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Nondestructive Activation Analysis of Environmental Samples

RICHARD DAMS and JOHN A. ROBBINS

JOHN W. WINCHESTER
Project Director

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THE UNIVERSITY OF MICHIGAN
GREAT LAKES RESEARCH DIVISION

Technical Report

NONDESTRUCTIVE ACTIVATION ANALYSIS OF ENVIRONMENTAL SAMPLES

Richard Dams
John A. Robbins

John W. Winchester
Project Director

ORA Projects 08903 and 12341

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I. Neutron Activation Analysis.

Introduction

The qualitative and quantitative determination of minute quantities by a technique known as activation analysis has become increasingly popular and is now generally recognized as a useful technique in various areas of scientific investigation and in an ever growing number of industrial control applications.

In this technique a nonradioactive substance is made radioactive by particle bombardment, and its decay characteristics are then measured. The nature of these characteristics allow an identification and the magnitude is a measure of the concentration. The sensitivity is extremely high for a large number of elements. The specificity is usually excellent. A number of textbooks describing the principles, the possibilities and a number of applications are available (1-5).

1. Principles.

Activation analysis is the determination of the weight of a particular element in a sample by measuring the radioactivity induced by irradiation, usually with thermal neutrons, but in some cases with fast neutrons or even with photons or charged particles.

The most commonly used and by far most intense neutron sources are provided by neutron chain reactors utilizing the fission reaction. Fast neutrons (>1 Mev) produced by the fission of uranium are moderated to epithermal (~ 0.4 ev) and further to thermal (<0.4 ev) energies. These thermal neutrons assure continuation of the fission reaction but can also be used to produce artificial radioactivity induced when a target material is exposed to thermal energies a chance exists that another nuclide will be created by neutron capture, immediately followed by emission of electromagnetic radiation.

The reaction is as follows

Examples are ${}_{11}^{23}\text{Na} (n, \gamma) {}_{11}^{24}\text{Na}$
 ${}_{27}^{59}\text{Co} (n, \gamma) {}_{27}^{60}\text{Co}$

The nuclides formed, ${}_{Z}^{A+1}\text{M}$, ${}_{11}^{24}\text{Na}$, ${}_{27}^{60}\text{Co}$ are often unstable and disintegrate, usually by emission of beta rays to an excited state of another isotope which deexcitates immediately by the emission of gamma rays. Beta rays are electrons or positrons. The beta rays from a given radioactive species are emitted with a continuous energy distribution extending from zero up to a maximum value. The gamma rays emitted by an excited state of a nuclide consist of electromagnetic radiations and have well defined energies $E = h\nu$ (ν is the frequency of the electromagnetic radiation). Gamma and beta rays can be detected with special counting equipment. More details about decay mode and interaction of radiation with matter can be found in the excellent books by Friedlander et al. "Nuclear and Radiochemistry" (6), and B. Harvey, "Introduction to Nuclear Physics and Chemistry" (7).

2. Formation and decay rate of radioactive species.

A target material A exposed to neutrons leads to the formation of nuclei B, which disintegrate to produce C as follows:

The rate of disintegration per atom of B is a constant, λ , specific for nucleus B. Indeed within a fixed time (half-life = $t_{1/2}$) half of the radioactive nuclides B will disintegrate into C. Thus $\lambda = \ln 2 / t_{1/2} \text{ (sec}^{-1}\text{)}$. The rate of formation of B depends on: (1) the number of atoms A present, namely N_A ; (2) the neutron flux ϕ , the number of neutrons per second passing through an area of 1 square centimeter (i.e. $\phi \text{ cm}^{-2}\text{-sec}^{-1}$); (3) the activation cross section of A, namely σ , expressed in barns ($1 \text{ barn} = 10^{-24} \text{ cm}^2$).

The rate of accumulation of B is given by the rate of formation minus the rate of disintegration.

$$\text{Thus: } \frac{dN_B}{dt} = \sigma \phi N_A - \lambda N_B \quad (1)$$

After integration assuming that before irradiation $N_B=0$, one finds that at the end of irradiation t_i , the number of atoms of B present equals:

$$N_B^{t_i} = \frac{\sigma \phi N_A}{\lambda} [1 - \exp(-\lambda t_i)] \quad (2)$$

The number of disintegrations per second, or the radioactivity at the end of irradiation t_i equals:

$$\text{Act}_B^{t_i} = N_B^{t_i} \lambda = \sigma \phi N_A [1 - \exp(-\lambda t_i)] = \sigma \phi N_A S \quad (3)$$

where $S = 1 - \exp(-\lambda t_i)$ is called the saturation factor. The radioactivity present at time t_d after the end of irradiation will be smaller and is given as:

$$\text{Act}_B^{t_d} = \text{Act}_B^{t_i} \exp(-\lambda t_d) \quad (4)$$

Thus knowing σ , λ or $t_{1/2}$, ϕ and N_A the radioactivity for each irradiation time and cooling time can easily be calculated. Approximate values for σ and $t_{1/2}$ are tabulated (6, 8). The neutron flux can be determined and is usually approximately known for each irradiation site. Neutron fluxes vary between 10^8 n cm⁻² - sec⁻¹ up to several times 10^{14} n cm⁻² - sec⁻¹ depending on the power of the nuclear reactor and on the irradiation site in the reactor. The number of atoms is given as

$$N_A = \frac{0.602 \times 10^{24} \times g \times \theta}{\text{M.W.}} \quad (5)$$

where g is the weight in grams
M.W. is the molecular weight of the target material
 θ is the isotopic abundance

3. Activation Analysis.

Inversely one obtains the weight of the element from the activity measurement by substituting equation (5) in (3)

$$g = \frac{\text{Act}_B^{t_i} \times \text{M.W.}}{0.602 \times 10^{24} \times \sigma \times \phi \times S \times \theta} \quad (6)$$

This technique is called absolute activation analysis. The values for σ are however not known accurately, a precise determination of ϕ is time consuming and an absolute measurement of the radioactivity is difficult. Therefore, a comparator method is usually used in activation analysis. A standard containing a well known

amount of the element is irradiated simultaneously with and as near as possible to the unknown sample. A relative counting of both samples is performed and the unknown weight is calculated as follows:

$$(g)v / (g) S = (Act)v / (Act)S \quad (7)$$

where (g)v and (g)S are the weights of unknown and standard (Act)v and (Act)S are countrates of unknown and standard.

Many sources of errors can be eliminated by irradiating the sample with a standard of similar composition. When the composition of a substance is unknown, a preliminary irradiation has to be performed and can be used as a qualitative analysis in order to fabricate a suitable standard. Care should be taken that the sample and standard are approximately the same weight, shape and thickness.

4. Sensitivity-Standard Deviations.

Neutron activation analysis is inherently a very sensitive technique for determination of a large number of elements. In the analytical and radiochemical literature several mathematical expressions are used as definitions for the limit of detection, ranging from one to ten times the standard deviation of the background and employing a widely varying terminology. Thus in order to draw valid conclusions with respect to detection capabilities from published data, it is necessary to examine carefully the definition and confidence level accepted by the author.

As can be seen from equation (6) the sensitivity of the analysis of an element by neutron activation is determined by its cross section (σ), its isotopic abundance (θ), the half-life of the isotope produced (λ), the irradiation time (t_i), and the neutron flux (ϕ). Furthermore, the sensitivity is determined by the efficiency of the detector and its background signal.

The following example shows how the sensitivity for an element can be calculated from equation (6).

Assume the following realistic values: $\sigma = 1$ barn

$$\theta = 1 \text{ (100\%)}$$

$$\text{M.W.} = 100$$

$$t_i = t_{1/2} \text{ thus } S = 0.5$$

$$\phi = 10^{12} \text{ n/cm}^2\text{-sec}$$

If the count rate necessary is 1 count per second with a detection efficiency of 33 percent the sensitivity for such an element is given as:

$$g = \frac{3 \times 100}{0.6 \times 10^{24} \times 2 \times 10^{12} \times 0.5 \times 1 \times 10^{-24}} = 10^{-9} \text{ grams}$$

If gamma counting is performed the abundance of the gamma rays in the decay scheme must be included.

Jenkins and Smales (9) calculated sensitivities assuming an irradiation at $10^{12} \text{ n/cm}^2\text{-sec}^{-1}$ to saturation or up to one month (which ever is shorter) and counting 2 hours after irradiation to allow for chemical separation. The lower limits of precise measurements were taken to be 100 dis/min, for beta emitters and 100 photons/min for gamma emitters. Table 1 summarizes the results.

As a matter of fact practical sensitivities may be much less favorable if a number of elements are determined simultaneously because irradiation cooling and counting time cannot be ideal for each isotope. An appropriate compromise must then be applied.

Radioactive disintegration being a statistical phenomenon the standard deviation of a counting equals the square root of the number of counts. If the net number of counts N is obtained by subtracting the background B from the total number of counts T. The standard deviation is calculated as follows:

$$\sigma_N = \sqrt{\sigma_T^2 + \sigma_B^2} = \sqrt{T + B}$$

5. Counting-Gamma Spectrometry.

When a gamma ray strikes a scintillation counter (NaI(Tl)) it produces light flashes which release photoelectrons from a photosensitive electrode.

Element	Estimated sensitivity, grams
Eu, Au, Ho,	10^{-12}
As, Ir, In, Kr, La, Mn, Pr, Re, Sm, Se, Tb, Tm, W, Yb	10^{-11}
Sb, Ar, Br, Cs, Co, Cu, Er, Gd, Ga, Hf, Nd, Pd, P, Rb, Na, Ta, Th, U, Y	10^{-10}
Ba, Cd, Ce, Cl, Ge, I, Hg, Os, Pt, K, Ru, Ag, Te, Sn, Xe, Zn	10^{-9}
Bi, Mo, Ni, Se, Si, S, Tl,	10^{-8}
Ca, Cr, Fe, O, Sr, Zr,	10^{-7}
Pb	10^{-6}

Table 1. Sensitivity of activation analysis as calculated by Jenkins and Smales (9).

These are multiplied in a photomultiplier to produce a pulse which then can be further amplified and recorded.

When a gamma ray strikes a semiconductor diode (Ge(Li)) a number of electrons are lifted from the valence band to the conduction band and swept through the material under the influence of an applied electric field so that an electric pulse is obtained. This pulse can also be amplified and recorded.

Whenever the output from a detector-amplifier system is proportional to the energy dissipation in the detector the measurements of pulse heights may be very useful. Pulse height analysis is made versatile and rapid by the use of a very important tool in activation analysis, namely the multichannel analyzer. In the analogue to digital converter (A.D.C.) the pulses are sorted according to size and immediately recorded as counts in the appropriate channel of the memory unit. The resultant plot of the number of counts as a function of channel number is called a "gamma spectrum". The number of channels of an analyzer is determined by the capacity of the analyzer memory. Pulse height analysers with 50 up to 8192 channels are in use. The time required for sorting and storing a pulse is appreciable and the analyzer cannot process a new pulse during this "dead time".

Provision is usually made to lengthen the counting time because of these dead time losses. A multichannel should also include the ability to display the gamma-spectrum on an oscilloscope and to drive an automatic printer, plotter or magnetic tape unit so that the content of each channel can be appropriately recorded.

Gamma rays emitted by radionuclides being monoenergetic, it is possible to detect several isotopes in presence of each other if a response proportional to the original gamma-ray energy can be obtained from a detection unit. Thus if a scintillation crystal or a solid state detector is large enough to contain the entire path of the gamma ray, it can measure its total energy. This effect is called "photoelectric effect" and the resulting pulses corresponding to the total gamma energy form the so-called "photopeak". In addition to this there will be a number of "Compton" effects. This is an incomplete recovery of the gamma energy by the crystal and results in the appearance of a continuous distribution of smaller energies corresponding to the total energy minus the energy which escaped the crystal. This is called the "Compton continuum." High energy gamma rays striking a detector material can give rise to pair production. This involves the production of a positron-electron pair (energy necessary is $2 mc^2 = 1.02 \text{ Mev}$). The positron is not stable and by the reverse process, positron-electron "annihilation", the same amount of energy is emitted as photons, namely two gamma rays with an energy of 0.511 Mev. If one of these or both escape the detector a peak is recorded with an energy equal to the gamma ray minus 0.511 or 1.022 Mev. They are called double or single escape peaks.

The most important characteristic of the gamma spectrum is the full energy peak resulting from photoelectric absorption of the gamma ray. Thus the detection efficiency for a full energy peak and the ability to resolve it from another peak with a slightly different energy are of prime importance. The energy resolution is defined as the "full width at half maximum". NaI(Tl) detectors have high inherently full energy detection efficiencies and can be obtained in large sizes

but unfortunately the energy resolution is rather poor (7.5% of the 0.662 Mev gamma ray of Cs¹³⁷) and is energy dependent. The Ge(Li) detectors on the other hand have lower intrinsic full energy detection efficiency and cannot yet be obtained in sizes larger than 100 cm³. The detection efficiency of a 30 cm³ Ge(Li) detector may be 7 times lower at 0.2 Mev and 25 times lower at 1.3 Mev than the efficiency of a 3"x3" NaI(Tl) detector, but the energy resolution is much better, especially for high energy gamma rays. If coupled to good electronic apparatus an energy resolution of 2.5 Kev at 1.3 Mev is now easily obtainable. The resolution is only slightly energy dependent. Thus the resolution at 0.2 Mev is 8 times and at 1.3 Mev 25 times better than obtainable with NaI(Tl).

More detailed information about gamma spectrometry can be found in the following works, "Gamma Spectrometry" by Heath, R.L. (10) and "Applied Gamma-ray Spectrometry" by C. Crouthamel, F. Adams, and R. Dams (11).

6. Nondestructive Activation Analysis.

The very high energy resolution obtainable in the Ge(Li) detectors allows the simultaneous specific measurement of the radiation emitted by several isotopes. Thus it becomes possible to distinguish and determine isotopes in complex mixtures without any chemical separation. The resultant instrumental or nondestructive activation analysis is attractive for different reasons.

1) The sample remains essentially unchanged, and can be used for subsequent investigations after the decay of the induced activity. It also allows inter-laboratory comparison of results on the same specimen.

2) Time is gained. This allows the analysis of large numbers of samples, which is very important in environmental research in order to draw valid conclusions.

3) The instrumental approach allows the development of completely automated systems using computer handling of data. The analysis can be performed by an essentially unskilled operator.

4) Isotopes with very short half-lives can be determined, whereas during chemical separations many short-lived isotopes decay essentially completely before the count is begun.

II. Procedure for Non-Destructive Neutron Activation Analysis of Environmental Samples.

Experimental

1. Apparatus used.

Nuclear Reactor: The irradiations are performed in the Ford Nuclear Reactor of the University of Michigan, operating at 2 Megawatts. The reactor is a swimming pool type, where the water acts as coolant, moderator and reflector. Short-lived isotopes are produced sufficiently by short irradiations (less than 30 minutes) in the pneumatic tube facility (neutron-flux $2 \times 10^{12} \text{ n/cm}^2 \text{ - sec}$). The transit time of the sample is approximately 3 seconds and the sample holder is a special nylon tube (Rabbit). Longer lived isotopes are produced by longer irradiations (more than 1 hour) in a facility (H-5) in the immediate neighborhood of the core (neutron-flux $1 \times 10^{13} \text{ n/cm}^2 \text{ - sec}$). The samples are packed in a polyethylene bottle and are lowered in the pool by means of aluminum wire. Holes drilled in the side of the bottle provide sufficient cooling of the contents.

Counting Equipment: The samples are counted by high resolution gamma-ray spectrometry. The gamma spectrometer consists of,

Detector: Nuclear Diodes - True coaxial germanium-lithium drifted diode. It has a capacity of 27.5 pf. The detector is positioned in a vacuum chamber at 12 mm distance from a 0.5 mm aluminum window. Its performances are: a full width at half maximum (F.W.H.M.) of 2.5 Kev for the gamma transition of Co^{60} at 1332.3 Kev, and a detection efficiency for this photpeak of 4% relative to a 3"x3" NaI(Tl) scintillation detector. This results in a peak to Compton ratio of 18.

Preamplifier: Nuclear Diodes-Model 101 is fixed to the detector chamber.

Linear Amplifier: Tenelec Model 202 - an inverted unipolar pulse shape is used. It can be differentiated once or twice.

Multichannel Analyzer: Nuclear Data 2200 Series - 4096 channel analyzer consisting of: Master Control; Analog to Digital Converter; Data reduce/integrate; Read in/out display; Memory Scaler; Memory Drivers; Memory array; Preset Counter; Magnetic

Tape Read; Magnetic Tape Control

Oscilloscope: Hewlett-Packard

Printer: Monroe-Data/Log - Model MC-13-80. It prints 20 channels per second.

Magnetic Tape Recorder: Kennedy 1400R - Incremental Magnetic Recorder 200 BPI.

It is adapted for use with Nuclear Data 2200 Series.

Counting Room: The equipment is used in a temperature and humidity controlled room.

Computer: IBM 360/67, with 2 CPU's and other on-line accessories, at the University of Michigan Computing Center.

2. Identification of Isotopes-Qualitative Analysis.

High resolution gamma-ray spectrometry is very valuable for identification of gamma-emitting isotopes. Indeed gamma-ray energies can be determined very precisely if proper care is taken to avoid instabilities and deviation from linearity of the analysing and amplifying equipment. Comparison of the gamma-ray energies with a compilation of energies of radionuclides formed by neutron irradiation (12, 13) generally allows an immediate identification of the gamma emitting isotopes present without further investigation of other decay properties, or radiochemical behavior. Because the production reaction(s) of these isotopes in a reactor are also given in these compilations, the identity of the original target atom is usually very obvious.

The spectrometer can be energy calibrated using a set of radionuclides with precisely known gamma-ray energies. Table 2 lists a number of calibration lines. For most applications a gain setting of 1 Kev/channel is very appropriate. Figure 1 shows the deviation from this setting for the actually used gamma-ray spectrometer in an energy range from 55 up to 2800 Kev, a range which covers more than 95% the gamma rays emitted by reactor produced isotopes. By visual inspection of the oscilloscope display the location of sufficiently intense peaks can easily be estimated with an accuracy of better than 0.1 F.W.H.M., which is here equivalent to approximately 0.25 Kev.

<u>Isotope</u>	<u>Energy (Kev)</u>
²⁴¹ Am	59.568 $\pm$ 0.017
¹³¹ I	80.166 $\pm$ 0.009
¹⁵³ Gd	97.43 $\pm$ 0.02
¹⁵³ Gd	103.18 $\pm$ 0.02
¹⁷⁷ Lu	112.97
¹⁴¹ Ce	145.44 $\pm$ 0.05
¹³⁹ Ce	165.84 $\pm$ 0.03
¹⁷⁷ Lu	208.36
²⁰³ Hg	279.12 $\pm$ 0.05
¹³¹ I	364.47 $\pm$ 0.005
¹⁹⁸ Au	411.776 $\pm$ 0.01
Annihilation	511.006 $\pm$ 0.02
²⁰⁷ Bi	569.65 $\pm$ 0.10
¹³⁷ Cs	661.59 $\pm$ 0.07
⁹⁵ Nb	765.83 $\pm$ 0.07
⁵⁴ Mn	834.84 $\pm$ 0.07
⁸⁸ Y	898.01 $\pm$ 0.07
²⁰⁷ Bi	1063.82 $\pm$ 0.28
⁶⁰ Co	1173.13 $\pm$ 0.04
⁶⁰ Co	1332.39 $\pm$ 0.05
²⁴ Na	1368.40 $\pm$ 0.04
²⁰⁸ Tl(ThC'') (D.E.)	1592.3 $\pm$ 0.13
²⁴ Na(D.E.)	1731.6 $\pm$ 0.16
⁸⁸ Y	1836.1 $\pm$ 0.07
²⁴ Na(S.E.)	2242.6 $\pm$ 0.14
²⁰⁸ Tl(ThC'')	2614.3 $\pm$ 0.09
²⁴ Na	2753.6 $\pm$ 0.12

Table 2. Energy Calibration Lines

From comparison with a calibration curve (Figure 1) an energy determination is immediately obtained with an accuracy of mostly better than 0.5 Kev. For more accurate energy determinations or for routine identification purposes a computer assisted qualitative analysis is performed. From a large number of calibration points a relation between gamma-ray energy and peak location is obtained and approximated by a nth degree polynomial. The degree of the polynomial and its

coefficients are calculated by a least squares fitting computer routine. Small differences in gain and zero threshold are corrected by recalibration with a high energy and a low energy gamma ray. The identification is then performed by comparing the experimental energies of the peaks of a complex gamma-ray spectrum with the gamma-ray energies of a library present in the memory of the computer. This library contains 2000 gamma-radiation energies of about 250 reactor produced isotopes (12). For the identification, also the half-life of the isotope is compared to the cooling time (time between end of irradiation and time of counting) of the sample.

Chapter 3 about computer handling gives more detailed information about this qualitative analysis.

3. Quantitative Analysis of Germanium Lithium Gamma-Ray Spectra.

The quantitative determination of the count rate due to a certain isotope from the gamma-ray spectrum involves the measurement of the area included under its full energy peak. This determination must not necessarily be quantitative. It is indeed sufficient to measure only a fraction of this area, provided that the same fraction of the peak area in the standard sample is summed. This can easily be carried out as follows: (see Figure 2). If A_0 is the count rate in the channel in which the highest number of counts is recorded, the area P under the peak is given as follows:

$$P = A_0 + \sum_{i=1}^n A_i + \sum_{i=-1}^{-n} A_i$$

However the observed peak will usually be interfered by a contribution due to the Compton continuum of more highly energetic full energy peaks. The area Q due to the Compton continuum under the peak can be approximated as follows:

$$Q = Q' + Q'' = \sum_{i=n+m}^{2n+m} A_i + \sum_{i=-(n+m)}^{-(2n+m+1)} A_i$$

The net peak area N is calculated as,

$$N = P - Q$$

and bears (within the standard deviation of the counts obtained in the channels) a constant relationship to the total area contained in the peak. The method can be optimized in terms of experimental conditions such as the calibration and the resolution of the spectrometer and the values chosen for n and m .

