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MEASUREMENTS OF RADIONUCLIDES IN THE ENVIRONMENT SAMPLING AND ANALYSIS OF PLUTONIUM IN SOIL

A. INTRODUCTION

Paragraph (e) of § 20.106, "Concentrations in effluents to unrestricted areas," of 10 CFR Part 20, "Standards for Protection against Radiation," provides that the Commission may limit the quantities of radioactive materials released in air or water by licensees during a specified period of time if it appears that the daily intake of radioactive materials from air, water, or food by a suitable sample of an exposed population group, averaged over a time period not exceeding one year, would otherwise exceed specified quantities. Section 20.201, "Surveys," of 10 CFR Part 20 requires that a licensee conduct surveys of levels of radiation or concentrations of radioactive material as necessary for compliance with AEC regulations in Part 20. Paragraph (c) of § 20.1, "Purpose," of 10 CFR Part 20 states that every reasonable effort should be made by AEC licensees to maintain radiation exposures, and releases of radioactive materials in effluents to unrestricted areas, as far below the limits specified in Part 20 as practicable, i.e., as low as is practicably achievable, taking into account the state of technology, and the economics of improvements in relation to benefits to the public health and safety and in relation to the utilization of atomic energy in the public interest.

This guide describes procedures acceptable to the Regulatory staff for sampling and analysis of plutonium in soil with the sensitivity and accuracy needed to adequately monitor plutonium in soil in the environs of fuel reprocessing and fuel fabrication facilities.

B. DISCUSSION

The Regulatory staff has reviewed and evaluated the data on plutonium in environmental and biological samples and has concluded that plutonium concentrations in these media are generally low and often below the detection limit of state-of-the-art equipment, and should be of little significance in terms of exposure to humans. Nevertheless, the long half-life (24,390 years) of the predominant plutonium isotope, Pu-239, coupled with its high relative radiotoxicity, make it desirable to document and periodically reassess its distribution and fate in the environment.

A soil sampling and analysis program provides the most direct means of determining the concentration and distribution of radionuclides in the environs of nuclear facilities. Hence, it would be desirable to include in environmental monitoring programs, a program for sampling and analyzing soil for plutonium. A soil analysis program would have the most significance for the preoperational monitoring program since it would serve to establish baseline concentrations of plutonium prior to operation of the facility. Soil analysis, although useful in special cases involving unexpected releases, is a poor technique for assessing small incremental releases and is therefore not recommended as a method for monitoring routine releases of radioactive material. Nevertheless, because soil is an integrator and a reservoir of long-lived radionuclides, and serves as an intermediary in several of the plutonium pathways of potential importance to humans, for example, resuspension and plant uptake, knowledge of the buildup of plutonium and other long-lived radionuclides in soil is essential. A soil-monitoring program conducted annually should be adequate to assess the cumulative deposit of plutonium in soil.

C. REGULATORY POSITION

The sampling and analytical procedures described in the appendices to this guide are acceptable to the Regulatory staff as bases for meeting the performance standards required to adequately inventory the plutonium deposited in the environs of nuclear facilities. Other procedures selected for sampling and analyzing plutonium in soil should conform to similar standards of performance.

APPENDIX A

SOIL SAMPLING AND SOIL SAMPLE PREPARATION

No single soil-sampling method is adequate to sample all soil types at all locations. For example, a method designed to sample cohesive sandy loam soil may not be suitable for sampling the dry loose soil common to some arid areas of the U.S. Rocky soils present problems for all sampling methods. It is necessary, therefore, that each situation be handled on a case-by-case basis and that the procedure be adjusted appropriately in a given situation. Two soil-sampling procedures are described here—the method described in HASL 300, [1] which can be used for most soil types, and a more specialized procedure for sampling dry sandy soil. [2] The techniques and principles embodied in these procedures are generally applicable to most situations and should be used as guides for sampling soils at specific sites.

[1] "Procedure Manual," HASL 300, Health and Safety Laboratory, U.S. Atomic Energy Commission, 376 Hudson Street, New York, New York.

