

~~CONFIDENTIAL~~

AEC RESEARCH AND DEVELOPMENT REPORT

6.1.16
HW-54913
COPY No. 155 A

0053629

**MEASUREMENT OF I¹³¹ AND OTHER FISSION
PRODUCTS ON VEGETATION BY
GAMMA-RAY SPECTROMETRIC ANALYSIS**

R. W. PERKINS

FEBRUARY 10, 1958

Official Classification of this report is
UNCLASSIFIED
with the exception of information that
may have been derived from all-source
intelligence gathering activities of
the United States Intelligence Community
Date 8/3/59

HANFORD LABORATORIES

HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND, WASHINGTON

GENERAL  ELECTRIC

RESTRICTED DATA

THIS DOCUMENT CONTAINS RESTRICTED DATA AS DEFINED
IN THE ATOMIC ENERGY ACT OF 1954. THE INFORMATION
RELATES TO THE CIVILIAN APPLICATIONS OF ATOMIC ENER-
GY. ITS TRANSMITTAL OR THE DISCLOSURE OF ITS CON-
TENTS IN ANY MANNER TO AN UNAUTHORIZED PERSON IS
PROHIBITED.

~~UNCLASSIFIED~~
~~CONFIDENTIAL~~

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, express or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission to the extent that such employee or contractor prepares, handles or distributes, or provides access to, any information pursuant to his employment or contract with the Commission.

~~CONFIDENTIAL~~

HW-54913

~~This document classified by~~
J. M. Nielsen

C-4, Chemistry-General
(M-3679, 20th Ed., Rev. 1)

This document consists
of 56 pages. Copy No. 155
of 170 copies. Series A

MEASUREMENT OF I¹³¹ AND OTHER FISSION PRODUCTS
ON VEGETATION BY GAMMA-RAY SPECTROMETRIC ANALYSIS

By

R. W. Perkins
Chemical Research
Chemical Research and Development Operation

February 10, 1958

HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND, WASHINGTON

Work performed under Contract No. W-31-109-Eng-52 between
the Atomic Energy Commission and General Electric Company

~~RESTRICTED DATA~~

This document contains Restricted Data as defined in the Atomic
Energy Act of 1954. Its transmittal or the disclosure of its con-
tents in any manner to an unauthorized person is prohibited.

Route To:	P. R. No.	Location	Route Date	Signature and Date

Printed in USA. Charge 95 cents. Available from the U. S. Atomic
Energy Commission, Technical Information Service, P. O. Box 1001,
Oak Ridge, Tennessee. Please direct to the same address inquiries
covering the procurement of other classified AEC reports

~~CONFIDENTIAL~~

TABLE OF CONTENTS

	<u>Page</u>
INTRODUCTION	3
SUMMARY AND CONCLUSIONS	4
THEORETICAL	4
INSTRUMENTATION.	11
EXPERIMENTAL.	14
Calibrations	15
Measurement of Ce ¹⁴¹⁻¹⁴⁴ and Ru ¹⁰³⁻¹⁰⁶ Isotope Ratios	21
Disintegration Rate Measurements	23
Natural Radioactivity	28
Observed Concentrations and Estimated Detectable Concentra- tions of I ¹³¹ and the Fallout Radioisotopes	28
DISCUSSION	30
ANALYTICAL PROCEDURE	31
ACKNOWLEDGEMENTS.	32
REFERENCES	33
APPENDIX A - Gamma Ray Spectra of the Fallout Radioisotopes	34
APPENDIX B - Mutual Interference Corrections in a Four Component System	43
APPENDIX C - Sampling Zones and Isotopic Concentrations Found	47
ABSTRACT	53

MEASUREMENT OF I¹³¹ AND OTHER FISSION PRODUCTS
ON VEGETATION BY GAMMA-RAY SPECTROMETRIC ANALYSIS

INTRODUCTION

Measurements of I¹³¹ and the "total beta" radioactivity on vegetation samples have been used for a number of years to estimate the extent of contamination from accidental releases of radioisotopes in plant operations. The interpretation of these measurements has become more difficult in recent years due to the increased background radioactivity from fallout fission products. A more sensitive method of measuring I¹³¹ was required, since this isotope is the one most frequently released by faulty plant operation, and has a relatively low permissible concentration on vegetation. The method previously used for the I¹³¹ determination required a chemical separation which involved leaching of the I¹³¹ from a vegetation sample with a hot, dilute HNO₃ solution followed by a carrier precipitation of I¹³¹ as silver iodide.⁽¹⁾ The I¹³¹ was then measured by beta counting. This procedure was found lacking because of its low and erratic yield, the occasional contamination of the I¹³¹ with other fallout radioisotopes and the relatively low sensitivity (about 6×10^{-6} $\mu\text{c/gm}$) which it provided. The "total beta" measurement (actually called a "non-volatile analysis" since only those isotopes which remained after ashing a vegetation sample were measured) was also inadequate since only a gross beta measurement was obtained, which provided no information as to the radioisotopic species being measured.

This paper describes a sensitive procedure based on gamma-ray spectrometric analysis for the quantitative measurement of I¹³¹ and several of the gamma-ray emitting fallout radioisotopes on vegetation samples.

SUMMARY AND CONCLUSIONS

A procedure based on gamma-ray spectrometric analysis has been developed for the determination of I^{131} and several of the long-lived fallout radioisotopes on vegetation. Quantitative analyses of I^{131} , $Ce^{141-144}$, $Ru^{103-106}$, Zr^{95} - Nb^{95} , and Ba^{140} - La^{140} are obtained from the measurement.

It was possible to measure I^{131} (in the presence of existing levels of fallout radioisotopes) with a sensitivity of about 1×10^{-6} $\mu\text{C/gm}$ on a 150 gram sample by making a 10-minute measurement with a multichannel analyzer, 3 in. x 3 in. NaI(Tl) scintillation spectrometer. The procedure was somewhat less sensitive for the other radioisotopes. Experiments with larger samples and crystals indicated that the I^{131} sensitivity could be improved by a factor of two to three by doubling the sample size and using a larger, 3 in. x 5 in. diameter NaI(Tl) crystal. This latter sensitivity represents a 10-15 fold increase over that obtained with the former chemical separations technique. ⁽¹⁾

The concentration of the various radioisotopes in a vegetation sample are calculated from the "photopeak counting rates" in the gamma-ray spectrum, after making the necessary Compton corrections.

The determination requires a ten minute counting measurement plus 10 to 15 minutes for disintegration rate calculations. For these short counting intervals the reproducibility appears to be about what one would predict from counting statistics.

THEORETICAL

Gamma rays resulting from the decay of radioisotopes are emitted with discrete energy values and undergo only slight attenuation in materials with low atomic numbers. These properties have permitted quantitative gamma-ray spectrometric analysis of certain mixtures of radioisotopes in samples of tissue and bone from experimental animals, in various food materials and also in humans. ^(2, 3, 4) By similar methods of analysis it is

possible to quantitatively measure the radioisotopes, resulting from fallout or other sources, on samples of vegetation. The problem of measuring radioactive fallout is simplified by the fact that the mixture of the fallout radioisotopes is generally restricted to rather definite compositions.

The instrumentation requirements and the principles involved in the gamma-ray spectrometric analysis of mixed gamma-ray emitters have recently been summarized. ⁽⁵⁾ The equipment used in this study is described in detail in the instrumentation section. The basic instrument used was an RIDL 100-channel analyzer equipped with a 3 in. x 3 in. solid NaI(Tl) crystal detector. A coincidence spectrometer employing two 3 in. thick x 5 in. diameter crystals was used for the occasional measurements of the Ru¹⁰³ to Ru¹⁰⁶ ratio.

The gamma-ray spectrum of "fresh fallout" radioisotopes, present on vegetation three to four days after fission (see Figure 1), 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 from a vegetation sample (see Figure 2). After a three to four week decay period, the short-lived radioisotopes have decayed to insignificant concentrations and the gamma-ray spectrum of a vegetation sample possesses five characteristic photopeaks (see Figure 3) due to Ce¹⁴¹-Ce¹⁴⁴, I¹³¹, Ru¹⁰³-Ru¹⁰⁶, Zr⁹⁵-Nb⁹⁵, and Ba¹⁴⁰-La¹⁴⁰. From this time until the isotopes have decayed below detection, it is possible to measure them with reasonable accuracy from the gamma-ray spectrum of the sample. The concentrations of I¹³¹ and Ba¹⁴⁰-La¹⁴⁰ on "fast growing" vegetation decrease more rapidly than would be predicted from their half-lives and their characteristic γ -ray energies are not usually present in the gamma-ray spectrum of vegetation samples collected two months after fallout. However, the concentrations of the isotopes in the

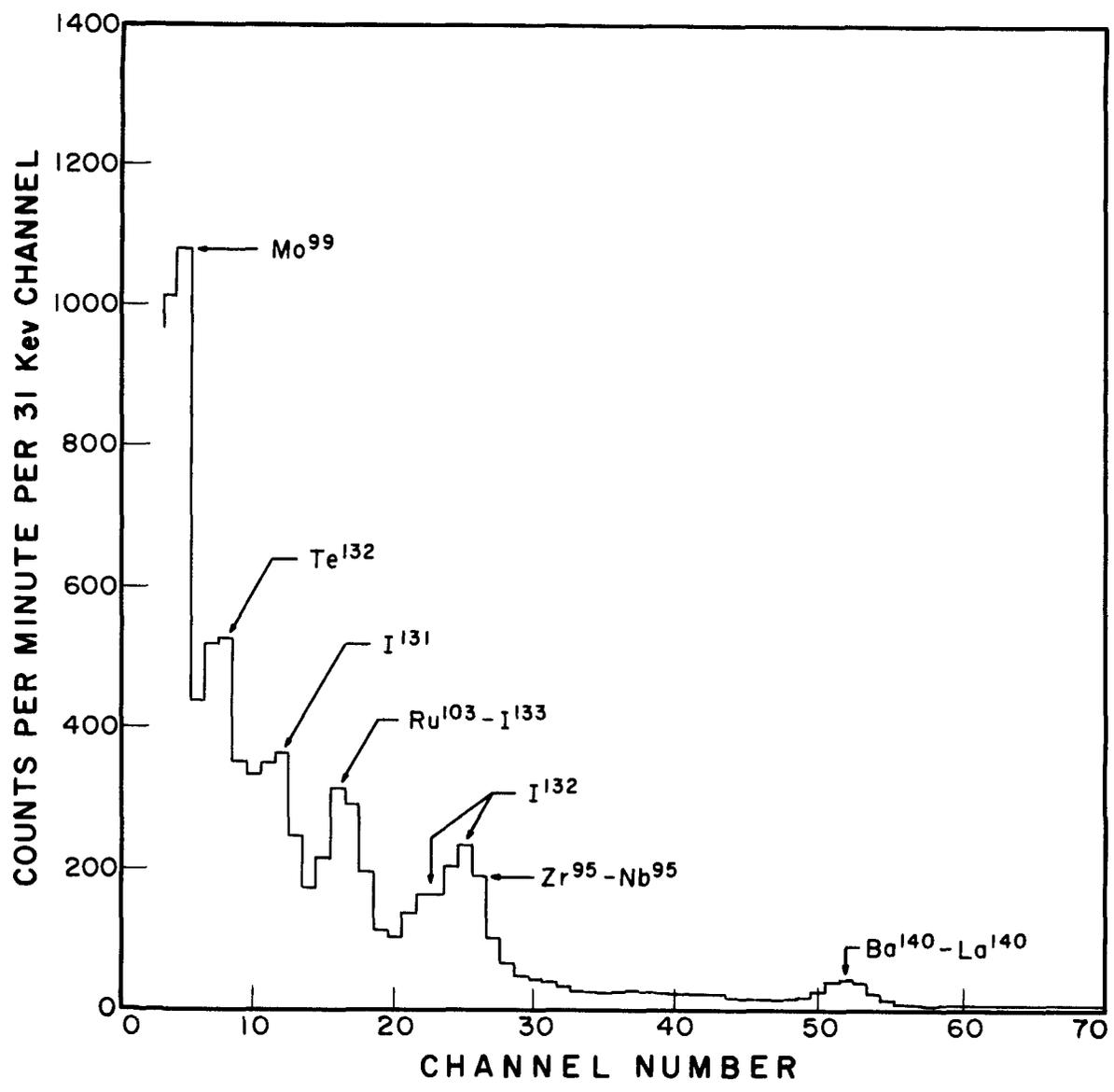


FIGURE - 1
GAMMA-RAY SPECTRUM OF A VEGETATION SAMPLE
CONTAINING 3-4 DAY-OLD FALLOUT.

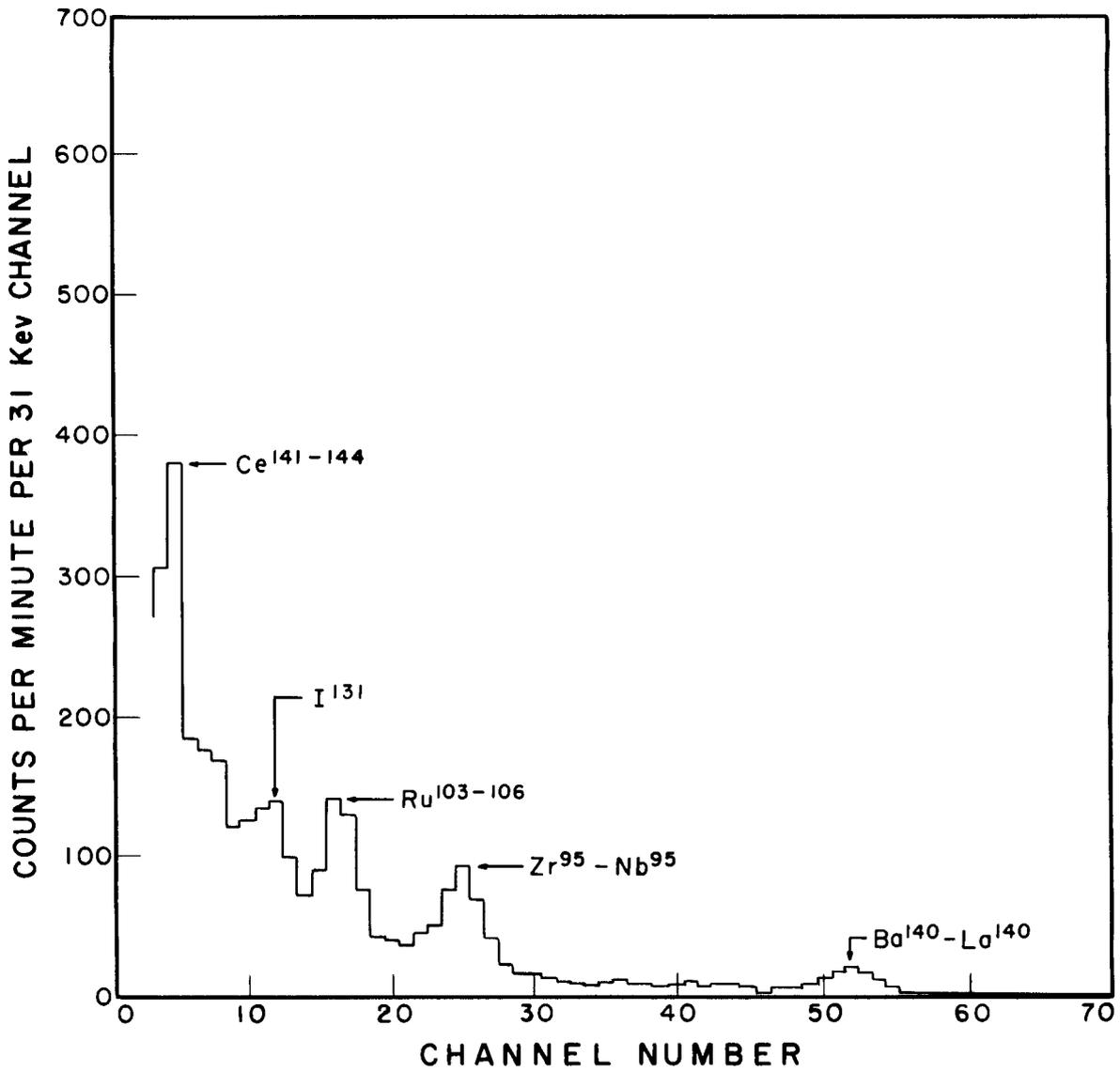


FIGURE -2
GAMMA-RAY SPECTRUM OF A VEGETATION SAMPLE
CONTAINING WEEK-OLD FALLOUT.

