GAMMA-RAY SPECTROMETRIC SYSTEMS OF RADIOACTIVE FALLOUT ANALYSIS

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THESIS

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ABSTRACT

The mechanism of the fallout radionuclide transfer from the atmosphere to rain is studied by using a gamma-ray spectrometric system of analysis. The quantitative measurement of several long-lived "fallout radioisotopes" in both air and rain samples are determined. The system of analysis for the measurement of these isotopes was devised using a set of simultaneous equations for making the necessary mutual corrections. A computer program for their solution is included.

The gamma-ray spectra of the samples (collected at daily intervals) are presented. The instrumentation requirements, method of isotope and instrument calibration, and the calculation of the isotope concentrations from the spectra are discussed.

The analysis of about 135 daily air samples and 45 rain samples from the Lansing area are presented. Gross beta analyses of the air and rain samples are described and their relation to the gamma-ray analysis is graphically presented. Finally, a hypothesis on the transfer mechanics of the radionuclides is proposed based upon the evaluation of the field study.

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By

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INTRODUCTION

The presence of fission product radioactivity has been reported in streams and waterways, foodstuffs, and sewage treatment plants. This world wide distribution of radioactivity does not represent an immediate health hazard, but it may result in long term injury to human health from internal deposition.

Presently, the atmospheric radioactivity levels are but a fraction of the level labeled "safe" by the U. S. Government. However, the "safe" level is merely a temporary standard and tending to be reduced by further understanding of the biological effects of radiation. Also, with expected increases in the use of radioisotopes in industry, medicine, and research institutions; mining and chemical processing of uranium ore; nuclear reactors for power production and research; chemical processing of spent reactor elements for the recovery of nuclear fuel; and fallout due to the use of nuclear testing devices; the presence of fission product radioactivity in our environment will increase. It may possibly increase into the dangerous levels of concentrations.

Even the present "low" level of environmental radiation may present a serious problem. The long term injury possible through genetic effects may be the most severe of problems to future generations. Radiation is known to be the cause of mutations. Some of these mutations are for the good but the majority are for the worse. In the past, nature allowed only the fit to survive and thus weeded out the unwanted mutations. However, today with the advanced state of the medical profession, the weaker, unwanted mutations may survive. With the undesirable mutations being in the majority, the long term effect may be devolutionary. For these reasons,

studies such as that presented in this thesis are desirable.

The identification of the radioactive materials present in the fallout is necessary for a meaningful assessment of the potential health hazard of the contamination present. The more difficult and yet important problem concerning the radioactive pollution of our environment is to understand, quantitatively, transfer mechanisms of the radio nuclides in the environmental media, in particular, from air to rain.

This thesis is to investigate the mechanism of radionuclide transfer from the atmosphere to the rain. Gamma scintillation spectrometry is applied as a possible means of analyzing the transfer mechanism of individual radionuclides. It is attempted first to develop a method for the quantitative analysis of mixtures containing unknown amounts of gamma-ray emitting radioisotopes. Secondly, the mechanism of the fallout radionuclide transfer from the atmosphere to the rain is studied.

In the laboratory, both gross beta count and gamma scintillation spectrographs of air and rain samples are analyzed. A definite correlation is found between rain concentration of a certain radionuclide and atmospheric concentration of the same nuclide for all the radionuclides analyzed in this study.

In addition to the direct air to rain activity relationship, additional parameters of important influence have been investigated, such as wind velocity, cloud height, climatological conditions and others.

This study is based on daily average samples, observed from September 1962 to July 1963 on the Michigan State University campus. Also, the species of the radionuclides analyzed are limited to the four long half life fission products: $Ce^{141-144}$, $CsBa^{137}$, $Ru^{103-106}$, and $ZrNb^{95}$.

LITERATURE STUDY

In 1957, Greenfield (13) made a theoretical analysis of the rain scavenging of radioactive particulate matter from the atmosphere. A two step process was involved in the analysis for the collection of particles by water-cloud droplets and subsequent deposition by rainfall. It was considered that (a) a certain percentage of particles would be captured by water-cloud droplets, and (b) a certain percentage of the droplets would be subsequently captured by falling raindrops.

It was concluded that, as a result of the collision of falling raindrops with particles, particles with diameters less than two microns would be removed in only minute amounts, depending on the amount of rainfall.

Also, the probability was very small that water-cloud droplets would pick up significant quantities of particles with diameters greater than 0.08 microns, and the mechanism of droplet capture by raindrops would occur in a manner similar to the direct capture of radioactive particles.

Assuming a mean particle diameter of two microns, Greenfield showed theoretically that over 97 percent of that radioactivity is scavenged by direct interaction with raindrops. For all practical purposes, the removal is by direct impingement of falling raindrops with particulate matter.

Itagaki and Koemuma (14) interpreted the Greenfield analysis to mean that the predominant transfer mechanism was collisions between raindrops and radioactive particles during the process of raindrop descent. They measured the radioactivity of seven rains at five different elevations and found that the specific activity decreased with altitude.

Kruger, et. al., (15) measured the concentration of Sr-90 in precipitation resulting from large scale uplift and convective storms. They

interpreted Greenfield's analysis as meaning that the predominant deposition mechanism was by cloud-droplet capture and subsequent rainout. This is in apparent contradiction to the work of Itagaki and Koemuma. By examining three rains they found evidence of correlation between cloud ceiling and the concentration of Sr-90 in the rainwater. The cloud ceiling was interpreted to be a measure of the concentration of the radioactivity resulting from the evaporation of the raindrop in the unsaturated layer of air near the ground.

Collins (16) explains this phenomenon on the basis of certain meteorological hypotheses; briefly, that precipitation that originates from air with less moisture can produce higher concentrations than air with greater moisture since more air is needed to make the same depth of rainfall. The assumption is inherent that arid regions obtain rain from drier air and that raindrops falling in arid regions undergo more evaporation than in humid areas, thus enriching the Sr-90 in the remaining liquid water.

Miyake, et. al., (17) have proposed an equation for the deposition of radioactivity in terms of the concentration in the air and the amount of precipitation. Their data are accurately represented by the equation when atmospheric activity is taken as a constant over an extended period of time. However, in works by Norris (18) large fluctuations in the concentrations of radioactive dusts are observed. Fluctuations of rain' activity may be due to these fluctuations in the concentrations of radioactive dust in the air. Norris concludes that in the case of summer rains, Greenfield's model does not apply. No rain was extensive to the degree that the percentage of particles removed from a vertical cylinder was equal to the percentage of particles removed from the entire radioactive cloud. The specific activity of rain did not appear to be related

to either cloud height or rain intensity. This tends to support Grenfield's proposition that the fraction scavenged may be considered to be independent of rainfall intensity. Norris proposes a relationship, between specific rain activity and the specific activity of air at the time of the rain, of the form $C_R = 223\ C_A$.

At the University of Michigan, Dingle (19), has undertaken to model the rain process in somewhat more detail, accounting for the size spectra of cloud droplets, of raindrops, and of contaminant particles in integrating the rain-generation and scavenging processes layer by layer through a hypothetical atmosphere. Unfortunately, the collection of data is not sufficient at this time to make comprehensive analyses. Dingle claims his effort holds a great deal of promise and plans to present further details and results in the near future.

Kruger says that in general, the fallout radioactivity concentration in precipitation which reaches the ground is likely to be dependent upon the parameters listed below. (15)

1. The height of the precipitation generating level.

2. The precipitation generation and growth mechanisms in the cloud.

3. The amount od radioactivity initially in the air masses participating in the precipitation process.

5. The previous precipitation experience of the air at the generating level.

6. The descent experience of the precipitation from the cloud, in which precipitation originates, to the ground

The data of Kruger (15), show that in each case the ground level Sr-90 concentration follows closely the trends in the height of the cloud

ceiling. A poorer correlation is noted for the average precipitation rate, and very little correlation is noted for the influence of previous precipitation experience at the generating level. Variations are seen in the individual showers. The largest Sr-90 concentrations are in the rain which falls after the cloud has achieved its greatest vertical development. Showers which occur further from frontal storm systems show lower concentrations but still with variations due to the changing height of the precipitation generating level.

In all the works performed to date; Sr-90 or gross beta analyses are used. With the vast amount of debris deposited in the atmosphere by nuclear tests, many other isotopes are present. Also because of natural decay the relative amounts are constantly changing. It is thus possible that a study of the individual isotopes will be more revealing than the gross type analyses. Gamma scintillation spectrometry offers a possible means of analyzing the transfer mechanism of air activity to the rain.

ATMOSPHERIC RADIOACTIVITY AND ITS TRANSFER TO RAIN

Beginning with the work of Greenfield, a number of authors have prepared plausible sounding hypotheses on the transport of the radionuclides in air to rain. However, nearly all authors have restricted their viewpoint to one basic parameter: Itagaki, with transfer via direct collisions between raindrops and radioactive particles; Kruger, with cloud-droplet capture and subsequent rainout; Miyake, with the correction for the wind velocity.

In examining Greenfield's work, the author feels that the emphasis is placed on the direct collision approach as furtherd by Itagaki and Koenuma. Itagaki and Koenuma's proof is dependent upon the observation that the specific radioactivity of rain was found to decrease with altitude. However, as described by Collins, the observation does not automatically imply such a generalization. Raindrops may begin their descent with an initial concentration. The change in concentration observed by Itagaki and Koenuma may be dus to the amount of evaporation a raindrop experiences during its fall. Thus, the specific activity of the rain would decrease with increasing sampling altitude. If the direct collision of raindrops with radioactive particles is an important mechanism, the specific activity of rain should be dependent upon the specific activity of the air as measured at the time of the rain. Experimental evidence will be presented later which tends to strengthen this proposition. Itagaki, Koenuma, and Kruger all observed an influence of cloud height upon the final specific rain activity. Their observations are unfortunately only qualitative. Although their results are ech used for proving opposite viewpoints, it is important to establish or disprove any relationship. Further study on this

mechanism is presented in this report.

If Kruger and Collins are correct, a correlation between specific rain activity and specific humidity should exist. To judge such a correlation it would be necessary to know the specific activity of the rain at the cloud height. With only ground level observations available, this would be impossible. However, the relative humidity idea may be of assistance in qualitatively explaining extreme points found in the experimental studies.

Under the assumption that the basic mechanism of transport is via direct collisions, Norris proposed a correction for differences in ground level wind speeds. However, the number of rains was small. More importantly, the velocities used were daily averages measured five miles distant from the rain sampling location. It is quite probable that air turbulence increases the possibility of a raindrop - particulate matter collision and that wind velocities may be used as an indication of this effect. However, the data and results presented by Norris were insufficient and a closer look at this mechanism is justified.

The amount of precipitation is listed by Miyake as an important parameter. Although this would not be so according to Greenfield, its effect may actually exist. For this reason, the amount of presipitation and its effect upon the specific activity of rain water is measured and will be discussed later.

The previous experience of the air at the generating level was presented by Kruger as a possibly important parameter. At present, it is only possible to look at this effect qualitatively. However, it may be of some importance and therefore its effect will be determined.

In the process of evaluating the possible parameters, both gross beta and individual isotope analyses will be used. Gross beta counts are made of all beta emitting isotopes. Individual isotopes may be more revealing since the amount and proportions of each radioisotope changes because of the addition of new radioactive debris and natural decay of the old.

THE GAMMA SCINTILLATION SPECTROMETER

The gamma scintillation spectrometer is used to analyze the spectra of gamma-ray emitting radionuclides. The common sensing unit used for the detection of radiation in a gamm scintillation spectrometer is a thallium activated sodium iodide crystal. In being absorbed in such a crystal, gamma photons transfer all or a portion of their energy to orbital electrons of the molecules composing the crystal. These electrons lose their energy by exciting and ionizing the molecules composing the crystal. The energy received by the molecules is, in turn, given off in the form of pulses of light, part of which are collected on the photocathode of a multiplier tube which is optically coupled to the crystal. The combination of crystal, photomultiplier tube, and amplifier act as a scintillation counter. (5) A diagram of the detector and accompanying photomultiplier tube is shown in Figure 1.

Gamma-radiation is absorbed in the crystal by three principle processes: (a) Photoelectric, (b) Compton scattering, and (c) Pair production. These processes are scheatically diagramed in Figure 2.

Absorption of gamma photons in the crystal by the photoelectric process results in current pulses essentially representing the energy of the incident photons. In the photoelectric effect, most of the energy of the gamma-ray is given to an inner shell atomic electronwhich excapes from the parent atom. This atomic electron leaves the parent atom with an energy less than the energy of the gamma photon (less by an amount equal to the binding energy of the atomic shell from which it escaped). The photon energy is converted to an electrical pulse by the scintillation crystal photomultiplier system. The parent atom, being left in an excited

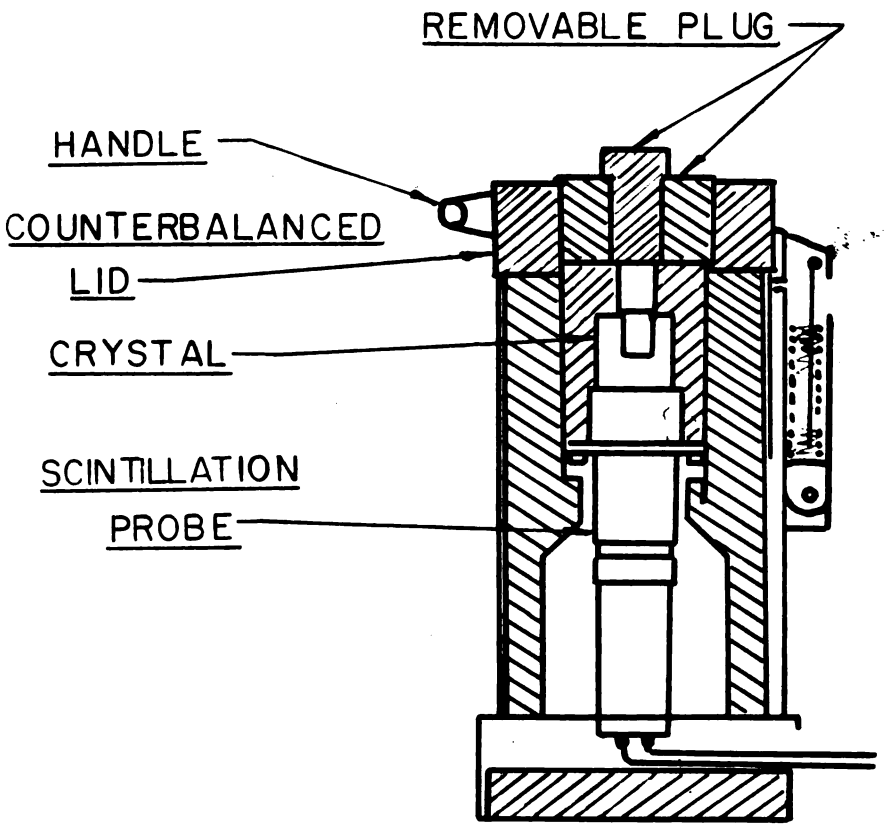


FIGURE - 1
CROSS-SECTION VIEW OF THE
SCINTILATION WELL DETECTOR

state, soon gives off the remaining energy when an electron falls into the shell from which the photo-electron came.

In Compton scattering, some of the energy of the incident gamma photon is transfered to an orbital electron, the remainder of the energy being carried away by the scattered photon. The scattered photon may or may not be further absorbed in the crystal. Again, because of the relatively long resolving time of the detection equipment, any series of Compton interactions followed by a final photoelectric capture within the crystal will produce the signal proportional to the energy of the incident photon. The photomultiplier tube can not distinguish between two seperate light flashes if they are simultaneous, and in these instances a current pulse representing their sum results. (1) Thus, Compton scattering accompanied by photoelectric absorption of the scattered photon may yield a pulse representing the energy of the original gamma radiation. In general, however, absorptions involving Compton scattering yield a spectrum of pulses with a maximum energy less than the energy of the incident gamma photon.

In pair production, a positron and an electron are created. This creation requires 1.022 Mev energy, the remaining of the incident gamma photon appearing as kinetic energy shared between the positron and the electron. The positron and the electron lose their kinetic energy through collisions with molecules composing the crystal. After it is brought to rest, the positron annihilates with a nearby electron giving rise to two photons, each possessing 0.511 Mev energy. If the two annihilation photons escape from the crystal, the resulting pulse represents the energy of the incident gamma photon minus 1.022 Mev. If both are absorbed within the

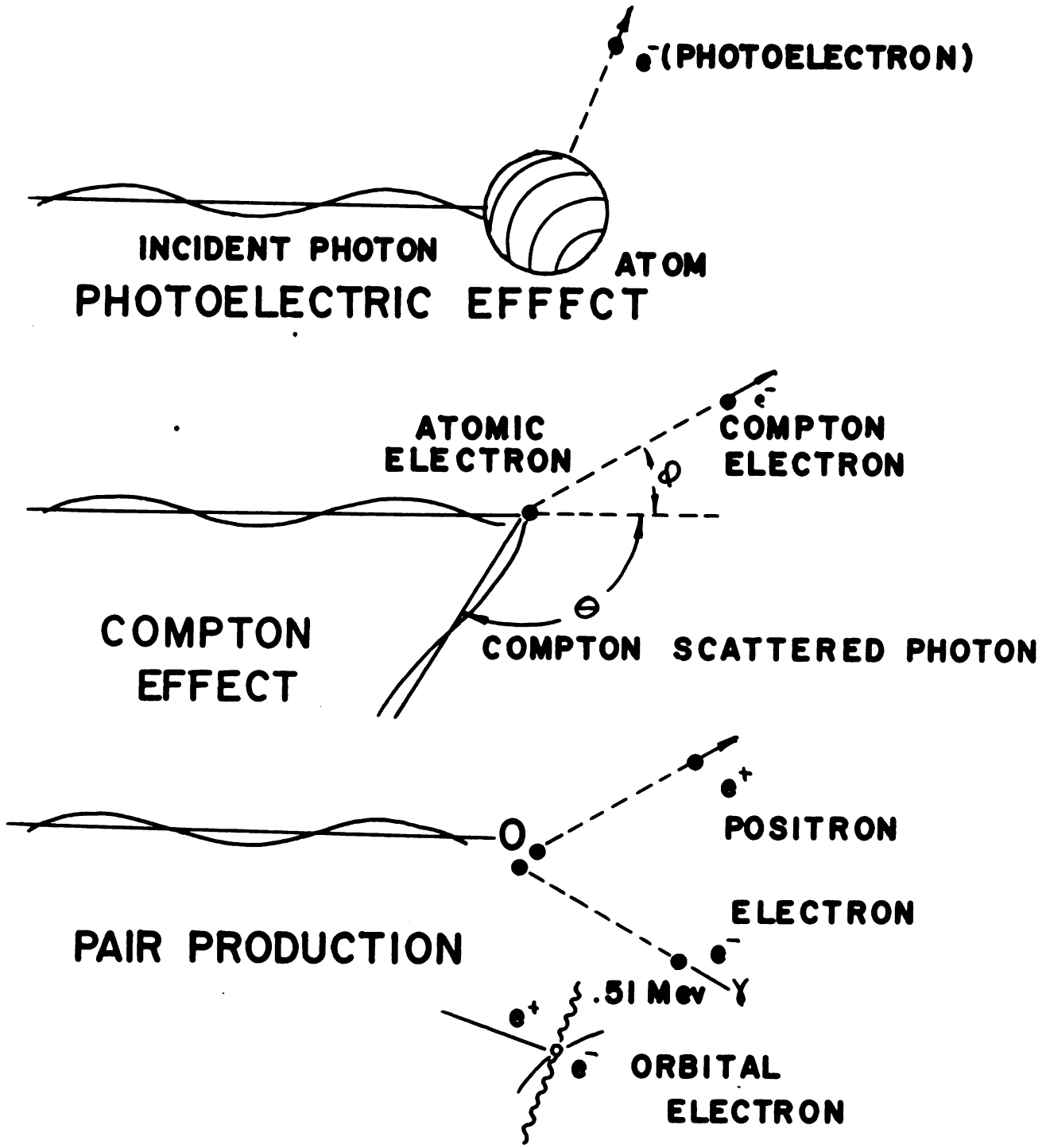


FIGURE - 2

PROCESSES OF ABSORPTION OF GAMMA RADIATION BY THE CRYSTAL DETECTOR

crystal, the pulse will represent the energy of the incident gamma photon.(9)

Through use of suitable electronic circuits and discriminators, it is possible to count only pulses within a given energy range. Such an instrumental set-up constitutes a gamma scintillation spectrometer. Its basic components are shown schematically in Figure 3.

