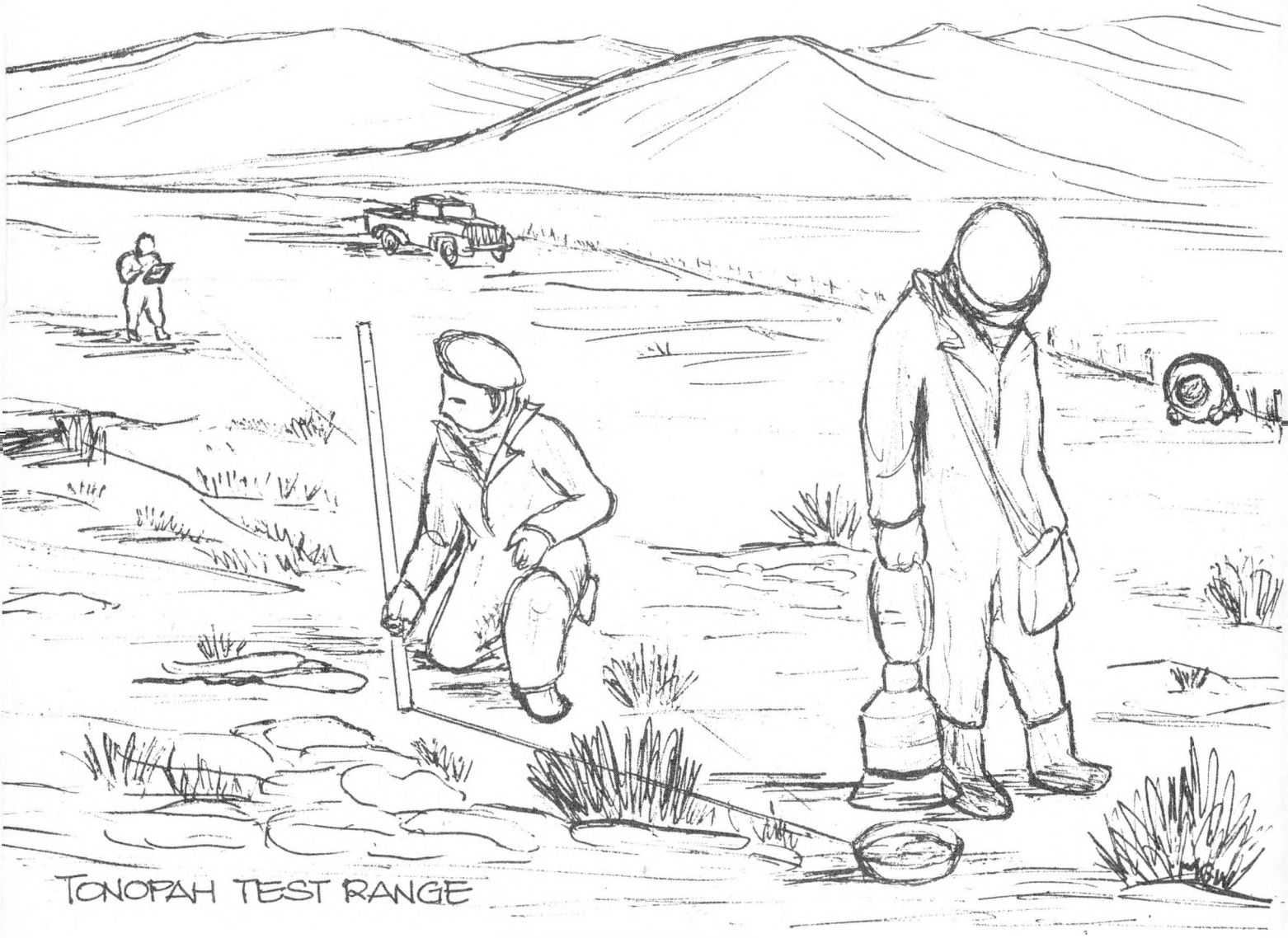
NVO-166
VOLUME 1 of 2

NEVADA APPLIED ECOLOGY GROUP

PROCEDURES HANDBOOK

FOR ENVIRONMENTAL TRANSURANICS

OCTOBER 1976



UNITED STATES
ENERGY RESEARCH & DEVELOPMENT ADMINISTRATION
NEVADA OPERATIONS OFFICE
Las Vegas, Nevada

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FOR ENVIRONMENTAL TRANSURANICS

OCTOBER 1976

EDITED BY

M. G. White & P. B. Dunaway

UNITED STATES

ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION

NEVADA OPERATIONS OFFICE

Las Vegas. Nevada

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*Envelope inside rear cover of Volume II.

Preface

PREFACE

The activities of the Nevada Applied Ecology Group (NAEG) integrated research studies of environmental plutonium and other transuranics at the Nevada Test Site have required many standardized field and laboratory procedures. These include sampling techniques (see illustration on cover), collection and preparation, radiochemical and wet chemistry analysis, data bank storage and reporting, and statistical considerations for environmental samples of soil, vegetation, resuspended particles, animals, and others.

Of course, improvements and/or modifications of the procedures were considered as the studies developed. However, every attempt was made to hold to the most practical procedures which would afford the best comparable results, within the funding allocated. Therefore, the procedures used by NAEG were standardized early in the program operation, as the best methods emerged.

This document, printed in two volumes, includes most of the Nevada Applied Ecology Group standard procedures, with explanations as to the specific applications involved in the environmental studies. Where there is more than one document concerning a procedure, it has been included to indicate special studies or applications perhaps more complex than the routine standard sampling procedures utilized.

The emphasis in these procedures is on applied environmental sampling and laboratory procedures. Nine of the documents included in the handbook are original papers prepared especially for this publication. Other procedure papers are reprinted from previous publications in the open literature.

Many persons have been responsible for contributions to NAEG standard procedures. Some of them are authors of the documents included in this publication, others are not named. Our gratitude is extended to all those headquarters, field, and laboratory people who have contributed to the accomplishment of the goals of the Nevada Applied Ecology Group. Their combined efforts in NAEG workshops, laboratories, and field operations have resulted in marked advances in knowledge of the movement of plutonium and other transuranics through man's environment. Our special thanks for continued encouragement and support are expressed to Gordon Facer, DMA, ERDA/HQ; Mahlon Gates, Manager, ERDA/Nevada Operations Office; Roger Ray, AMES, ERDA/NV; Richard O. Gilbert, BNWL; to the Reynolds Electrical & Engineering Co., Inc. (REECo), personnel assigned to the Nevada Applied Ecology Group operations at the Nevada Test Site; to H. B. Gayle, P. G. Noblitt, J. E. Sanchez, and the Word Processing Center of Holmes & Narver, Inc.; and to Winnie Howard and Don L. Wireman of the NAEG staff.

Mary G. White
Scientific Program Manager
Nevada Applied Ecology Group

Paul B. Dunaway
Chairman, Steering Committee
Nevada Applied Ecology Group

SOIL SURVEYS OF FIVE PLUTONIUM-CONTAMINATED AREAS ON THE TEST RANGE COMPLEX IN NEVADA

Verr D. Leavitt
National Environmental Research Center--Las Vegas

(Ed. Note: Previously published in NAEG Report NVO-142, pp. 21-27.)

ABSTRACT

This report discusses soils in five areas located on the Test Range Complex, Nye County, Nevada. This survey was undertaken as part of the Nevada Applied Ecology Group (NAEG) plutonium studies. A complete report is currently in preparation to be published as NERC-LV-539-28.

Most of the surface soils in the areas have a gravelly texture and are typically classified as gravelly sandy loam. The majority of the surveyed land is either floodplain or alluvial fan with deep soils having well-developed profiles and platy structure. All of the soils are alkaline, ranging in pH from 7.0 to 9.0. Two general categories of vegetation are found in the study areas, low and high desert shrub. The low desert shrubs are predominantly creosotebush (*Larrea divaricata*) and white bursage (*Franseria dumosa*). The high desert shrubs are mostly fourwing saltbush (*Atriplex canescens*), winterfat (*Eurotia lanata*), and bud sagebrush (*Artemisia spinescens*).

INTRODUCTION

The purpose of this soil survey was to gather information on soil and vegetation of Clean Slate 1, 2, and 3, Double Track, GMX, Areas 11 and 13 of the Test Range Complex in Nevada. This was accomplished in conjunction with the NAEG plutonium study. (See report, "Soil Survey of Five Plutonium Contaminated Areas on the Test Range Complex in Nevada," NERC-LV-532-28, in preparation.)

The areas surveyed are located in the south-central and eastern parts of Nye County in southern Nevada (see Figure 1). These areas are semiarid, with annual precipitation ranging from 4 to 8 in (Maxey and Jameson, 1948). All areas have native vegetation, and in the past have been used for grazing livestock. Approximately 7,560 acres, or two-thirds of the acreage, are potential agricultural land if water were available.

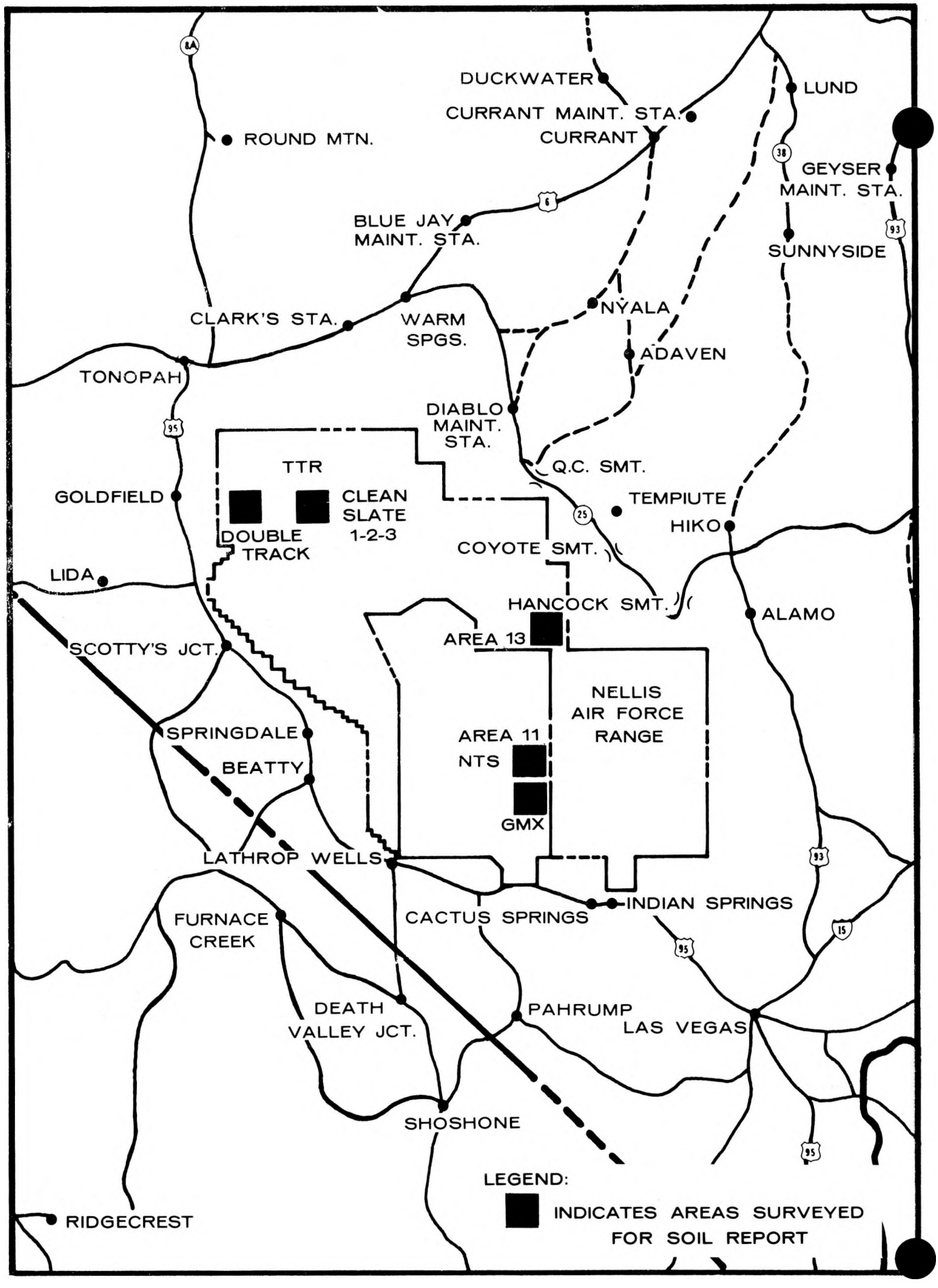

Figure 1. Areas Surveyed

The NAEG intensive site study areas are contaminated with plutonium and are, therefore, the object of investigations regarding the movement of plutonium in the environment.

METHODS

In each soil type, one soil pit, approximately 1 m^2, was dug into the C horizon (generally 1 m deep). From these pits, soil profiles were described by visual inspection. Auger holes or shallow pits were dug approximately every 100 m to confirm the soil type.

Soil order designations were made according to the 7th Approximation published by the Soil Conservation Service (1970). Soil colors were determined by comparison with the Munsell Soil Color Chart (1954). The determination of soil texture (particle size) was made by feeling the soil, and accuracy is, therefore, dependent on the experience of the observer. Soil structure (the aggregation of primary soil particles into compound particles) was described according to the criteria of the United States Department of Agriculture Handbook No. 18 (1951). The pH was determined by using cresol red between pH 7.2 and 8.8, and thymol blue between pH 8.0 and 9.6. Effervescence (an indication of crystalline salts) was determined by adding a few drops of 0.1 N HCl to the soil.

Examples of Data

Table 1 is a summary of dominant soil factors of the five areas that are included in the final soil survey report currently in publication. Other tables are included in that report which compare the different areas and give specific details as to soil type and description. Figure 2 is a representative soil survey map from which Profile No. 401 was taken as an example of descriptive information obtained in the survey. Vegetation growth and possible uses of the land are indicated in each such profile.

AREA 13
Profile No. 401

Soil Order: Aridisol

Typifying Pedon: sandy loam.

Horizon	Depth	
A2	0-15 cm	Very pale brown (10YR 7/3) dry, sandy loam, brown (10YR 4/3) strong thick platy structure;

Table 1.

Summary of Dominant Factors of the Five Areas

	Clean Slate 1, 2, & 3	Double Track	Area 5 GMX	Area 11	Area 13
Surface Soil	gravelly sandy loam	gravelly sandy loam	gravelly sandy loam	gravelly loam	gravelly sandy loam
Horizons*	A_2, B_2, B_3, C_1	C_1, C_2	A_1, C_1, C_2	A_2, B_2, C_1, $C_{2_{cam}}$	A_2, B_2, C_1, C_2
Potential Agricultural Land Acres	5,000	0	0	0	2,560
Land Use Capability Units**	$VIII_c$-K, $VIII_w$-F	VII_s-4, VII_s-8	VII_s-4, VII_s-7	VII_s-4, VII_s-7, VII_s-8	VII_c-K
Surface Structure	platy	subangular blocky	platy	platy	platy
Erosion (water & wind)	moderate wind, slight water	slight water	slight to moderate wind and water	slight wind	slight to moderate wind and water
Range in pH	7.0 to 8.8	8.2 to 9.0	8.0 to 8.8	8.0 to 8.8	7.8 to 8.4
Water Permeability (in/hr)	moderate 0.8 to 2.5	rapid 5.0 to 10.0	rapid 2.5 to 10.0	moderate 0.8 to 2.5	moderate 0.8 to 2.5
Slope (percentage)	level 0 to 2	gently sloping 0 to 4	level 0 to 2	level to steep hilly 2 to 30	level 0 to 2
Dominant Vegetation (species)	fourwing saltbush	shadscale	white bursage	wolfberry	fourwing saltbush

*USDA Soil Survey Handbook No. 18 (1951).
**Soil Survey of the Pahranagat-Penoyer Area, Nevada (1968).

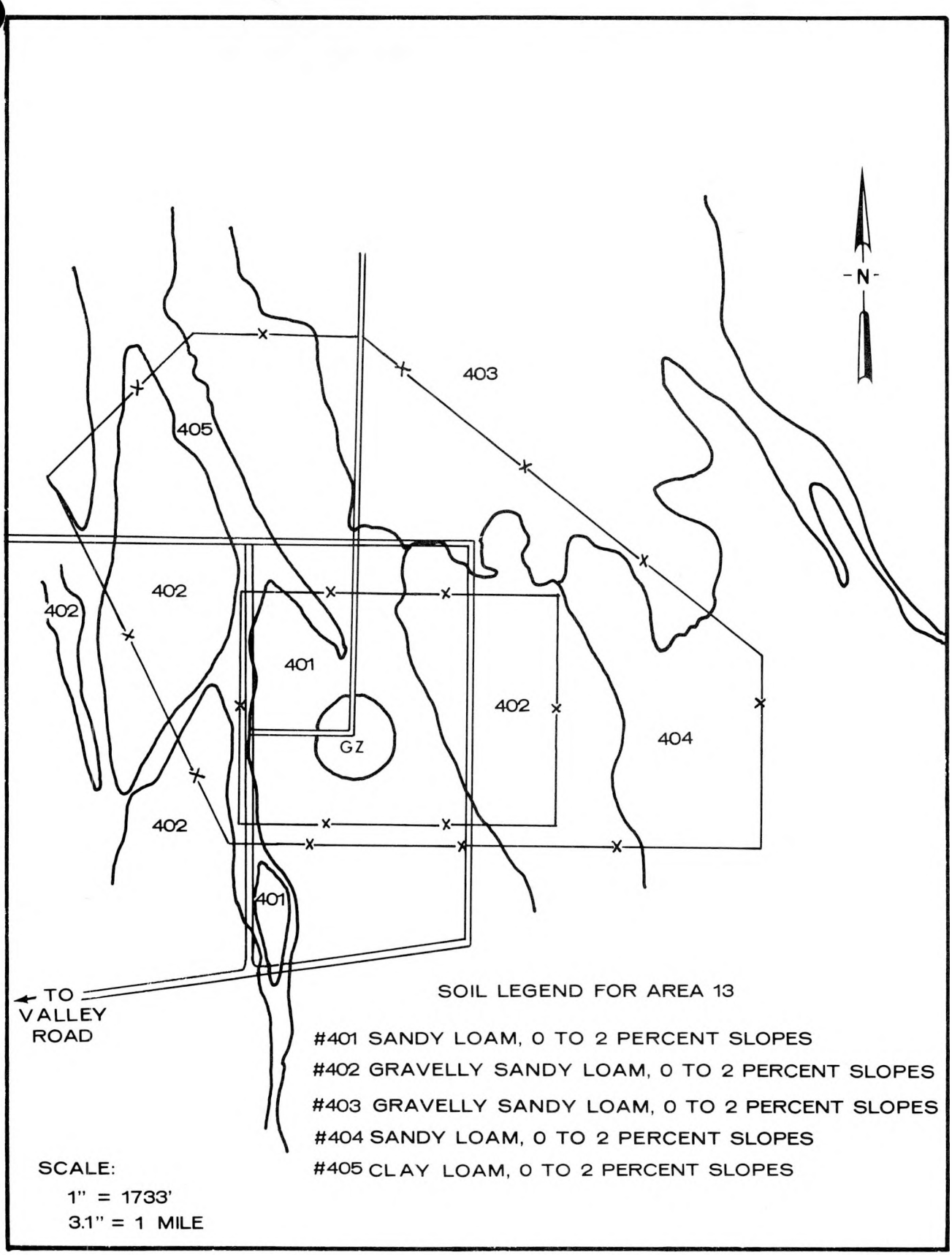

Figure 2. Soil Survey Map of Area 13

Horizon	Depth	
		soft, friable, nonsticky, nonplastic; many fine and medium vesicular pores; strongly effervescent, moderately alkaline (pH 8.0).
B2	15-45 cm	Reddish brown (5YR 5/4) dry, clay loam, reddish brown (5YR 4/4) moderate medium platy structure; slightly hard, friable, sticky, plastic; violently effervescent, moderately alkaline (pH 8.4).
C1	45-91 cm	Brown (10YR 4/3) loam, weak fine subangular blocky structure; soft, friable, slightly sticky, slightly plastic; violently effervescent, moderately alkaline (pH 8.4).
C2	91-152 cm	Brown (10YR 4/3) gravelly sandy loam, weak fine subangular block structure; soft, friable, nonsticky, nonplastic; slightly effervescent, moderately alkaline (pH 8.2).

The No. 401 soil occurs on level, or nearly level, alluvial fans and floodplains with slope gradients of 0 to 2%. The soil is formed in alluvium from quartzite, rhyolite, basalt, and limestone. The soil is well drained, with medium runoff, and moderate permeability (0.8 to 2.5 in/hr). Slight to moderate wind and water erosion is evident. The vegetation consists of fourwing saltbush, winterfat, bud sagebrush, Indian ricegrass, and horsebrush. The total plant density is about 10%. This soil supports good winter grazing for livestock and wildlife.

CONCLUSION

In the five areas surveyed, the wind has had a dominant influence on the surface texture of the soil. It has moved the fine textured soils, creating bare areas between the plants and depositing the fines around the base of the brush or vegetation. Most of the soils are potential agricultural lands if water were available. Of the 11,340 acres surveyed, 7,560 acres, or two-thirds, could be cultivated for crops.

The dominant plants are fourwing saltbush, winterfat, and galleta grass. Domestic livestock prefer the above vegetation to any other that grows on the desert range. Because a large portion of the land surveyed could be cultivated for crops or is potentially good winter range, these findings should be considered in any "cleanup" decision.

REFERENCES

1. Leavitt, V. D. In preparation. "Soil Surveys of Five Plutonium Contaminated Areas on the Test Range Complex in Nevada." NERC-LV-539-28, Las Vegas.

2. Maxey, G. B., and C. H. Jameson. 1948. "Geology and Water Resources of Las Vegas, Pahrump and Indian Springs Valleys, Clark and Nye Counties, Nevada." Bulletin No. 5, State of Nevada, Office of State Engineer Water Resources. Carson City.

3. Munsell Color Company, Inc., Baltimore, Maryland. 1954. Munsell Soil Color Chart.

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5. Soil Survey Staff Soil Conservation Service, USDA. 1970. "Soil Classification--A Comprehensive System 7th Approximation." Washington, D.C.

6. USDA. 1951. Soil Survey Manual. Agriculture Handbook No. 18. Washington, D.C.

Sample collection and preparation Nevada Test Site - NTS and Tonopah Test Range - TTR

SAMPLING AND FIDLER SURVEYS

Editor's Note: The following preliminary procedures are excerpts from correspondence and protocol prepared by R. O. Gilbert concerning the development of procedures for soil sampling and FIDLER surveys:

October 1972	Soil Sampling Protocol, Area 13 of NTS. R. O. Gilbert.
February 1973	Random Soil Locations, Area 5 (GMX), Letter, R. O. Gilbert to H. Kayuha.
October 1973	FIDLER Surveys--Tonopah Test Range and Plutonium Valley, Letter, R. O. Gilbert to P. B. Dunaway.

SOIL SAMPLING PROTOCOL--AREA 13 OF NTS*

October, 1972

R. O. Gilbert
Battelle Northwest Laboratories
Richland, Washington

On the day that soil samples are to collected, the field crew will be supplied with (i) field data sheets entitled "Soil Sampling for Inventory on Area 13--Field Data Sheet" (see attached example), and (ii) a map of Area 13 which gives the coordinates of each 400 x 400 ft. grid square as well as the boundaries of the numbered isopleths. A list of random locations where the soil samples are to be collected will be given in Columns 1 through 3 on the data sheet.

COLUMN 1: Isopleth No. (identified on map provided)

COLUMN 2: North coordinate

COLUMN 3: East coordinate

The exact spot to be sampled is determined by the coordinate numbers in Items 2 and 3. The number of samples to be collected will be less than the number of random locations listed on the sheet. This is to ensure that if for some reason a given random location is found to be unacceptable for obtaining a soil sample, then another sampling location is available (this point is discussed in greater detail below).

An attempt should be made to ensure that the north and east coordinate distances are measured off on true north and east directions, although a small error is not of serious concern. It is advisable that the field crew use good quality hand compasses for this purpose. It is important in order to avoid bias that the soil sample is taken at the exact spot indicated by the measuring tape, be it on desert pavement, under a bush, in a blow mound of sand, etc. If a bush will interfere with taking the soil sample, then the bush should be removed in such a manner that the soil to be sampled is not disturbed.

Before the soil sample is taken, FIDLER or SAM I readings (whichever is recommended by Eric Fowler) in the 60 KeV and 122 KeV channels will be taken at the proper height above the located point and recorded on the field data sheet (Columns 4 and 5). If a bush is growing over the spot so that there is the possibility that the bush would induce an error in the FIDLER reading, two FIDLER readings should be taken: one before the bush is removed and one after the bush is removed. The distance of the FIDLER

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*See Editor's Note, page 11.

above the located point should be the same for both readings. Once the FIDLER readings have been recorded, the soil sample should be taken accordir to the procedure approved by Eric Fowler. The depth to which the surface samples are to be collected will have been specified earlier by Eric Fowler; similarly for the proper packaging and transportation procedures. Note that on each page of the Field Data Sheet, the depth of the surface samples must be indicated.

If for some reason the soil sample cannot be taken at the exact spot indicated by the measuring process, it is not acceptable to sample at the "closest available spot," or some other convenient location since the location of the sample then becomes a subjective judgment on the part of the field crew. The recommended procedure is to take the sample at the next location on the list of random locations. In essence then, the field crew takes samples in the order in which they appear on the list and continue down the list until the designated number of samples is taken. The list of random locations will be sufficiently long to ensure that the required number of samples can be taken. The only legitimate reason for not taking the soil sample at the designated spot is if the soil is covered by an immovable object such as a slab of concrete, a large immovable rock, etc. It is important that the reason for not taking the soil sample be indicated in the "comments" column (Column 11). Any unusual characteristics relating to the spot sampled should be recorded in the "comments" column. Examples might include "middle of a road" or "in a wash-out area," etc. Whether or not the soil sample is collected, Columns 4-10 should be completed (note that this implies FIDLER readings are taken whether or not the soil sample is taken).

After the soil sample is taken, a permanent marker (stake?) should be driven into the ground to mark the spot. In addition, a permanent tag should be securely tied to the stake. This tag will have written on it (something that won't fade in sun and heat) the following information: (i) the random location number (Columns 1 through 3 on the Field Data Sheet), and (ii) the date the soil sample was collected.

BATTELLE PACIFIC NORTHWEST LABORATORIES

February 13, 1973

Mr. Henry Kayuha*
Nevada Operations Office
Atomic Energy Commission
P.O. Box 14100
Las Vegas, Nevada 89114

Dear Kiwi:

Enclosed are the random soil locations for Area 5 (GMX site) which you requested in our phone conversation of February 12, 1973. The locations are given in relation to ground zero. For example, location (N300, E34) is 300 feet north and 34 feet east of ground zero. You will note that Lee Eberhardt and I have used the FIDLER survey data obtained in October 1972 to divide Area 5 into five isopleths (strata) for purposes of sampling soil for inventory (see attached map).

We recommend that a total of 100 soil samples be taken from the entire area. However, if a lesser effort is required, we suggest that no fewer than 70 samples be collected. The allocation of the 100 or 70 samples (whichever is chosen) to the five strata should be as follows:

	Strata	Number of Soil Samples	
1	0- 5000 CPM	40	25
2	5000-25000 CPM	20	10
3	25000-50000 CPM	10	10
4	>50000 CPM	20	15
5	0-50000 CPM	10	10
		100	70

You will note that on the enclosed sheets a few extra random locations have been listed for each strata identified by a star. These are for use only if one or more of the unstarred locations cannot be used.

I suggest that you check with Eric Fowler at LASL regarding the manner he wants profile samples to be collected in GMX. He may have instructions concerning the microenvironment suitable for taking profiles, i.e., does he want profiles taken at random or, as was done in Area 13, taken only in desert pavement areas. It is probably reasonable to let 10% of the soil sample locations indicated on the enclosed sheets be profiles (subject to restrictions imposed by Eric).

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*See Editor's Note, page 11.

Vegetation samples obtained for purposes of estimating inventory in plants can be collected at random as is currently being done in Area 13. I am assuming here that the vegetation sampling protocol that I prepared for Area 13 is workable in the field. If the vegetation crews in Area 13 are finding it necessary to alter the protocol due to practical problems, then we can develop some other scheme. The important point here is that the shrubs must be a random sample from the area and this will not be the case if the shrub nearest to the random soil location is always harvested. If the vegetation protocol is not followed faithfully in Area 13, I would appreciate a call from you.

I'm glad to hear that the sampling is progressing so well in Area 13.

Sincerely,

R. O. Gilbert
Senior Research Scientist
Statistics Section
Systems Department

ROG:lib

cc: P. B. Dunaway, AEC, LV
B. Church, AEC, LV
I. Aoki, REECo, LV
L. L. Eberhardt, BNW
R. L. Hooper, BNW

BATTELLE PACIFIC NORTHWEST LABORATORIES

October 8, 1973

Mr. Paul Dunaway*
Office of Effects Evaluation
Nevada Operations Office
Atomic Energy Commission
P.O. Box 14100
Las Vegas, Nevada 89114

Dear Paul:

Enclosed you will find maps upon which are indicated my ideas regarding areas requiring more intensive Fidler surveys before isopleths are defined for Tonopah Test Range and Plutonium Valley. You will note I have recommended additional Fidler readings on grids of size 25, 50 or 100 feet, depending on the size of the area in question and the Fidler surveys already completed. If the indicated surveys are too costly or time consuming to be practical I suggest that any reduction of effort be made in the following regions:

a. the 100 ft. grid area in Clean Slate #1;

b. the 100 ft. grid area circled in red in Clean Slate #3; and/or

c. the 100 ft. grid area in Plutonium Valley (the 4th page of maps).

If further reductions in effort are required, the 100 ft. grid region in Clean Slate #3 could be further reduced and the 25 ft. grid regions in Sites B, C and D of Pu Valley could be expanded to 50 ft. grid size. (Map 5) I have not yet been supplied with the latest Fidler surveys of Clean Slate #2 in TTR. Hence that site is not included among the maps.

I am also sending copies of this letter and the maps to Sam Aoki so that he will know as soon as possible what I am recommending. He indicated to me at the information meeting just completed that he needed to have this information promptly so that he could plan this months field activities. Also, Lee and I will be coming to a decision shortly as to the type of sampling plan we feel would be appropriate for these areas. Once this is

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*See Editor's Note, page 11.

decided and we receive the results of the more extensive Fidler surveys we can choose the number and location of soil samples to be collected.

Best regards,

R. O. Gilbert
Senior Research Scientist
Statistics Section
Systems Department

ROG:ml1

Enclosure

cc: I. Aoki, REECo
E. B. Fowler, LASL
L. L. Eberhardt, BNW
H. J. Kayuha, LV

ACKNOWLEDGMENT

Editor's Note: The following REECo personnel have contributed to many of the studies (soil, soil mound studies, and other technique developments) reported in this procedures handbook:

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STANDARD NEVADA APPLIED ECOLOGY GROUP (NAEG) PROCEDURES FOR PREPARATION OF SOIL SAMPLES FROM NAEG INTENSIVE STUDY AREAS

D. L. Wireman
and
H. J. Kayuha

Reynolds Electrical & Engineering Co., Inc.
Las Vegas, Nevada

INTRODUCTION

Soil samples weighing 200-800 g which have been collected at field sites in plastic bags, are each counted with a stabilized assay meter (SAM-1) to determine the approximate ^{241}Am activity for radiation safety purposes prior to sample preparation. Readings with portable alpha, beta, and gamma instruments are also made for the same reason.

Samples consist mainly of surface or profile samples. All depth profile sections for a given profile sample are prepared by the same procedure for each section as for a surface sample. A library number and a library data card are assigned to each sample. The samples are weighed, dried, and reweighed. They are ball-milled and at this point in the procedure, percentage groups of the samples collected from the study area are selected at random and prepared by the following procedure:

Approximately 60% of the original number of samples are aliquoted for analysis from the ball-milled fraction. A soil fraction from approximately 35% of the original number of samples, after ball-milling, is passed through a 100-mesh sieve, and aliquots of the soil fraction passing the 100-mesh sieve are taken for analysis. For approximately 3% of the original number of samples, an aliquot of the soil fraction passing and an aliquot of the soil fraction not passing the 100-mesh sieve are taken for analysis, also the weight of the soil fraction passing the 100-mesh sieve and the weight of the soil fraction not passing the 100-mesh sieve are recorded for this 3% sample group. Approximately 2% of the original number of samples are prepared by the same procedures as the 3% group, except that only the aliquot of soil passing the 100-mesh sieve is taken for inter-intra laboratory comparison aliquots.

An aliquot of each sample is analyzed with a Ge(Li) system, for a short period, to determine the approximate ^{241}Am activity.

These values are forwarded to the laboratory or laboratories receiving aliquots for analysis. The sample fraction remaining is placed in the sample library by the library number, for future analysis.

EQUIPMENT

1. Ball mills, 1 gal can size, 140 rpm, closed system, vacuum exhaust.
2. Laboratory grinding balls, forged manganese steel, 1-in. diameter.
3. One-gallon empty paint cans, smooth exterior, with lids.
4. Drying oven, 0-500° F, commercial.
5. Balance, automatic tare, top loading, 0-1000 g range (i.e., Mettler P-1000).
6. Exhaust hood, 100 linear feet per minute minimum face velocity.
7. Sixteen-ounce polyethylene bottles with screw caps.
8. Plastic bags, 34 x 54 in. and 19 x 34 in.
9. Cardboard boxes.
10. Aluminum trays, 12 1/2 x 10 1/2 x 2 1/2 in. deep, and 8 1/2 x 6 x 2 in. deep.
11. Gloves, surgeon.
12. Tape, 1-in. filament type (i.e., scotch brand).
13. Tape, plastic electrical.
14. Plastic spoons, tea size.
15. Utility wipes.
16. Six-ounce paper cups.
17. Lab coats or paper coveralls.
18. Scissors.
19. One-gallon can lid fasteners.
20. Two-inch-diameter, 100-mesh sieves, with caps.
21. Eight-inch-diameter, 100-mesh sieves, with lids and receiving pans.
22. Vibration system for 100-mesh sieves (i.e., RO-Tap).

23. Sink with contaminated waste system.

24. Sixteen-ounce polyethylene bottle cap rings.

25. 2 x 3 in. manila envelopes.

PROCEDURE

Library Data Card

Assign library number and library data card to each sample. Record organization received from, date received, sample type, sample site location, and all other field input data, on library data card for each sample.

Initial Instrument Measurements for Radiation Safety

For each sample:

1. Place plastic bagged soil sample, containing between 200 and 800 g of soil, on calibrated stabilized assay meter detector.

2. Count for 0.1 minute.

3. Invert sample on stabilized assay meter detector and recount for 0.1 minute.

4. Estimate approximate ^{241}Am from highest net count rate.

5. If ^{241}Am estimated activity is greater than 5×10^3 dpm/g, do not prepare sample by these procedures.

6. Record estimated ^{241}Am dpm/g value.

7. Check sample at contact and determine portable instrument readings for alpha, beta, and gamma.

8. Record readings.

Weighing and Drying

For each sample:

1. Place plastic bag flat on bottom of hood for working surface.

2. Place small aluminum tray in hood. Label tray with library number.

3. Use gloves and lab coat or paper coveralls.

4. Place plastic bagged sample in hood.

5. Cut off neck of plastic sample bag.

6. Cautiously pour contents of bag into tray.

7. Tare balance with empty tray (with balance in hood).

8. Place tray containing sample on balance. Remove gloves and place them in plastic contaminated waste bag, in hood.

9. Record weight ± 1 g, on library data card, as wet weight.

10. Place tray containing sample in drying oven. Wash hands. Clean hood.

11. Adjust drying oven temperature to 105° C for 24 hr.

12. Prepare 1-gal can by placing 10 grinding balls in can, label can and lid with library number.

13. Using gloves, place can in hood.

14. Remove sample from oven, cool to room temperature, and place in hood.

15. Weigh sample and record net weight ± 1 g, on library data card, as dry weight.

Ball Milling

For each sample:

1. In hood, carefully transfer sample from drying tray to can. Place tray in plastic contaminated waste bag in hood.

2. Place lid on can and secure. Wash off gloves.

3. Decontaminate outside of can. Remove gloves and place in plastic contaminated waste bag.

4. Place four lid fasteners, equidistant, on can.

5. Place can on ball mill and rotate at 140 rpm for 5 hr with vacuum system on.

6. Remove can from ball mill and place in hood.

7. Place large tray in hood.

8. Prepare 19 x 34" plastic bag as follows: cut about 2 in. off sealed corner of bag. Pass corner of bag through 16-oz poly bottle cap ring. Flange plastic over cap ring and screw cap ring on 16-oz poly bottle.

9. Using gloves, loosen lid, but do not remove from can.

10. In hood, place can inside prepared plastic bag.

11. Seal plastic bag with tape.

12. Invert can, and tap side to remove soil and grinding balls.

13. Place grinding balls back into can.

14. Place lid on can.

15. Transfer soil from plastic bag to attached 16-oz poly bottle.

16. Hold bottle over large tray and carefully remove cap ring from bottle. Set bottle aside.

17. Carefully remove cap ring from plastic bag and place cap ring in water bath.

18. Seal plastic bag with tape and place in contaminated waste container.

19. Tare balance with new 16-oz poly bottle (without cap).

20. Weigh bottle (without cap) containing sample and record weight, ± 1 g, as ball mill weight. Place cap on bottle, label bottle with library number and decontaminate exterior.

Sample Aliquoting

Contaminated waste (such as plastic spoons, paper cups, gloves, and aluminum trays) produced during aliquoting in hood is to be placed, in the hood, into plastic contaminated waste bags, which are to be sealed prior to removal from hood.

A. Sixty percent group:

1. Randomly select a group of surface and profile samples from ball milling step 20 (all profile depth sections for a given profile are to be prepared by same method), which is approximately equal to 60% of the number of samples collected from the study area, and continue preparation.

For each sample:

2. In hood, tare balance with new 6-oz paper cup.

3. In hood, using gloves and new plastic spoon, transfer 10 ± 0.1 g of soil (from poly bottle in ball milling step 20) into paper cup.

4. In hood, transfer soil from paper cup into new 2 x 3 in. manila envelope (do not contaminate exterior of envelope). Seal envelope with filament tape.

5. Label envelope with library number, "10 ± 0.1 g", and radioactive material sticker.

6. Using same spoon and cup, transfer 50 ± 0.1 grams of soil from same poly bottle into paper cup.

7. In hood, transfer soil from paper cup into new 16-oz poly bottle.

8. Label bottle with library number, "50 ± 0.1 grams" and "Ge(Li)."

9. By tare, weigh soil remaining in poly bottle to ± 1 g. Record weight on library data card as library storage weight.

10. Label bottle "store".

B. Thirty-five Percent Group:

1. As with the 60% group, randomly select a group of samples (from ball milling step 20) which is approximately equal to 35% of the number of samples collected from the study area and continue preparation.

For each sample:

2. In hood, using gloves, using a new plastic spoon, transfer sufficient soil into a new 2-in-diameter, 100-mesh sieve (which has cap on one end), such that after the vibration period, an excess of 50 g of soil will have passed through the 100-mesh sieve.

3. By tare, weigh remaining soil in poly bottle to ± 1 g. Record weight on library data card as library storage weight. Label bottle "store".

4. Place second cap on sieve, seal sieve with tape, and label with library number.

5. Decontaminate exterior of sieve and place on vibration system until an excess of 50 g of soil has passed through sieve.

6. Remove sieve from vibration unit and place in hood.

7. In hood, using gloves, remove cap from sieve and place cap in water bath.

8. Using new plastic spoon, by tare, weigh 50 ± 0.1 g of soil, which has passed through 100-mesh sieve, into new 6-oz paper cup, then transfer to new 16-oz poly bottle.

9. Label bottle with library number, "50 ± 0.1 g", and "< 100 mesh."

10. Decontaminate exterior of bottle and label bottle with radioactive material sticker. Seal bottle with plastic electrical tape.

(This aliquot is to be analyzed with Ge(Li) system prior to shipment).

C. Three Percent Group:

1. As with the 60% group, randomly select a group of samples, (from ball milling step 20), which is approximately equal to 5% of the number of samples collected from the study area and continue preparation.

For each sample:

2. In hood, place new 8-in.-diameter, 100-mesh sieve in receiving pan (decontaminated) and transfer entire soil sample (from ball milling step 20) into sieve.

3. Place lid, (decontaminated), on sieve and seal sieve with tape.

4. Label sieve with library number, decontaminate exterior, and place on vibration system until all soil which will pass through the sieve has passed through.

5. Remove sieve from vibration unit and place in hood.

6. In hood, using gloves, remove lid from sieve and place lid in water bath.

7. Remove sieve from receiving pan and carefully transfer soil, which did not pass through sieve, into tared aluminum tray.

8. Weigh soil which did not pass through sieve and record weight on library data card as < 100 mesh weight.

9. Using new plastic spoon, by tare, weigh 50 ± 0.1 g of soil, which did not pass through 100-mesh sieve, into new 6-oz paper cup, then transfer to new 16-oz poly bottle.

10. Label bottle with library number, "50 ± 0.1 g", and "< 100 mesh." Decontaminate bottle, label with radioactive material sticker, and seal bottle with plastic electrical tape. (This aliquot is to be analyzed with Ge(Li) system prior to shipment).

11. Transfer remaining soil from tray to 16-oz poly storage bottle. Label bottle with library number, "< 100-mesh," and "store". Decontaminate exterior of bottle.

12. In hood, using gloves, carefully transfer soil, which passed through 100-mesh sieve, from receiving pan to tared aluminum tray.

13. By tare, weigh soil which passed through sieve, to ± 1 g, and record weight on library card as "< 100-mesh weight."

14. Using new plastic spoon, by tare, weigh 50 ± 0.1 g of soil which passed through sieve, into new 6-oz paper cup, then transfer to new 16-oz poly bottle.

15. Label bottle with library number, "50 ± 0.1 g," and "< 100-mesh." Decontaminate bottle, label with radioactive material sticker, and seal with plastic electrical tape. (This aliquot is to be analyzed with Ge(Li) system prior to shipment).

16. Using gloves, using new plastic spoon, transfer remaining soil from receiving pan to 16-oz poly storage bottle. Label bottle with library number and "< 100-mesh." Decontaminate exterior of bottle. Place empty receiving tray in water bath.

D. Two Percent Group:

The same procedure is to be used as for the 3% group, except that the 50-g aliquot of soil not passing through the 100-mesh sieve is not taken.

^{241}Am Determination

For each sample:

1. Analyze prepared 50-g aliquot, in 16-oz poly bottle, with Ge(Li) system for 10 min.

2. Determine ^{241}Am in dpm/gram.

3. Correlate ^{241}Am activity with library number and report this data with aliquot at time of shipment to analytical laboratory.

4. The ^{241}Am activity is also to be used in conjunction with other radiation measurements for radioactive shipment requirements.

5. The $^{239\text{-}240}$Pu activity for shipment requirements is to be estimated from the ^{241}Am activity by using the current $^{239\text{-}240}$Pu/^{241}Am ratios for the study area soil.

REECo FIELD ACTIVITIES AND SAMPLE LOGISTICS IN SUPPORT OF THE NEVADA APPLIED ECOLOGY GROUP

H. J. Kayuha, I. Aoki, and D. L. Wireman
Reynolds Electrical and Engineering Co., Inc., Las Vegas

(Ed. Note: Previously published in NAEG Report NVO-142, pp. 17-19.)

ABSTRACT

The field activities and sample logistics of Reynolds Electrical and Engineering Co., Inc. (REECO), in support of the Nevada Applied Ecology Group (NAEG) plutonium studies in the Test Range complex, are discussed in this summary report. Field instrument measurements, determination of sampling sites, and procedures used in preparation of samples are included.

INTRODUCTION

Reynolds Electrical and Engineering Co., Inc. (REECo), is the prime contractor of the Nevada Test Site (NTS). It is through REECo that the Nevada Applied Ecology Group (NAEG) obtains its primary field support. There are four areas selected by the NAEG for initial investigation: Area 5 GMX; Area 11, Plutonium Valley or Project 56; Area 13, Project 57; and the Area 52 Tonopah Test Range (TTR) sites, which include Clean Slates I, II, III, and Double Track events. Portable field instrument measurements, determination of sample locations, as well as sampling methods and sample preparation are discussed, with a summary of completed activities.

METHODS

Past reports from the individual events, in addition to periodic surveys, are used to determine the boundaries of the plutonium-contaminated areas.

After the contamination boundary has been established, surface soil samples are collected along a single transect originating at ground zero (GZ), following the center of the cloud pattern and extending to the perimeter of the study area. These samples serve as advanced information concerning the soils' isotopic content. A grid system is then established to permanently mark the locations at which portable survey instrument measurements are taken. The primary portable instrument used at the immediate study sites is the FIDLER, a single-channel NaI counter which has a thin window for measuring low-energy gamma and x rays. After the FIDLER survey has been completed, isopleths are determined. The isopleths are normally assigned activity levels as follows: 0-1 k cpm, 1-5 k cpm, 5-10 k cpm, and greater than 50 k cpm. The activity given in cpm refers to the 60 kev ^{241}Am energy. A random collection scheme is then established for both soil and vegetation sample collection. When ^{241}Am is prevalent, a FIDLER measurement is taken one foot above each random soil sampling location, for correlation determinations with ^{241}Am and ^{239,241}Pu analytical data and for additional isopleth delineation information. The soil samples collected are of two types: surface and depth profile. A surface soil sample is defined as a soil sample taken with a 12.7-cm diameter penetration ring, to a depth of 5 cm. A depth profile soil sample is defined as a 10- x 10-cm soil sample taken in a 2.5-cm depth increments, to a total depth of 25 cm. Area 13, Project 57, microplot experimental depth profile soil samples were taken at 3-cm depth increments to a total depth of 21 cm. Following collection, the soil samples are prepared for analysis at the soil preparation facility in Mercury. Here, soils library numbers are assigned which serve to permanently identify the samples. Portable instrument measurements, primarily with a stabilized assay meter (SAM-1), are used to separate the samples into relative activity levels prior to preparation. The soil samples are dried for 24 hours at a temperature of 105° C.

It has been determined, however, that no significant comparative weight increase is found when desert soil samples (500 grams) are dried at temperatures as low as 65° C, for durations as short as 12 hours. After drying, the samples are each transferred to a one-gallon paint can containing 10 one-inch diameter steel grinding balls and placed in a ball mill where they are rotated at 140 rpm for approximately five hours. A 50-gm aliquot of each sample is analyzed with a Ge(Li) 2048-channel analyzer system at the REECo Radiological Measurements Laboratory to estimate the ^{241}Am activity for shipping criteria. Aliquots, for radiological analysis, are packaged and distributed to designated analytical laboratories. The remaining sample is packaged and stored in the soils library.

Vegetation samples are collected from sites near to the random soil sample locations. They are placed in one-gallon paint cans, transported to the REECo preparation facility at Mercury, assigned library numbers, and prepared for shipment. They are then shipped to the LFE Environmental Analysis Laboratory, Richmond, California, where they are ashed, put into solution, and subsequently distributed to designated analytical laboratories.

Small animal samples are collected by a research group from the University of Nevada, Las Vegas. Preliminary preparation of these samples is performed by this research group at the facility in Mercury. Some of these samples are then shipped to LFE Environmental Analysis Laboratory for ashing and analysis. The balance are shipped to the Oak Ridge National Laboratory for histo-pathological examination.

Radiological monitoring support is provided by REECo for the Resuspension Element: loading and handling of the microcarbon air samples previous to resuspension operations; and the packaging and shipping of the samples following operations. The equipment, operation, and maintenance of the electrical power supply (generators) for the air sampling units are also provided by REECo.

SUMMARY

To date, the REECo preparation facility has processed 5,758 soil samples and 400 vegetation samples, including the NAEG samples. Also, there are 2,528 samples stored in the sample library, in addition to 36 4- x 4- x 4-foot pallets of samples collected by K. Larsen at and around NTS between the mid-50s-early 60s.

The field activity status of the present NAEG intensive study areas is as follows: fences enclosing the contaminated areas have been erected; single transect soil samples have been collected from all study areas and prepared for analysis; grid systems have been completed in all study areas; FIDLER surveys are complete in Areas 5 and 13, and are 75 percent complete at TTR and Area 11; soil and vegetation sampling are complete from Areas 5 and 13.

Future REECo support activities for FY 1974 are to complete the FIDLER surveys at TTR and Area 11; establish random sampling schemes; support the collection of soil and vegetation sample effort; prepare the samples for analysis; continue to support the resuspension and population census effort; and to computerize NAEG analytical bank.

NAEG SOIL MOUND PROCEDURES

Editor's Note: The following procedures are excerpts of correspondence, notes of meetings, and protocols prepared by R. O. Gilbert, E. B. Fowler, E. H. Essington, P. B. Dunaway, H. Kayuha, D. L. Wireman, D. N. Brady, A. E. Bicker, and M. G. White concerning the development of and procedures for NTS soil mound studies.

March 1974	Sampling of Mounds at NTS, E. H. Essington and E. B. Fowler.
March 1974	Sampling of Mounds, Letter, R. O. Gilbert to E. B. Fowler.
September 1974	NAEG Soil Mound Protocol . . . Preliminary Study of Area 11, NTS, P. B. Dunaway *et al.*
November 1974	Results of Preliminary Study of Soil Mound . . . in Area 11, NTS, D. N. Brady.
January 1975	Blow Sand Mounds on NAEG Intensive Study Sites, Letter, R. O. Gilbert to P. B. Dunaway.
August 1975	Mound Study No. 2 Protocol, Attachment to a Letter from E. H. Essington to NAEG Files.
October 1975	Laboratory Sample Preparation Protocol for NAEG Mound Samples, Letter, E. H. Essington to P. B. Dunaway.
November 1975	NAEG Soil Mound Study No. 2 (Revised), E. H. Essington *et al.*
January 1976	^{241}Am GE(Li) Analysis of Area 13 Soil Mound Test Samples, Excerpt from a Letter, A. E. Bicker to R. O. Gilbert.
January 1976	Recommendations for Mound Soil Preparation, Letter, E. H. Essington to P. B. Dunaway.
March 1976	Soil Mound Study Protocol, Letter, A. E. Bicker to M. G. White.
March 1976	Sample Preparation (Revised 3/4/76), Excerpt from a Letter, E. H. Essington to P. B. Dunaway.

SAMPLING OF MOUNDS AT NTS

March, 1974

E. H. Essington and E. B. Fowler
Los Alamos Scientific Laboratory*

As part of the NAEG effort to delineate Pu inventories in the intensive study areas, mounds were designated to be sampled in a random fashion similar to desert pavement. During the February 1974 Soils Element meeting, it became apparent that mound sampling was not being carried out as originally specified and that the technique of mound sampling has not been thoroughly investigated. Recommendations from participants of the meeting were requested regarding the best way to sample a given mound for the purpose of determining the distribution of Pu on and within the mound and the total inventory of Pu in the mound.

This presentation suggests a possible way of sampling mounds to satisfy the above criteria. The procedure has not been tried in field conditions and, therefore, is presented only as a recommendation for consideration and possible trial in a controlled field situation.

The theory behind the proposed sampling scheme involves sampling the mound in such a way that three-dimensional contours of both mound shape and Pu activity can be formulated. Field sampling consists of (1) several two-dimensional profiles, and (2) a stereo pair of photographs of sufficient resolution to contour the surface via photointerpretive means.

PROFILE SAMPLING

For purposes of discussion, assume the typical mound to be a metre in diameter and about one-half metre high with a substantial desert shrub growing from the mound. The field sampling procedure might be as follows:

Equipment

1. Stereo camera
2. Shrub clippers
3. Hand saw
4. Sampling scoop (10 cm long x 5 cm deep x 1 cm wide)
5. Thin smooth stainless steel sheets

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*See Editor's Note, page 33.

one, about 15 cm x 100 cm
two, about 50 cm x 100 cm sharpened edge
many, about 12 cm x 5 cm

6. Sample bags, etc.

7. Spatula, about 1 cm x 10 cm

Procedure

1. Select a mound for trial purposes.

2. Remove at ground level (top of mound) all vegetation by cutting or sawing, taking care not to disturb the mound in any way.

3. Photograph the mound with stereo camera preferably from a point perpendicular to the vertical axis of the mound and from such a distance to include the whole mound and part of the desert pavement surrounding the mound.

4. Select three to six radials about the mound for profile sampling depending on the uniformity of the mound (Fig. 1).[1]

5. Carefully dampen the surface centimeter or so of one of the radials.

6. Carefully insert one of the large steel sheets into the mound vertically to ground level. Then insert the other sheet so that a minimum of 12 cm remains between the two sheets at all positions.

7. Carefully insert vertically the 15 x 100 cm sheet at the end of the run as shown in Fig. 2A.

8. Carefully wet soil to sampling depth.

9. Choose sampling location according to sampling order (refer to Fig. 2B) and insert scoop upside down symmetrically within the sides of the baffles into soil to be collected (refer to Fig. 2C).

10. Carefully remove the soil from between the scoop and the sides to the depth of the scoop and discard. Invert scoop and slide into defined soil mass. Slide a 12 x 5 cm piece of steel under the scoop to isolate the soil underneath from cross contamination while collecting the next sample.

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[1] Due to mound complexity, only one radial was finally selected per mound (reference results of preliminary mound study, D. N. Brady, June 23, 1976).

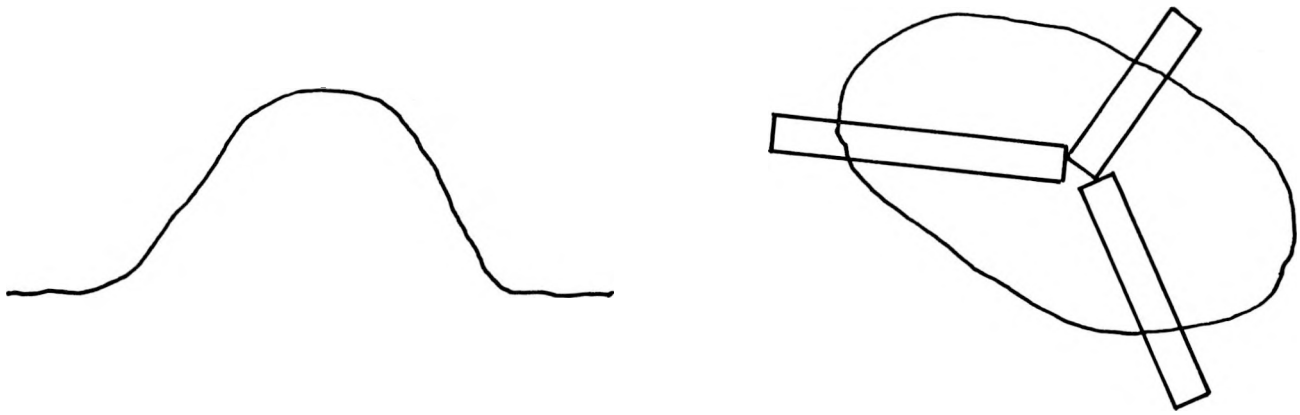
11. Bag, record mound location, radial number, radial aspect, sample number, and other pertinent information.

It is felt that samples collected in this manner will produce Pu values which can be contoured in three dimensions and, using the added variable of mound shape, a total inventory of Pu as well as distribution within the mound can be evaluated.

It is recommended that all samples thus collected from the first several mounds be analyzed to provide sufficient data to test the procedures and generate parameters to be used to suggest random sampling as a routine procedure.

Valuable information regarding the degree of downward movement of Pu both through and adjacent to the mound could be obtained by extending the depth of sampling to a depth comparable to existing profile specifications and extending the radial out beyond the mound into desert pavement several tens of centimeters.

A. UNIFORM MOUND



B. NONUNIFORM MOUND

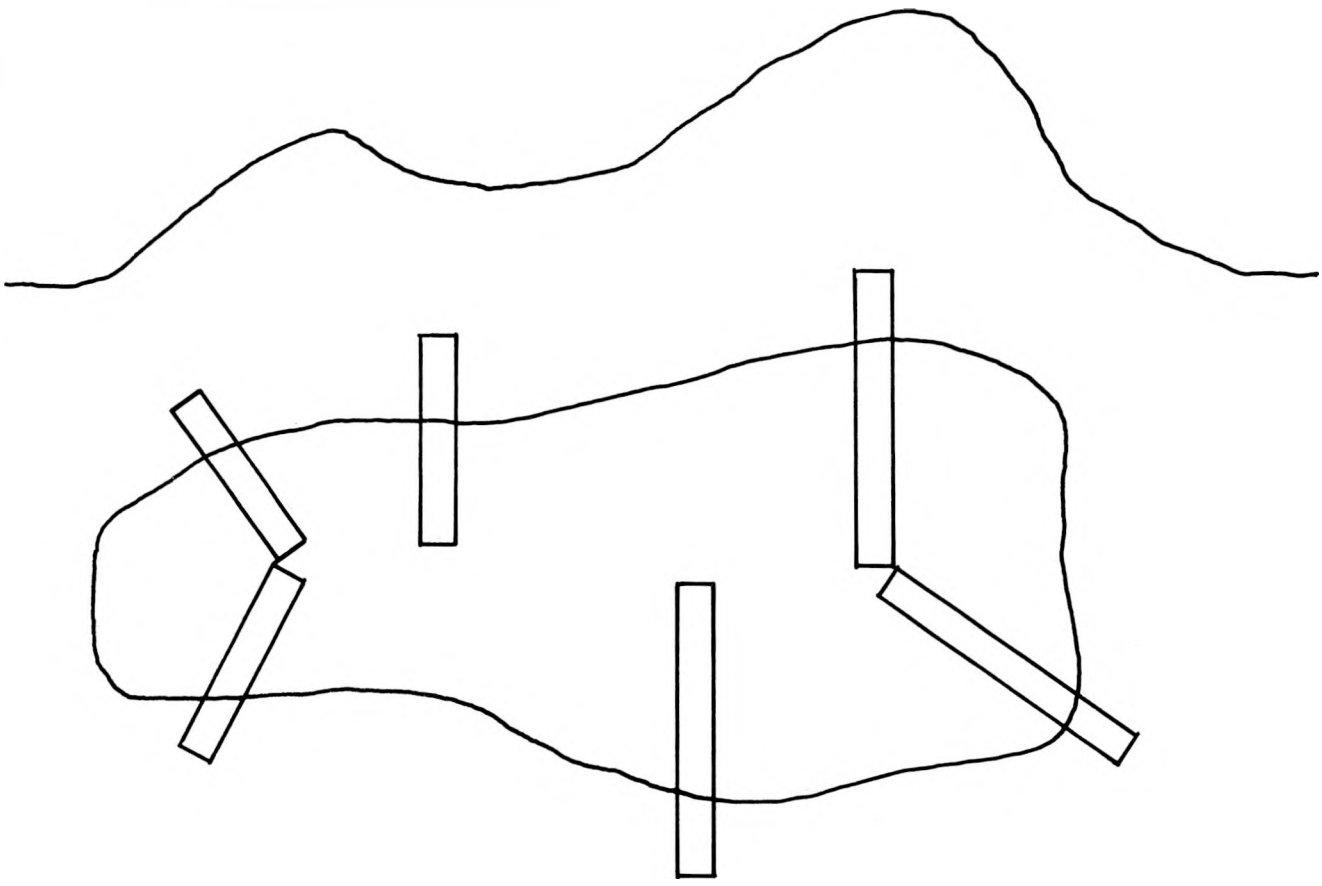
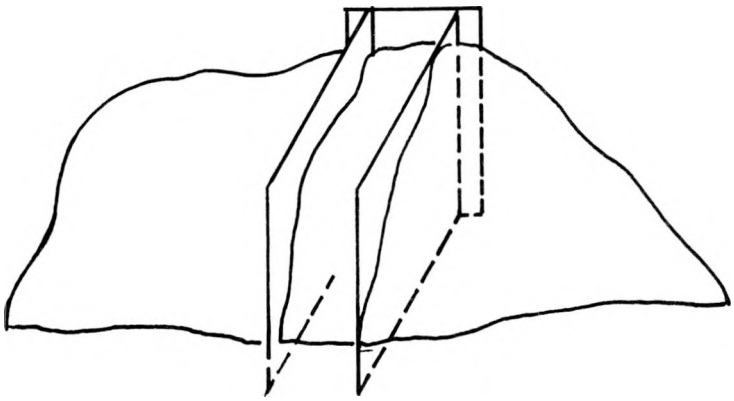
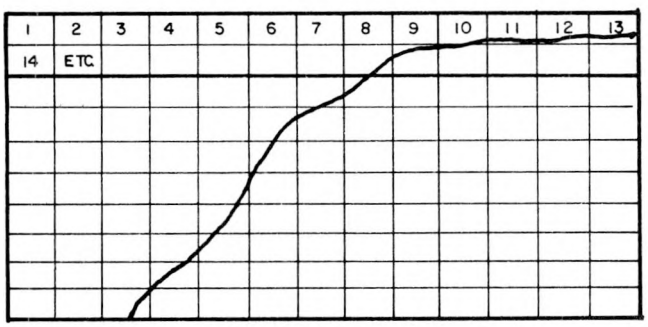


FIGURE 1.
SELECTION OF RADIALS FOR SAMPLING

A. POSITION OF BAFFLE PLATES



B. SAMPLING SEQUENCE (THE NUMBER OF SAMPLES IS YET TO BE DETERMINED.)



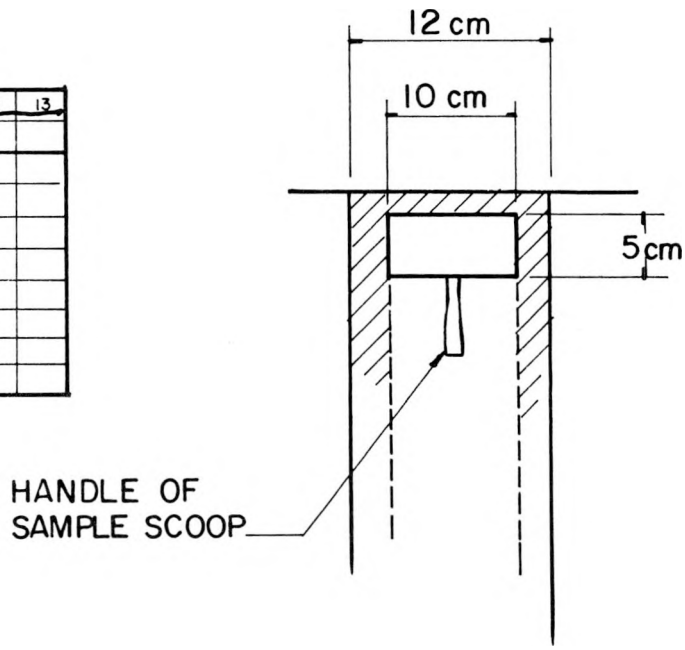


FIGURE 2.
SAMPLING PROFILES

BATTELLE PACIFIC NORTHWEST LABORATORIES

March 28, 1974

Dr. Eric B. Fowler, H-7*
University of California
Los Alamos Scientific Lab.
P.O. Box 1663
Los Alamos, New Mexico 87544

Dear Eric:

Thank you for the packet of information including the minutes of the Soils Element Meeting on February 25, 1974, your and Ed's ideas on the sampling of mounds, etc.

Concerning the minutes, I have looked them over and have enclosed copies of those pages requiring changes. I hope you can figure out my inserts. The rest of this letter is really addressed to both you and Paul Dunaway so I am sending both of you copies.

I have been thinking about the sampling of mounds. Our telephone conversation today (March 26) helped me visualize this a little better. My inclination at the present time is to have two separate sampling programs for mounds, one to estimate total inventory down to 5 cm below the surface of desert pavement, and the other for determining the 3-dimensional distribution of Pu in mounds.

First, concerning inventory, it seems reasonable to choose random locations over the entire field to be inventoried just as is being done at present. But for those that fall on a mound, a "mound surface sample" would be defined as a circular column of 12.7 cm diameter (same width as presently being used for surface samples) and with depth equal to the distance from the surface of the mound at the random location point to 5 cm below desert pavement level (Figure 1). The "mound surface sample" would need to be thoroughly ball-milled before one or more aliquots are analyzed, and, of course, we would need the dry weight of the entire mound surface sample as well as the dry weight of soil passing through the 100 mesh screen (assuming the sample is sieved). In the present mound sampling scheme the depth of sample is just the top 5 cm from the surface of the mound.

Now, a "desert pavement profile sample" would be defined as at present, i.e., samples taken at 2.5 cm increments from the surface down to 25 cm. A "mound profile sample" would consist of: (i) the same 2.5 cm increment samples as in the desert pavement profile sample, i.e., from the surface level of desert pavement down to 25 cm, and in addition, (ii) 2.5 cm increment soil samples from the desert pavement surface up to the top of the mound (Figure 1). The field crew might have some trouble taking the 2.5 cm

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*See Editor's Note, page 33.

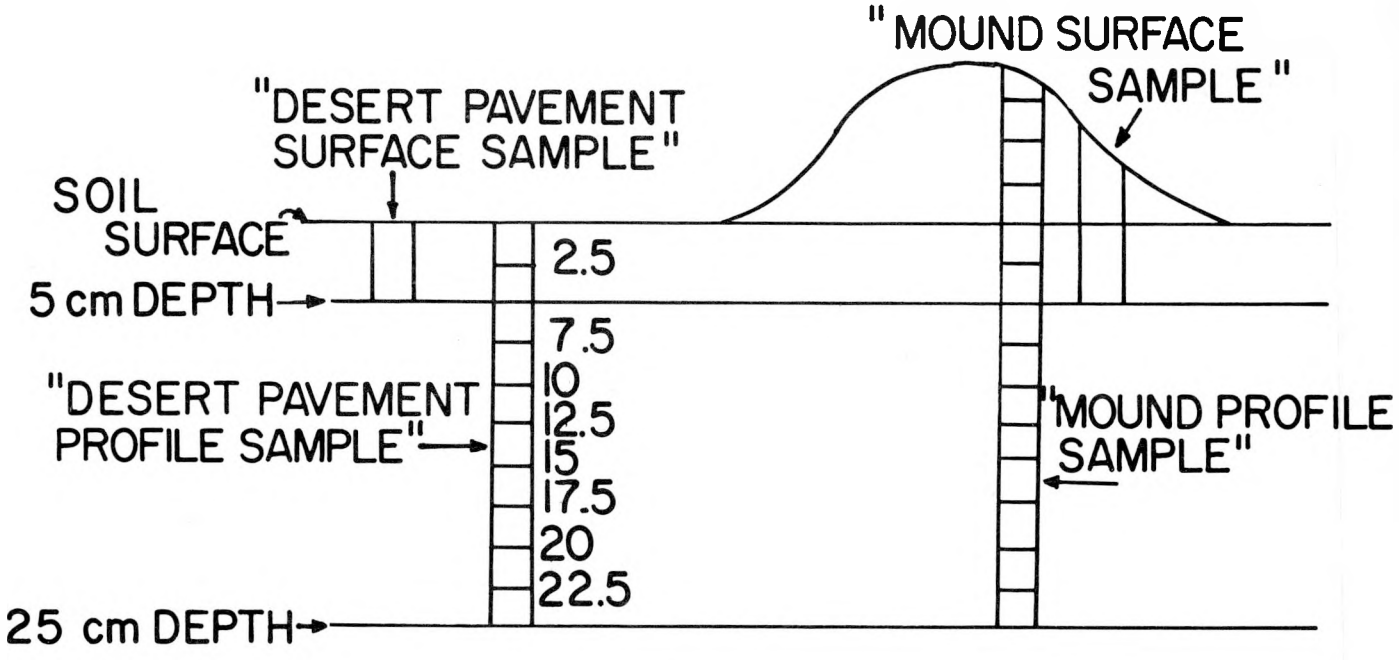
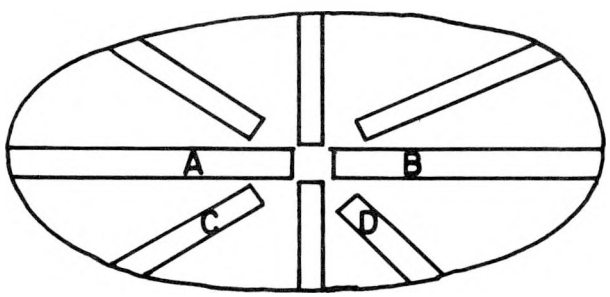


FIGURE 1

TOP VIEW OF MOUND



PREVAILING WIND DIRECTION

FIGURE 2

samples in the mound proper since they would have to be taken in 2.5 cm increments up from the desert pavement surface. Also, the topmost profile sample would in most cases be less than 2.5 cm deep.[1]

Please consider the above approach only as a continuing discussion of the whole mound sampling problem. I'm certainly not tied to it. It has at least one disadvantage in that the "mound surface sample" as defined above and drawn in the illustration would not yield results comparable to the mound surface samples taken in Areas 13, 5 (GMX) and TTR since these were taken from the surface of the mound to 5 cm depth.

Now considering the sampling of mounds to determine the 3 dimensional distribution of Pu. Again, my thoughts here as just extensions or rewording of ideas that came out in our phone conversation on March 26 with you and Ed. First we know that prevailing wind direction has probably had an important influence on the distribution of Pu in the mound. Also, since at this stage of our ignorance we are exploring the unknown, I suggest we try to sample several mounds in the same way; systematically if you will. Some kind of randomization might be required if we were going to estimate some characteristic of the mound, but that is not our purpose here. We are simply trying to determine the pattern of disposition of Pu throughout the mound, and are particularly interested, I believe, in detecting differences between various sections of the mound. I suggest we sample each mound as intensively as possible. One approach is diagramed in Figure 2. Each mound would have samples taken in the same positions relative to prevailing wind direction.[2] We can look upon this problem as one of looking for significant differences between say radials A and B or between C and D (Figure 2). To estimate precisely differences that are small would require, I fear, rather a large number of mounds to be sampled (20 or more considering the particle problem). If we are interested in detecting only large differences, then fewer mounds would probably do. If a particular difference, say A-B, is of over-riding importance to detect, then we could dispense with some of the other radials so that more mounds could be random selected for sampling to estimate A-B.

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[1] No 2.5 cm increment profile samples were taken in soil mound No. 2 study, but were in mound study No. 1. Reference Dunaway "Soil Mound Protocol--Collection, Preparation and Ge(Li) Analysis for a Preliminary NAEG Study of Area 11, NTS," page 3, paragraph 1, dated September 19, 1974, and Essington's "NAEG Soil Mound Study No. 2 (Revised)," page 4, Items 11 and 13.

[2] Multiple radials were not sampled. Reference Brady's "Results of Preliminary Study of Soil Mound and Desert Pavement Vertical Profile Pairs" dated June 23, 1976.

In addition to looking for differences between specific radials within mounds, we may also be interested in comparing the average Pu concentration in "desert pavement surface samples" with that in "mound surface samples." We also discussed this over the phone on March 26. At this point in time I envision that a mound be randomly sampled down to the 5 cm depth below desert pavement surface and these results be compared with random surface samples to 5 cm in desert pavement adjacent to the mound. This procedure could be replicated using randomly chosen mounds. An important aspect of the design would be the pairing of a mound's data with the desert pavement data obtained adjacent to the mound. We would need to work out the optimum number of mounds and adjacent desert pavement to sample but a minimum of 10 such pairs seems necessary. As above, the size of difference important to detect affects the number of pairs that are sampled. Hence, depending on the number of random samples taken within and around the mound, the total number of aliquots analyzed could be fairly substantial. One last point: It might be a good idea to delay this experiment until after the extensive systemmatic sampling of mounds discussed above is completed. Those results could provide valuable information relative to the design (number of samples, their location within the mound, etc.) of the mound-desert pavement experiment.

There are a couple of other points I need to mention:

(1) Surface Area of a Field

In the most recent estimate of surface inventory obtained for Area 13, I assumed that the surface of the ground was essentially flat. However, since there are mounds present this is not actually true. The true surface area is hence larger than just the flat area within the boundaries of the outer fence. A correction for this could be made by choosing several plots (say 20 ft. by 20 ft.) at random and actually determining the true surface area within the plots. Then a correction could be made to the final estimate of inventory. If mounds cover, say, 25% or more of the fenced region and are reasonably high, then the corrected and uncorrected estimates could differ quite a bit.

(2) Counting Statistics

I have derived approximate formulas for the σ counting errors associated with the determination of ^{239}Pu when using ^{242}Pu as a tracer under various assumptions regarding the equality of counting times, efficiencies of counting devices, etc. I'll hold on to these until I get the computational procedures used by Lab X and LFE to compare my formulas with theirs.

(3) Detection Limits

I have looked over the papers by L. A. Currie, "Limits for Qualitative Detection and Quantitative Determination," Anal. Chem. 40, 586-593 (1968) and J. K. Hartwell, "Detection Limits for Radioisotopic Counting Techniques," ARH-2537, Atlantic Richfield Hanford Company (1972) (this latter paper is a summary of the former). Unless there are more

definitive papers on the subject of detection limits that have appeared more recently, it's probably best to use the definitions of "detection limit" and "critical level" as defined in those papers since they appear to be statistically sound. I do need to find out from Lab X and LFE how they define these terms. I understand that you, Eric, have asked them to send me this information.

Well, this letter is much too long already. Thanks for your help Eric and Ed. I hope this letter will help our thinking on these problems.

Sincerely,

R. O. Gilbert
Senior Research Scientist
Systems Department
Statistics Section

ROG:lt

cc: Paul Dunaway
Lee Eberhardt

NAEG SOIL MOUND PROTOCOL--COLLECTION, PREPARATION, AND Ge(Li) ANALYSIS FOR A PRELIMINARY NAEG STUDY OF AREA 11, NTS*

September, 1974

(Ed. Note: All indicated changes to this protocol were made in the field at the time sampling was conducted.)

The following protocol was developed from the proceedings of the September 11, 1974, meeting at AEC/NV. Those in attendance were P. B. Dunaway, AEC/NV; M. G. White, AEC/NV; E. H. Essington, LASL; R. O. Gilbert, BNWL; E. M. Romney, UCLA; C. E. Rosenberry, REECo/NTS; and D. L. Wireman, REECo/NV.

