

NEUTRON ACTIVATION ANALYSIS OF  
PLANTS AS POLLUTION INDICATORS

by

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Thesis submitted to the Graduate Faculty of the  
Virginia Polytechnic Institute and State University  
in partial fulfillment of the requirements for the degree of

MASTER OF SCIENCE

in

Nuclear Science and Engineering

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May, 1972

Blacksburg, Virginia

## ACKNOWLEDGEMENTS

The author wishes to express special appreciation to Dr. A. Keith Furr, his thesis advisor, for the many suggestions and aid provided during the course of this study. The author wishes to thank Dr. Milton C. Edlund and Dr. Andrew Robeson for serving on his committee.

The author would like to recognize \_\_\_\_\_ for his help in operating the reactor on weekends and \_\_\_\_\_ for his assistance in obtaining and preparing samples for analysis. He wishes to acknowledge the computer program originally written by \_\_\_\_\_ from which the present NAA program was obtained after many changes. He also wishes to express his gratitude to \_\_\_\_\_ for his assistance in the problems of programming and in the maze of Job Control Language required to run the program. Finally, the author wishes to express his gratitude to \_\_\_\_\_ for his assistance in operating the reactor and for his moral support throughout the course of this study.

The author wishes to thank \_\_\_\_\_ for her efforts in typing the manuscript.

The author would like to acknowledge the financial assistance provided by the Atomic Energy Commission in the form of a Traineeship under which all of this work was done.

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## I. INTRODUCTION

Neutron Activation Analysis has been developed recently to be an important tool in analysis of trace constituents in samples. However, most laboratories have concentrated on comparative measurements of a related group of standards to the same elements in the samples under investigation. In the VPI&SU Neutron Activation Analysis Laboratory, the approach has been to develop a program which will yield quantitative results for any element in which an isotope can conveniently be made radioactive by thermal neutron bombardment. In order to increase the acceptance of this technique, the program development in the present work has been carried out in conjunction with an experimental program of current interest, the problem of pollution due to automobile fumes. This work has been carried on in conjunction with the development of the NAA computer program. The data from this work, in addition to much other data, has provided excellent realistic test data for the program.

Environmental pollution has been a major area of study for many years. The effects of air pollution on vegetation have been studied quite extensively.<sup>1-9</sup> One aspect of this topic that has been studied only to a limited extent is that of using plants as indicators of air pollution. One such study<sup>10</sup> used plants as an indicator of the amount of fluorine emitted from an industrial site. The procedure used in this study was to determine the amount of fluoride concentration in the plants, and, from these results, obtain information about the fluoride emission rate for that site. A slightly different approach

is that of observing the affected plants in a qualitative manner instead of a quantitative one. Such an approach has several limitations.<sup>11</sup> Firstly, by observation of vegetation, only five major pollutants have been identified, namely sulfur dioxide, ozone, peroxyacyl nitrates (PAN), fluoride, and ethylene. Secondly, there are dependencies upon the existence of susceptible varieties, at a susceptible stage of growth, at the time of exposure which is sufficiently intense to affect them. Thirdly, and most important, the presence of a competent observer is needed in the field at the appropriate time to observe the symptoms after they have developed and before they have become obscured by other changes. The Tennessee Valley Authority (TVA) conducted a surveillance program of the vegetation around several of its coal burning plants.<sup>12</sup> Using sensitive natural indicator species, the pattern of injury associated with a given power station was found to be dependent upon the dispersion pattern for the power station. Thus, a knowledge of the dispersion pattern proved a useful tool in determining where and how much vegetation damage to expect under given conditions.

The study done in this work is a quantitative one, but only in a comparative sense, because absolute concentrations of pollutant elements found in vegetation are virtually meaningless. This study is similar to another done by R. H. Daines<sup>13</sup> which involved the concentration of lead in vegetation as a pollution indicator. The accumulated data of this study seemed to indicate that "the lead in the air along highways is related to the volume of automobile travel and



distance from the highway."<sup>14</sup> Both plant and soil samples were taken under controlled conditions. The plants were grown in acid washed sand in a greenhouse and at various stations along highways. The pots which contained the plants and soil were sealed so as to admit only filtered air.

A second study<sup>15</sup> was done in Los Angeles, California in contrast to the above study by R. H. Daines. In this study, annual bluegrass and petunias were used as monitors in an air pollution study. These plants were grown under controlled conditions and then taken to sampling areas for 24 hour periods. The plants were studied at the laboratory in filtered chambers for symptom development. The results of the study are stated as follows: "The injured leaves were measured and showed poor correlation with oxidant levels, suggesting that the unmeasured factors were affecting sensitivity or that the oxidant levels did not accurately measure the phytotoxic substances in the air."<sup>16</sup> A comparison of these two studies shows that using plants as indicators of pollution is not a well-defined procedure.

The study represented by the present work also makes use of plants as indicators of air pollution. The plant samples were taken at discrete distances away from the highway. The highways that were sampled ranged from the Pennsylvania Turnpike, an old, well-traveled highway, to a section of Virginia Interstate 81, which was recently opened to the public.

The most notable difference between the above mentioned studies and the present one is that the present one attempts to demonstrate

that natural growth may be used rather than especially grown plants. All plants sampled were taken from along highways without any previous control over the plants. The object of this approach is to develop a pollution indicator which can be used without any prior preparation of the plant samples. A further criteria with regard to the sampling areas is that they be located in rural areas only. The intent here is to eliminate all forms of air pollution other than that of automobile exhaust. Great care must be exercised in analyzing the data obtained in such a study as this because of the large number of parameters involved. Consider, for example, the possible variation in soil composition in a single sampling section along a highway. Plant samples were taken at discrete intervals away from the highway. The soil adjacent to the highway is often fill that is foreign to the immediate area. Farther away from the highway the soil may be a part of a farm and may contain fertilizer. Such a possibility is feasible since all samples were taken in rural areas, many in farm fields. It is also probable that soil types will vary drastically along different highways and in different parts of the country. A trend that was seen by Daines<sup>17</sup> in his study of lead concentrations in plants along highways was that the lead content in the leaves is more closely related to the lead content in the air and the content in the below-ground parts were more closely related to the lead content in the soil. For this reason, the analysis in the present work was limited to the above ground portions of the plant since natural soil conditions prevailed.

As stated above, the experimental program was carried out in

conjunction with the Neutron Activation Analysis Program. Neutron Activation Analysis would appear to be a valuable technique for an experimental program depending upon quantitative analysis because of its capabilities. Among the features of this technique are:<sup>18</sup>

1. It is a non-destructive method of analysis.
2. It requires little or no preparation.
3. It can be completely automated.
4. It is sensitive to a wide range of elements.
5. It is accurate and gives results that are highly reproducible.
6. The time required for a complete analysis of all elements seen is relatively short.
7. All elements, to which the method is sensitive, can be seen simultaneously.
8. The problems of contamination and interfering elements can be greatly reduced with the simplified sample preparation and the use of Ge(Li) detectors.

However, before advantage can be taken of this impressive list of features, a functional computer program must be available. A description of this program developed during the course of this work and the results obtained by the use of this program represent the remainder of this thesis.

## II. ANALYTICAL PROGRAM

The Neutron Activation Analysis computer program presently being used is a FORTRAN IV program written for the VPI&SU IBM 370 digital computer. It requires approximately 200K bytes of memory on the machine. A flow chart for this program is shown in Fig. 1. This program was written for use with a 1000 channel spectrum gathered from a Harshaw Ge(Li) detector. Only 1000 of the 4000 channels has been used on the Intertechnique multichannel analyzer due to a previous limitation of data transfer methods from the NAA laboratory to the computer. Prior to the use of magnetic tapes for transfer and storage of data, computer cards were used for this purpose. These cards were punched automatically on an IBM 26 keypunch machine which restricted the use of the multichannel analyzer to only 1000 channels due to a time limitation, i.e., nine minutes per 1000 channels. This situation has been remedied with the magnetic tape's requiring less than a second for data transfer of 1000 channels. Conversion to the use of all 4000 channels is pending the implementation of a more sophisticated peak-identification routine.

### Program Segmentation

The program consists of two steps. The first step reads in the elemental library and stores this basic data on disk. The second step contains the main program and subroutines. The main program reads in the time cards and the spectra from the magnetic tape. It matches up the tagwords from the spectra on tape with the corre-

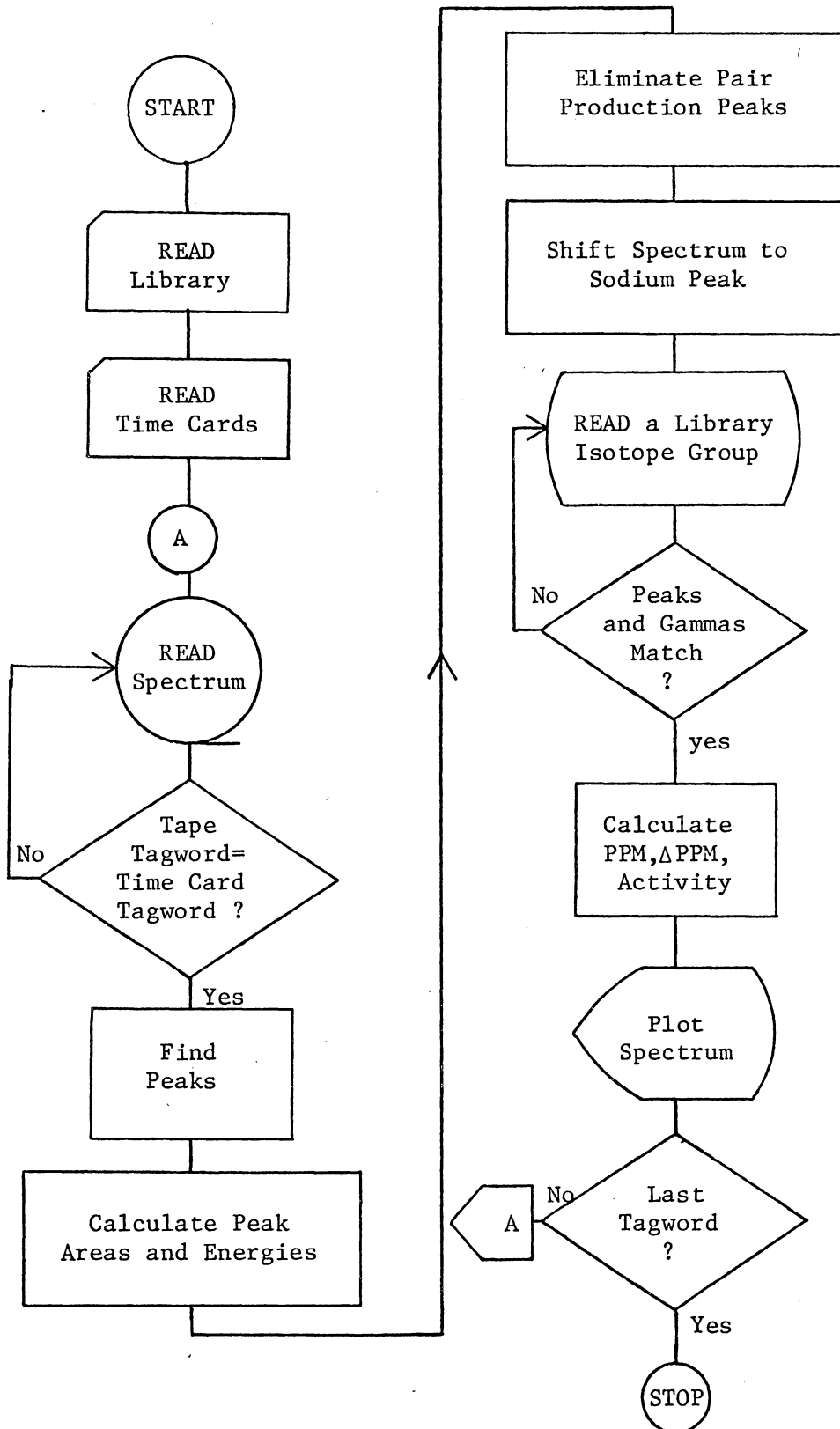


Fig. 1 Flow Diagram of NAA Computer Program

sponding tagwords from the time cards. The flow of operation is controlled in this program.

The first pair of routines, ICHANG and IC, prepares the data read from the tape for use in the program. The data is taken from the tape in packed BCD form and converted to hexadecimal by the assembler language routine, ICHANG; the function, IC, converts this data from hexadecimal to base ten form.

The first of the two major subroutines is IDEN. It finds the peaks in the spectrum and calculates their areas. The program steps through each of the one thousand channels in turn and searches for the peaks. A peak is initially found by having the sequence of at least two increasing points, i.e., each point being greater than its predecessor. The program continues stepping up the channels until the maximum and upper minimum points of the peak are found. The upper and lower minimum points of the peak are determined by either or both of the following two conditions: (1) the minimum points are truncated at a maximum of six points on the low energy side and of three points on the high energy side of the peak, and (2) a peak extremum is established at a certain point if the decrease in difference from that point to the next point is less than twenty per cent of the difference with that of the previous point. The limits used in the first criteria are based on the characteristics of the detector. Once these limits for each peak in the spectrum are established, a background line is determined in order to calculate the area of the peak. The line extends between the two minimum points of the peak. The peak area is

the total number of counts included in the background line. The statistical error in the peak area is the square root of the sum of the total number of counts and the number of background counts. In order to eliminate true peaks from statistical ones, there is a criterion in the program which throws out peaks whose areas are less than their error values.

The peak finding routine in IDEN is the most important segment of the program. This routine must be general enough to find peaks having many different shapes. The existing routine finds most peaks in spectra but it does have definite limitations in both finding the peaks and calculating their areas. The most notable limitation of this routine is that of resolving different peaks lying very close together. This routine has a minimum resolution capability of no less than one channel between peaks. The two increasing-point criterion for a peak is very inflexible and will reject some very narrow peaks. The method used for calculating the peak areas in this subroutine is very simple. The conversion to 4000 channels will greatly enhance the resolution of the peaks, but it has been found that many valid peaks are omitted due to statistical fluctuations in the broad tails of the peaks.

From subroutine IDEN, another smaller routine, GAUSS, is called to calculate the exact energy of each peak. The maximum and two adjacent points of the peak with their background values subtracted out are fitted to a Gaussian curve of the form  $y = Ae^{-(x-\bar{x})^2/s}$  where A is the amplitude, s the square of the standard deviation, and  $\bar{x}$  the

mean energy value of the peak. The energy values,  $X$ , which are determined by empirically noting the channels,  $I$ , containing calibration peaks are represented by  $x = 3.1681 \times I + .00677(I - 870)$ . This is only a close approximation since the gain of the system is not completely linear. However, the maximum discrepancy is only about 5 keV, partially due to errors in the literature values of the energy, so an acceptance window of 6 keV is employed. An area of the peak could be derived from these Gaussian fits, but since only three points are involved, the validity of these areas is suspect. When the entire 4000 channels is employed, more points can be used.

