y = \beta_2 x$, ([14], p. 167) where

$$\hat{\beta}_1 = \sum_{i=1}^{n} y_i \Big/ \sum_{i=1}^{n} x_i \qquad \text{and} \qquad \hat{\beta}_2 = \left(\sum_{i=1}^{n} y_i/x_i \right) /n \ ,$$

and the third is the usual unweighted linear regression model. Model 3 gives the best fit to these ^{235}U data and to the ^{238}U data as well (not shown). The poor fits using the commonly used ratio estimates $\hat{\beta}_1$ and $\hat{\beta}_2$ suggest that calculation of such ratios without first plotting the data and checking for adequate fits can give misleading results. Equally poor fits using $\hat{\beta}_1$ and $\hat{\beta}_2$ were also reported in [3] for $^{239-240}Pu$ concentrations. Model 3 was also fit to the logs of the data but the resulting fits were poor (correlations of 0.77 and 0.11 for ^{235}U and ^{238}U, respectively).

4. ESTIMATION OF PLUTONIUM CONCENTRATION CONTOURS

Estimates of $^{239-240}Pu$ concentration contours in surface soil have been obtained here using NEAR, QUAD, and TREND. Their relative performance is evaluated by computing two linear correlation coefficients: (i) ρ, the correlation between observed and estimated $^{239-240}Pu$ concentrations z_i and $\hat{z}_i$, respectively, at N soil sample locations[3] (i = 1, 2, ..., N), and (ii) ρ_B, the correlation between z_i and the residual $z_i - \hat{z}_i$. Ideally, ρ should be near 1 and ρ_B near 0. ρ measures the goodness-of-fit of $\hat{z}_i$ to z_i in that if $\rho = 1$, then $\hat{z}_i = z_i$ for all i = 1, 2, ..., N. ρ_B is a measure of an estimation bias found in [3] to be present when NEAR was used on soil and vegetation data of Project 57 and the GMX site. For this particular kind of bias, large positive residuals $z_i - \hat{z}_i$ tend to be associated with large z_i, and small positive (or negative) residuals with small z_i. When this effect is present, ρ_B tends to be greater than zero. As discussed in [3], the result of this bias is to exaggerate the extent of the estimated dispersion of plutonium away from the GZ area. Other kinds of bias can be present even when $\rho_B = 0$, so that the computation of ρ_B must be supplemented by plots of the residuals $z_i - \hat{z}_i$ versus the observed z_i.

Values of ρ and ρ_B obtained using NEAR, QUAD, and TREND are given in Table III. Cal-Comp plots of the estimated plutonium contours for analyses A1 through A4 are given in [3]. Analyses A6 and A10 are shown in Fig. 3.[4] QUAD and NEAR do about equally well in predicting observed Pu concentrations, considerably better than TREND, and rather impressive gains in goodness-of-fit can be achieved by working in $\log_e$ units rather than in the original scale for all three methods. TREND also tends to give wild estimates in the boundries of the study site (Fig. 3).

The smaller number of samples for QUAD relative to that for NEAR or TREND (Table III) occurs because a large proportion of grid node estimates $\hat{z}_g$ could not be made due to insufficient data points within the prescribed

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[3] The $\hat{z}_i$ are computed by SURFACE II by backward double linear interpolation from the estimated grid values $\hat{z}_g$. Note that $\hat{z}_g$ refers to Pu estimates at grid nodes, while the $\hat{z}_i$ are estimates at the random sample locations.

[4] The grid matrices A6, A8, A10, A11, and A13 were estimated on the transformed values $z' = \log_e z$ and contours plotted for specified values of z' such that $e^{z'} = 0.5, 1.0, 1.5, \ldots,$ etc., nCi/g.

distance from the grid node (footnote d, Table III). Since SURFACE II will print out the estimated grid matrix, the user can tell when insufficient data exists to meet the search criteria specified. R^2 (square of the multiple correlation coefficient) which gives the percent of the total variability of the observed plutonium values z_i explained by the estimated regression equation, is considerably larger when logarithms rather than untransformed data are used (Table III).

5. DISCUSSION

Stratified random sampling is more efficient than simple random sampling for estimating inventory at the safety-shot sites. Its effectiveness in other transuranic field studies in desert as well as less xeric climates for estimating a total or mean depends on whether strata can be constructed for which strata means vary widely and within-stratum variances are small relative to total variability. Stratified random sampling in conjunction with an appropriate algorithm such as QUAD, can also achieve reasonably good fits to observed data particularly when the data are transformed to logarithms. Further improvements in estimating contours will depend on the development of optimum computing algorithms (Kriging ?) and better sampling plans.

