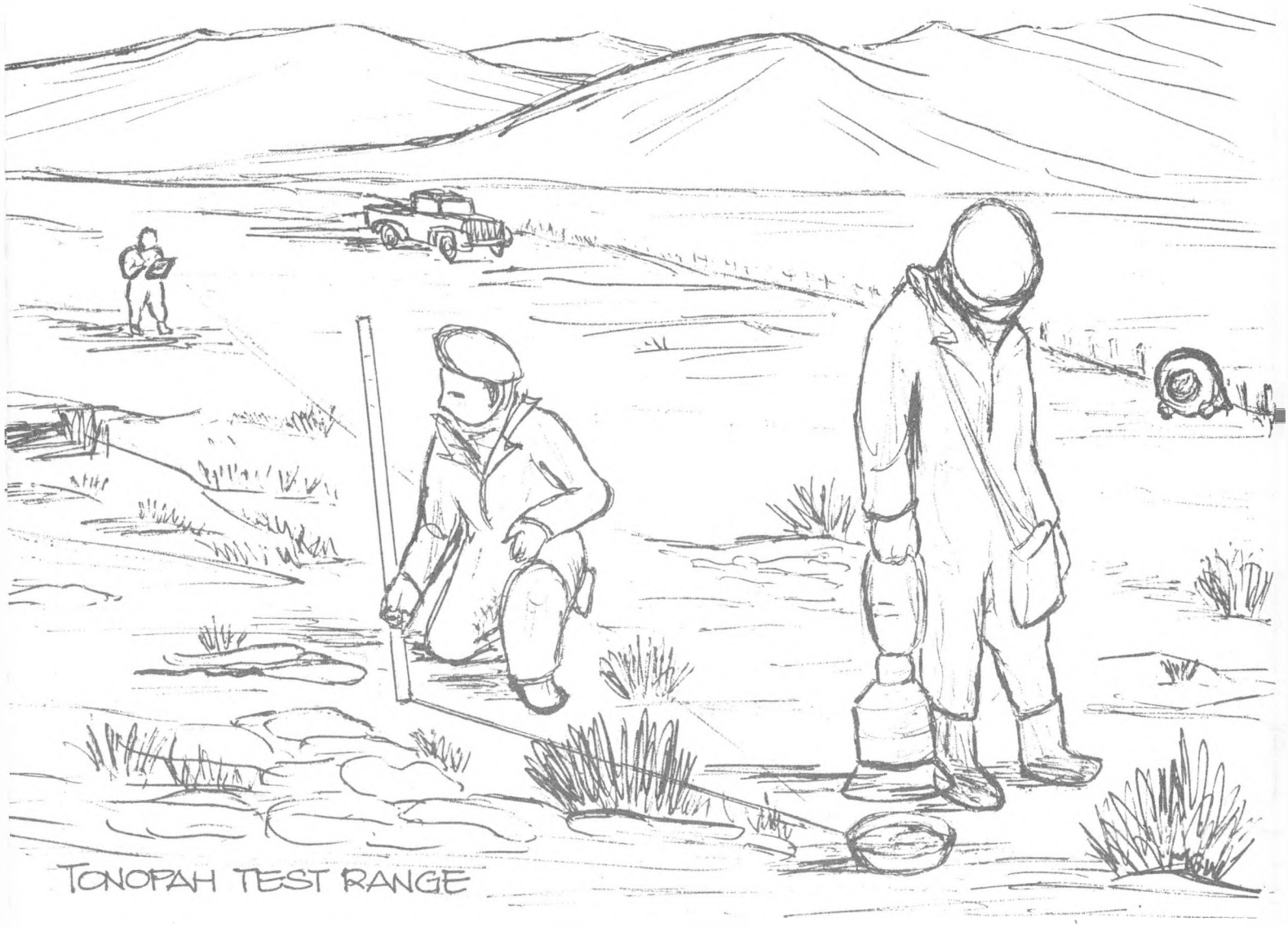
NVO-166
VOLUME 2 of 2

NEVADA APPLIED ECOLOGY GROUP

PROCEDURES HANDBOOK

FOR ENVIRONMENTAL TRANSURANICS

OCTOBER 1976



UNITED STATES
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Las Vegas, Nevada

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OCTOBER 1976

EDITED BY

M. G. White & P. B. Dunaway

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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
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Resuspension procedures and techniques

DEVELOPMENT OF SPECIALIZED INSTRUMENTS AND TECHNIQUES RESUSPENSION ELEMENT STATUS REPORT

Paul L. Phelps and Lynn R. Anspaugh

Bio-Medical Division
Lawrence Livermore Laboratory, California

(Ed. Note: Previously published in
NAEG Report NVO-142, pp. 221-233.)

ABSTRACT

An intensive study on the resuspension of plutonium at the Nevada Test Site has been initiated. The main thrust of the study is to develop a mathematical model for describing the concentrations of plutonium in air as a function of the source and driving forces. It is intended that the model be a basis for assessing potential health hazards and developing cleanup recommendations. In addition, the study is providing an opportunity to develop a generalized resuspension model, applicable in general for establishing environmental and biological assessment of nuclear projects involving plutonium.

Apparatus and experimental techniques for studying the dynamics of plutonium and soil particle behavior have been developed. This has included the development of ultrahigh-volume air samples ($1500 m^3/hr$) which allow collection of adequate samples of plutonium at worldwide air concentration levels in two hours of sampling time. We have also developed samplers for measuring the fraction of particles moving in creep and saltation. The Air Resources Laboratory, National Oceanic and Atmospheric Administration, has established an elaborate meteorological data-gathering system for measuring micrometeorological parameters, during sampling periods at the Nevada Test Site. Advanced optical methods for in-situ sizing and counting particles have also been employed. This technique allows observations of particle size and concentration of a few-minute time scale, such that rapid changes in meteorological parameters may be more directly correlated with the concentration of airborne particles and their size distributions.

The most intensive field program to date has been in the GMX area. Data have also been collected in Area 13 and Mercury. Ultrahigh-volume air samplers and high-volume cascade impactors were used to measure the concentration of plutonium in air and aerodynamic particle fractions of airborne soil and plutonium. Analysis of air samples collected by REECo from February, 1971, to July, 1972, shows that the GMX site, which was contaminated 17 years ago, still represents a significant

resuspension source. However, the average air concentration of resuspended ^{239}Pu outside the exclusion area is only a small fraction of the presently accepted maximum permissible concentration for occupational exposure. It was also concluded that the air concentrations of ^{239}Pu in Mercury may be influenced by the local NTS sources.

Measurements using cascade impactor studies indicate that there is no difference in the distribution of activity with particle size for the three species ^{238}Pu, $^{239,240}Pu$, and ^{241}Am. The total mass, however, was found to be distributed distinctly differently with a smaller median aerodynamic diameter and a larger geometric mean. The data also showed that the fraction of the resuspended plutonium aerosol at GMX which would be expected to undergo pulmonary depositions to be approximately 0.2 based upon the ICRP Task Group on Lung Dynamics model.

Experimental results have also shown that there is no obvious correlation of specific Pu activity with particle size. An average specific activity of 890 dpm/g, or about one-third of that found in the soil in close proximity to the cascade impactors, was measured. Particle data from the cascade impactors showed that the ratio of $^{239,240}Pu$ to ^{241}Am activity is the same as reported for soil in the vicinity of the cascade impactors. Preliminary results from the ultrahigh-volume air sampler runs indicate a gross correlation between many of the wind speed-related parameters and the concentration of resuspended plutonium.

Experience and data gathered at the GMX site were used to derive simple predictive models for air concentrations of plutonium due to resuspension on the Eniwetok Atoll.

INTRODUCTION

The Resuspension Element of the Nevada Applied Ecology Group (NAEG) was formed to undertake the task of studying the movement of plutonium at the Nevada Test Site (NTS) by wind-driven forces and assessing the potential biological hazards associated with airborne plutonium particles. The element also was to provide input to the Nevada Operations Office of the Atomic Energy Commission regarding cleanup of plutonium-contaminated areas. This involvement gave the Lawrence Livermore Laboratory (LLL) Bio-Medical Division an opportunity to study fundamental processes involved in the resuspension of plutonium particles from a soil surface, and to develop a time and spatial dependent mathematical model describing average concentration of airborne plutonium as a function of the source (geometrical configuration, soil surface characteristics) and driving forces. It is intended that the model will also provide the necessary input to models of lung dynamics of plutonium behavior in the human respiratory tract. The NAEG Resuspension Element's plan for meeting the above objectives is diagramed in Fig. 1.

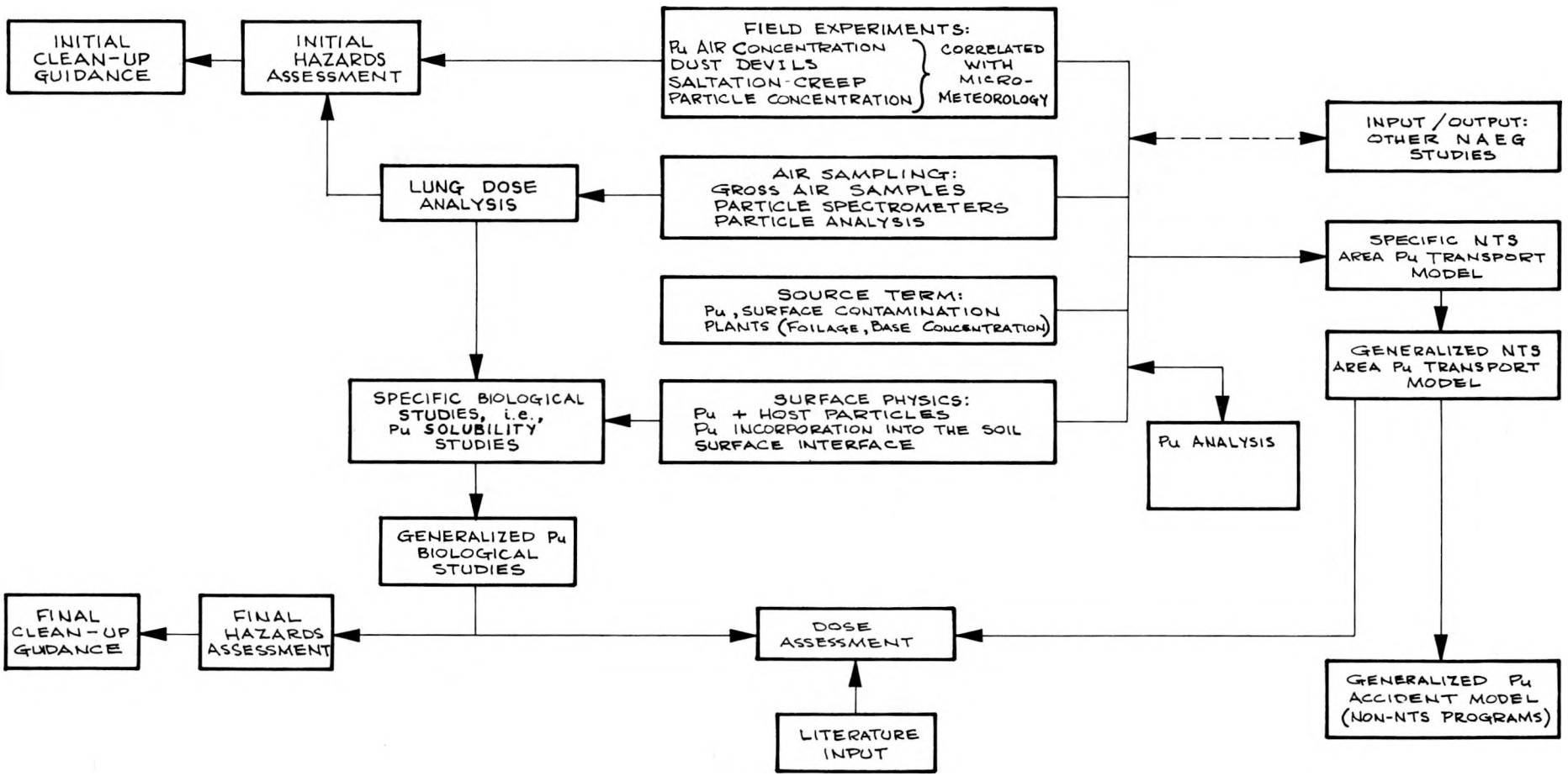

FIG. 1. THE INTERRELATIONSHIP BETWEEN THE FIELD EXPERIMENT (DATA GATHERING) AND OBJECTIVES OF NAEG.

Prior to LLL involvement in the NAEG program, Livermore had carried out resuspension studies related to nuclear cratering experiments and accidental releases from nuclear device tests. One of these studies was in conjunction with the Schooner cratering experiment (Anspaugh *et al.*, 1969; Anspaugh *et al.*, 1971; and Anspaugh *et al.*, in press).

Radioactivity measurements were made on an arc approximately 50 miles from the Schooner surface ground zero (SGZ). Some of the highest readings were obtained in the vicinity of the Queen City Summit outside the northern boundaries of NTS. Based upon conjecture that the high readings were caused by unique interactions between wind flow and topography at the Queen City Summit region which had existed during the Schooner cloud passage, it was hypothesized that similar conditions could have existed during atmospheric device testings in previous years at NTS which could have resulted in unusually high plutonium depositions at the same site.

Consequently, soil samples were analyzed for plutonium and data were obtained in support of the hypothesis. Plutonium levels in soil at Queen City Summit were found to be 100 times over the average background levels in U.S. soil. Subsequent measurements were made by the Environmental Protection Agency's National Environmental Research Laboratory, Las Vegas (formerly the U.S. Public Health Service), which confirmed the LLL's earlier findings. These findings added impetus to carry out resuspension studies at NTS.

Subsequent to starting the plutonium resuspension field studies, the opportunity arose to conduct a long-term experiment using the radioactivity deposited on the soil surface during the Baneberry accidental venting (Anspaugh *et al.*, in press). The incident occurred in December, 1970. The study was carried out over approximately a one-year period. The study was directed at observing the resuspension of radioactivity as a function of time.

Following the Baneberry study, efforts were directed toward designing a series of experiments at plutonium-contaminated areas on NTS. A joint proposal from LLL and NOAA/ARL outlining the experimental plan was submitted for AEC review. Early in 1972, research, development, and procurement were started on special apparatus needed to carry out the experiment. This included developing ultrahigh-volume air samplers, in-situ particle spectrometers, light-scattering-type particle counters, and establishing a micrometeorology field laboratory. A plutonium-contaminated site was selected and surveyed for layout of the experiment.

RESUSPENSION EXPERIMENT: GMX-AREA 5

The GMX Site, in Area 5, was selected for conducting the first plutonium resuspension experiment. This area had previously been used for carrying

out a series of experiments involving plutonium and high explosives. At no time were nuclear detonations involved. These plutonium-high explosive tests started in late 1954 and ended in early 1956. Thus, as of this date, the mean life of the plutonium source at GMX is 18 years. Following the 1954-1956 series of tests, the highly contaminated area was fenced off and remained essentially undisturbed.

Various groups within the NAEG program have gathered data which support the resuspension studies. These data include the areal distribution of plutonium, the distribution and concentration of plutonium within the soil, and a population census of vegetation covering the GMX study area.

The initial resuspension experiment was designed with the goal of developing a model to predict air concentrations of plutonium, given the source characteristics and relevant meteorological parameters. The objective was then to conduct a series of measurements within and downwind of the plutonium source. This was to be done during a period of time when the winds were consistent. Examination of several years of wind data showed that this was possible for a 2-4 hr period. The experiment was laid out as shown in Fig. 2. The line showing the sampling stations is located at 020° true, which corresponds to the mean direction of winds over 10 mph from March 24 to August 30, 1971, at the GMX site.

Micrometeorological measurements were directed at characterizing the driving forces; i.e., winds, turbulence, and related parameters that are considered to be most likely related to moving or picking up particles from the desert pavement. These measurements are discussed in detail by Kennedy and Booth (this report). The main micrometeorology station was established 1200 ft downwind from GZ, at R-1, as shown in Fig. 2. Air-sampling stations were established at GZ, 1200 ft, 2400 ft, and 4800 ft from GZ. In addition to the downwind sampling stations, control stations were established upwind from GZ, at Mercury, and in Livermore, California. These background stations utilized the identical type of ultrahigh-volume air sampler.

During a field experiment, which usually lasted from 1-4 hr, the various micrometeorological parameters were recorded and the ultrahigh-volume air samplers were operated. All the samplers were synchronized to start and stop together. Concurrent with these measurements, particle spectrum analyses at various heights above the ground were made using the Climet particle analyzer (Koval, this report). Also overlapping in time with these measurements, background air samples were collected 900 ft upwind from GZ; at Mercury, Nevada; and in Livermore, California.

Saltation/creep samplers have been installed for collecting particles that roll or bounce on the soil surface (Reichman, this report). These samplers as yet have not been integrated into the field experiments. It is planned to finish evaluation studies in 1974 and use the instruments shortly afterwards.

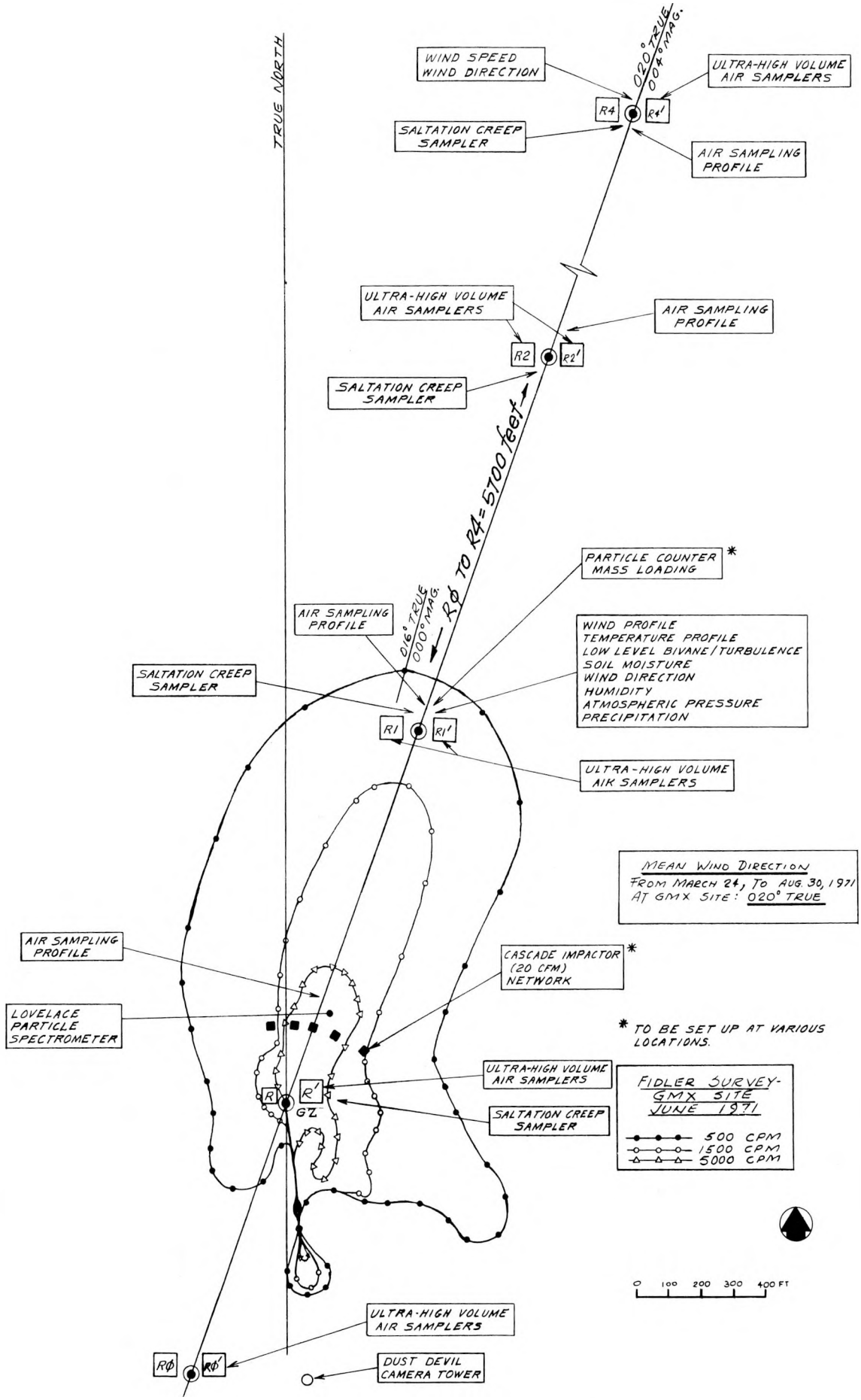

FIG. 2. LAYOUT OF THE RESUSPENSION EXPERIMENT AT GMX, AREA 5.

Cascade impactors were located on an arc 250 ft from GZ, and were run continuously for approximately 30-day periods. These instruments gave results on particle size fractions for plutonium and total airborne particulates. Results are given by Anspaugh and Phelps (this report).

A time-lapse camera on an 80-ft tower was located upwind from GZ such that the entire GMX experimental site was in its field of view. The camera has been operated during the daylight hours with a frame exposure every 5 sec. During the times of high dust devil activities (June-September), the camera was used to record dust devils traversing the GMX experimental site. This was done to qualitatively assess the impact of dust devils on transporting surface material into the atmosphere. The data which were available for the GMX experiment are presented in detail by Anspaugh and Phelps (this report). A preliminary analysis of the air concentration data collected by REECo prior to the initiation of this study is also presented by Anspaugh and Phelps (this report).

DEVELOPMENT OF SPECIALIZED INSTRUMENTS AND TECHNIQUES

A variety of specialized instruments had to be designed and fabricated before the plutonium redistribution experiments could be conducted at NTS. One of the major undertakings was the development of an air sampler capable of collecting adequate amounts of airborne plutonium particulates during a 2-4 hr sampling period. This time period corresponded to the time "window" during which consistent meteorology could be expected. Calculations indicated that even if the most sensitive radiochemical procedures were used in analyzing the air filters for plutonium, an air sampler with a through-put at 1500-2000 m^3/hr would be required. Samplers with these required flow rates were not available; therefore, development of an ultrahigh-volume air sampler meeting the desired specifications proceeded. Tests on the prototype sampler were very encouraging, indicating that even at global background levels, surface air could be sampled for a time period of 1 hr, and accurate determinations of plutonium air concentrations could be made. Fourteen ultrahigh-volume air samplers were eventually fabricated. They are deployed in Area 13, GMX, and Mercury. One unit is in operation at Livermore.

Micrometeorological forces that may cause resuspension of particles from the soil surface have significant transient behavior over very short periods of time, probably less than a second to a few minutes. The airborne particles resulting from resuspension normally are collected by use of a filter media -- airpump combination. The collected particles are then analyzed for plutonium, and the concentration of radioactivity in air is determined from the ratio of plutonium to the amount of air sampled. Therefore, since sampling must be done

over a minimum of 1 hr, which corresponds to the least time possible with the ultrahigh-volume air sampler previously mentioned, mechanisms related to pickup of particles from the soil surface cannot be studied in real time and with high resolution.

To overcome these limitations, two light-scattering instruments were adapted to measure the mass loading of particles in air and the number distribution as a function of particle size over very short periods of time. Both instruments give data in real time, such that it is possible to correlate micrometeorological events of short duration with the concentration of airborne particles. These instruments examine the particle spectrum in the region of 0.1 to 10 μm, and do not differentiate between radioactive and nonradioactive particles. Nevertheless, valuable data have been obtained on the mass loading of particles in air as a function of the forces causing pickup from the soil surface.

The first instrument employed was a light-scattering-type particle analyzer. This instrument, when used with a pulse-height analyzer, will give particle size histograms over a range of 0.5 to 10.5 μm. Sufficient data can be collected in periods of 2 min or less. Particle sizes as measured by this technique are related to their physical diameters. If the physical properties of the particles are known, aerodynamic or "effective" particle sizes can be determined. This is necessary for modeling of particle deposition in the human respiratory tract. Details of the instrument and its application to the GMX experiment are covered by Koval (this report). Another light-scattering device currently employed is the nephelometer. It has extremely fast response (< 1 sec); however, it does not give particle size distribution data, only relative particle concentration. It is an exceptional instrument for correlating very short-time duration meteorological events with mass loading of particles in the air.

A recently introduced high-volume (20 cfm) cascade impactor was adapted for use in the GMX experiment. This instrument has five stages which correspond to 1.1, 1.1-2.0, 2.0-3.3, 3.3-7.0, and greater than 7.0 μm. The filter papers may be analyzed for total plutonium per stage and total mass for all particles on each stage. Thus, the particles picked up from the soil surface may be characterized according to their aerodynamic sizes, and the relationship between plutonium and total mass may be established. Further refinements are being made on this instrument which will reduce reentertainment of particles and improve its particle intake characteristics.

Particles that roll or bounce along the soil surface (creep/saltation) may be responsible for the most significant net movement of surface mass compared with other processes. Existing instruments that had been used in agricultural erosion studies and for measuring the movement of sand in windstorms were evaluated. A series of laboratory studies were conducted at LLL on borrowed or copied instruments. Out of this work, a multistage sampler was developed, capable of collecting the particle fraction moving by creep and particles moving in saltation at various levels above the surface. Details on the design and preliminary results are covered by Reichman (this report).

Initial dust devil observations were made with a time-lapse camera adapted for use on an 80-ft tower. The camera was programmed to expose one frame every five sec. Experimentation with optimum camera location and lens selection resulted in ability to view the entire GMX experimental site. This system was developed as a survey tool in order to get an approximate idea of the dust devil population. A visual count in the GMX and surrounding area during a 1-hr period in July, 1973, showed a dust devil population of approximately one dust devil formation per minute, with an average lifetime of about 2 min.

An elaborate meteorological data-gathering system, capable of measuring wind and temperature profiles, fluctuations of the horizontal and vertical wind structure, soil moisture, soil temperature, humidity, rainfall, and other miscellaneous meterological measurements, was set up. This was under the direction of the Air Resources Laboratory-Las Vegas, and is covered in detail by Kennedy and Booth (this report).

FUTURE STUDIES

A number of problems dealing with understanding redistribution of radioactivity at NTS still remain. These problems are related to possible long-term health and safety issues, future land uses of NTS, and other AEC nuclear programs. So far, LLL studies have involved the movement of plutonium at the GMX site, and redistribution of gamma emitters released from the Schooner cratering event and the Baneberry device test. To date, the GMX studies have focused on plutonium particles carried into suspension by the force of the wind. Emphasis has been on collecting samples of airborne plutonium particles and measuring a number of micrometeorological parameters, such that the relationship between air concentrations of plutonium and the important driving forces may be understood, and a mathematical model constructed. The wind forces studied so far have been diurnal in nature, and occasionally those associated with dust storms.

Dust Devils

There is, however, another driving force: dust devils, which might be a significant mechanism under certain circumstances for transporting surface radioactive material into the atmosphere.

Because of the high wind speeds generated close to the ground and the large vertical structure, dust devils are capable of removing and transporting vertically a significant portion of loose surface material. Dust-devil measurements by Sinclair (1966, 1973) indicate that on the average, a typical dust-devil source region of 285 km^2 provides a total mean seasonal vertical dust transport of over 7×10^3 tons of desert sand. This is one to two orders of magnitude larger, for the same surface area, than pre-dust storm conditions (Gillette *et al.*, 1972)

and smaller by approximately the same amount within the most intense dust storms. Consequently, the vertical and horizontal transport of surface material by dust devils may in itself be an important component in the overall resuspension budget.

However, of more importance is the question of transport and ultimate fate of radioactive materials by dust devils and, in general, by atmospheric convective elements that originate in the surface boundary layer. LLL plans to make direct quantitative measurements of the amount, the distance, and the direction of dust transported from a known dust devil source region at NTS. The analysis of these data will provide estimates of the possible radioactive substances such as plutonium by dust devils.

Variation in Site Characteristics

The availability of plutonium for resuspension is related to a significant degree to the soil-plant surface characteristics of the plutonium-contaminated area. This is particularly true for aged sources that are found at NTS. A number of plutonium-contaminated sites exist at NTS. Each of the sites have more-or-less different and unique characteristics that influence the availability and movement of plutonium. The properties of importance are the initial physical-chemical form of the source, source age, source strength (surface related), source area and shape, physical-chemical makeup of the soil, soil surface structure, plant coverage, animal population, climatology, and land utilization. These properties are to be intensively studied.

An example of an area not yet studied is the Tonopah Test Range. This area is suitable for cattle grazing. A native species of grass, not found in the GMX area, is in abundance. Also, the physical "desert pavement" characteristics are significantly different than those so far studied. The Tonopah area affords an opportunity to study a plutonium-contaminated area with surface characteristics not heretofore studied.

Man-Made Effects on Redistribution

Displacement of surface soil by mechanical means can increase the rate of redistribution. The impact from future uses of NTS is of particular interest. In addition to continued device testing, future use may include radioactive fuel processing, radioactive waste storage, solar energy research, and agricultural uses. Coupled with these uses are a variety of ways that sufficient mechanical disturbances to contaminated sites can result. These include construction of facilities and roads, vehicular traffic over desert pavement, and grazing by cattle. A number of the areas contaminated from past device testing and nuclear cratering events should be studied. A natural follow-up experiment in the GMX area would be to introduce a variety of man-made disturbances to the soil surface and quantitate the change in resuspension of plutonium.

Source Term Analysis and Redistribution of Radioactivity at NTS

An evaluation of the total NTS contribution of dose to man is limited by the lack of accurate mapping of the resuspendable radioactive source material distributed throughout the site. However, elements within the NAEG program are compiling information on several radionuclides and their distribution in soil, which, if extended to all radionuclides and areas, could be the basis for input to such a mapping.

Assuming that a data bank on radionuclide distribution at NTS were established, the other necessary inputs would be a comprehensive modeling of the meso-scale meteorology related to wind flow at NTS, and an understanding of resuspension for the variety of source characteristics present throughout the site.

This study would provide information on the relationship between source and the dose to man from existing radioactivity and possible future uses.

ACKNOWLEDGMENTS

The authors wish to thank the numerous persons who have contributed to this study. Members of the Lawrence Livermore Laboratory who constructed and fielded the experimental apparatus, at times under very trying conditions, included J. Taylor, C. Fry, F. Green, V. Fowler, and B. Tuckey. R. Kauder of the Air Resources Laboratory-Las Vegas was a principal participant in the meteorological measurements and data handling efforts. We also wish to thank I. Aoki and A. Bicker of the Reynolds Electrical and Engineering Co., Inc., for supplying air-sampling data for the GMX area, and R. Cooper, who was extremely helpful and resourceful in maintaining the field apparatus. Several members of the U.S. Atomic Energy Commission, Nevada Operations Office, staff have contributed their support. They include P. B. Dunaway, M. G. White, R. Lease, and H. Kayuha. Finally, a special thanks to S. Lewis for typing the manuscript, D. Schilf for typing the numerous reports, and to A. Boyce, who did the illustrations.

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MEASUREMENTS OF METEOROLOGICAL PARAMETERS

Norman C. Kennedy and Howard G. Booth

Air Resources Laboratory--Las Vegas
National Oceanic and Atmospheric Administration

(Ed. Note: Previously published in
NAEG Report NVO-142, pp. 235-239.)

A part of the Air Resources Laboratory-Las Vegas support to the Resuspension Element has been the recording of meteorological information at air sampling locations. Early in the program, it was recognized that some, even if rather gross, measurements of wind direction, speed, and precipitation were desirable, and these have been made at REECo and LLL air sampling sites. These data are being used in the analysis of sample variation.

In conjunction with the more detailed GMX area resuspension experiments, a more elaborate meteorological data gathering system has been established. A brief outline of the wind, temperature, moisture, and other miscellaneous meteorological measurements made during sampling periods follows:

Wind

Mean wind direction is measured using standard vanes. A bivane is used to measure fluctuations of the horizontal and vertical directions with electronic computation of the resulting standard deviation of the azimuth and vertical angles.

Horizontal wind speed is measured at six levels (0.5, 1.0, 2.0, 4.0, 8.0, 10.0 m) using cup anemometers. Wind tunnel calibrations of the 3-cup anemometers are made periodically. Cup revolutions of the lowest 5 sensors are counted and converted to mean horizontal speed for the period of interest, usually a 10-min to 6-hr period. Analog recordings of speed and direction are made for the 10-m sensor to give a real time indication of wind characteristics and a record of gust speeds at that level.

Temperature

Temperature is sensed at and recorded for five levels. Two levels are at 1 and 3 cm below the soil surface and three levels are at 0.5, 2.0, and 8.0 m. Aboveground sensors are in aspirated shields.

Moisture

Relative humidity is measured using a hygrothermograph supplemented during experiments with observations using a sling psychrometer.

Soil moisture is obtained for representative times using the average of five approximately 100-g samples of the top 0.5-cm layer of the soil. Samples are weighed, then dried and reweighed.

Precipitation is measured using a recording rain gage.

Solar Radiation

Incoming and outgoing solar radiation are measured using a pyranometer and two radiometers. These measurements will be used primarily for dust devil studies.

Other Measurements

Atmospheric pressure is measured with an aneroid barometer at specific intervals.

Clouds are observed visually, as well as sensed by the pyranometer.

Visible weather phenomena, such as showers in the area, blowing dust over the dry lake, and dust devil activity, are recorded.

An upper air sounding (radiosonde) is available twice a day at standard observation times from a station about 10 miles to the northwest of the GMX area.

Discussion

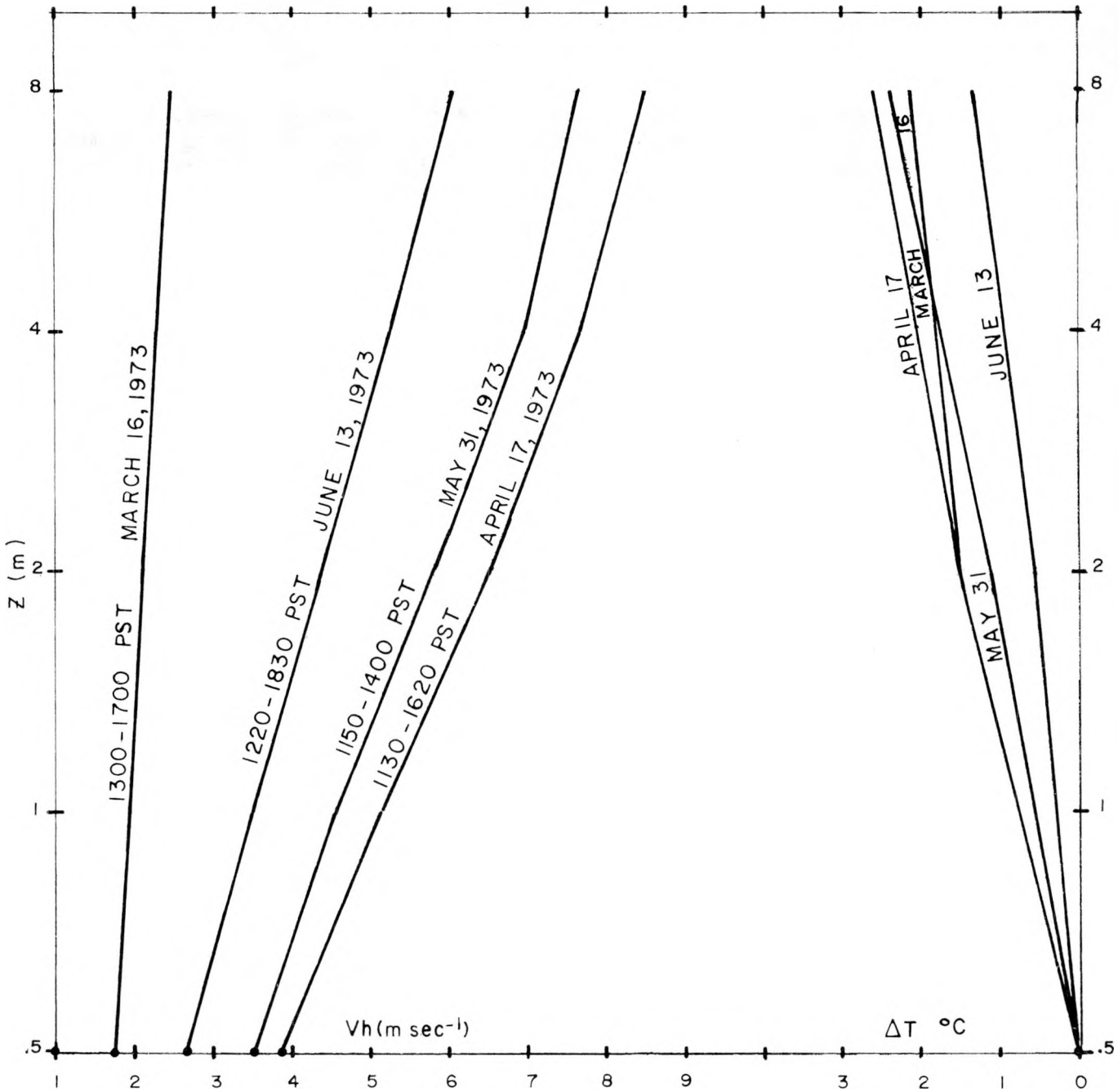
The system has been operated about 30 days over the past year for system checkout, during ultra-high volume air sample runs, and during particle analyzer runs.

Figure 1 shows the wind and temperature profiles for 4 high-volume air sampler runs. One profile is for light winds, one for strong winds, and the other two for the initial two runs for which the ^{239}Pu activity data has been received.

The wind speed profile is log height/linear speed diagram, using mean wind speed for the period the sampler was operating. The temperature diagram is similar but uses temperature differences between levels. Soil temperatures were about $10^{\circ}C$ higher. The wind speed profile gives a measurement of the vertical shear of the wind and an indirect estimate of eddy momentum flux (or stress) through computation of the friction velocity (U*). This is done with reservations during periods of extreme thermal instability.

Certain recorded and derived parameters are being observed in order to characterize the meteorology in ways that will relate it to the variation in the measurements of suspended material.

Wind direction is defined as the mean direction for the sample period. The sampler array is on a bearing of 200 degrees, which is close to the



DATE	TIME PST	WIND DIR DEG	MEAN GUST m sec^{-1}	SOIL MOISTURE %	CLOUD COVER	RICHARDSON'S NUMBER	σ_w (1 m) m sec^{-1}
730316	1300-1700	180	4.0	0.3	0	-0.958	.21
730613	1220-1830	210	9.8	0.3	.8	-0.025	.39
770531	1150-1400	209	13.9	0.3	.8	-0.033	.52
730417	1130-1620	216	13.9	0.4	.2	-0.030	.44

FIGURE 1. METEOROLOGICAL DATA FOR FOUR UHV AIR SAMPLER RUNS AT GMX SITE.

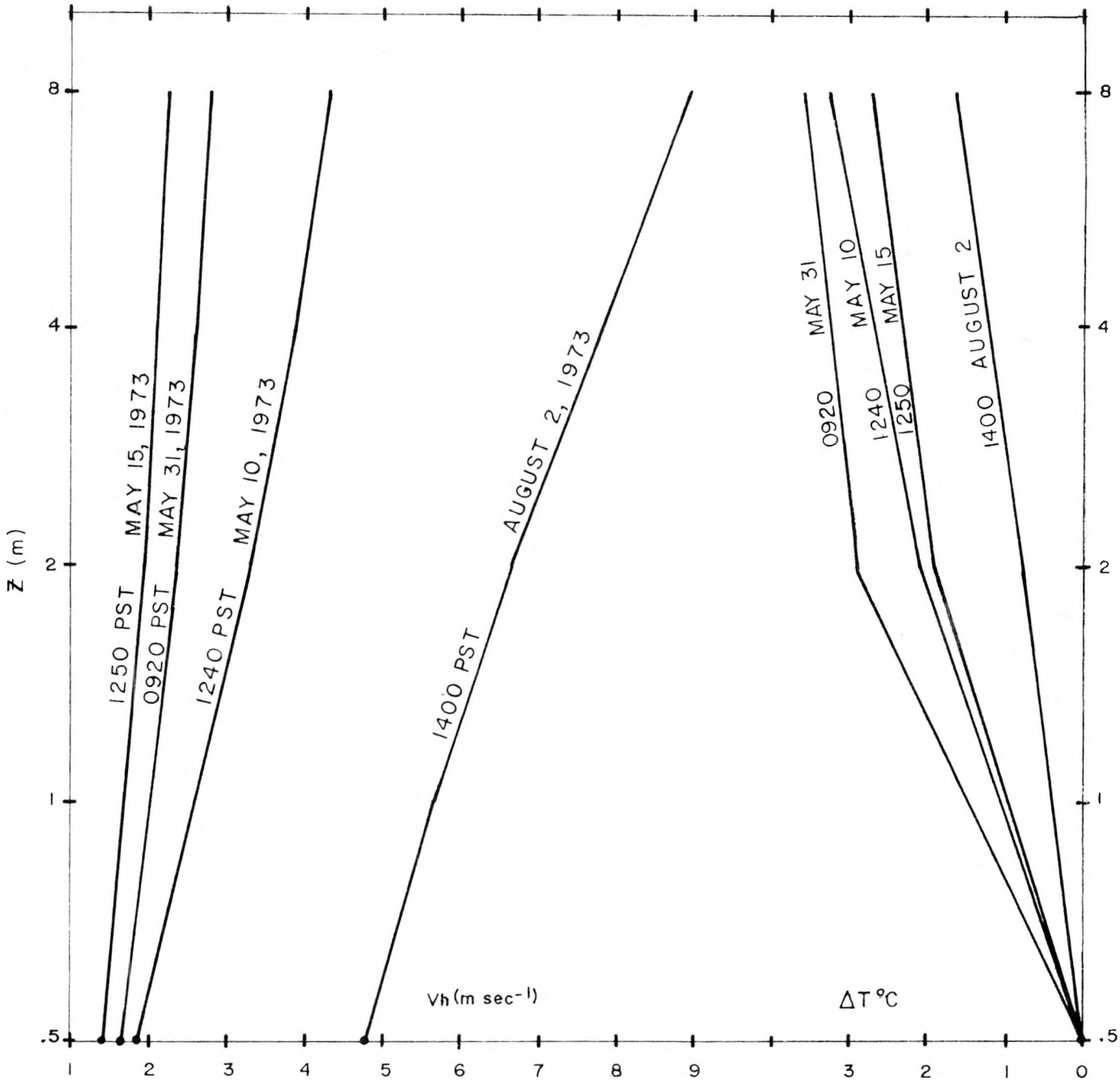
climatologically most frequently observed downwind direction. Mean gust indicates the mean of the peak gusts recorded for each 10-min increment during the sample period. Soil moisture has varied from abount 10% to 0.1%. Cloud cover is the mean of hourly values of low and middle cloud sky coverage.

The temperature profile is a measure of vertical stability and, in combination with the wind speed profile, gives a parameter which is a measure of the ratio of buoyant to inertia forces (Ri).

The mean and standard deviations of the horizontal component of the wind direction are indicators of the direction of travel and horizontal dispersion. The standard deviation of the vertical component of wind direction is an indicator of the vertical dispersion and also can be used to estimate eddy momentum flux.

Figure 2 shows information similar to Figure 1 for 10-min periods that correspond to particle analyzer run times.

In general, attempts are being made to measure some of the differing values for meteorological parameters that will assist in understanding variations in the air samples. It is planned to relate the measures of moisture to particle availability, wind pressure forces to initial particle motion, eddy momentum flux to suspension, and wind direction variance to dispersion.



DATE	TIME PST	WIND DIR. DEG.	MAX. GUST m sec⁻¹	SOIL MOISTURE %	CLOUD COVER	RICHARDSON'S NUMBER	σW m sec⁻¹
730515	1250	200	3.6	0.4	.4	- 1.20	.25
730531	0920	155	4.0	0.3	.1	- 0.675	.30
730510	1240	225	8.0	0.3	0	- 0.132	.29
730802	1400	185	17.4	m	.6	-0.0217	.75

FIGURE 2. METEOROLOGICAL DATA FOR FOUR PARTICLE ANALYZER RUNS AT GMX SITE.

ULTRA HIGH VOLUME AIR SAMPLER

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General Description of Air Sampler

An air sampler with a nominal volumetric flow rate of 1500 m^3/hr was developed in order that a measureable amount of ^{239}Pu could be collected in as short a time span as 2 hr. A photograph of the air sampler is shown in Fig. 1. The design of the unit is patterned after an air sampler built by Asikainen and Blomqvist (1970) which had a flow rate of 1000 m^3/hr. The air flow through the sampler is maintained by a centrifugal blower thats inlet is attached to one side of a large plenum. The filter, which measures 0.6 x 1.7 m, is mounted horizontally on top of the plenum at a height of 1.3 m above ground level. The blower exhausts through a 0.3-m diameter of 3-m-long sheet metal duct, which is inclined at an angle of 10^o with respect to the ground. The length of the duct precludes any interference of the exhaust flow stream with the air intake to the sampler. The duct is inclined so that the exhaust flow stream dissipates without directly striking the ground, which would artificially introduce aerosol into the air. The centrifugal blower is a 0.4-m-diameter wheel with radial blades and is driven by a 7.5-hp electric motor.

Filter Material

The filter material used is Delbag 99/98 Microsorban,* which is made of mats of polystyrene fibers with diameters of 1 μm and less. This material was chosen because of its low flow resistance, high particle collection efficiency, and ease of analysis for radionuclides. At the nominal flow rate of 1500 m^3/hr, the pressure drop across the filter is approximately 3000 Newtons (N)/m^2. For a face velocity of 0.42 m/sec, corresponding to the above nominal flow rate, the penetration of 0.3 μm diameter particle is 25%. At all other face velocities, the penetration is less than 0.25% (Lockhart *et al.*, 1964).

Due to poor mechanical strength of Microsorban, a backing material must be used beneath it. This combination is held in a hinged filter holder made of stainless steel. The filter material is supported on a 12.5-mm-mesh screen with a 1.5-mm-diameter wire. The filter holder is

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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 1. LLL UHV AIR SAMPLER

taped to the air sampler to eliminate any air leakage around edges of the holder. A sponge-rubber gasket is mounted on a hinged metal frame, which is used to clamp the filter holder firmly to the air sampler.

Flow Calibration

The volumetric flow rate of air through the filter is determined during each test run by the rate of rotation of an unloaded fan blade located in the exhaust duct. The unit consists of a modified cup-type anemometer with the cup rotor replaced with a 0.25-m-diameter, six-blade aluminum fan. An electromechanical counter records one count for every 60 revolutions of the fan blade. Given the number of counts/min, the air flow rate can be determined from a calibration curve such as that shown in Fig. 2. Notice that the rate of revolution of the fan blade is a linear function of flow rate. An independent measurement of the flow rate is obtained from the pitot tube located 1.2 m upstream from the fan blade.

Prior to deployment in the field, each air sampler is calibrated with a Meriam laminar flow element, which is a primary flow measuring element. A schematic of the calibration setup is shown in Fig. 3. The laminar flow element is connected to the sampler by a straight duct 10 diameters long, as recommended by the manufacturer, and by a transition piece above the filter, which is not present when the air sampler is in field use. The flow rate is determined by measuring the pressure drop across the laminar flow element to within 0.5% accuracy. The overall accuracy of the calibration technique is estimated to be within 5%. Both the counts/min from the modified anemometer and the pitot tube reading of the velocity pressure are calibrated against the laminar flow meter over a range of flow rates as shown in Fig. 2. Note that the velocity pressure as measured by the pitot tube is a function of the air density, as well as volumetric flow rate.

Experimental Procedure

Prior to each test run, the Microsorban filter material is placed in a decontaminated filter holder. The filter holder is then put in a sealed plastic bag for transportation to the site of the air sampler. The air sampler has previously been decontaminated and sealed by taping an aluminum cover in place. The aluminum cover is shown in the foreground of Fig. 1 in its storage location on the side of the sampler. Several hours before a test run, the filter holder is removed from its plastic bag and is placed in the air sampler by raising the hinged framework on which the aluminum cover is taped. After all of the edges of the filter holder have been taped in place, the hinged framework is lowered and secured. A thermal radiation shield is attached 80 mm above the aluminum cover to ensure that the Microsorban filter material does not exceed its maximum operating temperature of $60^{\circ}C$ while installed in the sampler.

At the start of the test run, the aluminum cover is removed and the centrifugal blower is turned on. The counts from the modified anemometer, as well as the pitot tube reading, the atmospheric pressure, and temperature, are recorded at the beginning and end of the test run, normally a period of 4-5 hr.

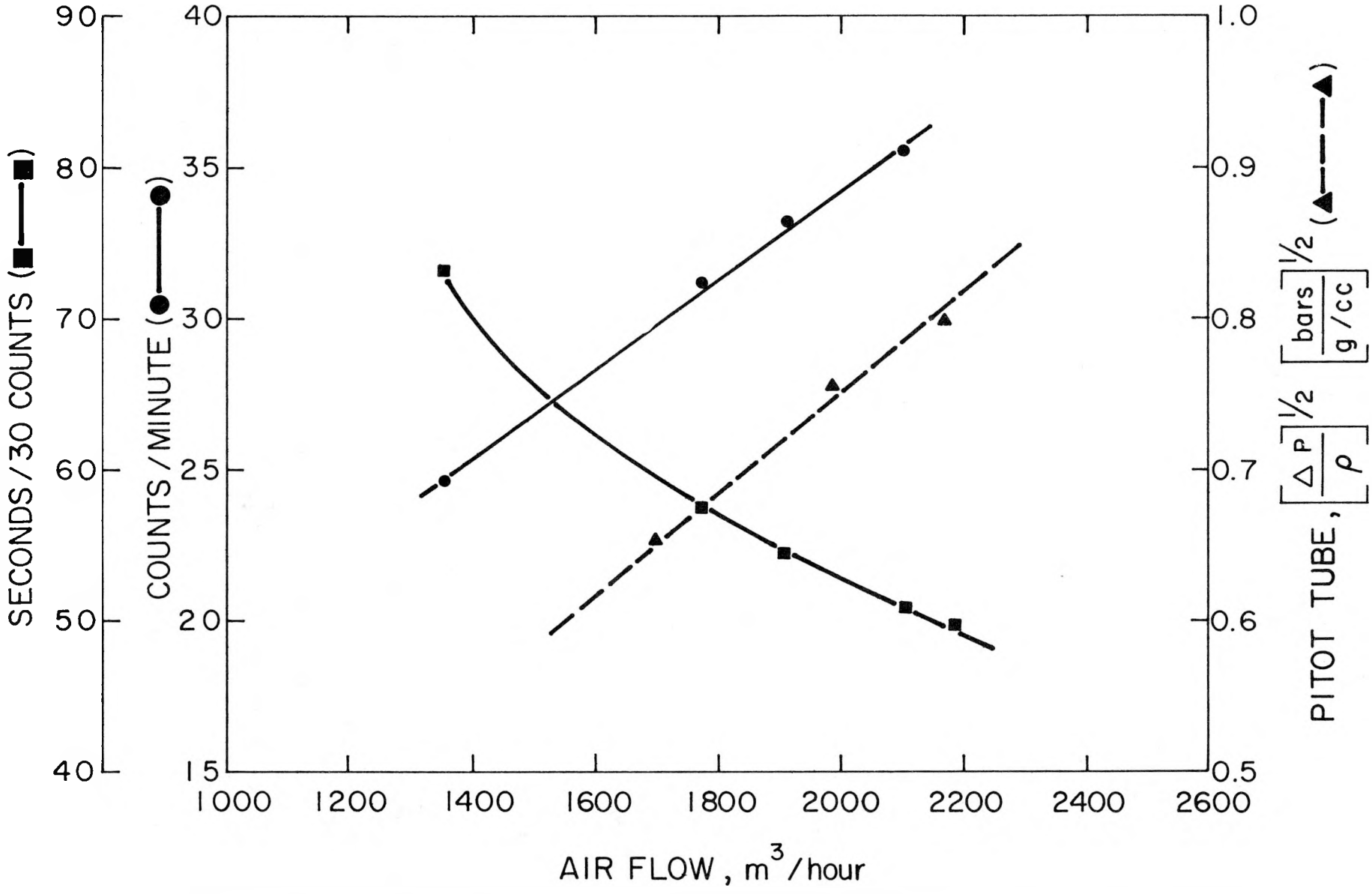

FIGURE 2. TYPICAL CALIBRATION CURVE FOR THE UHV AIR SAMPL

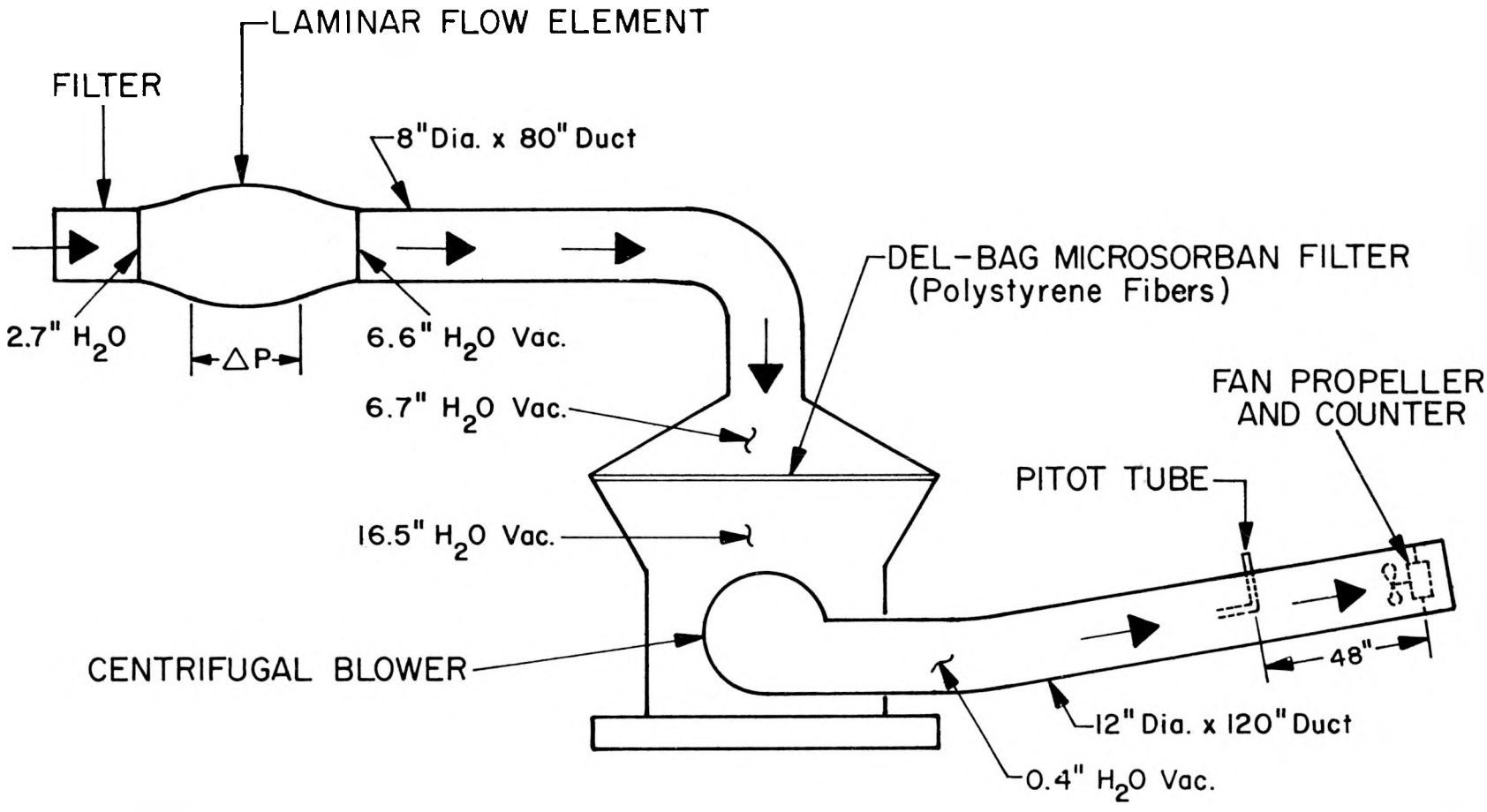

FIGURE 3.
SCHEMATIC OF CALIBRATION SETUP FOR THE ULTRA HIGH VOLUME AIR SAMPLER

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SALTATION AND CREEP SAMPLER

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INTRODUCTION

Motion of windblown sand close to the ground can occur by two mechanisms. The first of these mechanisms is creep, which is the rolling motion of sand along the ground. The second mechanism occurs when wind speed increases and sand "hops" along the ground in short, arching paths; this is saltation. Measurement of these mechanisms of sand movement, together with the resuspended sand, is necessary for the understanding of the total flux of windborne soil. In previous studies, various devices have been used to measure saltation and creep (Sehmel and Lloyd, 1972; Bagnold, 1954; Gillette and Goodwin, to be published). In order to develop an efficient sampling unit, several of the above devices were evaluated to determine the most efficient. Modifications then were made to further increase efficiency. Testing of the units was conducted under controlled conditions at Livermore in order that more reproducibility could be obtained than would be possible at the Nevada Test Site (NTS).