Pair production peaks are eliminated in the subroutine TRAPP. Peak energies 0.51 and 1.02 meV below any peak are set to 30,000 keV and are eliminated in IDEN when their energy values are determined to be greater than 10,000 keV. Both TRAPP and GAUSS can be easily bypassed in the program by converting their respective "CALL" statements into comment statements. Both of these statements are located in IDEN.

The second and final major subroutine, FIT, determines the elements contained in each sample and determines their parts per million and activity. A sodium peak is initially sought at  $2754 \pm 20$  keV, and if found, the entire spectrum energy calibration is shifted to position the sodium peak exactly at 2754 keV. Isotope gamma ray energies are then compared with each spectrum's peak energy value to determine the presence of an isotope. The existence of an isotope is confirmed by obtaining a one-to-one correspondence between each



library gamma energy of the isotope and a peak energy in the spectrum. It should be noted that the library does not contain gamma rays from any of the secondary daughter products. The number of parts per million and the activity for each gamma ray of the isotope is calculated by the equations:

$$\text{PPM} = \frac{\text{Area} * M \lambda}{f \epsilon b \sigma \phi W * .602} \frac{10^6}{(1 - e^{-\lambda t_a})(1 - e^{-\lambda t_c})e^{-\lambda t_{w60}}}$$

and

$$\text{ACT} = \frac{\text{Area} * 10^{-4}}{t_c * \epsilon * 60 * 3.7}$$

Also, the efficiency can be represented by the best empirical fit to a number of well known standards. The fit was derived from a non-linear fitting routine<sup>19</sup> and is given by

$$\epsilon = 8.9136 * \exp(-7.4096 * X) + 1.7446 * \exp(-.7124 * X) + .005863 \\ + .00000144 * \exp(-.001 * X),$$

where

f = the fractional abundance of the isotope being activated as part of the natural element,

$\epsilon$  = the counter efficiency for a particular gamma of energy E,

b = the branching ratio for the gamma being detected, i.e., the fraction of the activity resulting in a particular gamma,

$\sigma_a$  = the absorption cross-section of the isotope,

Area = peak area from the spectrum,

M = the molecular weight of one mole of the element,

$\lambda$  = the radioactive decay constant for the isotope in question,

$\phi$  = the neutron flux in neutrons/cm<sup>2</sup>/sec (in units of 10<sup>10</sup>)

W = mass of the sample

t<sub>a</sub> = activation time

t<sub>w</sub> = wait (decay) time

t<sub>c</sub> = count time.

The exponential factors take into account the activation, counting and decay times of the sample. If the exponent of any factor is less than 0.003, the exponent is expanded to  $e^{-\lambda t} = 1 - \lambda t$  since the difference becomes negligible. Any exponent with  $\lambda t \geq 12$  is automatically set to one for each gamma ray of an isotope. An error value of the parts per million is also computed using the error value of the peak area.

Subroutine SPLOT plots each spectrum on the CALCOMP plotter. Error bars are also plotted every fiftieth channel. These bars have vertical dimensions equal to the square root of the number of counts above and below the point. The plot is scaled such that the point in the channel containing the greatest number of counts is located on the upper limit of the vertical axis. The number of counts, C, on the "counts" axis of the plot is related to the actual number of counts, N, by the formula  $N = 8 * C / \text{Scale factor}$ . Also printed on the plot are the tagword, activation time, decay time, and count time. This subroutine can be bypassed by converting the "CALL SPLOT" statement located in the MAIN program to a comment statement.

A detailed description of the program symbols, input cards required to run the program, and output data are given in Appendices B and C.

### III. DATA AND RESULTS

The object of this plant study is to determine if plants can be used as an effective indicator of the volume of traffic on a highway. Bromine and aluminum concentrations in plant samples have been used as the indicators in this study although the possibility of aluminum in the dust from the highway may make the latter a poor choice. These two elements were chosen as indicators for several reasons:

1. Neutron Activation Analysis showed concentrations of approximately 15 and 9 PPM for bromine and aluminum respectively, in gasoline samples. A spectrum showing the aluminum peak at 1780 keV and the major bromine peak at 777 keV is in Fig. 2.

2. Both of these elements have appeared in all the plant tissue that has been analyzed in this study.

3. The 1780 keV peak of aluminum is generally a very large and easily distinguishable peak that is free of interference from other peaks.

4. Bromine has four prominent peaks at 554, 619, 698 and 777 keV as well as many other lesser peaks. The existence of several peaks helps to verify the existence of bromine in the sample. Since the concentration of bromine can be calculated from the area of each peak, the existence of several peaks allows an average value of the concentration to be calculated along with an error value. A spectrum of one of the plant samples is shown in Fig. 3. This spectrum consists of the sum of the spectra from the long and short activations in order to show all of the peaks.

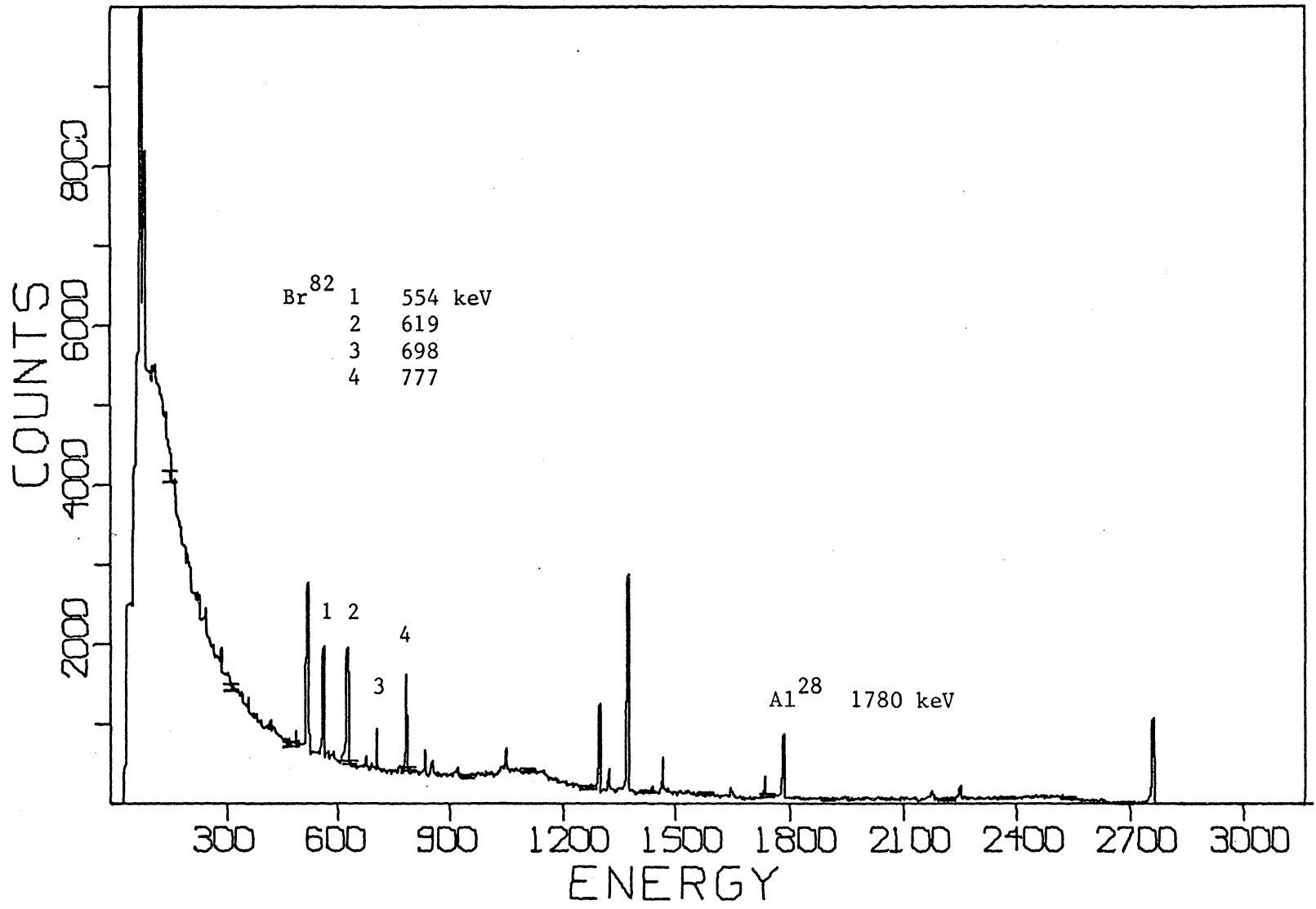


Fig. 2 Combined spectrum of a gasoline sample including the sum of the spectra from long and short activations.

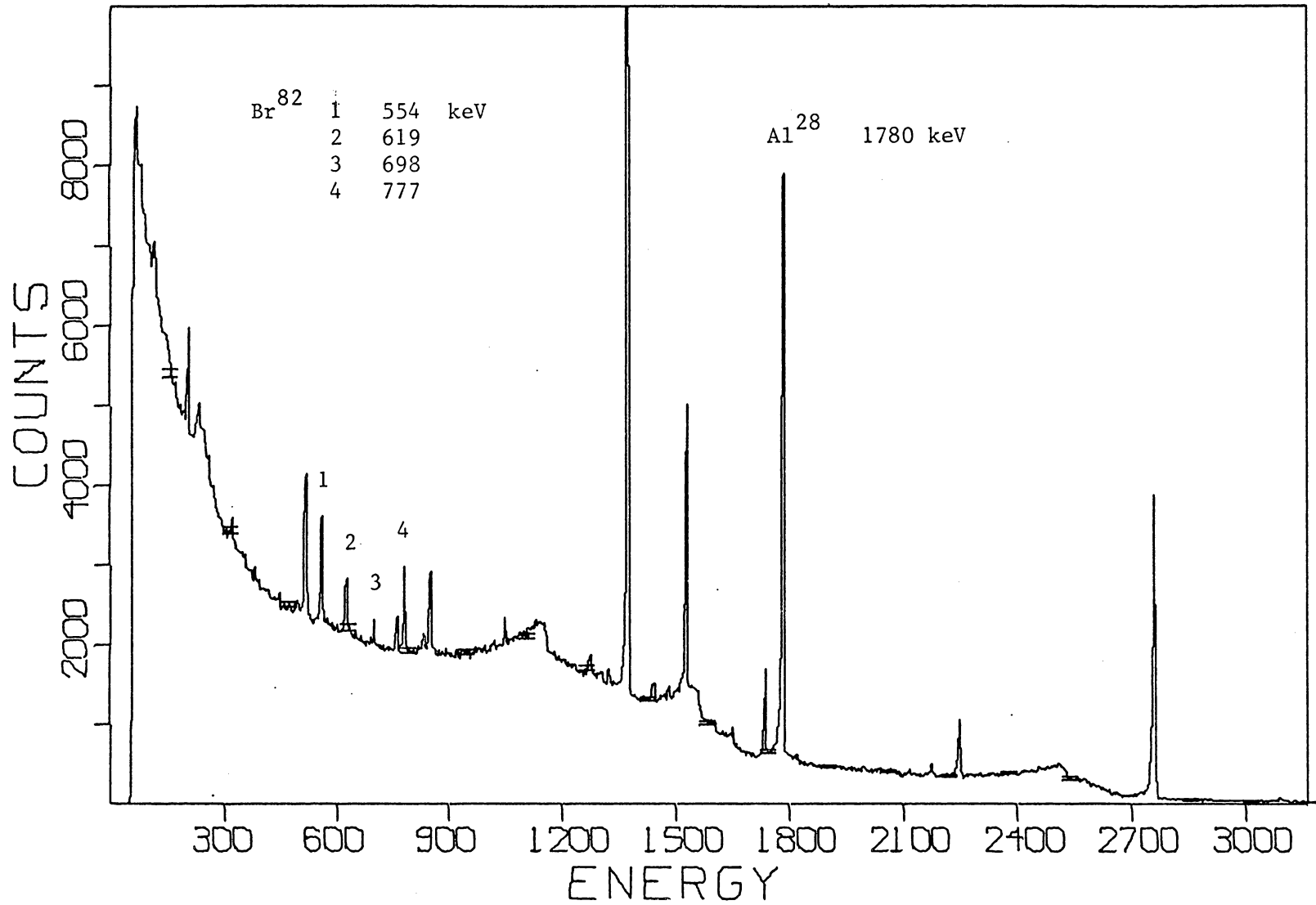


Fig. 3 Combined spectrum of a typical plant sample including the sum of the spectra from long and short activations.

Several different plant types have been used in this study. In the early stages of this study, a wide variety of plants was used with only one or two samples of each variety. This was also an attempt to determine an appropriate species. One set of samples was taken from the oldest of all the highways studied - the Pennsylvania Turnpike. The procedure was found to be inadequate because each species was not always present at every sampling location, thus making it difficult to compare element concentrations at different locations. An enlightening piece of data emerged from this set of samples which provided a much better approach to sampling procedures of that time. Bromine, vanadium, and titanium concentrations were significantly higher in maple leaf and grass samples than those in other plant samples. This result suggested the reasonable possibility that plants with large leaf areas will collect larger amounts of automobile fumes than those with smaller leaf areas.

A succeeding procedure incorporated pine needles and stems. The plants were blended and several samples were taken from the mixture. This approach was used to obtain an average concentration from the samples at each sampling location. Concentrations of the elements bromine, vanadium, and titanium were obtained from the samples. The sampling areas were located at discrete distances away from the highways. The concentrations of the elements in the samples were compared as a function of distance away from the highways. The results appeared to be random. A possible reason for this result is that, as stated above, automobile exhaust is not apt to be absorbed in pine

needles in large quantities because of the small surface area of the needles. This result is supported by the results of a study<sup>20</sup> in which fluoride levels in broad leaf grasses were consistently about 25% higher than those found for washed wire grass.

#### Experimental Procedure

Plant samples were obtained on March 30, 1972 along the highways, U. S. 460 and Interstate 81 near Christiansburg, Virginia. The samples were taken from six locations:

1. U. S. 460 in the Roanoke River Valley between Christiansburg and Roanoke on the eastern side of the highway.
2. Same location as (1) except on the opposite side of the highway.
3. Approximately 500 yards north along the highway from location (1) on the eastern side of the highway.
4. Approximately two miles in a northern direction from location (3) also on the eastern side of the same highway.
5. Interstate 81, approximately two miles southwest along the highway from the Christiansburg south exit. The samples were taken on the upwind side of the highway.
6. Interstate 81 at the south exit of Christiansburg on the downwind side of the road in a southeastern direction perpendicular to the highway.

The highway, U. S. 460, runs almost due north-south at all four sampling locations. Interstate 81 runs mainly east-west and only

slightly north-south at the two sampling locations on this highway. The prevailing wind direction in these areas is toward the south-east.