The major objectives of a comprehensive soil mound sampling program were established to be as follows:

1. Determine soil mound radioactivity desert pavement contribution to inventory.

2. Establish vertical radioactivity distribution in soil mounds, in 10 x 10 x 2.5 cm depth sections, to 25 centimeter total depth.

3. Determine radioactive levels of vegetation associated with soil mounds.

It was decided that a soil mound study should be conducted on a limited basis to determine whether or not a comprehensive study will be necessary, and if so, to provide preliminary information on appropriate techniques. The objectives of the preliminary study are as follows:

1. To determine radioactivity level differences in the vertical distribution of soil mound profiles vs desert pavement profiles.

2. To use the Ge(Li) method of analysis for soil and vegetation soil mound samples, so that rapid preliminary results can be obtained. The need or lack thereof, for a comprehensive soil mound program, can thereby be determined.

PROTOCOL

Sample Location and Collection

Reference a FIDLER survey map of Area 11 and determine the area encompassed by the fifth isopleth of C site. Select a large area of the fifth isopleth. Go to area with map and locate large area of fifth isopleth. Establish a

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*See Editor's Note, page 33.

line from Area 11-C, GZ, through the large area of the fifth isopleth. At the midpoint of this line, establish another line perpendicular to the first. Divide this second line into equal segments (each perhaps 10 feet long). Randomly select one segment. Establish the center of this segment as the center of a rectangle which is as large as possible within the confines of the fifth isopleth (perhaps 100 x 100 ft). Record the Nevada grid coordinates of the center point. Grid this square into equal sections (i.e., 10 ft x 10 ft). Number each square, left to right, top to bottom.

Randomly select, by random number table, five [1] sections within the rectangle such that each contains a soil mound which contains only one shrub and which contains a desert pavement area.

Locate a point on the desert pavement, in the first section selected, record the Nevada grid coordinates of this location, take a 35 mm photo of location, take a FIDLER reading at one foot height, and collect a vertical soil profile, to 25 cm depth, by standard NAEG procedures for collection of soil samples from NAEG intensive study areas (10 x 10 x 2.5 cm deep scoop method). Record Gilbert stake number as 1 for first grid section, 2 for second grid section desert profile, etc.

Within the first section selected, locate the center of the shrub at the highest elevation in the mound. Record the Nevada grid coordinates of this point.

If stereo camera is available, take top view and side view photos of mound. If stereo camera is not available, take top view and side view photos with 35 mm camera.

Using a random number table and the number series one (1) through 360, randomly select three degree numbers which will allow sufficient room to take three standard profile samples around the shrub.[2]

Beginning at the shrub center point, establish a line (sample line) to the outer edge of the mound at the number of degrees from true north first [3] selected. Record the degree reading. Select a point (sample point) on the sample line that is one-half the radius of the shrub canopy.

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[1] Later changed verbally by P. B. Dunaway to read 10.

[2] This paragraph was later changed to read, "Using the shrub center as a reference point, select a location about the shrub at which a vertical profile can be taken."

[3] The word "first" was later deleted and changed to read, "at which profile location was . . ."

Record the distance from the shrub center point to the sample point. Measure a distance of 10 cm [4] along the sample line, from the sample point, toward the mound edge. Take FIDLER reading at one foot height. At this point, vertically cut away the soil mound and expose a natural profile face which forms a vertical plane, perpendicular to the sample line. Vertically insert a thin smooth steel sheet (15 cm x 100 cm [5]) at the sample point, perpendicular to sample line, to a depth of 30 cm.[6]

Along each of two lines which are parallel to and about 5.5 cm [7] on opposite sides of the sample line, vertically insert a thin steel sheet (50 cm x 100 cm [8]) flush with the sample point sheet, to a depth of 30 cm.[6]

Saturate mound surface at sample point and surrounding area (about one square foot) with fine water spray (i.e., Hudson sprayer) and allow sufficient time for water to penetrate the soil to a depth of at least 3 cm.

Measure and record the height, in centimeters, from the nearest desert pavement surface on the sample line, to the sample point.

Collect vegetation from shrub previously selected, by standard NAEG procedures for collection of vegetation samples from NAEG intensive study areas.

Collect vertical soil profile, to 25 cm depth, by standard NAEG procedures for collection of soil samples from NAEG intensive study areas (10 x 10 x 2.5 cm deep scoop method).

In like manner, perform collection, measurements, and photography of the two other vertical soil profiles from same mound.[9]

Record Gilbert stake number as 1-1 for first [10] profile in first mound selected, 1-2 for second profile in first mound selected, and 1-3 for third

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[4] "10" later changed to read "12.5."

[5] "100" later changed to read "35."

[6] ". . . to a depth of 30 cm" was later changed to read ". . . through mound, to desert pavement surface."

[7] Later changed to read 7.5.

[8] Later changed to read 25 cm x 35 cm.

[9] Entire sentence later deleted.

[10] ". . . first . . ." later changed to read ". . . the . . ."

profile in first mound selected,[11] 2-1 for first [12] profile in second [13] mound selected, etc.

Using the lowest elevation desert pavement point previously selected, for measuring height from desert pavement to sample point, measure and record height, in centimeters, from this desert pavement point to the highest soil point on the mound. This is the total mound height.

Measure and record the height, in centimeters, from the other two desert pavement points, for the same mound, to the highest soil point on the mound.[14] The height measurement information from the three desert pavement points to the highest point on the mound can be used to determine the slope of the mound base.

Extensive field notes are to be taken, to include such things as near sample debris notations, hot particles detected by FIDLER near sample location, vegetation cover on mound, mound disturbances by animals, etc. If vegetation is found in a soil profile sample section, it is to be removed and placed in a separate collection container. The container is to be labeled to indicate the sample location information and the soil section from which the vegetation was removed.

Sample Preparation

Each soil and vegetation sample is to be assigned identification numbers.

The wet weight of each sample is to be taken and recorded. The sample is to be oven dried at 105° C. for 24 hours. The dry weight is to be taken at ambient temperature and recorded.

The vegetation samples are to be slow ashed in a muffle furnace to a temperature of 550° C.[15] The vegetation ash weight is to be taken at ambient temperature and recorded. The ash is to be transferred to a counting container as determined by the counting group.

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[11] ". . . 1-2 . . . and 1-3 . . . selected . . ." deleted.

[12] ". . . first . . ." later changed to read ". . . the . . ."

[13] ". . . in second . . ." later changed to read ". . . in the second . . ."

[14] First paragraph and this sentence later deleted and changed to read: "Measure and record the height, in centimeters, from the nearest desert pavement surface on the sample line, to the highest soil point on the mound. From each point, the lowest and highest point on the desert pavement about the mound, measure and record the height, in centimeters, to the highest soil point on the mound."

[15] Later changed to read 400° C.

A representative aliquot of each soil sample is to be transferred to a counting container. The container and aliquot size is to be determined by the counting group.

Sample Analysis

Each soil and vegetation sample is to be analyzed for ^{241}Am by a Ge(Li) system for sufficient time to give a 2 sigma counting error of less than plus or minus 5 percent or 30 minutes counting time, whichever is reached first.

Sample Storage

Each sample, the analyzed fraction, and the fraction which was not analyzed is to be stored for future analysis.

RESULTS OF PRELIMINARY STUDY OF SOIL MOUND AND DESERT PAVEMENT VERTICAL PROFILE PAIRS IN AREA 11, NTS*

NAEG MOUND STUDY PILOT PROGRAM

November, 1974

D. N. Brady
Reynolds Electrical & Engineering Co., Inc.

Introduction

Soil samples obtained from 10 soil mounds and 10 adjacent desert pavement locations near C site in Area 11 at NTS are discussed. These samples were obtained during September and October, 1974, by REECo personnel in support of a pilot study to determine the advisability of a more extensive mound sampling program in future.

Methods

A. Sampling Location Selection

A 100 foot by 100 foot square area was gridded into 10 foot squares near C site GZ in Area 11 (see accompanying diagram). Using a table of random numbers, 10 grid squares and one location in each square were selected with the limitation that each site must be desert pavement. A mound was then selected within a 10 foot radius of each of the desert pavement locations.

B. Mound Parameter Measurements

Each mound was carefully surveyed using a surveyor's transit and level. The C site GZ was arbitrarily selected as a common reference point, and measurements were obtained from a fixed distance above this point downwards for all measurements. The accompanying diagram defines all parameters. For purposes of clarity, all measurements have been converted from the field data sheets to a more comprehensible system. For each mound, a zero datum line was used as the reference point for all measurements associated with that mound. In every case, this line was the height of the lowest point of the mound base.

For intercomparison of mounds, tables of the heights of the zero datum lines, H_L, relative to the C site ground zero may be prepared.

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*See Editor's Note, page 33.

The mound sample locations were chosen by selecting a point perpendicular to the horizontal plane of the principal bush of the mound at a distance from the center of the bush equal to one half its average radius, R_B. The azimuths of these locations relative to the center of the bush were arbitrarily selected.

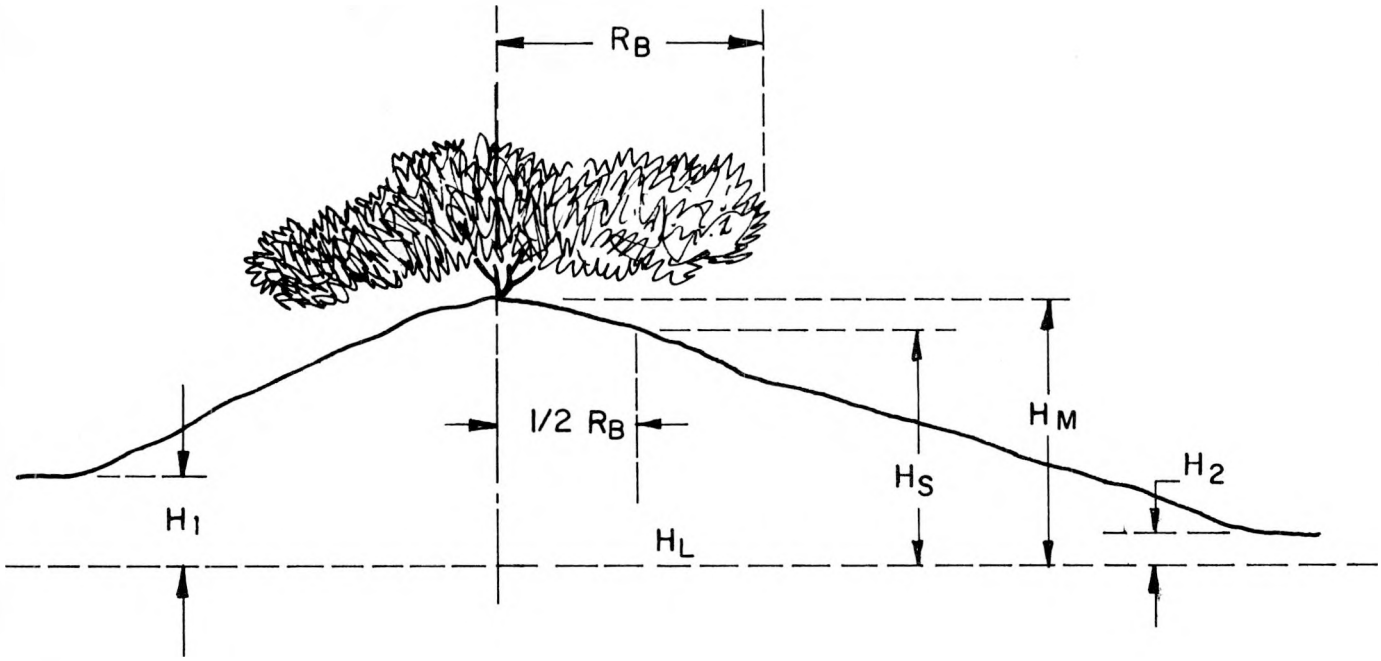
Ten soil samples were obtained from each mound and numbered consecutively from the top down, 1 through 10. Each sample was collected using the standard NAEG techniques, i.e., vertical plates were driven into the mound on three sides of the sampling location, and a scoop having internal dimensions of 10 cm x 10 cm x 2.5 cm was used to remove each sample starting from the top.

The desert pavement soil sample profiles were obtained using the standard trenching technique. These samples were numbered from the top down, 11 through 20.

FIDLER measurements were obtained at each mound and desert pavement sampling location.

Soil Sample Analysis and Results

Each soil sample was bottled in a 16 oz. Nalgene bottle without ball-milling, sieving, or other preparation, and counted for 30 minutes or until a ± 10% counting error was obtained. REECo's Environmental Sciences Department's automated Ge(Li) system was used to count these samples for the 60 keV ^{241}Am peak. In some instances, samples were recounted to double check the accuracy of the results.



H_L — HEIGHT OF LOWEST POINT OF BASE USED AS ZERO DATUM POINT.

H_1 — HEIGHT OF HIGHEST POINT OF BASE

H_2 — HEIGHT OF BASE IN LINE WITH SAMPLE AZIMUTH

H_S — HEIGHT OF SAMPLE BASE

H_M — HEIGHT OF MOUND

H_P — HEIGHT OF DESERT PAVEMENT AT PROFILE SAMPLING POINT (Not illustrated)

R_B — RADIUS OF BUSH CANOPY

A_Z — AZIMUTH IN DEGREES FROM TRUE NORTH TO SAMPLE POINT (Not illustrated)

W — WIDTH OF MOUND (Not illustrated)

L — LENGTH OF MOUND (Not illustrated)

FIGURE 1.
MOUND SAMPLING PARAMETERS

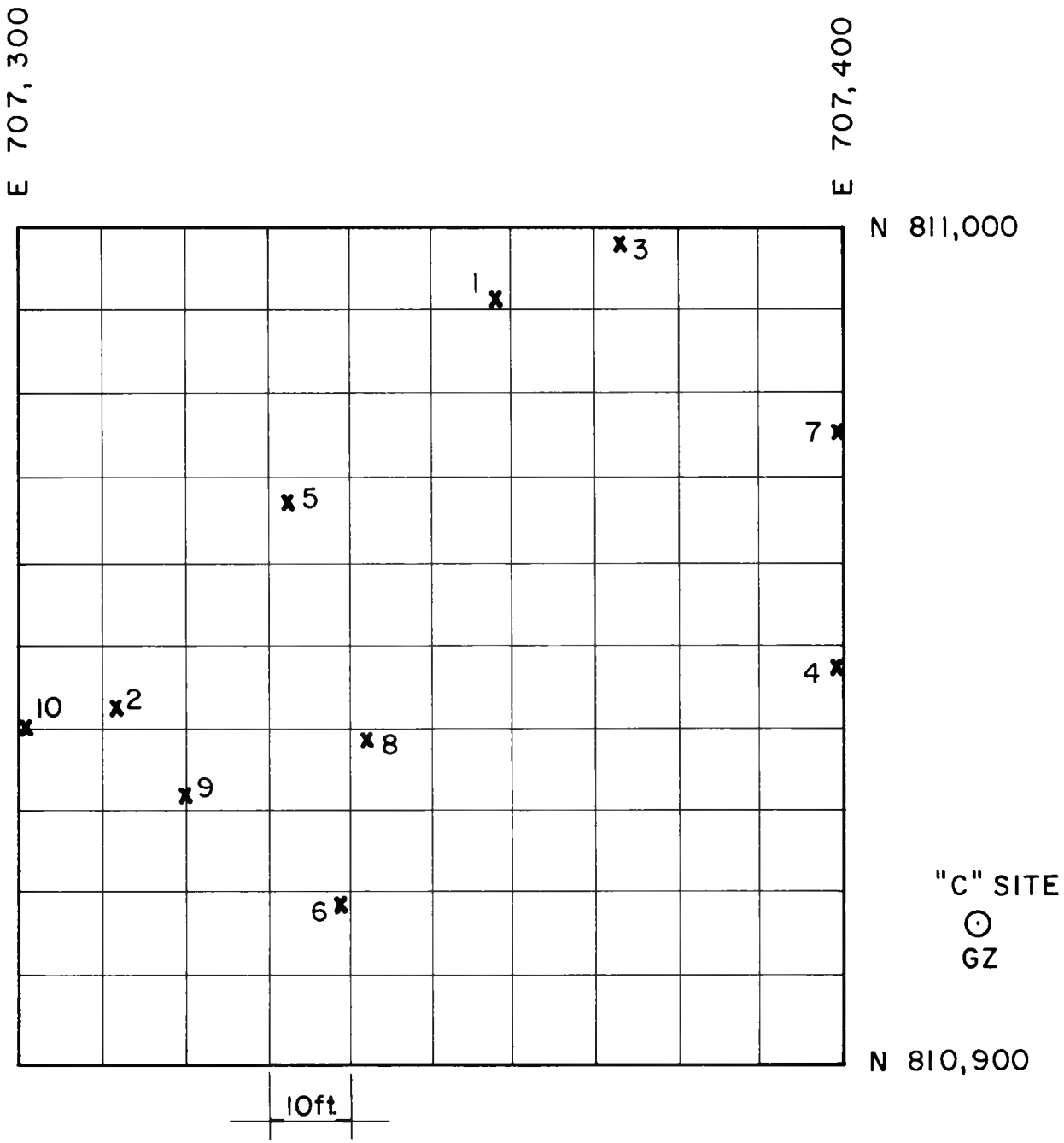
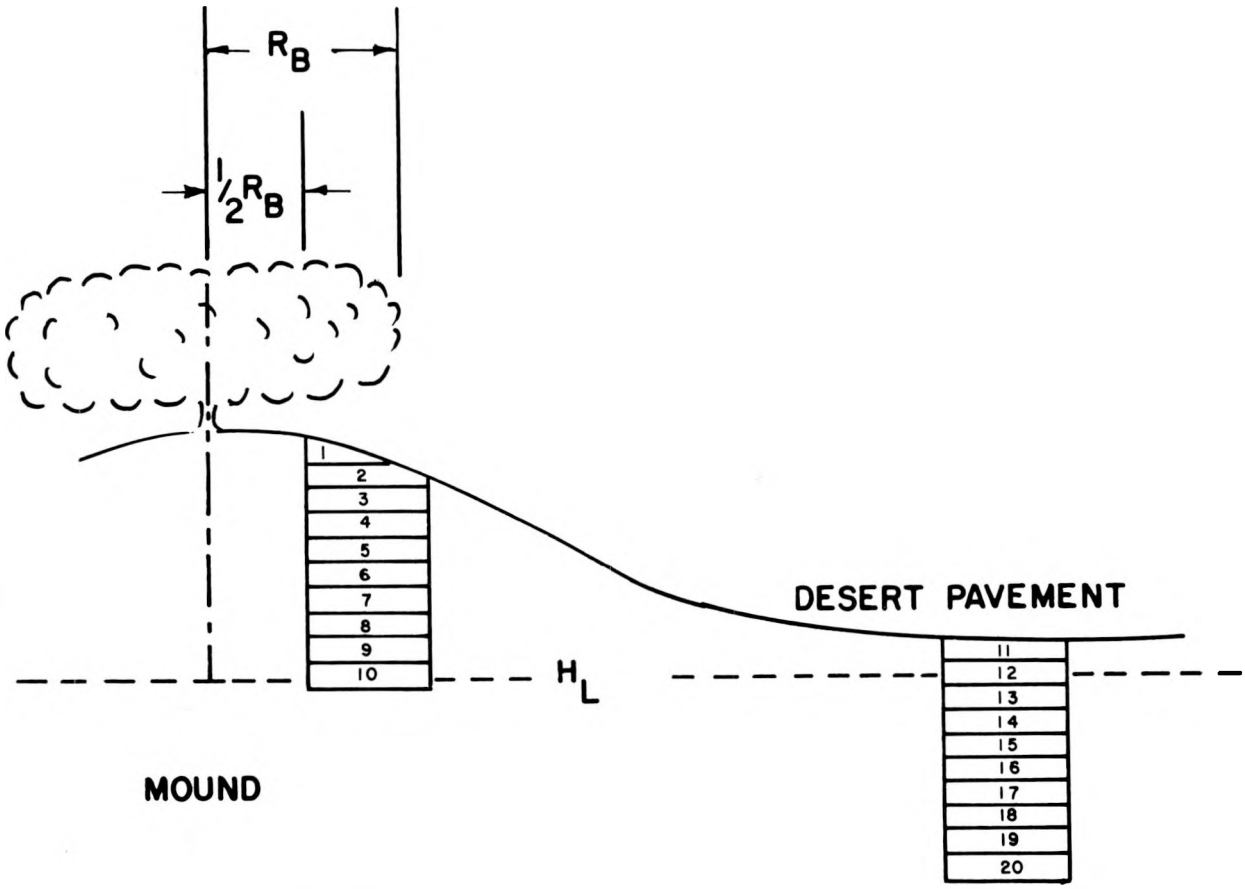


FIGURE 2.

PLAN VIEW: AREA-2 MOUND STUDY RANDOMLY SELECTED SOIL SAMPLING LOCATIONS - DESERT PROFILES.



SOIL SAMPLE POSITIONS AND NUMBERING SEQUENCE

FIGURE 3

BATTELLE PACIFIC NORTHWEST LABORATORIES

January 29, 1975

Mr. Paul B. Dunaway*
Office of Effects Evaluation
Nevada Operations Office
Atomic Energy Commission
P.O. Box 14100
Las Vegas, Nevada 89114

Dear Paul:

Some ideas are set down here for your consideration concerning the sampling of "blow-sand" mounds on the NAEG intensive study sites. These have evolved from the meeting we had in Las Vegas January 16-17, 1975, with Van Romney, Glen Bradley, and others, as well as from conversations since then with Lee Eberhardt, Eric Fowler, Van Romney, and others. My trip to Areas 13, 5 (GMX), and 11 January 17 was particularly helpful. Following a rather general discussion below, the sampling plan is summarized at the end of this letter.

First, I suggest we delay any attempt to sample specifically for estimating plutonium inventory in blow-sand mounds until we first sample to estimate the distribution of Pu throughout the mound. Sampling for Pu distribution should give us at least some information to decide whether a mound inventory sampling program is really necessary. It would be ideal, of course, if a single sampling plan could be used for estimating both inventory and distribution. But, the detailed sampling of a mound needed to estimate distribution necessarily (due to cost restraints) limits the number of different mounds that can be examined, which is undesirable if one wants to estimate inventory.

Coincident with a sampling plan for estimating Pu distribution within mounds there should be an effort to take field measurements of the kind that would be necessary in any future sampling for inventory purposes. To estimate Pu inventory in mounds, it is necessary to have information on the proportion of total land area covered by mounds, the total number of mounds, and the volume of individual mounds. Ways of estimating these parameters are suggested below. We should determine now the accuracy with which these parameters can be estimated and whether the suggested methods are practical to use in the field.

Due to the expense of collecting and analyzing large numbers of profile samples, I suggest that sampling for distribution of Pu within mounds be limited at least initially to 2 or 3 of the study sites. We talked about this briefly over the phone January 23. In a phone conversation with Van Romney on January 24, he suggested that Area 13 and either C or D site of

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*See Editor's Note, page 33.

Area 11 be chosen for study. He noted that Area 5 might not be the best choice since multiple tests were conducted there, and results obtained there might not be applicable to single-shot sites. He felt Area 13 should be studied since inferences could be made from Area 13 to the three Clean Slate sites on the Tonopah Test Range due to similarities in mound structure. The pilot mound study was done at C site, Area 11, and it might be well to build on that data base, although Van felt D site would be acceptable also.

There seemed to be general agreement at the January 16-17 meeting that sampling of mounds with rodent burrows should be separate from sampling for inventory and distribution within mounds. Hence, I suggest that only non-rodent mounds be chosen for the distribution study proposed in this letter.

At each of two study sites, say Area 13 and C site,[1] I suggest:

A. Select 5 locations in the inner two strata (nearest ground zero), each location being the center of a X by X foot square, where X would depend on the density of mounds. In Area 13 we might use X = 20 feet; for C site it appears X = 40 feet is more appropriate. The 5 sampling squares would not be located in disturbed (bladed, plowed, trampled) areas. I could choose these locations at my desk, but the field crew must use judgement in deciding if the designated area is disturbed. Several alternative locations could be provided the field crew for such eventualities.[2]

B. Within each of the 5 squares per study site the following counts should be made:

    1. Count the number of individual shrubs present (irrespective of species). Define a shrub to be in the square if at least half of its canopy is in the square.

    2. Count the number of "sampleable" mounds in the square. A "sampleable" mound is a "well formed" mound; one large enough so that several profile samples could be collected in the mound without danger of cross contamination between levels of a profile or between the profiles. There will usually be one or more shrubs growing in the mound. These sampleable mounds are those in which profile samples are to be taken (see below for details). The

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[1] The first two sites selected were CS3 (TTR) and Area 13, NTS (reference "Laboratory Sample Preparation Protocol for NAEG Mound Samples," E. H. Essington to P. B. Dunaway dated October 23, 1975).

[2] Actual size selected was 100 x 100 ft. plot, subdivided to 50 x 50 ft. sections, and then subdivided to 50 x 10 ft. sections (reference Essington "NAEG Soil Mound Study No. 2 (Revised)," November 4, 1975, page 3, Items 1, 2, and 3).

idea here is to distinguish between mounds to which the distribution sampling results apply from those smaller mounds that, practically speaking, cannot be sampled using the established profile method used to date in the NAEG program. This count data is not essential to the problem of estimating Pu distribution within the mound, but it is necessary if we are to get a rough handle on the total amount of Pu that might be in the sampling square. A "sampleable" mound is not a large mound area covered by grassy species. These grassy mounds should be studied separately, apart from the sampling program described in this letter.

3. Count the total number (n) of mounds present in the square, no matter what their size. This count will include the "sampleable" mounds tallied separately in 2 above. There may be a problem here in deciding whether a mound does or does not exist at the base of some of the small shrubs, especially since mounds can change in size with the season.

The information in 1, 2, and 3 above can, I suppose, be obtained by the field crew fairly rapidly and will be useful for future studies.

C. Within each of the 5 sampling squares choose a sampleable, non-rodent mound at random by numbering the non-rodent mounds from 1 to m and choosing a number from 1 to m from a random number table. Ideally, the mound should be large enough to take 4 profiles (Figure 1). If no such mound exists in the sampling frame, go to an alternative sampling frame (location supplied by myself). If none of the mounds in the area are sufficiently large to take 4 profiles, then take only profiles 1 and 3 or 1, 3, and 4 (Figure 1). The chosen mound must be large enough to take at least profiles 1 and 3.[3]

D. Remove at ground level (top of mound) all vegetation by cutting or sawing, taking care not to disturb the mound. Eric Fowler has suggested some of this vegetation be saved and dated to get some notion as to the age of the mound. The vegetation might also be saved and analyzed for plutonium (wet chemistry) and ^{241}Am (Ge(Li)).

E. Photograph the mound with stereo camera for purposes of computing the volume of the mound and to have a record of the mound shape.

F. Take FIDLER measurements over each of the 6 profile locations (1 foot from surface).[4]

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[3] Refer to "NAEG Soil Mound Study No. 2 (Revised)," November 4, 1975, for mound type criteria actually used.

[4] No profiles were taken in CS3 (TTR) and Area 13 (reference "NAEG Mound Study No. 2 (Revised)," November 4, 1975, pp. 3-6).

G. Take sufficient tape measurements (without disturbing the mound) in order to approximate the area and volume of the mound. Mounds are irregular in shape so that measuring the "length," "width," and "height of the mound may not be sufficient to get good estimates. Presumably a tape measure laid over the mound in several directions in combination with the usual length-width-type data would be useful. At the very least, measurements like those obtained on the mound pilot study in C site should be taken on all sampled mounds.

H. For the mound chosen in C. above, take 4 profile samples in the mound and 2 profile samples adjacent to the mound as shown in Figure 1. Note that mound profile 1 and 3 are on a line toward and away from ground zero to estimate the difference at these locations of Pu deposited at the time of the safety shot. Ed Essington (phone conversation January 24) suggested that a more important variable around which to center a mound distribution sampling program would be prevailing wind direction. He suggested in effect that profiles 1 and 3 be orientated upwind and downwind rather than in relation to ground zero. The orientation of each sampled mound relative to ground zero and wind direction will differ from mound to mound. In some cases profiles 1 and 3 might be orientated to GZ as shown in Figure 1 and simultaneously be orientated upwind and downwind. No matter what the orientation, profiles 1 through 4 should give some information on both GZ and prevailing wind effects. I am not suggesting the approach in Figure 1 is necessarily the best method, and would be willing to orient the mound sampling specifically to prevailing wind direction if the soils, vegetation, and resuspension elements agree it is the principal factor accounting for distribution of Pu in mounds. Norm Kennedy (phone conversation January 24) indicated that the general prevailing winds >8 mph (wind speeds below this would presumably not move an appreciable amount of soil) are from the south-southwest (measured from true north) in Area 11 and from the north in the vicinity of Area 13. This kind of information would obviously be needed in a "wind direction" orientated sampling program. One thought to keep in mind here is that the strata maps indicate the effects of original blast and fallout as well as redistribution of Pu via wind action. Hence, their general shape may be an indication of the best orientation of profiles 1 and 3.

I. Profiles 5 and 6 (Figure 1) would estimate Pu distribution with depth in desert pavement areas "close to" the mound, say 1 foot from the edge of the mound.

J. An ideal mound is shown in Figure 1. Actual mounds will be irregular in shape and have interfering vegetation so that exact placement of profile samples will necessarily be decided in the field. We should insist, however, that the agreed upon plan be adhered to as closely as possible.

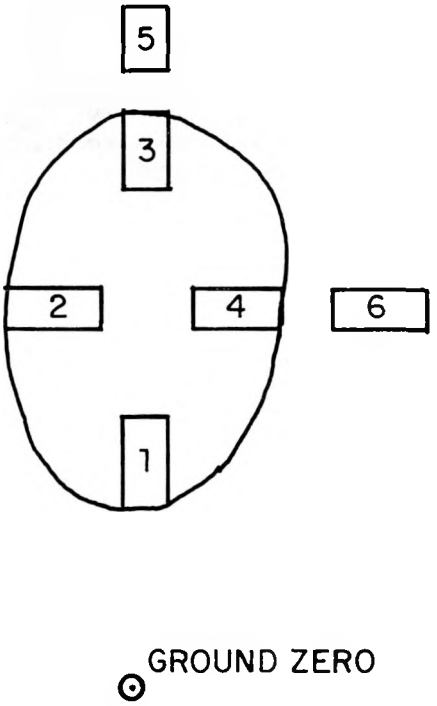

FIGURE 1

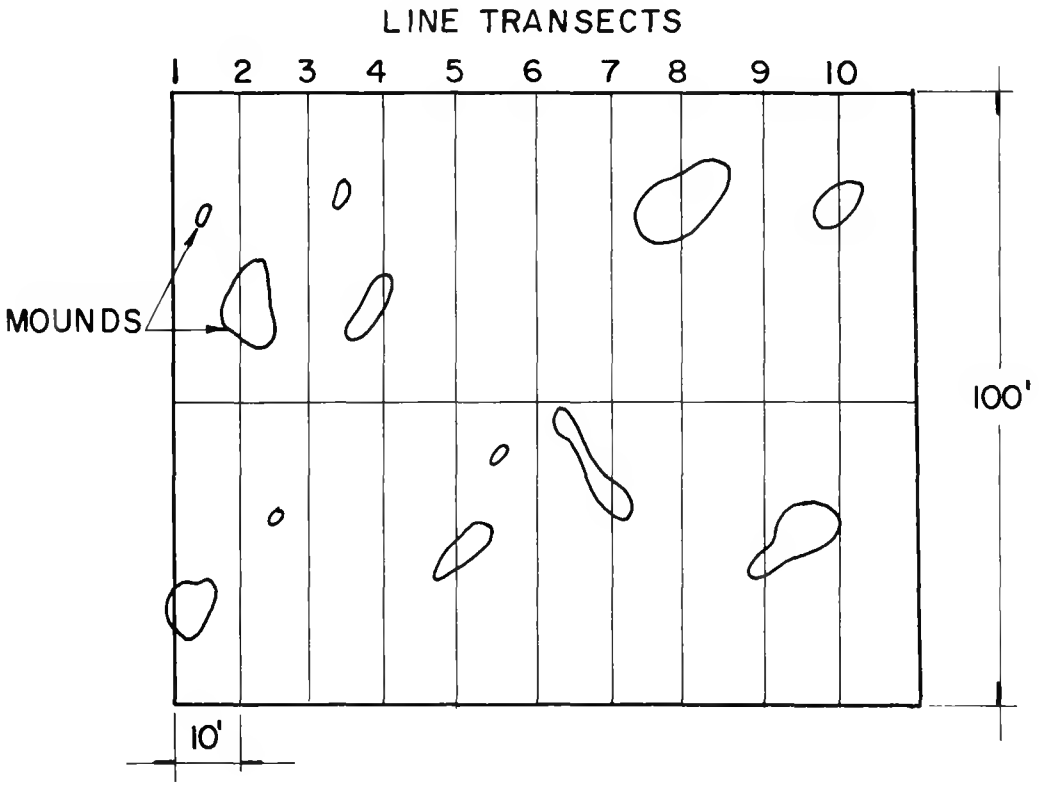

FIGURE 2

K. Take ten 2.5 cm increment samples in each of the 6 profiles associated with each mound. Ge(Li) scans for ^{241}Am would be conducted on all samples; $^{239-240}Pu$ (wet chemistry) analyses could be done on only levels 0-2.5 cm, 2.5-5 cm, and 10-12.5 cm. The Pu analysis on the 10-12.5 cm level would be done to check on the Pu/Am ratio with depth, i.e., is the distribution of Am throughout the mound really indicative of the distribution of Pu. For levels 0-2.5, 2.5-5, and 10-12.5 cm, or perhaps just the 10-12.5 level, you may want to consider longer counting times than usual for ^{241}Am so that the Pu/Am ratio can be estimated with sufficient accuracy to answer this basic question. A level below 12.5 cm could be chosen instead of 10-12.5 cm, but Am counting times might get longer to get as accurate a count.[5]

L. Based on past difficulties, I suggest none of the soil samples be sieved through a 100 mesh sieve before analyses. I presume ball-milling is required for those samples analyzed both by wet chemistry for Pu and Ge(Li) scans for ^{241}Am, but not for the samples only Ge(Li) scanned. It might be best to ball-mill all samples to eliminate variability due to that factor.

M. Lee Eberhardt suggested to me last week the possibility of using line transects to get information on mound cover. To obtain information on the proportion of field surface covered by mounds, I suggest that following the removal of profile samples from the 5 mounds in the study site, a 90 by 100 foot area be chosen at random in an undisturbed location. Along 10 line transects each 100 feet long and 10 feet apart, record the number of feet on the stretched tape measure that covers a mound (Figure 2). Record this information for each line separately so that we have 10 estimates with which to estimate the variance of the estimated proportion of land area covered by mounds. Line transects are frequently used to estimate canopy cover; here we would be measuring "mound cover." For this study, mounds of all sizes should be included in the measurements. Again, this is information required for future estimates of mound inventory. It is not necessary in the study of distribution of Pu within mounds.

In summary:

--Study distribution of Pu in mounds now; leave estimating Pu inventory for later.

--Study only Area 13 and C or D site of Area 11 now. Do other sites later if necessary.

--Select 5 mounds in each of the two study sites; remove vegetation; save for aging and analysis?

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[5] No profiles were taken in CS3 (TTR) and Area 13 (reference "NAEG Mound Study No. 2 (Revised)," November 4, 1975, pp.3-6).

--Take stereo picture of the mound.

--Take FIDLER readings over all 6 profile locations per mound.

--Take sufficient tape measurements to estimate the area and volume of the mound to be sampled.

--Take 4 profiles (10 increments of 2.5 cm per profile) in the mound and 2 profiles in adjacent desert pavement in an orientation toward GZ as shown in Figure 1. Alternatively take samples so that profiles 1 and 3 in all mounds are orientated upwind, downwind. If mounds are too small, we could eliminate profile 2 and then profile 4 if necessary.

--Within the sampling square surrounding each sampled mound count the number of (1) shrubs, (2) "sampleable" mounds, and (3) all mounds (including sampleable ones).

--In a 90 by 100 foot square, run 10 line transects each 100 feet long; on each measure the number of feet of the line covering a mound (of any size); record separately for each line (Figure 2).

--Do Ge(Li) scans for ^{241}Am on each sample. No ball milling required?

--Do Pu wet chemistry determination on levels 0-2.5, 2.5-5.0 cm; ball-milling required; no sieving.

--Analysis cost. There are 6 profiles per mound x 10 samples per profile x 5 mounds per study site x 2 study sites = 600 samples, each of which cost about $25 for a Ge(Li) scan. This comes to $15,000. Thirty percent of the 600 samples would have Pu analysis at $75 each. This comes to $13,500. Hence, total analysis cost = $28,500.

Turning for the moment to a rodent-mound study, the general features would seem to be (1) among those animals known to be resident in the study site, choose one or more at random and follow him to his burrow, (2) sample the mound according to the specific objectives of the study. I have not as yet had the opportunity to talk with Glen Bradly about specific objectives and until I do there isn't much point in describing a detailed plan. Rather than delay this letter any more I prefer to discuss the sampling of rodent mounds at a later time.

I hope, Paul, that the above discussion is useful. It follows the spirit of the mound study program suggested by Eric Fowler and Ed Essington last year. As laid out above, the distribution study would be confined to rather large, well formed non-rodent mounds. The applicability to other size mounds and rodent mounds would be unknown. Taking counts, mound measurements, and line transect data now should help in the planning of future inventory studies and perhaps allow us to get a very rough idea of inventory from the data collected here for distribution. I hope those people whose names I have used here will forgive me for any errors I may

have made in interpreting their words over the phone. I'm looking forward to receiving suggestions for improvement in the sampling program.

Best regards,

R. O. Gilbert
Senior Research Scientist
Statistics Section
Systems Department

cc: E. H. Essington, LASL
W. G. Bradley, UNLV
E. M. Romney, UCLA
M. G. White, AEC/EED, NV
L. L. Eberhardt, BNW
E. B. Fowler, LASL

ROG:lt

MOUND STUDY NO. 2 PROTOCOL*

August, 1975

E. H. Essington
Los Alamos Scientific Laboratory

Mission[1]

Determine contribution of plutonium in various types of mounds to total plutonium inventory of area down to 5 cm datum.

Determine percent of area covered by mounds of various types as opposed to desert pavement.

Determine Pu/Am for selected samples.

Clean Slate 3

A. General

Five types of mounds were identified which can be unique contributors to total inventory.

1. Grass Clump--a small lone mound developing under a clump of grass, such as Indian ricegrass. Mound may be 10-15 cm diameter and 1-2 cm high.

2. Shrub--a small lone mound developing under a single shrub, such as Atriplex. Mound may be 25-30 cm diameter and 3-5 cm high.

3. Shrub/Complex--large mound of varying shape 0.5-2 m containing various species of vegetation including grasses.[2]

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[1] Mission changed as indicated in Essington "NAEG Soil Mound Study No. 2 (Revised," November 4, 1975, page 1).

[2] Changed to read ". . . grasses and 10-40 cm high." (Reference Essington "NAEG Soil Mound Study No. 2 (Revised)," November 4, 1975, page 2, Item 3).

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*See Editor's Note, page 33.

4. Animal Dig--usually large mounds resulting from activities of either large burrowing animals or large colonies of small burrowi animals. The large colony type mounds will be excluded from considerations in this study since they are few in number. Their combined influence on plutonium inventory is expected to be very samll, and they are prime specimens for animal habitat studies.

5. Diffuse Grass--low flat mound of 0.5 m diameter up to 10 m across. Many small, low, grass tufts throughout mound area.

B. Data and Related Information Required

1. Coordinate (Nevada Grid) location of selected mound.

2. Category or type of mound as defined in A above.

3. Physical measurements--long axis, normal short axis using tape measure, height relative to desert pavement using level or scale.

4. Stereo pair of photos before vegetation harvested to show as much of mound as possible.

5. Stereo pair of photos after vegetation harvest from same aspect as in 4 but before sampling.[3]

6. Identity of vegetation and specifically of shrub from which sample is collected for age determination.

7. FIDLER reading of desert pavement surface sample.

8. FIDLER reading of mound after removal of vegetation.

9. FIDLER reading of desert pavement plane under mound after top of mound is removed.

C. Sampling[4]

1. Locate 100 x 100 ft plot using NG coordinates given.

2. Randomly select one of the four 50 x 50 ft quadrants.

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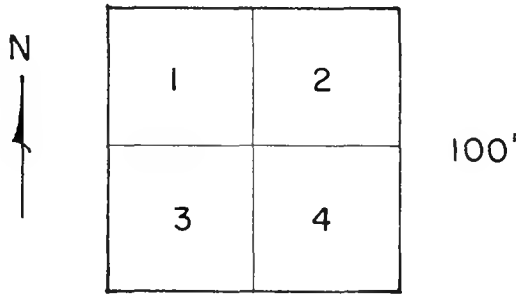
[3] ". . . but before sampling" was deleted (reference Essington "NAEG Soil Mound Study No. 2 (Revised)," November 4, 1975, page 3, first Item 5).

[4] This section changed as indicated in Essington's "NAEG Soil Mound Study No. 2 (Revised)," November 4, 1975, pp. 3-6, "Sampling" section).



3. Starting in the NW corner of the chosen quadrant number individual mounds categorized according to the list in A above (e.g., Grass Clump mounds numbered from 1 to N_1).

4. Choose two random numbers between 1 and N_1 which correspond to the required mounds to be sampled, and mark with surveyors ribbon.

5. Repeat step 3 and 4 for second type of mound recognized and for each remaining type. Make and record measurements outlined in B above, i.e., Items 1, 2, and 3.

6. From this point on work on one mound at a time.

7. Photograph mound in stereo (3 frames) approximately 10 cm apart at lens.

8. Harvest vegetation according to established NAEG procedures.

9. Photograph bare mound in stereo at same aspect as in 7 above (3 frames).

    a. Make FIDLER reading of mound according to established NAEG procedure.

10. Collect 12.5 cm diameter x 5 cm deep surface soil sample plus FIDLER reading from a desert pavement location 10 cm from edge of mound to closest edge of sample normal and to the left of prevailing winds using established NAEG procedures.

11. Collect mound to the original desert pavement area established with a level. Smaller mounds collected in total placed in sample container. Large mounds, which must be subsampled, weighed, and placed in mixer, in total, mixed, subsampled into storage container (e.g., 1 gallon paint can).

    a. Clean mixer with three loads of sand or uncontaminated soil.

    b. Take FIDLER reading from some vertical location at level of original desert pavement (according to Protocol No. 1 for Mound Sampling).

12. Install 5 cm wide flexible baffle plate outlining mound. Collect material under mound to 5 cm depth within boundary of baffle. Weigh, mix, and subsample as in 11 above for large samples. Small samples retained in total.

13. Repeat sampling procedures steps 7 through 12 for each mound type.[5]

Area 13

Sample in same manner as Clean Slate 3 identifying only three types of mounds:

1. Shrub/Complex

2. Shrub

3. Animal Dig

---

[5] Diffuse grass mound sampling.

1. Locate randomly a diffuse grass mound within the 50 x 50 square plot. Sketch mound on coordinate paper, number squares consecutively within boundary of mound, randomly select four squares as sampling point. Record location, photograph as in 9 above.

2. Collect desert pavement sample as in 10 above.

3. Collect mound sample also as in 10 above to desert pavement datum, then collect additional 5 cm depth, using ring, as sample below mound. Take appropriate FIDLER readings.

Stereo Aerial Photographs

The percentage cover that each mound type represents of the total intensive study site is a factor that must be resolved before further inventory studies are attempted. During a low-level flyover of areas at NTS and TTR at 100-1,000 ft above ground, observations were made which suggest that stereo color or infrared photographs at about 100-200 ft may be usable to determine the percent mound cover. The alternative would be to transect (on foot) the plot by many parallel lines and measure the percentage cover. Aerial photos may offer a much cheaper and direct means when considering the numbers of sites which would be so treated.

Several methods were suggested:

1. Contractor flyover using professional aerial photographic equipment. (Limitation--cannot fly low enough to produce desired resolution and access.)

2. Helicopter plus hand-held camera. (Limitation--access.)

3. Tethered weather balloon. Best method but requires ground crew and some camera modifications.

The various possibilities of contractor service are being pursued by NAEG (NVO).

The photographs should be overlapped so that there are three along the N-S line and three along the E-W line for a total of five [6] frames. Corner markers calibrated as to distance and differential elevation should be installed prior to aerial photography.

---

[6] Changed to read "6" (reference Essington's NAEG Soil Mound Study No. 2 (Revised)," November 4, 1975, page 10).

Present Study[7]

The present mound study was limited to providing information relative to inventory and not to plutonium migration mechanisms. However, future studies will be considered to evaluate or study the various effects mounds have on plutonium redistribution. Several are listed here:

1. Effect of mound in altering the horizontal distribution of plutonium.

    a. Upwind vs downwind concentrations on the mound.

    b. Upwind vs downwind concentrations on desert pavement and formation of plume.

2. Correlation of mound aspect to plutonium accumulation.

    a. Upwind vs downwind.

    b. Wind direction at time of detonation.

    c. Position and aspect relative to GZ.

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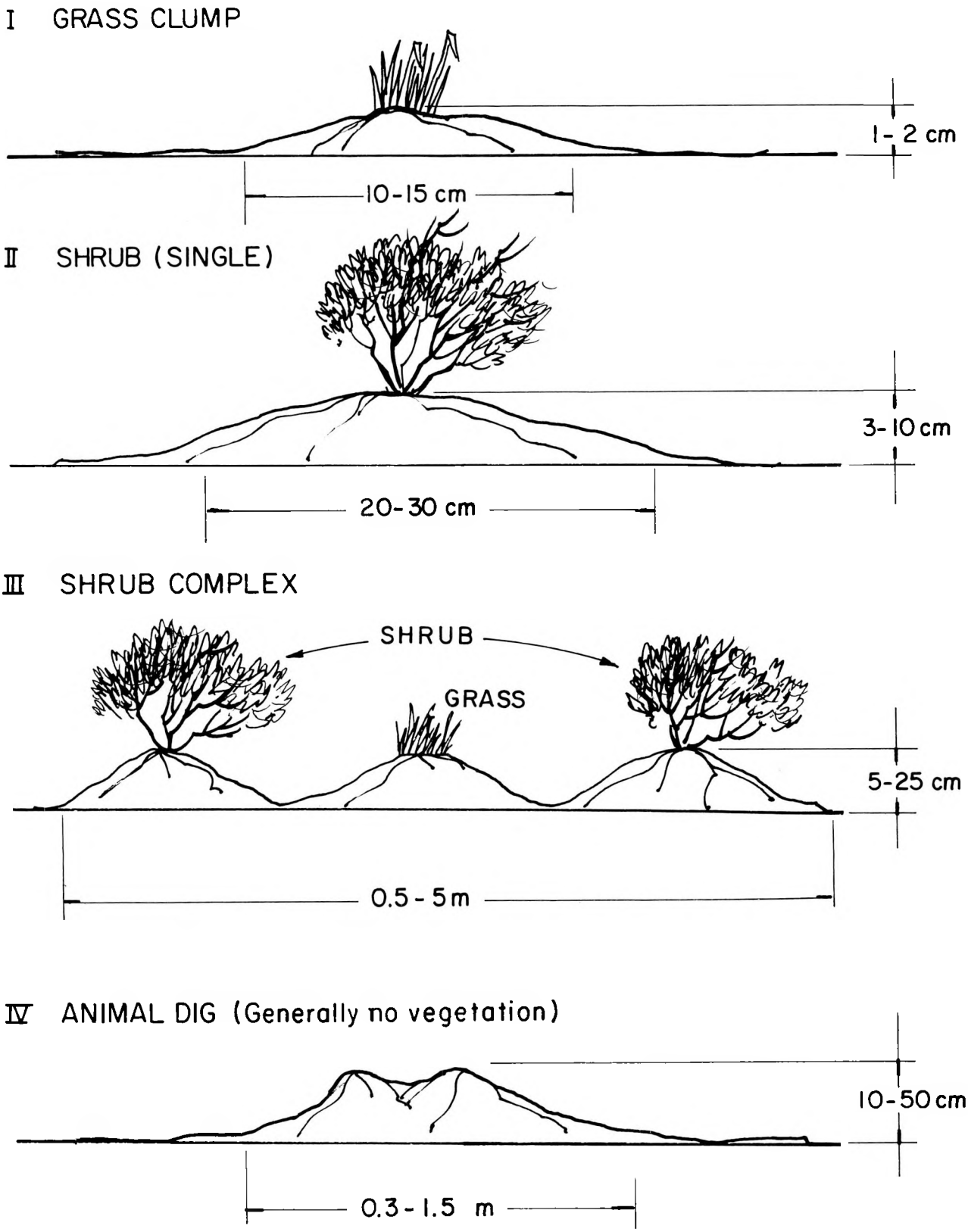
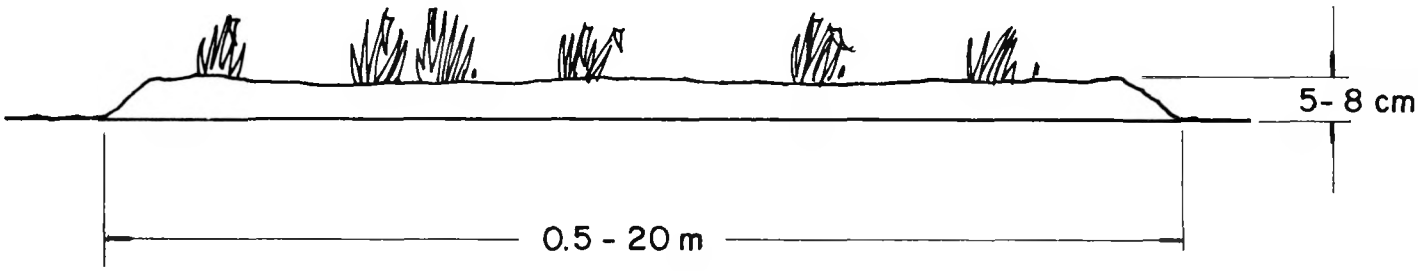
[7] Not included in Essington's "NAEG Soil Mound Study No. 2 (Revised)," November 4, 1975.

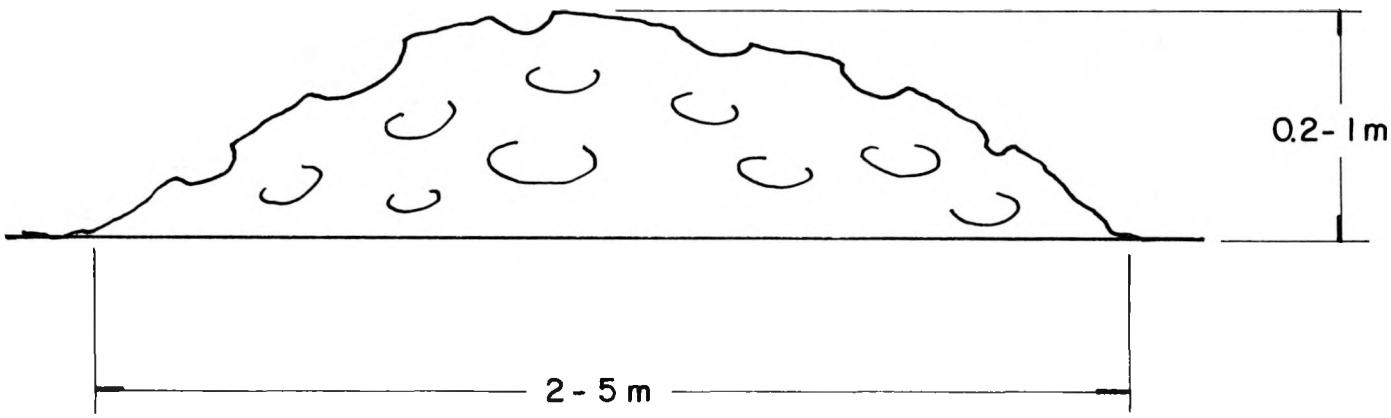

I GRASS CLUMP
1-2 cm
10-15 cm
II SHRUB (SINGLE)
3-10 cm
20-30 cm
III SHRUB COMPLEX
SHRUB
GRASS
5-25 cm
0.5-5m
IV ANIMAL DIG (Generally no vegetation)
10-50 cm
0.3-1.5 m

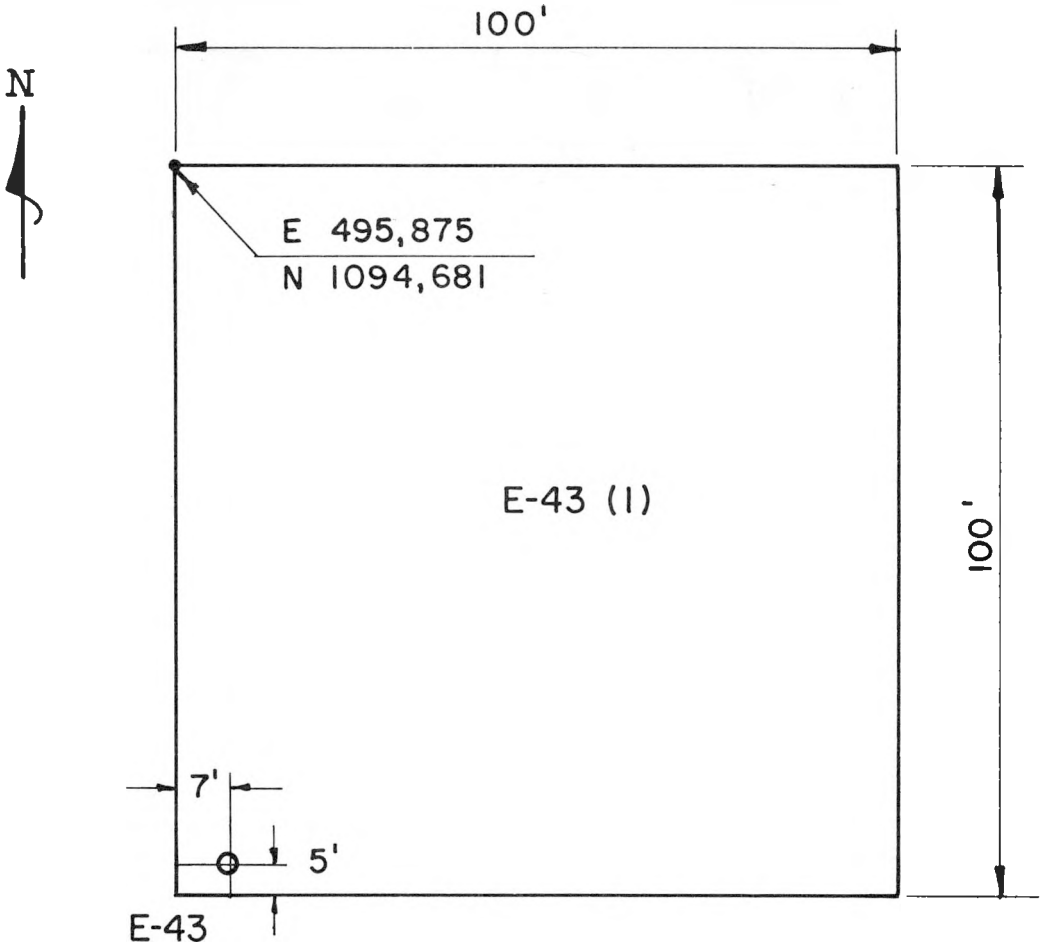
MOUND TYPES

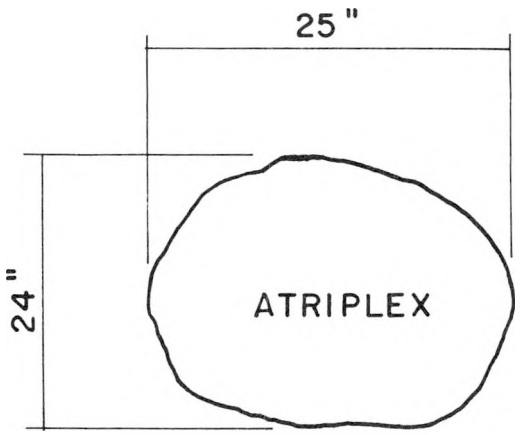
V DIFFUSE GRASS



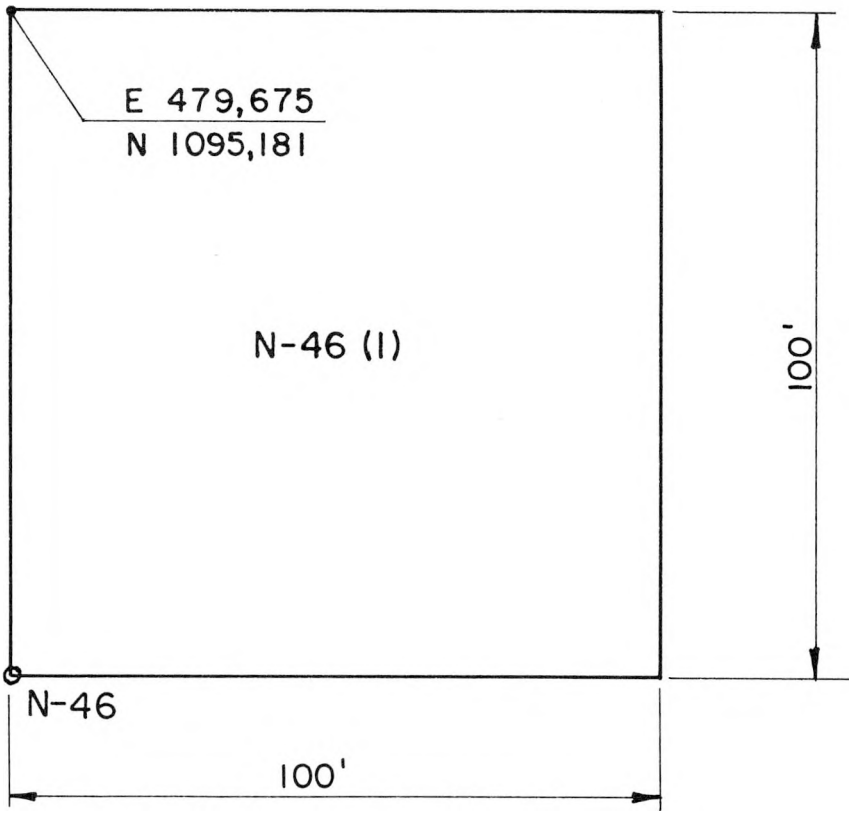
VI LARGE ANIMAL DIG (Generally colony of small diggings, no vegetation)

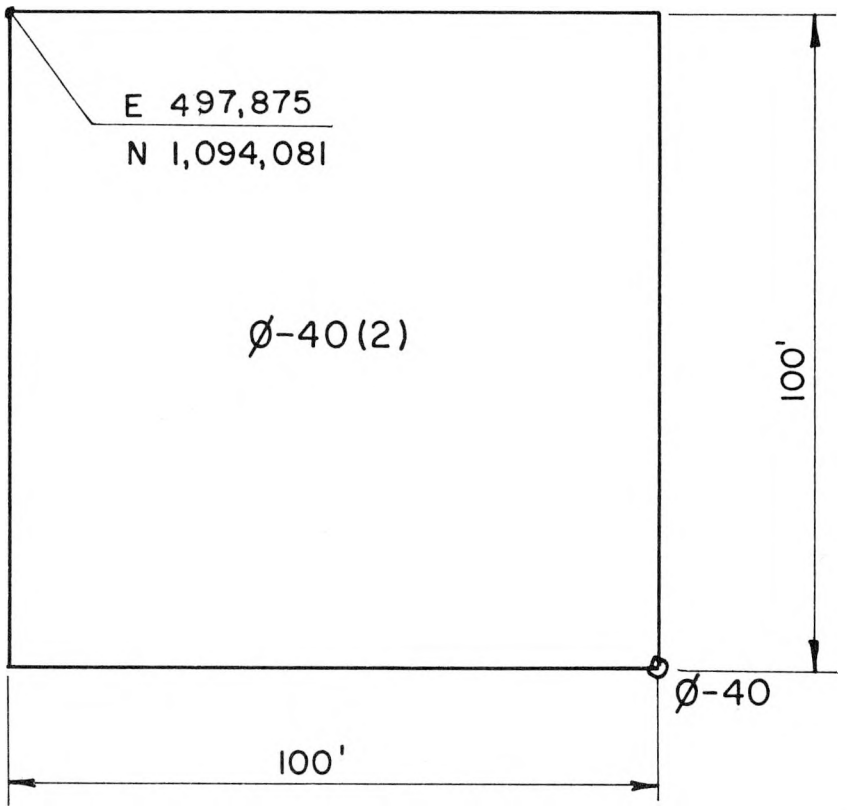


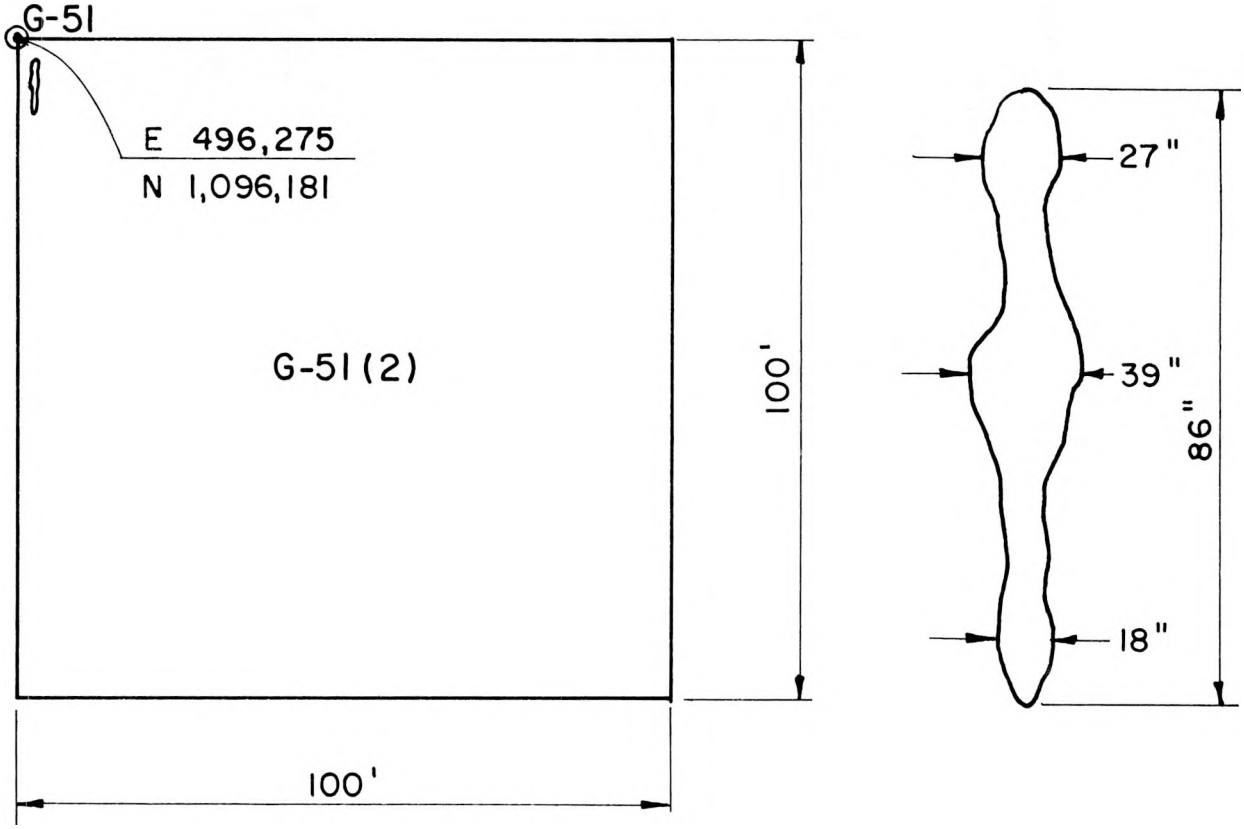




CLEAN SLATE 3 PLOT DESCRIPTION


E 479,675
N 1095,181
N-46 (1)
100'
N-46
100'


E 497,875
N 1,094,081
Ø-40 (2)
100'
Ø-40
100'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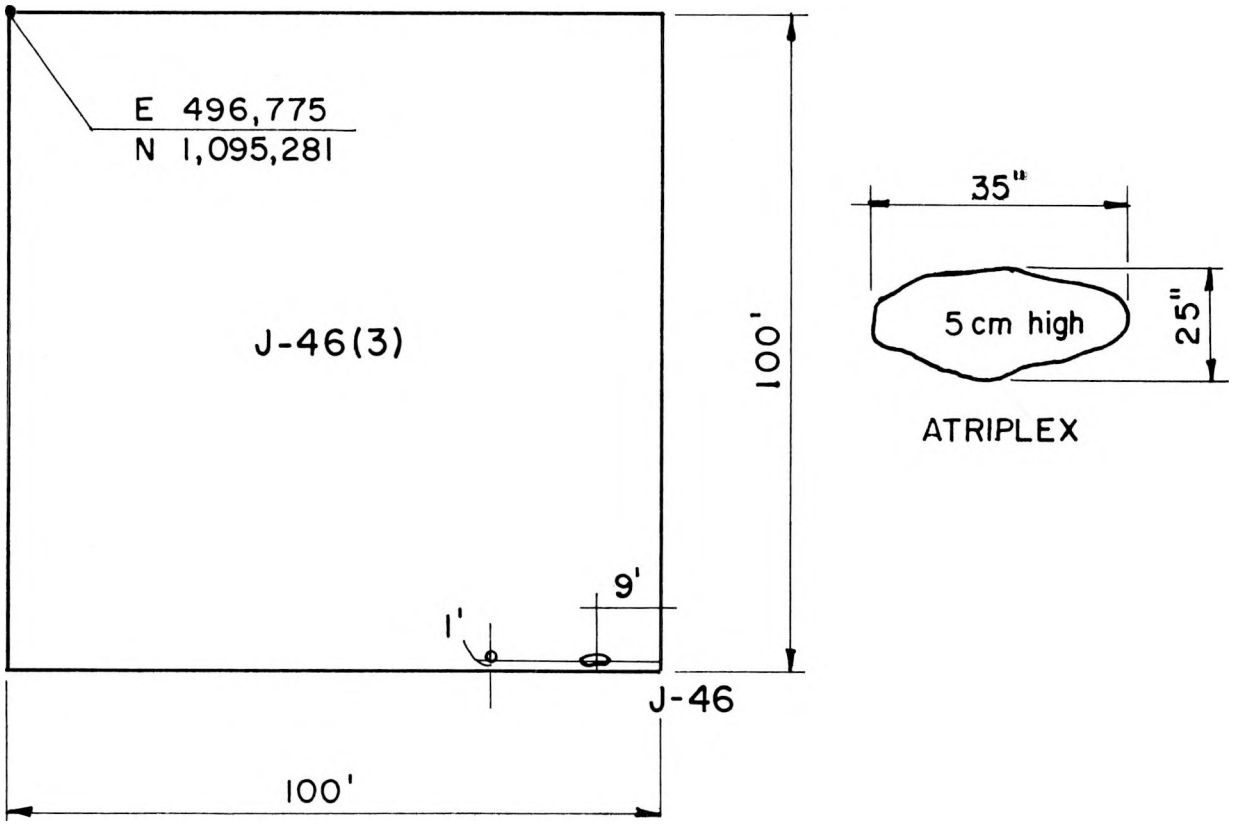
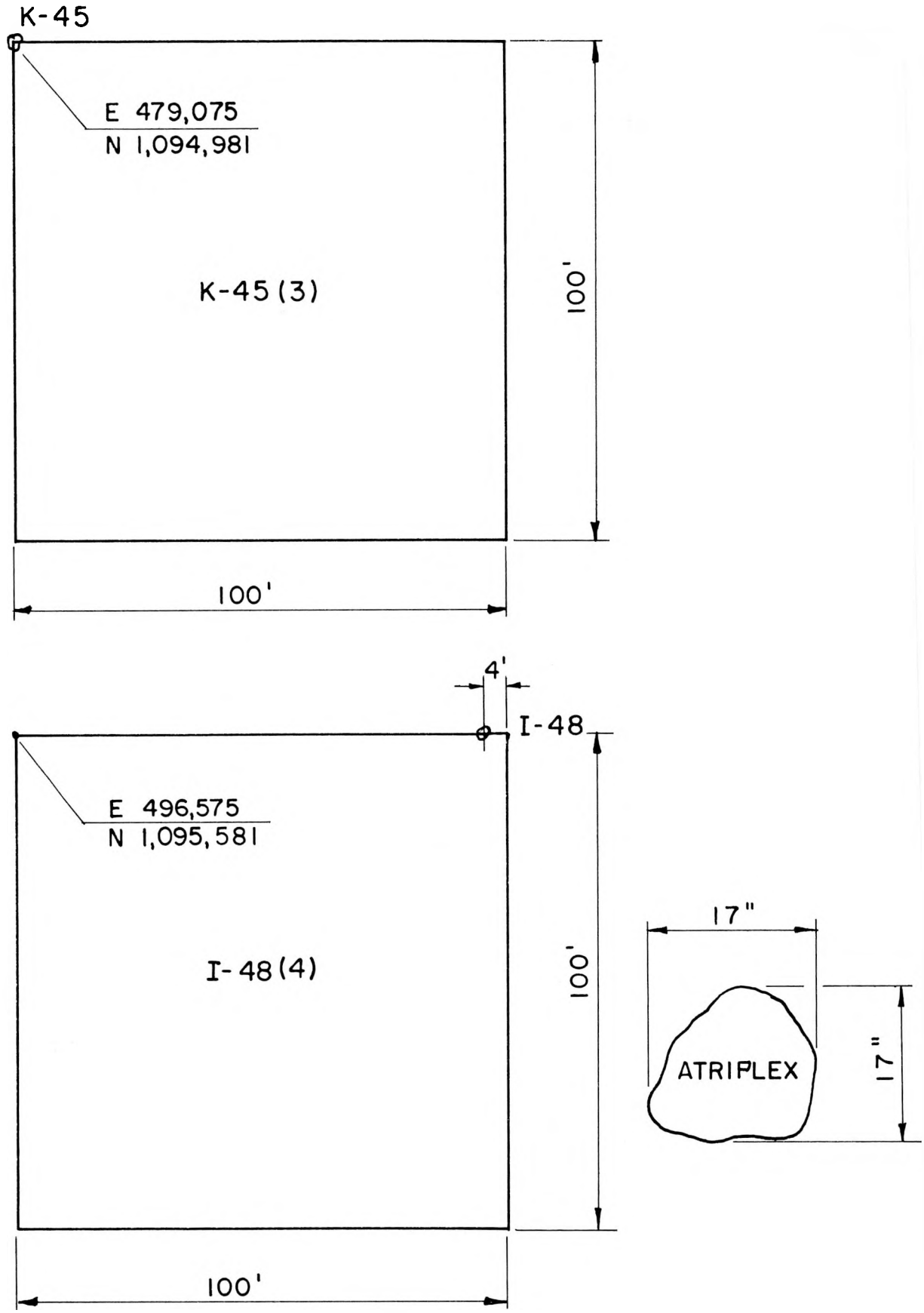
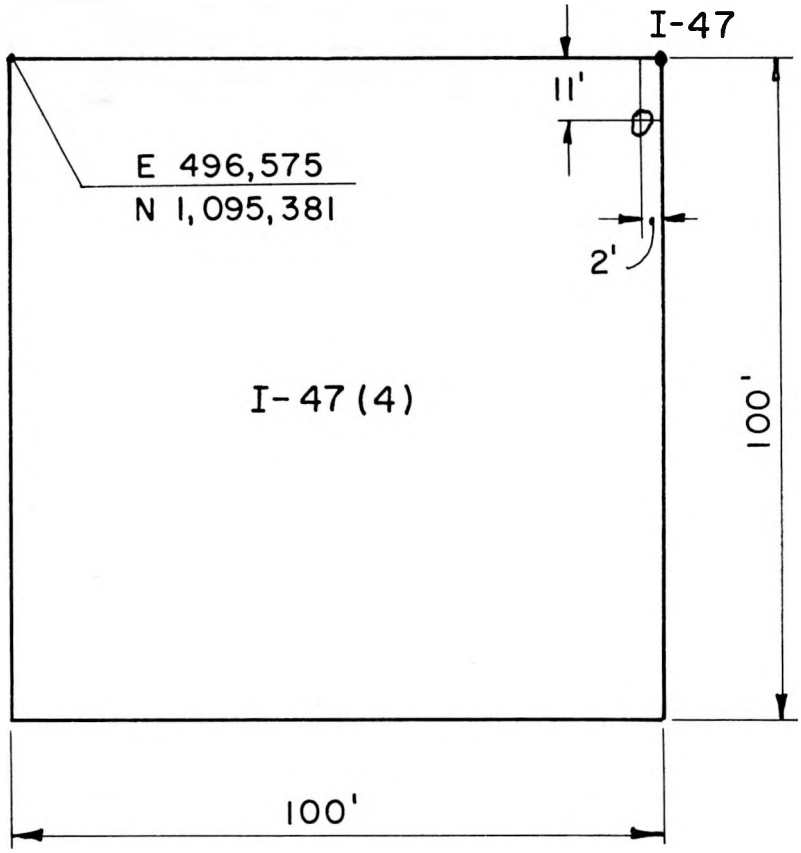
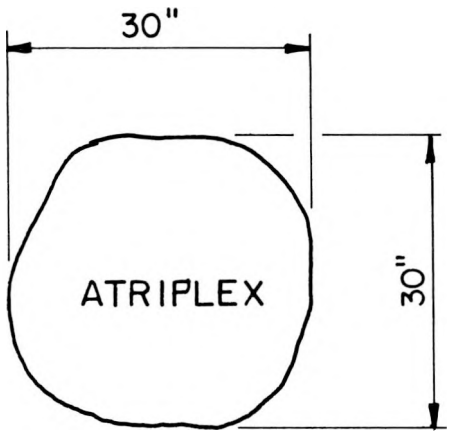
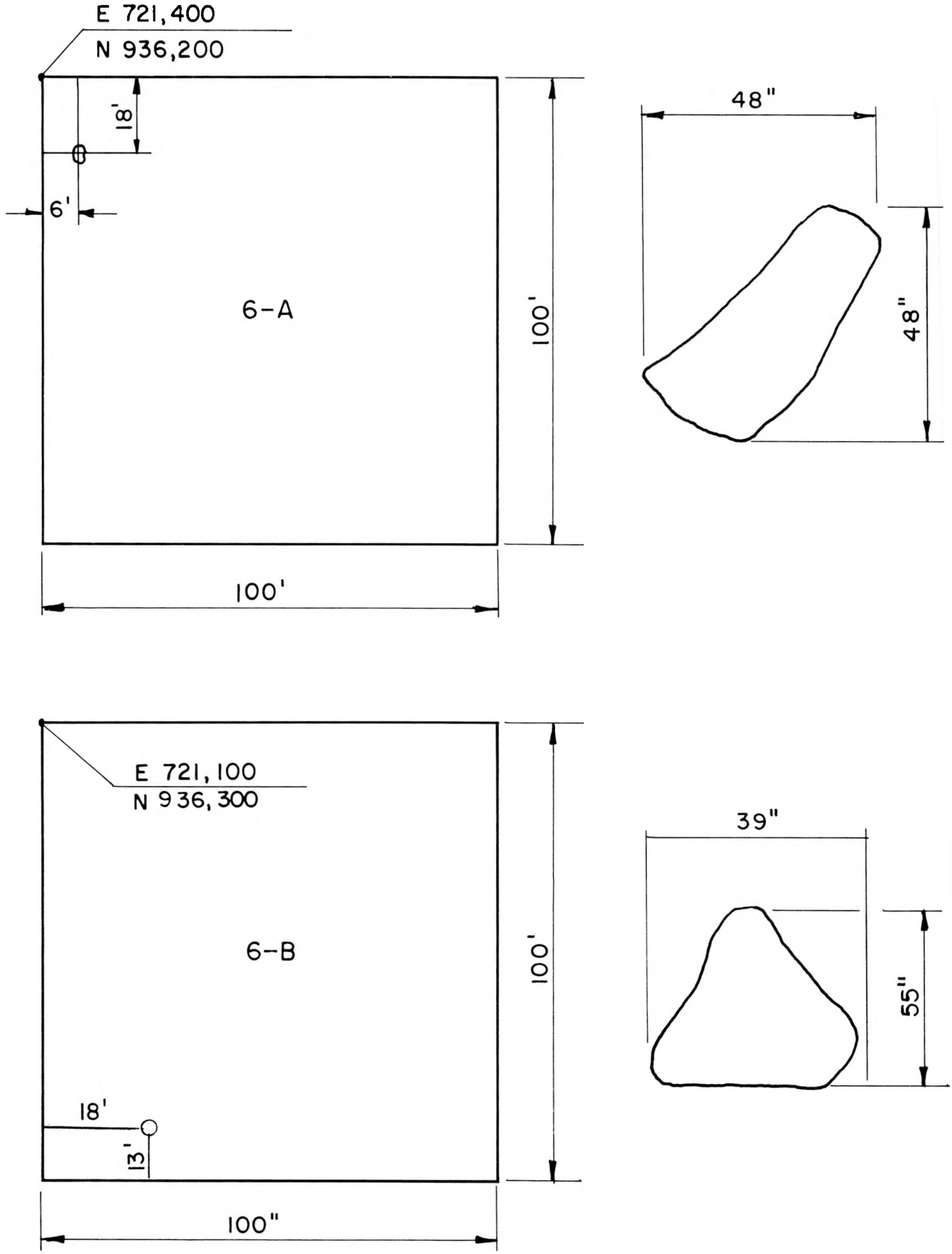
E 496,275
N 1,096,181
G-51(2)
100'
100'
27"
39"
18"
86"


E 496,775
N 1,095,281
J-46(3)
100'
9'
1'
J-46
100'
35"
5 cm high
25"
ATRIPLEX


K-45
E 479,075
N 1,094,981
K-45 (3)
100'
100'
4'
I-48
E 496,575
N 1,095,581
I-48 (4)
100'
100'
17"
ATRIPLEX
17"


I-47
11'
E 496,575
N 1,095,381
2'
I-47(4)
100'
100'


30"
ATRIPLEX
30"



AREA 13 PLOT DESCRIPTION

UNIVERSITY OF CALIFORNIA
LOS ALAMOS SCIENTIFIC LABORATORY

October 23, 1975

Dr. Paul B. Dunaway
Environmental Effects Division
ERDA/NVOO
P.O. Box 14100
Las Vegas, NV 89114

Subject: LABORATORY SAMPLE PREPARATION PROTOCOL FOR NAEG MOUND SAMPLES*

Attn: M. G. White

Dear Paul:

Attached is a copy of the section of the mound study protocol dealing with preparation and analysis of the mound samples. This protocol is to be followed for both Clean Slate 3 and Area 13 samples. Dr. Gilbert and I have consulted in the generation of this protocol.