DISCUSSION

Two types of samplers were tested, one was of the funnel type (Fig. 1), developed by Sehmel and Lloyd (1972). The second (Fig. 2) was of a type developed by Bagnold (1954) and was borrowed from Dale Gillette of the National Center for Atmospheric Research. The two samplers were of equal heights, but the funnels had a cross-sectional width of approximately 7 cm, while the Bagnold collector had a 1-cm width. Saltation was experimentally simulated by the use of a 1/2-hp blower with its nozzle angled down at 45^{o} to the ground. The blower was used to lift soil into the air where it was then carried by its own momentum and the momentum of

FIGURE 1. FUNNEL SAMPLER FABRICATED AT LLL BASED UPON SEHMEL DESIGN (1972).

FIGURE 2. BAGNOLD SAMPLER USED FOR COMPARISON TESTS (BORROWED FROM GILLETTE OF THE NATIONAL CENTER FOR ATMOSPHERIC RESEARCH).

the prevailing wind toward the two samplers. The samplers were spaced 6 ft apart, with the distance measured perpendicular to the wind, so that the wakes would not interfere with the flow pattern around each sampler. The blower was also situated in a manner such that each sampler was exposed to the same 2-phase flow. This experimental situation did not perfectly duplicate actual physical conditions of saltation. However, quantitative information concerning relative effectiveness of each sampler was obtained.

Tests were conducted with collection times varying from 45 to 90 min. As these tests were planned for comparison of the two samplers, the ratio of mass collected by each sampler was the only information required. However, since the funnel sampler results could conveniently be broken down into mass vs height data, this was also done. In Table 1, measurements with the funnel collector (mass collected vs height) are presented, together with the total mass for each sampler, for a typical run. For Livermore tests, the mass collected by the Bagnold collector was consistently .85 that collected by the funnels. However, because of the larger collection area of the funnel sampler, it appears to be approximately only 20% as efficient as the Bagnold collector. Tests conducted by Gillette and Goodwin (to be published) on the Bagnold sampler show the collection efficiency to be 60%, while Horikawa and Shen (1960) found an efficiency of 53%. Efficiency is defined as the actual mass of sand collected/mass of sand that should have passed through the collection area. The above results imply that the funnel sampler would be only 12-15% efficient; this is in contradiction to the initial assumption made about the funnel sampler (Sehmel and Lloyd, 1972). The results, however, agree with recent findings by Sehmel (personal communication, undated).

Other conclusions about the samplers can be drawn from Livermore experimental results which can be applied in the evaluation of new sampler designs. Livermore experiments show that a difficulty arises when a circular sampler is used, since there is a degree of uncertainty involved in determining the width of the collection area. This complicates the actual determination of the collection efficiency for the unit. It is also evident from Livermore observations that a detailed analysis of the particle paths would have to be performed to fully understand the funnel-type sampler. The experiments further point to a very practical consideration in designing a sampler, and that is the ease of sample retrieval in the field. In the case of the Bagnold collector, the sample was collected in a box below the unit; this box could be removed conveniently. In the case of the funnel collector, each of the 14 funnels had to be removed and sealed in a clean plastic bag. During field operations, this proved to be a time-consuming process.

Both of the above-mentioned units proved to have the same disadvantage: both units are more appropriate for soil erosion studies, and as a result are more suitable when at least 50 g of sand are collected. For a typical test conducted at Livermore with LLL experimental apparatus, on the order of 3 g of sand was collected by each sampler. For the

Table 1. Typical Collector Data Run. Length of Run, 60 min.

FUNNEL DATA	
Funnel Height (cm)	Mass (g)
3.81 cm	1.30
7.62	.56
11.43	.29
15.24	.37
19.05	.12
22.86	.11
26.67	.04
30.48	.03
34.29	.02
38.1	.01
41.91	.03
45.72	.01
49.53	.04
53.34	.04
TOTAL	2.97

BAGNOLD DATA	
Total	2.50 g

present resuspension study being conducted at NTS, it appears that even smaller quantities of sand than those found at Livermore would be moved by saltation and creep. The above conclusion is based on the type of ground cover at the GMX site. At this site, there is an abundance of large pebbles on the desert pavement, as well as a profusion of desert plants. From personal observation and comparison to Chepil's (1945) results, it appears that GMX has a stable soil. It, therefore, was appropriate to develop a modified sampler more suitable for collection of small amounts of sand.

A modified saltation sampler was designed and built at Livermore. The sampler, shown in Fig. 3, is basically a Bagnold type of sampler. Modifications have been made which enhance the particle flow through the collector and make the particle retrieval easier. The modified sampler developed for use at NTS is 30.5-cm high, compared with a height of 83 cm for the Bagnold unit. The shorter height was chosen because it has been found that even in strong dust storms, over 90% of the sand moved by saltation is collected below 30 cm^2. In order to increase collection efficiency, the modified unit has vented collection passages for the sand. By allowing the air to flow through the unit, the pressure in the unit more nearly approximates the ambient air. Therefore, particle paths are less affected. The vents are shown in Fig. 3. Each vent is covered with two layers of 300-mesh screen in order that no particles greater than 10μm can escape through the unit. Upon entering the unit, the sand is collected in containers, one for each collection slot and one for the creep slot. Upon termination of each run, the containers are capped and numbered. This collection process is simple and convenient, even on strong wind days. And, there is a minimum chance of losing the sample. Five of these units have now been built and are deployed in the GMX area at NTS.

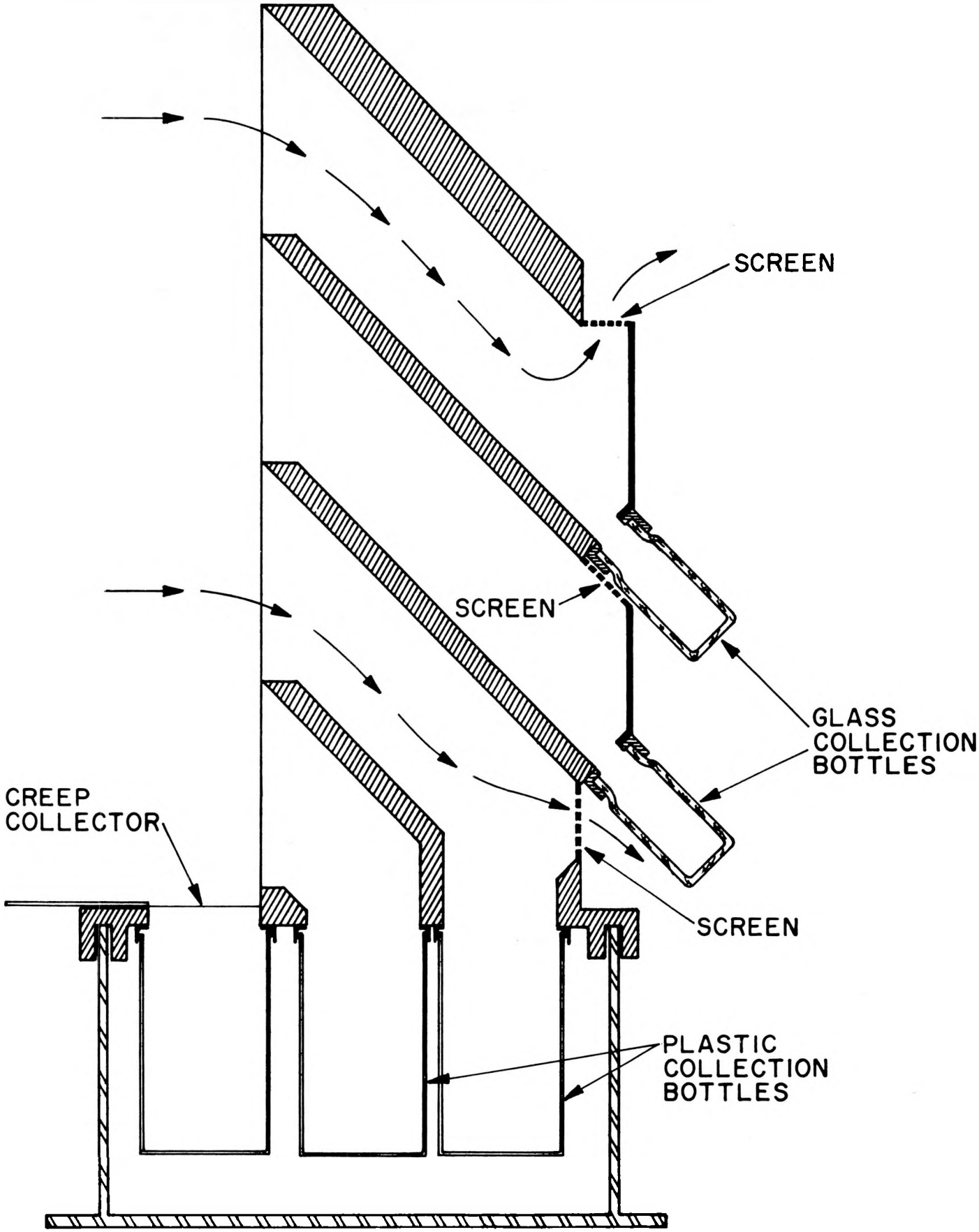

FIGURE 3. SCHEMATIC DIAGRAM OF THE LLL SALTATION AND CREEP SAMPLER.

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RESULTS AND DATA ANALYSIS: RESUSPENSION ELEMENT STATUS REPORT

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The long-range goal of the Nevada Applied Ecology Group (NAEG) resuspension studies is to produce a set of equations which can be used to predict the time-dependent average concentration of resuspended material downwind from a source of any geometrical configuration and soil surface characteristics. A great deal of work remains to be done before this goal can be accomplished. Results which are currently available are limited and/or have not been fully analyzed; they are presented in the nature of a status report and as representative of the types of measurements being made. (Preliminary results of the in situ optical size measurements are presented separately by Koval, this report.

These preliminary results have already found practical application in another AEC program which has the goal of evaluating the potential hazards associated with the reoccupation of the Eniwetok Atoll. The use of the Nevada Test Site (NTS) data and expertise to predict the levels of resuspended air activity arising from the aged Eniwetok source is treated in Appendix A of this report.

REECO AIR SAMPLING MEASUREMENTS

One of the short-range goals of the total NAEG program was to determine if resuspension created an occupational hazard at the fence boundaries of the GMX area. To accomplish this goal, Reynolds Electrical & Engineering Co., Inc. (REECo), collected and analyzed for ^{239}Pu* 503 air filter samples collected from February, 1971, to July, 1972 (Aoki, 1973); 200 control samples were also taken at Camp Mercury, NTS, at Building 214 during the same period. At GMX, two stations were maintained throughout this time period. Both were located on the fence perimeter, one to the northeast and one to the southwest of ground zero.

Two additional stations, located on the fence perimeter to the southeast and northwest of ground zero, were also operated from February through

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*Although results are frequently quoted here and elsewhere for ^{239}Pu, the usual methods of analysis cannot distinguish between ^{239}Pu and ^{240}Pu.

April, 1971. All samples were approximately 1 m above ground. The filter face was vertical and at each location was oriented toward ground zero. The sample collection period was generally 48 hr.

The measurement results are summarized in Table 1, and the cumulative distributions for the GMX NE, GMX SW, and Building 214 stations are plotted in Fig. 1. The data range over four orders of magnitude and are obviously not normally distributed. If the data were lognormally distributed, they should fall on a straight line when plotted as in Fig. 1. The measurements at GMX NE approach this condition, but data from the other two stations do not.

Unfortunately, most well-known statistical analysis procedures are not applicable to data that are not normally distributed, and it has been our experience that the use of such tests, when the data do not meet this criterion, frequently leads to erroneous conclusions. Therefore, these analyses have used only nonparametric, or distribution-free, statistical tests (Siegel, 1956).

The groups of samples which had detectable quantities of ^{239}Pu from the three major stations were tested to determine the statistical probability that the three groups of samples were drawn from the same population. The Kruskal-Wallis one-way analysis of variance was used and the result was $p = < 10^{-5}$. The same statistical test was also applied to the three combinations of sample groups taken two at a time. The results are given in Table 2 and indicate that the probability is negligible that any of the sample groups were drawn from the same population.

The conclusion is, therefore, drawn that the ^{239}Pu deposited at the GMX site more than 15 years prior to the measurements still represents a significant resuspension source. At the perimeter of the fence surrounding the exclusion area, however, the average air concentration of resuspended ^{239}Pu is only a small fraction of the presently accepted maximum permissible concentration for occupational exposure (Table 1).

The difference between the results for the GMX NE and GMX SW stations is of particular interest. A gross summary of the continuous meteorological data (Kennedy, 1973) taken during the same time period is shown in Fig. 2. The wind direction was most frequently toward the GMX SW station; however, the strong winds (> 10 mph) had a markedly different distribution with direction and most frequently were toward the GMX NE station. This gross correlation of air concentration with wind speed is predicted by theory (Bagnold, 1954; Healy and Fuquay, 1958) and has been reported for a single station in the vicinity of the ^{239}Pu contamination at the Rocky Flats Plant (Volchok, 1971; Sehmel and Orgill, 1973). It was not demonstrable, however, during LLL studies at NTS following the accidental venting of the Baneberry underground nuclear explosion (Anspaugh *et al.*, in press). More detailed studies using the wind velocity data and the individual measurements at the GMX NE station will be done to examine further the relationship between wind speed and air concentration of ^{239}Pu.

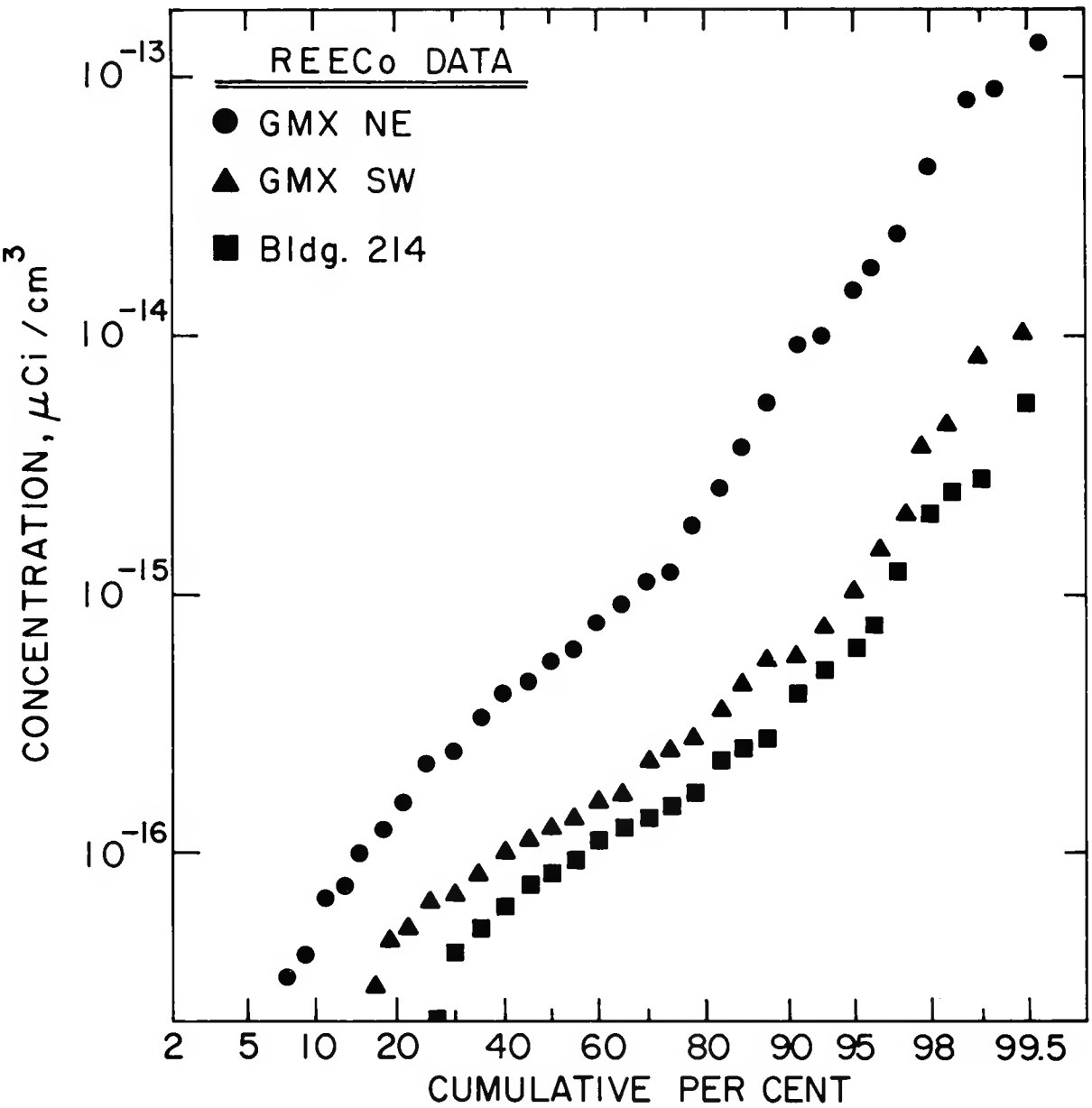

Fig. 1. Cumulative distributions of $^{239,240}Pu$ air concentration measured by REECo from February, 1971, to July, 1972, at 3 sites. Two sites were located at the northeast and southwest points of the perimeter force surrounding the GMX site in Area 5. The control location (Bldg. 214) is located in Mercury.

Table 1. Results of the REECo measurements for ^{239}Pu air concentration at the GMX area and the control location at Mercury (Bldg. 214). (Results are expressed in aCi/m^3; one aCi/m^3 equals 10^{-18} μCi/cm^3.)

Location	All Samples			Samples with detectable ^{239}Pu			
	N	Arith. Mean[a]	Median[a]	N	Arith. Mean	Geo. Mean	Median
GMX NE	254	6100	560	236	6600	790	620
GMX SE	31	870	200	23	1200	400	320
GMX SW	186	580	120	156	690	190	150
GMX NW	32	310	180	29	340	230	200
Bldg. 214	200	230	87	147	310	140	130

Occupational MPC[b] for 168-hr week (aCi/m^3):

Insoluble	10^7
Soluble	6×10^5

[a]Nondetectable results set equal to zero.

[b]ICRP Rept., 1960.

Table 2. Probability that the samples paired below were drawn from the same population. (The statistical test used was the nonparametric Kruskal-Wallis one-way analysis of variance.)

	GMX NE	GMC SW	Mercury (Bldg. 214)
GMX NE	1.0		
GMX SW	$< 10^{-19}$	1.0	
Mercury (Bldg. 214)	$< 10^{-27}$	0.009	1.0

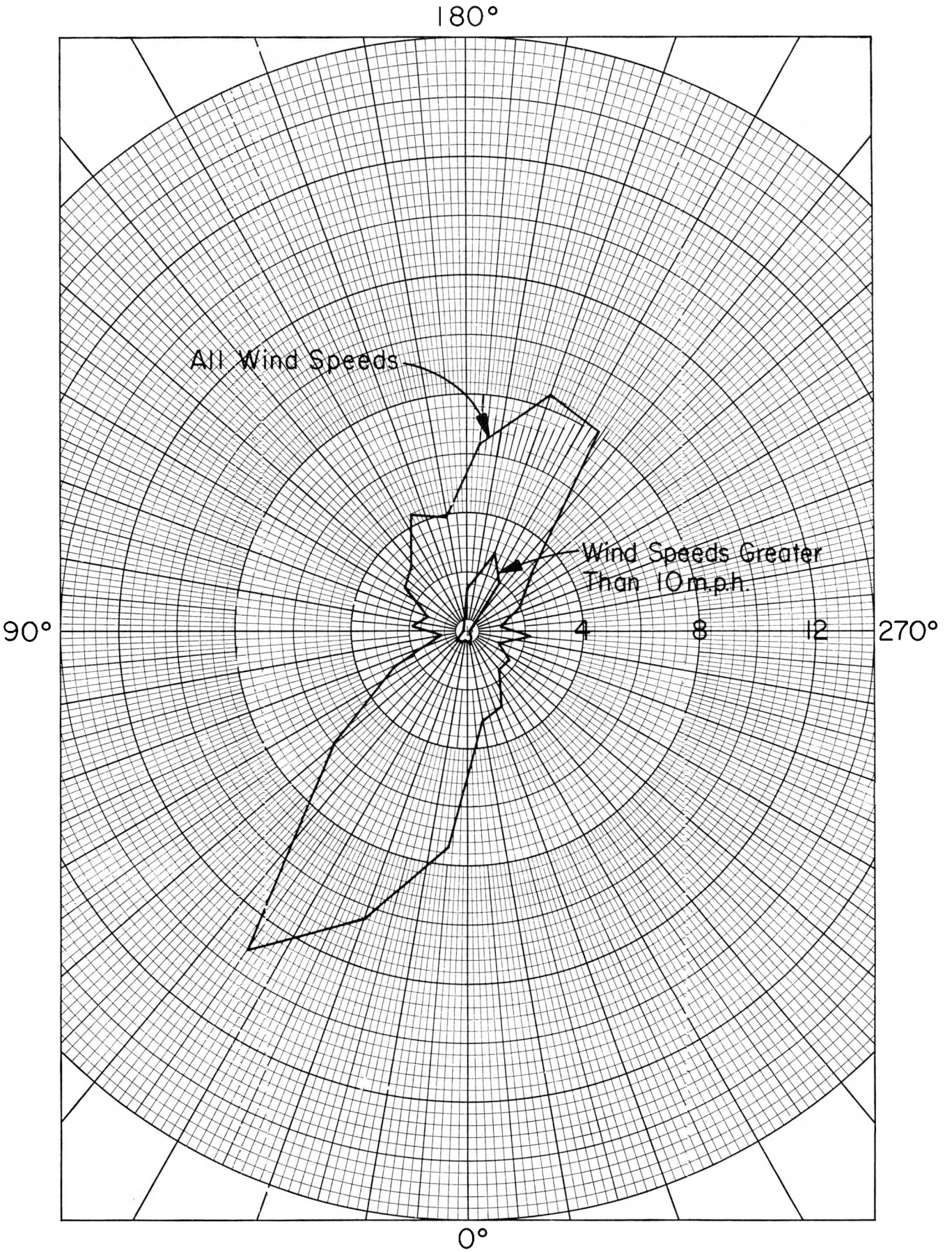

Fig. 2. Wind frequency distribution with direction at the GMX site from February, 1971, to July, 1972. This time period corresponds to the collection period of the REECo data shown in Fig. 1. The scale is percent occurrence within a 15° sector.

An additional question of considerable interest is whether the measurements at the control station (Building 214) are representative of worldwide background values, or are influenced by local NTS sources. Data for background surface air concentrations from several sources are tabulated in Table 3 and cover essentially the same time period as the Building 214 measurements. It is somewhat difficult to compare these measurements, as the sampling periods range from 48 hr to one month to one quarter, and the data spread for a single station appears to be inversely related to the sampling period. A statistical test which is sensitive to any difference in the distribution of samples is therefore undesirable. We can ask the more narrow question of whether the different groups differ in central tendency by applying the median test.

Four sets of data were considered: 52 measurements for nine stations of the EPA Radiation Alert Network, 96 measurements for six stations of the HASL Surface Air Sampling Program, 36 measurements for two sites of the LLL Environmental Monitoring Program, and 200 measurements for the REECo Building 214 station. For the latter set, all nondetectable values were set equal to zero. The median test was applied to the three sets of "background" data (EPA, HASL, LLL); the resultant probability that all three sets were drawn from the same population is > .90. For all four sets of data, the resultant probability is $< 10^{-4}$ that all four were drawn from the same population. Therefore, although there must be some reservations about the application of any statistical test to such disparate sets of data, the tentative conclusion is drawn that the air concentrations of ^{239}Pu in Mercury are influenced by the local NTS sources.

DISTRIBUTION OF MASS AND RADIOACTIVITY WITH PARTICLE SIZE

The distribution of mass and radioactivity as a function of particle size of the soil surface and materials moving in saltation and suspension is an important aspect of the resuspension research program. Such measurements are expected to help define the erodibility of different soil surfaces (Woodruff and Siddoway, 1965), to aid in the development of resuspension models (Slinn, 1973), and to provide input to models of lung dynamics (Morrow, 1966). Particle size distribution data for the GMX area soil are given elsewhere in this report by Tamura as part of the Soil Element; progress in developing a method to collect saltation particles for analysis is given by Reichman in this report as part of the Resuspension Element.

This section is concerned primarily with particle size distribution data for aerosols at the GMX site. Samples were collected by deploying five Andersen 2000* high-volume cascade impactors at the GMX in the

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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.

Table 3. Results of measurements of ^{239}Pu in surface air at other locations during January, 1971, to June, 1972. (With the exception of Rocky Flats, Colorado, these results should reflect the "background" levels of ^{239}Pu due to the injection of debris into the stratosphere by nuclear weapon tests.)

Location	^{239}Pu concentration, aCi/m^3			
	Minimum	Maximum	Arith. Mean	Geo. Mean
Radiation Alert Network, 48 Contiguous States (EPA)[a,b]				
Phoenix, Arizona	16	110	63	55
Denver, Colorado	17	125	65	56
New Orleans, Louisiana	19	75	40	36
Baltimore, Maryland	14	82	40	32
Buffalo, New York	16	104	48	37
Gastonia, North Carolina	20	120	59	51
Pierre, South Dakota	15	104	51	41
Austin, Texas	c	c	37	36
Seattle, Washington	10	131	41	29
All 9 Stations			50	40
Surface Air Sampling Program, Latitude 51° to 25°N (USAEC, HASL)[d,e]				
Moosonee, Ontario	c	c	54	44
New York, New York	14	135	51	40
Salt Lake City, Utah	33	315	98	76
Rocky Flats #1, Colorado	c	c	4600	4000
Sterling, Virginia	18	118	45	37
Miami, Florida	c	c	57	45
Bimini, Bahamas	c	c	76	62
6 Stations (Rocky Flats #1 excluded)			63	49
Lawrence Livermore Laboratory (LLL)[e,f]				
Main Laboratory	5	340	58	37
Site 300	14	110	52	44
Both Sites			55	40

[a]ORP, EPA, 1973.

[b]Quarterly composite samples.

[c]One or more results missing.

[d]Volchok *et al.*, 1973.

[e]Monthly composite samples.

[f]Gudiksen *et al.*, 1972; Gudiksen *et al.*, 1973.

location shown in Fig. 2 of the introduction to this report. The samplers were on an arc 250 ft from the GMX bunker with a spacing of 50 ft.

The Andersen high-volume cascade impactor has four stages plus an after-filter. At the design flow rate of 20 ft^3/min, the effective cutoff diameters of the four stages are 7.0, 3.3, 2.0, and 1.1 μm (Wood and Erickson, 1973). Each cascade impactor was mounted in a covered housing of the type recommended by the Environmental Protection Agency (1971) for the determination of suspended particulates in the atmosphere. Air intake into the housing was at a height of 1 m.

The first set of samples was collected by running the samplers continuously over the 36-day period from July 7, 1972, to August 12, 1972. This sampling period is longer than desirable, but was chosen to ensure that analyzable quantities of ^{239}Pu would be collected on each stage of the impactor. The pressure drop (corrected for altitude) across each impactor head was initially set to achieve a flow rate of 20 ft^3/min according to the manufacturer's calibration curve for each individual impactor unit. The pressure drop across each unit was periodically recorded during the run but was not readjusted.

The 25 samples were weighed at Livermore to determine the concentration of particulates. Other analyses were conducted which include the radiochemical determinations of ^{238}Pu, $^{239,240}Pu$, and ^{241}Am and the mass spectroscopic analysis of the separated plutonium from two of the after-filters.

The mass and radiochemical data for all stages of each sampler were combined in order to calculate the total air concentration for each species. These data are given in Table 4. The mean result for the concentration of $^{239,240}Pu$ is considerably higher than the REECo results, which is to be expected due to the proximity of the impactors to areas of greater ground deposition. A gross summary of the continuous wind data (Kennedy, 1973) taken during the high-volume cascade impactor run is shown in Fig. 3. The distribution of winds with direction is quite similar to that shown in Fig. 2, but the most frequent direction of strong winds was rotated about 10^{o} clockwise. This is consistent with the higher radionuclide concentrations indicated by samplers numbers 4 and 5.

The mass and radionuclide activities on each stage of one of the samplers are tabulated in Table 5. Data for the individual stages of each cascade impactor were used to determine the distributions of mass and activity as a function of particle size. This was accomplished by transforming the data with the dependent variable, the cumulative percent of the mass, or activity associated with particle sizes less than the effective cutoff diameter of the preceding stage. The logarithm of the effective cutoff diameter of the preceding stage was the independent variable. Because the data were to be fit to a lognormal distribution and 100% has no meaning in such a distribution, and because the effective cutoff diameter or largest size particle which enters the total sampling system is unknown, only four data points per sampler were available for

Table 4. Results of analyses for air concentrations summed over all particle sizes for the first high-volume cascade impactor samples. (Five impactors were run at the GMX site continuously for 36 days from July 7 to August 12, 1972.)

Sampler[a]	Mass $\mu g/m^3$	^{238}Pu $\mu Ci/cm^3$	$^{239,240}Pu$ $\mu Ci/cm^3$	^{241}Am $\mu Ci/cm^3$
1	69	1.9×10^{-16}	1.0×10^{-14}	1.0×10^{-15}
2	59	2.5×10^{-16}	1.2×10^{-14}	1.1×10^{-15}
3	71	3.8×10^{-16}	2.0×10^{-14}	2.4×10^{-15}
4	74	7.2×10^{-16}	3.9×10^{-14}	4.1×10^{-15}
5	81	9.5×10^{-16}	3.6×10^{-14}	3.5×10^{-15}
Mean	71	5.0×10^{-16}	2.3×10^{-14}	2.4×10^{-15}

[a]Samplers are numbered consecutively in a clockwise direction.

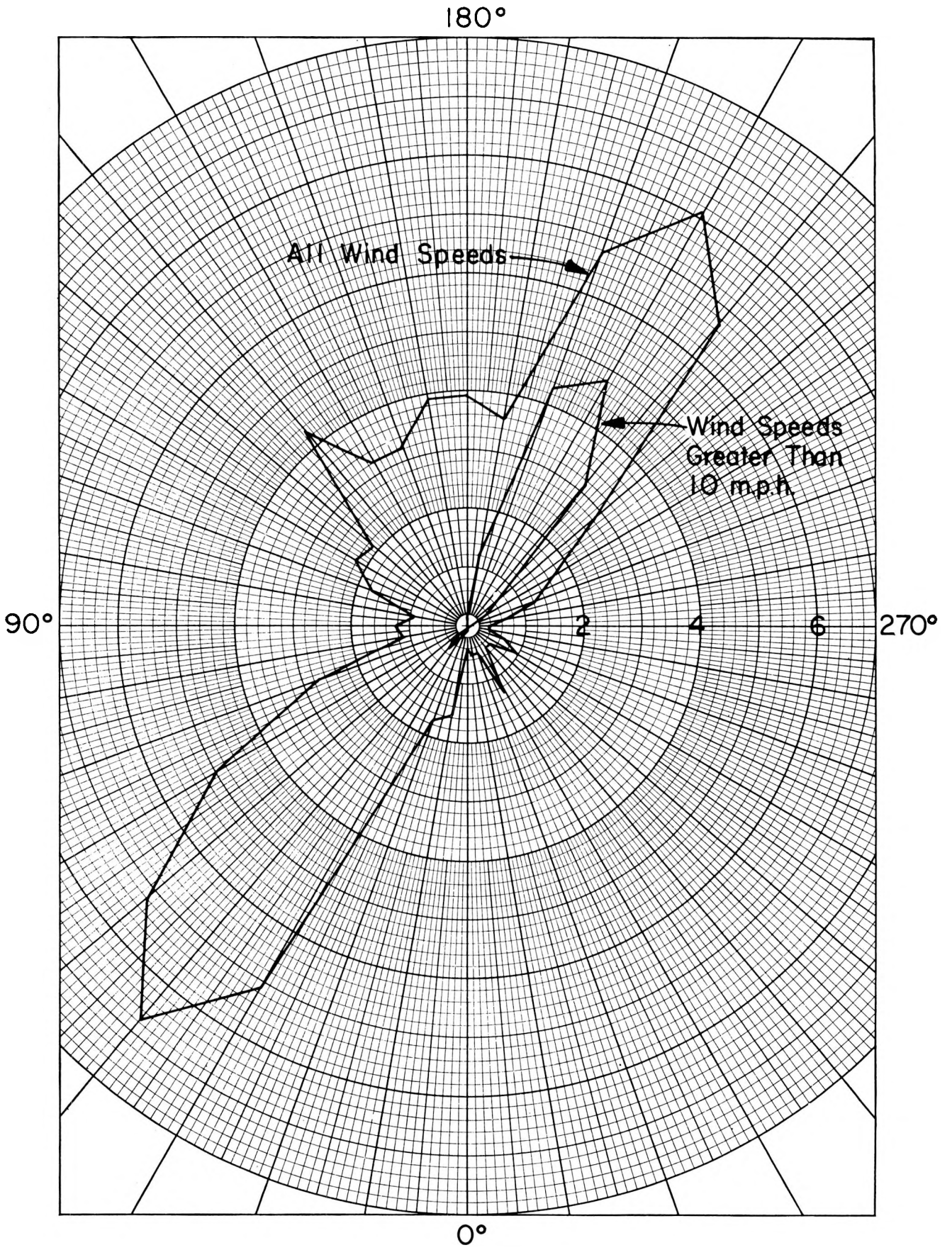

Fig. 3. Wind frequency distribution with direction at the GMX site from July 7, 1972, to August 12, 1972. This time period corresponds to the collection period of the first high-volume cascade impactor series. The scale is percent occurrence within a 10° sector.

Table 5. Mass and radionuclide activities on each stage of high-volume cascade impactor #4. (Values below are the totals accumulated during a continuous sampling from July 7 to August 12, 1972.)

Stage	Mass mg	^{238}Pu dpm	$^{239,240}Pu$ dpm	^{241}Am dpm
> 7μm	580	18. ± 2.	990 ± 70	100 ± 10
3.3 to 7.0μm	350	5.9 ± 0.5	310 ± 20	31 ± 4
2.0 to 3.3μm	140	5.6 ± 0.6	280 ± 20	31 ± 3
1.1 to 2.0μm	120	4.5 ± 0.4	230 ± 20	26 ± 3
Filter	990	13.0 ± 0.9	740 ± 40	77 ± 7
Total	2180	47 ± 2	2550 ± 90	260 ± 20

fitting. These four data points were fit to a lognormal distribution using Bevington's nonlinear least-squares fit routine, CURFIT (1969). The cumulative probability of a normal distribution needed for input to CURFIT was supplied by Bevington's AGAUSS routine modified to calculate the probability from $-\infty$ to x rather than from -x to x. Data were weighted by $1/\sigma^2$ of the gravimetric or radiochemical determination error. Two parameters are derived from this fitting process, the geometric mean particle size, $\bar{x}_g$ and the geometric standard deviation, σ_g. Sixty-eight percent of the total mass or activity is therefore between the particle sizes of $\bar{x}_g/\sigma_g$ and $\bar{x}_g\sigma_g$. If the data points are indeed well fit by a normal distribution, then $\bar{x}_g$ is also equal to the median diameter. Because the cascade impactor separates particles by inertial impaction and is calibrated for unit density spheres (Wood and Erickson, 1973), $\bar{x}_g$ is also equal to the mass median aerodynamic diameter (MMAD) or activity median aerodynamic diameter (AMAD). These are the standard parameters needed for input to models of lung dynamics (Morrow, 1966).

The results of the particle size distribution fits are presented in Table 6. The data were generally well fit by a lognormal distribution as measured by the reduced chi-square of the fit. Sampler 5, however, was an exception. In this case, the data for radionuclides were not well fit by a lognormal distribution because of a large amount of activity present on the fourth stage (1.1 to 2.0 μm). The difference between the five impactor units was most pronounced for the values of σ_g. Some of this difference is proably due to fluctuations in the flow rate which were recorded during the run; fluctuations were most pronounced for sampler 1.

In all cases, the distribution with particle size was so broad, as indicated by the large values of σ_g, that the fit for any individual impactor was very insensitive to σ_g, indicated by the high standard deviation of the determination of σ_g. With the goal of obtaining a better fit, the data from all five impactors were combined in two ways: 1) the total mass of activity on each stage for all five impactors was used to derive a new cumulative distribution and 2) the cumulative percent distributions for the five impactors were averaged. The results of fitting a lognormal distribution to these data are also shown in Table 6; these fits were more sensitive to σ_g.

The fits to the combined data, and generally the fits for any individual impactor, indicate that there is no difference in the distribution of activity with particle size for the three species ^{238}Pu, $^{239,240}Pu$, and ^{241}Am. The total mass, however, is distributed distinctly differently with a smaller median aerodynamic diameter and a larger geometric sigma. The mean results for mass and $^{239,240}Pu$ are plotted in Fig. 4.

The values of σ_g derived from these data are larger than those usually reported for aerosols of uranium and transuranium compounds (Mercer, 1964; Anderson and Nelson, 1967; Ettinger *et al.*, 1972; Vaane *et al.*, 1971) and are also larger than those explicitly considered in the ICRP Task Group of Lung Dynamics report (Morrow, 1966). There are

Table 6. Results of analysis of the distribution of airborne mass and radioactivity with particle size at the GMX area. (Data were obtained by running 5 Andersen 2000 high-volume cascade impactors continuously from July 7 to August 12, 1972. The ± values are the standard deviations of the fit.)

Sampler	Total Mass		^{239}Pu		$^{239,240}Pu$		^{241}Am	
	MMAD	σ_g	AMAD	σ_g	AMAD	σ_g	AMAD	σ_g
1	1.5 ± 0.3μm	30 ± 40	4. ± 1.μm	30 ± 30	3.5 ± 0.6μm	30 ± 20	4. ± 1.μm	30 ± 30
2	1.8 ± 0.4	20 ± 20	2.2 ± 0.3	8 ± 4	2.3 ± 0.3	9 ± 4	2.8 ± 0.4	9 ± 5
3	1.5 ± 0.2	6 ± 2	2.7 ± 0.3	6 ± 2	2.4 ± 0.2	7 ± 2	3.6 ± 0.3	4.1 ± 0.7
4	1.6 ± 0.3	20 ± 20	3.7 ± 0.4	8 ± 2	3.7 ± 0.3	9 ± 2	3.5 ± 0.5	8 ± 3
5	1.7 ± 0.3	20 ± 20	3.2[a]	3[a]	3.3[a]	5[a]	2.9[a]	3[a]
Total[b]	1.6 ± 0.1	15 ± 5	3.1 ± 0.2	5.4 ± 0.7	3.1 ± 0.1	7.4 ± 0.9	3.2 ± 0.2	5.4 ± 0.6
Mean[c]	1.6 ± 0.1	15 ± 5	3.0 ± 0.2	7 ± 1	3.0 ± 0.1	8 ± 1	3.2 ± 0.2	6.4 ± 0.9

[a]The reduced chi square of the fit indicates that the data are not well fit by a lognormal distribution.

[b]The materials on each stage were summed for the 5 impactors, and the cumulative percent distribution calculated.

[c]The cumulative percent distributions were averaged for the 5 impactors.

Table 7. The specific activity of $^{239,240}Pu$, dpm/g, on the stages of the 5 high-volume cascade impactors. (The samplers were run continuously from July 7 to August 12, 1972.)

	SAMPLER					
Stage	1	2	3	4	5	Mean
> 7μm	480	420	940	1700	1200	950
3.3 to 7μm	210	500	710	880	1200	700
2.0 to 3.3μm	420	1300	570	2000	880	1000
1.1 to 2.0μm	540	300	430	1900	3300	1300
After Filter	250	410	510	750	500	480
Mean	380	590	630	1400	1400	890

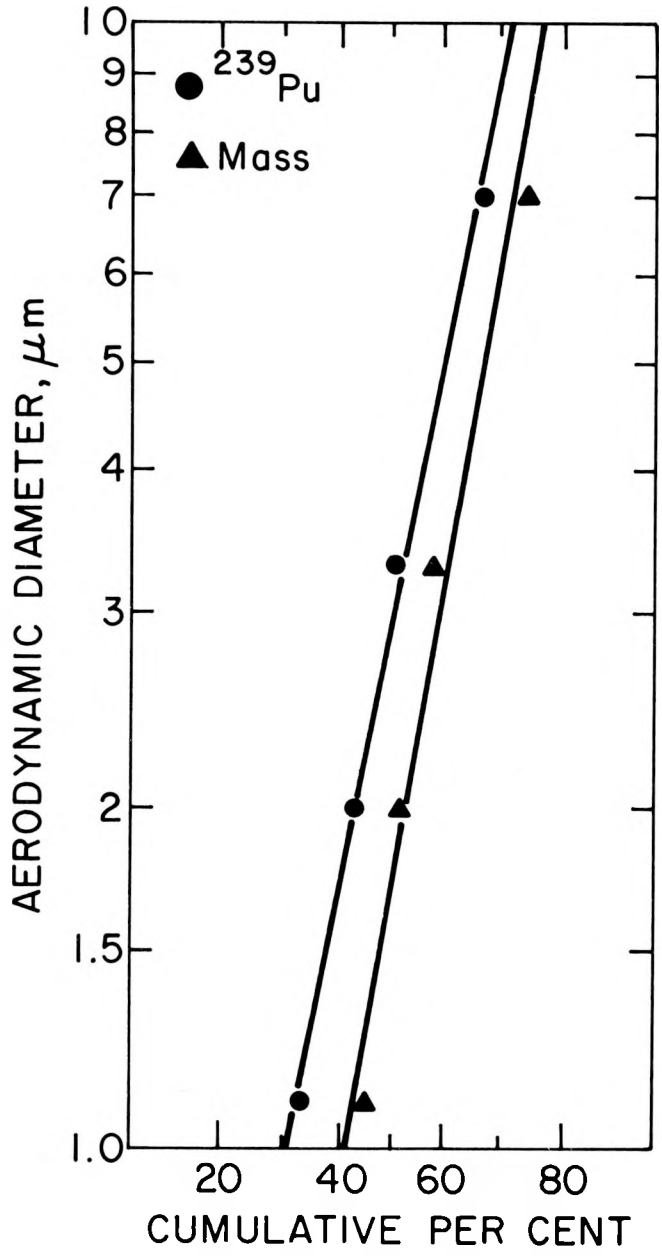

Fig. 4. The average cumulative distributions of mass and $^{239,240}Pu$ as a function of particle size as determined from the first high-volume cascade impactor run at the GMX site. The lines represent least-squares fits to lognormal distributions. Values of the derived parameters are given in Table 6.

several possible explanations for this difference. First, one must maintain some reservations about the performance of the Andersen 2000 cascade impactor itself. This is a relatively new product and has received only limited field evaluation (Wood and Erickson, 1973; Burton *et al.*, 1973; Sehmel, in press); Sehmel (in press) has indicated that there may be significant wall losses and reentrainment problems. Laboratory and field studies are presently being conducted to evaluate further these effects and their influence upon results derived from this impactor.

There are, however, very few data which are directly comparable to those reported in Table 6 for plutonium and americium. Nearly all results reported in the literature pertain to laboratory-generated aerosols or "field" aerosols. The word field is somewhat of a misnomer, however, as it has been used to describe aerosols in laboratories where plutonium is processed. A few results are available for more nearly equivalent situations from measurements made at NTS. Cascade impactors were used to study resuspended plutonium aerosols during Project 57 (Wilson *et al.*, 1960). A median diameter of about 1.5 μm was reported. No specific data were given for σ_g, but it was stated that the data were strongly biased because many samples with a σ_g of 10 or higher were discarded. Results of particulate studies were also reported for the initial cloud of plutonium aerosol produced by the nonnuclear Double Tracks and Clean Slate 1 events of the Roller Coaster Operation (Friend and Thomas, 1965). Much of the plutonium in these initial, explosion-produced clouds was associated with particles larger than could be sampled by cascade impactors, but it was stated that in the region where sizing by impactors is possible, a value for σ_g of 6 was obtained.

Even higher values of σ_g are given in Table 6 for mass of suspended particulates. Considerable data for comparison are available from studies conducted for the National Air Surveillance Networks (NASN) (Lee and Goranson, 1972; Lee *et al.*, 1972), and by Lundgren (1971). The results of these studies are that values of σ_g as large as 15 are not unusual in either polluted or nonpolluted atmospheres, even over time periods of a few hours. Distributions other than lognormal have been proposed for natural aerosols (Junge, 1963; Pasceri and Friedlander, 1965). The well-known Junge distribution (Junge, 1963), for example, predicts an essentially flat volume distribution as a function of particle size between 0.1 and 10 μm. Such a distribution is therefore compatible with large values of σ_g derived from a fit to a lognormal mass distribution.

The most immediate application of the results in Table 6 is the determination of the fraction of the resuspended plutonium aerosol at GMX, which would be expected to undergo pulmonary deposition. This fraction is approximately 0.2, based upon the ICRP Task Group of Lung Dynamics model (Morrow, 1966).

The possible effect of wall losses and reentrainment in the cascade impactor used would be to overestimate the relative amount of material in the smaller size ranges separated by the impactor (Sehmel, in press).

The possible bias in the respirable fraction is therefore conservative; the correct value may be lower, but it is very unlikely to be higher. Results have also been reported for the respirable fraction of the resuspended plutonium aerosol in the vicinity of the Rocky Flats Plant (Volchok and Knuth, 1972). In these studies, either a horizontal elutriator or a cyclone was used to remove all but the respirable fraction which was then collected on an after-filter. Results for the respirable fraction were 0.25 ± 0.09.

A question of considerable interest is whether the resuspended plutonium aerosols show a marked difference in specific activity as a function of particle size. This can be examined in an approximate way by calculating the specific activity (activity of $^{239,240}Pu$ per gram of total particulates collected) for each stage of the impactor. Such data for all five impactors are given in Table 7. There is no obvious correlation of specific activity with particle size. Considerable caution must be exercised in interpreting the data, however, because the resuspended aerosols are diluted by natural, inert aerosols transported into the area and which undoubtedly have a different distribution of mass with particle size. The average specific activity, 890 dpm/g, is about one-third of that found in the soil in close proximity to the cascade impactors. The specific activity concept has been expanded further in Appendix A of this report to derive an interim, approximate resuspension model applicable to aged plutonium depositions.

The $^{239,240}Pu$ specific activities in this environment are very low, relative to that of pure $^{239}PuO_2$. The specific activity of $^{239}PuO_2$ is 1.2×10^{11} dpm/g. The total activity present on all five stages of sampler 4 (Table 5) could have been contained in a single $^{239}PuO_2$ particle of 15 μm actual diameter, or 4 10 μm particles, or 30 5 μm particles, or 3500 1 μm particles. The aerodynamic diameter of such particles would be approximately 3.4 times larger.

One of the 25 results tabulated in Table 7 is anomalously high. This is for the 1.1 to 2.0 μm stage of sampler 5. The relative excess of $^{239,240}Pu$ activity on this stage is about 250 dpm. In order for a single, pure $^{239}PuO_2$ particle to produce this effect, it would require an actual diameter of 7 μm, which is equivalent to an aerodynamic diameter of about 24 μm. Theoretically, such a large particle should not be present on this stage, but may have been reentrained from a previous stage (Sehmel, in press).

Mass spectroscopy analyses were also performed on the plutonium separated from two of the cascade impactor backup filters. The results were given in Table 8. The weighted mean of the mass ratio of ^{240}Pu to ^{239}Pu is 0.0545. The equivalent activity ratio is 0.202. A more useful way of expressing the latter information is that 17 percent of the total $^{239,240}Pu$ activity at GMX, which is frequently referred to as only ^{239}Pu, is actually due to ^{240}Pu. The mass ratios in Table 8 plus the ^{238}Pu and $^{239,240}Pu$ activity measurements can be used to deduce the complete isotopic composition of the smaller airborne particles at the GMX site. These calculations were made and the results given in Table 9, where they are also compared to the typical plutonium

Table 8. Mass ratios of the plutonium separated from 2 of the cascade impactor backup filters. (The impactors were run continuously from July 7 to August 12, 1972.)

Sampler	Plutonium Mass Ratios		
	$^{240}/_{239}$	$^{241}/_{239}$	$^{242}/_{239}$
2	0.0547 ± 0.5%	0.00189 ± 2.5%	0.000161 ± 7.5%
3	0.0520 ± 2%	0.00195 ± 8%	0.000479 ± 33%
Weighted Mean[a]	0.0545	0.00190	0.000163

[a]Weighted by $1/\sigma^2$

Table 9. Isotopic composition of the plutonium separated from 2 of the cascade impactor backup filters. (The impactors were run continuously from July 7 to August 12, 1972. Results are calculated for the composition at time of collection.)

Isotope	Percent by Weight		
	Sampler 2	Sampler 3	Typical Plutonium[a]
^{238}Pu	0.0085 ± 0.0009	0.0073 ± 0.0006	0.04 ± 0.01
^{239}Pu	95.0 ± 8.	95.0 ± 6.	93.34 ± 0.5
^{240}Pu	5.2 ± 0.4	4.9 ± 0.3	6.0 ± 0.5
^{241}Pu[b]	0.18 ± 0.02	0.18 ± 0.02	0.58
^{242}Pu	0.015 ± 0.002	0.05 ± 0.02	0.04

[a]Del Pizzo *et al.*, 1970.

[b]Abundance is significantly influenced by radioactive decay during the time since deposition at the GMX site.

values published by Del Pizzo, Owen, and Putzier (1970). The calculated values for GMX plutonium agree quite well with the typical plutonium values, except for a deficiency of ^{238}Pu. The apparent deficiency of ^{241}Pu is partly due to radioactive decay of the aged GMX plutonium.

The absolute mass of the ^{241}Pu on the backup filters can be used to calculate the ^{241}Pu activity. This can be combined with the separately determined activity of ^{241}Am, which is the daughter product of ^{241}Pu, to calculate the age of the plutonium. It is in fact known, of course, how long the plutonium has been present at GMX, but such a calculation serves a useful purpose. It provides a check on the various types of analyses performed; and, if it is assumed that the analyses have no systematic errors, it provides a sensitive way of testing for the differential behavior of plutonium and americium in a natural environment.

The relevant equation is:

$$\frac{A_1}{A_2} = \frac{\lambda_2}{\lambda_2} \frac{\lambda_2 - \lambda_1}{1 - e^{-(\lambda_2 - \lambda_1)T}}$$

where A is activity, λ is the radiological decay constant, T is the age of the plutonium since purification, and the subscripts $_1$ and $_2$ denote the parent and daughter radionuclides, respectively. Such a calculation is highly dependent upon the half-lives of ^{241}Pu and ^{241}Am; published values of both have changed substantially in recent years. For ^{241}Am, the value used was 433 ± 2 years[36], and for ^{241}Pu 15.16 ± 0.19 years (Cabell and Wilkins, 1971). The calculated activity ratios and ages are given in Table 10. The actual weighted average time since deposition of the GMX plutonium is 16.7 years. The result for sampler 3 is statistically different from this at the 98% confidence level. As both calculated values are shorter than the true time since deposition, a possible interpretation is that americium has left that fraction of the upper soil layer which is resuspendible at a faster rate than has plutonium, or is preferentially associated with larger particles. The relative deficiency of americium is about 20%.

