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PROCEDURES FOR SAMPLING RADIUM-CONTAMINATED SOILS

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Technical Measurements Center

October 1985

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U.S. Department of Energy
Nuclear Energy Programs
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SUMMARY

Two procedures for sampling the surface layer (0 to 15 centimeters) of radium-contaminated soil are recommended for use in remedial action projects. These procedures were developed by the Technical Measurements Center, in support of the U.S. Department of Energy Division of Remedial Action Projects, with a view to providing the means of establishing uniform, consistent soil-sampling efforts among remedial action contractors.

Both procedures adhere to the philosophy that soil samples should have constant geometry and constant volume in order to ensure uniformity. In the first procedure, a "cookie cutter," fashioned from pipe or steel plate, is driven to the desired depth by means of a slide hammer, and the sample extracted as a core or plug. The second procedure requires use of a template to outline the sampling area, from which the sample is obtained using a trowel or spoon. Sampling to the desired depth must then be performed incrementally. Selection of one procedure over the other is governed primarily by soil conditions, the cookie cutter being effective in nongravelly soils, and the template procedure appropriate for use in both gravelly and nongravelly soils. In any event, a minimum sample volume of 1000 cubic centimeters is recommended.

The step-by-step procedures are accompanied by a description of the minimum requirements for sample documentation. Transport of the soil samples from the field is then addressed in a discussion of the Federal regulations for shipping radioactive materials. Interpretation of those regulations, particularly in light of their application to remedial action soil-sampling programs, is provided in the form of guidance and suggested procedures. Due to the complex nature of the regulations, however, there is no guarantee that our interpretations of them are complete or entirely accurate.

Preparation of soil samples for radium-226 analysis by means of gamma-ray spectroscopy is described in Appendix A; two procedures are provided, one for normal samples and one for large-volume samples. They are followed by a discussion and example of one-way analysis of variance, a technique recommended for use as a form of quality control. Finally, supplementary detail relative to the shipping regulations, including required calculations and determinations, is presented in Appendices B and C.

1.0 INTRODUCTION

1.1 PURPOSE AND SCOPE

This document presents procedures developed by the Technical Measurements Center (TMC) for the collection and preparation of radium-contaminated soil samples, in support of work performed for the U.S. Department of Energy (DOE) Division of Remedial Action Projects. Residues from uranium processing constitute the primary source of radioactive contamination at the majority of sites designated for cleanup under the four programs administered by the Division; those four programs are the Uranium Mill Tailings Remedial Action Project (UMTRAP), Surplus Facilities Management Program (SFMP), Formerly Utilized Sites Remedial Action Program (FUSRAP), and Grand Junction Remedial Action Program (GJRAP). It is estimated that 90 percent of the radioactive contamination at FUSRAP sites results from radionuclides of the uranium decay series, while at UMTRAP and GJRAP sites, these are the only radionuclides of interest. Of those radionuclides in the uranium series, radium-226 is of special concern because of its decay into short-lived daughters, especially radon-222, the presence of which constitute a human health hazard.

These procedures for sampling soils contaminated by radium-226 are modifications of those recommended in the U.S. Atomic Energy Commission Regulatory Guide (U.S. Atomic Energy Commission, 1974) for sampling soils contaminated by cesium-137, strontium-90, americium-241, and isotopes of plutonium. In addition to providing the actual step-by-step procedures, sample documentation is addressed, and available equipment is described, particularly in terms of its appropriate applications. Summaries of relevant standards and of soil-sampling methods used by other agencies are presented in an early section of this report; the step-by-step sample-preparation procedures are described in Appendix A.

Finally, the Federal regulations governing the transport of radioactive materials are summarized, and guidelines are presented relative to packaging and shipping of soil samples. Field procedures for determining the shipping category to which a sample belongs and the corresponding suitable method of sample transport are presented. Supplementary information on the Federal regulations and their applications is provided in Appendices B and C. It is important to note, however, that the regulations are not only complex, but are complicated by overlapping jurisdictions, and as such are open to a number of different interpretations. The guidelines and suggested procedures presented herein are based on one set of interpretations--those of the Technical Measurements

Center--and, though formulated only after careful investigation and consideration, may not necessarily ensure compliance.

1.2 SOIL SAMPLING IN SUPPORT OF REMEDIAL ACTIONS

Soil has been described as an integrator of long-lived radionuclides in terrestrial environments (Hardy and Krey, 1971). As such, soil is an agent in the transfer of radioactive contaminants to the hydrosphere, biosphere, and atmosphere. The emphasis on soil cleanup in remedial actions is therefore not unfounded. Soil sampling for purposes of characterization, estimation, and prediction is common to all phases of remedial actions, and generally addresses one or more of the objectives discussed below.

1.2.1 Distribution and Inventory of Contaminants

Soil sampling may be conducted to specify the extent of contamination. This may involve demarcating the geographic boundaries of contamination, and locating areas where the contaminant concentration exceeds the cleanup standard. Such information is necessary for developing engineering plans for cleanup. Estimation of the average contaminant concentration and/or total amount of contaminant in soil usually requires a carefully designed survey (see, for example, Gilbert and others, 1975; Weimer and others, 1981; and Krey, 1976).

1.2.2 Calibration of Instruments and Measurements

Soil sampling may be required to demonstrate agreement between in-situ field measurements and laboratory measurements, or for calibration of field instruments via regression analysis. Defining the regression relationship of laboratory measurements on field measurements is a prerequisite for applying double sampling, a technique used to optimize cost and precision when obtaining estimates of mean and variance for a site (Gilbert and Eberhardt, 1976).

1.2.3 Certification of Compliance

Soil sampling may play an integral role in verifying that cleanup standards have been met and that a site can be released for a specified level of use.

1.2.4 Radiologic Studies

Soil sampling is commonly used to some degree in site radiologic surveys (Goldsmith and others, 1981), in resuspension studies (Phelps and Anspaugh, 1974; Schwendiman and others, 1980), and in studies of plant uptake of contaminants (Hoffman and Keller, 1982).

1.3 DEFINITIONS

For purposes of this report, the following definitions are used:

- Gravel - That fraction of a soil, excluding soil aggregates, that will not pass through a 2-millimeter sieve.
- Soil - All unconsolidated material typically found on or near the surface of the earth including, but not limited to, silts, clays, sands, gravel, and small rocks [40 CFR 192.11(d)].

2.0 OVERVIEW OF SOIL SAMPLING STANDARDS AND METHODS

2.1 APPLICABLE STANDARDS

Standards applicable to the cleanup of sites contaminated by uranium mill tailings are described in 40 CFR Part 192 (U.S. Environmental Protection Agency, 1983). Section 192.12 states the following:

> "Remedial actions shall be conducted so as to provide reasonable assurance that, as a result of residual radioactive materials from any designated processing site:
>
> (a) The concentration of radium-226 in land averaged over any area of 100 square meters shall not exceed the background level by more than

(1) 5 pCi/g, averaged over the first 15 cm of soil below the surface, and

(2) 15 pCi/g, averaged over 15-cm-thick layers of soil more than 15 cm below the surface."

Section 192.20(b)(1) encourages compliance with the above standards through radiation surveys to the extent that is practical.

When radioactive contaminants other than radium-226 are present in amounts sufficient to constitute a health hazard, remedial action is required to reduce the residual radioactivity attributed to these sources to levels that are as low as reasonably achievable [40 CFR 192.22(b)]. Although the U.S. Environmental Protection Agency (EPA) has not set regulatory limits for radioactive contaminants other than radium-226, some guidance may be obtained from nonregulatory sources such as Mueller and others (1981) and Healy and others (1979). In addition, the U.S. Department of Energy provides guidance for cleanup of radioactive materials at FUSRAP and remote SFMP sites, including the establishment of limits for residual radioactivity in soil at those sites (U.S. Department of Energy, 1985).

2.2 REVIEW OF SOIL SAMPLING METHODS

Three well-documented procedures are reviewed in the succeeding subsections to provide background information on soil sampling methods. In each of these procedures, a fixed geometry is obtained by sampling a constant surface area through a specified depth. This is a desirable feature in soil sampling since variance arising from differences in sample volume will be minimized. This factor may be particularly important in soil sampling studies that involve comparisons of in-situ and laboratory measurements and comparisons of results from two independent sampling programs at a site.

2.2.1 Health and Safety Laboratory (HASL) Procedure

For a number of years, the Health and Safety Laboratory (HASL) conducted worldwide monitoring of radioactive fallout resulting from atmospheric bomb tests, as well as studying local radioactive releases from specific sites such as the Rocky Flats Plant near Golden, Colorado. Most of the sampling was conducted to estimate the amount of fallout deposited in a particular area. Contaminant concentrations were calculated as activity per unit area (e.g., mCi/km^2), requiring that sampling be performed in such a way that the weight of the sample could be related to the surface area actually sampled. Sampling sites were selected on the basis of a predetermined set of criteria in order to obtain unbiased estimates of the amount of fallout deposited.

The original soil-sampling procedure used by HASL is described in detail by Harley (1972). Generally, it involved collecting ten individual plugs or cores at each site, spaced 0.3 to 0.6 meter apart along a transect approximately 5 meters long. Fixed sample geometry was maintained by using both a "cookie cutter" and a barrel auger. The former, 9 centimeters in diameter, was used to remove a 5-centimeter-deep plug of sod, after which the auger, 8 centimeters in diameter, was used to retrieve the remainder of the sample to a depth of 15 to 25 centimeters. The ten samples collected in this way were composited to make a single large sample.

2.2.2 Nevada Applied Ecology Group (NAEG) Procedure

The Nevada Applied Ecology Group (NAEG) conducted an extensive sampling program to determine the distribution and inventory of plutonium, americium, and uranium in soil at various locations on or near the Nevada Test Site. The soil sampling method used by NAEG is summarized by Fowler and Essington (1977) and Fowler and others (1974). Contaminant concentrations were expressed as activity per unit area. Samples were collected such that surface area and volume were known.

In the NAEG method, the sampling tool consisted of a steel ring, 12.7 centimeters in diameter and 2.5 or 5 centimeters deep, on the upper end of which a lip was welded to provide rigidity and to ensure a constant depth of penetration. The ring was pressed into the soil to its upper edge, and the soil outside was removed to the base of the ring. Next, the soil inside the ring was removed to the base of the ring and placed in a bag. The ring was again pressed into the soil within the excavated hole, and the procedure repeated incrementally to the desired depth.

Gilbert and Eberhardt (1974) and Gilbert and others (1975) describe the use of random, stratified soil sampling in some of the NAEG plutonium studies. Strata were initially defined on a grid or radial traverse by means of instrument surveys. Once the strata were defined, sample-location coordinates within them were randomly selected prior to actual sampling. A list was prepared to specify primary sample locations, as well as contingency locations in the event that some primary locations could not be sampled. Field personnel were instructed to sample sequentially in the order that locations were listed. Prior to collecting each sample, in-situ radiation measurements were made and recorded on the data sheet. In addition, notes were made with respect to pertinent conditions encountered at each sampling location at the time of sampling.

2.2.3 Los Alamos National Laboratory (LANL) Procedure

No single soil-sampling method is endorsed by Los Alamos National Laboratory (LANL). The method described here was used by LANL personnel in a number of studies of vertical and horizontal distribution, temporal changes in distribution, and inventory of contaminants in alluvial sediments and soils (cf. Nyhan and Hakonson, 1976; Nyhan and others, 1976 and 1978). It was also used to a limited extent in a decontamination project (Alquist, 1981).

The sampling tool consisted of a short length of 2.5-centimeter-ID (inside diameter), schedule 89, polyvinylchloride (PVC) pipe with one end sharpened. An inside diameter of 2.5 centimeters is the maximum size that will permit retention of the core when sampling in loose soils or stream sediments (Nyhan and Hakonson, 1976, pp. 165-166). The pipe was driven into the ground and extracted to obtain a core. A stopper was placed in one end and the pipe was placed in a plastic bag. The pipe with the soil core was frozen in the field and subsequently sectioned into shorter lengths for preparation and analysis. Up to 10 percent compaction, integrated over the entire core, resulted from driving the tube into the ground.

3.0 PROCEDURES FOR SAMPLING SURFACE SOIL

3.1 SAMPLING REQUIREMENTS

To comply with 40 CFR 192.12(a)(1), surface samples of radium-contaminated soil must be collected to a depth of 15 centimeters. It is generally a good practice to collect all samples in the same way. This is accomplished in soil sampling by maintaining a fixed sample geometry, i.e., by sampling a constant surface area through a specified depth. Consequently, variance in the results arising from differences in volume among samples is minimized (for further discussion, see Section 3.6).