[2] Soil sampling method used by the Nevada Applied Ecology Group at Nevada Test Site, which has been modified by the Regulatory staff.

I. HASL Method for Soil Sampling and Soil Sample Preparation

A soil sampling and analysis program provides an acceptable method of assessing long-term buildup of long-lived radioactive contaminants in the environment. Surface soil analysis can also serve to define contamination contours or distribution patterns soon after a hypothetical acute airborne release of a contaminant. The latter would require sampling of only the top 5 cm of soil, including the vegetation. Experience indicates that attempts to sample a shallower depth result in less reproducible samples. In many areas, a site meeting the desired criteria has a root mat extending several centimeters into the ground, and it is rarely possible to remove an intact core less than 5 cm in depth.

A. Site Selection

When soil is sampled as part of a preoperational survey around a plant, it is desirable to select areas that can be resampled at a later time should it become necessary. Figure 1 shows a suggested distribution of

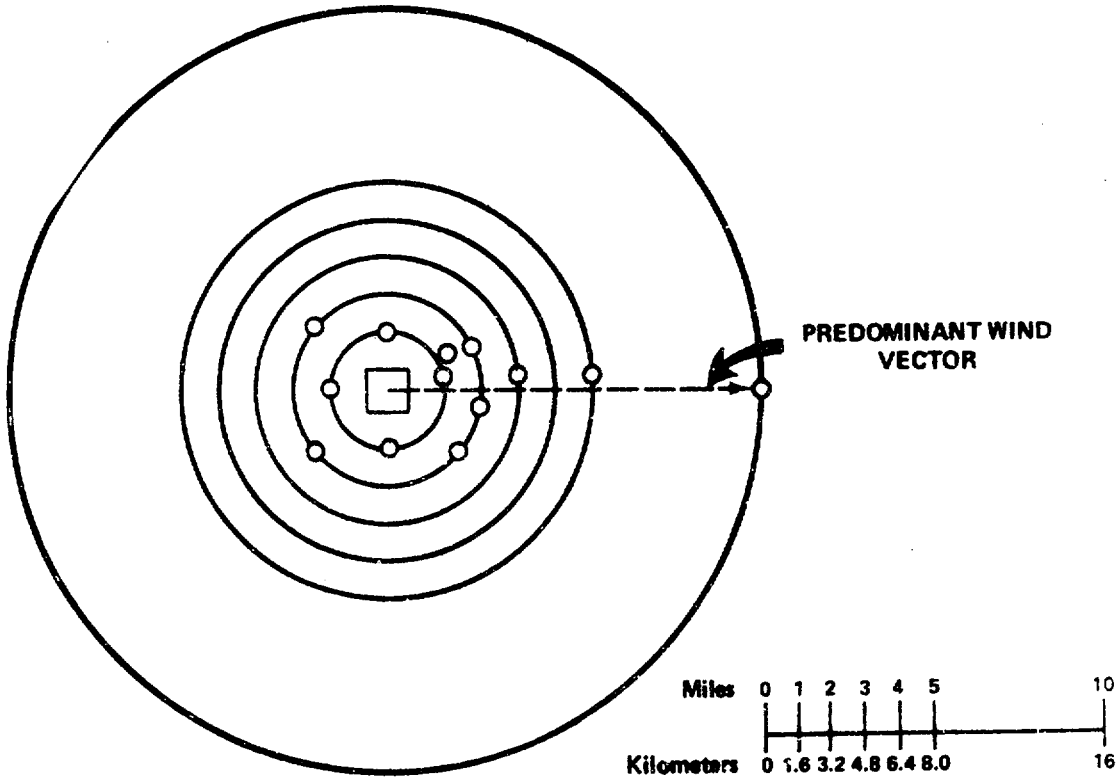

Figure 1. PRELIMINARY SOIL SAMPLING SITES NEAR A NUCLEAR FACILITY

sampling sites covering the area surrounding the plant, with emphasis on the downwind direction. About 13 sites, with the furthest extending 16 km (10 miles) downwind, should give an adequate picture of background levels in the environs of a plant. If necessary, sampling at this same array of sites would provide a preliminary picture of the contamination pattern following a release. It is also suggested that one or more samples be taken close to the center of the most heavily populated area in the vicinity of the plant.