Considerable attention has been devoted elsewhere in this volume to the determination of the ratio of $^{239,240}Pu$ activity to ^{241}Am activity in soil samples, and the correlation between the two measurements. The 25 data points for the cascade impactor samples were also examined in a manner similar to that used for soil samples. The correlation between the two measurements was calculated using the nonparametric Spearman rank method. The resulting value of r is 0.94 ($p = < 10^{-5}$). The data are shown in Fig. 5; the line is the least-squares fit to the data. The average value of the ratio is 10 ± 2, which is statistically indistinguishable from that reported for the GMX soil samples. The $^{239,240}Pu$ to ^{241}Am ratios for the cascade impactor samples were tested to determine if there is a significant difference in the ratio for the five particle-size classes.

Table 10. Results of the calculated activity ratio of ^{241}Pu to ^{241}Am and the calculated age of the plutonium present on 2 of the cascade impactor backup filters.

Sampler	$\frac{^{241}Pu\ Activity}{^{241}Am\ Activity}$	Age, years
2	30 ± 4	14.6 ± 1.4
3	35 ± 5	13.2 ± 1.5

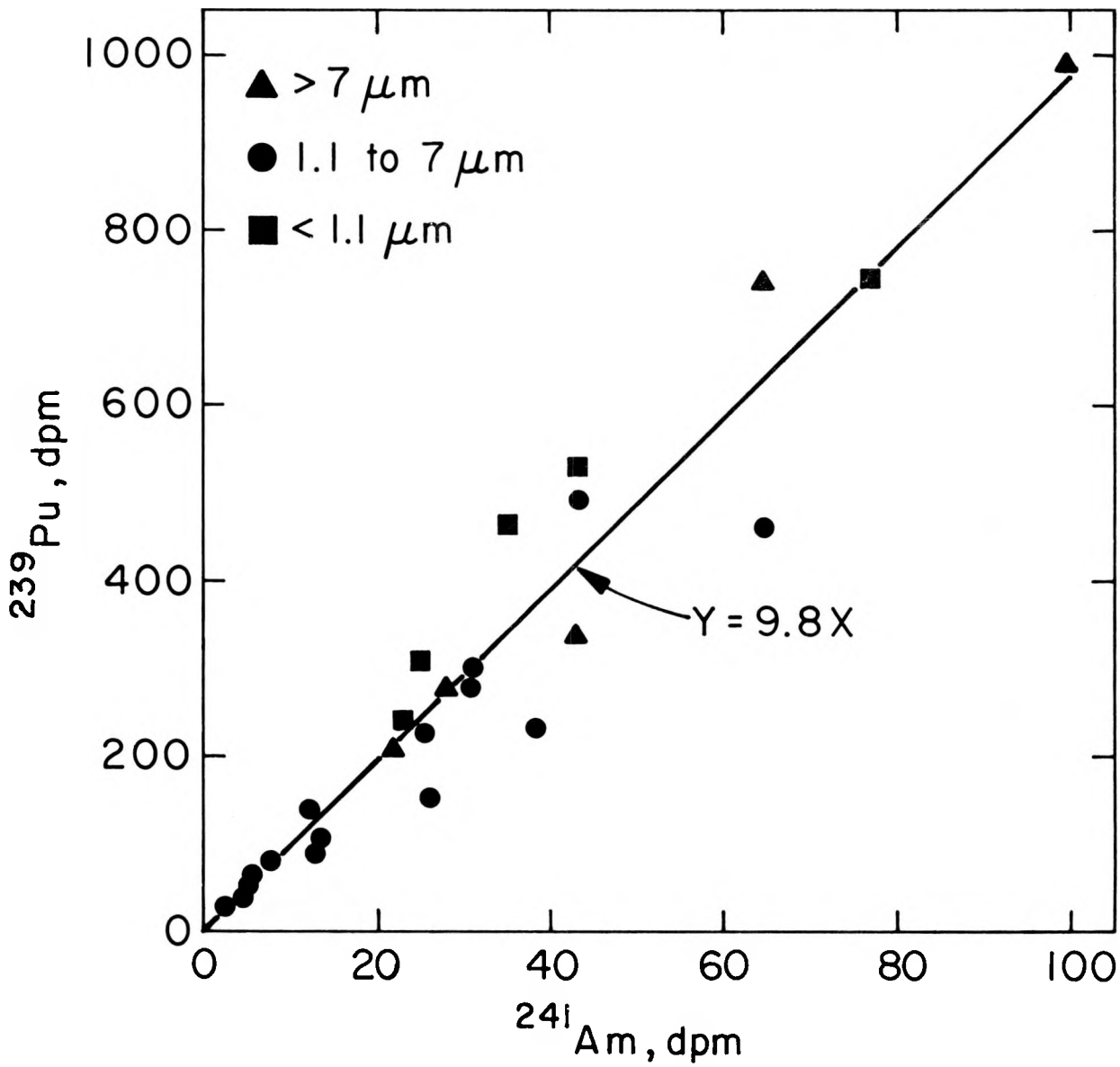

Fig. 5. The relationship between the total $^{239,240}Pu$ and ^{241}Am on individual stages of the 5 high-volume cascade impactor samplers. Data are for the first run at GMX on July 7 to August 12, 1972. The line represents a least squares fit of the data.

The data were considered as five groups of related samples and tested by the nonparametric Friedman two-way analysis of variance (Siegel, 1956). The result is that the null hypothesis cannot be rejected at the 0.05 level of significance.

A second set of samples was collected using the same five cascade impactors at the same positions. They were run continuously from April 24 to May 23, 1973, a period of 29 days. Complete results are not yet available for the activities of plutonium and americium; the gravimetric results are given in Table 11. The mass concentration during this sampling period was nearly a factor of 2 lower than the previous one, and the mass median aerodynamic diameter (MMAD) was also lower. Both observations are consistent with less material being resuspended from the soil surface; we anticipate that the concentration of resuspended plutonium was also lower. The values of σ_g derived from fits to a lognormal distribution are also a factor of 2 lower and imply a narrower distribution of mass with particle size. The mean data for the two runs are plotted in Fig. 6.

Additional cascade impactor samples are presently being collected. The sample collection periods have been shortened due to favorable experience in collecting more than adequate plutonium for analysis on every stage. One cascade impactor has been modified according to Sehmel's design (in press); this incorporates a rotating cowl which always faces the inlet into the wind. Results with this design will be compared with those using the EPA-recommended housing.

ULTRAHIGH-VOLUME AIR SAMPLING RESULTS

The primary method of studying the resuspension of plutonium is the utilization of ultrahigh-volume air samplers. The development of this sampler is described by Goluba, this report. This sampler was developed in order to collect an analyzable sample of airborne plutonium within a few hours. This is highly desirable because, in order to relate resuspended airborne plutonium to meteorological parameters, it is almost essential to collect samples during time periods which are shorter than significant changes in meteorological regimes.

Ten ultrahigh-volume air samplers have been installed in the GMX area. Two samplers were located at each of the five positions indicated in Fig. 2 of Phelps and Anspaugh, Introduction, this report. All five positions are on a single radial which was selected because it corresponds to the most frequent direction of high-speed winds. One location is upwind, one is at the approximate center of the highest concentration of plutonium ground deposition, and the other three are downwind. An additional control sampler is located on the outskirts of Mercury.

Table 11. Concentration of total suspended particulates and distribution with particle size for the second cascade impactor run at GMX. (The 5 impactors were run continuously from April 24 to May 23, 1973. Radiochemical results are imcomplete for this set of samples.)

Sampler	Concentration $\mu g/m^3$	MMAD μm	σ_g
1	38	1.21 ± 0.05	7.3 ± 0.6
2	37	1.13 ± 0.01	6.8 ± 0.1
3	47	1.28[a]	7.0[a]
4	47	1.06 ± 0.05	7.9 ± 0.8
5	37	1.17 ± 0.03	6.3 ± 0.4
Total[b]	41	1.17 ± 0.05	7.0 ± 0.6
Mean[c]	41	1.17 ± 0.05	7.0 ± 0.6

[a]The reduced chi square of the fit indicates that the data are not well fit by a lognormal distribution.

[b]The mass on each stage was summed for the 5 impactors, and the cumulative percent distribution calculated.

[c]The cumulative percent distributions were averaged for the 5 impactors.

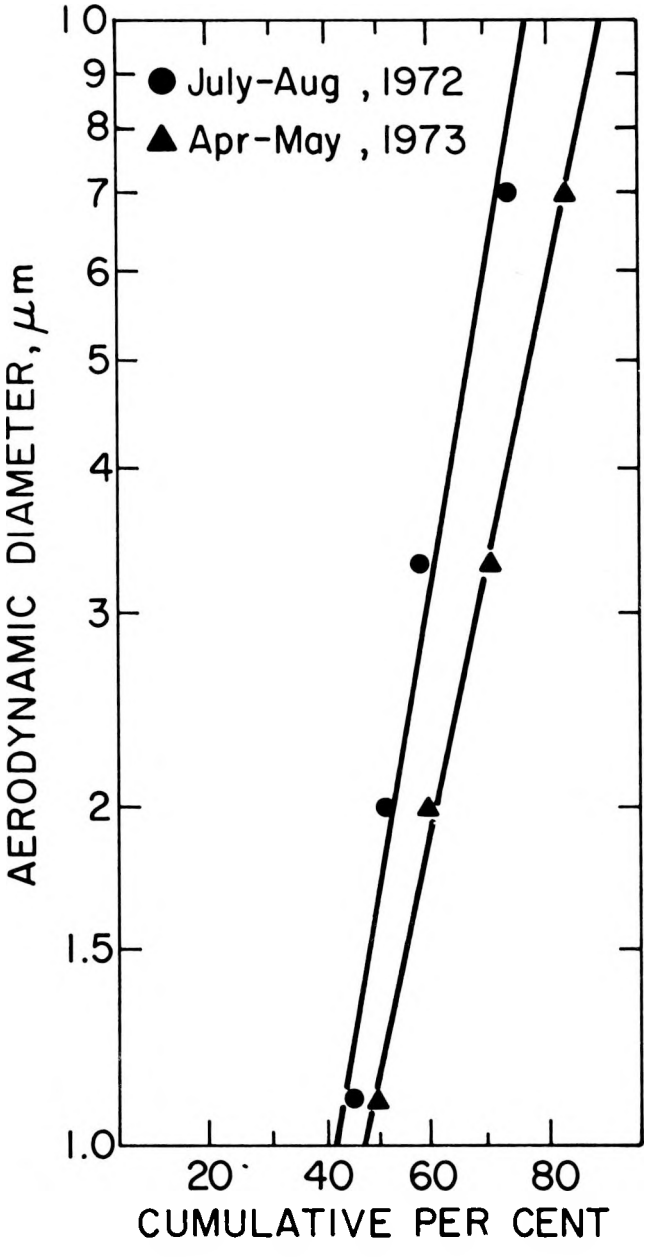

Fig. 6. The average cumulative distributions with mass as a function of particle size as determined from 2 high-volume cascade impactor runs at the GMX site. The lines represent least-squares fits to lognormal distributions. Values of the derived parameters are given in Tables 6 and 11.

The filter face of all samplers is parallel to the ground surface, and is at a height of 1.3 m. Duplicate samplers were initially installed in order to examine the reproducibility of results obtained over short time periods; they are about 5 m apart, perpendicular to the radial. If reproducibility proves to be satisfactory, the duplicate samplers will be deployed at other radials and/or heights to obtain additional information about concentration gradients.

The long-range goal in making such measurements is the derivation of a source term in terms of the fractional amount of activity injected into the air per unit area per unit time, as a function of meteorological variables. This also requires the use of a suitable meteorological transport model which can be used to derive such a source term, given the measured air concentrations and the geometrical configuration of the source, plus the relevant meteorological parameters. Measurements of mass loading alone, which can be made over periods as short as 1 sec, should greatly aid in deriving the relationships between the injected source term and meteorological parameters. It is, however, essential to demonstrate that any such relationships are consistent with measurements of resuspended plutonium concentration as well.

Several sample series were collected during the past year. Because of the large number of samples involved, and the higher priority given by the AEC community to the analysis of samples generated by the Eniwetok survey, complete results for only two series were available for inclusion in this report.

Results for the two available series are shown in Figs. 7 and 8, where the average of the duplicate samples is superimposed upon the sample locations and the ground deposition isopleths. Data for the individual samplers are tabulated in Table 12. The average wind speed at 8 m during the first run was 7.6 m sec^{-1}; during the second, it was 6 m sec^{-1}. Other basic and derived meteorological parameters for these runs are given in Fig. 1, Kennedy and Booth, this report. There is a gross correlation between many of the wind speed-related parameters and the concentration of resuspended plutonium. This is encouraging, but more data are obviously needed before a meaningful analysis can be attempted.

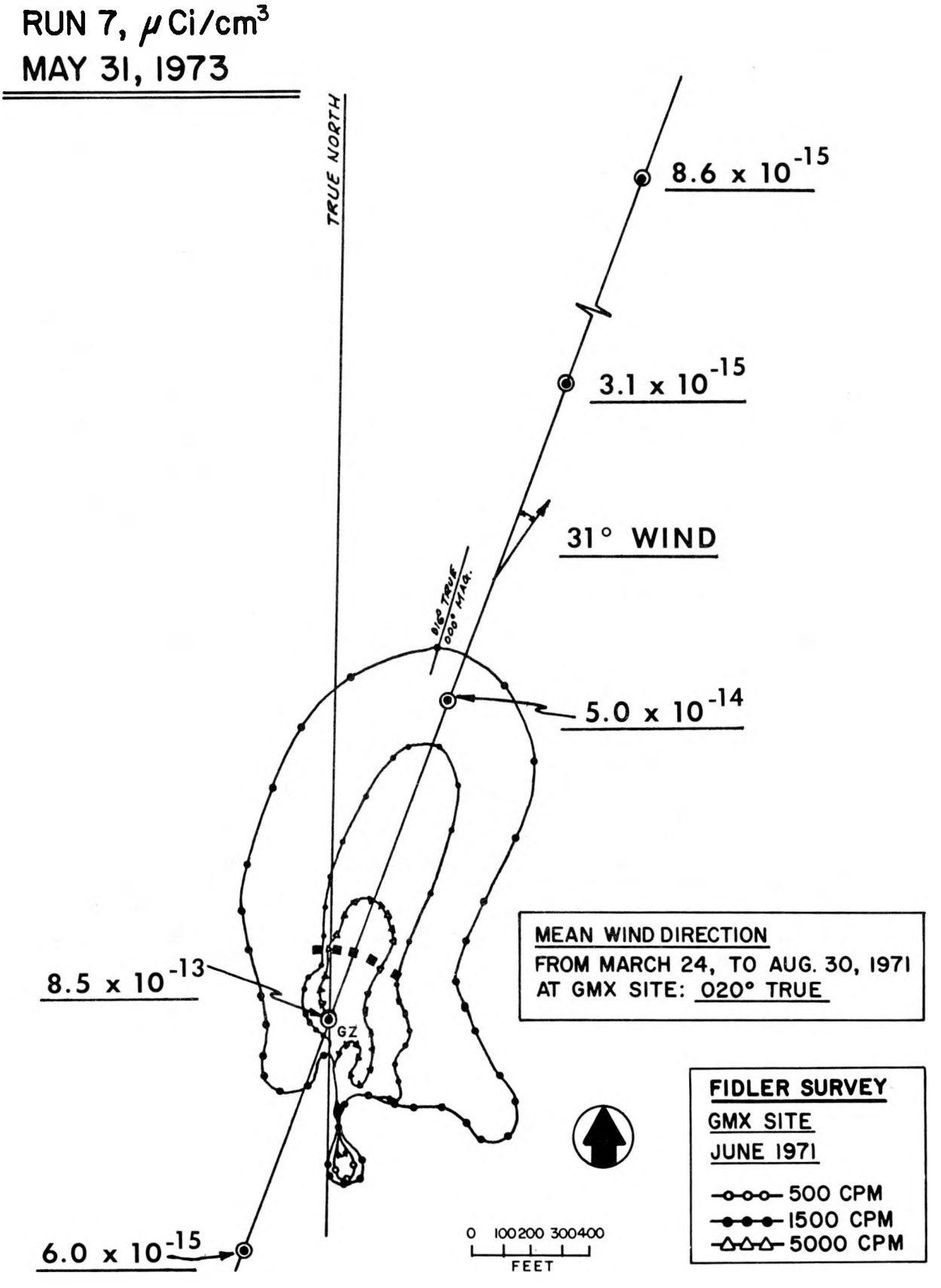

Fig. 7. $^{239,240}Pu$ air concentration at the GMX site derived from a set of ultrahigh-volume filter samples. The samplers were run from 1150 to 1400 PST on May 31, 1973. The numbers are averages of duplicate samples.

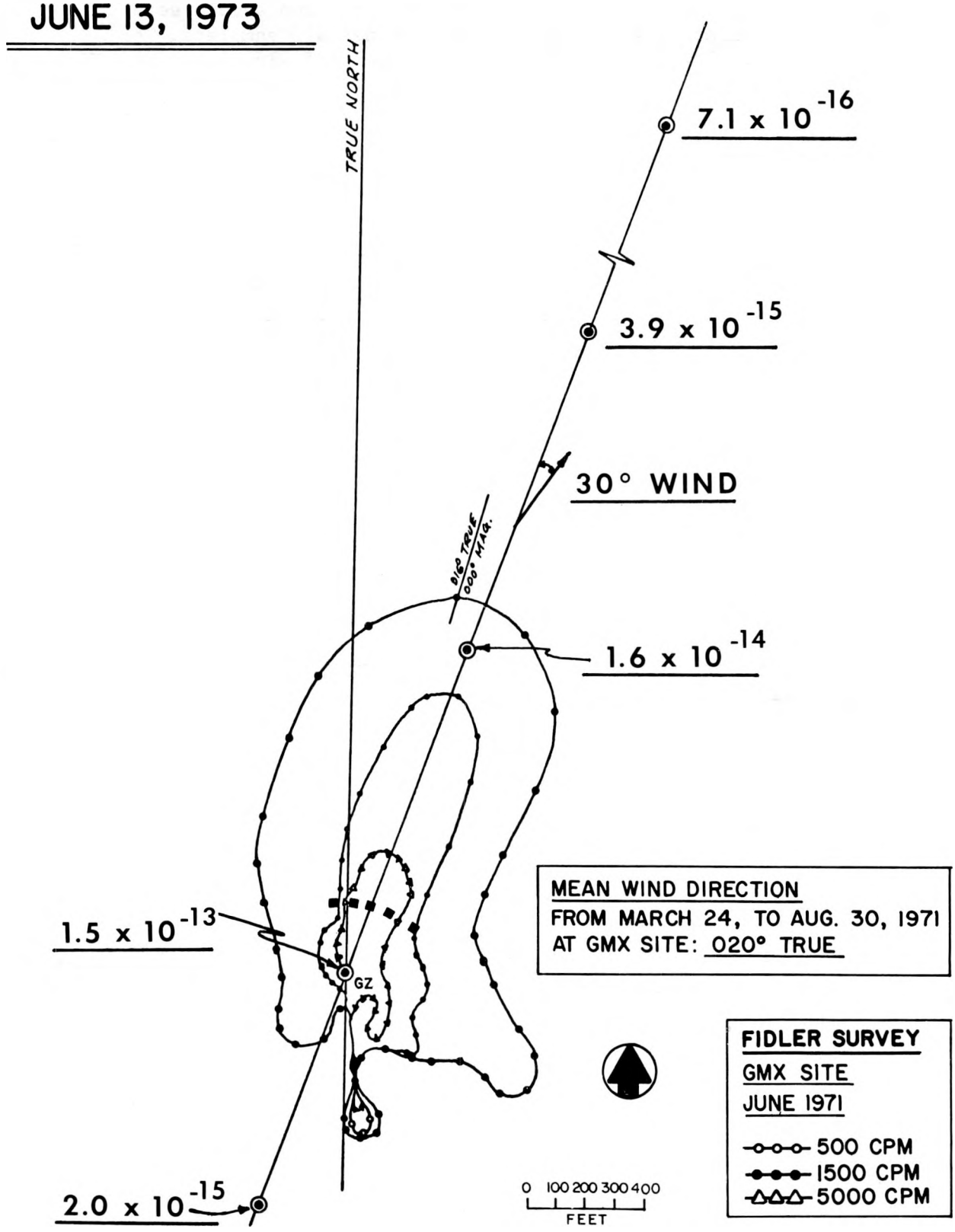

Fig. 8. $^{239,240}Pu$ air concentration at the GMX site derived from a set of ultrahigh-volume filter samples. The samplers were run from 1220 to 1830 PST on June 13, 1973. The numbers are averages of duplicate samples.

Table 12. $^{239,240}Pu$ concentration derived from two sample sets collected with the ultrahigh-volume air samplers. The samplers were run between the hours of 1150 and 1400 PST on May 31, 1973, and 1200 and 1830 PST on June 13, 1973. Some of the meteorological data recorded during these periods is given in Fig. 1, Kennedy and Booth, this report.

Location	$^{239,240}Pu$ Air Concentration, $\mu Ci/cm^3$			
	May 31, 1973		June 13, 1973	
	West	East	West	East
800 ft, Upwind	4.1×10^{-15}	8.0×10^{-15}	1.8×10^{-15}	2.2×10^{-15}
Ground Zero	5.6×10^{-13}	1.1×10^{-12}	8.2×10^{-14}	2.2×10^{-13}
1200 ft, Downwind	4.7×10^{-14}	5.3×10^{-14}	9.7×10^{-15}	2.3×10^{-14}
2400 ft, Downwind	3.1×10^{-15}	3.1×10^{-15}	5.1×10^{-15}	2.6×10^{-15}
4800 ft, Downwind	1.4×10^{-14}	3.1×10^{-15}	7.1×10^{-16}	[a]
Control	1.3×10^{-15}		[a]	

[a]Analysis not yet available.

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APPENDIX A, RESUSPENSION ELEMENT STATUS REPORT: THE USE OF NTS DATA AND EXPERIENCE TO PREDICT AIR CONCENTRATIONS OF PLUTONIUM DUE TO RESUSPENSION ON THE ENEWETAK ATOLL

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There are no general models that may be used with confidence to predict the resuspended air activity in the vicinity of an area contaminated with Pu. However, two approximate methods may be used--the resuspension factor approach and an argument based upon ambient air particulate concentrations with the assumption that the particulates are derived from the contaminated surface. The former method has been frequently used, but almost always in the context of a fresh surface deposit. The latter method is inappropriate to the fresh deposit situation, but should be reasonably valid after enough time has elapsed for the surface-deposited material to become fairly well mixed with a few centimeters of the soil surface.

RESUSPENSION FACTOR APPROACH

The resuspension factor, K, is defined as

$$K = \frac{\text{Air Concentration } (Ci/m^3)}{\text{Surface Deposition } (Ci/m^2)}$$

and thus has units of m^{-1}. It is almost always implied that both measurements are made at the same location. The difficulties with this approach are fairly obvious--no allowance is made for the geometrical configuration of the source, the particle size distributions of the contaminant and the soil surface, vegetation cover, etc. Stewart (1964) and Mishima (1964) have tabulated values of K from many experiments, including those involving laboratory floors as well as native soils. As would be expected, the tabulated values cover an enormous range and vary from 10^{-2} to 10^{-13}/m. Most of the high values, however, are derived from experiments with laboratory floor surfaces and/or with artificial disturbance.

For outdoor situations, Stewart (1964) suggests as a guide for planning purposes that a value for K of 10^{-6}/m be used "under quiescent conditions, or after administrative control has been established in the case of an accident." A value of 10^{-5}/m is suggested under conditions of moderate activity. Stewart states, however, that exceptionally higher values (mean of 10^{-5}/m) were observed during the Hurricane Trial (Monte Bello Islands) and credited this to the nature of the small islands exposed to sea breezes. Values approaching 10^{-3}/m when dust is raised by pedestrians and vehicles are also reported by Stewart.

Kathren (1968) has also considered the resuspension factor approach and has recommended the use of 10^{-4}/m as a conservative, but appropriate, value for setting standards for PuO_2 surface contamination.

Langham (1969, 1971) has suggested that a value of 10^{-6}/m is a reasonable average value to use in estimating the potential hazard of occupancy of a plutonium-contaminated area. At the same time, however, Langham notes that many measured values lie in the range of 10^{-5} to 10^{-7}/m, and reports that his own measurements in 1956 produced a value of 7×10^{-5}/m.

These recommended values, however, are all intended for application during the time period immediately following deposition. Numerous studies (Stewart, 1964; Langham, 1971; Shreve, 1958; Wilson *et al.*, 1960; Anspaugh *et al.*, in press) have shown that air concentrations of resuspended materials decrease with time. With the assumption that this decrease can be represented by a single exponential function, half-times of 35 to 70 days have been reported (Langham, 1971; Wilson *et al.*, 1960; Anspaugh *et al.*, in press). This decrease in air activity is not explainable by the relatively minor loss of material from the initial site of deposition (Stewart, 1964; Shreve, 1958), but is presumably caused by the migration of the initial surface-deposited material into the soil.

Attempts to use the resuspension factor approach to derive acceptable levels of soil surface contamination have included this "attenuation factor" as a simple exponential function with half-times of 35 (Langham, 1969) or 45 (Kathren, 1968) days. There are major uncertainties in such a formulation, however. The longest study of this "decrease with time" extended to only eleven months following the initial deposition (Anspaugh *et al.*, in press), which is extremely short compared to the half-life of a radionuclide such as ^{239}Pu. There are also published reports which indicate on experimental and theoretical bases that the decrease with time will not be adequately represented by a single exponential function, but that the rate of decrease itself will also decrease with time (Stewart, 1964; Shreve, 1958). Fortunately, the exact nature of this time dependence is not critical in determining the integrated exposure from the time of initial deposition, due to the fairly well-documented rapid decrease at early times. However, it is obviously the controlling factor for questions concerning the reoccupation of areas many years after the contaminating event.

As an illustration, the most conservative published model (Kathren, 1968) may be used to calculate a resuspension rate for material 15 years after deposition:

$$K = \frac{10^{-4}}{m} \exp\left[\frac{-\ 0.693 \times 15\ y \times 365\ d}{45\ d \qquad\qquad y}\right]$$

$$\simeq 10^{-41}/m$$

If, however, the resuspension rate asymptotically approached some finite value 10^{-6} of the original, then the resuspension rate 15 years later would obviously be $10^{-10}/m$; but the total integrated air activity (from t = 0 to ∞) for ^{239}Pu would be changed only by

$$A\ 10^{-4} \int_0^\infty \exp\ (-\ 0.693\ t/45\ d)\ dt + A\ 10^{-10} \int_0^\infty \exp\ (-\ 0.693t/24{,}400\ y)\ dt$$

$$= 6.5\ A \times 10^{-3} = 1.3\ A \times 10^{-3}$$

which is an increase of 20% and, more importantly, cannot be accumulated during an individual's life span.

Because the functional nature of the decrease in resuspension rate with time cannot be confidently extrapolated, previously published models should not be applied to the reoccupation of areas many years after the contaminating event.

The resuspension factor approach can be applied in an approximate way, however, if resuspension factors are used which were derived from measurements over aged sources. Perhaps the most relevant data are unpublished results from current resuspension experiments at the GMX site in Area 5 of the Nevada Test Site (NTS). The ^{239}Pu at this location was deposited following 22 high-explosive detonations during the time period from December, 1954, to February, 1956. Measurements of resuspended air activity levels at this site during 1971-1973 appear to be the only available data concerning resuspension of ^{239}Pu from a source of this age.

Data from two types of measurements are available and can be used to derive average resuspension factors. The first type of measurement (Anspaugh and Phelps, unpublished data) was accomplished by placing five high-volume cascade impactors (Wood and Erickson, 1973) within the most highly contaminated area, and running them for 36 days, from July 7 to August 12, 1972. The collected $^{239,240}Pu$ activity was distributed lognormally with particle size with an activity median aerodynamic diameter (AMAD) of 3.0 μm and a geometric standard deviation of 8.2. The $^{239,240}Pu$ concentration varied from 1.0 to 3.9 x 10^{-14} $\mu Ci/cm^3$ with an average of 2.3 x 10^{-14} $\mu Ci/cm^3$ for the five samplers. At the present time, only limited data are available regarding the soil activity in the area. Four soil samples of depth 0-3 cm from approximately the same location have been analyzed with results

(Eberhardt and Gilbert, 1972) of 2060 to 3550 dpm/g with a mean of 2700 dpm/g. Profile data from other locations at the same general site indicate that about 90% of the total deposition is contained within the top 5 cm of the soil (Gilbert and Eberhardt, this report). Measurements of soil density in the area average 1.8 g/cm^3 (Anspaugh and Phelps, unpublished data). The resuspension factor is therefore

$$\frac{2.3 \times 10^{-14}\ \mu Ci}{cm^3} \times \frac{g}{2700\ dpm} \times \frac{cm^3}{1.8\ g} \times \frac{0.9}{3\ cm} \times \frac{10^2\ cm}{m} \times \frac{2.22 \times 10^6\ dpm}{\mu Ci}$$

$$= 3 \times 10^{-10}/m$$

Additional air samples were taken by the Reynolds Electrical and Engineering Co. (REECo) on the edge of the contaminated area during the period of February, 1971, to July, 1972, with a sampling time of approximately 48 hr (Aoki, 1973). Measurements were made at four locations, but the most pertinent is the one which was most frequently in the direction of strong winds from the strongly contaminated area and where the highest air activities were recorded. Here, 254 individual air filter samples were collected and detectable results reported for 236. ^{239,240}Pu concentrations ranged from 3.5 x 10^{-17} to 6.3 x 10^{-13} μCi/cm^3, with arithmetic and geometric means of 6.6 x 10^{-15} and 7.9 x 10^{-16} μCi/cm^3, respectively. Results for four soil samples taken from approximately the same location range from 128 to 202 dpm/g with a mean of 160 dpm/g (Eberhardt and Gilbert, 1972). Because the arithmetic mean is a better representation of average lung exposure, it is used to derive a resuspension factor at this site:

$$\frac{6.6 \times 10^{-15}\ \mu Ci}{cm^3} \times \frac{g}{160\ dpm} \times \frac{cm^3}{1.8\ g} \times \frac{0.9}{3\ cm} \times \frac{10^2\ cm}{m} \times \frac{2.22 \times 10^6\ dpm}{\mu Ci}$$

$$= 2 \times 10^{-9}/m$$

This value is nearly an order of magnitude higher than the one previously calculated, and reflects some of the inherent difficulties in the resuspension factor approach, i.e., that no allowance is made for the geometrical configuration of the source and that higher ground activities may be present upwind.

It is obvious that this approach is subject to major uncertainties, but does serve as an order of magnitude indication of the resuspended air activities that may arise from a ^{239,240}Pu-contaminated area which has weathered for 15 to 20 years. The data discussed above also demonstrate unequivocally that resuspension of ^{239,240}Pu does in fact occur from such aged deposits, and at levels many orders of magnitude higher than would be expected if the often noted decrease with time were represented by a single exponential function with a half-time of 35 to 70 days.

MASS LOADING APPROACH

The other approximate prediction method is based upon measured or assumed levels of particulate matter in ambient air with the assumption that this material is derived from the contaminated soil. For fresh deposits, this approach is not valid because the freshly deposited debris is much more likely to be resuspended than the remainder of the weathered soil surface. After many years of weathering since the initial deposition, however, the contaminating material should be reasonably well mixed with a centimeter or two of soil such that the contaminant activity per gram of airborne particulate should approximate that in the upper soil. A major difficulty could arise, however, if for example $^{239,240}Pu$ were preferentially associated with the smaller particle sizes more likely to become airborne. For the NTS, such is not the case as determined by soil analyses (Tamura, 1973, and this report) and by the high-volume cascade impactor study. The latter study found an AMAD of 3.0 μm for $^{239,240}Pu$ whereas the total mass median aerodynamic diameter was 1.7 μm. The specific activity of the material collected on each stage can also be examined for a preferential association of Pu with particle size. Average data from all five samplers are:

Size (μm)	$^{239,240}Pu$ (dpm/g)
> 7	950
3.3 to 7	700
2.0 to 3.3	1030
1.1 to 2.0	1300
.01 to 1.1	480
All Stages	890
(Soil)	(2700)

Although there is considerable spread in these data, there is no indication of a preferential association of $^{239,240}Pu$ with a particular particle size; and as would be expected due to dilution by inert aerosol, the specific activity is lower than that of the soil.

If we assume that this is generally true, a general and conservative method of predicting resuspended air concentrations of contaminants would be to simply multiply the ambient air mass loading by the contaminant concentration in soil. A factor of some uncertainty for a specific calculation is what value to use for the ambient air mass loading in the absence of specific data. This becomes even more uncertain by the possibility that the people involved may be highly correlated with the source in the sense that children playing in sand, adults cultivating crops, etc., may generate their own "ambient air" which contains much more mass than would be recorded by a stationary sampler.

The lower and upper bounds of ambient air mass loading can be fixed rather easily for any site. There has been considerable interest in establishing a "background level" of mass loading, and this is generally believed to be on the order of 10 $\mu g/m^3$ (Porch *et al.*, 1970). The upper bound can be established in a reasonable way by the levels found in mine atmospheres which have led to a considerable prevalence of pneumoconiosis in the affected workers (Walton, Ed., 1970). Examination of these data indicates that current standards for occupational dust exposure ($\sim$ 1 -- 10 mg/m^3) have a very small or, perhaps, no margin of safety such that a reasonable upper bound can be taken as 1 mg/m^3. (British data (Jacobsen *et al.*, 1970) indicate that if the general public were exposed to dust levels in excess of 1 mg/m^3, the public health problem from the dust alone might be enormous. The presumption is therefore made that if $^{239,240}Pu$ were also in the dust at levels sufficient to achieve the maximum permissible concentration, the additional insult would be insignificant.) The reasonableness of the upper limit value of 1 mg/m^3 is also demonstrated by data which indicate that nonurban ambient air mass concentrations this high are usually associated with conditions described as dust storms (Spirtas and Levin, 1970; Hagen and Woodruff, 1973).

Measurements of ambient air mass loading can be used to further define a reasonable estimate for predictive purposes. The National Air Surveillance Network (NASN) has reported such results for several years. Data for 1966 (NAPCA, 1968) show that there were 217 urban and 30 nonurban stations reporting. The annual arithmetic average for the urban stations ranged from 33 (St. Petersburg, Florida) to 254 $\mu g/m^3$ (Steubenville, Ohio), with a mean arithmetic average for all 217 stations of 102 $\mu g/m^3$. For the nonurban stations, the range was from 9 (White Pine County, Nevada) to 79 $\mu g/m^3$ (Curry County, Oregon) with a mean arithmetic average for all 30 stations of 38 $\mu g/m^3$. No data in this report are available for nonurban locations on small islands similar to the Enewetak group; perhaps the closest analogue is the urban station at Honolulu, Hawaii, which had an annual arithmetic average of 35 $\mu g/m^3$.

More pertinent, but limited, data have recently been published for the island of Hawaii (Pueschel *et al.*, 1973; Simpson, 1972). Data are given for three locations: Mauna Loa Observatory located at a height of 3400 m, Cape Kumukahi, and the City of Hilo. NASN data for Hilo (for an unspecified period) are given as 18 $\mu g/m^3$, and nephelometer measurements varied from 18 $\mu g/m^3$ during the day to 26 $\mu g/m^3$ at night. At Cape Kumukahi, the nephelometer measurement was 9.2 $\mu g/m^3$. The greatest amount of data is available for Mauna Loa Observatory. Here, the NASN measurement was 3 $\mu g/m^3$, and the nephelometer measurements varied from 1.7 $\mu g/m^3$ at night to 6.5 $\mu g/m^3$ during the day. Additional measurements made by the USAEC Health and Safety Laboratory (HASL) were 3 $\mu g/m^3$. It is of interest in the present context that Simpson (1972) made the following comment concerning the HASL measurements: "The HASL filter samples contain substantial dust (3 -- 5 $\mu g/m^3$ of air sampled) because of the fact that the filter was located less than one meter above the ground surface near areas with substantial

personnel activity at the observatory site." Thus, while this method of measurement may not have coincided with Simpson's interest, it does indicate that ambient air mass loadings may be very low on such remote islands even when considerable human activity is occurring nearby.

On the basis of the above data, it would appear reasonable to use a value of 100 $\mu g/m^3$ as an average ambient air mass loading for predictive purposes. Indications are that this value should be quite conservative for the Enewetak Islands, and therefore allows room for the uncertainty involved because the people themselves may generate a significant fraction of the total aerosol. They may therefore be exposed to higher particulate concentrations than would be measured by a stationary sampler.

Supporting evidence that 100 $\mu g/m^3$ is a reasonable value to use for predictive purposes is provided by the National Ambient Air Quality Standards (EPA, 1971). Here, ambient air is defined as ". . . that portion of the atmosphere, external to buildings, to which the general public has access." The primary ambient air standards define "levels which . . . are necessary, with an adequate margin of safety, to protect the public health." The secondary standards define "levels which . . . (are) necessary to protect the public welfare from any known or anticipated adverse effects of a pollutant." These standards for particulate matter are given below:

National Ambient Air Quality Standards
for Particulate Matter
(in $\mu g/m^3$)

	Annual Geometric Mean	Max. 24-hr Concentration not to be exceeded more than once/yr
Primary	75	260
Secondary	60	150

Data to support these standards in terms of health effects, visibility restrictions, etc., have been provided (Middleton, 1969).

An arithmetic mean would be more desirable for predictive purposes. Data from 1966 (NAPCA, 1968) for nonurban locations indicate that the annual arithmetic mean is (on the average) 120% of the annual geometric mean.

REPRESENTATIVE CALCULATIONS

Because one of the primary objects is to derive an acceptable soil level for the Enewetak Islands, the approaches developed above were used to derive such levels for both soluble and insoluble ^{239}Pu. The derived values are given in Table 1. The two methods agree within a

factor of 2, at least for soil distributions like those found at NTS. (The ambient air mass loading at NTS during the cascade impactor run was measured to be 70 $\mu g/m^3$.)

Such derived values must, of course, be used with a great deal of discretion. They are based on simple model systems which are believed to be generally conservative, but individual situations can be imagined which could exceed the predictions.

OTHER CONSIDERATIONS

The above calculations relate only to the resuspended air activity in ambient air, and do not consider the additional problems of resuspension of material from contaminated clothing or the resuspension of material which has been transferred to homes.

Healy (1971) has considered these and other problems, and has provided tables of "decision levels" for surface contamination levels and home transfer levels. A decision level is based upon National Council on Radiation Protection and Measurements (NCRP) recommended dose limitations. Because the derivations are rather tenuous, Healy has used the phrase decision level and states that its use is to serve as a signal that further careful investigation is warranted.

Healy's decision levels for soluble ^{239}Pu are given in column 1 of Table 2. The values in column 2 are derived from these and an acceptable soil concentration of 1 nCi/g from Table 1 to give equivalent dirt (soil) contamination and transfer levels. The results are interpreted as indicating that the potential exists for greater dose contributions from these infrequently considered pathways than from the usually considered pathway of resuspension as calculated for ambient air. (This conclusion would be the same for insoluble ^{239}Pu.) Therefore, if dose calculations based on the usual resuspension pathway should appear limiting compared to other pathways such as food-chain transfer, these pathways considered by Healy need to be carefully evaluated for the specific Enewetak situation.

Table 1

Acceptable soil levels of ^{239}Pu for a source which has weathered for several years. (Values are approximate and are subject to uncertainty. The reference standard used in the derivations is 10% of the Maximum Permissible Concentration in Air for 168 hr occupational exposure $(MPC_a)^{25}$.)

	Insoluble	Soluble
Acceptable Air Concentration	10^{-12} μCi/cm^3	6×10^{-14} μCi/cm^3
Resuspension Factor Approach		
Assumed Resuspension Factor	10^{-9}/m	10^{-9}/m
Acceptable Soil Deposition[a]	10^3 μCi/m^2	60 μCi/m^2
Acceptable Soil Concentration[b]	20 nCi/g	1 nCi/g
Mass Loading Approach		
Assumed Mass Loading	10^2 μg/m^3	10^2 μg/m^3
Acceptable Soil Concentration	10 nCi/g	0.6 nCi/g

[a]Equivalent to approximately 10^4 μg of insoluble ^{239}Pu/m^2.

[b]Assumes same distribution of ^{239}Pu with depth and soil density as measured at NTS.

Table 2

Decision levels for soluble ^{239}Pu, and their equivalent in soil mass based upon the "acceptable soil concentration" from Table 1.

Pathway	Decision Level	Mass Equivalent
A. Direct Personal Contamination		
Direct Inhalation[a]	2×10^{-5} nCi/cm^2	1×10^{-5} g/cm^2
Direct Ingestion[b]	0.2 nCi/cm^2	0.2 g/cm^2
Skin Absorption[c]	8×10^{-4} μCi	0.8 g
B. Transfer (to Homes) Levels		
Resuspension[d]	0.01 μCi/day	10 g/day
Direct Inhalation	0.01 μCi/day	10 g/day
Direct Ingestion	100 μCi/day	10^5 g/day
Skin Absorption	0.03 μCi/day	30 g/day

[a] "The contamination level on clothing and skin that could result in inhalation of air at the MPC_a for the public." (Healy, 1971)

[b] "The contamination level on skin or clothing that could result in ingestion of a quantity of radioactive material equivalent to the ingestion of water at the MPC_w for an individual in the public." (Healy, 1971)

[c] "The total quantity of radioactive material maintained on the skin for 24 h/day that could result in absorption of a quantity equal to that which would be absorbed from the GI tract if water at the MPC_w for "soluble" isotopes for an individual in the public were ingested." (Healy, 1971)

[d] "The amount transferred per day that could result in air concentrations due to resuspension in a medium-sized home averaging at the MPC_a for an individual in the public." (Healy, 1971)

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Statistical analysis and data processing

RECOMMENDATIONS CONCERNING THE COMPUTATION AND REPORTING OF COUNTING STATISTICS FOR THE NEVADA APPLIED ECOLOGY GROUP*

by

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ABSTRACT

Recommendations based on work by Nicholson (1963) are made concerning the computation and reporting of count statistics for Group H8 at the Los Alamos Scientific Laboratory (LASL) relative to their analysis of environmental samples for plutonium, uranium, and other radionuclides for the Nevada Applied Ecology Group (NAEG). Computational formulas are given for (1) determining whether contamination above background level exists in the sample, (2) computing confidence intervals about the true net counting or disintegration rate of the sample, (3) determining the minimum true net disintegration rate that will be "detected" most of the time, and (4) reporting of net counts and their associated counting errors. Recommendations for answering these questions are made for four counting situations: (a) a sample (gross) count and a single background count, (b) a sample count with multiple background counts, (c) "base line" correction for background in gamma spectrometry, and (d) alpha spectrometry where a tracer is added to the sample. In addition, a "rule of thumb" (from Nicholson) is suggested for choosing between alternative methods of deciding whether contamination above background exists in the sample. An "exact" test is preferred when background counts are very low and/or the ratio of sample to background counting times is greater than 1. The concepts of "decision limits" and "detection limits" are discussed and mathematical derivations of the various computational formulas are given in an appendix. Tables are included for computing tests and confidence limits. Approximate counting error formulas for alpha spectrometry are given for various levels of generality relative to efficiencies of counting instruments and to equality of counting times for the sample, background, and tracer.

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CONTENTS

1. INTRODUCTION

In the course of statistical consultations with scientists engaged in radio-ecological research for the Nevada Applied Ecology Group (NAEG)[1], certain questions have arisen concerning the appropriate methods of computing and reporting "counting errors" associated with net counts of radionuclides in soil, vegetation, and other environmental samples. As an initial response to this need this paper has been prepared which discusses the reporting of count data, the computation of "decision limits" to decide whether "something" is in the sample other than background, and the calculation of the "detection limit". Consideration is first given to the simplist case where a single background and sample count are available. Attention is then directed toward the more complicated situations of (a) multiple independent background readings, (b) "base line" correction for background in gamma spectrometry, and (c) alpha spectrometry where a tracer is added to the sample to be counted.

This report is based primarily on the work of Nicholson (1963 and 1966), Currie (1968), and Hartwell (1972). Currie (1968) points out the present confusion surrounding the "limit of detection". He defines 3 types of limits ("decision limit", "detection limit", and "determination limit") and shows how they are used in the specification of lower limits for a measurement process. Hartwell (1972) discusses and endorses Currie's work and illustrates the computation of "decision limits" and "detection limits". These two papers are directed at the radiochemist while the paper by Nicholson (1963) discusses many of the same topics in a more classical statistical terminology. Nicholson (1963) studies several possible "decision limits" one of which is that suggested by Currie (1968). Nicholson's (1963) paper (summarized in Nicholson (1966)) is directed toward applications where background counts are appreciably large relative to the total (gross) counts. In this case, statistical procedures based on the normal distribution (appropriate when background counts are not a significant proportion of the total counts) may no longer be applicable.

Specific recommendations are made (summarized in Appendix A) concerning the computation and reporting of counting error information for the three cases given above. These recommendations (based largely on Nicholson's work) are directed toward counting procedures currently in use by Group H8 at the Los Alamos Scientific Laboratory (LASL). References of interest not covered in this report are those of Pasternack and Liuzzi (1968), who discuss the estimation of radionuclide concentrations in mixtures using gamma ray spectrometry, and Head (1972) on the "minimum detectable true area" of a photopeak in a Ge(Li) gamma ray spectrum.

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[1]The NAEG was established by the Atomic Energy Commission, Nevada Operations Office in July 1970 (now the Energy Research and Development Administration (ERDA)), for the purpose of determining and predicting the effects of radioactivity on biota, particularly with respect to food chains and environmental factors affecting man in the environs of the Nevada Test Site.

2. SPECIFICATION OF THE PROBLEM

Consider the simplist situation of a single background count. Let
x = total (gross) number of counts in the sample obtained in time t,
and y = background count obtained in time s, where x and y were obtained on count instruments with efficiencies e_s and e_B, respectively.[2] Then

$$\tilde{c} = x/e_s t - y/e_B s \qquad (1)$$

= estimated total (sample) disintegrations per minute (dpm)
- estimated background dpm
= estimated net dpm.

$\tilde{c}$ is an estimate of C, the true (unknown) net disintegration rate, and $y/e_B s$ estimates B, the true (unknown) background disintegration rate.

In this paper the following questions are considered:

Q1. How does one determine from $\tilde{c}$ whether or not $C > 0$? If it is decided that $C > 0$, then the conclusion is made that activity above background has been "detected".

Q2. How does one compute confidence intervals about C, the true net disintegration rate?

Q3. How are the $\tilde{c}$ and their counting errors to be reported to the scientific community for evaluation and statistical analysis for both the "detected" and "not detected" cases?

Q4. How does one determine the minimum true net disintegration rate C_o that we can be confident will be "detected" most of the time. (Nicholson terms C_o the "threshold" rate. Currie and Hartwell denote C_o by L_D.)

In section 3 the answers to these questions are considered for the simple case described above, while later sections deal with the more complicated counting situations mentioned in the Introduction.

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[2] In most, if not all, applications the gross and background counts will be determined on the same counting instrument in which case e_s will equal e_B. However, for the purpose of greater generality, most of the formulas given in this report are in terms of e_B and e_s. They are appropriate whether or not $e_B = e_s$.

3. A SINGLE BACKGROUND READING

3.1 "Decision Limits"

In this section the various formulas presented are in terms of counts per minute (cpm) rather than dpm in order to simplify the introduction of some of the statistical concepts. Later sections are in terms of dpm and hence involve the efficiencies e_s and e_B.

The answer to Q1 involves computing $\tilde{c}$ using Eq. (1) in addition to L_C, which Currie (1968) calls the "decision limit" and Hartwell (1972) refers to as the "critical level". In any case,

L_C = the net count rate which must be exceeded before a sample can be said to contain any material above background.

Hartwell (1972) suggests L_C be computed as follows:

$$\begin{aligned} L_C &= k_\alpha \, \sigma_B \, (1 + s/t)^{1/2} \\ &= k_\alpha \left[\frac{y}{s} \left(\frac{1}{t} + \frac{1}{s} \right) \right] 1/2 \\ &= k_\alpha \, \sigma_o \quad , \end{aligned} \tag{2}$$

where

σ_B = standard deviation of the background count rate based on one background count determination y
$= \sqrt{y}/s$,
σ_o = standard deviation of $\tilde{c}$ when in fact C = 0 (see Section (1.1) of Appendix B for a derivation; also see Nicholson (1963: p. 17, 18)), and
k_α = 100 (1-α) percentile of the unit normal distribution.

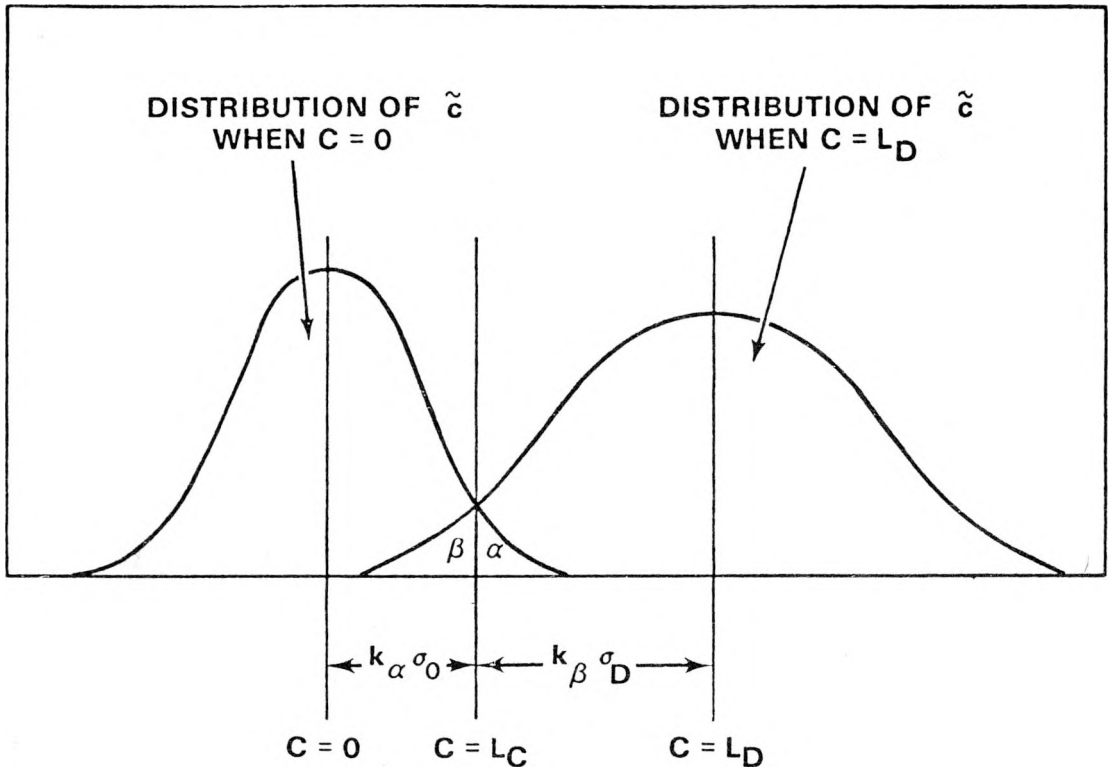
Then, computing $\tilde{c}$ using Eq. (1) the decision whether the true net count rate C is greater than zero is made by comparing $\tilde{c}$ with L_C as follows:

	Decision	Inference	
If $\tilde{c} > L_C$	"detected"	C > 0	
If $\tilde{c} \leq L_C$	"not detected"	C = 0	(3)

Thus, the solution to Q1 suggested by Currie (1968) and Hartwell (1972) is simply to compute $\tilde{c}$ from Eq. (1) and L_C from Eq. (2) and see whether $\tilde{c} > L_C$. Note that the decision of "detected" or "not detected" can be made only after the gross and background count have been determined. Note too that the decision is not being made whether or not C is detectable, but rather, whether it is "detected".

It is important to recognize that the above approach is actually a test of the null hypothesis H_0 : C = 0, where α is the "level of significance" or "Type I error" (α is discussed in the next paragraph; see Figure 1).

RELATIONSHIP BETWEEN THE "DECISION LIMIT" L_C AND THE "DETECTION LIMIT" L_D



α = TYPE I ERROR (LEVEL OF SIGNIFICANCE)

β = TYPE II ERROR

$1-\beta$ = "POWER" OF THE TEST

σ_0 = STANDARD DEVIATION OF $\tilde{c}$ WHEN C = 0

σ_D = STANDARD DEVIATION OF $\tilde{c}$ WHEN C = L_D

Figure 1

The recognition of this fact may help avoid confusion with the "detection limit" discussed in Section (3.4) below. Now, there may be some differen of opinion concerning whether Eq. (2) is the best way to compute L_C (in fact, a different formula is recommended below), but, hopefully, the basic notion of using a test of the hypothesis $H_o : C = 0$ to decide whether something has been "detected" can be accepted as a reasonable procedure.

It will be useful for later discussions to review the role of α in hypothesis testing. The "Type I error" (α) is the probability (specified by the experimenter) of wrongly concluding that something above background exists in the sample. Hence, if in fact $C = 0$, we would wrongly conclude that $C > 0$ about 100α percent of the time if the procedure in Eq. (3) is used. The particular value of α used should be decided upon (before counts are made) on the basis of the practical consequences of making a Type I error. In general, we want α to be small, and values of 0.01, 0.05 and 0.10 are often used. However, α would ordinarily never be greater than 0.10 since the risk (α) of falsely concluding that $C > 0$ would then be too high. The values of k_α in Eq. (2) for $\alpha = 0.01$, 0.05 and 0.10 are $k_{0.01} = 2.326$, $k_{0.05} = 1.645$ and $k_{0.10} = 1.282$, respectively.

The above approach to testing $H_o : C = 0$ is based on the assumption that $\tilde{c}$ is approximately normally distributed which allows us to use the particular values of k_α specified above. The study by Nicholson (1963:p. 3) indicates that even when the number of counts is quite low, the distribution of $\tilde{c}$ is still approximated quite well by the normal distribution.