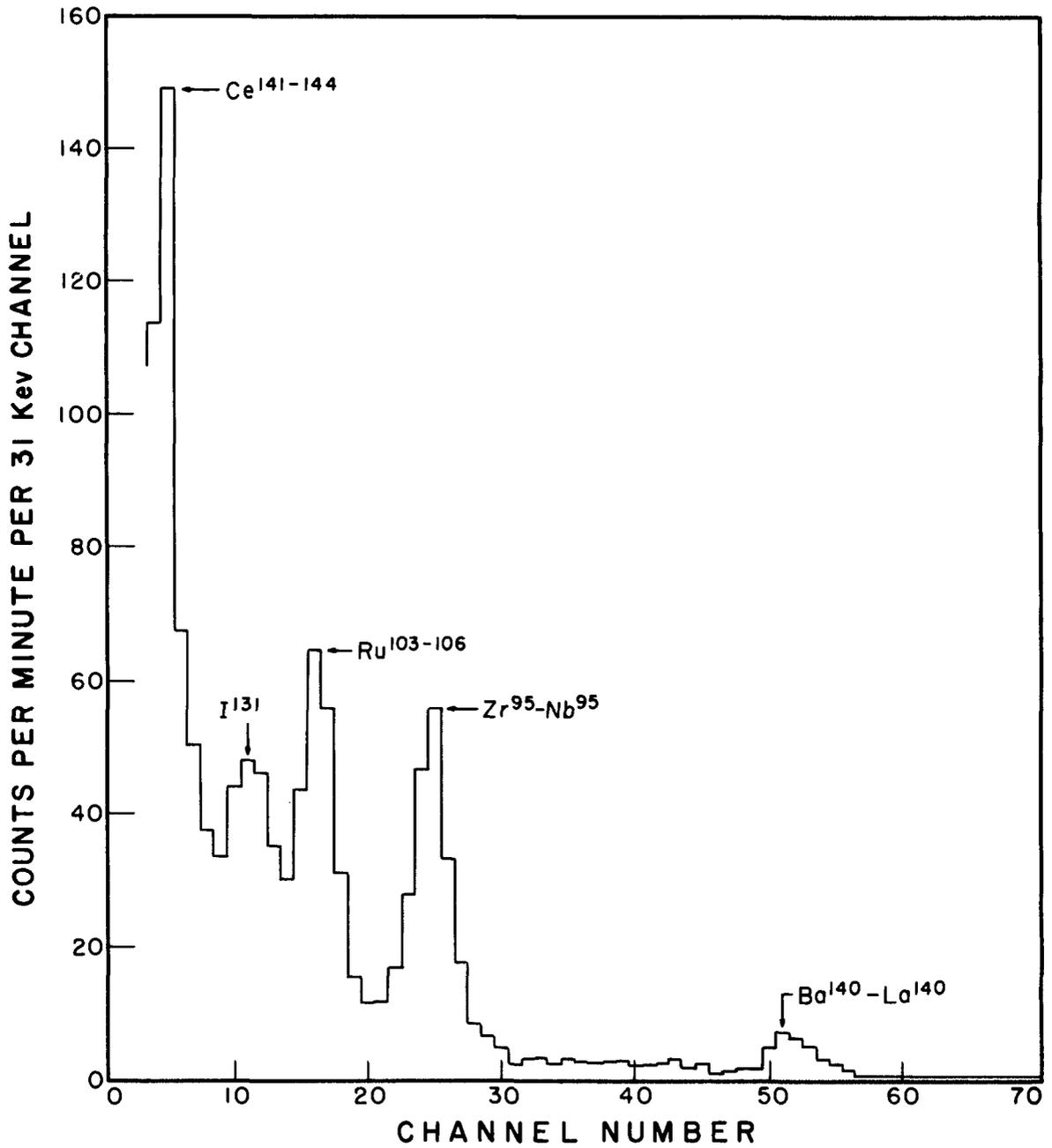


FIGURE - 3
GAMMA-RAY SPECTRUM OF VEGETATION SAMPLED
3 - 4 WEEKS AFTER FRESH FALLOUT

spectra observed here are in fair agreement with the isotope concentrations calculated⁽⁶⁾ for various times after fission.

In addition to occasional large amounts of "fresh fallout" from tests 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 radioisotopes, Ce¹⁴¹⁻¹⁴⁴, Ru¹⁰³⁻¹⁰⁶, and Zr⁹⁵-Nb⁹⁵, and does not usually contain significant amounts of Ba¹⁴⁰-La¹⁴⁰ or I¹³¹.

The general method employed for measuring the concentrations of the various radioisotopes is graphically illustrated in Figure 4. The counting rate at the Ba¹⁴⁰-La¹⁴⁰ photopeak is directly proportional to the amount of Ba¹⁴⁰-La¹⁴⁰ present. The "net counting rate" at the Zr⁹⁵-Nb⁹⁵ photopeak requires a correction for the contribution from Ba¹⁴⁰-La¹⁴⁰ and may also require a small correction for some of the other isotopes. Similar corrections are required for the counting rate measurements of Ru¹⁰³⁻¹⁰⁶, I¹³¹, and Ce¹⁴¹⁻¹⁴⁴ at their photopeaks. After these "Compton corrections" have been made, the "net counting rates" of the characteristic photopeaks are proportional to the amount of the radioisotope present.

In order to obtain a maximum counting efficiency for I¹³¹, with a minimum counting rate from the natural radioactive background plus the Compton from higher energy gamma-emitters in the sample, a rather narrow energy increment which contained only the center portion of the I¹³¹ photopeak was used. This energy increment and the energy increments used for measuring the other fallout radioisotopes are indicated in Figure 4.

A discussion of the specific conditions for making the γ -ray spectrometric measurements and the methods used for calibrating the equipment and calculating the disintegration rates are included in the experimental section.

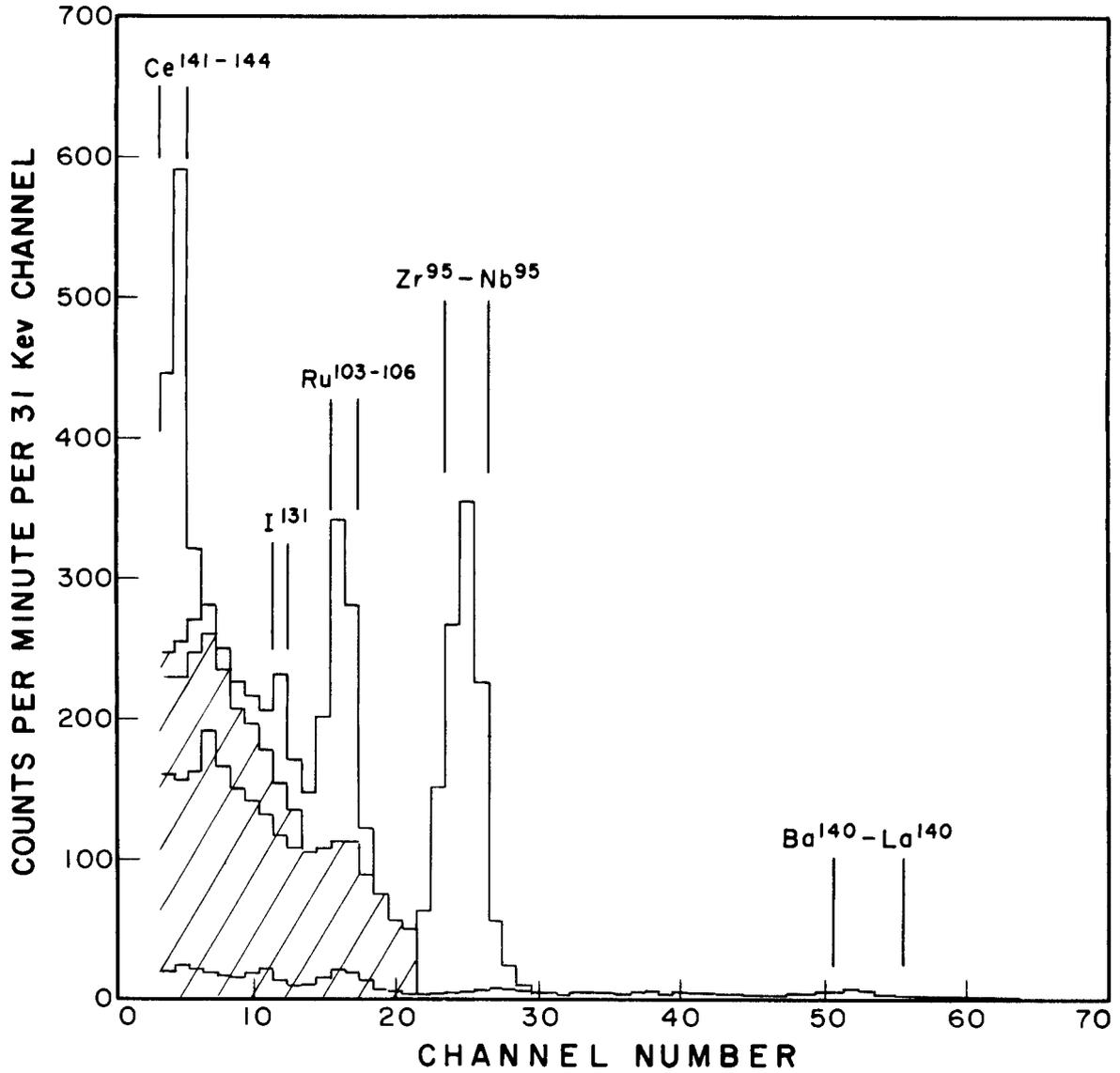


FIGURE - 4
GAMMA-RAY SPECTRUM OF THE FALLOUT
RADIOISOTOPES SHOWING THE ENERGY
INCREMENT AND COMPTON CORRECTION
FOR THE MEASUREMENT OF EACH ISOTOPE.

INSTRUMENTATION

An RIDL 100-channel analyzer equipped with a NaI(Tl) detector was used for the gamma-ray spectrometric measurements. A photograph of the analyzer, detector, and associated lead shield is shown in Figure 5. It will be noted that an RIDL pulse generator is also included in Figure 5. This instrument, although not used in the spectrometric measurements, was required for making sensitive zero adjustments and periodic linearity measurements on the analyzer. The general operations of the RIDL 100-channel analyzer is included in the instruction manual. ⁽⁷⁾

The detector-sample arrangement is also shown in Figure 5. The detector is a 3 in. x 3 in. NaI(Tl) crystal (Harshaw Chemical Company) mounted on a 6363 DuMont multiplier phototube. The sample container, a 9-ounce glass jar, is centered directly on top of the crystal.

The "zero setting" of the analyzer is made by adjusting its energy response to permit zero energy to coincide with the lower edge of channel one. This is achieved by feeding 90-volt pulses and then 10-volt pulses from the pulse generator into the analyzer and adjusting the "lower level setting" of the analyzer so these pulses fall at the lower edge of channels 90 and 10, respectively.

A high voltage of about 900 volts was used on the multiplier phototube and was adjusted to center the 0.356 Mev I^{131} photopeak in channel 12. This provides a channel width of about 0.031 Mev.

For making the Ru¹⁰⁶ measurement, a coincidence spectrometer employing two large NaI(Tl) detectors was used. The theory and operation of a coincidence spectrometer has been described previously. ⁽⁸⁾ The detectors and sample arrangement are illustrated in Figure 6. The detectors, two 3 in. x 5 in. diameter NaI(Tl) crystals mounted on 6364 DuMont multiplier phototubes, view the sample which is placed directly between them.

~~CONFIDENTIAL~~

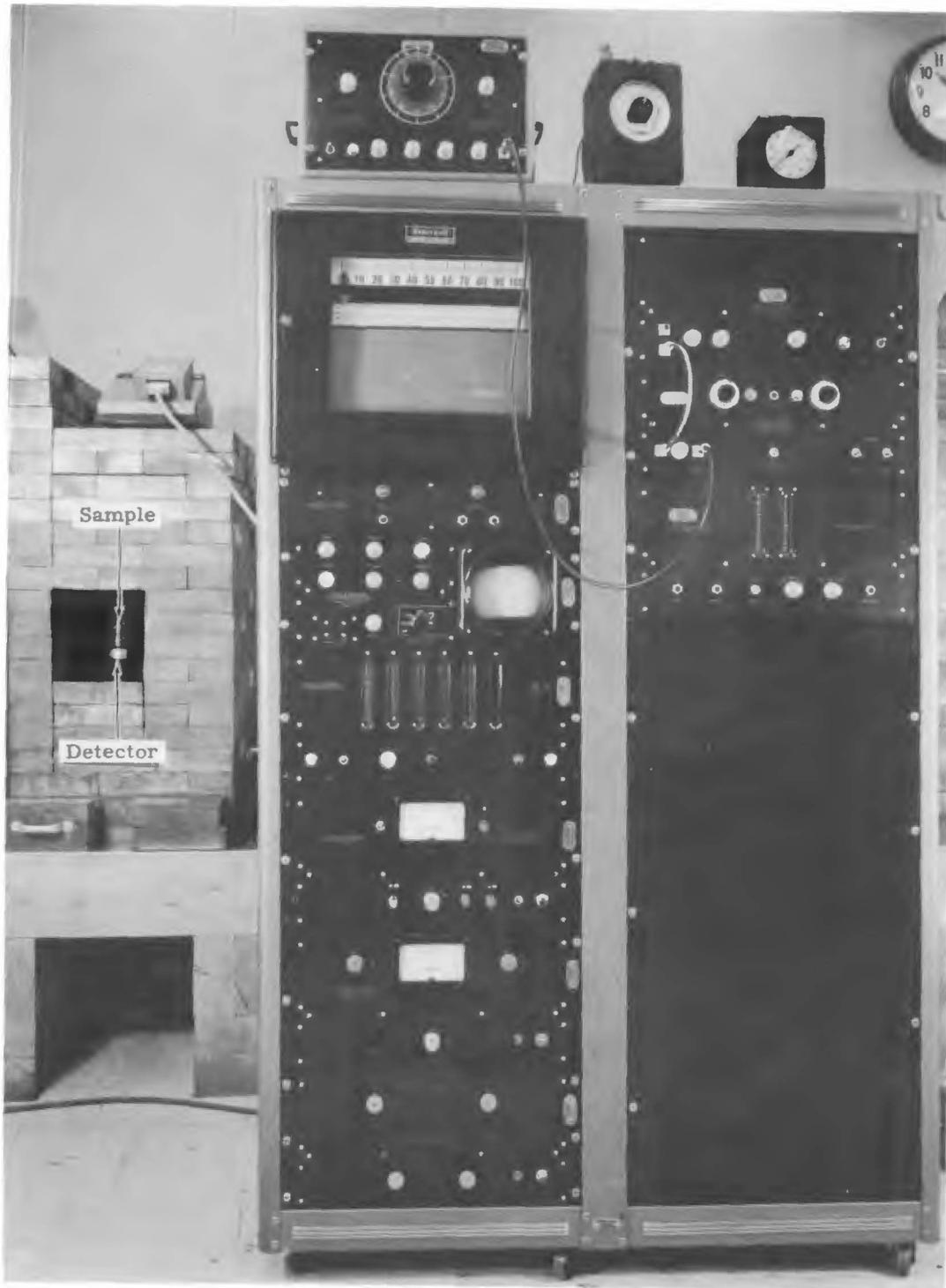


FIGURE 5
Spectrometer System Showing
Location of Detector and Sample in Lead Shield

~~CONFIDENTIAL~~

CONFIDENTIAL



FIGURE 6

Detector - Sample Arrangement
of the Coincidence Spectrometer

0
1
2
3
4
5
6
7
8
9
A
B
C
D
E
F
G
H
I
J
K
L
M
N
O
P
Q
R
S
T
U
V
W
X
Y
Z

The Ru¹⁰⁶ measurements were made using pulse-height analyzer window widths of three volts (about 0.090 Mev) with multiplier phototube voltages of about 900 volts, and amplifier gains sufficient to center the characteristic 0.513 and 0.624 Mev photopeaks of Rh¹⁰⁶ in these windows at pulse heights of 16.9 and 20.5 volts, respectively. The resolving time of the coincidence analyzer was 0.25 μ sec. The coincidence background under these conditions was 0.14 c/m. The resolutions of the two 3 in. x 5 in. diameter NaI(Tl) multiplier phototube detectors were 12 to 13 per cent at the Cs¹³⁷ photopeak. The counting efficiency (counting rate/absolute disintegration rate) for Ru¹⁰⁶ under these conditions was about 0.025 per cent.