The procedure described here is designed to obtain samples that will measure the total amount of an initially airborne contaminant that has fallen out in a given area. It will not evaluate the unusual case where excessive accumulations occur in low spots, at the foot of slopes, or in flooded areas.

The site should be nearly level with moderate to good permeability. There should be little or no runoff during heavy rains and no overwash at any time. Such a site is frequently found on smooth ridge crests, level virgin land, and well-kept lawns and grounds around institutional buildings. The site should not be near enough to buildings or trees to be sheltered during blowing rains. Soils having very high earthworm activity should be avoided because of uneven mixing of the soil to considerable depths.

B. Core Method

Experience has shown that a total sample area of 460 to 930 cm^2 (1/2 to 1 ft^2) will provide a reasonably good estimate of total deposit if the area consists of a composite of ten or more individual plugs or cores. A tool for taking samples may be anything that takes a core or plug that is of equal area throughout its entire depth. A good pair of sampling tools is an 8.9-cm-diameter (3-1/2-in.-diameter) topsoil cutter that takes a 5-cm-deep (2-in.-deep) sample and a 8.3-cm-diameter (3-1/4-in.-diameter) barrel auger that cuts an 8.9-cm-diameter (3-1/2-in.-diameter) sample.

The topsoil cutter is used to remove the sod to a depth of 5 cm, and the auger takes the remaining sample to the depth desired. The soil from ten cores sampled to a depth of 30 cm (12 in.) is composited to make a single sample weighing from 18 to 36 kg (40 to 80 pounds). If desired, the 0 to 5-cm sod samples may be kept separate, making a sample of higher concentration. The amount of contaminant found in the upper 5 cm and that found in the remaining subsurface are added to give the total for the 620 cm^2 (0.67 ft^2) of surface represented by the ten cores.

Powdery, dry, loose, single-grain soils cannot be sampled in a simple, satisfactory way by the core method. It is best to take samples when the soil contains enough moisture to be cohesive, even if this necessitates that the area to be sampled be wetted by sprinkling.

The procedure after the selection of an undisturbed site that meets the criteria previously discussed is as follows: Lay out a straight line transect about 4.6 m (15 ft) long. Since it may be desirable to resample the site at a later time, measure the coordinate distances to fixed landmarks to identify the relative position of the transect.

Press the 5-cm-depth topsoil cutter into the ground without twisting or disturbing the grass cover or surface soil. This may best be accomplished by stepping on the rim of the cutter with both shoe heels. If more force is required to press the cutter into the ground, a rubber mallet may be used. Gently twist the handle of the cutter to cleanly remove the topsoil plug. Take a total of ten topsoil cores in a straight line, about 30 cm apart, placing the cores in a plastic bag. (The total area sampled is 620 cm^2.)

Sometimes it may not be possible to remove a 5-cm-deep plug cleanly because of a thick root mat. If the topsoil and bottom soil are to be combined, a 10- or 15-cm-depth cutter may be used to remove the topsoil by pounding it partway into the ground with the rubber mallet until it is possible to remove the core intact. Topsoil depth cannot be measured accurately by this method.

C. Depth Profile

An area where there are no rocks and stones and very few pebbles is suitable for taking soil samples at various depths. A sandy loam type soil such as is found on Cape Cod or eastern Long Island can be sampled satisfactorily. These conditions, of course, are rarely found in the areas of interest. Consequently, the attempted use of the method described below runs the risk that subsoil layers may be contaminated with higher specific activity from upper layer soil.