Cost-effectiveness is fostered when the soil sampling methodology used is amenable to the satisfaction of multiple objectives, specifically distribution, inventory, and calibration.

3.2 SAMPLE DOCUMENTATION

A suggested format for sample documentation is presented in Figure 3-1. At minimum, the following information is required: sample number, project name or number, name of operator, date of collection, site-location information, depth of sample, sample type, sampling method, and sample volume (when applicable). Optional information includes time in and time out, elevation, geologic unit, soil series and texture, and comments.

The sample number should be unique to the sample, requiring that the numbering system guarantee no duplication of sample numbers either during the project or in subsequent projects. One approach is to use a six-character alphanumeric descriptor, consisting of three alpha characters ranging from AAA to ZZZ and three numerals ranging from 001 to 999. This approach has the capacity to generate 1.7×10^7 different sample numbers and therefore virtually guarantees uniqueness. It is also recommended that sample numbers be preprinted on the sample forms or tickets to avoid making clerical errors during fieldwork.

Site location may be described either by latitude/longitude, by township/range if applicable, or by some other grid. The locality name, county, and state should always be recorded, and the Federal Information Processing Standard (FIPS) code for county and state designation should be noted for subsequent data-base entry. When a suite of samples are collected at the same site and/or sample location, location information should be detailed on one sample form and that sample number referenced in the "same as" space on the forms for the rest of the samples in the suite. This will not only minimize clerical errors, but will save time in the field.

Time in and time out, though optional, provide data for planning man-hour requirements in future sampling surveys. The comments section should be used to record any pertinent information regarding the sample, such as exceptions to the prescribed sampling procedure or references to any in-situ measurements made at that location.

Where required, a chain-of-custody document should accompany samples during transport to provide a record of possession.

3.3 REVIEW OF AVAILABLE EQUIPMENT AND RECOMMENDATIONS

A successful soil-sampling program requires selection of equipment appropriate to the sampling procedure used, together with a view

Sample No.________________

Project Name and/or No.____

Operator's Name____

Date Collected__________________Time In__________Time Out_________

SITE LOCATION

Same as_____________________

Meridian____Twp_____NS Range_____EW Sec_____1/64____1/16____1/4___

Latitude: Deg_____Min_____Sec____Longitude: Deg_____Min_____Sec___

Locality: Name______________________County_________State__________

FIPS Code: County__________State_________

SAMPLE LOCATION

Same as_____________________

Grid_____________NS Grid_____________EW Grid Origin_____________

Elevation______________ Sample Depth_______ Geologic Unit_________

Soil Series__________________ Soil Texture_______________________

SAMPLE TYPE (check)

SOIL: Surface____ Profile____ Composite____ Core____ Cutting ____

ROCK: Grab___ Core___ Cutting___ SEDIMENT: Concentrate____ Bulk___

Sampling Method________________________________

Sample Volume__________________________________

Comments:

Figure 3-1. Suggested Format for Soil Sample Documentation toward cost-effective accomplishment of the purposes of the survey. No single tool will be adequate for every situation encountered in the field. Limitations on the usefulness of various tools are imposed by the objectives of the survey and by soil characteristics, such as bulk density, texture, and consistency. The various types of available equipment are reviewed in the following subsections; recommendations are then made with respect to the primary equipment used for sampling soils in support of remedial actions.

3.3.1 Augers

There are two basic types of auger, the screw-type or flight auger and the bucket or barrel auger. Hand-operated screw-type augers are generally not desirable when sampling for subsequent laboratory analysis for two reasons: First, the small diameter of the auger permits collection of only a small volume of sample; and second, the potential for cross-contamination exists when sampling different subsurface layers or depth intervals. The chief use of screw-type augers is in the examination of field properties (e.g., color, texture, etc.) of subsurface soil. They are also potentially useful for locating pockets or layers of tailings at shallow depths.

The bucket or barrel auger consists of a metal cylinder with an orchard bit welded to the bottom. Soil is forced up into the cylinder when the auger is turned, minimizing the potential for cross-contamination by eliminating contact of the sample with the sidewalls of the hole. Bucket augers vary in the diameter of the bucket and in the angle and closure of the cutting bit. Diameters of 5 and 7.5 centimeters are standard. Augers designed for use in hard clay soils have open bits, while those designed for use in sandy soils have greater closure. The "all purpose" design is intermediate. Some types are designed to cut a hole larger in diameter than the bucket to facilitate removal.

Bucket augers may be used to collect samples for subsequent laboratory analysis. The tendency for the soil to run out through the bit can be minimized by tamping the soil in the bucket with a broomstick prior to lifting the auger from the bottom of the hole. The effectiveness of a bucket auger is severely restricted, however, when sampling gravelly soils or dry soils of loose consistency; the presence of pebbles or rocks having diameters larger than 2.5 centimeters may prohibit its use entirely.

3.3.2 Tube Samplers

Tube samplers are designed to obtain small-volume cores, usually no greater than 2.5 centimeters in diameter. This type of sampler typically consists of a tube, attached to a handle and extension rod, with one side of the tube cut away for easy removal of the core. Some varieties without cutaway sides have a liner to facilitate core removal; caps may be placed on the ends of the liner for storing the core. Several types of cutting tips are available for different soil conditions, usually wet or dry. On some tube samplers, these tips are removable and interchangeable.

Core is obtained by manually pushing the tube into the ground. Some samplers have a foot pedal attached to the handle, which allows the operator to apply leg power when forcing the tube into the soil. Sampling depths of 2.5 to 3 meters are possible using a tube sampler, but depths of 1 to 1.3 meters are probably the practical limit.

As with the screw-type augers, the core-volume limitation makes use of tube samplers impractical for collecting samples for subsequent laboratory analysis. However, tube samplers are used extensively for soil mapping as a means of examining the field properties of soils, and they are potentially useful in remedial-action work for the same purpose. They may also be useful for locating pockets or layers of tailings at shallow depths.

3.3.3 Cookie Cutters

A cookie cutter is a metal cylinder or box that is driven into the ground to extract a large plug or core of soil. A design based on that described by Jamison and others (1950) and Robertson and others (1974) is shown in Figures 3-2 and 3-3. This design consists of a length of stainless steel pipe or tubing with a hardened-steel cutting tip pressed into one end. The cutting tip has a smaller inside diameter than the stainless steel cylinder to facilitate removal of the core. If desired, a liner can be fashioned and inserted in the cylinder to preserve the in-situ state of the core during removal. A cap with a one-half-inch steel plate forms the anvil for the slide-hammer assembly, which is used to drive the tube into the soil.

A box-type cookie cutter can be made from stainless steel plate. The bottom edge is sharpened (45-degree bevel) to provide a cutting edge. To avoid difficulty in removing the plug from the box, the soil should be sampled in depth increments of 5 to 7

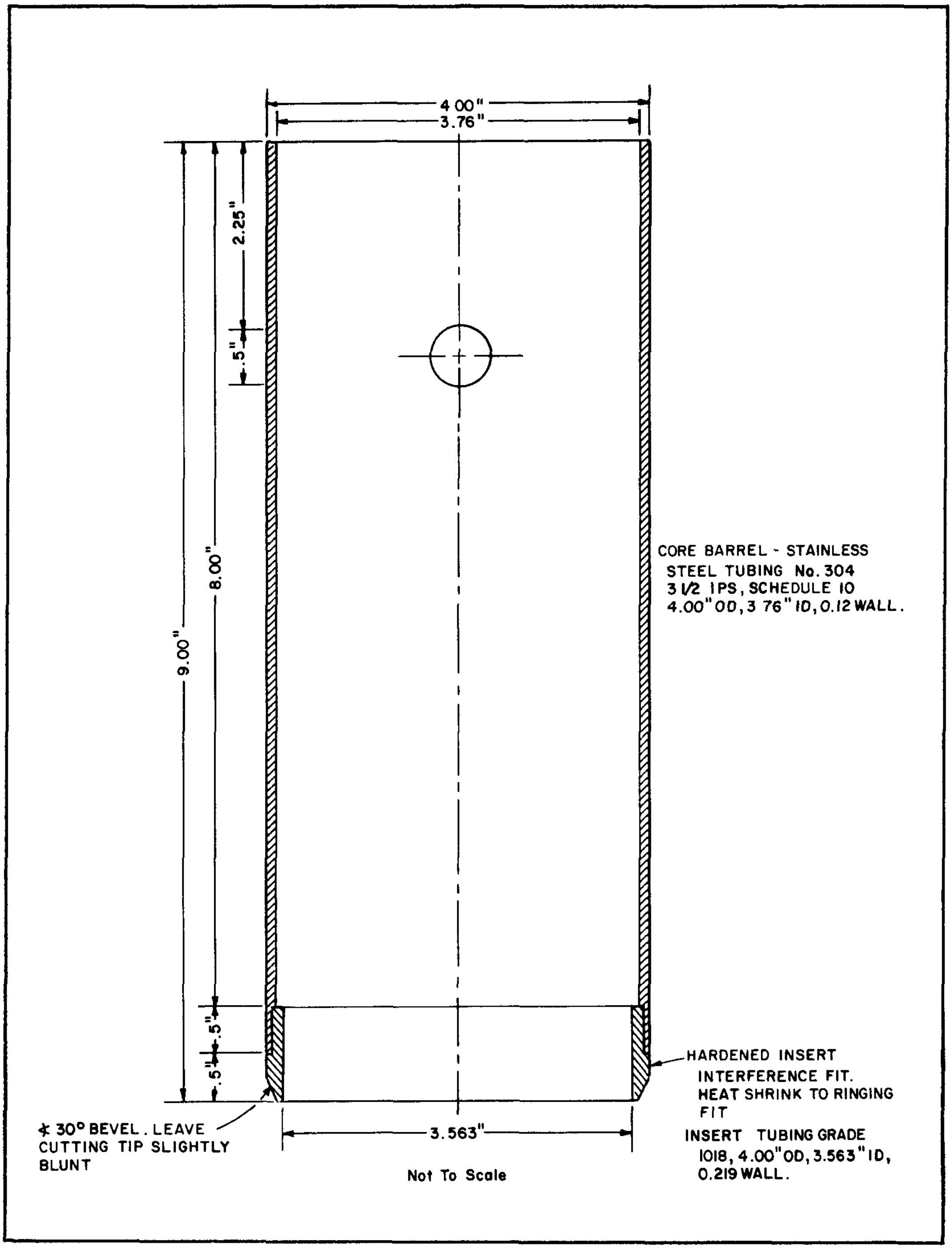
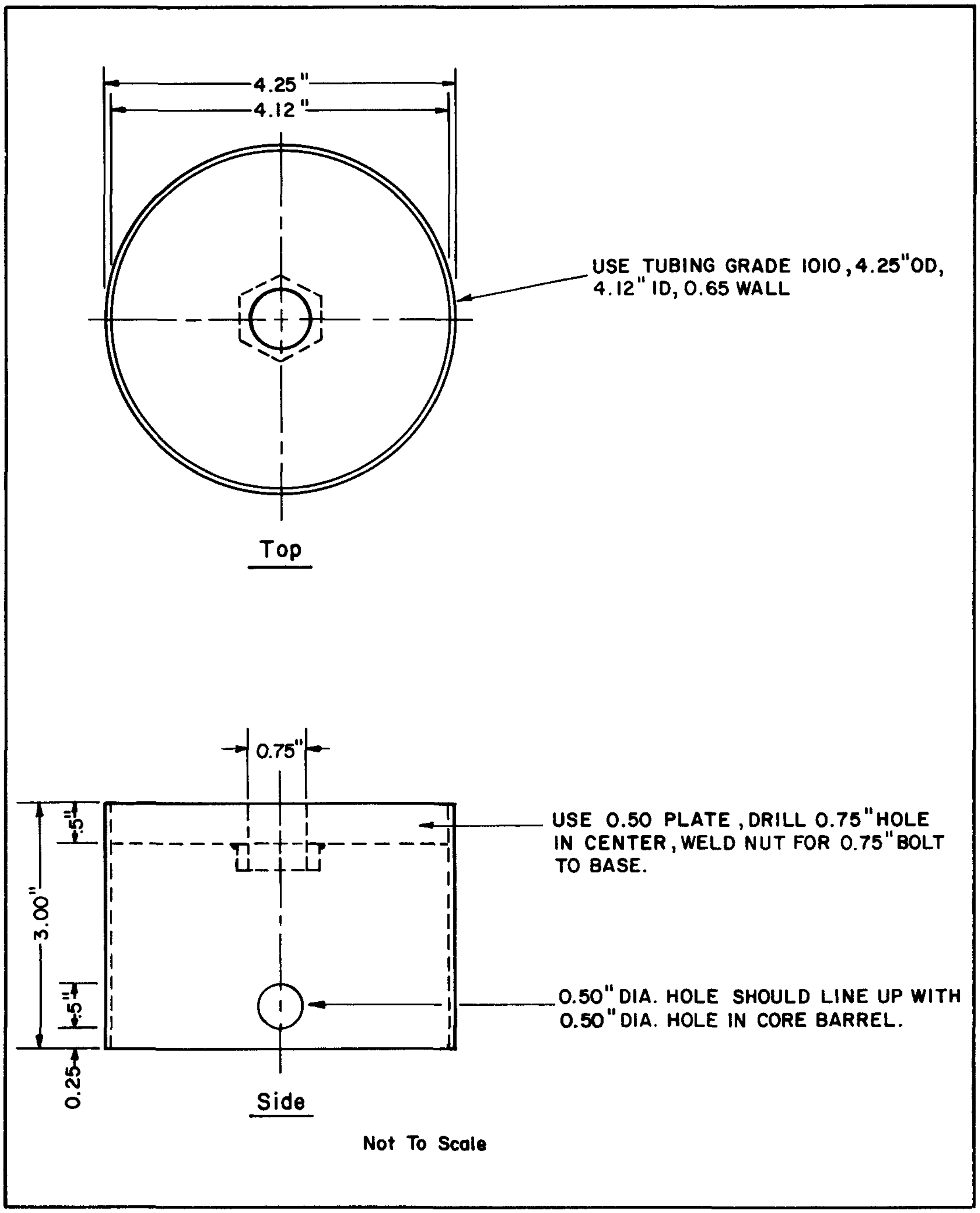