The electrical signal output of the photomultiplier tube is directly proportional to the event which caused the scintillation. However, further amplification is necessary because the pulse is of the order of millivolts, and the pulses are in the order of volts when used for the counting process. (9). As described here, the spectrum is a curve showing the numbe of pulses of any given energy as a function of energy. The analyzer contains two discriminators. Unless the voltage pulse is higher than the setting on the lower discriminator, the pulse is rejected. The second discriminator is set to reject voltage pulses above its setting. The circuits are arranged so that the second discriminator setting is a voltage slightly higher than the first discriminator setting. The second setting is referred to as the "channel width". The output of the pulse analyzer is connected to a scaler to record the number of pulses observed in a specified time.

To calibrate the curve in terms of Mev corresponding to any given pulse height setting, a radioactive sample with a known energy spectrum is used. C_s - 137 with its single peak at 0.661 Mev is a very common source material.

The N_a -24 spectrum is an excellent illustration of the priviously mentioned principles. A study of the N_a-24 spectrum reveals that there are peaks at energy levels of about 0.51, 1.02, 1.73, 2.23, and 2.75 Mev.

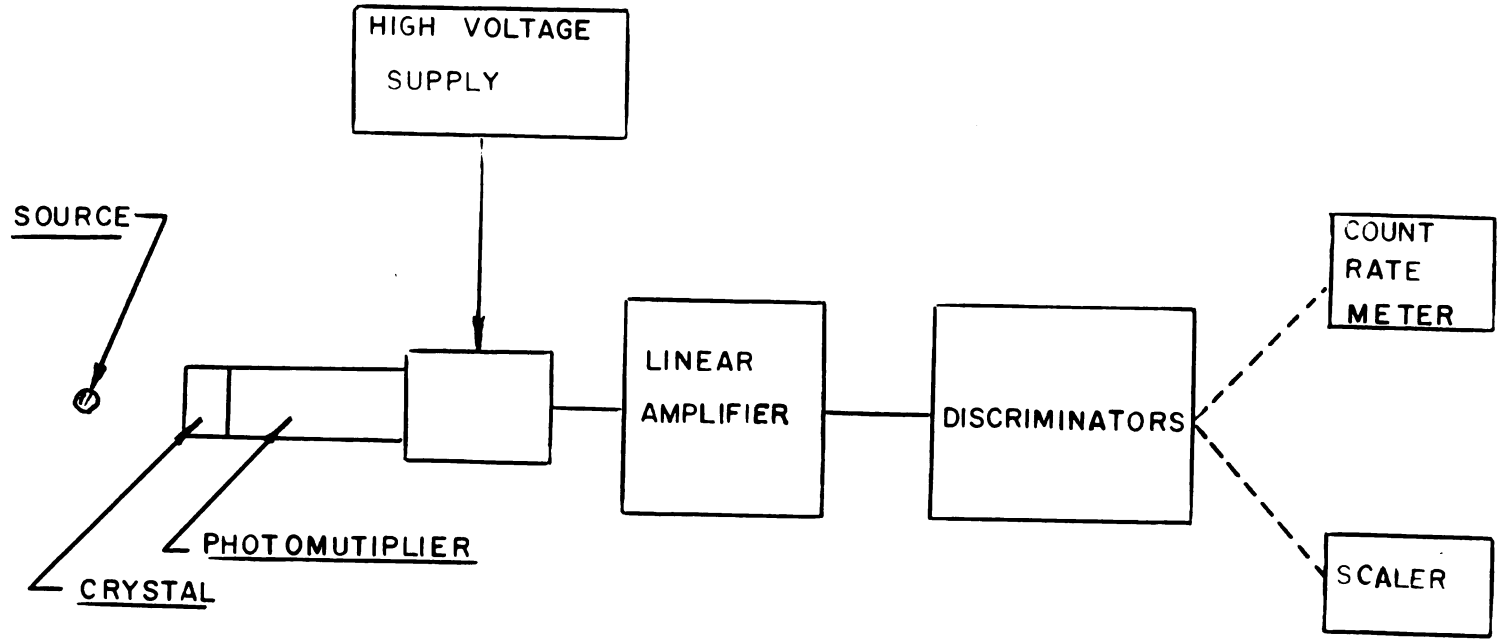


FIGURE - 3

GAMMA SCINTILLATION SPECTROMETER

The fact that there are peaks at these energies does not necessarily indicate that N_a-24 emits gamma photons with all these energies. It does indicate that the scintillation equipment observed a considerable number of events having these various energies. To explain the various peaks it is helpful to note that N_a-24 does emit gamma photons having energies of 1.36 and 2.75 Mev. The peaks at these energies indicate the likelihood that considerable numbers of Na-24 gamma-rays are completely absorbed in a Compton photoelectric series of events. The 0.51 Mev peak is produced as a result of pair production and subsequent annihilation; but where one of the photons escapes the crystal. The 1.02 Mev peak occurs with the absorption of both 0.51 Mev photons. The 1.73 Mev peak is the kinetic energy of the pair production from a 2.75 Mev gamma-ray (2.75 - 1.02 = 1.73). The 2.23 Mev peak is evidently the observation of a 0.51 event and a 1.73 event where both events occured in less than the resolution time of the equipment and thus were observed as a single event of energy 2.23 Mev. This is called a sum peak. (9)

The broadness of the peaks is attributed to a certain extent to Compton scattering where only a part of the energy was given up to the crystal while the remainder escaped and was not observed.

Because of the mechanism of its operation, the gamma scintillation spectrometer affords an excellent means for the study of the basic processes by which gamma radiation interacts with matter. (1) More importantly, the spectrometer makes possible an analysis which does not demand distruction of the sample. Also, a measurement of the peak area (at least for simple spectra) gives a direct indication of the isotope concentration. The mixing of a number of isotopes greatly complicates the analysis and will be discussed later in this report.

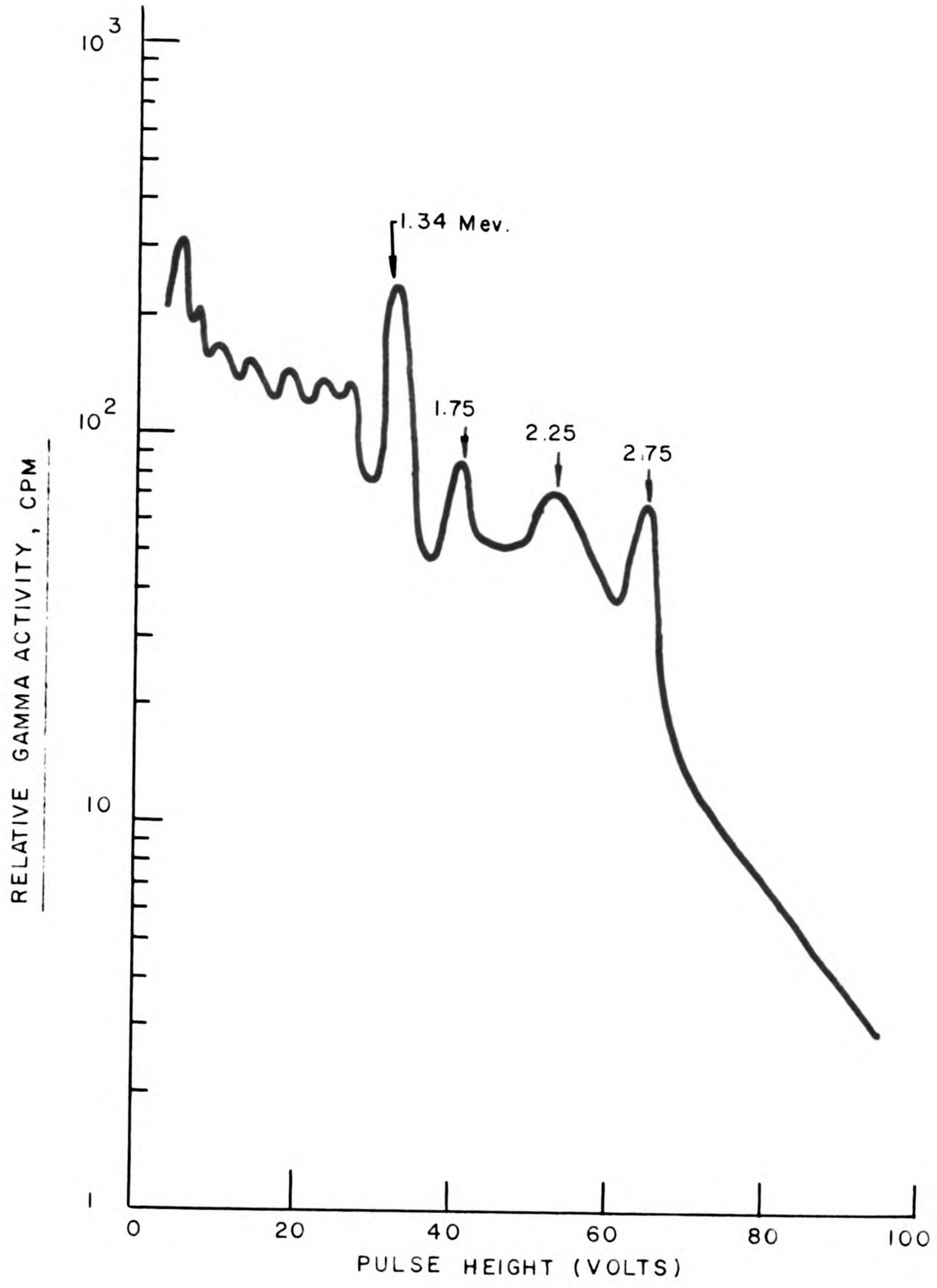


FIGURE-4

GAMMA SPECTRUM OF Na-24

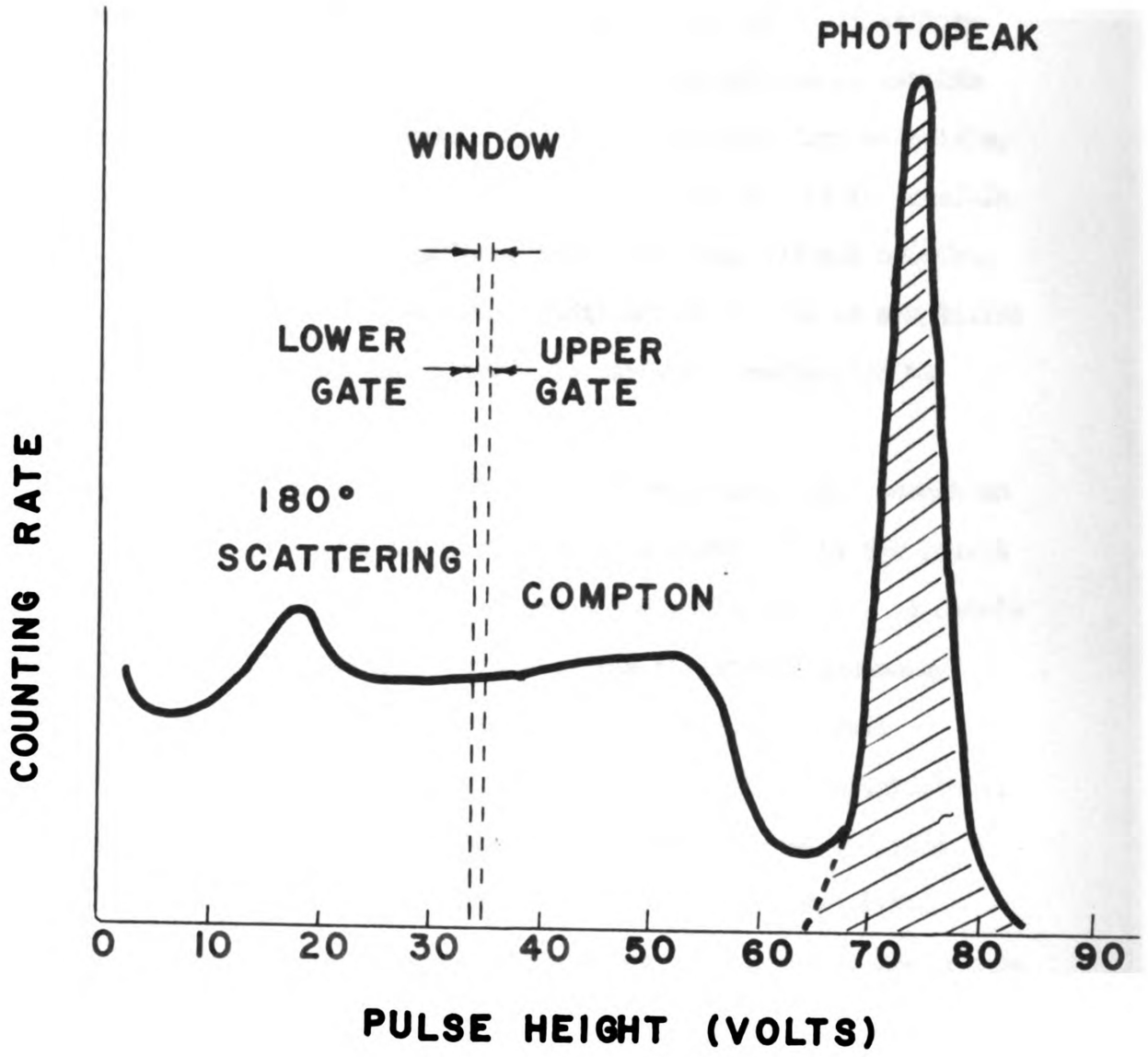


FIGURE-5

GAMMA-RAY SPECTROGRAPH

QUANTITATIVE ANALYSIS OF GAMMA SPECTROGRAPHS

Gamma-rays resulting from the decay of radioisotopes are emitted in discreet energy levels and undergo only slight attenuation in materials with low atomic numbers. These properties have permitted quantitative spectrometric analysis of certain mixtures of radioisotopes in samples of tissue and bone from experimental animals, in various food materials, and also in humans. (5,6) By similar methods of analysis it is possible to quantitatively measure radioisotopes resulting from fallout or other such sources. The problem of measuring radioactive fallout is simplified by the fact that the mixture of isotopes is generally restricted to rather definite compositions.

The gamma-ray spectrum of "fresh fallout" radioisotopes, present on vegetation three to four days after fission (see Figure 6) is the result of a complex mixture containing many short lived radioisotopes. A quantitative gamma-ray spectrometric analysis of the individual gamma-ray emitting radioisotopes in such a mixture would be very difficult.

About one week after fission, many of the short lived radioisotopes have decayed, and a less complex gamma-ray spectrum is observed.(see Figure 7) After a three to four week decay period, the short lived radioisotopes have decayed to insignificant concentrations and the gamma-ray spectrum possesses five characteristic photopeaks (see Figure 8), due to Ce^{141}, $CePr^{144}$, $Ru^{103-106}$, $ZrNb^{95}$, and $BaLa^{140}$. From this time until the isotopes have decayed beyond detection, it is possible to measure them with reasonable accuracy from the gamma-ray spectrum of the sample.

In addition to occasional large amounts of fresh fallout from tests

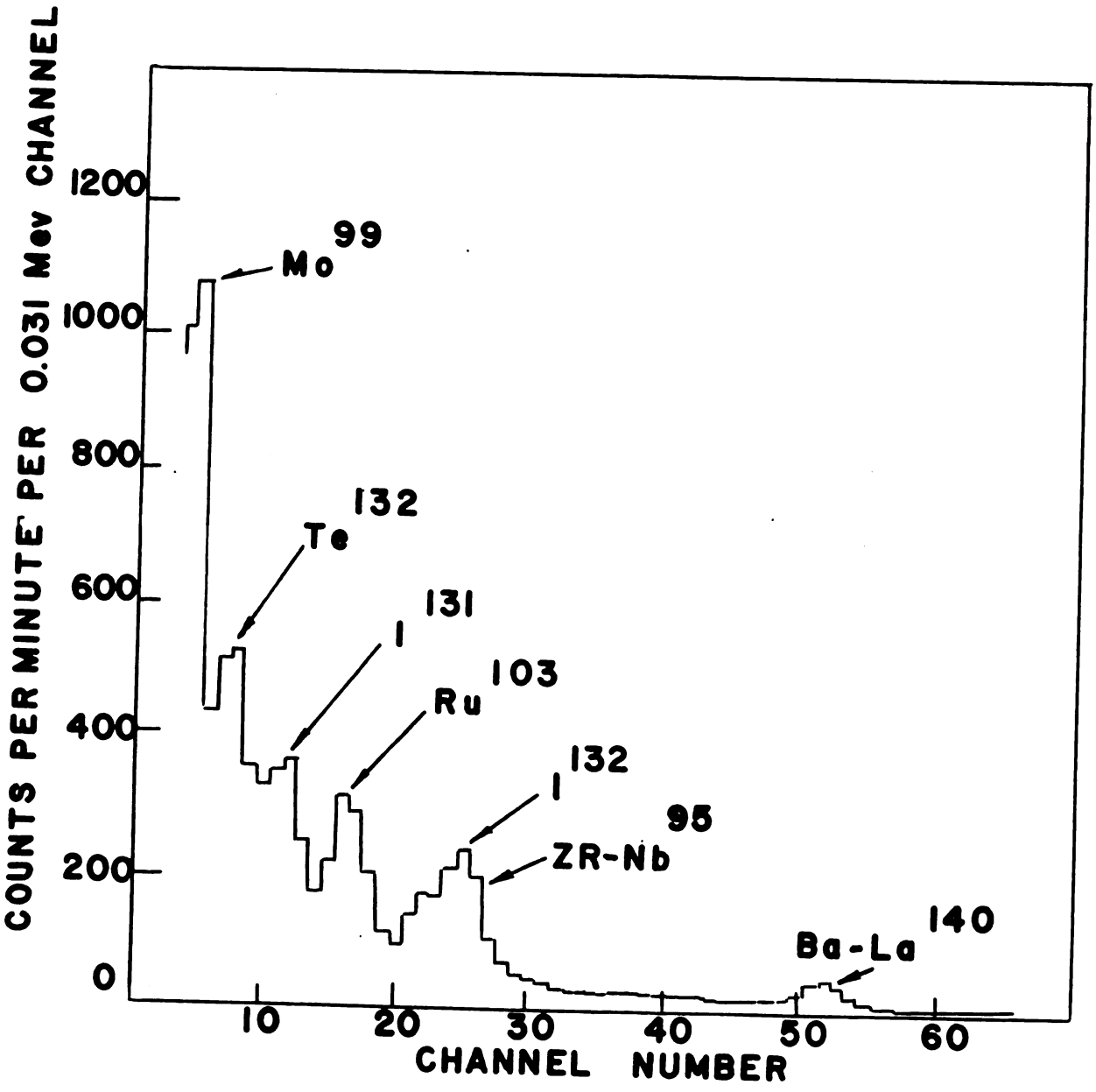


FIGURE- 6
GAMMA-RAY OF A VEGETATION SAMPLE CONTAINING 3-4 DAY- OLD FALLOUT

as in Nevada, there is a continual slow fallout of radioisotopes from atomic detonations at more remote locations on the globe. The material from these tests consists largely of the longer lived isotopes: $CsBa^{137}$, Ce^{141}, Ce^{144}, Ru^{103}, Ru^{106}, and $ZrNb^{95}$; and does not usually contain significant amounts of $BaLa^{140}$ or I^{131}. (7)

The general method employed for measuring the concentrations of the various isotopes is graphically illustrated in Figure 4. The counting rate at the $BaLa^{140}$ photopeak is directly proportional to the amount of this isotope present. The "net counting rate" at the $ZrNb^{95}$ photopeak requires a correction for the contribution from $BaLa^{140}$ as well as for some of the other isotopes. Similar corrections are required for the remaining isotopes. After these "Compton Corrections" have been made, the net counting rate of each characteristic photopeak is proportional to the amount of isotope present.

A discussion of the specific conditions for making the gamma-ray spectrograph and the methods used in calibrating the equipment are included in the experimental section.

The graphical method is not sufficient since it produces many additional errors. Another similar method might be to algebraically handle the "Compton Corrections."

In a case where eight isotopes are to be measured from a gamma-ray spectrum and each offers a contribution to the counting rate, the calculation of the net photopeak counting rates of the individual isotopes requires the solution of eight simultaneous equations for the eight unknowns.