EXPERIMENTAL

A 3 in. diameter by 3 in. thick NaI(Tl) crystal was chosen for this study since it provided good resolution, reasonable sensitivity, and was available in conjunction with a multichannel analyzer. A nine ounce bottle (2.9 in. diameter by 3.5 in. high) was used as a sample container. A container of this size will hold between 150 and 200 grams of sage brush greens.

A brief study with larger crystals and samples indicated that a higher sensitivity than that available with a 3 in. x 3 in. crystal and a nine ounce container was possible. The counting rate at the I¹³¹ photopeak was increased by a factor of about 1.7 by doubling the sample size and using a 16-ounce bottle as a container. The use of a larger crystal, 3 in. x 5 in. diameter, and the nine ounce container increased the counting rate by a factor of 2.3. The use of both the larger sample and crystal provide a four-fold increase in the counting rate at the I¹³¹ photopeak. These I¹³¹ photopeak counting efficiencies are comparable with the 3 in. x 3 in. crystal, nine ounce container combination in Table I.

TABLE I
RELATIVE COUNTING RATES AT THE I¹³¹ PHOTOPeAK
FOR VARIOUS CRYSTAL AND SAMPLE SIZES

<u>Sample Size</u>	<u>Detector Size</u>	<u>Relative Counting Rate</u>
9 ounce bottle (1 unit I ¹³¹)	3" x 3"	1
16 ounce bottle (2 units I ¹³¹)	3" x 3"	1.7
9 ounce bottle (1 unit I ¹³¹)	3" x 5" diameter	2.3
16 ounce bottle (2 units I ¹³¹)	3" x 5" diameter	4.0

The gamma-ray spectra obtained for these combinations are compared in Figure 7.

Although the large crystal does provide a higher counting efficiency it has the disadvantage of low resolution (about 12 per cent at the Cs¹³⁷ photopeak compared with 9.5 per cent for the three inch crystal) and about twice the background counting rate. The background spectra for the 3 in. x 3 in. and 3 in. x 5 in. crystals are compared in Figure 8.

In spite of the high background and low resolution obtained with a large crystal, it is apparent that a greater sensitivity, probably a factor of two to three, could be obtained if the larger crystal and samples were used.

CALIBRATIONS

The gamma-ray spectrometer was calibrated for the determination of fallout radioisotopes by measuring the counting efficiency of standardized aqueous solutions of the radiochemically pure isotopes in nine ounce bottles. It was felt that this method of calibration was better than trying to uniformly distribute a radioisotope over 150 grams of vegetation in a nine ounce bottle. Both water and vegetation are composed of low atomic number material;

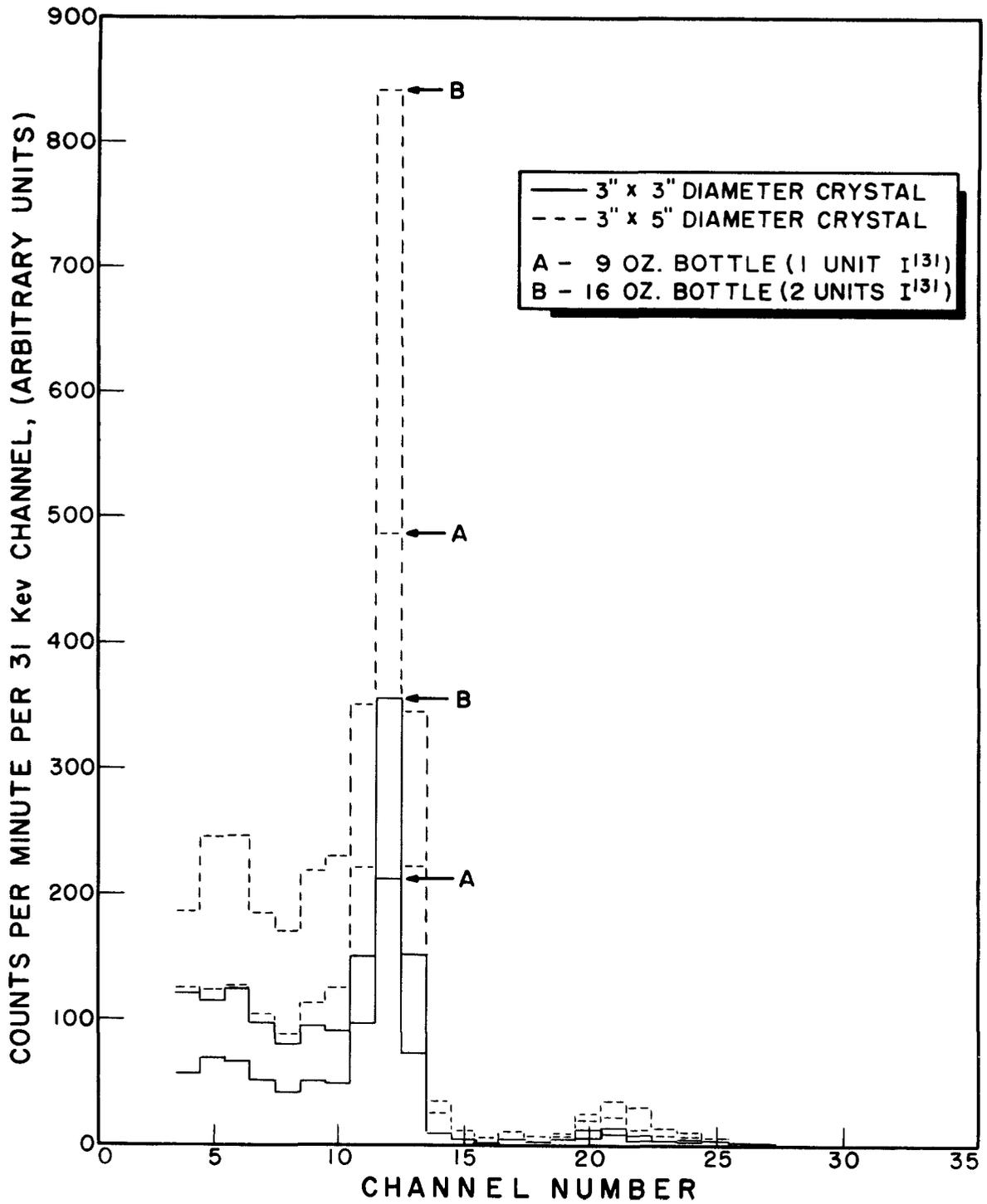


FIGURE -7
GAMMA -RAY SPECTRA OF DIFFERENT
SAMPLE AND CRYSTAL COMBINATIONS.

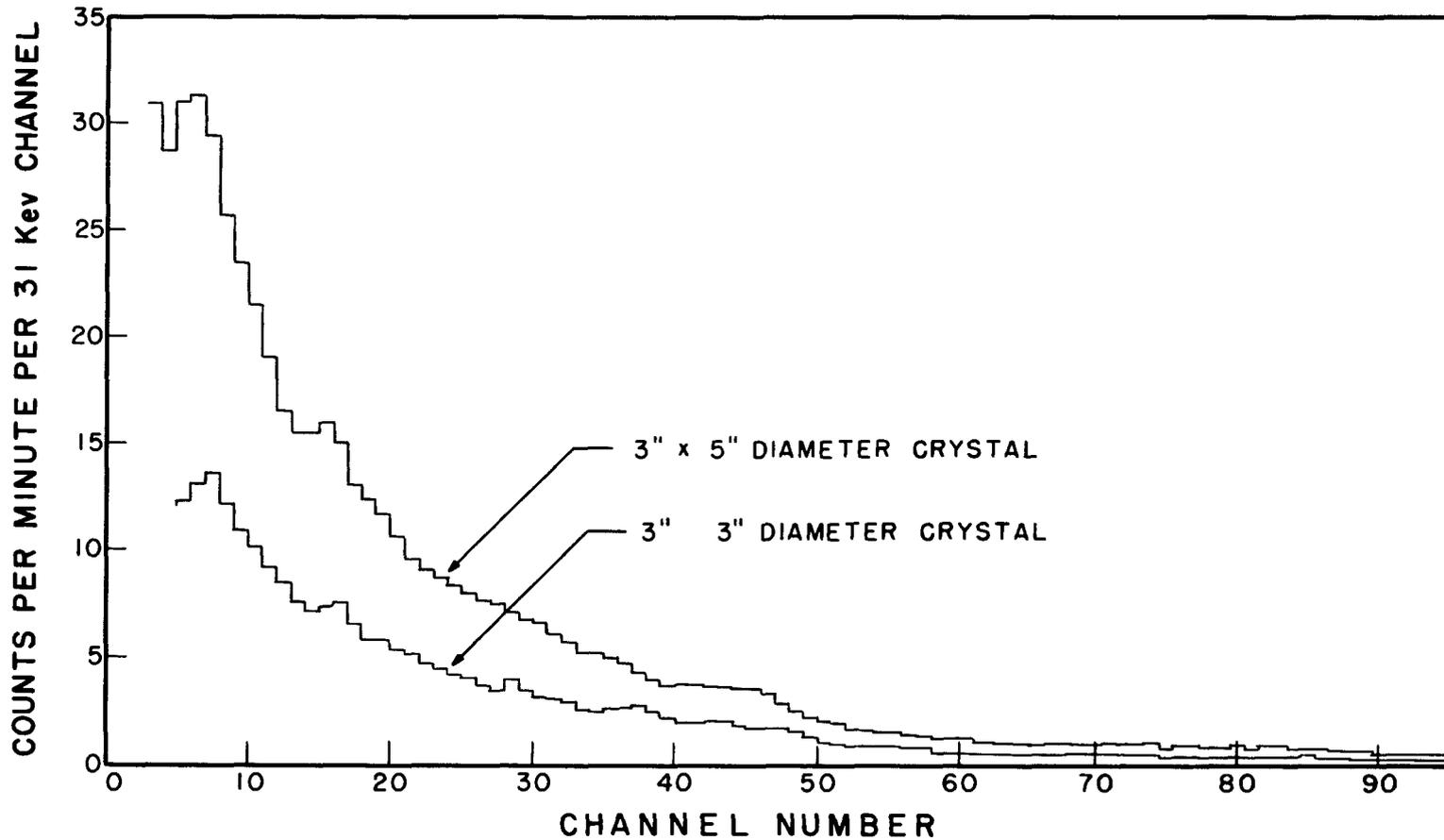


FIGURE - 8
COMPARISON OF BACKGROUND COUNTING
RATES NaI(Tl) CRYSTALS.

DECLASSIFIED

therefore, their attenuation of gamma-rays would be expected to be very small and about equal. This assumption was checked by measuring the gamma-ray spectrum of a 125 gram sample of vegetation in a nine ounce bottle, then adding 75 ml of water and re-measuring the spectrum. The two spectra agreed at every point within experimental error.

The gamma-ray spectra of radiochemically pure samples of isotopes found in fallout, measured as aqueous solutions in nine ounce bottles, are included in Appendix A. The area of the photopeaks used for the measurements of these isotopes is indicated.

The standardized radioisotopes used for these calibrations were calibrated by beta counting.

The gamma-ray counting efficiencies of these radioisotopes at their photopeaks are summarized in Table II. Also included in Table II are the fractions of the photopeak counting rates in other channel intervals where "Compton corrections" may be needed. The "photopeak counting efficiency factors" listed in Table II are equal to the absolute disintegration rates divided by the observed counting rates. A disintegration rate is thus calculated from the observed photopeak counting rate by multiplying by this factor after any necessary Compton corrections are made.

A similar compilation summarizing the photopeak counting efficiencies and Compton correction factors for mixtures of Ce^{141} - Ce^{144} and Ru^{103} - Ru^{106} is given in Table III.

TABLE II

PHOTOPEAK COUNTING EFFICIENCIES AND COMPTON CORRECTION FACTORS

Isotope	Half-life, Days	Photopeak Energy Mev ⁽⁹⁾	Photopeak Channels	Photopeak Counting Efficiency Factor, D/M C/M	Compton Correction Factors, $\frac{\text{Compton C/M}}{\text{Photopeak C/M}}$			
					Channels 4 - 5	Channel 12	Channels 16 - 17	Channels 24 - 26
Ce ¹⁴¹	32	0.134	4 - 5	34.98				
Ce ¹⁴⁴ -Pr ¹⁴⁴	290	0.145	4 - 5	223.21*		0.0110	0.0124	0.0161
I ¹³¹	8.05	0.364	12	51.33	0.593		0.0215	0.0156
Ru ¹⁰³	40.0	0.498	16 - 17	36.59	0.369	0.0822		
Ru ¹⁰⁶	365	0.513	16 - 17	197.62	0.686	0.152		0.0525
Zr ⁹⁵ -Nb ⁹⁵	63.3	0.754 and 0.764	24 - 26	42.37*	0.321	0.121	0.215	
Ba ¹⁴⁰ -La ¹⁴⁰	12.8	1.60	51 - 55	202.43*	1.877	0.545	1.735	0.599
K ⁴⁰	4.7 x 10 ¹¹	1.46	45 - 50	904.97	0.663	0.189	0.230	0.205

* Multiplying by this factor gives the D/M of the equilibrium mixture.

