Nondestructive Activation Analysis of Environmental Samples

RICHARD DAMS and JOHN A. ROBBINS

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Project Director

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Technical Report

NONDESTRUCTIVE ACTIVATION ANALYSIS OF ENVIRONMENTAL SAMPLES

Richard Dams
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TABLE OF CONTENTS

Chapter I. Neutron Activation Analysis

1. Principles ........................................... 1
2. Formation and Decay Rate of Radioactive Species .... 2
3. Activation Analysis .................................. 3
4. Sensitivity-Standard Deviations ....................... 4
5. Counting-Gamma Spectrometry ........................ 5
6. Nondestructive Activation Analysis ..................... 8

Chapter II. Procedure for Nondestructive Activation Analysis of Environmental Samples

1. Apparatus Used ....................................... 10
2. Identification of Isotopes-Qualitative Analysis ...... 11
3. Quantitative Analysis of Germanium Lithium Gamma-Ray Spectra ....................................... 13
4. Irradiation-Counting Scheme .......................... 15
5. Sensitivity-Detection Limits ............................ 24
6. Possible Interferences .................................. 26
7. Neutron Flux Mapping ................................... 29

Chapter III. Computerized Data Reduction

1. Introduction .......................................... 31
2. Tape Analysis
   A. Tape Structure ..................................... 34
   B. Tape Submission at U. M. Computing Center ....... 34
   C. Inventory Program (INVEN) .......................... 35
3. Data Analysis Programs
   A. Read and List Programs ............................. 39
      RDLST (main)
      RDTAPE
      MVL
      WRTMT
B. Base Spectrum Calculation and Peak Integration ... 43
   BASE
   PKBS
   WRTBS

C. Plotting ..................................................... 48

D. Peak Search Routine ........................................ 48
   PTST (main)
   PEAK
   SFZW
   FKZ
   CIJW

E. Component Identification ................................. 51
   IDTST (main)
   IDCOM
   GAMLIB (Data File)

F. Non-linear Least Squares Program ....................... 55
   NLLS
   ARG

4. Composite Program ........................................... 59
   SPAN (Main Spectrum Analysis Program)
   IM
   XM
   ECAL

References .................................................. 77

List of Figures ............................................. 78
<table>
<thead>
<tr>
<th>Program</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>INVEN</td>
<td>94</td>
</tr>
<tr>
<td>RDLST</td>
<td>95</td>
</tr>
<tr>
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<td>ECAL</td>
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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

\[ ^{A}_{Z}M + (n, \gamma) ^{A+1}_{Z}M \]
Examples are $^{23}\text{Na}(n,\gamma)^{24}\text{Na}$, $^{11}\text{Na}$, $^{59}\text{Co}(n,\gamma)^{60}\text{Co}$.

The nuclides formed, $^{A+1}\text{M}$, $^{24}\text{Na}$, $^{60}\text{Co}$ are often unstable and disintegrate, usually by emission of beta rays to an excited state of another isotope which deexcites 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 $E = h\nu$ ($\nu$ 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:

$$A \rightarrow n \rightarrow B \rightarrow \lambda \rightarrow C$$

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

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

Thus: $\frac{dN_B}{dt} = \sigma^{-} \phi N_A - \lambda N_B$  \hspace{1cm} (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:

$$\text{Act}_{B}^{t_i} = NB^{t_i} \lambda = \sigma \Phi N_A [1 - \exp (-\lambda t_i)] = \sigma \Phi N_A S \quad (3)$$

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 $\sigma$, $\lambda$, or $t \frac{1}{2}$, $\Phi$ and $N_A$ the radioactivity for each irradiation time and cooling time can easily be calculated. Approximate values for $\sigma$ and $t \frac{1}{2}$ are tabulated (6, 8). The neutron flux can be determined and is usually approximately known for each irradiation site. Neutron fluxes vary between $10^{13 - 2 - 1} n\ cm^{-2}\ sec^{-1}$ 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

$\text{M.W.}$ is the molecular weight of the target material

$\Theta$ 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 $\sigma$ are however not known accurately, a precise determination of $\Phi$ 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:

\[
\frac{(g)v}{(g)S} = \frac{(\text{Act})v}{(\text{Act})S} \tag{7}
\]

where \((g)v\) and \((g)S\) are the weights of unknown and standard \((\text{Act})v\) and \((\text{Act})S\) 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.


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 \((\sigma)\), its isotopic abundance \((\theta)\), the half-life of the isotope produced \((\lambda)\), the irradiation time \((t_i)\), and the neutron flux \((\phi)\). 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: 

$\sigma = 1 \text{ barn}$  
$\theta = 1 \text{ (100%)}$  
$M.W. = 100$  
$t_1 = t \frac{1}{2} \text{ thus } S = 0.5$  
$\phi = 10^{12} \text{n/cm}^2 \text{-sec}$

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:

$$\sigma_N = \sqrt{\frac{N^2 - N}{T} + \frac{B^2}{T - B}} = \sqrt{T + B}$$

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

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

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 $2 \cdot mc^2 = 1.02$ Mev).
The positron is not stable and by the reverse process, positron-electron "annihi-
lation", 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 Cs\textsuperscript{137}) 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 cm\textsuperscript{3}. The detection efficiency of a 30 cm\textsuperscript{3} 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).


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

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 (2). 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.


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:

$$ P = A_0 + \sum_{1}^{n} A_1 + \sum_{-1}^{-n} A_1 $$

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:

$$ Q = Q' + Q'' = \sum_{2n+m}^{2n+m+1} A_1 + \sum_{n+m}^{n+m} A_1 $$

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\( t_{\text{irr}} \approx t/2 \), \( t_{\text{cool}} < t/2 \) and \( t_{\text{count}} \approx 2t/2 \). 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, $^{116m}$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 ($2 \times 10^{12}$ n/cm$^2$-sec), the sample can easily be packed in a polyethylene 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 ($t_{1/2} = 110$ 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, $^{116m}$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.
Table 3. Nuclear properties of short-lived isotopes. The gamma-rays used in the determination are underlined.

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

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 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 (NH₄)₂SO₄ 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 $^{51}\text{Ti}$ ($t_{1/2} = 5.8$ min) produced by $(n,\gamma)$ 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 $^{51}\text{Ti}$, the same monitor can be used almost indefinitely. Indeed, if the rate of analysis does not
Table 4. Standard solutions of determination of elements giving rise to short-lived isotopes.