If the gain is set for 1 Kev/channel n is often taken equal to the F.W.H.M. of the peak. m is mostly chosen slightly higher. For the spectrometer used in this work the resolution varies from 2.2 to 3.0 Kev depending on the energy of the peak. During long counting times a small variation in gain, due to instability of the temperature in the counting room, is often experienced, resulting in an increased F.W.H.M. Therefore n can be chosen 2, 3 or 4 and for m a value of 4 is mostly appropriate. The choice of n , depends on the stability of the gain, the complexity of the spectrum, the counting statistics and the energy of the peak.

The precision of the determination of the peak area can be predicted if the peak is free of important interferences due to other peaks. The standard derivation is given as

$$\sigma_N^2 = \sigma_P^2 + \sigma_Q^2$$

$$\sigma_N = \sqrt{P + Q}$$

This method can however not always be applied:

1) Sometimes another photopeak is present immediately above or below the photopeak to be measured, interfering with the calculation of Q' or Q'' . The Compton contribution is then calculated at another energy region which is free of interferences.

2) If the photopeak itself is interfered by the presence of another photopeak it is advisable to determine the abundance of the nuclide from another photopeak. If this is not possible the interference may be reduced by decreasing n or the peak area may be calculated by doubling the area under the less interfered half of the photopeak. As a matter of fact the accuracy of such a determination is limited.

Because this nondestructive activation analysis procedure should be applied for large number of routine measurements, some sort of automatic data reduction is often necessary. Simple as these procedures for determination of count-rates under peaks may be, some sort of automation is sometimes desirable. Accuracy may be improved by the elimination of human errors in the transcription of data and in the operation of a desk calculator. However human judgment is to be maintained by incorporating into the calculation procedures suitable criteria for checking data.

A large number of methods and computer programs have been developed for the analysis of gamma-ray spectra obtained either with NaI(Tl) scintillation detectors or with Ge(Li) semiconductor detectors, but most of these programs involve thorough inspection and very complicated handling of data. Such a program is very expensive and thus less suitable for analysis of a very large number of quite similar samples. The next chapter describes the program for automatic data reduction applied in this work.

4. Irradiation-Counting Scheme.

High resolution gamma-ray spectrometry using germanium lithium drifted detectors allows the simultaneous measurement of a large number of isotopes. The ideal irradiation, cooling and counting times for the production and detection of an isotope are $t_{irr} \approx t_{1/2}$, $t_{cool} \leq t_{1/2}$ and $t_{count} \approx 2 t_{1/2}$. When a large number of elements have to be determined simultaneously by activation analysis, the irradiation-counting conditions can of course not be ideal for each isotope. Considering the elemental composition of typical environmental samples, the sensitivities for the determination of the elements present and the half-lives of the isotopes produced, a scheme, involving several irradiations and countings which allows the determination of a maximum number of elements was devised.

a) Analysis of elements giving rise to short-lived isotopes: Preliminary short neutron irradiations of some environmental samples and several countings soon after the end of irradiation showed the presence of several short-lived isotopes

such as ^{28}Al , ^{52}V , ^{49}Ca , ^{66}Cu , ^{37}S , ^{51}Ti , ^{38}Cl , ^{80}Br , ^{27}Mg , ^{56}Mn , $^{116\text{m}}\text{In}$, and ^{24}Na . Table 3 shows the half lives and most prominent gamma-rays, for the analysis of the elements giving rise to these isotopes. The following irradiation-counting scheme was developed. Considering the half-lives of the isotopes produced it appeared appropriate to perform only a short irradiation and to start counting soon after the end of irradiation. A five-minute irradiation performed in the pneumatic tube from which a fast return of the sample is ensured. For such short irradiation at a moderate neutron flux ($2 \times 10^{12} \text{ n/cm}^2\text{-sec}$), the sample can easily be packed in a poethylene vial with a regular cap or with a snap top. Even small amounts of liquid can be irradiated under these conditions. The vial containing the sample is placed in a special irradiation container (rabbit) and the experimenter himself can transfer his sample to the irradiation site through a pneumatic conveyor. Automatic or manual return after a predetermined time can be applied. Because the sample holder also becomes radioactive the sample is transferred to another vial before counting. If the sample contains an appreciable amount of air it is advisable to ventilate it before counting because neutron irradiation of the argon present in the air (0.9%) gives rise to the production of the isotope ^{41}Ar ($t_{1/2} = 110 \text{ min}$). For the same reason the irradiation container must be opened in a well ventilated hood. Three minutes after the end of irradiation a 400 second count is started. The bulk of the activity is then due to very short-lived isotopes such as ^{28}Al , ^{52}V , ^{49}Ca , ^{51}Ti , ^{66}Cu and ^{37}S . When these activities have died down to a lower level a second count is started for 1000 seconds at 15 minutes after the end of irradiation. This counting is favorable for the measurement of the isotopes ^{38}Cl , ^{27}Mg , ^{80}Br , $^{116\text{m}}\text{In}$, ^{56}Mn , and ^{24}Na . Typical gamma spectra obtained by counting an environmental sample after 3 minutes and 15 minutes cooling are shown in Figure 4-5. The fixed irradiation-counting scheme is also illustrated in Figure 3.

Element	Isotope Produced	Half-life	Most prominent γ rays Kev
Al	^{28}Al	2.30 min	<u>1778.9</u>
V	^{52}V	3.77 min	<u>1434.4</u>
Cu	^{66}Cu	5.1 min	<u>1039.0</u>
Mg	^{27}Mg	9.5 min	844.0; <u>1014.1</u> ;
Ca	^{49}Ca	8.8 min	<u>3083</u> ; 4071;
Na	^{24}Na	15.0 hr	<u>1368.4</u> ; <u>2753.6</u> ;
Mn	^{56}Mn	2.58 hr	<u>846.9</u> ; <u>1810.7</u> ; 2112.8; 2522; 2657; 2962; 3371;
In	$^{116\text{m}}\text{In}$	54.0 min	138.4; <u>417.0</u> ; 818.8; <u>1097.1</u> ; 1293.4
Cl	^{38}Cl	37.3 min	<u>1642.0</u> ; <u>2166.8</u> ;
Br	^{80}Br	18.0 min	<u>617.0</u> ; 640.3; 665.7; 704.3; 1256.7;
I	^{128}I	25.0 min	<u>442.7</u> ; 526.3; 743.5;
Ti	^{51}Ti	5.81 min	<u>320.0</u> ; 608.4; 928.5;
S	^{37}S	5.1 min	<u>3102.4</u>

Table 3. Nuclear properties of short-lived isotopes. The gamma-rays used in the determination are underlined.

Standards: In order to perform a quantitative analysis it is necessary to relate the count-rate of each isotope to the amount of the target element present. Therefore it is appropriate to prepare a number of standard samples containing well known amounts of each element. These amounts have to be balanced carefully to obtain a reasonable activity of all the respective isotopes measured.

Aqueous solutions were prepared from analytical grade salts of the elements to be determined. Table 4 shows the concentrations of the elements in these solutions. The solutions containing the metals consist of diluted nitric acid. The halogens on the other hand are present in diluted ammoniacal medium. From these solutions more diluted mixtures of the elements are prepared. Three mixtures respectively containing Al, V, Cu, Mg, Na, Ca, In, and Mn for solution 1; Cl, Br and I for solution 2; and a separate solution 3 for titanium and manganese because the titanium solution contains an unknown amount of chloride and because the large amount of magnesium present in solution 1 interferes with the determination of manganese. Because these three solutions are very diluted with respect to several

elements it is advisable to prepare new solutions each 3 months. Amounts ranging from 50 to 200 μ g as shown in Table 4 of these three solutions are spotted on respectively 3 pieces of ashless filter paper (4x5 or 4x2.5 cm). Solutions 1 and 3 are carefully dried under an infrared lamp while spotting. Due to the volatility of the halogens, even in this ammoniacal medium, this is not allowed for solution 2. These standard samples can be used several times provided that sufficient time is elapsed between two irradiations to allow for decay of the isotopes measured. Because sulphur is very insensitive by activation analysis a known amount of $(\text{NH}_4)_2\text{SO}_4$ can be weighed and irradiated. Amounts ranging from 50 to 200 mg are appropriate.

The filter papers containing the standard solutions are then irradiated following the same scheme as the unknown samples. Because some of the elements spotted (Na, Cl, Br, Mn) are also present as impurities in the ashless filter paper, a blank paper has to be irradiated and the amounts found in this paper are summed with the known amounts to obtain the total standard.

Flux monitor: Standard and sample cannot be counted simultaneously thus also the irradiation cannot be performed simultaneously. This assumes that the reactor flux does not vary significantly between the irradiation of standard and sample at the irradiation site. Frequent modifications in the composition of the reactor core makes this assumption however uncertain over a long period of time. Also the reproducibility of the irradiation position is unknown. Therefore a flux monitor is irradiated together with each sample and standard and used to correct for possible flux variations. A small foil of titanium (1 mg) is used as flux monitor and is counted at 13 minutes after the end of the irradiation for 20 seconds. The activity due to the isotope ^{51}Ti ($t_{1/2} = 5.8 \text{ min}$) produced by (n, γ) reaction is calculated and acts as a measure of the flux. This flux monitor is sealed in a polyethylene jacket. Due to the short half life of the isotope ^{51}Ti , the same monitor can be used almost indefinitely. Indeed, if the rate of analysis does not

Solution Number	Element	Concentration $\mu\text{g/ml}$	Volume Spotted λ	Amount Irradiated μg
1	Al	10.0	200	2.00
1	V	1.028	200	0.2056
1	Cu	25.12 ⁵	200	5.025
1	Mg	500.5	200	100.1
1	Ca	499.0	200	99.8
1	Na	51.4	200	10.28
1	Mn	1.214	200	0.2428
1	In	0.076	200	0.0152
2	Cl	264.0	50	13.2
2	Br	12.7 ³	50	0.636
2	I	12.8 ³	50	0.641 ⁵
3	Mn	24.28	100	2.428
3	Ti	415.0	100	41.5
4	S	100 mg (NH ₄) ₂ SO ₄	weighed	24.2 mg S

Table 4. Standard solutions of determination of elements giving rise to short-lived isotopes.

exceed one analysis per 35 minutes more than 98% of the activity has decayed for the next measurement.

Calculation of the Concentrations: The net-peak areas are calculated from the spectra obtained from standards unknown samples and flux monitor. The net activities induced in the standards and samples are divided by the activity in the simultaneously irradiated titanium flux monitor to obtain a relative activity per unit of integrated flux. The weights of the element (added and ashless filter paper blank) as present in the standards are then divided by the relative activities per unit of flux obtained for the respective isotopes. This yields a conversion factor for each element. This conversion factor is a constant and can be used for several months if the counting is performed in the same geometry conditions and if the performance and settings of the counting equipment do not vary appreciably. By multiplying the relative activities per unit of flux obtained from an unknown sample with this conversion factor, the total weight of the element (sample and blank) present in the sample is immediately obtained. By applying the

fixed irradiation scheme no decay corrections are necessary because all the samples are counted at the same time after irradiation as the standards.

The following formula shows this calculation.

$$(m + b)_i = (m + b)_s \cdot \frac{F_s}{A_s} \cdot \frac{A_i}{F_i} = K \cdot \frac{A_i}{F_i}$$

where: $(m + b)_i$ = the weight of the element present in sample i (sample + blank)
 $(m + b)_s$ = the weight of the element present in standard s (spotted + blank)
 A_i = the peak area of the isotope in the sample i
 A_s = the peak area of the isotope in the standard s
 F_i = the peak area of the flux monitor irradiated with the sample i
 F_s = the peak area of the flux monitor irradiated with the standard s
 K = the constant conversion factor for the element

b) Analysis of Elements Giving Rise to Longer-Lived Isotopes: Because a long irradiation favors the production of longer lived isotopes, a sample was irradiated in the pool of the reactor at a high neutron flux ($1.5 \times 10^{13} \text{ n/cm}^2\text{-sec}$) for several hours. At different times after the end of the irradiation gamma spectra of the sample were taken. Up to 1 hour after irradiation the spectrum mostly shows the very short-lived isotopes discussed in the previous section. Between 1 and 10 hours after irradiation the high activity due to ^{56}Mn and ^{24}Na greatly mask the presence of other isotopes. After 20 to 40 hours cooling time a number of photopeaks due to the following isotopes become visible:

^{82}Br , ^{42}K , $^{69\text{m}}\text{Zn}$, ^{64}Cu , ^{122}Sb , ^{140}La , ^{153}Sm , $^{152\text{m}}\text{Eu}$, ^{24}Na , and in some samples ^{76}As , ^{72}Ga , ^{187}W , ^{198}Au

Isotopes with half-lives longer than 10 days only become visible in the gamma-ray spectrum after the complete decay of the isotope ^{82}Br . Bromine is indeed usually abundant in environmental samples, it is very sensitive for activation analyses and the isotope ^{82}Br has a very complicated gamma-ray spectrum. Twenty to thirty days after the end of irradiation the following isotopes can be

detected, ^{51}Cr , ^{46}Sc , ^{60}Co , ^{59}Fe , ^{65}Zn , ^{75}Se , $^{110\text{m}}\text{Ag}$, ^{124}Sb , ^{141}Ce , ^{203}Hg and ^{233}Pa , and in some samples ^{58}Co .

For the quantitative analysis of the elements K, Zn, Br, Cu, Sb, La, Sm, Eu, Ga, W, Au, Cr, Sc, Co, Fe, Se, Ag, Ce, Hg, Th, and Ni a number of environmental samples (up to 15) and a standard containing well-known amounts of these elements are irradiated simultaneously for 2 or 5 hours at a neutron flux of $1.5 \times 10^{13} \text{ n/cm}^2\text{-sec}$ in the immediate neighborhood of the core of the reactor. The samples and the standard are heat sealed in polyethylene tubes. After irradiation the samples and standard are unpacked and transferred to the bottom of glass vials. After a cooling period of 20 to 30 hours the samples are counted for 2000 seconds for the measurement of the isotopes with half lives ranging from 8 to 50 hours. The isotopes with half-lives longer than 10 days are measured from a 4000 second count after 20 to 30 days cooling. Because the half-lives of the isotopes to be determined are sufficiently long as compared to the counting time a correction for the different decay times of the samples can easily be made. Table 5 shows the elements determined, the isotopes counted, their half lives and the energies of their gamma rays. Figures 7 and 8 show gamma spectra of both countings while the irradiation procedure is illustrated schematically in Figure 6.

Standards: Carefully weighed amounts of analytical grade salts of the elements are dissolved in dilute nitric acid. Well balanced amounts of these solutions are mixed to obtain reasonable activities of all the respective isotopes after irradiation. The solutions of gold contained some chlorine. Therefore it could not be mixed with the other elements (precipitation of silver and mercury). Also the addition of bromine and tungsten would cause evaporation (hydrobromic acid) or precipitation (tungstic acid H_2WO_4). Therefore these three elements are in a separate solution. The contents of these solutions and the concentrations of the elements are given in Table 6. Because the concentrations of several elements in these solutions are relatively low, new solutions should be prepared

Element	Isotope Produced	Half-life	Most Prominent γ -rays (Kev)
Na	^{24}Na	15.0 hr	<u>1368.4</u> ; <u>2753.6</u> ;
K	^{42}K	12.5 hr	<u>1524.7</u> ;
Zn	$^{69\text{m}}\text{Zn}$	13.8 hr	<u>438.7</u> ;
Cu	^{64}Cu	12.8 hr	β^+ (<u>511.0</u>);
Br	^{82}Br	35.9 hr	<u>554.3</u> ; <u>619.0</u> ; 698.3; <u>716.6</u> ; 827.8; <u>1043.9</u> ; <u>1317.2</u> ; <u>1474.7</u> ;
As	^{76}As	26.3 hr	559.2; <u>657.0</u> ; 767.5; <u>1228.8</u> ;
Ga	^{72}Ga	14.3 hr	601.1; 630.1; 786.4; 810.5; <u>834.1</u> ; <u>1800.1</u> ; <u>2201.4</u> ;
La	^{140}La	40.3 hr	<u>328.6</u> ; 486.8; 815.4; <u>1595.4</u> ;
Sm	^{153}Sm	47.1 hr	69.6; <u>103.2</u> ;
Eu	$^{152\text{m}}\text{Eu}$	9.35 hr	121.8; 344.2; 841.6; <u>963.5</u> ; 1315.0; 1388.9;
Sb	^{122}Sb	2.75 da	<u>504.0</u> ; <u>692.5</u> ; 1256.6;
W	^{187}W	24.0 hr	72.3; 134.3; 479.3; 551.4; 618.1; <u>685.7</u> ; 772.9;
Au	^{198}Au	2.70 da	<u>411.8</u> ;
Sc	^{46}Sc	83.9 da	<u>889.4</u> ; <u>1120.3</u> ;
Cr	^{51}Cr	27.8 da	<u>320.0</u> ;
Co	^{60}Co	5.24 yr	<u>1173.1</u> ; <u>1832.4</u> ;
Fe	^{59}Fe	45.1 da	<u>1098.6</u> ; <u>1291.5</u> ; 192.5; 143.0;
Ni	^{58}Co	71.3 da	<u>810.3</u> ; 863.5; β^+ (511.0)
Zn	^{65}Zn	245.0 da	<u>1115.4</u> ; β^+ (511.0);
Se	^{75}Se	121.0 da	96.7; 121.8; 136.0; <u>264.6</u> ; 279.6; <u>400.7</u> ;
Ag	$^{110\text{m}}\text{Ag}$	260.0 da	657.8; 706.4; 763.9; 884.5; <u>937.2</u> ; <u>1384.0</u> ;
Sb	^{124}Sb	60.9 da	<u>602.6</u> ; 645.7; 722.8; 1436.8; <u>1690.7</u> ; 2090.6
Ce	^{141}Ce	32.5 da	<u>145.4</u> ;
Hg	^{203}Hg	46.9 da	<u>279.1</u> ;
Th	^{233}Pa	27.0 da	<u>299.9</u> ; <u>311.8</u> ; 340.3;

Table 5. Nuclear properties of long-lived isotopes. The gamma rays used in the determinations are underlined.

Solution Number	Element	Concentration $\mu\text{g/ml}$
5	Zn	101.1
5	K	101.3
5	Cu	10.0
5	As	2.20
5	Ga	0.80
5	Sb	4.24
5	La	0.421
5	Sm	0.109
5	Eu	0.0216
5	Sc	0.826
5	Cr	42.1
5	Co	3.87
5	Fe	3,000.0
5	Ni	500.0
5	Se	3.53
5	Ce	10.0
5	Hg	10.3
5	Th	0.80
5	Ag	4.05
6	Br	4.71
6	W	0.37
6	Au	0.140

Table 6. Standard solutions of elements giving rise to long-lived elements.

each 3 months. 50 to 200 λ of this solution, depending on the irradiation time are spotted on a 4 x 2.5 cm or a 4 x 5 cm ashless filter paper. The amounts of the elements already present in the filter paper have to be summed with the amounts spotted to obtain the total standard concentration.

Calculation of the concentrations: After determination of the net peak areas and correction for decay the count rates of the samples are compared to the standard count rates. The concentrations of each element are calculated as follows.

$$(m + b)_i = (m + b)_s \frac{A_i}{A_s} \frac{\exp(+\lambda t_i)}{\exp(+\lambda t_s)} = (m + b)_s \frac{A_i}{A_s} \exp [\lambda (t_i - t_s)]$$

where: $(m + b)_i$ = the weight of the element present in sample i (sample + blank)

$(m + b)_s$ = the weight of the element present in standard s (spotted + blank)

A_i = the peak area of the isotope in sample i

- A_s = the peak area of the isotope in sample s
 λ = the decay constant of the element
 t_i = the cooling time of the sample i
 t_s = the cooling time of the standard s

5. Sensitivity-Detection Limits

In the irradiation counting system adopted a first detection limit is defined as the amount necessary to record at least 10 counts ($\sigma = 35\%$) under the photopeak or a number of counts equal to the background of the detector in that energy region. Practical detection limits are however mostly higher due to mutual interference of the isotopes. Indeed the sensitivity to measure a photopeak of an isotope is decreased when the peak is sitting on the Compton continuum of higher energetic gamma rays due to other isotopes. Thus a second detection limit may be defined which is defined as the amount of the element necessary to obtain at least a net photopeak activity equal to the standard deviation on its calculation. Thus if the net activity is given as $N = P - Q$, the detection limit is defined as $\sqrt{P - Q}$.

Column 5 on Tables 7 and 8 shows detection limits for typical air samples. As a matter of fact these values depend largely on the composition of the sample. For example a very high concentration of aluminum or chlorine may mask the presence of other short-lived isotopes. Large concentrations of sodium or bromine may interfere with the measurement of the isotopes with half-lives ranging from 8 to 50 hours. This results in an important decrease of the sensitivity.

The sensitivity of the analysis of an element in an environmental sample is further limited by the blanks. Indeed, abundant elements such as Cl, Br, Na, K, Zn, Fe, Mn, Al, Cu, Sb, Cr, etc. are liable to be present in the filter material, holder, or solvent in which the sample is collected.

In the present investigation an element is considered as being present if the net concentration exceeds twice the standard deviation of the blank value.