The plant samples consisted of common grass that is found growing wild in many fields and along highways. The samples consisted of both dead and green grass with the idea of obtaining a greater concentration of automobile exhaust. The samples were taken at three discrete intervals away from the highway in each case. The first group of samples was taken from an area within ten feet of the highway. These samples would not only be exposed directly to the automobile exhaust, but they would also come in contact with water as it is drained and splashed from the highway. The second group of samples was taken at a distance of approximately one hundred yards from the highway. The third and final group of samples was taken at distances of about 200 yards.

Some specific characteristics of the sampling locations are:

1. In all locations except the fourth one, the samples in the two groups at 100 and 200 yards from the highways were taken from farm fields and grazing land. The samples were picked along tree lines and paths so as to avoid the possible effects of cattle manure and fertilizer.
2. The fourth group of samples was taken on the downwind side of the highway in a tree lane through which ran electric power lines. This lane provided a wind tunnel which would probably produce greater concentrations of automobile exhaust.
3. The highway, U. S. 460, from which the first four samples



were taken had been a fairly heavily traveled highway until about four months ago. A new section of Interstate 81 was opened at that time and this new section of highway has taken most of the traffic from that section of U. S. 460. The last two samples were taken from a fairly new section of Interstate 81.

#### Preparation of the Samples

The plant samples from each location were homogenized in a blender along with a half pint of distilled water. Samples of this mixture were placed in three polyethylene vials. The blender was cleaned with soap and water between mixtures. The vials were weighed, identified, and sealed. Two of the three vials in each group contained both liquid and plant material from the mixture for all groups except the first six, namely A, B, C, D, E, and F. There was no further preparation required of the plant samples.

#### Irradiation and Counting

All samples were irradiated and counted twice, thus providing spectra from both long and short irradiations of the samples. The samples were first irradiated for a short period of time, namely one minute, in order to determine the aluminum concentration in the samples. The samples were counted for one minute after an average decay time of approximately 20 seconds between the end of irradiation and beginning of counting. The second irradiation consisted of a six hour activation for the samples with a 29 hour decay period before the counting of the first sample. These samples were counted for

five minute periods on an automatic counter with the 35.6 hour half-life  $\text{Br}^{82}$  as the isotope of interest in these spectra.

The long activation was done second in order to avoid the contribution of the long half-life elements on the short activation spectrum.

### Data and Results

The concentrations of aluminum and bromine in the plant samples as obtained through Neutron Activation Analysis are given in Tables III-A and B. This data has been plotted as a Gaussian distribution in Figs. 4-15. Each plot contains data from the samples taken from a common distance from each highway. From the concentration of the samples in each group, a mean and standard deviation were calculated. The Gaussian plots of probability vs. element concentration were drawn from these two values. The mean values and standard deviation in a condensed form are in Tables III-A and B. The results of Table III-C for both aluminum and bromine show that there is a substantial difference in the concentration of both elements between the first and next two locations away from the highway. A comparison of the concentrations at the second and third locations gives no results because of the large errors (standard deviations) in the mean values.

In Table III-B, the mean and standard deviation values were again computed with the exclusion of groups F, K, and L. An examination of Fig. 6 shows the aluminum concentrations of group F lie on the upper tail of the Gaussian distribution. This group is being excluded because the samples in this group were taken near a small stream and

Table III-A. Al<sup>28</sup> concentration of grass samples with samples taken from common distances from highway\*

Adjacent to highway	100 yards from highway	200 yards from highway
A	B	C
49.43	77.56	35.85
44.69	54.80	28.79
58.06	63.73	27.95
D	E	F
100.57	33.75	149.17
87.97	37.44	142.87
107.37	35.45	136.87
G	H	I
129.77	28.73	39.19
74.99	17.29	29.74
111.57	36.69	45.21
J	K	L
80.39	53.01	81.67
84.07	19.02	25.29
89.37	61.13	99.37
M	N	R
96.67	44.77	82.17
-	26.82	32.07
74.51	51.19	45.78
S	T	U
-	112.77	42.21
60.52	42.97	20.70
91.47	78.85	64.85

\*Groups A - L taken along Virginia 460 and groups M - U taken along Interstate 81.

Table III-B. Br<sup>82</sup> concentration of grass samples with samples taken from common distances from highway\*

Adjacent to highway	100 yards from highway	200 yards from highway
A	B	C
1.884	1.124	0.364
1.554	0.704	0.404
2.274	0.534	0.144
D	E	F
2.094	1.074	0.334
1.854	0.254	0.484
2.094	0.754	0.254
G	H	I
3.144	0.534	0.664
1.644	0.224	0.144
2.574	0.344	1.154
J	K	L
3.674	2.434	1.664
2.514	0.254	0.304
4.219	2.814	1.734
M	N	R
3.054	0.494	0.594
-	0.264	0.001
2.114	0.754	0.454
S	T	U
-	0.864	0.154
0.344	0.134	0.064
2.084	0.494	0.754

\*Groups A - L taken along Virginia 460 and groups M - U taken along Interstate 81.

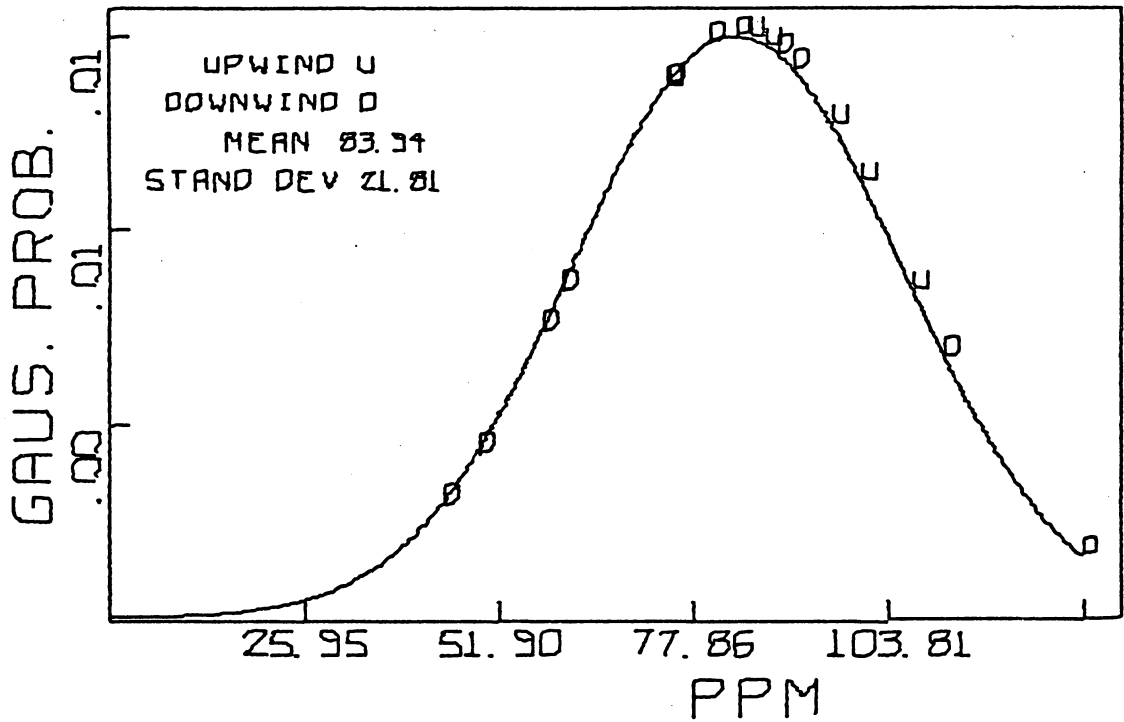


Figure 4. Probability vs.  $Al^{28}$  concentration (PPM) in plant samples taken adjacent to highway

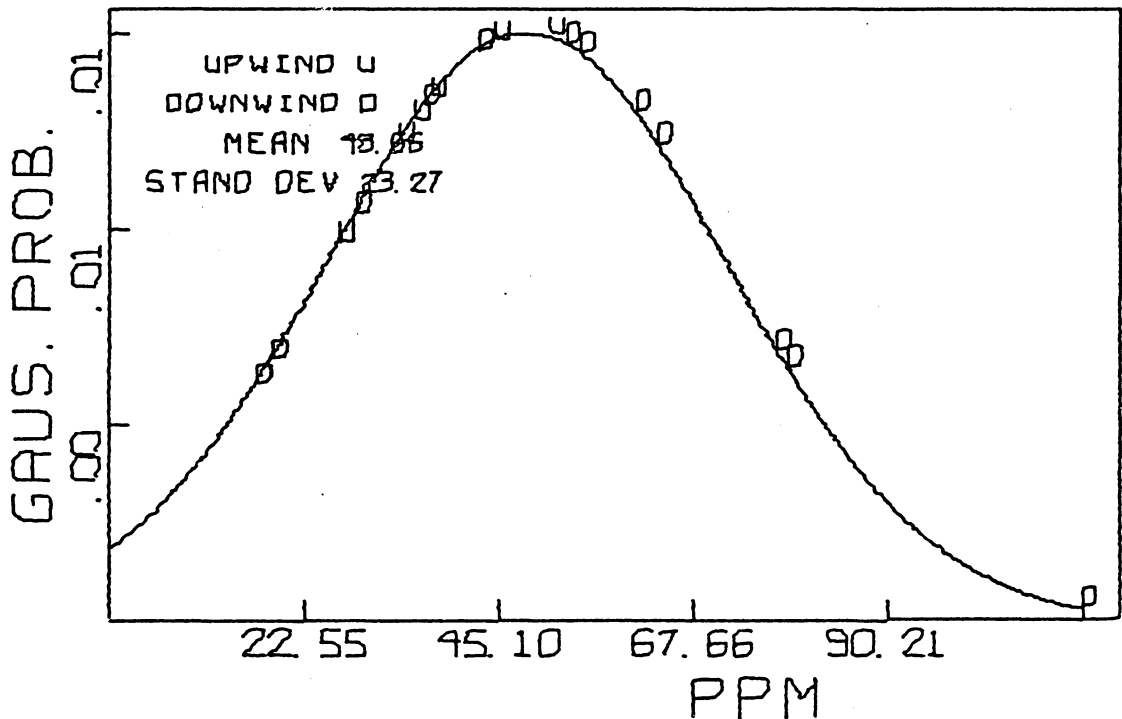


Figure 5. Probability vs.  $Al^{28}$  concentration (PPM) in plant samples taken 100 yards from highway

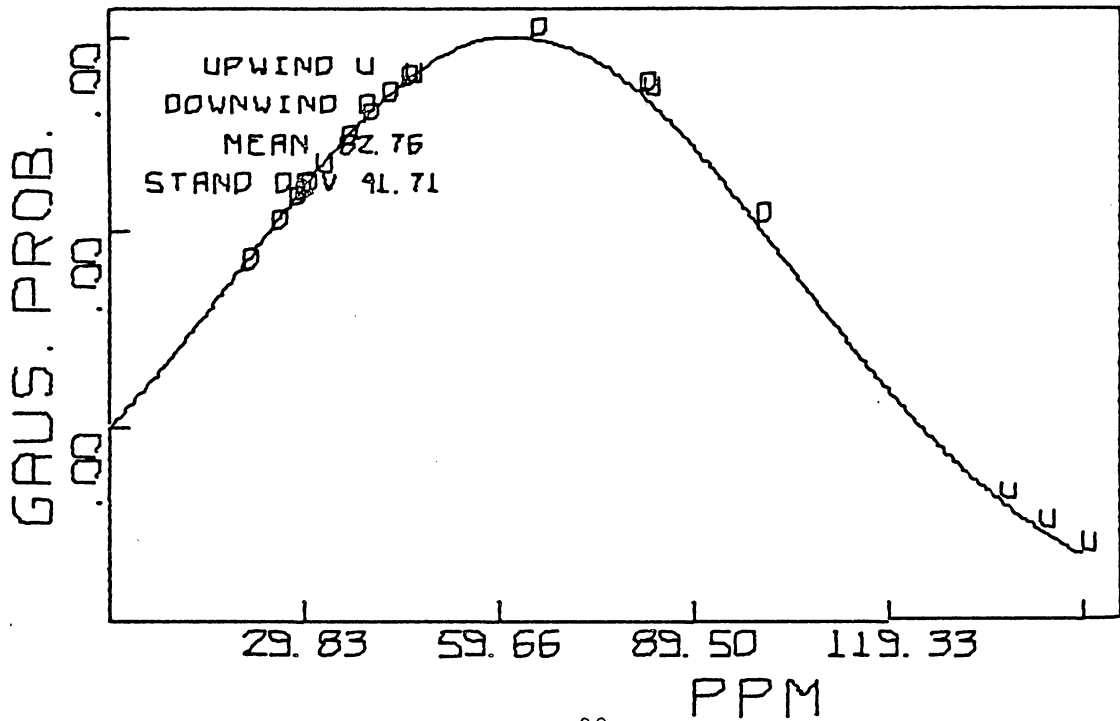


Figure 6. Probability vs.  $\text{Al}^{28}$  concentration (PPM) in plant samples taken 200 yards from highway

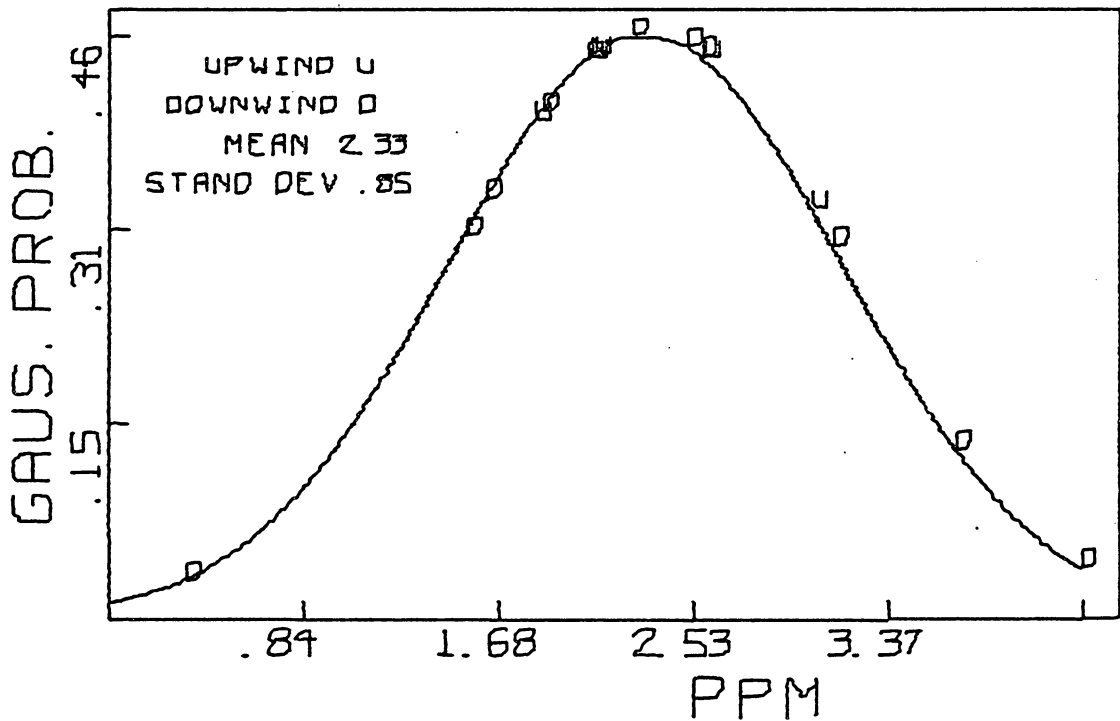


Figure 7. Probability vs.  $\text{Br}^{82}$  concentration (PPM) in plant samples taken adjacent to highway