One approach that might be investigated is the use of systematic sampling (perhaps on a grid) rather than random sampling within strata. Biased estimates might result, however, and estimating variances becomes more difficult (Chapter 8 in [5]). "Double sampling" ([5], Chapter 12) should also be considered as a method of increasing the efficiency of field sampling. The results in [4] are encouraging in this regard. There is a clear need for using inexpensive field detection instruments to gather information on the distribution of radionuclides before full scale sampling begins.

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TABLE I. ESTIMATED[a] CONCENTRATIONS OF $^{239-240}Pu$, ^{235}U, AND ^{238}U IN RANDOM SOIL AND ASSOCIATED VEGETATION SAMPLES FROM A SITE OF AREA 11, NTS

Isotope	Strata	Soil n	Soil Mean ± S.E.[b]	Soil Range	Vegetation n	Vegetation Mean ± S.E.	Vegetation Range		Veg. Mean / Soil Mean
$^{239-240}Pu$ (pCi/g dry)	1	12	5.92 ± 2.16	0.96 - 25.8	12	1.5 ± 0.57	0.056	7.3	0.25
	2	14	1.46 ± 0.335	0.12 - 4.1	18	0.64 ± 0.099	0.23	1.9	0.44
	3	9	32.4 ± 31.4	0.10 - 284	6	1.0 ± 0.38	0.13	2.8	0.032
	Total	35	11.0 ± 8.07	0.10 - 284	36	0.98 ± 0.21	0.13	7.3	0.089
^{235}U (ng/g dry)	1	14	30.0 ± 13.9	2.4 - 210	12	4.1 ± 1.3	0.795	13.1	0.14
	2	14	1,090 ± 454	52 - 5,500	20	49 ± 12	2.45	178	0.045
	3	10	17,200 ± 7,090	270 - 67,000	6	74 ± 16	37.8	135	0.0043
^{238}U (ng/g dry)	1	14	1,670 ± 198	155 - 2,630	12	41 ± 5.7	8.57	71.3	0.025
	2	14	2,120 ± 206	1,410 - 4,160	20	72 ± 8.8	18.2	158	0.034
	3	10	11,700 ± 3,610	1,820 - 36,300	6	81 ± 11	35.7	114	0.0069

[a] Preliminary estimates; 50% of results have been reported. [b] S.E. = standard error = $s/\sqrt{n}$.

TABLE II. ESTIMATED[a] INVENTORY OF ^{235}U, ^{238}U, AND $^{239-240}Pu$ IN SURFACE SOIL (0-5 cm DEPTH) AT A SITE OF AREA 11, NTS

Isotope	Strata	n	Natural Uranium + Safety-Shot Uranium: Mean ± S.E.	Estimated Inventory ± S.E.	Percent Inventory in Strata	Estimated 95% Confidence Limits On Inventory: Lower	Upper	c[b] of Inventory Estimate
			(grams/m^2)	(grams)		(grams)		
^{235}U	1	14	0.00142 ± 0.000698	178 ± 88	17	-12	368	1.8
	2	14	0.0434 ± 0.0168	335 ± 130	32	54	616	1.5
	3	10	1.12 ± 0.473	532 ± 225	51	24	1,040	1.3
	Total	38		1,045 ± 274	100	457	1,630	1.6
^{238}U	1	14	0.0754 ± 0.00910	9,470 ± 1,140	90	7,000	11,900	0.45
	2	14	0.0952 ± 0.00845	734 ± 65	7	594	874	0.33
	3	10	0.725 ± 0.249	344 ± 118	3	77	611	1.08
	Total	38		10,500 ± 1,150	100	8,020	13,000	0.68
			(μCi/m^2)	(Curies)		(Curies)		
$^{239-240}Pu$	1	12	0.265 ± 0.0955	0.0333 ± 0.0120	96	0.00688	0.0597	1.2
	2	14	0.0674 ± 0.0153	0.00052 ± 0.00012	1.5	0.00026	0.00078	0.86
	3	8	1.77 ± 1.73	0.000841 ± 0.000822	2.4	-0.00110	0.00279	2.8
	Total	34		0.0347 ± 0.012	99.9	0.0083	0.061	2.0

[a] Preliminary estimates; 50% of results are reported. [b] c = coefficient of variation = $[n(S.E.)^2]^{1/2}$/inventory estimate.