Element	$t_{irr.}$ flux	t_{count}	t_{decay}	Detection limit, ng, for typical air filter samples
S	5 min $2 \times 10^{12} n/cm^2\text{-sec}$	400 sec	3 min	25,000
Ca	"	"	"	1,000
Al	"	"	"	40
V	"	"	"	1
Cu	"	"	"	100
Ti	"	"	"	200
Br	"	1000 sec	15 min	20
In	"	"	"	0.2
Mn	"	"	"	3
Mg	"	"	"	3,000
Na	"	"	"	200
Cl	"	"	"	500
I	"	"	"	---

Table 7. Sensitivity of 13 elements by short irradiation in adopted irradiation-counting scheme.

Element	t_{irr} flux	t_{count}	t_{decay}	Detection limit, ng, for typical air filter samples
K	5 hour $1.5 \times 10^{13} n/cm^2\text{-sec}$	2000 sec	20-30 hours	75
Zn	"	"	"	200
Cu	"	"	"	50
Br	"	"	"	25
As	"	"	"	40
Ga	"	"	"	10
La	"	"	"	2
Sm	"	"	"	0.05
Eu	"	"	"	0.1
Sb	"	"	"	30
W	"	"	"	5
Au	"	"	"	1
Sc	"	4000 sec	20-30 days	0.3
Cr	"	"	"	20
Co	"	"	"	2
Fe	"	"	"	1,500
Ni	"	"	"	1,500
Zn	"	"	"	100
Se	"	"	"	10
Ag	"	"	"	10
Sb	"	"	"	8
Ce	"	"	"	20
Hg	"	"	"	10
Th	"	"	"	3

Table 8. Sensitivity of 23 elements by long irradiations in adopted counting scheme.

This standard deviation is determined by the accuracy of the blank determination and by the homogeneity of the blank material.

6. Possible Interferences

Besides the thermal neutrons there are always some fast or fission neutrons present at each irradiation site of a reactor. These high energy neutrons induce threshold reactions after collisions with atoms. Such threshold reactions are (n,p) , (n,α) , and $(n, 2n)$ reactions. Some determinations based upon an (n,γ) reaction induced by thermal neutrons may be interfered by such threshold reactions.

For example the determination of element ${}^A_Z M$ by reaction (1) is interfered by reactions (2), (3), and (4) on elements ${}^{A+1}_{Z+1} M$, ${}^{A+4}_{Z+2} M$, and ${}^{A+2}_Z A$.

To estimate the degree of interference it seemed appropriate to determine the ratio of the fast to thermal neutron flux.

Determination of Fission to Thermal Flux Ratio: Determination of the fission flux in a reactor site requires the measurement of the production rate of an isotope by a threshold reaction. Very often the reaction ${}^{32}\text{S}(n,p){}^{32}\text{P}$ is used to determine the value of an undisturbed fission flux. Thus from an amount of sulphur irradiated during a time t_1 and counted at a time t_2 after irradiation the fission flux is calculated as follows:

$$\phi_f = \frac{D_{32S} \exp(+\lambda t_2)}{\sigma_{32S} N_{32S} [1 - \exp(-\lambda t_1)]}$$

where ϕ_f = the mean fission flux in $\text{n/cm}^2\text{-sec}$.

D_{32S} = the absolute disintegration rate of ${}^{32}\text{P}$ produced by (n,p) reaction on sulphur

σ_{32S} = the mean fission cross section for the reaction ${}^{32}\text{S}(n,p){}^{32}\text{P}$

N_{32S} = the number of atoms ${}^{32}\text{S}$ present

λ = the decay constant of the isotope ^{32}P

This requires however an absolute measurement of the ^{32}P disintegration rate.

The interference due to a threshold reaction does not depend on the absolute value of the fission flux but only on its ratio to the thermal flux. Thus it is sufficient to compare the count rate of ^{32}P produced by the threshold reaction to the count rate of ^{32}P produced by the thermal reaction $^{31}\text{P}(n, \gamma)^{32}\text{P}$. This requires now only a relative measurement of ^{32}P . Thus from a simultaneously irradiated and counted amount of phosphorus the thermal neutron flux is given as:

$$\phi_{\text{th}} = \frac{D_{31\text{P}} \exp(+\lambda t_2)}{\sigma_{32\text{P}} \cdot N_{32\text{P}} [1 - \exp(-\lambda t_1)]}$$

where $D_{31\text{P}}$ = the absolute disintegration rate of ^{32}P produced by (n, γ) reaction on phosphorus

$\sigma_{32\text{P}}$ = the thermal neutron cross section for the reaction $^{31}\text{P}(n, \gamma)^{32}\text{P}$

$N_{32\text{P}}$ = the number of atoms ^{31}P present

The ratio of both fluxes equals now

$$\frac{\phi_f}{\phi_{\text{th}}} = \frac{D_{32\text{S}} \sigma_{32\text{P}} N_{32\text{P}}}{D_{31\text{P}} \sigma_{32\text{S}} N_{32\text{S}}} = \frac{\text{Act}_{32\text{S}} \sigma_{32\text{P}} N_{32\text{P}}}{\text{Act}_{32\text{P}} \sigma_{32\text{S}} N_{32\text{S}}}$$

where $\text{Act}_{32\text{S}}$ = the relative count rate of ^{32}P produced by (n, p) reaction on sulphur

$\text{Act}_{32\text{P}}$ = the relative count rate of ^{32}P produced by (n, γ) reaction on phosphorus

Experimental: Simultaneously known amounts of analytical grade $(\text{NH}_4)_2\text{SO}_4$ and $(\text{NH}_4)_3\text{PO}_4$ were irradiated at both irradiation sites used (pneumatic tube and reactor pool). Seven days after irradiation the salts were dissolved in diluted nitric acid and adequate amounts were counted by liquid scintillation using the Čerenkov effect. ^{32}P is a pure beta emitter with a maximum beta energy of 1.71 Mev and a half life of 14.2 days. The interfering activity due to ^{35}S produced by (n, γ) reaction on ^{34}S is a pure beta emitter with a maximum beta energy of only 105 Kev and a half life of 85 days. This energy is too low to generate Čerenkov scintillation. To check the purity of the samples a decay curve analysis was performed for 3 months. For all the samples half-lives ranging from 14.0 to 14.4

days were found and no indication of the presence of shorter or longer lived isotopes was detected. The results shown in Table 9, are calculated using the following generally accepted values for the cross sections (14,15). $^{31}\text{P}(n,p)^{32}\text{P}, \sigma = 0.19 \text{ barn}$
 $^{32}\text{S}(n,\gamma)^{32}\text{P}, \sigma = 0.005 \text{ barn}$

Irradiation Site	Activity	Activity	Ratio Activity(P) Activity(S)	Neutron Flux Ratio Thermal/Fission
	mg P c/min	mg S c/min		
Pneum. Tube Reactor Pool	206,480	8,823	23.40	7.6
	4,060,200	295,354	13.75	4.5

Table 9. Determination of thermal to fission flux ratio.

Calculation of the interferences: Based upon the experimentally found ratio of the fission to thermal neutron-fluxes and on the mean neutron fission cross sections as compiled by Neuerht and Zyp correction factors for possible interferences were calculated (Table 10- column 5). This correction factor equals the weight fraction of the interfering element which has to be subtracted from the element determined. Thus the corrected amount of element M present is given as

$$M = M_E - I_E K_{M.I.}$$

where M_E = the amount of element M experimentally found

I_E = the amount of interfering element I experimentally found

$K_{M.I.}$ = the correction factor for interference of element I on the determination of element M

The concentration ratios of interfering to interfered elements which give rise to an interference of 1% were also calculated. These ratios may be compared to concentration ratios found in actual air samples (Table 10, columns 6 and 7). It is obvious that in these samples a large correction (>5%) can only be expected in the analysis of magnesium, which is interfered by a threshold reaction on aluminum. Therefore this low interference was also determined experimentally by irradiating a solution of pure $\text{Al}(\text{NO}_3)_3$. It appeared that 100 micrograms of aluminum

Element Determined	Interfering Element I	Reaction	Irradiation Site	Correction Factor $K_{M.I.}$	Concentration ratio resulting in 1% interference I/M	Actual Concentration ratios in air I/M
Na	Al	$^{27}\text{Al}(n, \alpha)^{24}\text{Na}$	Pneum Tube	0.00015	65	2-8
Na	Mg	$^{24}\text{Mg}(n, p)^{24}\text{Na}$	"	0.00023	43	1-10
Mg	Al	$^{27}\text{Al}(n, p)^{27}\text{Mg}$	"	0.15	0.067	0.5-5
Mg	Si	$^{30}\text{Si}(n, \alpha)^{27}\text{Mg}$	"	0.00002	490	1-10
Al	Si	$^{28}\text{Si}(n, p)^{28}\text{Al}$	"	0.002	4.9	0.2-4
Al	P	$^{31}\text{P}(n, \alpha)^{28}\text{Al}$	"	0.00055	18	
K	Ca	$^{42}\text{Ca}(n, p)^{42}\text{K}$	React Pool	0.0013	7.6	0.5-5
Sc	Ti	$^{46}\text{Ti}(n, p)^{46}\text{Sc}$	"	0.000008	1,250	150-1500
Cr	Fe	$^{54}\text{Fe}(n, \alpha)^{51}\text{Cr}$	"	0.000014	720	50-500
Mn	Fe	$^{56}\text{Fe}(n, p)^{56}\text{Mn}$	Pneum Tube	0.0000083	1,200	10-50
Mn	Co	$^{59}\text{Co}(n, \alpha)^{56}\text{Mn}$	"	0.0000014	7,200	0.005-0.05
V	Cr	$^{52}\text{Cr}(n, p)^{52}\text{V}$	"	0.00002	510	0.5-5

Table 10. Nuclear interferences due to threshold reactions.

generates a false amount of magnesium equal to 20 ± 8 microgram. This results in a correction factor $K_{\text{Mg-Al}} = 0.20 \pm 0.08$ which is in fairly good agreement with the calculated value of $K_{\text{Mg-Al}} = 0.15$. Magnesium concentrations are therefore corrected with this factor (0.20) in actual analysis.

7. Neutron Flux Mapping

In the pool of the reactor up to 15 samples can be irradiated simultaneously in a polyethylene bottle with a diameter of 4 cm and a height of 8.5 cm. For the calculation of the concentrations it is assumed that all the samples receive the same neutron flux as the standard sample. However in the immediate neighborhood of a finite reactor core an important vertical and horizontal neutron flux gradient may exist. The samples are thus liable to undergo a different flux.

Experiment: An experiment was devised to determine the importance of this effect at the irradiation site used. An aluminum wire was stretched along the walls of the polyethylene bottle. After irradiation it was cut into pieces of 1

to 2 cm. (see Figure 9). These pieces were carefully weighed, counted and the specific activities were related to the neutron flux at the respective sites. The ^{72}Ga activity produced by the reaction $^{71}\text{Ga}(n, \gamma)^{72}\text{Ga}$ on the gallium impurity was counted as a measure of the thermal neutron flux and the ^{24}Na activity produced by the threshold reaction $^{27}\text{Al}(n, \alpha)^{24}\text{Na}$ on the aluminum matrix was counted as a measure of the fission flux. These isotopes were counted by selecting their most prominent photopeaks respectively at 834 and 1368 Kev by means of a single channel analyzer coupled to a NaI detector. The result of one of these experiments is illustrated in Figure 9. As expected the fast neutron flux is more subject to gradients. Indeed a further thermalization sums up with the geometrical effect. The total horizontal gradient (4 cm) appears to be between 6 to 10% for the thermal and 25 to 35% for the fast fission flux. The total vertical gradient (8 cm) on the other hand is between 16 and 21% for the thermal and fission flux.

All the elements, except one (Ni) are determined from mainly thermal neutron produced isotopes. However also the thermal gradients may cause non-neglectible errors. Therefore, only the lower 4 cm of the bottle are filled with one row of vertically stacked samples, with the standard in the middle. This largely reduces errors due to vertical gradients. Errors due to horizontal gradients are minimized by turning the bottle over approximately 180° at half of the irradiation time.

III. Computerized Data Reduction

1. Introduction.

From the foregoing discussion it can be seen that complete analysis of Ge(Li) gamma spectra involves the steps indicated in Fig. 10. Data from the storage volume must be transmitted to the appropriate processing device. In the case of hand analysis this would involve a paper tape listing of the spectrum channel by channel or an oscilloscope display. Paper tape printout of 4096 channels takes minutes even with high speed printout devices, while writing the data on magnetic tape for later processing requires about 30 seconds or less. For computer processing, spectral data stored magnetic tape must be read into fast memory core storage.

Next, all statistically significant peaks in the spectrum must be located. Doublets should be recognized and Compton edges should be discriminated against. While this process of peak recognition may be easily accomplished by eye, it is a non-trivial problem for computer. Various computer techniques have been devised such as the second difference method of Mariscotti,⁽¹⁶⁾ cross-correlation analysis of Black,⁽¹⁷⁾ the tangent method of Gunnink⁽¹⁸⁾ and others. In general, while all methods are successful in locating large isolated peaks, they may fail to work reliably in resolving doublets, discriminating between real peaks and Compton edges and in identifying small peaks which are marginally above the Compton continuum baseline.

A problem of comparable difficulty is that of determining net peak areas. Analysis of peaks which are large in comparison with a relatively flat baseline can be accomplished by summation over a specified interval centered on the peak maximum with subtraction of baseline counts estimated from channels on each side of the peak as mentioned in Chapter 2. In the case of peaks located near Compton edges where the continuum changes rapidly from channel to channel, this procedure can still be used successfully provided a judicious selection of channels is made for the estimate of the continuum under the peak. Difficulties arise in programming

computers to make the appropriate choices. Ralston and Wilcox⁽¹⁹⁾ have developed a procedure for calculating the baseline using an iterative spectrum smoothing routine discussed below. This baseline spectrum is probably a better approximation generally to the Compton continuum than the so-called straight line approach above but is limited in its ability to handle cases of broad peaks located near Compton edges. Additional problems arise in estimating net peak areas from resolved doublets. If the peaks are of comparable height, areas of individual components may be proportioned on the basis of height with limited accuracy. A more satisfactory approach corresponds to the method of Covell⁽²⁰⁾ developed originally for NaI spectra in which peaks are fitted by Gaussian functions to obtain resolved areas. If, however, a small peak resides on the shoulder of a larger one, the calculated net peak area of the smaller member is sensitive to the functional representation of the larger peak in the vicinity of the shoulder. At this laboratory a skewed Gaussian function combined with a linear background term has been used with some success in this type of situation but the increased running time in routine analysis makes it desirable to use alternative isolated peaks whenever possible. Fortunately, in Ge(Li) spectra, overlapping peaks are the exception rather than the rule so that alternative peaks can usually be found.

The correspondence between peak position and gamma energy, typically expressed in terms of an N^{th} order polynomial, enables the computer to locate peak positions if gamma energies are supplied or to identify radioisotopes possibly contributing counts under a given peak by various detection processes. From a knowledge of the experimental conditions it is generally obvious which isotope is primarily responsible for the presence of a given peak, but a thorough search of a gamma energy-isotope library can be helpful in identifying possible interferences which might otherwise be overlooked. In routine analyses of spectra with the same set of peaks recurring, it is of course unnecessary to use a peak identification option with each spectrum.

Net peak areas must be converted to isotope weights by means of appropriate factors which include a correction for the reduction in the number of counts due to the dead time of the multichannel analyzer. Most modern multichannel analyzers have "live time" clocks which correct for instrumental dead time by extending real time counting periods. Gavron⁽²¹⁾ has pointed out that in addition to the fact that these clocks are not always accurate, they can not make the proper dead-time compensation for short lived samples where no simple relation exists between the real time and the fractional dead time. An algorithm has been developed⁽²¹⁾ to handle this problem but it is desirable to keep counting rates sufficiently low that corrections having only approximate validity are unnecessary. In addition to conversion of net peak areas, the computer should make appropriate cooling and counting time decay corrections, flux normalizations, subtractions for interferences as indicated in Ch. 2 and calculate confidence levels in reported values.

It is readily seen that hand extraction of the useful information content of gamma spectra is a tedious and time consuming endeavor subject to a variety of human errors. When spectral data are complex or when a large number of spectra must be analyzed, as in the case of environmental sampling, it is especially desirable that data reduction be accomplished efficiently via computer processing with minimal human intervention. Because of the complexity of gamma spectra, computer programs have not yet been successful in taking over the entire job of data reduction, but the greatest share of the work can in fact be done automatically.

Numerous computer-based spectrum analysis methods are reported in the literature, especially for use with NaI spectra. The FORTRAN IV programs developed here for analysis of Ge(Li) spectra obtained from radionuclide mixtures present in suites of irradiated environmental samples, provide a broad data reduction capability. The operations outlined in Fig. 10 are written in terms of subroutines which may be incorporated into main programs meeting the specialized requirements of the investigator.

In the following section, details connected with storage of spectral data on magnetic tape and its subsequent retrieval by the computer are considered. Sufficient procedural information is presented to make this section hopefully a useful guide on how to get data from the analyzer into the University of Michigan IBM 360/67 computer. In section 3 the available subprograms and short main programs are described while in the final section the main program in current use with the analytical procedure outlined in Chapter 2 is described in detail.

2. Tape analysis:

A. Tape structure. Each spectrum is written on tape either as a set of $N/256$ records each of which is 258×6 BDC characters long or as one continuous record $(N+2) \times 6$ characters long where $N=1024, 2048, 4096$ channels. The writing mode is determined by a switch labeled (IRG/256) in the back of the ND 2200 series magnetic tape module. Existing programs expect the tape to be formatted in the latter way as one large record per spectrum. In this format the tagword is written twice at the beginning of the long record, while in the blocked format it is written twice at the beginning of each segment of 256 channels as shown in the ND 2200 series magnetic tape manual. For each channel, counts are represented on tape by a string of six BCD characters. Since tagword values have at most four digits, two of the six characters for the tagword contain no useful information.

B. Tape submission. Tapes should be submitted at the batch input window at the University of Michigan Computing Center, North University Building, Main Campus. Two labels are affixed physically to each reel submitted: (1) Computing Center ID (CCID) of the form Gxxx (e.g. G123) and (2) users tape ID (UTID) concocted for esoteric reasons by the user (e.g. SOURCE). In exchange for a tape the user receives a green card receipt having the appropriate Gxxx punched in the first columns. So as not to forget the association between the CCID and UTID it is desirable to write the UTID on the receipt card. Tapes so entered in the C.C. users library may be retrieved at the output window at a later time in exchange for the green card. Tapes may stay in the library so long as they are used at

least once a month. According to Computing Center personnel, tapes may not be retrievable if they are not used for several months or if the green card is lost! The UTID may be permanently associated with the tape while a new CCID will be assigned if the tape is withdrawn and subsequently resubmitted.

The green card itself is never used as part of a series of program input cards. The information about what tape to use is supplied on a \$RUN *MOUNT command card described below. For the protection of the user, the CCID and UTID are intercompared before the tape is mounted by the operator. If the association is incorrect, the tape will not be mounted.

C. Tape Inventory. Provided there are no parity or IRG "noise" errors on a tape, the string of BCD characters representing a spectrum can be translated into bytes and correctly grouped into integers giving counts per channel by the usual formatted FORTRAN read statement. Often the original tape has errors and very short records sometimes arising from user intervention in the write operation at the wrong moment. The simple read statement will not handle such troubles for, upon detection of errors, control is passed back to MTS.

A possible solution is to prepare an error-free tape using the MTS routine *TAPECOPY (Computing Center memo M 102) which can copy all records including those with parity errors. For faulty records the parity is re-calculated so as to be correct on the new tape. This program will not give information about the tagword and record length associated with each record.

The FORTRAN tape inventory program INVEN (p. 94) does have this capability but will reject any record which is not of correct parity. In fact the program as presently constituted will transcribe only records which are of correct parity, are "noise" free and of correct length (i.e. a multiple of 1024 channels). These more stringent requirements are adopted because it is not always clear how the record is in error if the parity is wrong or if the record is of improper size. An important advantage of having a tape inventory, is that it allows one to search

for a desired record on the basis of its position among many records rather than on the basis of its tagword. Even though it is desirable and customary to write record sequences with automatic tagword incrementing, it sometimes happens because of the human touch, that more than one record will have the same tagword, in which case record order is of clear importance.

The input card sequence for INVEN is shown on page 38.

Card 3: Beginning of MOUNT instructions.

Card 4: Mount tape with CCID G123 on a seven track unit (there are only two at the Computing Center), assign pseudo device name *R* to it and convert (N), 200 char. per inch (2), even parity (E) characters to bytes using a tape buffer storage size of 25,000 characters. The UTID is SOURCE. By default (i.e. omitting the statement ring = out) the tape ring is taken out which means no writing can be done on this tape (which is the original after all!) The Computing Center operator will put in or remove rings as per instruction on the \$RUN *MOUNT card. For additional information on *MOUNT consult Computing Center memo M67, M68 and CC News #145. (22)

Card 5: Same as 3 but one writes on this tape. Hence ring = IN. UTID = CLTP6

Card 6: End of MOUNT instructions.

Card 7: Load and run the compiled main program +MVL in the file INVEN.

FORTTRAN uses integers instead of pseudo device names; hence the correspondence, 2=*R*, 4=*W*, is made at load time. The program reads data from unit 2 and writes on tape mounted on unit 4.

Card 8: Name of the tape, perhaps UTID, written on new tape inventory sheet. (2A4) format.

Card 9: Any additional information to be printed out on the new tape inventory sheet (20A4 format).

Card 10: N, NSK in 2I4 format.

The inventory begins after NSK records on the source tape have been skipped

and continues transcribing through record with tagword N before quitting. In the automatic incrementing mode this number can usually be equal to the last tagword written on the tape. If automatic incrementing is not used, a dummy spectrum should be written at the very end with N=9999.