Figure 3-2. Schematic for the Barrel of a Cookie Cutter (based on Jamison and others, 1950; Robertson and others, 1974)



USE TUBING GRADE 1010, 4.25"OD, 4.12" ID, 0.65 WALL

USE 0.50 PLATE, DRILL 0.75"HOLE IN CENTER, WELD NUT FOR 0.75" BOLT TO BASE.

0.50" DIA. HOLE SHOULD LINE UP WITH 0.50" DIA. HOLE IN CORE BARREL.

Not To Scale

Figure 3-3. Schematic for the Cap of a Cookie Cutter (based on Jamison and others, 1950; Robertson and others, 1974). The cap acts as the anvil for the slide-hammer assembly. It may be "locked" to the barrel by inserting a rod through the aligned holes.

centimeters. Cylinder-type samplers can also be constructed without a cutting-tip insert and used in this way.

Two advantages of the cookie cutter are the rapid speed with which samples can be collected and the large volume obtained in a single sample. Moreover, all samples collected with a cookie cutter have identical geometry, and volumes can be calculated precisely. Drawbacks of the cookie cutter include the noisy slide hammer and the fact that its use requires physical exertion. Also, use of this sampling tool is severely limited in gravelly soils, in areas with shallow tree roots, and in dry soils with high bulk density or very hard consistency.

3.3.4 Templates

Ring templates of the type used by the Nevada Applied Ecology Group are readily fabricated from sections of pipe; square templates may be constructed from angle iron or aluminum. The template sampler offers perhaps the most versatility since use of this tool does not appear to be restricted by soil texture. Removal of the sample with spoons and trowels is tedious, however, and may require up to 20 or 30 minutes per sample.

3.3.5 Post-Hole Diggers

For certain purposes, post-hole diggers may be adequate for remedial-action sampling to depths of 15 centimeters. The hole geometry is reasonably constant, although sample volume must be estimated and will therefore be imprecise. Also, there is some danger of cross-contamination when using this type of sampling tool.

3.3.6 Recommendations

Cookie cutters for use in nongravelly soil and templates for use in gravelly or nongravelly soil are recommended as the primary equipment for soil sampling in support of remedial actions. These tools will not be suitable or desirable for all purposes, but they are best able to meet the criteria of fixed geometry and fixed volume for the specified sampling depth. The sampling procedures recommended for use with these two types of equipment are detailed in the sections that follow.

3.4 SAMPLING PROCEDURE FOR COOKIE CUTTER IN NONGRAVELLY SOIL

The following equipment and materials are needed for this procedure:

- Cookie Cutter and Slide Hammer
- Small Trowel or Hand Spade
- Medium-Sized Aluminum Cake Pan, 3 Inches Deep, or Plastic Tub
- Disposable Wipes
- Wire or Stiff-Bristled Brush
- Plastic Sample Bags and Ties
- Permanent-Ink Felt-Tip Marker
- Permanent-Ink Pen
- Field Data Forms or Sample Ticket Books

The sampling procedure for use of a cookie cutter in nongravelly soil consists of the following steps:

1. Record appropriate sample-location information on the field data form and/or sample ticket (cf. Figure 3-1).

2. With felt-tip marker, record sample-location number (e.g., grid coordinates) and sample number, if different, on the plastic bag that will hold the sample.

3. Place a paper tag, with the sample number written on it, in a second plastic bag.

4. Conduct in-situ radiation measurements and record raw data (optional). If no designated space is provided on the form, record these data in the comments section.

5. Scrape away <u>loose</u> vegetal debris or litter from the point to be sampled; do NOT scrape away mineral soil material. The sampling point should be directly below the position of the field detector used for radiation measurements, if such measurements were made.

6. Using the slide hammer, drive the cookie cutter to a depth of 15 centimeters. Detach the soil from surrounding material by either rotating the sampler or pushing the slide rod from side to side. Lift the sampler slowly from the hole, invert it over the sample tray, and push the sample out from the bottom.

   NOTE: Some compaction of the sample results from driving the sampler into the soil. If the sampler has no cutting-tip insert, extraction of the sample will be difficult. To avoid this problem, the sampler can be driven to approximately one-half the desired sampling depth, or 7.5 centimeters, and removed. After the sample is extracted, the sampler is carefully placed back in the hole and driven the remaining 7.5 centimeters.

7. Transfer the sample from the tray to the labeled sample bag using a trowel. Do not remove any organic matter or other material from the sample. Make sure a tag with the sample number is in the second bag. Place the bag containing the sample into the second bag, and tie.

8. Clean tools with the brush, and wipe with moistened disposable wipes.

9. Check labeling on the bag, field data form, and/or sample ticket book for accuracy of site information, especially location number and sample number.

3.5 SAMPLING PROCEDURE FOR TEMPLATE IN GRAVELLY OR NONGRAVELLY SOIL

The following equipment and materials are needed for this procedure:

- Sampling Template, Ring or Square
- Small Trowel or Hand Spade
- Straight Bar with Chisel End
- Spoon or Small Scoop
- Medium-Sized Aluminum Cake Pan, 3 Inches Deep, or Plastic Tub
- Wire or Stiff-Bristled Brush
- Disposable Wipes
- Plastic Sample Bags and Ties
- Permanent-Ink Felt-Tip Marker
- Permanent-Ink Pen
- Tape Measure
- Field Data Forms or Sample Ticket Books

The sampling procedure for use of a template in gravelly or nongravelly soil consists of the following steps:

1. Record appropriate sample-location information on the field data form and/or sample ticket (cf. Figure 3-1).

2. With felt-tip marker, record sample-location number (e.g., grid coordinates) and sample number, if different, on the plastic bag that will hold the sample.

3. Place a paper tag, with the sample number written on it, in a second plastic bag.

4. Conduct in-situ radiation measurements and record raw data (optional). If no designated space is provided on the form, record these data in the comments section.

5. Scrape away loose vegetal debris or litter from the point to be sampled; do NOT scrape away mineral soil material. The sampling point should be directly below the position of the field detector used for radiation measurements, if such measurements were made.

6. Press the template into the soil. Loosen the soil outside the template using a trowel or straight bar. Remove soil outside the template to lower edge at depth of 2.5 or 5 centimeters.

7. Place the labeled sample bag in the tray or tub. Remove the soil from the inside of the template with a spoon or scoop, and place the sample in the bag.

8. Repeat Steps 6 and 7 until a total depth of 15 centimeters is reached.

9. Transfer any soil remaining in the tray to the sample bag. Make sure a tag with the sample number is in the second bag. Place the bag containing the sample into the second bag, and tie.

10. Clean equipment with a brush and moistened disposable wipes.

11. Check labeling on the bag, field data form, and/or sample ticket book for accuracy of site information, especially location data and sample number.

3.6 SAMPLE VOLUME RECOMMENDATIONS

When planning a soil sampling program, questions generally arise concerning the number of samples to collect and the size of the sample volume. If statistical methods are used to answer these questions, prior knowledge is usually required, especially with respect to variance. Most basic statistics textbooks describe methods for determining the number of samples to collect. Sample volume and its effect on the data and statistics, however, are seldom addressed in the literature.

In some respects, the sample-volume problem is analogous to the aliquot-size problem encountered in laboratory analysis. That is, collecting field samples may be considered equivalent to subsampling some larger unit volume. Doctor and Gilbert (1978) studied the effects of aliquot size on various statistics for americium-241 analyses. Aliquot sizes ranged from 1 to 100 grams. Results for all sizes were right-skewed, but skewness decreased as size increased. The arithmetic mean was nearly constant, showing no trend with size of aliquot. The median and geometric mean, however, increased with aliquot size, converging on the arithmetic mean at the larger sizes. It was calculated that analysis of 1444 1-gram aliquots would be required to achieve the same precision (standard error) in the mean as that attained by analyzing twenty 100-gram aliquots.

The sensitivity of median and geometric mean to volume is to be expected on theoretical grounds for lognormal data (Michels, 1977), and this feature will probably be observed in any unimodal, right-skewed distribution. This fact suggests that when sample volumes are suspected to vary, comparison of data from different sampling surveys should be based on the arithmetic mean rather than the geometric mean or median. If the trend observed for standard deviation in laboratory analysis is applied to the field setting, one may reason that spatial (between-sampling-location) variability for some unit area can be reduced by collecting larger sample volumes. This will of course depend on source term and possibly other factors, and may not actually be true for all sites.

Nyhan and others (1981) conducted a study of the distribution of cesium-137 in sediments of Mortandad Canyon, a liquid-effluent-receiving area, and in soils in the nuclear fallout pathway at Trinity site to assess the influence of sample volume. At each site, ten locations, spaced approximately 1 meter apart, were sampled to depths of 5 centimeters using metal templates. Four samples of varying volume--25, 500, 2500, and 12,500 cubic centimeters (cm^3)--were collected at each location.

Summary statistics generated by this study are presented in Table 3-1. With respect to the arithmetic mean, estimates of cesium-137 concentration for the various sample volumes do not differ significantly at a site; sample volume has no effect. Similarly, there appears to be no relationship between standard deviation and volume at Mortandad Canyon, but, at Trinity site, standard deviation decreases with increasing volume of sample. An analysis of variance was performed to estimate components of spatial, aliquoting, and counting variation (Table 3-2). Spatial variance (between-location variation) is the largest source of variance at

Table 3-1. Summary Statistics of Cs-137 Concentrations in Surface Materials as a Function of Soil Sample Volume (modified from Nyhan and others, 1981)

Site	Parameter[a]	Cs-137 Concentration (pCi/g)			
		25-cm^3 Volume	500-cm^3 Volume	2500-cm^3 Volume	12,500-cm^3 Volume
Mortandad Canyon	Mean	70.1	76.7	72.0	73.8
	S.D.	5.67	6.55	6.74	5.36
	C.O.V.	0.0809	0.0854	0.0956	0.0726
Trinity Site	Mean	6.07	9.24	7.36	8.24
	S.D.	3.90	3.40	3.32	1.41
	C.O.V.	0.643	0.368	0.451	0.171

[a] Mean refers to arithmetic mean; S.D. is standard deviation; and C.O.V. is coefficient of variation, derived from dividing the standard deviation by the mean.

Table 3-2. Analysis-of-Variance Results for Cs-137 Concentrations at Mortandad Canyon and Trinity Site as a Function of Soil Sample Volume (modified from Nyhan and others, 1981)

Site	Source of Variance	Estimated Variance (pCi/g)			
		25-cm^3 Volume	500-cm^3 Volume	2500-cm^3 Volume	12,500-cm^3 Volume
Mortandad Canyon	Spatial	32.0	42.8	41.2	27.0
	Aliquoting	0.0	0.0	20.7	30.9
	Counting	0.135	0.135	0.134	0.134
Trinity Site	Spatial	15.4	11.5	10.4	1.95
	Aliquoting	0.0	0.0	0.559	0.556
	Counting	0.0016	0.0016	0.0016	0.0016

both sites. At Trinity site, spatial variance decreases with increasing volume. The fact that this trend is not manifested at Mortandad Canyon is attributed to the turbulent mixing of sediments in the stream channel, which results in a more homogeneous distribution of the contaminant (Nyhan and others, 1981, p. 8).