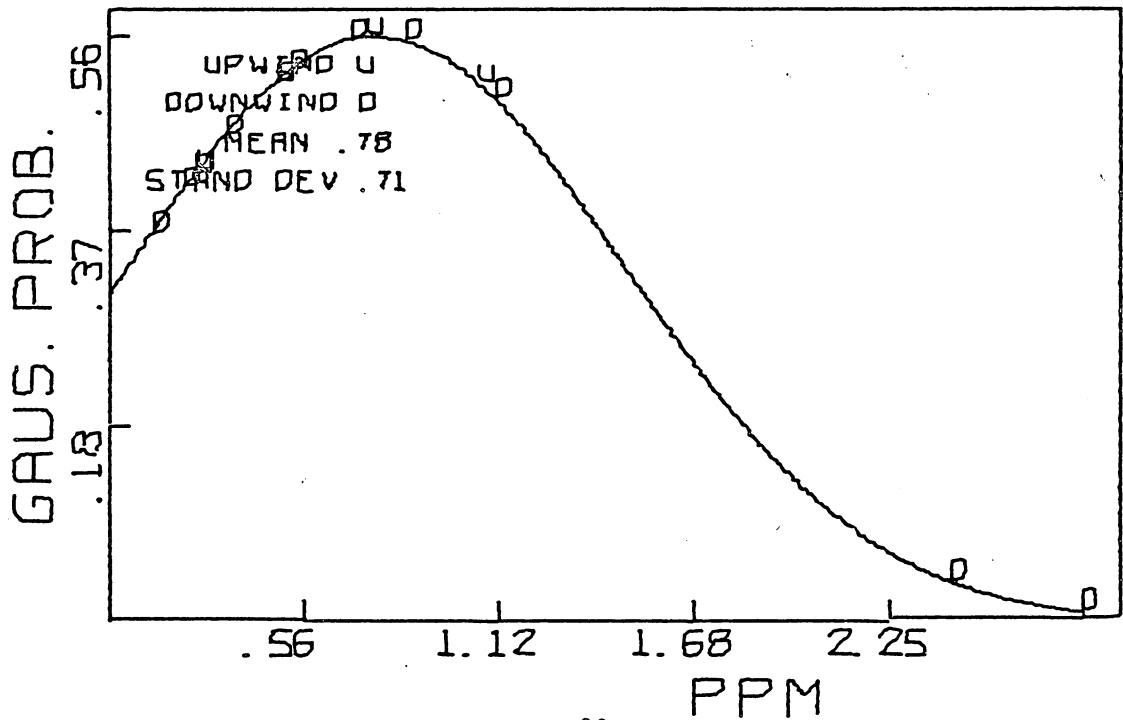


Figure 8. Probability vs.  $\text{Br}^{82}$  concentration (PPM) in plant samples taken 100 yards from highway

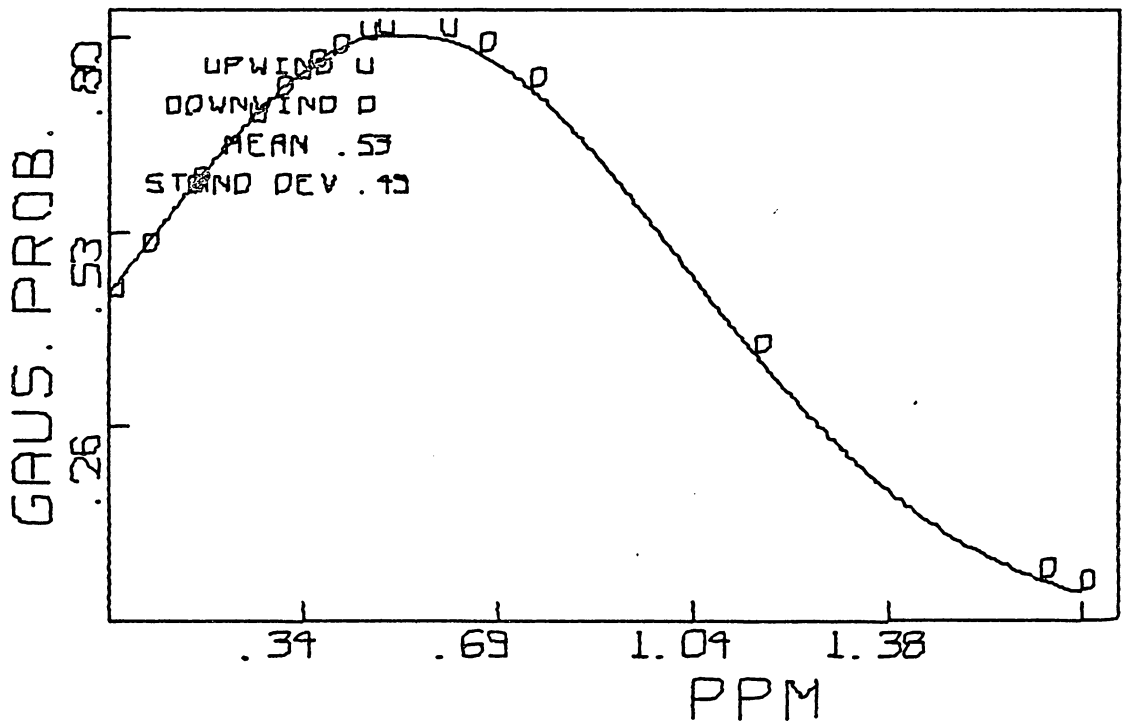


Figure 9. Probability vs.  $\text{Br}^{82}$  concentration (PPM) in plant samples taken 200 yards from highway

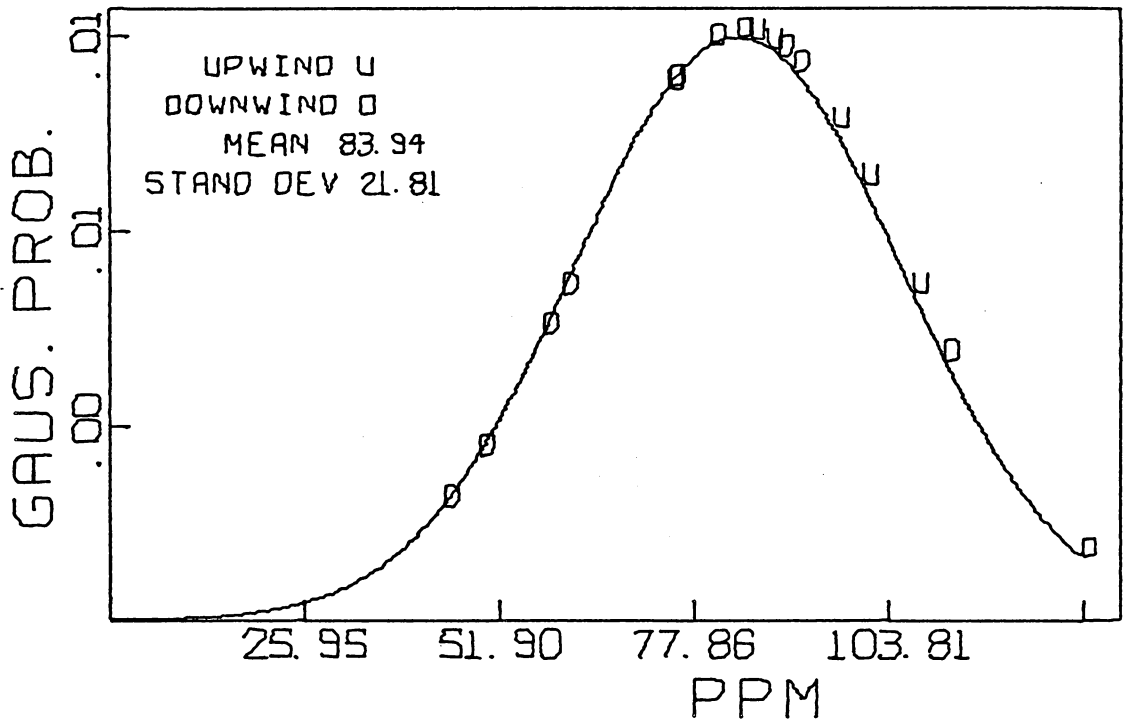


Fig. 10 Probability vs.  $\text{Al}^{28}$  concentration (PPM) in plant samples taken adjacent to highway. (Modified)

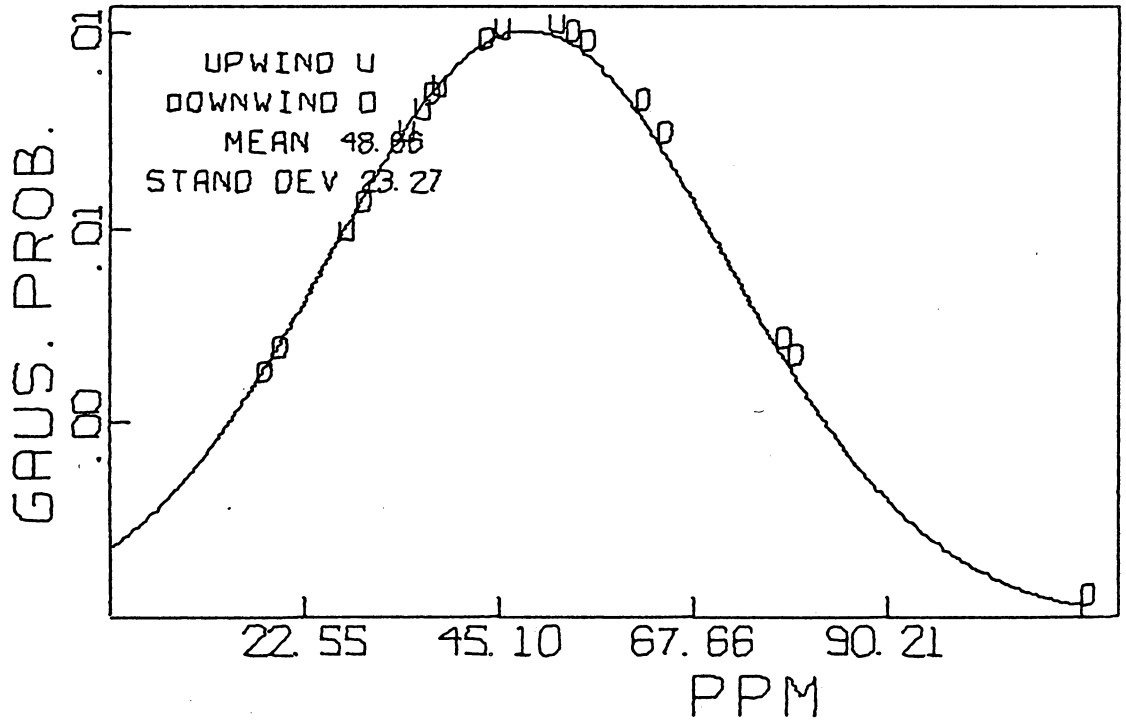


Fig. 11 Probability vs.  $\text{Al}^{28}$  concentration (PPM) in plant samples taken 100 yards from highway. (Modified)



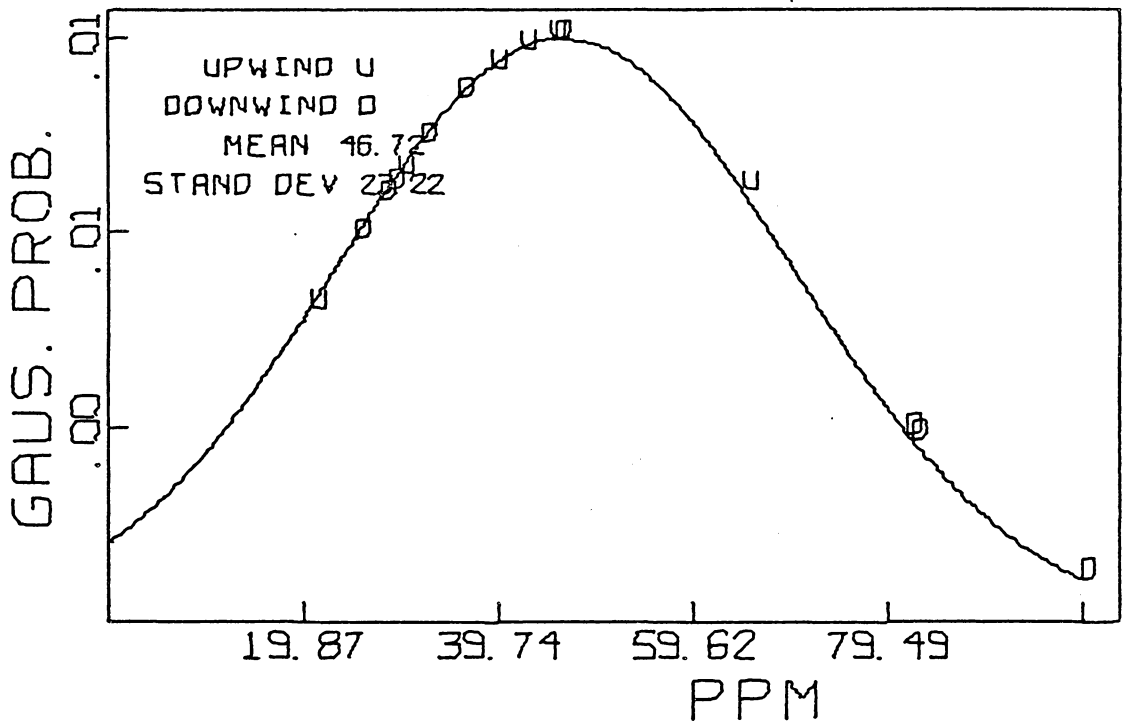


Fig. 12 Probability vs.  $\text{Al}^{28}$  concentration (PPM) in plant samples taken 200 yards from highway. (Modified)

