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Optimization of Sampling for the Determination of the Mean Radium-226 Concentration in Surface Soil

L. R. Williams
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Health and Safety Research Division

OPTIMIZATION OF SAMPLING FOR THE DETERMINATION OF THE MEAN RADIUM-226 CONCENTRATION IN SURFACE SOIL

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CONTENTS

	PAGE
ACKNOWLEDGEMENT	v
ABSTRACT	vii
INTRODUCTION	1
SUMMARY OF SOIL SAMPLE DATA	4
IDENTIFICATION OF EQUIVALENT SAMPLE SIZES	6
INCREASED ACCURACY AND PRECISION WITH INCREASED SAMPLING	9
USING SURFACE GAMMA MEASUREMENTS TO REDUCE SOIL SAMPLING	10
Predicting the mean Ra concentration from the mean gamma level	*10*
Use of gamma measurements to eliminate soil sampling in some cases	*11*
Use of gamma measurements to estimate the number of soil samples needed	*12*
Use of nearly average gamma levels to locate typical Ra contamination	14
SUMMARY AND CONCLUSIONS	15
Practical levels of accuracy and precision	*15*
Uniform vs. random spacing of soil samples and gamma measurements	*15*
Individual vs. composite samples	*15*
Use of gamma measurements to optimize soil sampling	*16*
Evaluating subsurface contamination	*16*
REFERENCES	19

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OPTIMIZATION OF SAMPLING FOR THE DETERMINATION OF THE MEAN RADIUM-226 CONCENTRATION IN SURFACE SOIL

ABSTRACT

In this report we describe the results of a field experiment designed to help identify an optimal method for determination of compliance with the U. S. Environmental Protection Agency's Ra-226 guidelines for soil. The primary goals of this study were (1) to establish practical levels of accuracy and precision that can be obtained in estimating the mean Ra-226 concentration of surface soil in a small contaminated region; (2) to obtain empirical information on composite vs. individual soil sampling and on random vs. uniformly spaced sampling; and (3) to examine the practicality of using gamma measurements in predicting the average surface radium concentration and in estimating the number of soil samples required to obtain a given level of accuracy and precision. Numerous soil samples were collected on each of six sites known to be contaminated with uranium mill tailings. Three types of samples were collected on each site: 10-composite samples, 20-composite samples, and individual or post hole samples. It was found that 10-composite sampling is the method of choice among these three approaches, in that it yields a given level of accuracy and precision for the least cost. It was also found that gamma measurements can be used to reduce surface soil sampling on some sites.

I. INTRODUCTION

Large quantities of processed uranium ore residues, called tailings, have been generated in the United States since the early 1940's. These tailings generally have been deposited in ponds or in huge stockpiles and allowed to dry. In some cases deposits were left unprotected for several years, allowing the tailings to be scattered by the wind over the surrounding area or to be used by nearby residents as sand substitutes or as backfill material.

In 1978, the U. S. Congress passed Public Law 95-604, the Uranium Mill Tailings Radiation Control Act (UMTRCA), requiring the federal government to perform remedial actions on inactive uranium mill tailings sites that had been federally supported and on vicinity properties that had been contaminated by the tailings. The U. S. Department of Energy (DOE) was assigned the responsibility for conducting remedial action at 24 sites, located primarily in the western United States. This assignment is being met through DOE's Uranium Mill Tailings Remedial Action Project Office (UMTRA-PO) in Albuquerque, New Mexico. The Radiological Survey Activities (RASA) group of Oak Ridge National Laboratory serves as an Inclusion Survey Contractor (ISC) for the UMTRA-PO and conducts radiological surveys of potentially contaminated properties in or near Grand Junction, Colorado, an area requiring perhaps the greatest amount of remedial action.

A primary responsibility of the UMTRA-PO is to certify that potentially affected properties are in compliance with current standards of the Environmental Protection Agency (EPA) for contamination of soil with Ra-226 (40 CFR Part 192)[1]. These standards require that the average Ra-226 concentration over a 100-m^2 area not exceed 5 pCi/g above background in the top 15 centimeters of soil or 15 pCi/g above background in 15-cm-thick layers more than 15 cm below the surface. Because of the huge number of properties involved, each property must be screened against the EPA standards as economically as possible. Thus, it is essential that survey procedures be designed to yield optimal information from a limited number of soil samples and field measurements.

In this report we describe the results of a field experiment designed to help identify the optimal method for sample collection within a radium-contaminated area. The goals are to find practical levels of accuracy and precision that can be obtained in estimating the mean radium concentration of a given deposit and to design a sampling protocol which adequately characterizes a deposit within certain constraints. (For the purposes of this study we define accuracy as the closeness of a measured or computed value to its true value and precision as the closeness of repeated measurements of the same quantity.[2])

The primary constraints are the time and cost of sampling and the feasibility of a method for a given deposit.

We have investigated the relative merits of three types of soil samples, distinguished by the method of collection. These are referred to as 10-composite samples, 20-composite samples, and individual or post hole samples. A 10-composite sample consists of 10 aliquots of soil weighing ~50 g each and taken at approximately uniformly spaced points over the deposit. A 20-composite sample consists of 20 aliquots weighing ~25 g each, collected at approximately uniformly spaced points over the deposit. An individual sample weighs ~500 g and is collected from an area of several hundred square centimeters. As a matter of practicality, all of the soil samples collected were taken from the top 15 cm of soil.

We have also examined the possibility of using surface gamma measurements to reduce the amount of soil sampling needed. In particular, we investigated the potential use of gamma measurements in predicting the average surface radium concentration and in estimating the number of soil samples required to obtain a given level of accuracy and precision. Gamma measurements were made at the ground surface at individual (post hole) sampling locations before the samples were taken and at the bottom of the holes after individual samples were removed.

The 20-composite, 10-composite, and individual samples were taken so that sampling points for each sample type encompassed each deposit in a fairly uniform manner (see Fig. 1). (Two exceptions arose for 10-composite samples; these are discussed later.) As nearly as practical, all samples were taken uniformly at depths from 0 to 15 centimeters. The composite samples were taken using a method referred to as the TAC drill method (Fig. 2), and the individual samples were taken with a post hole digger. The TAC drill method consists of a heavy duty carpentry drill with a long bit attached. A collection box with a bit-sized hole in one corner is used to contain the soil. The drill bit is put into the hole in the box and soil is pulled slowly up by the bit, through the hole and into the box (Fig. 3) The name TAC is an acronym for the Technical Assistance Contractor for the UMTRA Project, the Jacobs-Weston team, whose employee R. A. Nelson popularized the method.

Samples were collected on six test sites, each having a contiguous deposit of at least 15 square meters. Five of these sites were chosen from known contaminated sites in Grand Junction, Colorado, solely on the basis that extensive soil sampling would be permitted by the owners. The level of radium contamination was not considered in our selection process, but it was known from previous limited measurements that at least two of these sites could be regarded as marginally contaminated (that is, contamination was near guideline levels). The sixth site was

a 100-square-meter area selected on the original source of the contamination, namely, the tailings pile.

II. SUMMARY OF SOIL SAMPLE DATA

Soil sample data for the six sites are summarized in Table 1, which also gives an estimate of the true mean Ra concentration in surface soil on each of the deposits except the tailings pile. The degree of consistency of the three estimates of the mean Ra concentration corresponding to the three different types of sample collection varied somewhat with the site. The variation appeared to depend on the uniformity of contamination as indicated by the coefficient of variation (CV = standard deviation/mean) of the post hole samples, on the density of the various samples within the sampled area, and on whether all sampling techniques fully encompassed a deposit.