In the next few days the complete protocol package will be sent to you for your review. If there are any questions, please call.

Sincerely yours;

E. H. Essington
Soil Scientist
Environmental Studies
Group H-8

EHE/mlk

Attach. (a/s)

xc: M. G. White, ERDA/NVOO
D. Wireman, REECo

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*See Editor's Note, page 33.

Sample Preparation

1. Samples in 1-gal cans are to be oven dried and weighed to obtain "oven dry weight" of total sample. Samples in multiple 1-gal paint cans are to be treated in a similar manner but oven dry weight of total sample must be calculated from pooled data. In the case of subsampling in the field oven dry weight of the total mound before subsampling must be calculated from subsample oven dry weight, field wet weight, and proportion of total sample collected.

   a. Samples less than ∿200 g--transfer total sample to 1 qt can and ball-mill according to established NAEG sample preparation procedures.

   b. Samples between ∿200 and ∿800 g--replace in original 1 gal sample can and ball mill as per established NAEG procedures.

   c. Samples greater than ∿800 g--transfer to 3 gal or 5 gal can or appropriate inexpensive container and ball mill.

2. Should ball milling the large samples present problems the following test should be made. Immediate review of the results by Dr. Gilbert and E. Essington will be accomplished and a decision made as to the acceptability of the alternative method of sample preparation by step 3 below.[1]

   a. Locate a mound sample contained in two or more 1 gal cans so that the material in one can (A) has substantially higher ^{241}Am activity than the other (B). Mix the contents of the can thoroughly by rotating in all directions.

   b. Transfer up to one-half (or 300-400 g) of the material of can A to a clean 1 gal can (labeled can #1) and the other half (an exactly equal amount) to a second 1 gal can (labeled can #2).

   c. Transfer up to one-half (or 300-400 g) of the material in can B to can #1 and an exactly equal amount to a third 1 gal can (labeled can #3).

   d. Ball mill all three containers according to established NAEG procedures.

   e. From can #1 weigh out 25-10 g samples for Ge(Li) counting of each at the 60 keV ^{241}Am. Counting time should be adjusted to give 10% or less error at 0.1 level of confidence.

---

[1] From this point in this protocol forward, the procedures were changed as indicated in Essington's "NAEG Soil Mound Study No. 2 (Revised)," November 4, 1975, page 7, Item 2a forward.

f. Combine the contents (including balls) of cans #2 and 3 in a plastic bag capable of withstanding prolonged kneading.

g. Thoroughly mix the sample in the plastic bag by kneading for ∿10 minutes (observe progress of mixing whether or not presence of steel balls is advantageous and whether or not plastic bag holds up).

h. Weigh out 25-10 g samples from the plastic bag and count as in e. above. Immediately notify Dr. Gilbert and E. Essington of the results of this test.

i. Upon completion of this test, return all materials to the original containers (A and B) and set aside for further processing.

3. Divide total sample into a series of 1 gal cans (∿800 g each) and ball mill. Transfer contents of all cans to plastic bag and mix by kneading for ∿10 minutes. Store sample in as few 1 gal cans as possible.

Analysis of Samples

Vegetation samples will be held until further notice. Prepare a list of all vegetation samples including coordinate location of mound. Indicate type of vegetation sample available, tops, roots, or stem and provide copies of the listing for Dr. Romney, Dr. Gilbert, and Dr. Fowler.

All soil samples are to be analyzed for Pu and Am if funding is available, otherwise only one of the two replicate mounds will be committed to analyses at this time.

Each laboratory is to receive the usual aliquot of sample, unsieved. Analyses to be requested include:

1. Wet chemical $^{239-240}$Pu on all samples.

2. Ge(Li) analyses of all samples for ^{241}Am on same aliquot as wet chem Pu.

3. Those samples containing activity less than twice the detection limit for the Ge(Li) system in use are to be analyzed for ^{241}Am by wet chem (same aliquot as for Pu).

4. Randomly select 5% of those samples above twice the detection limit and analyze for ^{241}Am by wet chem (same aliquot as for Pu).

NAEG SOIL MOUND STUDY NO. 2 (REVISED)*

November 4, 1975

E. H. Essington, D. L. Wireman,
R. O. Gilbert, and E. B. Fowler

Mission

The NAEG Soil Mound Study was initiated with an NAEG Mound Study Pilot Program conducted in September and October, 1974, by REECo personnel to determine the advisability of a more extensive mound sampling program. The pilot study consisted of the collection of 10 soil profiles in selected mounds and 10 profiles in desert pavement adjacent to each sampled mound in Area 11-C. Only ^{241}Am was measured using a Ge(Li) detector. The data reflected the nonuniformity of ^{241}Am distribution within the mound from which it is assumed that plutonium distribution would be similarly nonuniform.

A second NAEG Soil Mound Study was initiated in August, 1975, to provide information relative to the following mission:

1. Determine contribution of plutonium in various types of mounds to total plutonium inventory of area down to the 5 cm datum.

2. Determine percent of area covered by mounds of various types as opposed to desert pavement.

3. Determine Pu/Am for selected samples.

Attachments

1. Field data forms and parameter descriptions. Letter to Fowler from White 9/25/75, BSD:DLW:237.

2. Stereo Aerial Photography.

3. Mound types--sketches.

4. Plot descriptions--sketches and coordinates.

Mound Types Identified

Five types of mounds were identified which can be unique contributors to total inventory.

See Editor's Note, page 33.

1. Grass Clump--small lone mound developing under a clump of grass, such as Indian ricegrass. Mound may be 10-15 cm diameter and 1-2 cm high.

2. Shrub--a small lone mound developing under a single shrub, such as Atriplex. Mound may be 25-30 cm diameter and 3-5 cm high.

3. Shrub/Complex--large mound of varying shape 0.5-2 m containing various species of vegetation including grasses and 10-40 cm high.

4. Animal Dig--usually large mounds resulting from activities of either large burrowing animals or large colonies of small burrowing animals. The large colony type mounds will be excluded from considerations in this study since they are few in number. Their combined influence on plutonium inventory is expected to be very small, and they are prime specimens for animal habitat studies.

5. Diffuse Grass--low flat mound of 0.5 m diameter up to 10 m across. Many small, low grass tufts throughout mound area.

Data and Related Information Required

1. Coordinate (Nevada Grid) location of selected mound.

2. Category or type of mound as defined above.

3. Physical measurements--long axis, normal short axis using tape measure, height relative to desert pavement using level or scale.

4. Stereo pair of photos before vegetation harvested to show as much of mound as possible.

5. Stereo pair of photos after vegetation harvest from same aspect as in 4.

6. Identity of vegetation and specifically of shrub from which sample is collected for age determinations.

7. FIDLER reading of desert pavement surface sample.

8. FIDLER reading of mound after removal of vegetation.

9. FIDLER reading of desert pavement plane under mound after mound is removed.

Sampling (Clean Slate 3)

1. Locate 100 x 100 ft plot using NG coordinates given for NW corner. Place marker at NW corner of plot that can be photographed from the air. Stake the plot into 50 x 50 ft squares.

2. Randomly select one of the 4 - 50 x 50 ft quadrants.

3. Divide the chosen quadrant into 5 quadrates using 6 - 50 ft lengths of rope or twine.

4. Starting in the NW corner of the quadrate number 1, number individual mounds categorized according to the list above throughout all 5 quadrates (e.g., GRASS CLUMP mounds numbered from 1 to N_1). Use Form No. 4 to record identification and counting of each mound type.

5. Choose 2 random numbers between 1 and N_1 which correspond to the required mounds to be sampled, and locate mounds using mound numbers assigned in Form No. 4.

6. Repeat step 3 and 4 for second type of mound recognized and for each remaining type.

7. From this point on, work on one mound at a time.

8. Photograph mound in stereo (3 frames) approximately 10 cm apart at lens.

9. Harvest vegetation according to established NAEG procedures. (In shrubbed mounds, collect a section of the shrub stem for age dating. This may have to be done while collecting mound material as stem is usually buried.)

10. Photograph bare mound in stereo at same aspect as in 7 above (3 frames).

11. Collect 12.5 cm diameter x 5 cm deep surface soil sample plus FIDLER reading (Form 3) from a desert pavement location 10 cm from edge of mound to closest edge of sampler normal and to the left of prevailing winds using established NAEG procedures.

12. Make measurements of mound as shown in Form No. 1. Record FIDLER readings at midpoint of mound (MP) and at the point where the highest reading occurs (H). Note the approximate location of H.

13. Collect mound to the original desert pavement datum established with a level. Smaller mounds are collected in total and placed in sample container. Medium mounds are collected in total but placed in 3 or 4 1 gal cans. Large mounds, which will not fit in 3 or 4 1 gal cans must be weighed, placed in concrete mixer, in total, mixed, subsampled into storage container (e.g., 1 gal paint can) and again weighed. Field wet weight of the total mound and of the subsample are important parameters to be used to determine total oven dry weight of the mound.

14. Clean mixer with 3 loads of sand or uncontaminated soil (if plastic liner is used, replace liner after each sample instead of cleaning with sand).

15. Record FIDLER reading from same vertical location (H) at level of original desert pavement.

16. Install 5 cm wide flexible baffle plate or string outlining mound. Collect material under mound to 5 cm depth within boundary of baffle or marker. Weigh, mix, and subsample as in 13 above for large samples. Small samples retained in total.

17. Repeat sampling procedures steps 8 through 16 for each mound (see note).

18. Return remainder of material in concrete mixer to mound location from which it was collected taking care not to disperse the material over the surrounding area.

NOTE: Diffuse grass mound sampling.

1. Locate randomly a diffuse grass mound within the 50 x 50 ft square plot. Sketch mound on coordinate paper, number squares consecutively within boundary of mound, randomly select 3 squares as sampling points. Record location, photograph for a stereo representation; no vegetation is to be collected unless large amounts of grasses occur at sampling point.

2. Collect desert pavement sample as in 11 above.

3. Collect mound sample also as in 11 above to desert pavement datum as one sample, then collect additional 5 cm depth, using 12.5 cm diameter ring, as sample below mound. Take FIDLER readings comparable to those above.

Sampling (Area 13)

Sample in same manner as Clean Slate 3, identifying only 3 types of mounds:

1. Shrub/Complex

2. Shrub

3. Animal Dig

Sample Preparation

1. Samples in 1-gal cans are to be oven dried and weighed to obtain "oven dry weight" of total sample. Samples in multiple 1-gal paint cans treated in similar manner but oven dry weight of total sample must be calculated from pooled data. In the case of subsampling in the field oven dry weight of the total mound before subsampling must be calculated from subsample oven dry weight, field wet weight, and proportion of total sample collected.

    a. Samples less than ∿200 g--transfer total sample to 1 qt can and ball-mill according to established NAEG sample preparation procedures.

b. Samples between ∿200 and ∿800 g--replace in original 1 gal sample can and ball milled as per established NAEG procedures.

c. Samples greater than ∿800 g--transfer to 3 gal or 5 gal can or appropriate inexpensive container and ball mill.

2. Should ball milling the large samples present problems, the following test should be made. Immediate review of the results by Dr. Gilbert and E. Essington will be accomplished and a decision made as to the acceptability of the alternative method of sample preparation by step 3 below.

a. Collect 800 g ± 50 g (air dry) soil from a mound within isopleth 5 or 6 of Area 13. Collect a similar sample of 800 g ± 50 g (air dry) soil from a similar type of mound from isopleth 1 of Area 13, so that the material in one can (A) has substantially higher ^{241}Am activity than the other (B). Oven dry soil and mix the contents of both cans thoroughly by rotating by hand in all directions.

b. Transfer up to one-half (or 300-400 g) of the material in can A to a clean 1 gal can (labeled can #1) and the other half (an exactly equal amount) to a second 1 gal can (labeled can #2).

c. Transfer up to one-half (or 300-400 g) of the material in can B to can #1 and an exactly equal amount to a third 1 gal can (labeled can #3).

d. Ball mill all 3 containers according to established NAEG procedures.

e. From can #1, weigh out 25-10 g samples for Ge(Li) counting of each at the 60 keV ^{241}Am. Counting time should be adjusted to give 10% or less error at 0.9 level of confidence.

f. Combine the contents (including balls) of cans #2 and #3 in a plastic bag capable of withstanding prolonged kneading.

g. Thoroughly mix the sample in the plastic bag by kneading for ∿10 minutes (observe progress of mixing whether or not presence of steel balls is advantageous and whether or not plastic bag holds up).

h. Weigh out 25-10 g samples from the plastic bag and count as in e. above. Immediately notify Dr. Gilbert and E. Essington of the results of this test.

i. Upon completion of this test, all materials may be discarded according to established radiological safety procedures.

3. Divide total sample into a series of 1 gal cans (∿800 g each) and ball mill. Transfer contents of all cans to plastic bag and mix by kneading for ∿10 minutes. Store sample in as few 1 gal cans as possible.

Analysis of Samples

Vegetation samples will be held until further notice. Prepare a list of all vegetation samples including coordinate location of mound. Indicate type of vegetation sample available, tops, roots, or stem and provide copies of the listing for Dr. Romney, Dr. Gilbert, and Dr. Fowler, through NAEG management.

1. All soil samples are to be analyzed for Pu and Am if funding is available, otherwise only one of the two replicate mounds will be committed to analyses at this time.

2. Each laboratory is to receive the usual aliquot of sample, unsieved. Analyses to be requested include:

   a. Wet chemical $^{239-240}$Pu on all samples.

   b. Ge(Li) analyses of all samples for ^{241}Am on same aliquot as wet chem Pu.

   c. Those samples containing activity less than twice the detection limit for the Ge(Li) system in use are to be analyzed for ^{241}Am by wet chem (same aliquot as for Pu).

   d. Randomly select 5% of those samples above twice the detection limit and analyze for ^{241}Am by wet chem (same aliquot as for Pu).

ATTACHMENT 2

Stereo Aerial Photography

The percentage cover that each mound type represents of the total intensive study site is a factor that must be resolved before further inventory studies are attempted. During a low-level flyover of areas at NTS and TTR at 100-1,000 ft above ground, observations were made which suggest that stereo color or infrared photographs at about 100-200 ft. may be useable to determine the percent mound cover. The alternative would be to transect (on foot) the plot by many parallel lines and measure the percentage cover. Aerial photos may offer a much cheaper and direct means when considering the number of sites which would be so treated.

Several methods were suggested:

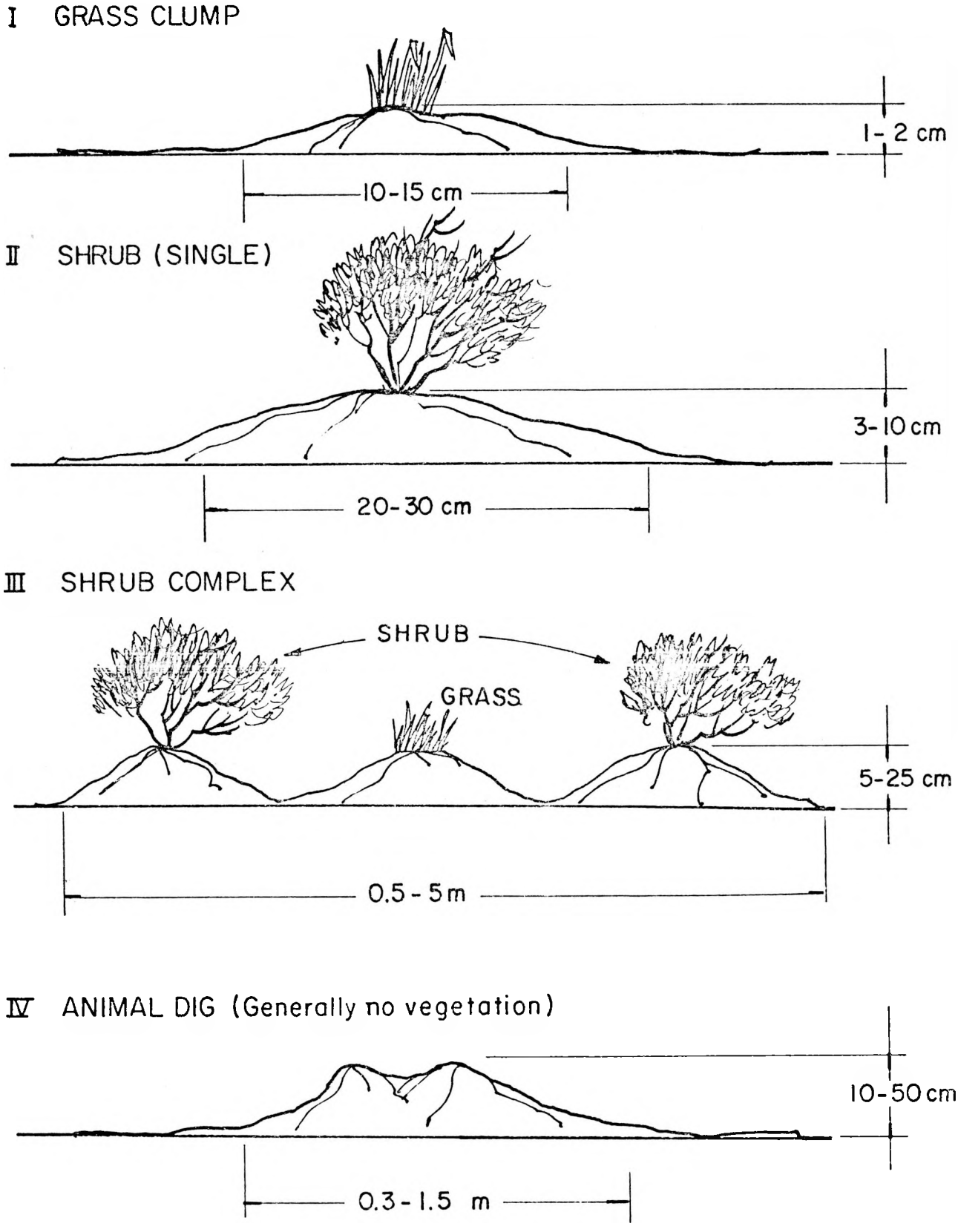
1. Contractor flyover using professional aerial photographic equipment. (Limitation--cannot fly low enough to produce desired resolutions and access.)

2. Helicopter plus hand-held camera. (Limitation--access.)

3. Tethered weather balloon. Best method but requires ground crew and some camera modifications.

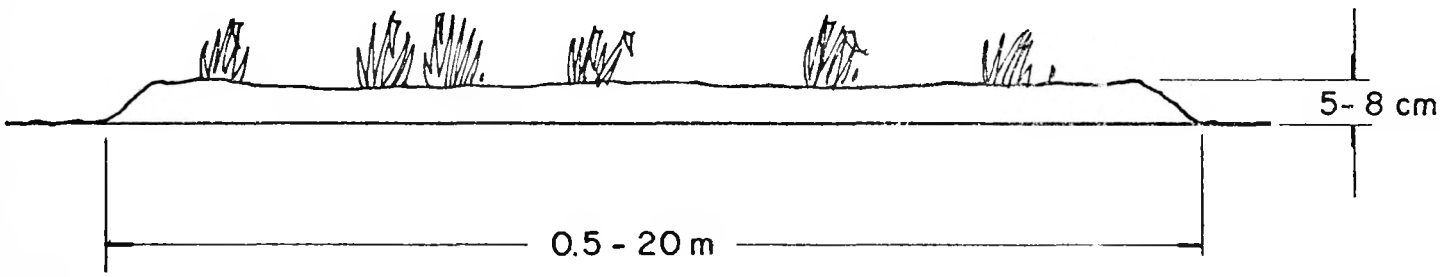
The various possibilities of contractor service are being pursued by NAEG (NVOO).

The photographs should be overlapped so that there are three along the N-S line and three along the E-W line for a total of 6 frames. Corner markers calibrated as to distance, size, and differential elevation should be installed prior to aerial photography.

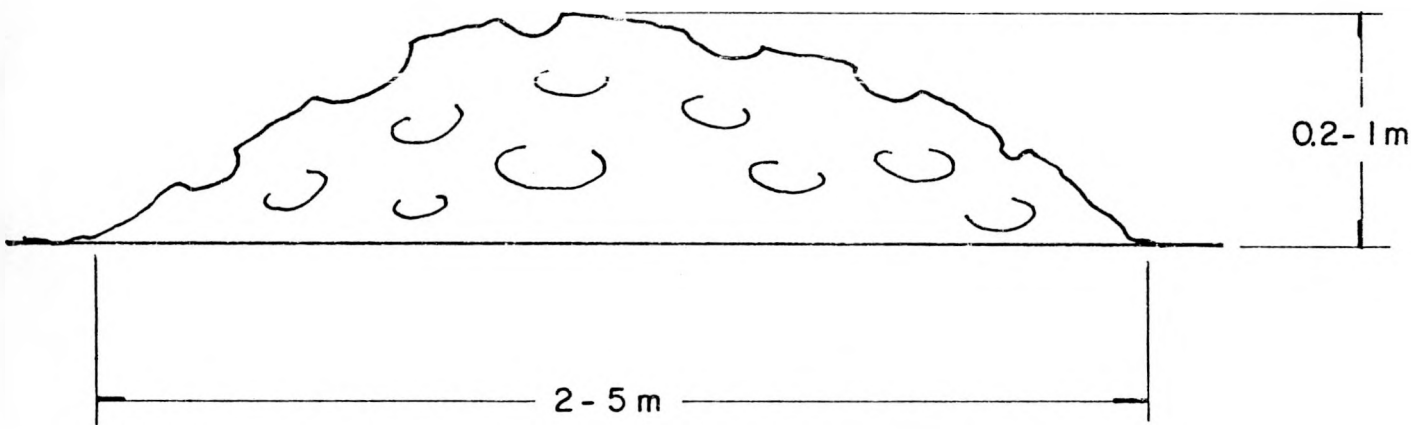

I GRASS CLUMP
1-2 cm
10-15 cm
II SHRUB (SINGLE)
3-10 cm
20-30 cm
III SHRUB COMPLEX
SHRUB
GRASS
5-25 cm
0.5-5m
IV ANIMAL DIG (Generally no vegetation)
10-50 cm
0.3-1.5 m

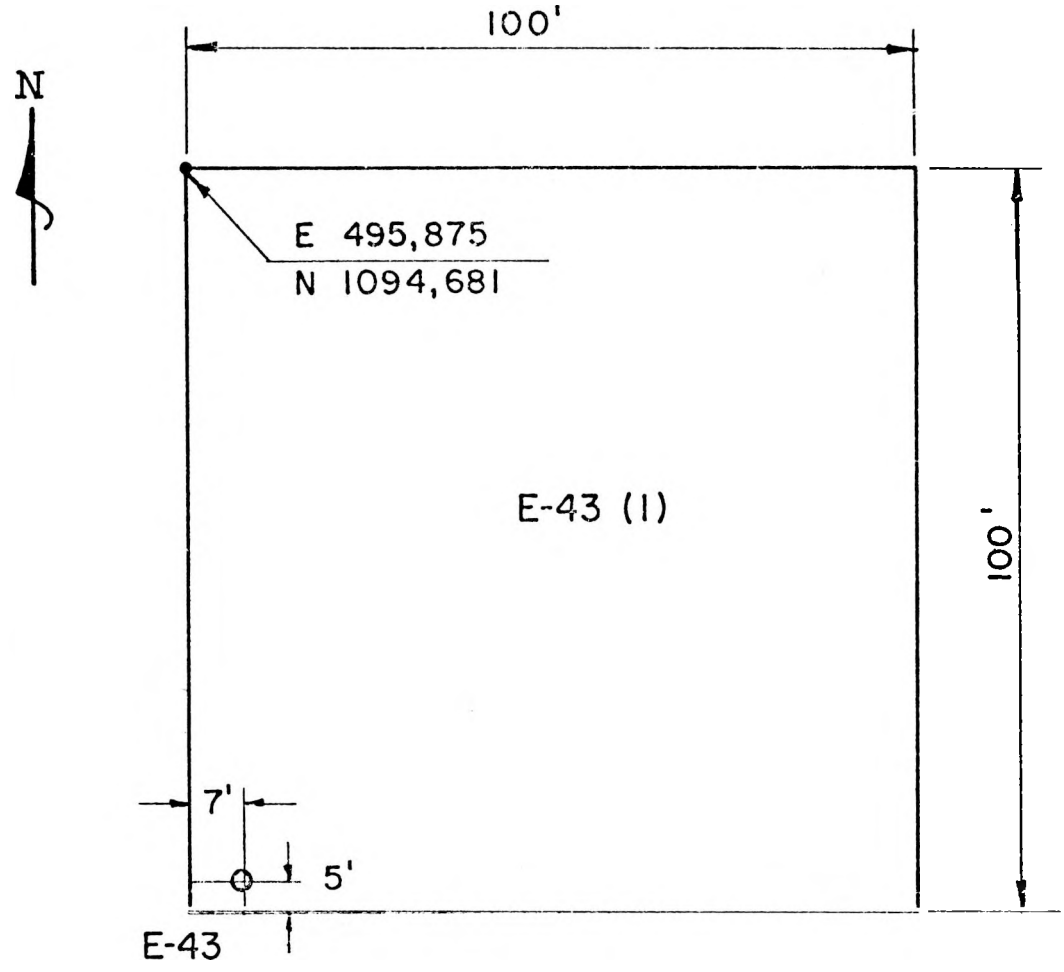
MOUND TYPES

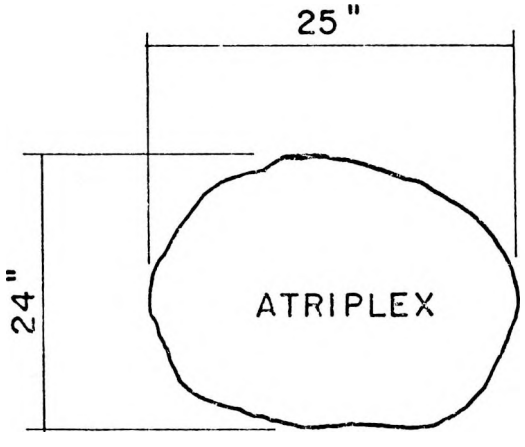
DIFFUSE GRASS



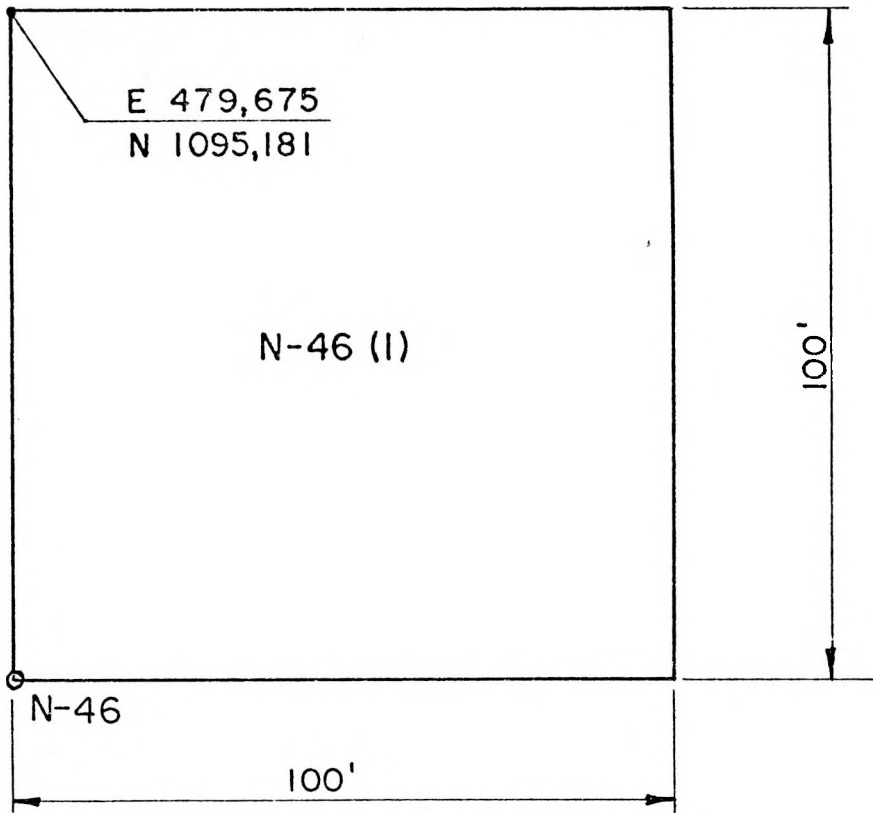
VI LARGE ANIMAL DIG (Generally colony of small diggings, no vegetation)

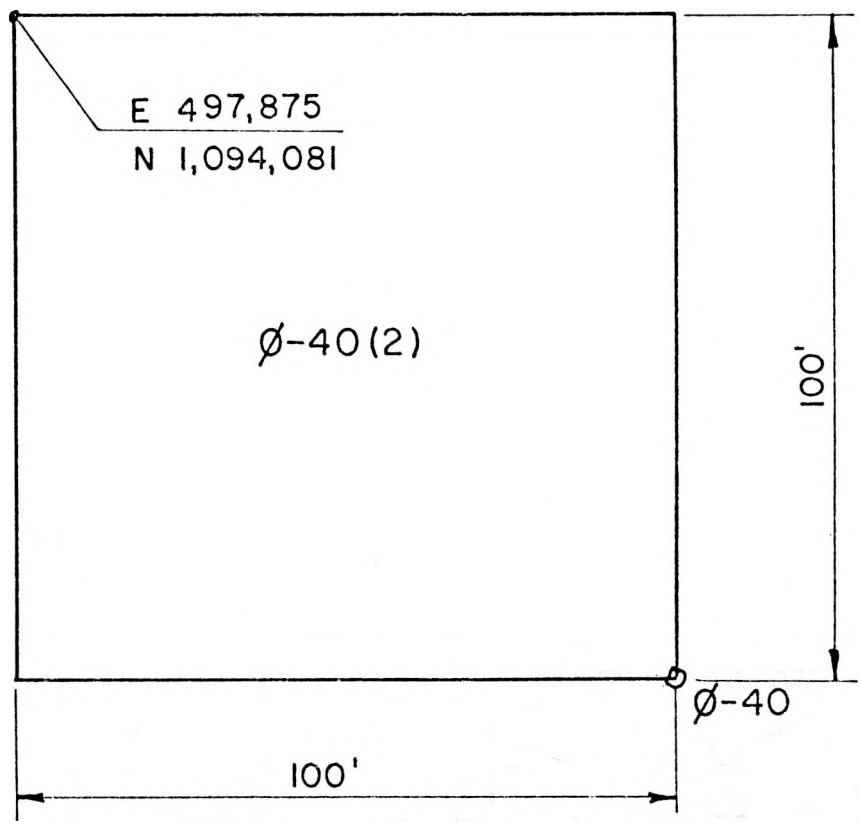


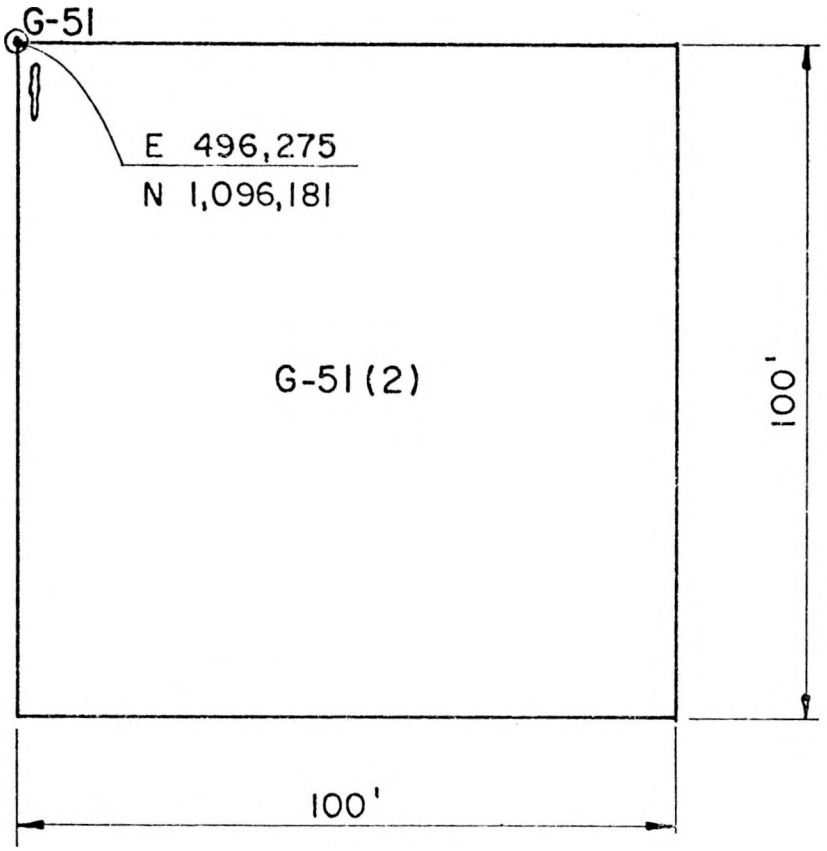


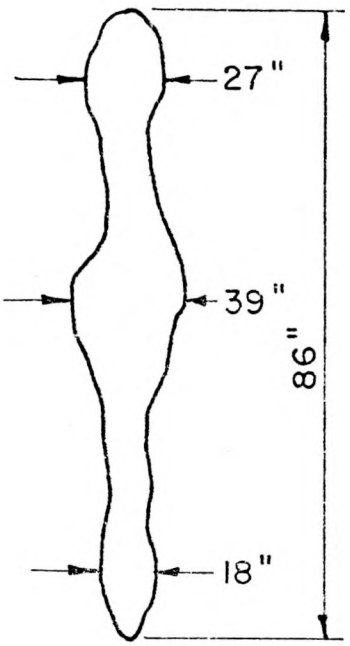


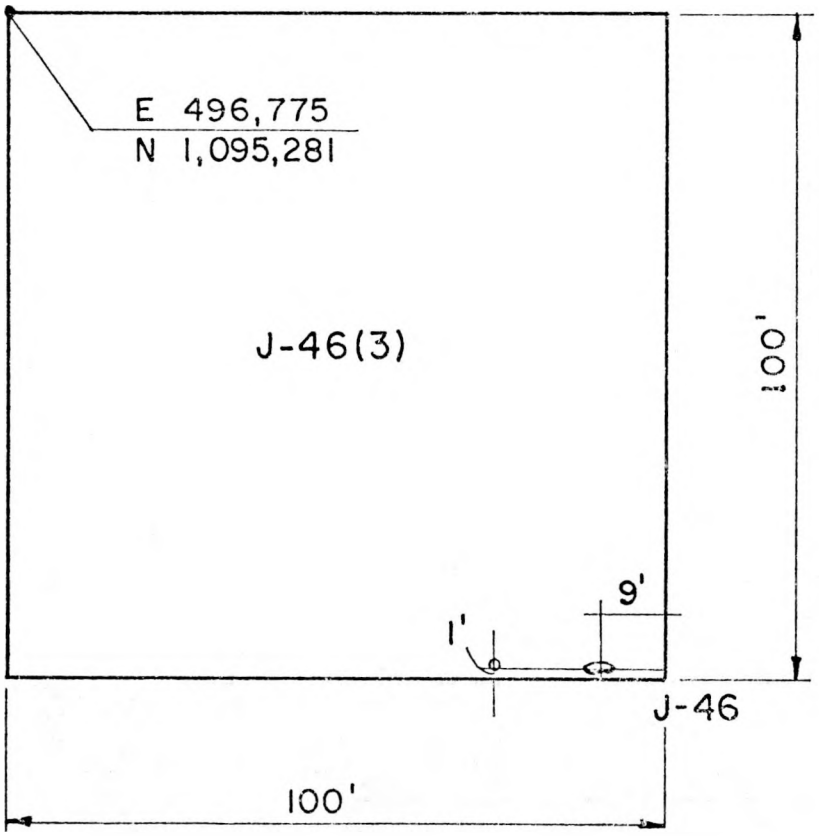
CLEAN SLATE 3 PLOT DESCRIPTION

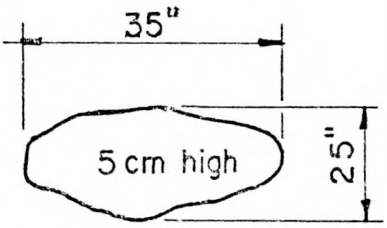

E 479,675
N 1095,181
N-46 (1)
100'
N-46
100'


E 497,875
N 1,094,081
Ø-40(2)
100'
Ø-40
100'

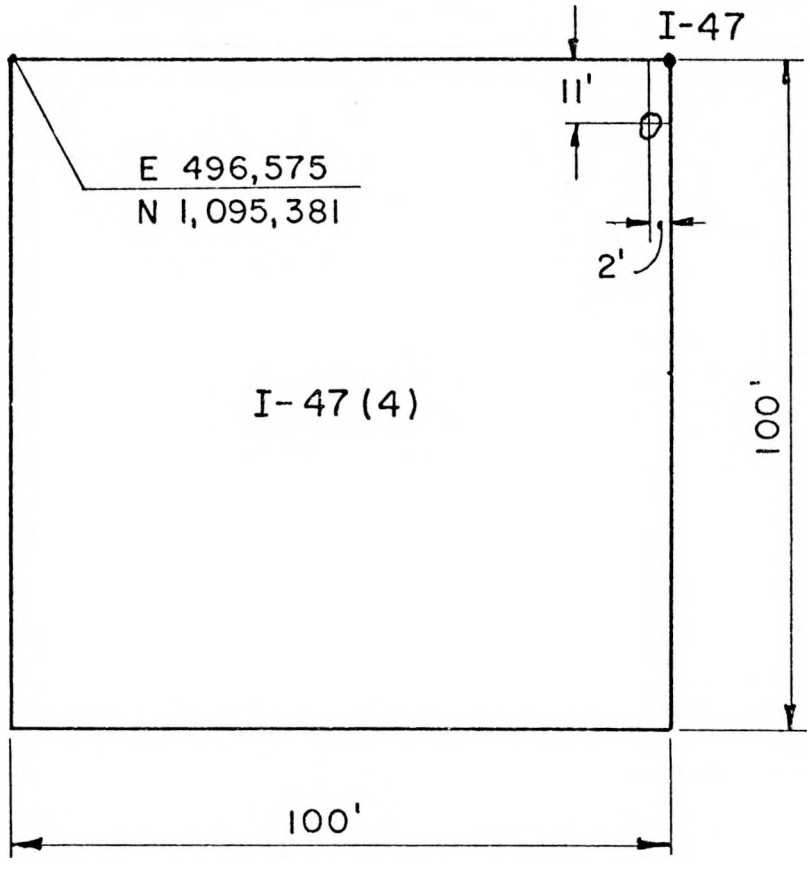

G-51
E 496,275
N 1,096,181
G-51(2)
100'
100'

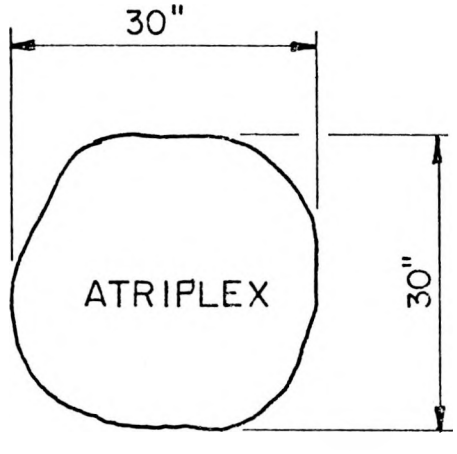

27"
39"
18"
86"

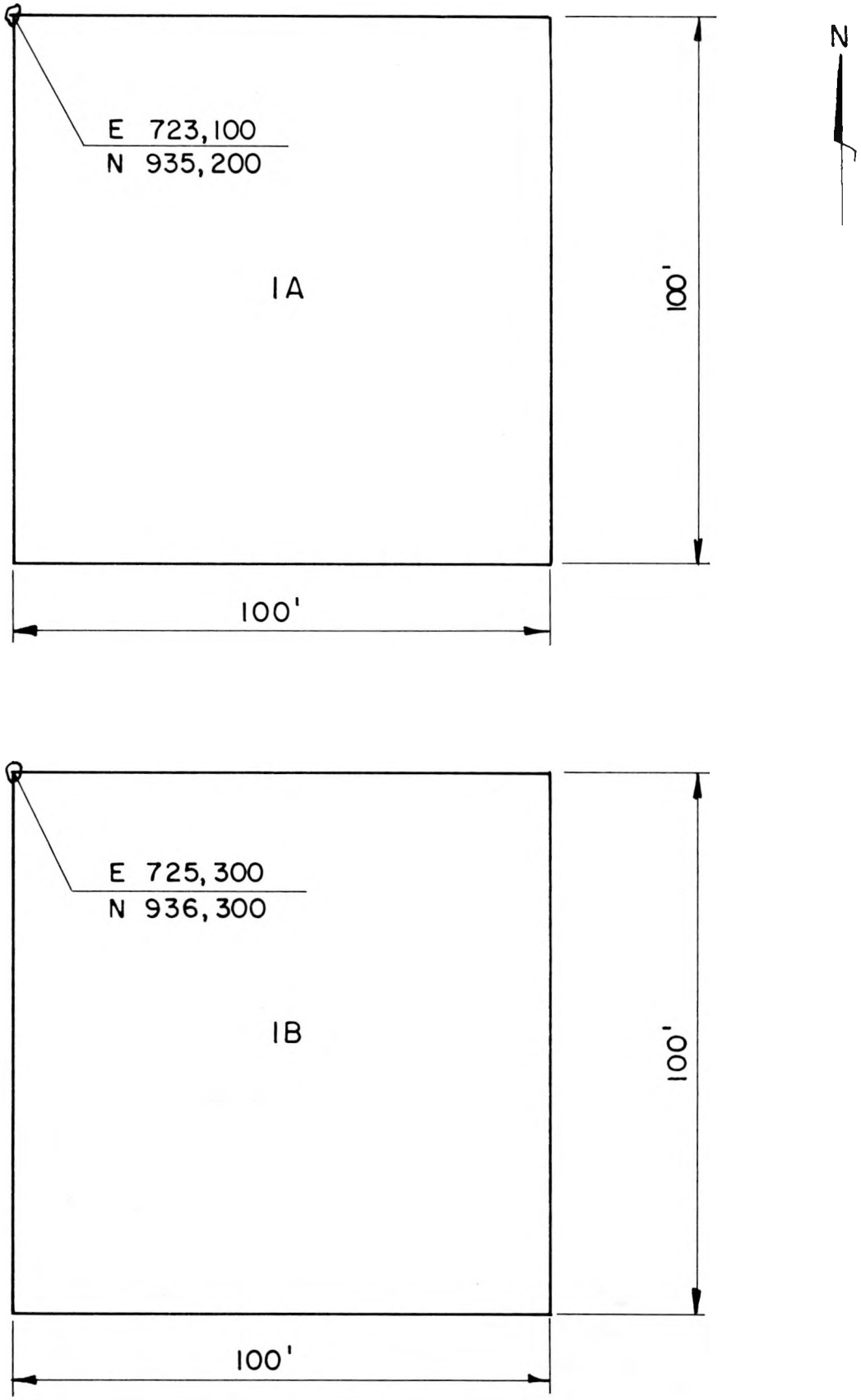

E 496,775
N 1,095,281
J-46(3)
100'
9'
1'
J-46
100'


35"
5 cm high
25"

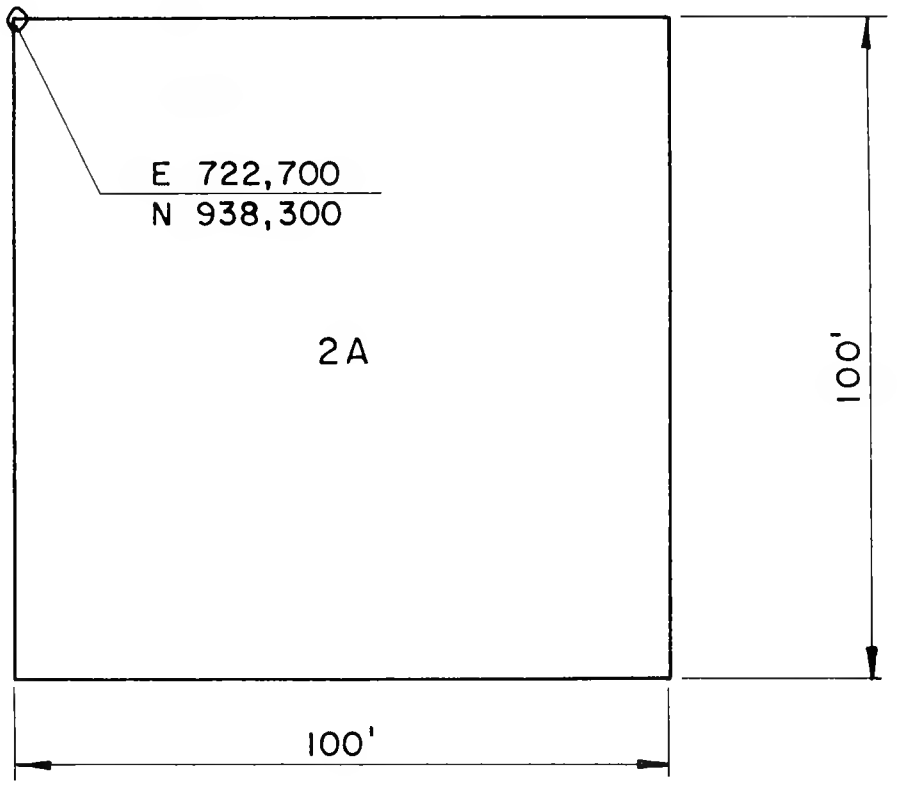
ATRIPLEX

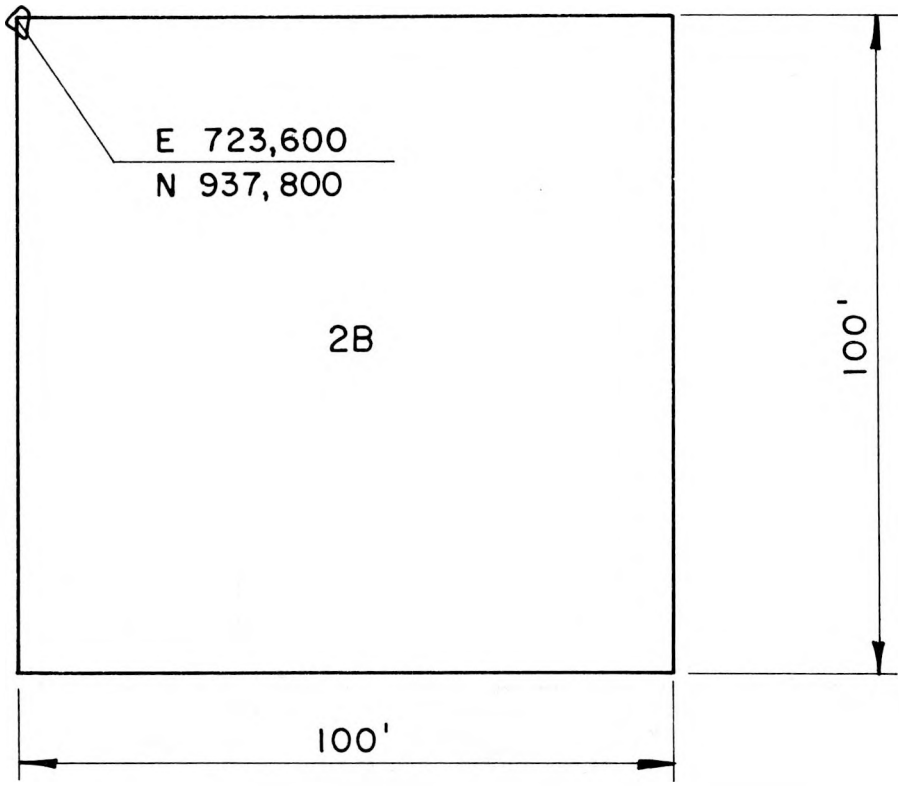

I-47
11'
E 496,575
N 1,095,381
2'
I-47(4)
100'
100'


30"
ATRIPLEX
30"

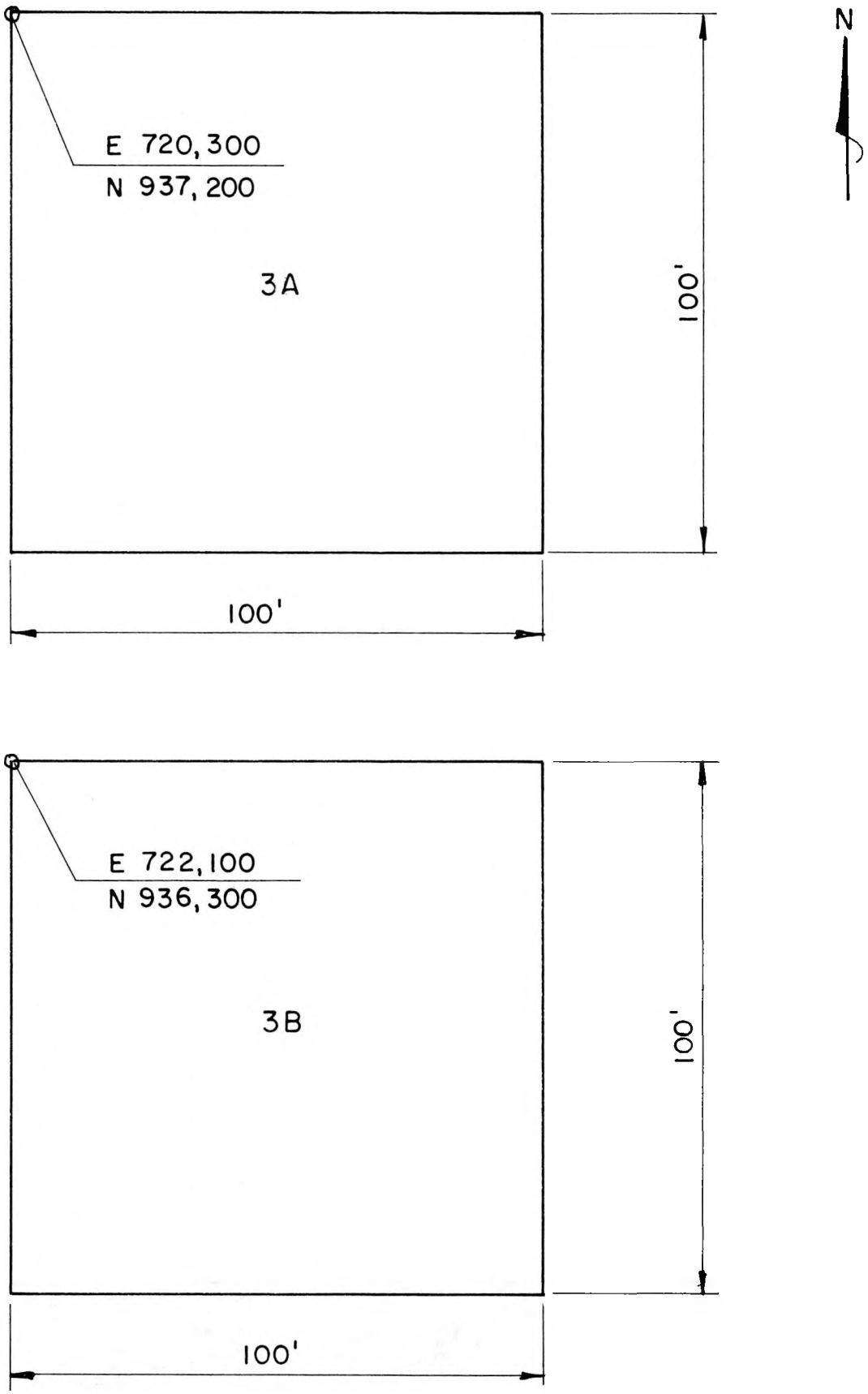


AREA 13

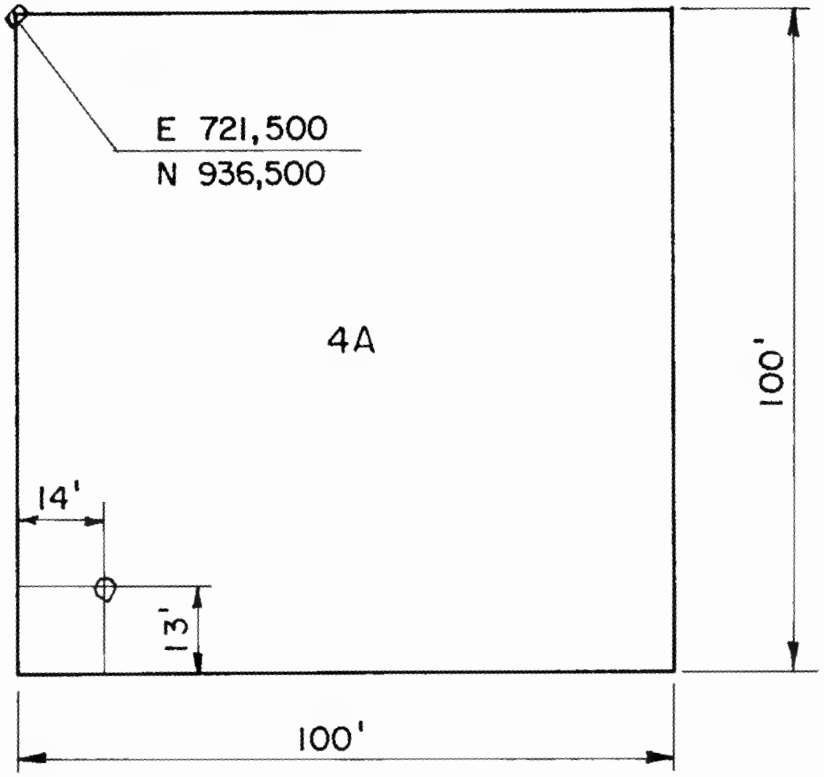


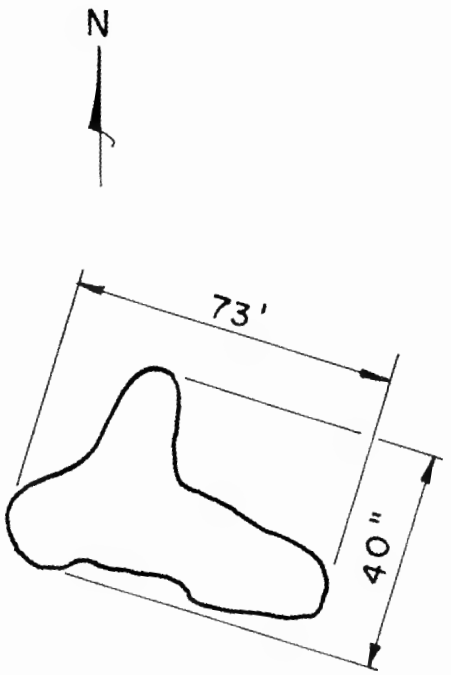


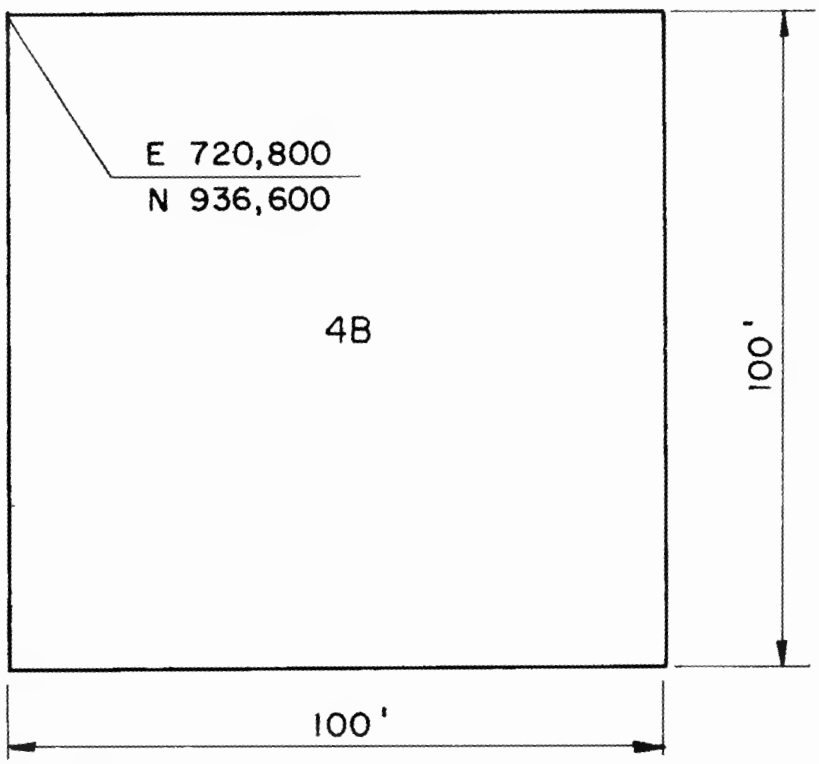
AREA 13



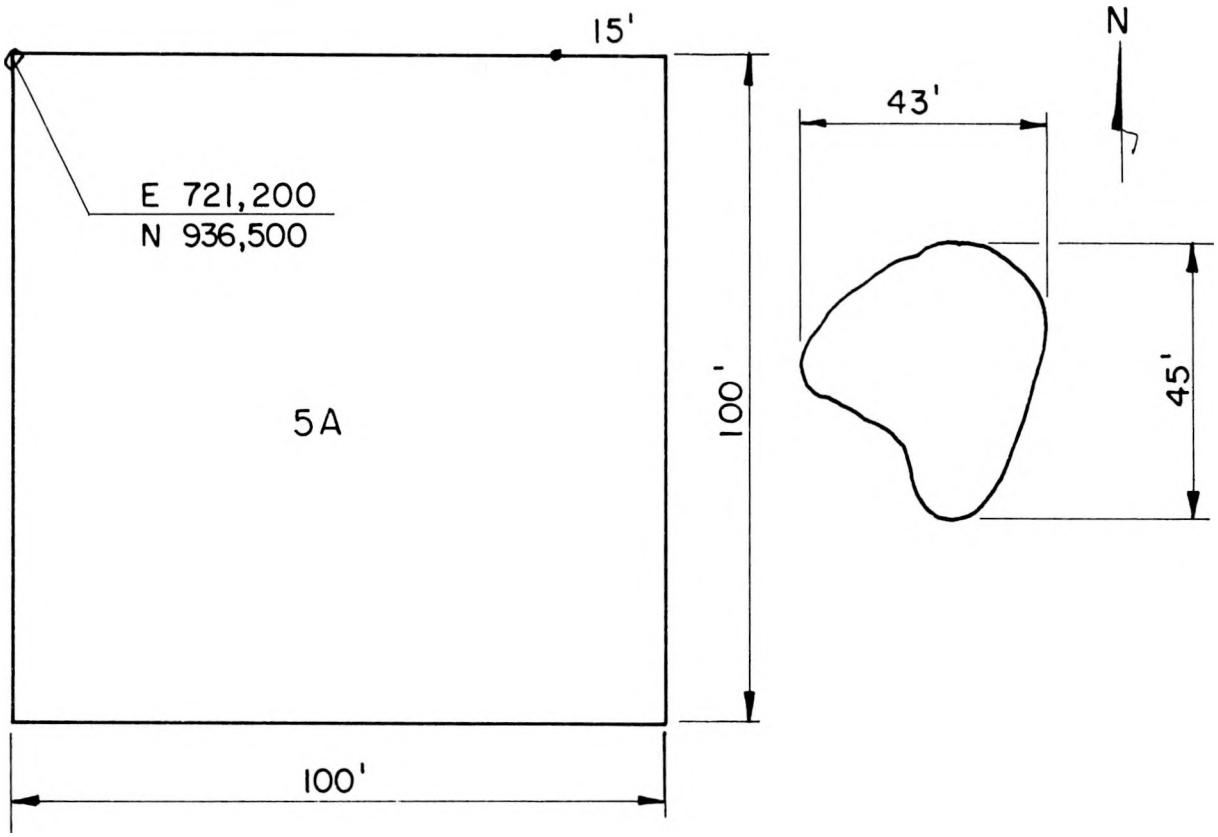
AREA 13

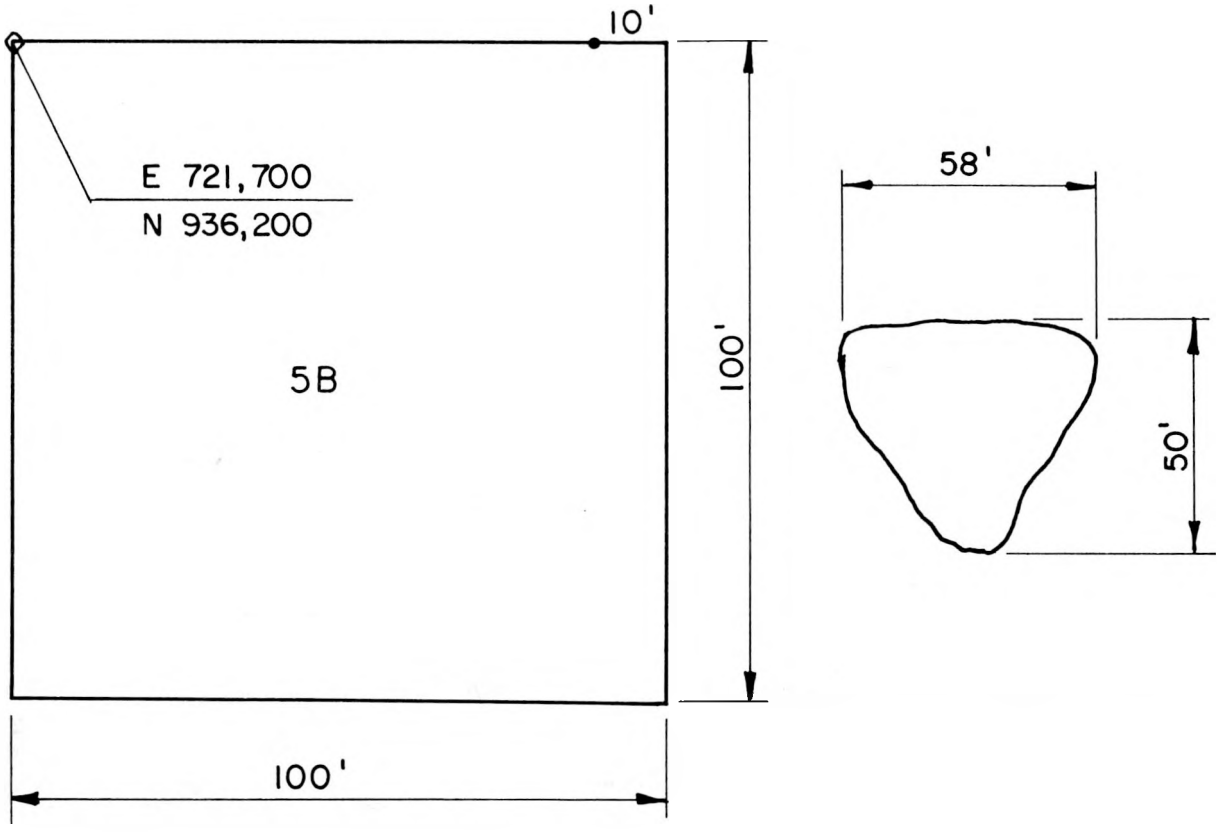

E 721,500
N 936,500
4A
100'
14'
13'
100'


N
73'
40"

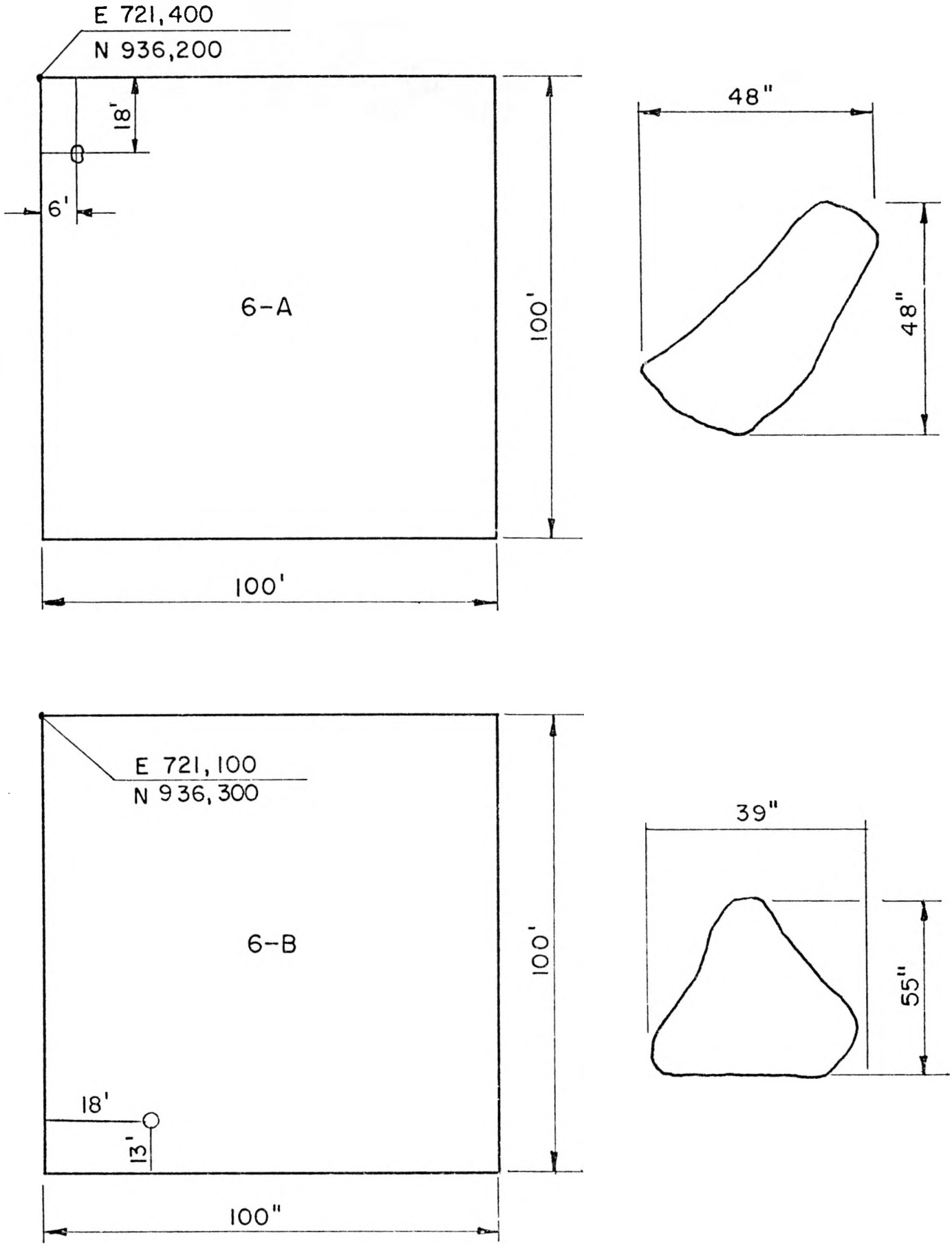

E 720,800
N 936,600
4B
100'
100'

AREA 13


15'
E 721,200
N 936,500
5A
100'
100'
N
43'
45'


10'
E 721,700
N 936,200
5B
100'
100'
58'
50'

AREA 13



AREA 13 PLOT DESCRIPTION

NAEG SOIL MOUND STUDY NO. 2
DATA CATEGORY SYMBOLS
AND DEFINITIONS

Mound Parameters

1. Date (MDY)--Month, day, and year mound parameters were recorded.

2. Area--Test Site numerical area designation, e.g., 52, 13, etc.

3. Event--Event designation within area, e.g., Clean Slate 3, Project 57, etc.

4. P--Sample plot number, e.g., sample plot number A1 is first plot, (A), selected in isopleth number 1.

5. Plot NW coordinates--North and East Nevada grid coordinates (NGC) of northwest corner of sample plot (100 ft x 100 ft).

6. Q1--Quadrant (50 ft x 50 ft) number:

   a. NW quadrant is number one (1).

   b. NE quadrant is number two (2).

   c. SW quadrant is number three (3).

   d. SE quadrant is number four (4).

7. Q2--Quadrate (10 ft x 50 ft) number: Within each quadrant, quadrates are numbered from North to South 1, 2, 3, 4, and 5.

8. M--Mound number; random number selected, from total number of mounds of one mound type, from one quadrant.

9. MT--Mound type; numerical designation assigned to each mound type. Those assigned are as follows:

   1 = grass clump mound.

   2 = shrub mound.

   3 = shrub/complex mound.

   4 = animal dig mound.

   5 = diffuse grass mound.

10. MP--Location of midpoint of mound; distance in feet South and distance in feet East that MP (intersection of WM and L) is from the northwest corner of the sample plot; also the Nevada grid coordinates of the intersection of WM and L.

11. L--Length of mound (in cm or ft) along longest axis.

12. L1--Length of line (in cm or ft) from HN (or HE if L is oriented exactly East and West) along L, to the first intersection of L and first width line (WM or W1 or W2, etc.).

13. L2--Length of line (in cm or ft) from WM, W1, W2, etc., and L line intersection (depending on how L1 was designated) to next width line intersection (or to HS or HW).

14. L3, L4, . . . Li--continuation as with L2, until HS (or HW) is reached.

    (NOTE: All width lines are taken at a 90-degree angle to L.)

15. WM--Width of mound (in cm or ft), taken at MP.

16. W1--Length of first width line (in cm or ft) from HN (or HE if L is oriented exactly East and West) unless WM is the only width line.