A general solution for the system may be calculated as follows:

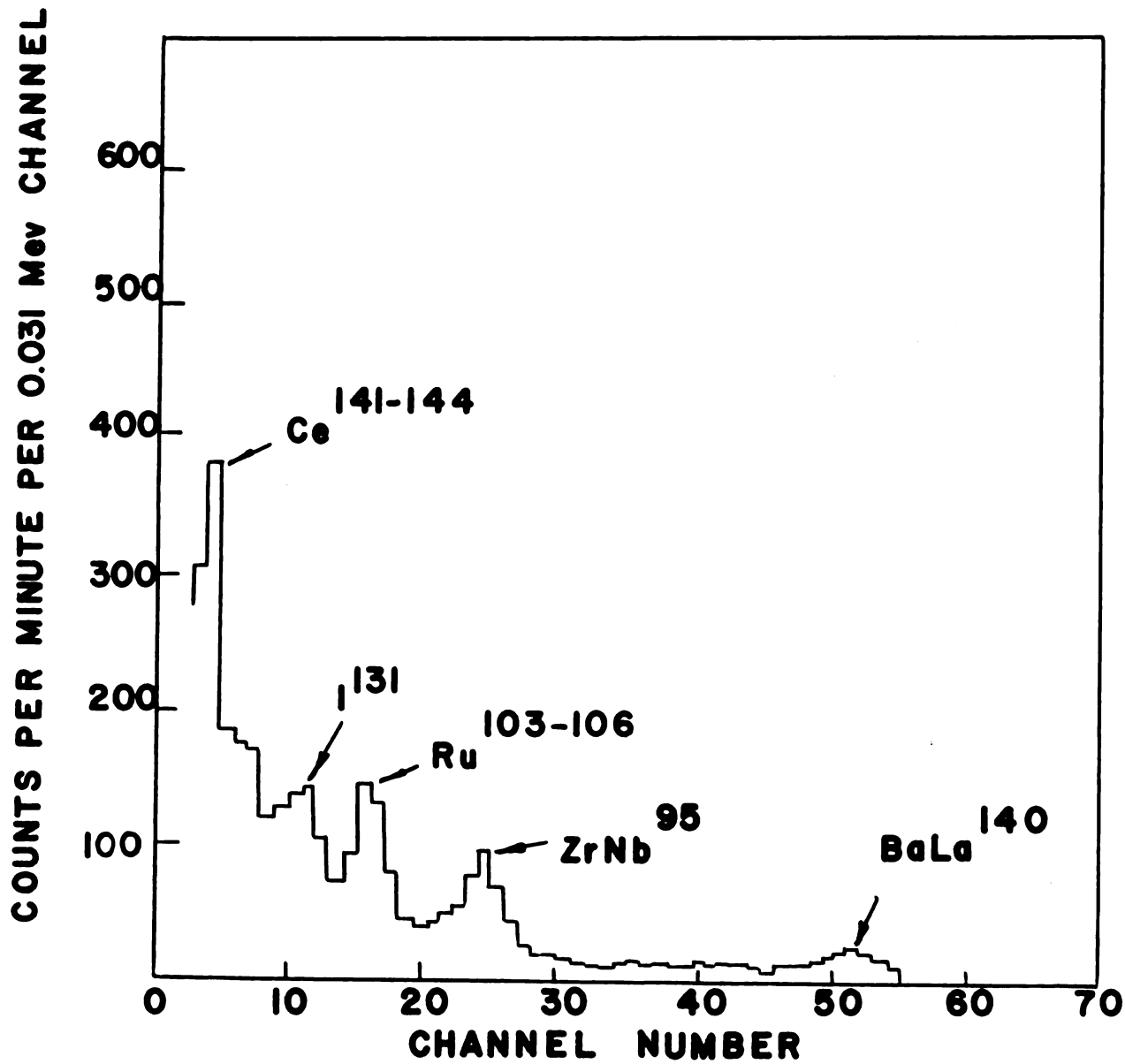


FIGURE-7
GAMMA-RAY SPECTRUM OF A VEGETATION SAMPLE CONTAINING WEEK-OLD FALLOUT

Let A,B,C,D,E,F,G, and H refer to the activity in micro micro curies for each of the eight radioisotopes present respectively.

P_{ij} will refer to the ratio of the count rate in channel "i" with the total activity of isotope "j", e.g.

$$P_{1A} = \frac{\text{Count rate in channel one as caused by isotope A}}{\text{Activity (uuc) of isotope A}}$$

Therefore eight simultaneous equations may be written and their solutions yielding A,B,C,D,E,F,G,and H. Let K1, K2, K3, K4, K5, K6, K7, and K8 represent the total count in channels 1,2,3,4,5,6,7,8 respecttively. Therefore:

K1 = P1a A + P1b B + P1c C + P1d D + P1e E + P1f F + P1g G + P1h H

K2 = P2a A + P2b B + P2c C + P2d D + P2e E + P2f F + P2g G + P2h H

K3 = P3a A + P3b B + P3c C + P3d D + P3e E + P3f F + P 3g G +P3h H

K4 = P4a A + P4b B + P4c C + P3d D + P4e E + P4f F + P4g G + P4h H

K5 = P5a A + P5b B + P5c C + P4d D + P5e E + P5f F + P5g G + P5h H

K6 = P6a A + P6b B + P6c C + P6d D + P6e E + P6f F + P6g G + P6h H

K7 = P7a A + P7b B + P7c C + P7d D + P7e E + P7f F + P7g G + P7h H

K8 = P8a A + P8b B + P8c C + P8d D + P8e E + P8f F + P8g G + P8h H

The value of P_{ij} must be obtained experimentally. For the spectrograph as used for this report, the values for P_{ij} are given in Figure 10.

Since all the samples analyzed for this report contained only four major isotopes, the process was further simplified by using only four simultaneous equations.

K1 = .5559A + .2598D + .4561F + .0864H

K4 = .0083A + .2646D + .3113F + .0250H

K6 = .0081A + .1469D + .6645F + .2149H

K7 = .0060A + .1275D + .8703F + .0002H

For ease in computation, a Fortran computer program was used in solving these equations. The program for both eight and four component systems is given in the appendix.

FIGURE 10

EXPERIMENTAL VALUES FOR THE COMPTON

AND PHOTOPEAK CORRECTIONS

Isotope	P1	P2	P3	P4	P5	P6	P7	P8
A Ce^{141}	.55589	.01477	.00916	.00830	.00770	.00814	.00596	.00550
B $CePr^{144}$	.36270	.04430	.03211	.02753	.02552	.03050	.02133	.01872
C I^{131}	.16891	.11521	.69832	.01457	.03036	.02119	0.0000	0.0000
D Ru^{103}	.11511	.10472	.04023	.25800	.14063	.01370	.01073	.00881
E $RuRh^{106}$	.25979	.19841	.14913	.26459	.02156	.01468	.01275	.01096
F $ZrNb^{95}$	.45610	.58592	.42387	.31127	.13731	.66453	.87030	.04390
G $BaLa^{140}$	.44854	.20364	.21337	.35052	.20153	.07705	.16516	.07982
H $CsBa^{137}$	.08639	.09369	.05907	.02501	.04424	.21488	.00018	.00032

LABORATORY INSTRUMENTATION AND PROCEDURE

Following are descriptions of the air sampling, rain sampling, and counting arrangements and procedures. The apparatus is shown in Figures 11, 12, and 13.

Air Sampling

Two techniques of air sampling were employed. One method was used for gross beta analyses while another for the gamma-ray spectrometric analyses.

For the gross beta analysis, approximately thirty cubic meters of air were sampled every twenty-four hours through a Millipore filter. This small volume results in a slightly reduced reliability due to counting statistics when compared to the "standard" high volume fibrous filter method. The fibrous filter introduces an error as the result of particulate matter which penetrates and is stored within the filter matrix. These particles are never counted when the gross beta method of analysis is used, and according to Cotton (10) this "effect cannot be accurately compensated by calculations." The Millipore filter has the characteristic of retaining on the surface all particles which are larger than the specified pore size. Therefore, there is negligible absorption of radioactivity by the filter matrix.

Air samples for beta counting were taken on a twenty-four hour basis at a flow rate of 25 liters per minute. This flow rate was then corrected for pressure drop. The filters were mounted and stored in a dessicator until counted.

For gamma-ray scintillation counting a large count-rate is necessary. For this reason a larger volume of air was used than that produced by the Millipore filter system. Instead a model (HAS-4 Atomic Products Corp.)

high volume air sampler with a (TFA 2133) felt filter was used.

Air samples for gamma counting were taken on a twenty-four hour basis at a flow rate of 35 cubic feet per minute. The filters were then placed in one ounce bottles for counting.

Rain Sampling

Under the assumption that a knowledge of the distribution of radioactivity in rain would be revealing, a collection basin was situated on the roof of the Engineering Building. The basin was designed of 16 gauge sheet metal with an area of 36 square feet. Rain samples were usually about thirty liters in volume.

One liter of the rain sample was evaporated and its residue placed on a counting planchet for beta counting. The procedure used was in all essentials the same as that described by Setter, Hagee, and Straub. (11) Techniques for preparing radioactive samples on planchets is described by the Nuclear Chicago Corporation. (12)

The remaining rain was used for gamma counting. This large volume of rain was evaporated in shallow pans, one meter square and heated by six 250 watt infra-red heating lamps. The residue was then washed into a small glass bottle for counting.

Counting

All beta counting was performed with a gas-flow proportional counter with a 2.5 millivolt sensitivity. A blanket counting efficiency of fifty percent was assumed for all samples.

The fifty percent was assumed so that the results could be compared, at the same order of magnitude, with other data. If "absolute disintegration rates" were needed, seperate efficiencies would have to be determined for each sample. However, fifty percent closely approxi-

mates the efficiency of gas flow counters of this type as well as agreeing with experimental values for individual isotopes.

The following procedure was used.

1. A background count of 500 counts was taken before each counting session. This was subtracted from the total counting rate for each sample.

2. One thousand counts were taken on each sample 4-7 days after sampling. Two or three additional counts were taken at 4-7 day intervals.

3. The sample counting rates were extrapolated back to the time of sample collection. The extrapolated counting rates were converted to micro micro curies per liter according to:

$$\text{uuc}/M^3 = \frac{\text{Counts per minute}}{(2.22)(\text{counting efficiency})(\text{volume of air in } M^3)}$$

$$\text{uuc}/l = \frac{\text{Counts per Minute}}{(2.22)(\text{counting efficiency})(\text{volume of rain sample in } l)}$$

A Nuclear Chicago single channel analyzer with a NaI (Tl) detector was used for the gamma-ray spectrometric measurements. A photograph of the analyzer, detector, associated lead shield, and scaler is shown in Figure 12. The general operations of the analyzer is included in the instruction manual. The detector is a three inch by three inch NaI (Tl) well crystal (Harshaw Chemical Company) mounted on a DuMont multiplier phototube. The sample container, a one ounce glass bottle, is placed directly within the crystal recess.

A high voltage of about 2000 volts was used on the multiplier phototube and was adjusted to center the 0.661 Mev Cs^{137} photopeak with a channel width of 0.060 Mev.

Known amounts, of each expected isotope, were counted with the spectrometer so as to determine the counting efficiency of each. The results of this analysis are shown in Figure 5. The procedure used

was essentially the same as used for beta counting, except 1000 counts were taken in each of the sixteen .060 Mev channels. Also, the gamma counting was performed only once, this being immediately after each sampling period ended.

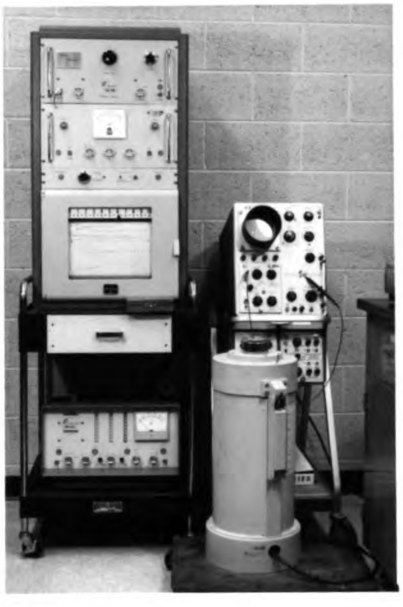
High Volume Air Sampler

Rain Trap

FIGURE 11

SAMPLING EQUIPMENT

Proportional Gas Flow Counter



Single Channel Analyzer

FIGURE 12

COUNTING EQUIPMENT

1 Liter Rain Samples for Beta Counting

Large Rain Samples for Gamma-ray Analysis

FIGURE 13

SAMPLE PROCESSING

COLLECTION EFFICIENCY OF THE FILTERING EQUIPMENT

The efficiency of any particular sampler, especially one used to detect radioactive matter, is ultimately determined by the type of filter material used. The detection of high charge, low penetration particulate matter such as alpha particles requires the use of a tight weave, fine grade filter while beta particles and gamma radiation are best counted from more porous filters permitting greater air flow.

Despite the availability of more efficient air sampling filters, felt filters were used for air sampling for gamma counting. Because of its comparatively low resistance to flow, it was selected over the cellulose papers. However, for a meaningful analysis to exist, the efficiency of the filtering system must be known.

This chapter describes work performed to determine the collection efficiency of TFA #2133 felt filters.

The AA Millipore filter was chosen as the standard of measurement because of its near perfect efficiency. However, allowances were necessary since the Millipore filter operates in the velocity range of 200 fpm while the felt filters in the velocity range of 400 fpm.

A. Goetz (10) used Cobalt Oxide fume particles of size 0.01 to 0.02 microns at the rate of ten liters per minute. With this he demonstrated that one particle in 500,000 passed the filter membrane. Merrill Eisenbut (11) measured the efficiency of the Millipore filter as 100 percent down to 0.1 micron.

Thus for particulate matter greater than 0.1 micron, one may use Millipore filters with confidence that its efficiency is 100 percent. However, for atmospheric fission products, the particle size distribution

appears to contain a percentage of matter which is smaller than the 0.1 micron limit. These particles may not be efficiently trapped by the Millipore filter matrix. Crude, qualitative measurements of these particles show that their effect is negligible. This is due, probably since the fine particles have insufficient inertia to penetrate the falling raindrop, and are instead merely pushed aside. Throughout this report the effect of these fine submicron particles is assumed negligible. Further and more exact work will be necessary to completely establish the true extent of the submicron particle's influence. At any rate, when compared to the Millipore filter's high efficiency; that determined for the felt filters should be meaningful.

Outline of Procedure

The felt filter was placed in an Atomic Products Corporation model HAS-4 high volume sampler and run at 35 cfm. An AA Millipore filter was placed in a filter holder, which was then placed directly in the high volume sampler's exhaust and run at 25lpm. Simultaneously, another Millipore filtering system was placed receiving fresh air from outside the Engineering Building.

Each filter was then run for twenty-four hours. The two Millipore filters were mounted and beta counted with a gas flow proportional counter.

Defining the filter efficiency as:

$$n = \frac{\text{cpm, filter no \#1} - \text{cpm, filter no. 2}}{\text{cpm, filter no. 1}}$$

For the arrangement used here, this efficiency was n = 87.6%.

ANALYSES AND DISCUSSION

Method of Analysis

Using the method of analysis as described in chapter five it was possible to quantitatively measure radioisotopes resulting from fallout in both air and rain samples. The laboratory setup meets all criteria of simplicity and adaptability. The data produced is both quantitatively and statistically reliable.

When young fission products are expected (such as in Figure 6), quantitative analyses become quite laborous since approximately eight unknowns exist. However, when analyses are made of long-lived radioactive fallout, only four radioisotopes are needed to describe the sample. With only four "unknowns" the resulting simultaneous equations may be solved directly using determenents. This results in relatively easy equations which yield the four unknown activities after a minimum of calculations. However, for more unknowns, calculations would approach the impossible and computer programs prove to be indispensible. In either case, the computer serves to save time and was used throughout this report.

To check the accuracy of this method, known amounts of seven isotopes were mixed. The sample was then treated as an unknown and the activity of each isotope calculated. The resulting errors are all within or near ten percent which is quite good considering the difficulty of preparing accurate samples.

This method is more than adequate for the quantitative analysis of mixtures containing unknown amounts of gamma-ray emitting radioisotopes.

Isotope	Activity (calculated)	Activity (actual)	Error (%)
Ce^{141}	24,794	22,600	9.7
$CePr^{144}$	18,010	18,200	-1.0
I^{131}	2,534	2,300	10.2
$RuRh^{106}$	9,989	10,120	-1.3
$ZrNb^{95}$	24,104	25,200	-4.3
$BaLa^{140}$	9,090	9,760	-6.6
$CsBa^{137}$	27,100	28,600	-5.3

Atmosphere and Rain Analysis

Itagaki, Koenuma, and Kruger each claim an influence of cloud height upon the final specific rain activity. Itagaki and Koenuma measured the radioactivity of seven rains at five different elevations and found that the specific activity decreased with altitude. On the other hand, the author has made 76 ground level observations; the specific activity of which is plotted versus cloud height. (Figure 35) This data shows no relationship between the specific rain activity and the cloud height. This apparent contradiction may be explained by either of two possible causes. First, the author used only cloud ceiling measurements, and since this makes no allowance for the cloud thickness, the possible error in measurements may cause the apparent discrepency. Secondly, as explained by Kruger, et. al., the distance a raindrop falls may be interpreted to be a measure of the concentration of radioactivity resulting from raindrop evaporation through the unsaturated layer of air near the ground. More precise cloud height measurements are needed to clear this point.

Norris' interpretation suggests that, for summer precipitation, an equation of the form

$$C_R = K_{CA} + kV^n C_A$$

might be more descriptive. In this equation, K_{CA} is a constant for the

specific atmospheric activity, and V is the wind speed, C_R and C_A are respectively the specific rain and air activities, and k and n are additional constants. Figure 33 confirms that some relationship exists between rain activity and wind speed. However, the relationship is linear in nature.

In addition, the specific activity of rain did not appear to be related to the amount of rainfall. This tends to support Greenfield's proposition that the fraction scavenged may be considered to be independent of rainfall intensity.

Figures 29 and 30 show a direct correlation between air activity and rain activity. The gross beta analysis shows two curves, one for early spring rains and another for the summer rains. The difference is probably due to the additional upper atmospheric turbulence as typified during the early spring months.

Since only spring rains were analyzed for the individual isotopes, they do not exhibit the two curve characteristic, see Figures 29 and 30. However, in each case the relationship between air and rain activity appears to be linear.

Thus it appears that rain activity depends basically upon two parameters, air activity and wind speed. Using the data of Figures 29 and 30, it appears this relationship is of the form

$$A_{Rain}/A_{Air} = (C_1 + C_2V).$$

If the velocity is measured in miles per hour, this equation becomes $A_R/A_A = 190 + 15.4V$. Unfortunately, the only wind speeds available were average daily values; the true wind speed may vary greatly with time. More closely measured velocities may show a better correlation

Using the average daily values, a better fit to the data is found if the equation is used as

$A_R/A_A = 324 + 10V$ (see Figure 36).

Using the field study as described above, it is possible to hypothesize about the transfer mechanism of fallout radionuclides from the atmosphere to the rain. Since the amount of radioactivity captured by the rain was found to be proportional to the amount of radioactive particles presented to the falling raindrop, it is justifiable to assume that the mechanism of transport is basically through direct collision. However, a certain percentage of particles may be captured by water cloud droplets. It does appear that this percentage is small. A more complete experimental program will be necessary to establish the actual amount. Also, the usage of daily averages has reduced the accuracy of the study. It is recommended that averages of smaller time duration be used in any further studies.

CONCLUSIONS

The following general conclusions are drawn from this study.

(1) The Gamma-ray Spectrometric system of analysis and the laboratory setup meets all criteria of simplicity and are both quantitatively useful and statistically reliable.

(2) The relationship between rain activity and air activity at the time of rain appears reasonably well described by $C_r = 334\ C_a$.

(3) Some relationship between rain activity and wind speed at the time of rain appears evident. However, daily mean velocities do not seem sufficient. Possibly, wind velocities averaged during the rainfall would be more descriptive. The data is described, quite accurately, by the relationship, $C_r = 334\ C_a + 10\ V$.

(4) The specific activity of rain did not appear to be related to either cloud height or rain intensity.

(5) The mechanism of fallout radionuclide transfer from the atmosphere to rain is basically through direct collisions between falling raindrops and the radioactive particulate matter.