1. Procedure

As in the core sampling method, the depth profile samples are taken so that the weight and depth of the material collected can be directly related to the surface area. To the extent that grass cover and terrain affect the choice of sampling area, the site selection criteria previously described apply. Lay a tarpaulin (about 2 m square) on the ground near the clipped area. Dig a trench about 60 cm wide by 90 cm long by 60 cm deep immediately adjacent to the clipped area, placing the removed dirt on the tarpaulin. Usually, the sod can be cut out in blocks, making it easy to replace after sampling. Smooth the long side of the trench adjacent to the clipped area with a flat blade shovel or mortar trowel, making it perpendicular to the surface. Take the first 5-cm increment by pushing a three-sided square pan with cutting edges on the open side (15 cm x 15 cm x 5 cm deep) into the side 5 cm below the ground surface. Use a sharp flat-bladed knife to score the edges, then

remove the first cut and seal in a small plastic bag. Cut away the topsoil on either side of the first cut to make a shelf about 45 cm long by 15 cm wide and 5 cm from the surface. Lightly brush away any particles that may have fallen on the shelf. Again, push the open-end cutting pan into the side, cut, and remove the next 5-cm-thick incremental sample. Continue this procedure until samples have been taken to the desired depth. The actual depth of a cut can be determined between cuts by placing a two by four on the surface and measuring the distance from the lower surface of the two by four to the subsurface.

When all of the samples have been taken, fill the trench with dirt from the tarpaulin, and replace the sod taken from the trench.

A depth profile is useful only for finding the relative vertical distribution of the radionuclide. Therefore, it is necessary to sample deeply enough that close to 100 percent of the radionuclide is measured. Since only 230 cm^2 of surface area at one spot is sampled when depth increments are taken, the integrated deposit is not necessarily representative of the area.

2. Equipment

Three-sided square pan with cutting edges on open side (15 cm x 15 cm x 5 cm deep made of 0.16-cm-thick cold-rolled steel, welded at the corners).
Plastic bags, 52 x 23 x 0.02 cm (20½ x 9 x 0.008 in.)
Mortar trowel
Long flat-bladed knife
Meter stick (or 24-in. ruler)
One 1.5 m length of two by four
Tarpaulin

D. Soil Sample Preparation

1. Procedure

a. Spread out sample on a plastic sheet and allow it to air dry. This may take three days or more.

b. Break up soil aggregates and pull apart the topsoil plugs (consisting of vegetation and root mat). With a pair of scissors, cut up the vegetation so that it can eventually be distributed homogeneously.

c. When the sample is completely dry, weigh the entire sample to ±50 grams.

d. Remove large rocks (> 2.5 cm), weigh separately, and discard. (For gravelly soil, it may be desirable also to screen out the greater-than-2-mm fraction after appropriate treatment of the sample to break up soil aggregates.)

e. Crush the entire soil sample, then blend for about 30 minutes.

f. Spread out the sample, mark off quarters, and take scoopfuls from each quarter consecutively until approximately 3 kilograms have been collected.

g. Pass this subsample of soil through a grinder or pulverizer and transfer to a one-gallon wide-mouth polyethylene bottle.

2. Equipment

Scale–Capacity of ~50 kg
Crusher–To precrush rocks to more suitable size for pulverizer
Pulverizer–For pulverizing to about 100 mesh
Blender–Capacity ~0.056 m^3 (2 ft^3)

E. Reporting of Data

Results should be expressed in nanocuries per gram of dry soil (total soil); the field bulk density should be recorded, as well as the area and depth sampled, to provide information necessary to also calculate and express contaminant activity per unit area.

II. Method of Nevada Applied Ecology Group for Sampling and Sample Preparation of Nevada Test Site Soil (modified for Regulatory purposes)

The method developed specifically for the Nevada Test Site under the auspices of the Nevada Applied Ecology Group should be applicable to other similar soils. The "ring" and "trench" methods have been used by the Nevada Applied Ecology Group for soil sampling on or around the Nevada Test Site. Either method can be used to obtain surface samples (defined as the upper 5 cm of soil) or profile samples.

A. Ring Method

In the ring method, a ring (12.7 cm ID x 2.5 cm deep) is pressed into the soil. Soil inside the ring is removed with a disposable plastic spoon to a depth of 2.5 cm and is bagged. Soil is next removed from outside the ring to the 2.5 cm depth, the ring is pressed into the soil another 2.5 cm, and another sample is taken with a second spoon. In this manner, profile samples can be taken to a desired depth; at lower depths where radionuclide concentrations may be low, it may be desirable to increase increments to multiples of 2.5 cm.