TABLE III. CORRELATIONS ρ AND ρ_B OBTAINED IN ESTIMATING $^{239-240}Pu$ CONCENTRATION CONTOURS

Analysis Number	Study Site	Method	Search Limits	Scale	No. of Samples N	Ecosystem Component	Correlation: z_i vs $\hat{z}_i$	Correlation: z_i vs $z_i - \hat{z}_i$	R^2
A1	Area 13	NEAR	None	Original	167	Soil	0.90	0.64	NA
A2	Area 13	TREND[a]	NA[c]	Original	168	Soil	0.58	0.81	0.34
A3	Area 13	NEAR	None	Original	133	Veg	0.93	0.81	NA
A4	GMX	NEAR	None	Original	113	Soil	0.72	0.79	NA
A5	GMX	NEAR	50-500[d]	Original	112	Soil	0.72	0.79	NA
A6	GMX	NEAR	None	Log	113	Soil	0.98	0.27	NA
A7	GMX	QUAD	100-200[d]	Original	82	Soil	0.70	0.81	NA
A8	GMX	QUAD	100-200[d]	Log	82	Soil	0.96	0.33	NA
A9	GMX	TREND[a]	NA	Original	113	Soil	0.54	0.84	0.29
A10	GMX	TREND[a]	NA	Log	113	Soil	0.93	0.36	0.87
A11	GMX	TREND[b]	NA	Log	113	Soil	0.90	0.44	0.81
A12	GMX	NEAR	None	Original	113	Veg	0.92	0.60	NA
A13	GMX	QUAD	100-200[d]	Log	78	Veg	0.98	0.34	NA

[a] 6th degree polynomial. [b] 4th degree polynomial. [c] NA = not applicable.

[d] For A5, maximum distance to nearest sample point must be ≤ 50 ft.; maximum search radius = 500 ft.

^{235}U AND ^{238}U CONCENTRATIONS IN SOIL PROFILES, A SITE, AREA 11

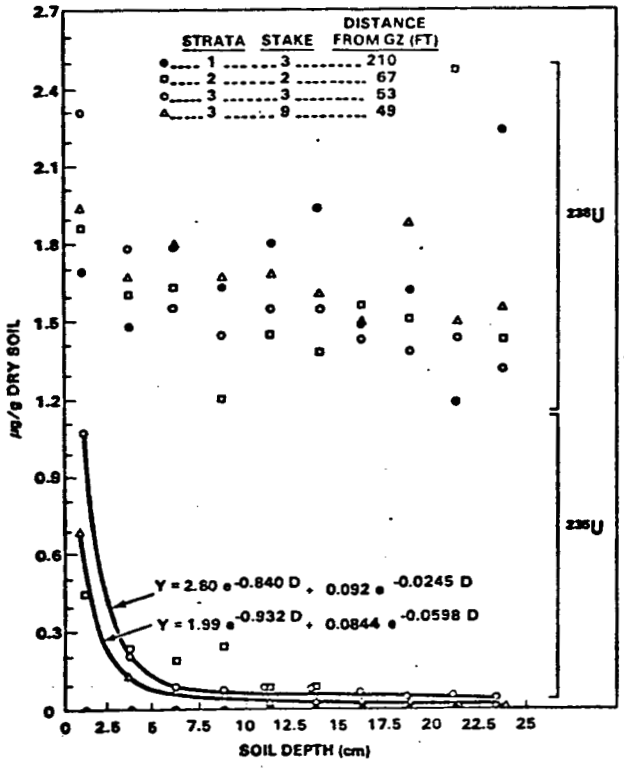

FIGURE 1

VEGETATION VS SOIL
^{235}U (µg/g DRY)