The three techniques yielded nearly identical estimates of the mean concentration on deposits GJ00762 and GJ12711. These deposits encompassed the smallest areas and had the most uniformly distributed surface contamination of the six sites, and sampling was more dense on these than on the other four sites. For these two deposits, any or all of the methods should yield a good estimate of the true mean Ra concentration. The true mean for deposit GJ12711 was estimated as the average of the means obtained by the three different methods. For deposit GJ00762 the true mean was estimated as the average of the means for the post hole samples and the 20-composite samples. The 10-composite sample mean was not used in this case because a substantial portion of this deposit was not sampled by the 10-composite method. (This differed from the study design, but the discrepancy was realized only after field and laboratory work had been completed.)

For the three largest deposits (GJ00870, GJ11088, GJ13660) the mean concentration for all post hole samples differed from the 20-composite mean by 20 to 30 percent. Some of this difference is to be expected because of the relatively large coefficient of variation of the post hole samples on these deposits (1.0 or greater). When the mean and standard deviation are approximately equal, 50% of the averages computed from samples of size 40, say, should differ from the true mean by at least 10%, and more than 20% should differ by at least 20%. We chose to use an unweighted average of means determined by the different methods in estimating a true mean concentration for each of these three deposits. In this way we can make comparisons of the three approaches without having a built-in bias toward a particular method. The 10-composite samples for GJ11088 were not used in the estimate of the true mean for that deposit, because a large portion of the deposit was not sampled by the 10-composite method. (Again, the discrepancy between study design and 10-composite sample collection was realized only after the field and laboratory work had been completed.)

For the tailings pile, the means for the two composite techniques are in reasonable agreement but are substantially lower than the mean for the post hole samples. The disagreement between composite and post hole means could have arisen from a difficulty with using a drill in unmixed, dry, sandy soil--the material tends to crumble and fall around the bit to some extent, perhaps resulting in a bias toward more shallow soil in the sample. If the top few centimeters on the sampled area of the tailings pile were substantially cleaner than deeper material, then post hole samples might yield a much higher estimate of the mean concentration than would composite samples collected with a drill. However, because we have no real way of knowing which of the sampling methods was most reliable for the tailings pile, and because the mean estimated from post hole samples was substantially different from the other two means, no attempt was made to estimate a true mean concentration for this site.

III. IDENTIFICATION OF EQUIVALENT SAMPLE SIZES

We compared the levels of accuracy and precision one can expect from a single 20-composite sample, a single 10-composite sample, and various numbers of post hole samples chosen (1) at random with replacement, (2) at random without replacement, or (3) uniformly spaced in the sense that a map of the deposit would be divided into some number N of regions of nearly equal area and one sample would be chosen at random from each of these regions. Random sampling with replacement (as carried out on a computer) has no precise analogy in the field but has some similarities to the situation in which sampling locations are chosen at random and no minimum distance is required between post holes. This would not be the same in the present study as random sampling without replacement, because on each site there was an effective minimum distance between post holes determined by the size of the site and the number of post holes.

The analysis in this section was made without regard to the gamma levels at the ground surface. In a later section we discuss the plausibility of using surface gamma measurements to reduce the amount of soil sampling needed.

The relative accuracies and/or precisions of different types of measurement will vary somewhat from site to site, and we cannot expect to find an equivalence rule without exceptions. However, the data indicate that the mean Ra concentration estimated from a single 10-composite sample taken at uniformly spaced points encompassing the deposit is about as reliable as the mean estimated from 10 random post hole samples with no duplication of sampling locations (sampling without replacement), or 13-15 random post holes with duplication allowed (sampling with replacement), or 8 post holes with each selected randomly from one of 8 subregions of approximately equal area covering the entire deposit. The approximate equivalence of these four approaches is indicated by the data in Tables 2 and 3. For example, from Table 2 it can be seen that the four approaches yield roughly the same levels of accuracy as measured by the portion of estimates of the mean that fall within a certain interval about the true mean for a site. From Table 3 it can be seen that the precision, as indicated by the standard deviation divided by the predicted mean, is also about the same for all four approaches.

The data for the six sites do not allow firm statistical statements concerning differences in accuracy and/or precision between the 10- and 20-composite sampling methods. For example, the values in Tables 2 and 3 do not indicate any significant differences between the two methods, although a greater precision of the 20-composite method, as an average, is suggested. As discussed in the next section, a detailed examination

of the data suggests that about equal numbers of 10- or 20-composite samples are needed to yield a relatively high degree of accuracy with a given confidence level (say, estimating within 10% of the true mean with a confidence level of 90%), but 20-composite samples are superior if less accuracy is required (say, estimating within 30% of the true mean with a confidence level of 90%). In this analysis the problem arose that the relatively small standard deviations for the 10-composite samples on two of the sites (GJ00762 and GJ11088) may be somewhat misleading when compared with those for the 20-composites. This is because some fairly large regions on each of these two deposits were not sampled by the 10-composite method, which may have led to slightly smaller variations than if the entire deposit had been encompassed, as it was in all cases for the 20-composite samples. Thus, it could be that there is slightly more advantage in the 20-composite method than is indicated by our analysis.

On a theoretical basis, one might expect 20-composite samples to yield a better estimate of the true mean concentration and to have a standard deviation that is roughly 30% smaller than that for 10-composite samples. It may be, however, that the larger plugs taken by the 10-composite method have some practical advantages over the 20-composite plugs. In particular, it may be that the 20-composite sampler is too small to produce a fairly uniform plug in some types of soil. Also, a single 50-g plug should give a better indication of the local condition than a single 25-g plug, just as a single post hole sample gives a better indication of local contamination than does a single 50-g plug. (Recall that 8 uniformly spaced post holes are roughly equivalent to a 10-composite sample over the same region.) Further comparisons of the 10- and 20-composite methods are made in the following section, where we examine the degree to which accuracy and precision are increased as the number of samples is increased.

All in all, we think that the 20-composite method would be slightly preferable to the 10-composite method if there were little additional time or expense involved with the former. However, we found that one 20-composite sample requires approximately twice as much field time as one 10-composite sample. Considering both collection and analysis of samples, we found that a 20-composite sample costs approximately 1.9 times as much as a 10-composite sample. It is evident that a 20-composite sample is not sufficiently superior to a 10-composite sample to warrant this much additional time and expense.

It requires about 8 uniformly spaced post hole samples to gain the accuracy yielded by a single 10-composite sample. Since collection and analysis of 8 post hole samples would cost about 3.5 times as much as a single 10-composite sample, the 10-composite sampling method would

appear to be the most economical and efficient for our purposes.

The problem arises that the TAC drill method is not particularly well suited to certain types of soil. For example, some problems are presented when drilling in rocky soil or in extremely loose soil. We think that the TAC drill method should be supplemented with another method of collecting 50-g aliquots of soil that are uniform with depth in types of soil for which the TAC drill method is not suited.

IV. INCREASED ACCURACY AND PRECISION WITH INCREASED SAMPLING

Accuracy and precision gained by increasing the sample size can be evaluated using the data in Table 4. We have restricted attention in this table to composite samples; general conclusions for post hole samples can be made using the equivalence rules discussed in the preceding section. It was assumed for this table that randomly selected 10- or 20-composite sample concentrations follow a normal distribution with mean equal to the true mean concentration for the region sampled and with standard deviation equal to the observed standard deviation for that region. Chi-square goodness-of-fit tests indicate that the assumption of normality is a reasonable one for the composite samples.

In the following discussion we will focus on typical values for the precision over all sites, for fixed levels of accuracy. As indicated by the ranges shown in Table 4, precision for either composite sampling method can vary markedly from one site to another, depending on the distribution of contamination. In a later section we discuss the use of the standard deviation of gamma measurements to help narrow in on the proper number of composite soil samples for a deposit for a given level of accuracy and precision.