<table>
<thead>
<tr>
<th>Solution Number</th>
<th>Element</th>
<th>Concentration µg/ml</th>
<th>Volume Spotted</th>
<th>Amount Irradiated µg</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Al</td>
<td>10.0</td>
<td>200</td>
<td>2.00</td>
</tr>
<tr>
<td>1</td>
<td>V</td>
<td>1.028</td>
<td>200</td>
<td>0.2056</td>
</tr>
<tr>
<td>1</td>
<td>Cu</td>
<td>25.12</td>
<td>200</td>
<td>5.025</td>
</tr>
<tr>
<td>1</td>
<td>Mg</td>
<td>500.5</td>
<td>200</td>
<td>100.1</td>
</tr>
<tr>
<td>1</td>
<td>Ca</td>
<td>499.0</td>
<td>200</td>
<td>99.8</td>
</tr>
<tr>
<td>1</td>
<td>Na</td>
<td>51.4</td>
<td>200</td>
<td>10.28</td>
</tr>
<tr>
<td>1</td>
<td>Mn</td>
<td>1.214</td>
<td>200</td>
<td>0.2428</td>
</tr>
<tr>
<td>1</td>
<td>In</td>
<td>0.076</td>
<td>200</td>
<td>0.0152</td>
</tr>
<tr>
<td>2</td>
<td>Cl</td>
<td>264.0</td>
<td>50</td>
<td>13.2</td>
</tr>
<tr>
<td>2</td>
<td>Br</td>
<td>12.73</td>
<td>50</td>
<td>0.636</td>
</tr>
<tr>
<td>2</td>
<td>I</td>
<td>12.83</td>
<td>50</td>
<td>0.6415</td>
</tr>
<tr>
<td>3</td>
<td>Mn</td>
<td>24.28</td>
<td>100</td>
<td>2.428</td>
</tr>
<tr>
<td>3</td>
<td>Ti</td>
<td>415.0</td>
<td>100</td>
<td>41.5</td>
</tr>
<tr>
<td>4</td>
<td>S</td>
<td>100 mg (NH₄)₂SO₄</td>
<td>weighed</td>
<td>24.2 mg S</td>
</tr>
</tbody>
</table>

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 \(s\) (sample + 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 \(^{56}\text{Mn}\) and \(^{24}\text{Na}\) 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:

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

Isotopes with half-lives longer than 10 days only become visible in the
gamma-ray spectrum after the complete decay of the isotope \(^{82}\text{Br}\). Bromine is
indeed usually abundant in environmental samples, it is very sensitive for activa-
tion analyses and the isotope \(^{82}\text{Br}\) has a very complicated gamma-ray spectrum.
Twenty to thirty days after the end of irradiation the following isotopes can be
detected, $^{51}\text{Cr}$, $^{46}\text{Sc}$, $^{60}\text{Co}$, $^{59}\text{Fe}$, $^{65}\text{Zn}$, $^{75}\text{Se}$, $^{110m}\text{Ag}$, $^{124}\text{Sb}$, $^{141}\text{Ce}$, $^{203}\text{Hg}$ and $^{233}\text{Pa}$, and in some samples $^{58}\text{Co}$.

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 $\text{H}_2\text{WO}_4$). 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
<table>
<thead>
<tr>
<th>Element</th>
<th>Isotope Produced</th>
<th>Half-life</th>
<th>Most Prominent $\gamma$-rays (Kev)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Na</td>
<td>$^{24}_{\text{Na}}$</td>
<td>15.0 hr</td>
<td>1368.4; 2753.6;</td>
</tr>
<tr>
<td>K</td>
<td>$^{42}_{\text{K}}$</td>
<td>12.5 hr</td>
<td>1524.7;</td>
</tr>
<tr>
<td>Zn</td>
<td>$^{69m}_{\text{Zn}}$</td>
<td>13.8 hr</td>
<td>438.7;</td>
</tr>
<tr>
<td>Cu</td>
<td>$^{64}_{\text{Cu}}$</td>
<td>12.8 hr</td>
<td>$\beta^+ (511.0)$;</td>
</tr>
<tr>
<td>Br</td>
<td>$^{82}_{\text{Br}}$</td>
<td>35.9 hr</td>
<td>554.3; 619.0; 698.3; 716.6; 827.8; 1043.9; 1317.2; 1474.7;</td>
</tr>
<tr>
<td>As</td>
<td>$^{76}_{\text{As}}$</td>
<td>26.3 hr</td>
<td>559.2; 657.0; 767.5; 1228.8;</td>
</tr>
<tr>
<td>Ga</td>
<td>$^{72}_{\text{Ga}}$</td>
<td>14.3 hr</td>
<td>601.1; 630.1; 786.4; 810.5; 834.1; 1800.1;</td>
</tr>
<tr>
<td>La</td>
<td>$^{140}_{\text{La}}$</td>
<td>40.3 hr</td>
<td>328.6; 486.8; 815.4; 1595.4;</td>
</tr>
<tr>
<td>Sm</td>
<td>$^{153}_{\text{Sm}}$</td>
<td>47.1 hr</td>
<td>69.6; 103.2;</td>
</tr>
<tr>
<td>Eu</td>
<td>$^{152m}_{\text{Eu}}$</td>
<td>9.35 hr</td>
<td>121.8; 344.2; 841.6; 963.5; 1315.0; 1388.9;</td>
</tr>
<tr>
<td>Sb</td>
<td>$^{122}_{\text{Sb}}$</td>
<td>2.75 da</td>
<td>504.0; 692.5; 1256.6;</td>
</tr>
<tr>
<td>W</td>
<td>$^{187}_{\text{W}}$</td>
<td>24.0 hr</td>
<td>72.3; 134.3; 479.3; 551.4; 618.1; 685.7; 772.9;</td>
</tr>
<tr>
<td>Au</td>
<td>$^{198}_{\text{Au}}$</td>
<td>2.70 da</td>
<td>411.8;</td>
</tr>
<tr>
<td>Sc</td>
<td>$^{46}_{\text{Sc}}$</td>
<td>83.9 da</td>
<td>889.4; 1120.3;</td>
</tr>
<tr>
<td>Cr</td>
<td>$^{51}_{\text{Cr}}$</td>
<td>27.8 da</td>
<td>320.0;</td>
</tr>
<tr>
<td>Co</td>
<td>$^{60}_{\text{Co}}$</td>
<td>5.24 yr</td>
<td>1173.1; 1832.4;</td>
</tr>
<tr>
<td>Fe</td>
<td>$^{59}_{\text{Fe}}$</td>
<td>45.1 da</td>
<td>1098.6; 1291.5; 192.5; 143.0;</td>
</tr>
<tr>
<td>Ni</td>
<td>$^{58}_{\text{Ni}}$</td>
<td>71.3 da</td>
<td>810.3; 863.5; $\beta^+ (511.0)$;</td>
</tr>
<tr>
<td>Zn</td>
<td>$^{65}_{\text{Zn}}$</td>
<td>245.0 da</td>
<td>1115.4; $\beta^+ (511.0)$;</td>
</tr>
<tr>
<td>Se</td>
<td>$^{75}_{\text{Se}}$</td>
<td>121.0 da</td>
<td>96.7; 121.8; 136.0; 264.6; 279.6; 400.7;</td>
</tr>
<tr>
<td>Ag</td>
<td>$^{110m}_{\text{Ag}}$</td>
<td>260.0 da</td>
<td>657.8; 706.4; 763.9; 884.5; 937.2; 1384.0;</td>
</tr>
<tr>
<td>Sb</td>
<td>$^{124}_{\text{Sb}}$</td>
<td>60.9 da</td>
<td>602.6; 645.7; 722.8; 1436.8; 1690.7; 2090.6</td>
</tr>
<tr>
<td>Ce</td>
<td>$^{141}_{\text{Ce}}$</td>
<td>32.5 da</td>
<td>145.4;</td>
</tr>
<tr>
<td>Hg</td>
<td>$^{203}_{\text{Hg}}$</td>
<td>46.9 da</td>
<td>279.1;</td>
</tr>
<tr>
<td>Th</td>
<td>$^{233}_{\text{Pa}}$</td>
<td>27.0 da</td>
<td>299.9; 311.8; 340.3;</td>
</tr>
</tbody>
</table>