Another effect of sample volume is seen in comparisons of in-situ radiometric assays and laboratory assays of soil samples. Such a comparison was made among assays of samples obtained from the inactive uranium mill tailings site at Monticello, Utah, a site included in the Surplus Facilities Management Program (Table 3-3). Three in-situ radiometric assays for radium-226 were made at each of 28 locations using a collimated Scintrex Gad-6 portable field spectrometer that was calibrated at the DOE Walker Field Airport calibration models in Grand Junction, Colorado (Ward, 1978; Stromswold and Kosanke, 1978). The three measurements were made approximately 1 foot apart, and, following each measurement, a sample was collected to a depth of 15 centimeters from the point measured by the detector. Sample volume was 500 cm^3 at the first point, 1500 cm^3 at the second, and 2500 cm^3 at the third. Table 3-4 presents results of a regression analysis of the data for the models

$$Y = b_0 + b_1X \qquad (3\text{-}1)$$

and

$$LnY = b_0 + b_1LnX \qquad (3\text{-}2)$$

where Y is the laboratory assay and X is the field assay; associated analyses of variance are also given in Table 3-4.

Examination of the regression residuals suggests that equation (3-2) is a better model, but, even so, both models exhibit parallel trends with increasing volume of sample. The correlation indicated by multiple R, the amount of variance explained in the regression indicated by multiple R^2, and the significance of the regression given by F increase with increasing sample volume. Conversely, the scatter about the line, given by the residual mean square (MSres), decreases with increasing sample volume. An analysis of variance of the three regression equations for each model indicates that there are no statistical differences arising from sample volume. That is, the three sample volumes exhibit essentially the same line. The largest volume, however, appears to give the most precise line.

In summary, sample-volume differences may affect statistical estimates when the geometric mean or median is used rather than the arithmetic mean. It is apparent from the above discussion, however, that the most important effect of sample volume is on variance estimates. As a rule, the larger the sample volume, the better, but there are clearly limits of cost-effectiveness in terms of time spent in sample collection, preparation, and analysis versus precision gained. In certain projects, optimization studies may be warranted to maximize precision against cost with respect to sample volume. At minimum, however, a sample volume of 1000 cm^3 is recommended. This will yield between 1.5 and 2.5 kilograms of sample in soils characterized by normal bulk densities.

Table 3-3. Radium-226 Concentrations from Paired Field (in-situ) and Laboratory Measurements as a Function of Soil Sample Volume (data obtained from inactive uranium millsite at Monticello, Utah)

	Radium-226 Concentration (pCi/g)					
Site	For 500-cm^3 Volume		For 1500-cm^3 Volume		For 2500-cm^3 Volume	
	Lab	Field	Lab	Field	Lab	Field
1	2.92	3.28	3.62	2.69	3.35	3.29
2	72.68	29.24	63.96	33.19	62.68	30.84
3	2.79	2.36	3.67	4.02	10.97	6.72
4	20.66	18.85	27.28	17.46	25.42	18.01
5	6.38	7.88	10.62	6.93	11.98	7.81
6	2.42	7.84	5.96	9.91	8.37	10.56
7	24.43	20.89	19.95	22.50	16.08	16.18
8	5.04	3.12	4.89	3.23	4.93	3.76
9	2.28	1.57	1.59	1.55	1.69	1.59
10	9.91	10.98	5.03	9.33	25.97	17.71
11	18.08	9.23	7.45	6.87	1.73	3.05
12	2.42	2.08	2.09	2.16	2.01	1.97
13	2.79	10.43	4.44	12.01	10.56	12.38
14	15.42	9.12	12.69	9.75	10.45	6.91
15	12.63	6.52	4.37	3.85	3.05	3.82
16	23.79	24.19	24.55	24.55	32.07	25.50
17	38.57	10.82	9.88	7.42	12.50	10.00
18	3.76	3.96	7.92	4.90	7.12	6.70
19	3.29	3.08	3.95	5.34	26.59	13.66
20	15.36	10.83	23.73	12.43	18.71	11.12
21	3.06	2.14	2.27	2.21	3.02	2.34
22	38.34	20.66	43.90	22.83	39.86	21.48
23	93.27	35.57	21.34	25.24	36.89	20.46
24	14.21	5.71	10.49	6.07	13.97	7.43
25	1.98	1.61	1.19	1.40	1.21	1.20
26	3.80	8.62	4.45	8.22	5.01	8.70
27	12.06	9.32	7.08	5.11	9.11	4.89
28	7.73	21.82	14.97	23.11	8.25	20.27

Table 3-4. Regression Analysis of Lab Assay (Y) on Field Assay (X) for Ra-226 as a Function of Soil Sample Volume

A. Model: $Y = b_0 + b_1X$

Sample Volume	b_0	b_1	Mult. R	MS Reg	SSRes	MSRes	F
All Data	-3.21	1.67	0.7052	16804	7024	86	196
500 cm^3	-5.08	1.996	0.6979	8790	3804	146	60
1500 cm^3	-1.80	1.37	0.7291	3934	1462	56	70
2500 cm^3	-2.45	1.62	0.7917	4460	1174	45	99

Parameter	Sum of Squares	DF[a]	Mean Square	F Ratio
Regression Over Groups	584.67	4	146.17	1.77
Residual Within Groups	6439.71	78	82.56	

B. Model: $LnY = b_0 + b_1LnX$

Sample Volume	b_0	b_1	Mult. R	MS Reg	SSRes	MSRes	F
All Data	0.05	1.04	0.8807	70.78	20.48	0.25	283
500 cm^3	0.13	1.01	0.8251	23.11	10.83	0.42	55
1500 cm^3	0.05	1.00	0.9017	21.74	5.00	0.19	113
2500 cm^3	-0.04	1.10	0.9259	25.89	4.31	0.17	156

Parameter	Sum of Squares	DF[a]	Mean Square	F Ratio
Regression Over Groups	0.346	4	0.087	0.335
Residual Within Groups	20.135	78	0.258	

[a] Degrees of freedom.

4.0 SHIPPING REGULATIONS

4.1 PURPOSE AND SCOPE

The purpose of this section is to acquaint readers with the Federal regulations governing transport of radioactive soil samples and describe the practical application of those regulations based on our interpretation of them. As noted earlier, the regulations are extremely complex and reflect overlapping jurisdictions of such Federal agencies as the Department of Transportation, Nuclear Regulatory Commission, and Department of Energy. Consequently, we offer no guarantee as to the accuracy or completeness of the interpretations and suggested applications presented herein.

The important features of the Department of Transportation regulations (49 CFR 173, dated 1 November 1983) are summarized in light of their application to radioactive soil samples contaminated with uranium mill tailings. In general, most soil samples collected at remedial action sites will not be classified radioactive for transport purposes and will therefore require no special attention to be considered in compliance. On the other hand, certain samples may fall into the categories for which special packaging and shipping restrictions are mandated. Guidelines for determining the category to which a particular soil-sample shipment belongs and for selecting a suitable mode of shipment and appropriate packaging are therefore also described in this section.

4.2 DEFINITIONS

The following definitions are summarized from those presented in 49 CFR 173.403:

- Natural Thorium - Thorium characterized by the naturally occurring distribution of thorium isotopes (essentially 100 weight-percent thorium-232).

- Natural Uranium - Uranium characterized by the naturally occurring distribution of uranium isotopes (approximately 0.711 weight-percent uranium-235 and the remainder essentially uranium-238).

- Specific Activity - In reference to a radionuclide, the activity of the nuclide per unit mass of the nuclide. The specific activity of a material in which a radionuclide is uniformly distributed is expressed as the activity per unit mass of the material.

- Radioactive Material - Any material having a specific activity greater than 0.002 microcurie per gram.

- Special Form Radioactive Material - Radioactive material that is either a single, solid piece or is obtained in a sealed capsule.

- Normal Form Radioactive Material - Radioactive material that is not Special Form Radioactive Material.

- Transport Index - The dimensionless number (rounded up to the first decimal place) marked on the label of a package to designate the degree of control to be exercised by the carrier during transport. Generally, the transport index is the number expressing the maximum radiation level, in millirem per hour, occurring at distance of 1 meter (3.3 feet) from the external surface of the package. Other considerations may apply to certain fissile materials.

4.3 QUANTITIES OF RADIOACTIVITY IN PACKAGES

The regulations impose limits on the total activity (i.e., specific activity times the weight of the package) contained within a package of radioactive material. With respect to Type A packages, the limits are expressed as two quantities, A_1 and A_2, which refer to the maximum permissible activity for radionuclides in Special Form and Normal Form radioactive materials, respectively. Since soil samples contaminated with uranium mill tailings fall into the latter category, the A_2 value sets the activity limits for packages of such samples. In those cases where contaminated-soil shipments are designated Low Specific Activity or Limited Quantity, some fraction of the A_2 value will normally apply (for further discussion, see Section 4.4).

Table 4-1 lists the A_1 and A_2 values cited in 49 CFR 173.435 for radionuclides of the uranium decay series. Values for radionuclides not listed in the regulations (e.g., lead-214, bismuth-214, and polonium-214) have been assigned in accord with the specifications set forth in Section 173.433(a)(2); details of the calculations are presented in Appendix B of this document.

Table 4-1. A_1 and A_2 Quantities for Radionuclides of the Uranium Decay Series

Isotope	Atomic Number	Decay	Half-Life[a]	Quantity in Curies	
				A_1	A_2
U-238	92	Alpha	4.5x10^{9} y	Unlimited	Unlimited
Th-234	90	Beta	24 d	10	10
Pa-234	91	Beta	18 m	--	--
U-234	92	Alpha	2.5x10^{5} y	100	0.1
Th-230	90	Alpha	7.5x10^{4} y	3	0.003
Ra-226	88	Alpha	1622 y	10	0.05
Rn-222	86	Alpha	3.8 d	10	2.0
Po-218	84	Alpha	3.05 m	2	0.002
Pb-214	82	Beta	26.8 m	0.09[b]	0.002[b]
Bi-214	83	Beta	19.7 m	0.09[b]	0.002[b]
Po-214	84	Alpha	10^{-4} s	2[b]	0.002[b]
Pb-210	82	Beta	22 y	100	0.2
Bi-210	83	Beta	5 d	100	4.0
Po-210	84	Alpha	138 d	200	0.2

[a] Abbreviations: y = years; d = days; m = minutes; s = seconds.
[b] Not listed in 49 CFR 173.435; calculation shown in Appendix B.

Determination of permissible quantities for soil that is contaminated with uranium mill tailings is more complicated than simple reference to the A_2 values, since such material contains a mixture of radionuclides. For such mixtures, two rules apply. A mixture of radionuclides from a single decay chain is considered to be a single radionuclide if the following criteria are met: 1) the parent and daughters are present in their naturally occurring proportions (for example, in secular equilibrium), 2) no daughter has a half-life longer than 10 days, and 3) no daughter has a half-life longer than the parent radionuclide. For such a mixture, the A_2 quantity to be applied is that of the parent radionuclide [Section 173.433(b)(2)]. Soil samples contaminated with uranium mill tailings do not meet these criteria, since uranium has been removed and since several daughters have half-lives longer than 10 days. Such material must therefore be considered a mixture of different radionuclides, and the permissible quantity determined via computation. For those mixtures in which the identity and activity of radionuclides is known, the permissible quantity for each radionuclide must be such that the following equation is true:

$$F_1 + F_2 + \ldots + F_k + \ldots F_n \leq 1$$

where F_k is the ratio of the total activity of the k^{th} radionuclide to its A_1 or A_2 quantity [Section 173.433(b)(3)].

4.4 SHIPPING CATEGORIES

Certain categories permit shipping of radioactive materials in other than specification packaging (i.e., Type A and Type B). These are of special interest since they may provide more latitude in the handling of field samples.

4.4.1 Low Specific Activity Materials

Low Specific Activity (LSA) materials include uranium and thorium ores, physical and chemical concentrates of these ores (e.g., yellow cake), unirradiated natural or depleted uranium or thorium, nonradioactive material externally contaminated with radioactivity that is not readily dispersible, and material in which the radioactivity is essentially uniformly distributed and does not exceed certain prescribed concentration limits. These limits for radionuclides of the uranium decay series, beginning with thorium-230, are listed in Table 4-2. In general, these concentrations will not be exceeded in uranium mill tailings nor in soil contaminated with such tailings.