REPRODUCED

TABLE III

PHOTOPEAK COUNTING EFFICIENCIES AND COMPTON CORRECTION FACTORS

FOR MIXTURES OF Ce¹⁴¹-Ce¹⁴⁴ AND FOR Ru¹⁰³-Ru¹⁰⁶

Ru ¹⁰³ : Ru ¹⁰⁶ Ratio	Photopeak Counting Efficiency Factor D/M C/M	Compton Correction Factor			Compton C/M Photopeak C/M Channels 24 - 26
		Channels	Channel	Channels	
		4 - 5	12	16 - 17	
3:1	46.3	0.388	0.0918		0.00305
2:1	50.6	0.395	0.0935		0.00445
1:1	62.1	0.419	0.0981		0.00820
1:2	80.4	0.455	0.105		0.0142
1:4	105.3	0.504	0.115		0.0223
1:10	141.3	0.574	0.130		0.0341
<u>Ce¹⁴¹: Ce-Pr¹⁴⁴</u>					
3:1	44.9		0.000690	0.000686	0.000891
2:1	50.1		0.000886	0.000998	0.00130
1:1	63.0		0.00164	0.00185	0.00240
1:2	83.7		0.00285	0.00322	0.00418
1:3	99.8		0.00340	0.00428	0.00555
1:4	112.5		0.00453	0.00511	0.00663
1:10	154.8		0.00701	0.00790	0.0103

MEASUREMENT OF Ce¹⁴¹⁻¹⁴⁴ AND Ru¹⁰³-Ru¹⁰⁶ ISOTOPE RATIOS

For gamma-ray spectrometric analysis of mixtures of Ba¹⁴⁰-La¹⁴⁰, Zr⁹⁵-Nb⁹⁵, Ru¹⁰³⁻¹⁰⁶, I¹³¹, and Ce¹⁴¹⁻¹⁴⁴ on vegetation, it is necessary to know the approximate ratios of Ru¹⁰³ to Ru¹⁰⁶ and of Ce¹⁴¹ to Ce¹⁴⁴-Pr¹⁴⁴. The gamma-ray energies responsible for the principal Ru¹⁰³ and Ru¹⁰⁶ photopeaks (0.498 and 0.513 Mev, respectively) and the Ce¹⁴¹ and Ce¹⁴⁴ photopeaks (0.145 and 0.134 Mev, respectively) are not resolved by the spectrometer and they appear in a spectrum as single peaks (see Figure 4); however, the spectra of Ru¹⁰³ and Ru¹⁰⁶ and of Ce¹⁴¹ and Ce¹⁴⁴ are somewhat different, and it is necessary to know the approximate ratios of these isotopes in the samples of interest if a high sensitivity for I¹³¹ is to be obtained. An occasional measurement of the ratios of Ce¹⁴¹ to Ce¹⁴⁴ and of Ru¹⁰³ to Ru¹⁰⁶ is sufficient since these ratios do not vary grossly in fallout over a given area, and a relatively large change in the ratios has only a small effect on the sensitivity of an I¹³¹ determination. The cerium isotopes are easily separated from vegetation samples by conventional radiochemical methods and the ratio of the Ce¹⁴¹:Ce¹⁴⁴ can be measured by beta counting techniques or by gamma-ray spectrometric analysis. The ruthenium radioisotopes are rather difficult to separate from large vegetation samples; however, it has been found possible to independently determine the Ru¹⁰⁶ in the nine ounce bottled samples by measuring the coincidence counting rate of the Ru¹⁰⁶ 0.513 and 0.624 Mev photon cascade.

The coincidence counting arrangement for the measurement of Ru¹⁰⁶ in a nine ounce bottle of vegetation with two 3 in. x 5 in. NaI(Tl) detectors is shown in Figure 6. The operation of the coincidence spectrometer and the required instrument setting are discussed in the instrumentation section. The overall coincidence counting efficiency of the instrument is measured by counting a standard aqueous sample of Ru¹⁰⁶ in a nine ounce bottle. The sample to be analyzed is then counted.

The coincidence counting efficiency for Ru^{106} is low, ~ 0.025 per cent. This is partly due to its decay scheme since only 11 per cent of the disintegrations include these coincidence photons⁽¹⁰⁾, but is also due to the obviously poor geometry afforded by such a large sample. The counting rate was high enough, however, to permit reasonably accurate Ru^{106} determination from an overnight count.

To determine the interference from the other fallout radioisotopes in the coincidence measurement of Ru^{106} , the coincidence counting rates of pure samples of each of these radioisotopes were measured. The coincidence counting efficiencies of these isotopes are compared with Ru^{106} in Table IV.

TABLE IV

COMPARISON OF THE COINCIDENCE COUNTING EFFICIENCIES
OF OTHER FALLOUT RADIOISOTOPES WITH Ru^{106}

<u>Isotope</u>	<u>Coincidence Counting Efficiency, $\times 10^4$</u>
Ru^{106}	2.48
Ba^{140} - La^{140}	0.716
Ru^{103}	<0.001
Zr^{95} - Nb^{95}	<0.001
I^{131}	<0.001
Ce^{141}	<0.001
Ce^{144}	<0.001

The data in Table IV indicate a negligible interference in the coincidence counting of Ru^{106} from all the isotopes except Ba^{140} - La^{140} . Since Ba^{140} - La^{140} is measured directly from the gamma-ray spectrum, a correction for its contribution can easily be made. This will only be necessary in relatively fresh fallout material.

The Ru¹⁰⁶ disintegration rate is calculated from the coincidence counting rate by the following equation.

$$D/M \text{ Ru}^{106} = \frac{D/M \text{ of Ru}^{106} \text{ standard} \times \text{"net coincidence C/M of sample"}}{\text{coincidence C/M standard Ru}^{106}} \quad (1)$$

$$\begin{aligned} \text{Net coincidence C/M of sample} &= \\ &= \text{observed C/M} - D/M \text{ Ba}^{140} \text{-La}^{140} \times (\text{Ba}^{140} \text{-La}^{140} \text{ coincidence counting efficiency}) \\ &= \text{observed C/M} - (D/M \text{ Ba}^{140} \text{-La}^{140} \times 7.16 \times 10^{-5}) \end{aligned}$$

DISINTEGRATION RATE MEASUREMENTS

The La¹⁴⁰ photopeak at 1.60 Mev is used as a measure of the Ba¹⁴⁰-La¹⁴⁰ content of the sample. The contribution from other fallout radioisotopes at this energy appears to be completely negligible; thus, the counting rate at 1.60 Mev peak is proportional to the Ba¹⁴⁰-La¹⁴⁰ content. The disintegration rate of Ba¹⁴⁰-La¹⁴⁰ is calculated by multiplying the observed counting rate in channels 51 to 55 by the Ba¹⁴⁰-La¹⁴⁰ counting efficiency factor (Table II).

$$D/M \text{ Ba}^{140} \text{-La}^{140} = 202.4 \times \text{C/M in channels 51 to 55.} \quad (2)$$

The method selected for calculating the Zr⁹⁵-Nb⁹⁵, Ru¹⁰³⁻¹⁰⁶, Ce¹⁴¹⁻¹⁴⁴, and I¹³¹ content of a sample may depend on the average age of the mixture being measured. For relatively fresh material the contribution to the gamma-ray spectrum of a sample from radoruthenium and cerium is almost entirely due to Ru¹⁰³ and Ce¹⁴¹. Since both Ru¹⁰³ and Ce¹⁴¹ have high efficiencies (see Table II) compared with Ru¹⁰⁶ and Ce¹⁴⁴, and since they do not emit gamma-rays of energies greater than 0.498 (and some 0.611) and 0.145 Mev, respectively, the radoruthenium and cerium offer almost no interference in the measurement of higher energy photopeaks. The I¹³¹ does offer a small interference at the Zr⁹⁵-Nb⁹⁵ and Ru¹⁰³⁻¹⁰⁶ photopeaks,

but this is negligible where the I^{131} photopeak is small compared with the Zr^{95} - Nb^{95} and $Ru^{103-106}$ peaks (see spectra, Appendix A). For such samples the observed counting rate measurement at each photopeak requires Compton corrections only for those radioisotopes which have higher energy photopeaks.

It will be shown later that even for samples that do contain considerable amounts of Ce^{144} and Ru^{106} , only a small error in the net counting rate measurements is produced by neglecting their contribution to higher energy photopeaks.

Where Ru^{106} and Ce^{144} are responsible for a major fraction of the ruthenium and cerium photopeaks, a more precise measure of the I^{131} content as well as the Zr^{95} - Nb^{95} , $Ru^{103-106}$, and $Ce^{141-144}$ content of the sample is obtained if an equation relating the mutual interference of these radioisotopes is used in calculating their disintegration rates.

The general problem of making interference corrections in a four component system is discussed in Appendix B. The equations for calculating the net counting rates of these isotopes involves the solution of four simultaneous equations with four unknowns. The general solutions of these equations are included in Appendix B. Although these equations are complex because of the large number of constants involved, they become very simple when the numerical values of these constants are substituted into the equations.

Substituting the "Compton correction value" (from Tables II and III) for Ru^{103} - Ru^{106} and Ce^{141} - Ce^{144} ratios of 2:1 and 1:3, respectively, (the ratios observed in this study) the equations for calculating net counting rates are:

$$\begin{aligned}
C/M \text{ Zr}^{95}\text{-Nb}^{95*} &= K \times C/M \text{ at Zr}^{95}\text{-Nb}^{95} \text{ photopeak - (contributions} \\
&\quad \text{from Ru}^{106} \text{ - I}^{131} \text{ - Ce}^{144}) \\
&= 1.004 \times C/M \text{ in channels 24 through 26 - (0.00113 C/M in} \\
&\quad \text{channels 16 and 17 + 0.01236 x C/M in channel 12 +} \\
&\quad 0.00552 \times C/M \text{ in channels 4 and 5)}
\end{aligned} \tag{3}$$

$$\begin{aligned}
C/M \text{ Ru}^{103-106*} &= 1.004 \times C/M \text{ in channels 16 and 17 - (0.2125 x} \\
&\quad C/M \text{ in channels 24 through 26 + 0.0165 x C/M in channel 12 +} \\
&\quad 0.00306 \times C/M \text{ in channels 4 and 5)}
\end{aligned} \tag{4}$$

$$\begin{aligned}
C/M \text{ I}^{131*} &= 1.004 \times C/M \text{ in channel 12 - (0.1012 x C/M in channels} \\
&\quad 24 \text{ through 26 + 0.0926 x C/M in channels 16 and 17 +} \\
&\quad 0.00246 \times C/M \text{ in channels 4 and 5)}
\end{aligned} \tag{5}$$

$$\begin{aligned}
C/M \text{ Ce}^{141-144*} &= 1.004 \times C/M \text{ in channels 4 and 5 - (0.1784 x C/M} \\
&\quad \text{in channels 24 through 26 + 0.3413 x C/M in channels 16 and} \\
&\quad 17 + 0.5851 \times C/M \text{ in channel 12)}
\end{aligned} \tag{6}$$

The disintegration rates of the above radioisotopes are calculated from their "net counting rates" by multiplying by their respective counting efficiency factors (Tables II and III).

For samples which contain relatively large amounts of Ru¹⁰³ and Ce¹⁴¹ compared with the Ru¹⁰⁶ and Ce¹⁴⁴, the contribution to higher energy peaks by radiocerium and ruthenium becomes negligible, and the equations assume the simplified forms which follow. The correction for any contribution from Ba¹⁴⁰-La¹⁴⁰ is included in these equations.

* If Ba¹⁴⁰-La¹⁴⁰ were present, a correction for its interference would be made before applying these equations (see equations 7 through 10).

$$C/M \text{ Zr}^{95}\text{-Nb}^{95} = C/M \text{ in channels 24 through 26} - (0.599 \times C/M \text{ in channels 51 through 55}) \quad (7)$$

$$C/M \text{ Ru}^{103-106} = C/M \text{ in channels 16 and 17} - (1.735 \times C/M \text{ in channels 51 through 55} + 0.215 \times C/M \text{ in channels 24 through 26}) \quad (8)$$

$$C/M \text{ I}^{131} = C/M \text{ in channel 12} - (0.545 \times C/M \text{ in channels 51 through 55} + 0.121 \times C/M \text{ in channels 24 through 26} + 0.0935 \times C/M \text{ in channels 16 and 17}) \quad (9)$$

$$C/M \text{ Ce}^{141-144} = C/M \text{ in channels 4 and 5} - (1.877 \times C/M \text{ in channels 51 through 55} + 0.321 \times C/M \text{ in channels 24 through 26} + 0.395 \times C/M \text{ in channels 16 and 17} + 0.593 \times C/M \text{ in channel 12}) \quad (10)$$

The constants in these equations are taken from Tables II and III, for the $\text{Ru}^{103-106}$ and $\text{Ce}^{141-144}$ ratios of 2:1 and 1:3, respectively.

Equations 3 through 6 involve no approximations and give the ultimate sensitivity in calculating disintegration rates from the gamma-ray spectrum of mixtures of these isotopes. These equations are obtained for other $\text{Ru}^{103-106}$ and $\text{Ce}^{141-144}$ mixture by substituting the proper Compton correction values from Table III into equation A-D of Appendix B.

The use of these precise equations does permit a more accurate measurement of the fallout isotopes and I^{131} ; however, the increased accuracy is not significant for most samples. The concentrations of $\text{Zr}^{95}\text{-Nb}^{95}$, $\text{Ru}^{103-106}$, I^{131} , and $\text{Ce}^{141-144}$ were calculated for a set of five typical samples by use of the precise equations 3 through 6, and also by use of the approximate equations 7 through 10. The ratios of Ru^{103} to Ru^{106} and Ce^{141} to Ce^{144} were about 2:1 and 1:3, respectively. The values obtained are summarized in Table V.

TABLE VCOMPARISON OF THE CONCENTRATIONS OF FALLOUT ISOTOPES
CALCULATED BY THE PRECISE AND APPROXIMATE EQUATIONS

($\mu\text{c}/\text{gram} \times 10^6$)

<u>Calculation Method</u>	<u>Zr⁹⁵-Nb⁹⁵</u>	<u>Ru¹⁰³⁻¹⁰⁶</u>	<u>I¹³¹</u>	<u>Ce¹⁴¹⁻¹⁴⁴</u>
Precise	22.2	7.43	4.01	50.1
Approximate	22.0	7.29	3.95	50.4
Precise	47.9	11.9	7.00	73.2
Approximate	47.7	12.1	6.61	75.8
Precise	76.0	29.9	6.74	149
Approximate	75.4	29.6	6.57	149
Precise	20.9	6.32	1.60	46.7
Approximate	20.7	6.23	1.54	46.9
Precise	20.7	6.38	1.12	40.7
Approximate	20.6	6.32	1.07	40.9

Table V shows that a very small error is produced in the measurement of these isotopes by use of the approximate equations. The errors in the I¹³¹ measurements appear relatively large, but these errors are small compared with the detection limit of $1 \times 10^{-6} \mu\text{c}/\text{g}$ which is discussed later.

NATURAL RADIOACTIVITY

The isotope K^{40} is present in vegetation at a relatively low specific activity and it is possible to estimate the concentration of this isotope in samples which contain no Ba^{140} - La^{140} . To obtain a measurement of the K^{40} content of sage brush, an 1800 gram composite sample of old sage brush from sampling zones through the area was ashed, the ash placed in a nine ounce bottle and the gamma-ray spectrum measured. A K^{40} specific activity of 1.18×10^{-5} $\mu\text{c}/\text{gram}$ was found which indicates a potassium content of 1.41 per cent of the fresh vegetation weight. This concentration of K^{40} offers only a small interference in the measurement of the fallout isotopes since only 11 per cent of the K^{40} disintegrations involve the emission of a gamma-ray. Neglecting the interference from K^{40} would produce an error of about 5×10^{-8} $\mu\text{c}/\text{gram}$ in an I^{131} measurement.

The counting efficiency and Compton correction factors for K^{40} are included in Table II. A gamma-ray spectrum of K^{40} is included in Appendix A.

OBSERVED CONCENTRATIONS AND ESTIMATED DETECTABLE CONCENTRATIONS OF I^{131} AND THE FALLOUT RADIOISOTOPES

The sensitivity for the measurement of I^{131} and each of the fallout radioisotopes in a vegetation sample depends on the concentrations of the other radioisotopes in the mixture, and it is therefore impossible to state detection limits which will cover all samples.