17. W2--Length of second width line from HN (or HE) unless WM is the only width line.

18. W3, W4, . . . Wi--Length (in cm or ft) of third, fourth, etc., width line from HN (or HE), unless WM is the only width line until HS (or HW) is reached.

19. TM--Length (in cm or ft) form MP to HMP.

20. T1--Length (in cm or ft) of longest section of W1 from L to mound edge.

21. T2, T3, . . . Ti--Length (in cm or ft) of longest section of W2, W3, etc., from L to mound edge.

    NOTE: A sketch of each mound, indicating data category symbols, is presented on each mound parameter field data sheet for specific clarification.

22. HN--Height (in cm or ft) of northernmost point of mound base along L.

23. HS--Height (in cm or ft) of southernmost point of mound base along L.

24. HE--Height (in cm or ft) of East point of mound base along L (HE is used in place of HN if L is oriented exactly East and West).

25. HW--Height (in cm or ft) of West point of mound base along L (HW is used in place of HS if L is oriented exactly East and West).

26. HMP--Height (in cm or ft) of mound base at intersection of TM and mound edge (HMP is zero (0) datum point).

27. HM--Height of highest point of mound.

    NOTE: Height measurements are recorded on field data sheets as actual observed measurements; for reporting purposes, however, all heights are relative to HMP (zero datum point).

28. LAZI--Azimuth degrees of L, taken clockwise from true North.

29. Mound Diagram--Sketch of mound on which all data category symbols are indicated for clarification of mound orientation.

30. Photo frames and aspect notation--Photographic film roll numbers and frame numbers used in photographing a mount; aspect--the condition of the mound at time photograph was taken, the direction toward which the camera was facing, the type of photos taken (e.g., single shot or stereo), and the angle along which the camera was sighted with respect to the horizon (i.e., from 0 to 90 degrees).

31. Field notes--Notes taken which reference unusual observations about the mound (i.e., dead vegetation cover, rodent burrows, no living vegetation on mound, large rock (in or on) mound, etc.

32. NGC--Nevada grid coordinates.

    NOTE: Photo and aspect information is noted on reverse of the mound parameter field data sheet to which it pertains.

Sample Parameters

1. Collection data (MDY)--Month, day, and year sample parameters were recorded and sample was collected.

2. Area, event, P, Q1, Q2, M, and T (mound type number)--same as for mound parameters.

3. SPXR--Desert pavement surface soil sample, 12.5 cm diam x 5 cm deep ring.

4. SPXR sample number designation = 30, if only one SPXR is taken relative to a mound; = 31 through 39, if more than one SPXR is taken relative to a mound.

5. SPXR location--Distance North or South (in cm or ft) and distance East or West (in cm or ft) from MP to SPXR sampler center for diffuse grass mounds; and ten (10) cm from HMP, along WM, to nearest edge of sampler, (away from mound), for all other mound types; also, NGC of sampler center location.

6. HPXR--Height (in cm or ft) at SPXR desert pavement surface ring sampler center.

7. SSIR--Surface soil sample, 12.5 cm diameter x depth to desert ring, taken on diffuse grass mound surface.

8. SSIR sample number designation = 10, if only one SSIR is taken on a mound; = 11 through 19, if more than one SSIR is taken on a mound.

9. SSIR location--Distance North or South (in cm or ft) and distance East or West (in cm or ft) from MP to SSIR sampler center; also, NGC of location.

10. HSIR--Height (in cm or ft) at SSIR surface ring sampler center.

11. SPIR--Subsurface soil sample, 12.5 cm diameter x 5 cm deep ring, taken from desert pavement surface (directly below SSIR location) to 5 cm below desert pavement.

12. SPIR sample number designation = 20, if only one SPIR is taken relative to a mound; = 21 through 29, if more than one SPIR is taken relative to a mound.

13. SPIR location--Same as SSIR for same location; also, NGC of location.

14. SMT--Soil sample that is composed of entire mound, taken to desert pavement surface.

15. SMT sample number designation = 40.

16. SMT location--same as MP location.

17. SMT original weight--wet weight of original total sample (in pounds or kilograms).

18. SPIT--Soil sample, area of mound base x 5 cm deep, taken from desert pavement surface (directly below SMT location) to 5 cm below desert pavement.

19. SPIT sample number designation = 50.

20. SPIT location--Same as MP location.

21. SPIT original weight--Wet weight of original total sample (in pounds or kilograms).

22. VEG (60-69)--Total leaves and stems from a single plant associated with the sampling of a mound.

23. VEG (60-69) sample number designation = 60, if only one plant is sampled from a mound (or no plants from the mound, but one from the SPXR); = 61 through 69, if more than one plant is sampled from same mound.

    NOTE: No vegetation location information or vegetation parameters are required for this study.

24. VEG (70-79)--Total trunk section (for plant aging) from same plant, for same mound as VEG (60-69).

25. VEG (70-79) sample number designation = 70, if from same plant as VEG (60-69) sample number 60; = 71, if from same plant as VEG (60-69) sample number 61, etc.

26. VEG (80-89)--Total roots from same plant, for same mound as VEG (70-79).

27. VEG (80-89) sample number designation = 80, if from same plant as VEG (70-79) sample number 70; = 81, if from same plant as VEG (70-79) sample number 71, etc.

28. Observations about sample--Indicated on sample parameter field data sheet, e.g., large rocks in sample, feces in sample, more vegetation in soil sample than usual, etc.

FIDLER Measurements

1. Measurement date (MDY)--Month, day, and year FIDLER measurements were taken with respect to a mound.

2. Instrument number--ERDA (or AEC) number on instrument package.

3. Probe number--ERDA (or AEC) number on FIDLER probe (detector).

   NOTE: All FIDLER readings are taken with probe face one (1) foot above measurement surface and with probe face parallel to measurement surface.

4. Area, event, P, Q1, Q2, M, and (T)--Same as for mound parameters.

5. FBKG (60KEV)--FIDLER background measurement in c/m with 60 KEV energy region window.

6. FBKG (122KEV)--FIDLER background measurement in c/m with 122 KEV energy region window.

7. SAMP H--Sample number designation for a FIDLER measurement taken at highest FIDLER activity point on mound.

8. SAMP MP--Sample number designation for FIDLER measurement taken at MP.

9. F60--FIDLER measurement in c/m with 60 KEV energy region window.

10. F122--FIDLER measurement in c/m with 122 KEV energy region window.

11. FN--FIDLER measurement net c/m at 60 KEV.

NAEG SOIL MOUND STUDY NO. 2
FIELD DATA SHEET
— MOUND PARAMETERS —

DATE (MDY) -___-___-___ REECO/ESD

AREA -_____ EVENT-_____

PLOT NUMBER (P) -_____

PLOT (100x100) NW COORD. (NGC) N _____ E _____

(Q1) QUADRATE NUMBER (50x 50) -_____ (1-4)

(Q2) QUADRATE NUMBER (10x 50) -_____ (1-5)

MOUND NUMBER (M) -_____

MOUND TYPE (MT) (CIRCLE ONE NUMBER):

1 (GRASS CLUMP MOUND): 2 (SHRUB MOUND):

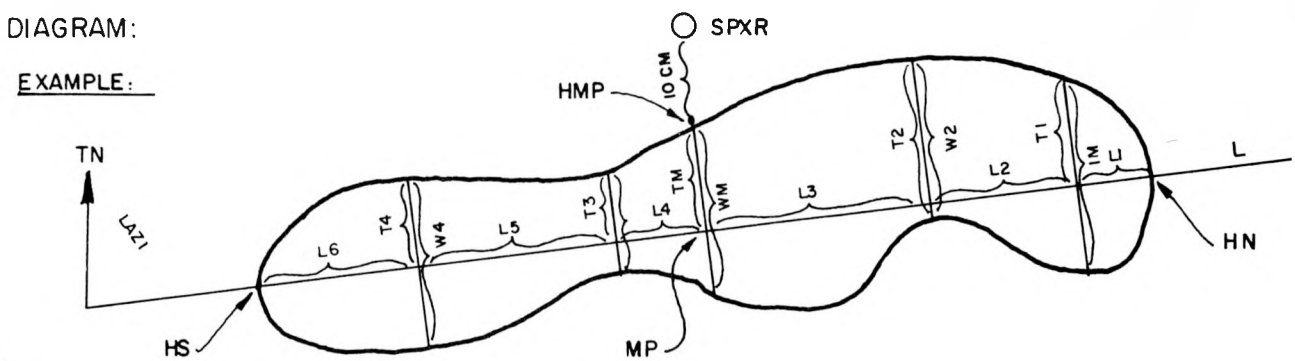
3 (SHRUB/COMPLEX): 4 (ANIMAL DIG): 5 (DIFFUSE GRASS)

MOUND (MP): N S E W _____ FT.; N S E W _____ FT.

(FROM NW PLOT COORD). = (NGC) N _____ E _____

L -- C F	L1 -- C F	L2 -- C F	L3 -- C F	L4 -- C F	L5 -- C F	L6 -- C F	L7 -- C I
WM -- C F	W1 -- C F	W2 -- C F	W3 -- C F	W4 -- C F	W5 -- C F	W6 -- C F	W7 -- C I
TM -- C F	T1 -- C F	T2 -- C F	T3 -- C F	T4 -- C F	T5 -- C F	T6 -- C F	T7 -- C I
HN -- C F	HS -- C F	HE -- C F	HW -- C F	HMP -- C F	HM -- C F	LAZI =_____ DEG	

MOUND DIAGRAM:



C = CM
F = FT

PHOTO FRAMES & ASPECT ON REVERSE: FIELD NOTES (YES) (NO)

FORM NO. 1
(REV. 1)

Mound Counting, Typing, and Random Selection Sheet (Form No. 4)

1. Area, event, P, Q1, Q2, NGC, and mound types are same as for mound parameter field data sheet.

2. Each block indicated is for the designation of the numerical order series of one mound type, e.g., shrub/complex, quadrate 1 block could be 1-13 = 13, indicating a series of mound numbers from 1 through 13, representing 13 mounds of that mound type found in quadrate 1 of an indicated quadrant, plot no., etc., e.g., shrub/complex, quadrate 2 block could be 14-21 = 8, etc. After the entire series for all five quadrates has been established, random number selection is performed to select two (2) numbers from each series, e.g., 2 and 11, which then become the mound numbers for a mound type within the quadrant, plot, etc.

NAEG SOIL MOUND STUDY NO. 2
FIELD DATA SHEET
-SAMPLE PARAMETERS-

COLLECTION DATE (MDY)__-__-__ REECO/ESD

AREA_____ EVENT_____

(P)_____-(Q1)_____-(Q2)_____-(M)_____(T) (_____)

SPXR(30-39): (S)_____: HPXR_____C F: NSEW_____C F; NSEW_____C F (FRM MP)

SPXR(30-39): (S)_____: HPXR_____C F: NSEW_____C F; NSEW_____C F (FRM MP)

SPXR(30-39): (S)_____: HPXR_____C F: NSEW_____C F; NSEW_____C F (FRM MP)

SPXR(30-39): (S)_____: HPXR_____C F: NSEW_____C F; NSEW_____C F (FRM MP)

SSIR(10-19): (S)_____: HSIR_____C F: NSEW_____C F; NSEW_____C F (FRM MP)

SSIR(10-19): (S)_____: HSIR_____C F: NSEW_____C F; NSEW_____C F (FRM MP)

SSIR(10-19): (S)_____: HSIR_____C F: NSEW_____C F; NSEW_____C F (FRM MP)

SSIR(10-19): (S)_____: HSIR_____C F: NSEW_____C F; NSEW_____C F (FRM MP)

SPIR(20-29): (S)_____: SMT(40) (TOTAL MOUND) (YES) (NO) ORIG. WT._____PK

SPIR(20-29): (S)_____: SPIT(50) (TOTAL 5CM BELOW) (YES) (NO) ORIG. WT._____PK

SPIR(20-29): (S)_____: VEG(60-69): (S)_____; (S)_____; (S)____; (S)____; (S)____

SPIR(20-29): (S)_____: VEG(70-79): (S)_____; (S)_____; (S)____; (S)____; (S)____

VEG(80-89): (S)_____; (S)_____; (S)_____; (S)_____; (S)_____

SPXR = DESERT PAVEMENT RING 10 CM FROM MOUND
SSIR = RING SAMPLE ON MOUND SURFACE
SPIR = DESERT PAVEMENT RING BELOW SSIR
SMT = TOTAL MOUND ABOVE DESERT PAVEMENT
SPIT = TOTAL DESERT PAVEMENT 5 CM BELOW MOUND
VEG(60-69) = LEAVES AND STEMS
VEG(70-79) = TRUNK SECTION
VEG(80-89) = ROOTS

OBSERVATIONS ABOUT SAMPLE:

C = CM; (S) = SAMPLE NO.;
F = FT; P = POUNDS;
K = KILOGRAMS

Form No. 2
9/75 (Rev. 2)

NAEG SOIL MOUND STUDY NO. 2
FIELD DATA SHEET
-FIDLER MEASUREMENTS-

MEASUREMENT DATE (MDY) __-__-__ REECO/ESD

INSTRUMENT NO.__________ PROBE NO.__________

AREA__________ EVENT__________

FBKG (60KEV)__________ C/M; FBKG (122KEV)__________C

(P)__________-(Q1)__________-(Q2)__________-(M)__________ (T)(______)

SAMP___H___: F60__________ C/M; F122__________C/M; FN__________C/M

SAMP___MP___: F60__________ C/M; F122__________C/M; FN__________C/M

SAMP__________: F60__________ C/M; F122__________C/M; FN__________C/M

SAMP__________: F60__________ C/M; F122__________C/M; FN__________C/M

SAMP__________: F60__________ C/M; F122__________C/M; FN__________C/M

SAMP__________: F60__________ C/M; F122__________C/M; FN__________C/M

SAMP__________: F60__________ C/M; F122__________C/M; FN__________C/M

SAMP__________: F60__________ C/M; F122__________C/M; FN__________C/M

SAMP__________: F60__________ C/M; F122__________C/M; FN__________C/M

SAMP__________: F60__________ C/M; F122__________C/M; FN__________C/M

SAMP__________: F60__________ C/M; F122__________C/M; FN__________C/M

SAMP__________: F60__________ C/M; F122__________C/M; FN__________C/M

SAMP__________: F60__________ C/M; F122__________C/M; FN__________C/M

REMARKS:

H = Highest activity point on mound
MP = Reading at mound midpoint

Form No. 3
9/75 (Rev. 1)

NAEG SOIL MOUND STUDY NO. 2
FIELD DATA SHEET
— MOUND SELECTION —

DATE (MDY) __-__-__ REECO/ESD

AREA _____ EVENT _____

PLOT NUMBER (P) _____

PLOT (100x100) NW COORD-(NOC) N _____ E _____

(Q1) QUADRANT NUMBER (50 x 50) - _____ (1-4)

QUADRATE (10 x 50)	SHRUB COMPLEX		SHRUB		ANIMAL DIG		GRASS CLUMP		DIFFUSE GRASS	
1		=		=		=		=		=
2		=		=		=		=		=
3		=		=		=		=		=
4		=		=		=		=		=
5		=		=		=		=		=
TOTAL										

Form No. 4

^{241}AM Ge(Li) ANALYSIS OF AREA 13 SOIL MOUND TEST SAMPLES*

January, 1976

(Letter, A. E. Bicker, REECo,
to R. O. Gilbert, BNWL)

Method

Soil was obtained in one-gallon cans collected from Area 13 Complex soil mounds. Can A was collected from the inner isopleth while Can B was collected from the outer isopleth.

The samples were dried in 800-gram aliquots for 24 hours at 105° C. and reconstituted. Cans A and B were thoroughly mixed by rotating in all directions. A total of 400 grams from Can A was transferred to a clean one-gallon can (labeled Can No. 1) and an exactly equal amount (400 grams) was transferred to a second clean one-gallon can (labeled Can No. 2).

A total of 400 grams of the material in Can B was transferred to Can No. 1 and another 400 grams was transferred from Can B to a third clean one-gallon can (labeled Can No. 3).

After aliquoting, Cans 1, 2, and 3 were ball-milled using ten steel balls per can for five hours. After ball-milling, twenty-five ten-gram samples were aliquoted from Can No. 1 for Ge(Li) ^{241}Am counting. (Ball-mill recovery weight from Can No. 1 was 790 grams.)

The contents of Cans No. 2 and 3 were combined (including steel balls) in a plastic bag which was sealed and enclosed in two additional plastic bags (one inside the other) leaving an air pocket in the sample bag to facilitate the mixing process. The plastic bags appeared to be durable enough for the mixing process as there was no leakage from the inner sample bag. Since the sample seemed to be thoroughly mixed during the first five minutes of mixing, the time was cut to five minutes. Apparently the mixing process was aided by leaving the steel balls in the bag, and no problems were encountered when the sample was removed from the bag.

After mixing, the sample from Cans 2 and 3 was removed from the bag, and twenty-five 10-gram aliquots were obtained for Ge(Li) ^{241}Am counting. (The ball-mill recovery weight from Cans 2 and 3 was 780 grams.)

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*See Editor's Note, page 33.

Soil Lab Measurements

Sample I.D.	Wet Wt. (gm)	Dry Wt. (gm)	FIDLER Meas. (c/m)*
Can A	3200	3020	4.32×10^4
Can B	3200	3033	1.85×10^4
Can No. 1	-	800	7.35×10^3
Can No. 2	-	400	6.59×10^3
Can No. 3	-	400	2.62×10^3

*All FIDLER measurements were obtained with can lids in place and the FIDLER probe in contact with the can lid.

UNIVERSITY OF CALIFORNIA
Los Alamos Scientific Laboratory

January 23, 1976

Dr. P. B. Dunaway
Environmental Sciences Div.
ERDA-NVOO
P.O. Box 14100
Las Vegas, NV 89114

SUBJECT: Recommendations for Mound Soil Sample Preparation*

REFERENCES:
1) Letter to P. B. Dunaway from E. H. Essington, Nov. 4, 1975 (H7-75-EHE-419)
2) Letter to E. H. Essington from A. E. Bicker, Jan. 2, 1976 (566-01-184)
3) Letter to E. B. Fowler from A. E. Bicker, Jan. 15, 1976 (566-01-194)

Dear Paul:

Reference 1 was a Mound Study Protocol in which two specific recommendations were made which required some independent sample preparation testing to be conducted. References 2 and 3 are REECo's results of those tests.

With regard to Reference 2, Dr. Fowler, Dr. Gilbert and I consulted upon statistical testing of the data populations and concluded that the alternate method described in Reference 1 "Sample Preparation" item 3 is an acceptable preparation technique for those large samples that cannot be ball-milled by standard NAEG techniques.

This conclusion is based on the fact that the two population variances were the same according to a standard "F" test of population variances.

Also stated in Reference 1 was the suggestion that samples smaller than about 200 g should be ball-milled in a 1 quart container. REECo had not had experience in ball-milling in 1 quart containers and attempted to compare ball-milling efficiencies of the standard 1 gallon can and 1 quart can. Reference 3 presents data concerning various combinations of ball-milling time and numbers of steel balls used. Dr. Fowler and I agree with the observation made in Reference 3 that to maintain sample consistency at the 100 mesh sieve separation comparable to the standard (400 g in 1 gallon can + 10 steel balls for 5 hrs) the combination of 100 g in 1 quart can plus 4 steel balls for 3 hrs is adequate.

Therefore, we recommend adoption of the procedure for kneading the several ball-milled sub-samples to reconstitute a "mixed" sample and the use of option C1 (Ref-3) for ball-milling in 1 quart containers. Adoption of

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*See Editor's Note, page 33.

these procedures is for Mound Study #2 only and should not be considered authorization for use as standard procedures.

If there are any questions, please call.

Sincerely,

E. H. Essington
Soil Scientist

EHE/mlk

xc: M. G. White, ERDA/NVOO
Don Wireman, REECo/NVOO

REYNOLDS ELECTRICAL & ENGINEERING CO., INC.

March 5, 1976

Dr. M. G. White, Assistant
Scientific Manager
Bioenvironmental Sciences Division
Nevada Operations Office
U.S. Energy Research &
Development Administration
Post Office Box 14100
Las Vegas, NV 89114

SOIL MOUND STUDY PROTOCOL*

Dear Dr. White:

The subject NAEG Soil Mound Study Protocol does not provide guidance for preparation of these samples in the Soils Laboratory. Therefore, we have contacted E. H. Essington by telephone on the morning of March 2, 1976, and we have received verbal instructions. Accordingly, the following steps are being incorporated in our soils preparation procedure for the TTR soil mound samples:

1. For the case where the entire mound constitutes a sample of large weight that was homogenized in the field using the cement mixer, the Soils Laboratory will aliquot 800 grams by taking equal amounts from each can delivered to the Soils Laboratory. This 800 gram sample will be dried, ball-milled, and a 50-gram aliquot sent to REECo's Counting Laboratory for 241-Am Ge(Li) scanning prior to shipping.

2. For the case where insufficient soil quantities precluded mixing in the field, all the soil in each can delivered to the Soils Laboratory will be dried, ball-milled in 800 gram quantities, recombined, mixed thoroughly, and a 50-gram aliquot extracted for counting and shipping in accordance with normal procedure.

In the case of aliquots shipped to LFE, 10 grams will be sub-aliquoted from the 50-gram aliquots submitted for shipping activity determination.

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*See Editor's Note, page 33.

We are requesting that all addendum to the subject protocol be forwarded to us as soon as may be convenient to formalize and document these changes in procedure.

Very truly yours,

Arden E. Bicker, Manager
Environmental Sciences Department

AEB:DNB:st

cc: P. B. Dunaway, ERDA/NV, M/S 505

bc: C. E. Rosenberry, M/S 235
E. R. Sorom, M/S 235
A. W. Western, M/S 235
D. L. Wireman, M/S 505

SAMPLE PREPARATION (REVISED 3-4-76)*

(Excerpt from a letter, 3/8/76,
to P. B. Dunaway from E. H. Essington.)

1. All samples in 1-gal cans brought in from the field are to be oven dried and weighed to obtain "oven dry weight" of total sample. Samples in multiple 1-gal paint cans are treated in a similar manner but oven dry weight of total sample must be calculated from pooled data. In the case of subsampling in the field (samples mixed in concrete mixer thence subsampled), oven dry weight of the total mound before subsampling must be calculated from subsample oven dry weight, field wet weight, and proportion of total sample retained.

2. Samples less than ∿200 g--transfer sample to 1 qt can and ball-mill according to sample preparation procedure as recommended in letter to Dunaway from Essington (H7-76-EHE-20, dated 1-23-76).

3. Samples between ∿200 and ∿800 g--replace in original 1 gal sample can and ball mill as per established NAEG procedure.

4. Samples greater than ∿800 g treat according to step 5.

5. For those samples greater than ∿800 g that were not thoroughly mixed in the field during collection (using a concrete mixer), dry the soil as per step #1 and mix thoroughly using either kneading or quartering.

   a. Kneading: Transfer all of the sample into an appropriately large double plastic bag. Seal the bag and mix soil thoroughly, actively, and continually for ∿10 min. Obtain aliquot as per step 6.

   b. Quartering: Spread a sheet of plastic (∿4 ft square) on a flat surface. Place all the sample in the center of the sheet. With a small scoop (200-300 g capacity) transfer a scoopful of soil to each of the four corners alternately until the entire center pile is distributed. Reverse the process, moving a scoopful of soil alternately from each corner to the center until all four corner piles are moved. Repeat process three times. Obtain aliquot as per step 6.

6. Using that small scoop, withdraw small portions of soil from either the plastic bag, the quartered material, or in the case of samples mixed in the field, alternate scoops from each can, and place soil in 1 gal paint can until an ∿800 aliquot has been transferred. Place a portion of the remaining soil sample in an appropriate container for storage noted "not ball milled." On 10% of the samples, collect 3 separate ∿800 g aliquots from the same sample in separate 1 gal cans and treat as separate aliquots for analysis; these will provide a

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*See Editor's Note, page 33.

measure of variability inherent in the procedure. Treat all ∿800 g aliquots as per step 3 above. Subsamples for analyses will be drawn from the less than 200 g (step 2) or 200-800 g (step 3) aliquots.

SOIL SAMPLING AND ANALYTICAL PROCEDURES EMPLOYED BY THE EPA FOR THE NAEG

Wayne Bliss
Environmental Monitoring and Support Laboratory, EPA
Las Vegas, Nevada

SAMPLING PROCEDURE

The soil sampling procedure was developed by a series of steps so that a procedure could be adopted which would give reliable data for defining the distribution and inventory of plutonium in soil around the Nevada Test Site (NTS). Some of the data generated and conclusions made during development are included below to illustrate the development of this procedure as well as to provide logic for developing other procedures which may be necessary when conditions differ from the desert environment.

Since desert soil is too dry and coarse to use "cookie cutter" or auger techniques, soil samples are collected by a "pit" technique. A pit is dug as deep as the maximum sampling depth required and one face is left vertical. A rectangular scoop is used to cut out each successive layer starting at the surface and working downward. After inserting the scoop into the face of the pit, a broad knife is used to close the open end of the scoop for easy removal of the soil. Where successively deeper layers are desired, the scoop and knife are left in place or a flat plate is inserted under the scoop before removal. The soil surrounding the scoop or plate is removed to the depth of the tool so that overlying soil will not fall into the succeeding layer. The scoop is designed to sample a surface area 10 cm by 10 cm so that 10 scoops would be greater than one square foot. Environmental sampling procedures used within ERDA prescribe this as an adequate area for determining deposit inventories (Harley, 1975).

In 1968, the Lawrence Livermore Laboratory collected soil samples at two sites northeast of the NTS which showed positive levels of plutonium. Those locations were chosen by the EPA for the first sampling sites in this survey. One profile sample was collected at each of the two sites. Layers of 0.5, 1.0, 2.0, 4.0, 8.0 and 8.0 cm thickness were collected from the corners and center of a 10 m square, resulting in a total area sampled of 500 cm^2. Later, the EPA discontinued collecting the 0.5 cm layer and the effort was reduced to two scoops per layer from one pit for each profile which gives a layer area of 200 cm^2. Eighteen sites around the NTS, plus Baker, California, and Kingman, Arizona, were selected for further profile sampling. Two profiles were collected from each of the 20 locations. The results showed that 90% or more of the plutonium resided in the top 3 cm of 86% of the profiles collected. All the detectable plutonium resided in the top 3 cm in 71% of the profiles collected. Not included in these figures is one profile where plutonium concentrations were below the minimum detectable quantity. Following this work, a meeting was held with other soil

sampling groups in the NAEG study and it was agreed that future profile layers would be collected in successive 2.5 cm increments so that profile layers could be related to surface area samples as defined below (Los Alamos Scientific Laboratory, 1972). It should be remembered, however, that the area of profile samples is small and not as representative of deposition as a sample area of one square foot.

Based on the analytical results of the profile samples and other soil sampling studies, a standardized sampling procedure was adopted to define area deposition. Scoops similar to those used in profile sampling were constructed to collect a surface area of 100 cm^2 to a depth of 5 cm. The depth of 5 cm was chosen to assure collection of nearly all the deposited plutonium. In field collection more concern is placed on the area sampled than on the depth sampled; however, experience has shown that depth is realistically accurate to within about half of a centimeter. Emphasis is placed on area because of its use in computing deposition. Because of the possibility of diluting a sample containing plutonium with underlying "clean" soil, units of area deposition are more meaningful than units of concentration in relating soil results produced by different sampling techniques. Determination of concentration requires that the report writer provides profile information and the depth of sampling for each survey.

Ten scoops are collected from each location and composited to make one sample. The scoops are selected from undisturbed nonvegetated areas as much as possible. There may be other areas which would yield more or less plutonium deposition such as blow sand under bushes, rocky areas, etc.; however, the open desert areas present the most standard area for sampling.

Sampling sites are selected from the intersections of an 8 by 8 km square grid adjusted to coincide with existing roads and trails to minimize equipment abuse, but allow the collection of as many samples as possible.

SAMPLE PREPARATION

Samples are bagged in the field and returned to the laboratory for preparation for analysis. The samples are oven-dried at 105^{o} C for at least 24 hours, weighed, and screened to 10 mesh. The coarse fraction is discarded and the smaller fraction retained for analysis. The sample is subdivided using a Jones sample splitter. An aliquot of about 300 g is analyzed for gamma emitters using either NaI(Tl) or Ge(Li) counting techniques and about 150 g is submitted for plutonium analysis.

SAMPLE ANALYSIS

Soil sample analytical procedures are presented in detail in the referenced literature (Talvitie, 1971; 1972; Johns, 1975). In summary, the 300 g portion is counted 40 minutes on a 10.2 by 10.2 cm NaI(Tl) crystal coupled to a 400 channel pulse height analyzer. Gamma-emitting radionuclides are

quantitated by a computer using a least squares technique. The 150 g portion is dried, ground, and mixed. A 10 g aliquot is ignited at 700° C and dissolved in nitric and hydrofluoric acids. Nitrate, fluoride, and silica are removed with hydrochloric acid and the plutonium is separated by ion exchange. The activity of plutonium is determined by alpha spectroscopy using ^{236}Pu as an internal reference standard. At two standard deviations, 0.005 pCi of ^{239}Pu per 10 g sample of soil may be detected, which is adequate to detect worldwide fallout. Results defining deposition are computed by multiplying the total weight of the portion of the total sample which was less than 10 mesh by the concentration, dividing by the area sampled, and applying the appropriate conversion factors to yield nCi/m^2.

REFERENCES

1. Harley, J. H., Ed. 1975 (Rev.). "HASL Procedures Manual." Health and Safety Laboratory, ERDA, New York, NY. p. B-05-02.

2. Johns, F. B., Ed. 1975. "Handbook of Radiochemical Analytical Methods." Report EPA-680/4-75-001. Environmental Monitoring and Support Laboratory, Las Vegas, NV.

3. Los Alamos Scientific Laboratory, Los Alamos, NM. October 12, 1972. Draft Minutes of Meeting With EPA, NERC-LV, 10/5/72. E. B. Fowler (ref. H7-EBF-335).

4. Talvitie, N. A. 1971. "Radiochemical Determination of Plutonium in Environmental and Biological Samples by Ion Exchange." *Analytical Chemistry* 43:1827-1830.

5. Talvitie, N. A. 1972. "Electrodeposition of Actinides for Alpha Spectrometric Determination." *Analytical Chemistry* 44:280-283.

VEGETATION SAMPLING PROTOCOL FOR INVENTORY--AREA 13 OF NTS

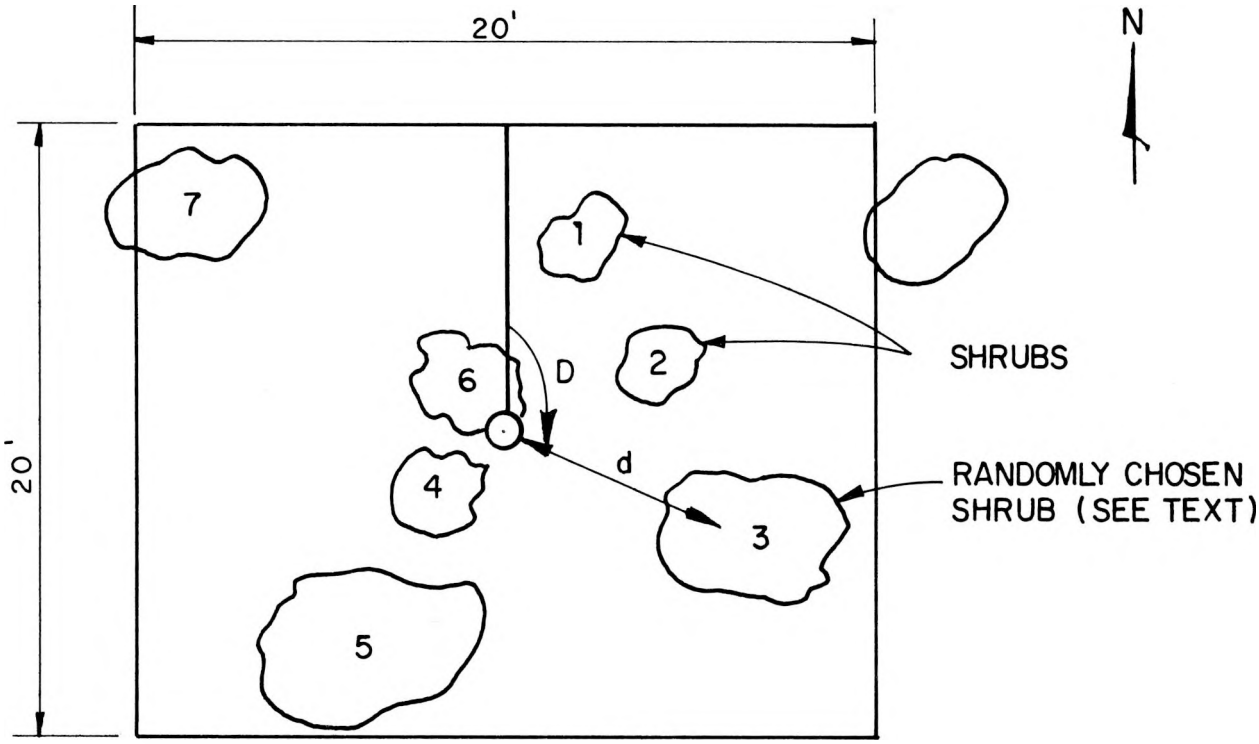
October, 1972

R. O. Gilbert
Battelle Pacific Northwest Laboratories
Richland, Washington

When sampling vegetation for inventory of $^{239-240}Pu$, we wish to obtain the vegetation samples within a prescribed distance from soil sampling locations. The vegetation sampling field crew will be supplied with a map of Area 13 as well as a list of random locations at which soil samples (surface or profile) have been taken. These random locations are identified by the north and east coordinates on the provided map of Area 13. These coordinates will be given in Columns 2 and 3 of the Vegetation Field Data Sheet (attached). If convenient, the soil and vegetation samples could be collected at the same time. This would eliminate the necessity of finding the random locations twice, once for the soil sampling crew and once for the vegetation people. The vegetation samples should not be taken before the soil samples are collected since it is always a possibility that a random location specified for a soil sample may not be usable (see "Soil Sampling Protocol for Inventory--Area 13 of NTS"). If vegetation samples are to be collected after the soil samples, then the vegetation sampling crew should double check with the soil sampling crew to determine the locations at which soil samples were actually taken.

A 20 x 20 ft. square with the soil sample location at the center will be the sample plot for vegetation associated with the randomly chosen soil location (Figure 1). This area should be enclosed with a suitable rope or ribbon tied to the four corner stakes on the square. The field crew must determine which of the several shrubs within this 20 x 20 ft. area (henceforth called the "sampling frame") will be sampled. The procedure used to determine this shrub is as follows:

> Each shrub within the sampling frame will be numbered from 1 to m, where m = number of shrubs in the sampling frame. A shrub is considered to be in the sampling frame if more than 1/2 of its canopy is inside the plot. That shrub in the northeast corner of the sampling frame which is closest to the true north line is given the number 1. The other shrubs are numbered consecutively from 2 through m as the field crew moves in a clockwise direction around the soil sampling location; i.e., the shrubs in the northeast corner of the square are numbered first followed by those in the southeast, southwest, and northwest corners in that order (Figure 1). It is recommended that identification tags or ribbons not be attached to the shrubs themselves in order to



⊙ RANDOMLY CHOSEN SOIL SAMPLING LOCATION

d: DISTANCE FROM CENTER OF SHRUB TO THE SOIL SAMPLE LOCATION (TO BE RECORDED UNDER ITEM 4 OF THE VEGETATION FIELD DATA SHEET).

D: DEGREES FROM TRUE NORTH (TO BE RECORDED UNDER ITEM 5 ON THE VEGETATION FIELD DATA SHEET).

FIGURE: 1:

EXAMPLE OF A 20 x 20 FT. VEGETATION SAMPLING FRAME SHOWING THE LOCATION OF SHRUBS.

avoid disturbing any Pu contamination on the surface of the shrub. The recommended procedure is to make a quick sketch of the sampling frame indicating the approximate location of the shrubs. The shrubs would then be numbered on the sketch. Once the shrubs are numbered, then a list of random numbers is used to pick one of the m shrubs at random. In order to avoid bias, it is important that the random number be chosen after the shrubs are numbered; i.e., the field crew should not know the number of the shrub to be sampled before the shrubs are numbered. Hence, it is recommended that a Xerox copy of a random number table be taken into the field with the crew and a 2-digit number be randomly chosen from this table after the shrubs are numbered.

In order to accurately record the location of the randomly chosen shrub in relation to the soil sample location, two measurements must be taken: (i) distance in feet from the center of the sampled bush to the soil sample location, and (ii) degree reading from true north relative to the soil sample location (see Figure 1). This information is recorded on the Vegetation Field Data Sheet in Columns 4 and 5, respectively. The degree reading can be obtained quickly and accurately using a good quality hand-held compass.

In Columns 6 through 10, the following information must be recorded for each shrub sampled:

Column 6: Total number of bushes in the sampling frame

Column 7: Genus of sample vegetation (first 2 letters)

Column 8: Species of sample vegetation (first 2 letters)

Column 9: Approximate size of sample (in grams)

Column 10: Record here any comments felt to be pertinent that would describe any unusual circumstances concerning the shrub sampled. Examples might be comments relating to the shrub's size or location with respect to other shrubs or the soil conditions, whether the shrub is half dead or healthy, etc.

The vegetation sample should be taken from the chosen bush in accordance with instructions from Van Romney. Since we are not pooling vegetation samples, the sample should be large enough so that a reasonable accurate $^{239-240}Pu$ count or concentration can be obtained from this one sample. The sample should be properly bagged and labeled. In particular, the following information should be secured to the vegetation sample: (i) north and east coordinate numbers of the soil sample associated with the vegetation sample, (ii) the distance and degree measurements recorded in Columns 4 and 5 of the Field Data Sheet, (iii) genus and species of the shrub from which the sample was taken, and (iv) date the sample was taken. This information is required because we want to be able to compare the vegetation sample with its associated soil sample.

The soil sampling crew has been instructed to refrain as much as possible from disturbing the vegetation around the soil sampling location. However, if the vegetation sampling crew feels that a shrub has been disturbed, they should not include that shrub in the list of shrubs from which the sample is to be collected. This may be necessary, e.g., if the soil crew finds it necessary to remove or cut away part of a shrub to enable them to obtain their soil sample.

Throughout this sampling protocol, we have assumed that one vegetation sample would be obtained in each sampling frame. It is possible that we may need to sample two or more samples per sampling frame. In such an event, the above procedures would remain unchanged except for obvious changes needed to randomly choose two or more shrubs and record the data for each on the Field Data Sheet.

At the time this protocol is being written, it has not been decided whether to take one or more vegetation samples at each soil sample location. That is, we may find that vegetation samples need be taken at only half as many locations as soil samples in order to obtain a precise inventory estimate. In this event, the vegetation field crew will be given the soil sampling locations for which vegetation samples are to be collected.

SHRUB VEGETATION SAMPLING FOR INVENTORY IN AREA 13 – FIELD DATA SHEET

Field Crews: _____

Date Field Collection: Month: _____ Day: _____ Year: _____

Approximate Size of Sample (in grams): _____

Dimensions of Sampling Square: _____

SOIL SAMPLE LOCATION			VEGETATION SAMPLE LOCATION		DESCRIPTION OF VEGETATION				
ISOPLETH NO.	NORTH COORINATE	EAST COORDINATE	DISTANCE FROM CENTER OF SAMPLED BUSH TO SOIL SAMPLE LOCATION (IN FT.)	DEGREE READING FROM TRUE NORTH RELATIVE TO SOIL SAMPLE LOCATION	NUMBER OF BUSHES IN SAMPLING SQUARE	GENUS OF SAMPLED VEGETATION	SPECIES OF SAMPLED VEGETATION	APPROXIMATE SIZE OF VEGETATION SAMPLE (IN GRAMS)	COMMENTS
1	2	3	4	5	6	7	8	9	10

ISOPLETH

1 <1,000
2 1,000 – 5,000
3 5,000 – 10,000
4 >10,000

STANDARD NEVADA APPLIED ECOLOGY GROUP PROCEDURES FOR COLLECTION OF VEGETATION SAMPLES FROM INTENSIVE STUDY AREAS

E. M. Romney
Laboratory of Nuclear Medicine, UCLA
Los Angeles, California

and

R. O. Gilbert
Battelle Pacific Northwest Laboratories
Richland, Washington

INTRODUCTION

The procedures described below can be adapted to meet most field situations and sampling purposes; however, they are outlined herein specifically for the sampling of shrub vegetation in desert areas at the Nevada Test Site (NTS) known to be contaminated with plutonium. The soil and vegetation sampling programs of the Nevada Applied Ecology Group (NAEG) studies are closely integrated in order to more efficiently coordinate and correlate the subsequent radiochemical analyses and data processing phases of the work.

Vegetation is a functional component of the desert environment involved in the transfer of radioactive elements from soil to grazing animals. Two different transport mechanisms are involved in the contamination of vegetation. They are: (1) uptake and concentration of radionuclides in plant parts through root systems, and (2) superficial contamination from radioactive fallout debris entrapped on the surface of plant foliage. In contaminated areas where plutonium is involved, this second mechanism appears to be the most important inasmuch as there is evidence of very low uptake of plutonium through plant roots.

The needs for collecting random vegetation samples within a prescribed distance from soil sampling locations were considered in developing these procedures along with the use of sampling methods compatible with radiological safety requirements. Work in contaminated study areas is performed under supervision of NTS Rad-Safe personnel.

FIELD SAMPLING PROCEDURES

1. The vegetation sampling team is supplied a map of the study area and a list of the random locations at which soil samples are taken within each activity isopleth. These random locations are identified by

north and east coordinates at which reference stakes are placed by the soil sampling team (refer to the procedure for collection of soil samples for the method of site selection and identification). The soil sample is collected at a distance of 3 feet and 180 degrees (from true north) from the reference stake before the vegetation sample is taken.

2. A 20-foot diameter circle, with the soil sample located at the center, is the sampling plot for vegetation associated with the randomly chosen soil location. A shrub is considered to be inside the sampling plot if more than half of its canopy lies within the circle. Shrubs are numbered clockwise from true north, and a list of random numbers is used to select one of the shrubs. In order to accurately record the location of the randomly chosen shrub in relation to the soil sample location, two measurements must be taken: (1) distance in feet from the center of the sampled shrub to the soil sample location, and (2) degree reading from true north relative to the soil sample location. This information is recorded on the vegetation field data sheet along with the isopleth number, the collection date, the north and east coordinates, the sample number, and the shrub genus and species.

3. From 300 to 500 grams of plant foliage are clipped from the sampled shrub and stuffed into a one-gallon, press-lid, metal can. The container lid is labeled with the study area code, the activity isopleth number, the sample number, and the date of collection.

4. Collected samples are packaged by Rad-Safe for transport to the processing laboratory.

GENERAL COMMENTS

Experience in sampling vegetation from intensive study areas on the Nevada Test Site and Tonopah Test Range disclosed certain situations that deviated from an ideal random selection of vegetation samples. In many instances, only one or two shrubs occupied space within the 20-foot diameter sampling plot. Where shrubs did not occur inside the sampling plot, the nearest shrub outside the plot was sampled. In those cases where grass species were the only vegetation inside the sampling plot, clippings from several different clumps within the plot were pooled together for a sample.

STANDARD NEVADA APPLIED ECOLOGY GROUP (NAEG) PROCEDURES FOR PREPARATION OF VEGETATION SAMPLES FROM INTENSIVE STUDY AREAS

E. M. Romney
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Los Angeles, California

and

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INTRODUCTION

Preparation procedures for handling 300 to 500 gram samples of woody vegetation were developed with an aim to avoid unnecessary transfer of plutonium-contaminated material from one container to another prior to analysis for ^{239}Pu and ^{241}Am. The necessary steps in sample preparation involve an assignment of a library identification number, packaging for transport, determination of vegetation sample weights, and dry-ashing for radiochemical analysis.

LABORATORY PREPARATION PROCEDURES

1. Vegetation samples are received from the field in sealed, one-gallon, press-lid metal cans. Information labeled on the container lid is verified with the field data collection sheet and the container, in turn, is labeled with a library identification number. Field weight of the vegetation sample is determined by difference in weight compared to an empty reference can. The sealed container is then enclosed in a plastic bag and packed for shipment to the radiochemistry laboratory.

2. Information from the field data sheet and the verified sample container is transferred to IBM cards for subsequent data processing. Included are the following data:

    a. Library identification numbers of the vegetation and corresponding soil sample.

    b. Study area and test event identity.

    c. Activity isopleth and vegetation sample number.

d. Reference stake number and north and east coordinates.

e. Distance (feet) and azimuth (degree from true north) of shrub relative to the soil sample location.

f. Estimate of sample activity (from soil sample ^{241}Am counts).

g. Field weight of sample.

h. Plant genus and species.

Samples received and logged in at the analytical laboratory must be further reduced in size in preparation for radiochemical analysis. This is done in the following steps.

3. The lid is removed from the sample container and the open end is covered with perforated foil. The container and sample is weighed, placed in an oven to dry at 110°C., and then weighed again to determine the oven-dry vegetation sample weight by difference.

4. The sample is carbonized at 250°C. in the drying oven overnight and allowed to cool. The foil is removed, a double walled plastic bag is fitted over the opening, and the carbonized vegetation sample is transferred to the plastic bag and crushed to a powder form.

5. The sample is now considerably reduced in size and ready for radiochemical analysis, the initial phase of which involves a transfer of the sample to a tared Pyrex glass beaker and subsequent ashing at 600°C. for 48 hrs. in an ashing furnace. The radiochemical procedures are describe by Major, *et al.* (1973).

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STANDARD NEVADA APPLIED ECOLOGY GROUP PROCEDURES FOR COLLECTION OF SMALL VERTEBRATES FROM INTENSIVE STUDY AREAS

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INTRODUCTION

Biological studies of the Nevada Applied Ecology Group (NAEG) have concentrated on the potential effects of residual plutonium (Pu) in the soil, plants, and biota of selected areas of the Nevada Test Site (NTS). Previous surveillance over a 10-year period has documented presence of this radionuclide in these areas (Romney *et al.*, 1970). Although research was initiated in the early years after the first atomic explosion, and much was known concerning its dispersal, deposition, and effect, upon biota after aboveground explosions (Wick, 1967), further study was warranted.

Studies related to Pu contamination have received high priority for current research by the NAEG. This group has entered into interdisciplinary research to further evaluate environmental and radiological health problems related to Pu contamination.

As a part of this research, ecological studies of vertebrates in Pu-contaminated areas of the NTS begun in March, 1972, continue to date (Moor and Bradley, 1974). During the initial phase of the studies, an important consideration was development of standardized procedures for inventory, census, and collection of small vertebrates. This methodology is described and discussed in the following report.

INVENTORY

The first consideration was a detailed inventory of vertebrate species encountered in the study sites. In general, the vertebrate biota of NTS is well known; checklists and brief species' accounts are readily available (Hayward *et al.*, 1963; Tanner and Jorgensen, 1963; and Jorgensen and Hayward, 1965).

Small mammals, birds, and lizards were inventoried as part of the overall census programs. In addition, incidental observations of other species (snakes, larger mammals, and some birds) were recorded in field notes. Both census and field note observations were used to develop a species inventory of vertebrates in each study site.

CENSUS AND COLLECTION

Whenever feasible, existing grid systems developed by the NAEG on the study sites were used as locations for census and collection of small vertebrates. Basic grid patterns consisted of numbered steel stakes installed in parallel lines at 400-foot intervals. In some instances, grids on a smaller scale were available.

Whenever possible, census was taken at regular intervals of approximately two months. In some instances, access to study sites could not be obtained with this regularity due to other scheduled NTS activities. The census period usually consisted of four days and three nights.

Census and collection techniques varied with ease of capture and observation, and are described in some detail for each group.

Reptiles

Census of lizards on the study sites was limited, and in some instances consisted of developing an index of relative abundance by season for each study site. Relative abundance was based upon observations of lizards along walking census strips of the basic grid pattern. Observation periods occurred at the time of day when soil and air temperatures were optimal for lizard activity for any particular season. Lizards, which were either active or flushed along these lines, were in almost all instances easily identifiable, especially in the open structure of desert shrub plant communities. Relative abundance was based upon a percentage of the total for each species observed during each census period. Line transect counts of lizards have been shown to be inadequate estimates of density (Degenhardt, 1966; Pianka, 1970; Medica *et al.*, 1971). This is also true of relative abundance indices since lizard species vary greatly in foraging patterns, reactions to the observer, and ease of detectability. However, even a crude index of relative abundance does provide considerable understanding of the composition of the lizard fauna, seasonal activity patterns, and the determination of dominant or important species for further study, including collection, autopsy, and radioanalysis.

An alternate census method employed was noosing, marking, and recapture. In general, procedures were as described by Medica *et al.* (1971). The noosing devices consisted of modified fiberglass fishing poles varying in length from 4 to 6 feet. Longer pole lengths were used for larger and more wary lizards, i.e., *Cnemidophorus tigris* and *Callisaurus draconoides*, whereas a shorter pole was employed to noose smaller lizards such as *Uta stansburiana*. A wire extension, approximately 1 ft. in length with a loop on the end, was attached to the tip of the pole. The loop consisted of a slip noose made of black surgical thread and was approximately twice the size of the lizard's head.

After noosing, the lizard was identified, sexed, and classified as hatchling or adult. Individuals were judged to be reproductively active by the presence of breeding coloration in males and by palpation in females. Captives were then marked by means of a toe clip and released. The numbering of toe clips is illustrated in Fig. 1. The system consists of cutting at least one toe, but never more than one from each foot. The individual is referred to as, for example, *Uta* LF1 (left front foot, toe number 1) or *Uta* LF1, RF1 (left front foot, toe number 1; and right front foot, toe number 1). After toe clipping, the toe clip combination is crossed off the toe clip chart for that species on the study site (Table 1). These toe clip charts are kept on a clipboard in the field and reduce the possibility of duplication of toe clips for each species.

The location of capture for each lizard was recorded in relation to grid location numbers. This allowed ease of recapture and some idea of movements and residence in the study sites.

Collections for autopsy and radioanalysis were made of lizards in the study sites. The collection technique employed was by noosing. Lizards were then placed in individual plastic bags with identification tags, kept in ice until returned to the Civil Effects Test Organization (CETO) Laboratory, and frozen. Individuals of established residence (previously marked at approximately the same location) were utilized for radioanalysis. In addition, males which exhibited territorial behavior were judged to be resident and were collected for autopsy. An alternate method which NAEG hopes to employ involves shooting 22-caliber bird shot. This method involves potential tissue damage and should be used with caution. However, lizards have been found to be only temporarily stunned, and with intact skin, if shot from a suitable distance and/or hit only at the edge of the shot pattern. Employment of this method would greatly reduce collection time and man-hours while increasing sample sizes for autopsy and radioanalysis.

The secretive and largely nocturnal habits of snakes coupled with excessive avoidance of trapping procedures present numerous collection difficulties. Collections for autopsy and analysis will be based upon incidental observation and capture. After initial sampling needs are met, attempts will be made to employ mark and recapture techniques to document residency. The marking technique employed will involve partial removal of ventral scutes.

Birds

Birds represent a significant segment of the small vertebrate fauna in NTS. They are especially abundant and have considerable impact upon desert scrub ecosystems during spring and fall migrations. However, few species are resident in the more arid desert ecosystems. Although an important part of the vertebrate fauna, they do present numerous problems due to both their seasonality and high mobility. Residence in the study areas can only be determined for breeding birds; therefore, they have not been used for autopsy or radioanalysis.

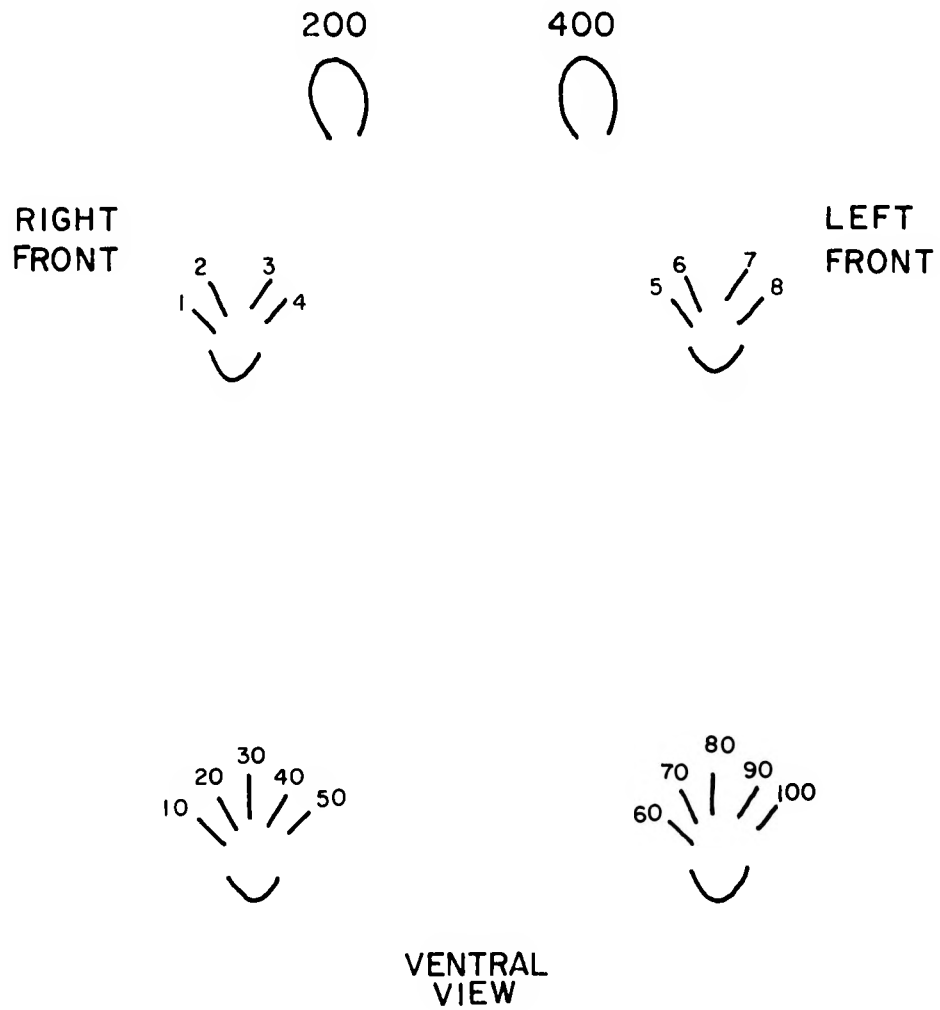

FIGURE 1.
ILLUSTRATION OF THE NUMBERING OF TOE CLIPS USED FOR IDENTIFYING INDIVIDUAL RODENTS.

Table 1. Toe Clip Chart Used to Indentify Individual Lizards amd Mammals in Study Areas at NTS

AREA _____ SPECIES _____

Toe Clip Chart

RF_1	LF_1	RH_1	LH_1
RF_2	LF_2	RH_2	LH_2
RF_3	LF_3	RH_3	LH_3
RF_4	LF_4	RH_4	LH_4
RF_5	LF_5	RH_5	LH_5
RF_1LF_1	RF_2LF_1	RF_3LF_1	RF_4LF_1
RF_1LF_2	RF_2LF_2	RF_3LF_2	RF_4LF_2
RF_1LF_3	RF_2LF_3	RF_3LF_3	RF_4LF_3
RF_1LF_4	RF_2LF_4	RF_3LF_4	RF_4LF_4
RF_1LF_5	RF_2LF_5	RF_3LF_5	RF_4LF_5
RH_1RF_1	RH_2RF_1	RH_3RF_1	RH_4RF_1
RH_1RF_2	RH_2RF_2	RH_3RF_2	RH_4RF_2
RH_1RF_3	RH_2RF_3	RH_3RF_3	RH_4RF_3
RH_1RF_4	RH_2RF_4	RH_3RF_4	RH_4RF_4
RH_1RF_5	RH_2RF_5	RH_3RF_5	RH_4RF_5
RH_1LF_1	RH_2LF_1	RH_3LF_1	RH_4LF_1
RH_1LF_2	RH_2LF_2	RH_3LF_2	RH_4LF_2
RH_1LF_3	RH_2LF_3	RH_3LF_3	RH_4LF_3
RH_1LF_4	RH_2LF_4	RH_3LF_4	RH_4LF_4
RH_1LF_5	RH_2LF_5	RH_3LF_5	RH_4LF_5
LH_1RF_1	LH_2RF_1	LH_3RF_1	LH_4RF_1
LH_1RF_2	LH_2RF_2	LH_3RF_2	LH_4RF_2
LH_1RF_3	LH_2RF_3	LH_3RF_3	LH_4RF_3
LH_1RF_4	LH_2RF_4	LH_3RF_4	LH_4RF_4
LH_1RF_5	LH_2RF_5	LH_3RF_5	LH_4RF_5

During the first year of the study, censusing was conducted briefly using the strip census method employed by Emlen (1971). Due to the short time period and low priority for this part of the project, sufficient data were not developed to estimate bird density in the study area. The short-term census and direct observations detailed in field notes provided some basis for developing a checklist of bird species for each area.

If it is later found desirable to use birds for autopsy or radioanalysis, more detailed studies to determine residence will be needed using techniques similar to those employed by Emlen (1971).

Mammals

Census techniques of mammals varied depending on size, relative importance, and ease of capture or observation. Census of larger mammals (rabbit size and larger) consisted of developing an index of relative abundance by season for each of the study areas. Methods included casual observation and indirect signs such as tracks, scat, burrows, and resting areas. The locations of sight or sign records in the field were recorded in relation to the grid location numbers previously mentioned.

Small mammal populations were estimated by using live trapping grids. Eight lines of 25 Sherman live traps (nonfolding traps, 3 x 3 x 10 in. made of .025-in. aluminum) located 50 feet apart, comprised the 3.9 hectare grids. Trapping grids were located at various distances from GZ, using established NAEG grid-system reference points. Mammals were trapped for four consecutive days every two months, or whenever access was permitted. Live traps were baited with rolled oats and checked approximately every four hours during the spring and summer breeding seasons. During the winter, when it was extremely cold, traps were checked twice in the evening and then closed to prevent animals from freezing. They were then opened early the next morning. During very hot summer weather, traps were closed late in the morning and opened early in the evening to prevent diurnal animals dying from the effects of high temperatures.

Mammals captured in live traps were marked with a toe clip (Fig. 1), using methods previously described for lizards, and then released. This method assigns a numeric value to each toe on each of the animal's feet. In addition, the animal's right and left ears are assigned values of 200 and 400, respectively, and may be clipped along with toe clips if additional marks are needed (i.e., the first toe of the right front foot is number 1; the first toe of the right hind foot is number 10). This method ensures that each animal is marked with a permanent identification. In this instance, however, the fifth digit on each foot was never clipped. Data recorded at time of capture included: the location of capture in the grid; identification of the animal, toe clip, and notation if the animal was a recapture; sex; relative age (immature, young adult, or adult); and reproductive condition (testes ascended or descended, pregnant or lactating, or nonreproductive).

The density of small mammals in the trapping grid was estimated using mark-recapture methods described by Hayne (1949) using the formula:

$$P = CM/R$$

where P is the population estimate, C is the number of animals captured during the last trapping period, M is the number of marked animals in the area, and R is the number of marked animals captured during the last trapping period. Alternative methods for estimating density of small mammals utilizing live trapping grids with assessment lines are discussed by Smith *et al.* (1974). We consider these techniques more accurate estimators of small mammal densities. These methods do, however, require more frequent and longer periods of accessibility to the grid areas.

The trapping grid was also used to gather animal movement data in order to establish home ranges (the area an animal travels during its daily activities) for each species in each study area during different seasons of a year. Home range estimates were based on recapture locations in the grid. A center of activity was determined for individual animals using the individual recapture locations and the relative frequency with which an animal is found at various locations in the grid (Hayne, 1949; Calhoun and Casby, 1958). The periphery of the home range was estimated by computing 1.96 standard deviations of recapture radii from the center of activity which theoretically includes 95% of the recaptures. The home range determined can then be examined in relation to the distance from GZ. Since the study areas have been surveyed for radioactivity, one can estimate the residual plutonium available within the animal's home range. Recapture radii were averaged by sex for each species and were used to estimate the effective trapping area of the grid. Criteria used for recapture radii of individual animals were as follows:

1. Animal must have been captured 10 or more times at three locations.

2. No obvious shifts in the animal's home range or no inclusion of relatively nonstationary home ranges.

When sufficient data to estimate recapture radii for a particular species were not available, NTS data from similar areas (Jorgensen and Hayward, 1965) were utilized to estimate home-range values. When sufficient grids have been established in each study area, animal populations of entire study areas can be estimated by extrapolation.

Density estimates were also utilized to compute species diversity indices. The Shannon formula (Shannon, 1948; Pielou, 1966; Lloyd, 1968) was used as a general index of species diversity in each study area: $[C/N (N \log 10 N - n_i \text{ Log } 10 n_i)]$, where C converts logs from base 10 to an arbitrary base, N is the total number of individuals of all species, and n_i is the number of the ith species. A high species diversity indicates a relatively large number of common species and a high complexity or diversity. These conditions may reflect a relatively high community stability and maturity (MacArthur, 1955; Margalef, 1963). These indices, therefore, are at least theoretically useful in comparing disturbed (contaminated) areas of NTS with relatively undisturbed desert areas.

In addition to estimates of home range, movements, population densities, and species components in the study areas, gathered data were utilized for other estimates. Aboveground activity of each species in each area is reflected by the relative number of individuals captured seasonally. Reproductive status and recruitment were estimated by the percentage of populations which can be described as either sexually active adults (testes descended, testes ascended, pregnant or lactating) or sexually inactive adults, young adults, or immatures.

Collections for autopsy and radioanalysis were made of small mammals in the study areas. Known resident animals (animals marked 2-6 months previously and recaptured in the same area) were periodically collected. These individuals were etherized, placed in individual plastic bags with a tag bearing appropriate data, taken back to CETO Laboratory, and placed in a freezer. Animals which were accidentally killed either by shock or from heat or cold were also placed in a plastic bag and taken to CETO Laboratory where they were prepared for histopathological examination.

Larger mammals will be shot or trapped with steel traps and placed in a freezer in the CETO building. These animals will also be prepared for radioanalysis and histopathological examination.

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STANDARD NEVADA APPLIED ECOLOGY GROUP PROCEDURES FOR PREPARATION OF SMALL VERTEBRATES FROM NAEG INTENSIVE STUDY AREAS FOR RADIOANALYSIS AND HISTOPATHOLOGICAL EXAMINATION

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INTRODUCTION

Literature on the uptake and toxicity of plutonium (Pu) in laboratory and field animals is extensive and was reviewed thoroughly by Wick (1967). It is apparent from this literature that while Pu uptake by animals is generally very low, even in heavily contaminated areas (Wilson *et al.*, 1960), the doses required to produce adverse physiological effects are also very low (Finkel and Biskis, 1962, and others). In addition, elimination rates are low. For example, $\frac{T}{2}$ equals 200 years in man (ICRP, 1959). These data indicate that intensive long-term study of the uptake and physiological effects of plutonium on animals which are indigenous to areas with residual Pu contamination is warranted.

Samples of native vertebrate animals from Pu-contaminated areas of the Nevada Test Site (NTS) and Tonopah Test Range (TTR) were taken as part of an ecological study of vertebrate populations in Pu-contaminated areas (Moor and Bradley, 1974). Sampling methods used during the above study have been reported (Moor *et al.*, 1974). In general, capture-recapture techniques were employed in the study of population dynamics of vertebrates in Pu-contaminated areas. Known resident vertebrates (animals marked 3-6 months previously) were sampled from study areas at various known distances from GZ, placed in individual plastic bags, and stored in a freezer in the Civil Effects Testing Organization (CETO) Laboratory, NTS.

RADIOANALYSIS

In the CETO Lab, Mercury, Nevada, animals individually wrapped in plastic bags and frozen were thawed and autopsied. Standard measurements and weights were recorded as follows: total length, tail length, hind foot length, ear length, and total body weights of mammals, and snout-vent lengths and total body weights of lizards. During autopsy, efforts were made to avoid cross contamination between animals and between tissues. Latex surgeon's gloves were worn and discarded after each animal was autopsied, and hands and surgical instruments were washed thoroughly after handling each tissue sample. Plastic-lined absorbent paper was placed on

the operating table and replaced after each autopsy. Small vertebrates were dipped into individual aluminum trays containing hot paraffin to minimize the possibility of cross contamination between pelt or outer skin layer and internal tissues (Lindberg *et al.*, 1955). Individuals were carefully skinned and skins were individually wrapped in plastic bags and sealed. An identification tag bearing the sample number, REECo library number, species identification, sex, date, investigator, and location of capture was placed in a plastic bag. The carcass was then thoroughly washed with running tap water. A medial-ventral incision through the body wall was made from the mandibular symphysis to the pubic symphysis. The gastrointestinal (GI) tract from esophagus to rectum or cloaca was removed intact; great care was taken not to rupture the tract, thereby spilling its contents and causing cross contamination of tissues. A gross examination of the animal was made at this time. Any unusual condition was recorded and reproductive status was determined (*i.e.*, pregnant, lactating, testes descended or ascended, or individual sexually inactive or immature). The GI tract and carcass were then placed separately in plastic bags with identification tags and data as described above. Three plastic bags containing the tissue samples for each individual were then placed in a plastic bag with appropriate identifying data clearly visible. These samples were then frozen and shipped in dry ice to the LFE Laboratories, where radioanalysis was performed. Also, all pertinent collected data were recorded in laboratory notebooks and retained by the investigators.

HEMATOLOGICAL STUDIES

The following procedures were used to obtain base line hematological data from animals collected off-site and in NAEG Intensive Study Areas.

Animals were captured alive utilizing Sherman live traps and returned to the laboratory, either CETO or UNLV, and examined within 24 hours of capture. Blood was obtained from rodents by cardiac puncture using ethyl ether as an anesthetic. In all instances, blood was collected in heparnized (ammonium sulfate) syringes. All analyses were done in duplicate, and any test exceeding a 1% difference was repeated. All analyses were done following standard methods (Schalm *et al.*, 1975).

Differential Stain

Blood smears were prepared on clean slides and allowed to air dry. Slides were then fixed with Wright's stain. One hundred cells were counted and identified using the four-field meander method, beginning at the margin of the smear and counting toward the center and then to the margin alternately.

Hemoglobin

The cyanmethemoglobin method was used for hemoglobin determination. To 5.0 ml Drabkin's solution, 0.02 ml of whole blood was added. Hemoglobin is converted to cyanmethemoglobin by the Drabkin's solution which contains ferricyanide and cyanide. The resultant color is indicative of hemoglobin

concentration which was read on a Fisher Flo-thru Hemophotometer in gm/100 ml or Bausch and Lomb Spectronic 20.

Packed Cell Volume

The packed cell volume (PCV) is a measurement of percentage of red blood corpuscles in whole blood. Precalibrated 75 mm heparinized micro-hematocrit tubes were centrifuged in an Adam's Readacrit Centrifuge at 8500 rpm for five minutes and PCV measured on the built-in reader (1-100%).

Erythrocyte Count

Blood was drawn into an RBC pipette (± 1%) and diluted with Haymen's dilution fluid (1:200). A Specto-Bright-line hemocytometer was used for counting erythrocytes. Numbers were estimated in millions/cm^3.

Leukocyte Counts

Blood was drawn into a WBC diluting pipette (± 1%) and diluted (1:20) with 1% glacial acetic acid. A Specto-Bright-line hemocytometer was used for counting leukocytes. Numbers were estimated in thousands/cm^3.

Plasma Protein

Plasma from the packed cell volume test was utilized to estimate concentration of plasma proteins using an American Optical total solids meter No. 10400 Refractometer. Total plasma proteins were read directly on the meter (g/100 ml).

Albumin concentration was determined colormetrically using an American Monitor buffered albumin dye No. 1007. The plasma albumin binds quantitatively to the dye 3, 3', 5, 5'-tetrabromo-m-cresolsulfonphthalein. The albumin dye combination produces an intense blue chromophore which was measured at 600 nm on a Bausch and Lomb Spectronic 20. A standard curve was developed for each sample series, and the albumin concentration calculated in gm/100 ml.

Glucose

Glucose concentration was determined using Harleco reagent and standard set No. 64147. To 10.0 ml reagent (ortho-toluidine 6% in glacial acetic acid), 0.1 ml of plasma was added. After boiling, the resultant green color was measured photometrically on a Bausch and Lomb Spectronic 20 at 630 nm, and the concentration (mg/100 ml ± 2 mg/100 ml) was determined from a standard curve.

Plasma Cholesterol

Plasma cholesterol was determined colormetrically using Harleco standard 7653B and reagent 7653A (acetic acid, acetic anhydride, and sulfuric acid). Plasma (0.1 ml) was added to 5.0 ml of reagent and incubated for 10 minutes at 37° C. The resultant color is proportional to the cholesterol concentration and read on a Bausch and Lomb Spectronic 20 at 625 nm.

HISTOPATHOLOGY

In the CETO Lab, frozen animals were thawed and autopsied. Specimens were washed thoroughly with running tap water and a "surface swipe" was taken and "surveyed" for alpha activity by REECo/Rad-Safe personnel. A medial-ventral incision through the body wall was made from the mandibular symphysis to the pubic symphysis. The diaphragm was carefully cut away, allowing the animal to be successfully spread apart so that all major internal organs could be readily observed. All gross abnormalities and pertinent reproductive data were recorded as previously described. A waterproof tag was placed on the animal's hind foot bearing the identifying information described under "Radioanalysis." Individuals were placed in polyethylene plastic gallon jars containing 10% formalin. For easy identification, sample numbers and the investigator's name were taped to the outside of the jars. For detailed histopathological examinations, samples were shipped to Dr. Gerry Cosgrove, Biology Division, Y-12, Oak Ridge National Lab, Oak Ridge, Tennessee.

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Analytical Procedures

PLUTONIUM PURIFICATION PROCEDURES

McClellan Central Laboratory
(MCL-LP-07-94)

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INTRODUCTION

The chemist should be familiar with the following information concerning plutonium chemistry before continuing with the procedures in this Laboratory Procedure.

1. Plutonium can exist in four oxidation states (III, IV, V, and VI) in aqueous solutions. These states are characterized by the stability of the Pu (III) state and the extensive oxidation (via $KBrO_3$) necessary to obtain the Pu (VI) state. The Pu (IV) state, however, is the principle (sic) oxidation state in aqueous solution.

2. This laboratory's plutonium procedure begins with an aliquot of the solution to be analyzed. It consists of two chloride form anion exchange resin column separations, a nitrate form anion exchange resin column separation and a lanthanum fluoride precipitation. The decontamination steps in the plutonium procedure are based on the following chemical behavior characteristics of the various oxidation states of plutonium ions:

   a. Pu (VI) is adsorbed on Dowex anion exchange resin (chloride form) from HCl concentrations greater than 6.0M HCl.

   b. Pu (IV) is adsorbed on Dowex anion exchange resin (chloride form) from HCl concentrations greater than 2.5M HCl.

   c. Pu (III) is not adsorbed on Dowex anion exchange resin (chloride form) at any HCl concentration.

   d. Pu (IV) is adsorbed on Dowex anion exchange resin (nitrate form) from concentrations greater than 4.0M HNO_3 and Pu (VI) is adsorbed on that same resin from concentrations greater than 6.0M HNO_3.

   e. Pu (III) and Pu (IV) will coprecipitate with LaF_3.

   f. Pu (V) disproportionates in acid medium to Pu (IV) and Pu (VI).

3. Plutonium presents an equilibration problem which is not encountered in other traced analyses performed by this laboratory. This problem exists because of the four possible oxidation states of plutonium in solution and the tendency of plutonium (IV) to polymerize in aqueous media. Specific steps must be employed to achieve equilibration of plutonium samples.

PROCEDURE A

PLUTONIUM PURIFICATION

The recovery of plutonium from the contaminated aliquot is accomplished in the following manner. Refer to Table 1 for the applicable reagents and see Figure 1 for the procedure flow diagram.

Table 1. Procedure A Reagents

REAGENT	REAGENT
$LaCl_3$ (alpha-free); 10 mg/ml	H_3BO_3 (saturated)
2M NH_4I	1M HCl
NH_4I (saturated)	10M HNO_3
4M HCl	10% $(NH_4)_2SO_4$ (pH 2)
5M $NH_2OH.HCl$	Zr (scavenge); 5 mg/ml
1.5M H_2SO_4	

--NOTE--

The La scavenge used in this procedure should be alpha-free to avoid the addition of any alpha-emitting isotopes.

1. Pipette the Pu tracer and sample activity into a 125 ml Erlenmeyer flask. Add 5 drops $LaCl_3$ scavenge and equilibrate the sample and tracer in the following manner:

   a. Add 10 ml $HClO_4$ and boil to dense $HClO_4$ fumes. Allow to cool.

   b. Dilute with 10 ml H_2O.

   c. Add 3 drops saturated NH_4I solution.

   d. Heat to boiling on a hot plate. Add HNO_3 dropwise until the solution becomes colorless. Again boil to dense $HClO_4$ fumes and repeat steps b, c, and d. Then boil the final solution to 2 to 3 ml. Transfer the sample to a 40 ml glass centrifuge tube with three 2 ml H_2O washes.

2. Precipitate $La(OH)_3$ with NH_4OH. Centrifuge and discard the supernate. Dissolve the precipitate in 3 ml HNO_3 and boil for one minute. Add 10 ml H_2O and reprecipitate $La(OH)_3$ with NH_4OH. Coagulate, centrifuge, and discard the supernate.

--NOTE--

Pu will not be adsorbed on an anion exchange resin column in the Pu (III) oxidation state. Thus, the sample must be boiled in HNO_3.

3. Dissolve the precipitate in 10 ml HCl which contains 1 drop HNO_3. Load the sample on a pre-equilibrated anion exchange resin column in the chloride form. Rinse the tube with a 1 ml wash of HCl and load the rinse on the column. Wash the column with 10 ml HCl.

--NOTE--

The resin bed is Dowex-1 X 8, 50 to 100 mesh, 4 cm X 12 mm diameter. Pre-equilibrate the bed with 15 ml HCl which contains 1 drop HNO_3.