(6) Daily averages of air activity, rain activity, wind speed, cloud height, and rain intensity are of too long a time duration. Further studies should use smaller time averages of the above parameters.

(7) Concentrations of submicron radioactive particle are present in the atmosphere. However, they are not scavenged by the rain and may therefore be neglected in considerations on the mechanisms of radionuclide transfer.

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APPENDIX A

GAMMA-RAY SPECTRA OF THE FALLOUT RADIOISOTOPES

The gamma-ray spectra of Ce^{141}, $CePr^{144}$, I^{131}, Ru^{103}, $RuRh^{106}$, $ZrNb^{95}$ and $BaLa^{140}$ are presented as Figures 16 through 23.

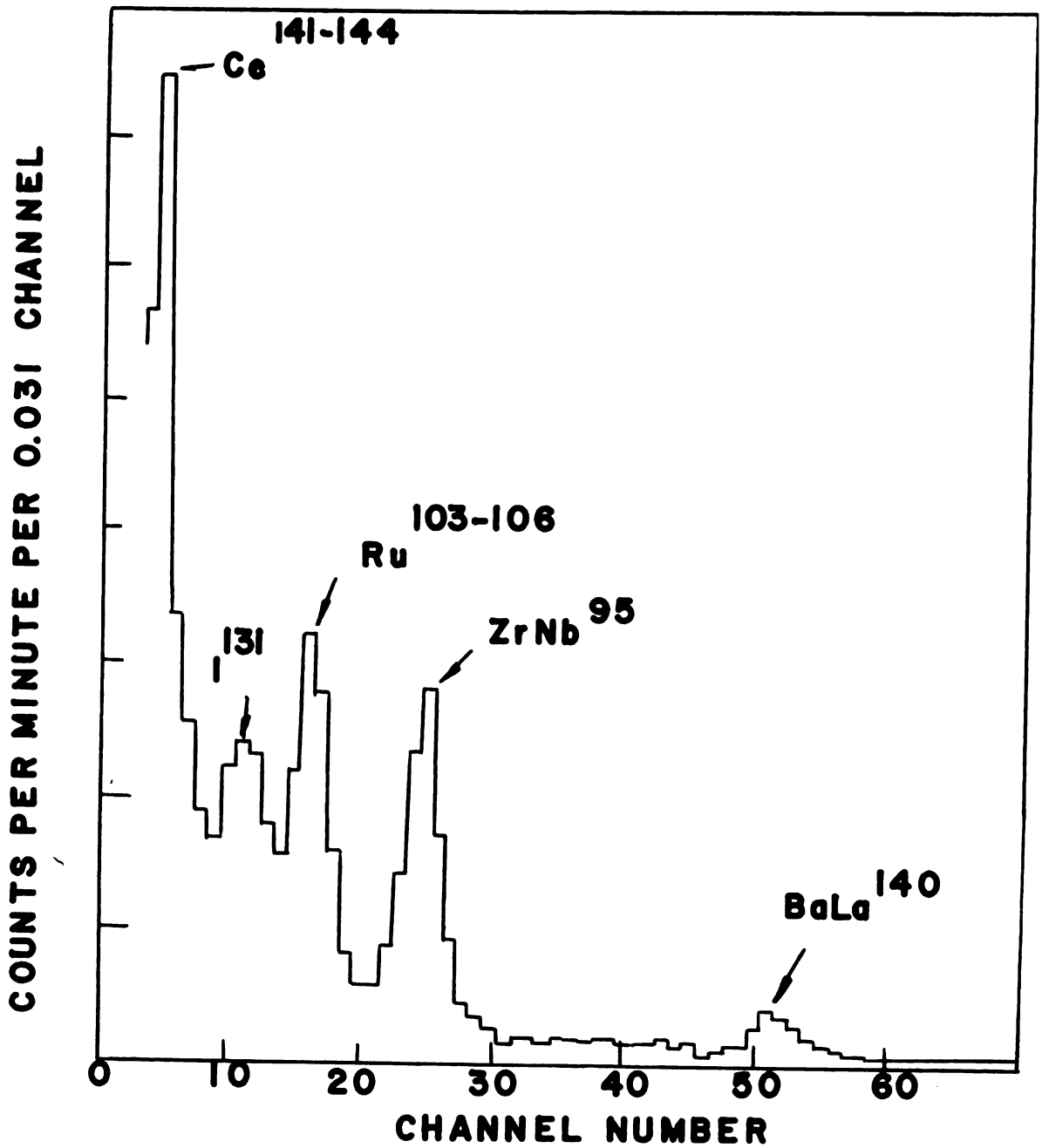

FIGURE - 8

GAMMA-RAY SPECTRUM OF VEGETATION SAMPLED 3-4 WEEKS AFTER FALLOUT

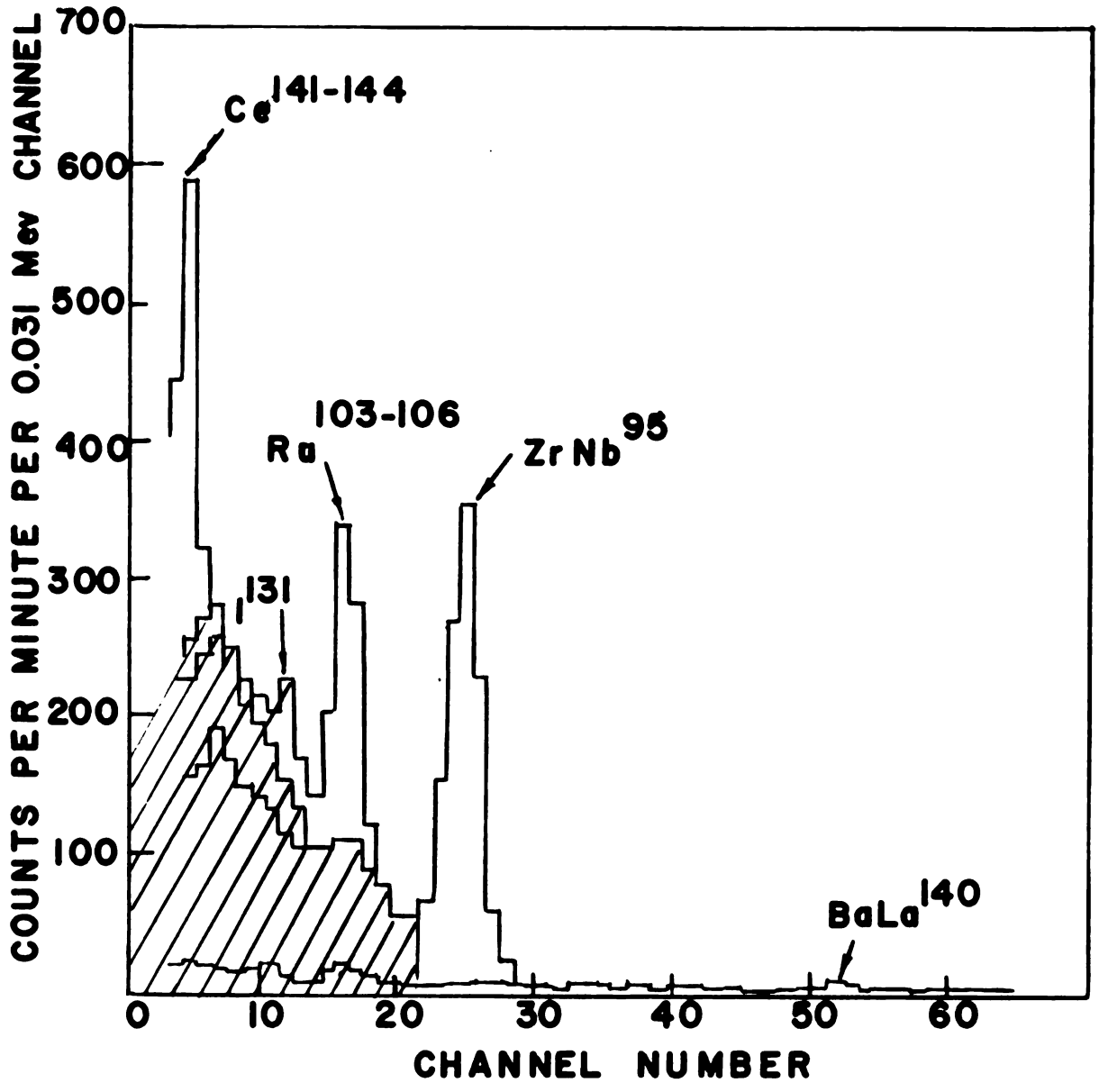

FIGURE - 9

GAMMA-RAY SPECTRUM OF THE FALLOUT RADIOISOTOPES SHOWING THE ENERGY INCREMENT AND COMPTON CORRECTION FOR THE MEASUREMENT OF EACH ISOTOPE

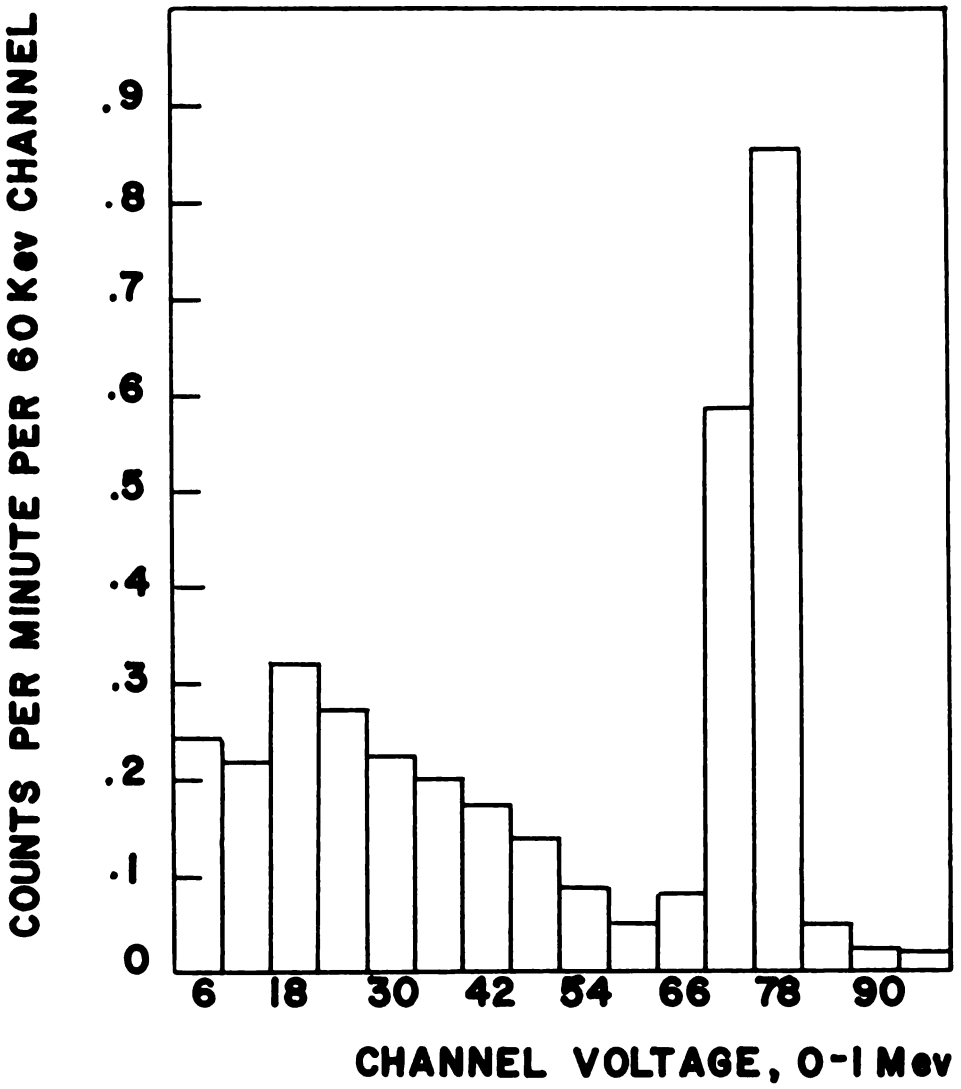

FIGURE - 16

Zr Nb - 95 EQUILIBRIUM MIXTURE
GAMMA-RAY SPECTRUM 1 oz.
AQUEOUS SOLUTION, 1uuc Zr - 95

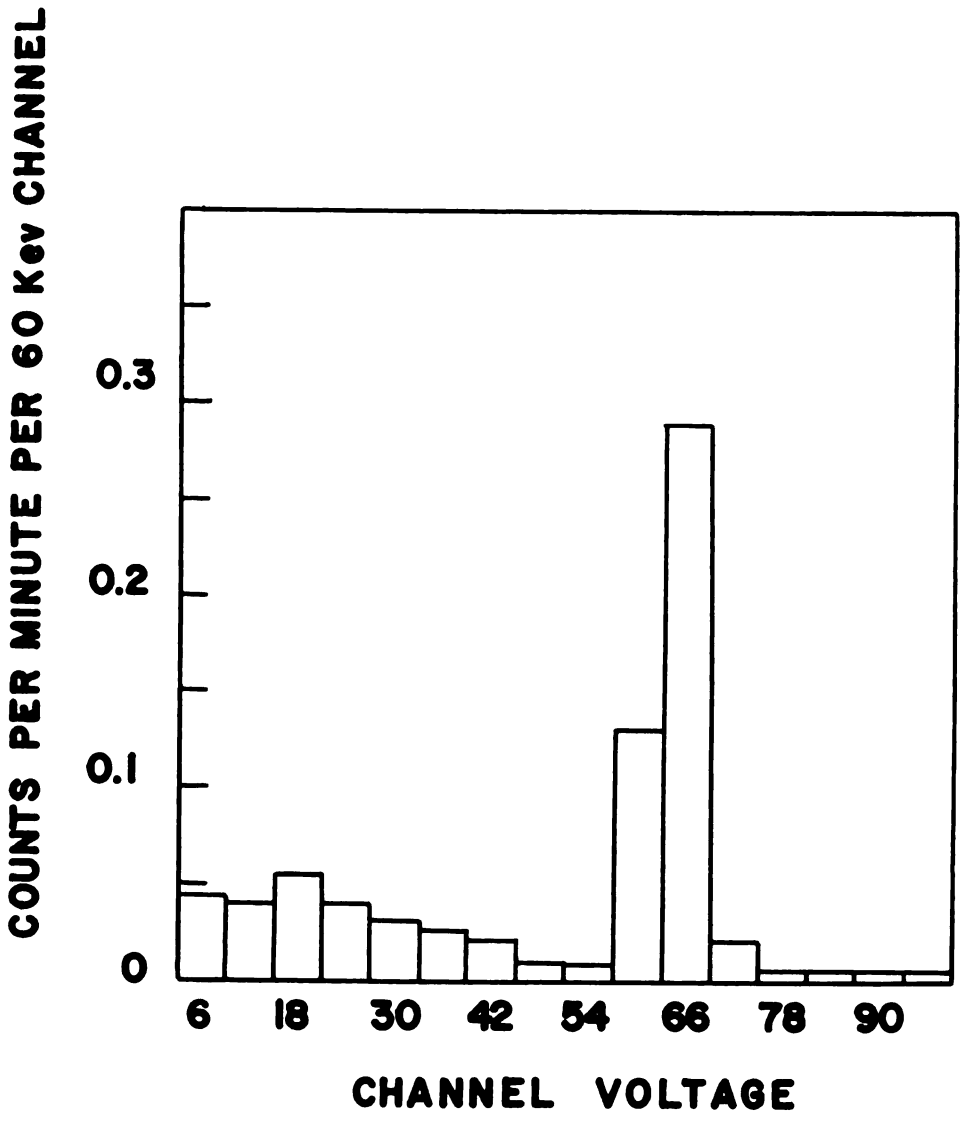

FIGURE - 17

Cs-137 GAMMA-RAY SPECTRUM
1 oz. BOTTLE AQUEOUS SOLUTION
1 uuc.

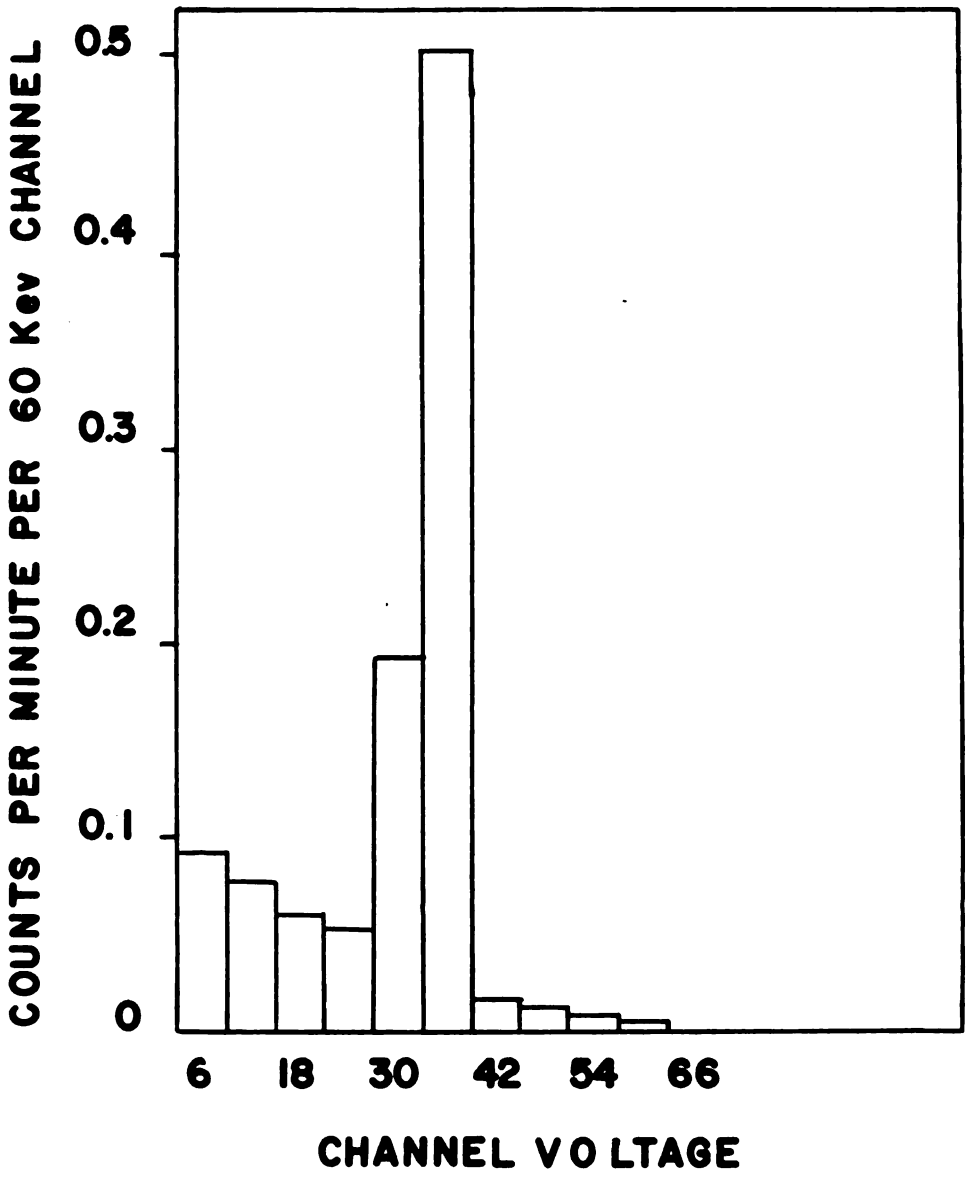

FIGURE - 18

I-131 GAMMA-RAY SPECTRUM
1 oz. BOTTLE AQUEOUS SOLUTION,
1 uuc.