Program Notes: Details can be understood by inspection of the FORTRAN listing (p. 94). A unformatted read subroutine is used in which the modifier m is defined (hex: z400000000) so as to transmit error messages to the program rather than to MTS. A detected error will result in repeated read-back-space operations by the tape drive. This behavior is curtailed by invoking a one record SKIP operation (Computing Center memo M80) whenever more than four unacceptable files (too short usually), four end-of-file marks or two permanent errors are detected in succession. This apparently arbitrary approach has greatly decreased the inventory time without impairing the efficiency of record retrieval. The most frequently encountered error is parity, code = 16. In the case of a tape drive failure, code = 24, the inventory will terminate. Experience has shown that if this error occurs, one should merely resubmit the job without changing any cards. Usually the second time around this error, which may arise because of the marginal compatibility of the Kennedy and the 360 tape drives, does not occur. Fortunately the code 24 error rarely occurs.

Having produced a new tape, the program rewinds it and inspects it in essentially the same way for errors. Thus an error-free inventory of this tape indicates that all is well for subsequent data analysis. Cost of the inventory: 10¢/4096 channels.

3. Output: Part of a source inventory is shown on page 40. EOF=End of file, FLN=File number, LEN=number of characters in the record, CHNLS= corresponding number of channels, CMRD=cumulative number of read operations, CRN=index of acceptable records written on new tape. Of the 42 records on this tape only one (Tg=0039) was found to be unacceptable. Thus the inventory of the new tape also

Input cards for Running the
compiled Fortran Program INVEN.

1. \$SIGNON SN87 T=IM 'NAME'
2. PASSWORD
3. \$RUN *MOUNT
4. G123 ON 7TP *R* MODE=2EN SIZE=25000 'SOURCE'
5. G308 ON 7TP *W* MODE=2EN RING=IN SIZE=2500 'CLTP6'
6. \$ENDFILE
7. \$RUN INVEN 2=*R* 4=*W*
8. CLTP6
9. WRITTEN ON 9/26/69
10. 00420000
11. \$SIG

listed on p. 40 displays the tagword and record length for 41 records. All extra end-of-file marks between records have been deleted.

Shown on p. 41 is part of a source inventory for an unusually bad case. The program detected two parity errors in a row after Tg=0041, decided thus to skip one record, immediately found a record far too short followed by another record with incorrect parity. After skipping a region with too many end of file returns, it detected a series of extremely short records which in the case of "Tg"=0213 it attempted to read 3 times before invoking skip. Notice that despite patches of trouble, no useful information was lost. The records were written in the automatic incrementing mode and each tagword is in fact present on the transcribed tape (CRN=14). Occasionally a desired spectrum is lost, as in the previous example (Tg 0039). The ratio of lost to saved spectra is about 1/50. It is desirable to write very important spectra twice to insure their retrievability. The Kennedy tape recorder accepts 1200 foot reels which can store about 100 4096 channel spectra. Thus one reel will take about 50 duplicate spectra of this size.

3. Data Analysis Programs.

Large multipurpose programs are built out of collection of subroutines having specific tasks, thus permitting rapid assembly of programs to fit specialized needs. In this section the subroutines are described as well as several short main programs which make use of them.

A. Tape read and list (RDLST). A set of routines read the tape (either the new tape produced by the inventory program or an error free original tape) and print out the contents of each channel. The main program (p. 95) (RDLST) makes use of five subroutines RDTAPE, MVL, WRTMT, BASE, and WRTBS. As seen from the FORTRAN listing of RDTAPE (p. 95) the tape read format is 3A4, 32(128I6). Reading in the first 12 characters as 4 byte words avoids complications connected with the special encoding of the tagwords. Namely:

Inventory Program Output :

CONTENTS OF TAPE BEING READ

ECF	1	FLN	1	TGW	CC01	LEN	12300	CHNLS	2048	CMRD	3	CRN	1
ECF	3	FLN	2	TGW	CC02	LEN	12300	CHNLS	2048	CMRD	6	CRN	2
ECF	4	FLN	3	TGW	CC03	LEN	12300	CHNLS	2048	CMRD	9	CRN	3
ECF	5	FLN	4	TGW	CC04	LEN	12300	CHNLS	2048	CMRD	12	CRN	4
ECF	6	FLN	5	TGW	CC05	LEN	12300	CHNLS	2048	CMRD	15	CRN	5
ECF	7	FLN	6	TGW	CC06	LEN	12300	CHNLS	2048	CMRD	18	CRN	6
ECF	8	FLN	7	TGW	CC07	LEN	12300	CHNLS	2048	CMRD	21	CRN	7
ECF	9	FLN	8	TGW	CC08	LEN	12300	CHNLS	2048	CMRD	24	CRN	8
ECF	10	FLN	9	TGW	CC09	LEN	12300	CHNLS	2048	CMRD	27	CRN	9
ECF	11	FLN	10	TGW	CC10	LEN	12300	CHNLS	2048	CMRD	30	CRN	10
ECF	12	FLN	11	TGW	CC11	LEN	12300	CHNLS	2048	CMRD	33	CRN	11
ECF	13	FLN	12	TGW	CC12	LEN	12300	CHNLS	2048	CMRD	36	CRN	12
ECF	14	FLN	13	TGW	CC13	LEN	12300	CHNLS	2048	CMRD	39	CRN	13
ECF	15	FLN	14	TGW	CC14	LEN	12300	CHNLS	2048	CMRD	42	CRN	14
ECF	16	FLN	15	TGW	CC15	LEN	12300	CHNLS	2048	CMRD	45	CRN	15
ECF	17	FLN	16	TGW	CC16	LEN	12300	CHNLS	2048	CMRD	48	CRN	16
ECF	18	FLN	17	TGW	CC17	LEN	12300	CHNLS	2048	CMRD	51	CRN	17
ECF	19	FLN	18	TGW	CC18	LEN	12300	CHNLS	2048	CMRD	54	CRN	18
ECF	20	FLN	35	TGW	CC39	LEN	1446	CHNLS	239	CMRD	117	CRN	39
ECF	21	FLN	40	TGW	CC40	LEN	12300	CHNLS	2048	CMRD	120	CRN	40
ECF	22	FLN	41	TGW	CC41	LEN	12300	CHNLS	2048	CMRD	123	CRN	41
ECF	23	FLN	42	TGW	CC42	LEN	12300	CHNLS	2048	CMRD	126	CRN	42

CONTENTS OF NEWLY WRITTEN TAPE CLT.P.6

WRITTEN ON 9/26/69.

FILE	NO.	1	TGNC	0001	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	1
FILE	NO.	2	TGNC	0002	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	2
FILE	NC.	3	TGNC	0003	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	3
FILE	NO.	4	TGND	0004	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	4
FILE	NO.	5	TGNC	0005	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	5
FILE	NO.	6	TGND	0006	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	6
FILE	NO.	7	TGNC	0007	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	7
FILE	NC.	8	TGNC	0008	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	8
FILE	AC.	9	TGND	0009	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	9
FILE	NO.	10	TGNC	0010	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	10
FILE	NO.	11	TGNC	0011	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	11
FILE	NO.	12	TGNC	0012	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	12
FILE	AC.	13	TGNC	0013	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	13
FILE	AC.	14	TGNC	0014	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	14
FILE	NO.	15	TGNC	0015	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	15
FILE	NO.	16	TGND	0016	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	16
FILE	NO.	17	TGNC	0017	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	17
FILE	NO.	18	TGND	0018	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	18
FILE	NC.	19	TGND	0019	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	19
FILE	NO.	20	TGNC	0020	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	20
FILE	NO.	21	TGNC	0021	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	21
FILE	NO.	22	TGNC	0022	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	22
FILE	NO.	23	TGNC	0023	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	23
FILE	NO.	24	TGNC	0024	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	24
FILE	NO.	25	TGNC	0025	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	25
FILE	NO.	26	TGNC	0026	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	26
FILE	NO.	27	TGNC	0027	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	27
FILE	NO.	28	TGNC	0028	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	28
FILE	NO.	29	TGNC	0029	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	29
FILE	NO.	30	TGNC	0030	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	30
FILE	NO.	31	TGNC	0031	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	31
FILE	NO.	32	TGNC	0032	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	32
FILE	NO.	33	TGNC	0033	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	33
FILE	NO.	34	TGNC	0034	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	34
FILE	NC.	35	TGNC	0035	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	35
FILE	NO.	36	TGNC	0036	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	36
FILE	NO.	37	TGNC	0037	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	37
FILE	NC.	38	TGNC	0038	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	38
FILE	NO.	39	TGNC	0039	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	39
FILE	NO.	40	TGNC	0040	LGNTM	12300	CHRR	CHNLS	2048	CUM	RDS	40
FILE	NO.	41	TGNC	0041	LGNTM	12300	CHRR	CH				

A Source Tape Listing of an Unusually Bad Tape :

CONTENTS OF TAPE BEING READ										
EOF 1	FLN 1	TGW 0035	LEN 24588	CHNLS 4096	CMRD 1	CRN 1				
EOF 2	FLN 2	TGW 0036	LEN 24588	CHNLS 4096	CMRD 4	CRN 2				
EOF 3	FLN 3	TGW 0037	LEN 24588	CHNLS 4096	CMRD 7	CRN 3				
EOF 4	FLN 4	TGW 0038	LEN 24588	CHNLS 4096	CMRD 10	CRN 4				
EOF 5	FLN 5	TGW 0039	LEN 24588	CHNLS 4096	CMRD 13	CRN 5				
EOF 6	FLN 6	TGW 0040	LEN 24588	CHNLS 4096	CMRD 17	CRN 6				
EOF 7	FLN 7	TGW 0041	LEN 24588	CHNLS 4096	CMRD 21	CRN 7				
ERR 1 CD 16										
ERR 2 CD 16 SKIP										
EOF 15	FLN 8	TGW 0042	LEN 317	CHNLS 50	CMRD 25					
EOF 16	FLN 9	TGW 0042	LEN 24588	CHNLS 4096	CMRD 27	CRN 8				
EOF 17	FLN 10	TGW 0043	LEN 24588	CHNLS 4096	CMRD 31	CRN 9				
ERR 3 CD 16										
ERR 4 CD 16 SKIP										
EOF 20										
EOF 21										
EOF 22										
EOF 23 SKIP										
EOF 24	FLN 11	TGW 0044	LEN 24588	CHNLS 4096	CMRD 38	CRN 10				
EOF 25	FLN 12	TGW 0045	LEN 24588	CHNLS 4096	CMRD 41	CRN 11				
EOF 26	FLN 13	TGW 0046	LEN 24588	CHNLS 4096	CMRD 43	CRN 12				
EOF 27	FLN 14	TGW 0047	LEN 24588	CHNLS 4096	CMRD 47	CRN 13				
EOF 28	FLN 15	TGW 0002	LEN 36	CHNLS 4	CMRD 48					
EOF 29	FLN 16	TGW 0213	LEN 27	CHNLS 2	CMRD 49					
	FLN 17	TGW 0213	LEN 27	CHNLS 2	CMRD 50					
	FLN 18	TGW 0213	LEN 27	CHNLS 2	CMRD 51					
QUEER FILE SKIP										
EOF 30	FLN 19	TGW 0048	LEN 81	CHNLS 11	CMRD 52					
	FLN 20	TGW 0048	LEN 24588	CHNLS 4096	CMRD 54	CRN 14

(I3 = 1234) thus is the correct tagword in A4 format. The short subroutine MVL (p. 95) converts I3 to I format so that tagword input information from cards is conveniently compared. Subroutine RDTAPE then will read one record of length NR+1 channels. NR (e.g.: 1023, 2047 or 2095) must be supplied by the program while the tagword, I3 (in alphameric mode), the $\left[\begin{smallmatrix} \text{live} \\ \text{actual} \end{smallmatrix} \right]$ time of data accumulation, IT, and the data array IR(J) (J=1,NR) are returned. IT is stored in the "zeroth" channel so that only NR channels contain pulse height data. Subroutine WRTMT (p. 95) will print out the values I3, IT and (IR(J), J=1, NR) obtained previously. BASE and WRTBS are described below.

The input card sequence required to print out portions of five of the forty-one spectra listed in the inventory of CLT6 p. 44 are shown below. Six values entered on one card are needed for each spectrum listed; NTG, NREC, NSK, IS, IE and NBS in 6I4 format. NTG = the tagword sought, NREC = the total number of channels in the spectrum (i.e. 1024, 2048, or 4096), NSK = the number of records to be skipped starting with the present position of the tape, before searching for the target record (i.e. that with the desired tagword). The search operation consists in successive formatted reads and comparison of the NTG with the tagword read, I3. If the tagword sought is less than the tagword read, a mismatch is detected and the program will stop. This feature can easily be changed so that the tape will backspace an appropriate number of records in its searching but this would be desirable only if automatic incrementing has been completely successful. In this connection it is important to note that the operation of skipping records is much faster than the formatted read operation. Hence it is desirable to choose NSK so that the very next record is the target record. This can be

done on the basis of the tape inventory which gives the correspondence between the record index and tagword on the newly written tape. IS and IE are respectively the first and last channels of the spectrum to be printed out. The listing gives 500 channels per page of output. $NBS \leq 0$ for a simple listing. The case for $NBS > 0$ is discussed below.

The first card in the data set, Card 7, causes the last 1048 channels of spectrum with tagword 1 to be printed out. Card 8 causes the first 2000 channels of the spectrum with tagword 7 to be printed. In this case 5 spectra are skipped prior to searching for 7. As mentioned previously if NSK had been taken = 0 the same output would have been obtained but the running time would have been increased because spectra 2 through 6 would have been read in in order to find 7. A portion of the first page of output for a spectrum is shown below. The time is labeled "live-time" by the program in as much as this is the common operational mode of the analyzer but the actual meaning of this number is of course determined by the position of the live-time switch on the analyzer itself. The effects of cards 9 and 10 can be seen by inspection of the inventory on p. 40. A blank card will cause RDLST to stop.

B. Base spectrum generation and peak integration. The subroutine BASE (p.95) provides a measure of the pulse height spectrum baseline on which peaks rest, thus enabling the computer to make a determination of net peak areas. The technique is described in detail elsewhere (Ralston & Wilcox 19) but, briefly, the program generates from the original spectrum, S^0 , with n_i counts in the i th channel, a smoothed spectrum $\bar{S}^0$ in which the number of counts in the i th channel, $\bar{n}_i$, is taken as the average over an interval of width $2l+1$ centered about i .

Thus

$$\bar{n}_i = \frac{1}{2l+1} \sum_{j=i-l}^{j=i+l} n_j$$

Card inputs to RDLST

1. \$SIGNON SN87T=IM
2. PASSWORD
3. \$RUN *MOUNT
4. G308 ON 7TP *R* MODE=2EN RING=OUT SIZE=25,000 'CLTP6'
5. \$ENDFILE
6. \$RUN RDLST 4 = *R*
7. 000120480000100020480000
8. 000720480005000120000000
9. 003820480032050010000000
10. 004020480000000120000000
11. 004220480001070009000030
12. BLANK CARD
13. \$SIG

The First Page of Output from RDLST .

END OF FILE											
END CF FILE											
END OF FILE											
TAGWORD= 0016 LIVE TIME= 1000 SEC											
Start Channel	Data										End Channel
1	1	2	1	1	2	1	1	2	3	8	10
11	18	29	37	49	81	118	144	193	224	314	20
21	403	615	644	598	604	559	604	602	620	568	30
31	555	510	514	553	518	525	522	590	587	620	40
41	505	465	439	437	463	432	408	414	455	454	50
51	450	419	422	436	437	443	444	437	448	435	60
61	410	486	545	633	487	359	364	390	367	412	70
71	331	369	355	377	332	362	403	418	421	376	80
81	324	331	332	320	364	326	295	342	329	315	90
91	317	346	337	318	323	314	319	349	326	346	100
101	314	374	377	355	321	294	327	320	313	342	110
111	328	322	321	341	340	316	335	309	311	323	120
121	298	306	307	355	319	309	309	315	334	299	130
131	339	300	315	286	274	289	299	298	288	297	140
141	301	293	290	298	294	282	286	308	259	290	150
151	324	313	303	283	272	280	285	312	296	266	160
161	252	267	246	279	271	259	280	273	297	318	170
171	273	253	284	263	270	260	263	262	248	270	180
181	251	270	266	264	251	287	240	268	274	247	190
191	244	254	276	270	245	238	237	249	267	255	200
201	249	286	261	285	267	254	250	255	296	220	210
211	257	247	238	231	253	239	259	224	218	233	220
221	229	248	240	255	242	269	221	211	237	232	230
231	230	205	205	222	237	204	210	231	219	234	240
241	213	212	209	206	182	204	217	201	199	200	250
251	202	215	190	210	191	171	174	174	197	194	260
261	199	187	198	166	175	184	175	165	175	187	270
271	191	149	176	176	175	167	173	163	205	181	280
281	185	161	181	181	170	155	171	156	177	175	290
291	184	188	186	165	173	145	161	155	165	179	300
301	187	157	181	165	155	137	176	177	140	174	310
311	156	171	184	154	169	171	169	181	167	137	320
321	170	161	166	168	163	170	157	162	154	144	330
331	181	161	159	157	164	170	156	134	151	140	340
341	155	144	147	142	158	157	152	157	164	151	350
351	152	155	136	159	154	154	138	140	157	129	360
361	154	157	162	134	130	152	153	137	156	142	370
371	127	146	145	135	141	163	138	142	134	127	380
381	147	156	156	143	126	133	135	149	142	161	390
391	123	127	142	139	137	140	132	123	118	135	400
401	112	116	139	152	139	132	131	127	146	150	410
411	129	131	147	136	107	122	126	144	116	128	420
421	105	127	126	128	131	129	151	139	104	129	430
431	133	114	122	126	123	128	125	101	160	162	440
441	150	121	122	110	108	133	124	128	105	93	450
451	123	117	125	125	135	119	133	109	122	125	460
461	122	122	123	131	139	98	116	117	124	108	470
471	130	99	119	117	118	118	119	111	119	135	480
481	110	114	122	130	114	128	113	122	103	115	490
491	134	110	120	141	115	128	100	138	134	121	500
501	128	116	110	160	125	123	123	118	148	170	510
511	225	243	214	143	129	104	118	111	112	113	520
521	109	109	119	111	122	123	125	116	118	117	530

S^0 and $\overline{S^0}$ are compared. Whenever n_i exceeds $\overline{n_i}$ by α standard deviations (i.e. by $\alpha \sqrt{\overline{n_i}}$), n_i is replaced by $\overline{n_i}$ thus generating a new spectrum S^1 .

The above steps of smoothing and replacement are repeated in turn using S^1 in place of S^0 and so on for n cycles with ever-diminishing values of α . ($\alpha_{i+1} < \alpha_i$). The resultant spectrum S^n is taken as the baseline. In subroutine BASE the initial value of α is taken as 9.2, decreasing in ten (i.e. $n=10$) equally spaced steps to 0.2. ℓ is taken to be 30 channels. The choice for ℓ results from a compromise between the use of a small ℓ (~ 10 channels) for which the Compton edges are well-followed and a large value which gives a better representation of the baseline under unusually broad peaks. An example of the effects dependent on the choice of ℓ is shown in Fig. 11. The spectrum was taken at a very high counting rate in order to exaggerate peak widths. With $\ell=30$ channels, the calculated baseline is largely insensitive to peak width variations encountered in actual measurements.

In the plot of a typical Ge(Li) gamma Spectrum (Fig. 12). The smooth curve follows the upper limit of the calculated baseline. Portions of the spectrum which do not exceed $0.2 \overline{n_i}$ are not replaced in forming the baseline. Thus only under the peaks is the baseline smooth. Elsewhere it often has the value of the original data. The first and last ($2\ell+1$) channels (61 in this case) are lost in the analysis. It will be seen that the baseline follows well the curve that would be drawn by eye except in the vicinity of Compton edges. While this program provides a means for doing routine analysis of peaks known to be away from Compton edges, it is the responsibility of the user to insure that this is in fact the case for his particular experimental conditions. Also note that BASE tends to underestimate the "baseline" associated with small peaks on the shoulder of a large one as in the case of peak labeled 10 situated on the shoulder of peak 11.

Arguments of subroutine BASE (p. 95):

L = sum half width = 30

IST = starting channel (usually = 1)

IND = final channel (usually = NR)

IR = data array

BS = returned base spectrum array

The subroutine PKBS listed on p.95, "integrates" the peak simply by summing counts over $2*NFW+1$ channels centered about channel ICH. Base spectrum counts obtained previously are summed over the same interval. The subroutine returns SBS= the net counts =

$$\sum_{J=ICH-NFW}^{J=ICH+NFW} \{ IR(J) - BS(J) \} \equiv N - B$$

the standard deviation, $SD = \sqrt{N + B}$, and the ratio = SD/SBS . In the expression for SD it is assumed that B has the Poisson- statistical uncertainty one would associate with real counts. For real peaks of about 3KeV FWHM, when the energy calibration is 1KeV/channel, the ratio SD/SBS as a function of NFW has a minimum around $NFW=2$. In order to compensate for possible line shape variations, NFW is typically taken = 3 which slightly increases SD in general (a few percent increase). PKBS requires the "peak" channel, ICH, as input. Thus any drift compensation must be done before calling PKBS.

A subroutine WRTBS (p.95) operates like WRTMT but lists the values of the base spectrum for each channel as well as the original data. In the read-list program RDLST, if $NBS > 0$, a base spectrum will be calculated from channel IS, to IE with $L=NBS$ (usually = 30) and WRTBS rather than WRTMT will be called. A sample portion of WRTBS output is shown on (p.49) for the last spectrum on CLTP6. This spectrum is produced from card 11 in the input (p. 44) to RDLST. This print out was originally obtained in order to examine the behavior of the computed baseline in the vicinity of the 846.9 KeV Mn⁵⁶ peak. It will be seen that the actual counts

in a given channel (e.g. the "peak" channel, 842) is the upper number of the pair (2144) while the lower one is the baseline count (261). Arguments of subroutine WRTBS:

IS = starting channel (usually 1)

IE = final channel (usually NR)

BS = previously calculated base spectrum

C. Plotting. A makeshift program has been written to product plots like that shown in Fig. 4. Limited to six consecutive plots, the program requires 5 values for each spectrum: NREC, ITG, NCUT, NSK, NCR.