A sample consists of soil taken from a minimum depth of 5 cm. A minimum of five separate samples should be taken along a straight line transect and composited for analysis. Since it may be desirable to resample the site at a later time, the coordinate distances of the transect should be measured to fixed landmarks to identify the relative position of the transect.

B. Trench Method

A rectangular trench of appropriate size is dug 15 to 25 cm deeper than the desired sampling depth. Samples are taken from a trench wall with a three-sided tray, 10 cm wide by 10 cm long by 2.5 cm deep.

The procedure for taking a sample is as follows: The tray is pushed in from the side of the trench with the top edges of the tray flush with the surface of the ground. After the tray is pushed into position, press down with a trowel or a thin, flat piece of metal approximately 15 cm wide above the open-sided front end of the tray. With the metal or trowel in place, the soil outside the tray is scraped off down to the depth of the tray. The separated soil is removed and bagged, and the process is repeated until blocks of soil have been removed to the desired depth. A sample consists of soil taken from a minimum depth of 5 cm. A minimum of five samples should be taken along a straight line transect, each from a separate trench, and composited for analysis. Since it may be desirable to resample the site at a later time, the coordinate distances of the transect should be measured to fixed landmarks to identify the relative position of the transect.

The trench method is often used for taking depth profile samples to obtain information on the distribution of a contaminant with depth.

C. Factors in Soil Sampling

If care is taken, either method works well in fine-textured soils; however, "stony" soils present difficulties. In stony soils, larger scoops are recommended. Larger scoops and hence larger soil volume will minify perturbations caused by stones, which interfere with the progress of the edges of the scoop as it is passes through the soil. A representative depth, however, is more important than a representative width.

Areas to be sampled should be undisturbed and should be well removed from dusty roads and from sites that show evidence of previous construction. A distance of no less than 120 m (400 ft) from the outer edge of construction is recommended. When it is desirable to sample soil for the measurement of environmental redistribution of radioactive materials by physical, chemical, and biological factors, a random sampling scheme is preferable to a biased selection of sampling sites. A practical method of selecting sampling sites is by locating them randomly by direction from grid points or other fixed points on an area.

Soils should be neither muddy nor dry at the time of sampling; soils with moisture content near field capacity sample best. A fine spray from a sprayer has been employed with success to provide optimum moisture for dry soil; if a spray is used, time (about 30 min) should elapse between its application and sampling to allow for equilibration of moisture.

D. Sample Preparation

Before a weighed portion of the soil is ground in preparation for analysis, it is desirable to oven-dry (110° C) for 24 hours and sieve it. A 0.6-cm-mesh (¼-in. mesh) screen removes larger stones and some organic debris such as roots, straws, etc.; the weight basis is, of course, the total soil. The plutonium contribution of stones larger than 0.6 cm (¼ in.) is generally negligible; however, if desired, the larger stones can be acid-washed and the washings added to the fraction that passes through the 0.6-cm (¼-in.) screen. If the quantity of organic material is not negligible (i.e., if it consists of more than a few strands of roots and/or a few leaves), it should be analyzed separately; the result should be weighted for the amount of organic matter relative to the total sample and added to the result for plutonium in the soil.

The volume or "nature" of soil adhering to roots and other debris may be such as to bias results; such debris may be be washed with a gentle spray from a wash bottle to remove the soil, and the washings may be added to the smaller-than-0.6-cm (¼ in.) fraction. This fraction, plus additions, should be oven-dried again and weighed.

E. Reporting of Data

Results should be expressed in nanocuries per gram of dry soil (total soil); the field bulk density should be recorded, as well as the area and depth sampled, to provide information necessary to also calculate and express contaminant activity per unit area.

III. Discussion

Both the HASL and Nevada Test Site procedures are intended to provide deposition and/or concentration data that are typical of a given area, and are thus based on criteria for obtaining samples that are most representative of that area. It is probable, however, that at some locations of nuclear facilities, these guides cannot be entirely met due to the special nature of the terrain. In such instances, each site should be handled on a case-by-case basis.