--SAFETY PRECAUTIONS--

When performing the following steps, handle the hydrofluoric acid with extra caution. This acid will cause severe and very painful skin burns if it contacts the skin. Immediate medical attention is required if HF is splashed onto the skin or into the eyes. A temporary first-aid measure involves washing the affected area with copious amounts of water and Hyamine 1622. Gloves will be worn during all operations involving the use of HF. For further details of the toxicology of HF refer to Sax, Dangerous Properties of Industrial Materials, Third Edition, Reinhold Publishing Corp, New York, 1968, pp 823.

4. Elute the zirconium fraction with 10 ml HCl which contains 1 drop HF followed by a 10 ml HCl wash.

5. Elute the Pu with 10 ml HCl which contains 5 drops saturated NH_4I solution into a 50 ml Nalgene tube. Wash the column with 10 ml HCl. RECORD THIS ELUTION TIME AS THE Pu-Np SEPARATION TIME.

6. Add 2 ml $5\underline{M}$ $NH_2OH \cdot HCl$, 5 drops $LaCl_3$ scavenge, 6 drops saturated NH_4I solution, $\overline{2}$ drops Zr scavenge and digest in a hot bath for five minutes. Add 2 ml HF, stir and place in a hot bath for five minutes. Centrifuge the LaF_3 and discard the supernate.

--NOTE--

The $NH_2OH \cdot HCl$ and the NH_4I are added to insure the complete reduction of the Pu to the Pu (III) oxidation state so that it will coprecipitate with LaF_3.

7. Slurry the precipitate with 5 ml saturated H_3BO_3 solution and then dissolve it in 2 ml HNO_3. Transfer the sample to a 40 ml tube with three 2 ml H_2O washes and precipitate $La(OH)_3$ with NH_4OH. Centrifuge and discard the supernate. Dissolve the precipitate in 3 ml HNO_3 and boil for one minute. Cool, add H_2O, 1 ml saturated H_3BO_3 solution and reprecipitate $La(OH)_3$ with NH_4OH.

8. Repeat the anion exchange resin column and plutonium elution sequence outlined in steps 3, 4 and 5. Collect the Pu in a leached 50 ml Erlenmeyer flask.

--CAUTION--

From this point on, use leached glassware only. All reagents are to be prepared with triply distilled H_2O and M/S distilled HNO_3. These precautions are necessary to prevent inert materials which may be present in ordinary reagents or on unleached glassware from being deposited on the final plated sample.

9. Add 5 ml HNO_3 and boil to near dryness. Repeat this process until the sample has been boiled to near dryness twice from a colorless solution.

--NOTE--

If the sample is not to be counted immediately, stop the analysis at this point. When the sample is to be counted start with step 10.

10. Add 3 ml HNO_3 and boil to near dryness. Cool. Add 2 ml 10$\underline{M}$ HNO_3 and heat gently to the appearance of vapors. Load the sample on a pre-equilibrated nitrate column. Perform 2 additional 2 ml 10$\underline{M}$ HNO_3 rinses of the flask and load the rinses on the column.

--NOTE--

The resin bed is Dowex-1 X 8, 100 to 200 mesh, 2 cm X 8 mm diameter. Pre-equilibrate the bed with two column volumes 10$\underline{M}$ HNO_3.

11. Wash the column twice with 5 ml 10$\underline{M}$ HNO_3. RECORD THE LAST DROP OF THE SECOND WASH AS THE Pu-Am SEPARATION TIME.

12. Wash the column with 1 ml H_2O and collect the H_2O wash in a clean 50 ml Teflon beaker. Elute the plutonium into the Teflon beaker with 5 ml 1M HCl which contains 3 drops 2M NH_4I followed by 5 ml 4M HCl.

--NOTE--

It is necessary to remove residual HNO_3 from the column before adding 1$\underline{M}$ HCl to avoid the formation of aqua-regia and to prevent the HNO_3 from reacting with NH_4I. Both of these events could prevent the NH_4I from reducing the Pu.

13. Add 2 drops 1.5$\underline{M}$ H_2SO_4 and evaporate the solution to the appearance of SO_3 fumes.

14. Transfer to a pre-plated cell with three 0.5 ml washes of pre-plated 10% $(NH_4)_2SO_4$ solution. Electrodeposit for one hour at 0.15 amperes. Make basic with NH_4OH before disconnecting the current. Flame and mount.

--NOTE--

The cell is pre-plated with 10% $(NH_4)_2SO_4$ solution for one hour to remove contaminates which otherwise would plate on the final mount form.

Add $LaCl_3$ and equilibrate tracer and activity (Para 1)

Precipitate $La(OH)_3$ with NH_4OH (Para 2) (Para 7)

1,2

Dissolve in HNO_3 and boil (Para 2) (Para 7)

1,2

Reprecipitate $La(OH)_3$ (Para 2) (Para 7)

1,2

Dissolve in HCl and load on a Dowex-1 anion exchange resin column (Para 3) (Para 8)

1,2

a. Wash with HCl-HF (Zr) (Para 4) (Para 8)

b. Elute Pu with HCl-NH_4I (Para 5) (Para 8)

1

Add $NH_2OH \cdot HCl$, NH_4I, Zr scavenge, $LaCl_3$ (Para 6)

1

Precipitate LaF_3 with HF (Para 6)

1

Dissolve LaF_3 with H_3BO_3 and HNO_3 (Para 7)

Convert the sample to the nitrate form (Para 9)

Transfer to a Dowex-1 nitrate column with 10M HNO_3 (Para 10) (Para 11)

a. Wash with 1 ml H_2O into 50 ml Teflon beaker (Para 12)

b. Elute Pu with 1M HCl-2M NH_4I (Para 12)

c. Wash with 4M HCl (Para 12)

Add H_2SO_4, fume, and electrodeposit (Para 13) (Para 14)

Figure 1. Flow Diagram for Procedure A

PROCEDURE B

PLUTONIUM RERUN

Further purification of plutonium samples from procedure A is accomplished in the following manner. Refer to Table 1 for the applicable reagents and see Figure 1 for the procedures flow diagram.

Table 1. Procedure B Reagents

REAGENT	REAGENT
$LaCl_3$ (scavenge); 10 mg $LaCl_3$ per ml	HF
4M HNO_3	H_2SO_4

1. Place the sample in a 50 ml Teflon beaker containing 5 ml HNO_3, 1 ml H_2SO_4, 5 drops $LaCl_3$, and 1 ml HF.

2. Boil to a small volume and transfer to a 40 ml glass centrifuge tube with 4M HNO_3.

3. Add 5 ml H_3BO_3 and precipitate $La(OH)_3$ with excess NH_4OH.

4. Proceed to the appropriate step of procedure A.

--NOTE--

The sample should be alpha counted before and after the stripping process, no more than 10% of the activity should remain on the plate. If there is more than 10% activity, repeat the stripping process.

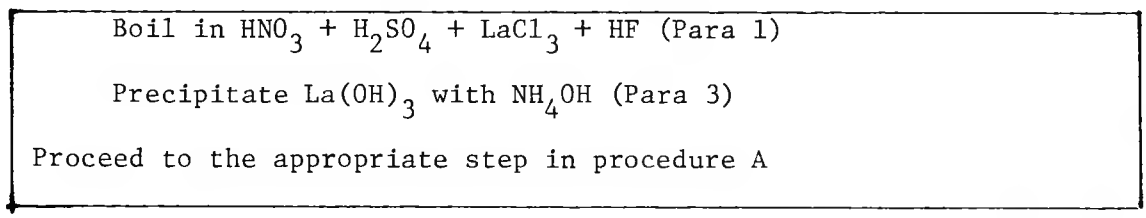

Figure 1. Flow Diagram for Procedure B

AMERICIUM (CURIUM) PURIFICATION PROCEDURE

McClellan Central Laboratory
(MCL-LP-7-95)

(Ed. Note: Previously published as
Americium (Curium) Purification,
McClellan Central Laboratory
Procedure MCL-LP-07-95 (1969).)

INTRODUCTION

Americium and curium exist in the Am(III) and Cm(III) oxidation states throughout the purification procedure used in this laboratory. These elements, when in the Am(III) and Cm(III) oxidation states, will not adhere to an anion exchange resin in the chloride form. They will, however, adhere to a cation exchange resin. The elution of americium and curium from a cation exchange resin column is accomplished by the use of a solution of pH 4.2 ammonium alpha-hydroxy isobutyrate (HIBA).

Americium and curium are separated from the rare earths by a concentration gradient elution using HIBA. The americium and curium bands are found between samarium and neodymium and are removed from the column at about 0.345M HIBA. It has been found that neodymium cannot be used as a marker for these samples because it trails into the americium sample and a contaminated plate will result. Praseodymium is used in place of neodymium to mark the bands.

The americium is separated from the curium by taking standard portions of the elutrient between the samarium and praseodymium markers. The successful electrodeposition of these samples depends upon the separation of the sample from the HIBA. This is done by adsorbing the sample onto a cation exchange resin column and then eluting it with 8M HNO_3.

Only one procedure is available for purifying americium (curium). This procedure is designed for the purification of americium (curium) from a solution of HIBA resulting from the separation of the sample on a cation exchange resin column.

PROCEDURE A

AMERICIUM (CURIUM) (FROM ALIQUOT)

REAGENTS

Sm scavenge (10 mg Sm/ml)	8M HNO_3
Pr scavenge (10 mg Pr/ml)	0.2M HCl
1.5M H_2SO_4	2M HCl

FLOW DIAGRAM

Sample + tracer + Pr and Sm scavenges

Pass through a Dowex-1x8 anion exchange resin column with HCl

Rare earth pre-column treatment

Rare earth Dowex-50x8 cation exchange resin column

Separate americium and curium fractions

Adjust acidity to 0.3M HCl

Pass solution through a Dowex-50 cation exchange resin column

a. Wash column with 0.2M HCl and H_2O
b. Elute sample with 8M HNO_3

Fume and electrodeposit

PROCEDURE

1. Add one ml each Sm scavenge and Pr scavenge to the Am^{243} and Cm^{244} tracers in a 40-ml glass centrifuge tube.

2. Dilute to 15 ml with H_2O. Precipitate a combined Pr and Sm hydroxide with NH_4OH. Centrifuge and discard the supernate. Wash with 5 ml H_2O.

3. Dissolve the precipitate in 10 ml HCl-saturated HCl and load onto an anion exchange resin column.[§] Wash the column with 10 ml HCl and collect the wash and the previous load in a 125-ml Erlenmeyer flask.

4. Treat the sample according to the rare earth pre-column procedure, column operation, and americium-curium separation steps as described in DECONTAMINATION AND DETERMINATION PROCEDURE FOR RARE EARTHS AND YTTRIUM, dated 4 November 1965.

5. Equilibrate a cation exchange resin column.[#$]

6. Add sufficient concentrated HCl to the complexed americium or curium to make the resulting solution 0.3M in HCl.[%] Mix thoroughly and load onto the cation exchange resin column. Wash the column with 15 ml 0.2M HCl followed by 3 ml H_2O.

[§]The resin bed is Dowex-1x8, 50-100 mesh, 4 cm long x 12 mm diameter. Equilibrate the resin bed with HCl.

[#]The resin bed is Dowex-50x8, 100-200 mesh, 1 cm long x 12 mm diameter and is equilibrated with 15 ml HCl, followed by 15 ml H_2O, 5 ml 2M HCl and finally 15 ml 0.2M HCl.

[$]The equilibration with concentrated HCl is designed to insure that cation resin is in the hydrogen form. The 2M HCl aids in the removal of traces of iron, manganese, and other elements which may affect the cleanliness of the final sample. The 0.2M HCl places the column at proper acidity for sample retention.

[%]Retention of americium and curium on this 1 cm column is based upon work done with Am^{241}. This indicated that the americium quantitatively adsorbs on the resin in the neighborhood of 0.2-0.3M HCl. At concentrations greater than 0.34M HCl some of the sample starts to elute because of the large volume of load solution (115-135 ml). It has been found that 10 ml of 2M HCl will begin elution of the sample.

7. Elute the sample into an acid-leached 125-ml flask using 20 ml 8M HNO_3.[§#]

8. Add 12 drops H_2SO_4 and boil to SO_3 fumes. Fume for 5 minutes on a low hot plate. Transfer to an electrolysis cell with water washes. Add one drop methyl red indicator and add NH_4OH dropwise until the solution just turns yellow. Add 1.5M H_2SO_4 dropwise until the solution just turns red. Then add one more drop of 1.5M H_2SO_4. Electrodeposit for at least one hour at 0.6 amp. Make solution basic with NH_4OH before disconnecting current. Disassemble cell and flame platinum disc.

[§]It has been found necessary to use extremely clean reagents and equipment throughout this procedure in order to obtain clean plates. The 8M HNO_3 should be with distilled HNO_3 and triple distilled water. The flask should be leached or boiled in HNO_3 to remove dirt, iron, manganese, and other contaminants.

[#]The 8M HNO_3 elutes the americium and curium very rapidly, and about 92% is eluted in 15 ml.

STRONTIUM PURIFICATION PROCEDURES
A AND B

McClellan Central Laboratory
(MCL-LP-07-38)

(Ed. Note: Previously published as
Strontium Purification, McClellan Central Laboratory
Procedure MCL-LP-07-38 (1975).)

INTRODUCTION

The chemist should be familiar with the following information concerning strontium chemistry before continuing with the procedures in this Laboratory Procedure.

1. Major contaminants of strontium are barium and calcium.

   a. Barium is primarily removed by the chromate scavenging of the acetate-buffered solution.

   b. Milligram amounts of calcium can be removed by precipitating strontium nitrate from 65% nitric acid solution.

   --NOTE--

   Increasing the strength of the nitric acid solution will cause increased carrying of the calcium by the strontium nitrate. Refer to Sunderman, D. N. Townley, C. W., The Radiochemistry of Barium, Calcium, and Strontium, National Academy of Sciences Publication NAS-NS-3010. 1960.

2. A final separation of strontium from yttrium is accomplished with an iron scavenge, and the time is recorded as the STRONTIUM-YTTRIUM SEPARATION TIME. The strontium must be mounted as soon after the separation time as possible to prevent any loss in the yttrium daughter caused by the yttrium not following the strontium sulfate. (Refer to MCL Technical Memo: Sr-60, 7 March 1956.)

3. The chemical properties of strontium used in purifying samples are as follows:

   a. Strontium nitrate is insoluble in 65% HNO_3.

   b. Strontium carbonate is insoluble in a basic media.

   c. Strontium sulfate is insoluble in 6M HCl.

   d. Strontium chromate is soluble in an acetate-buffered media.

e. Strontium samples are further purified by the use of $Fe(OH)_3$ scavenge.

f. The final mount form of the sample is $SrSO_4$.

4. There are two purification procedures for strontium, A and B.

a. Procedure A begins with an aliquot of the solution to be analyzed.

b. Procedure B is a rerun procedure if the sample from procedure A shows any form of contamination.

PROCEDURE A

STRONTIUM PURIFICATION

The recovery of strontium from the contaminated aliquot is accomplished in the following manner. Refer to Table 1 for the applicable reagents and see Figure 1 for the procedure flow diagram.

Table 1. Procedure A Reagents

REAGENT	REAGENT
Ba (scavenge); 50 mg/ml	1.5M Na_2CrO_4
6M HCl	Fe (scavenge); 5 mg/ml
$(NH_4)_2CO_3$ (saturated)	1 : 1 Ethanol - H_2O (v/v)
NH_4Ac-HAc Buffer (pH 5.4)	fHNO_3 (90%, fuming nitric acid)
NH_4OH (carbonate-free)	
1.5M H_2SO_4	

1. Aliquot the sample and Sr carrier into a 40 ml glass centrifuge tube, heat, and add 3 drops of Ba scavenge. Stir the solution and let it stand for 10 minutes. Quadruple the volume with fHNO_3, and cool in an ice bath for 30 minutes with occasional stirring. Centrifuge and discard the supernate.

--NOTE--

If milligram amounts of Ca are present, adjust the solution to 65% nitric acid concentration.

2. Dissolve the precipitate in 15 ml H_2O, add 2 drops Fe scavenge, and precipate $Fe(OH)_3$ with CARBONATE-FREE NH_4OH. Centrifuge and decant the supernate into a clean 40 ml tube. Add 5 ml saturated $(NH_4)_2CO_3$ solution to the supernate to precipitate $SrCO_3$ and $BaCO_3$. Digest the solution in a hot bath for 5 minutes. Centrifuge and discard the supernate.

3. Dissolve the precipitate in 5 ml 6M HCl and boil the solution to remove the carbonate from the solution. Perform an Fe scavenge using CARBONATE-FREE NH_4OH. Centrifuge and decant the supernate into a clean 40 ml tube.

4. Add 1 drop methyl red indicator and add 6M HCl until the solution just turns red. Add 3 ml, 5.4 pH, NH_4Ac-HAc buffer solution. Add 1 ml of

1.5M Na_2CrO_4. Digest for 5 minutes in a HOT bath, centrifuge, and decant the supernate into a 40 ml tube.

5. Add 3 drops Ba scavenge and repeat the chromate precipitation. Centrifuge and decant the supernate into a 40 ml tube.

6. Perform an Fe scavenge, using CARBONATE-FREE NH_4OH, and digest until the $Fe(OH)_3$ coagulates. RECORD THIS TIME AS THE STRONTIUM-YTTRIUM SEPARATION TIME. Filter the supernate through a Whatman No. 42 filter paper into a clean 40 ml tube.

--NOTE--

Once the Sr/Y separation time has been taken, the procedure should be completed AS SOON AS POSSIBLE.

7. Add 5 ml of NH_4OH and 2 ml of saturated $(NH_4)_2CO_3$ solution. Stir well and digest in a hot bath until the precipitate coagulates. Centrifuge and discard the supernate. Wash the pricipitate with 5 ml H_2O. Centrifuge and discard the wash solution.

8. Dissolve the precipitate in 2 ml 6M HCl and a few drops of H_2O, heat, and add 1 ml of 1.5 M H_2SO_4. Digest the solution in a hot bath until the precipitate coagulates. Centrifuge and discard the supernate. Wash the precipitate with 10 ml of a 1:1 mixture of ethanol and H_2O. Centrifuge and discard the wash. Slurry the precipitate onto a filter disc with ethanol. Ignite the $SrSO_4$ at 600°C for 30 minutes. Grind the precipitate to a fine consistency and wash it onto a tared filter disc with absolute ethanol. Use a back-up disc. Dry, weigh, and mount.

--NOTE--

$SrSO_4$ is relatively soluble; the volume must not exceed these limits. Absolute ethanol must be used to eliminate the presence of H_2O after the ignition of the $SrSO_4$.

Strontium carrier + activity + Ba scavenge (Para 1)

Precipitate $Sr(NO_3)_2$ and $Ba(NO_3)_2$ with $fHNO_3$ (Para 1)

Scavenge with $Fe(OH)_3$ precipitated with NH_4OH (Para 2)

Precipitate $SrCO_3$ and $BaCO_3$ with saturated $(NH_4)_2CO_3$ (Para 2)

Scavenge with $Fe(OH)_3$ precipitated with NH_4OH (Para 3)

Scavenge for Ba twice by precipitating $BaCrO_4$ (Para 4 and 5)

Scavenge with $Fe(OH)_3$ precipitated with NH_4OH(Sr/Y separation time) (Para 6)

Precipitate $SrCO_3$ with saturated $(NH_4)_2CO_3$ (Para 7)

Precipitate $SrSO_4$ with $1.5\underline{M}$ H_2SO_4 (Para 8)

Wash with 1 : 1 ethanol-H_2O (Para 8)

Ignite $SrSO_4$ at $600^{o}C$ for 30 minutes (Para 8)

Dry, weigh, and mount (Para 8)

Figure 1. Flow Diagram for Procedure A

PROCEDURE B

STRONTIUM RERUN

Further purification of strontium samples from procedure A is accomplished in the following manner. See Figure 1 for the flow diagram.

1. Disassemble the strontium sample from procedure A and dissolve the sample and filter paper with 5 ml HNO_3 and 3 ml $HClO_4$.

2. Reduce the volume to 2 ml and cool.

3. Proceed to the appropriate step of procedure A.

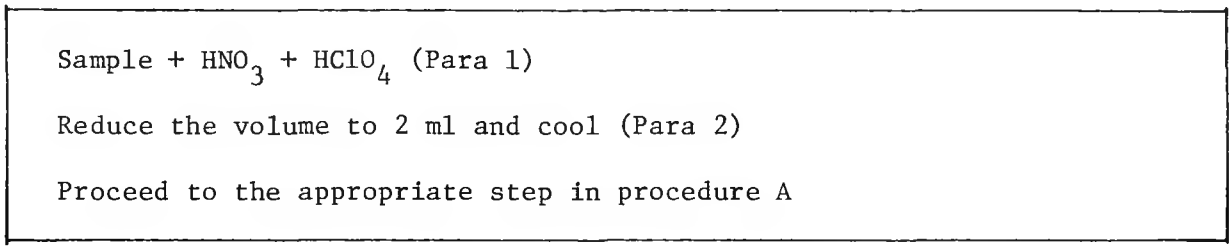

Figure 1. Flow Diagram for Procedure B

CESIUM PURIFICATION PROCEDURES A AND B

McClellan Central Laboratory
(MCL-LP-07-55)

(Ed. Note: Previously published as
Cesium Purification, McClellan Central Laboratory
Procedure MCL-LP-07-55 (1971).)

INTRODUCTION

The chemical properties of cesium which are used in the purification of samples include the insolubility of the chloroplatinate in 6M HCl, the insolubility of the perchlorate in ethanol, and the insolubility of the silicotungstate in 6M HCl. Additional purification is obtained by performing an iron scavenge.

Two procedures are available for purifying cesium. Procedure A begins with an aliquot of the solution to be analyzed. Procedure B is used when the sample is old enough so that all ^{86}Rb has decayed or when the cesium has been partially purified by elution from a cation exchange resin column with α-hydroxy isobutyric acid (HIBA). The only difference between these two procedures is that it is not necessary to repeat the precipitation of cesium silicotungstate in procedure B because the HIBA column provides decontamination against ^{86}Rb. Rhodium is co-eluted with cesium from the HIBA column but no special decontamination against rhodium is necessary because of the vast difference between the chemistry of cesium and that of rhodium.

PROCEDURE A

CESIUM (FROM ALIQUOT)

REAGENTS

6M HCl | 0.12M Silicotungstic Acid | 6M NaOH

10% H_2PtCl_6 | Fe Scavenge (5 mg/ml)

FLOW DIAGRAM

Cesium carrier + activity

Precipitate Cs_2PtCl_6 with 10% H_2PtCl_6

Precipitate $CsClO_4$ with absolute ethanol

Scavenge with $Fe(OH)_3$ precipitated with NaOH

Precipitate cesium silicotungstate twice with silicotungstic acid
[1,2]
Precipitate tungstic acid
[2]
Precipitate Cs_2PtCl_6 with 10% H_2PtCl_6

Dry at 110°C, mount and yield as Cs_2PtCl_6

PROCEDURE

1. Add 10 ml each HNO_3 and $HClO_4$ to the sample activity and cesium carrier in a 125-ml flask. Evaporate to dryness and bake the residue to remove the excess $HClO_4$.

2. Cool the flask and add 15 ml 6M HCl. Reduce the volume to 3 ml by boiling and transfer the solution to a 40-ml glass centrifuge tube with 6M HCl washes.

3. Add 1 ml 10% H_2PtCl_6 and stir well. It may be necessary to induce precipitation by lightly scratching the wall of the tube with a stirring rod. Allow to stand several minutes. Centrifuge and discard the supernate.

4. Wash the Cs_2PtCl_6 precipitate with 6M HCl. Centrifuge and discard the wash. Dissolve the precipitate in 3 ml HNO_3 and 3 ml $HClO_4$. Boil to reduce the volume to 3 ml[a/] and cool in an ice bath[b/].

---

a/ The dense white fumes of perchloric acid must be visible to insure that all HNO_3 has been volatilized.

b/ The sample must be cooled before adding ethanol. Explosive mixtures are formed when hot!!

5. Add absolute ethanol to obtain a final volume of 30 ml. Stir well and chill in an ice bath for 15 minutes. Centrifuge and discard the supernate.

6. Dissolve the precipitate in 5 ml H_2O and a few drops HNO_3 (heat if necessary). Add 3 dps Fe scavenge and precipitate $Fe(OH)_3$ with 6M NaOH[a]. Digest in a hot bath and filter into a clean tube through a Whatman #42 filter paper. Double the volume with HCl.

7. Add 1 ml 0.12M silicotungstic acid, stir, and digest for 5 minutes in a hot bath. Cool for 10 minutes in an ice bath. Centrifuge and discard the supernate.

8. Add 2 ml 6M NaOH, 2 ml H_2O, and boil to dissolve the precipitate. Add 10 ml 6M HCl and digest the sample in a hot bath with frequent stirring to precipitate the excess tungstic acid. The sample should be left in the hot bath until the precipitate coagulates and settles to the bottom of the tube. Centrifuge and decant the supernate into a clean tube. Add 5 ml 6M HCl to the excess tungstic acid precipitate and boil vigorously. Centrifuge and combine the wash with the supernate containing the cesium.

9. Repeat steps 7 and 8; then proceed to step 10[b].

10. Filter the final solution into a clean tube through a Whatman #42 filter paper. Add 1 ml 10% H_2PtCl_6 and stir well. It may be necessary to induce precipitation by lightly scratching the sides of the tube with a stirring rod. Allow the solution to stand for 3-5 minutes. Centrifuge and discard the supernate.

11. Add 2-3 ml H_2O and 2-3 ml 6M NaOH and bring the solution just to a boil[c]. Immediately centrifuge and discard the wash. Wash the precipitate with 10 ml 6M HCl. Centrifuge and discard the wash. Wash the precipitate with 10 ml ethanol. Centrifuge and discard the wash.

12. Transfer the precipitate to a tared filter paper with ethanol. Dry, weigh, and mount.

RE-RUN PROCEDURE

To re-run the sample, dissolve the filter paper and Cs_2PtCl_6 with 5 ml HNO_3 and 5 ml $HClO_4$. Reduce the volume to 3 ml and cool in an ice bath. Perform steps 5 through 12.

---

[a] If by mistake NH_4OH is used for the iron scavenge, add NaOH and boil the solution until all of the NH_3 has been expelled (i.e., fumes no longer turn litmus paper blue).

[b] In the analysis of old activity samples this step is not necessary, since the contribution of ^{86}Rb is usually negligible.

[c] Vigorous boiling will result in yield losses.

PROCEDURE B

CESIUM (HIBA COLUMN)

CO-ELUTION AND PRELIMINARY SEPARATION

Only rhodium is known to co-elute with cesium. Cesium is precipitated directly as the chloroplatinate and the supernate used for the rhodium separation.

REAGENTS

6M HCl | 6M NaOH | 0.12M Silicotungstic Acid

10% H_2PtCl_6 | Fe Scavenge (5 mg/ml)

FLOW DIAGRAM

Cs_2PtCl_6

Precipitate $CsClO_4$ with absolute alcohol

Scavenge with $Fe(OH)_3$ precipitated with NaOH

Precipitate cesium silicotungstic with silicotungstic acid

Precipitate tungstic acid

Precipitate Cs_2PtCl_6 with 10% H_2PtCl_6

Dry at 110°C, weigh and mount as Cs_2PtCl_6

PROCEDURE

1. Wash the Cs_2PtCl_6 precipitate with 6m HCl. Centrifuge and discard the wash. Dissolve the precipitate in 5 ml HNO_3 and 5 ml $HClO_4$. Boil to reduce the volume to 3 ml[a/] and cool in an ice bath[b/].

2. Add absolute ethanol to obtain a final volume of 30 ml. Stir well and chill in an ice bath for 15 minutes. Centrifuge and discard the supernate.

3. Dissolve the precipitate in 5 ml H_2O and a few drops HNO_3 (heat if necessary). Add 3 dps Fe scavenge and precipitate $Fe(OH)_3$ with 6M

---

a/ The dense white fumes of perchloric acid must be visible to insure that all HNO_3 has been volatilized.

b/ The sample must be cooled before adding ethanol. Explosive mixtures are formed when hot!!

NaOH[a]. Digest in a hot bath and filter the solution into a clean tube through a Whatman #42 filter paper. Double the volume with HCl.

4. Add 1 ml 0.12M silicotungstic acid, stir, and digest for 5 minutes in a hot bath. Cool for 10 minutes in an ice bath. Centrifuge and discard the supernate.

5. Add 2 ml 6M NaOH, 2 ml H_2O, and boil to dissolve the precipitate. Add 10 ml 6M HCl and digest the sample in a hot bath with frequent stirring to precipitate the excess tungstic acid. The sample should be left in the hot bath until the precipitate coagulates and settles to the bottom of the tube. Centrifuge and decant the supernate into a clean tube. Add 5 ml 6M HCl to the excess tungstic acid precipitate and boil vigorously. Centrifuge and combine the wash with the supernate containing the cesium.

6. Filter the final solution into a clean tube through a Whatman #42 filter paper. Add 1 ml 10% H_2PtCl_6 and stir well. It may be necessary to induce precipitation by lightly scratching the sides of the tube with a stirring rod. Allow the solution to stand for 3-5 minutes. Centrifuge and discard the supernate.

7. Add 2-3 ml H_2O and 2-3 ml 6M NaOH and bring the solution just to a boil[b]. Immediately centrifuge and discard the wash. Wash the precipitate with 10 ml 6M HCl. Centrifuge and discard the wash. Wash the pricipitate with 10 ml ethanol. Centrifuge and discard the wash.

8. Transfer the precipitate to a tared filter paper with ethanol. Dry, weigh, and mount.

---

[a] If by mistake NH_4OH is used for the iron scavenge, add NaOH and boil the solution until all of the NH_3 has been expelled (i.e., fumes no longer turn ligmus paper blue).

[b] Vigorous boiling will result in yield losses.

SEPARATION PROCEDURE FOR THE ELEMENTS
Sr, Ce, Cs,and Pu FROM NONCORALLINE SOIL

McClellan Central Laboratory
(MCL-LP-06-S-02)

INTRODUCTION

1. Two major problems are encountered in determining the content of radiospecies where the gross activity level is on the order of 10^{12} to 10^{10} curie/kilogram.

   a. First, the species of interest must be quantitatively leached from gram quantities of soil with minimal co-leaching of indigenous species which in large quantities would tend to complicate further analysis.

   b. Secondly, the isotopes of interest must be selectively separated from those interfering species which are co-leached.

2. Varying quantities of indigenous strontium and cerium are also leached, and must be corrected for via instrumental analysis. A typical 500 gram soil sample would contain interfering species in the amounts shown in Table 1.

3. Procedure S-02

4. RECOVERY PROCEDURES

   a. STRONTIUM. The recovery of strontium from soil is complicated by carbonate forming species, especially calcium. Calcium in concentrations of approximately 20 grams per 500 grams of soil can be quantitatively leached, and follows strontium throughout the separation. During the procedure, calcium is removed first by conversion to soluble bicarbonate, followed by a series of fuming nitric acid steps where $Sr(NO_3)_2$ is selectively precipitated. In some cases 700 milliliters per sample of HNO_3 is required to remove 95 percent of the interfering calcium. Indigenous strontium in soil is also leached and must be accounted for via instrumental analysis to correct gravimetric yields.

   b. CERIUM. The recovery of cerium from soil is complicated by hydroxide forming species, principally Ti, Al, and Mg. These elements are found in gram quantities per 500 grams in most soil and tend to follow cerium throughout the separation procedure. A procedure involving co-precipitation of cerium with lanthanum oxalate at pH 5.4 has been developed and used with success. The added lanthanum negated the effect of normal interferences. Iron, which presents a problem in cerium purification, may be removed by conducting numerous diethyl ether extractions. Indigenous cerium is also leached and must be corrected for.

Table 1. Typical 500 Gram Soil Sample

INTERFERING SPECIES	AMOUNT (in grams)
Aluminum	40.0
Iron	25.0
Calcium	18.0
Sodium	14.3
Potassium	13.0
Magnesium	10.5
Titanium	3.2
Strontium	0.09
Cerium	0.04

c. CESIUM. The recovery of cesium from soil is complicated by gram quantities of leachable alkali species having similar chemistry to cesium and the relatively high solubilities of most cesium compounds. To alleviate these problems, cesium can be co-precipitated with gram quantities of ammonium hexa-chloroplantinate. It has been stated in the publication "Chemical Analysis of Radioactive Materials" (1967), pp 271, that heretofore radioactive cesium could not be recovered from soil without complete destruction of the silicate matrices. Complete destruction of the silicate matrix, however, has been shown to introduce inert contaminants in such quantities as to prohibit further analysis of ordinary soils where the activity level is on the order of 10^{12} to 10^{10} curie/kilogram. Moreover, complete destruction of the silicate matrix has been found to be unnecessary.

d. PLUTONIUM. No major problems should be encountered during the recovery procedures of plutonium.

Table 2. Procedure S-02 Reagents

HBr	10M HNO_3	$LaCl_3$ (5 mg/ml)
HCl	4M HNO_3	H_2PtCl_6
HNO_3	pH 5.4 Buffer	$(NH_4)_2CO_3$ (Saturated)
$HClO_4$	H_2O_2 (30%)	NH_4I (Saturated)
HF	NH_4OH	EDTA (NH_4^+ form, 40 mg/ml)
6M HCl	Diethylether	

5. LEACH PROCEDURES

--SAFETY PRECAUTION--

Use EXTREME CARE when performing paragraph 5a(1) as 30% H_2O_2 will cause blistering of the skin. Irritation caused by H_2O_2 which does not subside upon flushing with water, SHOULD BE TREATED BY A PHYSICIAN.

--NOTE--

For further details on the toxicology of hydrogen peroxide see Sax, Dangerous Properties of Industrial Materials, Reinhold Publishing Corp., New York, 1963, pp 886-887.

a. PREOXIDATION

(1) Add 20 milliliters of H_2O_2, 50 milliliters of NH_4OH, and 200 milliters of H_2O to a 3000-milliliter glass beaker containing 500 grams of the soil to be analyzed.

(2) Stir well for 10 minutes and allow the solids to settle.

(3) Transfer the turbid supernate into several 40-milliliter tubes and centrifuge. Decant the supernates into a clean 2000-milliliter glass beaker. Wash any precipitate back into the original 3000-milliliter beaker using H_2O.

(4) Add an additional 150 milliliters of hot H_2O to the original beaker and again stir for 10 minutes.

(5) Allow the solids to settle, transfer the supernate into several 40-milliliter tubes and centrifuge. Decant the supernates into the 2000-milliliter glass beaker containing the initial wash supernates (refer to paragraph 5a(3)) and boil to dryness. Retain this beaker for use in paragraph 5b(3).

(6) Wash any precipitate remaining, into the original beaker (refer to paragraph 5a(3)) using H_2O. Retain this beaker for use in paragraph 5b(1).

b. COMPLEX FORMATION AND REMOVAL

(1) Add 200 milliliters of EDTA to the 3000-milliliter beaker containing the soil (retained in paragraph 5a(6)) and stir well for 10 minutes.

(2) Allow the soil to settle for one hour and transfer the supernate into several 40-milliliter tuhes.

(3) Centrifuge and decant the supernates into the 2000-milliliter beaker held in paragraph 5a(5).

(4) Wash any precipitate back into the original beaker using H_2O.

(5) Wash the soil with two 100-milliliter portions of hot H_2O to remove all EDTA; centrifuge; and add the wash supernates to the 2000-milliliter beaker.

(6) Boil the contents of the 2000-milliliter beaker to reduce the volume to approximately 200 milliliters, and perform a series of aqua salt destructions until frothing is minimized.

(7) Retain the 2000-milliliter beaker for use in paragraph 5c(4); retain the 3000-milliliter beaker for use in paragraph 5c(1).

c. AQUA REGIA/HF LEACH

(1) Place the 3000-milliliter beaker containing the soil residue in an ethanol/ice bath and carefully add 100 milliliters of HCl, and 100 milliliters of HNO_3.

--NOTE--

If severe frothing occurs, add 5 to 10 milliliters of aerosol solution.

--SAFETY PRECAUTION--

When performing the following procedure, handle the hydrofluoric acid with extreme caution. This acid will cause severe and very painful skin burns if it contacts the skin. Immediate medical attention is required if HF is splashed onto the skin or into the eyes. A temporary first-aid measure involves washing the affected area with copious amounts of H_2O and Hyamine 1622. Gloves will be worn during all operations involving the use of hydrofluoric acid.

--NOTE--

For further details on the toxicology of HF, refer to Sax, Dangerous Properties of Industrial Materials, Reinhold Publishing Corp., New York, 1963, pp 884-885.

(2) After frothing ceases add 10 milliliters of HF and remove the beaker from the ice bath and place on an electric hot plate (low setting).

(3) Boil the contents of the beaker for five minutes. Cool, allow the residue to settle, and transfer the supernate into several 40-milliliter tubes.

(4) Centrifuge and decant the supernates into the 2000-milliliter beaker retained in paragraph 5b(7).

(5) Wash any precipitate back into the original 3000-milliliter beaker using H_2O.

(6) Perform one additional aqua regia leach, and wash the soil residue with 100 milliliters of H_2O; centrifuge; and add the wash supernates to the 2000-milliliter beaker.

--NOTE--

An ice bath is not necessary when performing the second aqua regia leach.

(7) Retain the 3000-milliliter beaker for use in paragraph 5d(1). Retain the 2000-milliliter beaker for use in paragraph 5d(4).

d. HBr/HCl LEACH

--SAFETY PRECAUTION--

When using gaseous bromine, in paragraph 5d(1), use EXTREME CAUTION as this chemical acts as an irritant to the mucous membranes of the eyes and the upper respiratory tract.

--NOTE--

For further details on the toxicology of bromine refer to Sax, Dangerous Properties of Industrial Materials, Reinhold Publishing Corp., New York, 1963, pp 525.

(1) Add 40 milliliters of HBr and 100 milliliters of HCl to the soil residue in the original 3000-milliliter beaker with constant stirring.

(2) Carefully heat the beaker contents on an electric hot plate for ten minutes or until boiling occurs.

(3) Remove the beaker from the hot plate, allow the soil residue to settle, and transfer the supernate into several 40-milliliter tubes.

(4) Centrifuge and decant the supernates into the 2000-milliliter beaker retained in paragraph 5c(7).

(5) Wash the soil residue with 200 milliliters of hot H_2O; centrifuge; and add the wash supernates to the 2000-milliliter beaker.

(6) Discard the 3000-milliliter beaker containing the soil residue. Retain the 2000-milliliter beaker for use in paragraph 5e(1).

e. FINAL LEACH

--SAFETY PRECAUTION--

If violent frothing occurs during the initial perchloric reaction (paragraph 5e(1)), IMMEDIATELY remove the beaker from the hot plate and add approximately 50 milliliters of nitric acid to control the reaction. If this procedure is not adhered to a violent explosion may result.

(1) Add 100 milliliters of $HClO_4$ and 200 milliliters of HNO_3 to the 2000-milliliter beaker and boil to reduce the volume to approximately 80 milliliters.

--NOTE--

When HNO_3 is added to the solution Br will be evolved. Ensure that you are familiar with the safety precaution preceding paragraph 5d(1).

(2) Cool the contents of the beaker, and then transfer the contents into 1000-milliliter volumetric flasks. Reduce the volume to 1000 milliliters using H_2O. This solution constitutes the basic working stock.

6. SEPARATION PROCEDURES

a. GENERAL

(1) Aliquot 500 milliliters of the basic working stock (paragraphs 5e(1) and (2)) to a 1500-milliliter beaker containing the carriers and tracer of interest.

--NOTE--

Ba (50 milligrams) is added to insure a more quantitative separation of Sr.

(2) Add 50 milliliters of $HClO_4$ boil the solution until dense fumes begin to evolve.

(3) Cool the solution and add 100 milliliters of H_2O and 5 drops of saturated NH_4I.

--NOTE--

Ammonium iodide is required to reduce Pu (VI), Pu (V), and Pu (IV) to Pu (III) for purposes of equilibration. Equilibration is not required for tracer-free Pu.

(4) Boil the solution to dryness.

(5) Cool the solution and add 100 milliliters of HNO_3. Boil the solution to dryness.

(6) Transfer the beaker contents to approximately eight 40-milliliter tubes using 10 milliliters of 4M HNO_3. Retain these tubes for use in paragraph 6b(1).

(7) Retain the original beaker for use in paragraph 6b(1).

b. STRONTIUM

(1) Add 35 milliliters of fuming HNO_3 to each of the tubes retained in paragraph 6a(6); place the tubes in an ice bath for 20 minutes; centrifuge; and decant the supernates into the beaker retained in paragraph 6a(7).

(2) Combine the precipitate using approximately 20 milliliters of H_2O. Make basic with carbonate free NH_4OH.

(3) Centrifuge and decant the supernate into a clean 40-milliliter tube.

(4) Return the precipitate back to the original beaker. Hold for paragraph 6c(1).

(5) Add 20 milliliters of saturated $(NH_4)_2CO_3$ to the supernate and place in a hot water bath. The white precipitate consists of strontium (and barium) carbonate.

--NOTE--

If the precipitate contains excessive Ca, repeat the fHNO$_3$ precipitation.

(6) Continue with the strontium purification procedure, MCL-LP-07-38.

c. PLUTONIUM

(1) Boil the contents of the beaker retained in paragraph 6b(4) to dryness.

(2) Transfer the contents of the beaker to four 40-milliliter tubes using approximately 35 milliliters of 10M HNO_3 in each tube.

(3) Centrifuge and load the supernates on a pre-equilibrated nitrate column.

--NOTE--

Utilize Dowex 1 x 8, 100-200 mesh, 2 cm x 8 mm i.d., nitrate form, pre-equilibrated with 2 column volumes of 10M HNO_3.

(4) Return the precipitates to the original beaker using 6M HCl. Collect the effluent and return to the original beaker.

(5) Wash the column with 10 milliliters of 10M HNO_3 and add the wash effluent to the original beaker. Retain the beaker for use in paragraph 6d(1).

(6) Wash the column with 20 milliliters of H_2O and collect the effluent in a 125-milliliter Erlenmeyer flask.

(7) Add 5 drops of saturated NH_4I and 10 milliliters of 5M HCl to the column followed by a 10 milliliter 5M HCl wash.

(8) Collect both effluents in the 125-milliliter flask used in paragraph 6c(6). This flask contains a solution of plutonium chloride.

(9) Continue with the Pu purification procedure, MCL-LP-07-94.

d. IRON REMOVAL

(1) Place the beaker retained in paragraph 6c(5) on a hot plate and boil to dryness.

(2) Cool the contents, wash down the beaker walls with 40 milliliters of HCl and 2 milliliters of HBr, and boil to absolute dryness.

(3) Cool the contents, and transfer the contents of the beaker to a 500-milliliter separatory funnel using 100 milliliters of 6M HCl.

(4) Cool the funnel in an ice bath.

(5) Add 200 milliliters of anhydrous diethyl ether and mix thoroughly for 2 to 3 minutes. Discard the ether (top) layers containing Fe.

(6) Repeat the ether extractions until all of the yellow color (Fe) is removed from the 6M HCl layers.

(7) Add the HCl layer to a clean 500-milliliter beaker and hold for paragraph 6e(1).

e. CESIUM

(1) Add 10 boiling beads to the beaker retained in paragraph 6d(7).

--SAFETY PRECAUTION--

The ether burn-off, required in paragraph 6e(2), is necessary to prevent the formation of combustible deposits in the fume hood ducting. The burn-off must be performed with EXTREME CAUTION, and must be executed in an operating fume hood.

(2) Place the beaker on an electric hot plate (low setting) and ignite the ether vapors being evolved.

(3) Boil to reduce the volume to approximately 60 milliliters, and transfer the contents into two 40-milliliter tubes.

(4) Cool in an ice bath for 5 minutes and centrifuge.

(5) Add 2 drops of NH_4OH to each tube and decant the supernate into two clean 40-milliliter tubes. Discard any precipitate.

(6) Add 7 drops of 10 percent H_2PtCl_6 to each supernate; stir vigorously; and place in an ice bath for 5 minutes.

(7) Centrifuge and decant the supernates into four clean 40-milliliter tubes. Retain the supernates for use in paragraph 6f(1). The yellow precipitate consists of cesium (I) hexachloroplantinate, Cs_2PtCl_6 and $(NH_4)_2PtCl_6$.

(8) Continue with the Cs purification procedure, MCL-LP-07-55.

f. CERIUM

(1) Make the supernates, retained in paragraph 6e(7), basic with NH_4OH.

--NOTE--

Add NH_4OH until a white precipitate begins to form.

(2) Add 10 milliliters of 5.4 pH buffer and 1 milliliter of lanthanum chloride solution (5 mg/ml) to each 40-milliliter tube.

(3) Add 1 milliliter of saturated oxalic acid to each 40-milliliter tube and place in hot water bath. Centrifuge and combine the precipitates.

(4) The precipitate consists of cerium oxalate and lanthanum oxalate.

(5) Continue with the Ce purification procedure, MCL-LP-07-57.

DETERMINATION OF ^{241}AM IN SOIL USING AN AUTOMATED NUCLEAR RADIATION MEASUREMENT LABORATORY

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ABSTRACT

The recent completion of REECo's Automated Laboratory and associated software systems has provided a significant increase in capability while reducing manpower requirements. The system is designed to perform gamma spectrum analyses on the large numbers of samples required by the current Nevada Applied Ecology Group (NAEG) and Plutonium Distribution Inventory Program (PDIP) soil sampling programs while maintaining sufficient sensitivities as defined by earlier investigations of the same type. The hardware and systems are generally described in this paper, with emphasis being placed on spectrum reduction and the calibration procedures used for soil samples.

INTRODUCTION

This computerized system was developed during the years 1971 through 1973, with final debugging taking place in the spring and summer of 1974. Its function is to automatically control the laboratory instruments and to acquire and assemble data from these instruments on computer-compatible magnetic tape. The magnetic tape is then submitted to the AEC's remote Mercury terminal for transmittal over telephone lines to the NV computer center in Las Vegas.

At the Las Vegas computer center, the data is processed and stored in various tape files for future use. Results are transmitted back to the Mercury terminal, where reports are printed and delivered to the Environmental Sciences Department.

Although the laboratory system was operational in 1973, its full benefits were not experienced until spectrum reduction programs could be developed and existing software systems could be modified to accept the additional

spectrum results for each sample. With this phase completed, the nuclear instrumentation is kept busy 24 hr a day, 7 days a week. One technician is required on a half-time basis to load samples in the sample changers and to enter the required data into the computer system.

METHODS

Samples are collected on the test site and transported to the laboratory for analysis. At the laboratory, material for gamma analysis is placed in appropriate containers (usually 16-oz Nalgene bottles) and submitted directly to the Nuclear Radiation Measurements Laboratory (NRML). Data describing the material and where it was collected is transported to the laboratory with the sample. This information is entered directly into the computer system (through a laboratory keyboard terminal) or is punched on an IBM card.

Once a sample has been loaded into a sample changer in the NRML, and the various counting data entered (counting time, calibration factors and background), the counting procedure and data handling is automatic. The only human function is the structuring of computer runs and the transporting of data to the Mercury Remote Terminal.

At an intermediate processing point, an analyst reviews the data for possible errors and anomalies caused by machine malfunctions, unresolvable mixtures of isotopes, and insufficient data. The analyst uses knowledge of sample history, not available to the computer, to identify erroneous results and make the necessary alterations. When this data has been edited, it is resubmitted to the remote terminal for the final processing stage of the "Daily Report System."

All data submitted to the central computer passes through the hands of the job stream assembler. This technician actually programs the computer system by assembling a job stream with control cards. These cards direct the computer in properly applying the various programs (approximately 30 in number) to the data input of the various jobs.

AUTOMATED LABORATORY HARDWARE

The hardware system is presently comprised of a computer which controls and accepts data from four pulse-height analyzers, two alpha-beta counters, one gross-gamma counter, one liquid-scintillation spectrometer, and one analogue-to-digital converter.

A. PDP 11/20 Computer

This computer is the heart of the Automated Laboratory System (Figs. 1 and 2). Its function is to control the various nuclear instruments, accept data from those instruments and its manual input terminals, and assemble this data on industry-compatible magnetic tape.

The computer consists of a 16,384-word memory with a word length of 16 bits (16,384 x 16). It can also use the memory of a Canberra Model 8700 analyzer with 4,096 words as extended memory.

The data bus (UNI-BUS) of the computer connects the various pieces of peripheral equipment and instrumentation interfaces. The UNI-BUS is extended through one bus repeater.

B. Multichannel Pulse Height Analyzers

These instruments are used to accumulate spectrum data for the identification and quantitation of alpha- and gamma-emitting radionuclides. Their proper operation is routinely checked by the use of calibration source measurements, background measurements, and by monitoring the quality of data output before and after computer processing. Analyses for ^{241}Am in soil are accomplished with the use of germanium (GeLi) detector systems (Fig. 3).

Instruments presently used in the Automated Laboratory are:

1. GeoScience Model 7000 processor and Model 4000 two-parameter formater and display. Memory capacity is 4,096 x 20 binary or BCP. This is a dual input system with independent time base. Each of the two ADCs has a 100-mHZ clock and is automatically stabilized with a precision pulser. The system is presently used with a germanium detector of 16% efficiency and 37/1 peak to Compton ratio. This detector is mounted in a 25,000-lb lead shield and sample changer assembly. The changer will accommodate a maximum of 17 samples of varying size. The analyzer is equipped with a high-speed Franklin printer and a Moseley plotter for "off-line" use.

2. TMC analyzer with Model 438 display-plot control, 222 dual live timer, 437 control unit, 440 memory. Memory size is 4,096 x 20 BCD. This is a single-input configuration using a GEOS Model 8050 ADC which has a 50-mHZ clock and is automatically stabilized with a GEOS 2000 stabilizer and Canberra 8210 precision pulser. The system is presently used with a germanium detector of 12% efficiency and 16/1 peak to Compton ratio. This detector is mounted in a 25,000-lb lead shield and sample changer assembly. The changer will accommodate up to 34 samples contained in 16-oz Nalgene bottles. The analyzer is equipped with a high-speed Franklin printer for "off-line" use.

Figures 1 and 2 PDP 11/20 Computer

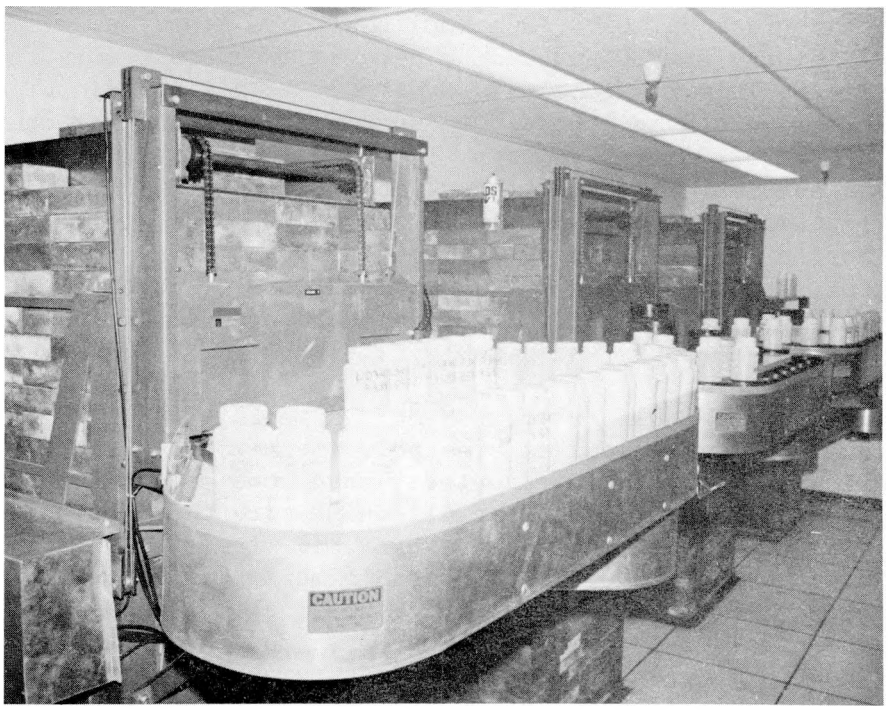

Figure 3 GeLi Detector Systems

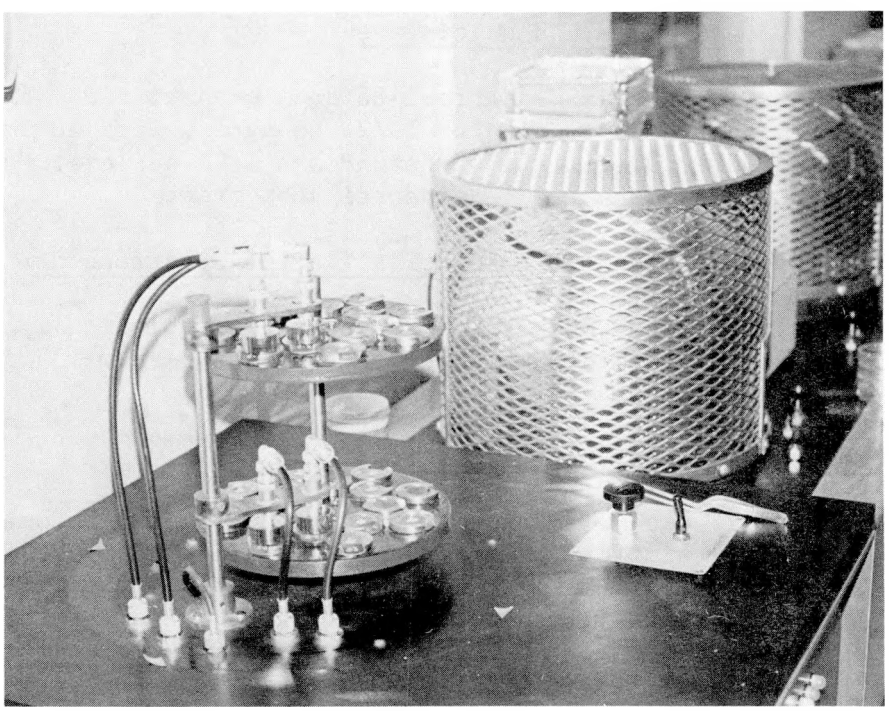

Figure 4 Silicon Detectors for TMC Converter

3. Canberra Model 8700 (Quantum) Analyzer. Memory size is 4,096 x 20 binary or BCD and can be used as extended memory for the PDP 11/20. This instrument is presently in a single-input configuration but can be expanded to up to eight inputs through the use of two 8220 mixer-routers. The 8060 ADC has a 100-mHZ clock and is automatically stabilized with an 8200 stabilizer and 820 precision pulser. The system presently uses a germanium detector of 18% efficiency and 37/1 peak to Compton ratio. The detector is mounted in a 25,000-lb lead shield and sample changer assembly. The changer will accommodate a maximum of 17 samples of varying geometry and size.

4. GEOS Model 7001 Analyzer. Memory size is 1,024 x 20 binary or BCD. This equipment is presently in an eight-channel configuration. Each channel is routed through one of two 2020 mixer-routers and consists of one Ortec 411 pulse stretcher, one Ortec 408 bias amplifier, and one Canberra 816 linear amplifier. The ADC is a GEOS Model 8060. No gain stabilization is employed. This system presently uses eight silicon barrier detectors which are ruggedized and have a 300-square-millimeter window. The detectors are mounted in groups of four in an automatic sample changer. Each group is enclosed with a vacuum bell jar and is mounted on a revolving table that accommodates eight samples (plated discs) for each detector (giving the changer a maximum capacity of 64 samples). A Digi-Data Model 1500 Mag-Tape is used for temporary data storage during "off-line" operation.

C. Canberra Data Scanner and Multiplexer

This system includes a Canberra 1488 data scanner and 1485 multiplexer which are interfaced to the PDP 11/20 computer. The 1485 multiplexer is used to interrogate each of four Model 1491 scalers. Each scaler accepts counting data from a separate subsystem.

D. Nuclear Chicago Mark I Liquid Scintillation Spectrometer

This instrument can count up to 150 samples automatically under computer control or in the "off-line" mode. A cesium-137 source is used for external standardization.

E. TMC Analogue-to-Digital Converter

This device is used with another alpha spectrometry automatic sample changer similar to the one used with the GEOS 7001 Analyzer. The capacity of this changer is 32 samples. It is one-bell-jar assembly with four silicon detectors (Fig. 4). The ADC deposits data directly into the computer memory where spectra are constructed.

GAMMA SPECTRUM REDUCTION SOFTWARE

A. Peak Detection

Peak detection is achieved with a one-pass program which performs the following functions:

1. Defines peak boundaries for truncation and peak area calculations.

2. Determines peak centroid in terms of channel number.

3. Sum counts and calculates dead-time correction.

4. Calculates peak area and percent counting error.

5. Tests peak for proper FULL WIDTH HALF MAX resolution.

Boundaries of a detected peak are defined when two adjacent channel pairs have a positive slope above the 2-sigma error level and again when the first maximum slope is positive and the trailing maximum slope is negative.

Thus: 1st boundary occurs when slope A_{min} GT 0 and slope B_{min} GT 0

2nd boundary occurs when slope A_{max} GT 0 and slope B_{max} LT 0

$$\text{Slope } A_{min} = \text{Chan}_{i+1} - \text{Chan}_i - 2(\text{Chan}_{i+1} + \text{Chan}_i)^{1/2}$$

$$\text{Slope } A_{max} = \text{Chan}_{i+1} - \text{Chan}_i + 2(\text{Chan}_{i+1} + \text{Chan}_i)^{1/2}$$

$$\text{Slope } B_{min} = \text{Chan}_i - \text{Chan}_{i-1} - 2(\text{Chan}_i + \text{Chan}_{i-1})^{1/2}$$

$$\text{Slope } B_{max} = \text{Chan}_i - \text{Chan}_{i-1} + 2(\text{Chan}_i + \text{Chan}_{i-1})^{1/2}$$

For area calculation, peak boundary widths are limited to those widths used in efficiency calculations from standard sample data.

The program uses dynamic smoothing (single-pass smoothing) when channel counts are less than 200. Three-point smoothing is used when counts are less than 100 and two-point smoothing is used between 100 and 200 counts. In this manner, obliteration of peaks is minimized while improving peak detection sensitivity. The pass is made from minimum to maximum channel number.

The peak centroid is determined by using the minimum and maximum breakover points in a symmetry extrapolation. An empirical correction for skewing was added (as a function of resolution).

Dead time is calculated as a function of channel count and channel number. Memory storage time and ADC clock time correspond to those used in the pulse-height analyzers (4 μsec and 100 Mhz).

Peak area is integrated between peak boundaries or boundary limits. Base area is always calculated from the detected boundaries. The calculation of base area is an estimation of background and, therefore, the counting error is an estimation. The base is calculated from a single channel outside each boundary and, therefore, is a linear extrapolation.

Resolution is checked at the FWHM level. If the measured value is less than one-half the stated value or more than twice the stated value, the peak is rejected. Resolution is expressed as a linear function of channel number.

B. Isotope Identification

This program receives peak centroid (peak channel number), peak net counts (peak area minus base area), and percent counting error from the peak detection program. With this information and additional files and tables, the following functions are performed:

1. Subtracts background counts from selected peaks corresponding to those peaks in the background matrix on file.

2. Calculates the energy of the peak centroid.

3. Identifies the isotopes that could produce the peak.

4. Determines probable isotopes and correct energy calibration.

Background spectra are accumulated and the output of the peak detection program is stored in a special background file. This data is then used to subtract background from corresponding peaks in the sample data originating from the same instrument.

Energy of the peak centroid is calculated with a second-order equation and calibration data (energy in centroid channel number) for three widely separated points in the spectrum (usually the peaks for ^{241}Am and ^{22}Na).

Identification of isotopes is accomplished from an isotope library which contains isotope identification, energy of each peak, and branching ratio for each peak. Branching ratio error was not included, since this information does not exist for many of the isotopes in the literature. An error of 5% is assumed for all. An energy window around each peak is defined with input data and is a linear function of channel number (usually 4 Kev wide). Isotopes competing for one peak are evaluated by "rank." The rank of a detected isotope is defined as the sum of the branching ratios of the detected peaks divided by the sum of the branching ratios for all peaks. The isotope having the lower rank is rejected.

When probable isotopes have been identified, an energy correction is calculated in an iterative process which uses the energies of

the assumed isotopes at each centroid to calculate new calibration data. This data is obtained from each detected peak using an average whose elements are weighed inversely to the distance between centroid and calibration point. If the correction exceeds 0.3%, the process is repeated with the redefinition of isotopes. For each iteration, the window (Del) is reduced by the equation:

$$Del = Del/(0.5\ C + 0.5),$$

where C is the number of iterations.

If the number of iterations required exceeds four, the process is halted and the program continues.

C. Peak Verification

Based on the isotope identifications from the previous program, this program determines that all peaks that should have been detected have been detected. Input data includes the original spectrum and the isotope library. A function defining the shape of the gamma efficiency curve is included as a table in a function subprogram.

If there is any evidence of a peak in the spectrum or if there is justification for not seeing the peak, such as excessive background or interference from other isotopes, a missing peak is considered detected. If a peak should have been detected and its absence cannot be justified, the corresponding branching ratio and all branching ratios for that isotope are set to zero, which is a flag to abort later activity calculations since isotope identification has not been verified. If more than three peaks have been detected for one isotope, the isotope ID is considered verified.

The 3-sigma counting error is used for each missing peak and 25% is added to this to account for accumulated errors in branching ratios and efficiency values. If this error is greater than or equal to 100%, the missing peak is justified.

Missing peak counts are determined by calculating back from the largest peak detected.

The output of this program is listed and given to the analyst for manual editing before further processing is performed. The analyst adds his knowledge of the sample to the processing cycle to accept or reject isotope selection and computer results. The analyst can reject the results in their entirety but cannot completely remove any portion of those results. If he rejects an isotope identification, he can only flag those results so that an activity is not calculated. The peaks will still be listed by possible identification and corresponding energy. Peaks which are not identified are listed as "XPEAKS."

D. Activity Calculations

This program is the last in the spectrum reduction process, and with it, the following functions are performed:

1. Peak ratios are checked.

2. Activities are calculated from valid peaks.

3. Data is integrated into the carry file of the laboratory's Daily Report System.

Activity is calculated from the best photopeak detected. This is the peak with the smallest error and one which has (for multipeak isotopes) at least one valid peak ratio. If no valid peak ratio can be found, the next best peak is selected and so on.

Specific activity is calculated using sample size as provided in the carry file. Half-life correction to midsampling time is accomplished with a half-life library which has been placed in one of the program's input files.

Data records are constructed to be compatible with daily report data. Where more than one data record is necessary, the computer indexes this record by incrementing the start sampling time. Isotope identification is contained in the comments portion of each record.

DAILY REPORT SYSTEM

This software system is a combined analytical and management information system. Its purpose is to calculate results and compile these results in a usable report form so that the daily functions of the Automated Nuclear Measurements Laboratory can be reviewed. Calibration (parameter) data is maintained and automatically edited, charge code data is assembled for budget management purposes, and routine bioassay sampling is managed by this system.

A. Processing Steps

The processing steps as defined by the various programs is as follows:

1. This process accepts data from our laboratory and separates spectrum data from other data. Spectrum card records are stored in a separate file for future processing. Other input data is sorted by sample number and each card record is listed.

2. Spectra are constructed from the card record spectrum data and stored in a separate file for future spectrum reduction.

3. Data in card record form is assembled into sample records. Counting data is matched to sample data where possible and appropriate calculations are made to obtain results which are stored to a maximum of five results per sample record. Comments are also matched with sample data and stored to a maximum of ten comments per sample record. At this step, various checks are made on the validity of the data and messages are listed when errors are detected.

4. Sample records are assembled on a carry file. Records are sorted by event code and type code. This is the file from which the daily report is printed. It lists all samples not yet completed and samples that are completed on the current run. At this phase, old sample data is combined with new counting data and old counting data is matched with new sample data. Appropriate calculations are made and results are stored in the sample record. Additional comments are also matched with the corresponding sample record. An editing feature enables the replacement of old data with new data. An editing table is used to designate samples to be dropped from the report.

5. Spectrum results are calculated and included in the carry file. Additional sample records are generated where necessary to accommodate the large volume of spectrum results (more than five results per sample and more than ten comments per sample).

6. The daily report is printed here and completed samples are segretated on a separate file. Completed samples are dropped from the carry file and the new file becomes an input file for the next job.

7. Completed samples are sorted by collection date and time. They are combined on a "Results File." This file will become a master file from which results will be selected for various reports.

8. During each week, a charge code is assigned and accumulated for each analysis. Codes are selected by analysis and sample type. The accumulated code credits are assigned to the appropriate budget through the event code of each sample. A report is automatically printed on a weekly basis and a monthly summary is printed on request (using a special input character). Each report lists the accumulated charge code credits and their assigned budget. A summary of all priority requests is also listed. Priorities range from zero through four and are also matched to the budget of the requester.

B. Table Editing

Table editing is usually performed sometime before step No. 3. Input data submitted for this phase include the following: current report

date, table of omission requests (samples to be omitted from the carry file), file control and listing flags, charge code table, budget code table, analysis description table, type description table, program description table, and a self-editing parameter table which provides background and efficiency information. An entry in this table will automatically be dropped if it is not used during a minimum of 20 successive runs.

C. File Editing and Report Programs

Numerous file editing programs are used to sort, merge, omit, or otherwise modify the data. Several report programs are also available with which to compile results from our result files. Since the design of these programs is not unique and the programs themselves are frequently changed, it is beyond the scope of this presentation to describe them here. The total system, however, is comprised of about 30 programs in all.

CALIBRATION FOR SOIL GAMMA ANALYSES

A. Preparation of Standard Soil Samples

Quantities of low-activity soil were collected from areas remote from contaminated terrain. This material was then dried and ball-milled to a particle size less than 10 mesh. A measured quantity of this material (about 1 kg) was placed in a 1-gal paint can. Five milliliters of a standard solution (NBS No. 4243-D) was added in such a way as to cover as much surface area as possible without coming in contact with the sides of the container. This mixture was then dried at room temperature for 72 hr. The paint can was then sealed and placed in a paint shaker where the soil was agitated for 6 hr. (This method had previously been proved successful in producing a homogeneous mixture.) This standard soil was then weighed into 16-oz Nalgene bottles in the amount of 100, 300, and 500 g. The bottles were capped and sealed, decontaminated, and delivered to the measurements laboratory for counting. Standards containing amercium-241 were prepared in the same manner with the exception that they were dried under a heat lamp. The americium solution was supplied by Dr. Eric Fowler of LASL.

B. Analysis of Efficiency

Standard samples were measured in pulse-height analyzers using Ge(Li) detectors. Parameters of counting time, channels per spectrum (2,000 chn/spect.), and energy calibration (1 kev per channel) were identical with those used in the measurement of soil samples. Spectrum listings were obtained through the laboratory computer system and peaks were selected manually.

C. Calculations

Calculations of photo peak gamma efficiency were performed using the most recently published data on branching ratios as supplied by Lawrence Livermore Laboratory (Prindle, various dates) and the Radiological Health Handbook (1970).

The photo peak at 279 kev which was produced by the isotope ^{203}Hg was not used, since it was apparent that some of the volatile mercury had been lost in the drying process. This effect had been expected, since mercury is very difficult to stabilize under the existing circumstances (see Fig. 5).

D. Energy Calibration

The energy calibration of each instrument is checked daily using a check source containing ^{57}Co and ^{22}Na. These isotopes produce photo peaks at 121, 511, and 1,274.5 kev. Each analyzer is also gain stabilized with a precision pulser, which obtains a resolution of about 4 kev FWHM at 1,274 kev.

E. Background

Machine background correction was found to be insignificant for soil, since naturally occurring isotopes in each soil sample produced a much greater radiation spectrum. Machine background ranges from 70 to 100 counts per minute, with most of the contribution in the thorium regions of the spectrum.

F. Sensitivity

The minimum detectable activity of nuclides in soil samples of 100 to 500 g range in the neighborhood of 10^{-1} picocuries per gram. The minimum detectable activity of ^{241}Am in soil samples with no significant interfering activities averages about 1.2×10^{-1} picocuries per gram.

The minimum detectable activity (Engstrom, 1964) is influenced by interfering isotopes in that these activities produce a background which is unique with each sample. The minimum detectable gamma intensity "A" is a function of the background counting rate as follows:

$$A = (3/q)\ (M^{-1/2}/T_b),$$

where

q is the instrument efficiency

M is the background counts collected in time T_b

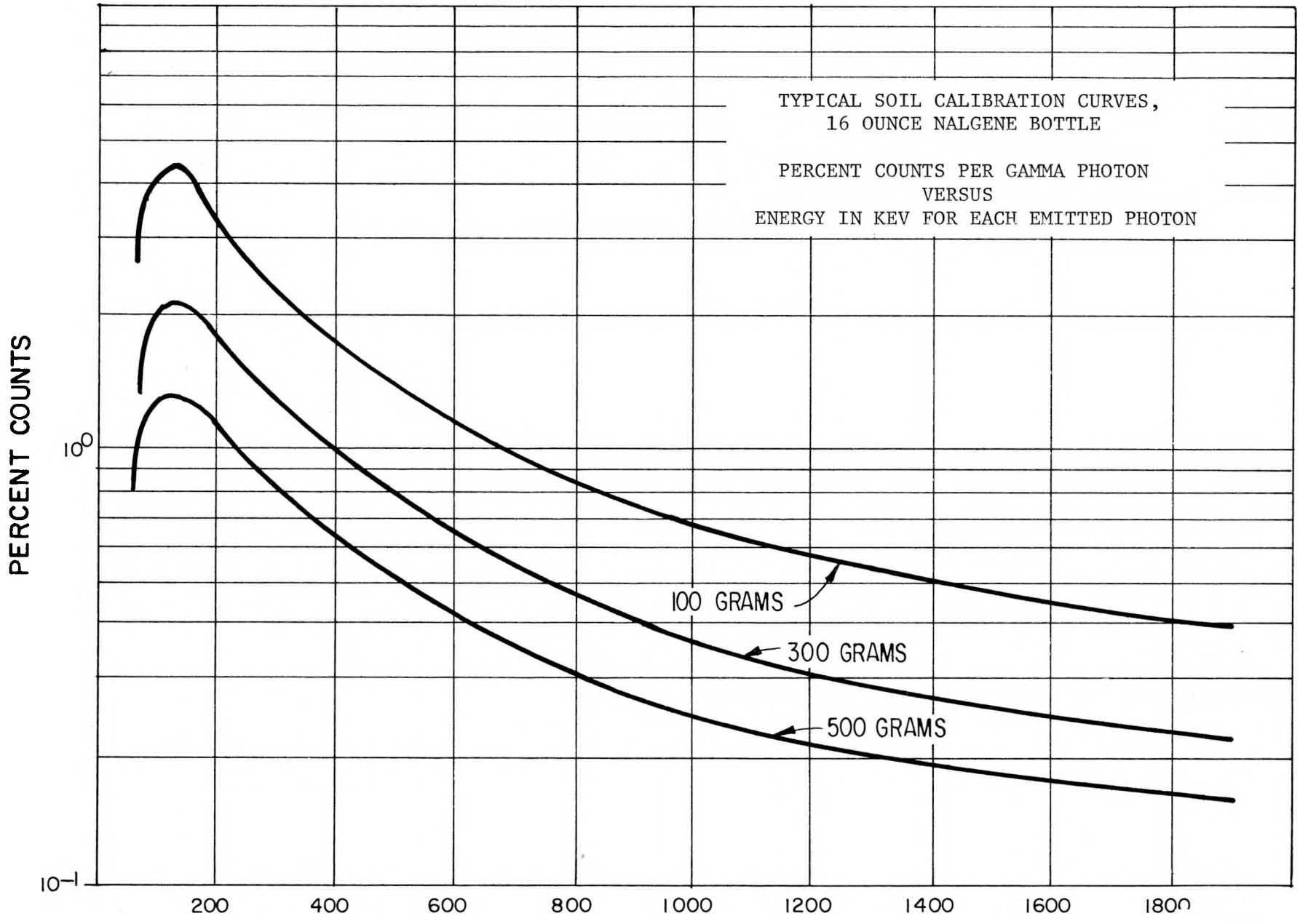
TYPICAL SOIL CALIBRATION CURVES,
16 OUNCE NALGENE BOTTLE
PERCENT COUNTS PER GAMMA PHOTON
VERSUS
ENERGY IN KEV FOR EACH EMITTED PHOTON
PERCENT COUNTS
10^0
10^{-1}
100 GRAMS
300 GRAMS
500 GRAMS
200
400
600
800
1000
1200
1400
1600
1800
ENERGY (KEV)

FIGURE 5

Therefore, it can be seen that for each decade of increase in interfering activity, the minimum detectable activity can be expected to increase by a little more than a factor of three.

The considerations for optimizing soil sample size were as follows:

1. Optimum size for the lower energy radiations of ^{241}Am was found to be considerably less than for the higher-energy gamma emitters encountered in the soils. If ^{241}Am was the only isotope of interest, then the sample size could be cut to about 100 g without seriously affecting sensitiviy (see Fig. 6). However, sensitivity for the higher-energy emitters would be optimized with a much larger volume. Therefore, samples were made as large as was practicable.

2. The 16-oz bottles were available in the laboratory and were a standard size for which there were already some calibration data.

3. The geometry of the 16-oz bottle used in the laboratory is ideal with respect to the geometry of the germanium gamma detectors. All the active sample material is in the detector window.

CONCLUSIONS

The use of this laboratory facility has been highly successful in analyzing large numbers of soil samples for ^{241}Am and other gamma-emitting nuclides. Present counting times of 100 min and present integral counts of 1,000,000 were found to be adequate in achieving sensitivities in the 0.1 picocurie per gram range which is consistent with similar projects conducted at Pacific test sites by the University of Washington (Nelson and Noshkin, 1973).

It was realized that differences in sample density would be a source of error; however, the simplified calibration method reduced the number of man-hours necessary for each analysis and increased the laboratory capability with regard to quantity. Minimizing this error was the fact that most soils collected were fine soils and closely approximated the standard. Another minimizing effect was a method of spectrum reduction which did not use spectrum stripping. The resulting differences were also considered a small part of the errors introduced by the physical sampling process and by the natural variations in soil concentration.