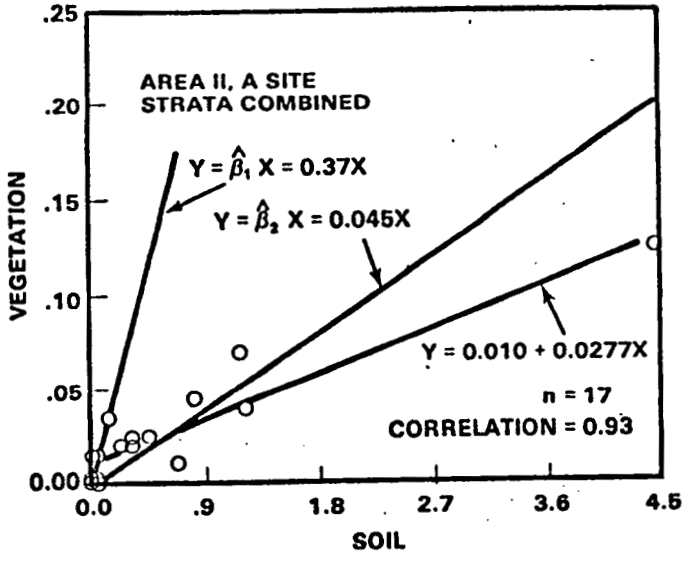

FIGURE 2

ESTIMATED ISOPLETH (CONTOUR) LINES OF $^{239-240}Pu$ (nCi/g) IN SOIL BASED ON RANDOM SAMPLES WITHIN STRATA, AREA 5 (GMX)

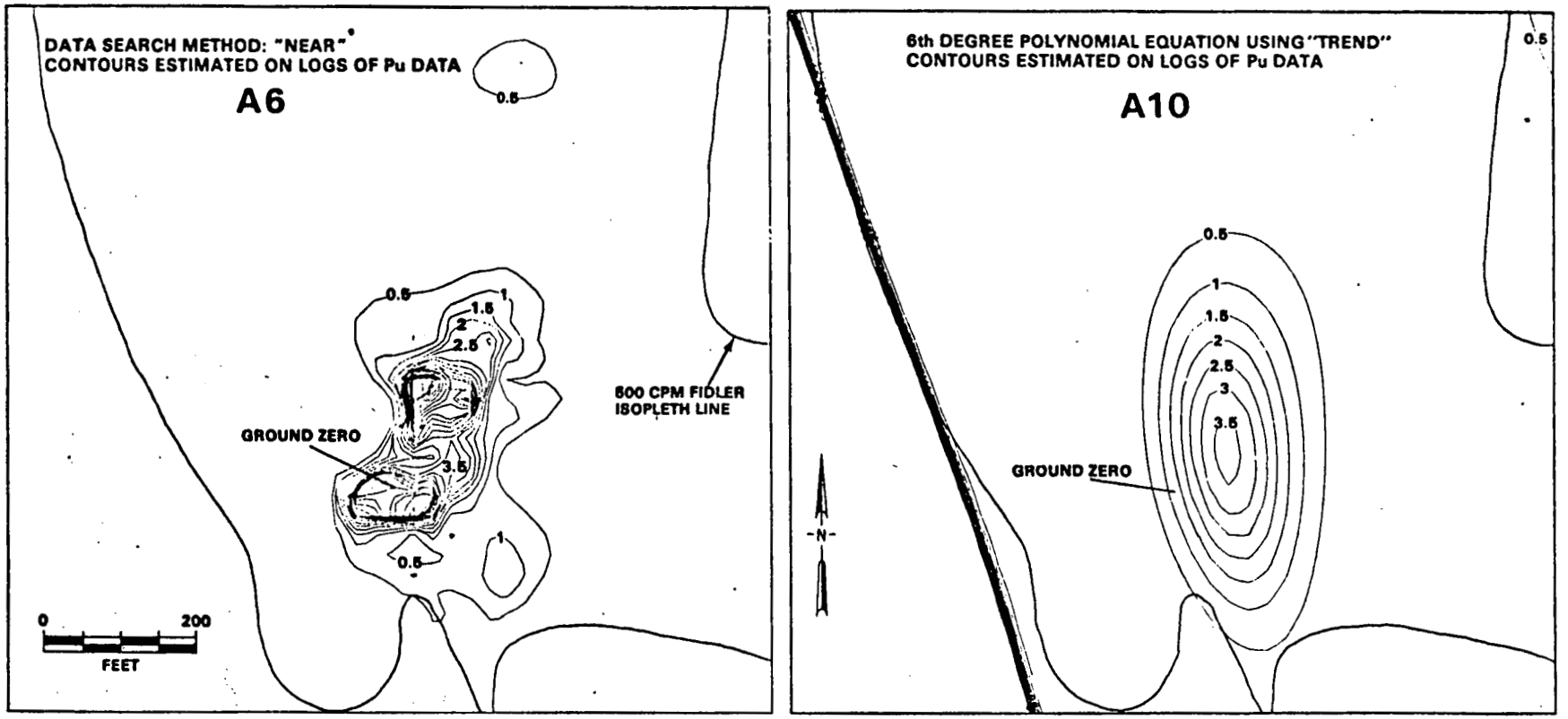

FIGURE 3