Nicholson (1963) compared the method of computing L_C using Eq. (2) with three other methods, one of which (Eq. (4) below) he found to be preferable. He suggests the testing procedure (Eq. (3)) be followed when L_C is calculated as

$$L_C = k_\alpha \left[(x + y)/ts\right]^{1/2} , \qquad (4)$$

which makes use of the gross sample count x in estimating σ_o (see Nicholson (1963: p. 19) for a derivation of Eq. (4)). His argument for preferring Eq. (4) to Eq. (2) is based on the fact that, if in reality the true net count rate C is zero, then x as well as y is an estimate of the background count and hence a better estimate of σ_o is obtained by using both x and y. Nicholson denotes the tests using Eqs. (2) and (4) as decision rules D_2 and D_3, respectively. The remainder of this section is devoted to a discussion of the merits of D_2, D_3, and an "exact" test D_e, leading to a rule of thumb (Eq. (5)) for deciding which rule to use in a given situation.

Nicholson's work indicates that the exact significance levels of the decision rule D_3 are closer to the stated level α than is the case for rule D_2. (The significance level of the tests D_2 and D_3 are not exactly α for any finite sample and background count since only when these counts are infinitely large does $\tilde{c}$ become exactly normally distributed.) In particular, he found that when Eq. (2) (rule D_2) was used, too many "detected" conclusions were reached. Table D.1 in Appendix D (an abbreviated version of Nicholson's Table 1) compares the exact significance

levels of D_2 and D_3 with the stated level α. As an example of its use, consider the case where $\alpha = 0.05$ and the expected background count (tB) is 2 counts in time t. If $\rho = t/s$ (ratio of sample count time t to background count time s) is 1/3, then when C = 0 we see that the decision "detected" is reached about 9.5% of the time when D_2 is used, which is considerably greater than the expected 5% rate. For rule D_3 (Eq. (4)) the exact level is elevated only slightly to 5.9%.

An inspection of Table D.1 shows that the difference between exact and stated significance levels of both tests varies depending on the count time ratio ρ and the expected background count tB. The exact levels of D_2 become extremely elevated for $\rho \geq 1$ when $tB \leq 5$. In general, the test D_3 is less affected than D_2, but extreme underestimation of α can occur when $tB \leq 2$ when $\rho \geq 1$.

Now, an evaluation of D_2 and D_3 must also consider their performance when $C > 0$, i.e., when we would be making a "Type II error" (see Figure 1 and the discussion in Section 3.4) if we did not reject $H_o : C = 0$. Nicholson's results (his figures 2, 3, 4) indicate that rule D_2 (Eq. (2)) is slightly more effective, i.e., is more powerful, at detecting $C > 0$ than is D_3 (Eq. (4)), but as pointed out above, at the expense of an elevated significance level. Hence, when C is greater than zero, Eq. (2) is somewhat better than Eq. (4) in detecting it, but when C = 0, Eq. (2) tends to wrongly conclude that $C > 0$ a greater proportion of the time than does Eq. (4). Thus, a decision as to whether Eq. (2) or Eq. (4) is to be used in testing $H_o : C = 0$ depends on the magnitude of these effects (which depend on ρ and tB) and to the importance attached to these two types of errors.

Figures B.1 - B.18 in Nicholson's report give the "power" (probability of rejecting $H_o : C = 0$ when in fact $C > 0$) of rule D_3 for various values of α, ρ, the expected background count tB, and the ratio of expected counts $k = tC/(tB)^{1/2}$. Five of these are reproduced here in Appendix D as Figures D.1 - D.4. These are for $\rho = 1$ and 1/2 when $\alpha = 0.01$ and for $\rho = 1/3$, 1/2 and 1 when $\alpha = 0.05$. Each curve in these figures corresponds to an expected background count tB and shows how for given tB the ability or "power" of rule D_3 using Eq. (4) to detect a positive net count increases as the ratio k increases. These figures also show that for a given tB and k, the power of D_3 increases as ρ increases, i.e., as the sample is counted for a longer time relative to the background count time.

Nicholson (1963: p. 6) derives an exact test D_e for use when Eq. (4) does not perform well. He suggests the following rule of thumb:

"For stated α equal to $\left\{\begin{matrix}0.10\\0.05\\0.01\end{matrix}\right\}$ [3] use decision rule D_3 when $\rho \leq 1$ and $y > \left\{\begin{matrix}5\\10\\15\end{matrix}\right\}$; use exact decision rule D_e when $\rho > 1$ and/or $y \leq \left\{\begin{matrix}5\\10\\15\end{matrix}\right\}$". (5)

This appears to be a reasonable guideline and its adoption by the participating laboratories in the NAEG program is recommended.

The rule D_e (Nicholson (1963: p. 24)) is exact in the sense that its significance level is exactly α rather than being elevated or depressed for certain values of ρ and tB as happens with D_2 and D_3. It is based on the binomial rather than the normal distribution and is an example of a randomized decision rule. The decision "detected" or "not detected" is made by comparing the gross count x with the number X, where tables of X and its associated probability P are given in Nicholson (1963; p. A.1-A.6). More specifically:

		Decision	Inference
Rule D_e:	If $x > X$	"detected"	$C > 0$
	If $x = X$	"detected" with probability P	$C > 0$ or $C = 0$ depending on outcome of random number draw (see text)
	If $x < X$	"not detected"	$C = 0$

Nicholson gives tables of the fixed numbers X and P for α = 0.10, 0.05 and 0.01 and values of x + y from 1 to 60 and ρ from 1/10 to 10. These are reproduced here as Table D.2 in Appendix D.

When x = X, rule D_e specifies that the decision "detected" or "not detected" depends on the "randomization" probability P. If this should occur in practice, i.e., if x = X, the decision can be reached by choosing at random a 3 digit number, say P', from a random number table (i.e., a table of randomly assorted digits from 0 to 9), dividing it by 1000 and then comparing it with the tabled P value for specified α, x + y and ρ. Thus, when x = X

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[3] "In the remainder of this statement, use the top quantity in each bracket with 0.10, the second with 0.05, etc."

	Decision	Inference
If $\frac{P'}{1000} > P$	"detected"	$C > 0$
If $\frac{P'}{1000} \leq P$	"not detected"	$C = 0$

where P is tabled in Table D.2.

As an example, suppose it is desired to make an $\alpha = 0.05$ level test when $\rho = 2$ and $y = 4$ so that according to the rule of thumb (Eq. (5)) the exact test D_e should be used. Suppose $x = 20$ so that $x + y = 24$. Entering Table D.2 for $\alpha = 0.05$, $x + y = 24$ and $\rho = 2$ we find $X = 20$ and $P = 0.762$. Since $x = X$, a random 3 digit number P' from a random number table is chosen and divided by 1000, giving say, $P'/1000 = 0.037$. Since 0.037 is less than the tabled P value of 0.762, we decide "not detected" and infer $C = 0$. The validity of the procedure depends on the 3 digit number P' being selected at random from the random number table so that any number between 000 and 999 is equally likely to be chosen.

The test is very simple to make when $x > X$ or $x < X$, since there is then no need to take the additional step of using a random number table. Random number tables are often given in standard statistical methods textbooks (e.g., Snedecor and Cochran (1967)). More extensive tables are given in "A Million Random Digits With 100,000 Normal Deviates" published by The Rand Corporation (1955). A more sophisticated approach would be to use a pseudo-random number generator on a digital computer to obtain a 3 digit number for use when $x = X$, but some assurance must be given that the random number generator does in fact generate truly random digits.

3.2 Confidence Intervals

Concerning Q2 in Section 2 on the computation of confidence intervals, it is recommended that these be computed in units of dpm, i.e., after correcting for the efficiencies e_s and e_B of the sample and background counting instrument(s), respectively. When $tB > 10$ and one background count determination is available, the recommended method for computing the upper and lower 100 $(1-\alpha)$ percent confidence limits on the true net disintegration rate C is

$$(x/e_s t - y/e_B s) \pm k_{\alpha/2} (x/e_s^2 t^2 + y/e_B^2 s^2)^{1/2} \quad . \qquad (6)$$

The quantity under the square root sign is an estimate of the variance of the estimated net dpm $(\hat{C})$ when $C \geq 0$ (see Section (1.1) of Appendix B for a proof). Values of $k_{\alpha/2}$ for $\alpha = 0.10$, 0.05 and 0.01 are $k_{0.10/2} = 1.645$, $k_{0.05/2} = 1.960$ and $k_{0.01/2} = 2.576$, respectively. For example, a 95% confidence interval on C would be obtained using $k_{0.05/2} = 1.960$. Eq. (6) (with $e_s = e_B = 1$)[4] is the formula used by Hartwell (1972: p. 10) to

[4]No counting instrument has an efficiency of 1. However, by setting $e_s = e_B = 1$ in Eq. (6), confidence limits are obtained on the net count rate rather than the net disintegration rate.

compute confidence limits on the net count rate when $\tilde{c} > L_C$. However, there is no necessity to limit the computation of a confidence interval to the situation where $\tilde{c} > L_C$. It is an equally valid computation when $\tilde{c} \leq L_C$, i.e., when "not detected" is the conclusion reached from testing H_o: $C = 0$. It is not recommended, however, that a confidence interval be reported on a routine basis by the laboratories (see Section (3.3) below).

Nicholson (1963: p. 5) considers an alternative method of computing confidence intervals which he recommends over Eq. (6) when y (the background count) lies between $2 + 3/2\rho$ and $6 + 12/\rho$. The form of the confidence interval on the true net count rate C is

$$\frac{L}{t} \leq C \leq \frac{U}{t} \tag{7a}$$

where L/t and U/t are the lower and upper $100(1-\alpha)$ percent confidence limits. His method may also be used to put confidence intervals on the true net disintegration rate C when $e_s = e_B$, in which case Eq. (7a) becomes

$$\frac{L}{te_s} \leq C \leq \frac{U}{te_s} \quad . \tag{7b}$$

In the unlikely event that $e_s \neq e_B$ then Eq. (6) must be used. The limits L and U depend on x, y, $\rho = t/s$ and α, and are given in Table D of Nicholson (1963: pp. D.1-D.99) for $1-\alpha = 0.90$, 0.95 and 0.99, for ρ between 2 and 1/10. Unfortunately, this table is too long to include in its entirety here, but in Table D.3 in Appendix D his tables for $1-\alpha = 0.95$ and $\rho = 1$ and 1/2 are given. From Nicholson (1963, p. 8) it appears that the differences between Eqs. (6) and (7a) become negligible for expected backgrounds >10. While he does not specifically discuss what happens when $e_s = e_B \neq 1$ (the case in practice) it is suggested here that Eq. (7b) be used in place of Eq. (6) whenever $e_s = e_B$ and $tB < 10$. If Table D.3 is not adequate or if Nicholson's Table D is not available, Eq. (6) may be used in any case. Nicholson (1963: p. 8) states that the use of either method is questionable if $y < 2 + 3/2\rho$.

3.3 Reporting of Data

Now, concerning question Q3 in Section (2) above on the reporting of data, we feel that "less than" numbers or the letters "ND", used in place of a numerical result to signify "not detected", should not be reported, regardless of the outcome of the recommended testing procedures in Section (3.1). Whether or not the decision "detected" has been reached, the net dpm and its counting error should be reported. All net disintegrations $\tilde{c}$ obtained are important even if they are negative or very near zero since they provide information about the total variability associated with a given counting system. Concerning negative numbers, if C = 0 we would in fact expect to obtain negative as well as positive net results since x and y are both estimating background levels. If the decision "not detected" is reached, this could be indicated on the output sheet by the symbol, say † (the symbol * should not be used since this customarily indicates statistical significance, which is opposite to the intended meaning here).

To be specific, the recommended reporting system is

$$\tilde{c}^{\dagger} \pm \text{"1 } \sigma \text{ counting error"} \quad , \tag{8}$$

which, when one background count determination is being used to estimate the background rate, is computed as

$$(x/e_s t - y/e_B s)^{\dagger} \pm (x/e_s^2 t^2 + y/e_B^2 s^2)^{1/2} \quad , \tag{9}$$

where the symbol † is used only if the test of $H_o : C = 0$ (see Section (3.1)) results in the decision "not detected". If desired, the absolute value of the percent counting error ($100\ \sigma/\tilde{c}$) could also be reported.

This method of reporting data is somewhat different from that suggested by Hartwell (p. 10). He recommends that if a net count is "detected", the 95% confidence interval on the net count rate (computed as in Eq. (6) above) be reported, i.e.,

$$\tilde{c} \pm k_{.05/2} (x/t^2 + y/s^2)^{1/2} \quad , \tag{10}$$

which is similar to the reporting system above except that he multiplies the right-hand side of Eq. (8) above by $k_{.05\ 2} = 1.96$. Now, when the decision "not detected" is reached, Hartwell and Currie recommend that the upper 95 percent one-sided confidence limit

$$U = (x/t - y/s) + 1.645\ (x/t^2 + y/s^2)^{1/2} \tag{11}$$

be computed and that the sample be reported as having a net count rate of "less than" the computed U with 95% confidence. We have noted above our objection to reporting "less than" numbers since it throws away information. While there is nothing statistically wrong with reporting results as in Eq. (10), the recommended method (Eq. (9)) is more flexible since confidence intervals for any desired level of significance (not just $\alpha = 0.05$) can be quickly computed by multiplying the right-hand side of Eq. (9) by $k_{\alpha/2}$. Note too that U in Eq. (11) can be easily computed from the information in Eq. (9) if the individual investigator so wishes. Also, Hartwell (p. 10) indicates that his reporting method can be confusing to those who do not adequately understand the method since it is likely that some positive values reported will be less than other "less than" reported values. The use of Eq. (8) above should avoid this kind of confusion.

3.4 "Detection Limit" or Threshold

Quoting from Hartwell (p. 7):

> "Suppose someone calls you on the phone and wants to know what your 'lower detection limit' is. He is really asking, 'If I send you a sample, how much material must be in that sample for you to be sure (95 percent sure) that you will find it?'".

Currie (1968) denoted that amount of material by L_D, the "detection limit", where

L_D = the smallest net count rate for which we can be 100(1-β) percent sure of detecting the material present in the sample using a given measurement process.

The "95 percent sure" in the above quote is the "100(1-β) percent sure" used in this definition of L_D. Currie showed how to compute L_D and Hartwell extended his results to the case where the sample count time (t) is different from the background count time (s). In the present discussion L_D is defined in terms of dpm rather than cpm.

In Section (1.2) of Appendix B it is shown that when working with disintegration rates, L_D is computed as follows:

$$L_D = L_C + \frac{k_\beta^2}{2e_s}\left[1 + \left(1 + \frac{4L_C e_s}{k_\beta^2} + \frac{4L_C^2 e_s^2}{k_\alpha^2 k_\beta^2}\right)^{1/2}\right] \qquad (12)$$

which reduces to

$$L_D = 2L_C + k^2/e_s \quad , \qquad (13)$$

when $k_\alpha = k_\beta = k$ (Hartwell (p. 9)). Figure 1 shows the relationship between L_C and L_D. The "decision limit" L_C in Eqs. (12) and (13) should be computed using Eq. (A.3) in Appendix A unless it is known without a doubt that the sample contains no activity above background. In the latter case L_C could be computed using the right hand side of Eq. (A.1) since both x and y will then estimate background. The point here is that σ_o^2 should estimate the variance of $\tilde{c}$ when C = 0, and this will not be the case if x is used in the estimation of σ_o^2 when (unknown to us) C > 0. An examination of Figure 1 may help here (it is also useful in understanding the proof of Eq. (12) in Section (1.2) of Appendix B).

The quantity k_β is the 100(1-β) percentile of the unit normal distribution where β is the "Type II error" mentioned above in Section 3.1 and is defined as the probability of wrongly concluding that nothing above background exists in the sample. Conversely, 1-β refers to the "power" or confidence we desire in being able to detect the net rate L_D. Values of k_β for β = 0.01, 0.02, 0.05 and 0.10 are the same as those given for k_α in Section 3.1, for α = 0.01, 0.02, 0.05 and 0.10, respectively.

Notice from Eqs. (12) and (13) that the value of the "detection limit" L_D changes depending on the background count y, the counting times t and s, the efficiencies e_s and e_B, and the values of k_α and k_β. Hence, the answer to the question quoted from Hartwell at the beginning of this section can only be given after these quantities are specified for a given measurement process. The purpose of computing L_D is to obtain some notion of the level of net disintegration rate that must be present in a sample before it will be "detected" a large portion (1-β) of the time by

a given measurement technique. If $1-\beta$ is increased, we see that k increases which implies L_D also increases. Hence, if we want to be 99% sure ($1-\beta = 0.99$) of detecting material present in the sample, L_D will be a larger fixed net disintegration rate than will be the case if we need to be only 80% sure ($1-\beta = 0.80$).

We have seen from Nicholson's study that the significance level associated with L_C computed using Eq. (2) tends to be greater than the stated level α, particularly for $tB \leq 5$ and $\rho \geq 1$. Referring to Figure 1 this implies the area α under Curve I is too large, i.e., L_C is farther to the left than it should be. This in turn implies that L_D is somewhat smaller than it should be relative to the stated α and β requirements. Unfortunately, we have no ready solution to this problem other than to point it out and to urge caution in the interpretations and applications of L_D for small background counts and $\rho \geq 1$. The magnitude of this problem when $e_s \neq e_B$ is unknown.

4. MULTIPLE INDEPENDENT BACKGROUND READINGS

If n background counts $y_1, y_2, \ldots, y_n$ counted for times $s_1, s_2, \ldots, s_n$, respectively, have been made to estimate the true background disintegration rate B, these should be utilized in testing $H_o: C = 0$ and in computing L_D. The generalizations of Eqs. (1) and (2) to the case $n > 1$ (assuming the n background readings all estimate the same true background level and all are counted with the same efficiency) are

$$\tilde{c} = x/t - \bar{y}/\bar{s} \tag{14}$$

and

$$L_C = k_\alpha \sigma_o = k_\alpha \left[(\bar{y}/\bar{s})\,(1/t + 1/n\bar{s})\right]^{1/2}, \tag{15}$$

where $\bar{y} = \left(\sum_{i=1}^{n} y_i\right) \Big/ n$

and $\bar{s} = \left(\sum_{i=1}^{n} s_i\right) \Big/ n$

so that $\bar{y}/\bar{s} = \sum_{i=1}^{n} y_i \Big/ \sum_{i=1}^{n} s_i$.†

†The estimate $\bar{y}/\bar{s}$, rather than the average rate $(1/n) \sum_{i=1}^{n} (y_i/s_i)$, is used to estimate B since $\mathrm{Var}(y_i)$ (which equals $E(y_i)$) is proportional to count time s_i (see, e.g., Armitage (1971: p. 278)).

There are several comments we wish to make at this point. First of all, we note that Eq. (15) reduces to Eq. (2) when n = 1. Secondly, the proof that $\sigma_o = [(\bar{y}/\bar{s})\ (1/t + 1/n\bar{s})]^{1/2}$ when C = 0 (see Section (1.1) of Appendix B) depends on the assumption that $\bar{s}$, the average background count time, is a fixed constant rather than a random variable. $\bar{s}$ would be a random variable e.g., if the background counts were obtained by letting the counting continue until a fixed number of background counts were obtained. Eq. (15) is not appropriate for that type of procedure, nor for that matter are any of the formulas given in this paper. Nicholson (1963: p. 5) notes that Birnbaum (1954) discusses random time procedures.

The "rule of thumb" given in Eq. (5) for n = 1 indicates Eq. (4) be used to test H_o : C = 0 unless the background count is very low and/or the ratio of count time $\rho = t/s$ is greater than 1. Nicholson (1963) did not consider the case n > 1, but using the method by which he derived Eq. (4) (weighting the $x, y_1, y_2, \ldots, y_n$ by the inverse of their variances) it is shown in Section (1.3) of Appendix B that Eq. (4) can be generalized for n > 1 to

$$L_C = k_\alpha \sigma_o = k_\alpha \left[\frac{\left(x + \sum_{i=1}^{n} y_i \right)}{\left(t + \sum_{i=1}^{n} s_i \right)} \left(\frac{1}{t} + \frac{1}{n\bar{s}} \right) \right]^{1/2} . \qquad (16)$$

Eqs. (2), (4), and (15) are all special cases of Eq. (16), since (16) reduces to (4) when n = 1 and to (15) when the gross count information x and t are not used. Eq. (2) is a special case of Eq. (15) when n = 1.

Analogous to the use of Eq. (4) when n = 1, a test of H_o:C = 0 is made by seeing whether $\tilde{c} = x/t - \bar{y}/\bar{s}$ is greater than L_C computed using Eq. (16): if so, we decide something has been "detected" and infer C > 0; otherwise we decide "not detected" and infer C = 0. We have not constructed an exact test of H_o:C = 0 analogous to D_e for n = 1 (although this could probably be done). Nor have we made any study of the exact significance levels of the test using Eq. (16) to see whether an exact test is even required. However, Nicholson (p. 26) proved when the gross and background counts are large for n = 1, the exact test D_e is equivalent to D_3 (the test using Eq. (4)). This suggests the significance levels of Eq. (16) may not be as biased as are those for D_3 since n > 1 background counts are pooled to make a larger total background count than when n = 1. In any case, based on Nicholson's study for n = 1, the significance level of the test using Eq. (16) should be uniformly closer to the stated level

than that of the test using Eq. (15). Hence, it is recommended that Eq. (16) be used to compute L_C when $n > 1$, with the cautionary note that for $\rho \geq 1$ and very low background counts too few "detected" decisions may be obtained when in fact $C = 0$.

The above discussion has been in terms of cpm rather than dpm. In Appendix A, Eqs. (14) and (16) are rewritten as Eq. (A.1) for the more general case where the background determinations were all taken on instruments with the same efficiency e_B, but e_B need not equal e_s, nor be equal to one. Hence, a test of $H_o : C = 0$ can be carried out in this more general case by using Eq. (A.1). As with the case where $n = 1$, if it should happen that $e_s = e_B$ then Eq. (16) may be used to make the test, i.e., the decision reached will be the same whether Eq. (16) or (A.1) is used. But the values of $\tilde{c}$ and the decision limit L_C themselves are not the same using the two equations.

Concerning the computation of a $100(1-\alpha)$ percent confidence interval on C when $n > 1$, the straightforward generalization of Eq. (6) is recommended, i.e.,

$$\left[\frac{x}{e_s t} - \frac{\sum_{i=1}^{n} y_i}{e_B \sum_{i=1}^{n} s_i}\right] \pm k_{\alpha/2} \left[\frac{x}{e_s^2 t^2} + \frac{\sum_{i=1}^{n} y_i}{e_B^2 \left(\sum_{i=1}^{n} s_i\right)^2}\right]^{1/2}, \quad (17)$$

where the standard deviation on the right hand side is derived in Section (1.1) of Appendix B. The data reported by the laboratories should be $\tilde{c} \pm 1\,\sigma$, i.e.,

$$\left[\frac{x}{e_s t} - \frac{\sum_{i=1}^{n} y_i}{e_B \sum_{i=1}^{n} s_i}\right]^{\dagger} \pm \left[\frac{x}{e_s^2 t^2} + \frac{\sum_{i=1}^{n} y_i}{e_B^2 \left(\sum_{i=1}^{n} s_i\right)^2}\right]^{1/2}, \quad (18)$$

where the symbol † is used only if $\tilde{c} \leq L_C$, where $\tilde{c}$ and L_C are computed as in Eq. (A.1) in Appendix A. The absolute value of the percent counting error $|100\,\sigma/\tilde{c}|$ could also be reported.

The limit L_D can be computed using Eqs. (12) or (13) (whichever is appropriate depending on whether or not $k_\alpha = k_\beta$), where L_C in these equations is computed using Eq. (A.3) in Appendix A rather than the right hand side of Eq. (A.1) for reasons discussed in Section (3.4) for the case $n = 1$.

5. "BASE LINE" CORRECTION FOR BACKGROUND IN GAMMA SPECTROMETRY

In gamma spectrometry the determination of background is not simply a matter of counting a background sample. In this case the technique known as "base line" correction can be used to estimate background levels. The computational formulas for L_C, L_D, and confidence intervals are slightly different than those given above.

In "base line" correction the "peak" channel is first located and a certain number of channels on either side are used to estimate the total (gross) count. Then channels on either side of these "peak" channels are averaged to estimate the background count. In this situation the counting time for both gross and background counts are necessarily equal and denoted here by t. Similarly there is only one counting instrument and hence one efficiency e_B. Let

$$\sum_{i=1}^{n_1} x_i = \text{total gross counts over } n_1 \text{ "peak" channels}$$

$$\bar{y} = \frac{1}{n_2} \sum_{i=1}^{n_2} y_i = \text{average background count over } n_2 \text{ channels bracketing the } n_1 \text{ "peak" channels.}$$

Then

$$\tilde{c} = \frac{1}{te_B} \sum_{i=1}^{n_1} x_i - n_1 \left(\frac{1}{te_B n_2} \sum_{i=1}^{n_2} y_i \right) \qquad (19)$$

$$= \text{gross dpm} - (n_1)\text{x(average background dpm)}$$

$$= \text{estimated net dpm.}$$

Then for testing $H_o: C = 0$ we may compare $\tilde{c}$ computed using Eq. (19) with (see Section 2.2 in Appendix B for the derivation of Eq. (20))

$$L_C = k_\alpha \sigma_o$$

$$= \frac{k_\alpha}{te_B} \left[\frac{n_1}{n_2} \left(\sum_{i=1}^{n_1} x_i + \sum_{i=1}^{n_2} y_i \right) \right]^{1/2} , \qquad (20)$$

the decision "detected" being reached if $\tilde{c} > L_C$; otherwise we conclude "not detected". The $100(1 - \alpha)$ percent confidence interval about C is given by the limits

$$\tilde{c} \pm \frac{k_{\alpha/2}}{te_B} \left[\sum_{i=1}^{n_1} x_i + (n_1/n_2)^2 \sum_{i=1}^{n_2} y_i \right]^{1/2} , \qquad (21)$$

where

$$\frac{1}{t^2 e_B^2}\left(\sum_{i=1}^{n_1} x_i + (n_1/n_2)^2 \sum_{i=1}^{n_2} y_i\right) \qquad (22)$$

is an estimate of the variance of $\tilde{c}$ when $C \geq 0$. The data reported by the laboratories should be

$$\tilde{c}^{\dagger} \pm [\mathrm{Var}(\tilde{c})]^{1/2} \quad ,$$

where $\dagger$ is used only if $\tilde{c} \leq L_C$, where $\tilde{c}$ is given by Eq. (19), and $\mathrm{Var}(\tilde{c})$ by Eq. (22). The computation of L_D is according to Eqs. (12) or (13) with L_C computed as

$$\frac{k_\alpha}{e_B t}\left[\frac{n_1\,(n_1 + n_2)}{n_2^2} \sum_{i=1}^{n_2} y_i\right]^{1/2} . \qquad (23)$$

Note that L_D now depends on $\bar{y}$, e_B, t, n_1, n_2, k_α and k_β. The derivations of the various variance formula given here for "base line" correction are given in Sections 2.1 and 2.2 of Appendix B.

6. APPROXIMATE TESTS AND CONFIDENCE INTERVALS WITH ALPHA SPECTROMETRY

When ^{239}Pu is determined using alpha spectrometry where a tracer, say ^{242}Pu, is added to the sample, the estimation problems can become rather complicated. This complexity arises because of the need to (i) correct for contamination of the sample by ^{239}Pu introduced with the ^{242}Pu tracer, (ii) to take into account the yield of the ^{242}Pu obtained from the sample, and (iii) to allow for different counting times and efficiencies of the various counting instruments used. For example, using the notations defined in Appendix (C) it is stated there (Eq. (C.2)) that the estimated disintegration rate of ^{239}Pu is given by

$$\tilde{c} = \frac{1}{e_T}\left[\frac{\left(\frac{x}{t} - \frac{b_x}{s}\right)\left(\frac{z}{t_T} - \frac{b_z}{s_T}\right)}{\left(\frac{w}{t} - \frac{b_w}{s}\right)} - \left(\frac{v}{t_T} - \frac{b_v}{s_T}\right)\right] . \qquad (24)$$

An approximate expression for $\mathrm{Var}(\tilde{c})$ can be obtained using a technique commonly known as "propagating the errors" or the "delta method", which uses the first few terms of a Taylor series expansion of $\tilde{c}$. In Appendix C three levels of generality are considered relative to the equality of counting times for sample and background count determinations and to whether the ^{242}Pu tracer is contaminated with ^{239}Pu. For each case we

derive by "propagating the errors" the appropriate formula for (i) computin the "1 σ counting error" for reporting with the net dpm, and for (ii) computing the variance of $\tilde{c}$ used to calculate L_C for use in computing L_D and for testing whether the "true" net count is zero. Since these approximate expressions are rather lengthy and are given in Appendix C, we do not repeat them here. These results are also summarized in Appendix A.

The accuracy of the approximate variance formulae given in Appendix C will depend on the magnitude of the relative variances [Var(count)]/(count)^2 of the various counts, which will be large when counting times are short and/or the count rate is low. One approach that could be used to study their accuracy would be a computer simulation study. The approximate variance formulas would be evaluated for various sample, background, and tracer count levels, and for different counting times and yields. Another more direct approach would be to try and enlarge upon the work of Goodman (1962) who found an exact expression to replace the approximate (propagation of errors) formula for the variance of the product of two means. In the present context we would need to consider in addition the ratio of means. Until such studies have been made, care should be used in the interpretation of tests of significant or in the "detection limit" L_D computed when sample and background counts are few in number.

Concerning L_D, the approximate variance expressions in Appendix C indicate that the "detection limit" depends on not only the sample and background counts, but also on the yield, efficiency, counting time and the amount of ^{239}Pu contamination present in the tracer. Hence, the utility of computing such a limit would seem to be reduced if the computed L_D varies widely as these variables change. Some consideration should also be addressed to whether the formulae for L_D given in Eqs. (12) and (13) are the most appropriate when working with alpha spectrometry. We have not looked at the problem but suspect that a more complicated expression may be required.

7. DISCUSSION

In this paper certain recommendations have been made relative to the computation and reporting of count statistics for counting procedures currently practiced by Group H8 at LASL. While the topics discussed here are necessarily limited in scope, future work could be directed toward counting procedures currently in use at other NAEG participating laboratories. The estimation techniques discussed by Pasternack and Liuzzi (1968) might also be applied to gamma ray spectrometry estimation problems at the various laboratories.

Hopefully the discussion here of the "decision limit" L_C and the "detection limit" L_D will result in a better understanding of the different roles played by these two quantities. A mutual understanding of these concepts might contribute to more uniform methods of deciding whether something above background is in the sample and to more uniform reporting methods.

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APPENDIX A

SUMMARY OF RECOMMENDATIONS

APPENDIX A

1. NOTATION

x = total (gross) sample counts in time t

y_i = i^{th} background count in time s_i, $i = 1, 2, \ldots, n$ $(n \geq 1)$

C = true (unknown) net disintegration rate (dpm)

B = true (unknown) background disintegration rate (dpm)

$\tilde{c}$ = estimated net disintegration rate (dpm)

$\rho = t/s$ = ratio of sample to background count time when $n = 1$

k_α = $100(1 - \alpha)$ percentile of the unit normal distribution

n = number of independent background count determinations $(n \geq 1)$

n_1 = number of "peak" channels in "base line" correction for background

n_2 = number of "background" channels in "base line" correction for background

e_s = efficiency of the counter on which x is obtained

e_B = efficiency of the counter on which each of the n background counts were obtained.

e = efficiency of the counter in "base line" correction for background

2. RECOMMENDATIONS

Q1: How to test $H_o: C = 0$, i.e., whether $\tilde{c}$ indicates something above background has been "detected".

A: One or more independent background readings $(n \geq 1)$.[5]

Decide "detected" if

---

[5] Use decision rule D_e rather than Eq. (A.1) when $\rho > 1$ and/or $\sum_{i=1}^{n} y_i \leq \begin{Bmatrix} 5 \\ 10 \\ 15 \end{Bmatrix}$. See "rule of thumb" (Eq. (5)).

$$\left(\frac{x}{e_s t} - \frac{\sum_{i=1}^{n} y_i}{e_B \sum_{i=1}^{n} s_i} \right) > k_\alpha \left[\frac{\left(x + \sum_{i=1}^{n} y_i \right)}{\left(t + \sum_{i=1}^{n} s_i \right)} \left(\frac{1}{e_s^2 t} + \frac{1}{e_B^2 n \bar{s}} \right) \right]^{1/2} . \qquad (A.1)$$

Otherwise decide "not detected".

B: "Base line" correction

"Detected" if

$$\frac{1}{te} \left[\sum_{i=1}^{n_1} x_i - (n_1/n_2) \sum_{i=1}^{n_2} y_i \right] > \frac{k_\alpha}{te} \left[\frac{n_1}{n_2} \left(\sum_{i=1}^{n_1} x_i + \sum_{i=1}^{n_2} y_i \right) \right]^{1/2} . \qquad (A.2)$$

Otherwise decide "not detected".

C: Alpha spectrometry (see Appendix C for approximate formulas and notation).

	Case	"Detected" if
1	(most general)	Eq. (C.2) > k_α [Eq. (C.12)]
2	($t = s$, $t_T = s_T$)	Eq. (C.5) > k_α [Eq. (C.13)]
3	($t = s$, $t_T = s_T$, $v = 0$, $b_v = 0$)	Eq. (C.7) > k_α [Eq. (C.14)]

Otherwise decide "not detected".

Q2: How to Compute a $100(1-\alpha)$ percent confidence interval on C.

A: One or more independent background readings ($n \geq 1$)

A.1 $e_s = e_B$ and $tB < 10$

$$\frac{L}{te_s} \leq C \leq \frac{U}{te_s}$$

where L and U are tabled by Nicholson (1963: Tables D.1, D.2, D.3) for $1 - \alpha = 0.90$, 0.95 and 0.99 for various values of ρ, x and y. See Table D.3 in the present report for his tables when $1 - \alpha = 0.95$ and $\rho = 1$ and 1/2.

A.2 $e_s \neq e_B$

$$\left[\frac{x}{e_s t} - \frac{\sum_{i=1}^{n} y_i}{e_B \sum_{i=1}^{n} s_i}\right] \pm k_{\alpha/2}\left[\frac{x}{e_s^2 t^2} + \frac{\sum_{i=1}^{n} y_i}{e_B^2 \left(\sum_{i=1}^{n} s_i\right)^2}\right]^{1/2}$$

B: "Base line" correction.

$$\frac{1}{et}\left[\sum_{i=1}^{n} x_i - (n_1/n_2) \sum_{i=1}^{n} y_i\right] \pm \frac{k_{\alpha/2}}{et}\left[\sum_{i=1}^{n} x_i + (n_1/n_2)^2 \sum_{i=1}^{n} y_i\right]^{1/2}$$

C: Alpha spectrometry (all results are approximate)

Case	Approximate Confidence Interval
1	Eq. (C.2) $\pm k_{\alpha/2}$ [Eq. (C.4)]
2	Eq. (C.5) $\pm k_{\alpha/2}$ [Eq. (C.6)]
3	Eq. (C.7) $\pm k_{\alpha/2}$ [Eq. (C.8)]

Q3: How are data to be reported?

1. Never report "less than" numbers or "ND".
2. Always report the estimated net disintegration rate $\tilde{c}$ along with its 1σ counting error as follows:

   $\tilde{c}^{\dagger} \pm 1\sigma$ counting error,

   where the symbol † is present only when the decision "not detected" has been reached using the methods summarized above under Q1. Percent counting error ($100\ \sigma/\tilde{c}$) may be reported if desired.

A: Multiple background readings ($n \geq 1$)

$$\left[\frac{x}{e_s t} - \frac{\sum_{i=1}^{n} y_i}{e_B \sum_{i=1}^{n} s_i}\right] \pm \left[\frac{x}{e_s^2 t^2} + \frac{\sum_{i=1}^{n} y_i}{e_B^2 \left(\sum_{i=1}^{n} s_i\right)^2}\right]^{1/2}$$

B: "Base line" correction

$$\frac{1}{et}\left[\sum_{i=1}^{n_1} x_i - (n_1/n_2) \sum_{i=1}^{n_2} y_i\right] \pm \frac{1}{et}\left[\sum_{i=1}^{n_1} x_i + (n_1/n_2)^2 \sum_{i=1}^{n_2} y_i\right]^{1/2}$$

C: Alpha spectrometry

Case	Data to be Reported
1	Eq. (C.2) ± Eq. (C.4)
2	Eq. (C.5) ± Eq. (C.6)
3	Eq. (C.7) ± Eq. (C.8)

Q4: How do we determine the minimum true net disintegration rate L_D that we can be sure will be "detected" $100(1-\beta)$ percent of the time for significance level α?

General Formula

(1) $k_\alpha \neq k_\beta$

$$L_D = L_C + \frac{k_\beta^2}{2e_s}\left[1 + \left(1 + \frac{4L_C e_s}{k_\beta^2} + \frac{4L_C^2 e_s^2}{k_\alpha^2 k_\beta^2}\right)^{1/2}\right]$$

(2) $k_\alpha = k_\beta = k$

$$L_D = 2L_C + k^2/e_s$$

The quantity L_C in these expressions is computed as follows:

A: Multiple background counts ($n \geq 1$)

$$L_C = k_\alpha \left[\frac{\sum_{i=1}^{n} y_i}{\sum_{i=1}^{n} s_i} \left(\frac{1}{e_s^2 t} + \frac{1}{e_B^2 n \bar{s}} \right) \right]^{1/2} \qquad (A.3)$$

B: "Base line" correction

$$L_C = \frac{k_\alpha}{et} \left[\frac{n_1(n_1 + n_2)}{n_2^2} \sum_{i=1}^{n_2} y_i \right]^{1/2}$$

C: Alpha spectrometry (all results are approximate)

Case	L_C used to compute L_D
1	k_α [Eq. (C.12)]
2	k_α [Eq. (C.13)]
3	k_α [Eq. (C.14)]

APPENDIX B

DERIVATIONS OF VARIANCE FORMULAS FOR $n \geq 1$ BACKGROUND READINGS

APPENDIX B

1.1 One or More Independent Background Determinations ($n \geq 1$)

1.1 Derivation of general variance formulae for σ_0^2 and σ_D^2.

When the n background determinations have the same efficiency e_B, the estimated dpm is

$$\tilde{c} = x/e_s t - \sum_{i=1}^{n} y_i \; / \; e_B \sum_{i=1}^{n} s_i \quad .$$

Therefore,

$$\text{Var}(\tilde{c}) = E(x) \; / \; e_s^2 t^2 + \sum_{i=1}^{n} E(y_i) \; / \; e_B^2 \left(\sum_{i=1}^{n} s_i \right)^2$$

$$= (C + B)t \; / \; e_s^2 t^2 + B \; / \; e_B^2 \, n\bar{s} \quad .$$

$$= C \; / \; e_s^2 t + B \left(1/e_s^2 t + 1/e_B^2 n\bar{s} \right) \qquad \text{(B.1)}$$

When $C = 0$ then $\text{Var}(\tilde{c})$ is given by the second term in Eq. (B.1) which is estimated by

$$\hat{\sigma}_0^2 = \frac{\sum_{i=1}^{n} y_i}{\sum_{i=1}^{n} s_i} \left[1/e_s^2 t + 1/e_B^2 n\bar{s} \right] \qquad \text{(B.2)}$$

which is used in the computation of L_D (Eq. (12) or (13)) when $n \geq 1$. When $C \geq 0$, an estimate of Eq. (B.1) is

$$\hat{\sigma}_D^2 = x/e_s^2 t^2 + \sum_{i=1}^{n} y_i \; / \; e_B^2 \left(\sum_{i=1}^{n} s_i \right)^2$$

which is used in computing confidence intervals on C (Eq. (6) for $n = 1$; Eq. (17) for $n > 1$).

1.2 Derivation of the "Detection Limit" L_D (Eqs. (12) and (13))

From Currie (1968: p. 589) and Figure 1 we have that

$$L_D = L_C + k_\beta \sigma_D \ .$$

Using Eq. (B.1), L_D in units of dpm may be written as

$$L_D = L_C + k_\beta (L_D/e_s + L_C^2/k_\alpha^2)^{1/2} \qquad \text{(B.3)}$$

or

$$(L_D - L_C)^2 = k_\beta^2 \ (L_D/e_s + L_C^2/k_\alpha^2) \ .$$

Expanding the left hand side and combining terms, we find

$$L_D^2 - L_D \left(2L_C + \frac{k_\beta^2}{e_s}\right) - \left(\frac{k_\beta^2}{k_\alpha^2} L_C^2 - L_C^2\right) = 0 \ .$$

Using the quadratic formula to solve for L_D gives

$$L_D = L_C + \frac{1}{2}\left\{\frac{k_\beta^2}{e_s} \pm \left[\left(4L_C^2 + \frac{4L_C k_\beta^2}{e_s} + \frac{k_\beta^4}{e_s^2}\right) + 4L_C^2\left(\frac{k_\alpha^2}{k_\beta^2} - 1\right)\right]^{1/2}\right\}$$

$$= L_C + \frac{k_\beta^2}{2e_s}\left[1 + \left(\frac{4L_C e_s}{k_\beta^2} + \frac{4L_C^2 e_s^2}{k_\alpha^2 k_\beta^2}\right)^{1/2}\right]$$

when the negative sign is ignored. When $k_\alpha = k_\beta = k$, this reduces to

$$L_D = L_C + \frac{k^2}{2e_s} + \frac{k^2}{2e_s}\left(1 + \frac{4L_C e_s}{k_\beta^2} + \frac{4L_C^2 e_s^2}{k_\alpha^2 k_\beta^2}\right)^{1/2}$$

$$= L_C + \frac{k^2}{2e_s} + \left[\left(\frac{k^2}{2e_s} + L_C\right)\left(\frac{k^2}{2e_s} + L_C\right)\right]^{1/2}$$

$$= 2L_C + \frac{k^2}{e_s} \ .$$

1.3 Derivation of σ_o on Eq. (16)

We want to test $H_o: C = 0$ using a variance estimate of $\tilde{c}$ which makes use of $x, y_1, \ldots, y_n$ when $C = 0$, where $\tilde{c} = x/te_s - \sum_{i=1}^{n} y_i \,/\, e_B \sum_{i=1}^{n} s_i$. When $C = 0$, x/te_s and each $y_i/s_i e_B$ estimate the true background disintegration rate B, i.e.,

$$E(x/te_s) = E(y_i/s_i e_B) = B \text{ for all } i.$$

Also,

$$\text{Var}(x/te_s) = Bte_s/t^2 e_s^2 = B/te_s$$

$$\text{Var}(y_i/s_i e_B) = Bs_i e_B/s_i^2 e_B^2 = B/s_i e_B \quad .$$

Following Nicholson (1963: p. 19), the minimum variance, unbiased, linear estimate of B is given by the weighted mean

$$\hat{B} = \frac{\omega_x (x/te_s) + \sum_{i=1}^{n} \omega_i (y_i/s_i e_B)}{\omega_x + \sum_{i=1}^{n} \omega_i} \quad , \qquad \text{(B.4)}$$

where the weights ω are the inverse of the variances of x/te_s and the $y_i/s_i e_B$, $i = 1, 2, \ldots, n$. Substituting into Eq. (B.4) we find

$$\hat{B} = \frac{\frac{x/t}{B/t} + \sum_{i=1}^{n} \frac{y_i/s_i}{B/s_i}}{\frac{1}{B/t} + \sum_{i=1}^{n} \frac{1}{B/s_i}}$$

$$= \left(x + \sum_{i=1}^{n} y_i\right) \Big/ \left(t + \sum_{i=1}^{n} s_i\right) \quad .$$

Substituting $\hat{B}$ into the second term of Eq. (B.1), which is $\text{Var}(\tilde{c})$ when $C = 0$, gives

$$\hat{\sigma}_o = \left[\frac{\left(x + \sum_{i=1}^{n} y_i \right)}{\left(t + \sum_{i=1}^{n} s_i \right)} \left(\frac{1}{e_s^2 t} + \frac{1}{e_B^2 n \bar{s}} \right) \right]^{1/2}$$

which is used in Eq. (16).

2.0 "Base Line" Correction for Background

2.1 Derivation of general variance formulae for σ_O^2 and σ_D^2

Eq. (19) is

$$\tilde{c} = \frac{1}{te} \left[\sum_{i=1}^{n_1} x_i - (n_1/n_2) \sum_{i=1}^{n_2} y_i \right] \quad . \tag{B.5}$$

Therefore,

$$\mathrm{Var}(\tilde{c}) = \frac{1}{(te)^2} \left[\sum_{i=1}^{n_1} \mathrm{Var}(x_i) + \left(\frac{n_1}{n_2} \right)^2 \sum_{i=1}^{n_2} \mathrm{Var}(y_i) \right]$$

$$= \frac{1}{(te)^2} \left[E\left(\sum_{i=1}^{n_1} x_i \right) + \left(\frac{n_1}{n_2} \right)^2 E\left(\sum_{i=1}^{n_2} y_i \right) \right]$$

$$= \frac{1}{(te)^2} \left[(C + n_1 B)te + \left(\frac{n_1}{n_2} \right)^2 (n_2 B)te \right]$$

$$= \frac{1}{te} \left[C + n_1 B (1 + n_1/n_2) \right] \tag{B.6}$$

When C = 0, then

$$\mathrm{Var}(\tilde{c}) = \sigma_o^2 = (n_1 B/te) \quad (1 + n_1/n_2) \tag{B.7}$$

which is estimated by

$$\hat{\sigma}_o^2 = \left(n_1 \sum_{i=1}^{n_2} y_i \; / \; t^2 e^2 n_2 \right) (1 + n_1/n_2) \quad .$$

This $\hat{\sigma}_o^2$ is used to compute L_C (Eq. (23)) which is used in the computation of L_D (Eq. (12) or (13)).

When $C \geq 0$, Eq. (B.6) is estimated by substituting Eq. (B.5) for C and and $\sum_{i=1}^{n_2} y_i/tn_2$ **e** for B, resulting in

$$\hat{\sigma}_D^2 = \sum_{i=1}^{n_1} x_i/t^2e^2 + (n_1/n_2)^2 \sum_{i=1}^{n_2} y_i/t^2e^2$$

which is used in computing confidence intervals on C (Eq. (21)).

2.2 Derivation of σ_o in Eq. (20)

We wish to test H_o:C = 0 using a variance estimate of $\tilde{c}$ that makes optimal use of the x_i and y_i in Eq. (B.5). When C = 0 we have

$$E\left(\sum_{i=1}^{n_1} x_i/t\right) = (1/t) \sum_{i=1}^{n_1} tB = n_1 B$$

$$E\left(\frac{n_1}{tn_2} \sum_{i=1}^{n_2} y_i\right) = (n_1/tn_2) \sum_{i=1}^{n_2} tB = n_1 B$$

$$\mathrm{Var}\left[\sum_{i=1}^{n_1} x_i/t\right] = (1/t^2) \sum_{i=1}^{n_1} E(x_i) = n_1 B/t$$

$$\mathrm{Var}\left[\frac{n_1}{tn_2} \sum_{i=1}^{n_2} y_i\right] = (n_1/tn_2)^2 \sum_{i=1}^{n_2} E(y_i) = n_1^2 B/tn_2 \quad .$$

Following Nicholson (1963:p. 19) the minimum variance, unbiased, linear estimate of $n_1 B$ is

$$n_1 \hat{B} = \frac{\left(\dfrac{\sum_{i=1}^{n_1} x_i/t}{n_1 B/t} + \dfrac{\dfrac{n_1}{tn_2} \sum_{i=1}^{n_2} y_i}{n_1^2 B/tn_2} \right)}{\left(t/n_1 B + tn_2/n_1^2 B \right)}$$

$$= \frac{n_1 \left(\sum_{i=1}^{n_1} x_i + \sum_{i=1}^{n_2} y_i \right)}{t\,(n_1 + n_2)} \quad .$$

Substituting $n_1 \hat{B}$ into Eq. (B.7) gives the $\hat{\sigma}_o^2$ used in Eq. (20).

APPENDIX C

PROPAGATION OF ERRORS IN ALPHA SPECTROMETRY

APPENDIX C

1. Notation

$\tilde{c}$ = estimated disintegration rate (dpm) of ^{239}Pu from the sample (after correction for tracer and yield)

x = total (gross) counts of ^{239}Pu in the sample (includes ^{239}Pu contamination from the tracer).

w = total (gross) counts of ^{242}Pu tracer recovered in the sample.

z = total (gross) counts of ^{242}Pu tracer added to the sample

v = total (gross) counts of ^{239}Pu contamination in the ^{242}Pu tracer.

b_x, b_y, b_z, b_v = background counts for x, y, z and v, respectively.

t = count time for x and w

s = count time for b_x and b_w

t_T = count time for z and v (T stands for "tracer")

s_T = count time for b_z and b_v.

e_s = efficiency of counter on which x, w, b_x and b_w are determined

e_T = efficiency of counter on which z, v, b_z and b_v are determined.

2. Estimation of $\tilde{c}$ and its Approximate Counting Error for Calculating Approximate Confidence Intervals

Case 1: (most general)

$$\tilde{c}_1 = \frac{\left(\frac{x}{te_s} - \frac{b_x}{se_s}\right) - \left(\frac{\frac{v}{t_Te_T} - \frac{b_v}{s_Te_T}}{\frac{z}{t_Te_T} - \frac{b_z}{s_Te_T}}\right)\left(\frac{w}{te_s} - \frac{b_w}{se_s}\right)}{\left(\frac{\frac{w}{te_s} - \frac{b_w}{se_s}}{\frac{z}{t_Te_T} - \frac{b_z}{s_Te_T}}\right)} \qquad (C.1)$$

$$= \frac{1}{e_T}\left[\frac{\left(\frac{x}{t} - \frac{b_x}{s}\right)\left(\frac{z}{t_T} - \frac{b_z}{s_T}\right)}{\left(\frac{w}{t} - \frac{b_w}{s}\right)} - \left(\frac{v}{t_T} - \frac{b_v}{s_T}\right)\right] \quad , \qquad (C.2)$$

where the second term in the numerator of Eq. (C.1) is the amount (expressed as a rate) of ^{239}Pu contamination in the ^{242}Pu tracer added to the sample, and the quantity in parentheses in the denominator of Eq. (C.1) is the "yield", i.e., the proportion of the total tracer added that is recovered.

Using the approximate "propagation of errors" formula (see, e.g., Hahn and Shapiro (1967:p. 231)):

$$\begin{aligned} \mathrm{Var}(\tilde{c}_1) \approx & \left(\frac{\partial \tilde{c}_1}{\partial x}\right)^2 \mathrm{Var}(x) + \left(\frac{\partial \tilde{c}_1}{\partial b_x}\right)^2 \mathrm{Var}(b_x) + \left(\frac{\partial \tilde{c}_1}{\partial w}\right)^2 \mathrm{Var}(w) \\ & + \left(\frac{\partial \tilde{c}_1}{\partial b_w}\right)^2 \mathrm{Var}(b_w) + \left(\frac{\partial \tilde{c}_1}{\partial v}\right)^2 \mathrm{Var}(v) + \left(\frac{\partial \tilde{c}_1}{\partial b_v}\right)^2 \mathrm{Var}(b_v) \\ & + \left(\frac{\partial \tilde{c}_1}{\partial z}\right)^2 \mathrm{Var}(z) + \left(\frac{\partial \tilde{c}_1}{\partial b_z}\right)^2 \mathrm{Var}(b_z) \end{aligned} \qquad (C.3)$$

we find that when counts are Poisson distributed, i.e., when Var(x) = x, Var(w) = w, etc.,

$$[\mathrm{Var}(\tilde{c}_1)]^{1/2} = \text{"1}\sigma \text{ counting error"}$$

$$\approx \frac{1}{e_T}\left[\frac{\left(\frac{z}{t_T}-\frac{b_z}{s_T}\right)^2}{\left(\frac{w}{t}-\frac{b_w}{s}\right)^2}\left(\frac{x}{t^2}+\frac{b_x}{s^2}\right)\right.$$

$$+\frac{\left(\frac{x}{t}-\frac{b_x}{s}\right)^2\left(\frac{z}{t_T}-\frac{b_z}{s_T}\right)^2}{\left(\frac{w}{t}-\frac{b_w}{s}\right)^4}\left(\frac{w}{t^2}+\frac{b_w}{s^2}\right) \qquad \text{(C.4)}$$

$$\left.+\left(\frac{v}{t_T^2}+\frac{b_v}{s_T^2}\right)+\frac{\left(\frac{x}{t}-\frac{b_x}{s}\right)^2}{\left(\frac{w}{t}-\frac{b_w}{s}\right)^2}\left(\frac{z}{t_T^2}+\frac{b_z}{s_T^2}\right)\right]^{1/2} .$$

Case 2: $(t = s,\ t_T = s_T)$

$$\tilde{c}_2 = \frac{1}{e_T t_T}\left[\frac{(x - b_x)(z - b_z)}{(w - b_w)} - (v - b_v)\right] , \qquad \text{(C.5)}$$

and

$$[\mathrm{Var}(\tilde{c}_2)]^{1/2} \approx \frac{1}{e_T t_T}\left[\frac{(z - b_z)^2}{(w - b_w)^2}(x + b_x)\right.$$

$$+\frac{(x - b_x)^2 (z - b_z)^2}{(w - b_w)^4}(w + b_w) \qquad \text{(C.6)}$$

$$\left.+(v + b_v)+\frac{(x - b_x)^2}{(w - b_w)^2}(z + b_z)\right]^{1/2} .$$

Case 3: $(t = s,\ t_T = s_T,\ v = 0,\ b_v = 0)$

$$\tilde{c}_3 = \frac{(x - b_x)(z - b_z)}{e_T t_T (w - b_w)} \quad , \tag{C.7}$$

and

$$[\mathrm{Var}(\tilde{c}_3)]^{1/2} \approx \tilde{c}_3 \left[\frac{(x + b_x)}{(x - b_x)^2} + \frac{(w + b_w)}{(w - b_w)^2} + \frac{(z + b_z)}{(z - b_z)^2}\right]^{1/2} . \tag{C.8}$$

We note that

(1) The efficiency e_s does not enter into these approximate counting error formulas.

(2) The formulas when $t = s = t_T = s_T$ can be obtained using Eqs. (C.5) and (C.6).