The detectable amount of each isotope in a specific sample can be calculated from the variance of the measurements. Where a small interference, such as natural radioactivity from K^{40} , is present but is neglected in calculating the radioisotope concentrations, the magnitude of the interference must be considered in estimating the detection limit. Also, since the exact ruthenium and the cerium isotope ratios for each sample are not used in making a Compton correction, the error resulting from this source must also be considered.

To determine the average sensitivities of the radioisotope measurements and at the same time obtain data on the concentrations of the fallout isotopes on vegetation, a series of about 90 vegetation samples were obtained from routine sampling locations and analyzed for I¹³¹ and the fallout radioisotopes. The samples were collected from the various sampling zones in and around the Hanford Atomic Product Operations (see map in Appendix C). Ten vegetation aliquots from each zone were composited into 150 grams samples for gamma-ray spectrometric analysis. The results are included in Appendix C. The approximate sensitivity levels, at the 97.5 per cent confidence level, are given in Table VI. The values are the quantities of the radioisotopes which would give an average counting rate higher than the background (Compton counting rate plus background) 97.5 per cent of the time.

TABLE VI
APPROXIMATE SENSITIVITY LEVELS
FOR THE FALLOUT RADIOISOTOPES

<u>Radioisotope</u>	<u>Detection Limit, $\mu\text{c}/\text{gram} \times 10^6$</u>
Ba ¹⁴⁰ -La ¹⁴⁰	2.5
Zr ⁹⁵ -Nb ⁹⁵	0.64
Ru ^{103-106*}	1.1
I ¹³¹	0.87
Ce ^{141-144**}	3.3

* Ru¹⁰³ : Ru¹⁰⁶ ratio of 2:1.
** Ce¹⁴¹ : Ce¹⁴⁴ ratio of 3:1.

DISCUSSION

It is essential that any analysis of vegetation for I^{131} and the fallout isotopes be a flexible one, since large changes in concentrations as well as composition may be produced by localized or generalized fallout from weapons testing locations in Nevada or other locations on the globe. Gamma-ray spectrometric measurements provide an ideal method of analysis, since the addition of a significant amount of any gamma-emitting radioisotope to the vegetation, either from fallout, plant operation, or other sources, will be observed in the gamma-ray spectrum of the sample. Also, it is usually possible to identify and estimate the concentration of such isotopes. Examples of such cases were the observations of Cs^{137} in two vegetation samples from zone B (see Appendix C). It was possible to attribute this Cs^{137} to plant operation.

The detectable concentration of I^{131} would of course increase if the concentrations of the other radioisotopes were to increase. In fresh fallout, this is not a problem since large amounts of I^{131} are associated with the fallout material and a low I^{131} sensitivity is not required.

As shown in the Experimental section (see Table I), the counting efficiency of I^{131} is increased by using a larger crystal or sample. These data indicate that the sensitivity of the I^{131} measurement might be increased by a factor of two to three by use of a 300 gram sample (16 ounce jar) and a 3 in. x 5 in. diameter crystal.

The ultimate sensitivity for I^{131} measurement could be obtained by using a large well crystal, perhaps 10 in. x 10 in. with a three-inch well for counting a vegetation sample. If such a crystal could be obtained with good resolution properties, the sensitivity for the I^{131} measurement might be increased by a factor of ten.

The quantitative measurement of I^{131} and the fallout radioisotopes in large vegetation samples requires that the radioisotopes be reasonably well distributed throughout the sample. If the majority of the radioactivity were concentrated in a "hot particle" a gross error in the measured radioisotope concentrations could result, however, the relative concentrations would be essentially correct. The measurement of several duplicate vegetation samples (see Appendix C) showed excellent agreement indicating that the radioisotopes were reasonably well distributed on vegetation samples.

ANALYTICAL PROCEDURE

A quantity of 150 grams of sage brush or other types of vegetation to be measured is cut and placed in a nine ounce jar. It is convenient to use a hand press of some type to aid in packing the vegetation into the jar. The zero setting of the 100-channel analyzer is checked as described in the instrumentation section, and the phototube high voltage is adjusted to center the I^{131} photopeak (0.364 Mev.) in channel 12. The gamma-ray spectrum of the sample is then measured.

The background in the channels of interest is subtracted and the Ba^{140} - La^{140} is calculated by use of equation 2. The $Ce^{141-144}$, I^{131} , $Ru^{103-106}$, and Zr^{95} - Nb^{95} are calculated by use of equations 3 through 6, or by use of the approximate equations 7 through 10.

The average Ce^{141} - Ce^{144} ratio is determined by periodically separating the Ce from a few samples and measuring its isotope ratio by either beta or gamma counting techniques.

The average Ru^{103} - Ru^{106} ratio is determined periodically from coincidence counting measurements of the Ru^{106} on a few of the samples. The over all counting efficiency of the coincidence spectrometer for Ru^{106} is first measured by counting a standard solution of Ru^{106} in a nine ounce bottle for one hour (about 10,000 counts). The standard is then removed and the sample counted overnight.

REFERENCES

1. Reid, D. L. and R. J. Morrow, "¹³¹I Vegetation Analysis", (Unpublished Procedure).
2. Davis, J. J. and R. W. Perkins, (Unpublished Work).
3. Anderson, Ernest C., Robert L. Schuch, William R. Fisher, and Wright Langham, "Radioactivity of People and Foods", Science, 125, No. 3261, (June 28, 1957), pp. 1273-78.
4. Miller, C. E., Measurement of Gamma Rays in Vivo, ANL-5596, June 1956, and ANL-5755, June 1957 (Unclassified).
5. Connally, R. E., "Instrumental Methods of Gamma-Ray Spectrometry", Analytical Chemistry, 28, No. 12, (December 1956), pp. 1847-53.
6. Operating Instructions for Model 3300 Pulse Height Analyzer, Radiation Instrument Development Laboratory, Chicago, Illinois.
7. Hunter, H. F. and N. E. Ballou, "Fission-Product Decay Rates", Nucleonics, 9, No. 5, (November 1951), pp. C1 - C7.
8. Perkins, R. W. and J. D. McCormack, The Determination of Cu⁶⁴ in Reactor Effluent Water by Coincidence Counting of the Positron Annihilation Radiation, HW-45636, October 12, 1956 (Confidential).
9. Sullivan, William H., Trilinear Chart of Nuclides, Washington, D. C., U. S. Government Printing Office, 1957.
10. Hollander, J. M., I. Perlman, and G. T. Seaborg, "Table of Isotopes", Rev. Modern Physics, 25, No. 2, (April 1953), p. 499.

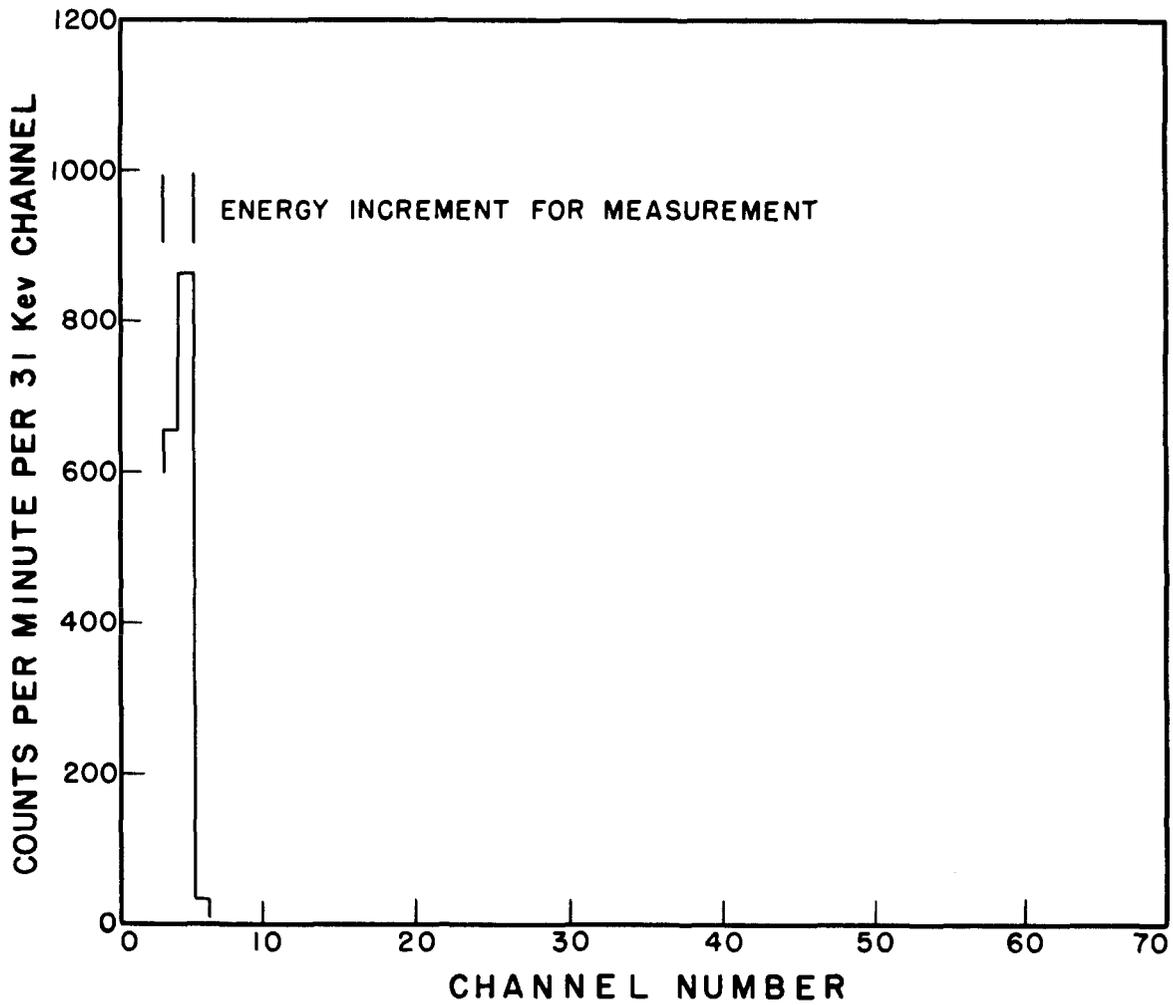


FIGURE - 9
Ce¹⁴¹ GAMMA-RAY SPECTRUM
9 oz. BOTTLE AQUEOUS SOLUTION,
5.355 x 10⁴ d/m.

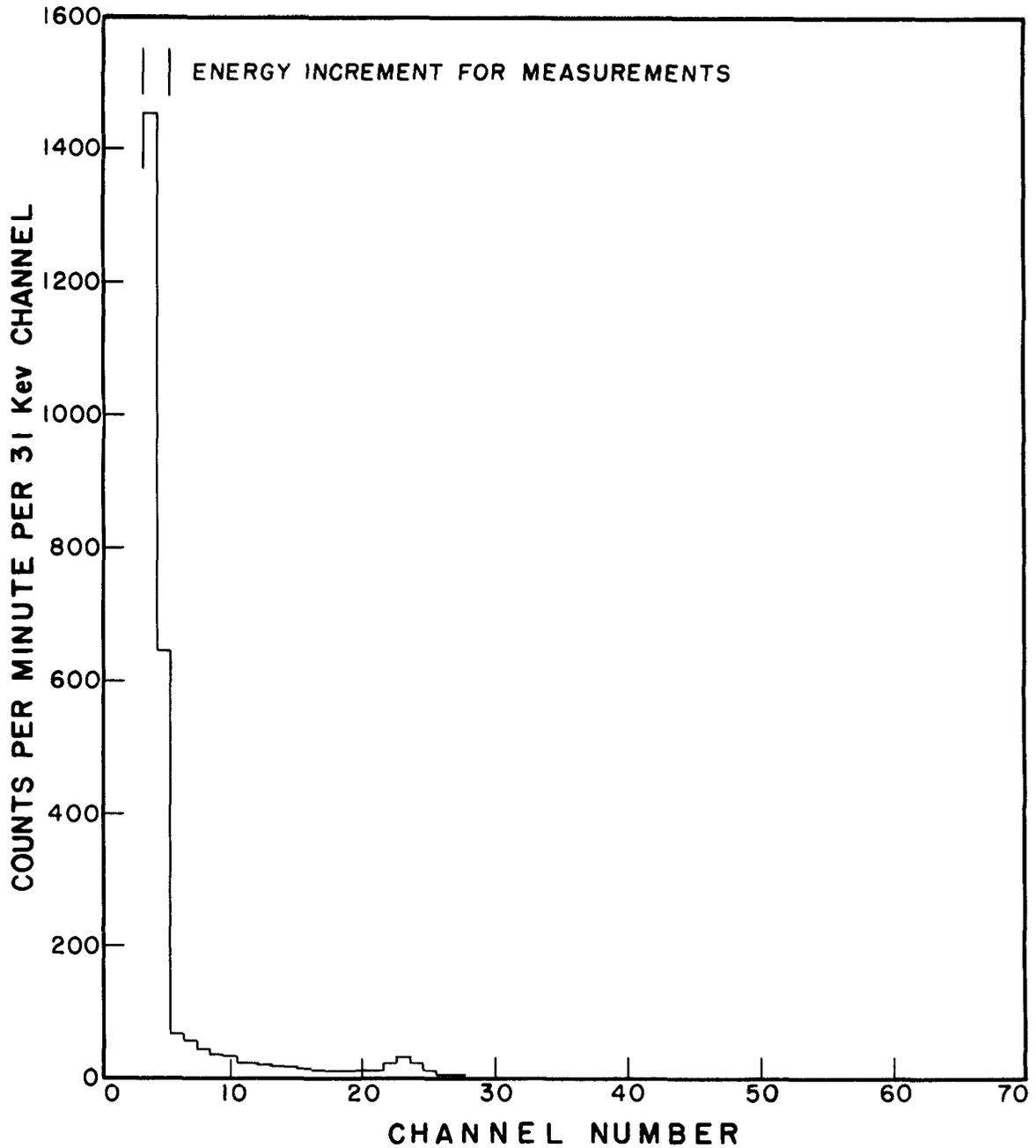


FIGURE - 10
Ce¹⁴⁴ - Pr¹⁴⁴ EQUILIBRIUM MIXTURE
GAMMA-RAY SPECTRUM 9 oz. BOTTLE
AQUEOUS SOLUTION, 4.693×10^5 d/m.