Table 4-2. LSA Concentration Limits for Radionuclides of the Uranium Decay Series [from 49 CFR 173.403(n)(4)]

Nuclide	A_2 Value (curies)	Estimated Average Concentration	
		Microcuries/Gram	Curies/Kilogram
Th-230	0.003	0.1	0.0001
Ra-226	0.05	0.1	0.0001
Rn-222	2.0	300	0.3
Po-218	0.002	0.1	0.0001
Pb-214	0.002	0.1	0.0001
Bi-214	0.002	0.1	0.0001
Po-214	0.002	0.1	0.0001
Pb-210	0.2	5.0	0.005
Bi-210	4.0	300	0.3
Po-210	0.2	5.0	0.005

Details for shipping LSA materials are described in 49 CFR 173.425. The chief advantage of shipping under the LSA category arises when the shipment is consigned "Exclusive Use," that is, under the supervision or direction of a single consignor from point of origin to final destination [Section 173.403(i)]. When packaged shipments of LSA materials are consigned as Exclusive Use, the shipment is exempt from specification packaging, labeling, and marking. Requirements that must be met include the following [Section 173.425(b)]:

1. The shipment must be consigned Exclusive Use, unloaded from the conveyance in which it was originally loaded, and accompanied by written instructions for the maintenance of Exclusive Use shipping controls. LSA materials can not be shipped by air.

2. The material must be packaged in strong, tight packages that will not leak under normal conditions of transport, and each package must be marked "Radioactive - LSA".

3. The package does not exceed the limits for removable radioactive contamination and radiation level (see Section 4.5 of this report).

4. The transport vehicle must be appropriately placarded, unless the shipment consists solely of unconcentrated uranium and thorium ores.

4.4.2 Limited Quantities of Radioactive Material

Limited Quantity shipments of radioactive material must meet the requirements specified in 49 CFR 173.421 and 173.423. If the activity per package does not exceed 10^{-3} A_2 quantity of the radionuclide, then it is exempt from specification packaging and from the associated shipping-paper, marking, and labeling requirements. Instead, the package need only comply with the following requirements:

1. The material must be packaged in strong, tight packages that will not leak under normal conditions of transport. The outside of the inner packaging, or the outside of the packaging itself when there is no inner packaging, must be marked "Radioactive".

2. The package does not exceed the limits for removable radioactive contamination and radiation level (see Section 4.5 of this report).

3. The package must be certified as being acceptable for transport by having a notice enclosed in or attached to the package, stating the name of the consignor and including the statement:

> This package conforms to conditions and limitations specified in 49 CFR 173.421 for excepted radioactive material, limited quantity, n.o.s., UN2910.

One advantage of Limited Quantity shipments is that there are no restrictions on air transport. [Guidelines specified in the International Civil Aviation Organization (ICAO) regulations should be consulted for air shipments of radioactive materials.] The major disadvantage is the low concentrations and quantity weights permissible for mixtures of different radionuclides. Figure 4-1, for example, shows the weight limits for various radium-226 activities, applicable to soil samples contaminated with uranium mill tailings assuming equilibrium among thorium-230 and its daughters. As the graph indicates, a package with an activity of 1100 picocuries per gram (pCi/g) may not exceed approximately 5 pounds. Similarly, a package with average activity near 200 pCi/g (the maximum concentration to be considered nonradioactive) may not exceed about 30 pounds. The calculations and assumptions used to generate this curve are presented in Appendix C.

4.5 RADIATION AND CONTAMINATION CONTROL

Measurements of radiation level (dose rate) and of nonfixed (removable) radioactive contamination are conducted on radioactive-material shipments to control exposure to radioactivity. Inspectors of such shipments can readily determine whether the packages comply with the regulations summarized below.

4.5.1 Radiation Level

Radiation level is the radiation dose-equivalent rate expressed in millirem per hour (mrem/h) [Section 173.403(v)]. Permissible radiation levels for various shipping categories are described below:

- Limited Quantity Packages - The radiation level at any point on the external surface of the package may not exceed 0.5 mrem/h [Section 173.421(b)].

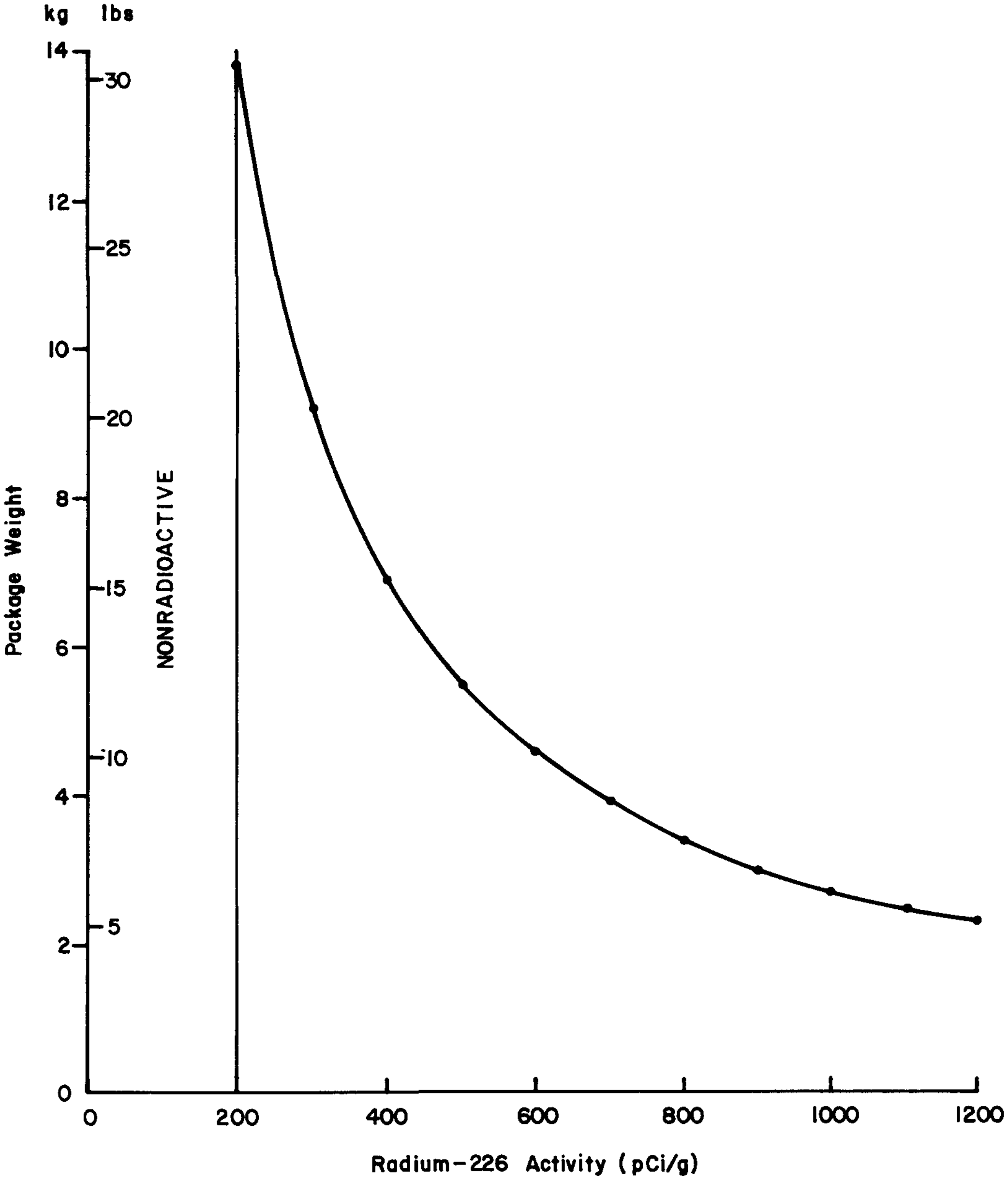


Figure 4-1. Activity and Weight Limits for Packages of Tailings-Contaminated Soil Shipped as Limited Quantity Material (also see Appendix C)

- LSA Packages - The radiation level of LSA packages transported as Exclusive Use shipments shall not at any time during transport exceed any of the following limits [Section 173.441(b)]:
  - -- 200 mrem/h on the accessible external surface of the package; or 1000 mrem/h if the following criteria are met: the shipment is made in a closed transport vehicle, the package is secured so that its position remains fixed during transport, and no unloading/loading operations occur between the beginning and end of transport.
  - -- 200 mrem/h at any point on the outer surface of the transport vehicle.
  - -- 10 mrem/h at any point 2 meters (6.6 feet) from the outer surfaces of the transport vehicle.
  - -- 2 mrem/h at any normally occupied position in the transport vehicle. This provision does not apply to private motor carriers when personnel operate under a radiation protection program.
- Other Packages - The radiation level must not exceed 200 mrem/h at any point on the external surface of the package, and the transport index must not exceed 10.

4.5.2 Contamination Control

Nonfixed (removable) radioactive contamination is defined as radioactive contamination that is easily removed from the surface by wiping with an absorbent material [Section 173.403(r)]. The maximum permissible limits for removable radioactive contamination, as set forth in 49 CFR 173.443(a), Table 10, are summarized below; these limits apply to any area of 300 square centimeters.

- 10^{-5} pCi/cm^2 or 22 disintegrations per minute (dpm)/cm^2, for beta/gamma-emitting radionuclides, all radionuclides with half-lives less than 10 days, natural uranium, natural thorium, uranium-235, uranium-238, thorium-232, and thorium-228 and -230 when contained in ore and physical concentrates.
- 10^{-6} pCi/cm^2 or 2.2 dpm/cm^2, for all other alpha-emitting radionuclides.

Exclusive Use consignments of radioactive material may not exceed these limits at the beginning of transport, and may not exceed 10 times the limits at any time during transport (Section 173.443).

4.6 FIELD APPLICATION OF REGULATIONS FOR SHIPMENT OF SOIL SAMPLES

4.6.1 Determination of Nonradioactive/Radioactive Status

4.6.1.1 Procedure

1. Assay sample or package of soil samples for Ra-226 concentration (pCi/g) using suitably calibrated field instruments according to approved procedures.

2. Multiply the Ra-226 concentration by 10 to obtain the specific activity of the packaged material (see Discussion, Section 4.6.1.2).

3. If the value is less than 2000 pCi/g, the package may be considered nonradioactive for transport purposes. If the value exceeds 2000 pCi/g, refer to Section 4.6.2 below.

4.6.1.2 Discussion. The factor 10 is the number of daughters of U-234. Equilibrium is assumed to exist among Th-230, Ra-226, etc., in the remainder of the decay chain. This assumption is generally not valid in field settings. An alternative would be to conduct disequilibrium measurements at a site and correct the Ra-226 and Th-230 activities accordingly. This too has its shortcomings. Note that the limit for Ra-226 concentration is 200 pCi/g. In this determination, uranium is assumed not to support the activity of Th-230 and its daughters, a generally valid assumption since mill records suggest that average recoveries were better than 80 percent. The short half-lives of Th-234 and Pa-234 preclude their support of the activity of Th-230 after extraction of uranium. It is further assumed that the contribution from the Th-232 series and the U-235 series is negligible. This is generally true for U-235 (isotopic abundance approximately 0.7 percent), but may vary for Th-232 depending on the source of the ore.

4.6.2 Determination for Limited Quantity Shipments

4.6.2.1 Procedure

1. Weigh package containing radioactive material.
2. Refer to Figure 4-1 to determine the maximum activity of Ra-226 permissible in the package.
3. If the Ra-226 activity of the package does not exceed the permissible activity, measure the exposure rate at the surface of the package. If this value does not exceed 0.5 mrem/h, go on to Step 4. If the exposure rate exceeds 0.5 mrem/h, repackage in a larger package, or refer to Section 4.6.3.
4. Measure the surface alpha activity with a portable field alpha detector. If the surface activity does not exceed the limits for removable radioactive contamination specified in Section 4.5.2, go on to Step 5. If the surface alpha activity exceeds the limits, decontaminate the package using approved methods, and resurvey. If the limits are still exceeded, repackage the contents in a different container. Dispose of the original container appropriately.
5. Label the outside of the inner packaging, or the outside of the package if there is no inner packaging, "Radioactive". Complete shipping papers. Maintain records of all field measurements, instrument numbers, etc.

4.6.2.2 Discussion. The curve depicted in Figure 4-1 assumes that soil material can be regarded as a mixture of four radionuclides, namely, Th-230, Ra-226, Pb-210, and Po-210. The calculations and assumptions used to generate the curve are described in Appendix C. If the quantity limit shown in Figure 4-1 is exceeded, but the surface radiation level does not exceed 0.5 mrem/h, it may still be reasonable to ship the material as Limited Quantity, given the inherent inaccuracy of the field assays. Moreover, this fact lends appeal to the strategy of packaging samples so that the limiting dose rate is not exceeded, regardless of the soil activity and package weight. Some allowance should be made, however, for ingrowth of radon and radon daughters if the shipment will require more than a few days to reach its destination.