It appears from Table 4 that it may not be practical to set as a goal an estimate that is within 10% of the true mean if a high level of confidence is desired, even in the case of composite sampling. For example, the average of as many as five 10-composite samples might be within 10% of the true mean only about two-thirds of the time.

It is considerably easier to obtain a high level of confidence if an accuracy of only 20% or 30% is required. For example, it appears that a single 10-composite sample would be within 30% of the true mean about three-fourths of the time, and the average of two 10-composite samples would be within 20% of the true mean about three-fourths of the time. It may require only 2 composite samples of either type to estimate within 30% of the mean about 90% of the time and 3 samples to estimate within 30% about 95% of the time.

V. USING SURFACE GAMMA MEASUREMENTS TO REDUCE SOIL SAMPLING

Predicting the mean Ra concentration from the mean gamma level

We investigated the relationship between the Ra-226 concentration, y, in post hole samples and the gamma level, x, measured at the surface before the sample was removed (Fig. 4). The concentration y can be expressed as

$$y = A + Bx + E ,$$

where A and B are constants determined by a best fit to the data and E is a random error term. The usefulness of the gamma level x in predicting the Ra concentration y depends on the variability of the random error term E and whether the assumed linear model is appropriate. For a single gamma measurement x_i, the associated random error E_i can be quite large, so that the gamma level frequently is not a good indicator of the Ra concentration at a specific location (see Fig. 5). On the other hand, a similar equation (with the same values of A and B) can be used to predict the average surface Ra concentration y over an entire deposit from a carefully determined average x of surface gamma levels. This is because the use of the average gamma level reduces the variance of the random error E considerably (ideally by as much as the inverse of the sample size).

Data for the tailings pile were excluded from the analysis because the high "background" gamma level for that site would lead to a predictive equation (more specifically, a Ra-intercept) that would not be representative for deposits with uncontaminated surroundings. For the other five sites there is a high correlation between the surface gamma level and the post hole Ra concentration. The best-fitting line changed only moderately from one site to another, and R^2 ranged between 0.65 and 0.90. Recall that the correlation coefficient R for a set of pairs (x_i, y_i) is a measure of the linear relationship between x and y, and that a high value of R does not indicate that every pair (x_i, y_i) is related in some predictable fashion. With a value of R=0.90, say, 19% = $(1.0-0.9^2)$x100% of the total variation in y is unrelated to x.

By combining the data from all five sites and fitting to those points for which the gamma level did not exceed 30,000 cpm (because almost all of the higher values are from a single site and also because a small number of apparent anomalies at high levels influence the slope and intercept somewhat) we obtained the linear regression equation

$$\text{Radium concentration (pCi/g)} = -16.1 + 2.75 \text{ Gamma (1000 cpm)}, \quad \text{(Eq. 1)}$$

with an R^2 of 0.77. Hence about three-fourths of the variation in Ra concentration is "explained" by the observed variation in gamma values.

This regression line and the individual pairs (Gamma,Ra-226) for all gamma levels and all sites excluding the tailings pile are shown in Fig. 4. To eliminate meter dependency in future applications of this equation, we have taken the calibration for the instruments used in this field study to convert the equation to units of μR/hr:

$$\text{Radium concentration (pCi/g)} = -21.7 + 1.63 \text{ Gamma } (\mu\text{R/hr}). \quad \text{(Eq. 2)}$$

Substitution of the average gamma level for a deposit into Eq. 2 (or Eq. 1) yielded close estimates of the true mean Ra concentration on each of the sites excluding the tailings pile (see Table 5). The maximum observed error is 20%. Thus, it would appear that the average gamma level is a good indicator of the average Ra concentration in surface soil; however, this should be further verified with additional measurements. Moreover, the possibility of a small amount of upward concavity in the relation between measured gamma and radium, stemming perhaps from a slight "saturation" effect in the gamma scintillator, should be examined through further measurements (see Fig. 4). If such concavity exists, the prediction equation should be modified accordingly.

Use of gamma measurements to eliminate soil sampling in some cases

It may be possible in some cases to conclude on the basis of gamma measurements alone whether surface Ra-226 contamination on a site is above or below guideline levels. (Of course, investigation of subsurface contamination may still be in order even if the top 15 cm of soil is below guideline levels.) In the following we discuss how gamma measurements might be used to reach such conclusions. In this discussion we are suggesting the use of conservatively high confidence levels to allow for potential inaccuracies in assumptions made in estimating the standard error of prediction when using an average of gamma values to predict an average Ra concentration.

The surveyors should first mark off a one-meter grid on a map of the site and then determine the location and area of the contaminated region using surface gamma measurements. The number N of grid points which fall in the contaminated region will be a reasonable estimate of the area of the region. (If N > 100 then replace N with 100 in the following discussion.) Surface gamma measurements should be recorded in 1000 cpm at each grid point in the contaminated region. The mean, m_g, and standard deviation, s_g, of the surface gamma measurements in 1000 cpm at grid points in the contaminated region can then be calculated.

If BG is the background value for the Ra concentration, then the surface Ra concentration will be above the current guideline level of 5 pCi/g provided it is greater than 500/N + BG. If s_p denotes the

standard error of prediction, then we can be 99% confident that the true average surface Ra concentration does not differ from the predicted average surface Ra concentration by more than $2.576 \times s_p$. (A 99% confidence interval is too stringent to set as a goal if we are estimating the average Ra concentration directly from soil samples. We have used 99% in this case for conservatism, because we doubt that the actual precision in estimating average Ra indirectly from gamma measurements is quite as great as our current limited data for our small number of sites would indicate.) Table 5 gives our calculated 99% confidence intervals for the average Ra concentration, based on average gamma levels, for the five sites. The true mean Ra concentration on site GJ13660 does not fall within the predicted 99% confidence interval, but 30% of the gamma measurements on this site exceeded 30,000 cpm, a level above which the regression equation is not considered valid.

If the estimate of the average Ra concentration minus $2.576 \times s_p$ exceeds $500/N + BG$, the site should be designated as being above the guideline level. Similarly, if the estimated average Ra concentration plus $2.576 \times s_p$ is less than $500/N + BG$, the surface Ra concentration can be considered to be below the guideline value.

Using the regression analysis for gamma vs. Ra and a correlation analysis for predicted Ra concentrations at adjacent post hole locations, we estimate that s_p does not exceed the square root of $1.12+142.7/N$, where N is the number of gamma measurements in the contaminated region. (Recall that N is also the area of the contaminated region in square meters.) Thus, a site should be considered above the guideline level for surface contamination provided

$$2.75 \times m_g - 16.1 - 2.576(1.12+142.7/N)^{1/2} \geq 500/N + BG$$

or

$$m_g \geq \{(500/N)+BG+16.1+2.576(1.12+142.7/N)^{1/2}\}/2.75 ,$$

where m_g is measured in 1000 cpm using the instruments and calibration factors of this study. As an example, with BG=2.0 pCi/g and a contaminated region of 100 m^2, the above inequality reduces to

$$m_g \geq 9.9 \ (\times 1000) \text{ cpm}$$

or, equivalently,

$$m_g \geq 20.2 \ \mu R/hr.$$

Use of gamma measurements to estimate the number of soil samples needed

For many deposits, the average gamma level will probably be sufficiently high to indicate that the site contamination is above guideline values or sufficiently low to indicate that *surface soil* is below

guideline values without soil sampling (and with a high degree of confidence). However, there will be sites with marginal average gamma levels on which soil sampling is needed to estimate the surface Ra concentration with sufficient accuracy. Even on a site with an average surface soil concentration below the guideline level, subsurface sampling may still be in order to decide whether remedial action is needed.