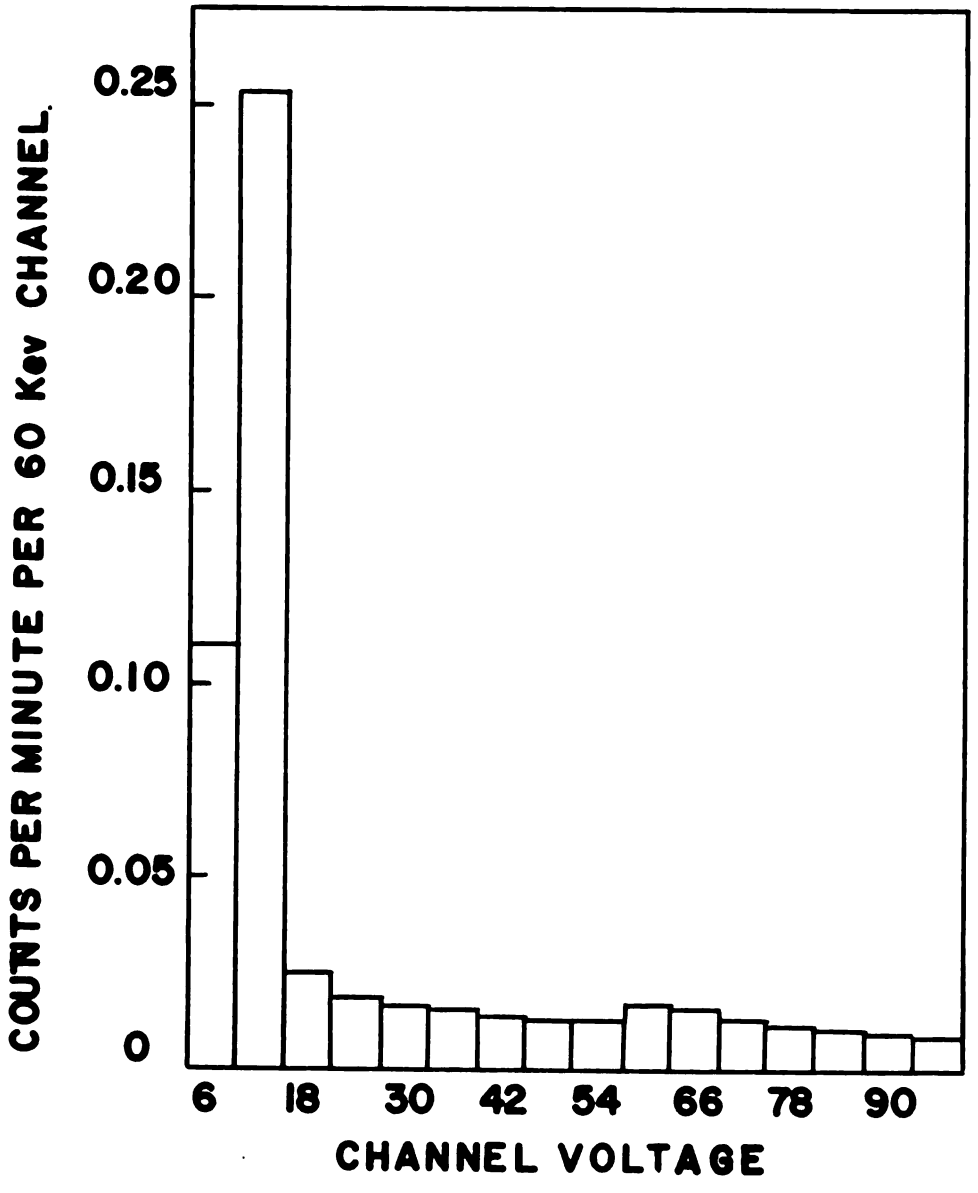

FIGURE - 19

CePr-144 EQUILIBRIUM MIXTURE GAMMA-RAY SPECTRUM, 1 OUNCE AQUEOUS SOLUTION, luuc Ce-144

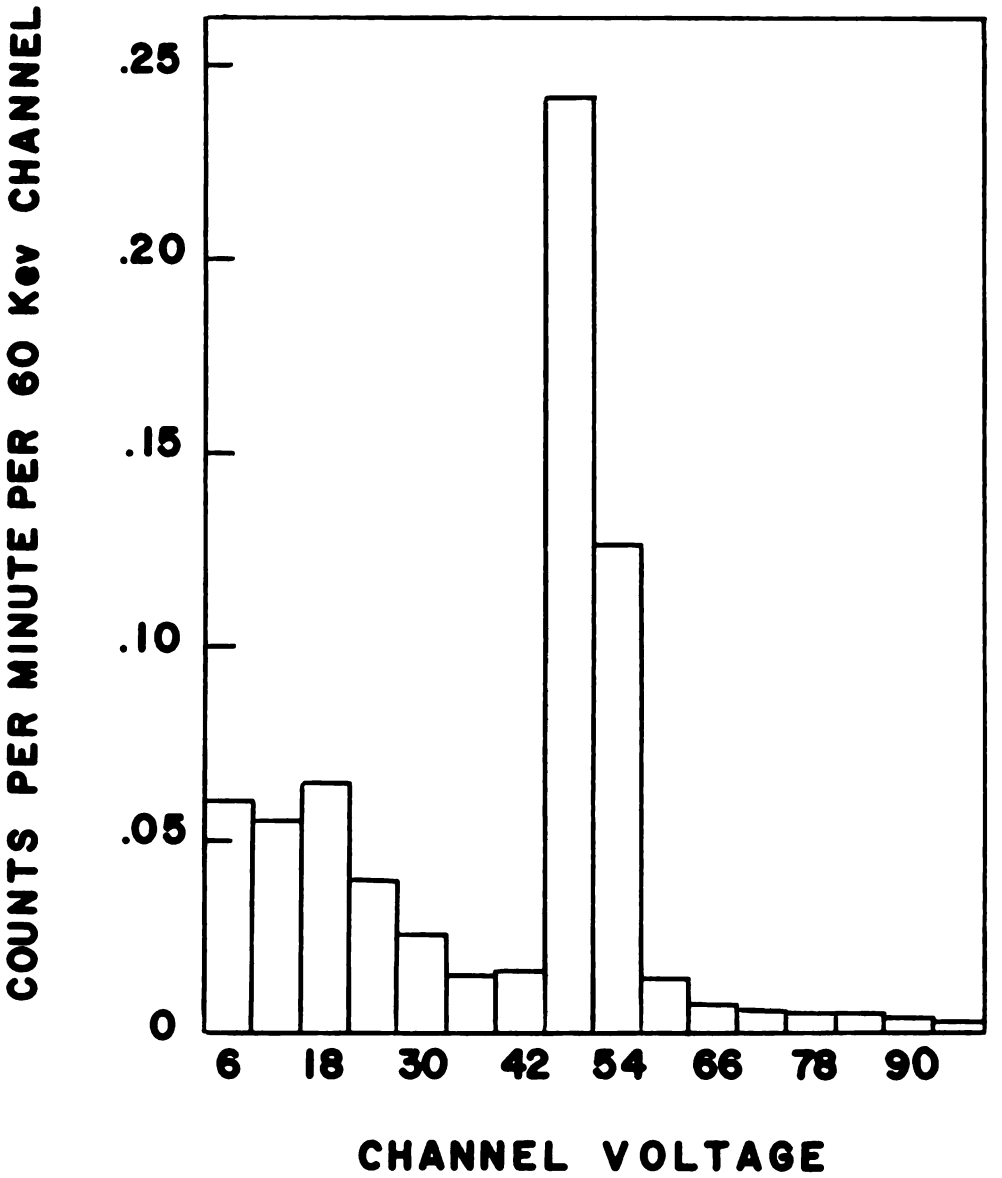

FIGURE-20

Ru-103 GAMMA-RAY SPECTRUM

1 oz. BOTTLE AQUEOUS SOLUTIO

1 uuc

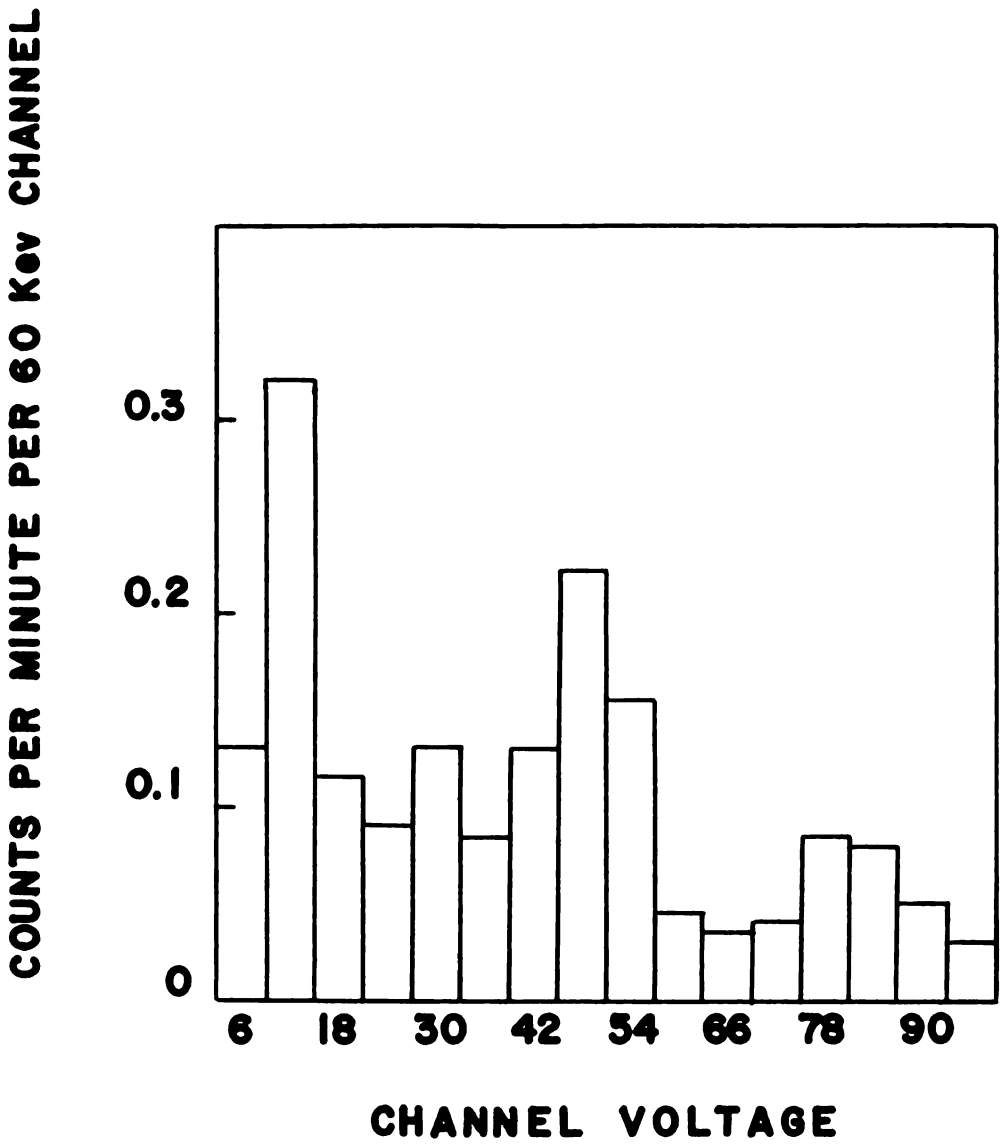

FIGURE - 21

BaLa-140 EQUILIBRIUM MIXTURE GAMMA-RAY SPECTRUM
1oz. AQUEOUS SOLUTION, luuc Ba-140

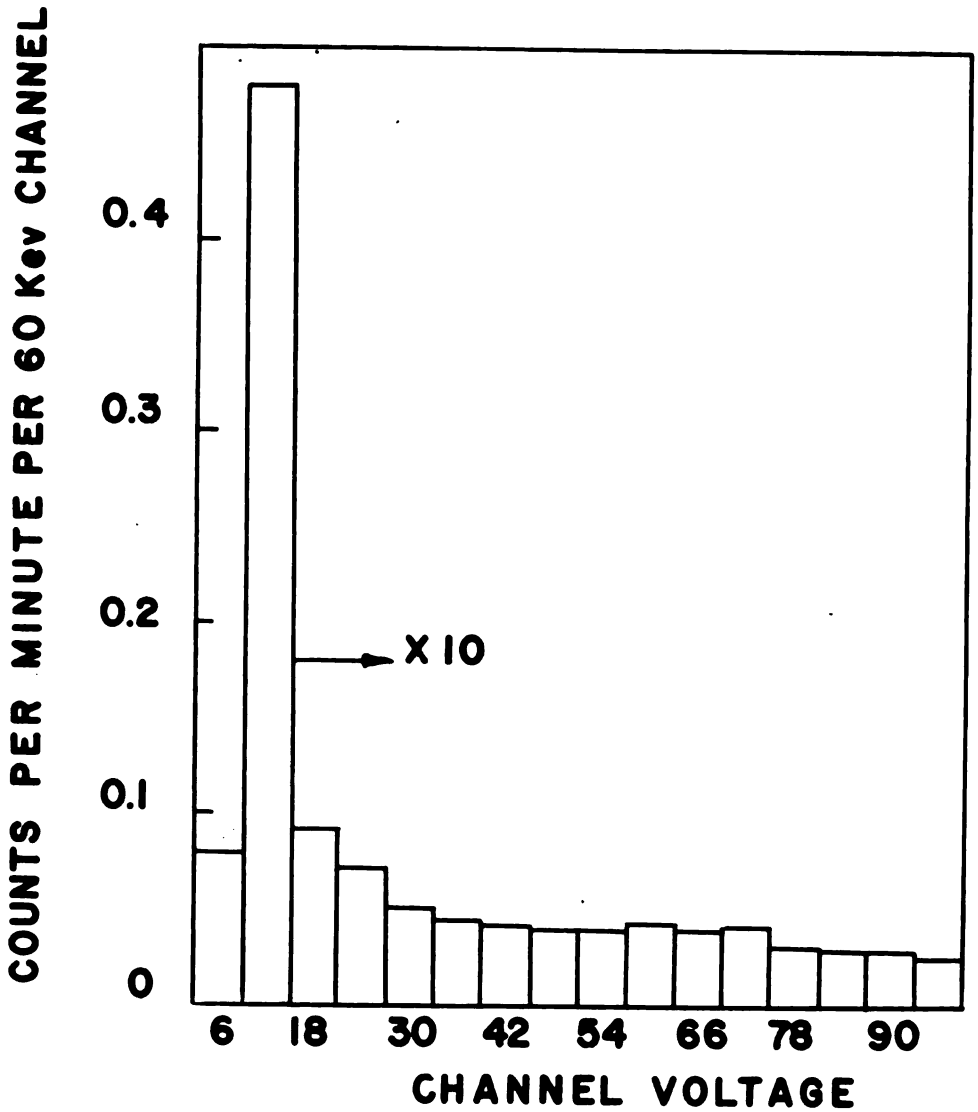

FIGURE - 22

Ce-141 GAMMA-RAY SPECTRUM
1 oz. BOTTLE AQUEOUS 1uuc
SOLUTION.

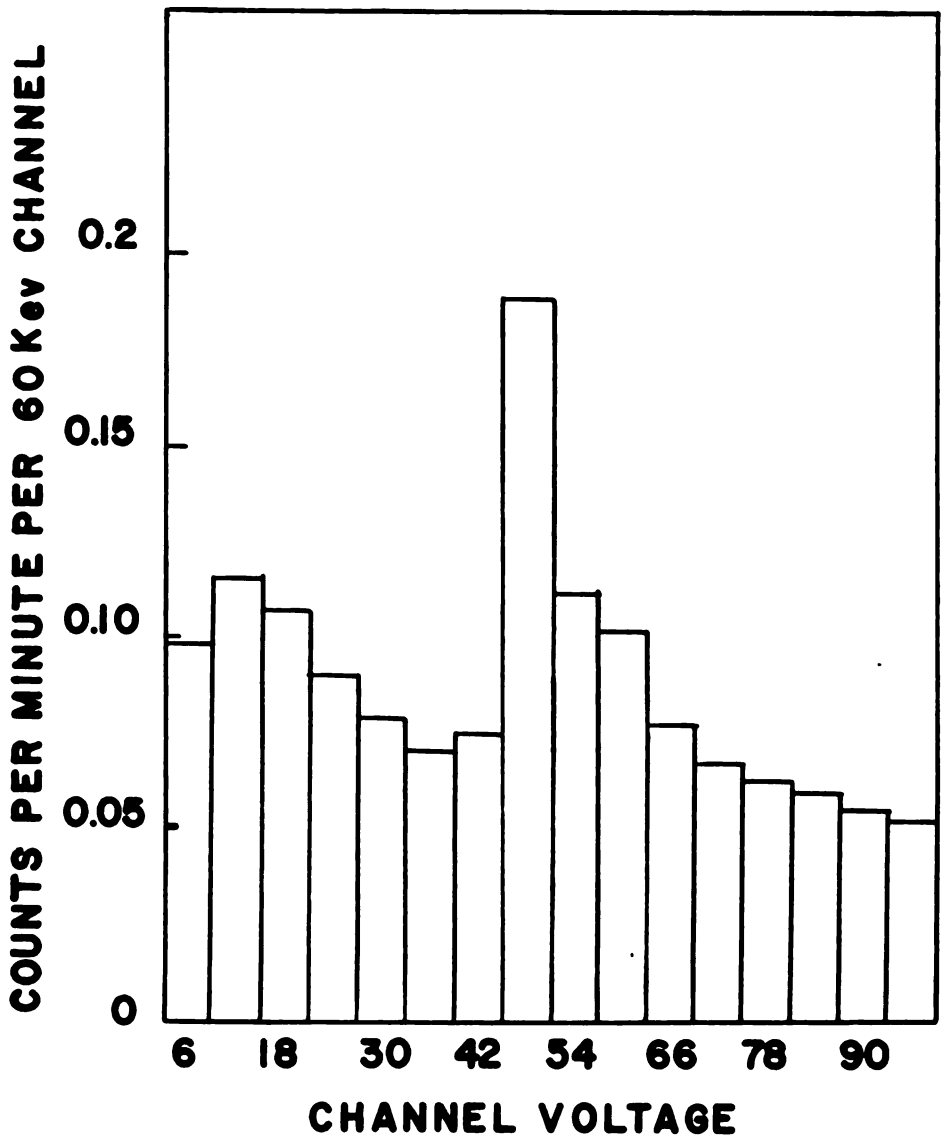

FIGURE - 23

RuRh-106 EQUILIBRIUM
MIXTURE GAMMA-RAY SPECTRUM
1 oz. BOTTLE AQUEOUS SOLUTION
1 oz. Ru-106

APPENDIX B

COMPUTER PROGRAMS FOR SOLUTION OF EIGHT AND FOUR COMPONENT SYSTEMS

```
*       PROGRAM TO SOLVE 8 X 8 SIMULTANEOUS EQUATIONS  HAUGEN
C       PROGRAM TO SOLVE 8 X 8 SIMULTANEOUS EQUATIONS
        COMMON A
        DIMENSION Z(8,9), C(N,8), A(8,9)
      3 FORMAT(F10.4,F10.4,F10.4,F10.4,F10.4,F10.4,F10.4,F10.4)
        READ 3, ((Z(I,J), J=1,8), I=1,8)
        READ 3,((C(I,J),J=1,8),I=1,N)
        PRINT 5
        PRINT 6
        PRINT 8
        DO 100 L5=1,N
        DO 20 I=1,8
        DO 20 J=1,8
     20 A(I,J)=Z(I,J)
        DO 30 I=1,8
     30 A(I,9)=C(L5,I)
        CALL SIMEQ
      5 FORMAT(30HAUGEN-SIMULTANEOUS EQUATIONS)
      6 FORMAT(1HO)
      7 FORMAT(1H2,I3,6X,F10.3,2X,F10.3,2X,F10.3,2X,F10.3,2X,F10.3,2X,
     1F10.3,2X,F10.3,2X,F10.3)
      8 FORMAT(1HO,2X,1HI,15X,1HA,11X,1HB,11X,1HC,11X,1HD,11X,1HE,11X,
     11HF,11X,1HG,11X,1HH)
        PRINT 7,15,(A(L1,9),L1=1,8)
    100 CONTINUE
        STOP
        END
C       SUBPROGRAM SIMEQ
        SUBROUTINE SIMEQ
        COMMON A
        DIMENSION A(8,9),B(8,9)
        IF ACCUMULATOR OVERFLOW 2,2
      2 IF DIVIDE CHECK 23,23
     23 NR=8
        NC=NR+1
        DO 1 M=1,NR
        MM=M
        NN=M+1
     21 DO 18 N=M,NC
     18 B(M,N)=A(M,N)
        DO 4 IT=NN,NC
        A(M,IT)=A(M,IT)/A(M,M)
        IF DIVIDE CHECK 3,22
     22 IF ACCUMULATOR OVERFLOW 3,4
      4 CONTINUE
        A(M,M)=1.
        GO TO 20
      3 DO 19 IT=M,NC
     19 A(M,IT)=B(M,IT)
        MM=MM+1
        IF(MM-NR) 5,5,6
      5 DO 7 N=M,NC
        T=A(M,N)
        A(M,N)=A(MM,N)
```

```
 7 A(MM,N)=T
   GO TO 21
 6 PAUSE 01
   RETURN
20 DO 1 N=1,NR
   IF (N-M) 8,1,8
 8 DO 9 IT=MM,NC
   A(N,IT)=A(N,IT)*A(N,M)
   IF ACCUMULATOR OVERFLOW 6,9
 9 CONTINUE
   A(N,M)=0.
 1 CONTINUE
   RETURN
   END
```

IMPORTANT:

In lines 4, 7, and 11: Replace letter 'N' with the number of days of data to be analyzed.