NREC = no. of channels

ITG = tagword desired

NSK = skip NSK records before looking for ITG

NCR = 0 do not calculate base

>0 calculate base spectrum and plot

NCUT = no. of channels to be plotted

NREC = 0 stop

The channel axis must be ≤ 10 inches. For 4096 channels this gives $FT = .0025$. While the log counts axis can be up to 24 inches. But for a fit on 8 1/2" x 11" paper, assuming 10^4 cts maximum counts in any one channel, $FT = 2$ gives a maximum height or $8" = 2 \log 10^4$.

The plot shown in Fig. 4 was produced from the series of input cards shown on p. 50. For a description of the MTS control cards, obtain the MTS plotting manual⁽²³⁾.

D. Peak search routine.

A peak finding routine, PEAK, (p.97), is available which has been tested and found to work well except in the case of doublets. This subroutine which in turn calls a series of subroutines FKZ, CIJWZ, SFWZ, locates peaks using a second difference operator function described in detail by Mariscotti.⁽¹⁶⁾ A FORTRAN listing of a test main program, PKTST, and the above subroutines is on p. 97. The twenty-five

Partial Output from RDLST with BASE Spectrum Option (NBS=30)

761	279 279	308 281	308 281	291 278	310 281	281 281	268 268	250 250	289 278	265 265	770
771	295 278	307 278	247 247	276 276	267 267	288 279	261 261	252 252	302 278	292 279	780
781	248 248	279 279	279 279	289 280	255 255	301 280	282 282	312 281	279 279	307 281	790
791	328 282	306 282	274 274	285 285	280 280	277 277	314 282	306 283	267 267	280 280	800
801	277 277	274 274	308 283	294 284	293 284	272 272	270 270	256 256	265 265	297 284	810
811	291 283	297 284	285 285	300 283	330 283	401 283	422 283	371 282	302 281	280 280	820
821	305 279	310 279	311 277	300 277	306 276	289 275	308 274	294 273	285 272	326 271	830
831	337 270	337 269	313 269	303 268	296 267	307 266	362 265	435 265	492 264	870 263	840
847 KEV $M_{A^{25}}$											
841	1814 262	2144 261	1543 260	1344 259	1261 257	777 256	390 255	271 253	224 224	222 222	850
851	224 224	236 236	222 222	237 237	242 242	207 207	247 242	220 220	225 225	219 219	860
861	240 240	221 221	229 229	247 234	231 231	249 232	230 230	226 226	214 214	219 219	870
871	239 227	263 228	210 210	224 224	224 224	200 200	230 221	205 205	243 220	206 206	880

Input Card Listing for Plot Program.

```
SSIGNON SN87 T=IM
PASSWORD
$CREATE PLOT SIZE=1000
$RUN *MOUNT
G308 ON 7TP *W* MODE=2EN RING=OUT SIZE=25000 'CLTP6'
$ENDFILE
$RUN *FORTRAN
.
.
.
FORTRAN LISTING P. 96
.
.
.
$ENDFILE
$RUN-LOAD/++*SOURCE*++PLOTSYS 4=*W* 9=PLOT
.
.
.
COMPILED SUBROUTINES (RDTAPE, MVL BASE)
.
.
.
$ENDFILE
4096000818000006000
BLANK CARD
$RUN *CCQUEUE PAR=PLOT
$RUN *PERMIT PAR=PLOT
$SIG
```

labeled lines shown in the plot (Fig. 12) were identified as peaks by the program. It will be seen that even a small peak on the shoulder of a large one is currently detected as in the case of pair 3-4 or 10 and 11. Also Compton edges are recognized and excluded on the basis of behavior of the second difference function. The sensitivity of peak detection is adjustable. In the present case, peaks labeled (A) and (B) were not identified but would have been if the sensitivity had been increased.

The peak finding routines have not as yet been incorporated into composite programs for routine use because in quantitative analysis of environmental samples the same set of peaks generally recur in each spectrum. Further information on this program is available on request.

E. Component Identification. The subroutine IDCOM (p.98) makes use of a list of 243 gamma-emitting nuclides and their energies⁽¹²⁾ to determine which nuclides may contribute by various detection processes to a gamma spectrum peak observed in a channel corresponding to the energy E. If the fwhm of the peak is $2\delta E$, a gamma ray of energy, E_γ , will contribute provided

$$E_\gamma = E \pm \delta E$$

for complete energy absorption in the detector ("main" peaks) or

$$E_\gamma - 511\text{KeV} = E \pm \delta E \text{ when } E > 1022 \text{ KeV}$$

for single-escape peaks or

$$E_\gamma - 1022 \text{ KeV} = E \pm \delta E \quad E > 1022 \text{ KeV}$$

for double-escape peaks, or provided two gammas from the same nuclide sum to the correct energy

$$E_{\gamma 1} + E_{\gamma 2} = E \pm \delta E.$$

The nuclide data is contained in a private file GAMLIB but is available on cards. It includes a complete list of slow-neutron produced (n, γ) nuclides as well as some fast-neutron produced species and background nuclides such as Cs^{137} and K^{41} . Actually the file contains two lists: (1) A main list with nuclides

index, name, half-life (min.) and up to five energies of the most intense gammas emitted (primary gammas) and (2) an auxiliary list containing up to 100 secondary gamma energies. The division into primary and secondary groups is somewhat qualitative following the classification of Adams and Dams. (12)

IDCOM requires a set of measured gamma energies, (ES, in KeV) the total number in the set, IPK, and the time (in minutes, TIME) elapsed between the end of irradiation and the start of counting. The subroutine expects to treat an entire spectrum. Thus if one finds no "main" peaks associated with the primary gammas of a certain nuclide, one certainly should not expect to find any S.E., D.E., sum or secondary peaks from that nuclide in the spectrum. This might not be true if certain peak energies were missing from the set. IDCOM first makes a list of those nuclides for which there is in each case at least one main peak found and for which the elapsed time is less than ten half-lives. Other nuclides are not considered further. For each nuclide on this list the subroutine calculates S.E., D.E. and sum energies from the primary gamma energies, obtains secondary peak energies from the auxiliary GAMLIB list and looks for equalities between these values and the energies $ES \pm \delta E$. Each nuclide name and coded detection process attributed to a given peak is indexed and stored for eventual transmission out of the subroutine. Finally for all j with $ES(j) < 100$ KeV, the subroutine looks for an $ES(j^1)$ satisfying the relation, $\frac{ES(j^1)}{ES(j)} = 1.132$, which holds for the $K\alpha$ and $K\beta$ X-ray pair. If such a pair is found, the Z of the associated atomic nucleus is given by

$$Z = f(E) = 28.553 + 0.9485 E - 0.003085E^2$$

The value of the energy spread, $\delta E \approx \frac{FWHM}{2}$, is obtained from a function subroutine EF(E). IDCOM Arguments: (values returned by IDCOM; not intermediate values.):

$N(j)$ = number of nuclide-process attributes associated with the jth peak of energy $ES(j)$, $NI(K,J)$ = process code associated with the Kth attribute and Jth peak. Thus:

- NI = 1 main peak
- 2 double escape peak
- 3 single escape peak
- 4 sum peak
- 5 secondary peak
- 6 X-ray pair member

IDX(K,J) = Nuclide index $1 \leq \text{IDX} \leq 243$ associated with the K^{th} attribute and J^{th} peak. If NI = 6, IDX is not the isotope index, but rather the Z of the X-ray transition nucleus. $\text{IDX} = 0.5 + f(E)$.

NS(L, Loc) = Nuclide symbol for the L^{th} nuclide on the nuclide list. Two locations required for the symbol. LOC=1,2.

NPP = total number of nuclides in the list.

IP(L) = Nuclide index associated with the L^{th} nuclide. This value is needed for finding the correct secondary gamma energy list.

INDX(L) = Actual number of primary gammas (excluding the 511 KeV gamma) associated with the L^{th} nuclide.

NPKL (L) = Number of primary gammas (excluding 511 KeV) associated with the L^{th} nuclide for which a main peak was present. $\text{NPKL}(L) \leq \text{INDX}(L)$.

ER(L,k) = The primary gamma energies associated with the L^{th} nuclide. $K=1, 5$.

If $N(j) = 0$ the j^{th} peak is said to be "not attributed".

Subroutine WRTID (FORTRAN p.98). This subroutine will output information from IDCOM. It must be called for each peak, of energy ES(j). From a data array IZ, WRTID picks the correct atomic element symbol to go with the calculated Z when NI=6.

Although in typical use, IDCOM and WRTID would be used in connection with the peak finding routines PEAK etc. (p.97) using values of ES obtained from it, a small main program IDTST (p.98) exists which takes data from cards and provides a summary of isotopes possibly present.

PEAK NO. 1	ENERGY 74.1 KEV		PEAK NO. 17	ENERGY 2168.5 KEV	
	13 TB161	MAIN PEAK		3 CL38	MAIN PEAK
	16 2187	MAIN PEAK			
	21 U239	MAIN PEAK			
		X-RAY FPCF FB			
PEAK NO. 2	ENERGY 85.2 KEV		PEAK NO. 18	ENERGY 2242.0 KEV	
	11 EU155	MAIN PEAK		1 NA24	SINGLE ESCAPE PEAK
	14 TR17C	MAIN PEAK			
	20 T2235	MAIN PEAK			
		X-RAY FPCF FB			
PEAK NO. 3	ENERGY 346.0 KEV		PEAK NO. 19	ENERGY 2322.0 KEV	
	9 EU152	MAIN PEAK		5 MN56	SPALL PEAK
	10 EU152P	MAIN PEAK			
	12 G0159	MAIN PEAK			
	15 PF181	MAIN PEAK			
	18 PT197M	MAIN PEAK			
	25 BA141	MAIN PEAK			
	1 NA24	COUPLE ESCAPE PEAK			
PEAK NO. 4	ENERGY 511.0 KEV		PEAK NO. 20	ENERGY 2657.0 KEV	
	8 SE83	MAIN PEAK		5 MN56	SUP PEAK
	17 CS190M	MAIN PEAK		5 MN56	SPALL PEAK
	19 TL202	MAIN PEAK			
	22 RU106	MAIN PEAK			
	16 W107	SMALL PEAK			
PEAK NO. 5	ENERGY 847.0 KEV		PEAK NO. 21	ENERGY 2753.0 KEV	
	5 MN56	MAIN PEAK		1 NA24	MAIN PEAK
	24 I134	MAIN PEAK			
PEAK NO. 6	ENERGY 1144.0 KEV		PEAK NO. 22	ENERGY 2962.0 KEV	
	23 SR92	MAIN PEAK		5 MN56	SPALL PEAK
	3 CL38	COUPLE ESCAPE PEAK			
PEAK NO. 7	ENERGY 1173.1 KEV		PEAK NO. 23	ENERGY 3068.0 KEV	
	6 CO60	MAIN PEAK		3 CL38	SLM PEAK
PEAK NO. 8	ENERGY 1332.4 KEV				
	6 CC6C	MAIN PEAK			
	7 CO60M	MAIN PEAK			
PEAK NO. 9	ENERGY 1368.0 KEV				
	1 NA24	MAIN PEAK			
PEAK NO. 10	ENERGY 1524.7 KEV				
	4 K42	MAIN PEAK			
PEAK NO. 11	ENERGY 1642.0 KEV				
	3 CL38	MAIN PEAK			
PEAK NO. 12	ENERGY 1655.0 KEV				
	3 CL38	SINGLE ESCAPE PEAK			
PEAK NO. 13	ENERGY 1731.0 KEV				
	1 NA24	COUPLE ESCAPE PEAK			
	24 I134	SUP PEAK			
PEAK NO. 14	ENERGY 1778.5 KEV				
	2 AL28	MAIN PEAK			
PEAK NO. 15	ENERGY 1810.0 KEV				
	5 MN56	MAIN PEAK			
PEAK NO. 16	ENERGY 2112.0 KEV				
	5 MN56	MAIN PEAK			

Output from IDCOM Test Program IDTST.

Card input is evident from the FORTRAN listing (p. 98). The first card contains the "cooling" time in minutes. Subsequent cards contain one gamma energy each (in KeV). Up to 100 energies may be entered. The last card in this series should be blank. Sample output is shown on page 54. Each peak is first listed separately with its attributes. Thus peak 3 has seven possibly contributing nuclides. The larger number of attributes at low energies is in part due to the assumption of a constant peak FWHM of $2\delta E = 4\text{KeV}$. A form used in later runs was

$$\delta E = 0.7 + .001E \text{ (KeV)}$$

IDTST provides a summary table in the form of a list of elements found, with the number of main peaks in the library compared with those found in the actual spectrum. The print out also includes the main gamma energies in the library. This table is helpful in determining which elements are really present. Generally speaking if not all main peaks are present for a given isotope its presence may be doubted.

F. Non-linear least squares routine.

Let the set (Y_j, X_j) $j = 1, N$ be N observations on Y and X which are presumed to be related by $Y_j = f(X_j, \beta_1 \dots \beta_m)$ where the β_i 's ($i = 1, M$) are M adjustable parameters. The subroutine NLLS determines a parameter set β_i , starting from initial guesses for β (say β_i^0), which minimizes the residual

$$R \equiv \sum_{j=1}^N W_j (y_j - f(x_j, \beta_1, \dots, \beta_m))^2$$

with respect to each β_i and thus produces a "best" fit according to this least squares criterion. That is, a set β_i is found such that $\frac{\partial R}{\partial \beta_i} = 0$ for all i . W_j is the weight attached to the j^{th} observation. If there is more than one parameter set which minimizes R , the solution obtained by the program will depend upon the starting point, i.e. the guesses for β_i . Moreover the functions must be bounded

in β_1^n in order to insure convergence. Despite such limitations the routine is of great usefulness and power.

Its application to the analysis of Ge(Li) spectra is two-fold.

- 1) Energy Calibration
- 2) Resolution of Doublets

The algorithm by which the minimization is accomplished is described briefly in Reference 24. Since a solution cannot be obtained in closed form generally, the equation is "linearized" by calculating small increments, δ_i , to initial values of β_i^0 , which will tend to minimize R, that is new values of β_i ,

$$\beta_i^1 = \beta_i^0 + \delta_i^0$$

are calculated for which

$$f(X_j, \beta_i^1 \dots) \approx f(X_j, \beta_i^0 \dots) + \sum_{i=1}^M \left. \frac{\partial f}{\partial \beta_i} \right|_{\beta_i = \beta_i^0} \delta_i^0$$

such that

$$\frac{\partial}{\partial \delta_k^0} \sum_{j=1}^N w_j (y_j - f(X_j, \beta_i^0 \dots))^2 - \sum_{i=1}^M \frac{\partial f}{\partial \beta_i} \delta_i^0 = 0$$

A solution of these M linear equations for each δ_i^0 allows one to correct β_i^0 in the direction of minimum R. The new values of β_i , β_i^1 , constitute new guesses for β_i which themselves may be corrected by calculation of a new δ_i^1 . This iterative procedure may be repeated until a desired accuracy in calculating each β_i is achieved. That is, until

$$\left| \frac{\beta_i^n - \beta_i^{n-1}}{\beta_i} \right| \leq \epsilon$$

for all i where ϵ is small.

Equation can be seen to be a matrix equation. Defining $F_j = (Y_j - f(X_j, \beta_1 \dots))$ taking the partial with respect to δ_k , gives,

$$\sum_{j=1}^N w_j (F_j - \sum_{i=1}^M \frac{\partial f}{\partial \beta_i} \delta_i) \frac{\partial f}{\partial \beta_k} = 0$$

which gives

$$Y_k \equiv \sum_{j=1}^N w_j F_j \frac{\partial f}{\partial \beta_k} = \sum_{i=1}^M \sum_{j=1}^N w_j \frac{\partial f}{\partial \beta_i} \frac{\partial f}{\partial \beta_k} \delta_i \equiv A_{ik} \delta_i$$

where A_{ik} is a symmetric matrix with elements given by

$$\sum_{j=1}^N w_j \frac{\partial f}{\partial \beta_i} \frac{\partial f}{\partial \beta_k}$$

symbolically the solution for δ_i is then

$$\delta_i = A_{ik}^{-1} Y_k$$

where A_{ik}^{-1} is the inverse matrix associated with A_{ik} . The standard error associated with each parameter β_i is given by

$$\sigma_{\beta_i} = \sqrt{\frac{1}{N-M} \left(\sum_{j=1}^N w_j F_j^2 \right) |A_{ik}^{-1}|}$$

The arguments of the FORTRAN listing for NLLS are adequately described by the comment cards (p. 99). Matrix inversion is accomplished by the subroutine INV called from the UM function library. The functional relationship $f(X_j, \beta_1 \dots)$ is supplied by the user as the subroutine ARG. The fortran listing of ARG on p. 99 is for a power series,

$$f(X_j, \beta_1 \dots \beta_m) = \beta_1 + \beta_2 X_j + \beta_3 X_j^2 \dots$$

Where U is the value of X_j for a specified J . ARG must be called for each data point. The dummy, $P(1)$, contains the number of parameters = the polynomial order +1. V is the value of $f(X_j, \beta)$ returned. The current values of β are stored as $G(I)$, $I=1, M$, and the derivatives, $\frac{\delta f}{\delta \beta_j}$, are returned by the subroutine as $D(I)$ $I=1, M$. Thus a linear least squares fit would be obtained by using NLLS-ARG with $P(1) = 2$.

Since the correspondence between the energy of a detected gamma ray and the position of its peak in the analyzer spectrum is very nearly linear for the Nuclear Data 2200 unit, the addition of a small quadratic "correction" term suffices for most analytic work undertaken on this instrument. Thus a value of $P(1) = 3$ is customarily used but the capability exists of making higher-order fits for gamma energy determinations. Experience has shown that with reasonable initial guesses (within 20% of correct values) the quadratic fit using up to perhaps 10 lines, converges in 2 or 3 iterations with $\epsilon = 0.1\%$.

For purposes of energy calibration less powerful techniques of course, may suffice but, in addition, NLLS provides a means of analyzing doublets. Given the functional form, Gaussian or skewed Gaussian, of each of two peaks which are superposed NLLS can estimate their contributions under a broader range of conditions than may be done well by eye. Some experience has been gained in application of this program but its need in analysing typically encountered samples is as yet not great.

4. Composite Program

The program SPAN (FORTRAN p.100) is designed to obtain weights of elements present in samples deposited on various substrates and irradiated according to the analytical scheme discussed in Chapter 2. For each of the four combinations of irradiation and counting modes, the program is supplied with a list of gamma energies for which it estimates the position of corresponding peaks in the analyzer spectrum at hand and calculates net peak areas. The general features of the program sequence can be seen from the abbreviated flow diagram (Fig. 13.) A sample input card set is shown on p. 61. Data on these cards were developed and actually used in analysis of air particulate samples. Up to 50 gamma energies may be contained in each of the four blocks of isotope data accepted by the program. Two cards are required for each gamma energy entered. In the program the four run modes are indexed as follows:

ITY	Irradiation	Counting Live Time
1	5 minutes	400 sec
2	"	1000
3	5 hours	2000
4	"	4000

For the short irradiations (ITY=1,2) the isotope cards have the following format:

CARD	1	NS	E	SM	FK1	DF1	FK2	DF2
		2A4	2X	F10.5	F10.5	F10.5	F10.5	F10.5
	2	POL	DP	SOR	DS	POR	DR	
		F10.5	F10.5	F10.5	F10.5	F10.5	F10.5	

NS = the isotope symbol (e.g. CA49) (ITY=1)

E = gamma energy (KEV) (e.g. 3083.0 KEV)

SM = weight of element (i.e. isotopic mixture not the individual radionuclide) in the standard. (Arbitrary units e.g. 99.8 µg)

FK1 = the factor converting net peak areas (counts) to element weight in units of SM based on some standard flux measure. (Say unit: 0.211 µg/count)

DF1 = the standard error in FK1. (.0275 $\mu\text{g}/\text{count}$)

FK2, DF2 are similarly defined for a second counting arrangement differing from that used above. (e.g. FK2 determined for a larger distance between the sample and the detector).

It will be seen that as an analytical technique is developed FK1 and DF1 are not initially known. If the program is used with those values omitted, the output will consist of unconverted net peak areas. These areas can be used to obtain the FK's which can in turn be entered in the appropriate spaces on the card. The computer could have been programmed to make this calculation itself except that (1) once determined, FK's should not have to be re-computed, and (2) the estimate of DF should involve not only the Poisson-statistical uncertainty of individual estimates but also the uncertainties in reproducing the value of FK experimentally. Thus FK and DF should result from a set of measurements for which are most conveniently analyzed once by "hand" using the computer to provide net peak areas.

The second of the two isotope cards contains substrate data.

POL = the amount of element in units of SM present in the substrate on which the unknown is deposited per standard substrate area or volume. (i.e. $\mu\text{g Ca}$ on a full-circle polyethylene collector sheet.)

DP = the uncertainty in the value POL.

The pairs SOR, DS and POR, DR are similarly defined, so that the program as presently written allows substrate or blank subtraction to be made for three different materials. If no data is entered on this card, the program will operate properly assuming zero for blank subtraction. It will be seen that given FK1 or FK2, the activity of the blank material can be found initially by running the program for blanks as samples with zeros for the blank data. The weighted averages and standard deviations resulting from measurement of a number of duplicate blanks and analysis by computer, enables these zeros to be replaced.