The mean and standard deviation of uniformly spaced gamma measurements can be used to estimate an optimum number of soil samples. Suppose that we want to be 90% confident that our estimate of the mean Ra concentration is within a fraction F of the true mean (for example, F=0.2 means within 20% of the true mean). Since the composite samples will be approximately normally distributed, 90% of the sample means from random samples of size K will be within F of the true mean provided

$$K > [(1.645 \times s)/(F \times m)]^2$$

where s is the population standard deviation and m is the true mean. Neither s nor m is known *a priori* and hence both must be estimated. From the regression of post hole Ra concentrations on surface gamma readings we find that the standard deviation of the Ra concentrations is approximately 1.63 times the standard deviation of gamma readings in μR/hr. Also, a rough estimate of composite sample standard deviation can be obtained by multiplying post hole sample standard deviation by 0.25. Thus, if m_g and s_g denote the mean and standard deviation of the surface gamma readings, we obtain the following estimate of the smallest number (K) of composite samples needed to be 90% confident that our Ra estimate is within a fraction F of the true mean:

$$K = \{[1.645 \times 1.63 \times 0.25\ s_g]/[F \times (-21.7 + 1.63\ m_g)]\}^2$$
$$= \{(0.67 \times s_g)/[F \times (-21.7 + 1.63\ m_g)]\}^2. \qquad \text{(Eq. 3)}$$

If the mean and standard deviation of the gamma measurements are given in terms of 1000 cpm using the current calibration, Eq. 3 would be replaced by

$$K = \{(1.13 \times s_g)/[F \times (-16.1 + 2.75\ m_g)]\}^2. \qquad \text{(Eq. 4)}$$

Table 6 compares the resulting estimates of the number of samples needed on the sites in this study (excluding the tailings pile) with the number actually needed to be 90% confident that the error is within 20% of the true mean. The large predicted values for site GJ00870 are due to the inclusion of gamma measurements over a large region of uncontaminated soil. If attention is restricted to areas that are clearly contaminated on this site, the number of 10- or 20-composite samples needed would have been estimated as five. On the other four sites, the

predicted number of composite samples needed ranges from two to five. Perhaps five 10-composite samples should be considered as a practical upper bound, even if a higher number is indicated by the above formula.

Use of nearly average gamma levels to locate typical Ra contamination

The method currently used by the RASA group to reduce soil sampling is that of analyzing two post hole samples taken at points where the gamma level at the surface is judged by the surveyors to be roughly average for the deposit. In order to examine the reliability of this method, we considered radium concentrations at all points on each site showing "nearly average" gamma levels, which we defined to be within 1500 cpm of the true average gamma level as determined from a large number of uniformly spaced measurements. This corresponded to a maximum error in the estimate of the true mean gamma level that ranged from 7% to 18%, depending on the site.

As shown in Table 7, relatively few of the soil samples taken at points showing nearly average gamma levels had Ra concentrations within 20% of the true mean concentration. An exception was site GJ12711, on which two-thirds of the soil samples at nearly average gamma levels were within 20% of the true mean. At the other extreme, on site GJ00870 only 1 of 12 soil samples at nearly average gamma levels showed a concentration within 20% of the true mean, the other 11 samples showed Ra concentrations ranging from 0.25 to 0.77 times the true mean. The method is not necessarily improved a great deal by increasing the number of samples collected at nearly average gamma levels. For example, for site GJ00870, 96% of the averages of 5 samples selected randomly (without replacement) from nearly average gamma locations would be at least 30% below the true mean and 60% would be at least 40% below.

VI. SUMMARY AND CONCLUSIONS

Practical levels of accuracy and precision

It may not be practical to set as a goal an accuracy as high as 10%. Perhaps an accuracy of 20% with a confidence level of 90% would not be unreasonable for surface soil, particularly if conclusions can be drawn for many sites from the average gamma level.

Uniform vs. random spacing of soil samples and gamma measurements

In the collection of soil samples or gamma measurements, approximately uniform spacing is preferable to totally random selection of sampling locations. It would not be worthwhile, however, to take the time to lay off an exact grid. It should suffice for surveyors to make rough approximations of distances, perhaps by stepping off distances from a few well established reference points.

Individual vs. composite samples

It appears that fairly high levels of accuracy and precision can be achieved much more efficiently using composite samples rather than individual samples. Moreover, 10-composite samples appeared to be superior to 20-composite samples when cost-effectiveness was considered.

A problem arises that some types of soil cannot be sampled properly with the TAC drill method. This drill method should be supplemented with another composite sampling method that would be effective in extracting approximately 50 g of soil, uniformly to a depth of 15 cm, from any region of a deposit not suitable for the TAC drill. For example, if a 10-m^2 deposit had an area of 5 m^2 that could not (or should not) be tested with the TAC drill, then five samples gathered by other methods (with each reduced to 50 g of soil if greater amounts were taken) might be combined with five 50-g plugs gathered with the TAC drill to form a single 10-composite sample.

For the purposes of this study it was useful to have each composite sample taken from points uniformly spaced over the whole deposit. For an actual field survey the collection of composite samples can be done more efficiently. If a given number, K, of composite samples are to be taken from the surface soil on a site, the contaminated region should be divided into K regions of approximately equal area and one composite sample should be taken from each of the K regions. The surveyor will save steps and may be better able to judge approximately uniform spacing over the smaller area than over the whole deposit.

Use of gamma measurements to optimize soil sampling

A carefully determined average gamma level at the surface over an entire deposit can be used, together with a regression equation based on a history of pairs (gamma,Ra-226), to obtain a reasonable estimate of the mean Ra concentration in the top 15 cm of soil in the deposit. This could help reduce soil sampling substantially. Once overall goals for accuracy and precision have been chosen, we could determine site-specific values $G_1<G_2$ such that an average gamma level less than G_1 would indicate that the surface Ra concentration falls below guideline levels and an average gamma level greater than G_2 would indicate that remedial action is in order. If the average gamma level is between G_1 and G_2, the determination of the mean surface Ra concentration would be made by soil sampling and analysis. The values G_1 and G_2 will be a function of the desired accuracy and precision, the background Ra concentration, and the area of the deposit.

The present Ra vs. gamma regression line should be improved by collection of several pairs of points (x,y), where x is the average gamma for points at which a composite sample was taken and y is the radium concentration in that composite sample. In the future, surface gamma readings should be recorded at each point from which a 10-composite plug is taken. A regression equation of 10-composite Ra concentration against average gamma (taken at the 10 points) with the composite samples taken from distinct (non-overlapping) regions would yield a much preferred regression equation. With this improved equation, we would not have to make the conservative estimate of the standard error of prediction when using the average gamma level to predict the average Ra concentration.

The number of soil samples required for the desired accuracy and precision can be estimated from the standard deviation of the gamma measurements as discussed earlier. However, some practical limit on soil sampling should be set, such as five 10-composite samples. More samples may be in order if the area of the deposit is substantially greater than the largest deposit in this study.

Evaluating subsurface contamination

Determination of the level of contamination in subsurface soil presents considerable practical problems, and it would not be reasonable to set the same goals for accuracy and precision in estimating the average subsurface Ra concentration at a given depth that one would set for surface contamination. It may be reasonable, for example, to attempt to determine the average Ra concentration from 15 to 30 cm with an accuracy of 50% at a 90% confidence level. Although the present study was not designed to gather direct information about a sampling technique for subsurface soil, the equivalences among the various sampling techniques

determined for surface soil might be assumed to apply also to subsurface soil. For the sites in this study (excluding the tailings pile), the number of 10-composite samples needed for an accuracy of 50% at a 90% confidence level (as predicted by Eq. 4) ranges from 0.25 to 2. (Of course, anything less than one composite sample should be interpreted as one.) Using our estimated equivalence of one 10-composite sample and 8 uniformly spaced post hole samples, we estimate that the number of post holes required for 50% accuracy at a 90% confidence level would range from 2 (0.25x8) to 16 (2x8) for the sites in this study.