FIGURE 24

COMPUTER PROGRAM FOR EIGHT

COMPONENT SYSTEM

```
*       PROGRAM TO SOLVE 4 X 4 SIMULTANEOUS EQUATIONS     HAUGEN
C       PROGRAM TO SOLVE 4 X 4 SIMULTANEOUS EQUATIONS
        COMMON A
        DIMENSION Z(4,5), C(N,4), A(4,5)
      3 FORMAT(F10.4,F10.4,F10.4,F10.4)
        READ 3, ((Z(I,J), J=1,4), I=1,4)
        READ 3,((C(I,J),J=1,4),I=1,N)
        PRINT 5
        PRINT 6
        PRINT 8
        DO 100 L5=1,N
        DO 20 I=1,4
        DO 20 J=1,4
     20 A(I,J)=Z(I,J)
        DO 30 I=1,4
     30 A(I,5)=C(L5,I)
        CALL SIMEQ
      5 FORMAT(30 HAUGEN-SIMULTANEOUS EQUATIONS)
      6 FORMAT(1HO)
      7 FORMAT (1H2,I3,6X,F10.3,2X,F10.3,2X,F10.3,2X,F10.3)
      8 FORMAT(1HO,2X,1HI,15X,1HA,11X,1HB,11X,1HC,11X,1HD)
        PRINT 7,L5,(A(L1,5),L1=1,4)
    100 CONTINUE
        STOP
        END
C       SUBPROGRAM SIMEQ
        SUBROUTINE SIMEQ
        COMMON A
        DIMENSION A(4,5),B(4,5)
        IF ACCUMULATOR OVERFLOW 2,2
      2 IF DIVIDE CHECK 23,23
     23 NR=4
        NC=NR+1
        DO 1 M=1,NR
        MM=M
        NN=M+1
     21 DO 18 N=M,NC
     18 B(M,N)=A(M,N)
        DO 4 IT=NN,NC
        A(M,IT)=A(M,IT)/A(M,M)
        IF DIVIDE CHECK 3,22
     22 IF ACCUMULATOR OVERFLOW 3,4
      4 CONTINUE
        A(M,M)=1.
        GO TO 20
      3 DO 19 IT=M,NC
     19 A(M,IT)=B(M,IT)
        MM=MM+1
        IF(MM-NR) 5,5,6
      5 DO 7 N=M,NC
        T=A(M,N)
        A(M,N)=A(MM,N)
      7 A(MM,N)=T
        GO TO 21
```

```
 6 PAUSE 01
   RETURN
20 DO 1 N=1,NR
   IF (N-M) 8,1,8
 8 DO 9 IT=MN,NC
   A(N,IT)=A(N,IT)-A(M,IT)*A(N,M)
   IF ACCUMULATOR OVERFLOW 6,9
 9 CONTINUE
   A(N,M)=0.
 1 CONTINUE
   RETURN
   END
```

IMPORTANT:

In lines 4, 7, and 11; Replace letter 'N' with the number of days of data to be analyzed.

FIGURE 25

COMPUTER PROGRAM FOR FOUR

COMPONENT SYSTEM

APPENDIX C

ANALYSIS OF SAMPLES

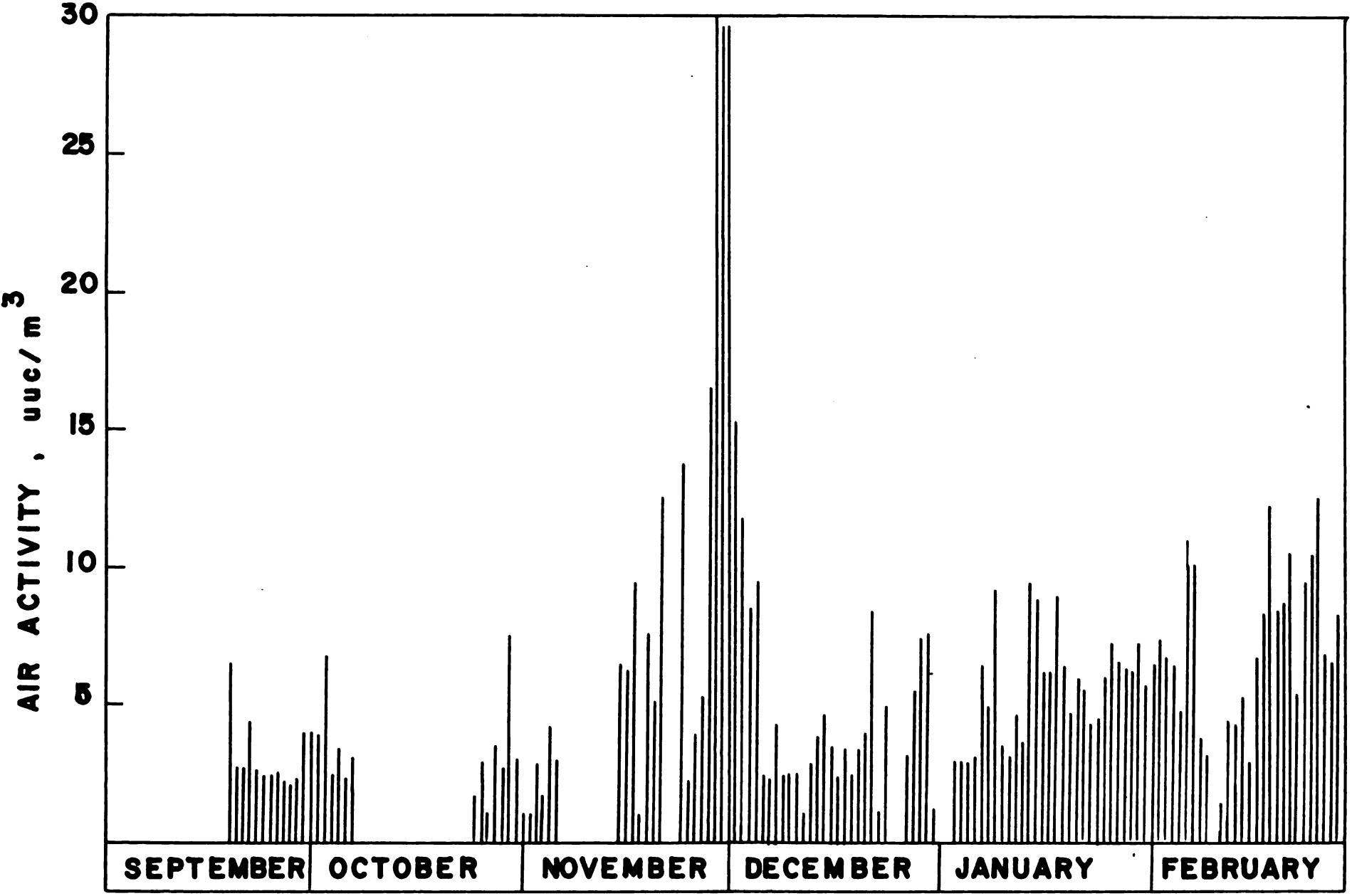

FIGURE - 26

DAILY (gross beta) AIR ACTIVITY

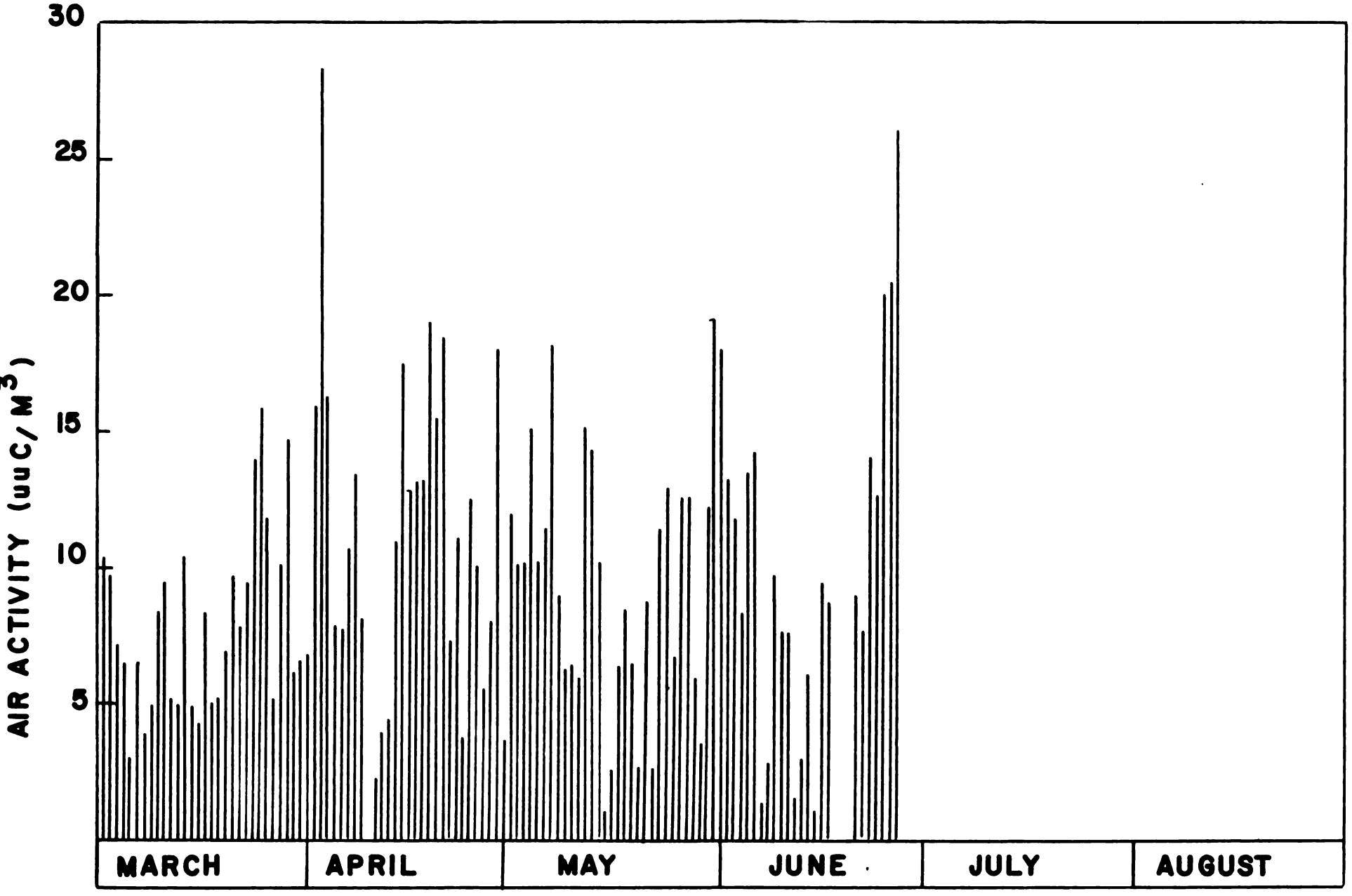
30
25
20
15
10
5
AIR ACTIVITY (uuC/M3)
MARCH
APRIL
MAY
JUNE
JULY
AUGUST