Table 5. Nuclear properties of long-lived isotopes. The gamma rays used in the determinations are underlined.
<table>
<thead>
<tr>
<th>Solution Number</th>
<th>Element</th>
<th>Concentration µg/ml</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>Zn</td>
<td>101.1</td>
</tr>
<tr>
<td>5</td>
<td>K</td>
<td>101.3</td>
</tr>
<tr>
<td>5</td>
<td>Cu</td>
<td>10.0</td>
</tr>
<tr>
<td>5</td>
<td>As</td>
<td>2.20</td>
</tr>
<tr>
<td>5</td>
<td>Ga</td>
<td>0.80</td>
</tr>
<tr>
<td>5</td>
<td>Sb</td>
<td>4.24</td>
</tr>
<tr>
<td>5</td>
<td>La</td>
<td>0.421</td>
</tr>
<tr>
<td>5</td>
<td>Sm</td>
<td>0.109</td>
</tr>
<tr>
<td>5</td>
<td>Eu</td>
<td>0.0216</td>
</tr>
<tr>
<td>5</td>
<td>Sc</td>
<td>0.826</td>
</tr>
<tr>
<td>5</td>
<td>Cr</td>
<td>42.1</td>
</tr>
<tr>
<td>5</td>
<td>Co</td>
<td>3.87</td>
</tr>
<tr>
<td>5</td>
<td>Fe</td>
<td>3,000.0</td>
</tr>
<tr>
<td>5</td>
<td>Ni</td>
<td>500.0</td>
</tr>
<tr>
<td>5</td>
<td>Se</td>
<td>3.53</td>
</tr>
<tr>
<td>5</td>
<td>Ce</td>
<td>10.0</td>
</tr>
<tr>
<td>5</td>
<td>Hg</td>
<td>10.3</td>
</tr>
<tr>
<td>5</td>
<td>Th</td>
<td>0.80</td>
</tr>
<tr>
<td>5</td>
<td>Ag</td>
<td>4.05</td>
</tr>
<tr>
<td>6</td>
<td>Br</td>
<td>4.71</td>
</tr>
<tr>
<td>6</td>
<td>W</td>
<td>0.37</td>
</tr>
<tr>
<td>6</td>
<td>Au</td>
<td>0.140</td>
</tr>
</tbody>
</table>

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 \(i\)
\[ A_s = \text{the peak area of the isotope in sample } s \]
\[ \lambda = \text{the decay constant of the element} \]
\[ t_i = \text{the cooling time of the sample } i \]
\[ t_s = \text{the cooling time of the standard } s \]

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 (\( Q = 35\% \)) under the photo-peak 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.
### Table 7. Sensitivity of 13 elements by short irradiation in adopted irradiation-counting scheme.

<table>
<thead>
<tr>
<th>Element</th>
<th>$t_{\text{irr. flux}}$</th>
<th>$t_{\text{count}}$</th>
<th>$t_{\text{decay}}$</th>
<th>Detection limit, ng, for typical air filter samples</th>
</tr>
</thead>
<tbody>
<tr>
<td>S</td>
<td>5 min 2x10^{12} n/cm^2-sec</td>
<td>400 sec</td>
<td>3 min</td>
<td>25,000</td>
</tr>
<tr>
<td>Ca</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>1,000</td>
</tr>
<tr>
<td>Al</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>40</td>
</tr>
<tr>
<td>V</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>100</td>
</tr>
<tr>
<td>Cu</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>200</td>
</tr>
<tr>
<td>Ti</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>20</td>
</tr>
<tr>
<td>Br</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>0.2</td>
</tr>
<tr>
<td>In</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>3</td>
</tr>
<tr>
<td>Mn</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>3,000</td>
</tr>
<tr>
<td>Mg</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>200</td>
</tr>
<tr>
<td>Na</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>500</td>
</tr>
<tr>
<td>Cl</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>---</td>
</tr>
</tbody>
</table>

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

<table>
<thead>
<tr>
<th>Element</th>
<th>$t_{\text{irr flux}}$</th>
<th>$t_{\text{count}}$</th>
<th>$t_{\text{decay}}$</th>
<th>Detection limit, ng, for typical air filter samples</th>
</tr>
</thead>
<tbody>
<tr>
<td>K</td>
<td>5 hour 1.5x10^{13} n/cm^2-sec</td>
<td>2000 sec</td>
<td>20-30 hours</td>
<td>75</td>
</tr>
<tr>
<td>Zn</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>200</td>
</tr>
<tr>
<td>Cu</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>50</td>
</tr>
<tr>
<td>Br</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>25</td>
</tr>
<tr>
<td>As</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>40</td>
</tr>
<tr>
<td>Ga</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>10</td>
</tr>
<tr>
<td>La</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>2</td>
</tr>
<tr>
<td>Sm</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>0.05</td>
</tr>
<tr>
<td>Eu</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>0.1</td>
</tr>
<tr>
<td>Sb</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>5</td>
</tr>
<tr>
<td>W</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>1</td>
</tr>
<tr>
<td>Au</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>0.3</td>
</tr>
<tr>
<td>Sc</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>0.3</td>
</tr>
<tr>
<td>Cr</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>20</td>
</tr>
<tr>
<td>Co</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>2</td>
</tr>
<tr>
<td>Fe</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>1,500</td>
</tr>
<tr>
<td>Ni</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>1,500</td>
</tr>
<tr>
<td>Zn</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>100</td>
</tr>
<tr>
<td>Se</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>10</td>
</tr>
<tr>
<td>Ag</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>10</td>
</tr>
<tr>
<td>Sb</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>8</td>
</tr>
<tr>
<td>Ce</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>20</td>
</tr>
<tr>
<td>Hg</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>10</td>
</tr>
<tr>
<td>Th</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>3</td>
</tr>
</tbody>
</table>
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 \((n,p), (n,\alpha), \) and \((n, 2n)\) reactions. Some determinations based upon an \((n,\gamma)\) reaction induced by thermal neutrons may be interfered by such threshold reactions.

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

\[
\begin{align*}
\text{A}_Z \text{M} & \text{ (n,} \gamma \text{) } \text{A}+1 \text{ M} \\
\text{A}+1 \text{ M} & \text{ (n,} p \text{) } \text{A}+1 \text{ M} \\
\text{A}+4 \text{ M} & \text{ (n,} \alpha \text{) } \text{A}+1 \text{ M} \\
\text{A}+2 \text{ M} & \text{ (n,} 2n \text{) } \text{A}+1 \text{ M}
\end{align*}
\]

(1) (2) (3) (4)

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 \(32S(n,p)32P\) 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 \(\phi_f\) = the mean fission flux in \(n/cm^2-sec\).