For example, if the facility site does not have suitable sampling areas, i.e., large flat open areas covered with grass, a greater number of soil cores should be taken for compositing than from a more suitable area. Using the HASL criterion of ten cores per sample from a good site as a base, a less suitable site may require sampling and compositing of 15 or more cores to obtain a reasonably representative sample.

In areas where the ground is covered with tall grass, it may be necessary to separately sample the grass and

soil. One method for doing this is to measure off a minimum of one meter square of ground and to collect the grass within this area by cropping it to about a two-cm height. The soil within this area could then be sampled by taking five plugs and cores (one taken at each corner and one taken in the center) using the HASL core method described in this appendix. Two of these one-meter-square plots which are spaced at least ten feet apart should provide the necessary number of plugs and cores needed for compositing. The grass could be analyzed separately or added to the soil and analyzed together with the soil. If the latter technique is used, it would be necessary to process the grass first by grinding and/or ashing and then adding a proportionate aliquot of the grass to the soil. Since the primary objective of soil analysis is to obtain representative measurements of contaminants deposited on the ground, it is essential that the vegetation growing on the soil, which usually contains some of the deposited material, also be included in the analysis. When the grass fraction is analyzed separately, the data should be normalized to the area of soil sampled and the result added to the soil data.

Interference from rocks is a common problem. It may be necessary in some instances to sample in a different area if the rock problem is severe in a given area. If moving to another area is not feasible, the sampling procedure should be modified to minimize the effect of the rocks. This may be done by sampling larger diameter cores to deeper depths. All rocks should be included in the sample. Rocks may be removed by sieving after the soil sample has been appropriately dried and weighed.

APPENDIX B

RADIOCHEMICAL ANALYSIS OF PLUTONIUM IN SOIL

The radiochemical analytical procedure described below is based on the procedure currently in use at Los Alamos Scientific Laboratory. Testing by a number of laboratories (Pacific Northwest Laboratory, Battelle Memorial Institute, Los Alamos Scientific Laboratory, Reynolds Electrical and Engineering Company) has shown the procedure described in this guide to be generally applicable for analyzing plutonium in soil, including Nevada Test Site soils. The main features of this procedure include the use of an acid-extraction mixture containing HF, HCl, and HNO_3, Pu-236 or Pu-242 tracer, electrodeposition of the plutonium, and counting by alpha spectrometry. Samples consisting of 10 to 50 g of soil can be readily analyzed by this procedure, using normally available laboratory equipment and materials. Soil samples much larger than this tend to be unwieldy because special equipment and materials such as large centrifuges, large PTFE beakers, etc., are usually required. The analysis of large soil aliquots is desirable, however, because larger aliquots usually provide a more representative sample. In general, it would be poor practice to use aliquot sizes containing less than 10 g of soil unless smaller than 10 g samples are replicated.

A. Principle

Plutonium is extracted from soil with a combination of nitric, hydrofluoric, and hydrochloric acids in the presence of Pu-236 (or Pu-242) tracer. Plutonium is isolated by anion exchange and electrodeposited onto a stainless steel disc for determination by alpha spectrometry.

B. Reagents

Ammonium hydroxide (28%)
Ammonium iodide
Boric acid
Dowex 1 x 4 (100-200 mesh, nitrate form) anion resin
Hydrochloric acid (38%)
Hydrofluoric acid (48%)
Iron carrier (10 mg/ml)
Nitric acid (70%)
Nitric acid (8N)
Octyl alcohol (Reagent grade)
Pu-236 tracer (or Pu-242)
Sodium bisulfite
Sodium hydroxide (50% solution)
Sodium nitrite

C. Special Equipment

Electrodeposition apparatus
Ion exchange column (~1.3 cm ID x ~ 15 cm)
Stainless steel disc (1.9-cm - 2.2-cm diam. polished on one side)
PTFE beakers (250, 400, 600 ml, etc.)
PTFE stirring rods (~0.3 cm x ~ 12 cm)

D. Extraction

1. Weigh a 10-g soil aliquot into a 250-ml PTFE beaker. Add an appropriate quantity of Pu-236 tracer. (Note 1 at the end of this section.)