(3) The formulas when $(t = s,\ t_T \neq s_T)$ or $(t \neq s,\ t_T = s_T)$ can be obtained by substitution of the approximate counting times into Eqs. (C.2) and (C.4).

3. Approximate 1σ Counting Errors for Computing L_C to Make Approximate Tests and for Computing L_D.

We wish to determine whether the estimated dpm of ^{239}Pu (excluding any contamination of ^{239}Pu added with the tracer) is sufficiently large to indicate something above background is in the sample and to compute an approximate "detection limit" L_D using Eq. (12) or (13). To do this we need an estimate of the variance of this estimated disintegration rate when only background is present. This variance can be approximated using the "propagation of error" approach used above. Turning to Eq. (C.3) we recall that the variances in that approximate expression can be replaced by expected values since we are dealing with Poisson random variables.

Actually we need concern ourselves only with E(x) since this is the only expected value in Eq. (C.3) containing the "true" net ^{239}Pu count rate (exclusive of ^{239}Pu contamination). Setting this "true" rate to zero will then yield an approximate estimate of $Var(\tilde{c})$ for use in testing $H_o: C = 0$ or for computing L_D.

Let

C_x = "true" net count rate of ^{239}Pu including any ^{239}Pu contamination added with the ^{242}Pu tracer

B_x = "true" background count rate for ^{239}Pu

C_{x1} = "true" net count rate of ^{239}Pu that does not include ^{239}Pu contamination

C_v = "true" net count rate of ^{239}Pu added with the ^{242}Pu tracer

C_z = "true" net count rate of ^{242}Pu tracer added to the sample

C_w = "true" net count rate of ^{242}Pu tracer recovered in the sample.

Then

$$C_{x1} = C_x - \frac{C_v}{C_z} C_w \quad . \qquad (C.9)$$

Now, since

$$E(x) = (C_x + B_x)t$$

we have

$$E(x) = (C_{x1} + \frac{C_v}{C_z} C_w + B_x)t \quad . \qquad (C.10)$$

Setting $C_{x1} = 0$ we find

$$E(x) = \left(\frac{C_v}{C_z} C_w + B_x\right) t$$

which is estimated by

$$\left[\frac{\left(\frac{v}{t_T} - \frac{b_v}{s_T}\right)}{\left(\frac{z}{t_T} - \frac{b_z}{s_T}\right)}\left(\frac{w}{t} - \frac{b_w}{s}\right) + \frac{b_x}{s}\right] t \quad . \qquad \text{(C.11)}$$

Substituting this expression for Var(x) in Eq. (C.3) gives the following approximate equations for the standard deviations $\tilde{c}_1$, $\tilde{c}_2$ and $\tilde{c}_3$ for Case 1, 2 and 3, respectively, when $C_{x1} = 0$:

Case 1: (most general case)

$$\sigma_0 = [\mathrm{Var}(\tilde{c}_1)]^{1/2} \approx \frac{1}{e_T}\left\{\frac{\left(\frac{z}{t_T} - \frac{b_z}{s_T}\right)^2}{\left(\frac{w}{t} - \frac{b_w}{s}\right)^2}\left[\frac{\left(\frac{v}{t_T} - \frac{b_v}{s_T}\right)}{t\left(\frac{z}{t_T} - \frac{b_z}{s_T}\right)}\left(\frac{w}{t} - \frac{b_w}{s}\right) + \frac{b_x}{s}\left(\frac{1}{t} + \frac{1}{s}\right)\right] + T_2 + T_3 + T_4\right\}^{1/2} , \qquad \text{(C.12)}$$

where T_2, T_3 and T_4 are the 2nd, 3rd, and 4th terms, respectively, under the square root in Eq. (C.4).

Case 2: ($t = s$, $t_T = s_T$)

$$[\mathrm{Var}(\tilde{c}_2)]^{1/2} \approx \frac{1}{e_T t_T}\left[\frac{(z - b_z)}{(w - b_w)}\,\frac{(v - b_v)}{t} + 2b_x\frac{(z - b_z)^2}{(w - b_w)^2} + T_2 + T_3 + T_4\right]^{1/2} , \qquad \text{(C.13)}$$

where T_2, T_3 and T_4 are the 2nd, 3rd and 4th terms, respectively, under the square root in Eq. (C.6).

Case 3: ($t = s$, $t_T = s_T$, $v = 0$, $b_v = 0$)

$$[\mathrm{Var}(\tilde{c}_3)]^{1/2} \approx \tilde{c}_3 \left[\frac{2b_x}{(x - b_x)^2} + \frac{(w + b_w)}{(w - b_w)^2} + \frac{(z + b_z)}{(z - b_z)^2} \right]^{1/2} . \qquad (C.14)$$

Multiplying Eqs. (C.12), (C.13) or (C.14) (whichever is appropriate) by k_α will yield an approximate value of L_C which may be compared with $\tilde{c}_1$, $\tilde{c}_2$ or $\tilde{c}_3$ for an approximate test of $H_o: C_{x1} = 0$. (see Section (6) of the text for further discussion).

APPENDIX D

TABLES OF DECISION CRITERIA

APPENDIX D:

These tables are reproduced (with permission) from the AEC Research and Development Report HW-76279 entitled "Fixed Time Estimation of Counting Rates With Background Corrections" by W. L. Nicholson, September 1963, Hanford Laboratories, Richland, Washington.

TABLE D.1

EXACT SIGNIFICANCE LEVELS FOR DECISION RULES D_2 AND D_3

Stated Level of Significance: 1% ($k_\alpha = 2.326$)

Expected Total Background Contribution: (tB)

$\rho = t/s$		1	2	3	5	10	15	20	30	40	50
0	D_2	0.018	0.017	0.012	0.014	0.014	0.019	0.013	0.015	0.014	0.012
	D_3	0.018	0.017	0.012	0.014	0.014	0.019	0.013	0.015	0.014	0.012
1/10	D_2	0.042	0.029	0.025	0.022	0.018	0.017	0.016	0.014	0.014	0.013
	D_3	0.024	0.022	0.020	0.017	0.015	0.014	0.014	0.013	0.013	0.012
1/5	D_2	0.041	0.038	0.031	0.023	0.020	0.018	0.017	0.016	0.015	0.014
	D_3	0.019	0.017	0.018	0.017	0.014	0.013	0.013	0.012	0.012	0.012
1/3	D_2	0.094	0.051	0.036	0.029	0.021	0.020	0.018	0.017	0.016	0.015
	D_3	0.021	0.019	0.016	0.014	0.012	0.013	0.012	0.012	0.012	0.012
1/2	D_2	0.113	0.072	0.052	0.037	0.026	0.022	0.020	0.018	0.017	0.016
	D_3	0.012	0.013	0.013	0.013	0.012	0.012	0.011	0.011	0.011	0.011
1	D_2	0.234	0.133	0.083	0.058	0.036	0.031	0.027	0.022	0.020	0.019
	D_3	0.000	0.003	0.006	0.009	0.009	0.010	0.010	0.010	0.010	0.010
2	D_2	0.383	0.319	0.216	0.109	0.058	0.046	0.039	0.030	0.027	0.025
	D_3	0.000	0.000	0.000	0.001	0.005	0.006	0.006	0.007	0.008	0.008

TABLE D.1 (Continued)

EXACT SIGNIFICANCE LEVELS FOR DECISION RULES D_2 AND D_3

Stated Level of Significance: 5% ($k_\alpha = 1.645$)

Expected Total Background Contribution: (tB)

ρ = t/s		1	2	3	5	10	15	20	30	40	50
0	D_2	0.080	0.053	0.084	0.068	0.049	0.053	0.052	0.046	0.053	0.056
	D_3	0.080	0.053	0.084	0.068	0.049	0.053	0.052	0.046	0.053	0.056
1/10	D_2	0.086	0.076	0.071	0.066	0.061	0.060	0.059	0.057	0.056	0.055
	D_3	0.067	0.064	0.063	0.060	0.058	0.056	0.055	0.054	0.054	0.054
1/5	D_2	0.107	0.086	0.077	0.069	0.065	0.065	0.060	0.058	0.057	0.057
	D_3	0.069	0.062	0.059	0.057	0.054	0.055	0.055	0.054	0.053	0.053
1/3	D_2	0.111	0.095	0.084	0.078	0.069	0.064	0.062	0.060	0.059	0.058
	D_3	0.094	0.059	0.056	0.057	0.055	0.054	0.054	0.053	0.052	0.052
1/2	D_2	0.166	0.122	0.107	0.083	0.071	0.069	0.066	0.063	0.061	0.060
	D_3	0.063	0.070	0.062	0.053	0.052	0.053	0.052	0.053	0.052	0.052
1	D_2	0.240	0.160	0.122	0.098	0.087	0.079	0.073	0.070	0.068	0.065
	D_3	0.030	0.050	0.050	0.052	0.049	0.050	0.050	0.050	0.051	0.050
2	D_2	0.383	0.320	0.224	0.139	0.116	0.095	0.090	0.081	0.076	0.072
	D_3	0.000	0.006	0.019	0.038	0.040	0.042	0.046	0.047	0.046	0.047

TABLE D.1 (Continued)

EXACT SIGNIFICANCE LEVELS FOR DECISION RULES D_2 AND D_3

Stated Level of Significance: 10% ($k_\alpha = 1.282$)

Expected Total Background Contribution: (tB)

$\rho = t/s$		1	2	3	5	10	15	20	30	40	50
0	D_2	0.080	0.143	0.084	0.133	0.083	0.125	0.112	0.089	0.092	0.092
	D_3	0.080	0.143	0.084	0.133	0.083	0.125	0.112	0.089	0.092	0.092
1/10	D_2	0.117	0.120	0.113	0.111	0.108	0.107	0.106	0.105	0.105	0.104
	D_3	0.114	0.113	0.113	0.107	0.106	0.105	0.103	0.103	0.102	0.102
1/5	D_2	0.129	0.127	0.128	0.118	0.116	0.109	0.109	0.107	0.107	0.106
	D_3	0.116	0.112	0.106	0.106	0.104	0.104	0.103	0.102	0.102	0.102
1/3	D_2	0.154	0.127	0.118	0.124	0.115	0.115	0.111	0.110	0.108	0.108
	D_3	0.112	0.115	0.115	0.105	0.104	0.101	0.103	0.102	0.101	0.101
1/2	D_2	0.183	0.155	0.146	0.134	0.121	0.117	0.114	0.111	0.111	0.110
	D_3	0.116	0.102	0.108	0.108	0.104	0.101	0.102	0.101	0.101	0.101
1	D_2	0.263	0.220	0.185	0.148	0.133	0.125	0.123	0.119	0.116	0.114
	D_3	0.104	0.124	0.115	0.101	0.101	0.101	0.101	0.101	0.100	0.099
2	D_2	0.384	0.324	0.241	0.179	0.155	0.141	0.140	0.134	0.127	0.123
	D_3	0.012	0.053	0.083	0.091	0.093	0.097	0.097	0.097	0.098	0.099

TABLE D.2

1% LEVEL OF SIGNIFICANCE CRITICAL POINTS $X_{.01}$ AND RANDOMIZATION PROBABILITIES $P_{.01}$ FOR DECISION RULE D_e

ρ = Sample - Background Counting Time Ratio (t/s)

Total Count	10		5		3		2		1		1/2		1/3		1/5		1/10	
x + y	X	P	X	P	X	P	X	P	X	P	X	P	X	P	X	P	X	P
1	1	.011	1	.012	1	.013	1	.015	1	.020	1	.030	1	.040	1	.060	1	.110
2	2	.012	2	.014	2	.017	2	.022	2	.040	2	.090	2	.160	2	.359	1	.010
3	3	.013	3	.017	3	.023	3	.033	3	.080	3	.270	3	.640	2	.077	2	.410
4	4	.014	4	.020	4	.031	4	.050	4	.160	4	.810	3	.130	3	.597	2	.175
5	5	.016	5	.024	5	.042	5	.075	5	.320	4	.143	4	.616	3	.207	2	.055
6	6	.017	6	.029	6	.056	6	.113	6	.640	5	.524	4	.162	3	.024	3	.807
7	7	.019	7	.035	7	.074	7	.170	6	.040	5	.081	5	.750	4	.511	3	.450
8	8	.021	8	.042	8	.099	8	.256	7	.194	6	.434	5	.250	4	.206	3	.247
9	9	.023	9	.051	9	.133	9	.384	8	.457	6	.050	5	.000	4	.026	3	.114
10	10	.025	10	.061	10	.177	10	.576	9	.924	7	.405	6	.400	5	.580	3	.018
11	11	.028	11	.074	11	.236	11	.864	9	.154	7	.039	6	.091	5	.270	4	.709
12	12	.031	12	.089	12	.315	11	.049	10	.423	8	.412	7	.629	5	.072	4	.453
13	13	.034	13	.106	13	.420	12	.145	11	.870	8	.045	7	.233	6	.740	4	.276
14	14	.037	14	.128	14	.561	13	.274	11	.158	9	.445	8	.962	6	.393	4	.145
15	15	.041	15	.154	15	.748	14	.450	12	.454	9	.067	8	.443	6	.163	4	.044
16	16	.045	16	.184	16	.997	15	.696	13	.925	10	.500	8	.128	7	.991	5	.839
17	17	.050	17	.221	16	.058	15	.010	13	.200	10	.103	9	.743	7	.582	5	.581
18	18	.055	18	.266	17	.128	16	.124	14	.533	11	.578	9	.328	7	.304	5	.389
19	19	.061	19	.319	18	.215	17	.272	14	.017	11	.154	9	.055	7	.104	5	.241
20	20	.067	20	.383	19	.323	18	.468	15	.276	12	.678	10	.610	8	.850	5	.124
21	21	.074	21	.460	20	.457	19	.731	16	.659	12	.221	10	.251	8	.508	5	.028
22	22	.081	22	.552	21	.628	19	.026	16	.087	13	.802	10	.001	8	.260	6	.811
23	23	.089	23	.662	22	.844	20	.164	17	.389	13	.305	11	.524	8	.073	6	.586
24	24	.098	24	.795	22	.031	21	.341	18	.834	14	.952	11	.198	9	.790	6	.408
25	25	.108	25	.954	23	.118	22	.570	18	.187	14	.408	12	.900	9	.479	6	.265
26	26	.119	25	.027	24	.222	23	.872	19	.543	14	.051	12	.466	9	.245	6	.147
27	27	.131	26	.069	25	.349	23	.091	19	.025	15	.531	12	.160	9	.062	6	.048
28	8	.144	27	.115	26	.503	24	.260	20	.322	15	.137	13	.834	10	.777	7	[illegible]3
29	9	.158	28	.168	27	.694	25	.474	21	.742	16	.676	13	.429	10	.480	7	[illegible]1
30	30	.174	29	.229	28	.930	26	.750	21	.145	16	.239	13	.135	10	.251	7	[illegible]1

TABLE D.2 (Continued)

1% LEVEL OF SIGNIFICANCE CRITICAL POINTS $X_{.01}$
AND RANDOMIZATION PROBABILITIES $P_{.01}$ FOR DECISION RULE D_e

ρ = Sample - Background Counting Time Ratio (t/s)

Total Count	10		5		3		2		1		1/2		1/3		1/5		1/10	
x + y	X	P	X	P	X	P	X	P	X	P	X	P	X	P	X	P	X	P
31	31	.192	30	.298	28	.070	26	.040	22	.496	17	.846	14	.793	10	.068	7	.332
32	32	.211	31	.377	29	.178	27	.207	23	.995	17	.360	14	.407	11	.797	7	.207
33	33	.232	32	.470	30	.307	28	.415	23	.300	17	.019	14	.121	11	.504	7	.101
34	34	.255	33	.576	31	.461	29	.678	24	.717	18	.502	15	.772	11	.274	7	.008
35	35	.281	34	.701	32	.648	29	.006	24	.137	18	.123	15	.398	11	.088	8	.793
36	36	.309	35	.845	33	.875	30	.174	25	.495	19	.667	15	.115	12	.844	8	.601
37	37	.340	35	.004	33	.054	31	.381	26	.994	19	.243	16	.766	12	.549	8	.440
38	38	.374	36	.057	34	.170	32	.639	26	.312	20	.858	16	.398	12	.314	8	.303
39	39	.411	37	.116	35	.306	33	.965	27	.739	20	.383	16	.117	12	.122	8	.185
40	40	.452	38	.182	36	.467	33	.157	27	.155	20	.037	17	.774	13	.914	8	.083
41	41	.498	39	.257	37	.660	34	.366	28	.529	21	.545	17	.409	13	.612	9	.981
42	42	.548	40	.341	38	.891	35	.624	28	.019	21	.159	17	.126	13	.369	9	.770
43	43	.602	41	.437	38	.065	36	.947	29	.352	22	.732	18	.792	13	.169	9	.590
44	44	.663	42	.546	39	.191	36	.152	30	.798	22	.299	18	.427	13	.002	9	.437
45	45	.729	43	.671	40	.338	37	.366	30	.199	23	.948	18	.142	14	.693	9	.304
46	46	.802	44	.814	41	.510	38	.627	31	.594	23	.461	19	.821	14	.439	9	.189
47	47	.882	45	.978	42	.714	39	.951	31	.064	23	.098	19	.454	14	.230	9	.087
48	48	.971	45	.054	43	.957	39	.160	32	.419	24	.647	19	.164	14	.054	10	.992
49	48	.013	46	.122	43	.101	40	.379	33	.891	24	.240	20	.860	15	.793	10	.789
50	49	.035	47	.198	44	.239	41	.647	33	.266	25	.860	20	.488	15	.526	10	.614
51	50	.057	48	.282	45	.399	42	.976	34	.688	25	.402	20	.193	15	.304	10	.462
52	51	.081	49	.377	46	.586	42	.178	34	.130	25	.054	21	.908	15	.117	10	.330
53	52	.106	50	.483	47	.806	43	.406	35	.511	26	.589	21	.529	16	.911	10	.213
54	53	.133	51	.603	47	.029	44	.681	35	.008	26	.198	21	.227	16	.628	10	.110
55	54	.162	52	.739	48	.161	44	.008	36	.356	27	.802	22	.964	16	.393	10	.017
56	55	.193	53	.893	49	.313	45	.207	37	.810	27	.364	22	.578	16	.194	11	.841
57	56	.226	53	.025	50	.489	46	.443	37	.217	27	.024	22	.268	16	.023	11	.665
58	57	.262	54	.097	51	.694	47	.729	38	.629	28	.552	22	.015	17	.748	11	.512
59	58	.300	55	.176	52	.934	47	.038	38	.092	28	.172	23	.634	17	.497	11	.377
60	59	.341	56	.264	52	.098	48	.245	39	.470	29	.767	23	.316	17	.284	11	.257

TABLE D.2 (Continued)

5% LEVEL OF SIGNIFICANCE CRITICAL POINTS $X_{.05}$ AND RANDOMIZATION PROBABILITIES $P_{.05}$ FOR DECISION RULE D_e

ρ = Sample - Background Counting Time Ratio (t/s)

Total Count x + y	10		5		3		2		1		1/2		1/3		1/5		1/10	
	X	P	X	P	X	P	X	P	X	P	X	P	X	P	X	P	X	P
1	1	.055	1	.060	1	.066	1	.075	1	.100	1	.150	1	.200	1	.300	1	.550
2	2	.060	2	.072	2	.088	2	.112	2	.200	2	.450	2	.800	1	.079	1	.252
3	3	.066	3	.086	3	.118	3	.168	3	.400	2	.058	2	.244	2	.653	1	.118
4	4	.073	4	.103	4	.158	4	.253	4	.800	3	.381	3	.983	2	.291	1	.022
5	5	.080	5	.124	5	.210	5	.379	4	.120	3	.028	3	.391	2	.090	2	.700
6	6	.088	6	.149	6	.280	6	.569	5	.366	4	.390	3	.094	3	.770	2	.446
7	7	.097	7	.179	7	.374	7	.854	6	.771	4	.036	4	.643	3	.414	2	.279
8	8	.107	8	.214	8	.499	7	.070	6	.135	5	.444	4	.262	3	.185	2	.155
9	9	.117	9	.257	9	.665	8	.204	7	.433	5	.073	4	.009	3	.015	2	.055
10	10	.129	10	.309	10	.887	9	.376	8	.893	6	.533	5	.518	4	.636	3	.882
11	11	.142	11	.371	10	.050	10	.604	8	.214	6	.136	5	.195	4	.358	3	.633
12	12	.156	12	.445	11	.144	11	.914	9	.571	7	.655	6	.890	4	.153	3	.447
13	13	.172	13	.534	12	.254	11	.114	9	.044	7	.222	6	.459	5	.969	3	.301
14	14	.189	14	.641	13	.387	12	.289	10	.348	8	.810	6	.159	5	.619	3	.182
15	15	.208	15	.770	14	.548	13	.510	11	.778	8	.333	7	.831	5	.362	3	.080
16	16	.229	16	.924	15	.747	14	.794	11	.173	8	.000	7	.436	5	.161	4	.971
17	17	.252	16	.032	16	.997	14	.067	12	.539	9	.471	7	.146	6	.989	4	.740
18	18	.278	17	.091	16	.109	15	.251	12	.026	9	.103	8	.815	6	.657	4	.555
19	19	.305	18	.157	17	.236	16	.475	13	.352	10	.637	8	.436	6	.403	4	.403
20	20	.336	19	.229	18	.383	17	.756	14	.792	10	.227	8	.149	6	.198	4	.275
21	21	.370	20	.309	19	.558	17	.051	14	.195	11	.834	9	.826	6	.027	4	.164
22	22	.407	21	.400	20	.767	18	.244	15	.584	11	.375	9	.454	7	.734	4	.065
23	23	.447	22	.502	20	.009	19	.477	15	.058	11	.033	9	.166	7	.473	5	.940
24	24	.492	23	.619	21	.135	20	.762	16	.411	12	.549	10	.860	7	.261	5	.739
25	25	.541	24	.753	22	.279	20	.057	17	.879	12	.168	10	.488	7	.081	5	.570
26	26	.595	25	.908	23	.444	21	.262	17	.262	13	.753	10	.195	8	.843	5	.426
27	27	.655	25	.033	24	.638	22	.505	18	.683	13	.326	11	.910	8	.571	5	.300
28	28	.721	26	.108	25	.866	23	.801	18	.131	14	.989	11	.534	8	.346	5	.189
29	29	.793	27	.190	25	.064	23	.083	19	.516	14	.509	11	.235	8	.156	5	9
30	30	.872	28	.279	26	.207	24	.300	19	.012	14	.140	12	.977	9	.931	6	7

TABLE D.2 (Continued)

5% LEVEL OF SIGNIFICANCE CRITICAL POINTS $X_{.05}$
AND RANDOMIZATION PROBABILITIES $P_{.05}$ FOR DECISION RULE D_e

ρ = Sample - Background Counting Time Ratio (t/s)

Total Count x + y	10		5		3		2		1		1/2		1/3		1/5		1/10	
	X	P	X	P	X	P	X	P	X	P	X	P	X	P	X	P	X	P
31	31	.959	29	.378	27	.370	25	.557	20	.370	15	.720	12	.594	9	.694	6	.804
32	31	.017	30	.488	28	.556	26	.866	21	.830	15	.308	12	.286	9	.454	6	.637
33	32	.048	31	.611	29	.773	26	.127	21	.240	16	.963	12	.031	9	.251	6	.492
34	33	.081	32	.749	29	.014	27	.357	22	.664	16	.500	13	.665	9	.074	6	.363
35	34	.115	33	.904	30	.158	28	.628	22	.121	16	.136	13	.348	10	.842	6	.248
36	35	.151	33	.035	31	.321	29	.954	23	.516	17	.720	13	.083	10	.585	6	.144
37	36	.189	34	.120	32	.505	29	.186	23	.012	17	.314	14	.748	10	.367	6	.049
38	37	.229	35	.212	33	.717	30	.431	24	.384	18	.972	14	.419	10	.177	7	.915
39	38	.271	36	.311	34	.961	31	.719	25	.851	18	.516	14	.145	10	.009	7	.744
40	39	.315	37	.421	34	.127	31	.035	25	.263	18	.151	15	.842	11	.740	7	.592
41	40	.363	38	.541	35	.290	32	.261	26	.700	19	.746	15	.501	11	.503	7	.457
42	41	.413	39	.674	36	.475	33	.522	26	.152	19	.339	15	.215	11	.298	7	.336
43	42	.468	40	.823	37	.684	34	.828	27	.564	19	.005	16	.948	11	.117	7	.226
44	43	.525	41	.988	38	.925	34	.110	27	.049	20	.552	16	.592	12	.920	7	.125
45	44	.587	41	.082	38	.108	35	.351	28	.439	20	.182	16	.294	12	.663	7	.031
46	45	.654	42	.177	39	.275	36	.629	29	.923	21	.793	16	.038	12	.440	8	.886
47	46	.725	43	.280	40	.461	37	.956	29	.324	21	.382	17	.693	12	.244	8	.726
48	47	.802	44	.391	41	.671	37	.199	30	.780	21	.041	17	.382	12	.070	8	.582
49	48	.885	45	.513	42	.910	38	.456	30	.217	22	.606	17	.115	13	.846	8	.452
50	49	.974	46	.646	42	.102	39	.753	31	.649	22	.229	18	.805	13	.603	8	.334
51	49	.028	47	.793	43	.272	39	.061	31	.117	23	.859	18	.479	13	.390	8	.225
52	50	.068	48	.955	44	.460	40	.301	32	.529	23	.440	18	.200	13	.201	8	.125
53	51	.109	48	.069	45	.672	41	.576	32	.023	23	.090	19	.928	13	.031	8	.032
54	52	.153	49	.168	46	.912	42	.894	33	.417	24	.676	19	.586	14	.789	9	.892
55	53	.198	50	.274	46	.106	42	.161	34	.897	24	.289	19	.293	14	.557	9	.737
56	54	.246	51	.389	47	.279	43	.418	34	.312	25	.941	19	.038	14	.351	9	.596
57	55	.297	52	.513	48	.471	44	.712	35	.769	25	.512	20	.702	14	.167	9	.468
58	56	.350	53	.649	49	.686	44	.033	35	.213	25	.152	20	.395	14	.000	9	.350
59	57	.405	54	.798	50	.928	45	.275	36	.650	26	.761	20	.128	15	.746	9	.242
60	58	.464	55	.961	50	.118	46	.550	36	.120	26	.363	21	.829	15	.522	9	.141

TABLE D.2 (Continued)

10% LEVEL OF SIGNIFICANCE CRITICAL POINTS $X_{.10}$ AND RANDOMIZATION PROBABILITIES $P_{.10}$ FOR DECISION RULE D_e

ρ = Sample - Background Counting Time Ratio (t/s)

Total Count x + y	10		5		3		2		1		1/2		1/3		1/5		1/10	
	X	P	X	P	X	P	X	P	X	P	X	P	X	P	X	P	X	P
1	1	.110	1	.120	1	.133	1	.150	1	.200	1	.300	1	.400	1	.599	0	.010
2	2	.121	2	.144	2	.177	2	.225	2	.400	2	.900	1	.100	1	.259	1	.554
3	3	.133	3	.172	3	.237	3	.337	3	.800	2	.283	2	.600	1	.074	1	.340
4	4	.146	4	.207	4	.316	4	.506	3	.150	3	.887	2	.233	2	.723	1	.205
5	5	.161	5	.248	5	.421	5	.759	4	.440	3	.332	3	.960	2	.401	1	.101
6	6	.177	6	.298	6	.561	5	.046	5	.900	4	.998	3	.473	2	.187	1	.009
7	7	.194	7	.358	7	.749	6	.202	5	.228	4	.427	3	.170	2	.018	2	.743
8	8	.214	8	.429	8	.998	7	.390	6	.592	4	.070	4	.840	3	.665	2	.538
9	9	.235	9	.515	8	.110	8	.632	6	.061	5	.562	4	.437	3	.399	2	.382
10	10	.259	10	.619	9	.232	9	.953	7	.386	5	.171	4	.149	3	.195	2	.256
11	11	.285	11	.743	10	.373	9	.156	8	.835	6	.735	5	.817	3	.024	2	.149
12	12	.313	12	.891	11	.539	10	.362	8	.223	6	.301	5	.441	4	.716	2	.053
13	13	.345	12	.026	12	.740	11	.613	9	.617	7	.948	5	.157	4	.456	3	.905
14	14	.379	13	.101	13	.988	12	.931	9	.083	7	.461	6	.841	4	.248	3	.703
15	15	.417	14	.180	13	.127	12	.158	10	.444	7	.102	6	.472	4	.072	3	.539
16	16	.459	15	.265	14	.273	13	.381	11	.924	8	.653	6	.185	5	.822	3	.400
17	17	.505	16	.358	15	.439	14	.647	11	.299	8	.254	7	.894	5	.555	3	.280
18	18	.555	17	.461	16	.631	15	.975	12	.732	9	.881	7	.524	5	.337	3	.174
19	19	.611	18	.577	17	.858	15	.195	12	.171	9	.432	7	.231	5	.150	3	.077
20	20	.672	19	.708	17	.065	16	.434	13	.572	9	.082	8	.970	6	.971	4	.969
21	21	.740	20	.857	18	.217	17	.717	13	.055	10	.641	8	.594	6	.689	4	.782
22	22	.814	20	.013	19	.386	17	.034	14	.434	10	.251	8	.292	6	.456	4	.622
23	23	.895	21	.101	20	.578	18	.258	15	.914	11	.883	8	.036	6	.257	4	.482
24	24	.984	22	.194	21	.800	19	.515	15	.310	11	.466	9	.680	6	.081	4	.359
25	24	.033	23	.294	21	.032	20	.818	16	.757	11	.095	9	.367	7	.856	4	.248
26	25	.073	24	.403	22	.190	20	.104	16	.198	12	.670	9	.102	7	.605	4	.146
27	26	.115	25	.522	23	.364	21	.344	17	.619	12	.280	10	.780	7	.389	4	.051
28	7	.157	26	.653	24	.559	22	.620	17	.094	13	.926	10	.454	7	.200	5	[illegible]3
29	8	.202	27	.799	25	.780	23	.945	18	.496	13	.491	10	.179	7	.031	5	7
30	29	.248	28	.961	25	.020	23	.194	19	.995	13	.134	11	.895	8	.783	5	.011

TABLE D.2 (Continued)

10% LEVEL OF SIGNIFICANCE CRITICAL POINTS $X_{.10}$ AND RANDOMIZATION PROBABILITIES $P_{.10}$ FOR DECISION RULE D_e

ρ = Sample - Background Counting Time Ratio (t/s)

Total Count	10		5		3		2		1		1/2		1/3		1/5		1/10	
x + y	X	P	X	P	X	P	X	P	X	P	X	P	X	P	X	P	X	P
31	30	.296	28	.074	26	.184	24	.452	19	.383	14	.730	11	.555	8	.548	5	.480
32	31	.347	29	.174	27	.363	25	.748	20	.854	14	.334	11	.267	8	.343	5	.361
33	32	.400	30	.281	28	.562	25	.059	20	.279	14	.001	11	.015	8	.161	5	.252
34	33	.457	31	.396	29	.786	26	.303	21	.727	15	.560	12	.668	9	.991	5	.151
35	34	.517	32	.520	29	.025	27	.579	21	.181	15	.194	12	.367	9	.735	5	.057
36	35	.580	33	.656	30	.194	28	.898	22	.610	16	.815	12	.104	9	.511	6	.937
37	36	.648	34	.804	31	.379	28	.169	22	.089	16	.410	13	.794	9	.313	6	.780
38	37	.721	35	.968	32	.584	29	.430	23	.503	16	.066	13	.478	9	.134	6	.638
39	38	.798	35	.083	33	.812	30	.727	23	.002	17	.651	13	.202	10	.950	6	.510
40	39	.881	36	.189	33	.044	30	.045	24	.403	17	.274	14	.933	10	.705	6	.391
41	40	.970	37	.303	34	.219	31	.295	25	.882	18	.922	14	.600	10	.488	6	.282
42	40	.032	38	.424	35	.410	32	.576	25	.309	18	.504	14	.311	10	.294	6	.180
43	41	.080	39	.554	36	.621	33	.895	26	.769	18	.149	14	.055	10	.117	6	.085
44	42	.129	40	.696	37	.855	33	.171	26	.220	19	.761	15	.734	11	.926	7	.989
45	43	.180	41	.849	37	.074	34	.439	27	.664	19	.371	15	.430	11	.688	7	.834
46	44	.233	41	.010	38	.256	35	.740	27	.136	19	.033	15	.162	11	.476	7	.693
47	45	.288	42	.118	39	.453	35	.056	28	.566	20	.617	16	.880	11	.284	7	.564
48	46	.345	43	.231	40	.671	36	.313	28	.055	20	.248	16	.560	11	.109	7	.444
49	47	.405	44	.350	41	.911	37	.600	29	.473	21	.890	16	.278	12	.915	7	.333
50	48	.468	45	.479	41	.116	38	.924	30	.968	21	.485	16	.026	12	.683	7	.229
51	49	.534	46	.616	42	.304	38	.196	30	.385	21	.133	17	.701	12	.474	7	.131
52	50	.603	47	.764	43	.508	39	.471	31	.863	22	.746	17	.405	12	.283	7	.038
53	51	.676	48	.925	44	.733	40	.779	31	.302	22	.362	17	.140	12	.108	8	.913
54	52	.753	48	.061	45	.981	40	.087	32	.765	22	.026	18	.854	13	.916	8	.770
55	53	.835	49	.174	45	.168	41	.352	32	.221	23	.614	18	.541	13	.686	8	.639
56	54	.922	50	.294	46	.362	42	.646	33	.672	23	.248	18	.264	13	.479	8	.516
57	54	.007	51	.421	47	.574	43	.976	33	.145	24	.893	18	.012	13	.289	8	.402
58	55	.059	52	.556	48	.805	43	.240	34	.583	24	.492	19	.689	13	.114	8	.295
59	56	.113	53	.701	48	.041	44	.523	34	.071	24	.141	19	.397	14	.925	8	.194
60	57	.168	54	.858	49	.228	45	.839	35	.499	25	.760	19	.133	14	.698	8	.099

TABLE D.3 CONFIDENCE INTERVAL END POINTS L AND U FOR CONFIDENCE COEFFICIENT .95 AND SAMPLE-BACKGROUND COUNTING TIME RATIO 1

TOTAL SAMPLE COUNT	TOTAL BACKGROUND COUNT -- Y									
	0		1		2		3		4	
X	L	U	L	U	L	U	L	U	L	U
0	0	0	0	3.01	0	2.18	0	1.36	0	0.53
1	0	5.67	0	4.83	0	4.00	0	3.17	0	2.34
2	0	7.29	0	6.46	0	5.62	0	4.78	0	3.94
3	0	8.82	0	7.98	0	7.14	0	6.29	0	5.45
4	0	10.29	0	9.44	0	8.59	0	7.74	0	6.90
5	0.30	11.71	0	10.86	0	10.00	0	9.15	0	8.30
6	1.13	13.09	0	12.24	0	11.38	0	10.52	0	9.67
7	1.96	14.45	0.16	13.59	0	12.74	0	11.87	0	11.01
8	2.79	15.79	1.00	14.93	0	14.07	0	13.20	0	12.34
9	3.61	17.11	1.83	16.24	0.26	15.38	0	14.51	0	13.65
10	4.44	18.41	2.67	17.54	1.10	16.68	0	15.81	0	14.94
12	6.10	20.98	4.34	20.11	2.78	19.23	1.32	18.36	0	17.49
14	7.76	23.50	6.01	22.63	4.47	21.75	3.02	20.87	1.63	20.00
16	9.41	25.99	7.68	25.12	6.16	24.23	4.72	23.35	3.34	22.47
18	11.07	28.46	9.35	27.57	7.85	26.69	6.42	25.81	5.05	24.92
20	12.73	30.89	11.01	30.01	9.55	29.12	8.13	28.24	6.77	27.35
22	14.38	33.31	12.60	32.43	11.24	31.54	9.84	30.65	8.49	29.76
24	16.04	35.71	14.20	34.82	12.94	33.93	11.55	33.04	10.21	32.15
26	17.70	38.10	15.81	37.21	14.62	36.31	13.27	35.42	11.93	34.52
28	19.35	40.47	17.44	39.57	16.26	38.68	14.99	37.78	13.66	36.88
30	21.01	42.83	19.08	41.93	17.91	41.03	16.71	40.13	15.39	39.24
34	24.32	47.51	22.40	46.61	21.24	45.71	20.07	44.81	18.86	43.91
38	27.64	52.15	25.76	51.25	24.60	50.35	23.45	49.44	22.29	48.54
42	30.95	56.77	29.14	55.86	27.99	54.95	26.85	54.05	25.70	53.14
46	34.27	61.35	32.56	60.44	31.42	59.54	30.28	58.63	29.14	57.71
50	37.58	65.91	36.00	65.00	34.86	64.09	33.73	63.18	32.59	62.27
54	40.89	70.45	39.46	69.54	38.33	68.62	37.20	67.71	36.07	66.80
58	44.21	74.97	42.94	74.06	41.81	73.14	40.69	72.23	39.56	71.31
?	47.52	79.47	46.44	78.56	45.32	77.64	44.19	76.72	43.07	75.80
›	50.83	83.96	49.95	83.04	48.83	82.12	47.72	81.20	46.60	80.2

E D.3 (Cont.) CONFIDENCE INTERVAL END POINTS L AND U FOR CONFIDENCE COEFFICIENT .95 AND SAMPLE-BACKGROUND COUNTING TIME RATIO 1

TOTAL SAMPLE COUNT	TOTAL BACKGROUND COUNT -- Y									
	5		6		7		8		9	
X	L	U	L	U	L	U	L	U	L	U
0	0	0	0	0	0	0	0	0	0	0
1	0	1.50	0	0.67	0	0	0	0	0	0
2	0	3.10	0	2.26	0	1.42	0	0.58	0	0
3	0	4.60	0	3.76	0	2.91	0	2.07	0	1.22
4	0	6.04	0	5.19	0	4.34	0	3.49	0	2.64
5	0	7.44	0	6.59	0	5.73	0	4.88	0	4.02
6	0	8.81	0	7.95	0	7.09	0	6.24	0	5.38
7	0	10.15	0	9.29	0	8.43	0	7.57	0	6.70
8	0	11.47	0	10.61	0	9.74	0	8.88	0	8.01
9	0	12.78	0	11.91	0	11.04	0	10.18	0	9.30
10	0	14.07	0	13.20	0	12.33	0	11.46	0	10.59
12	0	16.61	0	15.74	0	14.86	0	13.99	0	13.11
14	0.27	19.12	0	18.24	0	17.36	0	16.48	0	15.60
16	1.99	21.59	0.66	20.71	0	19.83	0	18.94	0	18.06
18	3.71	24.04	2.39	23.15	1.09	22.26	0	21.38	0	20.49
20	5.43	26.46	4.12	25.57	2.83	24.68	1.55	23.80	0.29	22.91
22	7.16	28.87	5.86	27.98	4.57	27.09	3.30	26.19	2.04	25.30
24	8.89	31.25	7.59	30.36	6.31	29.47	5.05	28.58	3.79	27.68
26	10.62	33.63	9.33	32.73	8.06	31.84	6.80	30.94	5.55	30.04
28	12.36	35.99	11.07	35.09	9.81	34.19	8.55	33.30	7.30	32.40
30	14.10	38.34	12.82	37.44	11.55	36.54	10.30	35.64	9.06	34.74
34	17.58	43.00	16.31	42.10	15.06	41.20	13.82	40.30	12.59	39.39
38	21.07	47.63	19.82	46.73	18.58	45.82	17.35	44.91	16.13	44.01
42	24.56	52.23	23.34	51.32	22.11	50.42	20.89	49.51	19.67	48.60
46	28.00	56.81	26.86	55.89	25.64	54.98	24.43	54.07	23.22	53.16
50	31.46	61.35	30.33	60.44	29.19	59.53	27.98	58.61	26.78	57.70
54	34.94	65.88	33.81	64.97	32.69	64.05	31.54	63.14	30.35	62.22
58	38.44	70.39	37.32	69.47	36.19	68.56	35.07	67.64	33.93	66.72
62	41.95	74.88	40.83	73.97	39.72	73.05	38.60	72.13	37.48	71.21
66	45.48	79.36	44.36	78.44	43.25	77.52	42.14	76.60	41.02	75.68

TABLE D.3 (Cont.) CONFIDENCE INTERVAL END POINTS L AND U FOR CONFIDENCE COEFFICIENT .95 AND SAMPLE-BACKGROUND COUNTING TIME RATIO 1

TOTAL SAMPLE COUNT	TOTAL BACKGROUND COUNT -- Y									
	10		12		14		16		18	
X	L	U	L	U	L	U	L	U	L	U
0	0	0	0	0	0	0	0	0	0	0
1	0	0	0	0	0	0	0	0	0	0
2	0	0	0	0	0	0	0	0	0	0
3	0	0.37	0	0	0	0	0	0	0	0
4	0	1.79	0	0.08	0	0	0	0	0	0
5	0	3.17	0	1.45	0	0	0	0	0	0
6	0	4.52	0	2.79	0	1.07	0	0	0	0
7	0	5.84	0	4.11	0	2.38	0	0.65	0	0
8	0	7.14	0	5.41	0	3.67	0	1.93	0	0.19
9	0	8.44	0	6.70	0	4.95	0	3.21	0	1.46
10	0	9.71	0	7.97	0	6.22	0	4.47	0	2.72
12	0	12.24	0	10.48	0	8.72	0	6.97	0	5.20
14	0	14.72	0	12.96	0	11.20	0	9.43	0	7.66
16	0	17.17	0	15.41	0	13.64	0	11.86	0	10.09
18	0	19.61	0	17.83	0	16.06	0	14.28	0	12.50
20	0	22.02	0	20.24	0	18.46	0	16.67	0	14.89
22	0.79	24.41	0	22.62	0	20.84	0	19.05	0	17.26
24	2.55	26.78	0.09	24.99	0	23.20	0	21.41	0	19.62
26	4.31	29.15	1.86	27.36	0	25.56	0	23.76	0	21.97
28	6.07	31.50	3.62	29.70	1.20	27.90	0	26.10	0	24.30
30	7.83	33.84	5.40	32.04	2.98	30.24	0.59	28.43	0	26.63
34	11.37	38.49	8.94	36.68	6.55	34.87	4.16	33.06	1.80	31.25
38	14.91	43.10	12.50	41.29	10.11	39.47	7.75	37.66	5.39	35.84
42	18.46	47.69	16.07	45.87	13.69	44.05	11.33	42.23	8.99	40.40
46	22.02	52.25	19.64	50.42	17.27	48.60	14.92	46.77	12.59	44.95
50	25.59	56.79	23.22	54.96	20.86	53.13	18.52	51.30	16.19	49.47
54	29.16	61.30	26.80	59.47	24.46	57.64	22.12	55.80	19.81	53.97
58	32.75	65.80	30.40	63.97	28.06	62.13	25.74	60.29	23.42	58.46
62	36.34	70.29	33.99	68.45	31.67	66.61	29.35	64.77	27.05	62.93
66	39.91	74.76	37.60	72.92	35.29	71.07	32.98	69.23	30.68	67.39

TABLE D.3 (Cont.)

CONFIDENCE INTERVAL END POINTS L AND U FOR CONFIDENCE COEFFICIENT .95 AND SAMPLE-BACKGROUND COUNTING TIME RATIO 1

TOTAL SAMPLE COUNT	TOTAL BACKGROUND COUNT -- Y									
	20		22		24		26		28	
X	L	U	L	U	L	U	L	U	L	U
0	0	0	0	0	0	0	0	0	0	0
1	0	0	0	0	0	0	0	0	0	0
2	0	0	0	0	0	0	0	0	0	0
3	0	0	0	0	0	0	0	0	0	0
4	0	0	0	0	0	0	0	0	0	0
5	0	0	0	0	0	0	0	0	0	0
6	0	0	0	0	0	0	0	0	0	0
7	0	0	0	0	0	0	0	0	0	0
8	0	0	0	0	0	0	0	0	0	0
9	0	0	0	0	0	0	0	0	0	0
10	0	0.97	0	0	0	0	0	0	0	0
12	0	3.44	0	1.68	0	0	0	0	0	0
14	0	5.89	0	4.12	0	2.35	0	0.57	0	0
16	0	8.31	0	6.54	0	4.76	0	2.98	0	1.19
18	0	10.72	0	8.93	0	7.15	0	5.36	0	3.57
20	0	13.10	0	11.31	0	9.52	0	7.73	0	5.94
22	0	15.47	0	13.67	0	11.88	0	10.09	0	8.29
24	0	17.82	0	16.02	0	14.23	0	12.43	0	10.63
26	0	20.16	0	18.36	0	16.56	0	14.76	0	12.95
28	0	22.50	0	20.69	0	18.89	0	17.08	0	15.27
30	0	24.82	0	23.01	0	21.20	0	19.39	0	17.58
34	0	29.43	0	27.62	0	25.81	0	23.99	0	22.17
38	3.05	34.02	0.72	32.20	0	30.38	0	28.56	0	26.74
42	6.65	38.58	4.33	36.76	2.02	34.93	0	33.10	0	31.28
46	10.26	43.12	7.95	41.29	5.64	39.46	3.35	37.63	1.05	35.80
50	13.87	47.64	11.57	45.80	9.27	43.97	6.98	42.13	4.70	40.30
54	17.49	52.13	15.19	50.30	12.90	48.46	10.62	46.62	8.34	44.78
58	21.12	56.62	18.83	54.78	16.54	52.94	14.26	51.10	11.99	49.25
62	24.75	61.08	22.47	59.24	20.18	57.40	17.91	55.55	15.64	53.71
66	28.39	65.54	26.11	63.70	23.83	61.85	21.56	60.00	19.30	58.15

TABLE D.3 (Cont.) CONFIDENCE INTERVAL END POINTS L AND U FOR CONFIDENCE COEFFICIENT .95 AND SAMPLE-BACKGROUND COUNTING TIME RATIO 1/2

TOTAL SAMPLE COUNT	TOTAL BACKGROUND COUNT -- Y									
	0		1		2		3		4	
X	L	U	L	U	L	U	L	U	L	U
0	0	0	0	3.39	0	2.94	0	2.49	0	2.04
1	0	5.67	0	5.21	0	4.76	0	4.32	0	3.87
2	0	7.29	0	6.84	0	6.39	0	5.94	0	5.48
3	0.28	8.82	0	8.37	0	7.91	0	7.46	0	7.01
4	1.01	10.29	0.12	9.83	0	9.38	0	8.92	0	8.46
5	1.74	11.71	0.86	11.25	0.09	10.79	0	10.34	0	9.88
6	2.47	13.09	1.60	12.63	0.84	12.18	0.12	11.72	0	11.26
7	3.20	14.45	2.22	13.99	1.59	13.53	0.88	13.07	0.20	12.61
8	3.94	15.79	2.87	15.33	2.28	14.87	1.65	14.41	0.97	13.95
9	4.67	17.11	3.53	16.65	2.95	16.19	2.38	15.73	1.74	15.26
10	5.40	18.41	4.22	17.95	3.64	17.49	3.07	17.02	2.50	16.56
12	6.86	20.98	5.64	20.52	5.07	20.05	4.50	19.59	3.93	19.12
14	8.33	23.50	7.10	23.04	6.53	22.57	5.97	22.11	5.41	21.64
16	9.79	25.99	8.60	25.53	8.03	25.06	7.47	24.60	6.91	24.13
18	11.26	28.46	10.12	27.99	9.57	27.52	9.01	27.05	8.45	26.59
20	12.72	30.89	11.68	30.43	11.12	29.96	10.57	29.49	10.01	29.02
22	14.18	33.31	13.25	32.85	12.70	32.38	12.15	31.91	11.60	31.44
24	15.65	35.71	14.84	35.25	14.29	34.78	13.75	34.31	13.20	33.84
26	17.11	38.10	16.45	37.63	15.91	37.16	15.36	36.69	14.81	36.22
28	18.58	40.47	18.08	40.00	17.53	39.53	16.99	39.06	16.44	38.59
30	20.04	42.83	19.72	42.36	19.17	41.89	18.63	41.41	18.08	40.94
34	22.97	47.51	23.03	47.04	22.48	46.57	21.94	46.09	21.40	45.62
38	25.90	52.15	26.37	51.68	25.83	51.21	25.30	50.74	24.76	50.26
42	28.83	56.77	29.76	56.29	29.22	55.82	28.68	55.35	28.14	54.87
46	31.75	61.35	33.17	60.88	32.63	60.40	32.09	59.93	31.56	59.45
50	34.68	65.91	36.60	65.44	36.07	64.96	35.53	64.49	35.00	64.01
54	37.61	70.45	40.06	69.98	39.53	69.50	38.99	69.02	38.46	68.55
58	40.54	74.97	43.54	74.49	43.00	74.02	42.47	73.54	41.94	73.06
62	43.47	79.47	47.03	79.00	46.50	78.52	45.97	78.04	45.44	77.56
6	46.39	83.96	50.54	83.48	50.01	83.01	49.48	82.53	48.95	82.05

TABLE D.3 (Cont.) CONFIDENCE INTERVAL END POINTS L AND U FOR CONFIDENCE COEFFICIENT .95 AND SAMPLE-BACKGROUND COUNTING TIME RATIO 1/2

TOTAL SAMPLE COUNT	TOTAL BACKGROUND COUNT -- Y									
	5		6		7		8		9	
X	L	U	L	U	L	U	L	U	L	U
0	0	1.59	0	1.14	0	0.70	0	0.25	0	0
1	0	3.41	0	2.96	0	2.51	0	2.06	0	1.61
2	0	5.03	0	4.58	0	4.13	0	3.67	0	3.22
3	0	6.55	0	6.10	0	5.64	0	5.19	0	4.73
4	0	8.01	0	7.55	0	7.09	0	6.64	0	6.18
5	0	9.42	0	8.96	0	8.51	0	8.05	0	7.59
6	0	10.80	0	10.34	0	9.88	0	9.42	0	8.97
7	0	12.15	0	11.69	0	11.24	0	10.77	0	10.31
8	0.30	13.48	0	13.03	0	12.57	0	12.10	0	11.64
9	1.09	14.80	0.44	14.34	0	13.88	0	13.41	0	12.95
10	1.87	16.10	1.23	15.63	0.59	15.18	0	14.71	0	14.25
12	3.36	18.66	2.79	18.19	2.19	17.73	1.56	17.27	0.95	16.80
14	4.84	21.18	4.28	20.71	3.72	20.25	3.16	19.78	2.56	19.32
16	6.36	23.66	5.80	23.20	5.24	22.73	4.68	22.26	4.12	21.79
18	7.90	26.12	7.34	25.65	6.79	25.19	6.23	24.72	5.68	24.25
20	9.46	28.55	8.91	28.09	8.36	27.62	7.80	27.15	7.25	26.68
22	11.05	30.97	10.49	30.50	9.94	30.03	9.39	29.56	8.84	29.09
24	12.65	33.36	12.10	32.90	11.55	32.43	11.00	31.96	10.45	31.49
26	14.26	35.75	13.72	35.28	13.17	34.81	12.62	34.34	12.08	33.86
28	15.90	38.12	15.35	37.64	14.81	37.17	14.26	36.70	13.72	36.23
30	17.54	40.47	17.00	40.00	16.45	39.52	15.91	39.06	15.37	38.58
34	20.86	45.15	20.32	44.67	19.78	44.20	19.24	43.73	18.70	43.26
38	24.22	49.79	23.68	49.31	23.14	48.84	22.60	48.36	22.06	47.89
42	27.61	54.40	27.07	53.92	26.53	53.45	25.99	52.97	25.46	52.50
46	31.02	58.98	30.49	58.50	29.95	58.03	29.42	57.55	28.88	57.08
50	34.46	63.54	33.93	63.06	33.40	62.58	32.86	62.11	32.33	61.63
54	37.93	68.07	37.39	67.59	36.86	67.11	36.33	66.64	35.80	66.16
58	41.41	72.59	40.88	72.11	40.35	71.63	39.81	71.15	39.28	70.68
62	44.91	77.09	44.38	76.61	43.85	76.13	43.32	75.65	42.79	75.18
66	48.42	81.57	47.89	81.09	47.36	80.61	46.83	80.14	46.30	79.66

TABLE D.3 (Cont.)

CONFIDENCE INTERVAL END POINTS L AND U FOR CONFIDENCE COEFFICIENT .95 AND SAMPLE-BACKGROUND COUNTING TIME RATIO 1/2

TOTAL SAMPLE COUNT	TOTAL BACKGROUND COUNT -- Y									
	10		12		14		16		18	
X	L	U	L	U	L	U	L	U	L	U
0	0	0	0	0	0	0	0	0	0	0
1	0	1.16	0	0.26	0	0	0	0	0	0
2	0	2.77	0	1.86	0	0.96	0	0.05	0	0
3	0	4.28	0	3.37	0	2.46	0	1.55	0	0.64
4	0	5.73	0	4.81	0	3.90	0	2.99	0	2.07
5	0	7.13	0	6.22	0	5.30	0	4.38	0	3.47
6	0	8.51	0	7.59	0	6.67	0	5.75	0	4.83
7	0	9.85	0	8.93	0	8.02	0	7.09	0	6.17
8	0	11.18	0	10.26	0	9.34	0	8.41	0	7.49
9	0	12.49	0	11.57	0	10.64	0	9.72	0	8.80
10	0	13.79	0	12.86	0	11.94	0	11.01	0	10.08
12	0.34	16.34	0	15.41	0	14.48	0	13.55	0	12.62
14	1.96	18.85	0.75	17.92	0	16.99	0	16.06	0	15.13
16	3.57	21.33	2.39	20.40	1.21	19.46	0.04	18.53	0	17.60
18	5.12	23.78	4.01	22.85	2.87	21.91	1.70	20.97	0.54	20.04
20	6.70	26.21	5.60	25.27	4.49	24.34	3.37	23.40	2.21	22.46
22	8.29	28.62	7.20	27.68	6.10	26.75	5.00	25.81	3.89	24.87
24	9.91	31.02	8.81	30.08	7.72	29.13	6.62	28.20	5.53	27.25
26	11.53	33.39	10.44	32.45	9.35	31.51	8.26	30.57	7.17	29.63
28	13.17	35.76	12.08	34.82	11.00	33.87	9.91	32.93	8.82	31.99
30	14.82	38.11	13.74	37.17	12.65	36.22	11.57	35.28	10.48	34.33
34	18.15	42.78	17.07	41.84	15.99	40.89	14.91	39.95	13.83	39.00
38	21.52	47.42	20.45	46.47	19.37	45.52	18.29	44.57	17.22	43.63
42	24.92	52.02	23.85	51.07	22.78	50.12	21.70	49.17	20.63	48.22
46	28.35	56.60	27.28	55.65	26.21	54.70	25.14	53.75	24.07	52.80
50	31.79	61.15	30.73	60.20	29.66	59.25	28.59	58.30	27.53	57.34
54	35.26	65.69	34.20	64.73	33.13	63.78	32.07	62.83	31.00	61.87
58	38.75	70.20	37.69	69.25	36.63	68.29	35.56	67.34	34.50	66.38
62	42.25	74.70	41.19	73.74	40.13	72.78	39.07	71.83	38.01	70.87
[illegible]	45.77	79.18	44.72	78.22	43.66	77.27	42.60	76.31	41.54	75.35

TABLE D.3 (Cont.)