FIGURE - 26

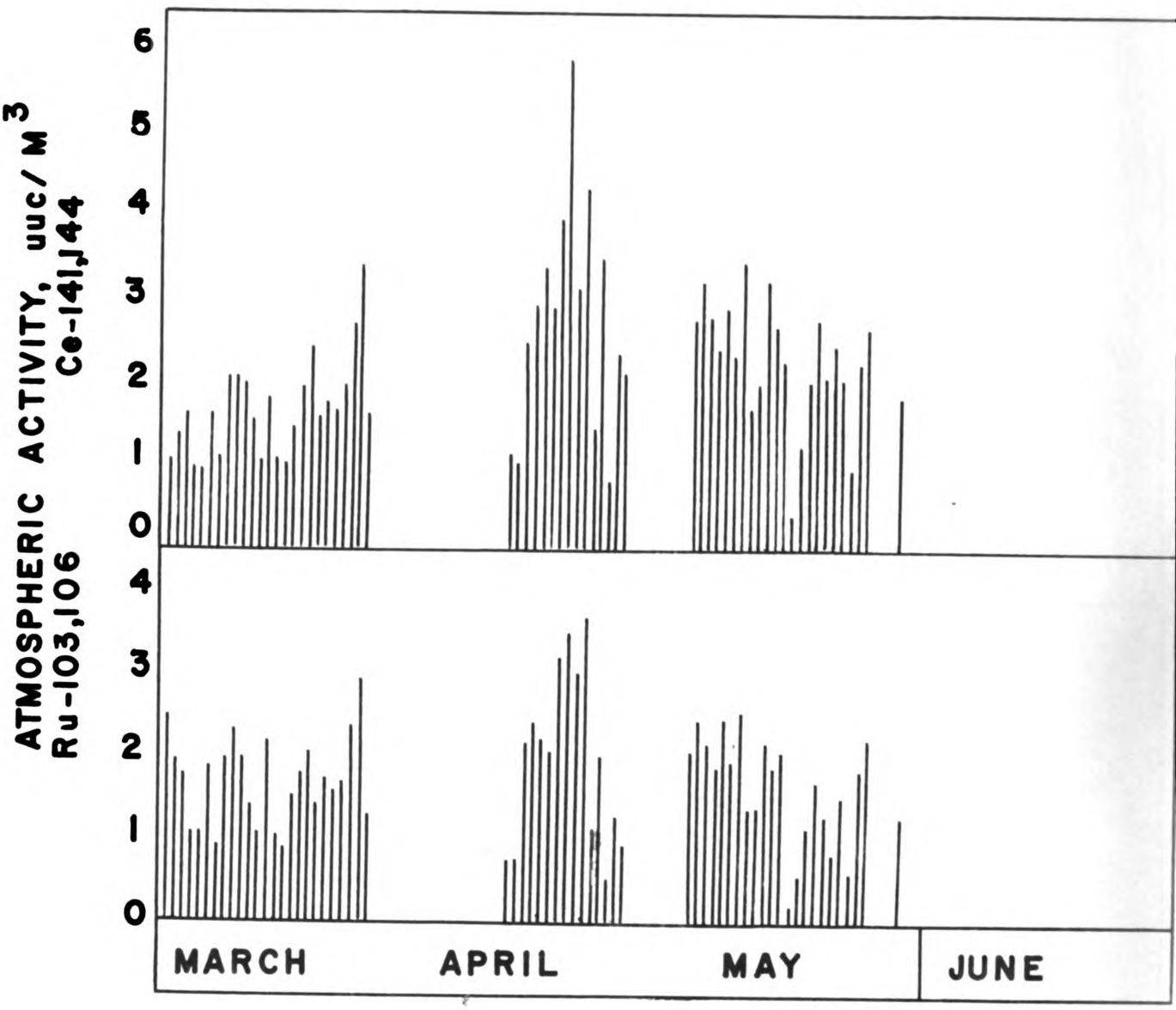

FIGURE - 27
DAILY AIR ACTIVITY
Ce-141,144 & Ru-103,106

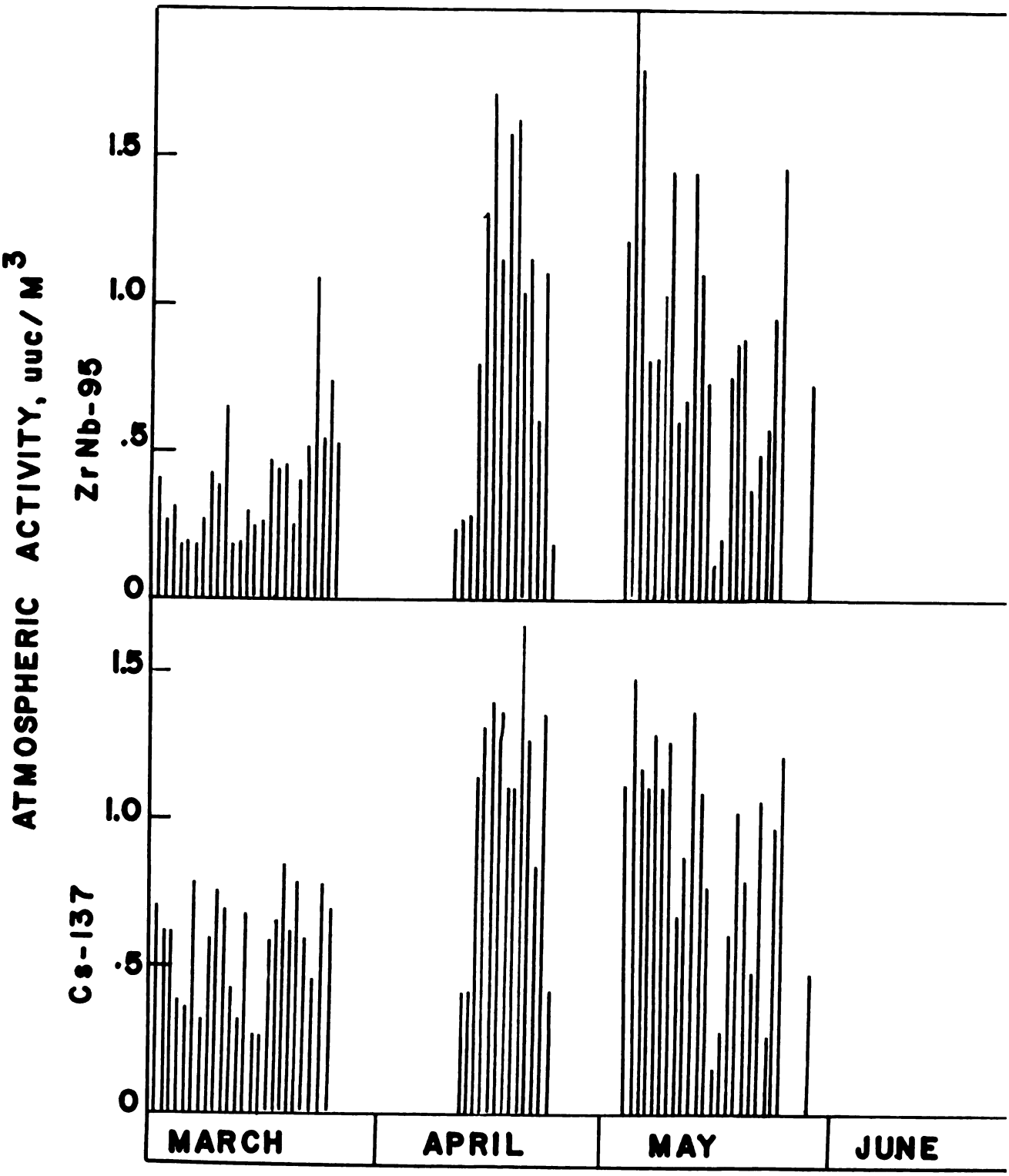

FIGURE - 28
DAILY AIR ACTIVITY
Cs-137 & ZrNb-95

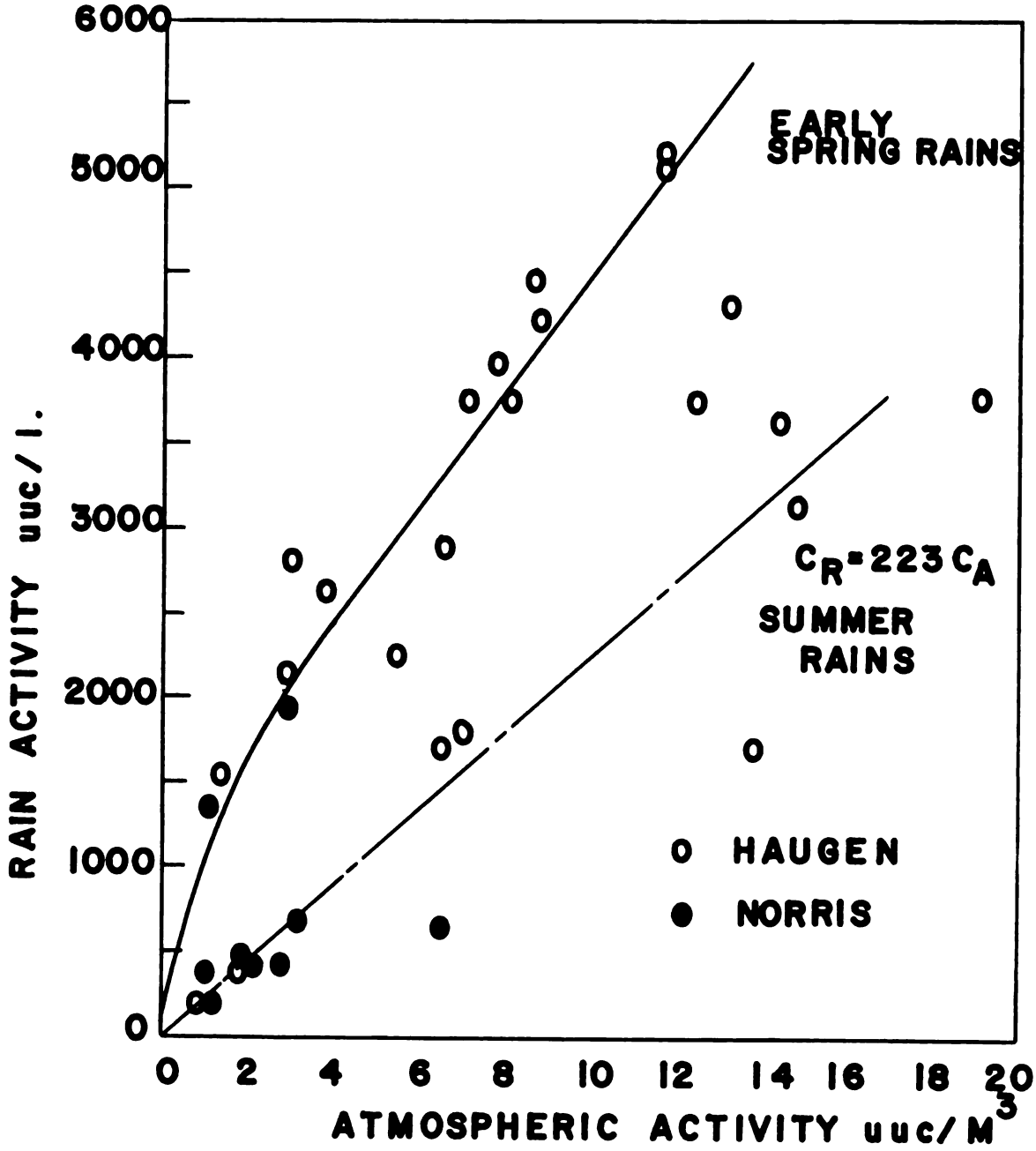

FIGURE - 29
RAIN VERSUS AIR ACTIVITY
GROSS BETA

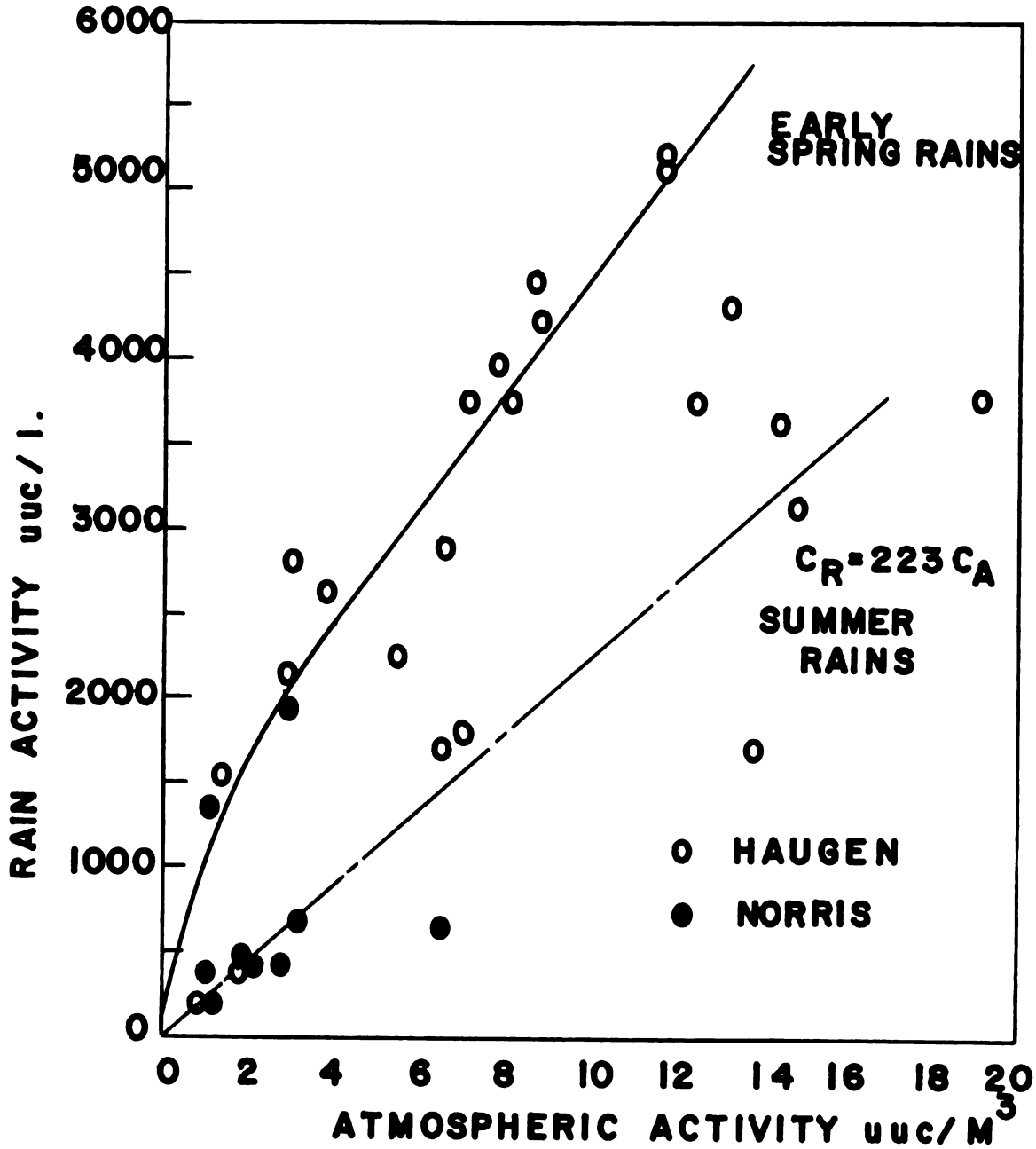

FIGURE - 29
RAIN VERSUS AIR ACTIVITY
GROSS BETA

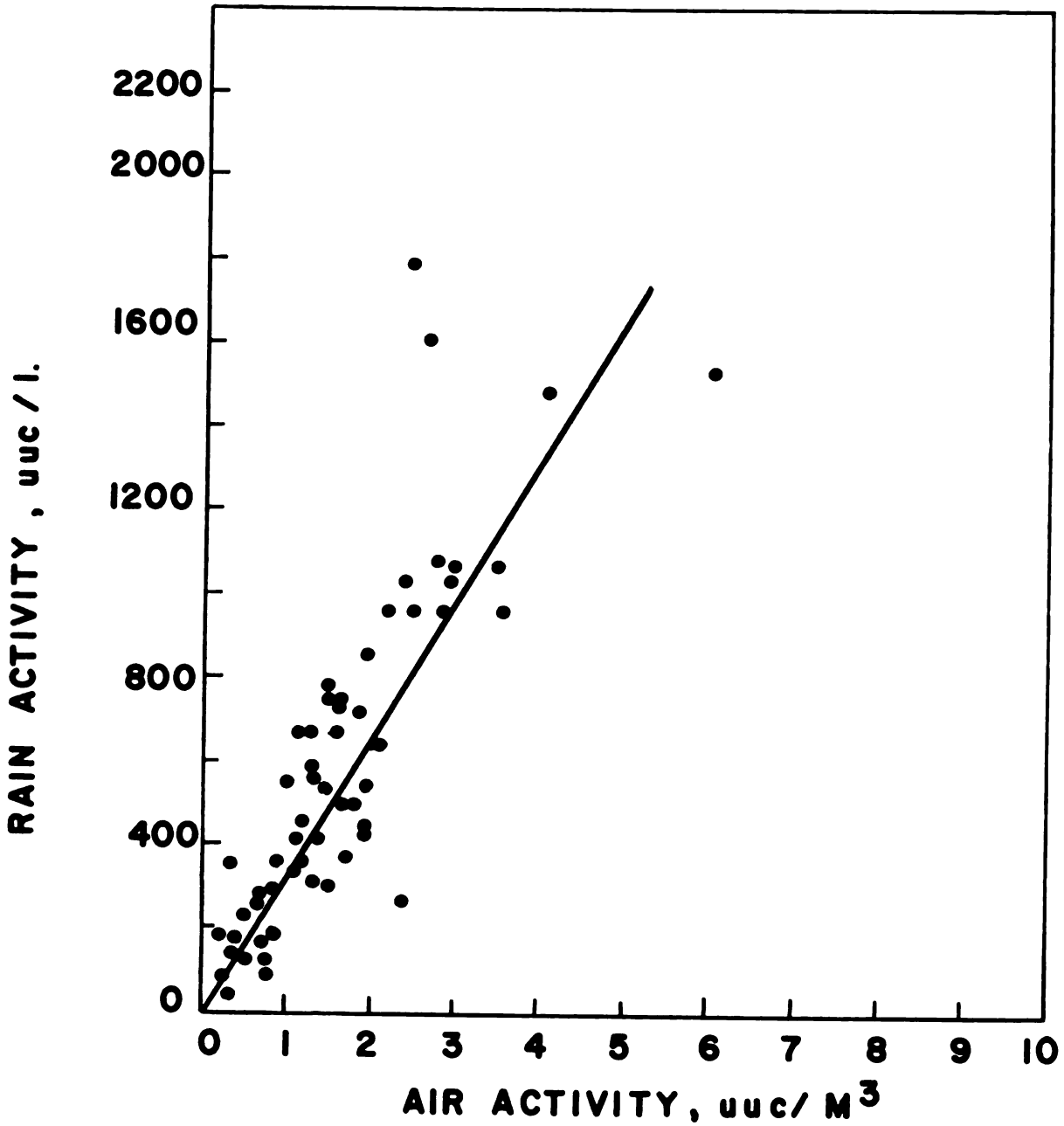

FIGURE - 30
RAIN VERSUS AIR ACTIVITY
(INDIVIDUAL ISOTOPES)

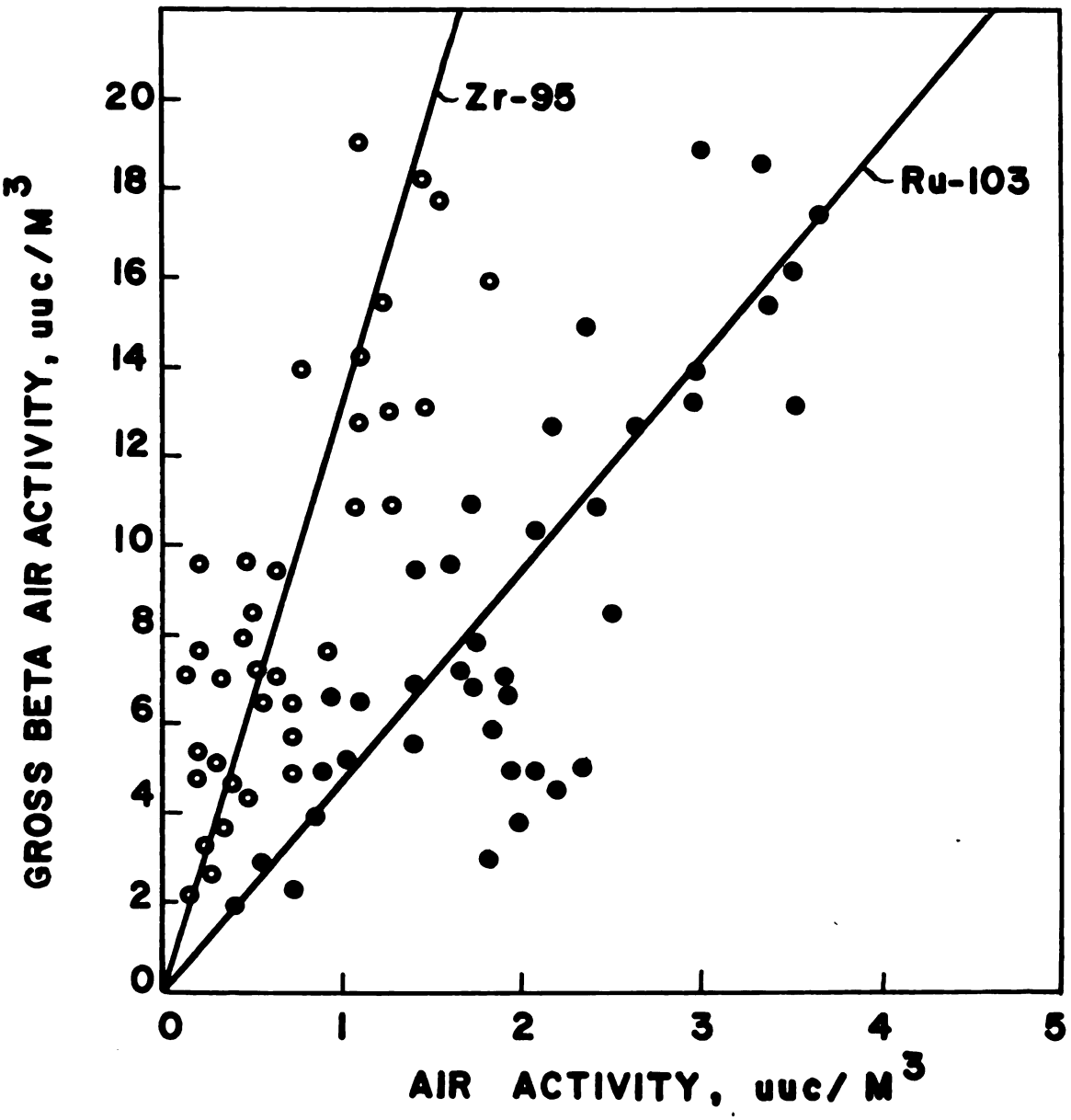

FIGURE-31

GROSS BETA AIR ACTIVITY VERSUS Zr-95 & Ru-103 AIR ACTIVITY.

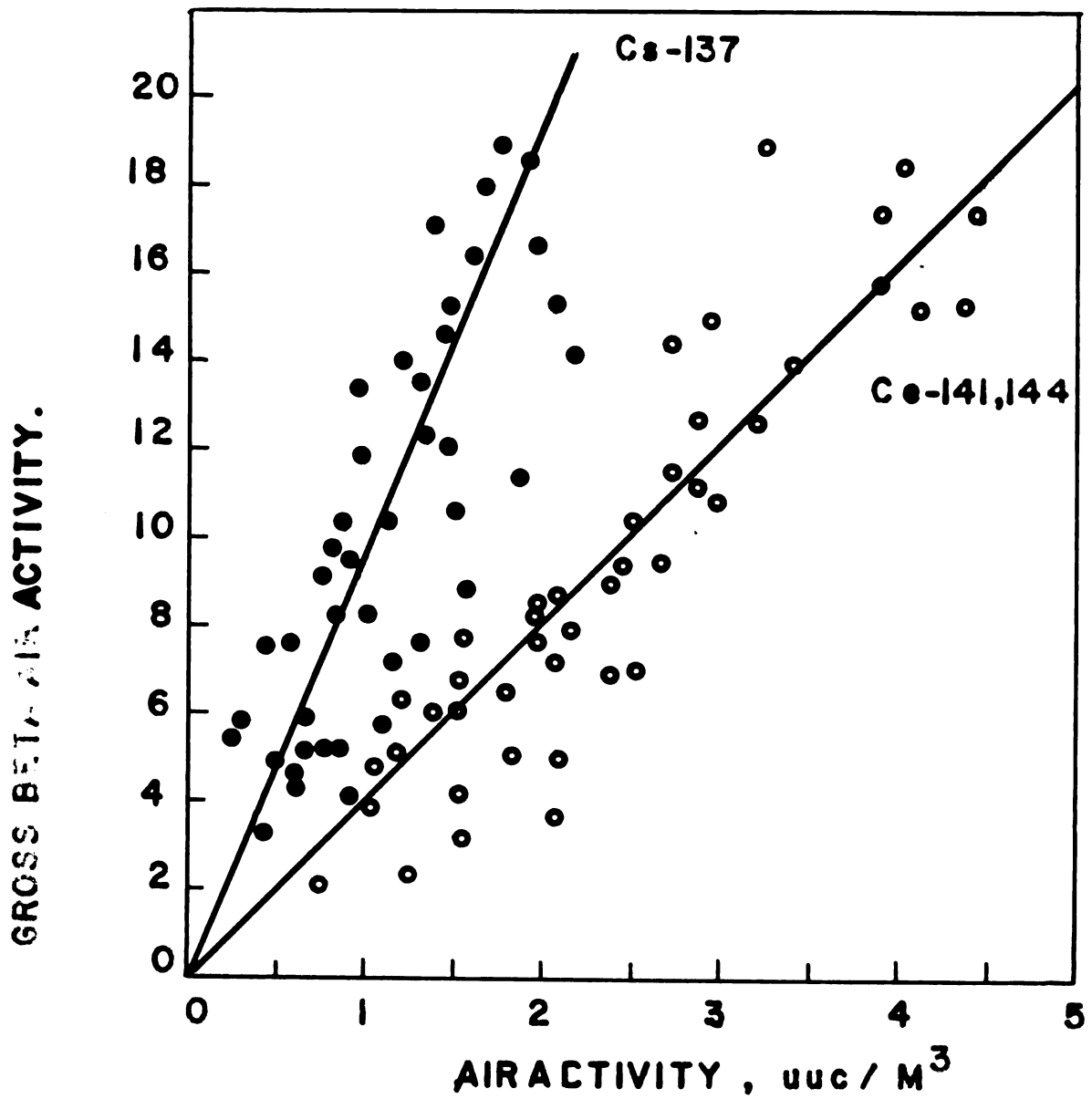

FIGURE-32

GROSS BETA AIR ACTIVITY VERSUS Cs-137 & Ce-141,144 AIR ACTIVITY.