- \(D_{32S}\) = the absolute disintegration rate of \(32P\) produced by \((n,p)\) reaction on sulphur
- \(\sigma_{32S}\) = the mean fission cross section for the reaction \(32S(n,p)32P\)
- \(N_{32S}\) = the number of atoms \(32S\) present
\[ \lambda \text{ = the decay constant of the isotope } ^{32}\text{P} \]

This requires however an absolute measurement of the \(^{32}\text{P}\) 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 \(^{32}\text{P}\) produced by the threshold reaction to the count rate of \(^{32}\text{P}\) produced by the thermal reaction \(^{31}\text{P}(n, \gamma)^{32}\text{P}\). This requires now only a relative measurement of \(^{32}\text{P}\). Thus from a simultaneously irradiated and counted amount of phosphorus the thermal neutron flux is given as:

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

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

\(\text{Act}_{32\text{P}}\) = the relative count rate of \(^{32}\text{P}\) produced by \((n, \gamma)\) 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. \(^{32}\text{P}\) 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 \(^{35}\text{S}\) produced by \((n, \gamma)\) reaction on \(^{34}\text{S}\) 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. \(^{31}\text{P}(n,p)^{32}\text{P}, \sigma = 0.19 \text{ barn}\), \(^{32}\text{S}(n,\gamma)^{32}\text{P}, \sigma = 0.005 \text{ barn}\).

<table>
<thead>
<tr>
<th>Irradiation Site</th>
<th>Activity (\text{mg P/c min})</th>
<th>Activity (\text{mg S/c min})</th>
<th>Ratio Activity(P) Activity(S)</th>
<th>Neutron Flux Ratio Thermal/Fission</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pneum. Tube</td>
<td>206,480</td>
<td>8,823</td>
<td>23.40</td>
<td>7.6</td>
</tr>
<tr>
<td>Reactor Pool</td>
<td>4,060,200</td>
<td>295,354</td>
<td>13.75</td>
<td>4.5</td>
</tr>
</tbody>
</table>

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 \(\text{Al(NO}_3)_3\). It appeared that 100 micrograms of aluminum
Table 10. Nuclear interferences due to threshold reactions.

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

generates a false amount of magnesium equal to $20 \pm 8$ 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 $^{71}$Ga(n,$\gamma$)$^{72}$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 $^{27}$Al(n,$\alpha$)$^{24}$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-negligible 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 Ce(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 258X6 BCD characters long or as one continuous record (N+2)X6 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.

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.

FORTRAN 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 with-
out 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. 99 (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

<table>
<thead>
<tr>
<th>LNF</th>
<th>CHNLS</th>
<th>CMRD</th>
<th>CRN</th>
<th>CHK</th>
<th>TYP</th>
<th>CHILS</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>12345</td>
<td>68</td>
<td>123</td>
<td>456</td>
<td>789</td>
<td>01234</td>
</tr>
<tr>
<td>2</td>
<td>23456</td>
<td>78</td>
<td>234</td>
<td>567</td>
<td>890</td>
<td>02345</td>
</tr>
<tr>
<td>3</td>
<td>34567</td>
<td>89</td>
<td>345</td>
<td>678</td>
<td>901</td>
<td>03456</td>
</tr>
<tr>
<td>4</td>
<td>45678</td>
<td>90</td>
<td>456</td>
<td>789</td>
<td>012</td>
<td>04567</td>
</tr>
<tr>
<td>5</td>
<td>56789</td>
<td>01</td>
<td>567</td>
<td>890</td>
<td>123</td>
<td>05678</td>
</tr>
<tr>
<td>6</td>
<td>67890</td>
<td>23</td>
<td>678</td>
<td>901</td>
<td>234</td>
<td>06789</td>
</tr>
<tr>
<td>7</td>
<td>78901</td>
<td>34</td>
<td>789</td>
<td>012</td>
<td>345</td>
<td>07890</td>
</tr>
<tr>
<td>8</td>
<td>89012</td>
<td>45</td>
<td>890</td>
<td>123</td>
<td>456</td>
<td>08901</td>
</tr>
<tr>
<td>9</td>
<td>90123</td>
<td>56</td>
<td>901</td>
<td>234</td>
<td>567</td>
<td>09012</td>
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<tr>
<td>10</td>
<td>12345</td>
<td>67</td>
<td>123</td>
<td>456</td>
<td>789</td>
<td>01234</td>
</tr>
</tbody>
</table>

## CONTENTS OF NEWLY WRITTEN TAPE CLIPS

<table>
<thead>
<tr>
<th>LNF</th>
<th>CHNLS</th>
<th>CMRD</th>
<th>CRN</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>12345</td>
<td>68</td>
<td>123</td>
</tr>
<tr>
<td>2</td>
<td>23456</td>
<td>78</td>
<td>234</td>
</tr>
<tr>
<td>3</td>
<td>34567</td>
<td>89</td>
<td>345</td>
</tr>
<tr>
<td>4</td>
<td>45678</td>
<td>90</td>
<td>456</td>
</tr>
<tr>
<td>5</td>
<td>56789</td>
<td>01</td>
<td>567</td>
</tr>
<tr>
<td>6</td>
<td>67890</td>
<td>23</td>
<td>678</td>
</tr>
<tr>
<td>7</td>
<td>78901</td>
<td>34</td>
<td>789</td>
</tr>
<tr>
<td>8</td>
<td>89012</td>
<td>45</td>
<td>890</td>
</tr>
<tr>
<td>9</td>
<td>90123</td>
<td>56</td>
<td>901</td>
</tr>
</tbody>
</table>

* Short Record:
A Source Tape Listing of an Unusually Bad Tape:

## CONTENTS OF TAPE READING READ

<table>
<thead>
<tr>
<th>FLN</th>
<th>TGW</th>
<th>LEN</th>
<th>CHNLS</th>
<th>CMRD</th>
<th>CRN</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>TGW 0035</td>
<td>24588</td>
<td>4096</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>2</td>
<td>TGW 0036</td>
<td>24588</td>
<td>4096</td>
<td>4</td>
<td>2</td>
</tr>
<tr>
<td>3</td>
<td>TGW 0037</td>
<td>24588</td>
<td>4096</td>
<td>7</td>
<td>3</td>
</tr>
<tr>
<td>4</td>
<td>TGW 0038</td>
<td>24588</td>
<td>4096</td>
<td>10</td>
<td>4</td>
</tr>
<tr>
<td>5</td>
<td>TGW 0039</td>
<td>24588</td>
<td>4096</td>
<td>13</td>
<td>5</td>
</tr>
<tr>
<td>6</td>
<td>TGW 0040</td>
<td>24588</td>
<td>4096</td>
<td>17</td>
<td>6</td>
</tr>
<tr>
<td>7</td>
<td>TGW 0041</td>
<td>24588</td>
<td>4096</td>
<td>21</td>
<td>7</td>
</tr>
<tr>
<td>8</td>
<td>TGW 0042</td>
<td>317</td>
<td>50</td>
<td>25</td>
<td></td>
</tr>
<tr>
<td>9</td>
<td>TGW 0042</td>
<td>24588</td>
<td>4096</td>
<td>27</td>
<td>8</td>
</tr>
<tr>
<td>10</td>
<td>TGW 0043</td>
<td>24588</td>
<td>4096</td>
<td>31</td>
<td>9</td>
</tr>
<tr>
<td>11</td>
<td>TGW 0043</td>
<td>24588</td>
<td>4096</td>
<td>41</td>
<td>11</td>
</tr>
<tr>
<td>12</td>
<td>TGW 0044</td>
<td>24588</td>
<td>4096</td>
<td>43</td>
<td>12</td>
</tr>
<tr>
<td>13</td>
<td>TGW 0045</td>
<td>24588</td>
<td>4096</td>
<td>47</td>
<td>13</td>
</tr>
<tr>
<td>14</td>
<td>TGW 0046</td>
<td>24588</td>
<td>4096</td>
<td>51</td>
<td></td>
</tr>
<tr>
<td>15</td>
<td>TGW 0046</td>
<td>24588</td>
<td>4096</td>
<td>52</td>
<td></td>
</tr>
</tbody>
</table>

**QUEER FILE SKIP**
Tagword #1  Tagword #2

\[
\begin{array}{cccc}
0 & 0 & 1 & 2 \\
3 & 4 & 0 & 1 \end{array}
\]

\[
\begin{array}{cccc}
1 & 2 & 3 & 4 \\
1 & 2 & 3 & 4 & 5 & 6 & \ldots \end{array}
\]

I1  I2  I3  IT

3A4  I6  etc.