CONFIDENCE INTERVAL END POINTS L AND U FOR CONFIDENCE COEFFICIENT .95 AND SAMPLE-BACKGROUND COUNTING TIME RATIO 1/2

TOTAL SAMPLE COUNT	TOTAL BACKGROUND COUNT -- Y									
	20		22		24		26		28	
X	L	U	L	U	L	U	L	U	L	U
0	0	0	0	0	0	0	0	0	0	0
1	0	0	0	0	0	0	0	0	0	0
2	0	0	0	0	0	0	0	0	0	0
3	0	0	0	0	0	0	0	0	0	0
4	0	1.16	0	0.24	0	0	0	0	0	0
5	0	2.55	0	1.64	0	0.72	0	0	0	0
6	0	3.91	0	2.99	0	2.07	0	1.16	0	0.23
7	0	5.25	0	4.33	0	3.41	0	2.49	0	1.56
8	0	6.57	0	5.64	0	4.72	0	3.80	0	2.87
9	0	7.87	0	6.94	0	6.02	0	5.09	0	4.17
10	0	9.15	0	8.23	0	7.30	0	6.38	0	5.45
12	0	11.69	0	10.77	0	9.84	0	8.90	0	7.97
14	0	14.19	0	13.26	0	12.33	0	11.40	0	10.46
16	0	16.66	0	15.73	0	14.79	0	13.86	0	12.92
18	0	19.10	0	18.17	0	17.23	0	16.30	0	15.36
20	1.06	21.53	0	20.59	0	19.65	0	18.71	0	17.77
22	2.75	23.93	1.61	22.99	0.47	22.05	0	21.11	0	20.17
24	4.44	26.31	3.30	25.37	2.17	24.43	1.04	23.49	0	22.55
26	6.08	28.69	5.00	27.74	3.88	26.80	2.75	25.86	1.63	24.92
28	7.74	31.04	6.65	30.10	5.57	29.16	4.47	28.21	3.35	27.27
30	9.40	33.39	8.32	32.44	7.24	31.50	6.15	30.56	5.07	29.61
34	12.76	38.05	11.68	37.10	10.60	36.16	9.52	35.21	8.45	34.26
38	16.14	42.68	15.07	41.73	13.99	40.78	12.92	39.83	11.85	38.88
42	19.56	47.27	18.49	46.32	17.42	45.38	16.35	44.42	15.28	43.47
46	23.00	51.84	21.93	50.89	20.86	49.94	19.79	48.99	18.73	48.04
50	26.46	56.39	25.39	55.44	24.33	54.49	23.26	53.53	22.20	52.58
54	29.94	60.92	28.88	59.96	27.81	59.01	26.75	58.06	25.69	57.10
58	33.44	65.43	32.38	64.47	31.32	63.52	30.25	62.56	29.19	61.61
62	36.95	69.92	35.89	68.96	34.83	68.01	33.77	67.05	32.72	66.09
66	40.48	74.39	39.42	73.44	38.36	72.48	37.31	71.52	36.25	70.57

TABLE D.3 (Cont.) CONFIDENCE INTERVAL END POINTS L AND U FOR CONFIDENCE COEFFICIENT .95 AND SAMPLE-BACKGROUND COUNTING TIME RATIO 1/2

TOTAL SAMPLE COUNT	TOTAL BACKGROUND COUNT -- Y									
	30		34		38		42		46	
X	L	U	L	U	L	U	L	U	L	U
0	0	0	0	0	0	0	0	0	0	0
1	0	0	0	0	0	0	0	0	0	0
2	0	0	0	0	0	0	0	0	0	0
3	0	0	0	0	0	0	0	0	0	0
4	0	0	0	0	0	0	0	0	0	0
5	0	0	0	0	0	0	0	0	0	0
6	0	0	0	0	0	0	0	0	0	0
7	0	0.64	0	0	0	0	0	0	0	0
8	0	1.95	0	0.10	0	0	0	0	0	0
9	0	3.24	0	1.39	0	0	0	0	0	0
10	0	4.52	0	2.66	0	0.81	0	0	0	0
12	0	7.04	0	5.18	0	3.32	0	1.46	0	0
14	0	9.53	0	7.66	0	5.80	0	3.93	0	2.06
16	0	11.99	0	10.12	0	8.25	0	6.37	0	4.50
18	0	14.42	0	12.55	0	10.67	0	8.80	0	6.92
20	0	16.83	0	14.96	0	13.08	0	11.20	0	9.32
22	0	19.23	0	17.35	0	15.47	0	13.59	0	11.70
24	0	21.61	0	19.72	0	17.84	0	15.96	0	14.07
26	0.51	23.97	0	22.09	0	20.20	0	18.31	0	16.43
28	2.23	26.33	0.00	24.44	0	22.55	0	20.66	0	18.77
30	3.95	28.67	1.73	26.77	0	24.88	0	22.99	0	21.10
34	7.37	33.32	5.20	31.42	3.00	29.53	0.80	27.63	0	25.74
38	10.78	37.94	8.63	36.04	6.49	34.14	4.30	32.24	2.11	30.34
42	14.21	42.52	12.07	40.62	9.93	38.72	7.79	36.82	5.64	34.92
46	17.66	47.09	15.53	45.18	13.39	43.28	11.26	41.37	9.13	39.47
50	21.13	51.63	19.00	49.72	16.88	47.81	14.75	45.91	12.63	44.00
54	24.63	56.15	22.50	54.24	20.38	52.33	18.25	50.42	16.13	48.51
58	28.13	60.65	26.01	58.74	23.89	56.83	21.77	54.92	19.66	53.01
62	31.66	65.14	29.54	63.23	27.42	61.31	25.31	59.40	23.19	57.48
66	35.19	69.61	33.08	67.69	30.97	65.78	28.85	63.87	26.74	61.95

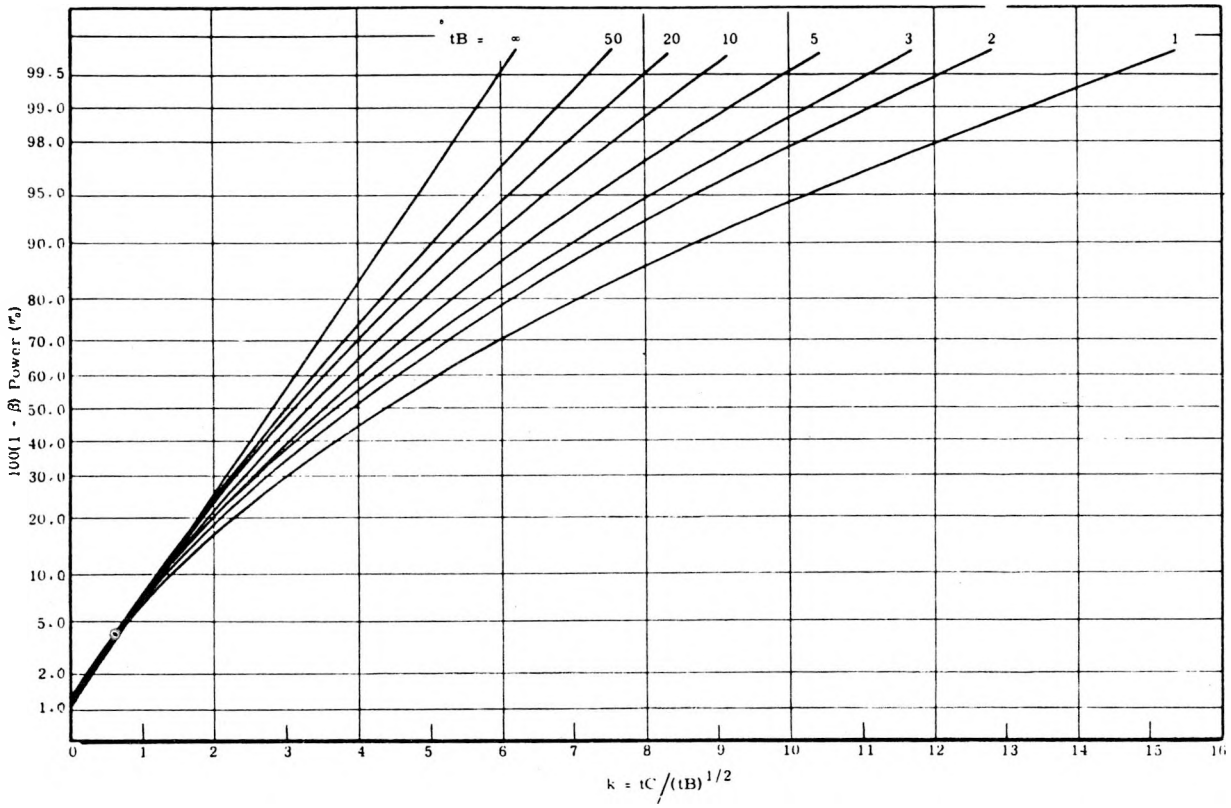

FIGURE D.1

Power of Rule D_3 (Using Eq. (4)) to Detect

$C > 0$ When $\alpha = 0.01$ and $\rho = t/s = 1/2$

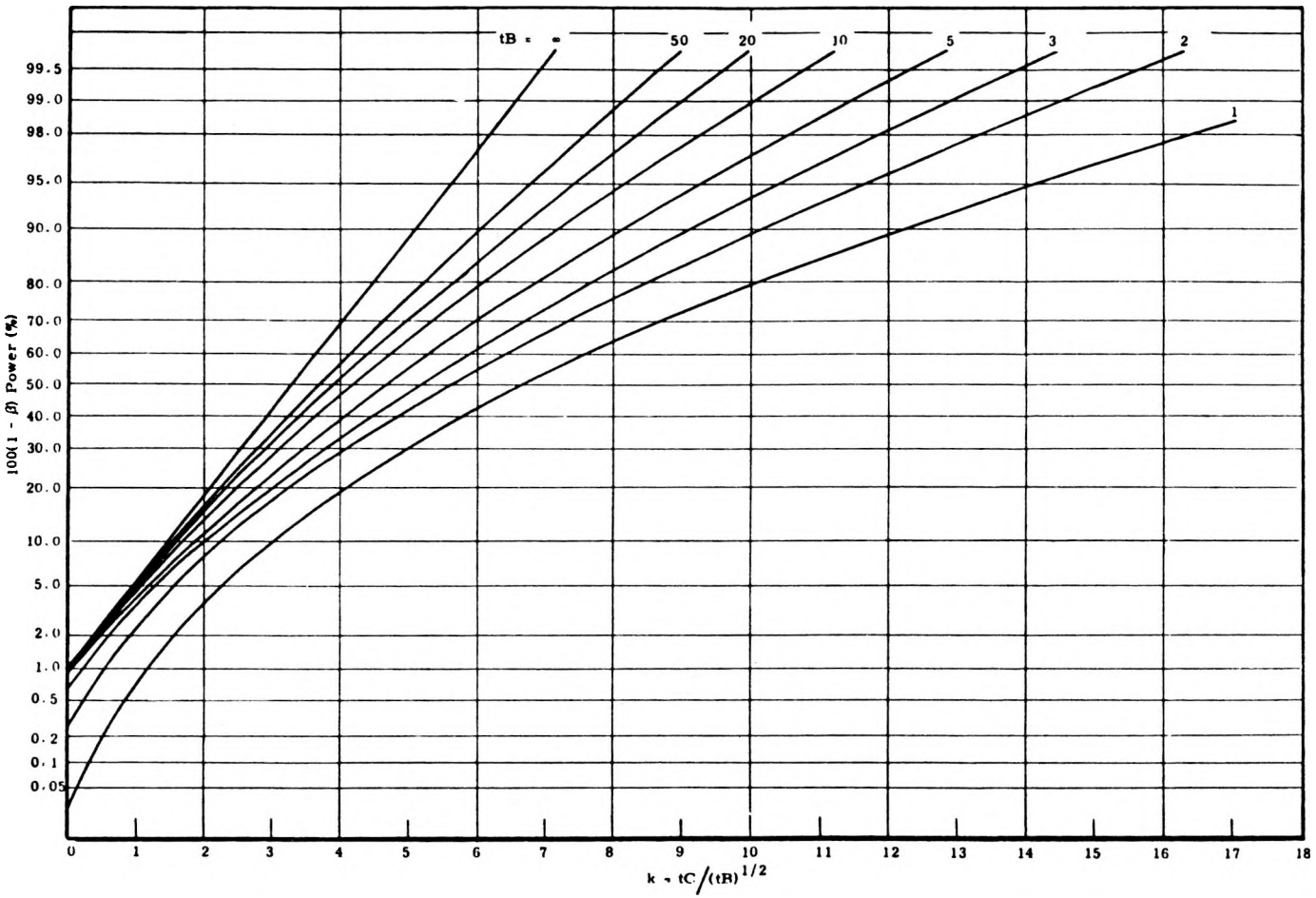

FIGURE D.2

Power of Decision Rule D_3 (Using Eq. (4)) to Detect

$C > 0$ when $\alpha = 0.01$ and $\rho = 1$.

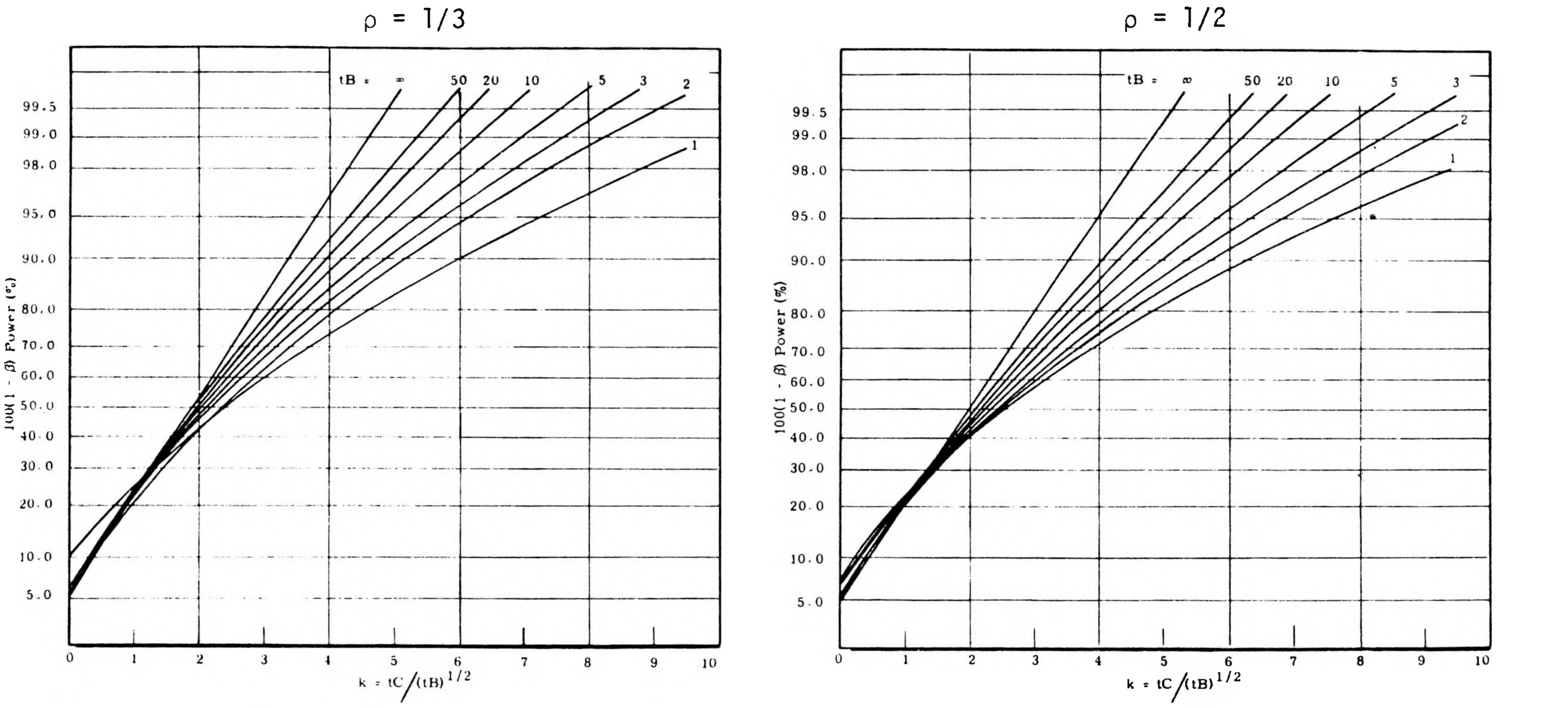

FIGURE D.3

Power of Decision Rule D_3 (Using Eq. (4)) to Detect $C > 0$ When $\alpha = 0.05$ and $\rho = 1/3$ and $1/2$.

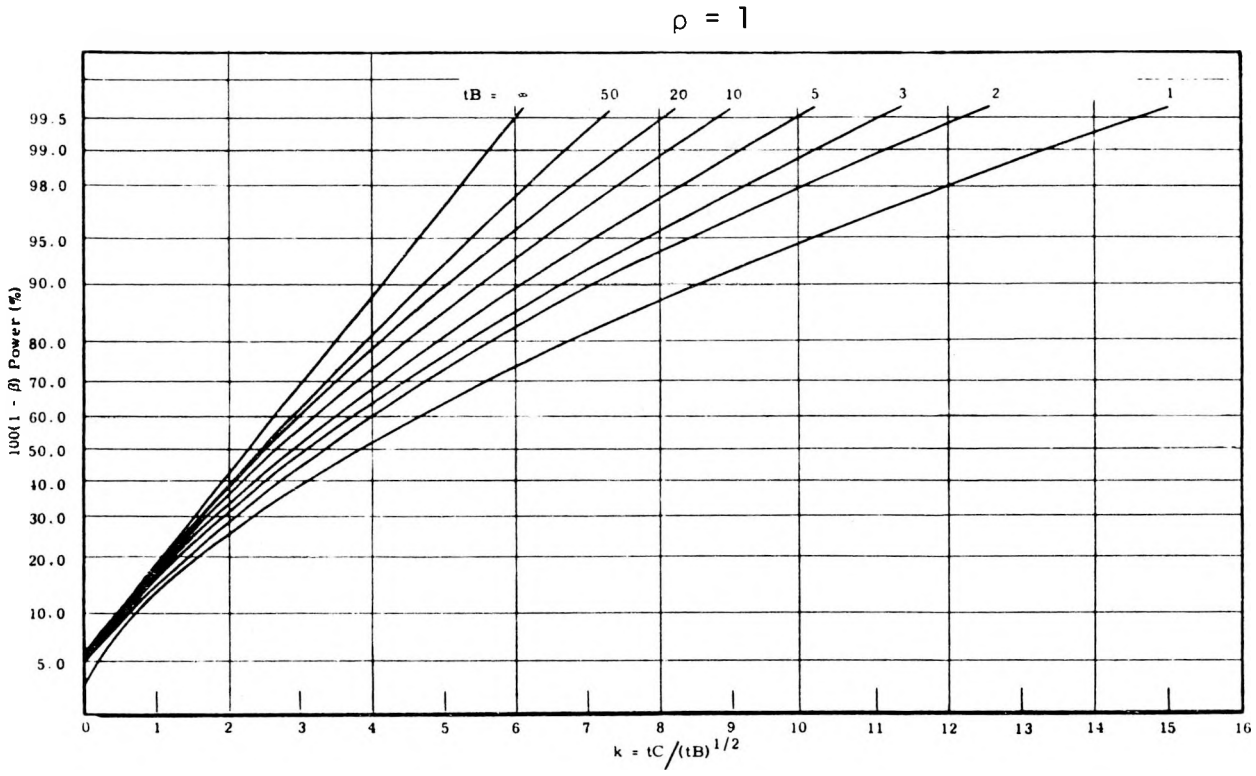

FIGURE D.4

Power of Decision Rule D_3 (Using Eq. (4)) to Detect C > 0
When α = 0.05 and ρ = 1.

PHILOSOPHY AND MECHANICS OF SYNTHESIZING DATA FROM DIFFERENT DATA BASES

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My basic philosophy on the synthesis of data as it relates to the Nevada Applied Ecology Group (NAEG) data bank can be summarized as follows: I believe the NAEG data bank should strive to store and retrieve information to provide (a) informative lists and tabulations of data, and (b) graphical and tabular descriptions of data sets. It should not attempt to compute estimates of parameters such as means and variances, estimate linear or nonlinear regression relationships, or perform other statistical manipulations. The data bank should not attempt statistical analyses primarily because a statistical analysis appropriate for one data set may not be appropriate for another data set. The responsibility for statistical analysis must rest with the scientist, not with the data bank. The data bank can perform a very valuable service, however, by providing tabular and graphical displays that help the scientist in his synthesis effort.

The synthesis or drawing-together of data from different data bases (soil, vegetation, small vertebrates, large vertebrates) must take into account spatial and temporal (time) relationships inherent in the data. Soil and vegetation samples, for example, may be collected adjacent to each other. Hence, their respective ^{239}Pu concentrations may also be related in some way. Or, consider a cow grazing in an area where the soil and vegetation are contaminated with plutonium. The Pu concentrations in the cow's tissue should be related to the length of time spent grazing in the area.

When we consider the kinds of information that must be stored in a data bank and retrieved in a concise and informative manner, it is important to remember that it is the relationship between *e.g.*, soil and vegetation or vegetation and cattle concentrations that is of greatest importance. Hence, we should strive to organize printouts that indicate relationships with clarity. This can be done in some cases with simple two-dimensional plots. For example, it is relatively easy to produce an on-line plot showing locations in the study area where soil and/or vegetation samples were collected. Sample locations are identified by their Nevada Grid Coordinates which uniquely identify each sampling location. It should also be possible to plot the capture locations of a small mammal that has been trapped over a given period of time. Since concentrations in tissues of small vertebrates may be related to soil or vegetation concentrations, the investigator may want a plot of soil or vegetation sample locations on the same map on which the capture locations of a single animal are plotted.

If the spatial pattern of radionuclide concentrations for soil or vegetation is needed, three-dimensional plots can be obtained on the Cal-Comp-Plotter or some other off-line plotting device (see Figure 1).

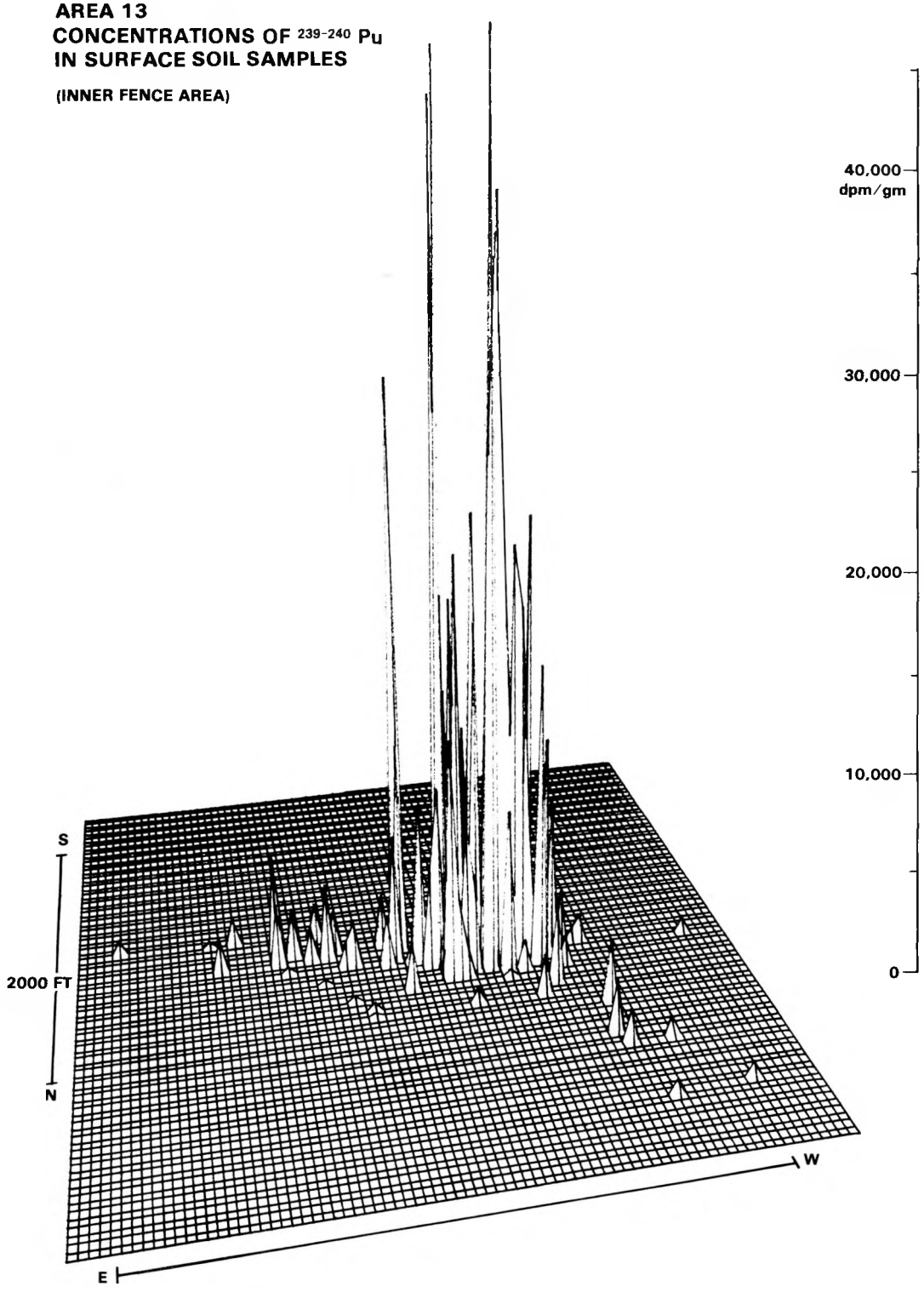
AREA 13
CONCENTRATIONS OF 239-240 Pu
IN SURFACE SOIL SAMPLES
(INNER FENCE AREA)
40,000
dpm/gm
30,000
20,000
10,000
0
S
2000 FT
N
W
E

FIGURE 1

The concentration is plotted on the vertical scale and the north-south, east-west coordinates on the horizontal (X,Y) scales. These simple location and concentration plots are very useful because they convey information about the relationship between geographical distance and radionuclide concentration, *i.e.*, where is the Pu? Another useful plot would be Pu concentration versus depth of the sample in the soil profile.

Once the scientist has a visual picture of where samples were collected and their relative radionuclide concentrations, he will probably want to see the actual data. For example, he may want a list of ^{239}Pu concentrations for surface soil samples obtained from Area 13 (Project 57), for strata 1, 2, and 3. This should be printed out in tabular form to make it easy for the requesting scientist to compute whatever statistics (means, standard deviations, etc.) may be needed. The headings for the columns could be as follows:

		NEVADA GRID COOR.			
STRATA	STAKE NO.	N	E	^{239}Pu	% COUNTING ERROR
1	1				
	2				
	.				
	.				
	n_1				
etc. for other strata					

Immediately before this listing would appear the information necessary to unambiguously define the tabular data; *e.g.*, AREA 13, PROJECT 57, SURFACE (0-5 cm) SOIL SAMPLES, UNITS: nCi/g dry weight. A similar listing could be made for vegetation samples. If vegetation and soil samples are collected adjacent to each other (so that the data are paired), another two columns could be added to the above example to include the vegetation concentration and its counting error. Other columns that might be added are for distance and direction between each soil, vegetation pair. Note that in the above example the stake number was given in addition to the Nevada Grid Coordinate. This stake number (which starts over at 1 within each stratum) should always be listed any time the Nevada Grid Coordinates are listed.

The units in which concentrations are listed should be at the discretion of the requestor. Soil results should be stored in terms of dry weight of the entire soil sample (before ball-milling). Vegetation data should be stored in terms of both dry and ash weights. Small and large vertebrate concentrations should be in terms of wet (received) weight as well as dry and ash weight. Each listing of data should be accompanied by a short comment defining what is meant by the terms "wet," "dry," or "ash" weight, or a reference to a published paper should be given where this information can be found.

Space should be provided in the computer to store comments that may be important in interpreting the data. Upon command, the computer should list these comments in tabular style for the requested data set. Example: AREA 13 (PROJECT 57), SPECIES X,

NEVADA GRID COOR.

ANIMAL NO.	N	E	COMMENTS
1			_____
2			_____
etc.			

From such a tabular printout the investigator can quickly scan the comments without going through pages of output. If no comments were stored this is also noted immediately by the requestor.

Radionuclide concentrations in tissues of grazing cows may be related to length of grazing, concentrations in the soil and vegetation in the grazing compound, age and breed of cow, etc. A scientist may request lists of concentrations for tissues in cows grazed for a specified length of time in a specified grazing compound. The data bank should be organized so that these lists can be obtained. One tabular layout is:

Tissue	Cow No.	^{239}Pu (nCi/g wet)	% Counting Error
Hide	3	-	-
	7	-	-
Muscle	3	-	-
	7	-	-
Etc.			

where this listing would be clearly identified as to Area, Compound grazed, length of grazing, species and breed of cow, etc. A list of comments should also be listed unless instructed specifically not to do so.

There is an additional graphic-tabular display commonly known as a Stem-and-Leaf display that would be a valuable addition to any list of data requested, where the list has, say, at least 10 data in it. A Stem-and-Leaf is basically a histogram, but the numerical values from which the histogram are constructed are retained and displayed. This display can be generated by calling a subroutine. The computer code package is on the CYBER 74 computer system at ERDA's Richland Operations Office. It was originally written at the National Bureau of Standards in 1973 and is available from them. The display is useful for describing any data set and for comparing the shape of its distribution with that of other data sets.

In summary, I feel that the NAEG data bank can increase its usefulness to the scientific community if the method of display of the stored data is organized to convey maximum information about each data set and the spatial

and temporal relationships between data. The examples given here can no doubt be expanded and improved upon. They are not meant to be the "final word."

ENVIRONMENTAL SCIENCES INFORMATION STORAGE AND RETRIEVAL SYSTEM

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(Ed. Note: Previously published in
NAEG Report NVO-153, pp. 471-472.)

ABSTRACT

Reynolds Electrical & Engineering Co., Inc. (REECo), has since 1970 accumulated information relating to the AEC's Nevada Applied Ecology Group (NAEG) programs at the Nevada Test Site (NTS).

These programs, involving extensive soil, vegetation, and small-animal studies, have generated informational data concerning the collecting, processing, analyzing, and shipping of sample materials to various program participants and contractors. Future plans include incorporation of Lawrence Livermore Laboratory's resuspension study data, REECo's on-site air data, and EPA's large-animal, off-site air, and off-site soil data.

INTRODUCTION

Since 1970, REECo has accumulated information relating to the AEC's NAEG programs at the NTS. These programs which involve extensive soil, vegetation, and small-animal studies have generated data concerning the collecting, processing, analyzing, and shipping of sample materials to various program participants.

In particular, the Distribution and Inventory Program Element generates large volumes of data. Moreover, organizing and maintaining a complete soils library has created additional information and responsibilities concerning soil sample histories and accountability.

Environmental Sciences Record-Keeping System

In order to establish the proper software system to adequately document these data, it was necessary to first index the information into four basic categories. These categories are defined as:

1. Field Data--Sample location, collection method, data collected, and sample volume.

2. Aliquot Data--How the sample was divided, processed, or otherwise changed.

3. Shipping and Receiving Data--Documentation of where, when, and to, or from whom the aliquot or aliquots were shipped, and an estimate of how much radioactivity was contained in each sample. (The radioactivity and radioisotope identification are entered as estimates to satisfy shipping requirements.)

4. Results--Documentation of analysis results of participating laboratories by filing and cross-referencing for future use.

Forms were designed so that information could be easily transferred to punch cards for EDP input. The aliquot card form was designed on the basic logic element in a pyramid-type logic structure which allows the computer to trace the sequence of events that leads to any aliquot in the system, tracing its history from its present form and quantity. A unique numbering system was devised which uses a key to identify the parent and offspring in each process. Each data category information is tied to the logic structure with the aliquot card number. These forms were also designed to handle results determined by other participating laboratories. A major function of the card form directs the computer in converting the result data input to a common data base.

Information Systems Department Participation

REECo's Information System Department programmed the information storage and retrieval system from the card forms, and the AEC central computer (CDC 6400) provides the hardware output. To date, one million data characters have been stored and processed into an initial report. It is anticipated that there will be 113,000 results stored in the near future; 20,000 PIDP and 93,000 NAEG. When these data are entered, there will be 127,000 results in storage. The system now uses eight different coded tables which are frequently being expanded. These tables contain the various names, descriptions, and other comments used routinely in various reports that are being generated.

FUTURE PLANS

A review and feasibility study is currently in progress by REECo's Information Systems Department, looking toward future expansion of the system and its data base to accommodate such projects as the LLL resuspension studies, REECo's on-site air data, and the EPA's past animal, off-site air, and off-site soil data. The study is scheduled for completion by mid-December, 1974.

Other NAEG methodology

PLUTONIUM UPTAKE BY A SOIL MICROORGANISM, *ASPERGILLUS NIGER*

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ABSTRACT

A common soil fungus, *Aspergillus niger*, was grown on malt agar and in broth containing soluble plutonium compounds. The transport factors derived from this series of experiments indicate that plutonium was concentrated in the mycelium and further transported to the aerial spores of this fungus. A new and simple spore collection technique was developed to prevent cross-contamination of the spores with mycelial fragments and by direct contact with the plutonium-containing agar medium.

If a similar process occurs in plutonium-contaminated soils, it could be an important link in the transfer of soil-deposited plutonium to man. It would also explain the apparent time-dependent increases in the uptake rate of plutonium by plants grown on contaminated soils reported by Romney and his associates.

INTRODUCTION

Nuclear power plants will become increasingly important as a source of electrical energy during the next few decades. The potential hazards derived from the use of plutonium in fast-breeder reactors will increase as such facilities are built (Cook, 1973). It has been estimated that by the year 2000, about 2,700 megacuries of radioactive wastes in the United States will have to be sequestered and about 400 megacuries of transuranic alpha emitters will be part of that waste. Plutonium-239 will be an important constituent of these transuranic alpha emitters and, since its half-life is 24,390 years, it will be a potential hazard for many thousands of years (Weinberg, 1972). Therefore, it is important that research be conducted to determine how environmental and biological factors influence the pathway of plutonium to man. Plutonium deposited on soil is believed to consist primarily of insoluble particles and of polymers absorbed on soil particles (Rhoads, 1957a and b). Resuspended plutonium-containing particles can enter man by inhalation or ingestion. The larger inhaled particles can be removed from the respiratory system by ciliary action and can then be transferred to the gastrointestinal tract for excretion. The smaller particles may be more dangerous because they can become deposited in the pulmonary spaces where they may be assimilated into the blood or retained in the lung, thus presenting a continuous source of radiation to the surrounding tissue.

Plutonium originally deposited on soil may be ingested by eating contaminate plants or animal products, such as vegetables, meat, and milk. Animals may become contaminated with plutonium through the inhalation of resuspended particles, the ingestion of dust particles deposited on vegetation, the ingestion of soil particles with the food and while grooming, and the ingestion of plants containing plutonium. Only that fraction of the plutonium that can be assimilated by the animal is of primary concern; therefore, the chemical nature of the ingested plutonium may be critical. It is probable that the plutonium absorbed by plant roots and translocated to a leaf will be more readily absorbed in animals' intestinal tracts than plutonium deposited on the leaf with resuspended dust.

Plants have been shown to assimilate plutonium from soil (Francis, 1973). The discrimination factor, defined as the ratio of plutonium disintegrations per minute per gram of dry plant material to the plutonium disintegrations per minute per gram of dry soil, has been reported to be on the order of 10^{-4} to 10^{-6}. Experimental evidence by Romney *et al.* (1970) indicated that the rate of plutonium uptake of ladino clover increased with time resulting in an increase of plutonium incorporation. This increase was explained as possibly being due to the continuing development of the plant roots which increased the number of contact points between the roots and the plutonium particles combined with an increasing biological availability of plutonium. It was suggested that plutonium availability might be enhanced by chelating materials present in the soil. Another possibility is that soil microorganisms may be involved in the transfer of plutonium from soil to plants.

As a first step in an evaluation of the impact of soil microorganisms on the uptake of plutonium by plants, this study was initiated to determine whether soil microorganisms assimilate plutonium and, if so, to quantify the amounts assimilated.

METHODS

For *in vitro* studies, *Aspergillus niger* was grown in plutonium-spiked malt extract broth and on malt agar. This fungus was selected because it is ubiquitous and its properties have been extensively studied. It also produces aerial spores atop a lengthy conidiophore, a feature well suited for spore collection and one which reduces the possibility of cross-contamination with the contaminated nutrient media. Since the plutonium concentration in the spores was expected to be low, it was necessary to use a collection method which provided a sufficiently large biomass to allow quantitation of the plutonium uptake. Also, the collection procedure had to preclude any contamination of the spores with mycelial fragments. For these reasons, the spore collection methods commonly used were not satisfactory (Barton *et al.*, 1972; Johnson *et al.*, 1968; and Kang *et al.*, 1965). A device (Figure 1) to meet these requirements was constructed. It consists of a hypodermic needle (18-gauge x 1 1/2-inch), with the beveled portion of the tip ground off. The needle is connected to a syringe filter holder 25 mm in diameter. This holder is fitted with a membrane filter of 0.22-µm pore size to retain the spores (Figure 2), and is connected with flexible tubing to an aspiration system consisting of a series of three 250-ml filtering flasks and an aspirator. Each filtering flask used during this

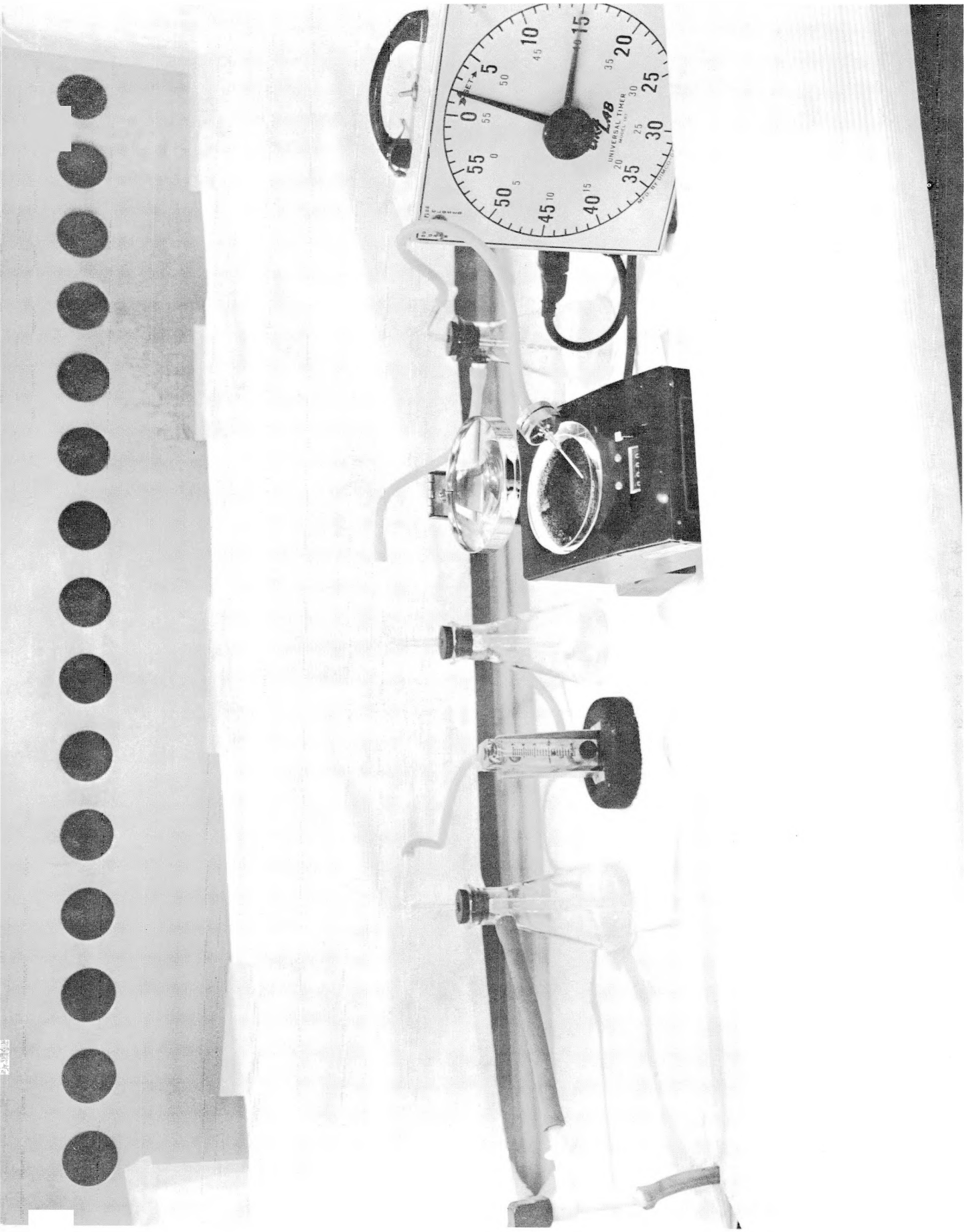

Figure 1. Apparatus Used to Collect Aerial Fungal Spores

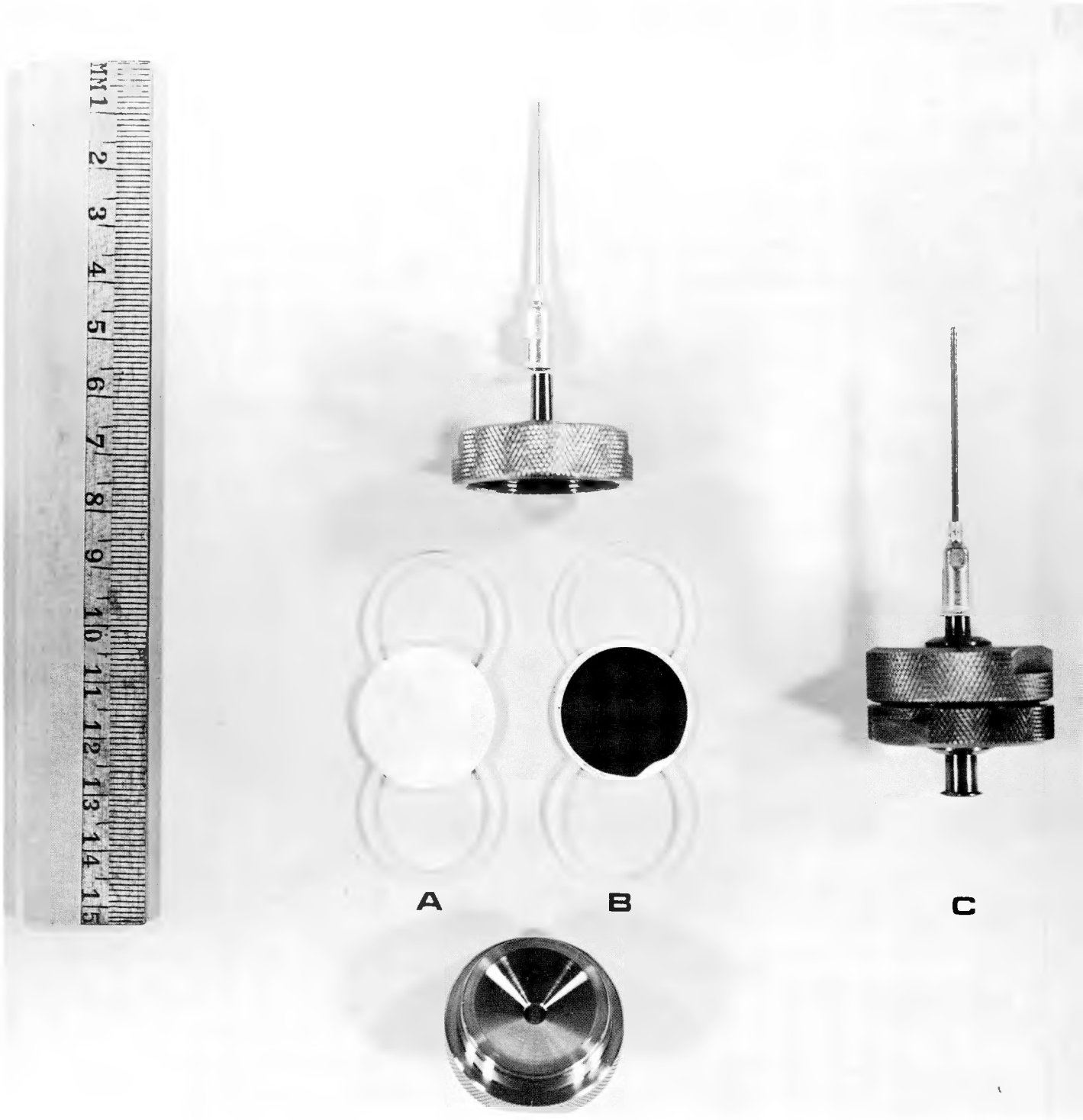

Figure 2. Disassembled Filter Holder and Modified Hypodermic Needle. (a) membrane without conidia, (b) membrane with conidia, (c) assembled unit

experiment contained 50 ml of water and two drops of antifoam agent. The flasks were used as a safety measure to trap any plutonium-containing material which might escape during an accidental rupture of the membrane filter.

With this apparatus, spores can be collected without any risk of contamination by mycelial fragments or by contact with the culture medium. Additional advantages of this collection device are its simplicity, low cost, ease of handling, and its capacity to collect relatively large amounts of material in a short time.

The spore harvesting technique employed involves placing the culture dish on a light box equipped with a magnifying lens. The spores were detached by gently touching them with the tip of the hypodermic needle and were simultaneously impacted onto the filter paper in the microsyringe holder. An air flow rate of 1.6 liters/min was used to vacuum up the spores after microscopic examination showed that no conidiophores were deposited on the filter at this flow rate. However, at air flow rates exceeding 2 liters/min, substantial portions of the conidiophores accumulated with the spores. A two-minute collection period was found to be optimal for massing the spores on the filter. Longer collection times resulted in an excessive accumulation of spores on the inner surface of the holder. An electric timer was used to control the lengths of collection periods; it was connected to the light box which was preset to terminate illumination after two minutes. Based on five two-minute collection periods per plate, the average spore recovery was estimated to be 95-98%.

EXPERIMENTAL DETAILS

In an initial experiment, malt-extract broth (15 g per liter), contained in 100-ml flasks, was spiked with a plutonium-238 citrate complex to give 223 nCi per flask, and inoculated with *Aspergillus niger*. The inoculum was an agar plug 4 mm in diameter cut from a two-week-old culture of this fungus. A total of six plutonium-spiked inoculated samples and six inoculated control samples was prepared. The first set consisting of three spiked and three control samples was agitated on a reciprocal shaker at room temperature for 14 days, the second equal set for 56 days. The fungal mats were then collected with glass rods, pressed to remove the broth, and washed three times with 25-ml volumes of 0.85% saline solution. The mats were oven-dried at 60° C., acid digested, and analyzed for plutonium-238 by standard alpha scintillation methods.

In a second set of experiments, malt agar (25 g agar plus 30 g malt extract per liter) was buffered to pH 2.5 with 0.1 M monopotassium phthalate and 0.1 M hydrochloric acid (Clark *et al.*, 1965). Plutonium-238 nitrate in 4 N nitric acid was added to aliquots of the buffered agar to obtain plutonium concentrations of 112, 224, and 448 pCi/ml. Twenty-ml aliquots of the spiked medium were pipetted into petri dishes to make three sets of four dishes with 2.24, 4.48, and 8.96 nCi per dish, respectively, containing 1.34 g of dry mass each. The dishes were inoculated with 0.3 ml of *Aspergillus niger* spore suspension per dish (10^8 spores per ml), placed in plastic bags, and incubated for 33 days at 25 ± 1° C. The aerial spores

were then collected using the collection device and technique described earlier, acid digested, and the plutonium activity determined by alpha spetrometry (Talvitie, 1972).*

Following collection of the spores, the agar with the mycelium from each dish was transferred to a glass beaker and melted on a hot water bath. The mycelial mat was carefully removed with a spatulum, placed on a Buchner funnel fitted with a #2 Whatman filter, and washed three times with 50-ml volumes of distilled water. The mat was oven-dried at 60° C., acid-digested, and the plutonium activity determined by alpha spectrometry (Talvitie, 1972). Three control samples were also treated in this manner.

In a third experiment, an attempt was made to remove any soluble plutonium which might have migrated to the spore surfaces. The plutonium-containing spores were treated for five minutes with an aqueous solution containing 0.85% NaCl, 0.1% Na_2 EDTA, and 0.1% of a 2% Tween 80 surfactant solution. The filtrate and the spores were then analyzed for plutonium (Talvitie, 1972).

No activity could be detected in the filtrate, whereas the spores showed the activity expected.

RESULTS AND DISCUSSION

The first set of experiments was designed to determine whether the soluble plutonium (added as a citrate complex to a malt extract broth culture medium) became translocated to the mycelium of *Aspergillus niger*. The results shown in Table 1 reveal that a substantial portion of the plutonium became associated with the mycelium in a way which resisted elution by washing. The specific activities of the mycelia determined after 14 and 56 days differed by a factor of two (Table 1, h). This time-dependent relationship suggests that plutonium was absorbed rather than adsorbed by the mycelium. When all other conditions are equal and concentration gradients within the media are excluded, adsorption depends only on the plutonium concentration and the surface area of the mycelium. The ratio of the dry weight to surface area of the mycelia can be assumed to be constant, and in these experiments, no significant concentration gradients could have been developed within the individual flasks, since plutonium was present as the soluble citrate complex (Cleveland, 1970a) and the broth was of low viscosity and was agitated continuously. If only adsorption had occurred, then the specific activities of the mycelia should have been equal in both treatments for adsorption is independent of the growth time. The fact that in these experiments the specific activities increased by a factor of two during the additional 42 days of incubation for the second group is indicative of plutonium absorption rather than adsorption. Experiments are in progress to further explore this aspect.

The second set of experiments was designed to determine the relationship between the plutonium concentrations in the culture media (agar) and the amount of plutonium translocated to the mycelia and spores. In these experiments, plutonium was added to the agar as a nitrate. To prevent hydrolysis of the plutonium and the formation of a colloid (which may start

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*In separate experiments, the moisture content of the spores was determined by placing them in desiccators over P_2O_5 immediately after collection. The total weight loss amounted to only 2-3%.

able 1. Plutonium Uptake of *Aspergillus niger* Mycelia From Plutonium Citrate Complex Broth and Plutonium Nitrate Agar Media

Culture Medium	Growth Period (days)	Total Pu Added (pCi)	Specific Activity of Medium (wet weight) (pCi/g)*	Specific Activity of Medium (dry weight) (pCi/g)	Mycelium Collected (dry weight) (g)**	Total Mycelium Activity per Sample (pCi)**	Specific Mycelium Activity (pCi/g)
Broth with plutonium citrate complex	14	0	0	0	0.43 ± 0.07	0	0
	14	2.23×10^5	2.23×10^3	1.49×10^5	0.43 ± 0.04	8.00×10^4	1.8×10^5
	56	0	0	0	0.30 ± 0.03	0	0
	56	2.23×10^5	2.23×10^3	1.49×10^5	0.31 ± 0.03	1.21×10^5	3.9×10^5
Agar with plutonium nitrate	33	0	0	0	0.12 ± 0.04	0.48	4.0
	33	2.24×10^3	1.12×10^2	1.67×10^3	0.09 ± 0.02	0.47×10^3	5.0×10^3
	33	4.48×10^3	2.24×10^2	3.34×10^3	0.11 ± 0.03	1.27×10^3	12.0×10^3
	33	8.96×10^3	4.48×10^2	6.69×10^3	0.11 ± 0.03	2.54×10^3	23.0×10^3

*Based on specific weight of culture media = 1.00

**Arithmetic means of three samples each

with the plutonium concentrations employed at a pH >2.8) (Cleveland, 1970b), the pH of the agar medium was kept at pH 2.5 by the addition of a buffer. The specific activity of the mycelia isolated in these experiments was found to be a linear function of the plutonium concentration in the agar (Table 1, h). The total amount of plutonium in the mycelium was about 25% of the amount added to the agar and was independent of the plutonium concentration originally added to the agar (Table 3, e). It should be noted that in both sets of experiments, the conditions were optimized for maximum plutonium uptake; *i.e.*, a soluble form of plutonium was evenly distributed throughout the media under conditions which prevented plutonium hydrolization and polymerization. In addition, the broth medium was continuously agitated to eliminate concentration gradients.

The specific activity of the spores (Table 2, f) was more than two orders of magnitude less than the specific activity of the mycelia grown on agar (Table 1, f). This indicates a translocation barrier between the mycelia and the spores. A nearly linear relationship was found to exist between the activity found in the spores and the activity added to the agar media. Only about 0.05% of the plutonium was translocated to the spores.