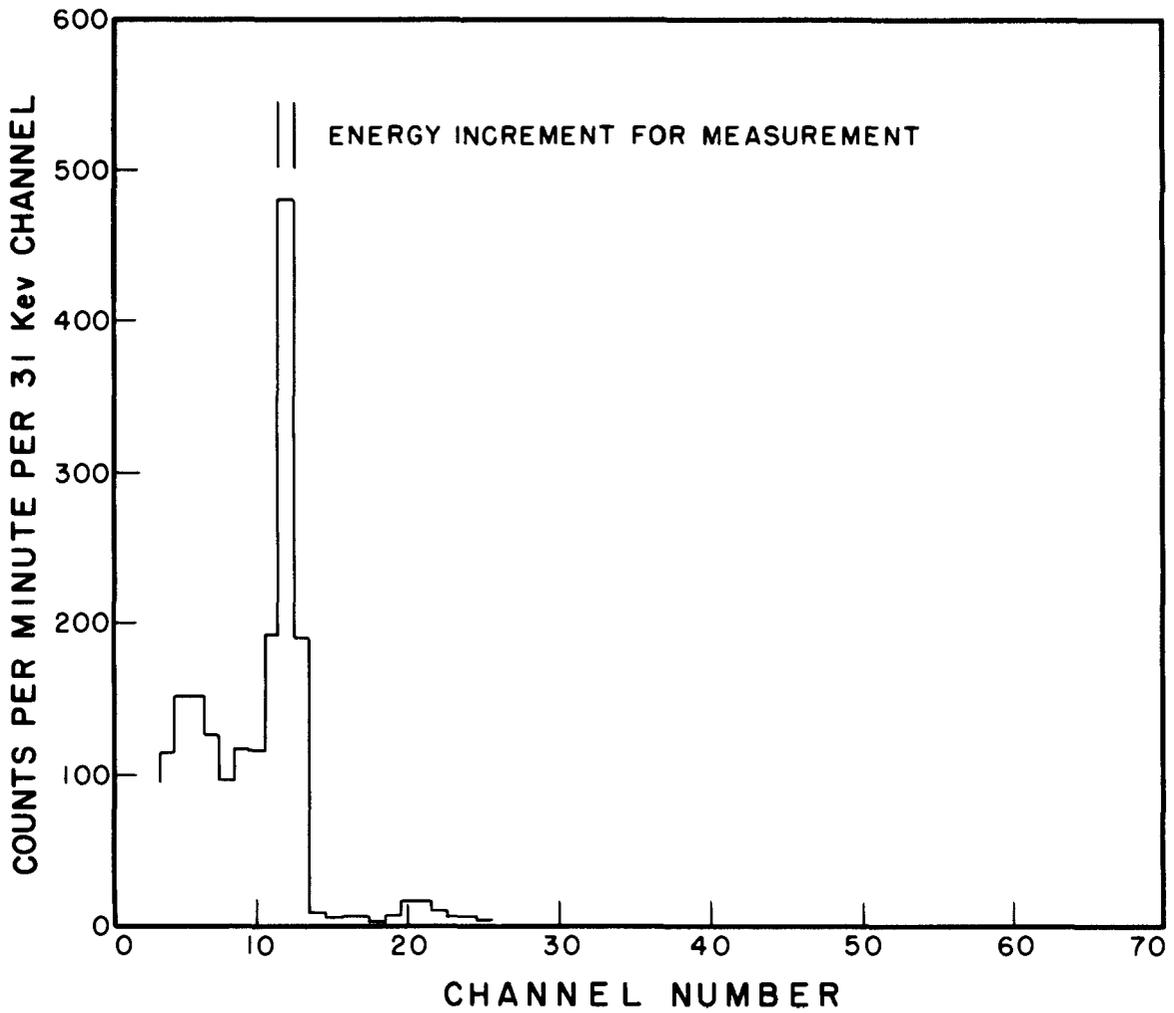


FIGURE - II
I¹³¹ GAMMA-RAY SPECTRUM
9oz. BOTTLE AQUEOUS SOLUTION,
2.472 x 10⁴ d/m.

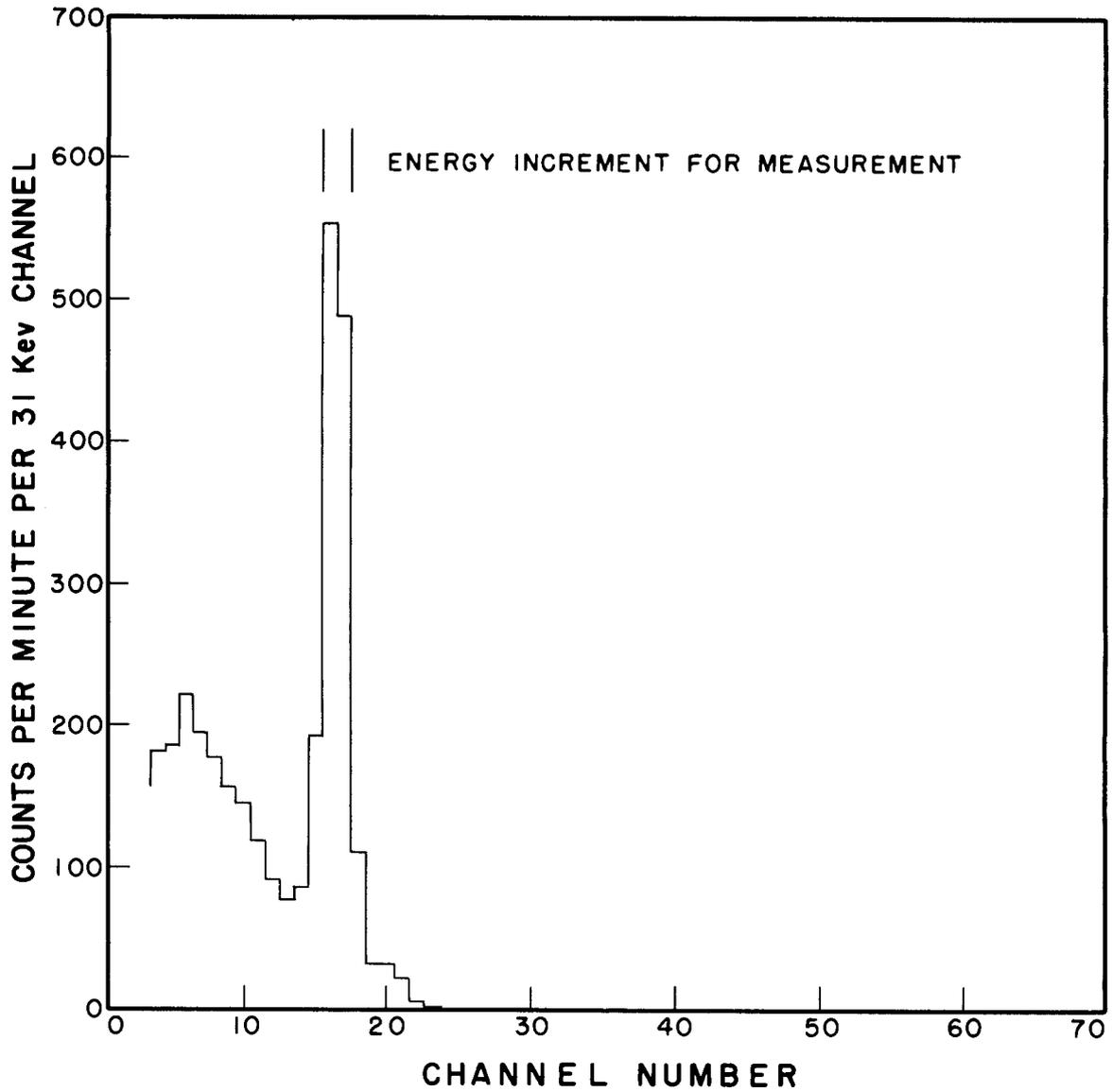


FIGURE - 12
Ru¹⁰³ GAMMA-RAY SPECTRUM
9oz. BOTTLE AQUEOUS SOLUTION,
3.785 x 10⁴ d/m.

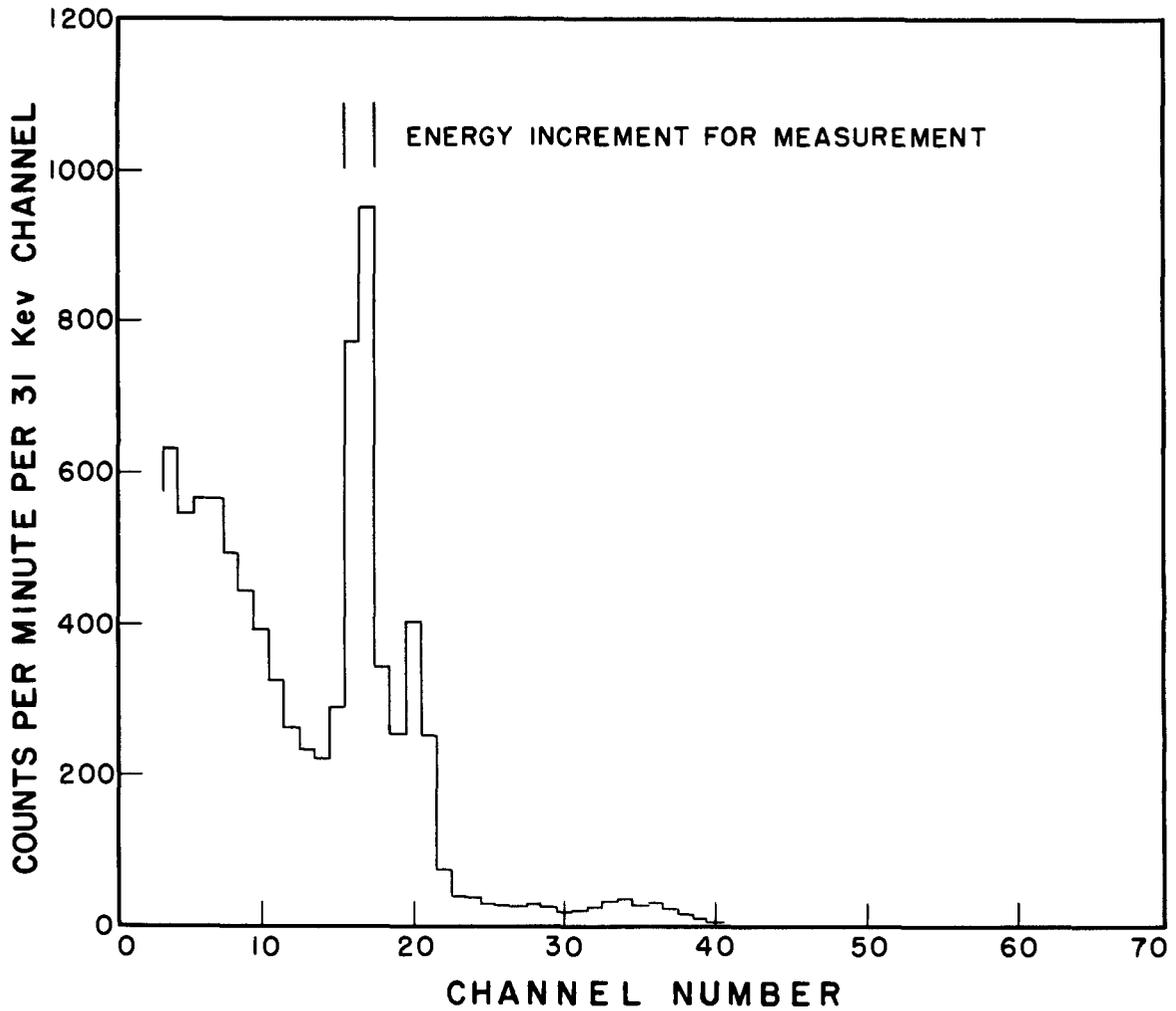


FIGURE - 13
Ru¹⁰⁶ GAMMA-RAY SPECTRUM
9oz. BOTTLE AQUEOUS SOLUTION,
3.406 x 10⁵ d/m.

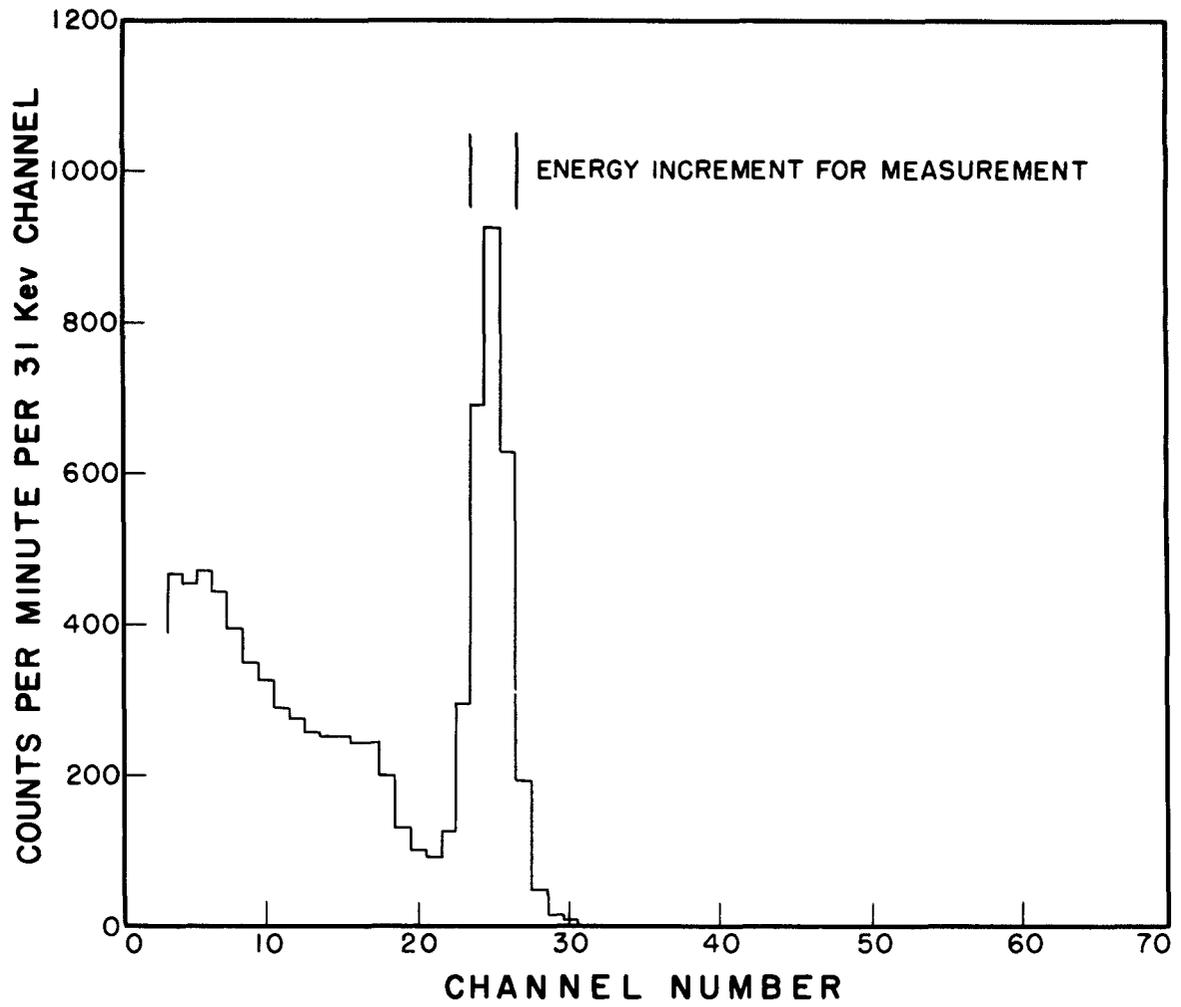


FIGURE - 14
Zr⁹⁵ - Nb⁹⁵ EQUILIBRIUM MIXTURE
GAMMA-RAY SPECTRUM 9 oz. BOTTLE
AQUEOUS SOLUTION, 9.490×10^4 d/m.