2. Add 60 ml of HNO_3 (70%) and 30 ml of HF (48%). Digest on a hotplate with frequent stirring for about an hour. (Notes 2 and 3)

3. Remove from the hotplate and cool somewhat before adding 30 ml each of HNO_3 and HF. Digest with some stirring for about an hour.

4. Remove from the hotplate and cool. Carefully add 20 ml of HCl (38%) and stir. Heat on a hotplate for 45 minutes with occasional stirring.

5. Add about 5 g of powdered boric acid, and digest for an additional 15 min. Stir occasionally.

6. Add about 200 mg of sodium bisulfite and digest on a hotplate. Continue heating and evaporate to a liquid volume of ~10 ml.

7. Add ~50 ml of water and digest on a hotplate with stirring for ~10 min to dissolve soluble salts.

8. Cool. Using a wash bottle, transfer approximately equal parts of the total sample into two 220-ml centrifuge bottles. (Note 4)

9. Add 1 ml of iron carrier solution (10 mg Fe/ml) to each centrifuge bottle and stir.

10. Add NaOH (50% solution) with stirring to each bottle to a pH of ~9. Add 5-10 ml excess of NaOH and stir for 1 min.

11. Centrifuge for ~5 min, decant, and discard the supernate.

12. Dissolve the precipitate with about 30 ml of 8N HNO_3 saturated with boric acid. Digest in a hot water bath for 10 min.

13. Cool and centrifuge for ~5 min. Decant the supernate into the original 250 ml PTFE beaker and save.

4. Wash the residue with 8N HNO_3 saturated with boric acid, centrifuge for 5 min, and combine supernates. Discard the residue.

5. Heat the supernate on a hotplate and evaporate to near dryness.

6. Add ~30 ml of water and heat to dissolve the salts. Cool, and transfer equal portions into centrifuge tubes.

17. Add NH_4OH, dropwise with stirring, to a pH of ~9.

18. Centrifuge and discard the supernate.

19. Dissolve the precipitate with a minimum of concentrated HNO_3 (70%) and transfer using 8N HNO_3 solution into a 250-ml beaker. Add 8N HNO_3 to a volume of approximately 75 ml. (Note 5)

20. Add ~200 mg of sodium nitrite ($NaNO_2$) crystals and stir with a stirring rod, bring to a quick gentle boil on a hotplate, and cool. Avoid prolonged heating.

21. Pass through an anion-exchange resin column previously pretreated with 8N HNO_3. Wash with six column volumes of 8N HNO_3. Let the HNO_3 just pass through the column before continuing the wash with six column volumes of 12N HCl. (Note 6)

22. Elute with four column volumes of NH_4I-HCl solution (1 ml 1M NH_4I solution to 20 ml 12N HCl), and collect in a 150-ml beaker.

23. Evaporate on a hotplate to 5 ml and add HNO_3 (70%) dropwise. Rinse down the sides of the beaker dropwise with HNO_3 and add six drops of HCl (38%). Evaporate to near dryness.

24. Add 50 ml of 8N HNO_3 solution and repeat steps 20 through 23, using a fresh anion-exchange resin column. (A smaller ion-exchange column may be used this time.)

25. Continue heating the sample just to dryness. Rinse down the sides of the beaker with concentrated HCl, and evaporate on a hotplate to approximately 1/2 ml. The sample is now ready for electrodeposition.

Notes:

1. An appropriate quantity of Pu-236 tracer is an activity level which is within an order of magnitude of the expected activity level of Pu-239 and Pu-240 in the sample. If Pu-238 determination is also required, it would be desirable to add no more Pu-236 tracer than the expected activity level of Pu-238 in the sample.

2. For larger soil aliquots, larger amounts of the acids (in about the same proportions) should be used. For example, for a 50 g sample, use 200 ml of HNO_3 and 100 ml of HF, etc.

3. For organic soils, first add HNO_3 only, in small portions with stirring. If the solution threatens to overflow as a result of froth generation, add a few drops of octyl alcohol and stir. Digest on a hotplate until the evolution of heavy reddish-brown fumes is reduced to a barely visible level. Cool to room temperature before carefully adding HF.