There are various approaches that could be applied to sample subsurface soil.

a. The present method could be continued, that is, one could select two points with average surface gamma levels and take post hole samples until a decrease in the gamma level is observed. This method does not appear to lead to a reliable estimate of the average Ra concentration in surface soil, and we would expect it to be at least as inaccurate for subsurface soil.

b. The 10-composite method could be applied to subsurface soil, with plugs taken at 15-cm intervals to a maximum obtainable depth over the deposit. Results from this study indicate that an accuracy of at least 50% with a precision of 90% can usually be obtained with a single 10-composite sample, although two samples might be preferable if the surface gamma measurements indicate extremely heterogeneous contamination. This approach would be facilitated somewhat if some method could be devised to determine the gamma level at depths from which composite samples are taken, so that sampling could be stopped once a relatively uncontaminated depth has been reached.

c. A method similar to method (b) could be used, except that a post hole digger would be used to obtain a number of uniformly spaced (but otherwise randomly selected) samples equivalent to the number of 10-composites indicated by Eq. 4. The accuracy and precision obtained would be the same as that with method (b). The advantages of this method are that the post hole digger will work in all types of soil and a decrease of contamination with depth could be measured in the post holes with a gamma scintillator. The disadvantage is that considerable digging would be required since eight uniformly spaced post hole samples are approximately equivalent to a single 10-composite sample.

d. We have reviewed data collected by ORNL from hundreds of UMTRA sites since 1984, including extensive comparisons of surface gamma

levels and Ra-226 concentrations in surface and subsurface soil at the same locations. Some of these locations showed typical surface gamma levels for a site, while others showed maximum levels for the site. We have drawn the tentative conclusion from these data, together with surface and subsurface gamma data from the present study, that a fairly conservative estimate of subsurface contamination usually can be obtained from subsurface samples taken beneath points showing highest surface gamma. Thus, it may be useful to employ a variation of method (a) in which post holes are dug at 2-3 points showing highest surface gamma levels rather than average gamma levels. Subsurface samples would be collected (for analysis) at 15-cm intervals as far down as practical or until gamma measurements suggest little or no further contamination. Since the Ra concentration in these samples is assumed to yield a conservative estimate for subsurface contamination, results could be used only to conclude that subsurface contamination is below guideline values. If the estimate is above guideline values, more extensive sampling of subsurface soil (say, by method (b) or (c)) would be indicated. Thus, this method would require revisits to a small percentage of the sites after the soil samples have been analyzed (considerably less than 15% of the sites, judging from the subsurface data for the hundreds of sites surveyed by ORNL since January, 1984). It would appear to be a fairly efficient method, however, since it would require a minimal number of subsurface measurements on most sites.

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[1] United States Environmental Protection Agency, 1983, Standards for Remedial Actions at Inactive Uranium Processing Sites; Final Rule (40 CFR Part 192). Fed. Regist. 48 (3):590-604 (January 5, 1983).

[2] Sokal, Robert R. and Rahlf, F. James, 1969, *Biometry*, W. H. Freeman and Company, p. 13.

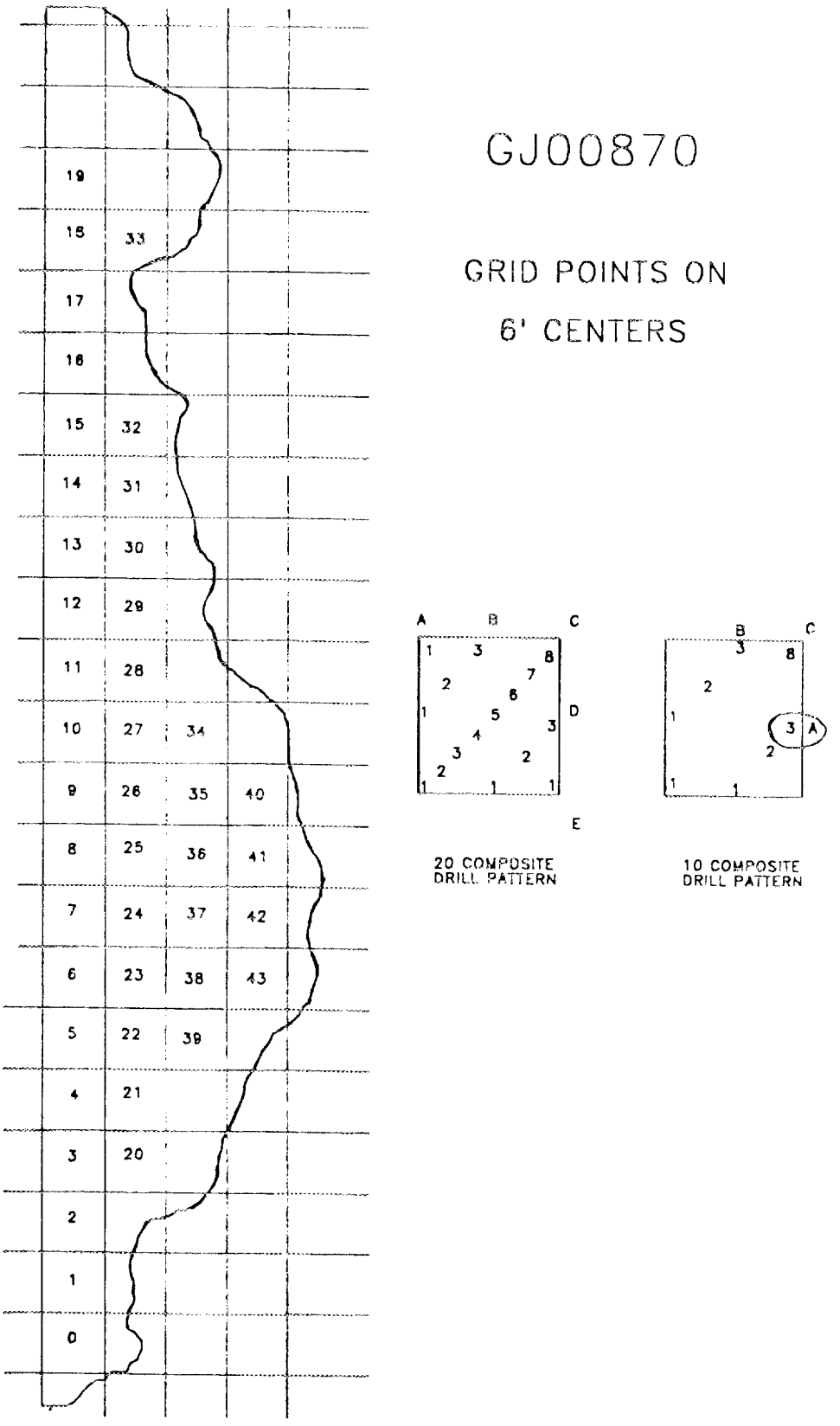

FIG. 1. Example of sampling grid pattern and deposit definition.