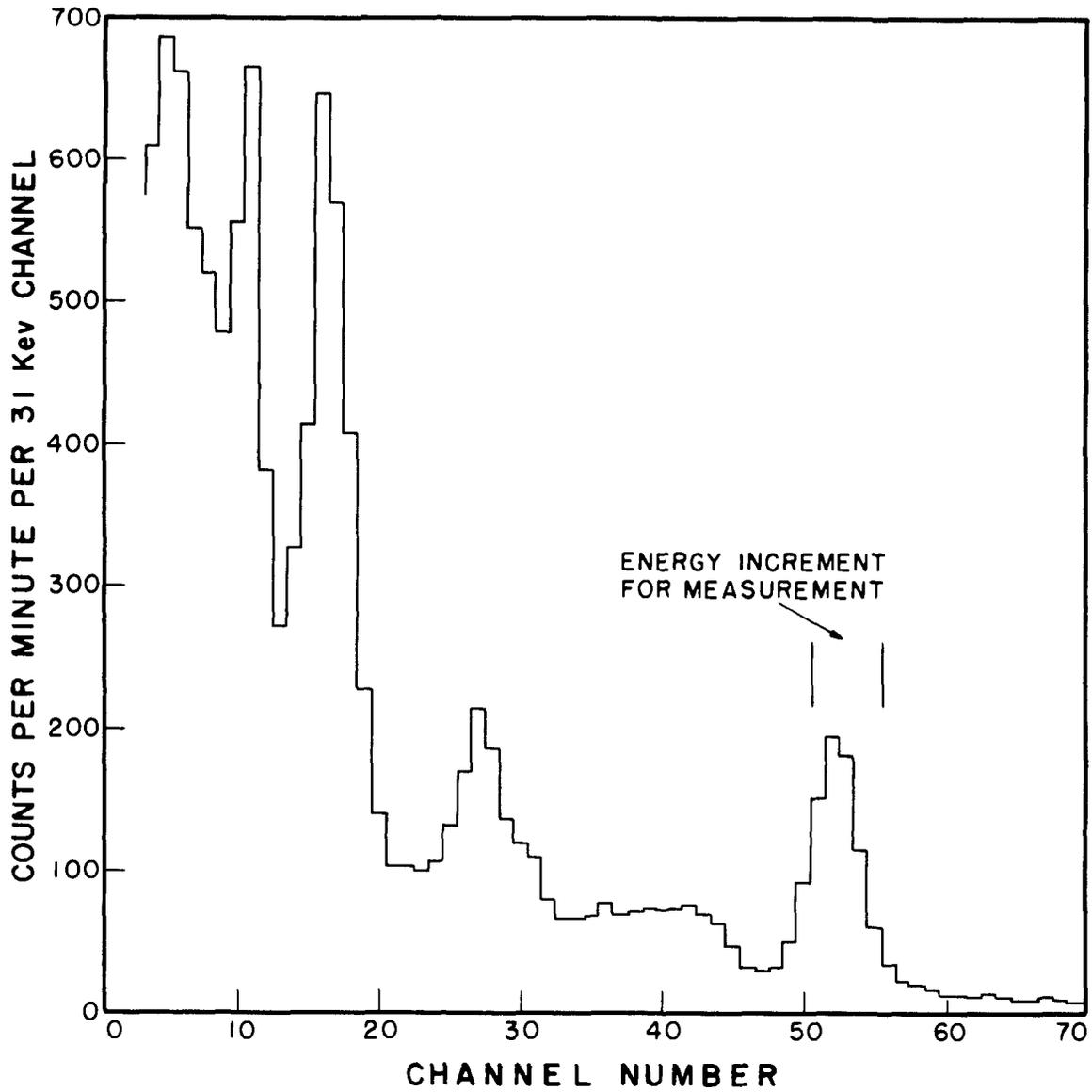


FIGURE - 15
B₀¹⁴⁰ - L₀¹⁴⁰ EQUILIBRIUM MIXTURE
GAMMA-RAY SPECTRUM 9 oz. BOTTLE
AQUEOUS SOLUTION, 1.417×10^5 d/m.

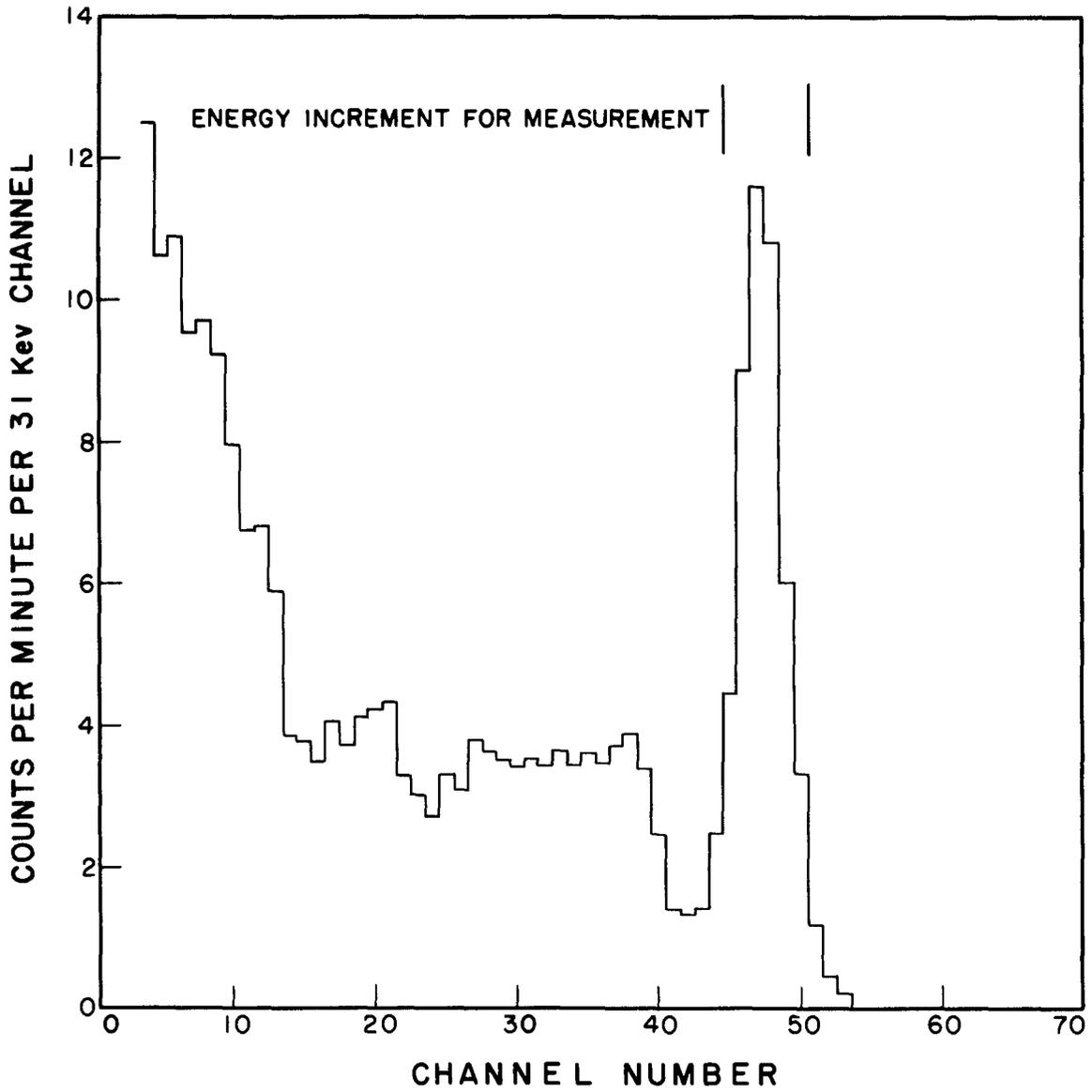


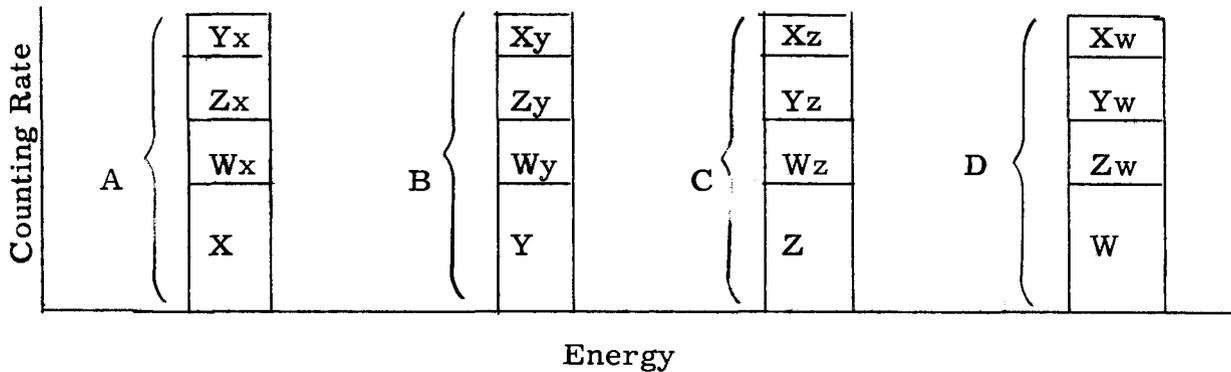
FIGURE - 16
K⁴⁰ GAMMA - RAY SPECTRUM
9oz. BOTTLE AQUEOUS SOLUTION,
3.97 x 10⁴ d/m.

APPENDIX B

MUTUAL INTERFERENCE CORRECTIONS IN A
FOUR COMPONENT SYSTEM

In a case where four radioisotopes are to be measured from a gamma-ray spectrum and each offers a contribution to the counting rate at the photopeaks of the other radioisotopes, the calculation of the "net photopeak counting rates" of the individual radioisotope requires the solution of four simultaneous equations for the four unknowns.

A general solution for the system may be calculated as follows: Let X, Y, Z, and W equal "net photopeak counting rates" due to $Ce^{141-144}$, I^{131} , $Ru^{103-106}$, and $Zr^{95}-Nb^{95}$, respectively, and A, B, C, and D equal the observed counting rates of the photopeaks for the mixture (see illustration).



The counting rates X, Xy, Xz, and Xw are the counting rates due to $Ce^{141-144}$ and are in a fixed ratio (for a given Ce^{141} to Ce^{144} mixture). The same is true for the Y's, Z's and W's.

The ratios of Xy/X , Xz/X , Yx/Y , etc., are thus known constants (and are given in Tables II and III).

Let these constants equal K_1 through K_{12} .

$$\begin{array}{lll} K_1 = \frac{Yx}{Y} & K_2 = \frac{Zx}{Z} & K_3 = \frac{Wx}{W} \\ K_4 = \frac{Xy}{X} & K_5 = \frac{Zy}{Z} & K_6 = \frac{Wy}{W} \\ K_7 = \frac{Xz}{X} & K_8 = \frac{Yz}{Y} & K_9 = \frac{Wz}{W} \\ K_{10} = \frac{Xw}{X} & K_{11} = \frac{Yw}{Y} & K_{12} = \frac{Zw}{Z} \end{array}$$

From inspection of the above illustration it is apparent that X, Y, Z, and W are equal to the following:

$$\begin{array}{l} X = A - (Yx, Zx, Wx) \\ Y = B - (Xy, Zy, Wy) \\ Z = C - (Xz, Yz, Wz) \\ W = D - (Xw, Yw, Zw) \end{array}$$

Substituting in the above K values and arranging the equations in the form suitable for solving by determinants, they become:

$$\begin{array}{l} X + K_1 Y + K_2 Z + K_3 W = A \\ K_4 X + Y + K_5 Z + K_6 W = B \\ K_7 X + K_8 Y + Z + K_9 W = C \\ K_{10} X + K_{11} Y + K_{12} Z + W = D \end{array}$$

Solving the above equations for X, Y, Z, and W by determinants gives the following equations:

$$\begin{array}{ll} X = \frac{\Delta x}{\Delta} & \text{Equation A} \\ Y = \frac{\Delta y}{\Delta} & \text{Equation B} \\ Z = \frac{\Delta z}{\Delta} & \text{Equation C} \\ W = \frac{\Delta w}{\Delta} & \text{Equation D} \end{array}$$

The Δ values are in terms of A, B, C, D, and the constants:

$$\Delta = [K_5 (K_9 K_{11} - K_8) + K_{12} (K_6 K_8 - K_9) + 1 - K_6 K_{11}] - K_4 [K_2 (K_9 K_{11} - K_8) + K_{12} (K_3 K_8 - K_1 K_9) + K_1 - K_3 K_{11}] + K_7 [K_2 (K_6 K_{11} - 1) + K_5 (K_1 - K_3 K_{11}) + K_{12} (K_3 - K_1 K_6)] - K_{10} [K_2 (K_6 K_8 - K_9) + K_5 (K_1 K_9 - K_3 K_8) + K_3 - K_1 K_6]$$

$$\Delta_x = A [K_5 (K_9 K_{11} - K_8) + K_{12} (K_6 K_8 - K_9) + 1 - K_6 K_{11}] - B [K_2 (K_9 K_{11} - K_8) + K_{12} (K_3 K_8 - K_1 K_9) + K_1 - K_3 K_{11}] + C [K_2 (K_6 K_{11} - 1) + K_5 (K_1 - K_3 K_{11}) + K_{12} (K_3 - K_1 K_6)] - D [K_2 (K_6 K_8 - K_9) + K_5 (K_1 K_9 - K_3 K_8) + K_3 - K_1 K_6]$$

$$\Delta_y = A [K_{12} (K_4 K_9 - K_6 K_7) + K_5 (K_7 - K_9 K_{10}) + K_{10} K_6 - K_4] + B [K_7 (K_{12} K_3 - K_2) + K_{10} (K_2 K_9 - K_3) + 1 - K_9 K_{12}] + C [K_{12} (K_6 - K_3 K_4) + K_5 (K_{10} K_3 - 1) + K_2 (K_4 - K_6 K_{10})] + D [K_5 (K_9 - K_7 K_3) + K_2 (K_7 K_6 - K_4 K_9) + K_4 K_3 - K_6]$$

$$\Delta_z = A [K_4 (K_8 - K_9 K_{11}) + K_7 (K_6 K_{11} - 1) + K_{10} (K_9 - K_6 K_8)] + B [K_7 (K_1 - K_3 K_{11}) + K_{10} (K_3 K_8 - K_1 K_9) + K_9 K_{11} - K_8] + C [K_4 (K_3 K_{11} - K_1) + K_{10} (K_1 K_6 - K_3) + 1 - K_6 K_{11}] + D [K_4 (K_1 K_9 - K_3 K_8) + K_7 (K_3 - K_1 K_6) + K_6 K_8 - K_9]$$

$$\Delta_w = A [K_4 (K_{11} - K_8 K_{12}) + K_5 (K_8 K_{10} - K_7 K_{11}) + K_7 K_{12} - K_{10}] + B [K_7 (K_2 K_{11} - K_1 K_{12}) + K_{10} (K_1 - K_2 K_8) + K_{12} K_8 - K_{11}] + C [K_5 (K_{11} - K_1 K_{10}) + K_{12} (K_1 K_4 - 1) + K_2 (K_{10} - K_4 K_{11})] + D [K_5 (K_1 K_7 - K_8) + K_4 (K_2 K_8 - K_1) + 1 - K_2 K_7]$$