04-09-70	04-09-70	04-09-70	04-09-
SIGNON SM8 T=2M			
PASSWD	SE75	264.6	.176
SRIN *MPRINT	SE75	400.7	.176
GS45 ON 7TP *T* MODE=2EN RING=OUT SIZE=25000 *TAPE271			.015 2904
SENDFILE	MG203	279.1	.515 .006 .03 .1125
SRIN SPAN ***T*	CNA0	1173.1	.015 .086 .01 .000
S37 3102.4 21.9 .01066 .00011 .0393 .003R	.001	.0005	.193 .001 .00015 .000
CA49 3083.0 99.8 .211 .0275 .R18 .04	CNA0	1332.4	.193 .001 .01 .9999
AL2R 1778.9 2.0 .00085 .000055 .0035 .00017	.001	.0005	.00015 .000
Y52 1434.4 2.056 .00004313 .0000027 .000152 .0000152	FE59	1291.5	150.0 .5 .0 .1082
.00091 .0002 .00037 .001	FE59	1098.6	150.0 .5 .0 .1082
CU66 1039.0 5.025 .00292 .00023 .0105 .00105	SC4A	889.4	.450 .3 .225 .04
.1 102 .06 .06 .01 .001	SC4A	1120.3	.0413 .002 .2013
T151 320.0 122.0 .002784 .00018 .01237 .001237	2MA5	1115.4	5.055 .1 .25 .5880
.24 .04 .25 .2 .05 .0342 .00342	.13	.05	.285 .05 .08 .015
NA24 1368.4 10.28 .00857 .00037 .0342 .00342	SR124	602.6	.212 .002 .01 .1461
.2 .02 .4 .1 .75 .075	.002	.0006	.011 .006 .00075 .000
NA24 2753.6 10.28 .01954 .001 .0807 .00807	SR174	1690.7	.212 .002 .01 .1461
.02 .4 .1 .75 .075	.002	.0006	.011 .006 .00075 .000
MNSA 846.9 2.428 .0000751 .0000036 .00029 .000029	CR51	320.0	2.1 .037 .1 .667
.0043 .0015 .0054 .01039 .00047 .0393 .00393	PA233	311.8	.025 .01 .015 .003
CL3R 1642.0 24.4 .01039 .00047 .0393 .00393	C05R	810.3	25.0 .12 .1 .5 1711
.44 .2 15.0 7.0 .49 .049	AG110M	937.2	.202 .025 .01 .6072
NA24 1368.4 10.28 .003195 .00015 .00682 .000682	AG110M	1384.0	.202 .025 .01 .6072
.2 .02 .4 .1 .735 .1	CE141	145.4	.5 0.0 .03 .780
NA24 2753.6 10.28 .007822 .00028 .0152 .00152			
.2 .02 .4 .1 .735 .1			
CL3R 1642.0 26.4 .00602 .00037 .0129 .00129			
.44 .2 15.0 7.0 .49 .08			
CL3R 2166.8 26.4 .00633 .00037 .0136 .00136			
.44 .2 15.0 7.0 .49 .08			
RR00 817.0 1.273 .000327 .00004 .000714 .0000714			
.041 .015 .065 .015 .024 .004			
MG27 1014.1 100.1 .0786 .0037 .161 .0161			
.1.03 .2 .5 .5			
MNSA 846.9 2.428 .0000333 .0000018 .0000667 .0000667			
.0043 .0015 .0054 .0006 .0024 .0004			
MNSA 1810.7 2.428 .000333 .000018 .000562 .0000562			
.0043 .0015 .0054 .0006 .0024 .0004			
MNSA 2112.8 2.428 .000726 .0000476 .000134 .000134			
.0043 .0015 .0054 .0006 .0024 .0004			
IN11AM 417.0 .0152 .00000336 .0000002 .0000083 .0000083			
.000045 .00001 .00019 .00003 .0000054 .0000023 .0000129 .00000129			
IN11AM 1097.1 .0152 .00000336 .0000002 .0000083 .0000083			
.000045 .00001 .00019 .00003 .0000054 .0000023 .0000129 .00000129			
112R 442.7 1.28 .0001625 .00004 .000325 .00008			
.02 .008			
NA24 1368.4 15.3			
CH4A 511.0 .50 .02 .025 12.8			
.0A .006 .05 .01 .005			
SM133 103.2 .00545 .0003 47.1 .0003			
ZMA0R 438.7 5.055 .1 0.025 13.8 .3			
.15 .05 .285 .05 .001 24.0 1.0			
W187 485.7 .0185 .001 24.0 1.0			
W187 479.3 .0185 .001 24.0 1.0			
RR02 554.3 .235 .062 .03 35.3 1.0			
.0X1 .015 .065 .015 .024 .004			
RR02 619.0 .235 .062 .03 35.3 1.0			
.041 .015 .065 .015 .024 .004			
RR02 776.6 .235 .062 .03 35.3 1.0			
.041 .015 .065 .015 .024 .004			
RR02 1043.9 .235 .062 .03 35.3 1.0			
.041 .015 .065 .015 .024 .004			
RR02 1474.7 .235 .062 .03 35.3 1.0			
.041 .015 .065 .015 .024 .004			
GA72 1860.6 .04 .01 14.3 .002			
GA72 2201.4 .04 .01 14.3 .002			
EUI52M 963.5 .00108 .00008 9.4 .00005			
AS76 559.2 .11 .007 26.3 .006			
AS76 657.0 .11 .007 26.3 .006			
AS76 1215.8 .11 .007 26.3 .006			
AS76 1228.8 .11 .007 26.3 .006			
K42 1524.7 5.065 .19 .25 12.5			
.07 .02 .1 .01 .075 .01			
LA140 1545.4 .021 .0004 .0001 40.2 .001			
LA140 328.6 .021 .0004 .0001 40.2 .001			
SR122 564.0 .212 .002 .01 67.2 .01			
.002 .0006 .011 .006 .00075 .0001			
SR122 692.5 .212 .002 .01 67.2 .01			
.002 .0006 .011 .006 .00075 .0001			

A List of Input Cards to SPAN.

The order of isotopes within a block is generally unimportant but in its present version the program corrects the total counts from $Mg^{26}(n, \gamma)Mg^{27}$ activity ($E=1014$ KeV) for the interference arising from the $Al^{27}(n,p)Mg^{27}$ reaction. In order to make the correction properly in the present program, the Al isotope cards must appear third in block with ITY=1 and the Mg isotope cards must appear sixth on the block with ITY=2. Moreover the analysis of the ITY=1 spectrum must precede analysis of the ITY=2 spectrum.

For the long irradiations (ITY=3,4) the isotope cards contain the following data:

Single	1.	NS		E		SM		ASH		DA		TAU		FMY
Isotope		2A4	2X	F10.5		F10.5		F10.5		F10.5		F10.5		F10.5
set		2. Same as card 2 for ITY=1,2												

NS, E, SM have the same meaning as before. (Say Zn 69M (ITY=3), 438.7 KeV, 5.055 μ g).

ASH = the amount of element present in a standard-sized portion substrate containing the standard. (units of SM 0.1 μ g)

DA = the uncertainty in ASH (0.025 μ)

TAU = the isotope halflife in hours. (13.8 hr)

There are two choices for standards with ITY=3,4. Either SM or SM*FMY. The choice is determined by a parameter in the control cards described below.

The ordering of isotope cards is again unimportant except:

1) For ITY=3 the first card must be NA 1368.4 KeV and the second CU 511.0. The program corrects the observed 511 KeV counts arising mainly for the $Cu^{66}\beta^+$ annihilation for a small contribution arising from the energetic Na^{24} 2753.6 KeV gamma ray. Experimentally:

$$\text{Net CTS Due to } Cu^{66} = \text{total CTS} - 0.093 * \text{COUNTS in } 1368 \text{ KeV } Na^{24} \text{ peak}$$

2) For ITY=4 the first card must be SE75 264.6 KeV while the third must be Hg203 279.1. Since the 279.1 line is the only one available for Hg 203, it must be

used if amounts of this element is to be measured but the, 279.6 KeV Se75 line is virtually coincident with it. This interference can be removed by subtracting the counts inferred to be due to Se75 based on the ratio of counts in the 264.6 KeV and 279.6 KeV peaks for pure Se.

Experimentally $\text{Se}(279.6)/\text{Se}(264.6) = 0.385$ so that counts due to Hg = gross counts - $0.385 \times \text{CTS}$ from Se 264.6.

These interference corrections which presently necessitate ordering of the data and spectrum processing in the case of Mg-Al involve minor modifications of the program and can be removed if calculation of weights of these elements (Mg, Hg and Cu) are not of interest.

The end of each of the four isotope sets is defined by two blank cards.

The isotope data cards are followed by four corresponding sets of gamma energies (ITY=1-4) for use with an internal energy calibration routine. The list consists of energies corresponding to recurrent, prominent, isolated peaks. Energies entered may not necessarily be those in the isotope data set. While gross changes in the calibration must be treated by supplying information externally from cards it has proved useful to compensate for small alterations in calibration (shifts of up to several kilovolts) by means of information in the spectrum itself. Expected peak positions calculated using the above lists locate an interval over which a search is made for the absolute maximum. If the maximum occurs at the ends of the interval, it is considered out of range for inclusion in the final list of peaks for calibration. If it is within the range its statistical significance is checked. If it qualifies statistically, the centroid is located using a parabolic interpolation routine XM. This routine has the effect of adding the correct decimal point to the channel with maximum counts. The subset of energies and identified centroids satisfying both position and statistical criteria is then used to update the calibration prior to spectrum analysis. If the number of energies left in the subset is less than the number of coefficients in the fitting polynomial (order+1), no update is made.

Once established, the above set (isotope data and internal energy calibration data) is included routinely with every tape analyzed. Cards specific for processing a given tape follow this set.

1. Label Card LAB1, LAB2, (2A4) label of the tape being treated (e.g. MT27)

2. Parameter Card

NFW	NDEL	NBASE	NCUT	LWD	SFLUX
I4	I4	I4	I4	I4	I10.5

For each gamma energy an interval $\pm$ NFW channels centered on the calculated peak position, ICH, is searched for the absolute maximum channel, IMX. The net peak area is calculated over an interval $\pm$ NBASE channels centered on the IMX. As in the case of the energy calibration, the maximum must be within the interval ICH $\pm$ NFW to be considered correctly located. If the maximum occurs at the ends of the interval a warning is generated.

A measure of the statistical significance of each net sum over the interval IMX $\pm$ NBASE, is computed and printed out. The statistical criterion is defined as $CRIT = ((N-B)/\sqrt{B})^2$ where N = total counts over the interval and B = the corresponding baseline sum. Thus CRIT is the square of the ratio of net peak counts to the standard deviation in baseline counts in the interval. This expression for CRIT can be related to the decision, detection and quantitative determination limits L_C , L_D and L_Q defined by Currie⁽²⁵⁾ in a careful discussion of signal detection thresholds. L_C is the value of that net signal which can just be detected in a single observation. L_D is the value of that true limiting mean signal which can be reliably detected in a given analytical procedure and L_Q is that level at which an individual signal is likely to be close to the limiting mean (somewhat arbitrarily taken as being within 10% with a 95% probability). "Working" expressions for the three limits are given⁽²⁵⁾ for both Gaussian and Poisson signal distributions. For the latter case

$$L_C = 2.33 \sqrt{\mu_B}$$

$$L_D = 2.71 + 4.65 \sqrt{\mu_B}$$

$$L_Q = 50 (1 + \sqrt{1 + \mu_B/12.5})$$

for "paired observations" of signal and background where μ_B = the limiting mean value of the background = σ_B^2 . For sufficiently large μ_B ,

$$\left(\frac{L_C}{\sigma_B}\right) \sim 2.33, \left(\frac{L_D}{\sigma_B}\right) \sim 4.65, \left(\frac{L_Q}{\sigma_B}\right) \sim \frac{50}{\sqrt{12.5}}$$

The approximations are correct to within 10% for $\mu_B \gg 70, 34, 1250$ counts respectively.

In the present case the net "signal" is N-B while the limiting mean background $\mu_B=B$ and $\sigma_B=\sqrt{B}$. Thus

$$\text{CRIT}_C = \left(\frac{L_C}{\sigma_B}\right)^2 \sim 6$$

$$\text{CRIT}_D = \left(\frac{L_D}{\sigma_B}\right)^2 \sim 20$$

$$\text{CRIT}_Q = \left(\frac{L_Q}{\sigma_B}\right)^2 \sim 200$$

From examples of isolated peaks having different values of CRIT, shown in fig. 14, it can be seen that there is qualitative agreement with the above results. For $\text{CRIT} \lesssim 5$ there is essentially no indication of a peak. The values of α are also measured and indicated, where

$$\alpha \equiv \frac{\text{Maximum count in the scan interval} - \bar{B}}{\sqrt{\bar{B}}}$$

where $\bar{B} = (B/\text{number of channels in the sum}) = \text{average baseline count per channel}$ in this region. Here $\alpha \sim 1$ indicating that the maximum peak count is comparable to the background fluctuations. For $\text{CRIT} \sim 10$ there is a detectable peak but little of quantitative use. The "peak" maximum is typically two standard deviations above the mean background ($\alpha \sim 2$). For $20 \lesssim \text{CRIT} \lesssim 200$ detection is reliable but fractional uncertainties in the peak area determination exceed 10%. These results are summarized below. The values entered apply with $\mu_B > 500$ counts.

Qualitative meaning of the statistical measure CRIT

CRIT	α	Comment
≤ 5	≈ 1	No detectable peak
5-20	~ 2	Detected with marginal reliability
20-200	3-10	Reliably detected but poor statistics
> 200	≥ 10	Statistics better than 10%

NDEL defines the interval for the internal energy calibration and for the titanium (320 KeV peak) flux monitor peak search. With 1KeV/channel calibration NDEL is typically taken as 5 channels.

NCUT = the number of channels out of the actual spectrum which will be treated by the program. Entering the minimum value consistent with retrieval of desired information will decrease running time. Gamma energy entered in the isotope data set with peaks in channels beyond NCUT will be deleted. (e.g. the 3083 KeV Ca^{41} peak should be analyzed. The baseline should be calculated correctly 2 ℓ +1= channels beyond this point so NCUT is taken as 3200).

LWD = the moving average half-width for the baseline calculation (BASE)
(e.g. LWD= ℓ =30)

SFLUX = the measure of reactor flux, in Ti^{320} KeV gamma counts, to which the values FK1, DF1, FK2, DF2 are referred. (e.g. 6000 counts. This number was obtained from computer analysis with NDEL=5).

3. The main control cards containing coded instructions for processing individual spectra appear next in the sequence.

ITG	NREC	NBG	ITY	NBE	NSK	NRW	NBKS	NCAL	FTR	CONV	TIME
I4			8I2							3F10.3	

ITG = tagword sought

NREC = 1,2,4 coded channel number 1=1024, 2=2048, etc.

NSK >0: skip NSK records

NBKS >0: backspace NBKS records

NRW >0: rewind the tape

ITG should not be entered for these operations. One instruction per card should be entered. At the end of execution of the instruction, the program expects another main control card.

NCAL <0: read energy calibration from cards in the following format:

EG	POS
2F	10.5

EG = actual gamma energy

POS = observed peak position

The order of the fit is - NCAL. Up to 100 such pairs may be entered. The end of this data set is indicated by a blank card. At the end of the calibration calculation, the program expects to read another main control card, so ITG should not be entered when NCAL<0.

NCAL>0: after reading in the spectrum with tagword ITG an internal energy calibration of order NCAL is made before analyzing.

No-operation values for the above quantities are thus zero. A totally blank card indicates the end of all data processing.

ITY = run type index defined earlier.

NBG = code describing the type of processing required for the spectrum.

		NGB				
		1	2	3	4	5
ITY	1	Sample Spectrum			Flux Monitor	Reset Flux
	2	Type of substrate			Spectrum	correction
	3	is			Standard *	Standard *
	4	POL	SOR	POR	Spectrum No Multitplier	Spectrum With Multiplier

*Blank is ASH

NBE = 0 use FK1, DF1 with ITY=1,2 not used with ITY=3,4

= 1 use FK2, DF2

FTR = fraction of substrate (POL, SOR, POR or ASH for the standard) actually used.

CONV = divides the calculated weight of sample to convert to desired units
(e.g. CONV = no of cubic meters of air sampled in which case output is in $\mu\text{g}/\text{m}^3$)

If ITY=3,4 and NBG=5, that is for a standard spectrum with modified standard values CONV = the multiplier of SM for those values of FMY in the isotope data set which are left blank.

For values of ITY=1,2 NBG=4,5

and ITY=3,4 NGB=4

CONV is not used.

TIME = time in the format hours.minutes for the onset of counting either the standard or the sample. This value is needed only for ITY=3,4.

The program calculates the correct fraction in hours from this value. Since the standard and the sample cannot be counted simultaneously a correction must be made for the decay of one relative to the other. Only the relative values of time are needed in the calculation.

Sample output. Computer output listed on pages 73 to 75 results from the main control data card sequence listed on p.61. The first card with NCAL=-1 produces a linear external energy calibration prior to reading any spectra using the energy cards which follow. The associated output gives the number of iterations required to produce a fit with $\epsilon=0.0011$, the linear coefficients with the intercept = -0.207 channels in this case and slope = 1.0037 channels/KeV. The five output columns are

E C_O C_C ΔC CRIT

E = isotope gamma energy

C_O = observed peak position

C_C = peak position-calculated from the best fit

ΔC = C_O - C_C

In the case of the external calibration CRIT is undefined and taken to be zero.

At the end of the external calibration a card with NSK=8 is read and 8 records are skipped. The spectrum with tagword 9 is sought and is processed as a flux monitor spectrum. The associated output for this analysis includes a printout of the main control card image, the current energy calibration parameters and

ICH = the expected peak location (319)

IMX = the observed maximum in the scan interval (320)

"FLUX" = the net peak area of the 320 KeV Ti^{51} gamma (6970.4) with the associated Poission-statistical uncertainty (89.5)

NBE = index of the set of conversion factors to be updated (in this case FK1)

SFLUX = the net peak area on which the conversion factors entered on the isotope data cards are based.

FL = the ratio SFLUX/FLUX

The flux monitor spectrum for the short runs (400 and 1000 sec.) must be analyzed first before treating the sample spectra. In typical measurements the flux spectrum is generated after the 400 second count and before the 1000 second count. Thus in order to analyze the 400 second spectrum the computer is instructed via the next card with NBKS=2 to backspace two records. Spectrum with tagword 8 is then sought and treated as a 400 second spectrum. (ITY=1)

Here NBG=2 indicating substrate type two (SOR) was used but only a fraction equal to 0.164 of the amount entered in the isotope data cards is to be subtracted.

CONV = 0.0053 = $.001 \times 5.3 \text{ m}^3$ of air were sampled by the substrate section and the output units are to be expressed in ng/m^3 while the conversion factors are in $\mu\text{g/count}$. (S is entered in mg/count so that the output units for this case is $\mu\text{g/m}^3$).

Following the listing of the control data in the output for tagword 8 are the results of analysis for each isotope present in the isotope data set within the range of NCUT. This table has the following entries

IND, ICH, IMX, NS, E, CT, DCT, W, DW, PCT, FC, CRIT

IND = the list index

ICH = expected peak channel

IMX = absolute maximum in the scan interval ICH+NFW

NS = element symbol

E = gamma energy

CT = net peak area (raw data counts)

DCT = Poission statistical uncertainty in CT

W = the corrected and converted net peak area (here in ng/m^3)

DW = the combined uncertainty in this value

PCT = the percent of substrate in the total sample weight

FC = the decay correction factor applied by the program. For ITY=1,2 FC=1.0

CRIT = the statistical criterion defined earlier.

It is assumed that the live time operating mode handles the dead time correction properly and that short-lived isotope dead time effects not correctable by live time operation are small. This is likely to be true if the overall dead time is kept low (e.g. <20%). Also it is assumed that the short-lived counting is for the same live-time period (400 and 1000 sec) as for the measurements of corresponding FK's. If not the correction must be included in the value of CONV.

W is given by

$$W = (CT \cdot FK - FTR \cdot BL) / CONV$$

where CT, FTR, and CONV are defined earlier. FK is either FK1 or FK2 and BL is the activity of the appropriate blank, POL, POR, etc. The error in W is given by

$$DW = \sqrt{\left(\frac{\partial W}{\partial FK}\right)^2 (DF)^2 + \left(\frac{\partial W}{\partial CT}\right)^2 (DCT)^2 + \left(\frac{\partial W}{\partial BL}\right)^2 (DBL)^2}$$

where DBL = DP, DS, etc.

For the first isotope, S37 (3102.4 KeV), the peak is not found within the 7 channel scan interval centered on channel 3112 but at the end channel, 3109. The net sum in the interval centered around 3109 is evaluated but the remaining calculation is deleted and a warning message printed. Inspection of the value CRIT suggests that the value is of doubtful statistical significance. This can be seen further from inspection of the original data which is presented below the isotope table. The contents of 23 channels centered on ICH are printed-out so that computer results may be readily checked by hand in those cases where any doubt about the analysis exists. The form of this table is

1. Index line 0 - 22
2. IND, IS, COUNTS (IS to IS+22) in I5 FORMAT
- .
- .
- .
- .

Thus in the case of S37 with IND=1 IS=3101, the column labeled 11 corresponding to IS+11=3112=ICH, is the expected peak position.