As a means to compare the uptake of plutonium by plants grown on plutonium-contaminated soil, the discrimination factor defined earlier is commonly used. Because the soil water content may fluctuate within wide limits, the discrimination factor is based on dry soil weight. When trying to apply the same concept to plutonium uptake of *Aspergillus niger* grown on culture media, it must be kept in mind that the soil used in plant growth serves a dual function: (1) it is a mechanical support to the plants, and (2) it is a source of nutrients. In culture media, such as agar and broth (or, for that matter, hydroponic solutions), water is an integral part of the mechanical support for the fungus, and is thus comparable to soil. Consequently, a discrimination ratio should be based on the wet weight of the culture medium.

The values calculated on this basis are listed in Table 3, d. They are several orders of magnitude higher than the discrimination factors found for plant uptake from soil (Francis, 1973). This may indicate a lower discrimination against plutonium absorption and translocation in the fungus, or a greater availability of plutonium in the agar system. However, it should be kept in mind that soil/plant and culture medium/fungus systems are entirely different. Plants assimilate only minor amounts of inorganics from the soil and, for practical purposes, no soil depletion occurs during one growth season. Plants synthesize their organic materials, whereas microorganisms depend solely on the culture medium for their inorganic and organic materials. This results in a significant mass transfer from the culture medium to the microorganism. The material balance, including water, is constant for such a system, except for small losses due to microbial metabolism. On the other hand, higher plants obtain all of their carbon from the atmosphere, and their metabolism, ion absorption, and translocation systems are vastly different to those found in fungi.

The percentage of the plutonium transferred to the microorganism could be used as another basis of comparison. These values are given in Table 3, e,

Table 2. Plutonium Translocation to *Aspergillus niger* Spores From Plutonium Nitrate Agar Medium

Plutonium Nitrate Added (pCi)	Growth Period (days)	Specific Activity of Culture Medium (wet weight) (pCi/g)*	Specific activity of Culture Medium (dry weight) (pCi/g)	Spore Mass Collected**(g)	Total Spore Activity per Sample (pCi)*	Specific Spore Activity Corrected for Background (pCi/g)
0	33	0	0	0.017 ± 0.004	0.09	5
2.24×10^3	33	1.12×10^2	1.67×10^3	0.021 ± 0.004	0.90	39
4.48×10^3	33	2.24×10^2	3.34×10^3	0.031 ± 0.008	1.73	53
8.96×10^3	33	4.48×10^2	6.69×10^3	0.043 ± 0.001	5.40	123

*Based on specific weight of culture media = 1.00

**Arithmetic means of three samples each

Table 3. Comparison of Discrimination Ratios, Percent Uptake, and Transport Factors for *Aspergillus niger*

Parent Medium	Tissue	Total Plutonium in Parent Medium (pCi)	Discrimination Factor Based on Wet Culture Medium Weight*	Percent Plutonium Transferred to Tissue	Transport Factor
Broth	Mycelium 14 days 56 days	 2.23×10^5 2.23×10^5	 81 175	 36 54	 1.2 2.6
Agar	Mycelium	2.24×10^3 4.48×10^3 8.96×10^3	45 54 51	21 28 28	3.0 3.6 3.4
Agar	Mycelium & Spores	2.24×10^3 4.48×10^3 8.96×10^3	38 40 37	21 28 28	2.5 2.7 2.5
Agar	Spores	2.24×10^3 4.48×10^3 8.96×10^3	0.35 0.24 0.27	0.04 0.04 0.06	2.3×10^{-2} 1.6×10^{-2} 1.8×10^{-2}
Mycelium	Spores	0.47×10^3 1.27×10^3 2.54×10^3	NA NA NA	0.2 0.1 0.2	7.8×10^{-3} 4.4×10^{-3} 5.3×10^{-3}

*Based on specific weight of culture media = 1.00

NA = Not Applicable

but this approach gives little, if any, information about the concentration or discrimination effects. As a solution to understanding plutonium transport, a "transport factor" (TF) is proposed which is applicable for culture media where the distribution of nutrients and pollutants is uniform. This factor (TF), which is concentration-independent, is defined as that fraction of the total plutonium that is transported from the media to the tissue, divided by the fraction of the total dry mass transported from the media to the tissue, or

$$TF = \frac{Pu_T/Pu_M}{M_T/M_M}$$

where Pu_T = total plutonium content of tissue (e.g., mycelium, spores)

Pu_M = total plutonium originally present in the parent medium

M_T = dry mass of tissue

M_M = dry mass originally present in the parent medium

The transport factor is identical to the ratio of the specific activities (mycelium/agar) or to the familiar discrimination factor when based on the dry mass of the culture medium. It immediately shows if accumulation of or discrimination against the pollutant has occurred. TF >1 indicates an accumulation; TF <1 defines discrimination against the pollutant.

The transport factors defining the movement of plutonium from substrate to mycelia and on to the spores are listed in Table 3, f. These values show that under the experimental conditions employed, an accumulation (TF >1) of plutonium occurred in the mycelium of *Aspergillus niger*. All transport factors based on agar media were essentially concentration-independent. On the other hand, the transport factor for plutonium from mycelia* to spores is smaller by more than two orders of magnitude (TF <1). This demonstrates that discrimination occurred against plutonium transport from mycelia to spores.

The implications of these findings are threefold. First, it is known that soil microorganisms play an important role in plant nutrition, chemically transforming substances that are unavailable to plants to forms available for plant uptake. This suggests that soil microorganisms may also be able to attack deposited plutonium making it more available to plants. Thus, years of microbial activity associated with plant root development would result in an increase of the plutonium uptake rate by plants with time. This would explain the experimental results of Romney *et al.* (1970) which showed an increase in the plutonium uptake rate by plants with time, particularly when considering that the rhizosphere is the region of intensive and extensive microbial activity. Consequently, once plutonium enters the soil, the importance of plant assimilation as a pathway to man may increase with time, and it may be that in several decades plutonium uptake by plants may likely increase to a higher level than is currently believed.

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*Mycelial dry mass and plutonium concentration values used in these calculations were those determined after spore collections.

Second, an additional impact of fungal uptake of plutonium may be imposed if plutonium is assimilated by fungi and deposited in their spores. Many soil fungi release their spores into the air. If inhaled by man and retained in the respiratory system, this will cause a prolonged radiation exposure of the surrounding tissue in the same manner as resuspended plutonium particles.

Third, the direct uptake of plutonium from soil by animals and man could also be affected by soil microbial activity. As pointed out in the introduction, the plutonium particles presently associated with dust and soil that can be inhaled and ingested by animals and man are generally insoluble. If, over the years, soil microorganisms change part of this plutonium into forms more biologically available to man and animals, then inhalation and ingestion of plutonium-containing soil particles will present an increasing problem for man.

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ABSORPTION, DISTRIBUTION, AND EXCRETION OF PLUTONIUM BY DAIRY CATTLE

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ABSTRACT

In order to obtain information on the significance of the milk link in man's food chain as a source of plutonium exposure and to gain additional information on plutonium deposition patterns in ruminants, a series of metabolism studies with dairy cows was initiated. This preliminary report outlines the first two studies in the series and, while some of the laboratory assays have not been completed, several comparative observations can be reported at this time.

Two groups of Holstein dairy cows, four cows in each group, were studied to examine the physiological transport of ingested plutonium citrate and plutonium dioxide. Approximately 3 mCi of plutonium citrate per animal was administered in an acute treatment to the first group, while 1 mCi of plutonium dioxide was given daily to each animal for 19 consecutive days in the second group. Samples of blood, milk, urine, and feces were taken during and after oral dosing, while tissue collections were made at necropsy 6 to 13 weeks after treatment.

As expected, the major portion of plutonium activity (approximately 96% in Group I and slightly less than 100% in Group II) was excreted in the feces. However, recovered activity in urine and milk following both the acute dose of plutonium citrate and the multiple doses of plutonium dioxide confirmed the physiological uptake and transport of both chemical forms. Total plutonium transport to milk was not great and, on a percent of oral dose basis, was observed to be 2×10^{-4} and 2×10^{-5} following the plutonium citrate and plutonium dioxide treatments, respectively. A complete report, including a comparison of gastrointestinal uptake, tissue deposition patterns, physiological reduction factors, and placental transfer, will follow once the laboratory analyses are finalized.

INTRODUCTION

The extent of plutonium transport from contaminated forage to bovine milk has not been established. Since dairy animals consume large quantities of food and water, and since milk and milk products form a direct link in man's food chain, bovine metabolism of ingested plutonium is of considerable importance for an analysis of exposure pathways to human populations. This research was undertaken to study the problem in lactating Holstein dairy cows, insofar as the relationship between activity ingested, uptake by systemic circulation, tissue retention, and transport to milk are concerned, and to investigate the transport of different chemical forms of plutonium administered under acute and chronic conditions.

In an early plutonium metabolism study on rats, Scott *et al.* (1948) observed that the average value for gastrointestinal absorption of plutonium, using three different valence states, was approximately 0.007%. Comparative studies on the intestinal uptake of plutonium nitrate revealed no significant differences in total absorption between the rat and pig (Weeks *et al.*, 1956). These results were quantitatively similar to those obtained in a rather extensive report by Katz *et al.* (1955) which, following a chronic oral plutonium treatment to rats, presented the mean gastrointestinal absorption and retention value at 0.003% of dose. Several studies in the literature have addressed the metabolism of plutonium once it enters the systemic circulation. Investigators have frequently noted that a large percentage of intravenously injected plutonium was lost from the blood, often removed in colloidal aggregates by the liver, shortly after injection (Painter *et al.*, 1946; Stover *et al.*, 1959). Plutonium remaining in the blood for longer periods was largely combined with protein (Painter *et al.*, 1946; Stover *et al.*, 1959; Beliayev, 1959). But while the physical state of injected plutonium clearly influenced the tissue distribution pattern, variations between species, treated under identical conditions, have been noted (Rosenthal *et al.*, 1972).

Stover *et al.* (1968) isolated the serum Pu IV protein complex and identified the protein as transferrin, the protein that transports iron. This study also noted that the distribution of Pu IV between transferrin and the low molecular weight constituents of blood was related to the extent of transferrin saturation with iron and suggested that this may be a factor in the observed variations in plutonium distribution and excretion. Relationships between iron and plutonium transport at the intestinal level are not known, except that with both iron and plutonium, soluble complexes appear to increase intestinal uptake. Current investigations on iron absorption are concentrating on an intracellular protein, the synthesis of which increases under conditions of iron deficiency (Forth and Rummel, 1973). Relationships with plutonium at this intracellular level have not been established.

While investigating the effects of plutonium on mice treated *in utero*, Finkel (1947) discussed the relative concentrations of the element which had been transported through both the placenta and through the milk after parturition. In spite of the early realization that plutonium could be transported by the systemic circulation to milk, relatively little work was done in the major milk producers, *i.e.*, ruminants. However, in 1964, Sansom reported the transfer of ingested plutonium oxide to bovine milk but observed that the results may have been influenced by fecal contamination. The need for additional investigation was further suggested by the fact that in the above-mentioned study, only two cows were used. One animal had been given a citrate-buffered solution of plutonium nitrate, while the other cow received small particles of plutonium oxide. McClellan *et al.* (1962a) injected two Suffolk sheep with citrate-buffered plutonium nitrate in order to establish a milk-to-plasma ratio. Although this study with sheep presented a reduction factor for plasma to milk (0.025), it did not discuss the anticipated larger reduction factor from gut to plasma for different chemical forms of plutonium administered under acute and chronic conditions.

The basis for this study is that man consumes large quantities of bovine metabolic products so that any evaluation of radiological hazards associated with a plutonium-contaminated countryside should consider transport to milk. The investigation was undertaken by (1) examining the amount of ingested activity transferred to the systemic circulation, (2) determining the amount of activity transported to milk, (3) establishing what portion of activity was retained in the tissues following initial absorption, and (4) observing the above phenomena after various chemical forms of the element were administered. In pursuing these objectives, samples of blood, milk, urine, and feces were taken from two groups of Holstein dairy cows during and after oral treatment with different chemical forms of plutonium. Tissue collections were made following subsequent necropsies.

METHODS AND MATERIALS

Two groups of four lactating dairy cows per group were given oral doses of plutonium. Treatment aliquots were initially placed in 1/2 oz gelatin capsules containing 5 g of starch. The 1/2 oz capsules were sealed in larger 1-oz capsules and subsequently given to the animals. A balling gun was used to administer the capsules.

Group I (mean weight 700 kg) received an acute treatment of 3 mCi plutonium citrate per animal, while Group II (mean weight 650 kg) received 1 mCi plutonium dioxide per animal per day for 19 consecutive days. Samples of blood, milk, urine, and feces were taken during and after dosing, while tissue collections were made at necropsy 7 to 13 weeks after treatment (Table 1).

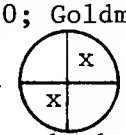
Table 1.

Collection Schedule for Urine, Milk, Feces, and Blood

Treatment	Approximate Animal Weight (kg)	Duration of Treatment	Urine	Milk	Feces	Blood
3 mCi plutonium citrate per animal	700 kg	One treatment	One collection per animal was taken on the treatment day, followed by twice a day collections through post-treatment day 24.	One collection per animal was taken on the treatment day, followed by twice a day collections through post-treatment day 24.	Fecal samples were taken two times a day from each cow for a period of 24 days after treatment. Duplicate samples were collected at each sampling period.	One sample per animal was taken daily for 24 days after treatment and again at necropsy on posttreatment day 93.
1 mCi plutonium dioxide per animal per day	650 kg	One treatment per day for 19 consecutive days	One collection per animal of urine was taken on treatment day one, followed by twice a day collections through post-treatment day 7.	One collection per animal was taken on treatment day one, followed by twice a day collections through post-treatment day 7.	Fecal samples were taken two times a day from each cow from the second treatment day through post-treatment day 7. Duplicate samples were collected at each sampling period.	One sample per animal was collected daily from treatment day one through recovery day 7 and again at the necropsies on posttreatment days 42 and 73.

Acute doses were prepared by adding 15 ml of citrate buffer to 0.5 ml of concentrated H_3PO_4 (Cleveland, 1967; Holley *et al.*, 1958) containing 15 mCi of plutonium-238. Doses (pH 3.3-3.5) were quantitated by comparing aliquots with plutonium-238 standard solutions using a liquid scintillation technique. The plutonium-238 dioxide microspheres were obtained from the Lovelace Foundation, Albuquerque, NM. These microspheres had a count median diameter, geometric standard deviation, and mass median diameter of 0.060041, 2.347549, and 0.533629 micrometers, respectively. The diameter of the average microsphere was 0178997 micrometers. Approximately 100 mCi of the particles were suspended in 50 ml of 2% Dowfax 2A-1 surfactant solution, and triplicate aliquots were then analyzed by liquid scintillation to determine the precise plutonium-238 concentration. Individual doses were prepared by removing a 500-µl aliquot from the suspension and placing it in the gelatin capsule.

Animals were placed in metabolism stalls a minimum of 5 hr before plutonium administration. An in-dwelling, inflatable catheter was used for urine collection and the cows remained catheterized from the time they entered the stalls until being transferred to outdoor pens two weeks after termination of the last dosing. Urine was collected in a 20-liter plastic bottle which was placed at the rear of the stall and connected to the catheter by polyethylene tubing. No formaldehyde was added to the urine samples.

Fecal samples, collected in a grid-covered pan lined with polyethylene sheeting, were mixed with sufficient formalin to increase the fecal weight by approximately 10%. Feces and formalin were subsequently mixed again using a spatula and heavy-duty mixer. Since a 12-hr accumulation of feces ranges in weight from 15 to 20 kg, considerable mixing was required to obtain homogeneity. Upon completion of the mixing, the feces were sampled as per the accepted cone and quartering method (Scott, 1950; Goldman, 1972). The contents of the tub were quartered  and the x'ed portions removed to a bucket. The contents in the bucket were mixed again, sectioned into eights, and 100-g samples removed as per the x's  and placed in a prelabeled polyethylene 400-ml container. Remaining fecal material was discarded as radioactive waste.

Milk samples were collected with a milking machine and, to minimize cross-contamination of milk with excreta, each animal was fitted with a waterproof udder support bag. The bag thus enclosed the teats and most of the udder in waterproof canvas which could be easily unfastened to allow access for milk collection and essentially prevented contamination from feces. The milk, which had been collected in individual

bucket milkers, was then poured into a prelabeled gallon cubitainer with 10 ml of formaldehyde. When the cows were released from the stalls and transferred to individual pens, milk collections were made with a portable milker.

Blood was collected by jugular venipuncture. Following centrifugation, the serum samples were separated from the formed elements. Tissue samples, collected at necropsy, included portions of the lung (apical and diaphragmatic lobes), liver, small intestine, spleen, kidney, skin, skeletal muscle, cardiac muscle, bone (femur, rib, and vertebra), rumen, abomasum, and pulmonary lymph nodes. All tissues removed for radiochemical analysis were weighed immediately.

Table 2 presents the analytical methods used to detect plutonium activity in the various biological samples. Blood and the necropsy tissue collections have not been analyzed for the multiple dose study. The majority of milk samples from the multiple dose phase have been analyzed; however, final interpretations will not be presented until all biological collections have been assayed.

The coprecipitation of plutonium with barium sulfate, an outline of which is presented below, was similar to the method reported by Sill (1969).

a. Duplicate 500- to 600-ml samples were weighed into 1,000-ml beakers. A 100-ml sample size was used for the blood.

b. The samples were digested with 70% HNO_3 and 30% H_2O_2 until all organic material was destroyed. Samples were then taken to dryness, but not baked.

c. One hundred milliliters of 38% HCl were added and the sample was again taken to dryness, but not baked. This step was repeated.

d. The salts were dissolved in 200 ml 2N HCl.

e. The sample was divided in half. One half was labeled SA and the other half was labeled SE.

f. One milliliter of 96% H_2SO_4 was added to all samples.

g. One-tenth milliliter of plutonium-238 standardized solution (approximately 100,000 dpm) was added to the SA sample.

h. One milliliter of 30% H_2SO_4 was added to all samples, which were then warmed gently.

i. Two grams $K_2S_2O_7$ were added to all samples and the samples were subsequently boiled for 2 min.

j. One milliliter of 0.5% $BaNO_3$ was added to all samples at the rate of 0.1 ml every 2 sec while the samples were stirred at boiling temperature. The samples were boiled for 1 min and the procedure was repeated with the addition of another 1.0 ml of 0.5% $BaNO_3$.

Table 2.

Analytical Methods to Determine Plutonium-238
Activity in Biological Collections

Treatment	Materials Collected				
	Milk	Blood	Urine	Feces	Necropsy Tissue
Acute dose of 3 mCi/animal of plutonium-238 citrate	Modified method of Sill (1969) Barium Sulfate	Modified method of Sill (1969) Barium Sulfate	Modified method of Sill (1969) Barium Sulfate	Modified liquid scintillation method of Butler (1968)	Modified method of Sill (1969) Barium Sulfate
One mCi/animal/ day for 19 consecutive days of plutonium-238 dioxide	Modified method of Sill (1969) Barium Sulfate	*Contract	Modified method of Sill (1969) Barium Sulfate	Direct quantitation of the 44 keV x-ray of plutonium using a Phoswich Detector	*Ion exchange and alpha spectrometric determination, Talvitie (1971-1972)

*Methods may be modified, since samples have not been analyzed.

k. The samples were cooled and filtered through a tared 0.2-0.8 μm membrane filter. Precipitates were washed with 30 ml distilled H_2O and then with 30 ml 90% ethanol.

l. The filter was dried for 4 hr and weighed.

m. Air-dried filters were mounted on a 4-in planchet and counted in an internal proportional counter.

n. Counting efficiency was achieved by correlating the SA sample values with the calculation of plutonium-238 activity in the SE sample.

The analytical method employing the modified Butler (1968) liquid scintillation technique included the following procedures:

a. Duplicate 50- to 100-g samples were weighed into 150-ml beakers.

b. Samples were digested with 70% HNO_3 and 30% H_2O_2 until all organic material was destroyed. Samples were then taken to dryness, but not baked.

c. The resultant salts were dissolved in 1N HNO_3, transferred to a 25-ml flask, and made to volume with 1N HNO_3.

d. Duplicate 10-ml aliquots were pipetted into 25-ml scintillation vials containing 15 ml of liquid scintillation mixture. One vial was labeled SA and the other vial was labeled SE.

e. One-tenth milliliter of a plutonium-238 standardized solution (containing approximately 100,000 dpm) was added to the SA sample.

f. Approximately 0.1 ml of distilled water was added to the SE sample.

g. The samples were dark-adapted for 24 hr and counted in a liquid scintillation counter.

h. Counting efficiency was achieved by correlating the SA sample values with the calculation of plutonium-238 activity in the SE sample.

If the plutonium-238 activity in the sample was less or was expected to be less than 100 dpm-ml, the above procedure was amended beginning with step c.

c^1. One hundred milliliters of 38% HCl were added to the HNO_3 digested and dried sample. The HCl solution was then evaporated to dryness, but not baked. The resultant salts were dissolved with 60- to 80-ml 8N NCl.

d^1. The hot sample (70 to 80°C) was extracted into 10 ml of 10% triisooctylamine (TIOA) in toluene ($\frac{v}{v}$), and the 10 ml of TIOA added to a scintillation vial containing 15 ml of scintillation mixture.

e^1. The sample was dark-adapted for 24 hr and counted in a liquid scintillation counter.

Finally, experience gained from the acute plutonium study indicated that a considerable amount of time and effort was necessary to measure the activity in biological samples by the barium coprecipitation method. In an intermediate unpublished study, Bretthauer (1973) analyzed 20 fecal samples by both the barium method and by direct counting with a Phoswich Detector. The Phoswich Detector counting system had exhibited a background level of 5.22 ± 0.75 cpm at an efficiency of 0.2%, resulting in a minimum detectable activity level of 1.51 nCi. Results indicated that the Phoswich Detector was acceptable for fecal analyses in the subsequent multiple dose study. Early fecal results were desirable for safety reasons, since the cows were to remain in the metabolism stalls until 99% of the original dose had been recovered.

Therefore, in the multiple dose study, 400-g fecal samples were transported to the counting facility, the outer bag removed, and the polyethylene counting container placed in contact with a 5-in Phoswich Detector. Samples were counted and the concentration calculated in reference to a fecal sample containing a known amount of plutonium-238 (approximately 100,000 dpm). Samples were then double-bagged and stored in case there was a later need for reanalyzing selected samples.

RESULTS AND DISCUSSION

Table 3 presents a summary of the results following the acute plutonium-238 citrate treatment. Sample collection and analyses continued for as long as activity was detectable (approximately 720 hr posttreatment). Transfer to milk was slight (2×10^{-4} percent of dose recovered) and, based on the limited data reported in the literature, not unexpected. The gastrointestinal absorption, however, was somewhat greater than anticipated, resulting in a mean serum activity of 2.5 pCi/g 24 hr after the acute treatment. Plutonium activity in urine and milk was essentially nondetectable 96 hr after dosing, which would appear to be in agreement with the serum half-time of 50 ± 12 hr. Peak activity, recorded as a percent of dose per kg, was approximately three times as high in urine as in milk, but the total percent recovered in these two compartments was surprisingly close, 2.5×10^{-4} and 2.0×10^{-4} for urine and milk, respectively.

The plutonium activity level in the systemic circulation is, of course, one of the primary physiological variables influencing subsequent transfers to both milk and urine. An indication of this is found during the first 72 hr following the acute ingestion of plutonium citrate. Twenty-four hours posttreatment, the mean serum plutonium activity was approximately two times the activity of the formed elements on a pCi/g basis (2.5 to 1.0, respectively). Serum activity (2.03) remained essentially twice that of the formed elements (1.03) at the 48-hr collection. However, by posttreatment hour 72, the two blood

Table 3.

Average Recovery and Kinetic Data from
Four Dairy Cows Following an Acute Oral Dose of
3 mCi Plutonium Citrate*

Compartment	Peak Concentration (% Dose/Unit Measure)	Time to Peak Concentration (hr)	Compartment Half-times ±SD (hr)	Percent Recovered
Feces	2.7 kg	24	15 ± 0.5 252 ± 74	96
Urine	1.3×10^{-5}/kg	24	9 ± 2 >100	2.5×10^{-4}
Milk	4.7×10^{-6}/kg	24	16 ± 3 >100	2.0×10^{-4}
Blood				
1. Serum	6.1×10^{-5}/g	24	50 ± 12 >250	- - - -
2. Formed Elements	2.9×10^{-5}/g	96	398 ± 77	- - - -

*Stanley *et al.*, in preparation.

compartments had nearly equal activities; *i.e.*, 1.2 and 1.0 pCi/g for serum and formed elements, respectively. It should be noted that the mean activity of the formed elements changed very little from 24 to 72 hr posttreatment, while serum activity decreased to one-half the peak value recorded 24 hr after dosing (Table 4). Observations on milk and urine during this first 72 hr revealed a drop to approximately one-fifth peak activity for milk (0.19 to 0.03 pCi/ml), and a drop to approximately one-fourth peak activity for urine (0.52 to 0.12 pCi/ml). Therefore, the initial rise to peak activity and subsequent fall in activity for both milk and urine did not correspond so much to changes of plutonium activity in the formed elements, but, as expected, to changes in serum activity (Fig. 1). The exchangeable portion for most heavy elements is contained in the plasma, and the activity associated with the formed elements of the blood is essentially bound and nonexchangeable. This is apparently the case following plutonium ingestion in dairy cattle, where a significant correlation ($P<0.05$) between serum and urine activity, as well as between serum and milk activity, was observed following the acute plutonium citrate treatment (Fig. 2). While correlations obviously do not establish cause and effect relationships, these results, along with results from future investigations on dairy cattle, should provide more insight into the specific transport processes.

Table 5 shows the distribution of plutonium-238 in cow tissues on both an activity per gram of tissue basis and as total activity per organ. These data are average values for the two animals sacrificed. The two cows weighed approximately the same (652 and 677 kg) and both sacrifices were accomplished three months after dosing. In addition, one cow contained a 115-day fetus, and the fetal activity levels are also presented. On a pCi per organ basis, 70% of the bovine carcass activity was recovered in bone, with the concentration in the vertebrae being about five times higher than that noted in the femur. Plutonium was also observed to be transmitted across the bovine placenta, where the fetal concentration was in excess of 1.3×10^{-6}% of the total dose. Finkel (1947) presented one of the earliest reports on placental transport of plutonium and observed that following injections (0.06 to 0.016 μCi/g) to pregnant mice, the transferred activity varied inversely with the original dose. Wilkinson and Hoecker (1953) also observed the apparent inverse relationship following plutonium injection and noted that the placental content of plutonium-239 was higher than the corresponding fetal value. This situation was not observed in the one fetus sampled during the acute phase in dairy cattle, where the fetal liver had a greater activity than the placenta (0.09 and 3.0 pCi/g for the bovine placenta and fetal liver, respectively). In a relatively recent investigation, Sikov and Mahlum (1968) injected intravenous doses of plutonium-239 citrate and polymeric plutonium into rats and also ovserved that the fraction of injected plutonium which reached the fetus was very small, being approximately 0.03 and 0.001% of the administered dose per fetus for the ionic and polymeric forms, respectively.

Table 4.

Mean Blood Plutonium Activity in Four Holstein Dairy Cows Following an Acute Oral Dose of 3 mCi Plutonium Citrate

Posttreatment Time (hr)	Mean Blood Plutonium Activity (pCi/g)	
	Serum	Formed Elements
24	2.5	1.0
48	2.0	1.0
72	1.2	1.0
96	1.4	1.1
120	0.9	1.0
144	0.9	0.9
168	0.8	0.8
192	0.6	0.8
216	0.5	0.7
240	0.5	0.8
264	0.6	0.8

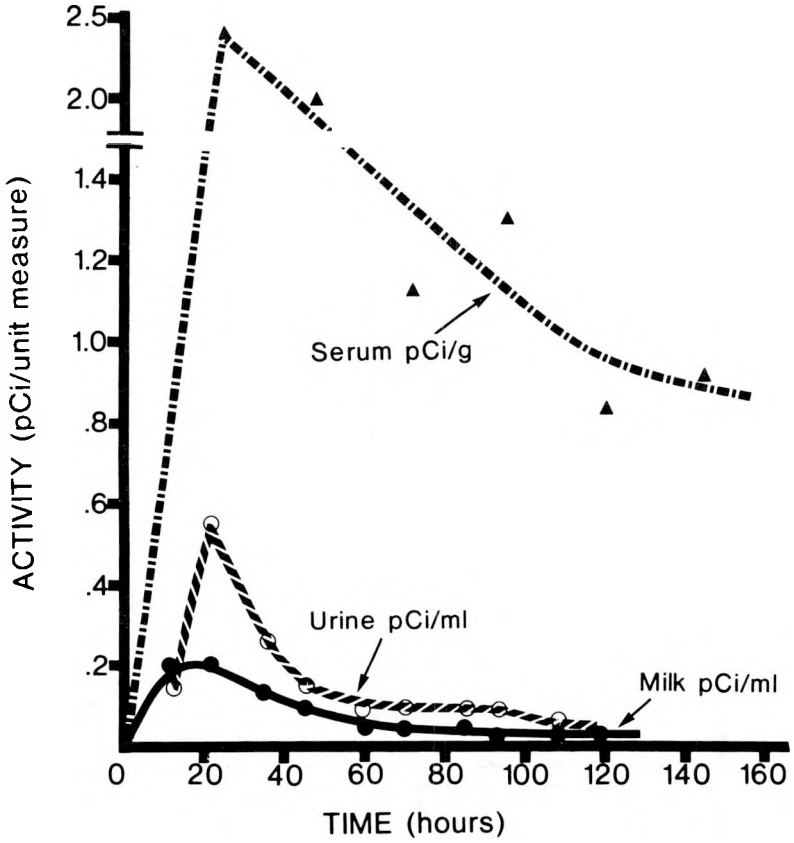


Figure 1. The relationship between plutonium activity in milk, urine, and serum, plotted on the vertical axis in pCi per unit measure, and time in hours plotted on the horizontal axis. All values represent a four-cow average following an oral plutonium citrate treatment of 3 mCi per animal.

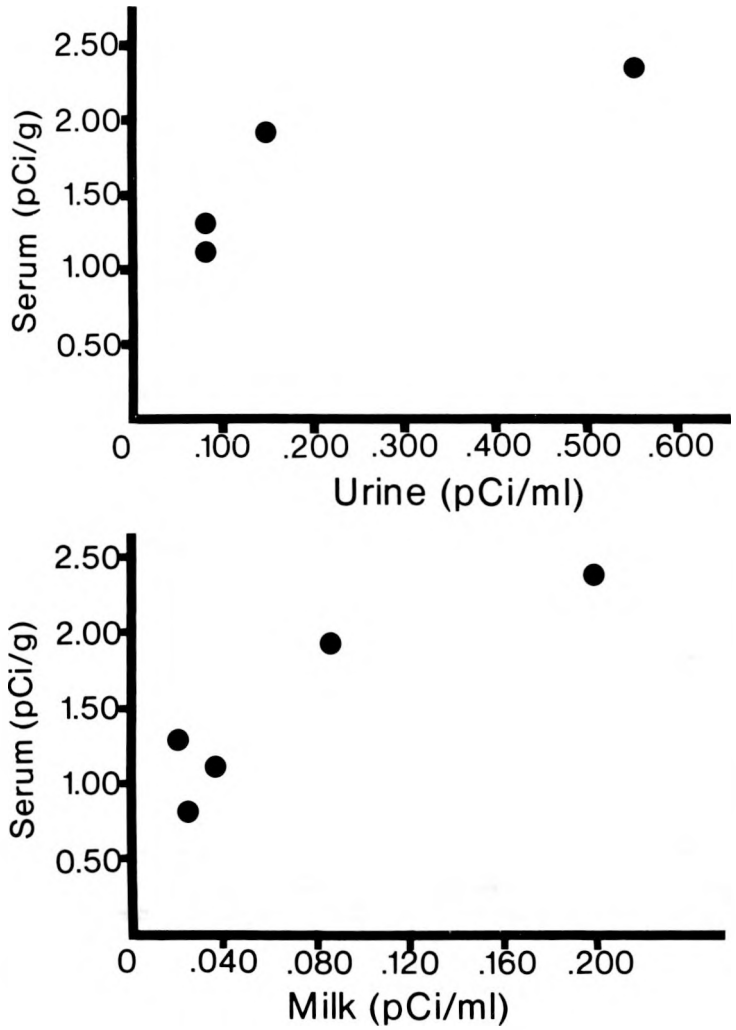


Figure 2. The relationship between either urine activity or milk activity, plotted in pCi/ml on the horizontal axis, and the serum activity plotted in pCi/g on the vertical axis. All values represent a four-cow average following an oral plutonium citrate treatment of 3 mCi per animal.

Table 5.

Mean Tissue Distribution of Plutonium-238
in Holstein Dairy Cows Three Months
After an Acute Oral Dose of 3 mCi Plutonium Citrate*

	Number of Animals	Mean Weight	Tissues	Concentration (pCi/g)	Recovery (pCi/organ)
Mature Animals	2	665 kg	Bone	7.0	725,000
			Liver	11.0	105,000
			Muscle	0.2	60,000
			Small Intestine	6.0	42,000
			Rumen	0.9	4,600
			Heart	0.9	2,800
Fetus	1	793 g	Fetal Fluid	0.02	-
			Fetal Placenta	0.09	-
			Fetal Liver	3.0	50
			Fetal Bone	0.4	-

*Stanley *et al.*, in preparation.

Data from the multiple dose study (Table 6) must be considered as preliminary at this stage because the results from the quality control program, a process of reanalyzing random samples by an alternate method (Talvitie, 1972) to ensure reproducibility, have not been received. In addition, the fact that no blood samples nor necropsy tissues have been analyzed further restricts metabolic interpretations. However, it is possible to report at this time the transfer of chronically ingested plutonium dioxide to milk, albeit in small quantities (approximately 2.0×10^{-5}% recovered in milk). There was considerable variation in milk activity between animals on a day-to-day basis (13-830 pCi/day), as well as some variation in milk production (12-30 liters per day), but plutonium activity was noted in all milk collections from treatment day nine through the first posttreatment day. Urine activity, which essentially reached a plateau on day three, attained maximum value on treatment day 15 and demonstrated a precipitous decline on the second posttreatment day. The percentage of total dose recovered in urine was approximately 1.7×10^{-4}. Total fecal contamination, which closely approximates 100% of dose, reached a somewhat constant activity plateau on treatment day four and, as expected, did not decline until the second posttreatment day. An outline of these data is presented in graphic form in Fig. 3 to illustrate relative concentrations in terms of total activity.

During the multiple dose treatments, the air conditioner, normally in operation to ensure appropriate temperatures throughout metabolic studies, temporarily stopped functioning and placed the four cows in some degree of heat stress. European breeds of cattle, such as the Holsteins used in this project, are not considered to be physiologically well adapted to hot desert climates (Schmidt-Nielsen, 1964). As the temperature rises, cattle will consume increased amounts of water and there is a parallel increase in urine output, with increased frequency of urination and decreased concentrations (Thompson *et al.*, 1949). A slight increase in urine output was observed as the multiple dose study progressed, but most of the daily excretions were within the accepted normal range of 9.2 to 23.5 liters (Dukes, 1955). Mean daily urine output during the chronic treatment was 20 liters, with quantities ranging from 13.4 to 25.1 liters. However, mean fluctuations noted in urinary activity, recorded as pCi per liter, were somewhat reduced when these data were expressed in terms of total activity per day.

A comparison of the acute study and chronic study in dairy cows reveals that, as expected, the major portion of plutonium activity (approximately 96% in Group I and slightly less than 100% in Group II) was excreted in the feces. However, recovered activity in urine and milk following both the acute dose of plutonium citrate and the multiple doses of plutonium dioxide confirmed the physiological uptake and transport of both chemical forms. Total plutonium transport to milk was not great and, on a percent of oral dose basis, was observed to be 2×10^{-4} and 2×10^{-5} following the plutonium citrate and plutonium dioxide treatments, respectively (Table 7). At peak milk activity during the chronic plutonium dioxide treatment, contamination

Table 6.

Average Recovery and Kinetic Data from Four Dairy Cows
Following Chronic Ingestion of Approximately
1 mCi Plutonium Dioxide for 19 Consecutive Days

Compartment	Peak Concentration (% Dose/kg or l)	Time to Peak Concentration	Compartment Half-times ± SD* (hr)	Percent Recovered
Feces	5.8×10^{-1}	treatment day 4	9 ± 0.9 150	approaching 100
Urine	1.3×10^{-6}	treatment day 15	9 ± 3	1.7×10^{-4}
Milk	2.7×10^{-7}	treatment day 11		2.0×10^{-5}

*NOTE: Compartment half-time calculated beginning on posttreatment day 1.

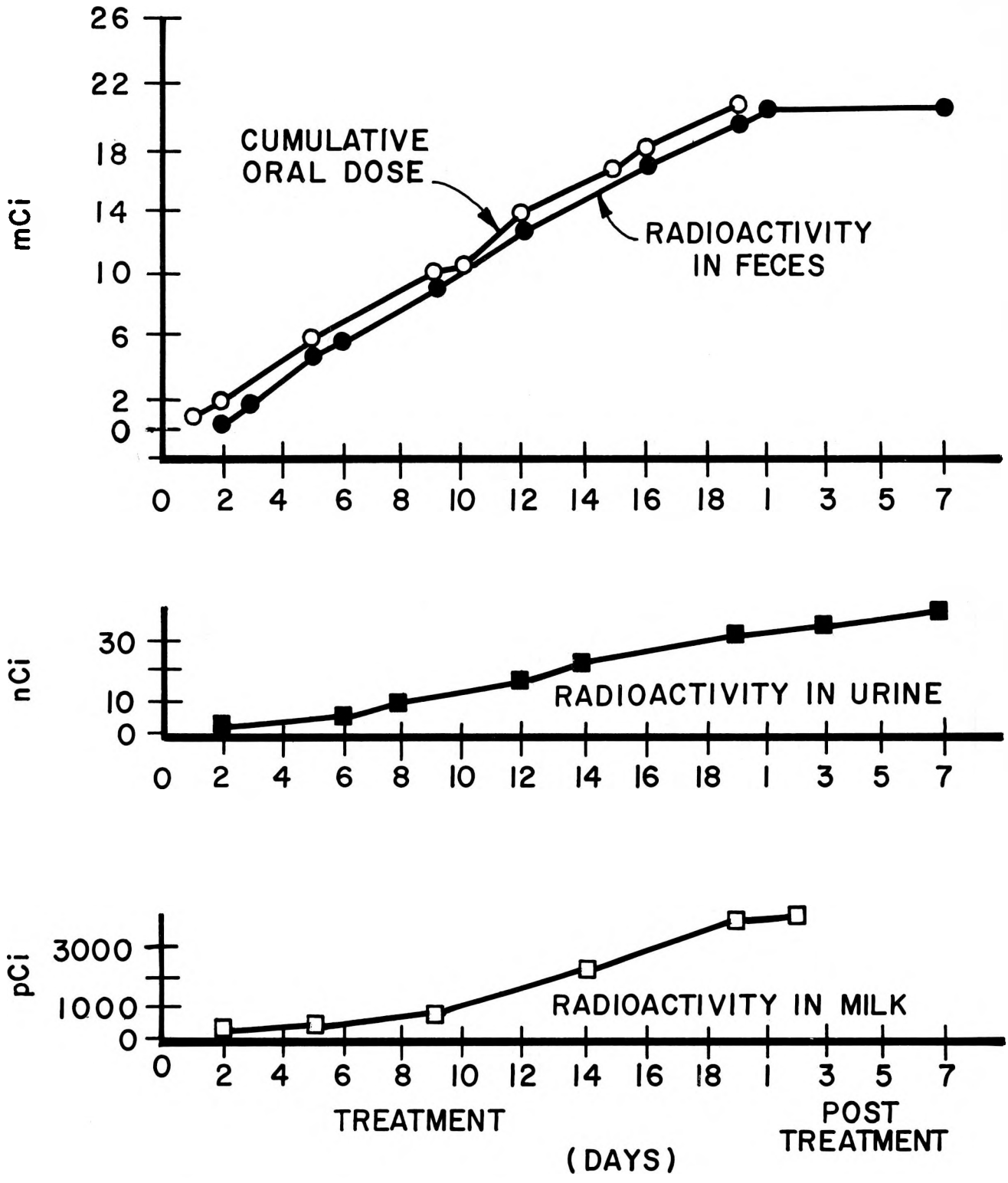


Figure 3. The relationship between total recovered radioactivity in feces, urine, and milk, plotted in mCi, nCi, and pCi, respectively, on the vertical axis, and time in days plotted on the horizontal axis. The total oral dose of 1 mCi plutonium dioxide per day is also plotted in mCi on the vertical axis.

Table 7.

Comparative Kinetic Results for Milk Following
Acute Ingestion of Plutonium Citrate and Chronic
Ingestion of Plutonium Dioxide by Holstein Dairy Cattle

Oral Treatment	Duration of Treatment	Number of Animals	Mean Weight	Mean Activity in Milk: Percent of Dose Recovered	Mean Activity in Milk: Peak Concentration (% Dose/Liter)
3 mCi Plutonium Citrate	One Treatment	4	700 kg	2.0×10^{-4}	4.9×10^{-6}
1 mCi Plutonium Dioxide	One Treatment Per Day For 19 Consecutive Days	4	650 kg	2.0×10^{-5}	2.7×10^{-7}

with approximately 10 mg of feces per liter of milk would have accounted for all of the activity detected in the milk. However, as noted in the methods section of this paper, every reasonable precaution was taken to prevent the occurrence of cross-contamination and it is considered unlikely. Furthermore, cross-contamination of urine with fecal material was also unlikely under the collection techniques employed, and the subsequent urine/milk activity ratios noted in this report were in agreement with results from past studies on other heavy elements; *e.g.*, iron (Mullen *et al.*, 1971). It is not known whether the observed plutonium activity in bovine milk resulted from intracellular incorporation by the secretory epithelial cells or, among other possibilities, was bound to a portion of the cell membrane that was subsequently discharged along with the intracellular contents. The blood supply to the bovine mammary gland during lactation is profuse and amounts to approximately 400 volumes of blood to one volume of milk produced (Graham *et al.*, 1936). Therefore, bloodborne plutonium becomes available for clearance or removal by the mammary gland and, along with the other milk constituents, probably gains admission to the epithelial cells lining the alveoli. If the chemical nature and quantity of plutonium entering and leaving the mammary gland were known, along with blood flow, it would be possible to present a more definitive statement. However, this has been historically a difficult physiological problem (Smith, 1959), since in addition to the many veins leaving the gland, there is also an abundant lymph drainage.

The occurrence of plutonium activity in milk has been observed in other domestic animals. In lactating Suffolk sheep (McClellan *et al.*, 1962), following a single plutonium nitrate injection, milk activity reached peak levels approximately 12 hr after treatment, a situation not unlike the rapid appearance of peak activity in bovine milk 12 to 24 hr after acute ingestion of plutonium citrate. However, in the case of chronically ingested plutonium dioxide, the bovine milk activity reported in this paper was not consistently detectable until the ninth treatment day. This slightly delayed transfer is somewhat in agreement with Sansom (1964) who reported that, following an acute oral plutonium dioxide dose in dairy cattle, peak milk activity did not occur until the fourth posttreatment day.

Early studies on a lactating cat which received an intraperitoneal plutonium injection of 0.097 mCi (0.03 μCi/g) eight days after parturition (Finkel, 1947) produced milk containing 0.003 μCi/ml. As with the bovine observations reported in this paper, the subsequent milk levels were extremely low, but they were not totally negligible due to the relatively high plutonium retention noted in some juvenile animals that have ingested contaminated milk. A kitten receiving the above-mentioned milk (0.003 μCi/ml) for 16 days was retaining, at the time of sacrifice 24 days postpartum, 0.0016 μCi with each gram of body weight gained.

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ABSORPTION, DISTRIBUTION, AND EXCRETION OF PLUTONIUM BY DAIRY CATTLE

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ABSTRACT

In order to obtain information on the potential transport of ingested plutonium and to gain additional information on plutonium deposition patterns in ruminants, a series of metabolism studies with dairy cows have been conducted. This paper, which reviews the results on tissue uptake following oral plutonium administration, serves as an addendum to the more comprehensive report presented at the 1973 Meeting of the Nevada Applied Ecology Group. Some of the information from the previous report has been incorporated into this paper to establish the overall objectives and to make certain comparisons; however, the more complete documentation of this investigation has already been published (Stanley, *et al.*, 1974).

Two groups of Holstein dairy cows, four cows in each group, were studied to examine the physiological transport of ingested plutonium citrate and plutonium dioxide. Approximately 3 mCi of plutonium citrate per animal was administered in an acute treatment to the first group, while each animal in the second group received 1 mCi of plutonium dioxide daily for 19 consecutive days. Samples of blood, milk, urine, and feces were taken during and after oral dosing, while tissue collections were made at necropsy 6 to 13 weeks after treatment.

As expected, the major portion of plutonium activity (approximately 96% in Group I and slightly less than 100% in Group II) was excreted in the feces. However, recovered activity in urine and milk following both the acute dose of plutonium citrate and the multiple doses of plutonium dioxide confirmed the bovine gastrointestinal uptake and transport of this radionuclide. In both Groups I and II, the total deposition of plutonium was greatest in bone, liver, and skeletal muscle. The duodenum and other portions of the small intestine also had fairly high levels, but unabsorbed plutonium may have contributed to these values. While the results following the different plutonium treatments are not strictly comparable, these data have shown tissue uptake and retention of plutonium following oral administration of both chemical forms.

INTRODUCTION

Because dairy animals consume large quantities of food and water, and since animal products form a direct link in man's food chain, bovine metabolism of ingested plutonium is of considerable importance for an analysis of exposure pathways to human populations. A research project was therefore undertaken to study the problem in lactating Holstein dairy cows, insofar as the relationship between activity ingested, uptake by systemic circulation, tissue retention, and transport to milk are concerned, as well as to investigate the transport to different chemical forms of plutonium administered under acute and chronic conditions. This paper, which reviews the results on bovine tissue uptake following oral plutonium exposure, serves as an addendum to the more comprehensive report presented last year at the Nevada Applied Ecology Group Meeting.

Several studies in the literature have addressed the metabolism of plutonium once it enters the systemic circulation. Investigators have frequently noted that a large percentage of intravenously injected plutonium was lost from the blood, often removed in colloidal aggregates by the liver shortly after injection (Painter *et al.*, 1946; Stover, *et al.*, 1959). Plutonium remaining in the blood for longer periods was largely combined with protein (Painter *et al.*, 1946; Stover *et al.*, 1959; Beliayev, 1959). But while the physical state of injected plutonium clearly influenced the tissue distribution pattern, variations between species, treated under identical conditions, have been noted (Rosenthal *et al.*, 1972).

Basic physiological studies have established that amphibians, reptiles, birds, and mammals possess the cellular elements to directly or indirectly affect the exchange of ions between bone and the major extracellular fluid compartments. It is also well known that plutonium is deposited in bone and that variations exist between the uptake for different bones (i.e., comparison of vertebrae with femur) as well as at different sites on the same bone. Membranous tissue surrounding the bone (periosteum), as well as that lining the marrow cavities and haversian canals (endosteum), has been shown to play a part in this pattern of plutonium distribution. Hamilton (1947) was the first to describe this superficial deposition of plutonium on the periosteum, endosteum, and the covering of the trabecular bone. More recent reports (Arnold and Jee, 1957; Jee, 1972) have discussed the sequence by which plutonium accumulates in the bone cells. Apparently, the initial deposits of plutonium are removed from the bone surfaces by osteoclastic resorption. The plutonium-labeled osteoclasts break up and a portion of the plutonium is released into the blood while some is phagocytized by hemosiderin-laden macrophages. Evidence also suggests that hemosiderin and plutonium-containing macrophages may subsequently be retained in the bone marrow.

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Schubert *et al.* (1961) reported the tissue distribution pattern following I.V. injections of monomeric and polymeric ^{239}Pu in mice and noted the relatively even distribution of monomeric plutonium between liver and bone (values calculated from the mean femur content). For mice injected with the polymeric solution of plutonium, the amount deposited in the liver was more than twice that of bone and, relative to the monomeric retention pattern, more of the radioelement was retained in the liver. Results from longer-lived animals have emphasized the potential hazard of liver-deposited plutonium. Stover *et al.* (1959) discussed the metabolism of intravenously injected ^{239}Pu in beagle dogs and noted the long-term liver retention. Further studies on the beagle (Stover *et al.*, 1968; Stover *et al.*, 1971) demonstrated that there was a dose level effect, associated with radiation damage, on the hepatic uptake in canines after different injected doses of plutonium.

The current investigation with dairy cows was undertaken by (1) examining the amount of ingested activity transferred to the systemic circulation, (2) determining the amount of activity transported to milk, (3) establishing the portion of activity retained in the tissues following initial absorption, and (4) evaluating the above phenomena after various chemical forms of the element were administered. In pursuing these objectives, samples of blood, milk, urine, and feces were taken from two groups of Holstein dairy cows before, during, and after oral treatment with different chemical forms of plutonium. Tissue samples were collected at necropsy.

Some of the results and literature citations from the 1974 Nevada Applied Ecology Group Progress Report have been incorporated into this paper to establish the overall study objectives. However, for a more complete outline of this investigation as well as the results on plutonium excretion and transfer to milk, the reader is referred to Stanley *et al.* (1974).

METHODS AND MATERIALS

Two groups of four lactating dairy cows per group were given oral doses of plutonium. Treatment aliquots were initially placed in 1/2-oz gelatin capsules. The 1/2-oz capsules were sealed in larger 1-oz capsules and subsequently administered by use of a balling gun.

Group I cows (mean weight 700 kg) received an acute treatment of 3 mCi plutonium citrate per animal while Group II cows (mean weight 650 kg) received 1 mCi plutonium dioxide per animal per day for 19 consecutive days. Samples of blood, milk, urine, and feces were taken during and after dosing, while tissue collections were made at necropsy 6 to 13 weeks after treatment (Table 1).

Acute doses were prepared by adding 15 ml of citrate buffer to 0.5 ml of concentrated H_3PO_4 (Cleveland, 1967; Holley *et al.*, 1958) containing 15 mCi of plutonium-238. Doses (pH 3.3-3.5) were quantitated by comparing

Table 1. Collection Schedule for Urine, Milk, Feces, and Blood

Treatment	Approximate Animal Weight (kg)	Duration of Treatment	Urine	Milk	Feces	Blood
3 mCi plutonium citrate per animal	700	One Treatment	One collection per animal was taken on the treatment day, followed by twice-a-day collections through post-treatment day 24.	One collection per animal was taken on the treatment day, followed by twice-a-day collections through post-treatment day 24.	Fecal samples were taken two times a day from each cow for a period of 24 days after treatment. Duplicate samples were collected at each sampling period.	One sample per animal was taken daily for 24 days after treatment and again at necropsy on post-treatment day 93.
1 mCi plutonium dioxide per animal per day	650	One treatment per day for 19 consecutive days	One collection per animal of urine was taken on treatment day one, followed by twice-a-day collections through post-treatment day 7.	One collection per animal was taken on treatment day one, followed by twice-a-day collections through post-treatment day 7.	Fecal samples were taken two times a day from each cow from the second treatment day through post-treatment day 7. Duplicate samples were collected at each sampling period.	One sample per animal was collected daily from treatment day one through recovery day 7 and again at the necropsies on post-treatment days 42 and 73.

aliquots with plutonium-238 standard solutions using a liquid scintillation technique. Plutonium-238 dioxide microspheres were obtained from the Lovelace Foundation, Albuquerque, New Mexico. These microspheres had a count median diameter, geometric standard deviation, and mass median diameter of 0.0600, 2.3476, and 0.5336 mm, respectively. The diameter of the average microsphere was 0.1790 mm. Approximately 100 mCi of the particles were suspended in 50 ml of a 2% solution of Dowfax 2A-1- distilled water surfactant. Aliquots were then analyzed by liquid scintillation to determine the plutonium-238 concentration. Individual doses were prepared by removing 500 µl aliquots from the suspension and placing them in gelatin capsules.

Animals were placed in metabolism stalls a minimum of 5 hr before plutonium administration. An in-dwelling inflatable catheter was used for urine collection; the cows remained catheterized from the time they entered the stalls until being transferred to outdoor pens two weeks after termination of the last dosing. A discussion of fecal, urine, milk, and blood collection procedures has been presented previously. At time of sacrifice, complete tissue weights were taken of all organs, except skin. This obviously was not always practical in the case of some other tissues; e.g., muscle, bone, skin, blood, etc. Under these conditions, estimates of tissue weight were made based on a percentage of the total body weight.

Table 2 presents an outline of the tissues taken at necropsy. It should be noted that the Group II cows were sacrificed at different times after treatment. Therefore, for purposes of a plutonium tissue comparison, Table 2 shows three groups of animals. Standard necropsy procedures (Olafson, 1954) were used to collect the various tissues, but for a more detailed anatomical description, the reader is referred to Sisson and Grossman (1938).

Analytical methodology (Table 3) has been discussed previously with the exception of the methods used for Group II blood and necropsy tissues. Assay procedures for these Group II collections are outlined below. So that appropriate comparisons could be made between Groups I and II, especially with reference to plutonium tissue distribution patterns, a series of quality control samples were submitted for analysis by both methods. Differences in the results were negligible, thereby permitting a direct comparison between the samples obtained from the animals in both groups.