4. If large centrifuge tubes are not available, it might be expeditious to perform the precipitation in a beaker first, to allow the precipitate to settle somewhat, to decant the supernate, and then to complete the separation by centrifugation.

5. If the volume of the hydroxide precipitate is considerably greater than should be expected from the 10 mg of Fe added, the final volume should be brought up to ~100 ml with 8N HNO_3 or, alternatively, the dissolved hydroxides should be evaporated to salts first before addition of the 8N HNO_3 solution. The final normality of the HNO_3 solution is not extremely critical, but should be in the range of 7-9.

6. In the absence of an excessive amount of salts (and this should be the case with 10-g soil samples), a resin column with dimensions of approximately 1.3 cm ID by 10 cm of wet, settled resin should be adequate.

E. Electrodeposition

1. Add 1-1/2 to 2 ml of 4N HCl into the beaker and, using a "disposable pipette" (2-ml glass eye-dropper type, with 2-ml bulb), rinse down the sides of the beaker with the sample solution. Transfer the solution into a plating cell.

2. Add another 1-1/2 to 2 ml of 4N HCl solution into the beaker, rinse as above, and add to the plating cell.

3. Repeat the above step with ~1 ml of H_2O.

4. Add a drop of thymol blue indicator solution and add concentrated NH_4OH dropwise until the color changes to yellow.

5. Add 2N HCl solution dropwise to a salmon-pink end point.

6. Electroplate at 1.5 A for 20 min.

7. At the end of 20 min, add 2-3 ml of concentrated NH_4OH (two shots with 2-ml disposable pipette), and leave the current on for another 20 sec.

8. Turn the current off, rinse out the solution into a beaker with H_2O, and dismantle the cell. Rinse the disc with H_2O, and dry it on a hotplate at medium heat for 5 min. The sample is ready for counting.

F. Counting

A properly electrodeposited sample should be free of residue. Normally, three plutonium peaks are distinguishable, the Pu-239/Pu-240 peak at 5.11-5.17 MeV, the Pu-238 peak at 5.46-5.50 MeV, and the Pu-236 peak at 5.72-5.77 MeV (or Pu-242 peak at 4.86-4.90 MeV). Since Pu-238 is readily resolved from the other plutonium isotopes, it is often advantageous to use the Pu-238 to Pu-239/Pu-240 ratio as a possible tag for identifying the various sources from which the plutonium may have been released. In those cases where Pu-238 activity is very low, it is well to remember that Am-241 has essentially the same alpha energy (5.44-5.49 MeV) as Pu-238 and will therefore add to the Pu-238 count if present. Although Am-241 is chemically removed from the plutonium fraction by this procedure, fresh Am-241 continues to grow in from the Pu-241 present. Depending on the amount of Pu-241 in the sample and the ingrowth period, Am-241 activity could add significantly to the Pu-238 peak. It is desirable, therefore, that the plutonium be counted as early as possible after its isolation.

Traces of Pu-238 may be present in some Pu-236 sources; because of the relatively short half-life of Pu-236 (1,041 days), this problem worsens with age. Each Pu-236 source should be checked for potential contamination by other plutonium isotopes. Each plutonium isotope should be accurately determined as a fraction of the Pu-236 activity and should be corrected for in the analysis. Older Pu-236 sources also contain U-232 and Th-228 daughters with alpha energies in the 5.3 - 5.4 MeV range. Therefore, the plutonium fraction should be chemically isolated before a check is made for other plutonium isotopes in the Pu-236 source. Plutonium-236 should be corrected for decay if the decay period exceeds 15 days.

Plutonium-242 tracer, if available, could be used instead of Pu-236. There are several advantages to using Pu-242. First, its half-life is long (3.87×10^5 yr), obviating the necessity for decay corrections. Second, since its alpha energy (~ 4.9 MeV) is below the energy of either Pu-238 or Pu-239/Pu-240, the potential problem of "tailing" of a tracer peak into a lower-energy peak region in the detection system is eliminated.