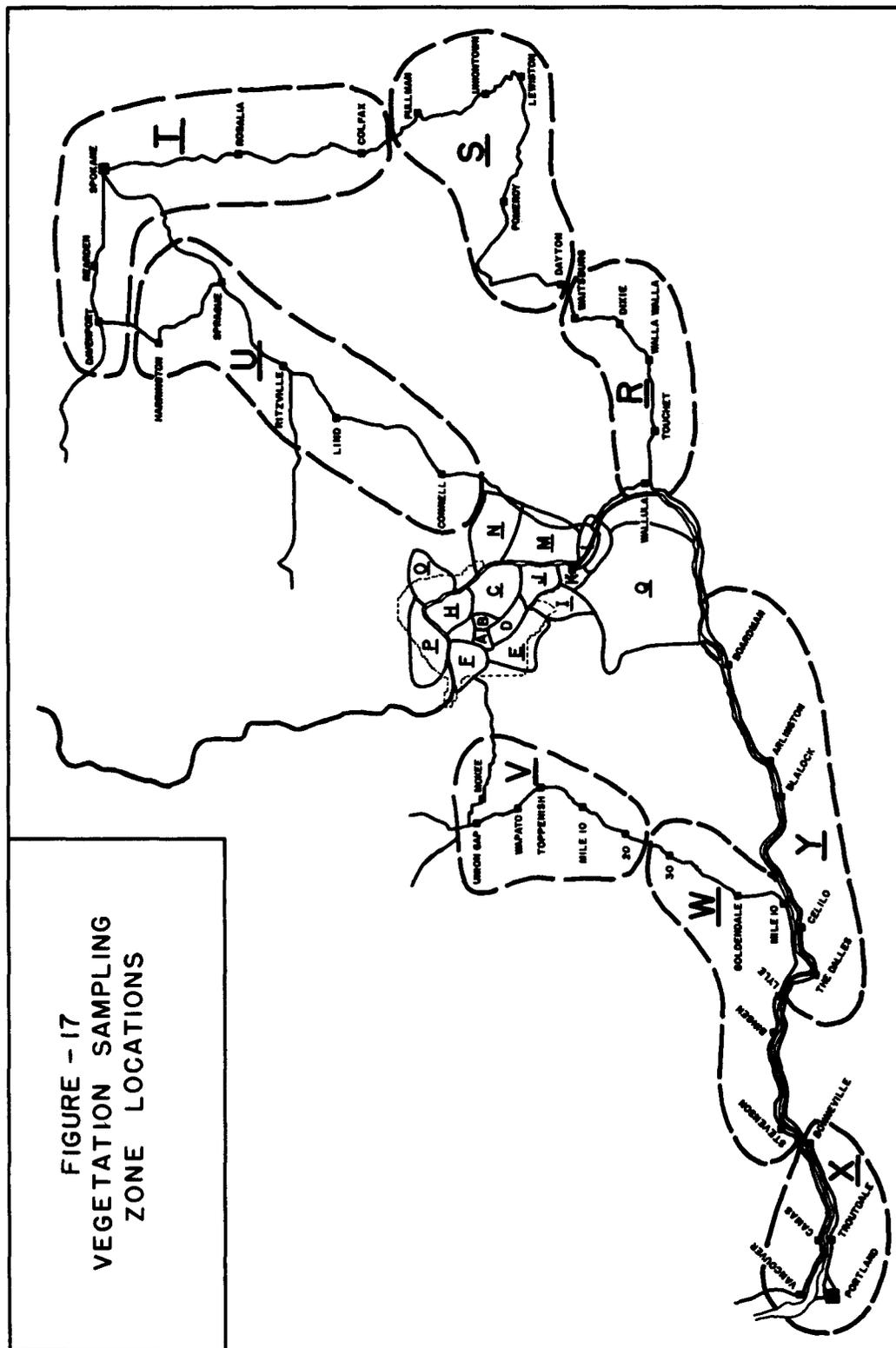
The K values given in Tables II and III (for Ru¹⁰³-Ru¹⁰⁶ and Ce¹⁴¹-Ce¹⁴⁴ ratios of 2:1 and 1:3) are:

K ₁	0.593	K ₅	0.0935	K ₉	0.215
K ₂	0.395	K ₆	0.121	K ₁₀	0.00555
K ₃	0.321	K ₇	0.00428	K ₁₁	0.0156
K ₄	0.00340	K ₈	0.0215	K ₁₂	0.00445

Substituting these values into the above equations gives these comparatively simple equations

$$\begin{aligned}X &= 1.004 A - (0.5851 B + 0.3413 C + 0.1784 D) \\Y &= 1.004 B - (0.00246 A + 0.0926 B + 0.1012 D) \\Z &= 1.004 C - (0.00306 A + 0.0165 B + 0.2125 D) \\W &= 1.004 D - (0.00552 A + 0.01236 B + 0.00113 C).\end{aligned}$$

These equations are presented in terms of the radioisotopes, counting rates, and channel numbers on page 25.



CONFIDENTIAL

TABLE VII

CONCENTRATIONS OF RADIOISOTOPES FOUND ON VEGETATION SAMPLES

($\mu\text{c}/\text{gram} \times 10^6$)

$\text{Ru}^{103} : \text{Ru}^{106}$ Ratio of 2:1

$\text{Ce}^{141} : \text{Ce-Pr}^{144}$ Ratio of 1:3

Date	Location, Zone	$\text{Zr}^{95}\text{-Nb}^{95}$	$\text{Ru}^{103}\text{-Ru}^{106}$	I^{131}	$\text{Ba}^{140}\text{-La}^{140}$	$\text{Ce}^{141}\text{-Ce}^{144}\text{-Pr}^{144}$
7-2-57	O	17.1	8.92	2.61	18.2	46.0
7-2-57	P	23.8	6.27	0.673	18.5	40.0
7-8-57	C	22.4	7.49	4.40	16.2	49.2
7-8-57	F	28.8	8.99	1.98	15.6	47.1
7-8-57	I	15.0	5.06	1.27	7.54	25.4
7-9-57 *	B	31.5	14.9	7.56	19.7	38.1
7-9-57	G	37.9	13.1	2.03	21.5	77.2
7-9-57	J	25.3	7.70	2.07	12.4	50.1
7-9-57	M	7.81	3.81	0.145	4.92	20.4
7-9-57	N	11.1	3.30	2.35	5.08	45.2
7-9-57	O	26.3	7.32	1.41	17.3	60.9
7-9-57	P	21.1	5.92	1.31	14.0	38.4
7-10-57	A	39.5	9.15	2.57	18.1	72.6
7-10-57	D	27.2	6.52	1.53	13.8	43.5
7-10-57	H	22.1	8.27	0.949	14.9	44.6
7-10-57	K	17.2	5.32	0.679	11.9	35.9
7-10-57	L	5.19	1.47	0.124	2.31	9.77
7-10-57	R	15.8	5.22	0.330	6.71	28.8
7-10-57	S	9.67	2.67	--	8.68	21.9
7-10-57	T	11.8	3.03	--	4.62	20.6
7-10-57	U	10.5	3.23	0.178	8.77	27.0

* $\text{Cs}^{137} = 150$

CONFIDENTIAL

CONFIDENTIAL

TABLE VII (contd.)

$\text{Ru}^{103} : \text{Ru}^{106}$ Ratio of 2:1

$\text{Ce}^{141} : \text{Ce-Pr}^{144}$ Ratio of 1:3

Date	Location, Zone	$\text{Zr}^{95}\text{-Nb}^{95}$	$\text{Ru}^{103}\text{-Ru}^{106}$	I^{131}	$\text{Ba}^{140}\text{-La}^{140}$	$\text{Ce}^{141}\text{-Ce}^{144}\text{-Pr}^{144}$
7-11-57	E	16.1	3.26	0.217	7.74	24.1
7-11-57	Q	6.29	1.52	--	4.78	10.5
7-15-57	C	27.9	6.08	1.62	8.64	36.5
7-15-57	F	30.3	7.29	0.123	13.5	44.9
7-15-57	I	19.9	5.44	0.0583	7.61	28.0
7-16-57	G	24.8	10.9	1.89	15.9	57.4
7-16-57	J	13.0	4.04	0.604	7.67	31.0
7-16-57	M	7.86	2.75	0.532	4.96	20.4
7-16-57	N	12.0	4.45	0.402	3.38	17.6
7-16-57	O	28.2	8.78	0.218	15.9	52.8
7-16-57	P	26.4	11.9	1.30	11.7	51.7
7-17-57	A	48.5	12.4	5.68	13.4	80.0
7-17-57*	B	53.2	20.7	7.69	21.0	95.2
7-17-57	H	24.1	6.12	1.19	14.2	48.7
7-17-57	K	10.8	3.63	0.178	7.27	27.7
7-17-57	L	8.03	2.18	--	5.83	18.0
7-17-57	Z	9.83	3.47	--	6.36	22.0
7-18-57	D	17.7	9.15	1.35	6.26	29.7
7-18-57	Q	7.35	1.89	--	3.36	13.3
7-19-57	E	28.6	5.28	0.667	10.8	41.9
7-22-57	C-1	26.2	6.89	2.23	12.4	53.3
7-22-57	C-2	24.8	8.54	1.50	10.5	53.9
7-22-57	F	36.1	9.58	0.877	11.1	62.3
7-22-57	I	18.7	4.81	0.715	7.33	31.3
7-23-57	B-1	76.8	30.1	7.52	14.1	134
7-23-57	B-2	80.4	22.9	8.63	14.3	127

* Cs^{137} - 270

CONFIDENTIAL

CONFIDENTIAL

CONFIDENTIAL

TABLE VII (contd.)

$\text{Ru}^{103} : \text{Ru}^{106}$ Ratio of 2:1

$\text{Ce}^{141} : \text{Ce-Pr}^{144}$ Ratio of 1:3

Date	Location, Zone	$\text{Zr}^{95} - \text{Nb}^{95}$	$\text{Ru}^{103} - \text{Ru}^{106}$	I ¹³¹	$\text{Ba}^{140} - \text{La}^{140}$	$\text{Ce}^{141} - \text{Ce}^{144} - \text{Pr}^{144}$
7-23-57	G	32.8	11.7	1.50	10.3	49.2
7-23-57	J	16.0	3.10	0.886	4.49	25.6
7-23-57	M	12.7	3.00	0.444	4.17	19.0
7-23-57	N	6.97	0.975	0.937	4.12	13.0
7-23-57	O	15.5	4.97	1.33	6.10	25.0
7-23-57	P	19.5	4.52	--	5.83	27.7
7-23-57	R	6.54	1.42	--	1.96	3.01
7-23-57	S	4.63	2.69	0.607	6.32	13.2
7-23-57	T	12.6	1.59	0.247	3.45	13.6
7-24-57	A-1	24.0	6.88	2.37	5.41	28.3
7-24-57	A-2	23.8	5.51	1.29	5.41	28.8
7-24-57	D-1	21.1	6.37	1.78	8.45	45.7
7-24-57	D-2	21.0	6.42	1.26	8.02	40.0
7-24-57	H	24.2	10.8	2.64	11.6	63.1
7-24-57	K	20.0	3.74	1.03	5.24	26.2
7-24-57	L	7.18	2.37	0.0360	2.77	17.4
7-24-57	U	22.0	5.36	0.280	8.57	40.5
7-25-57	Q	4.75	2.28	0.901	--	12.1
7-26-57	E	12.7	2.94	0.303	5.63	19.9
7-29-57	C-1	21.4	5.27	1.82	5.42	38.1
7-29-57	C-2	30.0	9.54	3.33	12.0	59.7
7-29-57	F	27.8	5.03	0.190	5.81	30.5
7-29-57	I	30.4	5.34	0.679	4.23	33.7
7-30-57	A-1	26.7	6.26	2.08	6.67	30.7
7-30-57	A-2	24.3	5.26	3.69	5.16	30.6
7-30-57	E	19.0	3.90	0.229	4.43	25.9
7-30-57	G	43.6	8.42	--	7.27	48.6
7-30-57	J	12.2	1.95	1.05	2.99	17.7
7-30-57	M	9.74	1.59	0.712	3.04	13.5
7-30-57	N	11.8	2.63	1.01	2.31	20.0

CONFIDENTIAL

CONFIDENTIAL

CONFIDENTIAL

TABLE VII (contd.)

$\text{Ru}^{103} : \text{Ru}^{106}$ Ratio of 2:1

$\text{Ce}^{141} : \text{Ce-Pr}^{144}$ Ratio of 1:3

<u>Date</u>	<u>Location, Zone</u>	<u>Zr⁹⁵-Nb⁹⁵</u>	<u>Ru¹⁰³-Ru¹⁰⁶</u>	<u>I¹³¹</u>	<u>Ba¹⁴⁰-La¹⁴⁰</u>	<u>Ce¹⁴¹-Ce¹⁴⁴-Pr¹⁴⁴</u>
7-30-57	O	18.6	5.11	1.15	4.60	28.3
7-30-57	P	22.3	4.73	0.336	4.60	28.3
7-30-57	V	7.62	2.11	0.321	2.34	11.5
7-30-57	W	1.79	0.664	--	1.19	2.70
7-31-57	D-1	17.0	3.99	0.520	4.71	22.3
7-31-57	D-2	15.0	3.45	0.628	4.57	20.5
7-31-57	H	21.9	5.35	0.538	5.90	34.1
7-31-57	K	8.98	1.95	0.299	2.36	13.2
7-31-57	L	3.38	1.39	0.456	1.76	4.52
7-31-57	X	5.50	0.260	--	1.80	4.12
7-31-57	Y	6.42	2.04	0.245	1.52	8.65
8-1-57	B-1	31.9	7.50	2.26	7.11	45.6
8-1-57	B-2	26.1	7.78	4.02	5.84	33.1
8-1-57	Q	7.03	1.26	0.342	2.13	10.5

CONFIDENTIAL

CONFIDENTIAL

CONFIDENTIAL

INTERNAL DISTRIBUTION

Copy Number

1	B. V. Andersen
2	G. J. Behling
3	R. J. Brouns
4	L. P. Bupp - H. M. Parker
5	A. C. Case - D. E. Warner
6	H. V. Clukey
7	J. J. Davis
8	J. W. Healy
9	A. R. Keene
10	H. A. Kornberg
11	J. M. Nielsen
12	D. W. Pearce
13	R. W. Perkins
14	W. H. Reas
15	D. L. Reid - H. A. Treibs
16	L. C. Schwendiman - H. G. Rieck
17	300 File Copy
18	Record Center
19 - 24	Extra

EXTERNAL DISTRIBUTION

25 -26	Aerojet-General Corporation
27	AFPR, Curtiss-Wright, Clifton
28	AFPR, Lockheed, Burbank
29 - 30	AFPR, Lockheed, Marietta
31	Air Technical Intelligence Center
32	Alco Products, Inc.
33	Allied Chemical and Dye Corporation
34 - 37	Argonne National Laboratory
38	Armed Forces Special Weapons Project (Sandia)
39	Army Chemical Center
40 - 43	Atomic Energy Commission, Washington
44 - 45	Atomics International
46	Babcock and Wilcox Company (NYOO-1940)
47	Babcock and Wilcox Company (SOO-274)
48	Battelle Memorial Institute
49 - 50	Bettis Plant (WAPD)
51	Brookhaven National Laboratory
52	Bureau of Aeronautics

EXTERNAL DISTRIBUTION (contd.)Copy Number

53	Bureau of Ships
54	Bureau of Yards and Docks
55	Chicago Patent Group
56	Columbia University (Hassialis)
57	Combustion Engineering, Inc. (CERD)
58	Curtiss-Wright Corporation
59	Division of Raw Materials, Washington
60	Dow Chemical Company (Rocky Flats)
61 - 63	duPont Company, Aiken
64	duPont Company, Wilmington
65	Fluor Corporation
66	Foster Wheeler Corporation
67	General Atomic Division
68 - 70	General Electric Company (ANPD)
71 - 72	Goodyear Atomic Corporation
73	Hanford Operations Office
74	Iowa State College
75 - 76	Knolls Atomic Power Laboratory
77 - 78	Los Alamos Scientific Laboratory
79 - 81	Mallinckrodt Chemical Works
82	Marquardt Aircraft Company
83	Martin Company
84	Mound Laboratory
85	National Advisory Committee for Aeronautics, Cleveland
86	National Bureau of Standards
87	National Lead Company, Inc., Winchester
88	National Lead Company of Ohio
89	Naval Air Development Center
90	Naval Medical Research Institute
91	Naval Research Laboratory
92	New Brunswick Area Office
93	New York Operations Office
94	Nuclear Development Corporation of America
95	Nuclear Metals, Inc.
96	Office of Naval Research
97	Patent Branch, Washington
98 - 101	Phillips Petroleum Company (NRTS)
102	Pratt and Whitney Aircraft Division
103	Public Health Service
104	Sandia Corporation
105	Sandia Corporation, Livermore