(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 alphabetic mode), the \[ \text{live} \text{ actual} \] time of data accumulation, IT, and the data array \( \text{IR}(J) \) (\( J=1, NR \)) 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 \( \text{IR}(J), J=1, NR \) 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 CLTH6 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 < 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 in-
creased 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 oper-
ational 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', with nᵢ counts in the ith
channel, a smoothed spectrum S₀ in which the number of counts in the ith channel,
\( \bar{n}_i \), is taken as the average over an interval of width 2l+1 centered about i.
Thus

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

<table>
<thead>
<tr>
<th>Start Channel</th>
<th>TAGWORD 0016</th>
<th>LIVE TIME 1000 SEC</th>
<th>End Channel</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1</td>
<td>2</td>
<td>3</td>
</tr>
<tr>
<td>11</td>
<td>10</td>
<td>29</td>
<td>37</td>
</tr>
<tr>
<td>31</td>
<td>595</td>
<td>310</td>
<td>553</td>
</tr>
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<td>41</td>
<td>505</td>
<td>405</td>
<td>430</td>
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<td>61</td>
<td>410</td>
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<td>422</td>
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<td>71</td>
<td>321</td>
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<td>91</td>
<td>317</td>
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<td>101</td>
<td>314</td>
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<tr>
<td>111</td>
<td>328</td>
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</tr>
<tr>
<td>121</td>
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<td>131</td>
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<td>141</td>
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<tr>
<td>201</td>
<td>199</td>
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So and $S^0$ are compared. Whenever $n_i$ exceeds $\bar{n}_i$ by a standard deviations (i.e. by $\alpha \sqrt{\frac{\bar{n}_i}{n_i}}$), $n_i$ is replaced by $\bar{n}_i$ 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 $\alpha$. ($\alpha_{i+1} < \alpha_i$). The resultant spectrum $S^n$ is taken as the baseline. In subroutine BASE the initial value of $\alpha$ 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 $L=30$ 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 $\bar{n}_i$ 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 $L+1$) 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):

$L = \text{sum half width} = 30$

$\text{IST} = \text{starting channel (usually = 1)}$
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

\[ SBS = \text{the net counts} = \]
\[ \sum_{J = ICH - NFW}^{ICH + NFW} \{ IR(J) - BS(J) \} = N - B \]

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 Mn$^{56}$ 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)

<table>
<thead>
<tr>
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<th>308</th>
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Input Card Listing for Plot Program.

$SIGNON 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 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, $E_\gamma$, 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 $(n,\gamma)$ 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 Adams (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 ± δE. 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) satisfying the relation, \( \frac{ES(j)}{ES(j)} = 1.132 \), which holds for the Ka X-ray pair. If such a pair is found, the Z of the associated atomic nucleus is given by

\[
Z = f(E) = 28.553 + 0.9485 E - 0.003085 E^2
\]

The value of the energy spread, δE = \( \frac{FWHM}{2} \), is obtained from a function subroutine EF(E). IDCOM Arguments: (values \textit{returned} by IDCOM; \textit{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 ≤ IDX ≤ 243) associated with the Kth attribute and Jth peak. If NI = 6, IDX is not the isotope index, but rather the Z of the X-ray transition nucleus. IDX = 0.5 + f(E).

NS(L, Loc) = Nuclide symbol for the Lth 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 Lth 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 Lth nuclide.

NPKL(L) = Number of primary gammas (excluding 511 KeV) associated with the Lth nuclide for which a main peak was present. NPKL(L) ≤ INDX(L).

ER(L,k) = The primary gamma energies associated with the Lth nuclide. K=1, 5.

If N(j) = 0 the jth 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.
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 $2\delta E = 4$ KeV. A form used in later runs was

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

IDIST 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 $Y_j = f(X_j, B_1 \ldots B_m)$ where the $B_i$'s ($i = 1, M$) are $M$ adjustable parameters. The subroutine NLLS determines a parameter set $B_i$, starting from initial guesses for $B$ (say $B_{i0}$), which minimizes the residual

$$R = \sum_{j=1}^{N} W_j (Y_j - f(X_j, B_1 \ldots B_m))^2$$

with respect to each $B_i$ and thus produces a "best" fit according to this least squares criterion. That is, a set $B_i$ is found such that $\frac{\partial R}{\partial B_i} = 0$ 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 $B_i$. Moreover the functions must be bounded
in $\beta_1^n$ 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, $\delta_1$, to initial values of $\beta_1^0$, which will tend to minimize $R$, that is new values of $\beta_1$,

$$\beta_1^1 = \beta_1^0 + \delta_1^0$$

are calculated for which

$$f(X_j, \beta_1^1) \approx f(X_j, \beta_1^0) + \sum_{i=1}^{M} \frac{\partial f}{\partial \beta_i} \bigg|_{\beta_i = \beta_i^0} \delta_1^0$$

such that

$$\frac{\partial}{\partial \delta_i} \sum_{j=1}^{N} \omega_j (y_j - f(x_j, \beta_1^0)) - \sum_{i=1}^{M} \frac{\partial f}{\partial \beta_i} \delta_i^0 = 0$$

A solution of these $M$ linear equations for each $\delta_i^0$ allows one to correct $\beta_1^0$ in the direction of minimum $R$. The new values of $\beta_1, \beta_1^1$, constitute new guesses for $\beta_1$ which themselves may be corrected by calculation of a new $\delta_1^1$. This iterative procedure may be repeated until a desired accuracy in calculating each $\beta_1$ is achieved. That is, until

$$\left| \frac{\beta_1^n - \beta_1^{n-1}}{\beta_1} \right| \leq \epsilon$$

for all $i$ where $\epsilon$ is small.
Equation can be seen to be a matrix equation. Defining \( F_j = (Y_j - f(X_j, \beta_1 \ldots)) \), taking the partial with respect to \( \delta_k \), gives,

\[
\sum_{J=1}^{N} w_j \left( F_j - \sum_{l=1}^{M} \frac{\partial f}{\partial \beta_l} S_l \right) \frac{\partial f}{\partial \beta_k} = 0
\]

which gives

\[
\sum_{J=1}^{N} w_j F_j \frac{\partial f}{\partial \beta_k} = \sum_{l=1}^{M} \sum_{J=1}^{N} w_j \frac{\partial f}{\partial \beta_l} \frac{\partial f}{\partial \beta_k} S_l = A_{ik} S_i
\]