Upon completion of analysis of spectrum 8, 48 records are skipped and spectrum 59 is sought. This is treated as a long-irradiation standard spectrum (ITY=3, NBG=4). Comparison of the card input with the control line print out (p. 74) indicates that that time entered as 9h.15 min. has been correctly converted to 9.25 hrs. Also a new energy calibration update has been made which is not indicated in the write up, hence the new linear calibration coefficients - 0.522 and 1.002. Output for the standard spectrum has a slightly different form:

IND, ICH, IMX, NS, E, CT, DCT, FKS, DFS, PCT

where all entries have been defined previously except

FKS = calculated conversion factor ($\mu\text{g}/\text{count}$)

DFS = Poisson statistical uncertainty in CT combined with the blank uncertainty DA.

FKS = $(SM + ASH \cdot FTR) / CT$

and $DFS = \sqrt{\left(\frac{\partial FKS}{\partial ASH}\right)^2 (DA)^2 + \left(\frac{\partial FKS}{\partial CT}\right)^2 (DCT)^2}$

and it is assumed that there is no uncertainty in the value of SM or FTR.

It should be noted that while DF1 is based on a series of measurements indicating the overall reproducibility of FK1, DFS is based on a single measurement. While it includes the established uncertainty in the substrate activity (DA) it is not strictly analogous to DF1.

$PCT = \text{the percent of ASH in the total activity} = 100. \times \text{ASH} / (\text{SM} + \text{ASH})$

With analysis of the standard spectrum completed, one spectrum is skipped and spectrum 61 is analyzed as a sample spectrum (NBG=2, ITY=3) (p. 75). At the end of this analysis the job terminates.

Further information on all programs can be obtained from Dr. John Robbins, Great Lakes Research Division, I.S.T., North University Building, Room 1112, Telephone 764-2420.

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FLUX CAL.: ICH= 319 IMX= 320 *FLUX*= 6970.4 89.5 NBE= 1 SFLUX= 6000.0 FL= 0.86078

TAPE ID: MT27 TGM= CC08 LIVE TIME= 400 SEC

CONTROL LINE: 8 4 2 1 0 C 0 0 0 FTR 0.1640 CONV 0.0053 0.0 Energy Calibration Coefficients: -2.068460 1.003741

IND	ICH	DOX	NS	E	CT	DCT	W	DM	PCT	FC	CRIT
1	3112	3109	S37	3102.40	4.3	3.1	PEAK NOT WITHIN 7 CHANNEL SCAN INTERVAL				6.75
2	2086	2084	S37DE	2080.40	6.4	9.1	13.939	20.104	0.0	1.0000	1.05
3	3092	3092	CA49	3083.00	82.4	9.5	3036.112	535.378	1.5	1.0000	1873.11
4	1783	1783	AL28	1778.90	5403.8	99.5	1414.226	92.859	0.2	1.0000	353396.25
5	1438	1437	V52	1434.40	879.9	58.9	6.718	0.622	0.2	1.0000	599.22
6	1041	1041	CO66	1039.00	1112.9	92.5	3163.232	252.791	0.1	1.0000	30527.79
7	319	320	T151	320.00	535.3	91.3	256.571	48.728	2.9	1.0000	73.41
8	1371	1370	NA24	1368.40	149.0	49.8	213.562	76.200	5.5	1.0000	19.05
9	2762	2760	NA24	2753.60	57.7	9.1	187.380	33.224	6.2	1.0000	269.50
10	848	848	MN56	846.90	6568.0	102.9	92.632	4.730	0.2	1.0000	26751.95
11	1646	1646	CL38	1642.00	224.3	36.5	-51.003	227.537	112.3	1.0000	90.72

IND	IS	0	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22
1	3101	0	0	1	1	0	2	1	2	1	0	1	0	0	0	0	1	0	0	0	0	0	1	0
2	2075	6	4	5	4	3	7	7	6	4	9	8	7	4	6	8	7	3	7	3	10	6	6	6
3	3081	1	0	1	0	0	2	4	3	5	13	24	26	9	8	1	1	0	0	0	1	0	0	1
4	1772	91	75	57	112	123	128	122	197	407	998	2148	2760	2064	980	297	84	36	27	22	14	16	21	19
5	1427	190	181	178	177	174	190	189	224	281	379	440	362	283	203	180	187	186	164	167	209	164	170	183
6	1030	200	230	216	211	230	231	244	260	421	877	1743	2161	1337	262	238	171	164	161	179	164	144	144	162
7	308	568	577	592	560	602	632	609	588	615	600	611	643	683	665	600	637	589	574	548	535	581	585	559
8	1360	163	171	183	179	141	165	178	175	170	176	215	196	189	193	178	195	180	151	165	155	161	154	171
9	2751	3	3	2	0	5	2	4	3	14	16	14	9	10	3	1	0	4	4	3	1	3	2	0
10	837	322	280	327	307	335	305	414	506	624	1165	2035	2450	1552	642	315	231	244	217	238	225	219	222	195
11	1625	76	94	121	105	83	86	52	82	96	97	126	152	120	92	96	70	87	75	67	81	75	71	72

A Sample Long-Irradiation Standard Output from SPAN.

TAPE ID: MT27 TGM= OC59 LIVE TIME= 2000 SEC STD A (2)

CONTROL LINE: 59 4 4 3 0 0 0 0 0 0 PTR CONV TIME Energy Calibration Coefficients
 1.0000 1.0000 9.2500 -0.522490 1.002185

IND	ICH	IMX	NS	E	CT	DCT	PKS	DPS	PCT	CRIT
1	103	103	SM153	103.2C	5780.2	255.8	0.94288E-06	0.41734E-07	0.0	1119.70
2	439	439	ZN69M	438.70	3481.7	150.8	0.14806E-02	0.64110E-04	1.94	1259.69
3	687	687	W167	685.70	1392.2	109.6	0.13289E-04	0.10461E-05	0.0	365.01
4	480	480	W187	479.30	1866.2	140.0	0.99129E-05	0.74348E-06	0.0	392.98
5	555	554	BR82	554.3C	11838.8	163.4	0.25087E-04	0.91292E-06	20.88	18848.21
6	620	619	BR82	619.00	6192.2	136.9	0.47964E-04	0.19319E-05	20.88	6111.63
7	778	777	BR82	776.6C	7881.2	133.1	0.37685E-04	0.14195E-05	20.88	12627.73
8	1046	1045	BR82	1042.90	1585.3	114.8	0.14960E-03	0.10010E-04	20.88	704.25
9	1477	1477	BR82	1474.70	755.7	55.7	0.39300E-03	0.31861E-04	20.88	485.95
10	1864	1864	GA72	1860.40	63.1	53.0	0.63409E-03	0.53312E-03	0.0	2.89
11	2206	2207	GA72	2201.40	234.5	57.0	0.17056E-03	0.41419E-04	0.0	36.56
12	965	964	EU152M	963.50	1155.7	105.3	0.93451E-06	0.785164E-07	0.0	268.83
13	560	559	AS76	559.2C	5713.7	156.4	0.11324E-04	0.18231E-06	0.0	12802.25
14	658	657	AS76	657.00	654.8	109.1	0.16798E-03	0.27976E-04	0.0	76.31
15	1218	1217	AS76	1215.80	323.7	82.4	0.33978E-03	0.86524E-04	0.0	32.39
16	1231	1230	AS76	1228.80	90.1	77.2	0.12213E-02	0.10473E-02	0.0	2.76
17	1528	1527	K42	1524.70	1748.6	63.2	0.30053E-02	0.10879E-03	3.62	2721.48
18	1598	1599	LA140	1595.40	633.4	54.3	0.33157E-04	0.28447E-05	0.0	345.91
19	329	328	LA140	328.60	950.4	180.7	0.21203E-04	0.38682E-05	0.0	61.97
20	565	564	SB122	564.00	6972.5	146.7	0.30692E-04	0.64595E-06	0.93	6677.44
21	693	692	SB122	652.50	472.4	104.8	0.45300E-03	0.10046E-03	0.93	42.50

IND	IS	0	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22
1	92	4372	4388	4363	4210	4235	4345	4344	4271	4362	5026	6032	6501	5201	4254	4243	3981	4029	4186	4168	4170	4048	4065	4235
2	428	1430	1314	1453	1444	1497	1470	1448	1387	1576	1919	2380	2458	1900	1521	1351	1303	1338	1312	1334	1325	1295	1281	1320
3	676	759	765	791	756	749	773	760	638	847	1030	1160	1210	918	799	738	787	889	837	843	831	850	1078	1579
4	469	1231	1254	1237	1322	1285	1283	1214	1305	1296	1531	1818	1886	1562	1381	1255	1283	1339	1558	1666	1537	1363	1297	1267
5	544	1085	1175	1156	1158	1143	1205	1327	1471	1773	2814	4315	4262	2704	1936	2491	3378	2989	1970	1616	2240	2965	2682	1653
6	609	954	942	918	985	965	975	939	1188	1345	2006	2693	2577	1579	1078	860	881	872	934	906	905	1032	1085	1056
7	767	674	690	775	720	795	811	865	519	1353	2295	3193	2728	1467	845	723	647	650	693	731	746	701	701	723
8	1035	815	795	777	821	789	792	786	845	906	1178	1400	1345	1029	879	816	804	820	884	803	848	795	820	838
9	1466	185	169	158	160	169	194	165	179	171	214	301	396	372	279	198	173	175	199	183	171	180	184	189
10	1853	195	201	208	213	230	180	210	192	200	204	206	215	189	210	214	175	237	221	186	185	185	209	209
11	2195	209	219	186	214	208	208	213	213	226	246	257	249	277	264	218	228	209	216	213	204	207	243	236
12	954	682	707	727	731	713	745	734	725	746	873	1110	1030	914	726	704	735	744	699	702	717	705	719	734
13	549	1209	1327	1471	1773	2814	4315	4262	2704	1936	2491	3378	2989	1970	1616	2240	2965	2682	1653	1127	1044	978	959	1029
14	647	833	847	776	815	800	772	805	763	811	961	1062	976	895	806	726	831	783	761	766	821	820	802	798
15	1207	469	492	533	511	472	511	507	495	493	463	548	535	484	538	481	428	444	435	401	428	431	389	439
16	1220	538	481	428	444	435	401	428	431	389	439	458	432	440	439	424	468	394	397	412	408	392	401	416
17	1517	164	148	174	171	164	182	202	203	257	456	659	649	404	244	177	151	153	167	169	165	146	157	163
18	1587	174	158	184	169	188	175	177	177	185	177	196	288	360	310	256	206	157	156	149	152	186	173	164
19	318	2526	2621	2643	2550	2283	2321	2398	2290	2364	2435	2624	2448	2401	2258	2316	2270	2303	2381	2258	2274	2212	2230	2204
20	554	4315	4262	2704	1936	2491	3378	2989	1970	1616	2240	2965	2682	1653	1127	1044	978	959	1029	1009	1019	975	1014	1001
21	682	780	838	847	1030	1160	1210	918	799	738	787	889	837	843	831	850	1078	1579	1697	1361	978	817	720	726

A Sample Long-Irradiation Unknown Output from SPAN.

TAPE ID: MT27				TGV= C061	LIVE TIME= 2000 SEC		6-11-72					
CONTROL LINE: 61 4 2 3 0 C 0 0 0				PTR 0.3280	CONV 0.0113	TIME 9.9000	Energy Calibration Coefficients -0.522490 1.002185					
IND	ICH	IMX	NS	E	CT	DCT	W	DW	PCT	FC	CRIT	
1	103	103	SM153	103.20	4168.6	328.6	0.322	0.029	0.0	1.0096	334.75	
2	439	438	ZN69M	438.70	754.9	187.9	90.498	23.778	8.4	1.0332	36.62	
3	687	688	W187	685.70	158.7	144.6	0.175	0.160	0.0	1.0189	2.43	
4	480	483	W187	479.30	1162.6	184.3	PEAK NOT WITHIN 7 CHANNEL SCAN INTERVAL					82.42
5	555	555	BR82	554.30	14142.5	205.8	27.301	1.224	6.5	1.0128	14175.67	
6	620	620	BR82	619.00	8013.9	185.3	29.735	1.532	6.0	1.0128	4879.70	
7	778	777	BR82	776.60	10432.2	171.0	30.455	1.398	5.8	1.0128	11581.68	
8	1046	1045	BR82	1043.90	2410.6	154.6	27.781	2.784	6.4	1.0128	540.95	
9	1477	1478	BR82	1474.70	912.7	77.8	27.620	3.498	6.4	1.0128	324.44	
10	1864	1863	GA72	1860.40	110.6	75.7	5.880	6.375	0.0	1.0320	4.35	
11	2206	2207	GA72	2201.40	50.3	76.6	1.291	1.140	0.0	1.0320	2.82	
12	965	964	EUI52M	963.50	806.0	139.6	0.066	0.013	0.0	1.0491	69.52	
13	560	557	AS76	559.20	13228.4	203.5	PEAK NOT WITHIN 7 CHANNEL SCAN INTERVAL					12421.52
14	658	658	AS76	657.00	161.3	151.8	2.239	2.139	0.0	1.0173	2.27	
15	1218	1216	AS76	1215.80	421.3	114.8	11.828	4.412	0.0	1.0173	27.82	
16	1231	1229	AS76	1228.80	243.7	109.9	24.588	23.823	0.0	1.0173	10.04	
17	1528	1528	K42	1524.70	3522.3	93.8	888.474	40.060	0.3	1.0367	4701.98	
18	1598	1599	LA140	1555.40	679.7	76.7	1.840	0.262	0.6	1.0113	177.62	
19	329	328	LA140	328.60	3441.4	247.6	5.982	1.175	0.2	1.0113	409.49	
20	565	564	SB122	564.00	2242.8	174.0	5.309	0.485	5.7	1.0067	358.85	
21	693	696	SB122	692.50	2798.7	152.6	PEAK NOT WITHIN 7 CHANNEL SCAN INTERVAL					765.10

IND	IS	0	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22
1	92	7905	7588	7557	7515	7201	7464	7587	7519	7570	7747	8625	8819	8381	7660	7276	7433	7181	7357	7323	7283	7124	7447	7313
2	428	2548	2525	2600	2411	2502	2483	2509	2403	2587	2536	2752	2748	2602	2421	2432	2411	2451	2445	2401	2347	2395	2418	2395
3	676	1583	1461	1517	1536	1480	1520	1474	1549	1511	1482	1469	1549	1559	1464	1520	1486	1498	1522	1472	1530	1639	1762	2392
4	469	2368	2410	2392	2337	2357	2278	2340	2335	2420	2352	2383	2468	2456	2420	2549	2449	2523	2697	2940	2719	2604	2547	2548
5	544	2114	1970	2028	2067	2119	2131	2207	2234	2590	3775	5843	6350	4363	2822	2509	2729	2700	2379	2247	2348	2545	2382	2316
6	609	1973	1968	1848	1914	2012	1976	1998	2105	2401	3037	4229	4278	3090	2217	1923	1953	1841	1833	1953	1906	1913	1920	1927
7	767	1375	1491	1395	1398	1427	1405	1419	1488	2002	3293	4681	4146	2613	1606	1302	1338	1342	1288	1279	1342	1340	1385	1378
8	1035	1567	1501	1468	1569	1578	1561	1525	1564	1648	2000	2253	2224	1853	1611	1557	1469	1535	1557	1539	1514	1667	1536	1509
9	1466	365	371	332	337	372	351	406	360	393	411	514	651	659	511	396	338	401	401	401	387	361	366	360
10	1853	376	407	405	384	379	430	394	421	425	393	455	405	420	404	379	400	402	436	412	402	412	400	414
11	2195	393	402	390	404	399	364	420	406	421	430	428	415	453	418	447	391	445	420	428	419	416	421	438
12	954	1332	1388	1286	1336	1325	1385	1337	1336	1383	1480	1601	1511	1495	1345	1339	1344	1370	1373	1287	1312	1337	1308	1312
13	549	2131	2207	2234	2590	3775	5843	6350	4363	2822	2509	2729	2700	2379	2247	2348	2545	2382	2316	2043	1965	2017	2030	1848
14	647	1748	1784	1707	1789	1746	1675	1733	1592	1632	1621	1705	1779	1646	1574	1640	1561	1556	1582	1622	1609	1600	1583	1496
15	1207	920	923	983	913	926	938	1026	515	979	1024	998	930	930	924	956	917	867	947	901	888	874	878	918
16	1220	924	956	917	867	947	901	888	874	878	918	898	879	822	841	823	748	785	850	811	792	779	806	804
17	1517	426	361	384	381	393	387	434	432	562	937	1361	1364	974	564	399	365	360	354	336	360	347	359	398
18	1587	363	354	382	362	402	391	377	377	358	385	407	523	563	517	474	412	373	345	376	389	371	357	373
19	318	4474	4527	4679	4515	4570	4526	4603	4446	4646	4608	4839	4793	4602	4430	4316	4538	4521	4490	4419	4206	4209	4141	4033
20	554	5843	6350	4363	2822	2505	2725	2700	2379	2247	2348	2545	2382	2316	2043	1965	2017	2030	1848	2012	2012	1999	2027	1983
21	682	1474	1549	1511	1482	1465	1549	1559	1464	1520	1486	1498	1522	1472	1530	1639	1762	2392	2719	2449	1801	1521	1454	1465

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Fig. 1

Fig. 2

Fig. 3

Irradiation and Counting Scheme for Short-lived Isotopes.

Fig. 4

Fig. 5

Fig. 6

Irradiation and Counting Scheme for Long-lived Isotopes.

Fig. 8

Fig. 9

Fig. 10. A General Data Reduction Schematic Diagram

Fig. 11 Dependence of the calculated baseline on the moving-average half width, l .

Fig. 12. Gamma spectrum showing the calculated baseline and computer-located peaks.

An Abbreviated Flow Diagram for SPAN .

Fig. 13.

Fig. 14 Sample spectrum peaks versus the statistical criterion, crit. (Arrows indicate expected peak positions.)

APPENDIX

The Main Tape Inventory Program INVEN :

IN IV. G. COMPILER	MAIN	03-28-70	16151.11	PAGE 0001 IN IV. G. COMPILER	MAIN	03-28-70	16151.11	P
C MESSY TAPE READ, INVENTORY AND SUPER DUPER CLEAN WRITE				CALL READ(15,LEN,M,LN,5,C0,C01,C01,C01,C01,C01)				
C 7 TRACK BCD READ AND WRITE				MEN=LN				
C READ FROM PS0N 2 WRITE ON PS0N 5				MEN=(MEN/6)-2				
INTEGRAL LEN				WRITE (6,100) J,15(1),LEN,MEN,J				
DIMENSION IS(1000)				100 FORMAT(10X,'FILE NO. ',14,' TEND ',14,' LGTH= ',16,' C				
CEPSSICK LA(20)				1' CHNLS ',14,' CUM RES ',14)				
DIMENSION LB(2)				GO TO 8				
DATA M/24000000P				80 WRITE(6,524)				
READ (5,104) LB(1),LB(2)				525 FORMAT(5X,' END OF FILE 1)				
104 FORMAT(24X)				GO TO 8				
READ (5,707) (LACJ),J=1,201				81 WRITE (6,526)				
707 FORMAT(20X)				526 FORMAT(5X,' THERE IS AN ERROR ON THE NEWLY WRITTEN TAPE ')				
READ (5,407) N,N5K				8 CONTINUE				
400 FORMAT(21X)				WRITE(6,537)				
IF(N5K)6,6,7				537 FORMAT(1M,1				
7 CONTINUE				CALL REWIND (4)				
CALL SKIP(7,N5K,2)				STOP				
6 CONTINUE				2 CONTINUE				
KSF=0				GO TO 1				
KNTF=0				10 KEOP=KEOP+1				
KNT=0				KNTF=KNTF+1				
KF=0				WRITE (6,200) KEOP				
KEOP=0				200 FORMAT(20X,'END ',14)				
KEOP=0				IF(KNTF=81),1,5				
KCUR=0				5-WRITE(6,503)				
WRITE(6,601)				503 FORMAT(1M,30X,'SKIP')				
601 FORMAT(1M,'27X,' CONTENTS OF TAPE BEING READ ','')'				CALL SKIP(0,1,2)				
1 CONTINUE				KNTF=0				
KCUR=KCUR+1				KNT=0				
CALL READ(15,LEN,M,LN,2,C10,C11,C12,C13,C14,C15)				GO TO 1				
MEN=LEN				11 MC=8				
MEN=(MEN/6)-2				GO TO 16				
KF=KF+1				12 MC=12				
101 FORMAT(14X,'LN ',13,' TEND ',14,' LEN ',16,' CHNLS ',14,				GO TO 16				
1' CMRD ',13)				13 MC=16				
KNTF=0				GO TO 16				
IF(MEN-1724)31,21,22				14 MC=20				
22 IF(MEN-2044)31,21,23				GO TO 16				
23 IF(MEN-4096)31,21,31				15 MC=24				
21 KREC=KREC+1				WRITE(6,1773)				
WRITE (6,500) KREC				1003 FORMAT(20X,' TAPE MACHINE FAILURE CODE 24 ')				
500 FORMAT(1M,'5X,' CUM ',13)				STOP				
CALL WRITE(15,LEN,0,LN,4)				16 KERR=KERR+1				
KSF=0				WRITE(6,300) KERR,MC				
KNT=0				300 FORMAT(20X,'ERR ',14,' CO ',14)				
IF(MVLT(151)-41),3,3				KNT=KNT+1				
C MVLT IS FUNCTION TO CONVERT 4 TO 1 MODE USING BYTE-WISE SHIFTING				IF(KNT=21),4,4				
3 CALL REWIND(4)				4 CONTINUE				
WRITE (6,524) LB(1),LB(2)				CALL SKIP(0,1,2)				
524 FORMAT(1M,14X,' CONTENTS OF NEWLY WRITTEN TAPE ',24X,'')				KNTF=0				
WRITE(6,800) (LACJ),J=1,201				KNT=0				
800 FORMAT(40X,2044,'')				WRITE(6,502)				
DO 4 J=1,KREC				502 FORMAT(1M,37X,' SKIP')				
				GO TO 1				
				31 KSF=KSF+1				
				IF(KSF=41),32,32				
				32 WRITE(6,531)				
				531 FORMAT(20X,'QUEER FILE SKIP')				
				CALL SKIP(0,1,2)				
				KSF=0				
				GO TO 1				
				END				