The large quantities of lead shielding used with gamma detectors enabled reduction of cosmic background to levels compatible with laboratories at lower elevations. Therefore, no sensitivity was sacrificed due to this source.

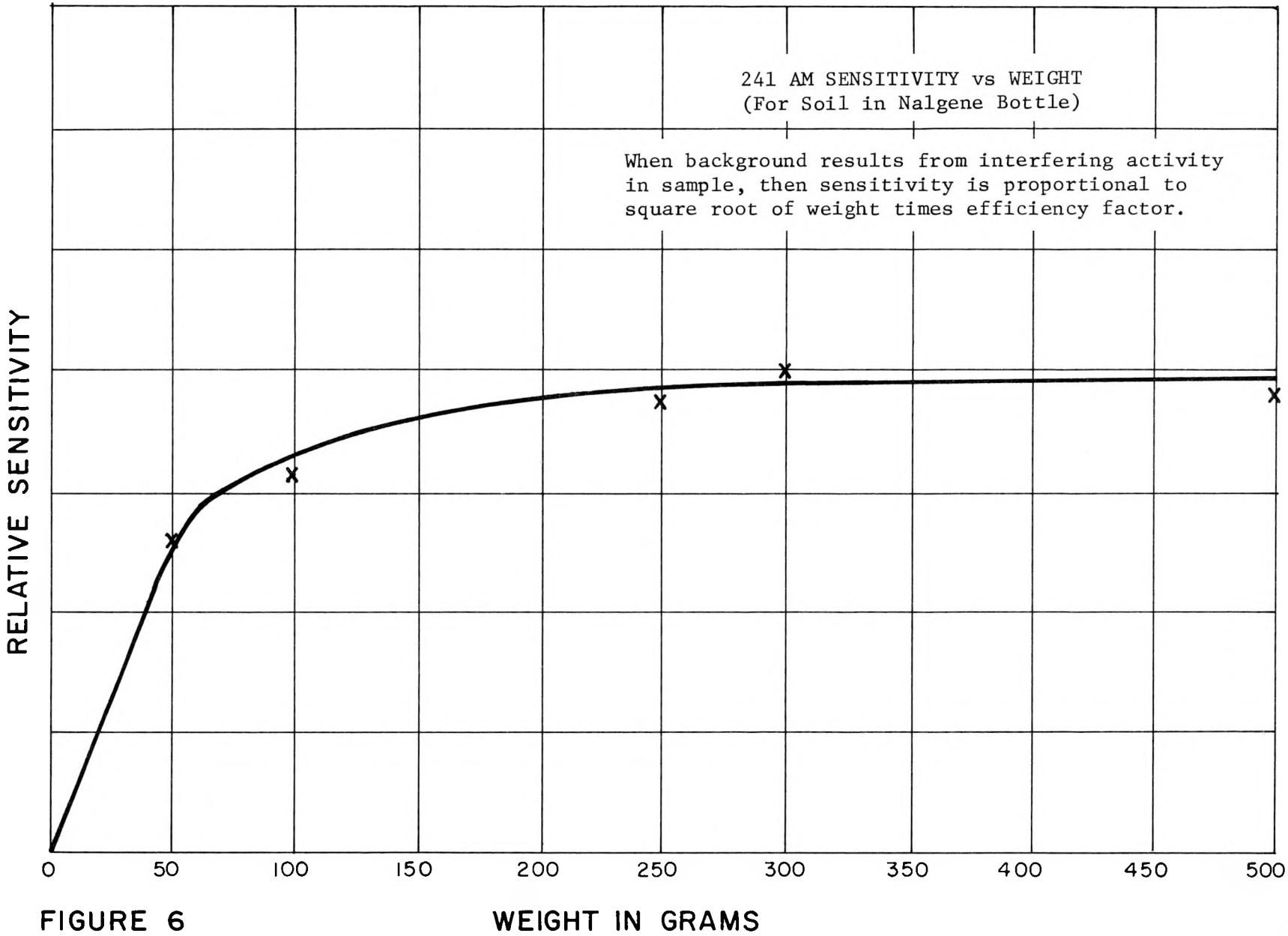
241 AM SENSITIVITY vs WEIGHT
(For Soil in Nalgene Bottle)
When background results from interfering activity in sample, then sensitivity is proportional to square root of weight times efficiency factor.
RELATIVE SENSITIVITY
0
50
100
150
200
250
300
350
400
450
500
WEIGHT IN GRAMS

FIGURE 6

In the past six months, this system has produced results for over 7,000 analyses. Of these, 3,000 have been gamma spectrum analyses of soils.

This system has eliminated the jobs of two full-time technicians and has kept expensive instrumentation in operation 24 hr a day and 7 days a week without the added expense of overtime. Reliability has been excellent and downtime is far less than that experienced by the central computer.

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DETERMINATION OF ^{238}U, $^{235-236}U$, and $^{233-234}U$ IN SOIL AND VEGETATION SAMPLES (TENTATIVE)

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OBJECTIVE

Samples of soil and vegetation containing small amounts of U in the presence of relatively large amounts of Pu and Am are treated by this procedure to isolate an alpha spectrometrically pure U sample. The main interfering element is Fe which is naturally present in all the sample matricies to such an extent that serious alpha spectrum degradation occurs if Fe is not removed. Alpha interferences from such sources as Pu, Am, Th, and almost all other alpha emitters (except U) are either removed from the sample or have alpha energies that do not interfere with the various natural or weapon U isotopes. The procedure includes (1) sample digestion, (2) removal of the matrix mass by Fe scavenging, (3) ion exchange separation of U from almost all other elements which might interfere, (4) ether extraction of the Fe, and (5) electrodeposition of the U for alpha spectrometry using ^{232}U as a chemical yield tracer. The procedure is outlined in Fig. 1.

REAGENTS AND EQUIPMENT

A. Preparation of Soil

1. 250 ml Teflon beaker with Teflon cover
2. HNO_3, 70%
3. HCl, 38%
4. HF, 48%
5. H_3BO_3 - crystal
6. $FE(NO_3)_3$ (25 mg Fe/ml)
7. ^{232}U tracer $\sim$ 10 d/m/ml

B. Preparation of Vegetation

1. Muffle furnace
2. 600 ml pyrex beaker with pyrex beaker cover

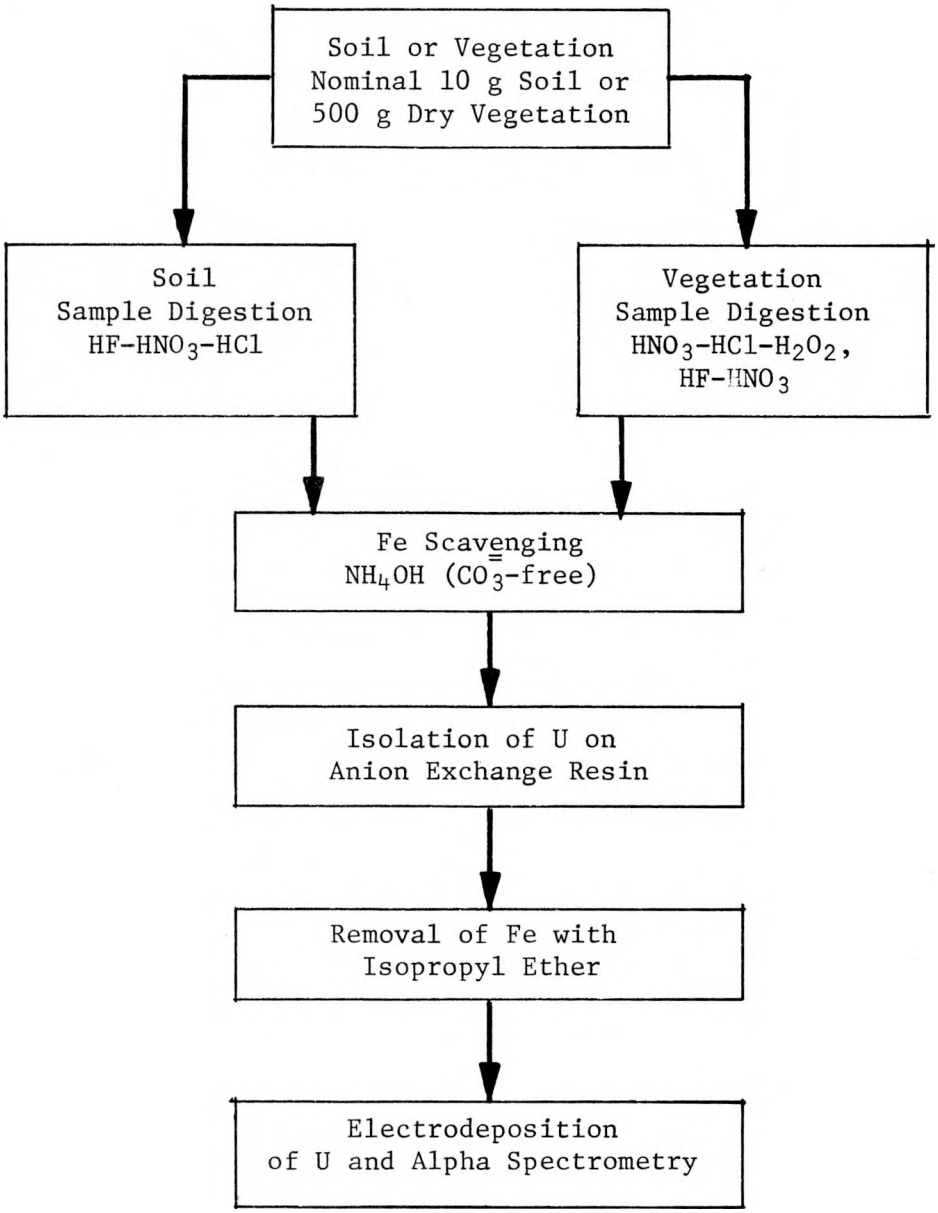

Figure 1. Flow Chart of Tentative Procedures to Isolate Alpha Spectrometrically Pure U

3. HNO_3, 7%

4. HCl, 38%

5. HF, 48%

6. H_2O_2, 30%

7. H_3BO_3 - crystal

8. $Fe(NO_3)_3$ (25 mg Fe/ml)

9. Filtering Apparatus

10. ^{232}U tracer $\sim$ 10 d/m/ml

C. Fe Scavenging

1. NH_4OH 28% ($CO_3^{=}$-free)

2. 8M HNO_3 (saturated with H_3BO_3)

3. 100 ml polypropylene round bottom centrifuge tubes

4. Stirring rods, polyethylene or Teflon--able to withstand centrifugation

D. Isolation of U

1. Resin columns, 1.27 cm (1/2 in.) diameter by 10 cm with $\sim$ 250 ml reservoir and Teflon stopcock.

2. Anion exchange resin--Bio Rad AG 1 x 8 50-100 mesh

3. Acid washed sand 100-200 mesh

4. Pyrex wool

5. 100 ml pyrex beaker

6. 250 ml polyethylene bottle

7. HCl, 48%

8. 2M NaOH

9. $HCl \cdot NH_4I$ (230 ml HCl + 72 ml H_2O + 18 ml 1M NH_4I)

10. $HCl \cdot HNO_3$ (240 ml HCl + 2.4 ml HNO_3)

11. 9M $HCl \cdot HNO_3$ (350 ml HCl + 136 ml H_2O + 12 ml HNO_3)

12. 5M HCl

13. 0.25M HCl

E. Fe Removal

1. separatory funnel--60 ml

2. isopropyl ether--alcohol free

3. 8M HCl

4. 0.25M HCl

5. HNO_3, 70%

6. HCl, 38%

F. Electrodeposition

1. H_2SO_4 (6 ml H_2SO_4/100 ml H_2O)

2. 0.18M H_2SO_4

3. thymol blue--.02% (.02 g thymol blue dissolved in 10 ml absolute ethanol then diluted to 100 ml with H_2O)

4. NH_4OH, 1:9

5. NH_3

6. $NH_4NO_3 \cdot NH_4OH$ (10 g NH_4NO_3 + 10 ml NH_4OH per 1).

7. ethanol pH 8

8. electrodeposition cell and apparatus

9. electropolishing solution (200 ml H_2O + 450 ml H_3PO_4 + 350 ml H_2SO_4)

10. plate, stainless steel 20 mm dia. by .010 in. (.254 mm)

11. Pt electrode ∿ 3. mm dia.

PROCEDURE

A. Preparation of Soil

1. Weigh up to 10 g dry soil into a 250 ml Teflon beaker. Add 2 ml $Fe(NO_3)_3$ and an appropriate amount of ^{232}U tracer. (Normally ∿ 10 d/m/samples are used but should ^{238}Pu be very high in the sample, larger amounts of ^{232}U will be needed.)

2. Add to the sample 30 ml HNO_3, mix thoroughly to completely suspend the sample, add 30 ml HF. Mix sample thoroughly, cover with Teflon beaker cover and heat to near boiling for 1 hour.

3. Remove beaker cover and add 30 ml HNO_3/30 ml HF, mix and heat for 1 hour.

4. Remove beaker from hot plate, cool, and add 10 ml HCl, cover beaker with Teflon cover and heat until red-brown fumes no longer are evolved. Remove beaker cover and evaporate sample to about 25 ml.

5. Add about 5 g H_3BO_3, mix, and heat for 15 minutes. Proceed with Fe scavenging.

B. Preparation of Vegetation--Procedure from W. J. Major *et al.* (1973)

1. Sample in 1 gal paint can is covered with perforated aluminum foil, dried at 110°C, weighed, and carbonized at 250°C for 12 hrs. Sample is transferred to double-walled plastic bag, crushed, and placed in tared pyrex beaker. Sample is ashed at 600°C for 2 days, and weighed.

2. Ash is leached with hot HNO_3-HCl and H_2O_2.

3. Any residue is filtered, ashed, and treated with HF-HNO_3 and H_3BO_3. This solution is added to the solution from step 2 above. The sample is now ready for Fe scavenging. It may be desirable to sub-aliquot at this point.

C. Fe Scavenging

1. Transfer dissolved sample or aliquot of dissolved sample to a centrifuge tube using 8M HNO_3 saturated with H_3BO_3 for washing. To the sample add an appropriate amount of ^{232}U tracer and 2 ml $Fe(NO_3)_3$.

2. Slowly add $CO_3^=$ free NH_4OH to the first permanent Fe floc. Add an additional 10 ml NH_4OH, adjust volume with H_2O to about 70 ml, mix well with stirring rod, and centrifuge, discarding supernate.

3. Dissolve precipitate with about 10 ml HNO_3 saturated with H_3BO_3 and repeat step 2 above. After the second execution of step 2 above the sample is ready for isolation of U on anion exchange resin.

D. Isolation of U. Procedure modified from Crouch and Cook (1956).

1. Prepare a resin column as follows:

a. Pack a 1 cm plug of pyrex wool into the bottom of the pyrex column, then load with 8 cm of settled anion exchange resin followed by 1 cm of washed sand.

b. Charge resin by washing with:

(1) 3 x 10 ml HCl

(2) 3 x 10 ml H_2O

(3) 4 x 10 ml $HCl \cdot NH_4I$

(4) 1 x 10 ml H_2O

(5) 3 x 10 ml 2M NaOH

(6) 2 x 10 ml H_2O

(7) 2 x 10 ml 1M HNO_3

(8) 2 x 10 ml 3M HNO_3

(9) 2 x 10 ml 8M HNO_3

(10) 2 x 10 ml $HNO_3 \cdot HCl$
(160 ml 8M HNO_3 + 5 ml HCl)

(11) 6 x 10 ml $HCl \cdot HNO_3$

(12) 6 x 10 ml 9M $HCl \cdot HNO_3$

c. Immediately after wash number (12) (above) add sample to resin column. Prepare sample by dissolving precipitate in enough HCl to make about 9M then add about 30 ml of 9M $HCl \cdot HNO_3$ solution. Centrifuge to remove undissolved solids and decant supernate into resin column. Wash centrifuge tube and contents 3 times with 10 ml portions of 9M $HCl \cdot HNO_3$ centrifuging and each time decanting supernate to resin column.

d. Wash resin column with the following:

(1) 6 x 10 ml 9M $HCl \cdot HNO_3$,

(2) 8 x 10 ml $HCl \cdot NH_4I$, and

(3) 15 x 10 ml 5M HCl.

e. Elute U into a 250 ml polyethylene bottle with 15 x 10 ml .25M HCl.

f. Evaporate sample to dryness in 100 ml beaker, wash beaker sides with 5 ml HNO_3, evaporate to dryness, wash beaker sides with 5 ml HCl and evaporate to dryness. Repeat column isolation of U a second time on a fresh but similar resin column (a smaller column can be used since only Fe and U are expected to be present in any quantity). Sample is now ready for removal of Fe.

E. Fe Removal

1. Place about 12 ml isopropyl ether in a 60 ml separatory funnel.

2. Wash ether with 2 x 10 ml portions of 8M HCl with vigorous shaking for 15 sec.

3. Transfer sample to separatory funnel with 2 x 4 ml and 1 x 2 ml portions of 8M HCl. Extract Fe with vigorous shaking for 20 sec and transfer aqueous phase back to original 100 ml beaker.

4. Add 10 ml 8M HCl to separatory funnel and gently swirl. Add aqueous phase to original sample. Evaporate sample to dryness.

5. Wash ether with 2 x 10 ml portions of .25M HCl then with 2 x 10 ml portions of 8M HCl and proceed with next extraction.

6. Repeat ether extractions until the Fe has been completely removed as evidenced by disappearance of the yellow coloration. Three extractions are usually sufficient.

F. Electrodeposition of U. Procedure modified from Talvitie (1972).

1. To the evaporated sample add 10 ml of H_2SO_4 (6 ml H_2SO_4/100 ml) washing sides of beaker. Evaporate to SO_3 fumes.

2. Add 3 ml .18M H_2SO_4 and 2 drops of thymol blue indicator. Neutralize to a salmon-pink color with NH_3.

3. Polish a stainless steel plate by electrolysis in 10 ml of electro-polishing solution with the plate as the cathode and a Pt anode (about 10 minutes at 1 ampere, 12-15 volts). Wash cell thoroughly with H_2O.

4. Transfer sample to electrodeposition cell using 18M H_2SO_4 to wash beaker--2 ml rinse, 2 ml + police, and 2 ml + police.

5. Readjust indicator color to a salmon color with NH_3.

6. Electrodeposit U for about 60 minutes at 1 ampere and 12-15 volts with the plate as the anode.

7. Before interrupting the current add to the cell 10 ml of 1:9 NH_4OH and continue electrodeposition for 1 minute.

8. Remove the cell from the electrodeposition apparatus and wash cell with 3 small portions of $NH_4NO_3 \cdot NH_4OH$ (F-6). Disassemble the cell and wash plate with ethanol. Drain dry and heat on hot plate at 250°C for 15 minutes. Sample is now ready for alpha spectrometry.

ACKNOWLEDGEMENT

The authors wish to thank Dr. Gerald Ellis for preliminary investigations and literature search.

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PROCEDURE FOR ANALYSIS OF ^{90}SR IN SOIL AND VEGETATION SAMPLES

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STRONTIUM

Strontium-90 is isolated from digested vegetation or soil samples as the carbonate, is purified by cation exchange, and is beta counted as the carbonate. It is assumed that the Nevada Applied Ecology Group's (NAEG) samples are very low in ^{89}Sr (52.7d-half life) and devoid of ^{85}Sr (64.0d-half life), therefore, direct counting of the $SrCO_3$ with appropriate efficiency corrections will yield an acceptable ^{90}Sr value for the sample. The procedure is listed in the Appendix. The $SrCO_3$ precipitate resulting from the purification of the Sr is mounted on a filter pad with a diameter of 24 mm, and counted on a gas flow proportional beta counter with a thin mylar window. The efficiency of the beta detector is measured by processing a known amount of ^{90}Sr as a sample, preparing the $SrCO_3$, and counting the resulting precipitate in the same geometry as an unknown sample.

It is usually difficult to separate a pure ^{90}Sr fraction from a sample, prepare the ^{90}Sr as $SrCO_3$, and count the prepared sample in a short enough time to avoid a significant ^{90}Y ingrowth. An alternative is to wait about 2 weeks until the ^{90}Sr - ^{90}Y essentially have attained secular equilibrium. An attempt is made to correct for the ^{90}Y ingrowth knowing the time elapsed from the purification step to the counting step.

Starting with the general decay equation for parent and radioactive daughter (Lapp and Andrews, 1963),

$$\Delta N_y = (\lambda_s N_s - \lambda_y N_y)\, \Delta t \,, \qquad (1)$$

where: N_y = number of ^{90}Y atoms,

N_s = number of ^{90}Sr atoms,

λ_y = decay constant for ^{90}Y,

λ_s = decay constant for ^{90}Sr, and

t = elapsed time from ^{90}Sr purification to count time.

The number of ^{90}Sr atoms that have decayed during the elapsed time is given by

$$N_s = N_s^o \, e^{-\lambda_s t} \quad , \tag{2}$$

where N_s^o = initial number of ^{90}Sr atoms present.

Substituting (2) into (1) gives

$$\Delta N_y = \left(\lambda_s N_s^o e^{-\lambda_s t} - \lambda_y N_y \right) \Delta t. \tag{3}$$

Integrating (3) results in the number of atoms of ^{90}Y that are present at time t,

$$N_y = N_s^o \frac{\lambda_s}{\lambda_y - \lambda_s} \left(e^{-\lambda_s t} - e^{-\lambda_y t} \right) . \tag{4}$$

But since $A_y = N_y \lambda_y$ the equation can be rewritten in terms of activity as follows:

$$A_y = A_s^o \frac{\lambda_y}{\lambda_y - \lambda_s} \left(e^{-\lambda_s t} - e^{\lambda_y t} \right). \tag{5}$$

where: A_s = actual ^{90}Sr activity in the sample (d/m), and

A_y = actual ^{90}Y activity in the sample (d/m) at count time (t).

Assuming that $A_s = A_s o$ and λ_s is very small because of the short time involved relative to the ^{90}Sr half-life,

$$A_y = A_s \left(1 - e^{-\lambda_y t} \right) . \tag{6}$$

If T is the measured net ^{90}Sr + ^{90}Y activity then

$$T = S + Y \tag{7}$$

where S = measured ^{90}Sr, and Y = measured ^{90}Y activities respectively. However,

$$S = \frac{A_s E_s}{\overline{SA}_s} \text{ and } Y = \frac{A_y E_y}{\overline{SA}_y} \quad , \tag{8}$$

where: E_s and E_y are the respective efficiencies of infinitely thin ^{90}Sr and ^{90}Y sources, and

$\overline{SA}_s$ and $\overline{SA}_y$ are the respective self-absorption correction factors.

Substituting (8) into (7) gives

$$T = \frac{A_s E_s}{\overline{SA}_s} + \frac{A_y E_y}{\overline{SA}_y} \quad , \tag{9}$$

and finding A_y from (6) gives

$$T = \frac{A_s E_s}{\overline{SA}_s} + \frac{E_y A_s}{\overline{SA}_y} \left(1-e^{-\lambda_y t}\right). \tag{10}$$

By measurement $SA_y \simeq 1$. Therefore,

$$A_s = T/ \left[\frac{E_s}{\overline{SA}_s} + E_y \left(1-e^{-\lambda_y t}\right)\right]. \tag{11}$$

It was determined that the counting efficiency of an infinitely thin sample of ^{90}Sr and ^{90}Y was essentially the same. This was determined by counting different weights of $SrCO_3$ precipitate of purified ^{90}Sr as soon after purification as possible then recounting the same samples 14 days later. The difference in count rate was directly attributed to ^{90}Y. Self-absorption curves for ^{90}Sr and ^{90}Y are plotted in Figure 1. Note that the self-absorption curve for ^{90}Y supports the assumption used in (11) that $SA_y \simeq 1$.

An example of the influence of time between ^{90}Sr separation and counting on the net count rate using (11) is shown in Table I. Beginning with 100 d/m ^{90}Sr as a constant source the net count rate at time t is given.

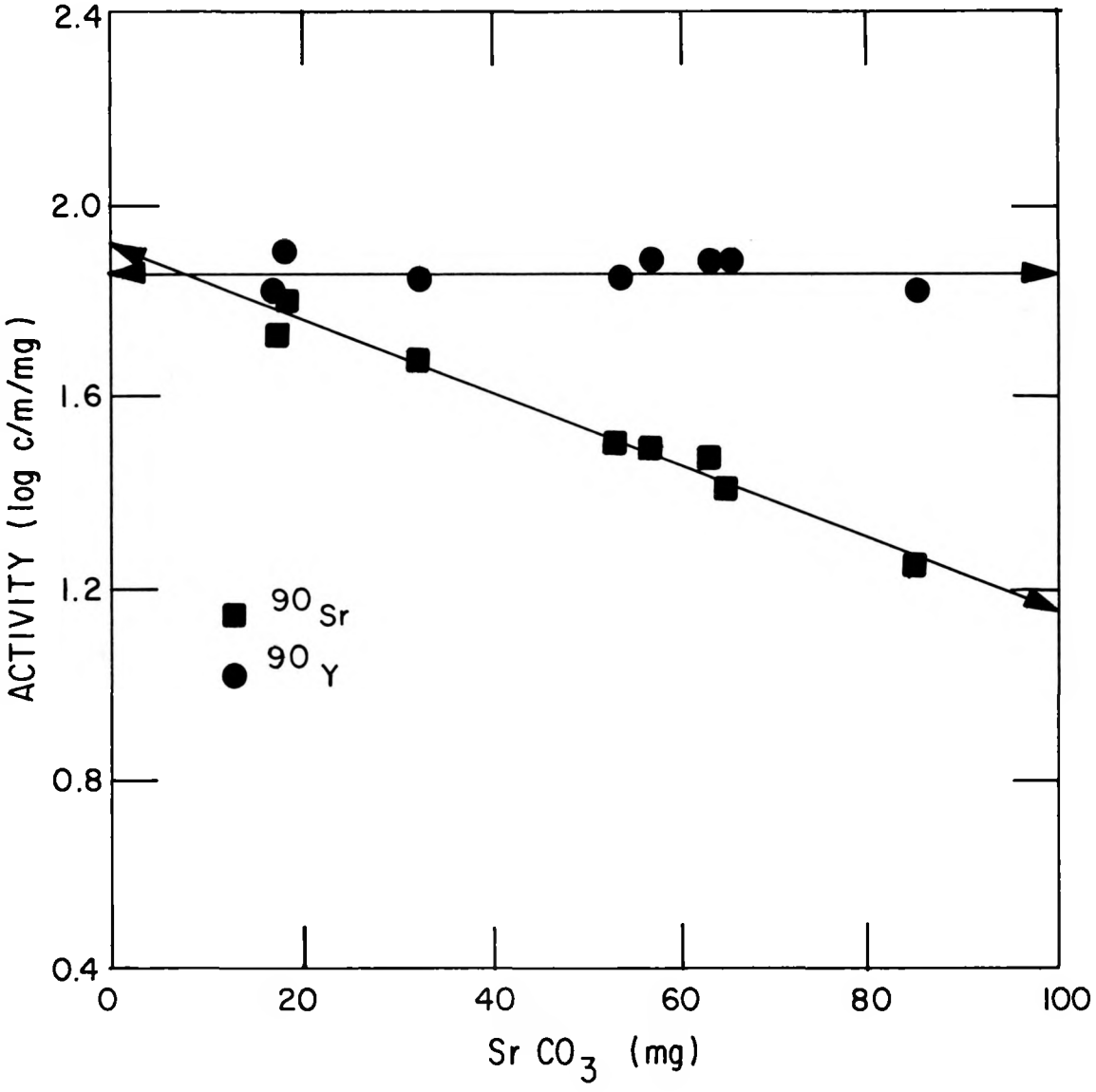

FIG. 1 Self-absorption of ^{90}Sr and ^{90}Y beta emissions by $SrCO_3$

TABLE I

Effect of ^{90}Y Ingrowth on ^{90}Sr

Net Count Rate

Time (hr)	Net Count Rate (c/m)*
0	13.5
1	13.7
4	14.4
9	15.5
16	17.0
25	18.7

*Initial ^{90}Sr present = 100 d/m.

REFERENCE

1. Lapp, R. E., and H. L. Andrews. 1963. *Nuclear Radiation Physics.* Prentice-Hall, Inc., p 82.

APPENDIX

LASL PROCEDURE FOR ^{90}SR IN SOIL AND VEGETATION
(Porter *et al.*, 1967)

Reagents and Apparatus

1. Dowex 50W x 8 (50-100 mesh) cation exchange resin in sodium form.
2. Glass column (1 cm dia. ID) with 250 ml reservoir and to contain 10 ml resin.
3. Prepare resin by washing the column with 150 ml 4 M NaCl and then with 50 ml H_2O.
4. Beaker, Griffin, 250 ml.
5. Centrifuge tube, conical, glass, 90 ml.
6. Centrifuge tube, polypropylene, 100 ml.
7. Strontium carrier, 24.09 g $Sr(NO_3)_2$ + 2.9 ml HNO_3 diluted to 500 ml with H_2O.
8. HNO_3, 70%
9. 8 M HNO_3
10. 0.1 M HNO_3
11. NH_4OH, 28%
12. Ethanol absolute
13. Thymol blue indicator - .02% (Dissolve .02 g thymol blue in 10 ml absolute ethanol then dilute to 100 ml with H_2O)
14. 6% EDTA solution, 65 g/l Na_2 EDTA dissolved in H_2O
15. EDTA pH 5.1 rinse solution. Mix 50 ml 6% EDTA solution and 100 ml H_2O. Adjust pH to 5.1 with NH_4OH.
16. Prepare pH 4.6 buffer by dissolving 200 g sodium acetate in 50 ml water, adding 385 ml glacial acetic acid, and diluting to 1 liter, adjust pH to 4.6 with NaOH or acetic acid using a pH meter.
17. 4 M NaCl (dissolve 240 g NaCl/l)

18. 1.5 M HCl

19. Ammonium carbonate reagent--$(NH_4)_2CO_3$(3 M) in NH_4OH (3 M); 200 ml NH_4OH + 288 g $(NH_4)_2CO_3$/1.

Soil

1. Weight 10 g soil into a 90 ml glass C-tube. Add Sr carrier and 20 ml HNO_3, digest 1 hr. at boiling.

2. Centrifuge, transfer supernate to 250 ml beaker.

3. Add 20 ml HNO_3, digest .5 hr., centrifuge, combine supernatants.

4. Add 20 ml 8 M HNO_3, digest .5 hr., centrifuge, combine supernates and discard residue.

5. Evaporate combined supernates to near dryness to reduce acid content to a minimum.

6. Transfer contents of beaker to centrifuge tube (100 ml) using 0.1 M HNO_3 to wash beaker.

7. Add NH_4OH dropwise to supernate to precipitate Fe, centrifuge, and decant supernate back into 250 ml beaker.

8. Dissolve Fe precipitate in minimum HNO_3 and reprecipitate with NO_4OH. Centrifuge, and combine supernates in the 250 ml beaker of step 7. Repeat once more. Discard Fe precipitate. Proceed to "Separation of Sr."

Vegetation (dissolved in 8 M HNO_3)

1. Aliquot desired amount of solution into a 250 ml beaker.

2. Add Fe carrier, Sr carrier, and excess Ca. Evaporate to near dryness to remove excess acid. Transfer sample to 100 ml centrifuge tube using 0.1 M HNO_3 to wash beaker.

3. Add NH_4OH dropwise and continue with step 7. above.

Separation of Sr

1. Add a few drops of thymol blue indicator and then add NH_4OH to beaker to the blue endpoint. Add an excess 10 ml of NH_4OH. Add 5 ml ammonium carbonate reagent to precipitate alkaline earth carbonates. Let stand overnight. Filter through membrance and filter and wash beaker and filter with minimum of H_2O discarding filtrate. Transfer filter and carbonate precipitate back into 250 ml beaker, washing filter funnel and sides of beaker with 100 ml 6% EDTA solution.

2. Adjust solution from 1. to about pH 4.6 with a minimum of NH_4OH or HCl, add 5 ml buffer solution.

3. Transfer solution to resin column and let flow at 5 ml/min.

4. Wash beaker and resin column with 150 ml of pH 5.1 EDTA solution. Record time at end of elution as beginning of ^{90}Y ingrowth.

5. Wash column with 50 ml water, discard all effluents.

6. Wash column with 10 ml 1.5 N HCl, discard effluent.

7. Elute Sr with 100 ml 1.5 N HCl at a flow rate of 2 ml/min, collect effluent in a clean 250 ml beaker.

8. Regenerate resin with 150 ml 4 M NaCl and wash with 50 ml H_2O.

9. To the Sr eluate add 25 ml NH_4OH to the thymol blue endpoint (blue), slowly add 5 ml ammonium carbonate reagent. Let stand for 30 minutes with periodic stirring. Filter through a tared glass fiber filter. Wash beaker with 3-10 ml portions of water, policing thoroughly. Wash filter with absolute ethanol. Dry filter and weigh for yield determination and count precipitate for ^{90}Sr content.

REFERENCE

1. Porter, C. R., M. W. Carter, G. L. Rehnberg, and E. W. Pepper. "Determination of Radiostrontium in Food and Other Environmental Samples." 153rd Meeting American Chemical Society, Miami Beach, Fla. Apr. 9-14, 1967.

PARTICLE SIZE ANALYSIS OF SOILS

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The segregation technique for particle size analysis, described herein, differs from the conventional technique, primarily in the pretreatment of the soil sample. The usual pretreatments optimize dispersion of the particles to determine the ultimate size of the soil particles. These pretreatments include hydrogen peroxide to destroy organic matter, addition of a dispersant such as sodium carbonate or sodium phosphate, and mechanical agitation in a shaker or mixer (Day, 1955). The only aid used to disperse the particles in this size analysis is gentle stirring of the suspension with a glass rod covered with a rubber policeman. The reason for the minimal pretreatment is to retain the association of the plutonium with the soil particles. Using this technique, several samples were given 5-min ultrasonic treatment to induce dispersion and the results were compared with nonultrasonified replicates. The results show that the sand content decreased by approximately 10% and the silt and clay content increased proportionately. The sand contributed about 25% of the soil radioactivity without dispersion; with ultrasonic treatment, sand contributed about 8% (Tamura, 1975). The technique therefore is indicative of the association of the nuclide or pollutant of interest present in the soil; it should not be used as a method for determination of the ultimate soil particle size. All treatments should be carried out in hoods or glove boxes with forced air (100 cfm) and filtered discharge (HEPA filter).

Procedure

1. Air dry the soil sample in glove box or chemical hood approved for transuranic handling. Samples should be placed in photographic trays lined with plastic sheets.

2. When dried (determined by an aliquot whose weight remains constant), the gravel particles are separated from the sand, silt, and clay fraction with ASTM Sieve No. 10 (2 mm opening). The soil is shaken for about 5 min; to prevent loss of dust, a cover and bottom receiver are used. The gravel and remaining soil sample are weighed, their weight recorded, and the gravel kept in a container.

3. Different sand-size sieves are assembled in descending order of size; the smallest size is connected to the receiver (Note 1).

4. A 100-g aliquot of less than 2-mm particles is weighed on a plastic sheet.

5. The aliquot is carefully poured into the top sieve; the cover is placed, and the sieve assembly tapped and shaken for about five minutes to foster flow of the particles downward. This operation is conducted in a glove box or approved chemical hood.

6. A funnel is placed in the neck of a 1-gal size bottle. The funnel size should be large enough so that the sieve will "sit" midway in the funnel. For 3-in.-diam sieves, a 6-in. lip diameter funnel serves well.

7. The sieve assembly from step 5 is placed on a plastic sheet. The bottom receiver is removed from the assembly, the contents are wetted carefully to prevent dust escaping and then poured into the bottle aided by washing.

8. The finest size sieve (No. 270,53 μm opening) is then placed over the funnel. The contents are carefully wetted. The underside of the sieve is also wetted to permit flow. The particles are then washed using a wash bottle; the finer particles passing the no. 270 sieve are collected in the bottle. When washing this sieve (and the others also), a rubber policeman is used to gently stir the particles. Wash until the outflow water is relatively clear. Set the sieve in the funnel.

9. The next sized sieve (No. 120, 125 μm opening) is brought to the funnel and wetted as described in step 8. Be sure during the wetting procedure that any liquid flowing out of the sieve passes over and through the finer sized sieve(s) already treated. Wash the particles using the rubber policeman. Caution: The wash water from the larger sieves flowing into smaller sieves has tendency to collect in the smaller sieves unless the suspensions are stirred.

10. The third sieve (No. 60, 250 μm opening) is treated identically as in step No. 9.

11. The fourth sieve (No. 20, 840 μm opening) is treated similarly to the previous sieve. This being the last sieve, the washing with the policeman is continued until the wash water flowing through the sieve is clear. Caution: With three finer size sieves beneath, attention must be paid to the accumulation problem noted in step 9 in the finer sieves. During the washing, check water level in the finer sieves and stir to prevent overflow.

12. Remove the fourth sieve from the assembly, inspect bottom of sieve, and wash any particles adhering to the bottom walls of the sieve. Place the sieve on a section of aluminum foil, fold the edges of the foil to cover the bottom of the sieve. Place the foil-protected sieve in a 40°C. oven for drying.

13. The third sieve is washed until water flowing through the sieve is clear. This sieve is removed and treated in the same manner as the earlier sieve in step 12.

14. The second sieve is treated as in step 13.

15. The first sieve is treated as in step 13.

16. The funnel is washed down with distilled water. The bottom contains particles less than 53 μm in diameter (silt and clay). The suspension is stored for further segregation.

17. After the sieves are dried, they are reassembled in the glove box or hood as in step 3. The cover is placed on the top sieve and the receiver beneath the bottom sieve and the entire assembly tapped to complete segregation. In the wet sieving process, the effective diameter of the sieve openings is reduced by the water film and some particles smaller than the sieve size are retained. By dry sieving, the remaining finer particles are segregated. Tapping is accomplished using the handle end of a spatula. Material passing the sieve is dry sieved at the next lower sieve size.

18. Remove the particles from each screen, weigh, and store in capped containers. In removing the particles from the screen, the sieve is carefully inverted and the particles collected in an evaporating dish. With the sieve inverted, any particles adhering to the screen holes are gently pressed out into the evaporating dish.

19. Each screen is treated as in step 18.

20. The receiver will contain particles finer than 53 μm in diameter. The contents of the receiver are carefully wetted and added to the bottle of step 16.

21. To segregate the 53 - 20 μm diameter particles, gravity sedimentation is used. A 250-ml tall form beaker marked at the 10-cm height is required. The suspension in the bottle is stirred, and the suspension is poured into the beaker up to the 10-cm mark. The appropriate time of settling is determined by the temperature, particle specific gravity, height of fall, and size of particles. For the 20-μm size 10-cm fall, 2.65 g/cm^3 specific gravity, and 20°C., the time of settling is 4.5 min. The suspension in the beaker is stirred until all particles are swirling; the stirrer is then reversed to stop the swirling motion and the timer started. After 4.5 min, the suspended portion is poured into another 1-gal container (Note 2).

22. Another aliquot of the original suspension is added to the tall form beaker to 10-cm height. The beaker is stirred as in step 21 and timed for 4.5-min settling. The suspended portion is poured into the bottle containing the less than 20 μm particles. This operation is continued until all the less than 53 μm suspension has been transferred into the beaker.

23. The last aliquot will not fill the beaker to 10 cm. The bottle is washed with distilled water and the washing added to the beaker. This is repeated until the 10-cm mark is reached. The contents of the beaker is stirred, timed, and the suspension poured off as in the earlier steps.

24. The particles at the bottom of the beaker represent the 53 - 20 μm size fraction. This fraction is to be washed three times with distilled water filled to the 10-cm mark, stirred, and timed.

25. The settled material in the tall form beaker is placed in the oven to dry at 40°C. After drying, it is weighed, and stored in a capped container.

26. The remaining 1-gal bottle contains particles less than 20 μm diameter. The segregation of the 20 - 5 μm size fraction is accomplished using a refrigerated No. 2 International Type centrifuge. Under the same conditions as outlined in step 21, except that centrifuge tubes are used, the sedimentation time is 3.3 min at 300 rpm.

27. Steps 2 through 25 are followed substituting centrifuge tubes in place of the tall form beaker. To speed up the operation, two centrifuge tubes are used in the separation. The timing of the sedimentation is started when the centrifuge reaches 300 rpm. With experience, 300 rpm is rapidly reached by manipulating the speed control.

28. The suspension which is collected in 1-gal bottle contains particles less than 5 μm in diameter. To separate the 5 - 2 μm size, centrifugation is used except that the centrifuge is operated at 750 rpm for 3.3 min under conditions outlined in step 21.

29. The remaining suspension contains particles less than 2 μm (clay size). In desert soils the amount is about 1 - 2% and no further size segregation is made.

30. To concentrate the clay fraction, 2 ml of 1.0 M $CaCl_2$ is added to the suspension. The volume of the suspension is about 2 liters. If the suspension does not flocculate, the container is heated on a hot plate. If the flocculation appears incomplete (turbidity in the liquid phase after settling of floccules), 1 ml of $CaCl_2$ is added, stirred, and the suspension allowed to settle (Note 3).

31. The clear supernate is decanted into a 1-gal bottle and stored for analysis. The remaining floccules and water are transferred to centrifuge tubes and centrifuged at 2000 rpm for 5 min. After the final volume has been centrifuged and the supernate saved, distilled water is added, the floccules stirred, and centrifuged again at 2000 rpm for 5 min. This treatment is repeated and the less than 2-μ particles on the bottom of the tube are dried in a 40°C. oven.

32. After drying, the clay particles are removed from the centrifuge tubes, weighed, and saved in capped bottles. In most cases, clay

particles will adhere to the walls of the centrifuge tube. A brush is used to dislodge the particles and the particles saved with the original clay sample.

NOTE 1: Sieves come in different diameter and mesh sizes. The 3-in.-diam sieve has been a convenient size for the 100 g of soil used. The number of sand size fractions to be segregated depends upon need and convenience. The sand size ranges selected for the sandy desert soil was, in part, limited by the sieves available in the laboratory. The four sand sizes and sieve numbers were 2 mm - 840 μ (No. 20), 840 - 250 μ (No. 60), 250 - 125 μ (No. 120), 125 - 53 μ (No. 270).

NOTE 2: Gravity and centrifugal sedimentation procedures have been described by M. L. Jackson of the University of Wisconsin. For details of the procedures refer to M. L. Jackson, 1956. Soil chemical analysis, advanced course, University Wisconsin, Madison, Wisonsin.

NOTE 3: The supernate of several soils from the "safety shot" areas were analyzed for plutonium after $CaCl_2$ flocculation. The activity was insignificant. For a different source of plutonium, the effect of $CaCl_2$ must be evaluated.

REFERENCES

1. Day, P. R. 1955. Particle Fractionation and Particle Size Analyses." *In: Methods of Soil Analysis*, Part I (C. A. Black, Editor-in-Chief), American Society of Agronomy, Madison, Wisconsin, pp. 545-567.

2. Tamura. 1976. "Physical and Chemical Characteristics of Plutonium in Existing Contaminated Soils and Sediments." IAEA-SM-199/52. *In: Transuranium Nuclides in the Environment*. 1976. IAEA, Vienna.

IN SITU OPTICAL PARTICLE SIZE ANALYSIS OF AMBIENT AEROSOL

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(Ed. Note: Previously published in
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DESCRIPTION

The Climet Particle Analyzer* is a light-scattering instrument capable of sizing and counting particles in the range 0.5 to 10 μm in diameter. Fig. 1 shows a simplified drawing.

The small, carbon-vane pump draws particle-laden air into the unit at a fixed rate of 0.25 cfm. Clean "purge air," which has been filtered to remove particles above 0.1 μm, is introduced as a sheath around the incoming stream.

Particles next enter the view volume, defined by the cross section of the particle air stream and vertical extent of an intense light beam. The Climet view volume occupies approximately 6×10^{-4} cm^3 and is located at one focus of an elliptical mirror. For concentrations below about 35 per cm^3, particles can be assumed to enter the view volume individually.

As the particle passes through the view volume, it scatters light in all directions. A portion of the light scattered in the near-forward direction is reflected by the mirror to its other focus, where a photomultiplier tube is positioned. A light-absorbing cone blocks the direct beam and allows virtually no light to reach the photomultiplier when particles are not present in the view volume.

The intensity of each light pulse collected by the tube is a function of particle size, shape, and refractive index. Each pulse from the photomultiplier is amplified and applied to a network which linearizes the relationship between pulse height and particle size (assuming uniform spherical shape and constant index of refraction).

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*Reference to a company or product name does not imply approval or recommendation of the product by the University of California or the U.S. Atomic Energy Commission to the exclusion of others that may be suitable.

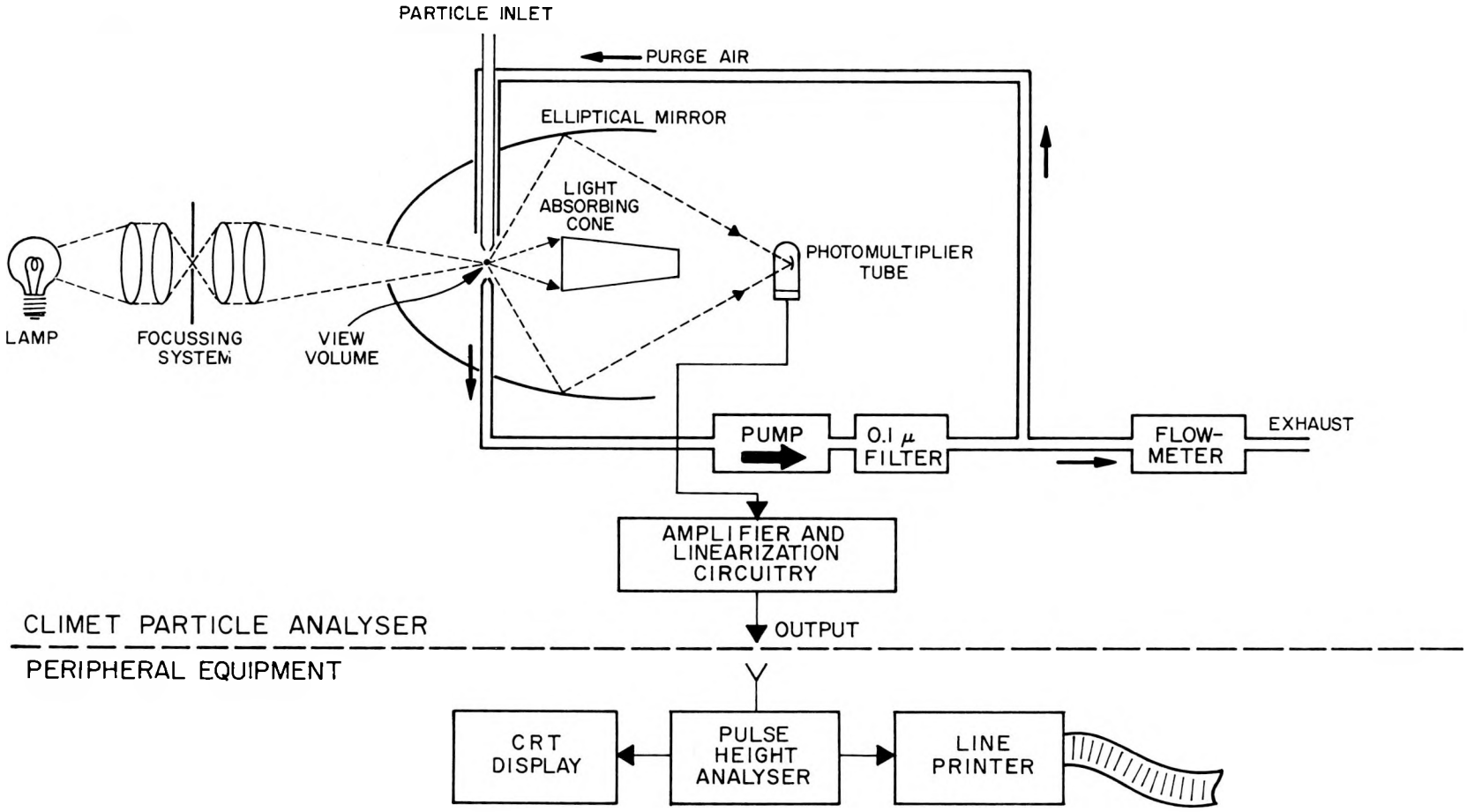

FIGURE I. SIMPLIFIED SCHEMATIC OF THE CLIMET PARTICLE ANALYZER AND ASSOCIATED EQUIPMENT.

For particle counting, the Climet pulse output is fed into a multi-channel pulse height analyzer. This instrument classifies each pulse according to amplitude, and maintains a running total in 200 channels which represents the size range 0.5 to 10.5 μm. Fig. 2 represents a typical display as seen on the height analyzer's CRT. In addition to the CRT display, a small line printer provides hard-copy output of the channel-by-channel particle count.

CALIBRATION

Although not shown on Fig. 1, the Climet Particle Analyzer includes a built-in system for field calibration. Light pulses of known magnitude are fed via light-pipe to the photomultiplier tube. In the calibration mode, output from the tube is displayed on a front-panel meter. Photomultiplier high voltage is adjusted to obtain the correct output.

Since the field calibration is only a relative one and serves to compensate for component aging, some absolute calibration is also required. For this purpose, various sizes of polystyrene latex spheres were employed in conjunction with a Royco aerosol generator. Monodisperse aerosols in seven sizes were produced and individually fed to the Climet Analyzer. Each of these aerosols were displayed on the CRT as a nearly Gaussian distribution of particle sizes. The center of each distribution was chosen as the representative size for that aerosol. A plot of the calibration curve derived from this procedure is shown in Fig. 3. The straight-line fit is reasonably good, and use of the linear relationship simplifies data reduction.

FIELD EXPERIMENTS

The Climet Particle Analyzer is configured for data collection at the Nevada Test Site (NTS), as shown in Fig. 4. The optical components, lamp, and photomultiplier are mounted separately from the air pump, flow regulators, and electronics. The two enclosures are joined by approximately 15 ft of air tubing and electrical cable. Separation of the two units allows sampling at heights up to 20 ft off the ground while still permitting convenient flow adjustment and calibration checks.

The optical unit is fitted with a wind vane coupled to the air inlet tube. Although the inlet is kept pointing into the wind by this vane, there is no simple way to maintain the inlet velocity equal to the wind speed. Maintaining equal velocities (isokinetic sampling) ensures

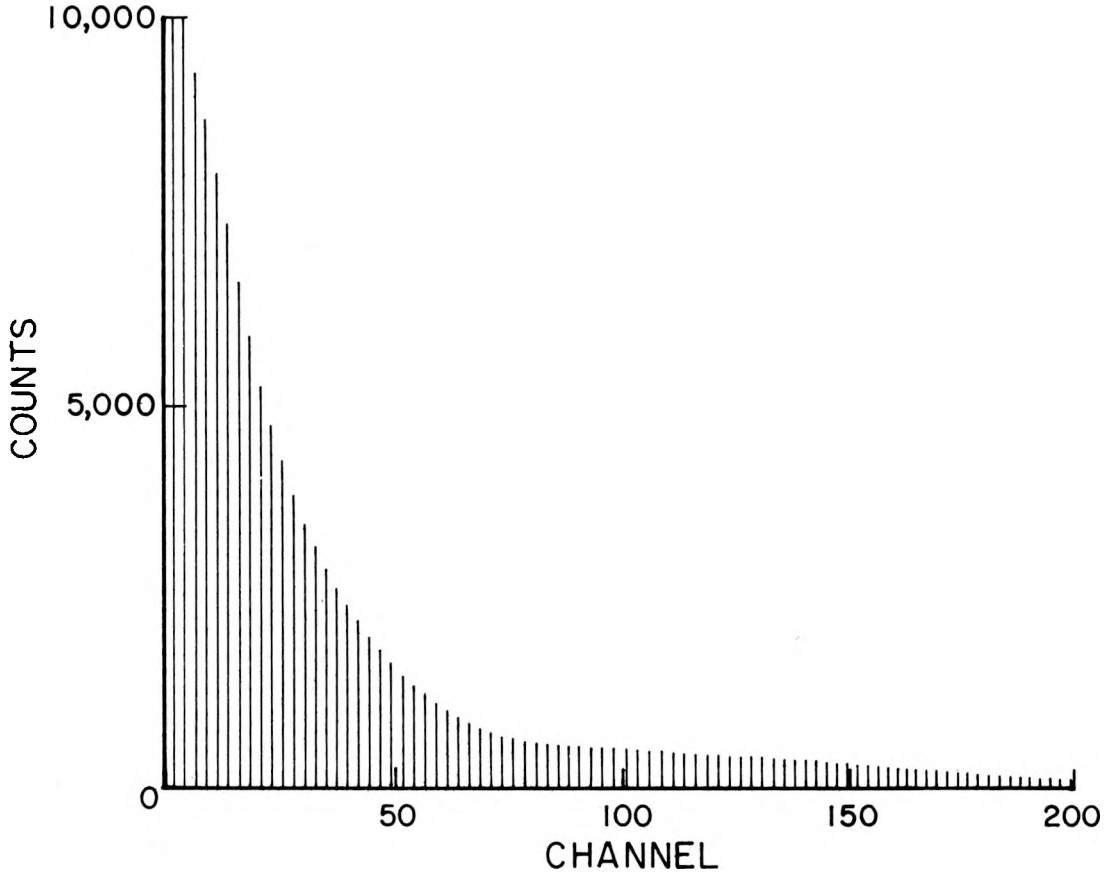

FIGURE 2. TYPICAL CRT DISPLAY AFTER A CLIMET PARTICLE ANALYZER RUN.

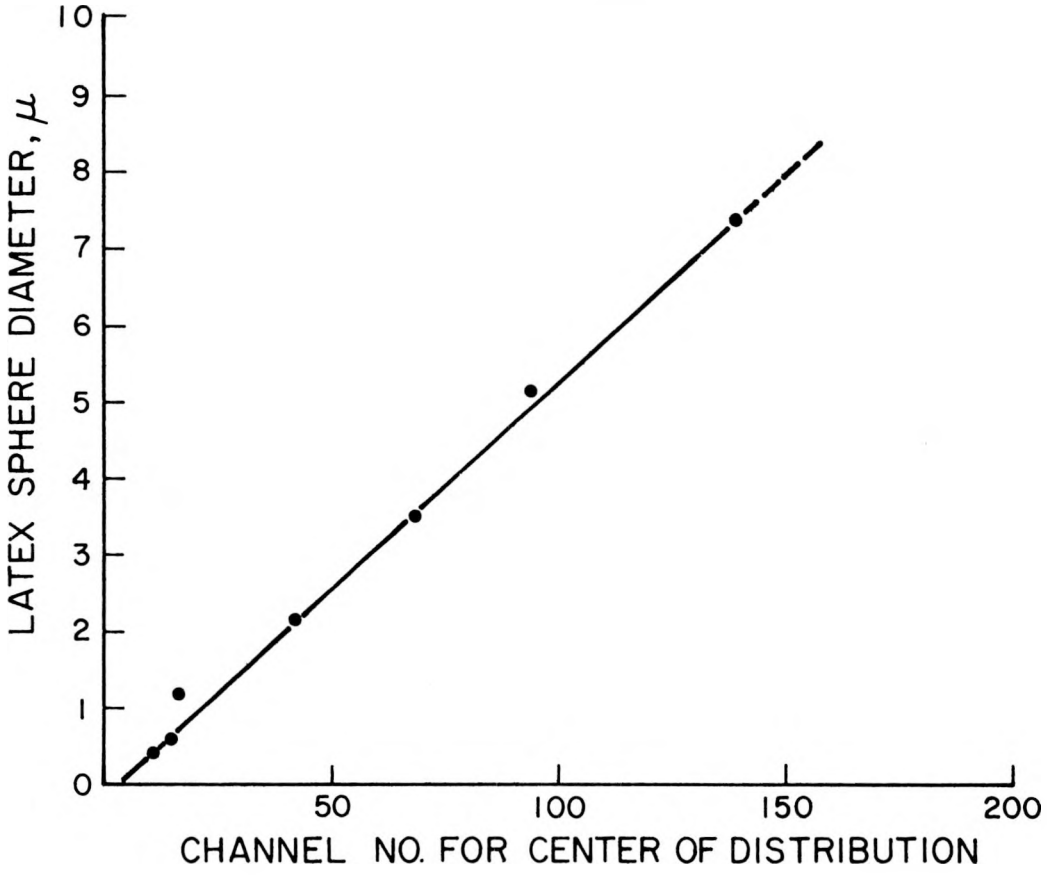

FIGURE 3. CLIMET CALIBRATION CURVE OF LABORATORY MEASUREMENTS USING SEVEN SIZES OF POLYSTYRENE LATEX SPHERES, 0.48 TO 7.3 μ DIAMETERS.

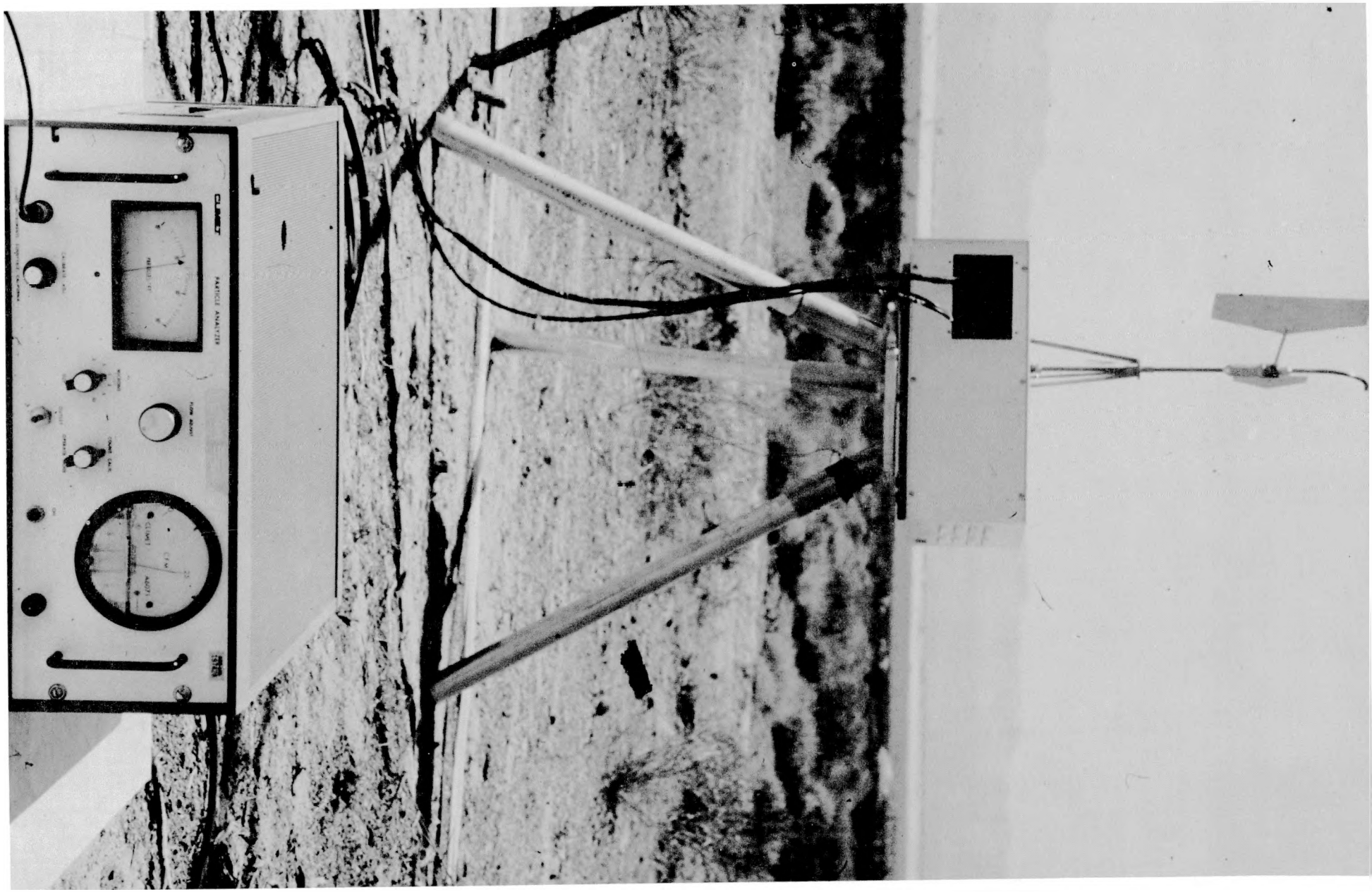

FIGURE 4. TYPICAL FIELD SETUP AT NTS FOR THE CLIMET PARTICLE ANALYZER.

that the particles which enter the inlet tube are a true representation of ambient concentrations. If the inlet velocity is significantly lower than the wind speed, a greater proportion of large particles than are actually present per unit volume of air will be collected. Similarly, fewer large particles are collected when the inlet speed exceeds that of the wind.

Since the Climet flow rate is only slightly variable about a fixed value, the intake velocity cannot be adjusted by varying the flow rate. A varying flow rate would also make it difficult to determine the volume of air sampled.

The other possibility for changing the inlet velocity involves changing the inlet diameter while holding flow rate constant. Unfortunately, attempting to accomplish this on a real-time basis as the wind speed changes is a complex problem. The present setup, described below, makes only limited use of this principle.

The wind vane is constructed to accept three different-sized copper tubing inlets. At the normal flow rate, the tubes provide intake velocities of approximately 2.5, 4, and 7 m/sec. During field operation, an attempt is made to fit the vane with the tube which has an intake velocity most closely approximating the mean wind speed. If a significant wind change occurs, the inlet tube is changed accordingly. Inlet size is recorded for each data run.

When sampling at NTS, the two Climet units are placed in an open area, usually with the main chassis on the ground and the sensor unit on a tripod. About 75 ft of coaxial cable connect the Climet to the pulse height analyzer, CRT, and printer mentioned above. These latter components are housed in a small trailer.

Following a 30-min warm-up for all equipment, the Climet field calibration and flow rate are checked and adjusted. At this time, the applicable inlet tube is installed. After all pulse height analyzer channels are cleared, pulse counting is initiated. Normally, particle counts are collected for 120 sec--timed automatically by the equipment. At the completion of the run, cumulative counts in each of the 200 channels are printed on paper tape by the line printer. The channels are again cleared, and the equipment is ready for the next run. Runs are usually scheduled each 10 min to correspond with the 10 min intervals during which meteorological data is being collected. Pertinent data, such as time, sampling height, and inlet size, are recorded manually on the paper tape for each run.

DATA REDUCTION

During the time between April 18 and August 2, 1973, data from 132 Climet Particle Analyzer runs were collected at NTS. Since the paper

tape output of the line printer does not allow any direct means of automated processing, data from all tapes were keypunched onto punch cards. Each run is now characterized by 30 cards--29 for the channel-by-channel counts, and one which contains date, time, sampling height inlet size, and any appropriate comments included by the operator.

To date, only a very preliminary data reduction effort has been completed. Computer programs are being written to operate on the large volume of particle data. Several areas of investigation are planned using the Climet data alone. These include log-normal distribution fits and analysis of concentration change with height. By far, the most important phase of the data analysis, however, is the study of correlation between the particle data and the meteorological measurements made at the same time.

Significant meteorological parameters which correspond in time with each Climet run have also been put on computer cards. Storing both the particle and meteorological data bases in the computer will allow fast, efficient manipulation of the data. The primary goal of this data analysis will be detection of correlations between particle concentration and one or several meteorological parameters. As yet, the programs required for this effort have not been completed. We have, however, manually assembled some of the data.

Fig. 5 displays data from three Climet runs which represent the full range of particle concentrations encountered thus far. The meteorological parameters listed are typical of those to be used in the correlation studies. While quantitative conclusions cannot be drawn from these limited data, it is apparent from the figure that, at least in a general sense, there exists a correlation between various wind parameters and particle concentration. It is our intent to identify and model the quantitative relationships which exist between particle loading and micrometeorology.

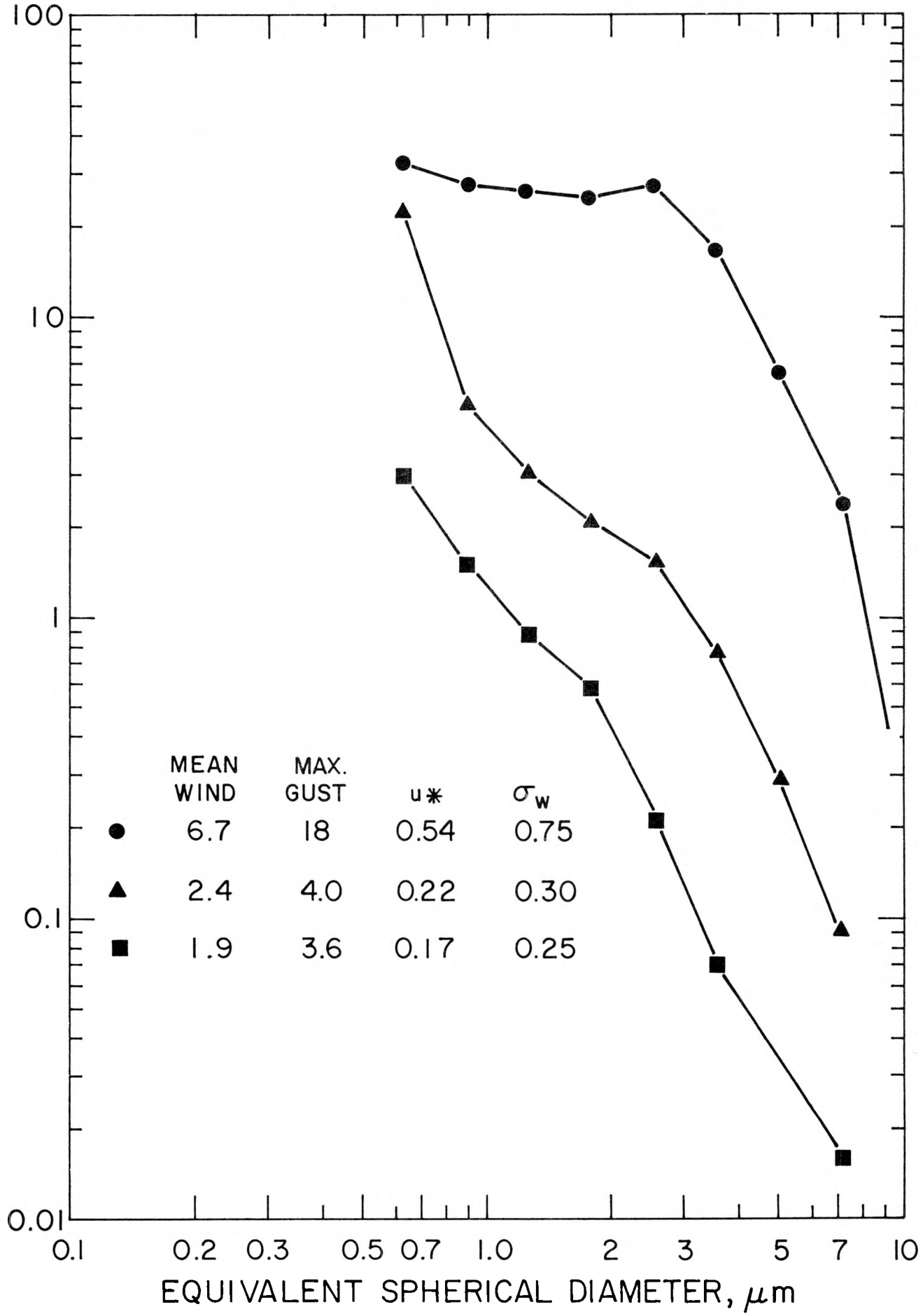

FIGURE 5. CONCENTRATION DATA FROM THREE CLIMET RUNS REPRESENTING THE RANGE ENCOUNTERED BETWEEN APRIL 18 AND AUGUST 2, 1973.

CHARACTERIZATION OF PLUTONIUM IN SURFACE SOILS FROM AREA 13 OF THE NEVADA TEST SITE*

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(Ed. Note: Previously published in
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ABSTRACT

Total plutonium was determined in nine surface soil samples (0-5 cm) from Area 13 in the Nevada Test Site (NTS). Particle size segregation was performed, and each particle size fraction of seven samples was analyzed for plutonium. The coarse silt fraction [53-20 micrometers (μm)] contained the highest percentage of plutonium in the soil (about 65%). Evidence of erosional translocation of plutonium was observed in one sample and corroborative evidence was noted in describing the soil type.

Tests with 8 molar (M) nitric acid showed that about 13% of the plutonium was leached from the NTS sample, about 70% from sediments at Oak Ridge, and about 83% from sediments at Mound Laboratory. In 0.1 M citric acid, about 1% of the plutonium was extracted from an NTS sample, 25% from Oak Ridge samples, and 44% from Mound. Implications of these results on transport of plutonium within the NTS are discussed.

INTRODUCTION

Several NTS areas have been used for testing accidental detonation of atomic devices. These high-explosive detonations, with no or little fission, dispersed plutonium over the immediate landscape. One of these tests (Project 57) is in Area 13 and is being utilized as an ecological study area. An outer fence encloses approximately 1,000 acres in Area

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*Research sponsored by the U.S. Atomic Energy Commission under contract with the Union Carbide Corporation. Publication No. 633, Environmental Sciences Division, Oak Ridge National Laboratory.

13 and an inner fence, enclosing an area of about 250 acres, surrounds the more highly contaminated area.

Soil at the site has served as the primary repository of dispersed plutonium. Since initial deposition, the area has remained relatively undisturbed for about 17 years. The area is covered with desert shrubs at 5-10% plant density and rainfall is approximately 4-8 in. per year. Experimental studies are being conducted in the area on the behavior of the plutonium, including resuspension by wind action, uptake by the desert vegetation, and concentration in mammals living and feeding in the area. To better understand plutonium translocation by these processes, information is necessary on the amount, distribution, and forms of soil-deposited plutonium.

This progress report covers results of studies of nine surface soil samples taken from a north-south transect in Area 13. Results of similar studies of several sediment samples contaminated with plutonium in the waste disposal area of Oak Ridge National Laboratory (ORNL) and Mound Laboratory (Miamisburg, Ohio) are also presented for comparative purposes.

MATERIALS AND METHODS

The locations where nine surface samples (0-5 cm) were taken initially in Area 13 are shown in Fig. 1. The samples were taken by members of Reynolds Electrical & Engineering Co., Inc. (REECo), as part of a coordinated sampling program of the Nevada Applied Ecology Group (NAEG). These samples differed from those supplied to other investigators in that they were not ball-mill ground. This allowed determination of particle size distribution. Later, three additional samples were taken at intermediate points between sampling points 6 and 7 (labeled 6A, 6B, and 6C). Analyses of these samples have not been completed at this time.

Ten-gram aliquots were used to determine total plutonium in these samples. The extraction technique used in the HASL-LASL technique described elsewhere (Tamura, 1974). Approximately 75 g of the sample were used to make size segregation of the soil. In order to minimize disturbance of the possible bonding association of plutonium with soil particles as it exists in the field, size segregation was made only with water and gentle trituration with a rubber policeman during screening. Sizes below 53 μm* were segregated by centrifugation. The fractions recovered were dried in a 40°C oven; the clay fraction, however, was obtained by freeze drying.

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*For the sake of simplicity here, particle size in micrometer (μm) refers to mass median diameter of particles in a given soil fraction.

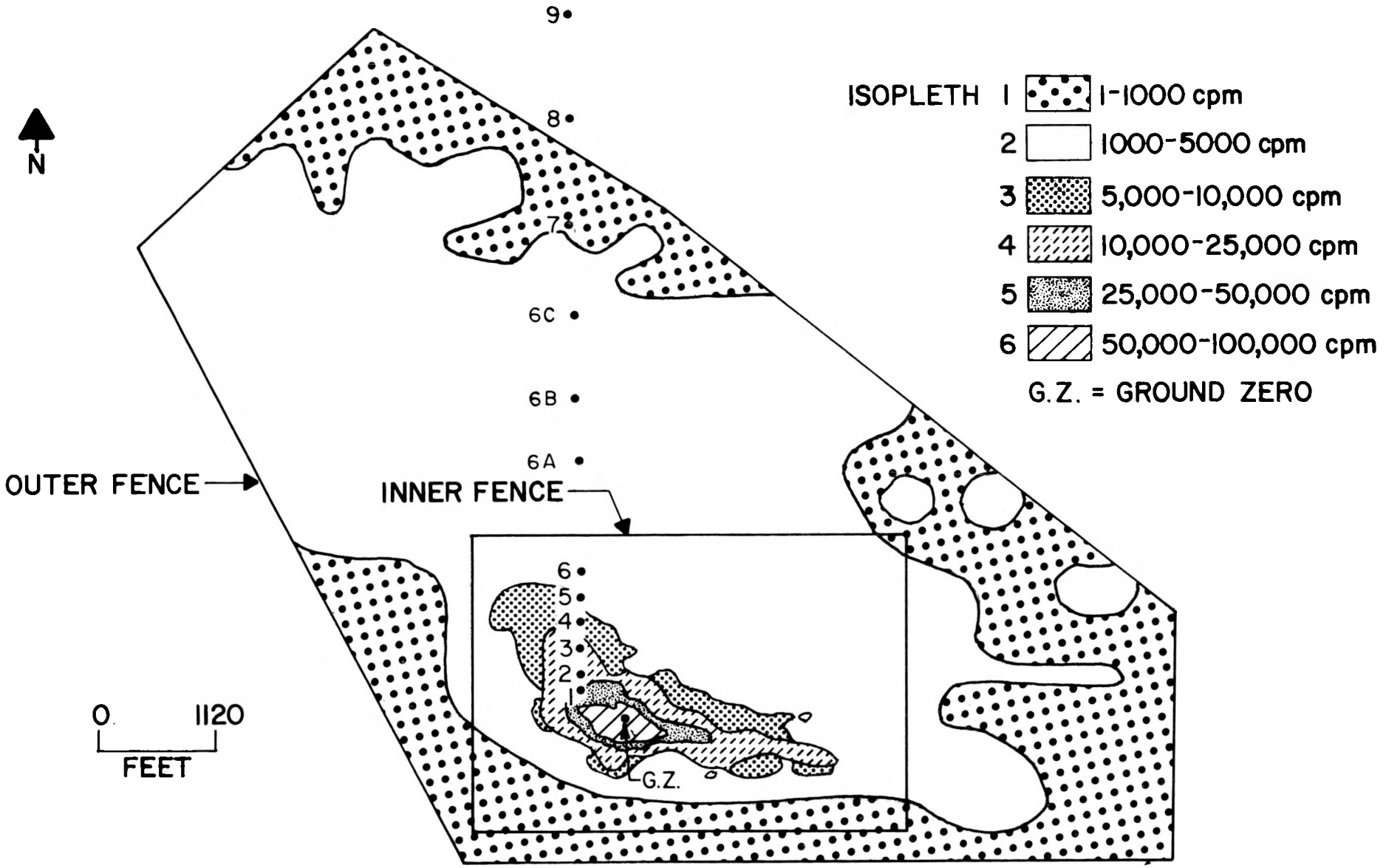

FIGURE 1.
ACTIVITY ISOPLETH & SAMPLE LOCATION IN AREA 13. LATER SAMPLINGS WERE MADE BETWEEN SAMPLE POINTS 6 & 7.