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

\[
\sum_{J=1}^{N} w_j \frac{\partial f}{\partial \beta_k} \frac{\partial f}{\partial \beta_l} \]

symbolically the solution for \( \delta_i \) 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 \( \beta_i \) is given by

\[
\sigma_{\beta_i} = \sqrt{\frac{1}{N-M} \left( \sum_{j=1}^{N} w_j F_j^2 \right) \left| A_{ii}^{-1} \right|}
\]
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 \( f(X_j, \beta_1\ldots) \) is supplied by the user as the subroutine ARG. The fortran listing of ARG on p. 99 is for a power series,

\[
f(X_j, \beta_1\ldots\beta_m) = \beta_1 + \beta_2 X_j + \beta_3 X_j^2 \ldots
\]

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 \( \beta \) 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:

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<tr>
<th>ITY</th>
<th>Irradiation</th>
<th>Counting Live Time</th>
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<tr>
<td>1</td>
<td>5 minutes</td>
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<tr>
<td>2</td>
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<tr>
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<td>4000</td>
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For the short irradiations (ITY=1,2) the isotope cards have the following format:

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<th>CARD</th>
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<td>1</td>
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<table>
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<tr>
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<th>DF1</th>
<th>FK2</th>
<th>DF2</th>
</tr>
</thead>
<tbody>
<tr>
<td>2A4</td>
<td>2X</td>
<td>F10.5 F10.5 F10.5 F10.5 F10.5 F10.5</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
```