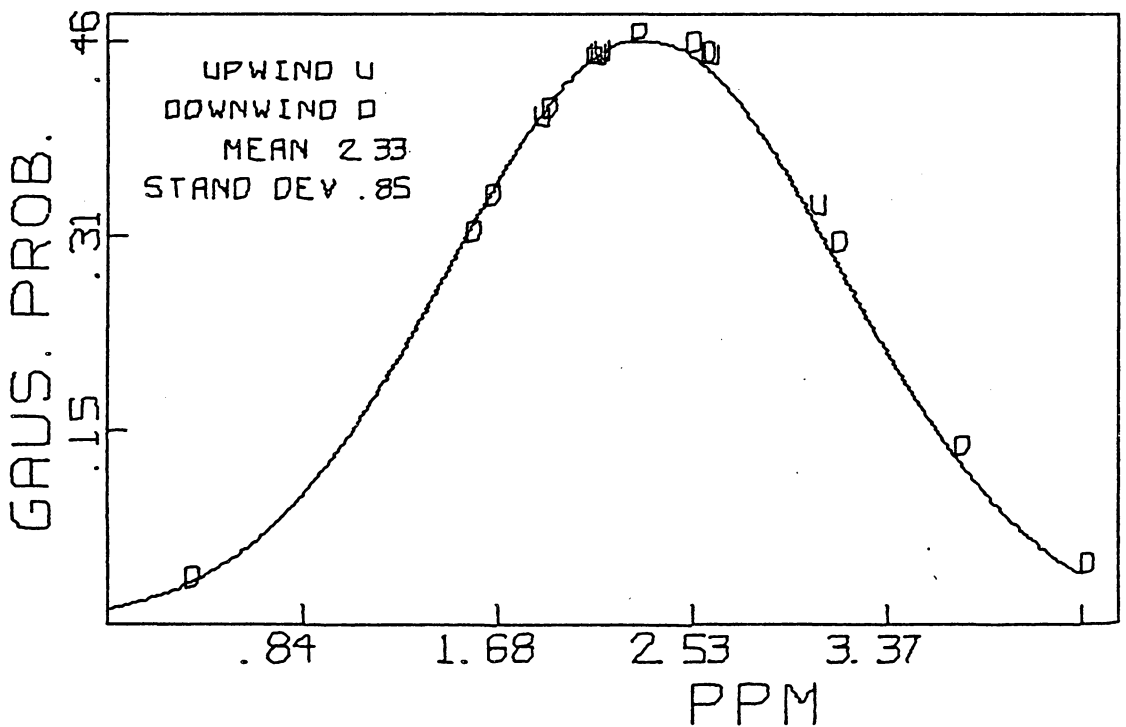


Fig. 13 Probability vs.  $\text{Br}^{82}$  concentration (PPM) in plant samples taken adjacent to highway. (Modified)

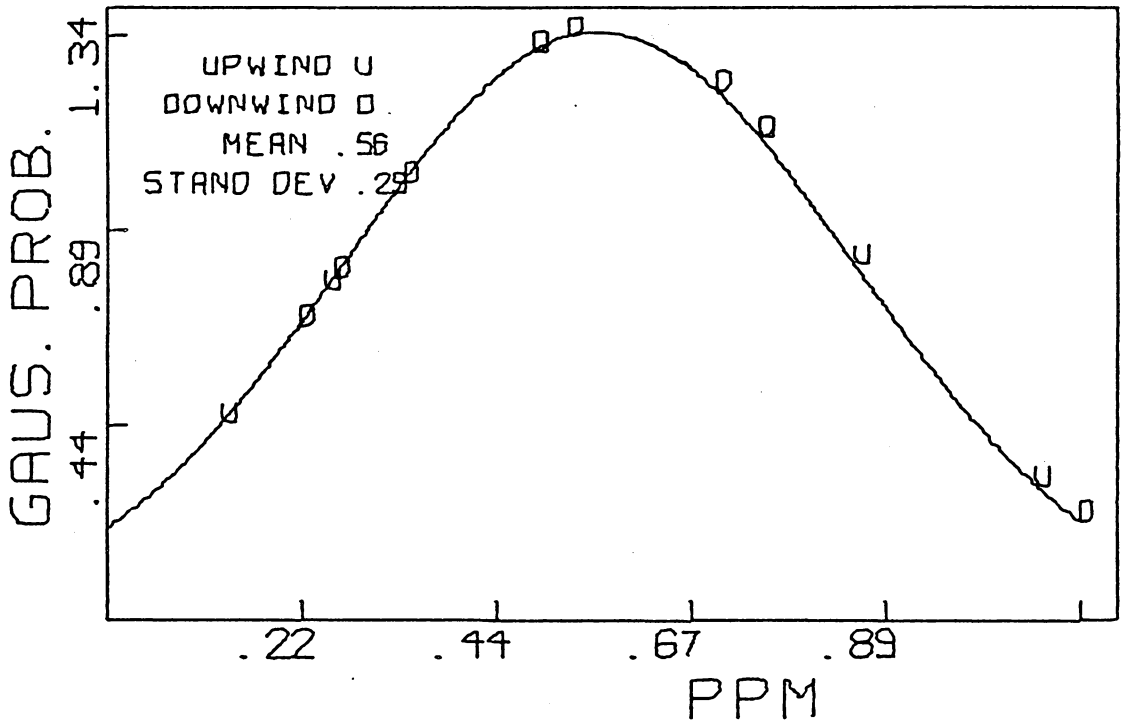


Fig. 14 Probability vs.  $\text{Br}^{82}$  concentration (PPM) in plant samples taken 100 yards from highway. (Modified)

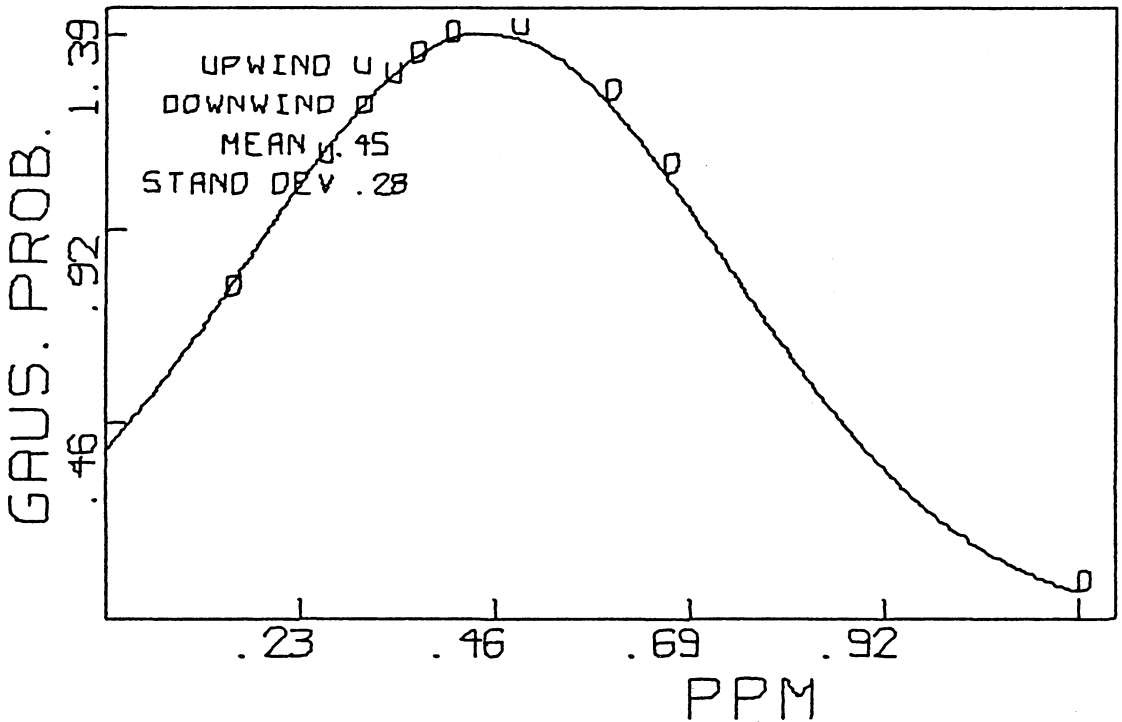


Fig. 15 Probability vs.  $\text{Br}^{82}$  concentration (PPM) in plant samples taken 200 yards from highway. (Modified)

Table III-C. Mean and standard deviation of all values in Tables  
III-A and B

---

	Aluminum		Bromine	
	<u>Mean</u>	<u>St.Dev.</u>	<u>Mean</u>	<u>St.Dev.</u>
1. Adjacent to highway	83.94	21.81	2.33	0.85
2. 100 yards from highway	48.66	23.28	0.78	0.71
3. 200 yards from highway	62.76	41.71	0.53	0.49

Table III-D. Mean and standard deviation of the values in Tables  
III-A and B excluding groups F, K, and L

---

	Aluminum		Bromine	
	<u>Mean</u>	<u>St.Dev.</u>	<u>Mean</u>	<u>St.Dev.</u>
1. Adjacent to highway	83.94	21.81	2.33	0.85
2. 100 yards from highway	48.66	23.27	0.56	0.29
3. 200 yards from highway	46.72	23.22	0.45	0.28

this stream may have contributed to this large concentration of aluminum. Such a possibility is very likely because the stream may have flooded its surrounding area, including the sampling area. Such a contribution is not the result of air contamination and will not be included in this set of data.

Groups K and L were excluded because the samples in these groups were taken in a tree lane which perhaps acts as a wind tunnel and, as can be seen in Figs. 8 and 9, the result is a large concentration with respect to the mean values. Eliminating these three groups from the data greatly reduces the standard deviation values and also changes the mean values considerably. The results in Table III-D with these modifications show a decrease in concentration at an increasing distance, but this decrease is partially obscured because of the large standard deviations.

As can be seen in Figs. 4-15, samples were plotted in two groups with respect to being up or downwind of the highway. The correlation between the two groups appears to be random.

The computer program that has used the data of this study as a test case has given very reproducible results as shown in Tables III-A and B. The concentration values given in these two tables are quite comparable within each group of three values. It should be noted that the first six groups A-F in each table contain data from samples of both plant tissue and liquid; whereas, all other groups contain data from samples with both plant tissue and liquid in the first and third vials and only liquid in the second vial. A significant difference in

concentration of both elements is seen between the first and third samples and the second sample in most cases. The results indicate that the aluminum and bromine are concentrated in the plant tissue and not on the surface of the plant.

The aluminum in the samples may not be due to automobile exhaust, but rather to the dust that lies on the highway.<sup>21</sup> The large concentration of aluminum in the plant samples may result from the scattering of dust from the highway by traffic. This dust will collect on plants near the highway but not on ones farther away. It is still possible that some of the results do indicate concentrations of aluminum from automobile exhaust, but these results are not as significant as those from the bromine data because of the presence of aluminum in dust.

#### IV. SUMMARY

The purpose of this study has been to determine if Neutron Activation Analysis can be effectively applied to a realistic study of plant analysis as a pollution indicator. The results of this study have shown that Neutron Activation Analysis of plants has determined that there is a correlation between highway traffic and the location of plants with respect to that traffic. Bromine has been found to be a good indicator of automobile exhaust pollution; whereas, aluminum is questionable because of its presence in dust that lies on highways. The aluminum may only be indicating a concentration of dust adjacent to the highway and not the concentration of automobile exhaust along the highway. An added element of importance to this study has been the fact that it has been conducted using natural growth, rather than laboratory grown plants. This fact adds to the usefulness of this procedure for determining the amount of pollution from a highway. This study has been only a preliminary one which has shown  $\text{Br}^{82}$  and possibly  $\text{Al}^{28}$  to be useful indicators of pollution. More work must be done to develop the procedures used in this study in order to make it into a more accurate and more reliable technique for the indication of automobile pollution.

This plant study as a test case for the Neutron Activation Analysis computer program has revealed many good and bad qualities of the program. The uncomplicated level of the program makes it easy for many people to use and understand it. The program has been found through extended use to yield very consistent results.

Two major weaknesses of the program are its peak finding and area calculation routines. The simplified routine in the program presently finds peaks and calculates their areas well for most cases, but there are many other cases when more refined routines are required. These are the two areas of the program that require the most improvement.

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## VI. APPENDIX A: NAA LAB PROCEDURES

The Neutron Activation Analysis Laboratory at Virginia Polytechnic Institute and State University is composed of three sections: the reactor facility, the detection and counting system, and the data processing facility.

The laboratory has facilities able to handle samples requiring both short and long activation times. Samples requiring short activation times are transferred to the reactor by means of a pneumatic transfer system, otherwise known as the "rabbit." This system is a high pressure system using nitrogen gas to force the capsules to and from the reactor. The transfer capsules presently being used are approximately 1 5/8" long by 5/8" in diameter and the samples are contained within 1/2" x 1" polyethylene vials. The use of two containers for sample transferral provides added protection against leakage or breakage. The use of polyethylene vials has several advantages: the elements in polyethylene have low activities, the polyethylene is soft so that some identification can be written on the vial with a scribe, and it has a low melting point so that the vials can easily be sealed with a soldering gun.

The capsules are transferred to and from the reactor with approximately 35 pounds/in<sup>2</sup> of nitrogen gas. The transfer time is about one second or less each way. The "rabbit" tubing goes into the core through the south-eastern off-center stringer. The neutron flux in this stringer is  $1.2 \times 10^{12}$  neutrons/cm<sup>2</sup>/sec.

The "rabbit" facility allows elements in samples to be activated

and found with minimum half-lives of an order of magnitude of as small as one minute. This minimum half-life time limit is imposed as a result of the time of transfer between the sample receiving station (from the reactor) and the detector. An average of 25 seconds is lost between the times of the arrival of the sample at the receiving station and the starting of the counting system. This situation leaves a great deal of room for improvement in the system so that elements with half-lives in the seconds can be found.

Samples requiring long period activations are inserted in either of two places in the reactor - the south beam port or the central stringer. There are other experimental facilities in the reactor such as the thermal column, north beam port, shield tank, and the remaining three off-center stringers - but these are seldom used for neutron activation work. The flux in the south beam port is  $0.67 \times 10^{12}$  neutrons/cm<sup>2</sup>/sec. Numerous samples may be irradiated at one time in this facility. The central stringer provides the experimental facility with the largest flux -  $1.3 \times 10^{12}$  neutrons/cm<sup>2</sup>/sec.

The detection and counting system consists of the detector, pre-amplifier, linear amplifier multichannel analyzer, and peripheral devices. A block diagram of this system is shown in Fig. 16. The detector is a Harshaw solid state Ge(Li) unit with a rated resolution of less than 3.5 keV at moderate counting rates.

The preamplifier and linear amplifier input the signal from the detector and amplify and shape the signal for input to the multichannel analyzer. This unit is an Intertechnique 4000 channel analyzer.