In addition to obtaining total plutonium of unsegregated soils as described above, selected samples were extracted with 8 M HNO_3 for 1 hr at room temperature. Eight M nitric acid was used since this extraction is similar to that developed by REECo (1972), thus permitting subsequent determination of plutonium without further analytical development. The technique differs only in that REECo recommends 4 hr of extraction at 90-95°C.

Several samples were also extracted with 0.1 M citric acid for 30 min at room temperature. This reagent was selected since the more soluble forms of plutonium are extractable in citric acid. Subsequent to extraction in citric acid, the solution was acidified to 1 to 4 M with nitric acid. The plutonium was extracted in tertiary amine nitrate, stripped in perchloric acid, and reextracted and counted in a scintillator solution (PBBO-naphthalene-HDEHP in toluene) using a high-resolution liquid scintillation detector. The extraction steps following acidification to 1 to 4 M HNO_3 were developed by McDowell *et al.* (1974). Spiking tests with known plutonium standards verified that acidification of the citric acid with nitric acid enabled the tertiary amine nitrate to extract soluble plutonium from the citric acid media.

In addition to NTS samples, several sediment cores were obtained from a former impoundage on White Oak Creek at ORNL and from a canal receiving drainage water from Mound Laboratory (Miamisburg, Ohio). The sediment cores were air dried and selected segments were analyzed to compare their behavior with NTS soil samples.

RESULTS AND DISCUSSION

Total Plutonium and Plutonium According to Particle Size Distribution

Prior to segregating the soils, 10 g of each sample were analyzed for total plutonium. After segregating the soil sizes in seven of the nine samples, each fraction was analyzed for total plutonium. Particles greater than 2,000 μm were not analyzed and in calculating plutonium concentration in soil, this fraction was assumed to contain no plutonium. Total plutonium in the unsegregated and segregated soil was averaged. The mean value for each sample is shown in Table 1. Since aliquots of the same original sample were analyzed by Los Alamos Scientific Laboratory and the LFE corporation, their results are also included in Table 1.*

*Data taken from letter dated November 13, 1972, from E. B. Fowler, LASL, To R. O. Gilbert, Battelle--Pacific Northwest Laboratory.

Table 1. Plutonium concentration in nine surface soil samples from north-south transect in Area 13. Activity in dpm/g of soil and standard deviation of the mean.

Sample No.	Grid Coordinate*	Distance from Ground Zero	Analysis by**		
			ORNL	LASL	LFE
1	936400 N	500	7267 ± 4.6%	6416 ± 7.5%	6720 ± 2%
2	936550 N	602	2666 ± 12.0%	1067 ± 15%	2050 ± 3%
3	936800 N	806	915 ± 14.8%	-	787 ± 4%
4	937050 N	1031	480 ± 23.1%	213 ± 39%	866 ± 6%
5	937300 N	1265	334 ± 5.7%	396 ± 15%	705 ± 10%
6	937550 N	1504	210 ± 29.0%	163 ± 31%	254 ± 4%
7	940800 N	4718	22 ± 4.5%	41 ± 24%	25 ± 1%
8	941800 N	5715	20	11 ± 21%	30 ± 5%
9	942800 N	6713	18	13 ± 19%	16 ± 2%

*Listed coordinate is north-south; east-west coordinate is 721000 E. Coordinates for ground zero (GZ) are 936098.8 N and 721402.9 E.

**ORNL refers to analysis by Oak Ridge National Laboratory (author); LASL refers to Los Alamos Scientific Laboratory; and LFE refers to LFE Corporation in Richmond, California.

Plutonium levels are seen to decrease as the distance from GZ increases (Table 1). Furthermore, plutonium levels are consistent with the Fidler* readings taken in the field (see Fig. 1). However, variability in analytical results from the different laboratories is large, and variability is ascribed to particle size of plutonium in samples. This variability as related to the particle size is discussed more fully later.

Particle-size analyses of the nine samples are given in Table 2. Normally, particle-size analysis in soils is reported for particles less than 2000 μm; however, since plutonium analyses by other participants in the program is reported for soil, particle-size percentages are reported on the entire sample for consistency of reporting. The size analyses show that the soils are high in sands (2000-53 μm) and low in clay (< 2 μm). Highest content of sands, i.e., approximately 87%, is in samples 7 and 8, and the highest clay content (4.5%) is in sample 6. Sample 6 also contains the lowest sand content (53%).

Concentrations of plutonium in particle size ranges less than 2000 μm, are presented in Table 3. Alpha activity expressed as dpm/g in each size fraction is in row A. Because of low concentration of plutonium in samples 8 and 9 and the similarity in plutonium distribution, these size fractions were not analyzed. The fraction with the highest level of plutonium is in the 20-5 μm size of sample 1; however, the highest contributor (rows B and C) to total soil plutonium is the 53-20 μm size (coarse silt). Contribution to total soil plutonium takes into account the weight distribution of each fraction given in Table 2. Although plutonium concentration in the medium silt fractions (20-5 μm) of samples 1 and 7 are higher than in the coarse silt, the lower weight of these fractions decreases their contribution to total soil plutonium.

An interesting distribution of plutonium is seen in sample 6. In all other samples the clay fraction contributed less than 2% to total soil plutonium; in sample 6, the clay fraction contributed 9%. Sample 6 was also the highest in clay content (Table 2). An inspection of the soil survey map of Area 13 (Leavitt, 1974) reveals the possible cause of this plutonium distribution (Fig. 2). Note that sample 6 is taken in an area where the soil type is labeled 405. According to the soil description, soil type 405 is similar to 401 except for a heavier texture and evidence of wind and water erosion. If erosion had removed the coarser materials and thereby enriched the clay content in the sample, plutonium in the clay fraction would be expected to be between 48 and 10 dpm/g (between the concentrations in samples 5 and 7). The extracted plutonium is 536 dpm/g which suggests that highly contaminated clay particles must have been transported to the site of sample 6. It would be interesting to analyze additional samples in soil type 405 to confirm the distribution and to attempt to establish the extent to which soil erosion may have transported plutonium from near GZ to sample point 6 and beyond through the water course.

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*The Fidler is a portable field instrument which measures the ^{241}Am gamma radiation which is correlated with plutonium content.

Table 2. Particle size distribution of nine surface soil samples from Area 13. Results expressed in percentage by weight.

Size Range (μm)	Sample No.								
	1	2	3	4	5	6	7	8	9
> 2000	1.60	6.80	10.35	7.04	10.08	8.50	3.51	3.55	3.47
2000-840	3.69	7.64	8.17	5.81	6.99	7.41	4.66	6.14	4.25
840-250	26.41	26.86	26.65	22.51	23.12	15.48	32.34	33.31	24.04
250-125	27.37	22.95	20.01	22.05	15.78	17.01	29.92	29.04	31.42
125-53	22.99	17.95	19.62	24.23	16.69	20.67	20.74	18.44	18.82
53-20	7.67	7.02	6.07	8.67	8.73	10.29	4.05	5.89	4.39
20-5	3.92	4.14	3.51	3.71	9.52	10.39	1.37	1.65	5.70
5-2	2.17	2.44	2.14	2.64	5.05	6.26	0.65	0.82	4.02
< 2	3.74	3.10	1.89	3.28	3.34	4.53	2.72	1.30	3.90
Total	99.29	98.90	98.41	99.94	99.30	100.54	99.96	100.14	100.01

Table 3. Distribution of plutonium in size fractions and contribution of size distribution to total soil plutonium.

Sample No.	Activity*	Size Range (μm)							
		2000-840	840-250	250-125	125-53	53-20	20-5	5-2	< 2
1	A	123	114	606	7386	37,341	47,852	9052	2872
	B	4	30	116	1698	2864	1875	196	100
	C	0	0.4	2.4	24.5	41.3	27.0	2.8	1.4
2	A	52	26	306	2563	20,609	6859	1384	1360
	B	4	7	70	460	1447	284	34	42
	C	0.2	0.3	3.0	19.6	61.6	12.1	1.4	1.8
3	A	27	32	29	950	11,084	3968	992	896
	B	2	7	6	186	673	139	21	17
	C	0.2	0.7	0.6	17.7	64.0	13.2	2.0	1.6
4	A	22	24	12	14	5824	1672	256	144
	B	1	5	3	3	505	62	7	5
	C	0.2	0.9	0.4	0.5	85.4	10.5	1.2	0.8
5	A	56	2	2	85	3074	637	92	48
	B	4	1	0	14	268	61	5	2
	C	1.1	0.1	0	4.0	75.9	17.2	1.3	0.4
6	A	9	1	4	6	1936	408	44	536
	B	1	0	1	1	199	42	3	24
	C	0.2	0.1	0.2	0.4	73.4	15.6	1.0	9.0
7	A	0	2	3	2	328	512	24	10
	B	0	1	1	1	13	7	0	0
	C	0	2.4	4.0	1.8	58.6	30.8	0.9	1.3

*A = dpm/g in size fraction; B = contribution in dpm to 1-g soil, activity rounded off to nearest whole number; C ercentage contribution of activity in size fraction to soil.

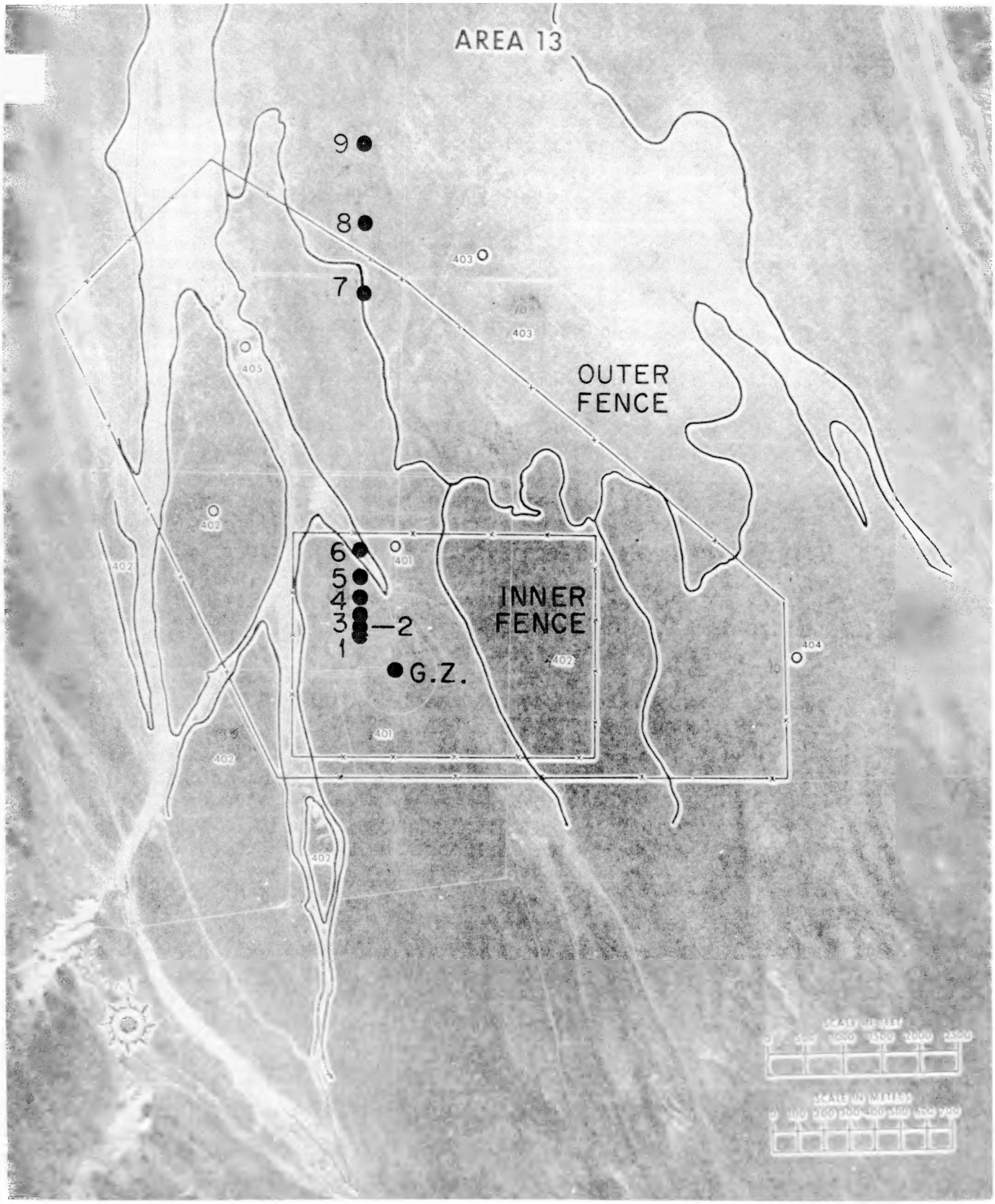

Figure 2. Soil Sample Locations Relative to Soil Type Distribution (Soil Map by Leavitt, V. D., 1974).

It should be noted that particle size segregation down to 53 μm is made by screen sieving; but size segregations at 20, 5, and 2 μm are made by application of Stoke's law. Stoke's law of settling velocity involves the density of minerals; with soils, the value of 2.65 g/cm^3 is normally used for determining the time required to segregate specified particle sizes. PuO_2 has a density of about 11.5 g/cm^3; thus the settling rate is faster for PuO_2 than for the same sized silicate minerals. Consequently, the 53-20 μm fraction which was separated on the basis of a density of 2.65 g/cm^3 could contain PuO_2 particles with diameters ranging between 53 and 8 μm.

The 53-20 μm fraction contributes a large fraction to total soil plutonium. One might, therefore, use this fraction to illustrate the inherent variability in plutonium analysis. Take the plutonium activity in sample 7 of Table 1 as an example. A value of of 22 dpm/g of soil is shown. Data in Table 3 show that about 60% of the activity is contributed by plutonium in the 53-20 μm fraction. If one assumes that plutonium exists as the oxide and that the plutonium particle has a diameter of 8 μm (particles are assumed at a lower size range which may be reasonable since the next size range contributes about 30% of the activity), the activity of a pure 8 μm sperical $^{239}PuO2$ particle would approximate 360 dpm. The value of 328 dmp/g (Table 3) was based on a 2.0-g sample; hence, the measured activity was 656 dpm/2 g. This activity level would suggest that 1.8 particles were present. If, in taking a 2-g sample the aliquot contained three $^{239}PuO_2$ particles, the activity contributed by the fraction would be 24 dpm instead of 13 dpm, and the total soil plutonium would be 34 dpm/g. On the other hand, if the sample had one particle, the activity contributed by the fraction would be 8 dpm and the total soil plutonium would be 18 dpm/g. In taking a 10-g sample of unsegregated soil for analysis, the 53- to 20-μm fraction would make up 0.4 g. In only 0.4 g there is a likelihood that no particles of plutonium would be present; in this case, assuming other size fraction contributions remain constant, the total soil plutonium could be as low as 10 dpm/g. At soil plutonium levels of this magnitude and with only a few particles to work with, there is a need for analyzing larger samples. For example, in analyzing 2 g of the 53- to 20-μm fraction, this was equivalent to analyzing 50 g of unsegregated soil. Furthermore, this helps to explain the wide range of values reported by the three laboratories.

Selected Leaching Tests of Plutonium

Earlier work with NTS soil samples suggested that most of the plutonium extracted is plutonium oxide of high specific gravity (Tamura, 1974). During exploratory field work at ORNL, plutonium was extracted from sediment samples taken in an old former lake bed. In contrast to plutonium in NTS soils, which is associated primarily with silt size particles, ORNL samples showed that a substantial amount (40%) of total sediment plutonium was associated with clay. In addition, a contaminated core was taken from the north canal at Miamisburg, Ohio, and slices of the core representing different depths were taken. In order to compare

differences in the behavior of plutonium in these three types of environmental samples, selected samples of each were leached with 8 M nitric acid for 1 hr at room temperature (28°C). The fractions leached in the acid are shown in Table 4; results show that the plutonium in ORNL and Mound sediments are more soluble than that in NTS soils in strong acid. It should be mentioned that plutonium in the NTS and ORNL samples is primarily $^{239-240}Pu$; in the Mound samples it is ^{238}Pu.

To further characterize the behavior of plutonium in NTS, ORNL, and Mound samples, citric acid was used to extract plutonium. Extraction data in Table 5 reinforce the results of the nitric acid leach; viz., that "NTS soil plutonium" is less soluble than "ORNL and Mound sediment plutonium." Note also that the plutonium in the ORNL sample is about 20 times more soluble than the NTS sample in citric acid. Furthermore, in comparing the citric and nitric acid leaches, plutonium in NTS samples is about 10 times more soluble in 8 M nitric acid than in 0.1 M citric acid; but, in ORNL samples, the plutonium is only about two to three times more soluble in 8 M nitric than in 0.1 M citric acid. These differences suggest that the chemical form of the plutonium is different in the two types of samples. If size distribution of the plutonium were the only difference, citric acid solubilization in clay size fractions (< 2 μm) of the two samples would have shown similar extractable percentages (not 3.9 and 15.6% as seen in Table 5). Further characterization of these samples and studies will be needed to define the chemical and physical forms of the plutonium in these environments.

Implications of Findings in Area 13

The relatively uniform distribution of plutonium in the coarse silt fraction of all seven samples and the relatively constant fraction leached by 8 M nitric acid suggest that the uptake factor* for the same plant species in the area should be relatively constant. An increased uptake may be expected in plants growing in areas where soil is similar to sample 6 because of the comparatively high concentration of plutonium in the clay size fraction. Heavier textured soil of sample 6 contained 9% of the plutonium activity in the clay fraction as compared to less than 1.8% in other samples.

Distribution of plutonium in different particle sizes should be useful in interpreting resuspension phenomena. Plutonium concentrations in larger particles, such as the coarse silt and sands, would be useful in understanding plutonium accumulation under bushes by saltation and creep phenomena. Plutonium concentrations in the finer sizes, which could become airborne, would be valuable in interpreting the potential hazard of plutonium which might be moving off site.

Leaching data obtained with mineral and organic acids both on the NTS, ORNL, and Mound samples should be useful in evaluating the potential

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*Uptake factor is defined as the ratio of activity per gram of plant to that per gram of soil.

Table 4. Plutonium leachability of selected soils and sediments in 8 M nitric acid. One-hour contact at room temperature (28°C).

Site Location	Soil Designation	Activity Leached (dpm/g)	Total Activity in Sample (dpm/g)	Fraction Leached (%)
Nevada Test Site	No. 2	306	2666	11.5
	No. 6	31	210	14.7
Oak Ridge National Laboratory	IRB-1	347	452	76.8
	EO-S180-1	122	196	62.2
	W45-S302-1	143	201	71.1
Mound Laboratory	747-8C (0-3 cm)	492	608	80.9
	747-8C (7-9 cm)	906	1102	82.2
	747-8C (43-47 cm)	4375	5079	86.1

Table 5. Extraction of plutonium from selected soils and sediments in 0.1 M citric acid. Thirty minutes' extraction at room temperature (28°C).

Site Location	Soil Designation	Activity Leached (dpm/g)	Total Activity in Sample (dpm/g)	Fraction Leached (%)
Nevada Test Site	No. 3 (unsegregated)	12	915	1.3
	No. 3 (53-20 μm)	124	11,084	1.1
	No. 3 (20-5 μm)	37	3,968	0.9
	No. 3 (< 2 μm)	35	896	3.9
Oak Ridge National Laboratory	EO-S180-1	32	135	23.7
	IRB-1 (53-20 μm)	78	313	24.9
	IRB-1 (20-5 μm)	75	301	24.9
	IRB-1 (5-2 μm)	70	329	21.3
	IRB-1 (< 2 μm)	79	506	15.6
Mound Laboratory	747-8C (43-47 cm)	2168	4760	45.5
	747-8C (0-3 cm)	248	577	43.0

hazard of plutonium as it exists in these environments. Lower leachability in nitric acid and lower solubility in citric acid of NTS samples, as compared with the other two samples, would suggest a lower uptake factor in plants grown in NTS soils.

FUTURE PLANS

Plans for future studies include the following:

1. Complete segregation and analysis of three surface samples (6A, 6B, and 6C) taken between samples 6 and 7 of the north-south transect and which lie between the inner and outer fence in Area 13. These samples will provide needed data to define mathematically the relationship of activity with distance from GZ.

2. Continue chemical characterization studies of soil plutonium using NTS soils as well as other environmental samples representing different sources of plutonium. These studies will enable better prediction of the long-term behavior of plutonium in the environment and more reliable assessment of the potential hazard.

3. Continue physical characterization of soil plutonium in Area 13, especially with regard to nearness to the site of detonation. Preliminary results suggest that the plutonium on particles near ground zero (600 ft or closer) are more tightly bonded to soil particles than plutonium in soils farther away.

4. Study the distribution of plutonium in soil as a function of depth. Since surface plutonium occurs largely in the coarse silt sizes, size distribution of plutonium at greater depths would provide information on the mode of downward transport. Leach tests of plutonium as a function of depth and particle size would also aid in understanding the mechanism of downward movement.

5. Sample and analyze plutonium in soil type 405 from Area 13 to establish and ascertain the extent of movement of plutonium by erosion.

ACKNOWLEDGMENTS

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DISTRIBUTION AND CHARACTERIZATION OF PLUTONIUM IN SOILS FROM NEVADA TEST SITE

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ABSTRACT

This study was undertaken to determine the distribution and characterization of plutonium in soil fractions of the Nevada Applied Ecology Group (NAEG) intensive site study area samples. This report discusses analytical results obtained on three selected surface soil samples from two areas at Nevada Test Site.

Analytical methods are described for determination of total plutonium content, plutonium distribution in different particle size fractions, and short-time digestion leachability by HNO_3. Leaching with HNO_3 revealed that 65 to 91% of the plutonium could be leached. The leaching results suggest the possibility of using the acid extraction as a means of predicting the "availability" of plutonium in soils. Preliminary data suggest that plutonium in the coarser size fractions is PuO_2, whereas plutonium associated with the finer size particles possibly is a hydrous PuO_2.

INTRODUCTION

A comprehensive environmental studies program of plutonium is being conducted by the Nevada Applied Ecology Group (NAEG) at the Nevada Test Site (NTS). This program has as its ultimate objectives the determination of the plutonium hazard existing at NTS and making recommendations of necessary cleanup of particular areas. To meet these objectives, extensive studies are being conducted on the inventory of plutonium at NTS, resuspension and redistribution of plutonium primarily by wind action, and redistribution through food chains and animal behavior.

This study is part of the effort of the soils group, whose major responsibility is the inventory of plutonium. In addition, since soil plutonium is the source term in resuspension, the reservoir for uptake by plants, and since soil is part of the habitat of animals and

microbes, information beyond inventory is desirable. The objectives of this phase of the study are (1) to identify and characterize the plutonium in the soils of NTS; (2) to define differences of behavior of the plutonium in the different areas as related to soil types, to distance and nature of the detonation device, and to different forms of plutonium; and (3) to determine the potential movement of the plutonium into the soil and/or off the site of deposition and into plants.

This report discusses results obtained on three selected surface soil samples (0- to 3-cm depth) from two areas at NTS. Both areas contain plutonium which was released to the environment by a series of "safety shots." These areas are low in fission products, since the shots were not designed to provide conditions necessary for the fission reaction.

SAMPLE DESCRIPTION AND METHODOLOGY

For these initial studies, two samples were collected from the GMX site (Area 5) and one from Area 13. The GMX site was first considered for intensive study, and several studies were initiated at this site; however, considerations of available facilities and need for larger grazing areas determined future studies to be in Area 13. The soils of these two desert areas are in the aridosol or entisol order.[a] Previous analysis had shown that about 80 to 90% of the plutonium was in the 0- to 3-cm depth zone; hence, samples were taken down to 3 cm for these exploratory studies. In GMX, one sample was taken from the bare area characterized by a weak desert pavement; the field survey instrument gave a reading of 15,000 cpm.[b] The field coordinate for this sample is 250 ft north of ground zero (GZ) and in radian No. 7, according to the grid system laid out by staff of Reynolds Electrical and Engineering Co., Inc. (REECo). The second sample was taken within 10 ft of the desert pavement sample in the same coordinates. The second sample was taken under a creosote bush; the field survey instrument gave a reading of 21,000 cpm. The soil under the bush forms a mound strikingly different from the desert pavement by the absence of gravel particles. This type of soil is commonly referred to as "blow sand."

The sample from Area 13 (grid notation F-4) was taken in an enclosed, covered area called "microplot." Vegetation studies are being conducted in the plot. The field survey instrument gave a reading of 11,000 cpm.

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[a] Soil classification by V. D. Leavitt, Soil Scientist, Environmental Protection Agency.

[b] The field survey instrument used is called the "FIDLER," which measures the 60 kev gamma rays of americium-241; the latter is correlated with plutonium content.

The sample did not contain as much gravel as the desert pavement sample, and it was more cohesive than the loose, sandy texture of the blow sand.

The soils were returned to the laboratory and placed in glove boxes. The samples were then mixed, aliquots taken, and passed through a 2-mm sieve to remove and record the gravel content. Further analyses were performed using the less than 2-mm particles. For total plutonium analysis in the soil, 10-g samples were used. For mechanical separation of particle sizes, 100-g samples were used. Particle sizes greater than 53 microns (270 mesh size) were separated by sieves. The soil was suspended in water and stirred with a rubber policeman in order to break weakly aggregated particles. No dispersant was used in order to retain the particle distribution close to that occurring in the soil. After passing the soil through the series of sieves arranged in successive sizes, the screens were washed with water from a wash bottle. The individual screens were dried in an oven at 40°C.

Particles less than 50 μ were separated by gravity sedimentation and centrifugation; cuts were made at 20-, 5-, and 2-μ diameter sizes (Jackson, 1956). The silt samples were also dried in the oven; the less than 2-μ clay size sample was freeze dried. The size of sample used for plutonium analysis differed depending on amount recovered after separation. The sand size samples which were most abundant permitted using up to 10 g; on the other hand, the fine silt and clay size samples permitted only about 0.5 g for analysis.

The analytical method used was that furnished by Mr. Gerald Hamada, formerly of REECo and now with the regulatory branch of AEC. The technical details of the method were described at the Plutonium Information Meeting held on October 3-4, 1972, at Las Vegas, Nevada. A report was submitted which contained the procedures and test results using the technique (Tamura, 1972). Briefly, the technique utilizes a mixture of concentrated HNO_3 and HF to digest the soil, followed by concentrated HCl to form aqua regia, with the remaining HNO_3 to decompose any organic matter. The acids are neutralized with NH_4OH after about 3 hr of digestion. The plutonium is dissolved in 8 M HNO_3 saturated with boric acid. After several extractions with the HNO_3-boric acid solution, the solution is neutralized with NH_4OH, the precipitate is saved, redissolved with HNO_3, and reprecipitated with NH_4OH. Final dissolution is accomplished with 8 M HNO_3. The plutonium is adsorbed after oxidation and reduction treatments on strong acid anion exchanger, eluted with NH_4I-HCl solution, evaporated to appropriate volume, and, finally, electroplated. The electroplated plutonium is counted using a solid state detector. By introducing a known amount of ^{236}Pu into the soil, the efficiency of recovery is determined, and appropriate corrections are made of the total plutonium.

Since there is question whether the acid digestion can remove all the plutonium in the soil, several runs were made by redigestion of the residue. In the three soils tested in this manner, the results showed that the first treatment removed over 95% of the plutonium from these soils. It appears that plutonium in the soil which was deposited under "safety shot" conditions is extractable by the HF-HNO_3 treatment.

Several tests were also made using a milder acid treatment. In these tests, the soils were treated for 30 min at 93°C with 8 M HNO_3, the solution was then ion exchanged on an anion resin, eluted, boiled down, and electroplated. This procedure is basically the same as REECo's method (1972), except for the shorter time involved.

Density gradient separation technique was used on two particle size samples of the microplot soil. The technique of density gradient separation has been described previously. The technique consists of preparing a solution whose density increases with depth; the sample is placed on the solution and then centrifuged to permit the minerals of different densities to reach their isodensity level. Samples recovered in this manner are referred to as bands. The bands were washed, and the relative concentration of plutonium was assayed by direct counting with a survey meter. Several fractions were also examined by a scanning electron microscope in an attempt to see if plutonium had been sufficiently concentrated for viewing and analysis by x-ray fluorescence technique.

RESULTS AND DISCUSSION

The plutonium concentrations in the soil and in the different size fractions are presented in Tables 1, 2, and 3. In addition, the percentages of the different particle size fractions are given. In regard to the total soil, the results show that the concentration of plutonium is relatively uniform. The results are expressed in disintegrations per minute (dpm) per gram of total soil, including the gravel size particles. This mode of expression is consistent with that of the total inventory studies in which the soil is mechanically ground without prior removal of gravels.

In Table 1, the particle size distribution and the plutonium in the different size fractions are reported for the desert pavement sample. The soil contains about 30% gravel particles; the remainder of the particles are primarily sand size. The highest concentration of plutonium is in the coarse silt (53-20 μ) size fraction. Although no analysis was made on the coarse sand (2,000-840 μ) and gravel (>2,000 μ) size fractions, it is believed that they contain only traces of the plutonium in this sample. Although the coarse silt contains the highest concentration, the highest contribution to the soil is from the very fine sand (125-53 μ) fraction; this is because the soil contains about 20% of very fine sand, but only 6.6% coarse silt. Together, they contribute over 75% of the activity in the soil.

The blow sand sample location is within 10 ft of the desert pavement sample. Results are shown in Table 2 for the blow sand. The obvious difference between the two samples is the absence of gravels in the blow sand. (Ignoring the gravel content, both soils contain

Table 1.

Distribution of Plutonium in Desert Pavement Soil
(Total Soil = 2,725 ± 370 dpm/g)

Size fraction (μ diameter)	Abundance (%)	Activity in fraction (dpm/g)	Activity in soil (dpm/g)	Contribution to soil (%)
>2,000	30.7	--	--	--
2,000-840	2.1	--	--	--
840-250	19.7	50	10	0.4
250-125	17.1	2,632	451	17.5
125-53	19.8	6,056	1,198	46.6
53-20	6.6	11,850	776	30.2
20-5	2.6	4,032	105	4.1
5-2	0.83	2,480	20	0.8
<2	0.67	1,698	11	0.4
Totals	100.1		2,571	100.0

Table 2.

Distribution of Plutonium in Blow Sand under Shrubbery
(Total Soil = 2,747 ± 412 dpm/g)

Size fraction (μ diameter)	Abundance (%)	Activity in fraction (dpm/g)	Activity in soil (dpm/g)	Contribution to soil (%)
>2,000	1.3	--	--	--
2,000-840	3.3	755	25	0.8
840-250	26.2	1,008	264	8.5
250-125	34.7	3,240	1,125	36.2
125-53	24.9	4,836	1,202	38.7
53-20	6.2	5,764	357	11.5
20-5	0.71	9,816	70	2.3
5-2	0.58	8,400	49	1.6
<2	0.81	2,028	16	0.5
Totals	98.7		3,108	100.0

Table 3.

Distribution of Plutonium in Microplot Soil (Area 13)
(Total Soil = 2,458 ± 310 dpm/g)

Size fraction (μ diameter)	Abundance (%)	Activity in fraction (dpm/g)	Activity in soil (dpm/g)	Contribution to soil (%)
>2,000	5.0	--	--	--
2,000-840	7.4	44	3	0.1
840-250	27.3	44	12	0.5
250-125	24.6	60	15	0.6
125-53	20.7	2,944	610	23.2
53-20	6.6	18,712	1,235	47.0
20-5	4.5	14,804	666	25.3
5-2	1.4	4,336	61	2.3
<2	1.6	1,680	27	1.0
Totals	99.1		2,629	100.0

predominantly sand-size particles.) The highest concentration of plutonium in the blow sand is in the medium silt (20-5 μ) fraction; the next highest concentration is in the fine silt (5-2 μ) size. This distribution is in contrast to the desert pavement, which had the highest concentration in the coarse silt, followed by the larger-sized very fine sand. A possible explanation for this difference may be in the nature of the soil; the desert pavement has a developed platy structure, whereas the blow sand is extremely loose. Thus, in the segregation process, the size fractions of the desert pavement may represent aggregates formed by finer sizes, whereas the blow sand represents individual mineral particles.

The small amounts of silts and clay in the blow sand sample make their contributions of plutonium to the total soil activity relatively small. The very fine sand (125-53 μ) and fine sand (250-125 μ) about equally contribute 75% of the total activity in the soil. The silts and clay of the blow sand contribute 16% of the total activity; the silts and clay of the desert pavement contribute 36% of the total.

In Table 3, results are presented for the microplot sample. This sample was taken in Area 13, approximately 40 miles north of the two samples from Area 5. Like the soils from Area 5, this soil is also very sandy (Table 3). The highest concentration of plutonium is in the coarse silt fraction, followed by the medium silt fraction. Although the amount of these size fractions are low, they contribute 72% of the total activity.

In comparing the size distribution of plutonium in the three soil samples, the amount of plutonium contributed by the silts and clays follows the order, microplot (76%) > desert pavement (36%) > blow sand (16%). It is interesting to note that studies conducted of samples from Yucca Flat, NTS, showed that most of the plutonium was associated with particles greater than 44 μ in diameter (Mark, 1970). The field survey instrument gave readings of 11,000, 15,000, and 21,000 cpm, respectively, for microplot, desert pavement, and blow sand. The analysis of plutonium gave 2,458, 2,725, and 2,747 dpm/g, with standard deviations of approximately 15%. A possible explanation of the difference in the field instrument readings and the concentration of plutonium in the soils may be in the nature of the plutonium and the associated ^{241}Am. Since the microplot sample contains 76% of the total plutonium in the silt and clay size fractions; the desert pavement 36%; and the blow sand 16% in these fractions, the smaller particles of plutonium with the associated ^{241}Am would occupy interstices of the sand grains. And, it is possible that the measured 60 kev x-rays of americium originating from the small particles is attenuated more than the americium associated with the larger plutonium particles.

Another aspect of this study is to describe the behavior of the plutonium. Studies are being conducted on the uptake of plutonium by vegetation in the desert. In this regard, it would be useful to have an index of "availability" of the plutonium using milder extraction

procedures, and to relate the extracted amounts with amounts found in the plants. For this preliminary evaluation, 8 M HNO_3 was used; the soil was digested for 30 min at approximately 93°C. This procedure is a modified REECo procedure (1972) which normally requires digestion for 2 hr. The results of the 30-min digestion is given in Table 4. In addition to the percentage leached from the 8 M HNO_3 extraction, the percentage of plutonium in the particles less than 125 μ, and that in particles less than 53 μ, are included for comparison. The results show that extraction is a function of the amount of finer-sized plutonium particles present in the sample. The higher surface area of smaller particles would be more susceptible to dissolution. Since "availability" is a function of particle size, it appears that this test might be used to predict the "availability" of plutonium in these soils which have a similar source term.

Two particle size fractions from the microplot samples were further investigated by density gradient separation. The 20- to 5-μ size and the less than 2-μ sizes were selected; the results are shown in Table 5. In the 20- to 5-μ size, the bulk of the particles had densities less than 2.8 g/cc, but contained only about 15% of the plutonium. The high content of plutonium in the heaviest fraction suggests that the plutonium exists separately as the oxide or as oxide coating on dense material. In the clay size fraction, however, the bulk of the particles are less than 2.2 g/cc, and the plutonium is associated with these particles. This suggests that the plutonium is adsorbed on the clays, probably as the polymeric form of the oxide.

Since the high density fractions contained most of the plutonium, an attempt was made to see if particles of plutonium might be visible by scanning electron microscopy. The micrographs in Fig. 1 were taken from the heavy fraction of the 53- to 20-μ fraction of the desert pavement soil. Like the microplot sample, the heavy fraction of this particle size sample was also high in plutonium. The particles as seen by the scanning electron microscope is shown in (a) of Fig. 1. The remaining micrographs are elemental analysis of the same particles which were analyzed by x-ray fluorescence using the imaging mode technique. The three elements most abundant in the sample were selected for the figure. They are labeled (b) iron, (c) calcium, and (d) silicon. Also detected were titanium, aluminum, chromium, manganese, and nickel. No measurable plutonium was noted in fluorescence spectra. Plans are under way to concentrate the plutonium even further using a solution heavier than 2.8 g/cc. It is hoped that this will permit analysis of plutonium.

SUMMARY AND CONCLUSIONS

Three surface soil samples, contaminated with plutonium from the test shots at the Nevada Test Site, were analyzed for total plutonium

Table 4.

Comparison of Plutonium Extracted by Leaching with HNO_3 and Size Distribution of Plutonium

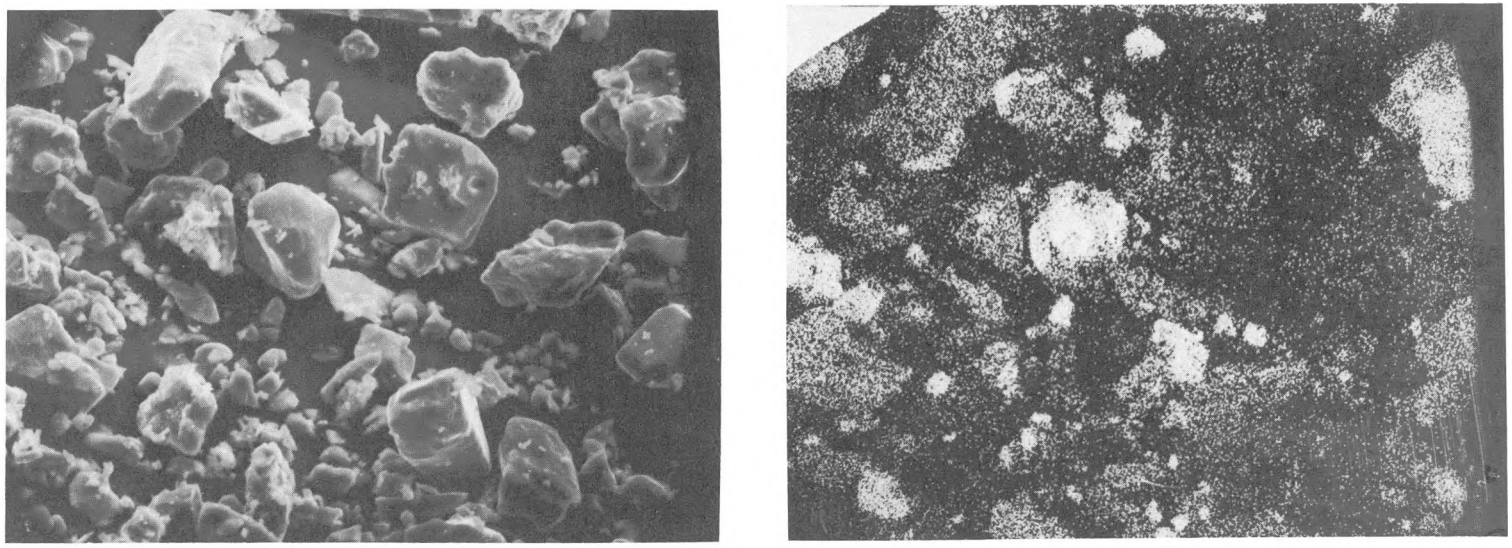
	HNO_3 (dpm/g)	Total (dpm/g)	Percentage leached	Plutonium activity in particles less than	
				53 μ	125 μ
Microplot	2,230	2,458	91	76	99
Desert Pavement	2,180	2,725	80	36	82
Blow Sand	1,780	2,747	65	16	55

Table 5.

Density Separation and Plutonium Distribution in 20-5 μ and <2 μ Fractions of Microplot Soil (Area 13)

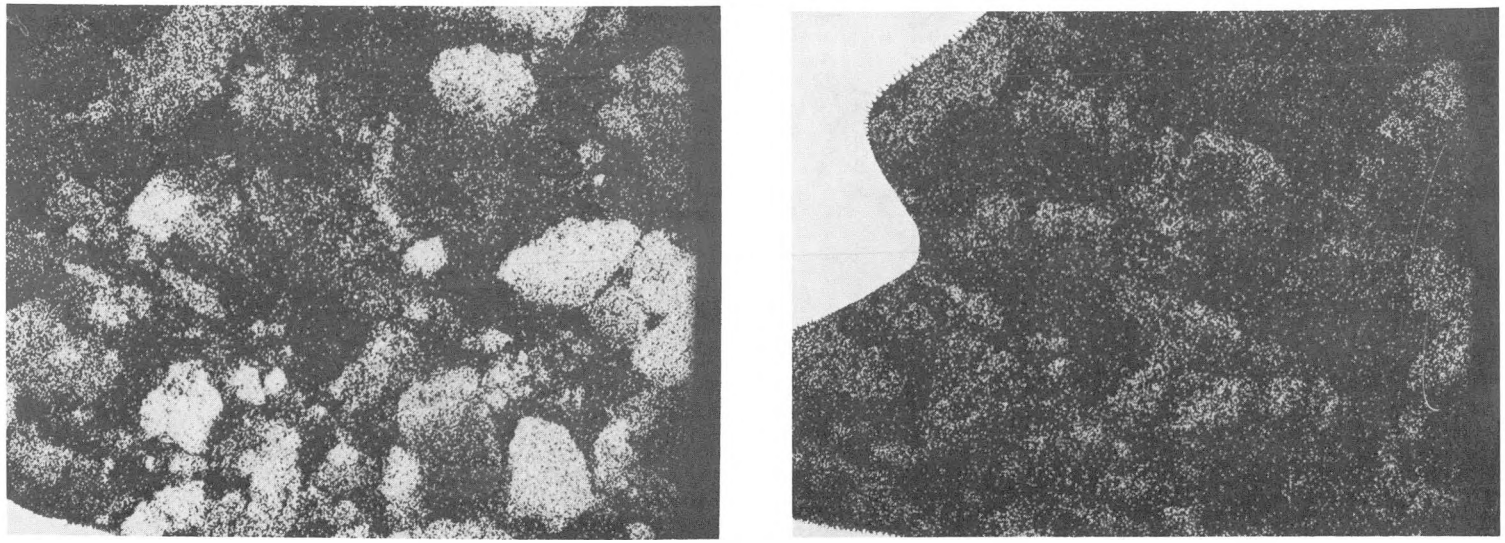
Band No.	Density (g/cc)	Amount (%)	Activity (%)
	20-5 μ		
1	<1.8	2.4	0.2
2	2.3-2.5	32.2	5.3
3	2.5-2.8	59.6	9.3
4	>2.8	5.8	85.2
	<2 μ		
1	<1.8	1.0	0.4
2	2.1-2.2	97.4	98.6
3	2.2-2.7	1.3	0.8
4	>2.8	0.3	0.2

PHOTO 2133–73



(a) MICROGRAPH (b) IRON

50μ



(c) CALCIUM (d) SILICON

Figure 1. Heavy Fraction ($>2.8\ g/cm^3$) of Desert Pavement Soil.

content, plutonium distribution in different particle size fractions, and leachability by HNO_3. Although the field survey instrument had indicated that the plutonium content might differ by a factor of 2, analysis revealed that the plutonium content was relatively uniform. The highest concentration of plutonium in the microplot soil was in the coarse silt (53 to 20-μ diameter) fraction (47% of the total plutonium), and 76% of the plutonium was in the silt (53-2 μ) and clay (<2 μ) fractions. In the desert pavement soil, the highest plutonium concentration (47% of the total) was in the very fine sand (125-53 μ) fraction; the silt and clay size fractions contained 36% of the total plutonium. In the blow sand sample, the highest concentration (39% of the total) was also in the very fine sand (125-53 μ) size fraction; the silt and clay size fractions contained 16% of the total plutonium. The size distribution of plutonium was offered as an explanation for the different readings of the field survey instrument.

Leaching with 8 M HNO_3 for one-half hour at 93°C revealed that 91% of the plutonium was leached from the microplot soil, 80% from the desert pavement soil, and 65% from the blow sand. The order of leaching is consistent with the amount of plutonium found in the greater surface area particles of silts and clays in these soils.

Density gradient separation of silt and clay fractions revealed that the plutonium in the silt fraction was associated with particles greater than 2.8 g/cc (heaviest fraction), whereas in the clay fraction, the plutonium was in the 2.1 to 2.2 g/cc fraction. This distribution suggests that the plutonium in the silt size fraction is probably present as an oxide, whereas in the clay size fraction, it may be a polymeric form adsorbed on lighter clay minerals.

Scanning electron microscope studies of the heavy fraction of the silt size particles did not reveal detectable amounts of plutonium. The heavy fraction contained primarily iron, calcium, and silicon. The leaching results suggest the possibility of using the acid extraction as a means of predicting the "availability" of plutonium in soils.

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DETERMINATION OF ^{239}PU AND ^{241}AM IN LARGE NAEG* VEGETATION SAMPLES

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ABSTRACT

A method has been developed at this laboratory for analyzing ^{239}Pu and ^{241}Am in various types of woody vegetation from NAEG* collection sites in amounts ranging from 300 to 500 g dry weight. Special dry ashing techniques are used initially to eliminate carbonaceous material. A one-gallon metal paint can, covered with perforated aluminum foil, is used as a disposable container to perform initial drying (110°C) and carbonization steps (250°C). Ashing is then completed in a Pyrex glass beaker (600°C), also covered with perforated foil. The sample ash is treated with HNO_3-HCl plus H_2O_2. Any insoluble residue is filtered and treated with HF and HNO_3 in a soil-type dissolution procedure, since tests show that a variable amount of undissolved plutonium and americium remains in the residue. The vegetation is thus reduced from its large, irregular bulk to a small volume of homogeneous solution. All or a portion of the dissolved sample is transferred to a counting vial for instrumental measurement of ^{241}Am via its 60-kev gamma emission. Uncertainties in counting such low-energy gammas in inhomogeneous samples are essentially eliminated and a standard counting geometry is achieved.

Radiochemical isotope dilution analysis is performed for ^{239}Pu using ^{236}Pu tracer. Also, if ^{241}Am is too low for instrumental measurement or a confirmation of the instrumental measurement is required, isotope dilution analysis for ^{241}Am is performed using ^{243}Am tracer. Comparisons are made between radiochemical and instrumental analyses of ^{241}Am.

Plutonium is isolated from a sample aliquot on an anion exchange resin column. Americium is isolated from another aliquot in an HNO_3-methanol anion exchange resin column. Plutonium and americium are finally electrodeposited on stainless steel and measured by alpha spectroscopy. Tracer recoveries for plutonium range from 60 to 80%, with americium slightly lower.

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INTRODUCTION

It is estimated that by 1980, 25% of the nation's electrical generating capacity will be supplied by nuclear reactors, many of the plutonium breeder type. As a result of this and the "cleanup" programs to turn nuclear test site lands back to public domain, extensive environmental surveillance programs are being conducted at these sites to determine traces of residual radioactivity. A primary function of the program is the collection of vegetation and soil samples for instrument and radiochemical analysis. Investigators are particularly interested in determining the amounts of ^{239}Pu and ^{241}Am contained by vegetation in the nuclear test areas, because grazing animals in the food chain are involved. Only the branches and foliage of the vegetation are collected, as the roots are not usually eaten. Large or composite samples are usually taken to provide a more representative sample.

A method was recently developed at this laboratory for analyzing ^{239}Pu and ^{241}Am in woody vegetation samples from nuclear test sites, ranging from 300 to 500 g dry weight. In this method, special dry ashing techniques are used initially to eliminate carbonaceous material. The vegetation sample is carbonized and ashed under controlled conditions, and the ash is completely dissolved. All or a portion of the dissolved sample is gamma-counted directly for measurement of ^{241}Am. Uncertainties in counting such low-energy gammas in inhomogeneous samples are essentially eliminated and a standard counting geometry is achieved. Plutonium and americium are assayed by radiochemistry and alpha spectroscopy on separate aliquots. Comparisons are made between radiochemical and instrumental analyses of ^{241}Am.

Several hundred NAEG vegetation samples, up to 500 g dry weight, were analyzed for ^{239}Pu and ^{241}Am by the method described. Total radioactivity in the branches and foliage was determined, and no attempt was made to determine the amount of radioactivity associated with each.

PROCEDURE

Composites or individual vegetation samples collected in the field for instrument or radiochemical analysis are bulky and, by necessity, often compacted into a large container, such as gallon-sized metal paint cans used for NAEG samples. Samples could be ground to reduce the size, but this presents a problem as the grinder must be decontaminated after each operation. Also, the vegetation sample must be further reduced in size to prepare it for analysis. In this laboratory's method, the lid is removed from the paint can sample and the

open end covered with perforated aluminum foil (Fig. 1). The vegetation sample is dried in a drying oven at 110°C and weighed by difference from a previously tared can. The sample is carbonized at 250°C in the drying oven overnight. The sample is cooled, foil removed, and a double-wall plastic bag is fitted over the can opening. The carbonized vegetation is transferred to the bag and crushed to a powder form. The sample, now considerably reduced in size, is placed in a tared Pyrex glass beaker and ashed at 600°C for two days in an ashing furnace. The beaker is rotated periodically to expose unreacted carbon and decrease ashing time. After the weight of ash is obtained, the ash is leached with hot HNO_3-HCl and H_2O_2. A residue usually remains at this point and the mixture is filtered. The filter is ashed and dissolved with HF, HNO_3 plus HF, HNO_3 + H_3BO_3, HCl, and finally HNO_3 (Major *et al.*, 1971; Major *et al.*, 1964). All or a portion of the dissolved sample is transferred to a 7-cm counting vial and the 60-kev gamma ray of ^{241}Am is measured by pulse height analysis on a 3-in NaI(Tl) wafer low-background detector. The system is calibrated with a 6N HNO_3 standard solution of ^{241}Am in the 7-cm counting vial.

An aliquot of the dissolved sample is taken for ^{239}Pu analysis (Fig. 2). Sample activity is equilibrated with ^{236}Pu tracer and the solution poured onto a Dowex 1 x 4 anion exchange resin column. The column is washed with HCl, 6N HNO_3, and HCl. Plutonium is eluted with HCl-NH_4I. Another aliquot (Fig. 3) is equilibrated with ^{243}Am tracer and poured onto a Dowex 1 x 4 HNO_3-methanol anion exchange resin column. Iron passes through and the americium is eluted with 6N HNO_3. Plutonium and americium are finally electrodeposited on stainless steel for alpha spectrometry measurement. The detection limit for ^{241}Am by instrument analysis is 30 dpm. The detection limit for ^{239}Pu and ^{241}Am by radiochemistry is 0.05 dpm.

EXPERIMENTAL

It is known that plutonium resists leaching by mineral acids from high-fired soil samples. It was thought this might also be true of vegetation samples from nuclear test sites subjected to furnace ashing at high temperatures. To determine this, 13 large vegetation samples from a nuclear test site were ashed and leached with hot HNO_3-HCl and H_2O_2. The solutions were filtered and the filtrate and residue were equilibrated with ^{236}Pu tracer and assayed for ^{239}Pu by the soil dissolution and radiochemical method of this laboratory.

To test our method for determination of ^{241}Am in vegetation, five large vegetation samples were ashed and leached with hot HNO_3-HCl and H_2O_2. The solutions were filtered and the filters ashed, dissolved, and added to the filtrate. An aliquot was equilibrated with ^{243}Am tracer and assayed by the radiochemical method described earlier.

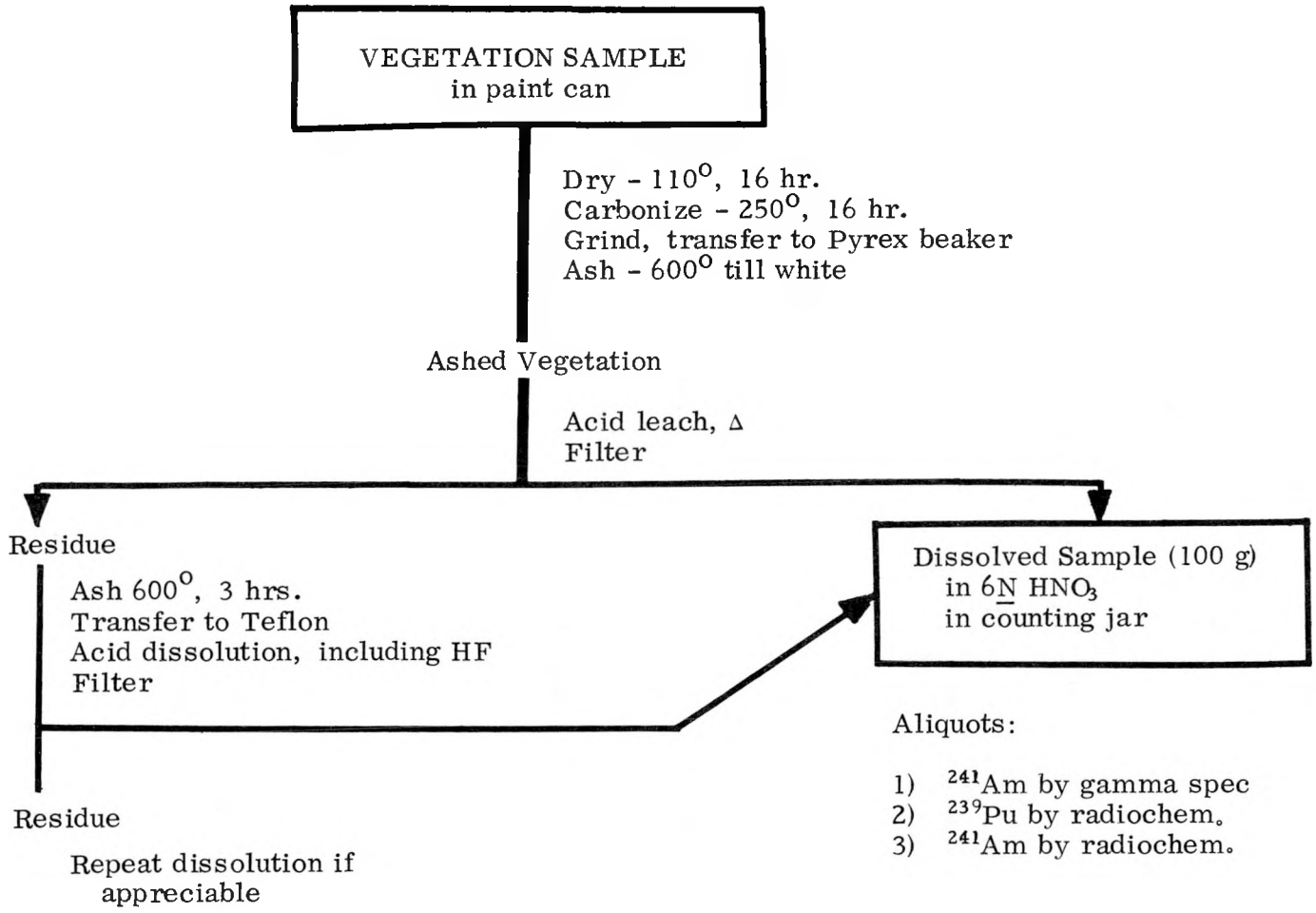

Figure 1. Sample Preparation

Figure 2. Plutonium Radiochemistry

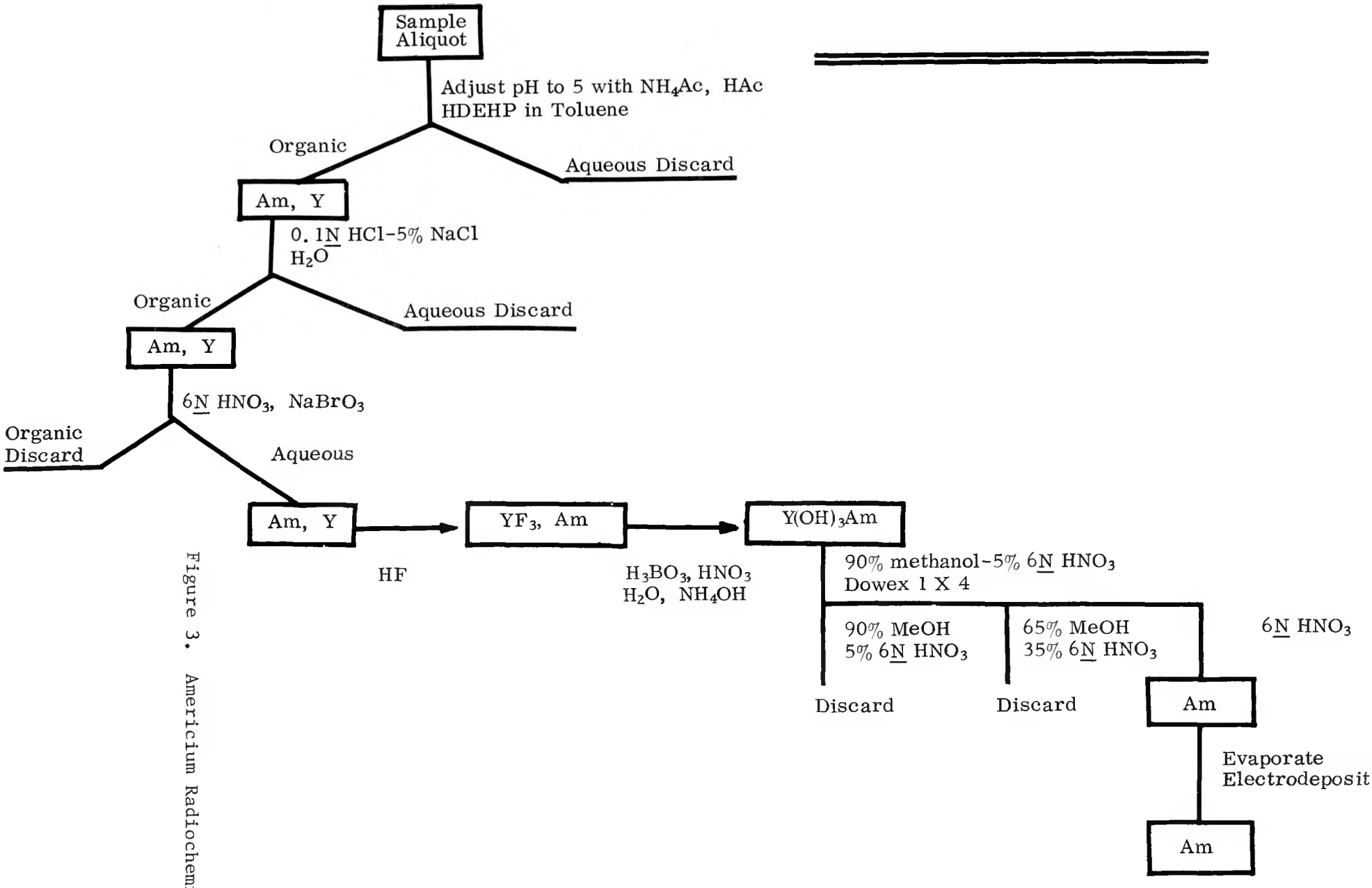

Figure 3. Americium Radiochemistry

To determine the best method for accurately measuring ^{241}Am by instrument analysis, a dissolved rather than a solid sample was analyzed, as we had difficulty obtaining a reproducible geometry of the latter. A 100-g aliquot of each of the samples analyzed for ^{241}Am by radiochemistry was transferred to a gamma counting vial and counted on the 3-in NaI wafer. The dpm was calculated by comparing with a standard consisting of a ^{241}Am spike in 100 g of 6N HNO_3.

RESULTS AND DISCUSSION

Results of the leaching test for plutonium are given in Tables 1 and 2. As shown, an appreciable amount of plutonium is retained by the residue and is quite variable, ranging 0 to 41%. Dry sample weights ranged from 120 to 440 g and ash weights ranged from 10 to 34 g. There was no apparent correlation of quantity of ash with the amount of plutonium retained in the undissolved residue. The routine procedure adapted for these vegetation samples incorporates a leach of the residue followed by a total dissolution of any remaining material.

It was not determined why part of the plutonium is retained in the vegetation residue. It may be that silicious material (known to entrain plutonium) was present either from windblown dust or in the plant cellular structure. Certainly, the residues were easily solubilized and the plutonium content determined by our soil dissolution and radiochemical method. The source of the radioactivity, whether from fallout or uptake through the root system, was not identified in this work.

Results of the method test for ^{241}Am are given in Table 3. Comparison of instrument and radiochemical values shows agreement between the two procedures is good within the counting statistics. Comparing data of duplicate samples 4a and 4b show ^{241}Am values can be reproduced by instrument and radiochemical methods with good results.

SUMMARY

Carbonizing the vegetation first reduces it to brittle charcoal, which is easily crushed into a smaller volume. Transferring directly to a plastic bag prevents escape of any charred vegetation. Each ashing furnace could hold five samples, but the ashing process is relatively slow. It is recognized that different ashing procedures and equipment could result in more rapid ashing. However, a major objective was to prevent formation of smoke and soot and resultant losses

Table 1.

Plutonium Left in Vegetation Ash
After Acid Leaches*

Pu in Leach (dpm/g ash)	Pu in Residue (dpm/g ash)	Pu in Residue (% of Total)
118	22	16
47	19	29
151	37	20
44	0	0
63	3	5
231	66	24
354	249	41
69	3	4
54	0	0
174	20	10
284	5	2
326	0	0
355	6.2	2

*Leached with HNO_3-HCl and H_2O_2.

Table 2.

Plutonium Left in Vegetation Ash
After Acid Leaches*

Pu Left in Residue (%)	Number of Samples
0 - 5	7
5 - 20	3
20 - 30	2
30 - 50	1
<50	0

*Leached with HNO_3-HCl and H_2O_2.
Ash weights range 10 to 34 g.

Table 3.

Americium-241 Instrumental Vs.
Radiochemical Results

Sample No.	Instrumental (dpm ^{241}Am per sample)	Radiochemical (dpm ^{241}Am per sample)	Ratio (Instru/R'chem)
1	124 ± 25%	(81 ± 12%)*	(1.5)
2	635 ± 9%	791 ± 10%	0.80
3	653 ± 2%	706 ± 6%	0.93
4 a**	388 ± 3%	444 ± 5%	0.87
4 b**	388 ± 3%	438 ± 4%	0.88

*No. 1 has low tracer yield.
**No. 4 a and b are duplicate aliquots.

Note: Instrumental on 95% aliquots, radiochemistry on 20% aliquots.

of plutonium and americium entrained in their carbon particles. This was accomplished by decomposing the vegetation during the carbonizing and ashing processes, and outgassing decomposition products below ignition temperature in inexpensive ashing containers which were discarded after one use.

No attempt was made to quantitatively determine the ^{241}Am in either the filtrate or residue portion of the vegetation samples. A few residues were assayed for ^{241}Am by instrument analysis and the results indicated the residue contains ^{241}Am in amounts similar to that of ^{239}Pu.

The determination of ^{239}Pu and ^{241}Am in large NAEG vegetation samples requires a method which results in complete solubilization of the ash. The instrumental method for ^{241}Am appears to be valid, as there is good agreement between radiochemical and instrument measurements. However, the instrumental method is known to be subject to more variables and is known to be less sensitive than the radiochemical method.

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ANALYSIS OF VEGETATION COVER IN CERTAIN Pu-CONTAMINATED AREAS USING AERIAL PHOTOGRAPHY

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ABSTRACT

Two methods of estimating vegetation cover were developed using aerial photographs; both are less expensive and do not contribute to disturbance of the areas compared to standard methods of measuring vegetation cover on the ground. Cover values for five Pu-contaminated areas at Nevada Test Site and Tonopah Test Range are presented.

INTRODUCTION

Vegetation cover in the Pu-contaminated areas at Nevada Test Site (NTS) is of interest for at least two reasons. First, shrubs accumulate fallout particulates at their bases as a result of characteristics of the air movement among them. Second, reclamation or decontamination of these areas is more complex because of vegetation.

Values for vegetation cover can be obtained in a number of ways. Traditionally, they require extensive field work for the required measurements. However, ground work is time-consuming, disruption of the environment may be significant, and there may be hazards to personnel, as in Pu-contaminated areas.

This report provides data on vegetation cover derived from two methods of analysis of aerial photography, which greatly reduces work on the ground. It also estimates error sources in providing these values and indicates other advantages and disadvantages of the method.

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METHODS

Over each area studied (Area 5-GMX, Tonopah Test Range--Roller Coaster, Area 13, and Area 11) near midday, aerial photographs were taken with a four-camera system. The cameras photographed in standard color, in the green band, in the infrared (IR) produced as black and white, and in infrared false color. Comparison proved the IR black and white record most useful; it showed almost as much information as color and is much cheaper to produce. Certain localities were chosen as characteristic of the particular vegetation of an area; and from the black and white IR images, 5- by 7- or 9- by 9-in transparencies were made so that the resulting enlargements produced an image of about a 1000:1 scale. These transparencies were examined by two experimental methods for determining the area covered by vegetation.

Analysis with a Calibrated-Stage Microscope

In an 8- by 10-cm template made of brass sheet stock about 0.4-mm thick, five circular holes with mean diameters of 9.5180 mm ± 0.0094-mm SD were milled for making measurements by readings of microscope stage displacement. This template was laid over the transparency, and the two were placed on the movable stage of a Hauser P320 microscope whose movements in both the X and Y axes are calibrated for displacements of 0.0001 in (0.00254 mm).

By thus studying the template and film transparency through the microscope, the numbers of shrubs in each 9.5-mm circle could be counted and their diameters measured. Shrubs more than half inside the circle were counted; more than half outside were not. From the diameters, the areas covered by the shrubs could be estimated (assuming the shrubs to be circular, viewed from above). The areas of all the shrubs within a circle were added and the total was divided by the areas of the circle and multiplied by 100. This provided a percentage of the area covered by the vegetation within each circle. Since the scale of the aerial photographs was known from REECo surveys of the Pu-contaminated areas, these measurements could, therefore, be calculated as area on the ground, as well as relative areas on the film image. A discussion of possible errors in the process follows.

On the basis of a photograph on the scale of 1000:1, a template with 9.5180-mm-dia holes allows counting shrubs in an area of 71.15 m^2 on the ground. (On almost all photographs, the scale was such that the areas counted were about 100 m^2.)

Because replicate areas were utilized, the values could be reported with standard deviations, thereby providing a statistical approach for showing differences between areas.

There are several sources of error in providing absolute numbers for shrub measurements. A very samll error may arise from the ground surveys on which the ground area scale was based. Photographic optics might also contribute, and paper stretching in print production may be a factor. Since these are probably small contributions, they are not discussed in detail. Moreover, once data are reported as percent cover, errors of scale are canceled. Even where these data are reported in units of area, the error will probably not be any larger than those inherent in measuring shrub size on the ground by conventional field methods. Suffice to say that if the measurements by microscope stage incremental movement were the sole source of error on a photograph of 1000:1 scale and the error in measuring shrub size is the same as measuring a machined opening in a template, that is ±0.0094 mm for a 9.5180-mm opening, the distance on the ground surface becomes ±0.099%, which equals ±9.4 mm in 9.5 m.

Isodensitometer Analysis

This method utilized an instrument produced by Spatial Data Systems (Goleta, California) called Datacolor Isodensitometer System. Two models were used: an older instrument at LLL, and a new one at EG&G, Las Vegas Operations. Any transparency having a relatively wide range of gray scales can be used on this instrument, provided only that there is not more range in the scale of background grays than among the grays of the images of interest. The aerial photograph is viewed by the densitometer by transmitted light through a video camera, and the different intensities of grays on the photograph are converted electronically into vivid colors. The areas covered by a particular gray, now converted to color, can be read out as a percent of the total area. Since shrub vegetation is usually somewhat darker or lighter than background soil, the colors corresponding to the areas covered by shrubs can be summed up as the fraction of the total area being considered. Where there are sufficient differences among the green shades of various species (interpreted as grays on photos, and color by Datacolor), the area covered by a particular species relative to other vegetation and to the total area can be determined.

This method is fast and can be utilized for large areas if there is sufficient resolution of individual shrubs or shrub clumps. However, it is subject to the same errors in optics as the previous method. While it avoids some sources of error, it is particularly subject to misinterpretation by an operator who is not aware of ground conditions. In addition, strong shadows and highlights from early morning or late evening photography may provide misleading values, either greater or smaller.

The dark areas about very old shrubs resulting from large accumulations of organic matter may still remain after the shrub is dead, thereby registering a gray interpretable as a living shrub. The method also allows misinterpretation as a result of seasonal phenology; that is, leafy or bare shrubs would still cover the same area and this cannot be readily shown by aerial photography. Because of the large number of uncontrolled variables, Datacolor operation is somewhat art rather than science.

It also has certain advantages, however. No assumptions need be made as to shape of shrubs for area measurements because photos record exact shapes, which the densitometer also reads. For both methods, scale need not be known, since the data are read in percentages of the total; but if scale is known, then ground surface areas can be given with the microscope methods. Ground areas cannot be given readily with the Datacolor system. Datacolor is useful, however, for determining other landscape features which may be of ecological interest, as, for example, drainage patterns, where the substratum is usually of different color or texture and is, hence, readable. Blown-out areas or other barren areas within vegetation can be readily assessed.

Because the Spatial Data System instrument arrived late in the fiscal year, there was not time to provide estimates of vegetation cover by both methods. A few areas, therefore, have only the microscope estimates.

The methods of analysis utilized here are new. Since they were developed primarily by using aerial photography at Area 5-GMX, much more data and details of vegetation are presented for this area than for the others.

RESULTS

Area 5--GMX

GMX lies on the eastern part of Area 5, NTS. The region of interest is about 3,600 by 3,200 ft. It is generally flat at an elevation of 2,975 ft. Elevation increases slowly to the north. The soil is alluvial and the region is crossed by a broad southward drainage system.

A relatively few species of shrubs provide most of the vegetation cover. The dominant species in most of the area around GMX is *Larrea divaricata* (creosote bush), although in the southeastern side of GMX, *Atriplex canescens* replaces *Larrea* in the drainage pattern. *Larrea* is extremely variable in size. The largest shrubs may reach three meters in height in the southeastern sector, while elsewhere, mature shrubs are generally less than one meter.

Among other shrubs which occur frequently are the following:

1. *Atriplex canescens* is, after *Larrea*, the largest shrub in the area. It sometimes exceeds the smaller *Larrea* in size. In some sectors it is abundant, and in the eastern sector (outside the fence) of the drainage pattern, it occurs in an almost pure stand.

2. *Lycium andersonii* is a shrub of intermediate size which occurs more frequently in the western sector of the fenced area than in the eastern sector.

3. *Acamptopappus shockleyi* and *Franseria dumosa* are smaller shrubs of the Compositae which occur widely, but more frequently in the southern and western sectors.

4. *Dalea fremonti* occurs in the wash on the western side, especially in the drainage ways. *Krameria parvifolia* is often associated with it.

The percentages of the area covered by shrub vegetation as evaluated by the two methods are presented in Table 1. The numbers measured by both methods appear equally valid, and the values do not differ more than a few percent.

The most sparsely covered region at GMX is that in broad drainage pattern to the east and southeast of GZ. This region has an almost pure stand of *Atriplex canescens*, except on its peripheries, where *Larrea* increases in frequency (see Fig. 1, Plot I). The microscope analysis gives a cover value of 4.17% ± 1.00, and the isodensitometer gives a mean value of 6.5% for four readings.

This was one area in which there was sufficient difference in the gray scales of the images produced by *Larrea*, relative to other species. The isodensitometer showed a value of 2.0 percent for the cover attributable to *Larrea*.

The next higher cover value occurs in Plot IV, Fig. 1, where the shrubs are *Larrea-Franseria* and *Atriplex confertifolia*. There is 1.5 to 2 times the cover here compared to the *Larea-Atriplex canescens* covered locations. This area also allowed an isodensitometer estimation of the cover for *Larrea*. The value was 0.9%.

Plot III, which is within the GZ fence, and Plot II, north of GZ, showed a cover value of around 12% by microscopic analysis. The isodensitometer system showed a mean of 9.1% for Plot II. Plot II was also of interest because of the large number of shrubs, with perhaps three times as many individuals per unit area as at other sites. Plot III was not determinable by the isodensitometer, because the large differences in the soil color background were greater than the differences in gray scale produced among shrubs.

To conclude, the shrub cover around and within the fenced areas of GMX is relatively sparse. The least cover, 4 to 6%, occurs in the drainage area to the east and southeast. The highest value was about 12% cover.

Tonopah Test Range--Roller Coaster

Three of the Roller Coaster tests were done in desert grassland communities known as Cactus Flats, and the fourth in a typical Mojave Desert Shrub community in the adjacent valley to the west on the Tonopah Test Range (TTR). The area is typical Basin-and-Range Province with block fault mountain ranges paralleling the valley. From the lower slopes of the mountains are numerous alluvial fans which converge to form relatively flat valley floors. The elevation at the

Table 1.