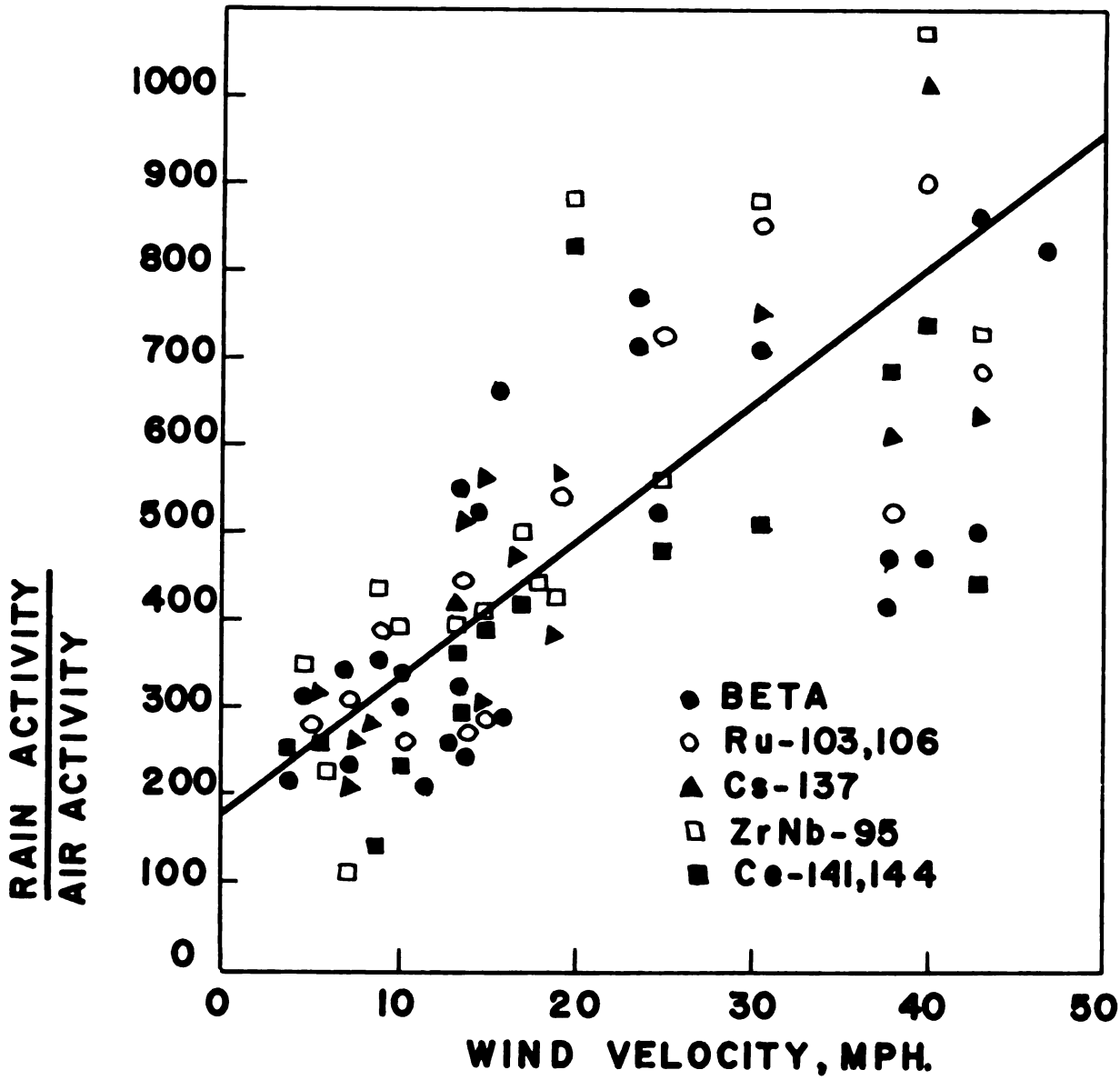

FIGURE - 33
RAIN/AIR ACTIVITY VERSUS
WIND SPEED

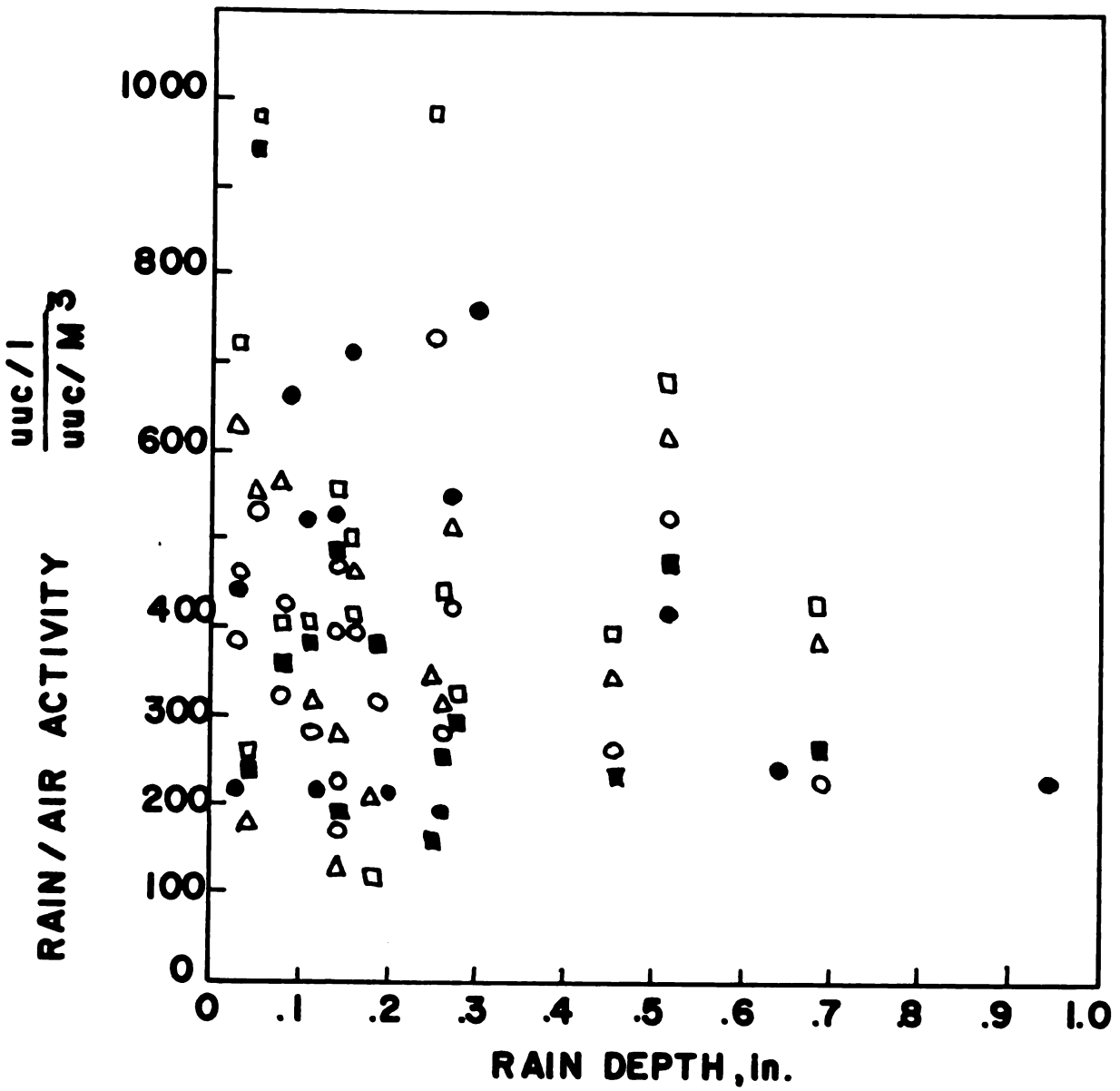

FIGURE-34

RAIN/AIR ACTIVITY VERSUS

RAIN DEPTH

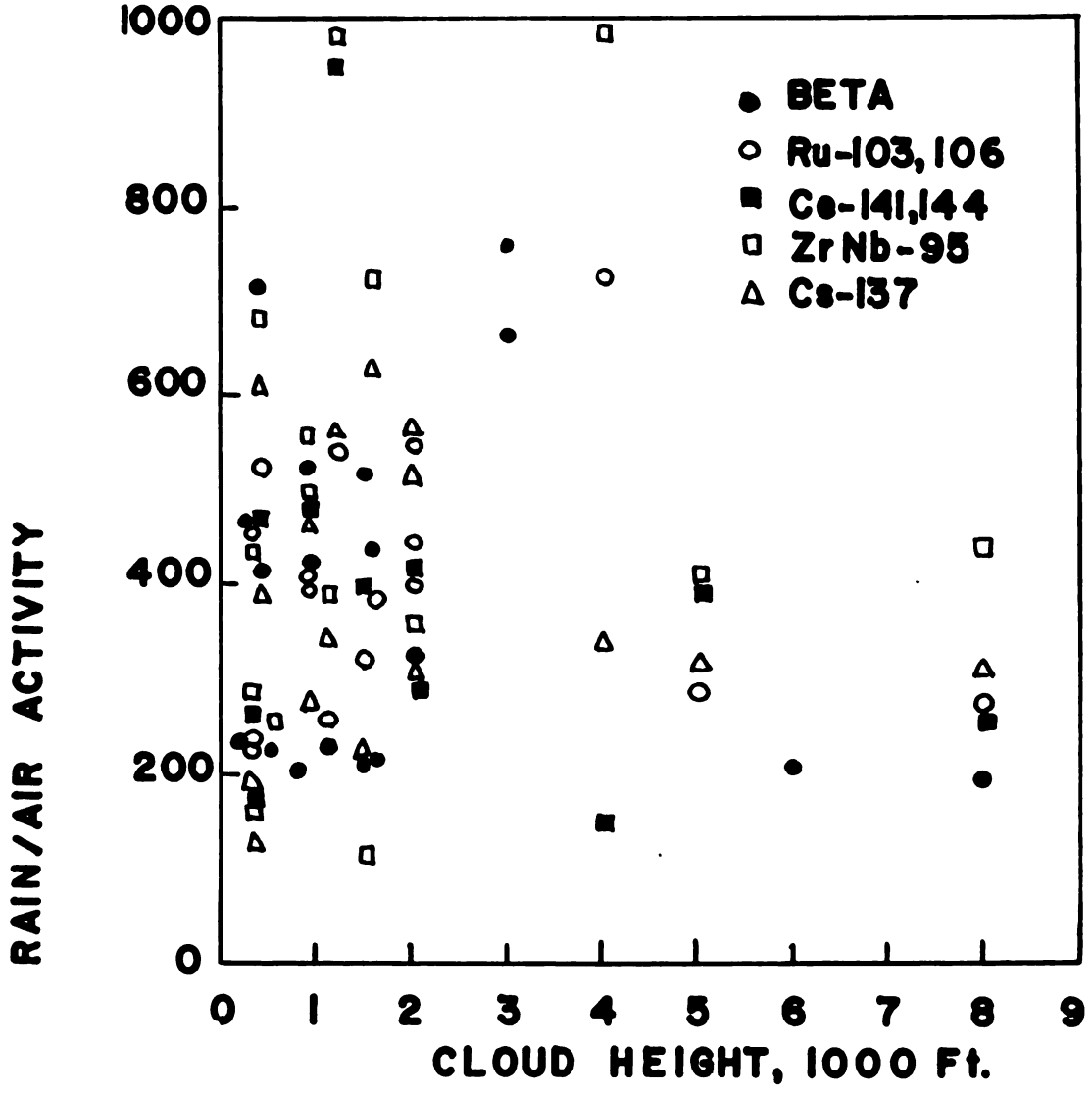

FIGURE-35
RAIN/ACTIVITY / AIR ACTIVITY
VERSUS CLOUD HEIGHT

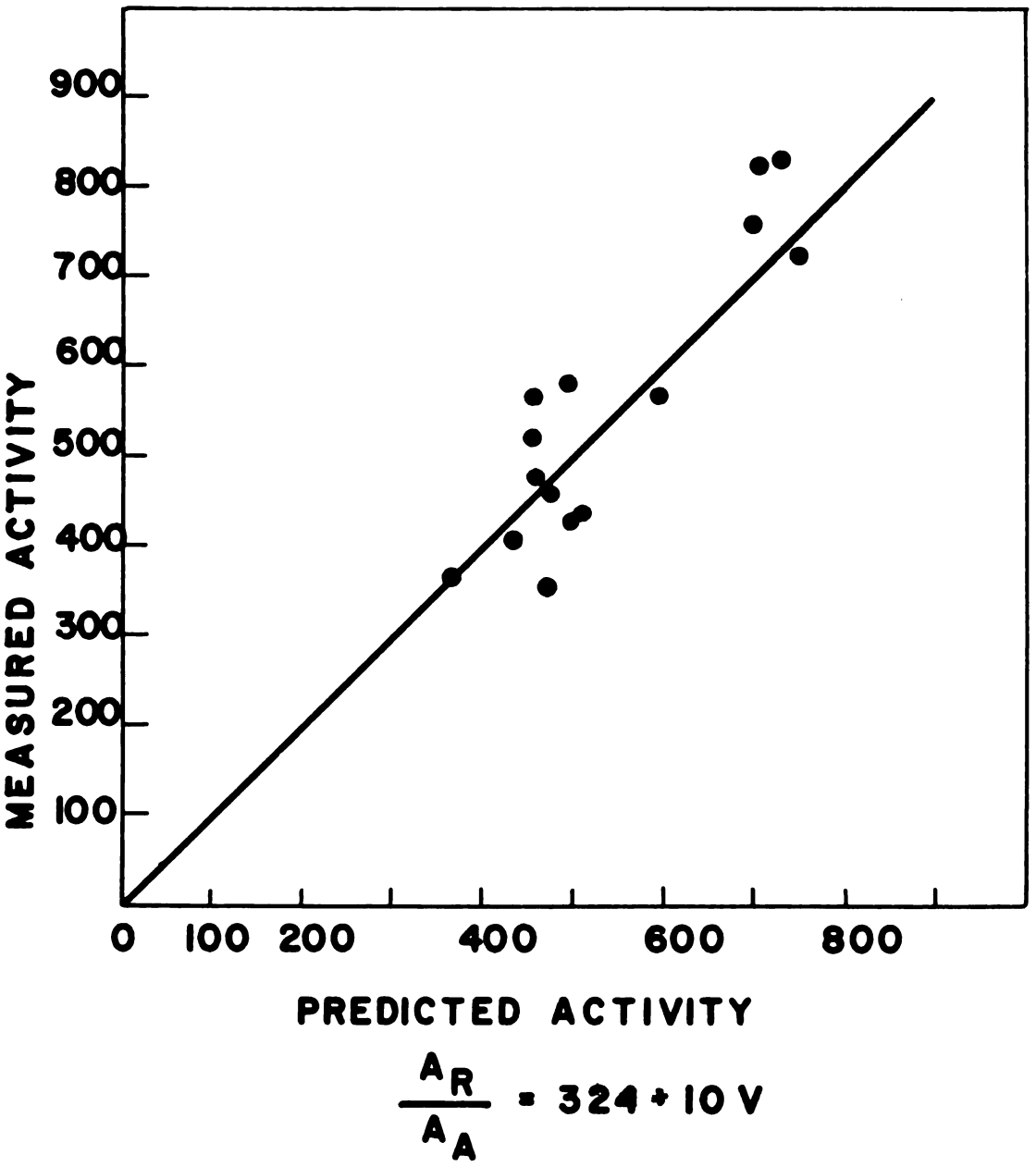

FIGURE -36

MEASURED VERSUS PREDICTED RAIN/AIR ACTIVITIES

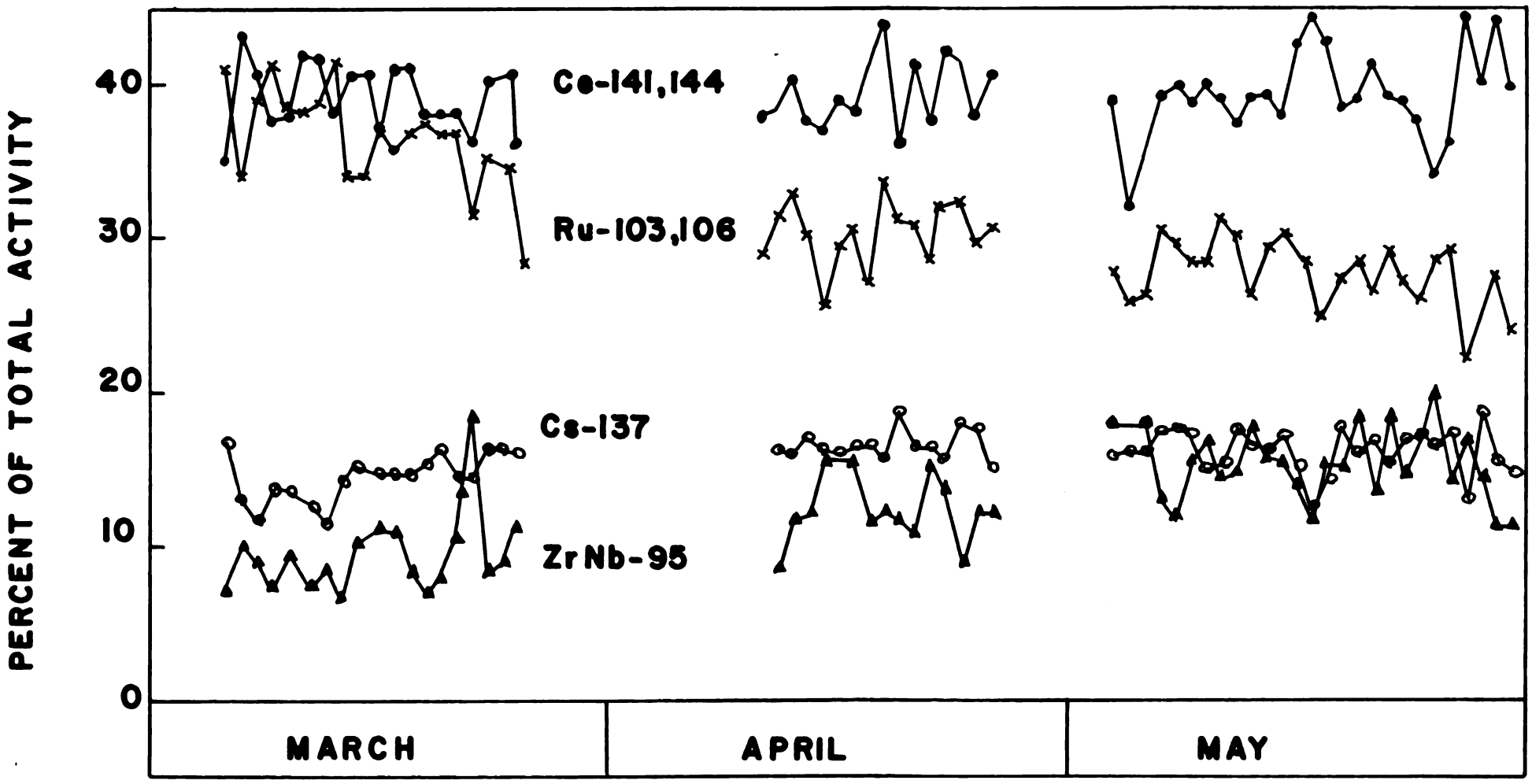

FIGURE - 37

DAILY ISOTOPE PERCENTAGES

	Wind Speed, mph	Cloud Height	Rain Depth, in	Air Activity uuc/M^3 Beta	$Ru^{103,106}$	$Ce^{141,144}$	Cs^{137}	Zr^{95}	Rain Activity uuc/l Beta	$Ru^{103,106}$	$Ce^{141,144}$	Cs^{137}	Zr^{95}
March 6	40	300	.14	8.6	1.88	1.63	0.77	0.33	3780	425	283	100	57
11	25	900	.14	8.6	1.09	1.23	0.46	0.26	4490	432	591	128	60
19	38	400	.51	5.5	1.43	1.60	0.59	0.27	2280	755	753	361	185
20	43	1600	.03	7.0	1.85	1.88	0.89	0.40	1500	720	830	294	142
29	47	2000	.26	8.1									
April 17	16.1	4000	.52	13.2	2.96	4.02	1.14	1.60	4330	1190	1448	647	668
19	13.4	8000	.26	12.2	3.52	5.95	2.05	1.61	3750	963	1538	640	712
22	13.7	2000	.27	6.9	2.36	3.48	1.29	1.12	3780	1050	1033	672	762
23	16.9	900	.15	11.1	0.67	0.87	0.38	0.18	1800	274	362	179	89
May 4	14.2	1500	.18	15.0	2.33	2.91	1.47	2.34	10500	762	1020	304	268
9	8.9	4000	.25	6.1	2.48	3.50	1.28	1.46	1800	1810	5130	440	545
10	18.7	300	.68	6.4	1.37	1.70	0.67	0.63	3600	311	450	196	274
13	14.7	5000	.11	14.3	1.92	2.69	1.10	1.11	1710	555	1048	350	449
17	10.2	1100	.45	6.4	1.21	1.87	0.62	0.66	4240	326	430	213	262
21	19.0	1200	.05	8.8	1.44	2.06	0.97	0.81		786	1960	552	294
28	13.8	300	.04	5.8	1.29	1.85	0.73	0.75		584	484	132	185
June 11	4.1	500	.16	8.1	1.12	1.73	0.63	0.52		392	640	193	210

FIGURE 38

LIST OF AIR AND RAIN DATA

APPENDIX D

EQUIPMENT USED

Sample Preparations

1. Type AA, 47 mm diameter Millipore filters and holder
2. TFA #2133 "High Volume" felt filters
3. Rotometer, 30 liters per minute capacity
4. Manometer
5. Building Vacuum system
6. H-4 "high Volume" air sampler
7. Rain Collection Basin
8. Rain evaporation pans
9. 250-watt infra-red lamps

Counting

1. Nuclear Chicago single channel analyzer model #1810
2. Nuclear Chicago Model 186 scaler, C-110B automatic sample changer and C-111B printing timer.
3. Gas flow proportional counter with preamplifier and micromil window, N. C. model #180
4. Sample mounts, copper sample pans, and one ounce bottles of glass