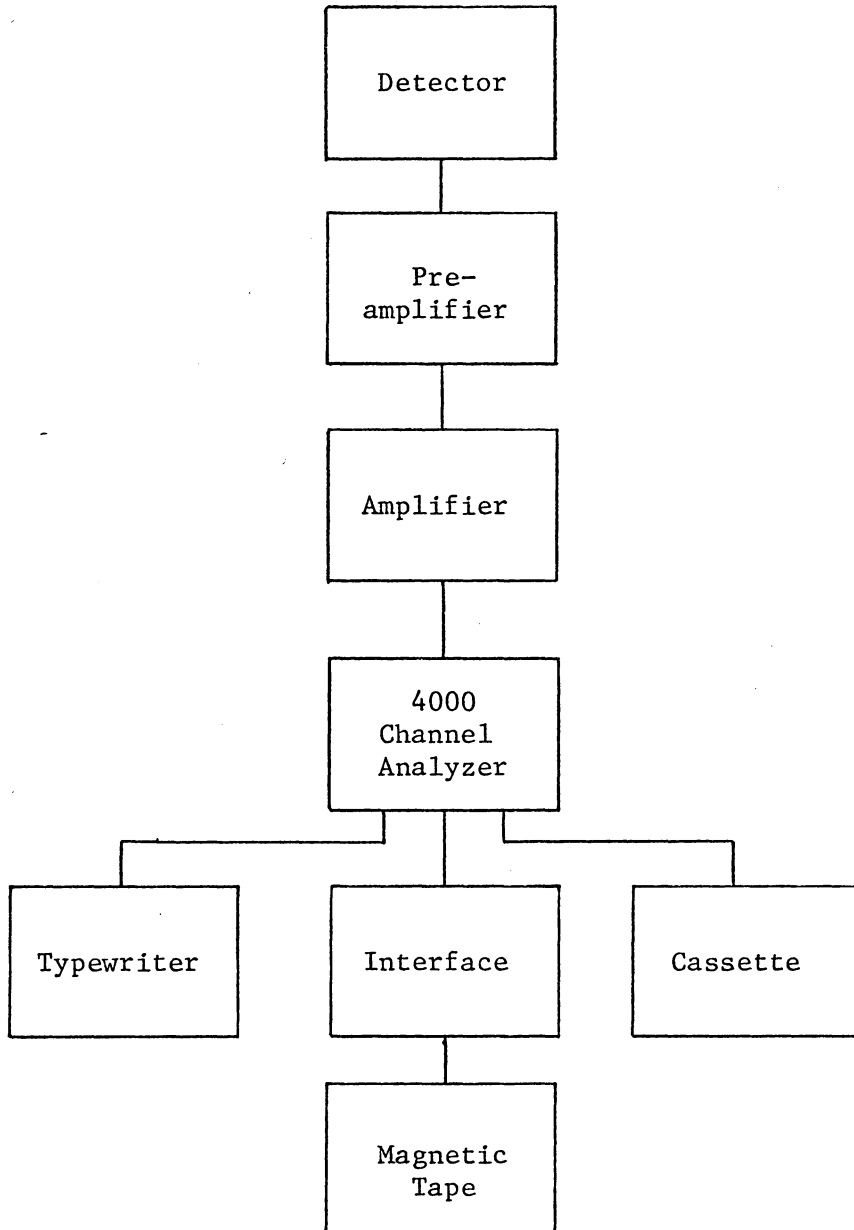


Fig. 16. Block Diagram of NAA Counting System

Presently only 1000 of the 4000 channels is being used in data processing. This analyzer is a very versatile piece of equipment. Some of its useful capabilities are listed below:

1. Addition, subtraction, multiplication, and division of two spectra manually and automatically. One use of this feature permits the automatic subtraction of a background spectrum from input spectra.
2. Summing capabilities within a spectrum. This feature allows all of the counts in a peak to be summed with the result's being the area under the peak.

The output of the analyzer can go to any of three peripheral devices - typewriter, cassette, or magnetic tape. The typewriter is capable of printing out the contents of all 4000 channels or any included combination of channels. The cassette unit is used only for temporary storage of spectra. It is often used for transferring spectra from one magnetic tape to another. The Ampex magnetic tape unit is connected to the analyzer through the Intertechnique interface unit. This unit adds the six digit tagword to each spectrum as it is stored on the magnetic tape. Each tagword and spectrum combination are stored on tape as one file and each file occupies approximately nine inches of tape. Magnetic tapes have several good characteristics as data storage devices:

1. The transfer time from analyzer to tape is less than one second.
2. Each tape can store several thousand spectra.

3. Specific spectra can easily be found on the tape by means of the tagword.

These magnetic tapes provide the means of transferring the spectra from the NAA laboratory to the university computing center.

Before samples are to be counted on the detection and counting system, the analyzer must be calibrated. The standard calibration procedure that is presently used with 1000 channels is as follows:

1. Obtain the  $\text{Co}^{60}$  -  $\text{Cs}^{137}$  source and place it on the detector.
2. Turn the "intensification" knob to "channel B."
3. Turn the two channel selectors, "channel A" and "channel B" located on the upper right-hand side of the analyzer face to "1" and "420" respectively.
4. Turn down the "brightness" knob until the single dot at channel 420 is brighter than all of the other dots.
5. Begin the standard counting procedure by depressing the "store" button.
6. Adjust the "Fine Gain" knob on the Tennelec TC 200 amplifier until the bright dot on the CRT display is in the maximum channel of the highest in energy of the three major peaks in the spectrum.

The third section of the NAA procedure is that of processing the data by means of the computer. The details of this procedure are explained in detail in the following appendices.

## VII. APPENDIX B: NAA PROGRAM OPERATION

The explanation of the Neutron Activation Analysis Program given in this appendix is intended for someone who has some knowledge of the FORTRAN IV computer program language. All of the explanations and symbols are described in the order that they appear in the program. The FORTRAN listing of the program is given in Appendix C.

### MAIN

1. Read in all time cards containing activation, decay, and count times, sample mass, flux, identification, and tagword. See Fig. 18 for a sample time card. These values are stored for use later in the program.

IMAX = Number of channels  
TID = Time in reactor, day of year  
TIH = Time in reactor, hour  
TIM = Time in reactor, minute  
TIS = Time in reactor, second  
TOD = Time out of reactor, day of year  
TCD = Time begin counting, day of year  
TSD = Time stop counting, day of year  
ATA = Activation time in minutes  
ATW = Decay time in minutes  
ATC = Count time in minutes  
FLUX = Neutron flux  
NTAG = Tagword

A1,A2,A3 = Sample identification

LASTAG = Tagword of last timecard

WT = Sample mass

IO = Number of time cards

2. The assembler language subroutine, ICHANG, manipulates the data, IDATA, read from tape into a form useable by the computer.

IFILE = File number of the spectrum read from tape. IFILE = 1 is the file containing the first spectrum read in the program.

ITAG = Tape tagword

3. The subroutine, IC, converts the data, in the array IDATA from ICHANG into decimal form from hexadecimal form.

4. The DO loop 405 matches tagwords from time cards and tape.

5. The shift routine moves every spectrum down x number of channels, where x appears in statement number 52 as  $LL = LLM - x$ . The value of x must be inserted directly into the program. It can have a range of values  $0 \leq x \leq 5$ .

ID = Array containing the number of counts as a function of channel number

6. The spectra are written out in block form.

7. Subroutine IDEN

8. Subroutine FIT

9. Subroutine SPLOT



IDEN

1. Four IF statements locate each peak (11, 14, 19, 25). Each peak contains a maximum of six points below the peak and three points above. A peak minimum is established if a point drops less than 20% of the drop of the previous point from its predecessor.

J = Peak number

IST,KF = minimum channel numbers of peak on low and high energy side.

12 = Peak channel number

ICHECK = Loop counter for the checking of 20% of reductions in count differences to find the peak minima.

2. The 2725 Loop calculates peak energies, areas, and error in area. The area is the sum of all the counts in the peak less the background counts. The background consists of the area under a line drawn between the two minimum points of the peak.

DEL = Total peak width

SLOPE = Slope of the background line

SUM = Sum of all the counts in the peak

SUMB = Sum of all background counts in the peak

AR = Area of each peak excluding background

3. Subroutine GAUS

IX = Peak channel number

I = Peak number

YO = Number of counts in the initial channel, ISTI, of the peak

POS = Mean energy value (KeV) of each peak.

4. ERRAR = Error in the area of each peak.

5. Subroutine TRAPP. If POS = 10000, the peak is rejected.

(See TRAPP write-up)

6. ERRAR = AR if the error in the area is greater than the area of the peak, the peak is rejected as being only statistical.

7. J = IN. Number of peaks remaining

#### TRAPP

Peaks which are 0.51 and 1.02 MeV below a given peak are rejected by setting their energy values, POS, equal to 30000 KeV. The two pair production peaks must both be equal the upper peak less 0.51 and  $1.02 \pm .002$  MeV.

#### GAUS

The maximum and two adjacent point of each peak are fitted to a Gaussian distribution from which the mean channel no.XMEAN, standard deviation, SDEV, and amplitude, AMP, are calculated.

#### FIT

This subroutine compares the experimental peaks with the library gamma ray energy values and determines the elements in each sample along with the corresponding concentration and activity.

1. Loop 530 finds the sodium peak at or near  $2754 \pm 20$  KeV, FNAPK, if there is such a peak in the spectrum. If there is a sodium peak found, the shift factor, SHFT, is computed and used to shift the

entire spectrum by the factor SHFT.

DDI = The area of the largest peak in the vicinity of 2754 KeV.

POSN = Energy values of the shifted peaks.

2. Read in isotope groups from disk and compare the gamma ray energies with the peak energies in the spectrum.

NO = Number of gamma rays of each isotope.

ELE = Element symbol.

ISOT = Isotope mass number.

HF = Halflife.

BRCH = Branching ratio.

ABUND = Abundance.

SIGMA = Absorption cross section.

IFACT = Time unit: 0 = minutes

1 = hours

2 = days

3 = years

ALAM = Decay constant.

4. The program requires a one-to-one correspondence between gamma ray energy values and spectrum peaks in order to find an isotope.

5. The efficiency, EFF, equation is based upon the characteristics of the detector.

6. Elements having exponents, ALTA, ALTC, and ALTW, greater than 12 are rejected and those with values less than .003 are approximated by a first order Taylor series expansion of the exponent.

7. PPM = Parts per million.

ERPPM = Error in parts per million.

ACT = Activity

### SPLIT

This subroutine plots a spectrum with the number of counts of the vertical axis channel number on the horizontal axis.

IFIRST, ITOT = Plot number and total number of plots.

AX = Array proportional to the energy. Proportionality constant = 0.01.

BY = Array proportional to the array of counts, ID. Proportionality constant =  $10^{-4}$

X,C,Y,D = Symbols used in printing hash marks on the axes of the plot.

DY = Scale Factor.

For N = Actual number of counts and

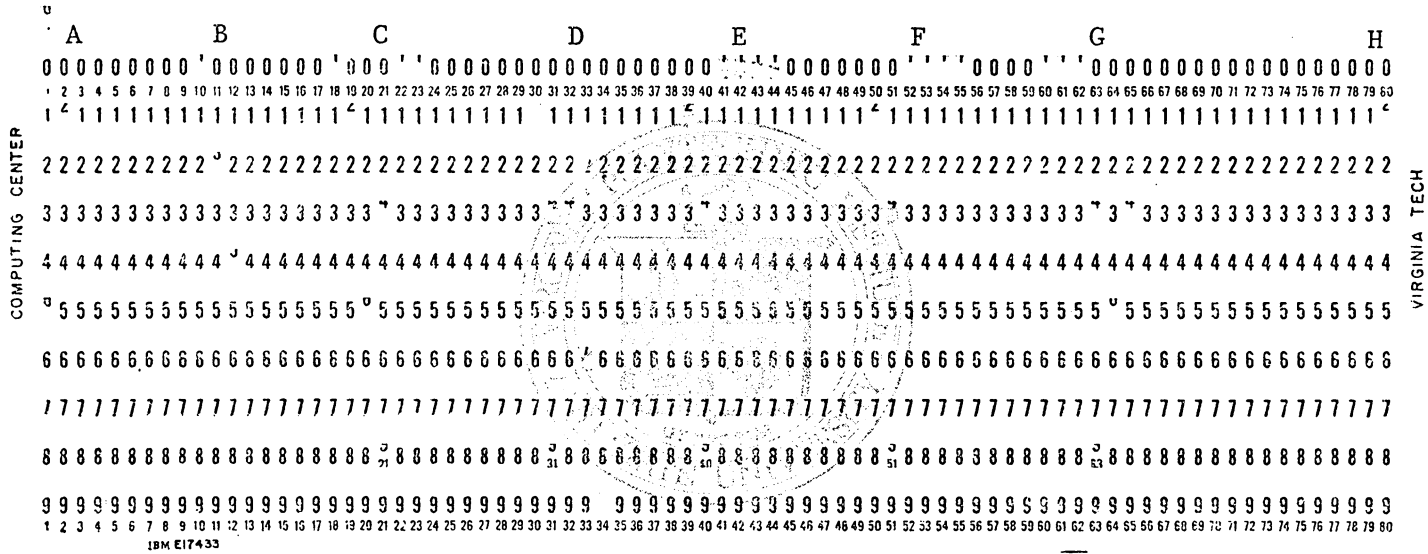
C = Number of counts on the "counts" axis of the plot

$N = (8/DY)*C.$

### Input Data

The NAA program was designed in order to make the input data as uncomplicated as possible for the user. The data sets required for standard operation of the program are the library, spectra, time cards, and file cards. The library data are arranged into isotope groups. A typical isotope group is shown in Fig. 17. Changes in this data set are not required for standard operation of the program.

NA	024	015.00	2.754	1.0000	1.0000	000.53	1
NA	024	015.00	1.369	1.0000	1.0000	000.53	1



- |                                 |   |
|---------------------------------|---|
| A = element symbol              | F = abundance   |
| B = isotope atomic weight       | G = absorption cross section<br>(in units of $10^{-24} \text{cm}^2$ ) |
| C = half-life (for units see H) | H = half-life unit code: 0 minutes<br>1 hour<br>2 days<br>3 years     |
| D = gamma ray energy            |   |
| E = branching ratio             |   |

Fig. 17. Typical Library Data Card Set for one Isotope with two gamma rays

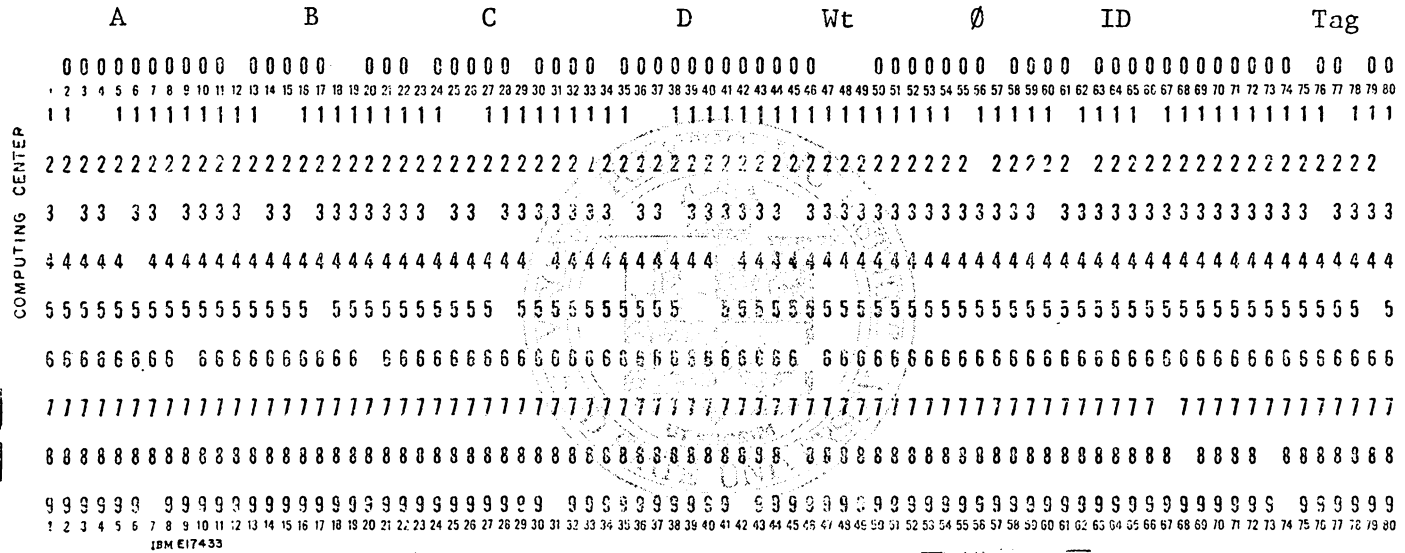
The time cards contain data recorded at the times of activation and counting. A sample card is shown in Fig. 18. The cards in this data set may be arranged in any order with the exception of the last card. The last time card should correspond to the last spectrum on the magnetic tape with respect to the included group of time cards. After the spectrum corresponding to the final time card has been processed, the program operation is terminated.