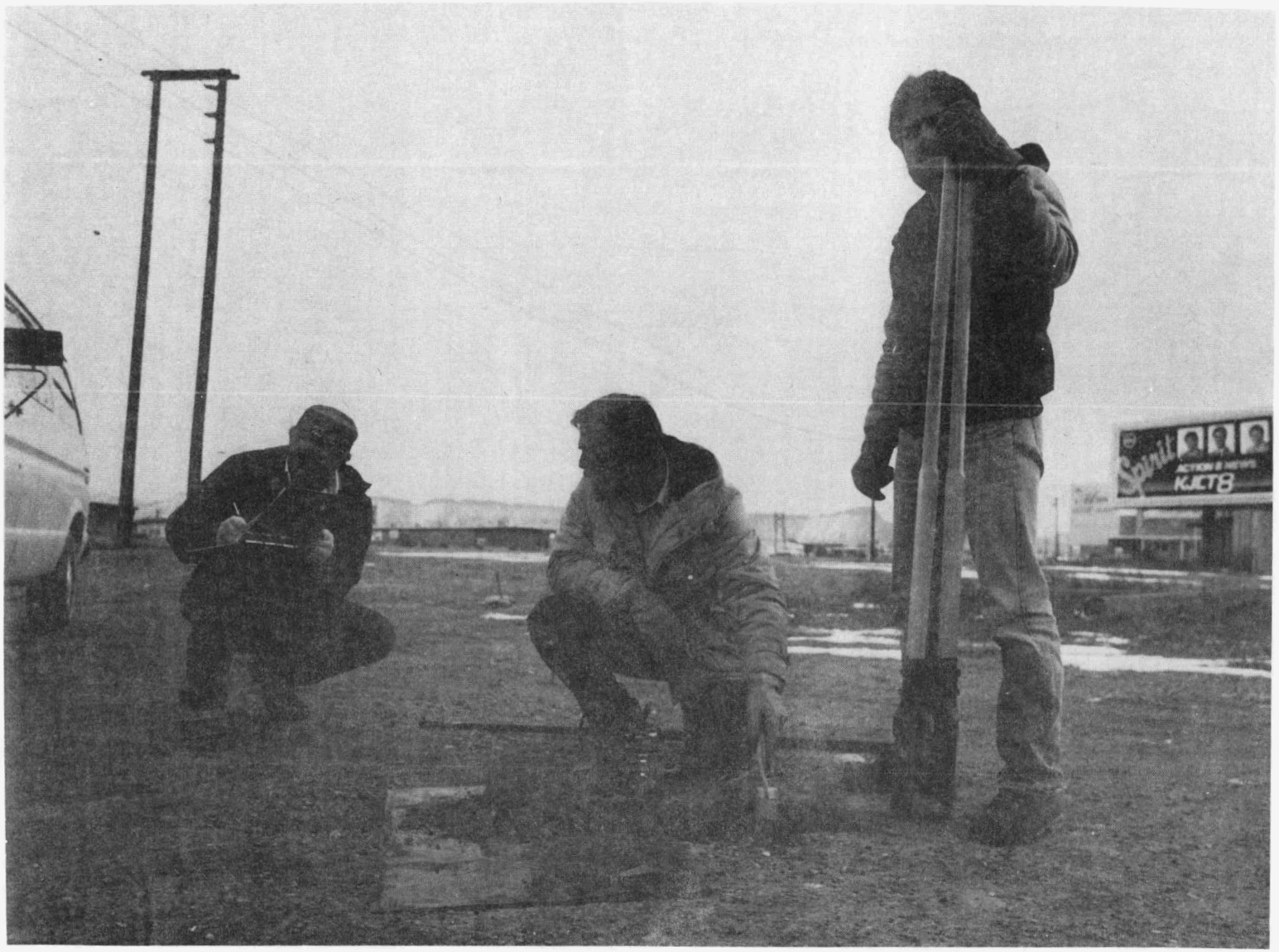

FIG. 2. Current sampling protocol---two post hole samples at average gamma.