Methods employed for the Group II tissue anlaysis were essentially those of Talvitie (1971, 1972). Whole samples were dry ashed in a muffle furnace with the temperature controlled at 550°C or less, to avoid fusion of the ash. Resultant ash was ground, homogenized, and an aliquot removed for further analysis. After adding plutonium-236 tracer, the sample was digested with concentrated nitric acid to remove residual carbon. The digested residue was dissolved in 9M hydrochloric acid, and passed through an anion exchange column which retained the plutonium. Interfering iron was removed from the column by a nitric acid wash, and the plutonium was selectively eluted with dilute hydrochloric acid. Plutonium was then electroplated from a sulfate solution on stainless steel discs. The discs were counted on an alpha spectrometer for both plutonium-238 and

Table 2. Background Information for the Discussion of Plutonium Tissue Retention Patterns in Dairy Cattle

Treatment	Duration of Dosing	Number of Animals	Mean Wt. (kg)	Sacrifice Time	Tissues Collected
Oral dose of plutonium-238 citrate	One dose 3 mCi per animal	2	665	93 days post-treatment	Liver, bone, kidneys, spleen, skeletal muscle, cardiac muscle, skin, rumen, abomasum, duodenum, small intestine, bile, blood, and lung.
Oral doses of plutonium-238 dioxide	1 mCi per animal per day for 19 consecutive days	2	603	42 days post-treatment	Liver, bone, kidneys, spleen, pulmonary lymph nodes, skeletal muscle, cardiac muscle, skin, rumen, abomasum, duodenum, small intestine, bile, blood, and lung.
Oral doses of plutonium-238 dioxide	1 mCi per animal per day for 19 consecutive days	2	733	73 days post-treatment	Liver, bone, kidneys, spleen, pulmonary lymph nodes, skeletal muscle, cardiac muscle, skin, rumen, abomasum, duodenum, small intestine, bile, blood, and lung.

Table 3. Analytical Methods to Determine Plutonium-238 Activity in Biological Collections

Treatment	Materials Collected				
	Milk	Blood	Urine	Feces	Necropsy Tissue Samples
Acute dose of 3 mCi/animal of plutonium-238 citrate	Modified method of Sill (1969) Barium Sulfate	Modified method of Sill (1969) Barium Sulfate	Modified method of Sill (1969) Barium Sulfate	Modified liquid scintillation method of Butler (1968)	Modified method of Sill (1969) Barium Sulfate
One mCi/animal/ day for 19 consecutive days of plutonium-238 dioxide	Modified method of Sill (1969) Barium Sulfate	Sample wet ashed, extracted, electroplated, followed by spectrometric determination	Modified method of Sill (1969) Barium Sulfate	Direct quantitation of the 44-keV X ray of plutonium using a Phoswich Detector	* Ion exchange and alpha spectrometric determination, Talvitie (1971, 1972)

plutonium-236. Sample activity was calculated by using the total plutonium-238 counts adjusted by the percent recovery of the plutonium-236 tracer.

Blood samples for the Group II animals were analyzed on contract with Eberline Instrument Corporation. A 10-g aliquot of formed elements, or 20-g aliquot of serum, was initially spiked with plutonium-242 tracer and then wet ashed with concentrated nitric acid until the ash was white. This was followed by dissolution of the sample in nitric acid and a sodium nitrite valence adjustment to Pu IV. Plutonium was then extracted into a 30% Aliquot-336 xylene solution and eluted with a perchloric acid-oxalic acid mixture. The eluate was evaporated and plutonium converted to the chloride form with concentrated hydrochloric acid and reevaporated. A hydrochloric acid-ammonium oxalate solution was then used to dissolve the residue. The plutonium was electroplated from this solution. The planchet was counted on an alpha spectrometer for both plutonium-238 and plutonium-242. Sample activity was calculated by using the total plutonium-238 counts adjusted by the percent recovery of the plutonium-242 tracer.

RESULTS AND DISCUSSION

Table 4 presents the percent of total plutonium dose retained by selected bovine tissues following both the acute plutonium-238 citrate administration and the multiple doses of plutonium-238 dioxide. Percentages noted in the table were based on the highest value found in the two cows sampled. Therefore, this information suggests an estimated upper range of retention since bone, skeletal muscle, skin, and blood calculations were based on a fraction of the total body weight. While the tissue results following the two different plutonium treatments are not strictly comparable, these data do show absorption and retention of plutonium following exposure to both chemical forms. There is also a similar plutonium tissue distribution pattern for both chemical forms.

As expected, bone and liver contained fairly high concentrations of the retained plutonium activity in animals from all sacrifice groups. Total skeletal muscle activity closely approximated the liver activity in some cases but was, of course, based on a much greater mass of tissue. Blood, taken at time of sacrifice, was also relatively high in plutonium content. The duodenum and other portions of the small intestine had fairly high concentrations, but unabsorbed plutonium may have contributed to these values. Detailed tissue results following multiple doses of plutonium dioxide are presented in Appendix I.

Sikov *et al.* (1968) reported on the passage times of ingested plutonium oxide particles which had been administered by stomach tube to adult and juvenile rats. These rats were serially sacrificed, at which time sections

Table 4. Approximate Percentage* of Oral Plutonium Dose Retained in Selected Bovine Tissues at Time of Sacrifice

Tissue	Multiple Doses of Plutonium Dioxide 1 mCi/Animal/Day for 19 Consecutive Days		Acute Dose of Plutonium Citrate 3 mCi/Animal
	Sacrifice 42 Days Post-Treatment (2 Cows)	Sacrifice 73 Days Post-Treatment (2 Cows)	Sacrifice 93 Days Post-Treatment (2 Cows)
Liver	5.4×10^{-4}	6.0×10^{-4}	3.5×10^{-3}
Bone	3.1×10^{-3}	3.2×10^{-3}	2.4×10^{-2}
Kidney	8.7×10^{-6}	1.3×10^{-5}	2.9×10^{-5}
Spleen	1.1×10^{-5}	8.3×10^{-6}	3.3×10^{-5}
Blood	5.6×10^{-5}	5.2×10^{-5}	1.1×10^{-4}
Lung	3.0×10^{-5}	2.2×10^{-5}	7.7×10^{-5}
Skeletal Muscle	8.8×10^{-5}	1.4×10^{-4}	2.0×10^{-3}
Cardiac Muscle	4.7×10^{-6}	4.1×10^{-6}	9.6×10^{-5}
Rumen	8.6×10^{-6}	8.9×10^{-6}	1.5×10^{-4}
Abomasum	8.1×10^{-6}	3.6×10^{-6}	7.0×10^{-6}
Duodenum	2.5×10^{-5}	1.0×10^{-4}	8.3×10^{-5}
Small Intestine	2.0×10^{-3}	1.2×10^{-4}	2.0×10^{-3}

*Percentages are based on the highest tissue value found in the two cows sampled. Therefore, these percentages suggest an upper range of retention and not a two-cow average. Furthermore, if complete organ weights could not be taken (blood, bone, skeletal muscle, etc.), estimates were made based on a percentage of the total body weight.

of the digestive tract (including contents) were collected. A progressive reduction in activity with time after dosing was noted, but a few very high and very low values were also reported. The current study on dairy cows did not analyze intestinal contents, and the intestinal segments were superficially washed with water before being placed in the collection containers. However, isolated hot spots of unabsorbed plutonium may have been present. Since the quantity of absorbed plutonium was the primary concern, these activity concentrations noted in the bovine gastrointestinal tissues should be viewed as being peripheral to the study objectives.

Since the liver was a major organ of plutonium deposition in these dairy animals, it was of interest to examine the activity of plutonium found in bile at the time of sacrifice. Biliary plutonium ranged from nondetectable to 5.6×10^{-1} pCi/g when samples from both treatment groups were compared (Table 5). An estimated percentage of plutonium excreted in the bile of these animals would probably be of doubtful physiological significance. Several variables including blood flow to the liver and the digestive state of the animal affect the volume of bile produced; and, some question remains on the mechanism and rate of plutonium transfer from liver or blood to bile. Ballou and Hess (1972) demonstrated the predominant role of bile in the early postinjection phase of intestinal plutonium excretion and suggested that at least part of the injected radionuclide entered the biliary system directly from the blood. However, the studies on dairy cattle reported here do not allow for an estimate of the total fecal activity that resulted from bile. Intravenous plutonium doses to dairy cows would produce more definitive information on this point.

Bone marrow samples were not consistently taken; however, marrow samples (126 and 123 grams) were collected from the femur of the two plutonium dioxide dosed animals that had been sacrificed 73 days post-treatment. Plutonium concentrations of 0.091 and 0.021 pCi/g were noted in these marrow collections. This corresponded to activity levels of 0.90 and 0.98 pCi/g, respectively, observed in the mineralized portion of the bones. With this limited information, no attempt was made to establish a ratio of marrow activity to total retained skeletal activity. Furthermore, the cellular distribution pattern of plutonium within various bovine tissues was not within the overall objectives of this study and more refined techniques would, in most cases, be required to provide such information.

Many investigators have examined the cellular and subcellular distribution patterns of plutonium in various species and this topic is definitely of current research interest. Boocock *et al.* (1970), utilizing differential centrifugation and centrifugation through a sucrose density gradient, showed that the subcellular distribution of plutonium in the rat liver was predominantly associated with the lysosomes. Studies with beagle dogs have also noted that hepatic plutonium concentrations were highest in the mitochondrial-lysosomal

Table 5. Activity in Bile at Time of Sacrifice in Animals Receiving Oral Doses of Plutonium

Treatment	Time of Sacrifice	Number of Animals	Individual Activity Levels (pCi/g)	Total Activity (pCi)
Acute dose of 3 mCi/animal of plutonium citrate	93 days post-treatment	2	5.0×10^{-2}	1
			nondetectable	--
One mCi/animal/day for 19 consecutive days of plutonium dioxide	42 days post-treatment	2	2.0×10^{-1}	126
			6.8×10^{-2}	46
One mCi/animal/day for 19 consecutive days of plutonium dioxide	73 days post-treatment	2	5.6×10^{-1}	406
			6.4×10^{-2}	48

fractions and microsomal fractions (Bruenger *et al.*, 1971). Differences in the amount of hepatically deposited plutonium and differences in the intraorgan distribution pattern have also been observed following injections of monomeric and polymeric material. Autoradiographs of beagle tissue have indicated that initially monomeric plutonium is found largely in random clusters in reticuloendothelial cells (Bruenger *et al.*, 1972). Based on studies using mice intravenously injected with plutonium-239, Rosenthal *et al.* (1972) calculated the proportion of total skeletal plutonium found in the bone marrow. For monomeric, midrange polymeric and highly polymeric plutonium, these investigators determined the percentage of skeletal plutonium in marrow (5-6 days after injection) to be 2, 7-15, and 62%, respectively.

It was obviously important during the current study to ensure that the bovine skin collections did not contain any external contamination. In this regard, most of the administered plutonium activity had been excreted before the cows were moved from the metabolism stalls to the outdoor pens. The cows were also washed with water just prior to leaving the metabolism stalls. Furthermore, at the time of sacrifice, skin samples were shaved and superficially washed before being placed in the sample containers. In spite of these precautions to prevent external contamination, the values listed below for the skin should probably be viewed with caution. For the plutonium citrate treated animals, skin activity values ranged from 0.2 to 0.4 pCi/g, while values ranging from 5.3×10^{-2} to 3.8 pCi/g were noted for animals that received the dioxide doses. The 3.8 pCi/g sample was taken from one of the cows sacrificed 73 days after the last treatment. If this value were representative of a theoretical total skin collection, the extrapolated activity would suggest that approximately 1.0×10^{-3}% of the ingested dose was retained by the skin which would appear to be abnormally high in light of the other tissue retention observations.

The number of bovine pulmonary lymph nodes varies from one cow to another but an observed nodal activity ranging from 7.6×10^{-1} to 3.3×10^{-2} pCi/g confirmed the lymphatic uptake of plutonium following the oral dioxide doses. This was not totally unexpected, since the walls of the lymphatic capillaries offer little resistance to the flow of fluids or particulate matter from the interstitial spaces into the lymphatic channels. Following intradermal implants, other investigators have noted varying levels of plutonium activity in the lymph nodes depending on the node location in the drainage pattern (Cable *et al.*, 1962; Johnson *et al.*, 1970). Inhalation exposures have also resulted in significant plutonium concentration in the lymphatic system (Langham *et al.*, 1962).

Plutonium activity was also noted in the blood immediately after the initiation of oral plutonium dioxide dosing. As expected, there was a dramatic rise in serum activity which reached somewhat of a plateau by the third dosing day (Fig. 1). This serum plateau level

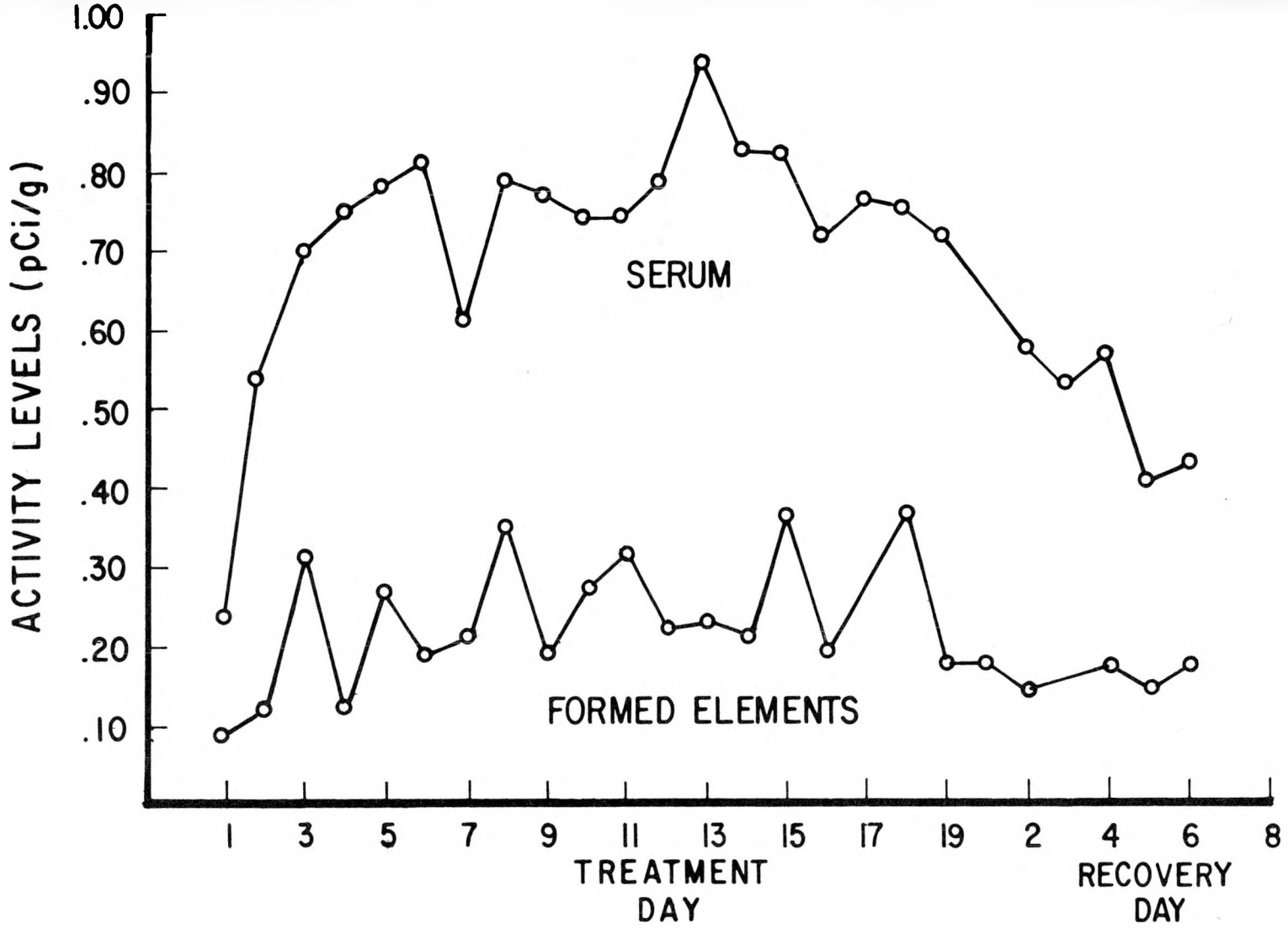

FIGURE 1. MEAN PLUTONIUM ACTIVITY CONCENTRATIONS IN THE BLOOD OF FOUR HOLSTEIN DAIRY COWS RECEIVING ORAL DOSES OF 1 mCi PLUTONIUM-238 DIOXIDE PER ANIMAL PER DAY FOR 19 CONSECUTIVE DAYS.

was essentially maintained throughout the treatment period until day 19, at which time a definite reduction in activity occurred. For comparative purposes, the plutonium excretion in urine during this time is also presented (Fig. 2). Mean serum plutonium concentrations reached a peak of 0.94 pCi/g on treatment day 13, while the peak mean formed element activity (0.36 pCi/g) was reached on day 18. By the sixth postexposure day, the mean serum and formed element activities had decreased to 0.42 and 0.17 pCi/g, respectively. There was no immediate explanation for the occasional high values noted in the formed elements during certain collections, but the general trend (excluding isolated outliers) appeared relatively consistent. While no attempt was made to establish a biological half-life for the plutonium activity noted in the formed elements, the biological half-life for the serum activity (based on recovery day samples only) was 5.5 days.

Plutonium was also observed to be transported across the bovine placenta where the fetal tissue concentration (excluding placenta and fetal fluid) was in excess of 1.7×10^{-6}% of the oral adult dose for the fetus in the plutonium citrate group and approximately 1.8×10^{-5}% of dose for the fetus collected from the plutonium dioxide treated animal. Collections were made of fetal fluid, fetal liver, and placenta while the individual fetal carcasses were divided into quarters for further analysis. Any attempt to establish comparative relationships between quantity of ingested dose and fetal uptake is somewhat complicated by the fact that (1) different chemical forms of plutonium were administered to the adults, (2) one cow received an acute treatment and the other multiple exposures, (3) animals were sacrificed at different times after treatment was terminated, and (4) the animals were in slightly different stages of pregnancy. However, in both cases, the fetal liver contained the highest concentrations of recovered activity.

Finkel (1947) presented one of the earliest reports on placental transport of plutonium and observed that following injections (0.06 to 0.016 μCi/g) to pregnant mice, the percentage of transferred activity varied inversely with the original dose. Wilkinson and Hoecker (1953) also observed the apparent inverse relationship following plutonium injection and noted that the placental content of plutonium-239 was higher than the corresponding fetal value. This situation was not observed in either fetus sampled during the study with dairy cattle where the fetal liver had a greater concentration of plutonium than the placenta in both cases (Table 6). In a relatively recent investigation, Sikov and Mahlum (1968) injected intravenous doses of plutonium-239 citrate and polymeric plutonium into rats and also observed that the fraction of injected plutonium which reached the fetus was very small, being approximately 0.03 and 0.001% of the administered dose per fetus for the ionic and polymeric forms, respectively.

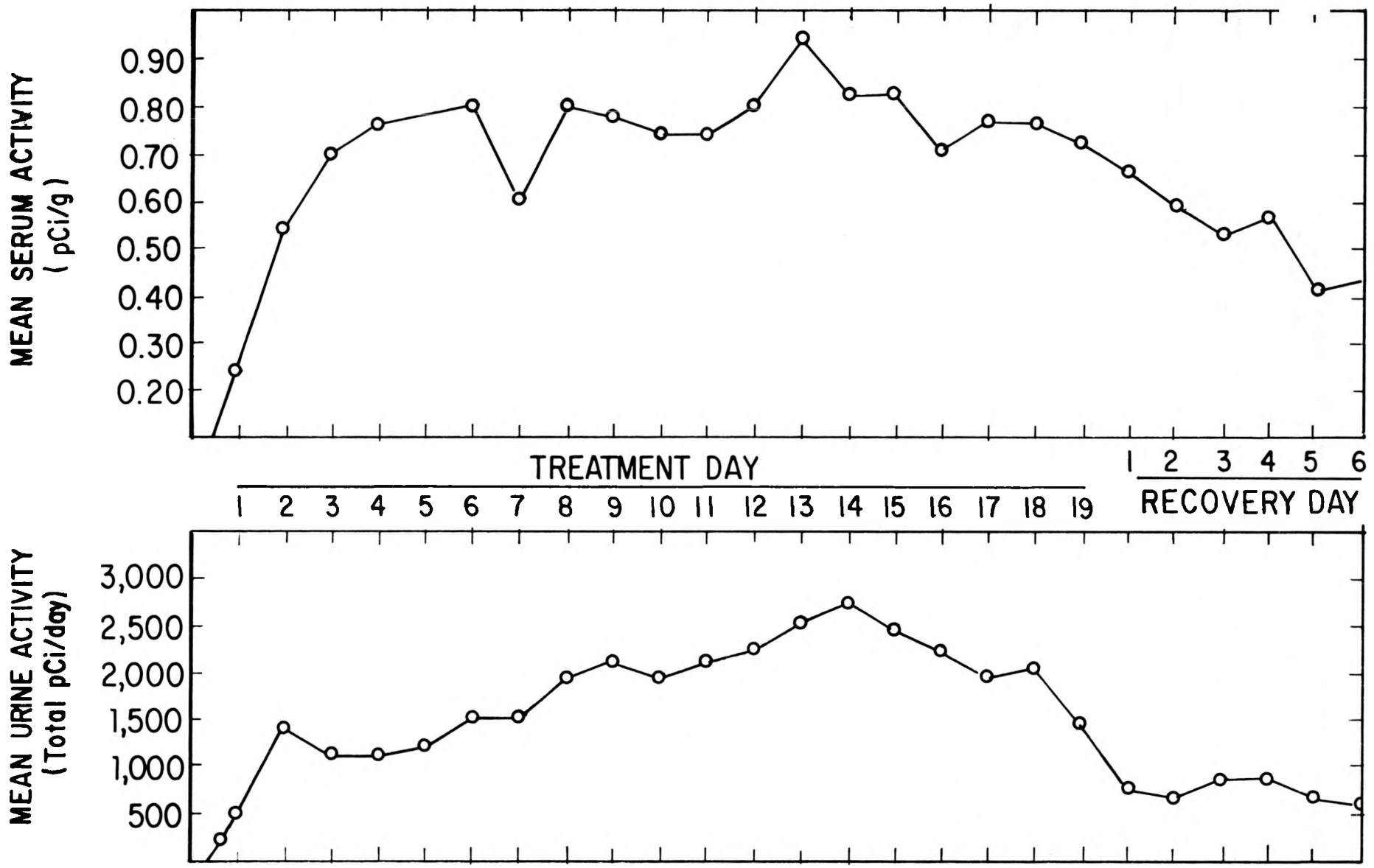

FIGURE 2. MEAN PLUTONIUM ACTIVITY CONCENTRATIONS IN BLOOD AND URINE OF FOUR HOLSTEIN DAIRY COWS RECEIVING ORAL DOSES OF 1 mCi PLUTONIUM-238 DIOXIDE PER ANIMAL PER DAY FOR 19 CONSECUTIVE DAYS.

Table 6. Physiological Transport of Ingested Plutonium to Selected Fetal Tissues in the Holstein Dairy Cow

Exposure Period	Weight of Adult Animal (kg)	Fetal Weight (g)	Recovered Plutonium Activity (pCi/g)		
			Fetal Liver	Fetal Fluid	Placenta
One fetus collected at time of sacrifice from an adult dairy cow which had received an acute oral dose of 3 mCi plutonium citrate 93 days prior to sacrifice.	677	792	3.0×10^{0}	2.0×10^{-2}	9.0×10^{-2}
One fetus collected at time of sacrifice from an adult dairy cow which had received multiple doses of plutonium dioxide (1 mCi/day for 19 consecutive days). Treatments had terminated 42 days prior to sacrifice.	547	1894	2.9×10^{1}	7.0×10^{-5}	6.6×10^{-1}

FUTURE PLANS

I. Biological Transfer of Plutonium-238 via *In vivo* Labeled Milk

A series of investigations will be conducted to provide additional information on relationships between the concentration of plutonium in the systemic circulation and its subsequent transfer of bovine milk. In addition, the project should provide some definitive data on the relative biological availability of *in vivo* plutonium-238 labeled milk as opposed to the *in vitro* labeled doses commonly used in gastrointestinal uptake studies.

Preliminary Study

One lactating Holstein dairy cow, confined to a metabolism stall, will be injected intravenously with doses of plutonium-238 nitrate, buffered with citrate. During the first portion of this range-finding effort, the cow will be injected intravenously with progressively higher concentrations of plutonium. Acute injections will be made on every third or fourth day until three dose levels have been administered to the cow (approximately 9 to 12 days' duration). Subsequent determinations will be made on the amount of plutonium transferred to milk, as well as the activity remaining in the blood at various time intervals after plutonium injection.

During the next portion of this preliminary effort, the cow will be given a second series of intravenous plutonium doses. These injections, administered once per day on a consecutive daily basis, will each contain approximately the same concentration of plutonium; i.e., a chronic exposure. Milk collected from the plutonium treated adult cow will be placed in suitable buckets and fed to two calves (eight liters/animal/day). Prior to feeding, portions of this milk will be analyzed at the Nevada Test Site farm using a Phoswich detector. Calves will be individually sacrificed at 24 hr and at 7 days after the last oral dose. Samples of liver, skeletal muscle, bone, spleen, blood, and kidney will be collected from each calf at time of sacrifice.

The main purpose of the preliminary effort is to establish doses for the following definitive study. The definitive phase, using four adult cows and a total of eight calves, will be required before significant statements can be made on the relative biological availability of *in vivo* and *in vitro* labeled milk. Supporting information on the plasma clearance, tissue deposition pattern, and fecal activity in dairy cattle following plutonium injections will also be gained during this definitive phase.

Definitive Study

Four lactating cows having similar milk production will be dosed intravenously for five consecutive days with plutonium-238. The injected plutonium concentration will have been determined in the feasibility effort. Cows will be milked twice daily and frequent blood samples will be collected. Appropriate portions of the milk will be radioanalyzed and the remainder fed to four calves, each weighing approximately 200 lb. Each calf will be assigned to a specific cow and will be given the *in vivo* contaminated milk for five days. Four additional calves will be fed *in vitro* plutonium-238 labeled milk. The plutonium-238 concentration will be adjusted to the same concentration as that fed in the *in vivo* phase. Plutonium nitrate buffered in citrate will be added to pooled milk obtained from the Nevada Test Site herd.

Calves that ingest *in vitro* labeled milk will be sacrificed at the same time post-treatment as the calves that receive *in vivo* labeled milk. Tissue analysis will therefore allow for comparisons of physiological uptake and retention between the *in vitro* and *in vivo* exposure groups. Furthermore, two of the adult cows used to produce the *in vivo* labeled milk will be sacrificed for tissue collection 30 days after the last injection, and the remaining two adult cows will be sacrificed 60 days after the last treatment. Frequent blood samples will continue to be taken from the adult cows until time of sacrifice. Plasma-to-tissue plutonium concentration ratios will be established following radioassay of the necropsy samples.

II. Comparative Biological Availability and Physiological Transport of Ingested Plutonium-238 and Plutonium-239 Dioxide in Lactating Dairy Cows

Future studies with dairy cows will also address potential isotopic differences in the metabolism of plutonium. While details of this project are not complete, the basic objectives have been established and were subdivided into three closely related parts.

A. To determine if the ratio of ingested plutonium-238/plutonium-239, administered in the dioxide form, remains the same in samples of blood, milk, urine, and selected tissues of lactating Holstein dairy cows.

B. To determine if the presence of one plutonium isotope (plutonium-238/plutonium-239) will modify the gastrointestinal absorption, tissue retention, and/or excretion rate of the other.

C. To quantitate and compare differences, if any, in the absorption, retention, and excretion of plutonium-238 and plutonium-239 administered as the dioxide.

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*Acknowledgments apply to the entire project, a portion of which is presented here.

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Table Analysis of Necropsy Tissues from Mature Holstein Dairy Cows Sacrificed at 42 and 73 Days, Respect y, Following Ingestion of 19 mCi Plutonium-238 Dioxide per Animal. Oral Treatments Consisted of 1 mCi . -- Animal (Mean Weight 650 kg) per Day for 19 Consecutive Days

Sacrifice 42 Days Post-Treatment					Sacrifice 73 Days Post-Treatment			
TISSUE	Cow Number	Sample Weight (g)	^{238}Pu Activity (pCi/kg)	Counting Error 2σ	Cow Number	Sample Weight (g)	^{238}Pu Activity (pCi/kg)	Counting Error 2σ
LIVER	122	8083	1.0×10^4	7.2×10^1	117	8667	1.1×10^4	7.6×10^2
	185	6854	1.0×10^4	6.9×10^2	134	330	1.1×10^4	7.6×10^2
			Avg. 1.0×10^4 Range 0 ±S.D. 0				Avg. 1.1×10^4 Range 0 ±S.D. 0	
FEMUR	122	863	4.1×10^2	2.9×10^1	117	834	9.0×10^2	2.9×10^1
	185	645	5.3×10^2	3.5×10^1	134	557	9.8×10^2	3.2×10^1
			Avg. 4.7×10^2 Range 1.2×10^2 ±S.D. 1.1×10^2				Avg. 9.4×10^2 Range 8.4×10^1 ±S.D. 7.1×10^1	
RIB	122	940	2.5×10^3	1.7×10^2	117	974	2.3×10^3	3.8×10^1
	185	1148	6.4×10^3	4.8×10^1	134	947	3.4×10^3	4.6×10^1
			Avg. 4.5×10^3 Range 3.9×10^3 ±S.D. 3.5×10^3				Avg. 2.9×10^3 Range 1.1×10^3 ±S.D. 9.7×10^2	

NOTE: Standard deviation - ±1 standard deviation of the population is based on the range of the sample observations (Snedecor, 1956). Snedecor, G. W., 1956. *Statistical Methods*, Iowa State College Press, Ames, Iowa, pp. 37-38.

APPENDIX I

Table 1. (Cont.) Analysis of Necropsy Tissues from Mature Holstein Dairy Cows Sacrificed at 42 and 73 Days, Respectively, Following Ingestion of 19 mCi Plutonium-238 Dioxide per Animal. Oral Treatments Consisted of 1 mCi per Animal (Mean Weight 650 kg) per Day for 19 Consecutive Days

	Sacrifice 42 Days Post-Treatment				Sacrifice 73 Days Post-Treatment			
TISSUE	Cow Number	Sample Weight (g)	^{238}Pu Activity (pCi/kg)	Counting Error 2σ	Cow Number	Sample Weight (g)	^{238}Pu Activity (pCi/kg)	Counting Error 2σ
VERTEBRAE	122	803	3.9×10^3	4.7×10^2	117	743	5.5×10^3	5.1×10^1
	185	712	6.3×10^3	3.8×10^1	134	672	4.1×10^3	4.9×10^1
			Avg. 5.1×10^3 Range 2.4×10^3 ±S.D. 2.1×10^3				Avg. 4.8×10^3 Range 1.4×10^3 ±S.D. 1.2×10^3	
KIDNEY	122	1602	7.6×10^2	3.8×10^0	117	1563	1.2×10^3	8.7×10^1
	185	1600	1.0×10^3	7.6×10^0	134	1208	6.4×10^2	4.4×10^1
			Avg. 8.8×10^2 Range 2.4×10^2 ±S.D. 2.1×10^2				Avg. 9.2×10^2 Range 5.6×10^2 ±S.D. 5.0×10^2	
SPLEEN	122	1002	9.1×10^2	6.2×10^1	117	1005	1.1×10^3	8.2×10^1
	185	860	2.2×10^3	1.5×10^2	134	1061	1.5×10^3	1.0×10^2
			Avg. 1.6×10^3 Range 1.3×10^3 ±S.D. 1.1×10^3				Avg. 1.3×10^3 Range 4.0×10^2 ±S.D. 3.5×10^2	

Table 1. (Cont.) Analysis of Necropsy Tissues from Mature Holstein Dairy Cows Sacrificed at 42 and 73 Days, Respectively, Following Ingestion of 19 mCi Plutonium-238 Dioxide per Animal. Oral Treatments Consisted of 1 mCi per Animal (Mean Weight 650 kg) per Day for 19 Consecutive Days

	Sacrifice 42 Days Post-Treatment				Sacrifice 73 Days Post-Treatment			
TISSUE	Cow Number	Sample Weight (g)	^{238}Pu Activity (pCi/kg)	Counting Error 2σ	Cow Number	Sample Weight (g)	^{238}Pu Activity (pCi/kg)	Counting Error 2σ
PULMONARY LYMPH NODES	122	13.6	3.3×10^5	4.9×10^3	117	11.6	7.6×10^2	7.1×10^1
	185	15.9	8.3×10^3	6.7×10^2	134	5.0	1.5×10^4	1.7×10^3
			Avg. 1.7×10^5 Range 3.2×10^5 ±S.D. 2.9×10^5				Avg. 7.9×10^3 Range 1.4×10^4 ±S.D. 1.3×10^4	
LUNG, UPPER RIGHT, LEFT APICAL	122	1148	7.6×10^2	2.0×10^1	117	1014	5.6×10^2	9.3×10^1
	185	904	1.0×10^3	6.6×10^1	134	984	6.1×10^2	4.3×10^1
			Avg. 8.8×10^2 Range 2.4×10^2 ±S.D. 2.1×10^2				Avg. 5.8×10^2 Range 5.0×10^1 ±S.D. 4.4×10^1	
LUNG, LOWER RIGHT, RIGHT DIAPHRAG-MATIC	122	1556	5.9×10^2	3.3×10^0	117	665	6.7×10^2	4.6×10^1
	185	1227	5.7×10^2	3.9×10^1	134	843	5.4×10^2	3.7×10^1
			Avg. 5.8×10^2 Range 2.0×10^1 ±S.D. 1.8×10^1				Avg. 6.1×10^2 Range 1.3×10^2 ±S.D. 1.2×10^2	

Table 1. (Cont.) Analysis of Necropsy Tissues from Mature Holstein Dairy Cows Sacrificed at 42 and 73 Days, Respectively, Following Ingestion of 19 mCi Plutonium-238 Dioxide per Animal. Oral Treatments Consisted of 1 mCi per Animal (Mean Weight 650 kg) per Day for 19 Consecutive Days

	Sacrifice 42 Days Post-Treatment				Sacrifice 73 Days Post-Treatment			
TISSUE	Cow Number	Sample Weight (g)	^{238}Pu Activity (pCi/kg)	Counting Error 2σ	Cow Number	Sample Weight (g)	^{238}Pu Activity (pCi/kg)	Counting Error 2σ
SKELETAL MUSCLE	122	4871	2.8×10^3	1.7×10^1	117	1540	6.6×10^1	6.0×10^0
	185	5060	6.2×10^1	4.5×10^0	134	1059	6.5×10^1	7.1×10^0
			Avg. 1.4×10^3 Range 2.7×10^3 ±S.D. 2.4×10^3				Avg. 6.5×10^1 Range 1.0 ±S.D. 0.9	
CARDIAC MUSCLE	122	2606	2.3×10^2	1.6×10^1	117	2329	2.3×10^2	2.2×10^1
	185	2339	3.4×10^2	2.2×10^1	134	96	1.6×10^2	1.5×10^1
			Avg. 2.9×10^2 Range 1.1×10^2 ±S.D. 9.8×10^1				Avg. 1.9×10^2 Range 7.0×10^1 ±S.D. 6.2×10^1	
SKIN	122	1195	5.3×10^1	3.9×10^0	117	1167	1.1×10^2	7.8×10^0
	185	1183	1.5×10^2	1.2×10^1	134	37.2	3.8×10^3	2.5×10^2
			Avg. 1.0×10^2 Range 9.7×10^1 ±S.D. 8.6×10^1				Avg. 2.0×10^3 Range 3.7×10^3 ±S.D. 3.3×10^3	

Table 1. (Cont.) Analysis of Necropsy Tissues from Mature Holstein Dairy Cows Sacrificed at 42 and 73 Days, Respectively, Following Ingestion of 19 mCi Plutonium-238 Dioxide per Animal. Oral Treatments Consisted of 1 mCi per Animal (Mean Weight 650 kg) per Day for 19 Consecutive Days

	Sacrifice 42 Days Post-Treatment				Sacrifice 73 Days Post-Treatment			
TISSUE	Cow Number	Sample Weight (g)	^{238}Pu Activity (pCi/kg)	Counting Error 2σ	Cow Number	Sample Weight (g)	^{238}Pu Activity (pCi/kg)	Counting Error 2σ
RUMEN	122	5809	9.0×10^{1}	6.4×10^{0}	117	1195	9.0×10^{1}	6.8×10^{0}
	185	4463	2.5×10^{2}	1.6×10^{1}	134		1.3×10^{2}	2.1×10^{0}
			Avg. 1.7×10^{2} Range 1.6×10^{2} ±S.D. 1.4×10^{2}				Avg. 1.1×10^{2} Range 4.0×10^{1} ±S.D. 3.5×10^{1}	
ABOMASUM	122	1789	8.6×10^{2}	1.1×10^{1}	117	1297	2.0×10^{2}	2.3×10^{1}
	185	1658	7.4×10^{2}	6.0×10^{1}	134	826	3.8×10^{2}	3.3×10^{1}
			Avg. 8.0×10^{2} Range 1.2×10^{2} ±S.D. 1.1×10^{2}				Avg. 2.9×10^{2} Range 1.8×10^{2} ±S.D. 1.6×10^{2}	
DUODENUM	122	172	2.8×10^{4}	2.0×10^{2}	117	170	9.6×10^{4}	2.4×10^{2}
	185	265	1.4×10^{3}	1.1×10^{2}	134	219	6.7×10^{4}	8.4×10^{3}
			Avg. 1.5×10^{4} Range 2.7×10^{4} ±S.D. 2.4×10^{4}				Avg. 8.2×10^{4} Range 2.9×10^{4} ±S.D. 2.6×10^{4}	

Table 1. (Cont.) Analysis of Necropsy Tissues from Mature Holstein Dairy Cows Sacrificed at 42 and 73 Days, Respectively, Following Ingestion of 19 mCi Plutonium-238 Dioxide per Animal. Oral Treatments Consisted of 1 mCi per Animal (Mean Weight 650 kg) per Day for 19 Consecutive Days

	Sacrifice 42 Days Post-Treatment				Sacrifice 73 Days Post-Treatment			
TISSUE	Cow Number	Sample Weight (g)	^{238}Pu Activity (pCi/kg)	Counting Error 2σ	Cow Number	Sample Weight (g)	^{238}Pu Activity (pCi/kg)	Counting Error 2σ
SMALL INTESTINE	122	4635	7.0×10^4	7.8×10^3	117	1659	4.3×10^3	2.9×10^2
	185	3844	8.4×10^4	9.1×10^3	134	890	2.5×10^3	1.9×10^2
			Avg. 7.7×10^4 Range 1.4×10^4 ±S.D. 1.2×10^4				Avg. 3.4×10^3 Range 1.8×10^3 ±S.D. 1.6×10^3	

PROCEDURES FOR DATA BASE ON THE ENVIRONMENTAL ASPECTS OF THE TRANSURANICS*

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ABSTRACT

The Data Base on the Environmental Aspects of the Transuranics is a computerized information file containing indexed and abstracted references to literature on biological and medical studies of the effects of transuranics, biological and ecological availability, turnover, and food chain dynamics, analysis of environmental materials, environmental transport mechanisms, waste disposal, monitoring, and regulations and standards for environmental levels. Tabulated data on plutonium in mammals that were compiled by the Comparative Animal Research Laboratory staff are being merged with the bibliographic information from the data base, thus a more detailed retrieval in that subject area is possible. Services from the data base include annotated bibliographies and searches tailored to the researcher's needs.

Information centers have become an integral part of scientific research needs. Founded in January 1972, the Data Base on Environmental Aspects of Transuranics continues under the direction of Paul B. Dunaway, Acting Director of the Bioenvironmental Sciences Division of the Nevada Operations Office, to offer an effective, easy-to-use link between the researcher and the relevant literature.

Literature on transuranics is now so large and so diverse that keeping abreast of even a single, well-defined speciality has become cumbersome and time-consuming for individual scientists. Literature within the scope of the Data Base on the Environmental Aspects of the Transuranics comes from many disciplines and from worldwide sources. More than one-sixth of the

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*Operated by Union Carbide Corporation for the U.S. Energy Research and Development Administration.

references in the data base are from countries other than the United States. Several indexes to foreign and translated literature are systematically searched, for example, Plutonium-Dokumentation from the Kernforschungszent Karlsruhe, German Federal Republic, and Translations from the Scientific Literature from the National Science Foundation. Documents are selected for the data base because they meet a criterion of potential value to current research and management.

The information is distilled from the documents with the specific needs of the researcher in mind, such as numeric results, new methods, or instrumentation. The major areas of evaluation are: (1) biological and medical studies on the effects of the transuranics, particularly as these elements relate to health considerations in man; (2) biological and ecological availability, turnover, and food chain dynamics; (3) analysis of environmental materials; (4) environmental transport mechanisms, including resuspension; (5) waste disposal as related to environmental concerns; (6) monitoring; and (7) regulations and standards for environmental levels.

Indexed and abstracted references are entered into and are retrievable from a dynamic, computerized information file (or data base) that is easily modified to reflect changing research needs. Annotated references are indexed to allow easy access for retrieving references to documents of interest to the user. The goal is to furnish users all the references related to their specific query and few that are unrelated. This is done by liberal use of subject categories and key terms. The studies on man and animals are divided into the subject categories of medical and biological aspects. Table I lists those aspects that are considered important when the document is analyzed. These data are sought for in the article, included in the abstracts, and indexed with key terms. This checklist is one of the ways the Data Base on the Environmental Aspects of the Transuranics strives to give users a reference to every paper in their subject area.

The subjects of plutonium and neoplasms are used as an example of how the data base is indexed. To narrow the search, neoplasms are noted as being either malignant or benign. Choosing malignant neoplasms, key terms such as adenocarcinomas, lymphosarcomas, fibrosarcomas, and osteosarcomas cound be used to find a specific disease. It is possible to be even more specific, for example, separating out articles with the animal tested, dose, intake route, isotope, chemical form, or those containing a mathematical model.

The personnel of the Data Base on the Environmental Aspects of the Transuranics have been working closely with staff of the Comparative Animal Research Laboratory (The University of Tennessee and U.S. Energy Research and Development Administration) to develop a single, common data base containing tabulated data on plutonium in mammals in conjunctions with the abstracted and indexed references. The extracted data from approximately 1400 documents were separated into defined fields for tabular display by the Comparative Animal Research Laboratory mammalian research staff and were designed to merge with the bibliographic information on plutonium in mammals from this data base to avoid duplication of effort. The tabular data system has the advantage of supplying detailed retrievals; for example, it is possible to retrieve only those studies on fetal uptake of Pu-239 in

Table I. Environmental aspects of the transuranics checklist for indexing.

1. TYPE STUDY

   Clinical
   Field
   Laboratory
   Theoretical

2. ORGANISM

   Common name--variety, group, wild, etc.
   Scientific name
   Age, size, developmental stage, sex
   Larger group (animals, plants, mammals, primates, microorganisms)

3. MATERIAL

   Form--Chemical--Compound
   Physical--Particle size, aerosol
   Nuclide

4. METHOD

   Administration route
   Dose
   Number of injections
   Observations
   Tissues
   Excretion

5. WHERE

   If data are place specific

6. WHEN

   If data are time (date) specific

7. EFFECTS

   Where
   What kind--pathological designations (acute, chronic)

8. ECOSYSTEM

   Terrestrial
   Freshwater
   Marine
   Estuary

the +4 citrate form given to rats by intraperitoneal injection in doses greater than one microcurie but not exceeding seven microcuries. The collaborative file of the Comparative Animal Research Laboratory and the Data Base on the Environmental Aspects of the Transuranics can be searched by the tabular data fields (number of animals, age of animals, intake time intervals, etc.) as well as, or in combination with the bibliographic data (author, title, publication description, and publication date) or by descriptors. This tabular data base on plutonium in mammals will soon be available for searches from the Data Base on the Environmental Aspects of the Transuranics and will also be published by the Comparative Animal Research Laboratory as a handbook of tabular data.

The Data Base on the Environmental Aspects of the Transuranics is part of the Ecological Sciences Information Center, which is one of the component centers of the Information Center Complex (ICC) at the Oak Ridge National Laboratory. Other centers in the ICC are concerned with energy, life sciences and human health, environmental impact, trace contaminants, land use and planning, and ecosystem modeling and analysis. The staff of the Data Base on the Environmental Aspects of the Transuranics also works with staffs of other computerized data bases including the Technical Information Center, where literature from the U.S. Energy Research and Development Administration and international literature on nuclear science are broadly indexed and abstracted, and the Nuclear Safety Information Center, where technical information involving the licensing of nuclear facilities and operational data relating to their safe operation are analyzed.

Abstracts in the Data Base on the Environmental Aspects of the Transuranics come from the original documents, most of which are available at the information center to on-site users. The scientific staff that analyze documents for the data base have advanced degrees in branches of science which overlap with the areas of research covered (physiology, radiobiology, biochemistry, medicine, and pharmacology). Additional expertise is utilized from researchers who use the data base, such as Nevada Applied Ecology Group contractors and Oak Ridge National Laboratory staff. These users help in obtaining better service by (1) explaining their needs; (2) alerting the data base to new documents as well as to historic documents that may have been declassified and never referenced in secondary sources or may be classified; and (3) serving as a resource to answer other users' questions.

Bibliographies which are arranged by subject and indexed by author, title, key terms, publication description, taxonomic name of the organism studied, and geographic location of field research are published periodically. Spceific searches on the Data Base on the Environmental Aspects of the Transuranics tailored to the researchers needs are available without charge to ERDA staff and unfunded students. These bibliographies and specific searches of the data file may be obtained by writing to Director, Nevada Applied Ecology Information Center, Building 2029, P.O. Box X, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830, or by telephoning (615) 483-8611, Ext. 3-6524; FTS (615) 805-6524.

List of bibliographies prepared to date by the NAEIC:

ORNL-EIS-71-10
ORNL-EIS-72-21 (NVO-AEIC-72-21)
ORNL-EIS-73-21 (NVO-AEIC-73-21)
ORNL-EIS-74-21 (NVO-AEIC-74-21)
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The Distribution and Excretion of Plutonium Administered Intravenously to the Rat

1947
J. Biol., Chem., 171(1), 273-283

SUBJECT CATEGORY: Biological Aspects

KEYWORDS: PLUTONIUM; RATS; EXCRETION; BONES; LIVER; SPLEEN; KIDNEYS; FECES; ABSORPTION; URINE; INJECTION, INTRAVENOUS; LABORATORY STUDIES; METABOLISM; GASTROINTESTINAL TRACT; PLUTONIUM; DISTRIBUTION; ACCUMULATION; VALENCE
CHEMICALS: Plutonium nitrate; Plutonium citrate; Plutonium chloride

ABSTRACT: The effect of citrate ion concentration, valence state, and various other factors on the body distribution and excretion of plutonium following its intravenous administration to the rat was studied. The first day following intravenous injection of $PuC2(NO3)2$ the urinary excretion of plutonium was substantially higher as compared to excretion after intravenous injection of plutonium chloride, plutonium nitrate, and tetravalent plutonium citrate complex. Fecal excretion during the first day was correspondingly low following injection of $PuC2(NO3)2$. On the 30th day following injection no significant differences were noted in either urinary or fecal excretion of plutonium administered in the four different forms. The skeleton was the major site of deposition regardless of the form in which plutonium was injected. The percent deposition in the skeleton and liver varied with the form of plutonium injected, but the form did not affect the deposition of plutonium in kidney and spleen. The size of the injected dose of tetravalent citrate complex did not affect the percent of the dose excreted in the feces or urine or the percent of injected material in the various tissues six days following injection. In the absence of citrate ion, absorption of plutonium from the gastrointestinal tract was quite low. Plutonium absorbed via this route gave a higher deposition in the skeleton than when administered intravenously as above. (KM)

<8>
Christenson, C.W.; Thomas, R.G.
Los Alamos Scientific Laboratory, Los Alamos, New Mexico

Movement of Plutonium through Los Alamos Tuff

1962, March
TID-7628, Part of Second Ground Disposal of Radioactive Wastes Conference Atomic Energy of Canada Limited, Chalk River, Canada, Sept. 26-29, 1961, (p. 248-281), 635 p.

SUBJECT CATEGORY: Waste Disposal and Management; Ecological Aspects

GEOGRAPHIC LOCATION: United States (SW), New Mexico, Los Alamos

KEYWORDS: PLUTONIUM; HAZARD ANALYSIS; SOILS, TUFF; SOILS, CLAY; GEOLOGY; WASTES, RADIOACTIVE; SEEPAGE PITS; PERMEABILITY; SOIL TRANSPORT; SITE EVALUATION

ABSTRACT: Uncontrolled and uncontained ground disposal of plutonium can be dangerous under field conditions. Plutonium species penetrated at least 28 feet in Los Alamos tuff. Moisture data, rates of flow of liquid and physical inspection indicated that this penetration takes place along fissures. The amount of activity sorbed at any one point in depth is dependent upon the chemical and physical nature of the substructure in that area. High percentages of clays, deposited by local weathering, will sorb plutonium species and result in a localized area of high alpha activity. As the species percolates through the soil, changes in valence state may also occur with changes in chemical environment; sorption or even solution may result. (KM)

<9>
Scott, K.G.; Axelrod, D.J.; Fisher, H.; Crowley, J.F.; Hamilton, J.G.
Crocker Radiation Laboratory, Divisions of Radiology and Medicine, San Francisco, California; University of California, Division of Medical Physics, Berkeley, California

The Metabolism of Plutonium in Rats Following Intramuscular Injection

1948
J. Biol. Chem., 176, 283-293

SUBJECT CATEGORY: Biological Aspects

KEYWORDS: PLUTONIUM 239; METABOLISM; RATS; INJECTION, INTRAMUSCULAR; BONES; VALENCE; ACCUMULATION; RADIOAUTOGRAPHY; TISSUES; LIVER; KIDNEYS; SPLEEN; GASTROINTESTINAL TRACT; ABSORPTION; RETENTION; LABORATORY STUDIES

ABSTRACT: Detailed metabolic studies of Pu 239 in the III, IV, and VI valence state administered to rats intramuscularly are presented. Plutonium was not found to be absorbed from the gastrointestinal tract to any extent. The skeleton was the main organ of deposition of plutonium, and the degree of retention in this organ was very great. No significant differences were observed in the metabolic properties of plutonium absorbed by the body for its three valence states. Radioautographs demonstrate the deposition of plutonium in the region of the endosteum, periosteum, and the endosteal covering of the trabecular bone. (Auth)

COMMENTS: Tables of deposition in tissues for various valence states noted.

<638>
Auerbach, S.I.; Cromroy, H.L.; Digregorio, D.; Dunaway, P.B.; Holt, B.P.; Lindquist, E.W., Jr.; McCormick, P.; Murphy, P.G.; Reichle, D.E.; Schmidt, C.H.; Stannard, B.; Styron, C.E.; Van Hook, P.I., Jr.; Woodwell, G.M.; Gamble, J.F.

Committee 6. Radioecological Effects of Fallout Radiation

December, 1971
CONF-700909, Proceedings of a Symposium held at Brookhaven National Laboratory, Long Island, New York, Sept. 15-18, 1970, (p. 643-644)

SUBJECT CATEGORY: Ecological Aspects

KEYWORDS: EXPLOSIONS, NUCLEAR; FALLOUT; ENVIRONMENT; BETA PARTICLES; COMMUNITIES; MAN; RADIATION EFFECTS

ABSTRACT: A committee reported to the symposium on the question of how environmental systems would interact in the nuclear war context in terms of essential recovery based on three time phases: (1) immediate effects and their possible impact on man, (2) long term impact, and (3) very-long term impact. More information is needed on effects of beta particles on natural communities and community structre. Thinking should be focused on total ecosystem response to radiation fallout. (BBM)

COMMENTS: Report from a committee included in Appendix A of the proceedings

<639>
Walter, G.H.; Burdett, A.K., Jr.; Paston, B.M.; Gaut, H.R.; Greene, J.C.; Griffin, S.A.; Hall, N.S.; Kittel, W.; Mistry, U.B.; Neal, W.T.L.; Sand, P.F.; Sharmon, E.C.; Simpson, P.E.; Stangler, M.J.; Strope, W.E.; Warner, E.G.

Committee 7. Use and Requirements for Research Data in Agricultural Defense Planning

December 1971
CONF-700909, Proceedings of a Symposium held at Brookhaven National Laboratory, Long Island, New YOrk, Sept. 15-18, 1970, (p. 645-646)

SUBJECT CATEGORY: Ecological Aspects; Postattack Assessment

KEYWORDS: RADIATION, GAMMA; BETA PARTICLES; LIVESTOCK; CROPS; FALLOUT; EXPLOSIONS, NUCLEAR

ABSTRACT: Recommendations were presented to the symposium by a committee concerned with use and requirements for research data in agricultural defense planning. It was felt future research should concentrate on effects of reason- able combinations of fallout beta and gamma irradiation using realistic fallout decay rates on growing crops of wheat, corn, soybeans, alfalfa, and grain sorghums, and on cattle, hogs, and chickens. Research on ingestion should concentrate on radioactive iodine. Research on salvage of irradiated livestock and combined beta-gamma research is needed. It is suggested that a glossary of terms be developed and used for all nuclear research. (BBM)

COMMENTS: Report from a committee included in Appendix A of the proceeding

<640>
Evans, R.D.
Massachusetts Institute of Technology, Cambridge 38, Massachusetts

Remarks on the Maximum Permissible Deposition of Plutonium in Man, and the Safety Factors in the Pivot Point Radiation Protection Guide of 0.1uc of Radium in Man

1962
Health Physics, 8, 751-752

SUBJECT CATEGORY: Medical Aspects

KEYWORDS: MAN; MAXIMUM PERMISSIBLE BODY BURDEN; PLUTONIUM; RADIUM

ABSTRACT: The relationship between the maximum permissible deposition of plutonium in man and the experience with radium deposition in man is described, with emphasis on the "hidden" safety factors in the radium limit which are, in turn, reflected in the plutonium limit. (Auth)

<641>
Swanberg, P., Jr.
General Electric Company, Hanford Laboratories, Richland, Washington

Comparison of Urinary Excretion Data from Selected Plutonium-Exposure Cases at Hanford

1962
Health Physics, 8, 761-765

SUBJECT CATEGORY: Medical Aspects

KEYWORDS: MAN; CASE HISTORIES; PLUTONIUM; EXCRETION; TIME FACTOR; INHALATION; EQUATIONS, DERIVED; INJECTION

ABSTRACT: Eight case histories relating observed plutonium excretion rate data are presented. These data were selected from the records of Radiation Protection Operation at Hanford to show the effects of individual differences on mathematical expressions describing the excretion rate of the radionuclide as a function of time. Both inhalation and injection modes of entry are considered, with particular emphasis on the inhalation mode. (Auth)

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