The spectra on magnetic tape, as mentioned above, constitute another input data set for the program. The tagwords corresponding to the respective spectra are also stored along with each spectrum. Once the desired spectra have been recorded on the tape, all that is required of the user is to transfer the tape to the tape library at the computer center.

The file cards are Job Control Language (JCL) cards which dictate which files are to be read from the data tape. These cards will be discussed further in the following section.

A final input parameter to the program is a shift constant. Occasionally, entire spectra have been shifted up as many as five channels. The cause for this shifting has been the temporary transferring of spectra to and from cassettes. The shift parameter statement is located approximately in the middle of program MAIN. The statement is "LL = LLM - X," where X is the shift parameter. The maximum value X can have is 5.

031134936 031135006 031135049 031135549 .6000 120 CAS 178 81031052



A = start time of irradiation

Wt = sample weight

B = 031 day number  
13 hours  
49 minutes  
36 seconds

∅ = neutron flux  
067 south beam port  
120 rabbit tube  
130 central stringer

C = start time of counting

ID = sample identification

D = end time of counting

Tag = tagword of spectrum on tape

Fig. 18. Typical Time Card

Data Printout

The printed output is contained in four different sections. The first section lists all of the data from the time cards exactly as it appears on the cards. A printout of each of the three remaining sections is given for each spectrum. The second section contains a list of all the peaks found in the spectrum along with the corresponding lower, center, and upper peak channel limits, energy, area, and error in the area. The final section gives the major output of the program - the elements found in the sample and their respective concentrations. Along with each isotope is printed the natural abundance of the isotope, the concentration, error in concentration, activity, and experimental and theoretical energies of each of the associated gamma rays. The elements listed in this section are not always the ones that are truly contained in the sample. This fact stems from the limited capability of the peak finding routine. The lack of precision in this routine requires that broad acceptance limits be used for matching element gamma rays with the peak energies in the spectrum. Such limits often result in one peak's being matched with several different elemental gamma rays. A feature of this program which helps to alleviate this problem is that of matching more than one peak for those elements with several emitted characteristic gamma rays, e.g., the four peaks located at 777, 554, 619, and 698 keV in Fig. 3 must be present in order to have the program find Br<sup>82</sup>. In addition, a visual comparison of the experimental and theoretical gamma energies are often enough to discard most of the extraneous identification.



Control Cards

The Job Control Language cards required to run the program are shown in Fig. 19. The tape identification is given as "XXXXX." A maximum number of 95 pairs of "GO.FT10FNNN" cards may be used for one program. The numbers NNN must increase sequentially without missing any number. The "DD" card file numbers, "FFF" must be in ascending order but the numbers do not need to be consecutive. There is a 100 card limit on the number of time cards that may be included. There must be a blank card following the final data card.

```

//JOB CARD
/* SETUP UNIT=TAPE9, DDNAME=FT10F001, ID=(XXXXX, NCRING, SAVE, NL)
/* MAIN REGION=(, 200, ,, 200), LINES=30, TIME=(5, 45)
//STEP1 EXEC FORTGC
//FORT.SYSIN DD *
        FORTRAN DECK TO READ IN LIBRARY
// EXEC FORTFIX
//GO.FT08F001 DD DSN=&FURR, VOL=SER=WCRK01, UNIT=SYSDA,
//          DISP=(NEW, PASS), DCB=(RECFM=FB, LRECL=80, BLKSIZE=7200),
//          SPACE=(TRK, (10))
//GO.SYSIN DD *
        LIBRARY
//STEP2 EXEC FORTGC
//FORT.SYSIN DD *
        MAIN DECK
// EXEC ASMF, PARM='LOAD, NODECK'
//ASM.SYSGO DD DSN=&LOADSET, UNIT=SYSSQ,
// DISP=(MOD, PASS)
//ASM.SYSIN DD *
        ASSEMBLER LANGUAGE PROGRAM
// EXEC FORTFIX, LIB1=PLCLIB
//GO.FT08F001 DD DSN=&FURR, VOL=SER=WCRK01, UNIT=SYSDA,
//          DISP=(OLD, PASS), DCB=(RECFM=FB, LRECL=80, BLKSIZE=7200)
//GO.FT10FNNN DD UNIT=TAPE9, DISP=CLD, DCB=(BLKSIZE=768, RECFM=U, DEN=2),
//          VOL=(, RETAIN, SER=XXXXX), LABEL=(FFF, NL)
//GO.SYSIN DD *
        TIME CARDS
        BLANK CARD

/*
//

```

Fig. 19. Job Control Statements for NAA Computer Program

VIII. APPENDIX C: NAA PROGRAM LISTING

```
C*****SET UP LIBRARY*****C
C
C
      DIMENSION A(20)
5     READ(5,10,END=20) A
      WRITE(8,10)A
10    FORMAT(20A4)
      GO TO 5
20    CONTINUE
      END FILE 8
      STOP
      END
```

C\*\*\*\*\*MAIN PROGRAM\*\*\*\*\*C

C  
C

C  
C

```
COMMON ID(1023),POS(200),AR(200),ERRAR(200)
DIMENSION AFLUX(200),ATA(200),ATW(200),ATC(200),AWT(200),MTAG(200)
DIMENSION A1(100),A2(100),A3(100)
DIMENSION ICATA(1020)
INTEGER TID,TIH,TIM,TIS,TOD,TOH,TOM,TOS,TCO,TCH,TCM,TCS,
*TSD,TSH,TSM,TSS
```

C  
C  
C  
C

```
READ TIME ALL CARDS AND STORE ACT, DECAY, AND COUNT TIMES, WT, FLUX,
SAMPLE ID, AND TAGWORD.
```

```
IG=0
IFILE=0
IMIN=1
IMAX=1000
```

20

```
READ(5,369) TID,TIH,TIM,TIS,TOD,TOH,TOM,TOS,TCO,TCH,TCM,TCS,
*TSD,TSH,TSM,TSS,WT,IFLUX,W1,W2,W3,NTAG
IF(NTAG.EQ.000000) GO TO 5
IO=IO+1
WRITE(6,700) TID,TIH,TIM,TIS,TOD,TOH,TOM,TOS,TCO,TCH,TCM,TCS,
*TSD,TSH,TSM,TSS,WT,IFLUX,W1,W2,W3,NTAG
FLUX=IFLUX
IF(FLUX.EQ.0.) FLUX=130.
AFLUX(IO)=FLUX*10.**10
SI=TID*1440.+TIH*60.+TIM+TIS/60.
SO=TOD*1440.+TOH*60.+TOM+TOS/60.
SC=TCO*1440.+TCH*60.+TCM+TCS/60.
SS=TSD*1440.+TSH*60.+TSM+TSS/60.
ATA(IO)=SO-SI
ATW(IO)=SC-SC
```

```

      ATC(IO)=SS-SC
      AWT(IO)=WT
      MTAG(IO)=NTAG
      A1(IO)=W1
      A2(IO)=W2
      A3(IO)=W3
      LASTAG=NTAG
      GO TO 20
369  FORMAT(4(I3,I2,I2,I2,2X),F8.5,2X,I3,2X,2A4,A2,5X,I6)
C
C   READ SPECTRUM FROM TAPE AND SEARCH THE TIME CARDS FOR A MATCH IN
C   TAGWORDS.
C
700  FORMAT(1X,4(I3,I2,I2,I2,2X),F8.5,2X,I3,2X,2A4,A2,5X,I6)
5    READ(10,10,END=999) ITAG,(IDATA(II),II=1,1020)
100  FORMAT(100A3,100A3,56A3,/,3X,100A3,100A3,55A3,/,
      13X,100A3,100A3,55A3,/,3X,100A3,100A3,55A3)
999  IFILE=IFILE+1
      ITAG=ICHANG(ITAG)
      ITAG=IC(ITAG)
      DO 405 IT=1,10
      IF(ITAG.NE.MTAG(IT))GO TO 405
      GO TO 410
405  CONTINUE
      GO TO 5
410  DO 15 I=1,1000
      IDATA(I)=ICHANG(IDATA(I))
15  ID(I)=IC(IDATA(I))
C
C   SHIFT ROUTINE FOR 1000 CHANNEL SPECTRA
C
      DO 2478 LLM=6,1006

```

```

LL=LLM-0
ID(LL)=ID(LLM)
2478 CONTINUE
DO 2468 LLL=1000,1023
ID(LLL)=0
2468 CONTINUE
C
C
IPIK=0
IIDEN=1
111 FORMAT(1X,120)
1112 WRITE(6,1003) ITAG,(IZZ,IZZ=1,10)
1003 FORMAT(1H1,10X,' TAG,WORD =',I8,/,12X,10I10)
DO 1001 III=1,103
IZ=(III-1)*10
IZTEN=IZ+10
IZP1=IZ+1
IF(III.EQ.103) IZTEN=1023
WRITE(6,1000) IZ,(ID(JJ),JJ=IZP1,IZTEN)
1000 FORMAT(10X,I5,10I10)
1001 CONTINUE
WRITE(6,2) IFILE
2 FORMAT(' FILE NUMBER=',I3)
CALL IIDEN(J)
TA=ATA(IT)
TW=ATW(IT)
TC=ATC(IT)
WT=AWT(IT)
W1=A1(IT)
W2=A2(IT)
W3=A3(IT)
FLUX=AFLUX(IT)

```

```
      NTAG=MTAG(IT)
      CALL FIT(J,TA,TW,TC,FLUX,WT,NTAG,W1,W2,W3)
C
C      CALL SPLOT(IT,IMIN,IMAX,NTAG,TA,TW,TC,IO)
C
      IF(ITAG.EQ.LASTAG) GO TO 30
      GO TO 5
30    CONTINUE
      STOP
      END
```

```

        SUBROUTINE IDEN(J)
        COMMON   ID(1023),POS(200),AR(200),ERRAR(200)
        INTEGER*2   I2(200),KF(200),IST(200)
102   FORMAT(2(F15.2,I10))
C     THIS SUBROUTINE FINDS PEAKS IN A SPECTRUM THE CORRESPONDING AREAS,
C     AND THE ENERGY OF THE PEAKS.
        L1=0
        J=0
C     FIND PEAKS
        I=1
2     I=I+1
        IF(I.EQ.1024) GO TO 2000
        IF(ID(I).LE.ID(I-1)) GO TO 2
4     I=I+1
        IF(I.EQ.1024) GO TO 2000
        IF(ID(I).LE.ID(I-1)) GO TO 2
        J=J+1
        IB=I-2
3     I=I+1
        IF(I.EQ.1024) GO TO 2001
        IF (ID(I).GT.ID(I-1)) GO TO 3
        I2(J)=I-1
        DEL1=IC(I-1)-ID(I-2)
        IDEL1=.2*DEL1
        DO 300 ICHECK=2,7
        IDEL2=ID(I-ICHECK)-ID(I-ICHECK-1)
        IF(IDEL2.LT.IDEL1) GO TO 310
        DEL1=IDEL2
        IDEL1=.2*DEL1
300   CONTINUE
310   IST(J)=I2(J)-ICHECK+1
2710  I=I+1

```



```

IF(I.EQ.1024) GO TO 2001
IF(ID(I).LT.ID(I-1)) GO TO 2710
IPEAK=I2(J)
DEL1=ID(IPEAK)-ID(IPEAK+1)
IDEL1=.2*DEL1
DO 320 ICHECK=2,4
IDEL2=ID(IPEAK+ICHECK-1)-ID(ICHECK+IPEAK)
IF(IDEL2.LT.IDEL1) GO TO 330
DEL1=IDEL2
IDEL1=.2*DEL1
320 CONTINUE
330 KF(J)=I2(J)+ICHECK-1.
2720 GO TO 4
2001 J=J-1
2000 WRITE(6,2918)
2918 FORMAT(' I PEAK LOWER CH UPPER CH PEAK ENERGY ',
* ' AREA AREA ERRCR')
IN=0
C FIND PEAK AREAS--ADD UP ALL COUNTS IN THE PEAK AND SUBTRACT OUT
C BACKGROUND. BACKGROUND IS A STRAIGHT LINE BETWEEN MIN POINTS OF
C PEAKS, HIGH AND LOW.
DO 2725 I=1,J
KF1=KF(I)
IST1=IST(I)
DEL=KF1-IST1
SLOPE=(ID(KF1)-ID(IST1))
SLOPE=SLOPE/DEL
AR(I)=0.
SUMB=0.
SUM=0.
DO 2730 IJ=IST1,KF1
VID=ID(IJ)

```

```

VIDO=ID(IST1)
VIJ=IJ
VIST1=IST1
SUM=SUM+VID
BACKG=SLOPE*(VIJ-VIST1)+VIDO
SUMB=SUMB+BACKG
2730 AR(I)=AR(I)+VID-BACKG
      B=I2(I)
      IX=I2(I)
      YO=ID(IST1)
      CALL GAUS(IX,I,YO,SLOPE,IST1,B)
      POS(I)=3.1681 * B+ .00677 * (B-870)
      ERRAR(I)=SQRT(SUM+SUMB)
      IF(I.EQ.1) GO TO 2725
C     CALL TRAPP(I)
2725 CONTINUE
      DO 125 I=1,J
      IF(POS(I).GT.10000.) GO TO 125
      IF(ERRAR(I).GT.AR(I)) GO TO 125
      IN=IN+1
      WRITE(6,425) IN, I2(I),IST(I),KF(I),PCS(I),AR(I),ERRAR(I)
      POS(IN)=PCS(I)
      AR(IN)=AR(I)
      ERRAR(IN)=ERRAR(I)
125  CONTINUE
425  FORMAT(4(I4,5X),3(E12.4,5X))
      J=IN
      RETURN
      END

```

```

      FUNCTION IC(N)
C     CONVERT FROM HEXADECIMAL TO BASE TEN
      IHTH=N/16**5
      ITTH=(N-IHTH*16**5)/16**4
      ITH=(N-IHTH*16**5-ITTH*16**4)/16**3
      IH=(N-IHTH*16**5-ITTH*16**4-ITH*16**3)/16**2
      IT=(N-IHTH*16**5-ITTH*16**4-ITH*16**3-IH*16**2)/16
      IU=N-IHTH*16**5-ITTH*16**4-ITH*16**3-IH*16**2-IT*16
      IC=IHTH*10**5+ITTH*10**4+ITH*10**3+IH*10**2+IT*10+IU
      RETURN
      END
      SUBROUTINE TRAPP(I)
      COMMON      ID(1023), POS(200), AR(200), ERRAR(200)
C
C     TRAP ROUTINE--PAIR PRODUCTION.  WE THROW OUT PEAKS .51 AND 1.02
C     MEV BELOW ANY PEAK.
C
      JP=0
      IM1=I-1
      DO 5 IM=1,IM1
      IF(POS(IM).LE.600.) GO TO 5
      DEL51=POS(IM)-POS(I)
      DEL5=ABS(DEL51)-510.
      DEL5=ABS(DEL5)
      DEL102=ABS(DEL51)-1020.
      DEL102=ABS(DEL102)
      IF(DEL102.GE.2.) GO TO 20
      JP=JP+1
      JPIM=IM
20     IF(DEL5.GE.2.) GO TO 5
      IF(JP.NE.1) GO TO 10
      POS(JPIM)=30000.