4.6.3 Determination for Exclusive-Use LSA Shipments

4.6.3.1 Procedure

1. The Ra-226 concentration of the material may not exceed 0.1 microcurie per gram.

2. Measure the surface alpha activity with a portable field alpha detector. If the surface activity does not exceed the limits specified in Section 4.5.2, go on to Step 3. If the surface alpha activity exceeds the limits, decontaminate the package using approved methods, and resurvey. If the limits are still exceeded, repackage the contents in a different container. Dispose of the original container appropriately.

3. Label each package "Radioactive - LSA".

4. Measure the dose rate on the accessible external surface of the package. After loading in the transport vehicle, measure the dose rate on the external surface of the vehicle, at 2 meters from the external surface of the vehicle, and in normally occupied positions within the vehicle. The measurements should comply with the limits specified in Section 4.5.1 of this report.

5. Complete shipping papers and placard the vehicle appropriately. Maintain records of all field measurements, instrument numbers, etc.

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Appendix A

SAMPLE PREPARATION GUIDE FOR RADIUM-226 ANALYSIS OF SOIL

INTRODUCTION

The specific procedure used for preparing a sample for analysis depends on the nature of the sample, the requirements of the analytical procedure, and the types of equipment used at the sample preparation facility. A highly specific protocol would be inappropriate, given the number of laboratories involved in analyzing samples in support of remedial actions. Instead, a generalized step-by-step procedure is presented herein, with a view to ensuring a minimum acceptable standard of sample for analysis; a procedure for handling large-volume samples is also described. In order to satisfy quality-control requirements associated with sample preparation, it is recommended that one-way analysis of variance be performed. An example of its application is presented in the final section of this appendix.

The goal of any sample preparation procedure is to obtain an aliquot that is (1) suitably prepared to meet the requirements of the specified analytical method, and (2) representative of the bulk composition of the original sample. It is therefore imperative that the preparation procedure be formulated in such a way as to ensure that the sample will not be contaminated during handling, and that a homogeneous mixture will result from which an unbiased aliquot may be drawn.

In this discussion of sample preparation procedures (both for normal and large-volume samples), it is assumed that the soil sample being prepared is to be analyzed for radium-226 by gamma spectroscopy. Specifications for other types of analysis (e.g., alpha spectrometry, x-ray diffraction, etc.) may require that these generalized procedures be modified and/or expanded.

DEFINITIONS

- Aliquot - A part of a quantity (sample) that divides the quantity without leaving a remainder. The aliquot is that subsample of the prepared sample that is analyzed by the laboratory.
- Fines - That fraction of a soil that passes through a 2-millimeter sieve.
- Gravel - That fraction of a soil, excluding soil aggregates, that will not pass through a 2-millimeter sieve.
- Reject - The remaining portion of a prepared soil sample that is not used for laboratory analysis.

- Soil - All unconsolidated material typically found on or near the surface of the earth including, but not limited to, silts, clays, sands, gravel, and small rocks [40 CFR 192.11(d)].

GENERALIZED PROCEDURE FOR PREPARATION OF NORMAL SAMPLES

EQUIPMENT AND MATERIALS

- Balance
- Sample Trays
- Sieve, 2-millimeter
- Riffle Splitter
- Oven
- Jaw Crusher
- Rotary Grinder and/or Ball Mill
- Blender
- Plastic Bags or Suitable Containers for Sample Storage
- Marking Pens
- Labels
- Cans, Vials, or Other Suitable Containers for Aliquots
- Small Masonry Trowel, Garden Trowel, or Large Spoon

PROCEDURE

A schematic of this generalized sample-preparation procedure for normal samples is presented in Figure A-1. The special notes cited in certain of the procedural steps are explained in the section that follows the procedure.

Sample-Receiving Documentation

1. Complete and file chain-of-custody form.

2. Assign and record laboratory sample number if different from field sample number.

3. Record appropriate sample information.

Pre-Preparation

4. Scan sample in its bag using a scintillometer. Record count rate, or counts if a fixed count time is used [optional, Note 1].

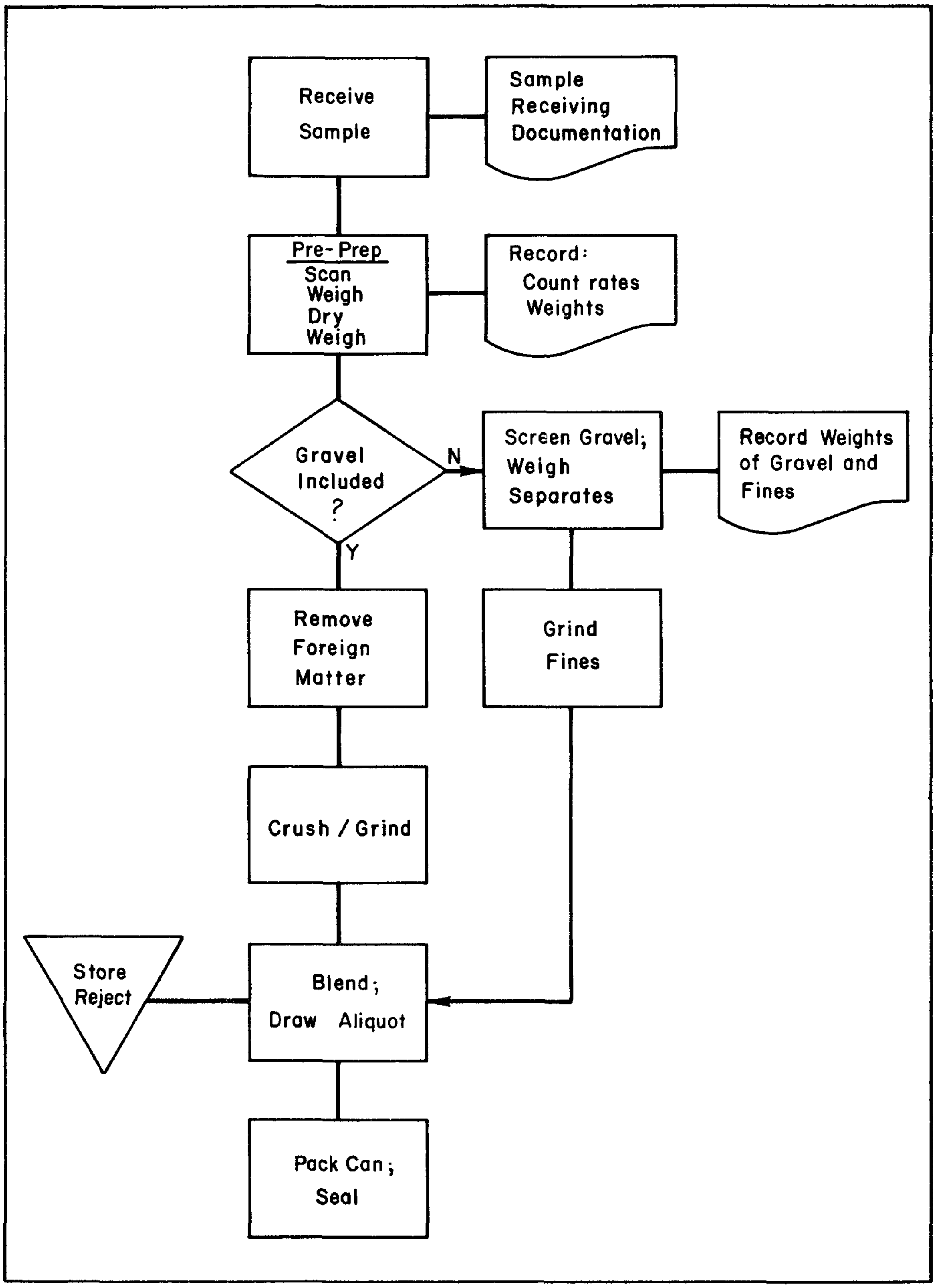

Figure A-1. Schematic of the Generalized Sample-Preparation Procedure for Normal Samples

5. Transfer sample from sample bag to a tared sample tray.
6. Spread sample evenly over tray. Break up large aggregates of soil. Remove and discard leaves, twigs, roots, and other vegetal debris.
7. Weigh sample in tray. Record gross weight.
8. Place tray in oven and dry sample for 24 hours at 100° to 110°C.
9. Remove tray from oven. Allow sample to cool to room temperature.
10. Weigh sample in tray. Record gross oven-dry weight.
11. Calculate and record net dry weight and moisture loss on drying (LOD) [Note 2].

Preparation

12. If gravel is to be included for analysis, go to Step 13. If gravel is to be excluded [Note 3], perform the following tasks:
    a) Sieve sample on 2-mm sieve. Lightly crush soil aggregates and re-sieve. Repeat until only gravel remains on screen.
    b) Place gravel and fines in tared trays. Weigh and record weights of gravel and fines.
    c) Go to Step 15. Continue processing fines.
13. If gravel is to be included for analysis, crush sample in jaw crusher until material is of a suitable size for processing in a ball mill or rotary grinder.
14. Clean jaw crusher thoroughly with compressed air.
15. Place a small amount of sample (20 to 30 grams) in rotary grinder or ball mill and pulverize. Discard [Note 4].
16. Pulverize remainder of sample in rotary grinder or ball mill to -28 mesh.
17. Clean rotary grinder or ball mill with compressed air.

18. Blend sample in mechanical blender.

19. Obtain can or other container which will hold the aliquot for counting. Affix paper label to container. Record laboratory sample number on label.

20. Weigh container to nearest 0.1 gram. Record weight on label.

21. Withdraw aliquot from blended sample. Place in container and hand-pack. Add additional sample until container is full [Note 5].

22. Seal container. Record date and time on label [Note 6].

23. Weigh sealed container to nearest 0.1 gram. Record gross weight on label. Calculate net weight.

24. Let sample stand for 21 days to permit establishment of equilibrium between Ra-226 and its principal gamma-emitting daughters.

Storage

25. Place reject in plastic bag or container labeled with laboratory sample number and field sample number.

26. Place tag marked with laboratory sample number and field sample number in bag or container [Note 7].

27. If gravel was not processed, place gravel in plastic bag or container, and label as in Steps 25 and 26.

NOTES ON PROCEDURE

Note 1. The scintillometer scan is optional but may be beneficial in several ways. For one, it identifies highly radioactive samples that may require special handling for radiation safety purposes. It also provides a basis for sorting samples into groups of similar radiation level for batch processing, either for preparation or laboratory analysis, when cross-contamination is known or suspected to be a problem (cf. Fowler and Essington, 1977, p. 9). It is not desirable to prepare or analyze a high-level sample next to a low-level sample; and the effects of cross-contamination are minimized by handling samples in groups of similar radiation activity. Categories may be defined as order-of-magnitude levels such as 10, 10-100, 100-1000 counts per minute (cpm), or they may be defined on the basis of prior information about the samples (e.g., from in-situ field measurements) or about the site (e.g., from previous reports or analyses). Processing should then proceed from lowest to highest radiation level, giving rigorous attention to cleaning equipment between batches.

Note 2. For certain purposes, it is desirable to express radionuclide concentration in units of activity per unit area. This is facilitated by collecting samples according to a fixed geometry. In remedial action work, laboratory analytical results are often reported as activity per gram. To make the conversion from activity per mass to activity per unit area, it is necessary to know the net dry weight of the sample. Also, net dry weight should be routinely recorded to monitor sample loss during handling. It should be noted that the LOD measurement is seldom an accurate measure of field moisture conditions when obtained from samples shipped in plastic bags. There are instances, however, when even an estimate is useful.

Note 3. The EPA definition of soil includes gravel. Thus, when sample preparation and analysis are conducted in support of remedial actions, the samples should be processed with gravel included. Screening may be necessary, however, to remove objects like nuts, bolts, and nails.

Note 4. This is a very effective method for removing contamination from a grinder before passing the main sample through it. As an alternative for small-volume samples, a similar amount of high-purity, low-radioactivity silica sand, such as Ottawa sand, may be used. Table A-1 presents data on the effectiveness of this decontamination method.

Note 5. A sample splitter, such as a Riffle Splitter, may be used at this point, although it will probably not be necessary.

Note 6. In certain cases, it may be desirable to re-dry the aliquot prior to sealing the container. The container should be completely full and well-packed to minimize the vertical sorting of various particle sizes that might occur as a result of vibration during subsequent handling. Depending on the geometry of the counting system, such vertical sorting can affect the assay.