```
<table>
<thead>
<tr>
<th>POL</th>
<th>DP</th>
<th>SOR</th>
<th>DS</th>
<th>POR</th>
<th>DR</th>
</tr>
</thead>
<tbody>
<tr>
<td>F10.5 F10.5 F10.5 F10.5 F10.5 F10.5</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
```

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 ug/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. ug 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.
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:

<table>
<thead>
<tr>
<th>Single Isotope set</th>
<th>1. NS</th>
<th>E</th>
<th>SM</th>
<th>ASH</th>
<th>DA</th>
<th>TAU</th>
<th>FMY</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>2A4</td>
<td>2X</td>
<td>F10.5</td>
<td>F10.5</td>
<td>F10.5</td>
<td>F10.5</td>
<td>F10.5</td>
</tr>
<tr>
<td>2. Same as card 2 for ITY=1,2</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

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

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

DA = the uncertainty in ASH (0.025 µ)

TAU = the isotope half-life 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$^{66}$ β+ annihilation for a small contribution arising from the energetic Na$^{24}$ 2753.6 KeV gamma ray. Experimentally:

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

\[
\begin{array}{ccccccc}
\text{NFW} & \text{NDEL} & \text{NBASE} & \text{NCUT} & \text{LWD} & \text{SFLUX} \\
14 & 14 & 14 & 14 & 14 & 110.5
\end{array}
\]

For each gamma energy an interval ± 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 ± NBASE channels centered on the IMX. As in the case of the energy calibration, the maximum must be within the interval ICH±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 DMX±NBASE, is computed and printed out. The statistical criterion is defined as

\[
\text{CRIT} = \left( \frac{N - B}{\sqrt{B}} \right)^2
\]

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

\[
\begin{align*}
L_C &= 2.33 \sqrt{\mu_B} \\
L_D &= 2.71 + 4.65 \sqrt{\mu_B} \\
L_Q &= 50 \left( 1 + \sqrt{1 + \mu_B/12.5} \right)
\end{align*}
\]
for "paired observations" of signal and background where \( \mu_B = \) the limiting mean value of the background = \( \sigma_B^{-2} \). For sufficiently large \( \mu_B \),
\[
\left( \frac{L_C}{\sigma_B} \right) \sim 2.33 \left( \frac{L_D}{\sigma_B} \right) \sim 4.65 \left( \frac{L_Q}{\sigma_B} \right) \sim \frac{50}{\sqrt{12.5}}
\]
The approximations are correct to within 10\% for \( \mu_B > 70 \), 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 CRIT \( \ll 5 \) there is essentially no indication of a peak. The values of \( \alpha \) are also measured and indicated, where
\[
\alpha = \frac{\text{Maximum count in the scan interval} - \overline{B}}{\sqrt{\overline{B}}}
\]
where \( \overline{B} = (B/\text{number of channels in the sum}) = \) 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 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 \( \ll \) CRIT \( \ll 200 \) 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

<table>
<thead>
<tr>
<th>CRIT</th>
<th>α</th>
<th>Comment</th>
</tr>
</thead>
<tbody>
<tr>
<td>&lt;5</td>
<td>1</td>
<td>No detectable peak</td>
</tr>
<tr>
<td>5-20</td>
<td>~2</td>
<td>Detected with marginal reliability</td>
</tr>
<tr>
<td>20-200</td>
<td>3-10</td>
<td>Reliably detected but poor statistics</td>
</tr>
<tr>
<td>&gt;200</td>
<td>&gt;10</td>
<td>Statistics better than 10%</td>
</tr>
</tbody>
</table>

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\textsuperscript{41} peak should be analyzed. The baseline should be calculated correctly 2\&\&+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=\&\&=30)

SFLUX = the measure of reactor flux, in Ti\textsuperscript{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:

\[
\begin{array}{|c|c|}
\hline
EG & POS \\
\hline
2F & 10.5 \\
\hline
\end{array}
\]

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.

<table>
<thead>
<tr>
<th>NGB</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
</tr>
</thead>
<tbody>
<tr>
<td>ITY</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>Sample Spectrum</td>
<td>Flux Monitor</td>
<td>Reset Flux correction</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>Type of substrate</td>
<td>Spectrum</td>
<td>Standard</td>
<td></td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>is</td>
<td>Standard</td>
<td>Spectrum</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>POL SOR POR</td>
<td>No Multiplier</td>
<td>With Multiplier</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*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 $\mu g/m^3$)

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 $\varepsilon=0.0011$, the linear coefficients with the intercept = -0.207 channels in this case and slope = 1.0037 channels/KeV. The five output columns are

\[ \begin{array}{cccc}
E & C_o & C_c & \Delta C \\
\end{array} \]

\[ \begin{array}{cccc}
E & = \text{isotope gamma energy} \\
C_o & = \text{observed peak position} \\
C_c & = \text{peak position-calculated from the best fit} \\
\Delta C & = C_o - C_c \\
\end{array} \]
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 uncertainly (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 NBC=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 = 0.001*5.3 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 µg/count. (S is entered in mg/count so that the output units for this case is µg/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

\[ W = \frac{CT \times FK - FTR \times BL}{CONV} \]

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

\[ DW = \sqrt{\left( \frac{\partial W}{\partial FK} \right)^2 \left( \frac{\partial W}{\partial CT} \right)^2 + \left( \frac{\partial W}{\partial BL} \right)^2 \left( \frac{\partial W}{\partial BL} \right)^2} \]

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 (µg/count)

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

FKS = (SM + ASH*FTR)/CT

and DFS = \[ \sqrt{\left(\frac{\partial FKS}{\partial ASH}\right)^2(\partial ASH)^2 + \left(\frac{\partial FKS}{\partial CT}\right)^2(\partial CT)^2} \]
and it is assumed that there is no uncertainty in the value of SM or FTR.

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

\[
PCT = \text{the percent of ASH in the total activity} = 100 \times \frac{ASH}{SM+ASH}
\]

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.
A Sample Short-Irradiation Output from SPAN.

**EXTERNAL CALIBRATION:**

<table>
<thead>
<tr>
<th>Iterations</th>
<th>Value 1</th>
<th>Value 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>-0.2085E 01</td>
<td>0.1837E 01</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Value 3</th>
<th>Value 4</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0</td>
<td>0.0</td>
</tr>
</tbody>
</table>

**Control Line:**

- FLUX: 4970.4
- NSE: 1
- SF: 6000.0
- FL: 0.86078

**Tape ID: MT27**

**Tape ID: MT27**

**Energy Calibration Coefficients:**

<table>
<thead>
<tr>
<th>Value 1</th>
<th>Value 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0</td>
<td>0.0</td>
</tr>
</tbody>
</table>

**Tape ID: MT27**

**Energy Calibration Coefficients:**

<table>
<thead>
<tr>
<th>Value 1</th>
<th>Value 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0</td>
<td>0.0</td>
</tr>
</tbody>
</table>

---

**Control Line:**

- FLUX: 4970.4
- NSE: 1
- SF: 6000.0
- FL: 0.86078

**Tape ID: MT27**

**Energy Calibration Coefficients:**

<table>
<thead>
<tr>
<th>Value 1</th>
<th>Value 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0</td>
<td>0.0</td>
</tr>
</tbody>
</table>

---

**Control Line:**

- FLUX: 4970.4
- NSE: 1
- SF: 6000.0
- FL: 0.86078

**Tape ID: MT27**

**Energy Calibration Coefficients:**

<table>
<thead>
<tr>
<th>Value 1</th>
<th>Value 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0</td>
<td>0.0</td>
</tr>
</tbody>
</table>

---

**Control Line:**

- FLUX: 4970.4
- NSE: 1
- SF: 6000.0
- FL: 0.86078

---

**Control Line:**

- FLUX: 4970.4
- NSE: 1
- SF: 6000.0
- FL: 0.86078

---

**Control Line:**

- FLUX: 4970.4
- NSE: 1
- SF: 6000.0
- FL: 0.86078

---

**Control Line:**

- FLUX: 4970.4
- NSE: 1
- SF: 6000.0
- FL: 0.86078
A Sample Long-Irradiation Standard Output from SPAN.

<table>
<thead>
<tr>
<th>IND</th>
<th>ICH</th>
<th>DMT</th>
<th>NS</th>
<th>E</th>
<th>CT</th>
<th>DCT</th>
<th>PKS</th>
<th>DPE</th>
<th>CRIT</th>
<th>PCT</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>554</td>
<td>11</td>
<td>17</td>
<td>16</td>
<td>11</td>
<td>2206</td>
<td>1207</td>
<td>1208</td>
<td>1209</td>
<td>1210</td>
</tr>
<tr>
<td>19</td>
<td>1477</td>
<td>1477</td>
<td>12</td>
<td>15</td>
<td>15</td>
<td>1220</td>
<td>1221</td>
<td>1222</td>
<td>1223</td>
<td>1224</td>
</tr>
<tr>
<td>18</td>
<td>1864</td>
<td>1864</td>
<td>16</td>
<td>17</td>
<td>17</td>
<td>1860</td>
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**TAPE ID: M727**  **TGW = 0059**  **LIVE TIME = 2000 SEC**

**CONTROL LINE:**

- **IND**: 59 4 4 3 0 0 0 0 0
- **ICH**: 1.0000
- **DMT**: 1.0000
- **NS**: 0.0000
- **E**: 9.2500
- **CT**: -0.0000
- **DCT**: 1.0000
- **PKS**: 1.0000
- **DPE**: 1.0000
- **CRIT**: 1.0000

---

**Energy Calibration Coefficients**

- **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
- **PCT**: 0.0000 0.0000 0.0000 0.0000 0.0000 0.0000 0.0000 0.0000 0.0000 0.0000 0.0000 0.0000 0.0000 0.0000 0.0000 0.0000 0.0000 0.0000 0.0000 0.0000 0.0000 0.0000 0.0000

---

**Sample Data**

- **IND**: 20
- **ICH**: 554
- **DMT**: 11
- **NS**: 17
- **E**: 13.22
- **CT**: 578.02
- **DCT**: 255.8
- **PKS**: 0.94284E-06
- **DPE**: 0.1734E-07
- **CRIT**: 0.0
- **PCT**: 1119.70

---

**Note:** The table contains numerical data and is structured to represent the output from SPAN, including various columns such as **IND**, **ICH**, **DMT**, **NS**, **E**, **CT**, **DCT**, **PKS**, **DPE**, **CRIT**, and **PCT**.
A Sample Long-Irradiation Unknown Output from SPAN.

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### Additional Data

**NOTE:** All values are given in arbitrary units.
17) W.W. Black, received through private communication.


22) Computing Center Memos relating to available service routines like *MOUNT and *TAPECOPY may be obtained from the Counseling Room filing cabinet, room 1013.


List of Figures.

Fig. 1 Measurement of Linearity of the Gamma-ray spectrometer.
Fig. 2 Determination of a net peak area.
Fig. 3 Irradiation-counting schema for short-lived isotopes.
Fig. 4 Gamma-ray spectrum obtained after 3 min. cooling time.
Fig. 5 Gamma-ray spectrum obtained after 15 min. cooling time.
Fig. 6 Irradiation-counting schema for long-lived isotopes.
Fig. 7 Gamma-ray spectrum obtained after 15 hours cooling time.
Fig. 8 Gamma-ray spectrum obtained after 15 days cooling time.
Fig. 9 Flux gradient at irradiation site in irradiation bottle.
Fig. 10 Ge Li Spectra General data reduction scheme.
Fig. 11 Dependence of the calculated baseline on the moving average half-width, e.
Fig. 12 Gamma-ray spectrum showing the calculated baseline and computer-located peaks.
Fig. 13 An abbreviated flow diagram for SPAN.
Fig. 14 Sample spectrum peaks versus the statistical criterion, CRIT.
Fig. 1

Channel Deviation From 1KEV/Channel

0 0.5 1.0 1.5 2.0 2.5 MEV
Fig. 2
Fig. 3

Irradiation and Counting Scheme for Short-lived Isotopes.

Sample plus Ti Flux Monitor

\[ t_{ir} = 5 \text{ min.} \; ; \; \phi = 2 \times 10^{12} \text{ n/cm}^2/\text{sec} \]

\( t_d = 13 \text{ min.} \)
\( t_c = 20 \text{ sec.} \)

Ti Flux Monitor

\( t_d = 3 \text{ min.} \)
\( t_c = 400 \text{ sec.} \)

\( t_d = 15 \text{ min.} \)
\( t_c = 1000 \text{ sec.} \)

Magnetic Tape

\( \text{Al}^{28} \)
\( \text{V}^{52} \)
\( \text{Cu}^{66} \)
\( \text{Ca}^{49} \)
\( \text{Ti}^{51} \)
\( \text{S}^{37} \)

\( \text{Br}^{80} \)
\( \text{Cl}^{38} \)
\( \text{Mn}^{56} \)
\( \text{Mg}^{27} \)
\( \text{In}^{116m} \)
\( \text{Na}^{24} \)
Sample Gl-31
Air Vol.: 5.65 m³
L₁ = 5 min.
L₂ = 3 min.

Fig. 4
Sample Gl-11
Airport, Gary, Indiana
Air Vol.: 5.65 m³
$E_{\gamma}$ = 5 min.
$E_{\gamma}$ = 15 min.

Log Counts / 1000 sec.

$E_{\gamma}$, MeV

Fig. 5
Fig. 6

Irradiation and Counting Scheme for Long-lived Isotopes.

Ten Samples plus Standard

$t_{ir} = 2-5$ hrs.; $\Phi = 1 \times 10^{13}$ n/cm$^2$/sec

$t_d = 20-30$ hrs. $t_c = 2000$ sec.

$t_d = 20-30$ days $t_c = 4000$ sec.

Magnetic Tape
Sample Gl-11
Airport, Gary, Indiana
Air Vol.: 11.3 m³
\( t_m = 2 \) hrs.
\( t_A = 15 \) hrs.

Fig. 7
Sample Gl-11
Airport, Gary, Indiana
Air Vol.: 45.3 m³

Fig. 8
Fig. 9

IRRADIATION CONTAINER

ALUM.

FLUX IOO%_REL.

THERMAL

FAST

100%

90

80

70

60

50

40

30

20

10

0

1 2 3 4 5 6 7 8 9 10 11 12 13

10 5 0 5 10 cm
Fig. 10. A General Data Reduction Schematic Diagram

- Data Storage → List Plot
  - Peak Location
    - Multiple Peak Analyses
      - Net Peak Areas
        - Energy Calibration
          - Component Identification
            - Conversion, Corrections
Fig. 11 Dependence of the calculated baseline on the moving-average half width, $l$. 

$C_{60, 1332 \text{ KeV}}$
Fig. 12. Gamma spectrum showing the calculated baseline and computer-located peaks.
An Abbreviated Flow Diagram for SPAN.

1. Read Isotope Data Set
2. Read Internal Calibration Energies
3. Read Interval Data
4. Read Run I.D.
5. Read Spectrum Control Data
   - Source Tape
   - Inventory
   - New Tape
6. Acquire Desired Spectrum from Magnetic Tape
7. Calculate Baseline
8. For each stored gamma energy appropriate to the specified run type, ITY, calculate:
   - Net Peak Area
   - Statistical Data
   - Interference Terms
9. Update long irradiation conversion factors.
   - Print Out
10. Short Irradiation
11. Flux NBG = 4
    - Normalize short irradiation conversion factors to the present flux measure.
12. Print Out
13. Calculate Total Weight
14. Make Appropriate Blank Subtraction
15. Calculate Statistical Quantities
16. Print Out

Fig. 13.
Fig. 14 Sample spectrum peaks versus the statistical criterion, crit. (Arrows indicate expected peak positions.)
The Main Tape Inventory Program INVEN:

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<td>Print contents of tape being read</td>
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<td>2</td>
<td>CONTINUE</td>
<td>Continue reading tape</td>
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<td>3</td>
<td>CALL Sll/10,1,2</td>
<td>Load Sll/10 into memory</td>
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<td>WRITE 16,50</td>
<td>Write contents of file 16</td>
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<td>STOP</td>
<td>End of program</td>
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Other relevant lines include:

- Line 24: Go to 16
- Line 25: Call Sll/10,1,2
- Line 26: Write file 16 contents
- Line 27: Continue reading tape

The program uses various subroutines for handling tape reading and file operations.
The RDLST and BASE Programs:

```
Programs:

MM_MIL_14
1077 M_MIL_14
75 Subroutine

1044 Subroutine

1043 Subroutine

1042 Subroutine

1041 Subroutine

1040 Subroutine

1039 Subroutine

1038 Subroutine

1037 Subroutine

1036 Subroutine

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A Simple Main Plotting Program:

FORTRAN IV G Compiler

```
DOUBLE PRECISION BS(4096)
DIMENSION SR(4096), CH(4096)
DIMENSION IR(4096)
FTY = 2.0
FTX = .0025
XMX = 66.
CALL PLTXMX(XMX)
XST = 11.0
999 CONTINUE
READ(5, 42) NREC, ITG, NCUT, NSK, NCR
CALL SKIPCO, NSK, 4
420 FORMAT(514)
IF (NREC .GT. 05, B55, 717)
717 CONTINUE
NR = NREC - 1
478 CONTINUE
CALL RTAPEINR, I3, IT, IR
IF (MVLT(I3) - ITG) 978, 799, 805
805 STOP
799 CONTINUE
XST = XST + 11.0
XND = XST + 10.0
CALL PENUP(XST, 0.5)
CALL PENDP(XND, 0.5)
CALL PENUP(XST, 0.5)
CALL PENDP(XST, 0.5)
CALL PENUP(XST, 9.01)
DO 1 J = 1, ACUT
SR(J) = IR(J)
IF (SR(J) .LT. 2.2) 2, 3
2 CONTINUE
SR(J) = 0.5
GO TO 987
3 SR(J) = 0.5 + FTY * ALOG10(SR(J))
987 CONTINUE
U = J
1 CH(J) = U + FTX * XST
CALL PLINE(CH(J), SR(J), NCUT, 1, 0, 0, 1)
IF (NC1998, 999, 998
998 CONTINUE
DO 51 J = 1, NCUT
BS(J) = SR(J)
51 CONTINUE
T = BS(J)
IF (T - 1.0152, 52, 53
52 T = 1.0
53 CONTINUE
SR(J) = 0.5 + FTY * ALOG10(T)
CALL PLINE(CH(J), SR(J), NCUT, 1, 0, 0, 1)
GO TO 999
END
```
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Isotope Component Identification Programs:

FUNCTION (FIA)

RETURN

END
Non-linear Least Squares Routines:

SUBROUTINE NLLS(X1,Y1,LI,G,P,SE,VAR,ERR,EPS,NIT,NP,KI)

C NLC-LINEAR LEAST SQUARES FITTING ROUTINE
C DATA PARS X1,Y1,G DESCRIPTED BY A CONTINUOUS AND DIFFERENTIABLE FUNCTION
C WHERE THE PARS ARE ADJUSTABLE PARAMETERS. NLLS STARTS AT GUESSES
C FOR G(J) AND RETURNS *BEST FIT* VALUES FOR THEM WHICH MINIMIZE THE
C SUM OF THE SQUARES OF THE DIFFERENCES BETWEEN Y AND F.
C NO<0 OF CATAPOYNTS X1,Y1,J1
C K<6 STATISTICAL WEIGHT ASSOCIATED WITH Y(J1). -1/STD. DEVIATION**2
C (G(J)=-G(J) /ERROR IN PARAMETER G(J))
C C<5 VARY PARAMETER COMMUNICATING WITH ARG
C P<10=TRUE LEAST SQUARES FIT, FALSE UNLESS DIFFERENTIABLE MATRIX =0.0, THEN ERR=1
C W<0 OF ITERATIONS ALLOWED
C K<6=0 ACTUAL NUMBER PERFORMED
C EPS<0 ALLOWED FRACTIONAL ERROR IN EVERY G(J) TO TERMINATE ITERATION
C NP<10 OF PARAMETERS G(J)

DOUBLE PRECISION T20.20

DIMENSION X1(100),Y1(100),G(20),SE(20),P(10)
DIMENSION C(20)

EPS=0.0
DC I=1,10
DC IP=1,2,1
2  W=1.0, 1 CONTINUE
END

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IV C COMPILER ANL 12-31-69 11126.11 PAGE 0001

SUBROUTINE ARGIV,V1,G1,P1

C PERMISION G(21), G(21), P(100)
N(1)
V=G1)
C=1.0, 1 CONTINUE
END

MEMORY REQUIREMENTS 0001 FA BYTES
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