```

```
5   PGS(IM)=30000.  
10  GO TO 10  
    CONTINUE  
    CONTINUE  
    RETURN  
    END
```

```

SUBROUTINE GAUS(IX,I,YO,SLOPE,IST1,XMEAN)
C
C GAUS FITS THE MAXIMUM AND TWO ADJACENT POINTS TO A GAUSSIAN CURVE.
C TO DISREGARD THIS SUBROUTINE CONVERT THE 'CALL GAUS( )' STATEMENT
C IN IDEN TO A COMMENT CARD.
C
COMMON      ID(1023),POS(200),AR(200),ERRAR(200)
ST1=IST1
B1=IX-1
B2=IX
B3=IX+1
Y1=ID(IX-1)
Y2=ID(IX)
Y3=ID(IX+1)
Y1=Y1-SLOPE*(B1-ST1)-YO
Y2=Y2-SLOPE*(B2-ST1)-YO
Y3=Y3-SLOPE*(B3-ST1)-YO
IF(Y1.LE.1..OR.Y3.LE.1..OR.Y2.LE.1.) GO TO 5
ARG2=Y2/Y1
DIF1=ARG2-1.1
IF(DIF1.LE.0.) GO TO 5
ALP=ALOG(Y3/Y1)/ALOG(Y2/Y1)
XMEAN=B1*B1-B3*B3-ALP*(B1*B1-B2*B2)
XMEAN=XMEAN/(2.*(ALP*(B2-B1)-(B3-B1)))
SDEV=((B1-XMEAN)**2-(B2-XMEAN)**2)/ALOG(Y2/Y1)
AMP=Y1*EXP(((B1-XMEAN)**2)/SDEV)
GO TO 10
5 XMEAN=B2
10 CONTINUE
RETURN
END

```

```
SUBROUTINE FIT (J,TA,TW,TC,FLUX,WT,NTAG)
COMMON ID(1023),POS(200),AR(200),ERRAR(200)
DIMENSION POSN(200),IK(10),EN(10)
DIMENSION IPEAK(50),ABRCH(50),ASIG(50),ABD(50),ALAM(50)
```

C  
C  
C  
C

```
FIT FINDS THE ELEMENTS IN EACH SPECTRUM AND THEIR CONCENTRATION,
ERROR IN CONCENTRATION, AND ACTIVITY.
```

```
FNAPK=2754.
```

```
DELTA=20.
```

```
DD=0.
```

```
DD1=0.
```

```
DO 530 NN=1,J
```

```
IIII=J-NN+1
```

```
DELE=POS(IIII)-FNAPK
```

```
IF(ABS(DELE).GT.DELTA) GO TO 530
```

```
DD=AR(IIII)
```

```
IF(DD1.GT.DD) GO TO 530
```

```
DD1=DD
```

```
III=IIII
```

530 CONTINUE

```
IF(DD.EQ.0.) GO TO 535
```

531 SHFT=FNAPK/POS(III)

```
DO 533 NNA=1,J
```

```
POSN(NNA)=POS(NNN)*SHFT
```

533 CONTINUE

```
WRITE(6,589) POS(III)
```

589 FORMAT(10X,' .....A SODIUM PEAK HAS BEEN DETECTED AT',F8.2,' KEV RA  
\*THER THAN',/,14X,' AT ITS TRUE ENERGY OF 2754 KEV. CONSEQUENTLY, A  
\*N ENERGY',/,14X,' CORRECTION HAS BEEN APPLIED BY MEANS OF A GAIN S  
\*HIFT.....',///)

```
DO 600 MMM=1,J
```

```

P=PCS(MMM)
PN=POSN(MMM)
IF(MMM.NE.1) GO TO 592
C WRITE(6,590)
590 FORMAT('          UNCORRECTED ENERGY          CORRECTED EN
*ERGY',/)
592 CONTINUE
C WRITE(6,591)MMM,P,PN
591 FORMAT(10X,12,12X,F8.2,18X,F8.2)
600 CONTINUE
GO TO 601
535 CONTINUE
DO 7871 I=1,J
POSN(I)=PCS(I)
7871 CONTINUE
WRITE(6,538)
538 FORMAT(1X,/,10X,'.....NO SODIUM PEAK HAS BEEN DETECTED. CONSEQUEN
*TLY NO CORRECTION',/,14X,' FOR DETECTOR GAIN SHIFT VARIATIONS HAS
*BEEN APPLIED TO THIS',/,14X,' SPECTRUM....',/)
601 CONTINUE
WRITE(6,8780) TA,TW,TC,WT,NTAG,W1,W2,W3
8780 FORMAT(' ACTIVATION TIME=',F10.3,' WAIT TIME=',F10.3,
1' COUNT TIME=',F10.3,/, ' WEIGHT=',F10.5,' TAG=',I6,3X,
*' ID=',5X,2A4,A2//)
WRITE(6,519)
519 FORMAT('          ELEMENT          ISOTOPE          NATURAL          CONCENTRA
*TION      ERROR      ACTIVITY      EX ENERGY      TH ENERGY',/20X,'
*          ABUNDANCE          (PPM)          (PPM)          MICRO-CURIES
*          (KEV)          (KEV)')
510 CONTINUE
READ(8,500,END=555) NO
500 FORMAT(I1)

```

```

IF(NO.EQ.0) GO TO 555
DO 501 II=1,NO
READ(3,502) ELE,ISOT,HF,ENRGY,BRCH,ABUND,SIGMA,IFACT
EN(II)=ENRGY
502 FORMAT(A2,7X,I3,3X,F8.2,6X,F5.3,2X,F8.4,5X,F6.4,2X,F11.5,11X,I1)
ABRCH(II)=BRCH
ASIG(II)=SIGMA
ABD(II)=ABUND
IF(IFACT.EQ.0) A=1.
IF(IFACT.EQ.1) A=60.
IF(IFACT.EQ.2) A=1440.
IF(IFACT.EQ.3) A=525600.
ALAM(II)=.693/(A*HF)
511 CONTINUE
501 CONTINUE
DEL=10.
DO 505 JJ=1,NO
ENRGY=EN(JJ)*1000.
DO 504 KK=1,J
DELEN=ENRGY-POSN(KK)
IF(ABS(DELEN).LT.DEL) GO TO 503
504 CONTINUE
GO TO 510
503 CONTINUE
IK(JJ)=KK
505 CONTINUE
DO 506 LL=1,NO
MM=IK(LL)
IPEAK(LL)=MM
CNTS=AR(MM)
ERCNTS=ERRAR(MM)
X=EN(LL)

```



```

ENERGY=X*1000.
EFF=3.9136*EXP(-7.4096*X)+1.7446*EXP(-.7124*X)+.005863+.00000144*E
*XP(-.001*X)
EFF=EFF/100.
ASGT=ISGT
ALTA=ALAM(LL)*TA
ALTC=ALAM(LL)*TC
ALTW=ALAM(LL)*TW
IF(ALTA.LE.12..AND.ALTC.LE.12..AND.ALTW.LE.12.) GO TO 8786
GO TO 510
8786 IF(ALTA .GE. .003) GO TO 8984
EXPA = ALTA
GO TO 8788
3984 EXPA=1.-EXP(-ALTA)
8788 IF(ALTC.GE..003) GO TO 8789
EXPC=ALTC
GO TO 888
3789 EXPC=1.-EXP(-ALTC)
888 IF(ALTW.GE..003) GO TO 905
EXPW=1.-ALTW
GO TO 910
905 EXPW=EXP(-ALTW)
910 PPM= (ASGT*ALAM(LL))/(ABD(LL)*EFF*ABRCH(LL)*WT*.602*ASIG(LL)
1*EXPA*EXPC*EXPW*60.*FLUX)
ERPPM=ERCNTS*PPM
PPM=CNTS*PPM
PPM=PPM*10.**6
ERPPM=ERPPM*10.**6
IF(PPM.GE.10.**6) GO TO 510
ACT =CNTS/(TC*EFF*60.*3.7)/10.**4
889 CONTINUE
IF(LL.NE.1) GO TO 8901

```

```
      WRITE(6,520) ELE,ISGT,ABD(LL),PPM,ERPPM,ACT,POSN(MM),ENERGY
520   FORMAT(17X,A2,9X,I3,9X,F5.3,3X,E10.4,3X,E10.4,3X,E12.4,7X,F7.2,
      *3X,F7.2)
      GO TO 506
8901  WRITE(6,523) ABD(LL),PPM,ERPPM,ACT,POSN(MM),ENERGY
523   FORMAT(40X,F5.3,3X,E10.4,3X,E10.4,3X,E12.4,7X,F7.2,3X,F7.2)
506   CONTINUE
      GO TO 510
555   CONTINUE
      REWIND 8
      RETURN
      END
```

SUBROUTINE SPLOT(IFIRST,NMIN,NMAX,NTAG,TA,TW,TC,ITOT)

C  
C  
C

SPLOT PLOTS EACH SPECTRUM.

COMMON ID(1023),POS(200),AR(200),ERRAR(200)

DIMENSION AX(1000),BY(1000)

IF(IFIRST.NE.1) GO TO 4

CALL PLCT(1.0,1.0,-3)

GO TO 2

4 CALL PLCT(1.0,0.0,-3)

2 CALL PLOT(0.0,8.0,2)

CALL PLOT(10.0,8.0,2)

CALL PLOT(10.0,0.0,2)

CALL PLOT(0.0,0.0,2)

DO 6 I =NMIN,NMAX

AX(I)=I/100.

BY(I)=ID(I)\*1.E-4

6 CONTINUE

MIN=NMIN+10

N=2

TAG=NTAG

K=50

Y=0.0

X=0.0

DO 7 I=1,9

X=I

CALL PLOT(X,0.0,3)

CALL PLCT(X,-.1,2)

C=I\*100.

CALL NUMBER(X-.2,-.3,.15,C,0.0,-1)

7

CONTINUE

DO 8 J=1,9

```

Y=.8*J
CALL PLCT(0.0,Y,3)
CALL PLOT(-.1,Y,2)
IF(J.NE.N) GO TO 8
D=1000.*J
CALL NUMBER(-.3,Y-.2,.15,D,90.,-1)
N=J+2
8 CONTINUE
CALL SYMBOL(4.0,-.6,.2,7HCHANNEL,0.,7)
CALL SYMBOL(-0.6,3.2,.2,6HCCUNTS,90.,6)
CALL SYMBCL(8.0,7.4,.1,10HTAGWORD =,0.,12)
CALL NUMBER(9.2,7.4,.1,TAG,0.,-1)
CALL SYMBCL(8.0,7.1,.1,12HACT. TIME =,0.,12)
CALL NUMBER(9.2,7.1,.1,TA ,0.,2)
CALL SYMBCL(7.8,6.8,.1,12HDECAY TIME =,0.,12)
CALL NUMBER(9.2,6.8,.1,TW ,0.,2)
CALL SYMBCL(7.8,6.5,.1,12HCOUNT TIME =,0.,12)
CALL NUMBER(9.2,6.5,.1,TC ,0.,2)
CALL SCALE(BY(NMIN),NMAX-NMIN+1,8.0,YMIN,DYY,1)
CALL SCALE(AX(NMIN),NMAX-NMIN+1,10.0,XMIN,DXX,1)
DY=1./DYY
CALL SYMBCL(1.0,7.4,.1,7HSCALE =,0.,7)
CALL NUMBER(1.8,7.4,.1,DY,0.,2)
CALL LINE(AX(MIN),BY(MIN),NMAX-MIN+1,1)
DO 5 I=NMIN,NMAX
IF(I.NE.K) GO TO 5
VID=ID(I)
B1=(VIC+VID*.5)*1.E-4
B2=(VID-VID*.5)*1.E-4
CALL PLOT(AX(I)-.05,B1/DYY,3)
CALL PLOT(AX(I)+.05,B1/DYY,2)
CALL PLOT(AX(I),B1/DYY,3)

```

```
CALL PLOT(AX(I),B2/DYY,2)
CALL PLOT(AX(I)-.05,B2/DYY,3)
CALL PLOT(AX(I)+.05,B2/DYY,2)
K=K+50
5 CONTINUE
IF(IFIRST.EQ.ITOT)GO TO 9
CALL PLOT(10.0,0.0,-3)
GO TO 3
9 CALL PLOT(0.,0.,-4)
RETURN
3 END
```

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NEUTRON ACTIVATION ANALYSIS OF  
PLANTS AS POLLUTION INDICATORS

by

Crispin A Schamp

(ABSTRACT)

The method of Neutron Activation Analysis has been used in a test case for the development of a technique for using trace elements in plants as pollution indicators for automobile exhaust effects. The plant study in the present work is a pilot program to develop sample collection, preparation, and analysis techniques. The results obtained indicate that natural growth plants can be used successfully as indicators of automobile pollution incorporating the methods of analysis of this study.

This work has been a test case for the computer program, which was used in the VPI&SU Neutron Activation Analysis Laboratory and was developed during the course of this study. The peak finding and area calculation routines incorporate uncomplicated techniques which have been shown by the test data to produce consistent results. A versatile feature of the program is that it can determine simultaneously, both qualitatively and quantitatively, the presence of the more than 60 different elements contained in the program library.

It has been shown that neutron activation analysis, employed in conjunction with the computer program described herein, using the analytical techniques developed in the VPI&SU Laboratory can provide

an effective tool for defining the environmental impact area of  
automobile traffic.