AL MEMORY REQUIREMENTS 020608 BYTES

L MEMORY REQUIREMENTS 0002CC BYTES

A Simple Main Plotting Program :

FORTRAN IV G COMPILER		MAIN	10-24-69	15:04.22	PAGE 06
0001		DOUBLE PRECISION BS(4096)			
0002		DIMENSION SR(4096),CH(4096)			
0003		DIMENSION IR(4095)			
0004		FTY=2.0			
0005		FTX=.0025			
0006		XMN=66.			
0007		CALL PLTXMX(XMN)			
0008		XST=-11.0			
0009	999	CONTINUE			
0010		READ(5,420) NREC,ITG,NCUT,NSK,NCR			
0011		CALL SKIP(0,NSK,4)			
0012	420	FORMAT(5I4)			
0013		IF(NREC)805,8C5,717			
0014	717	CONTINUE			
0015		NR=NREC-1			
0016	478	CONTINUE			
0017		CALL RDTAPE(NR,I3,IT,IR)			
0018		IF(MVL(I3)-ITG)478,799,805			
0019	805	STOP			
0020	799	CONTINUE			
0021		XST=XST+11.0			
0022		XND=XST+10.0			
0023		CALL PENUP(XST,0.5)			
0024		CALL PENDN(XST,0.5)			
0025		CALL PENDN(XND,0.5)			
0026		CALL PENUP(XST,0.5)			
0027		CALL PENDN(XST,9.0)			
0028		DO 1 J=1, NCUT			
0029		SR(J)=IR(J)			
0030		IF (SR(J)) 2,2,3			
0031	2	CONTINUE			
0032		SR(J)=0.5			
0033		GO TO 987			
0034	3	SR(J)=0.5+FTY*ALOG10(SR(J))			
0035	987	CONTINUE			
0036		U=J			
0037	1	CH(J)=U*FTX+XST			
0038		CALL PLINE(CH(I),SR(I),NCUT,1,0,0,1)			
0039		IF(NCR)998,999,998			
0040	998	CONTINUE			
0041		CALL BASE(30,1,NCUT,IR,BS)			
0042		DO 51 J=1, NCUT			
0043		T=BS(J)			
0044		IF(T-1.0)52,52,53			
0045	52	T=1.0			
0046	53	CONTINUE			
0047	51	SR(J)=0.5+FTY*ALOG10(T)			
0048		CALL PLINE(CH(I),SR(I),NCUT,1,0,0,1)			
0049		GO TO 999			
0050		END			

MEMORY REQUIREMENTS 00044C BYTES

IV 6 COMPILER	MAIN	10-27-65	C9158,25	PAGE 0001	IV 6 COMPILER	INDEX	10-27-65	C5158,27	PAGE 0001
C	MAIN	PRG C	T RUN	10CEP	FPEP	EATA	ON	CARES	
		DIMENSION	ESI(100),	NI(100),	NI(20,100),	IC(20,100),	NS(100,2)		
		DIMENSION	ISI(100),	INCHI(100),	INPK(100),	ER(100,3)			
		READ	19,1001	TIME					
		K=0							
		1 K=1							
		READ(13,100)	ESI(K)						
		100 FORMAT(12F10.3)							
		IF(ESI(K)12.2)							
		2 IPR=K-1							
		CALL	10CMTIME,ESI,IPR,N,NT,IDX,NS,NPP,IP,INC,NPKL,ER)						
		WRITE(16,201)							
		201 FORMAT(1M1)							
		GO	3 J=1, IPR						
		WRITE(16,200)	J, ESI(J)						
		200 FORMAT(201,/,/,) PEAK NO. '13,10R,ENERGY',F7.1, KEV,/,							
		CALL	NTIC(12,M,N1,102,IP,NS,NPP)						
		3 CONTINUE							
		WRITE(16,203)							
		GO	4 J=1, NPP						
		WRITE(16,300)	J, IPI(J),NS1(J),1,NS1(J,2),INCHI(J),INPK(J),						
		1 ER(I,J),K=1,5)							
		300 FORMAT(102,14,2X,14,2P,2A4,2X,14,2X,14,512X,F7.1))							
		4 CONTINUE							
		STOP							
		END							
IV 6 COMPILER	WPT ID	10-27-65	09158,35	PAGE 0001					
		SUBROUTINE	NTIC(IJ,N,N1,102,IP,NS,NPP)						
		DIMENSION	NI(100), NI(20,100), ICH(20,100)						
		DIMENSION	IP(100), NS(100,2), I(100)						
		DATA	12 /M,*,HE*,*,L*,*,BE*,*,B*,*,C*,*,M*,*,D*,*,F*,*,NE*,*,NA*,						
		1,*,MG*,*,AL*,*,SI*,*,P*,*,S*,*,CL*,*,A*,*,K*,*,CA*,*,SC*,*,TI*,*,V*,*,CR*,							
		2,*,H*,*,FE*,*,CC*,*,NI*,*,CU*,*,Z*,*,GA*,*,GE*,*,AS*,*,SE*,*,BR*,*,KR*,*,RB*,							
		3,*,SR*,*,Y*,*,Z*,*,R*,*,BI*,*,PK*,*,TC*,*,BU*,*,BN*,*,PO*,*,AC*,*,CO*,*,IN*,*,SM*,							
		4,*,SB*,*,BE*,*,I*,*,NE*,*,CS*,*,DA*,*,LA*,*,CE*,*,PR*,*,ND*,*,PH*,*,SH*,*,EU*,							
		5,*,GO*,*,TO*,*,OY*,*,MC*,*,ER*,*,TH*,*,YB*,*,LU*,*,PF*,*,TA*,*,W*,*,RE*,*,OS*,							
		6,*,TE*,*,FO*,*,AU*,*,HG*,*,FL*,*,PB*,*,BT*,*,PC*,*,AS*,*,RN*,*,FR*,*,RA*,*,AC*/							
		M=N(IJ)							
		IF(NJ15,3,6)							
		4 CONTINUE							
		GO	4 L=1, NJ						
		NR=N(IJ,L)							
		I=IDR(L,J)							
		GO	98 K=1,NPP						
		IF(I-IPI(L))99,99,98							
		98 CONTINUE							
		99 CONTINUE							
		GO	TO(201,202,2C3,204,2C5,206),NR						
		201 WRITE(16,301) K, NS(K,1),NS(K,2)							
		301 FORMAT(20X,14,5X,2A4,3X, MAIN PEAK*)							
		GO	TO 4						
		202 WRITE(16,302) K, NS(K,1),NS(K,2)							

Non-linear Least Squares Routines :

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IV G COMPILER      NLS      12-31-69      11:26.11      PAGE 0001

SUBROUTINE NLS(X,Y,W,D,G,P,SE,VAR,ERR,EPS,NIT,ND,NP,K)
C NCM-LINEAR LEAST SQUARES FITTING ROUTINE
C DATA PAIRS X(I),Y(I) DESCRIBED BY A CONTINUOUS AND DIFFERENTIABLE FUNCTION
C F(X,G(I)) WHERE G(I)'S ARE ADJUSTABLE PARAMETERS. NLS STARTS WITH GUESSES
C FOR G(I) AND RETURNS "BEST FIT" VALUES FOR THEM WHICH MINIMIZE THE
C SUM OF THE SQUARES OF THE DIFFERENCES BETWEEN Y AND F.
C ND=NO OF DATAPOINTS X(I),Y(I)
C W=STATISTICAL WEIGHT ASSOCIATED WITH Y(I). =1/STANDARD DEVIATION**2
C G(I)=GUESS PARAMETERS INITIALLY, =BEST FIT PARAMS. UPON RETURN FROM NLS
C P=DUMMY PARAMETER COMMUNICATING WITH ARG
C SE(I)=STANDARD ERROR IN PARAMETER G(I)
C VAR=VARIANCE OF FIT
C ERR=0 UNLESS DET OF INVERSE MATRIX =0.0. THEN ERR=1
C NIT=NC OF ITERATIONS ALLOWED
C K=ACTUAL NUMBER PERFORMED
C EPS=ALLOWED FRACTIONAL ERROR IN EVERY G(I) TO TERMINATE ITERATION
C AP=ND OF PARAMETERS C(I)
DOUBLE PRECISION T(20,20)
DIMENSION X(100),Y(100),W(100),G(20),SE(20),B(20),D(20),P(10)
DIMENSION A(20,20)
DIMENSION IP(40)
DIMENSION CC(20)
ERR=0.0
DO 1 I=1,ND
  IF(W(I))2,2,1
2 W(I)=1.0
1 CC(1)=0
  K=0
3 IF(K-NIT)4,11,11
4 K=K+1
  DO 5 J=1,NP
    B(I)=0.0
    DO 5 I=1,NP
5 A(I,J)=0.0
    DO 6 I=1,ND
      CALL ARG(V,X(I),D,G,P)
      DEL=(Y(I)-V)*W(I)
      DO 6 J=1,NP
        B(I)=B(I)+DEL*D(I)
      DO 6 L=J,NP
        A(L,J)=A(L,J)+D(L)*D(J)*W(I)
6 T(L,J)=A(L,J)
    DO 7 J=1, NP
      DO 7 L=J,NP
7 T(J,L)=T(L,J)
    CALL INVP(20,T,IP,20,T)
    DO 41 J=1, NP
      CC(J)=0.0
      DO 41 L=1, NP
      DUM=T(I,J)
41 CC(J)=CC(J)+B(L)*DUM
    DO 14 J=1, NP
      IF(DABS(T(J,J))-1.CCG-30)15,14,14
15 ERR=1.0
14 CC(1)=0
  IF(ERR)16,16,17
17 RETURN
16 CC(1)=0
  KCHK=0
  DO 8 J=1,NP
    IF(G(J))914,8,914
914 CC(1)=0
  IF(ABS(CC(J)/G(J))-EPS)8,8,10
10 KCHK=1
  8 G(J)=G(J)+CC(J)
  IF(KCHK)11,11,3
11 VAR=0.0
  DO 12 I=1,NC
    CALL ARG(V,X(I),D,G,P)
12 VAR=VAR+B(I)*(Y(I)-V)**2
  RGT=ND-NP
  IF(RGT)1001,1001,1002
1001 VAR=-0.0
  GO TO 1003
1002 CC(1)=0
  VAR=SQRT(VAR/RGT)
1003 CC(1)=0
  DO 13 J=1,NP
    DUM=DABS(T(J,J))
13 SE(J)=VAR* SQRT(DUM)
  RETURN
END

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IV G COMPILER ARG 12-31-69 11:26.07 PAGE 0001

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SUBROUTINE ARG(V,L,C,G,P)
DIMENSION D(20), G(20), P(100)
N=P(1)
V=G(1)
C(1)=1.0
DO 1 J=2,N
  C(J)=D(J-1)*L
1 V=V+G(J)*C(J)
RETURN
END

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MEMORY REQUIREMENTS 0001A BYTES

The Main Spectrum Analysis Program SPAN :

IN IV C COMPILER	MAIN	03-28-70	1A160.46	PAGE 0001	IV C COMPILER	MAIN	03-28-70	1A160.46	PAGE 0003
DOUBLE PRECISION B5(4000)					IF(1010,1,8				
DIMENSION NS(14,2,50),E(4,50),ASH(4,50),DA(4,50),POL(4,50)					4 IF(1010-1,10155,658,1666				
DIMENSION SDR(4,50),DS(4,50),SM(4,50),TAU(4,50)					1066 WRITE(6,1667)				
DIMENSION FK(4,50,2),CF(4,50,2),POR(4,50),DR(4,50)					1667 FORMAT(5X,' TAGNOMO DISCREPANCY')				
DIMENSION ENVI(4,50),NBR(4,50),ECAL(20),NCL(14),LVL(21),JLP(50)					STOP				
DIMENSION DP(4,50),IP(4,1),ID(4000),IT(4,50),VL(20)					658 CONTINUE				
DIMENSION X(100),Y(100),M(100),D(20),C(20),P(100),SE(20)					ITN=TIME				
DATA FL,SBSE,SDBF,SBSNA,SDRBA,DAVAL,AVALL,0.690,0/					TMP=ITN				
DO 4940 J=1,23					FP=10.0+ITN(TMP)/6.0				
4940 LVL(J)=J-1					TIME=TMP+FP				
WRITE(6,16001)					IF(INCUT-NR10124,9124,9125				
8001 FORMAT(1H,20X,' ISOTOPE AND BLANK DATA SUMMARY')					9124 KCUT=NCUT				
WRITE(6,224)					GO TO 9216				
DO 34 J=1,4					9125 KCUT=NR				
WRITE(6,16003) J					9216 CONTINUE				
8003 FORMAT(///,10X,' RUN TYPE INDEX= ',I4,/))					INCK=NF*LLWD-KCUT				
K=0					GO TO 1711,1712, KSET				
1 K=K+1					712 ICH=0.5*ECAL(320,ALOPD,G)				
GO TO (73,73,74,74), J					IND=ICH+200				
73 READ(5,102) NS(J,1),NS(J,2),K1,ET(J,K),SM(J,K),FR(J,K),DF(J,K,1)					CALL BASE(LWD,1,THO,TR,BS)				
1,FR(J,K,2),DF(J,K,2)					CALL IM(ICH,MODEL,IR,1P)				
WRITE(6,1022) NS(J,1),NS(J,2),K1,ET(J,K),SM(J,K),FR(J,K),DF(J,K,1)					CALL PKBS(TIME,MODEL,IR,BS,RO,SRO,SDDI				
1,FR(J,K,2),DF(J,K,2)					FL=SFLUX/SRO				
102 FORMAT(24X,23,'6F10.5)					GO TO 717				
1022 FORMAT(24X,23,'2F10.5,2X1,2F10.5,2X1)					711 CONTINUE				
READ (5,103) POL(J,K),CP(J,K),SOR(J,K),OS(J,K),POR(J,K),DR(J,K)					CALL BASE(LWD,1,NCUT,TR,BS)				
103 FORMAT(6F10.5)					IF(INCAL) 1754,1754,1755				
WRITE (6,1032) POL(J,K),CP(J,K),SOR(J,K),OS(J,K),POR(J,K),DR(J,K)					1755 NCAL=NCAL+1				
1 DR(J,K)					LORD=NCAL+1				
1032 FORMAT(18X,10,6)					P(1)=LORD				
GO TO 75					KOL=1				
74 READ (5,104) NS(J,1),K1,NS(J,2),K2,ET(J,K),SM(J,K),ASH(J,K),DA(J,K)					K=0				
1,TAU(J,K),FMV(J,K)					DO 1756 J=1, NC				
WRITE(6,1042) NS(J,1),K1,NS(J,2),K2,ET(J,K),SM(J,K),ASH(J,K),DA(J,K)					IC=0.5*ECAL(IECTTY,J),LST,G)				
1,TAU(J,K),FMV(J,K)					IF(C) INCK(4080,4080,1756				
1042 FORMAT(24X,24,'7F10.5)					4080 CONTINUE				
104 FORMAT(24X,23,'7F10.5)					CALL IM(ICH,MODEL,IR,1P)				
READ (5,103) POL(J,K),CP(J,K),SOR(J,K),OS(J,K),POR(J,K),DR(J,K)					IF(TABSTIC-INX1-NOEL)1760,1756,1756				
WRITE (6,1032) POL(J,K),CP(J,K),SOR(J,K),OS(J,K),POR(J,K),DR(J,K)					1760 CALL IM(INX1,IC,SP,NMC)				
1 DR(J,K)					IF(INCAL)1757,1757,1756				
75 IF(ET(J,K)12,2)					1757 CALL PKBS(TIME,1,IR,BS,RT,ST,SD)				
2 IPK(J)=K-1					CRIT=ST*RT				
34 CONTINUE					15CRIT=10.0/1756,1756,7762				
WRITE(6,8573)					7762 K=K+1				
8573 FORMAT(1H,///,2,' INTERNAL ENERGY CALIBRATION GAMMA ENERGIES',///)					X(K)=EC(ITY,J)				
DO 1762 J=1,4					V(K)=X(K)				
WRITE(6,8003) J					V(K)=X(K)				
K=0					1756 CONTINUE				
1762 K=K+1					IF(K-LORD)1756,1756,1759				
READ(5,103) ET(J,K)					1759 KOL=2				
IF(ET(J,K)11764,1764,1962					GO TO 1754				
1963 CONTINUE					32 KRD=0				
WRITE(6,8217) K, EC(J,K)					LORD=LORD+1				
8217 FORMAT(14X,12,2X,10,4)					P(1)=LORD				
GO TO 1763					356 KRD=KRD+1				
1764 NCL(J)=K-1					READ(5,103) X(KRD),Y(KRD)				
1762 CONTINUE					IF(X(KRD)1357,357,356				
201 FORMAT(24X)					357 K=KRD-1				
READ(5,200) NFWM,NOEL,NBASE,NCUT,LWD,SFLUX					1759 INCK=NF*W*W*D*G*P*SE,VAR,ERR,.001,20,K,LORD,ITER)				
200 FORMAT(15X,10,5)					1863 CONTINUE				
LWM=2*NOEL+1					LST=LORD				
LBASE=2*NBASE+1					WRITE(6,9999)				
LLWD=2*NOEL+1					9999 FORMAT(1H,1)				
LDEL=2*NOEL+1					IF(INCAL)1011,0111,9854				
WRITE(6,211) LFM,LDEL,LBASE,NCUT,LLWD					6111 CONTINUE				
211 FORMAT(1H,1, SCAN WIDTH ',I4,' MCWIDTH WIDTH ',I4,' SUM					WRITE(6,6112) ITER, (G(J), J=1, LORD)				
MCWIDTH ',I4,' SPEC CUT-OFF ',I4,' BASE SMOOTHING WIDTH ',I					6112 FORMAT(///,5X,'EXTERNAL CALIBRATION: ITERATIONS= ',I3,5X,TIME(12,5)				
2,///)					1,///)				
WRITE(6,6212) SFLUX					GO TO 9853				
6212 FORMAT(10X,' STANDARD FLUX = ',F10.1,' CTS',///)					9854 WRITE(6,6113) IS, ITER, (G(J), J=1, LORD)				
LST=2					6113 FORMAT(///,5X,'INTERNAL CALIBRATION: ICM= ',A4,' ITERATIONS= ',				
6117=0					113,5X,TIME(12,5),///)				
DO 33 J=1,4					9853 CONTINUE				
IP=IPK(J)					DO 9851 J=1, K				
DO 33 K=1, IP					FIT=ECAL(IE(J),LORD,G)				
33 ITR(J,K)=0.5*ET(J,K)					UZ=FIT-FIT				
3 READ(5,222) TG,NREC,NBG,ITY,NBF,NSK,NRW,NBKS,NCAL,PTX,CONV,TIME					WRITE(6,9852) X(J),Y(J),FIT,UZ,VL(J)				
KR=1					9852 FORMAT(20X,3F10.3,2X1,5X,F6.2,5X,F7.2)				
NSV=NBE					9851 CONTINUE				
222 FORMAT(14,812,3F10.5)					DO 3317 J=1,4				
NBE=NBE+1					IP=IPK(J)				
IF(INCAL)32,31,31					DO 3317 K=1,IP				
31 IF(INRW)4,4,5					3317 ITR(J,K)=0.5*ECAL(IE(J,K),LORD,G)				
5 REMIND 4					IF(INCAL)3,3,1754				
GO TO 3					1754 CONTINUE				
4 IF(NBKS) 14,14,15					717 CONTINUE				
15 CALL SKIP(10,-NBKS,4)					508 FORMAT(1H,1,OR,' TAPE TO: ',24X,' TCM= ',A4,' LIVE TIME= '				
GO TO 3					114,' SEC',///)				
14 IF(NBKS)333,333,668					WRITE(6,223)ITG,NREC,NBG,ITY,NSV,NSK,NRW,NBKS,NCAL,PTX,CONV,TIME				
668 CALL SKIP(10,NSK,4)					1,IC(1CON),LCOM=1,LORD)				
GO TO 3					223 FORMAT(1H,1,CONTROL LINE ',I4,2X,6(12,1X),3(2X,F6.4),7(2X,F10.6)				
333 CONTINUE					GO TO (1771,1772),KOL				
IF(ITY=21334,334,337					1772 WRITE(6,1773)				
334 IF(NBG=41337,336,335					1773 FORMAT(1X,' ENERGY CALIBRATION NOT MODIFIED')				
335 FL=1					1771 WRITE (6,224)				
WRITE(6,3334)					GO TO (1718,1719), KSET				
3339 FORMAT(20X,///,10,'NO FLUX CORRECTIONS',///)					719 WRITE(6,720)ICH,TRN,SRO,SDD,NBE,SFLUX,FL				
GO TO 3					720 FORMAT(1H,1,FLUX CAL',I, ICH= ',I4,5X,'INCK= ',I4,				
336 KSET=2					15X,'*FLUX= ',F10.1,2X,F6.1,5X,'NBE= ',I2,5X,'SFLUX= ',F10.1,				
337 CONTINUE					2, FL= ',F10.5)				
NB=1024*NREC-1					GO TO 3				
IF(NB=0,6,7					718 I=ITY				
6 STOP					IP=IPK(J)				
7 CONTINUE					DO 12 J=1, IP				
559 CONTINUE					IC=IC(1,IT,J)				
CALL ROTAPE(NA,13,17,IR)					IP=IPK(J)				
LT=NRV(13)					IF(ICH=INCK)17112,7112,7449				
					7449 IP(J)=1				

[illegible]