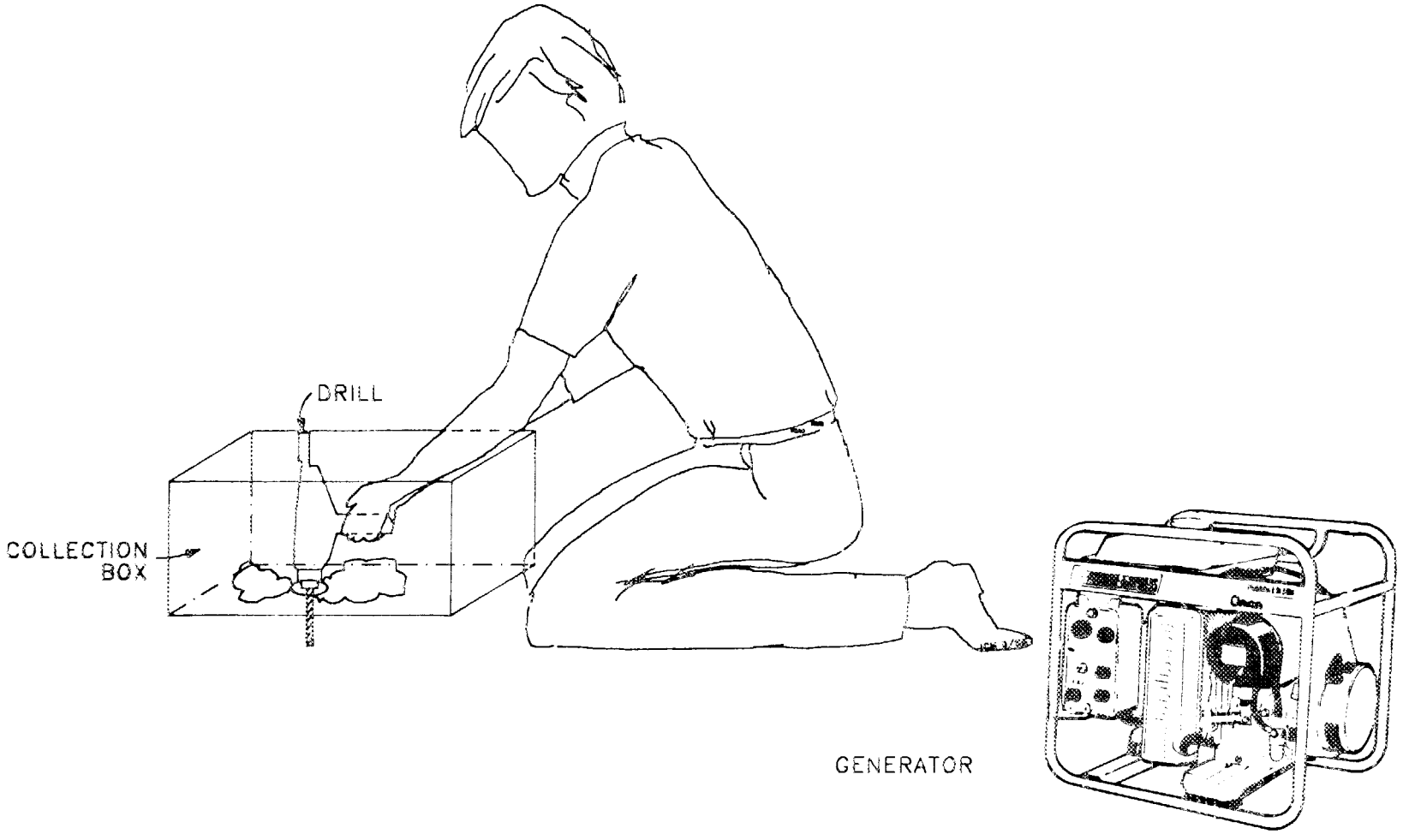

FIG. 3. TAC Drill Method

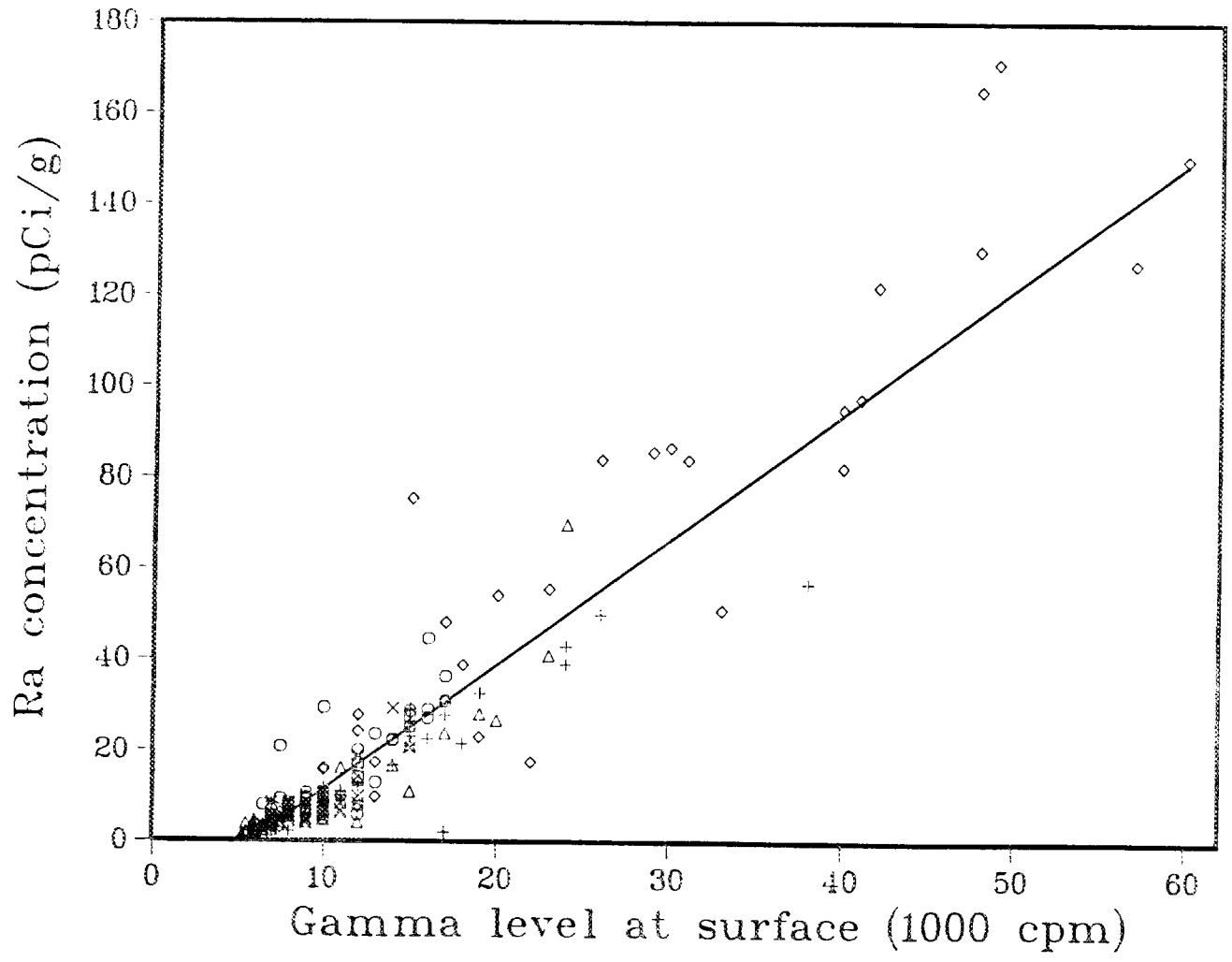

Figure 4. Ra-226 concentration in post hole sample versus gamma level at the ground surface. GJ00762-circle; GJ00870-triangle; GJ11088-plus sign; GJ12711-X; GJ13660-diamond.

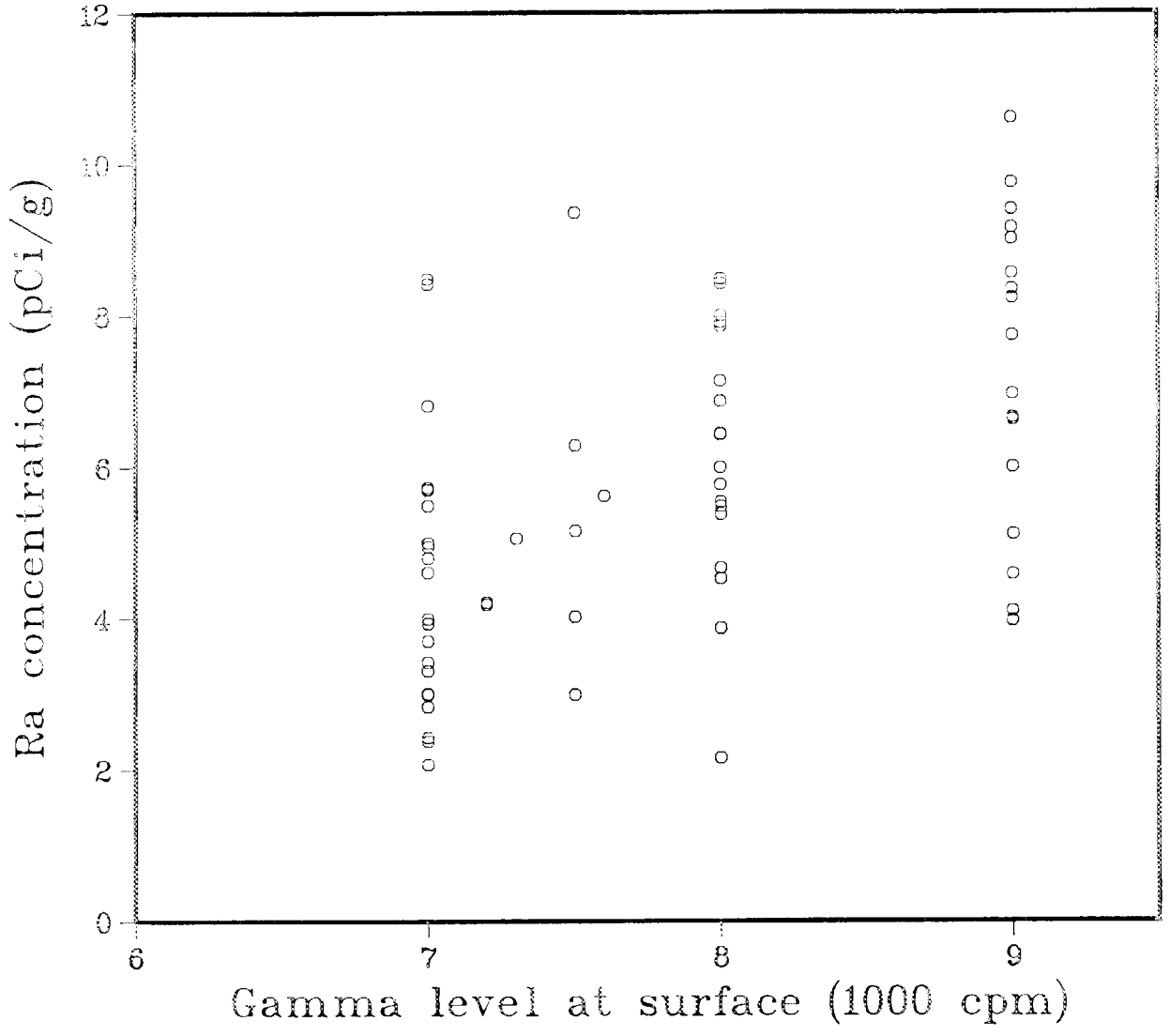

Figure 5. Ra-226 concentration in post hole sample versus gamma level at the ground surface for a narrow range of gamma levels.

Table 1. Summary of soil sample data for each site.