Note 7. This is a precaution against the erasure, obscuring, or separation of the external label from the sample bag or container.

Table A-1. Comparison of the Effectiveness of Two Cleaning Procedures Used to Decontaminate Sample Preparation Equipment[a]

Type of Equipment	Contamination (ppm U_3O_8) Air Cleanup	Ottawa Sand and Air Cleanup
Pulverizer		
Blank (Ottawa Sand)	1	1
After 1.5% U_3O_8 Ore	13	1
After 0.1% U_3O_8 Ore	2	<1
After 188-ppm U_3O_8 Sample	1	<1
Blank Ottawa Sand	<1	<1
Y Blender		
After 1.5% U_3O_8 Ore	1	2
Blank	<1	<1
After 0.1% U_3O_8 Ore	<1	<1
After 188-ppm U_3O_8 Sample	<1	1
Blank	<1	1

[a] Ottawa sand, containing about 1 ppm U_3O_8, was run through the equipment after each cleaning procedure and analyzed for U_3O_8 to determine the amount of contamination remaining.

PROCEDURE FOR PREPARATION OF LARGE-VOLUME SAMPLES

APPLICATION

For certain sampling designs, especially those involving composite sampling, samples with volumes exceeding laboratory-equipment capacity may be collected. As a consequence, the sample must be divided into several smaller parts to prepare it for analysis, a process which may make it difficult to obtain homogeneity in the bulk sample. The procedure described below for preparing large-volume samples is an adaptation of the splitting-mixture scheme reported by Nyhan and others (1981). This procedure is to be used in conjunction with the generalized sample-preparation procedure described above.

EQUIPMENT AND MATERIALS

Required equipment and materials are the same as those specified for the generalized sample-preparation procedure for normal samples.

PROCEDURE

A schematic of the preparation procedure for large-volume samples is shown in Figure A-2.

1. Complete sample-receiving documentation and pre-preparation (Steps 1 through 11 of the generalized procedure for normal samples) [Note 1].
2. Divide dry sample into parts I and II using Riffle Splitter.
3. Divide part I into parts 1 and 2, and part II into parts 3 and 4, using Riffle Splitter.
4. Crush, grind, and/or blend parts 1, 2, 3, and 4 separately, according to the generalized procedure for normal samples (Steps 12 through 18).
5. Place parts 1 and 2 together in large sample tray and mix by hand with trowel or spoon.
6. Divide combination 1-2 into mixtures A and B using Riffle Splitter. Mixtures A and B each consist of a mixture of parts 1 and 2.
7. Place parts 3 and 4 together in large sample tray and mix by hand with trowel or spoon.
8. Divide combination 3-4 into mixtures C and D using Riffle Splitter. Mixtures C and D each consist of a mixture of parts 3 and 4.
9. Blend mixtures A, B, C, and D separately in mechanical blender [Note 2].
10. Place mixtures B and C together in large sample tray and mix by hand with trowel or spoon.
11. Divide combination B-C into two parts using Riffle Splitter. The resulting parts are a mixture of parts 1, 2, 3, and 4.

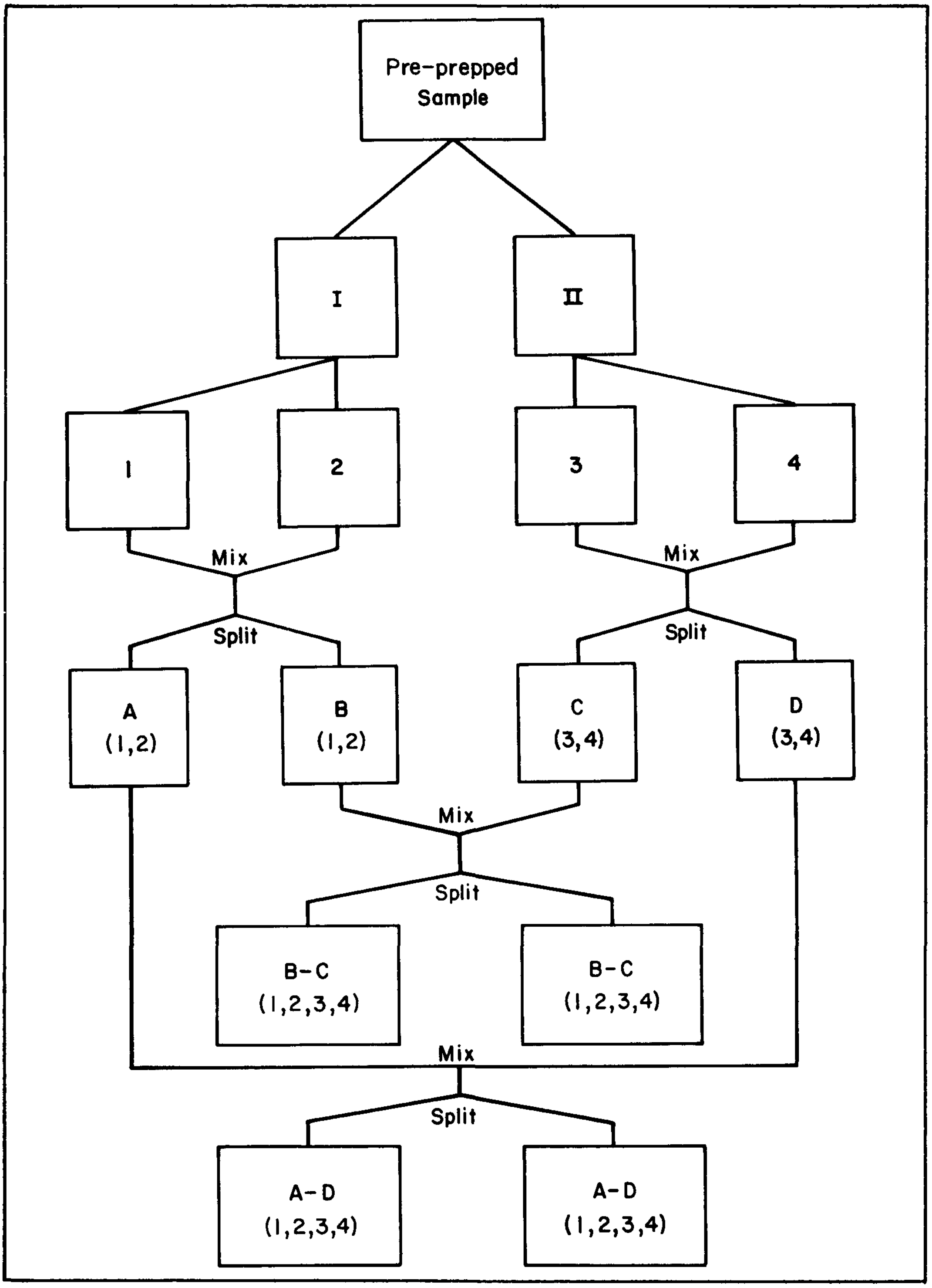

Figure A-2. Schematic of the Sample Preparation Procedure for Large-Volume Samples

12. Place mixtures A and D together in large sample tray and mix by hand with trowel or spoon.

13. Divide combination A-D into two parts using Riffle Splitter. The resulting parts are mixtures of parts 1, 2, 3, and 4.

14. Continue sample preparation beginning with Step 19 of the generalized procedure for normal samples [Note 3].

NOTES ON PROCEDURE

Note 1. It is desirable to proceed as far as possible with sample preparation before dividing the sample. Therefore, if possible, crush and grind the sample to -28 mesh before dividing.

Note 2. Blending mixtures A, B, C, and D in Step 9 may be unnecessary.

Note 3. It is desirable to reblend the final mixture separately before withdrawing the aliquot(s). The procedure for aliquoting should be given careful consideration. Composite samples are normally taken to obtain an average value of the several combined samples when time, expense, or other considerations do not permit analysis of the samples individually. Analyzing four aliquots drawn from the four mixtures defeats this purpose if the large-volume sample was a composite. (This should be done at least once, however, to test the effectiveness of the sample-splitting scheme.) One alternative is to mix the four mixtures together in a large sample tray and draw the aliquot from the whole sample, the drawback being that such a mixture may not be uniform.

PROCEDURE FOR DOCUMENTING THE ADEQUACY OF SAMPLE PREPARATION

GENERAL DISCUSSION

Sampling uncertainty is generally much greater than analytical uncertainty, but the latter is easier to specify and document. In documenting the analytical uncertainty of radium-226 analyses, at least two sources of error should be considered. One is the counting error, which may be estimated either experimentally by replicate counting of a sample or by theoretical calculations. The other is the subsampling or aliquoting error, which arises when a subsample is drawn from a soil sample that is not perfectly homogeneous. This source of error may be estimated by analyzing multiple aliquots of a sample. A sample preparation procedure may be deemed adequate for radium-226 analysis when there is no significant difference among the results obtained from aliquots drawn from a particular sample. The equality of aliquots may be tested by means of one-way analysis of variance, an application of which is described below.

EXAMPLE OF ONE-WAY ANALYSIS OF VARIANCE

Sample Selection and Counting

Four samples of varying radium-226 content were selected from a suite of samples collected at an inactive uranium mill tailings site. Field volume of the samples was approximately 2500 cm^3 each. They were collected in accord with the soil sampling procedures detailed in the body of this report (cf. Section 3), and were prepared for analysis in accord with the procedure for preparing normal samples presented in this Appendix A. Four aliquots of approximately 425 cm^3 each were drawn from each sample. Three 3500-second counts were made on each aliquot to obtain the concentration of radium-226.

The resulting data and simple statistics are presented in Table A-2. Counting errors, expressed as the coefficient of variation, range from almost 20 percent in sample 1 to 1 percent in sample 4. Intuitively, it seems reasonable to assume that subsampling error may become more apparent when counting error is small. In sample 4, for example, aliquot 1 "looks" different from aliquot 2, suggesting that the homogeneity of the prepared sample may be questionable.

Table A-2. Data and Statistics Obtained from Repeat Counts of Aliquots from Contaminated Soil Samples[a]

Parameter[b]	Radium-226 (pCi/g)			
	Aliquot 1	Aliquot 2	Aliquot 3	Aliquot 4
SAMPLE 1				
Count 1	3.26	2.76	3.29	4.10
Count 2	3.86	3.29	3.66	4.20
Count 3	4.10	3.80	4.70	3.46
$\bar{X}_j$	3.74	3.28	3.88	3.92
S_j	0.43	0.52	0.73	0.40
$S_j/\bar{X}_j$	0.11	0.16	0.19	0.10
SAMPLE 2				
Count 1	27.28	23.55	26.71	28.80
Count 2	27.99	26.38	26.88	28.93
Count 3	27.05	27.32	28.43	27.18
$\bar{X}_j$	27.44	25.75	27.34	28.30
S_j	0.49	1.96	0.95	0.98
$S_j/\bar{X}_j$	0.02	0.88	0.03	0.03
SAMPLE 3				
Count 1	59.81	66.29	65.25	62.66
Count 2	61.86	62.76	67.30	63.97
Count 3	64.81	62.50	62.87	63.71
$\bar{X}_j$	62.16	63.85	65.14	63.45
S_j	2.51	2.12	2.22	0.69
$S_j/\bar{X}_j$	0.04	0.03	0.03	0.01
SAMPLE 4				
Count 1	493.31	533.77	537.90	527.42
Count 2	513.88	526.41	515.25	519.05
Count 3	518.18	524.53	514.21	525.40
$\bar{X}_j$	508.46	528.24	522.45	523.96
S_j	13.29	4.88	13.39	4.37
$S_j/\bar{X}_j$	0.03	0.01	0.03	0.01

[a] The field volume of each sample was approximately 2500 cm^3.

[b] $\bar{X}_j$ = mean; S_j = standard deviation; and $S_j/\bar{X}_j$ = coefficient of variation.

One-Way Analysis of Variance

Each of the four samples was then examined separately using one-way analysis of variance to test the null hypothesis (H_0) of equal aliquot means, $\mu 1 = \mu 2 = \mu 3 = \mu 4$, against the alternate hypothesis (H_a) that at least one mean is different. An analysis-of-variance table is formatted as follows:

Source of Variation	Sum of Squares	Degree of Freedom	Mean Square	F-Value
Among Aliquots	SSA	DFA	MSA	
Within Aliquots	SSW	DFW	MSW	MSA/MSW
Total	SST	DFT	MST	

Let

X_{ij} = the concentration from the ith count of the jth aliquot

m = number of replicate counts performed on each aliquot

n = number of aliquots

N = mn

$\bar{X}$ = total mean calculated from the equation

$$\bar{X} = (1/N) \sum_{j=1}^{n} \sum_{i=1}^{m} X_{ij}$$

$\bar{X}_j$ = mean of the jth aliquot calculated from the equation

$$\bar{X}_j = (1/m) \sum_{j=1}^{n} X_{ij}$$

Calculate degrees of freedom using the following equations:

1. Total degrees of freedom calculated as

$$DFT = N - 1$$

In this case, DFT = 11.