Vegetation Cover Values for Area 5--GMX

PLOT NO.	SUBPLOT AREA	HAUSER MICROSCOPE ANALYSIS					SPATIAL DATA SYSTEM ANALYSIS	
		Number Shrubs/ Subplot	Shrub Mean Diameter (Meters)	±	Standard Deviation (Meters)	Percent Cover	Percent Cover	Percent Open
I	107.1 m^2	10	0.60	±	0.27	2.68	6.0	93.0
		7	0.97	±	0.86	4.80	7.2	92.5
		11	0.72	±	0.47	4.19	6.6	92.4
		13	0.47	±	0.11	2.06	6.4	----
		10	0.99	±	0.70	7.14	2.0*	----
	Mean Values	*10.2*	*0.75*			*4.17±1.00*	*6.5*	----
II	109 m^2	37	0.66	±	0.25	11.46	8.8	98.6
		37	0.67	±	0.36	12.08	10.0	88.1
		30	0.80	±	0.31	14.03	12.6	----
		30	0.70	±	0.32	10.60	7.9	90.6
		33	0.70	±	0.31	11.67	6.3	92.5
	Mean Values	*33.4*	*0.71*			*11.97±1.09*	*9.1*	----
III	**	12	0.77	±	0.54	11.54	***	***
		14	0.70	±	0.09	11.56		
		16	0.74	±	0.28	14.48		
		6	1.17	±	0.43	13.37		
		10	0.79	±	0.16	10.33		
	Mean Values	*11.6*	*0.84*			*12.25±1.79*		
IV	90.4 m^2	9	0.77	±	0.43	4.69	8.0	----
		8	0.66	±	0.16	3.00	8.8	----
		11	1.08	±	0.67	11.25	8.4	----
		13	1.18	±	0.60	15.79	7.1	----
		16	0.01	±	0.62	13.82	0.9*	----
	Mean Values	*10.4*	*0.94*			*9.71±5.62*	*8.1*	----

*Cover areas provided by *Larrea*. Only in these cases could the cover area provided by a single species be determined.
**Since scale was not determinable, subplot areas were not determinable.
***Not determinable by this method. Background spectrum too large.

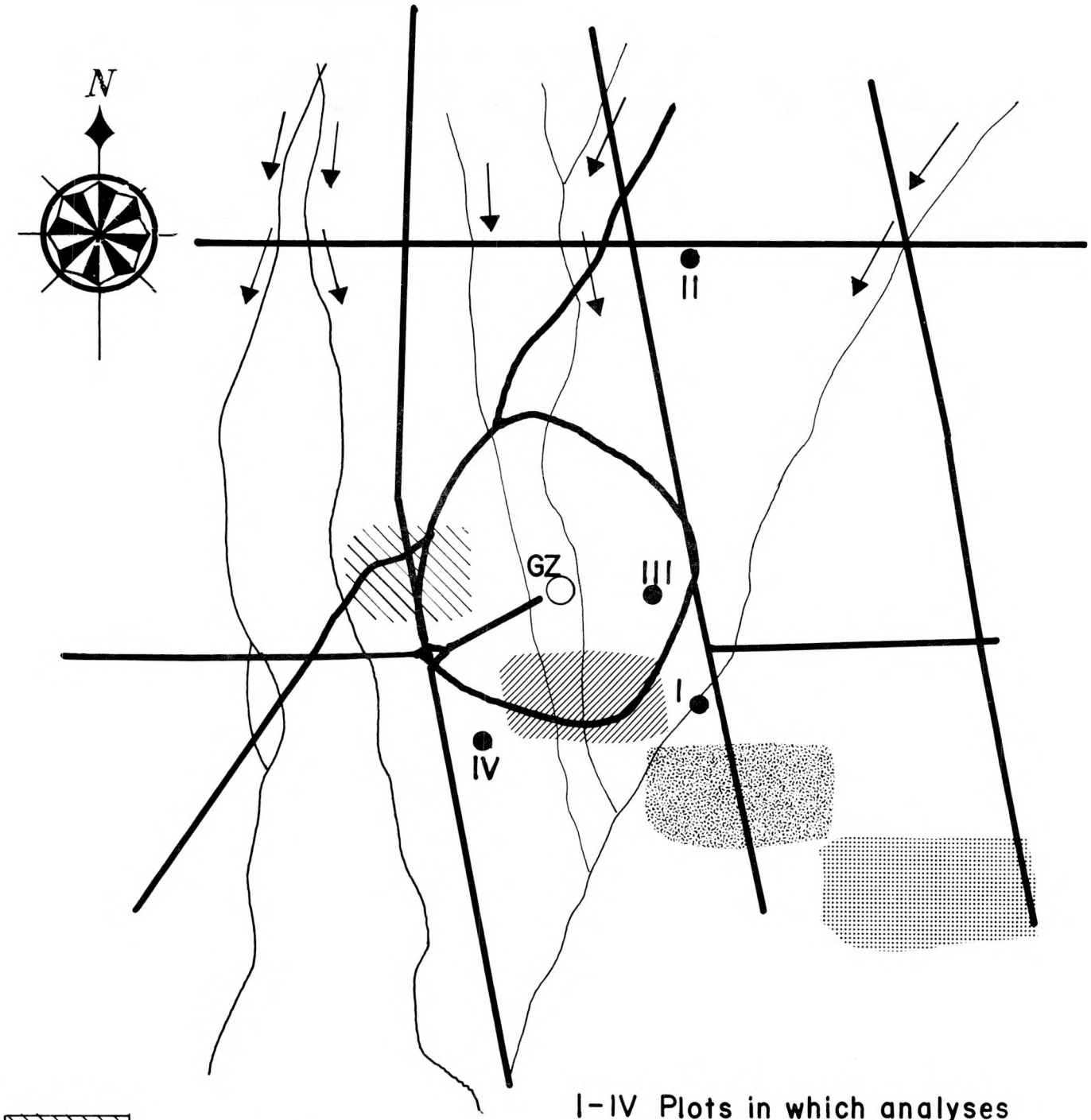

FIGURE-1 AREA 5-GMX

Clean Slate 1, 2, and 3 test areas is about 5,400 ft, along the floor of Cactus Flats. At the Double Track test areas in the next valley to the west (Stonewall Flat), the elevation is about 5,000 ft.

The bottom of Cactus Flats contains the usual playas, that is, the "dry lakes" which characterize the Basin-and-Range Province. In addition, there are dunes in the central part of the valley which appear to be fairly stable on the downwind (northeast) side of the playas. These are characterized by vegetation that is different in many ways from the rest of the valley, and these have not been considered here. At Clean Slate 1, in particular, there are large numbers of blown-out areas of from tens to hundreds of square meters. These are generally characterized by low ridges of relatively large shrubs in rows transverse to the wind. On the southwest boundary of Clean Slate 1 is a large nonvegetated playa. Calculation of shrub cover does not include an evaluation of nonvegetated areas, which are relatively large. Clean Slate 3 is also bordered on the west by a playa.

Vegetation in the Clean Slate areas is dominated by *Hilaria jamesii*, called Galleta grass, an important forage grass. The plant associations are *Hilaria-Chrysothamnus viscidiflorus* and *Hilaria-Atriplex confertifolia-Artemisia spinescens*. Other species occur with *Hilaria* also, with less frequency. Among them are *Eurotia lanata*, *Oryzopsis hymenoides* (rice grass), *Tetradymia* (two species), and *Grayia*.

At Double Track, grass is very infrequent and is usually *Oryzopsis hymenoides* or *Tridens pulchellus*. The shrubs are *Grayia spinosa*, *Lycium pallidum*, *Atriplex confertifolia*, *Artemisia spinescens*, *Lepidium fremontii*, and *Tetradymia glabrata*. *Ephedra* is also present.

Cover values for Roller Coaster locations are given in Table 2. Clean Slate 1, 2, and 3 locations were analyzed by the calibrated stage microscope method, Double Track by the Spatial Data Systems method.

In the course of investigation in the Clean Slate areas, certain systematic changes in the vegetation were noted between grazed and ungrazed areas. A preliminary evaluation of this condition is given in the Appendix.

Area 13

This area is just north of the NTS proper near the northeastern corner. It is in the bottom of a relatively wide alluvial valley bound on the east by the Groom Range and on the west by the Belted Range.

The vegetation is typical of Great Basin Desert, with relatively few species of perennial shrubs. Beatley (1969) described it as an *Atriplex confertifolia* association. Within the Pu-contaminated area where there are little differences in elevation, the slightly higher elevations are inclined to be *Atriplex canescens*, *Grayia spinosa*, along with *Tetradymia glabrata*, while the lower areas are more likely to be *Atriplex confertifolia*, *Kochia americana*, and *Eurotia lanata*. The only

Table 2.

Vegetation Cover Values for Roller Coaster Locations

Location	Hauser Microscope Analysis (percent cover)	Spatial Data System Analysis (percent cover)
Clean Slate 1*		
Plot 1-A	9.1 ± 1.1	---
Plot 1-B	14.7 ± 3.4	---
Plot 1-C	12.4 ± 3.1	---
Clean Slate 2**		
Plot 2-A	5.2 ± 0.4	---
Plot 2-B	2.2 ± 0.4	---
Plot 2-C	1.2 ± 0.3	---
Plot 2-D	2.3 ± 0.4	---
Plot 2-E	0.3 ± 0.2	---
Clean Slate 3*		
Plot 3-A	8.4 ± 2.5	---
Plot 3-B	5.6 ± 2.2	---
Plot 3-C	4.5 ± 0.6	---
Double Track		
9 plots evaluated	---	14.9 ± 3.5

*15 plots (approximately 100 m^2) were counted and measured for each value.

**5 plots were counted and measured for each value.

significant grass present is *Oryzopsis hymenoides*, which occurs with significant frequency only in the north end of the fenced area in a locale with a sandy soil which is unlike the alkaline clay soils of most of the fenced area. *Artemisia spinescens* also occurs.

Estimates of the vegetation cover are given in Table 3.

The conclusion is that Area 13 is probably the most densely vegetated area at or around the NTS where Pu contamination is a problem.

Table 3.

Vegetation Cover Values for Area 13

Aerial Photo-- UCLA Site Coordinates	Hauser Microscope Estimate		Spatial Data System Estimate	
	No. of Plots Analyzed	Percent Cover	No. of Plots Analyzed	Percent Cover
941,000 721,400	10	17.9 ± 1.9	3	18.2 ± 4.9
941,000 721,000	5	18.2 ± 1.4	8	15.0 ± 1.9
939,000 723,000	5	17.3 ± 3.0	8	16.9 ± 1.8
939,800 723,000	5	17.5 ± 2.5	4	14.0 ± 0.7
941,800 717,800	15	20.1 ± 0.2	3	24.0 ± 5.0

Area 11

This Pu-contaminated area is on the eastern boundary of NTS, an area called Plutonium Valley on USGS maps, at an elevation of about 3,400 feet. Beatley calls Plutonium Valley a *Grayia-Lycium-Tetradymia glabrata* association. It is, however, such a heterogeneous area that it might also be classified as a transitional area. The number of perennial shrub species here is probably larger than any other area investigated in this study. Seventeen species were tabulated and several species of the Cactaceae were not. It is difficult to attribute dominance to any single species or small number of species in much of the valley. The hills on the east contain both *Yucca brevifolia* and *Larrea tridentata*. *Lycium*, *Grayia*, and two species of *Atriplex* occur with some frequency.

Because the area is near the upper reaches of the valley, where several drainage ways converge, the area is marked by a complex drainage pattern.

Vegetation cover values are shown in Table 4.

Table 4.

Vegetation Cover Values for Area 11

Area 11	Hauser Microscope Estimate (percent cover)
Plot 11-A*	16.5 ± 4.4
Plot 11-B*	16.1 ± 5.0
Plot 11-C*	16.1 ± 5.0

*5 plots counted for each value.

APPENDIX

PRELIMINARY ASSESSMENT OF VEGETATION CONDITIONS IN THE VICINITIES OF CLEAN SLATE 2 AND 3 (ROLLER COASTER SERIES)

by William A. Rhoads and Robert K. Mullen

In the course of a ground-based vegetation survey of Roller Coaster, it was noted that all GZ enclosures were free from the pressures of cattle grazing. As we subsequently determined, cattle have been excluded from the GZ enclosures for about nine years, except for isolated instances when fences were temporarily down. Due in part to the dimensions of the enclosures, the apparent effect of cattle exclusion is most dramatic at Clean Slate 2 and 3. It is apparent even to a casual observer that the condition of the grasses (primarily *Hilaria* and *Oryzopsis*) within these enclosures is vastly better than outside these enclosures. Although the areas around Clean Slate 1, 2, and 3 are now covered by grasses and shrubs, it is likely that, before heavy grazing pressure, these areas were mostly grass covered. (It was informally estimated that 2,500 cattle now graze the area.)

Closer comparison of the vegetation within and without these enclosures shows more subtle differences in the range vegetation than just the condition of the grasses.

One difference, to be described preliminarily in the following, concerns the abundance of the shrub *Artemisia spinescens* (called "Bud Sage") in the enclosures relative to its abundance outside. That is, it appeared much less abundant in areas protected from cattle grazing compared to those areas subject to grazing. Measurements appear to support this observation. In the Clean Slate 2 GZ enclosure, *Artemisia* is, on the average, about 18 times less abundant than in areas in the vicinity of the enclosure which are accessible to cattle (Table A1). Within the Clean Slate 3 enclosure, similar preliminary measurements indicate *Artemisia* is 18 to 30 times less abundant than it is outside this enclosure (Table A2).

The codominant shrub at these sites is *Atriplex confertifolia*. Within the GZ enclosures, *Atriplex* is dominant almost to the exclusion of *Artemisia*, while outside of these enclosures, the two shrubs coexist in a condition where, although *Atriplex* appears dominant in terms of biomass, *Artemisia* contributes significantly to the biomass of the association. It is not immediately apparent why the *Artemisia* should be so infrequent inside the enclosures.

Table A1.

Artemisia spinescens/yd^2 at Clean Slate 2

Location of Area Sampled	Grazed	Not Grazed
Near GZ		
Graded	---	0.320*
Not Graded	0.320**	---
Arcs "B," "C"		
Graded	0.535†	---
Not Graded	0.535†	---

*368 yd^2 sampled.
**400 yd^2 sampled.
†200 yd^2 sampled.
†*Arthemisia* 1/3 to 1/4 smaller than in adjacent area not graded.

Table A2.

Artemisia spinescens/yd^2 at Clean Slate 3

Location of Area Sampled	Grazed	Not Grazed
Near GZ		
Graded	---	0.033*
Not Graded	0.641**	---
Arcs "B," "C"		
Graded†	0.585	---
	0.715	
Not Graded†	0.925	---
	0.540	

*360 yd^2 sampled.
**565 yd^2 sampled.
†200 yd^2 sampled for each value.

For this report, there was not sufficient time to learn the history of these areas. It is apparent from the aerial photographs that the GZs were graded, or the soil disturbed in some systematic way, which appears to have removed the shrub vegetation which was previously there. This apparently occurred in 1963. The GZs must also have received heavy traffic, which may not have been the case with the regions out 0.4 mile from GZ between the "B" and "C" arcs, where grading of some kind occurred and where the shrubs were also removed. In these areas remote to GZ, however, *Artemisia* has returned with the same frequency it occurs in adjacent areas which were not graded. Both areas were, of course, subject to grazing.

This preliminary investigation thus shows two things:

1. *Artemisia* is far less abundant in the bladed, but ungrazed, GZ enclosures than it is outside these enclosures.

2. There appears to be no significant difference in abundance of *Artemisia* between the bladed and nonbladed areas outside the GZ enclosures where both areas are subject to grazing.

Although grazing, or the lack of it, is superficially the most dramatic difference between the enclosed and open areas, it is difficult to conclude that this is causal. Within the enclosures, *Atriplex confertifolia* is much larger than outside, where it is accessible to cattle. It appears, however, not to be grazed. Nor is *Artemisia* grazed in the open areas. The primary forage appears to be *Eurotia* and the grasses, chiefly *Hilaria*. If neither *Artemisia* nor *Atriplex* are grazed, then how can the dramatic differences in *Artemisia* abundance and *Atriplex* size between the grazed and nongrazed areas be accounted for?

Some possible causes can be suggested, and other questions can be asked which may serve as guides for further investigation.

Halogeton, a toxic forb, appears within the enclosure at Clean Slate 2 (graded, but not grazed), but does not appear along the "B" and "C" arcs (graded and grazed). Is this a response to the greater disturbance which undoubtedly occurred near GZ? (This may have some bearing on methods to be considered in case a "cleanup" of the area is undertaken.) What is the interrelationship of grass and *Artemisia spinescens*, where grass is grazed or not grazed, in the presence of *Artemisia* which appears not to be grazed?

In summary, all the region around the Clean Slate series is presently being heavily grazed, except those few acres in the immediate vicinity of the GZs. *Artemisia spinescens* is one of the codominant shrub species (with *Atriplex confertifolia*) under conditions where grasses would likely be dominant without grazing. One of the GZ areas, which is now largely grass and *Atriplex*, also has a large population of *Halogeton*.

If cleaning up the areas is to be considered, a better understanding of the ecology of the area is needed in order to avoid the possibility of returning the area to a less useful condition.

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ANALYSIS OF ^{239}PU AND ^{241}AM IN NAEG LARGE-SIZED BOVINE SAMPLES*

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ABSTRACT

Analysis of environmental levels of ^{239}Pu and ^{241}Am in Nevada Applied Ecology Group (NAEG) large-sized bovine samples requires development of special procedure modifications to overcome the complexities of sample preparation and analyses. Also, special techniques are often employed to prepare and analyze different types of bovine samples, such as muscle, blood, liver, and bone.

Sample sizes range up to 4 kg of muscle and 1.5 kg of bone. Muscle and large tissue samples are cubed prior to ashing, and bone is sawed into small sections. Large samples are split between several 2-liter Pyrex beakers and decomposed on a hot plate to a charcoal appearance. Ashing is completed at 450°C in an ashing furnace. Ashing is done conservatively to outgas the sample without starting grease fires. The ash is dissolved in dilute HCl. Any insoluble residues are filtered, ashed, and dissolved with HF-HNO_3, H_3BO_3, and combined with the dissolved ash. Plutonium-236 and ^{243}Am tracers and Y carrier are added and equilibrated with sample activity.

In large samples, the Pu and Am are first carried on a mixed CAF_2-YF_3 precipitate. Plutonium and Am are separated on an anion column, and Pu is purified and determined by previously reported methods. Americium is purified by liquid-liquid extraction with HDEHP, then carried on a precipitate of YF_3. Residual Ca and Mg are removed in an acid oxalate step. Americium is finally purified on an ETOH-6N HNO_3 anion exchange column and electrodeposited on Pt for alpha spectrometer counting.

Plutonium and ^{241}Am detection limits vary with sample ash content, but typically are 10^{-4} dpm/g ash. Average tracer recovery for ^{236}Pu and ^{243}Am is 80 and 60%, respectively. Since ^{241}Am has a higher energy than ^{243}Am (5.49 vs 5.28 MeV), the tracer must be matched with the ^{241}Am for accurate measurement.

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INTRODUCTION

Several thousand samples containing a wide range of radioactive elements have been analyzed by our laboratory for the various programs sponsored by the Nevada Applied Ecology Group. These analyses included radiochemical and instrumental determination of such radioisotopes as ^{238}Pu, $^{239,240}Pu$, ^{241}Am, ^{55}Fe, ^{90}Sr, ^{210}Pb, ^{0}Fe, and total and isotopic U. Analyses were performed on a variety of matrices including soils, vegetation, glass fiber and microsorban filters, saltation samples, rodents, and large-sized bovine samples. Data generated was reported routinely in tabular form to the NAEG investigators for their evaluation. A special computer program was developed for reporting the data.

Classical radiochemistry procedures were employed where possible for these analyses. However, special methods and techniques often had to be developed, such as the radiochemical and instrumental procedures for determining ^{239}Pu and ^{241}Am in large vegetation samples (described at the 1973 NAEG Pu information meeting; Major *et al.*, 1973). Since that time, a radiochemical procedure was required and has been developed for determination of ^{239}Pu and ^{241}Am in NAEG large-sized bovine samples. It should be noted that only new containers and glassware are used to contain samples during analysis to prevent chance contamination. Similarities exist with radiochemical Am procedures used on other types of NAEG samples. However, significant changes were made for the bovine samples. Several NAEG bovine and rodent samples have been analyzed by this new procedure and reported to the respective NAEG investigators.

METHODS AND MATERIALS

Sample Preparation

Samples ranging up to 4 kg of tissue and 1.5 kg bone are usually received in a frozen condition in polyethylene bags and are allowed to thaw. Tissues are cut into 1-in. cubes, and bone is sawed into small sections. The samples are dried at 105°C for 24 hr in several 2-liter Pyrex beakers covered with perforated aluminum foil. The samples are weighed and dried for another 6 hr, or until a constant weight is obtained.

Samples are transferred to hot plates in hoods, H_2O_2 is added to reduce offensive odors, and the samples are then decomposed to a charcoal appearance. Samples are transferred to a charring oven, more H_2O_2 is added, and the sample is ashed at 200°C overnight to outgas carbonaceous tissue at a slow rate to prevent grease fires. Samples are transferred to a muffle furnace and ashed at 520°C to a white ash.

The ash is allowed to cool and is dissolved in dilute HCl. The solution is filtered, and the filter and residue are ashed. Ash is dissolved in HNO_3-HF and H_3BO_3, then combined with the filtrate. Plutonium-236, ^{243}Am tracers, and Y carrier are added and equilibrated with sample activity. A sequential scheme is given in Fig. 1.

Plutonium-239 and Americium-241 Analysis

Hydrofluoric acid is added to the solution after sample preparation, and Pu and Am are carried on the CaF_2-YF_3 precipitate. The precipitate is dissolved in HNO_3 acid. Pu and Am are then separated on a Dowex 1 x 4 anion exchange resin column. The Pu is purified and determined by previously reported methods. Effluent from the column, containing the Am, is adjusted to pH 3, and extracted with HDEHP in toluene. The Am is back-extracted with HCl and carried on a second mixed fluoride precipitate. There are residual macroimpurities present at this point, presumably Ca and Mg, which are removed by an acid oxalate precipitation step. The oxalate is ashed, the resulting oxide dissolved in nitric acid, and YF_3 and $Y(OH)_3$ are sequentially precipated. Americium is finally purified on an ETOH-6N HNO_3 anion exchange resin column, eluted, and electrodeposited on a platinum disc. A sequential scheme is given in Figs. 2, 3, 4, and 5.

Experimental

Plutonium-239 is separated on an anion exchange column and purified as mentioned above. Americium-241 in vegetation, filters, saltation samples, rodents, and soils are analyzed by instrumental or radiochemical methods similar to procedures reported at the 1973 NAEG meeting. A modified procedure, however, has been developed for analysis of ^{241}Am in large-sized bovine samples. The procedure eliminates interference of large residuals of Ca and Mg, as well as rare earths, and improves recovery of tracer. In this procedure, Am is extracted with HDEHP in toluene, as in the previous method, but concentration of HDEHP has increased from 20 to 50% to allow for the extraction competition from Ca and Mg. After checking spiked tissue samples with different quantities of HDEHP, 50% proved to be the optimum concentration. Spiked tissue samples were also analyzed by utilizing the acid oxalate step. Tracer yields were increased and electroplates were cleaner by use of this method.

In order to further improve tracer recovery in the bovine samples, the resin column loading solution was changed from 90% MeOH - 10% 6N HNO_3 to 60% ETOH - 40% 6N HNO_3 (Hagan and Arrhenius, 1963). Tracer yields of 40-70% resulted compared to 20-40% by the MeOH-6N HNO_3 procedure. In order to further improve quality of the electroplate, a 40-ml 75% MeOH - 25% HNO_3 wash was added to the 40 ml of 60% MeOH-40% HNO_3 wash. It was noted, after the double wash, that a clean electroplate was obtained, in comparison to some plates with a slightly milky haze after only the single wash. Table 1 shows experimental results of the new organic aqueous washes and 3N HNO_3 elution of the new resin column loading solution.

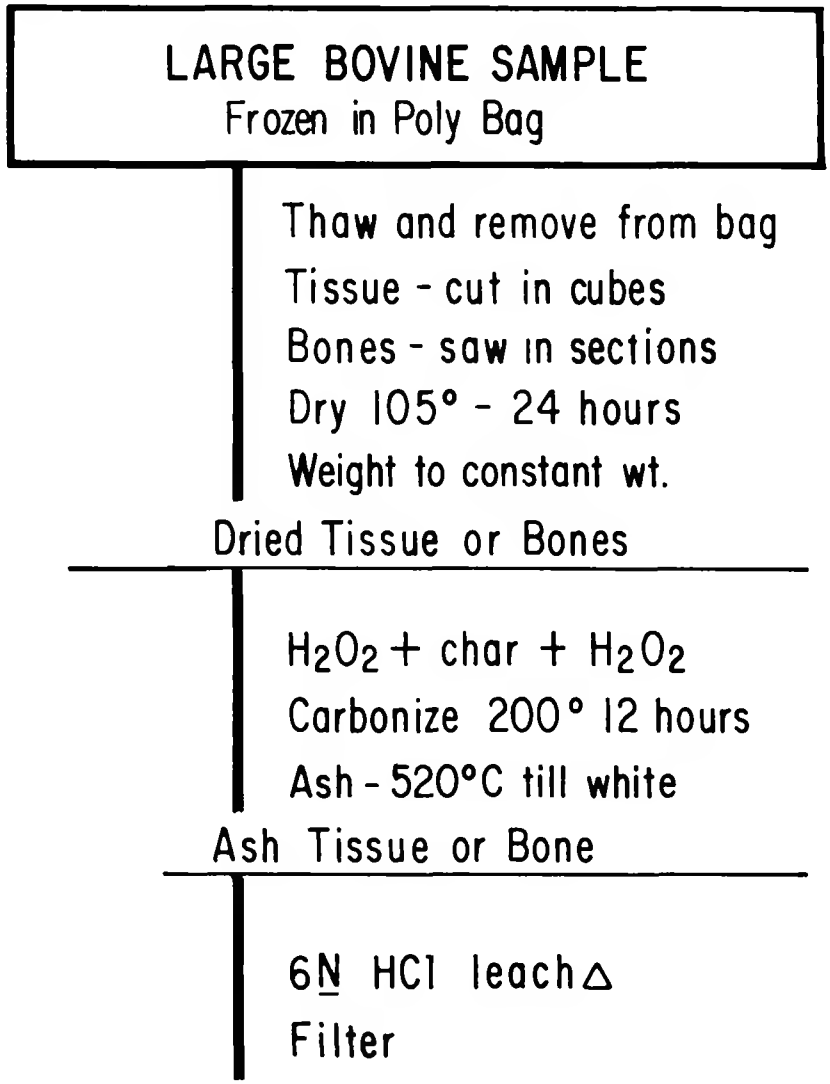

Fig.1 SAMPLE PREPARATION

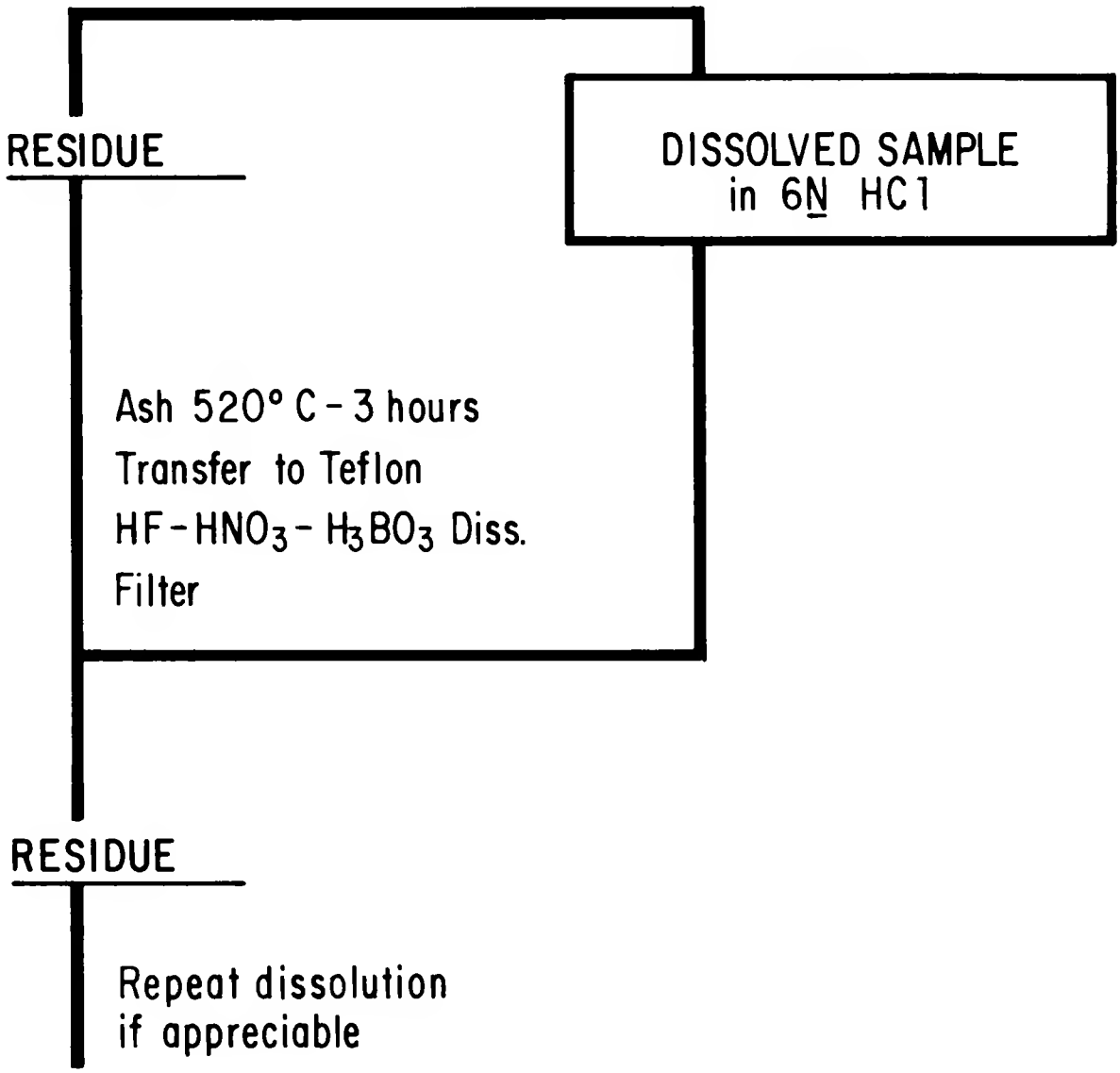

Fig. 1 SAMPLE PREPARATION (Continued)

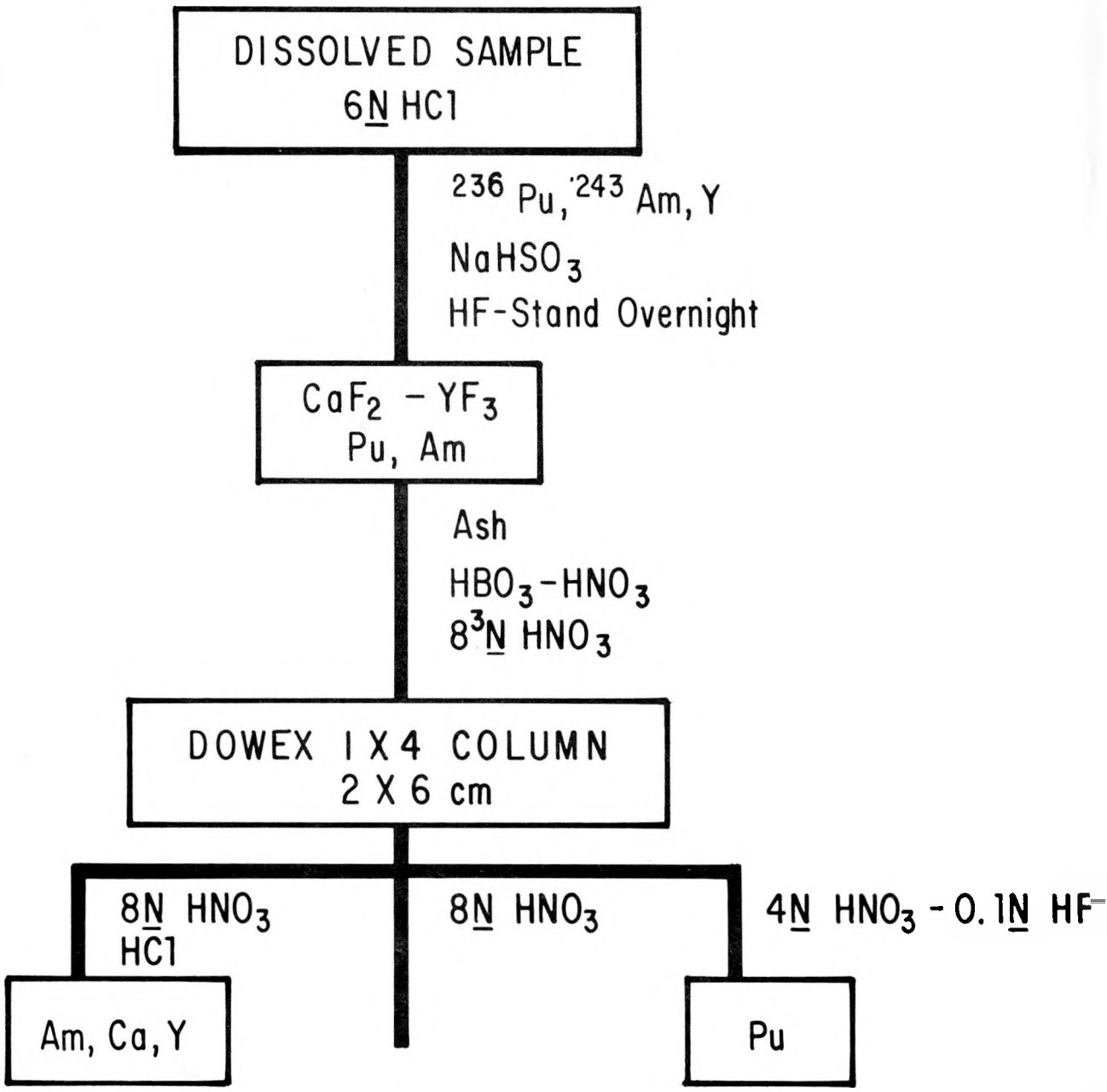

Fig. 2 PLUTONIUM AND AMERICIUM SEPARATION

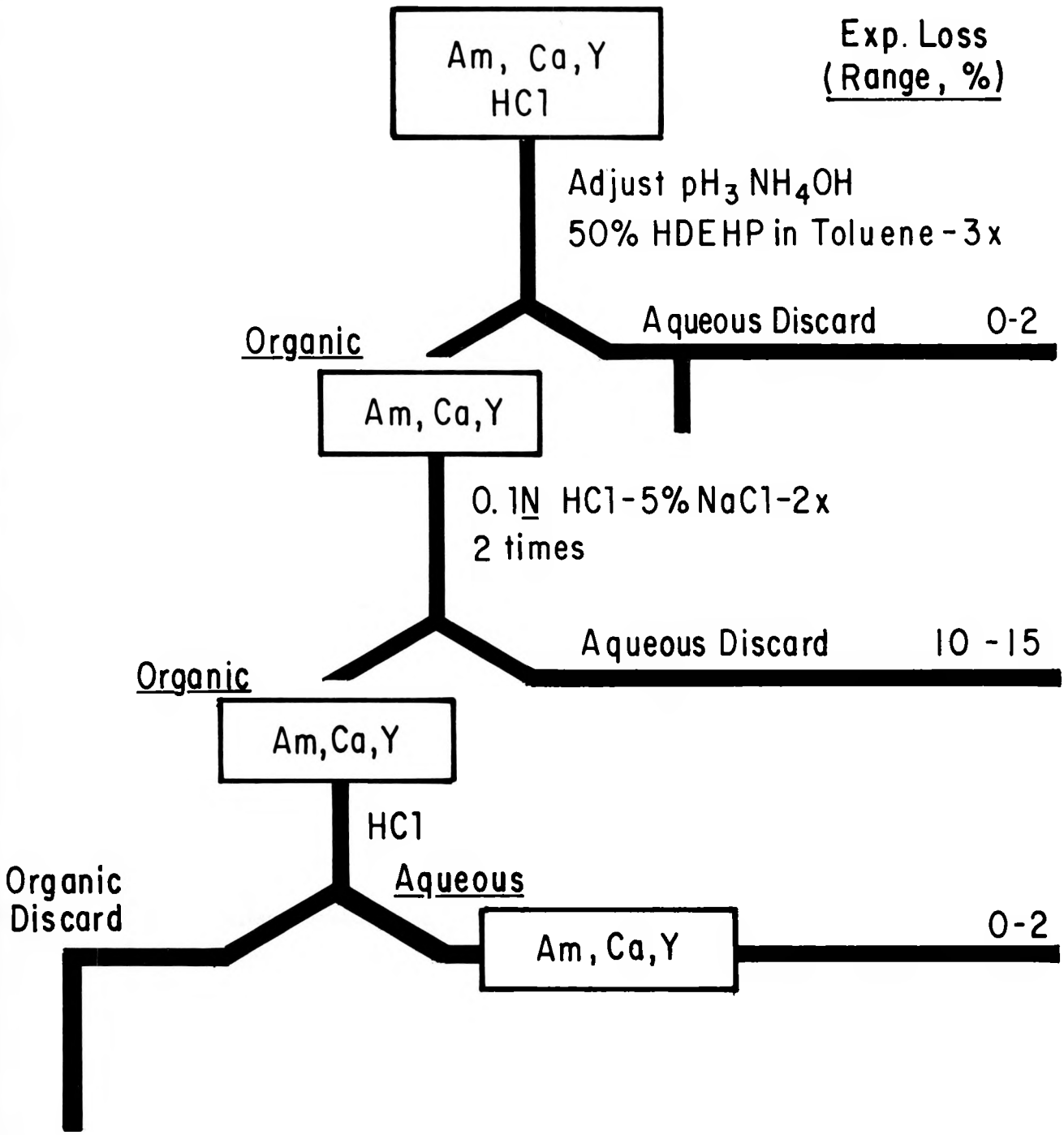

Fig. 3 AMERICIUM EXTRACTION

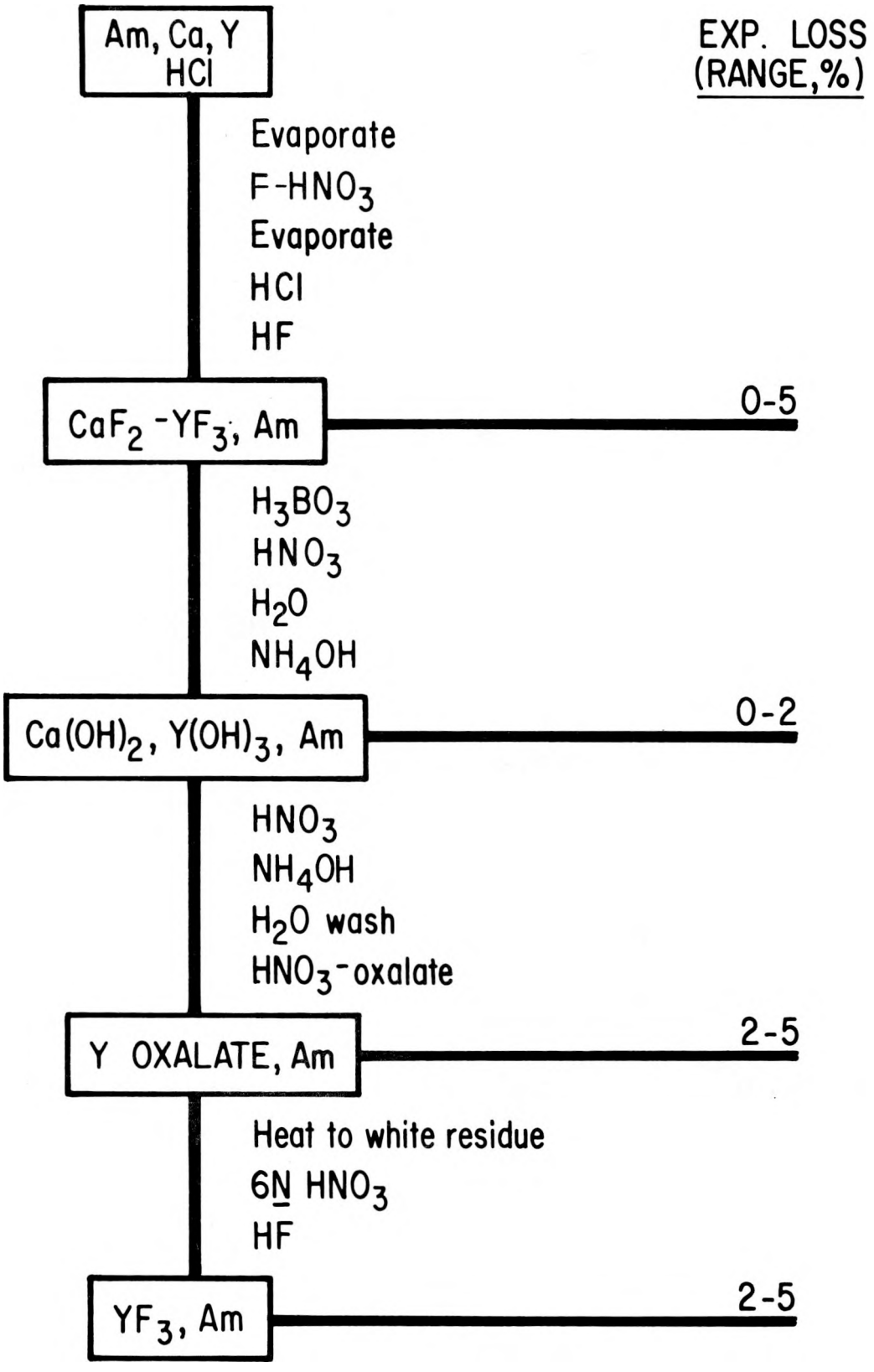

Fig. 4 AMERICIUM ISOLATION

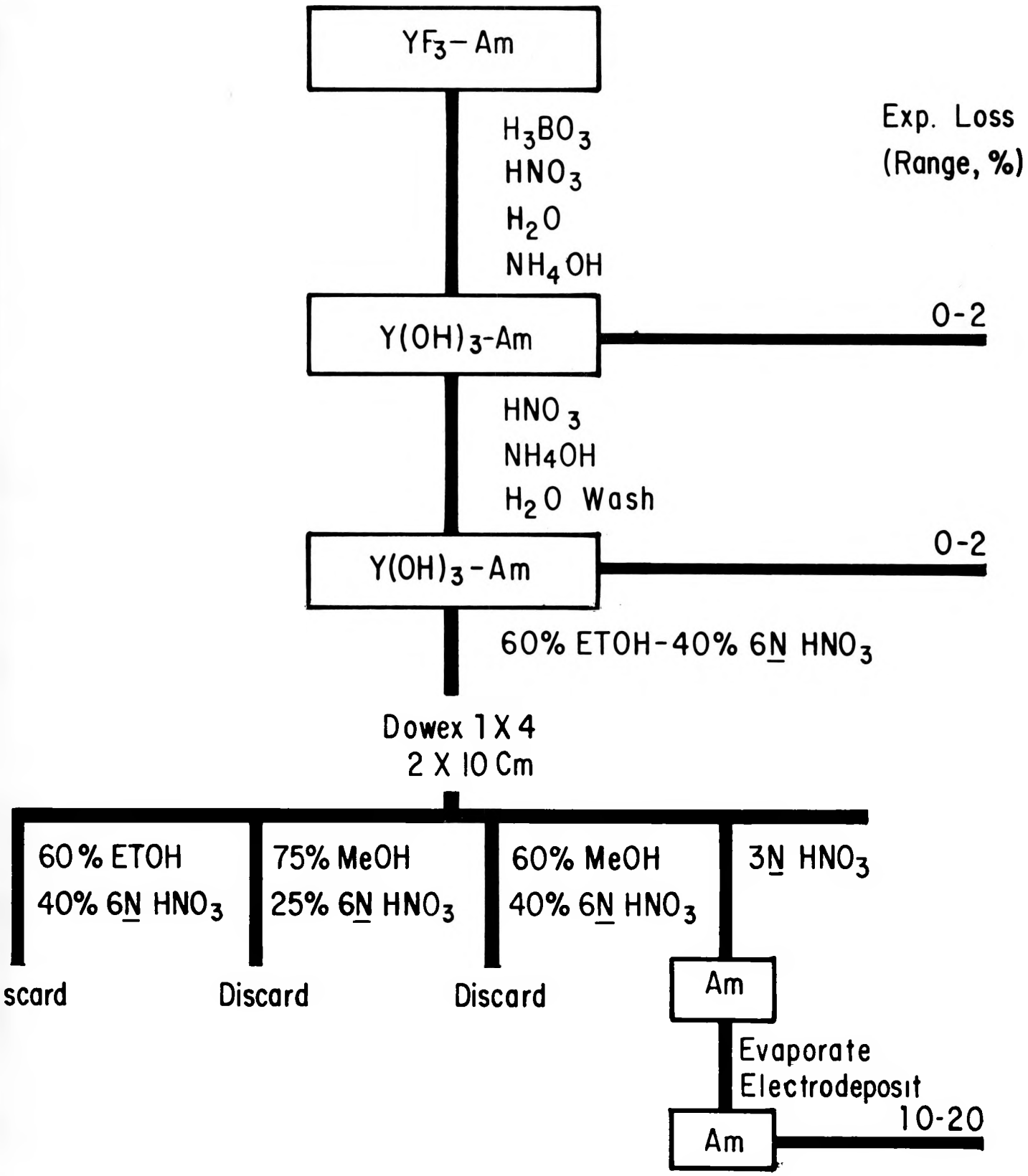

Fig. 5 AMERICIUM PURIFICATION

Table 1. Elution of Americium-241 from Dowex 1 x 4 Anion Resin Column by Organic-Aqueous Washes

A. 75% MeOH--25% 6N HNO_3				
Volume (ml)	Column A (cpm)	% Eluted	Column B (cpm)	% Eluted
5 - 40	0	0	0	0
B. 60% MeOH--40% 6N HNO_3				
Volume (ml)	Column A (cpm)	% Eluted	Column B (cpm)	% Eluted
5 - 25	0	0	0	0
30	250	1.0%	82	0.5%
35	255	1.0%	321	2.0%
40	726	2.8%	600	3.7%
C. 3N HNO_3				
Volume (ml)	Column A (cpm)	% Eluted	Column B (cpm)	% Eluted
5	22,800	88.3%	14,100	87.3%
10	1,700	6.6%	1,000	6.2%
15	65	0.3%	47	0.3%
20	37	0.1%	0	---

Note: Solution of ^{241}Am loaded with 5-ml 60% ETOH--40% 6N HNO_3

To determine losses through the Am procedure steps, spiked samples were analyzed. Results are shown in Figs. 3, 4, and 5.

Since the ^{241}Am has a higher energy than the ^{232}Am tracer, it was recognized that there might be some overlap in the alpha spectrum in higher-activity bovine samples. An experiment was performed to determine the optimum $^{241}Am/^{243}Am$. For an average quality electroplate, a ratio of four appears to be adequate, with a maximum allowable ratio of ten. Typical spectra covering the two ratios are shown in Figs. 6 and 7.

DISCUSSION

With reference to the weighing procedure in the sample preparation section, it was determined, at an interim NAEG meeting last spring, to be more desirable to report bovine data on a gram/dry weight basis due to difficulties in obtaining a standard wet or ash weight.

To reduce ashing time, samples are periodically removed from the furnace during high-temperature ashing and cooled with shaking in order to draw oxygen back into the beaker. In addition, after the initial ashing, the sample is solubilized, filtered, and the filter ashed, since it is extremely time-consuming to ash the last traces of carbon particles in the presence of bulk ash. Any insoluble residue on the filter is treated by further ashing, dissolved, and combined with the sample.

In the Pu and Am analysis, yttrium is added to carry Am after bulk Ca and Mg are removed. A strong reducing agent shown in the flow chart is used to speed up and complete the reduction of Pu. Protactinium and Np as well as Pu are separated from Am by absorption on the Dowex 1 x 4 anion column.

The HDEHP extraction of Am and NaCl wash remove the bulk M^{+2} ions, Th, and rare earths below Pm (NAS-NS 3050). Traces of Pm, Sm, and Eu carry through but are not present in macro amounts that would significantly degrade the quality of the electroplate. Since all three are primarily beta emitters, their radiations do not interfere with the alpha spectrum. The Ca plus Mg are separated after the bulk of these ions are separated by extraction, since any prior separation would not be quantitative. The ETOH-6N HNO_3 column separates rare earths above Eu, resulting in a final electroplating solution containing radiochemically pure Am and insignificant amounts of rare earths.

Americium yields from the MeOH-6N HNO_3 anion exchange column were severely influenced by slight changes in the column load. The hydroxide precipitate to be dissolved for loading on the column varied from

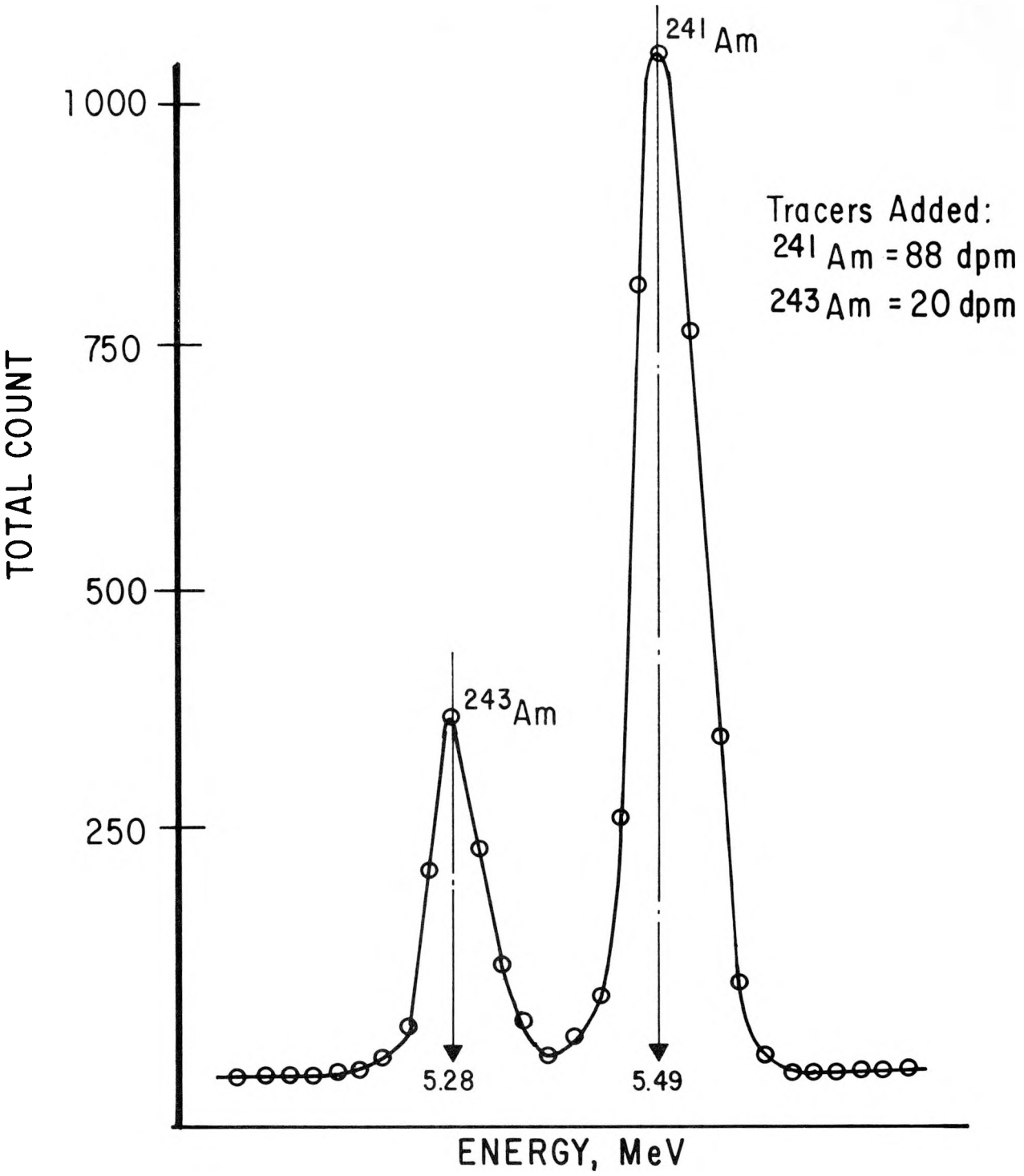

Fig. 6 ALPHA SPECTRUM OF $^{241}Am/^{243}Am = 4$

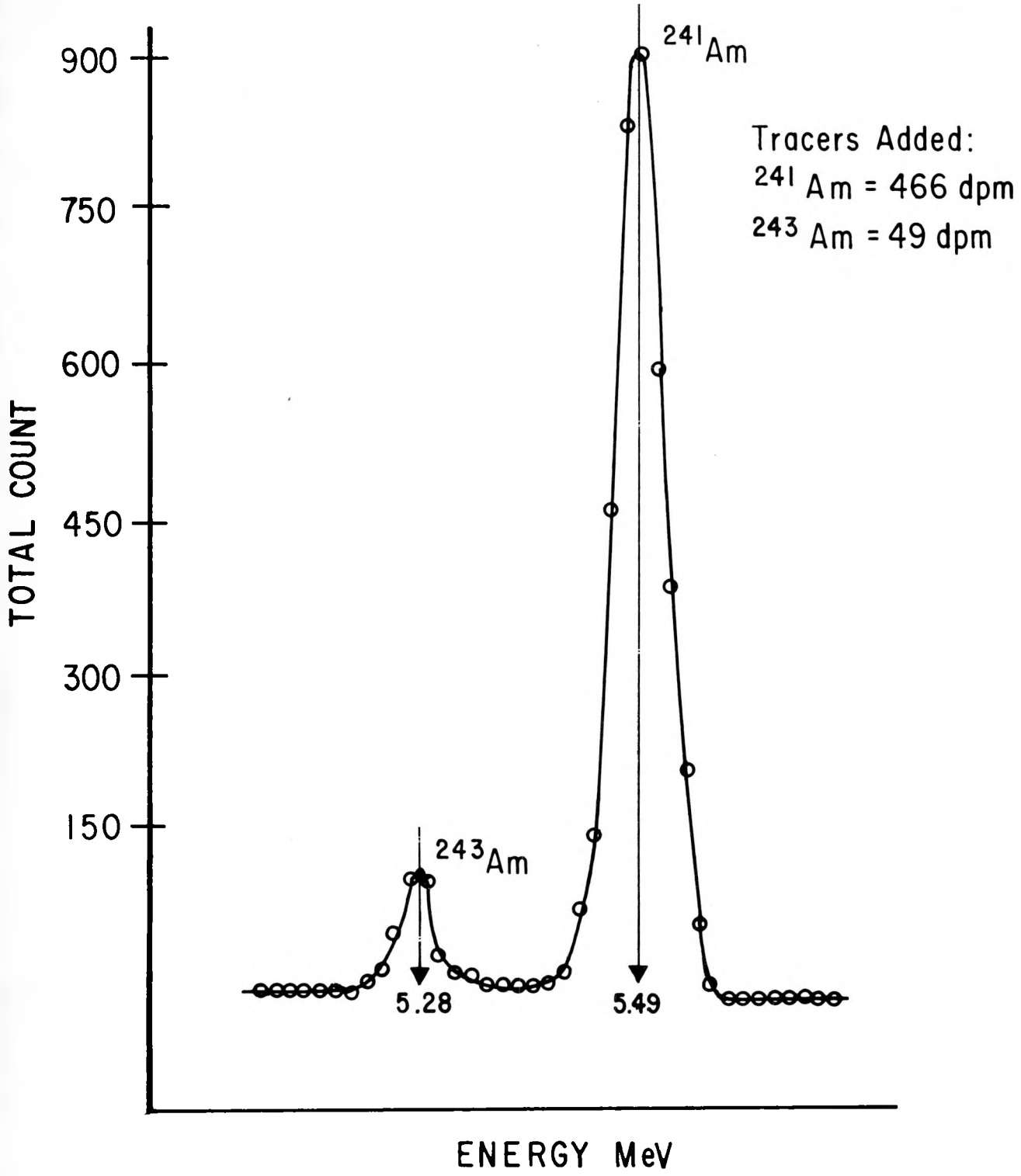

Fig. 7 ALPHA SPECTRUM OF $^{241}Am/^{243}Am = 10$

sample to sample. It was difficult to dissolve the precipitate in the specified amount of 90% MeOH - 10% 6N HNO_3 and keep loading conditions constant. Substitution of ETOH for MeOH permitted a higher percentage of HNO_3 to be used, while still achieving full absorption of AM on the column. Also, the higher HNO_3 concentration permitted more complete solubilization of Am from the hydroxide precipitate. The overall result of using 60% ETOH - 40% 6N HNO_3 was a higher and more consistent tracer recovery.

In Table 1, it is shown that the 75% MeOH - 25% 6N HNO_3 wash solution while removing rare earth elements leaves all the Am on the column. Also, it is seen the second wash has no effect on the Am elution until considerable wash solution has passed through. In the Am elution with 3M HNO_3, it is apparent the first few milliliters contain most of the Am.

Figures 3, 4, and 5 show that although the losses of Am through any given step are not severe, careful adherence to the procedure is important. This is especially essential in the ETOH-6N HNO_3 step to minimize losses. The amounts of cations and rare earths vary considerably for different samples; thus, allowances must be made.

Significance of the $^{241}Am/^{243}Am$ may be noted in Figs. 6 and 7. Since the energy difference between ^{241}Am and ^{243}Am is only 200 keV, it is apparent that the level of ^{241}Am can influence the ^{243}Am alpha spectrum. It has been determined that the peak-to-valley ratio of ^{241}Am is approximately 100, at best. Therefore, the tracer should not be less than 10% of the sample activity. Figure 6 shows the tracer at 25% with no overlap of alpha spectra; Figure 7 at 10% shows overlapping starting to occur.

In conclusion, the changes in Am procedural steps have resulted in considerable increase in tracer recovery in large bovine samples. Also, the quality of the electroplate and alpha spectrum is excellent, comparing favorably with those of the other NAEG-type samples.

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PROCEDURES FOR THE ANALYSIS OF NAEG SMALL VERTEBRATE SAMPLES

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INTRODUCTION

The methodology used in the analysis of NAEG vertebrate samples is described for the transuranic isotopes $^{239-240}Pu$, ^{241}Am, the fission product isotope ^{90}Sr, and the neutron activation produced isotope ^{55}Fe.

The methodology is similar to that used for the analysis of NAEG large vegetation samples (Ref. 1) and NAEG large-sized bovine samples (Ref. 2). The analytical procedures are summarized and more detailed procedural steps are included at the end of the summary.

SAMPLE PREPARATION

Small animal samples, typically reptile, birds, and mammals, have been previously autopsied and dissected as described by Moor and Bradley (1974). In the case of rodents, which have been most extensively analyzed, the animals are dissected into pelt, G.I. tract, and carcass samples and each are analyzed separately. The samples are received at the analytical laboratory in a frozen condition. The samples are thawed and dried in an oven and then ashed in a furnace. The dry and ashed weights are recorded.

CHEMICAL PROCESSING

The ash is dissolved by treatment with HNO_3, HCl, HF, and H_3BO_3. A solution of 100 grams of the dissolved ash is prepared.

The dissolved ash may be measured for ^{241}Am on a gamma spectrometer if there is sufficient activity. This is done before aliquots are taken for radiochemical processing. Radiochemistry isotope dilution analysis is performed for $^{239-240}Pu$ and also ^{241}Am and/or ^{90}Sr or ^{55}Fe as required by the analytical protocol. Plutonium-236 and ^{243}Am tracers are added and/or Sr and Fe carriers and steps are performed to equilibrate sample activity with tracers and carriers. An aliquot of the original dissolved ash is also analyzed for elemental Fe content by atomic absorption spectroscopy and the results are later used in the calculation of Fe carrier yield.

An anion exchange separation procedure is used to separate Pu from Am, Fe, and Sr. An ammoniacal iron hydroxide precipitation separates Sr from Fe and Am. Iron is extracted into isopropyl ether, leaving Am in the aqueous phase. Each fraction Pu, Sr, Fe, and Am is then further purified to remove interfering radioactive and chemical impurities.

The purified Pu, Am, and Fe are electrodeposited and the samples are submitted for radioactivity measurements. The yield of Sr carrier up to this point is determined by atomic absorption analyses. Yttrium carrier is added to the Sr sample solution which is set aside for three weeks to allow ingrowth of ^{90}Y radioactivity, after which the yttrium fraction is chemically separated and purified. It is then weighed as Y_2O_3 and submitted for beta activity measurements.

RADIOACTIVITY MEASUREMENTS AND CALCULATIONS

The $^{239-240}Pu$ and ^{241}Am alpha spectra are obtained, either on a Frisch grid ionization chamber, or a surface barrier detector, plus multichannel pulse height analyzers.

Integration of the alpha spectra is performed for each alpha peak of interest and the data is corrected for detector background and any tracer impurity. The error is calculated as one standard deviation due to counting, including errors due to the tracer peak, etc.

The yttrium planchet, containing the ^{90}Y daughter of ^{90}Sr, is counted on one of a battery of Tracerlab CE-14 low background beta detectors with anticoincidence background subtraction. The detector backgrounds are 0.4 to 0.6 cpm and the efficiency of each detector is 0.54 cpm/dpm for ^{90}Y betas in Y_2O_3 filtered precipitates mounted upon plastic planchets. The system has automated data output on perforated tape. Each yttrium planchet is counted 6 hours after Y_2O_3 filtration and also at least four more times within 12 days to check isotopic purity. The decay data is processed by least squares analysis on an IBM 1130 computing system. Corrections are made for chemical yields of Sr and of Y, sample self-absorption, aliquot, incomplete ^{90}Y ingrowth, and decay. The output from the computer is a data sheet and a plot of the decay.

The electroplated iron sample is counted on a thin NaI (Tl) wafer detector, optimized for low energy photon counting. A ^{55}Fe standard is counted before and after the sample has been counted to check for instrument drift. The photon spectrum, over the energy region, 0 to 12 keV, is taken in each case. The punched tapes are processed on the computer by least squares analysis techniques. The computer resolved dpm results are corrected for yield, aliquot, decay, precipitate absorption of the emitted X rays, and macro iron in the original sample.

The isotopic results for $^{239-240}Pu$, ^{241}Am, ^{90}Sr, and ^{55}Fe are reported on the basis of dpm and nCi of each isotope per gram of sample dry weight and also per gram of ash weight.

CHEMICAL PROCEDURES FOR SMALL VERTEBRATE SAMPLES

Preparation

1. Unpack and inspect sample, record sample numbers and other pertinent information about each sample into the sample receipt logbook. Transfer the sample to an appropriate sized Pyrex glass beaker which has been weighed for tare weight. Weigh the sample.

2. Dry the sample overnight in a 105° C oven and weigh until constant weight is obtained.

3. Char the sample on a hot plate or in a 250° C oven.

4. Heat the sample in a 450 to 525° C oven until a white ash is obtained. Weigh the beaker plus ash, transfer the ash and tare the beaker, and calculate the ash weight.

Dissolution

1. Dissolve the ash with HNO_3-HCl. Heat until all brown fumes disappear. Add HNO_3. Boil to 20 ml. At this point, if residue remains, add 20 ml H_2O, filter through a No. 42 Whatman filter, and return the residue to the beaker and ash and then continue with 5. Transfer the solution to a polycarbonate tube and add 2 ml HF. Heat in a water bath for 20 minutes.

2. Add 4 ml H_3BO_3 and heat in a water bath for 15 min. Transfer to a glass beaker and add 2 ml H_3BO_3. Evaporate to 15 ml.

3. Add 25 ml HCl and evaporate to 15 ml.

4. Add 25 ml HNO_3 and evaporate to 15 ml. Cool. Add 15 ml H_2O. Transfer to a plastic bottle. Dilute to 100 grams.

5. Transfer any undissolved ash with HNO_3 to a polycarbonate tube and continue with step 1 by adding 2 ml HF. When the ash is dissolved, adjust the solution to 100 g of 8N HNO_3.

Pu, Am Separation

1. Transfer the aliquot to be analyzed into a Teflon beaker. Add ^{236}Pu and ^{243}Am tracers. Add 5 ml HF and evaporate to a small volume. Add 5 ml HF and 15 ml HNO_3 and evaporate. Add 10 ml HNO_3 and 5 ml saturated H_3BO_3 and evaporate. Repeat the treatment with HNO_3 and H_3BO_3. Add 20 ml HCl and evaporate to half volume. Add 2 ml H_2O_2 and heat until the reaction ceases.

2. Add an equal volume of H_2O. Cool to room temperature. Dilute the solution to 120 ml with 8N HNO_3.

3. Add 3 ml 5% $NaNO_2$ solution, 1 ml at a time, 5 minutes apart.

4. Prepare the Dowex 1 x 4, 100-200 mesh resin column (1.5 inch x 1-inch diameter) by washing with 30 ml 8N HNO_3. Transfer half of the prepared resin to the sample beaker and let equilibrate with stirring for one hour.

5. Add 1 ml 5% $NaNO_2$. Stir and pass the solution through the column. Save the effluent and the 8N HNO_3 wash for Am analysis.

6. Wash the column with 40 ml 8N HNO_3, then wash with 40 ml HCl.

7. Elute Pu with 40 ml HCl-NH_4I solution (500 mg NH_4I dissolved in 400 ml HCl. Continue with the Pu plating procedure, step 1.

Pu, Am, Sr, Fe Separation

1. Transfer the aliquot to be analyzed into a Teflon beaker. Add ^{236}Pu and ^{243}Am tracers, Fe and Sr carriers. Take an aliquot of the solution for elemental Fe analysis by atomic absorption spectroscopy.

2. Equilibrate and separate Pu as in steps 1 through 5 of the Pu, Am separation.

3. Evaporate the effluent and wash from Pu separation column to a small volume. Add NH_4OH until $Fe(OH)_3$ forms and pH 8 is reached.

4. Filter the sample through a Whatman No. 42 filter. Record the Y-Sr separation time for Sr. Withdraw an aliquot of the filtrate for Sr yield determination by atomic absorption analysis. Continue with step 1 of the Sr procedure.

5. Dissolve the $Fe(OH)_3$ with HNO_3 and reprecipitate with NH_4OH. Filter through Whatman No. 42 filter and combine the filtrate.

6. Dissolve the $Fe(OH)_3$ with HNO_3 (if any residue cannot be dissolved, further treatment is necessary). Add NaOH to precipitate $Fe(OH)_3$. Wash the hydroxide precipitated with H_2O.

7. Dissolve the $Fe(OH)_3$ with HCl and adjust the HCl concentration to 8N. Extract Fe with isopropyl ether.

8. Wash the organic phase once with 8N HCl.

9. Back extract Fe with H_2O. Continue with step 1 of the Fe procedure.

10. Evaporate the aqueous phase from step 7 to a small volume and continue with Am purification procedure.

Pu Plating Procedure

1. Evaporate the HCl-NH_4I eluate from the anion columns to dryness while adding HNO_3. Continue until the solution is colorless as it approaches wet dryness.

2. Add 1 ml HNO_3 and 2 ml HCl and evaporate to dryness. Repeat.

3. Add 5 drops $HClO_4$ and evaporate until fumes stop.

4. Repeat step 2 twice.

5. Add 2 ml HCl. Evaporate to dry, being careful not to bake.

6. Add 2 ml HCl. Evaporate to 1/2 ml. Add 1 drop methyl red. Cool.

Am Purification

1. Boil down Pu wash to approx. 10 ml. Add 0.2 to 0.5 mg Y carrier. Transfer with H_2O or dilute HNO_3 to a 40-ml tube. Add excess NH_4OH. Heat in a water bath with frequent stirring for 10 minutes. Cool and centrifuge. Discard supernate.

2. Dissolve in 3 ml HNO_3. Add 1 drop methyl red. Titrate with NH_4OH until yellow. Back titrate with 6N HNO_3 until the color is red and the solution is clear.

3. Add 5 ml oxalic acid. Heat in a water bath with frequent stirring for 15 minutes. Cool. The solution should turn yellow. Centrifuge. Discard the supernate.

4. Heat the precipitate evenly over a flame until brown fumes stop and the precipitate is white.

5. Dissolve in 10 ml 6N HNO_3. Transfer to polycarbonate tube with 6N HNO_3. The final volume should be approx. 20 ml.

6. Add 4 ml HF. Digest in hot water bath for 20 minutes with stirring. Cool, centrifuge, discard the supernate.

7. Dissolve precipitate with 3 ml H_3BO_3, 2 ml HNO_3, and 5 ml H_2O. Heat with stirring for 10 minutes. Transfer back to a glass tube with H_2O.

8. Add excess NH_4OH. Heat in a water bath for 10 minutes with frequent stirring. Cool, centrifuge, discard the supernate.

9. Take up in approx. 3 ml HNO_3. Repeat step 8.

10. Wash the precipitate with 5 ml H_2O. Centrifuge. Discard the supernate.

11. Prepare anion exchange resin columns, AG 1 x 8 (100-200 mesh, 6-inch height x 8 mm diameter), by passing 20 ml 60% EtOH/40% 6N HNO_3 through the column.

12. Dissolve the $Y(OH)_3$ with 5 ml 60% ethanol-40% 6N HNO_3 and then load onto the preconditioned column.

13. Wash the column with 40 ml 75% methanol-25% 6N HNO_3 solution.

14. Wash the column with 40 ml 60% methanol-40% 6N HNO_3 solution.

15. Elute Am with 40 ml 8N HNO_3.

16. Evaporate and electroplate on a Pt. disc for 20 min.

Fe Procedure

1. To the Fe solution from the separation procedure, add NH_4OH until it is basic.

2. Centrifuge and discard the supernate.

3. Prepare to electroplate the Fe on a Cu disc. Label a copper disc clearly, then polish with a fine abrasive (e.g., "Ajax"). Rinse well with water, then with methanol. Dry with an absorbent tissue and weigh to the nearest 0.1 mg. Assemble a Tracerlab electroplating cell using the prepared copper disc. Cover the disc with methanol until ready to plate. Add a few drops HCl to the precipitate from step 2. Evaporate the Fe solution to wet dryness. Do not bake. Cool. Dissolve the residue with 2 ml water. Add 8 ml stock carbonate solution, and 1.5 ml stock phosphate solution (Note a.). Transfer to the prepared electroplating cell. Complete the transfer by washing with exactly 5 ml of stock carbonate solution. Electroplate for 2 hours at room temperature. Start the electrolysis at 150 milliamperes, and raise the current to 300 milliamperes (about 5-8 volts) after 10 minutes.

   a. Exact amounts of these solutions must be used to insure proper conditions for electroplating.

4. At the end of the plating period, quickly remove the anode from the cell and transfer the solution to a clean 40 ml centrifuge tube (Note b.). Save the solution until the yield of iron has been determined and found to be satisfactory. Disassemble the cell and wash the plate with water, then with methanol. Dry with absorbent tissue and weigh to the nearest 0.1 mg. Cover the iron deposit with a thin coating of "Krylon." Place the disc in a lined and labeled tin box for counting.

   b. The iron metal is soluble in the plating solution; so speed is necessary here to keep the amount dissolved to a minimum. Rinse the cell with water, then with methanol to remove all of the plating solution.

Sr Procedure (Y-Milk Procedure)

1. To the Sr solution from the separation, add HNO_3 until the solution is acid. Add Y carrier and set aside for at least 10 days to allow the ^{90}Y to grow in appreciably. Strontium-90-^{90}Y equilibrium is established about 21 days after removal of ^{90}Y in the Sr procedure. The ^{90}Y may be milked off any time after 10 days and the ^{90}Y activity corrected to the equilibrium value.

2. Heat the solution in a hot water bath, and dilute to 15 ml with H_2O (Note a.). Make ammoniacal to pH 8-10 with fresh NH_4OH. (Note time of $Y(OH)_3$ precipitation as Primary Separation Time.) Stir. Centrifuge and decant the supernate to a clean 40-ml tube and save. Wash the precipitate with 5 ml H_2O. Centrifuge, decant, and save with original supernate.

   a. The remaining steps should be carried out within one day.

3. Dissolve the $Y(OH)_3$ with 2 ml HCl and transfer to a polypropylene tube with 10 ml H_2O. Add 1 ml HF and digest for 15 minutes.

4. Centrifuge and discard supernate, wash precipitate with 1N HCl-0.1 N HF.

5. Slurry YF_3 with 3 ml H_3BO_3 and 2 ml HNO_3 and 5 ml H_2O. Heat to dissolve. Transfer with H_3O to glass tube. Add NH_4OH to precipitate $Y(OH)_3$.

6. Dissolve $Y(OH)_3$ with 1 ml HCl and 10 ml H_2O, add NH_3OH to precipitate $Y(OH)_3$, wash with 5 ml H_2O.

7. Dissolve the washed $Y(OH)_3$ in 3 drops 6 N-HCl. Continue adding HCl dropwise until dissolved and dilute to 10 ml with H_2O. Add 3 ml of saturated oxalic acid.

8. Stir and warm on a water bath 5 minutes, then cool 5 minutes in an ice bath. Wash the precipitate with three 5-ml portions of water and discard each wash. Add 5 ml of methanol and filter onto a 2.3 cm Whatman No. 42 filter paper using a stainless steel filter tower with vacuum.

9. Place the $Y_2(C_2O_4)_3$ and filter paper, in a Coors No. 00 porcelain crucible. Tilt the cover on the crucible and ignite to Y_2O_3 in a muffle furnace at 800° C for 1 hour.

10. Prepare a 2.3 cm Whatman No. 42 filter paper disc by washing with three 5-ml portions of methanol. Dry in an oven at 90-100° C for 10 minutes, and cool 10 minutes in a desiccator. Make a 1 minute timed weighing. Repeat weighing procedure to constant weight.

11. Moisten the oxide from step 10 with absolute methanol and carefully grind the oxide to a fine powder with a stirring rod. Add a few ml methanol, and filter the slurry onto the tared filter paper. Wash the

Y_2O_3 with three 5-ml portions of methanol. Dry the Y_2O_3 in an oven and weigh, as in step 11. Mount on plastic planchet and count after 6 hours.

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