Site	Type of sample	Number of samples	Number of samples or plugs per m^2	Sample mean Ra conc. (pCi/g)	Standard deviation	Estimated true mean Ra conc. (pCi/g)
GJ00762	individual	45	3	14.2	10.4	14.2
	20-composite	30	40	14.2	2.9	
	10-composite	30	40	13.7[a]	2.6	
GJ00870	individual	66	0.5	7.3	11.2	9.0
	20-composite	30	4	9.6	2.8	
	10-composite	30	2	10.2	4.2	
GJ11088	individual	60	0.3	19.3	21.5	17.3
	20-composite	30	2.5	15.2	4.1	
	10-composite	30	2.5	10.1[a]	3.6	
GJ12711	individual	55	1.7	7.9	5.2	7.8
	20-composite	30	18	7.7	0.6	
	10-composite	30	9	7.8	1.1	
GJ13660	individual	40	0.2	51.3	50.2	57.9
	20-composite	30	3.3	64.1	14.2	
	10-composite	30	1.6	58.3	12.1	
GJTPILE[b]	individual	95	1	60.8[c]	50.2	?
	20-composite	30	6	34.0[c]	9.6	
	10-composite	30	3	39.8[c]	10.3	

[a]Substantial area of deposit not sampled by this method; hence mean may not be representative of entire deposit.

[b]Tailings pile.

[c]Effective depth of sampling may have been different for composite samples and post-hole samples on tailings pile.

Table 2. Comparison of accuracy and precision produced by different methods and sample sizes.

Percent of true mean	Site	Fraction within given percent of true mean				
		single 20-comp	single 10-comp	15 random PH'S with replacement	10 random PH's without replacement	8 random PH's, nearly uniformly spaced
5	GJ00762	0.30	0.23[a]	0.23	0.18	0.21
	GJ00870	0.10	0.12	0.09	0.07	0.09
	GJ11088	0.13	0.07[a]	0.10	0.11	0.06
	GJ12711	0.57	0.27	0.22	0.18	0.16
	GJ13660	0.17	0.20	0.15	0.16	0.13
	MEAN	0.25	0.18	0.16	0.14	0.13
10	GJ00762	0.33	0.50[a]	0.37	0.36	0.40
	GJ00870	0.20	0.25	0.19	0.14	0.16
	GJ11088	0.20	0.10[a]	0.27	0.23	0.23
	GJ12711	0.73	0.4[illegible]	0.45	0.36	0.29
	GJ13660	0.27	0.37	0.33	0.29	0.32
	MEAN	0.35	0.34	0.32	0.28	0.28
20	GJ00762	0.70	0.83[a]	0.68	0.66	0.78
	GJ00870	0.50	0.59	0.35	0.28	0.33
	GJ11088	0.40	0.23[a]	0.46	0.38	0.42
	GJ12711	1.00	0.87	0.74	0.74	0.67
	GJ13660	0.50	0.60	0.58	0.54	0.52
	MEAN	0.62	0.62	0.56	0.52	0.54
30	GJ00762	0.83	0.93[a]	0.85	0.84	0.92
	GJ00870	0.70	0.72	0.52	0.43	0.48
	GJ11088	0.67	0.60[a]	0.62	0.60	0.60
	GJ12711	1.00	0.97	0.90	0.90	0.92
	GJ13660	0.73	0.87	0.78	0.73	0.72
	MEAN	0.79	0.82	0.73	0.70	0.73

[a]Part of deposit on this site not sampled by 10-composite method. Mean of area sampled was used in place of true mean for entire deposit.

Table 3. Comparison of coefficients of variation (standard deviation divided by the mean) obtained by different methods.

Site	single 20-comp	single 10-comp	15 random PH's with replacement	10 random PH's without replacement	8 random PH's, nearly uniformly spaced
GJ00762	0.20	0.19	0.19	0.21	0.19
GJ00870	0.29	0.41	0.39	0.45	0.41
GJ11088	0.27	0.36	0.29	0.31	0.32
GJ12711	0.08	0.14	0.17	0.19	0.20
GJ13660	0.22	0.21	0.25	0.26	0.26
GJTPILE	0.28	0.26	0.21	0.24	0.21
MEAN OF C.V.'s	0.22	0.26	0.25	0.28	0.27
S.D. OF C.V.'s	0.08	0.10	0.08	0.09	0.09

Table 4. Accuracy and precision obtained by averaging 1 to 5 composite samples, assuming normal distributions for composite samples. Values indicate fraction of estimates expected to lie within given percent of mean.[a]

Percent of mean[a]	Type of composite	Statistic for all 6 sites	One sample	Two samples	Three samples	Four samples	Five samples
10	20	Range	0.24-0.80	0.36-0.92	0.44-0.96	0.50-0.98	0.54-0.99
		Median	0.30	0.43	0.51	0.58	0.63
		Mean/S.D.	0.37/0.21	0.50/0.21	0.58/0.20	0.64/0.18	0.68/0.17
	10	Range	0.16-0.52	0.24-0.68	0.32-0.78	0.36-0.84	0.40-0.88
		Median	0.32	0.45	0.53	0.61	0.65
		Mean/S.D.	0.32/0.13	0.44/0.17	0.52/0.17	0.56/0.20	0.63/0.18
20	20	Range	0.50-0.98	0.66-0.99	0.76-0.99	0.82-0.99	0.86-0.99
		Median	0.58	0.74	0.84	0.89	0.92
		Mean/S.D.	0.64/0.18	0.77/0.12	0.85/0.09	0.90/0.07	0.92/0.05
	10	Range	0.36-0.84	0.50-0.94	0.60-0.98	0.66-0.99	0.72-0.99
		Median	0.61	0.77	0.85	0.90	0.93
		Mean/S.D.	0.59/0.18	0.73/0.17	0.81/0.15	0.86/0.14	0.89/0.11
30	20	Range	0.68-0.99	0.84-0.99	0.92-0.99	0.94-0.99	0.96-0.99
		Median	0.77	0.91	0.96	0.97	0.98
		Mean/S.D.	0.79/0.12	0.91/0.06	0.96/0.03	0.97/0.02	0.98/0.01
	10	Range	0.52-0.96	0.68-0.99	0.78-0.99	0.84-0.99	0.88-0.99
		Median	0.79	0.91	0.96	0.97	0.99
		Mean/S.D.	0.76/0.17	0.87/0.12	0.92/0.09	0.94/0.06	0.96/0.05

[a]"Mean" for 20- or 10-composite samples refers here to mean based on all available 20- or 10-composite samples, respectively. This may differ from true mean for deposit as determined from all types of measurements.

Table 5. Comparison of the radium concentration predicted by the regression and the true mean concentration for the deposit.

Site	Average surface gamma measured at post holes (μR/hr)	Ra conc. predicted from regression eq.	99% confidence error limits	True mean Ra conc.
GJ00762	21.0	12.5	± 5.4	14.2
GJ00870	17.8	7.3	± 4.7	9.0
GJ11088	25.9	20.5	± 6.3	17.3
GJ12711	19.3	9.8	± 5.0	7.8
GJ13660	41.6	46.1[a]	± 5.4	57.9

[a]30% of the gamma measurements on this site exceeded 30,000 cpm or 54 μR/hr.

Table 6. Predictions and observations of number of composite samples needed for 90% confidence that estimated Ra concentration is within 20% of true mean.

Site	Type of composite	Prediction	Observation
GJ00762	20	3 (2.2)	3 (2.8)
	10	3 (2.2)	3 (2.4)
GJ00870	20	13 (12.2)	6 (5.8)
	10	13 (12.2)	12 (11.5)[a]
GJ11088	20	5 (4.2)	5 (4.9)
	10	[b]	
GJ12711	20	2 (1.5)	1 (0.4)
	10	2 (1.5)	2 (1.4)
GJ13660	20	4 (3.6)	4 (3.3)
	10	4 (3.6)	3 (2.9)

[a]Some background area was included in the "deposit". This may have contributed to the high number of samples needed.

[b]Insufficient surface gamma measurements in sampled area.

Table 7. Summary of accuracy obtained by selecting one post hole sample at random from an area with a nearly average gamma level.

Site	Fraction within 20% of true mean
GJ00762	0.13
GJ00870	0.08
GJ11088	0.11
GJ12711	0.68
GJ13660	0.33

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