2. Aliquot degrees of freedom calculated as

$$DFA = n - 1$$

In this case, DFA = 3.

3. Within-aliquot degrees of freedom calculated as

$$DFW = DFT - DFA$$

In this case, DFW = 11 - 3 = 8.

Calculate the sums of squares using the following equations:

1. Total sum of squares calculated as

$$SST = \sum_{j=1}^{n} \sum_{i=1}^{m} (X_{ij} - \bar{X})^2$$

2. Aliquot sum of squares calculated as

$$SSA = \sum_{i=1}^{m} m(\bar{X}_j - \bar{X})^2$$

3. Within-aliquot sum of squares calculated as

$$SSW = \sum_{j=1}^{n} \sum_{i=1}^{m} (X_{ij} - \bar{X})^2$$

Calculate the mean squares using the following equations:

1. Total mean square calculated as

$$MST = SST/DFT$$

2. Aliquot mean square calculated as

$$MSA = SSA/DFA$$

3. Within-aliquot mean square calculated as

$$MSW = SSW/DFW$$

Calculate the F-statistic from the equation

$$F_{calc} = MSA/MSW$$

This value, F_{calc}, is compared with an F-value from prepared tables. It is necessary to choose a desired level of confidence, say 0.95, and read the value for (DFA, DFW) degrees of freedom.

If

$$F_{calc} \leq F_{table}$$

the null hypothesis cannot be rejected; that is, no difference can be discerned between the aliquot means on the basis of the data. It is inferred from this result that the preparation procedure yields a reasonably homogeneous sample.

The analysis of variance for the sample data cited in Table A-2 is presented in Table A-3. The tabulated F-value for the 0.95 confidence level and (3, 8) degrees of freedom is 4.07. For each of the four samples, the calculated F-value is less than this tabulated value. Thus, the sample preparation procedure seems adequate for radium-226 analysis.

CONCLUSION

It is recommended that analysis of variance or some similar testing procedure be performed routinely, either by the experimenter submitting samples or by the laboratory, to ensure that sample preparation is adequate. If for a number of samples H_0 is rejected, the sample preparation procedure should be modified to achieve homogeneity. For example, the sample may need to be ground to a finer particle size and/or blended for a longer period of time. The least desirable alternative from an economic standpoint is to analyze several aliquots from each sample to determine the sample mean.

Table A-3. Results of One-Way Analysis of Variance for Data Cited in Table A-2

Sample Number	Source of Variation	Sum of Squares	Degree of Freedom	Mean Square	F_{calc}[a]
1	Among Aliquots	0.77	3	0.26	
	Within Aliquots	2.31	8	0.29	0.90
	Total	3.08	11	0.28	
2	Among Aliquots	10.19	3	3.40	
	Within Aliquots	11.88	8	1.49	2.28
	Total	22.07	11	2.01	
3	Among Aliquots	13.56	3	4.52	
	Within Aliquots	32.39	8	4.05	1.12
	Total	45.95	11	4.18	
4	Among Aliquots	661.04	3	220.3	
	Within Aliquots	797.66	8	99.7	2.21
	Total	1458.7	11	132.6	

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[a] F_{calc} = F-calculated. For F-tabulated, F (0.95, 3, 8) = 4.07.

Appendix B

ASSIGNMENTS OF A_1 AND A_2 QUANTITIES FOR LEAD-214, BISMUTH-214, AND POLONIUM-214

SUMMARY OF THE REGULATIONS FOR CALCULATING ASSIGNMENTS

For radionuclides of known identity for which A_1 and A_2 quantities are not specified in 49 CFR 173.435, the quantities must be calculated in accord with the guidelines set forth in Section 173.433(a)(2). These guidelines are summarized below.

DETERMINATION OF A_1 QUANTITY

According to the requirements of 49 CFR 173.433(a)(2)(i), the A_1 quantity for a radionuclide that emits only one kind of radiation (gamma, X, beta, or alpha) is determined in accord with rules A, B, C, or D, respectively (see below). For those radionuclides that emit more than one kind of radiation, A_1 is taken to be the most restrictive (smallest) quantity of those determined for each kind of radiation emitted. If the radionuclide decays to a shorter lived daughter with a half-life less than 10 days, A_1 must be calculated for the parent and the daughter, and the more restrictive of the two assigned to the parent. In any case, A_1 shall not exceed 1000 curies.

Rule A - Gamma-Ray Emitters

A_1 = 9/gamma

where gamma is the gamma ray constant in roentgen-meters per hour-curie, approximately equal to 0.5N [E_{gamma}(average)] where N is the number of gamma quanta per disintegration and E_{gamma}(average) is the average gamma energy in MeV (U.S. Department of Health, Education, and Welfare, 1970, p. 33).

Rule B - X-Ray Emitters

For $Z < 55$, A_1 = 1000 curies
For $Z > 55$, A_1 = 200 curies

where Z is the atomic number of the radionuclide.

Rule C - Beta-Ray Emitters

A_1 is determined by the maximum beta energy, E_{max}, according to the following specifications:

Maximum Beta Energy (MeV)	A_1 Quantity (curies)
$E_{max} < 0.5$	1000
$0.5 \leq E_{max} < 1.0$	300
$1.0 \leq E_{max} < 1.5$	100
$1.5 \leq E_{max} < 2.0$	30
$E_{max} \geq 2.0$	10

Rule D - Alpha-Ray Emitters

$A_1 = 1000(A_3)$

where A_3 is determined from the relationships detailed below, based on the atomic number of the radionuclide, Z, and the half-life, $T_{1/2}$.

For $Z \leq 81$,

Range of Half-Life	A_1 Quantity (curies)
$T_{1/2} < 10^3$ days	3
10^3 days $\leq T_{1/2} \leq 10^4$ years	0.050
$T_{1/2} > 10^4$ years	3

For $Z \geq 82$,

Range of Half-Life	A_1 Quantity (curies)
$T_{1/2} < 10^3$ days	0.002
10^3 days $\leq T_{1/2} \leq 10^4$ years	0.002
$T_{1/2} > 10^4$ years	3

DETERMINATION OF A_2 QUANTITY

Pursuant to 49 CFR 173.433(a)(2)(ii), the A_2 quantity is the more restrictive of the following:

- The corresponding A_1 value.
- The value of A_3 as determined from Rule D above.

ASSIGNMENTS OF A_1 AND A_2 QUANTITIES FOR LEAD-214, BISMUTH-214, AND POLONIUM-214

DATA REQUIRED FOR CALCULATING A_1 QUANTITIES

Lead-214

- Emissions: Beta Rays, Gamma Rays, X Rays
- $T1/2$: 26.8 Minutes
- Feeds: Bi-214 ($T_{1/2}$ = 19.9 minutes)
- Z: 82
- $E_{(max)}$ (beta): 0.980 MeV (U.S. Department of Health, Education, and Welfare, 1970, p. 94)
- N_{gamma}: 22 (Kocher, 1981)
- E_{gamma} (average): 0.2807 MeV (Kocher, 1981)

Bismuth-214

- Emissions: Beta Rays, Gamma Rays, X Rays
- $T1/2$: 19.9 Minutes
- Feeds: Po-214 ($T_{1/2} = 1.6 \times 10^{-4}$ second)
- Z: 83
- E_{max} (beta): 3.20 MeV (Kocher, 1981)
- N_{gamma}: 182 (Kocher, 1981)
- E_{gamma} (average): 1.158 MeV (Kocher, 1981)

Polonium-214

- Emissions: Alpha Rays, Gamma Rays
- T1/2: 1.6×10^{-4} second
- Feeds: Pb-210 ($T_{1/2}$ = 22.26 years)
- Z: 84
- N_{gamma}: 3 (Lederer and Shirley, 1978)
- E_{gamma} (average): 0.797 MeV (Kocher, 1981)

A_1 AND A_2 ASSIGNMENTS

The assignments of A_1 and A_2 quantities for lead-214, bismuth-214, and polonium-214 are presented in Tables B-1 and B-2, respectively.

Table B-1. A_1 Quantities for Pb-214, Bi-214, and Po-214

Isotope	Calculated A_1 Quantity (curies)				Assigned A_1 Quantity (curies)
	Gamma	X	Beta	Alpha	
Pb-214	1.5	200	300	NA	0.09
Bi-214	0.09	200	10	NA	0.09
Po-214	10.8	NA	NA	2	2

Table B-2. A_2 Quantities for Pb-214, Bi-214, and Po-214

Isotope	A_2 Quantity (curies)
Pb-214	0.002
Bi-214	0.002
Po-214	0.002

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Appendix C

QUANTITY LIMITS FOR MIXTURES OF RADIOACTIVE MATERIALS

EQUATIONS FOR DETERMINING QUANTITY LIMITS

The derivation of equations for calculating quantity limits for packages containing mixtures of radioactive material is given below.

Let

R_k, $k = 1, n$	designate the k^{th} radionuclide in a material containing n different radionuclides.
Q_k	be the quantity in curies of R_k in the mixture.
$A_2(R_k)$	designate the A_2 quantity in curies of R_k.
C_k	designate the average concentration of R_k in curies per kilogram.
M	be the mass of radioactive material in kilograms.

Also, pursuant to 49 CFR 173.433(b)(3), let F_k designate the fraction relating the quantity of R_k to its A_2 quantity, defined by the relationship

$$F_k = \frac{Q_k}{A_2(R_k)} \qquad \text{(C-1)}$$

RULE: In the case of a mixture of radionuclides, where the identity and activity of each radionuclide are known, the permissible activity of each radionuclide, R_1, $R_2, \ldots R_n$, must be such that

$$1 \geq \sum_{k=1}^{n} F_k \qquad \text{(C-2)}$$

Since $Q_k = C_k M$, equation (C-2) becomes

$$1 \geq M \sum_{k=1}^{n} C_k/A_2(R_k) \qquad \text{(C-3)}$$

Equation (C-3) can be used to determine the permissible weight of radioactive material when the concentrations of component radionuclides are known.

LIMITED QUANTITY SHIPMENTS OF CONTAMINATED SOIL

For solid materials, the regulations specify quantity limits of $10^{-3}A_2$ (49 CFR 173.423). Equation (C-3) must therefore be modified to the form

$$1 \geq 10^3 M \sum_{k=1}^{n} C_k / A_2(R_k) \qquad \text{(C-4)}$$

The radioactivity of uranium mill tailings is primarily supported by thorium-230 since most of the uranium in the ore was removed. Assuming that thorium-230 and its daughters have equal activities (equilibrium), and treating the material as a mixture of ten different radionuclides, the package quantity limits for Limited Quantity shipments are restrictively small. One interesting approach that evolved from a discussion of this problem (N. Johnson, Eberline Instrument Corporation, personal communication) was to subdivide the thorium-230 chain into four subchains as follows.

Subchain	Components	A_2 Quantity (curies)
1	Th-230	0.003
2	Ra-226 Rn-222 Po-218 Pb-214 Bi-214 Po-214	0.05
3	Pb-210 Bi-210	0.2
4	Po-210	0.2

Subchains 2 and 3 may each be considered a single radionuclide since no daughter has a half-life that is longer than that of its parent nor longer than 10 days. If this scheme is valid, mill tailings and tailings-contaminated soil may be regarded as a mixture of four radionuclides, making shipment under the Limited Quantity exclusion less restrictive in terms of activity and weight.

Using equation (C-4) and the four A_2 values cited above, and assuming equilibrium, package limits for Limited Quantity shipments were calculated. The results are listed below, and plotted in Figure 4-1.

Radium-226 Concentration		Package Weight	
pCi/g	Ci/kg	kg	lbs
200	$2x10^{-7}$	13.8	30.4
300	$3x10^{-7}$	9.2	20.2
400	$4x10^{-7}$	6.9	15.2
500	$5x10^{-7}$	5.5	12.1
600	$6x10^{-7}$	4.6	10.1
700	$7x10^{-7}$	3.9	8.6
800	$8x10^{-7}$	3.4	7.5
900	$9x10^{-7}$	3.0	6.6
1000	$1x10^{-6}$	2.7	5.9
1100	$1.1x10^{-6}$	2.5	5.5
1200	$1.2x10^{-6}$	2.3	5.1