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Incorporated
P.O. Box 608, San Diego, California 92112

APPLICATIONS OF NEUTRON ACTIVATION ANALYSIS IN SCIENTIFIC CRIME INVESTIGATION

ANNUAL REPORT

JUNE 1, 1968 THROUGH MAY 31, 1969

Work done by:

D. E. Bryan
F. M. Graber
V. P. Guinn
R. P. Hackleman
R. D. Hamill
H. R. Lukens
J. K. MacKenzie
H. L. Schlesinger
T. Yamaguchi

Report written by:

V. P. Guinn
R. P. Hackleman
H. R. Lukens
H. L. Schlesinger

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ABSTRACT

This report describes forensic activation analysis research conducted at Gulf General Atomic, under a contract with the Division of Isotopes Development, U.S. Atomic Energy Commission, during the period, June 1, 1968 through May 31, 1969. During this period, a large number of test firings of revolvers and automatic pistols were performed, followed by hand sampling with paraffin and neutron activation analysis (NAA) of the paraffin handlifts for barium and antimony. Also, occupational "handblank" samples were obtained by the same procedure from persons of a number of additional occupations – and similarly analyzed for barium and antimony. A bivariate normal statistical treatment method was developed for the interpretation of the accumulated gunshot-residue and handblank data. As applied to the data obtained thus far, it is a promising method. An analogous multivariate normal statistical treatment method was investigated for treatment of the more than two variables observed in NAA data on paint, paper, and bullet lead.

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1. INTRODUCTION

This Annual Report covers the period, June 1, 1968 through May 31, 1969. However, notification of contract funding was not received until June 25, 1968, so the work reported was essentially performed during an 11-month (rather than 12-month) period. During this period, the manpower effort devoted to the investigation was approximately 2.5 man-years.

1.1 PRINCIPAL ACTIVITIES

Major efforts were expended on: (1) the development of sound statistical methods for the treatment of past and future gunshot-residue, handblank, paint, paper, and bullet-lead data, (2) the development of a statistically-designed experimental plan for the completion of the large-scale studies of these types of evidence materials, and (3) the obtaining and analyzing of a large number of additional gunshot-residue and occupational handblank samples. Lesser, but quite significant, efforts were devoted to: (4) the procurement of relevant background information from practicing criminalists (via a questionnaire) and from manufacturers (via visits), (5) the initial preparation of a comprehensive report, summarizing all the forensic activation analysis work carried out under this investigation since its inception (May 1, 1962) through May 31, 1968, (6) participation in relevant meetings, and (7) presentation of papers on the forensic activation analysis work at scientific meetings. Each of these activities is discussed in detail in later sections of this report.

1.2 GULF GENERAL ATOMIC FACILITIES EMPLOYED

Of the various irradiation, counting, and computing facilities available at Gulf General Atomic, those specifically utilized in this investigation

during the present report period included the following:

1. The 250 kW Mark I TRIGA nuclear reactor
2. One of the Cockcroft-Walton 14 MeV neutron generators
3. Various NaI(Tl) multichannel gamma-ray spectrometers
4. The UNIVAC 1108 computer

Sample and standard preparations, radiochemical separations, and most of the gamma-ray spectrometry measurements were carried out in the Activation Analysis Building, which is adjacent to the TRIGA Reactor Building and the Neutron Generator Bunker.

1.3 GULF GENERAL ATOMIC PERSONNEL ENGAGED IN STUDY

As in the earlier years of this investigation, the Principal Investigator during this report period was Dr. Vincent P. Guinn, Manager and Technical Director of the GGA Activation Analysis Department. However, a large fraction of the experimental work during this period was either performed by, or closely supervised by, Mr. Howard L. Schlesinger, a Staff Associate of the Activation Analysis Department, with previous forensic activation analysis experience in the U.S. Treasury Department group. All of the development of the statistical techniques was done by Dr. R. Paul Hackleman – a mathematician/statistician Staff Associate in the GGA Mathematics and Computing Department. Other radiochemists of the Activation Analysis Department who contributed significantly to this investigation during the present report period were: Mr. H. Richard Lukens (Staff Member), Mr. Francis M. Graber (Staff Associate), and Mr. Donald E. Bryan (Research Assistant). Assistance was also rendered by Technicians John K. MacKenzie, Tad Yamaguchi, and R. David Hamill.

2. STATISTICAL TREATMENTS DEVELOPED

Two basic statistical treatments were under development by Dr. R. P. Hackleman for use in this investigation: (1) a bivariate normal (BVN) treatment for use with barium and antimony gunshot-residue and occupational-handblank data, and (2) a multivariate normal (MVN) treatment for use with elemental compositional data on paints, paper, and bullet lead. These are both actually log-normal treatments. They are discussed below.

2.1 BIVARIATE NORMAL TREATMENT OF GUNSHOT-RESIDUE DATA

From many earlier experimental measurements on paraffin lifts from test firings of many types and calibers of revolvers and automatic pistols, and of occupational-handblank samples, a number of conclusions had been drawn:

1. Many variables (many of them, uncontrolled variables) are involved in the deposition of gunshot-residues on the back of the gunhand. These include: (a) type of gun, (b) condition of gun, (c) particular chamber fired, (d) caliber of gun, (e) brand of ammunition, (f) wind velocity and direction, (g) inclination of gun, (h) number of firings, (i) cleanliness of inside of gun barrel, etc.
2. Whereas persons in most occupations exhibit very low handblank levels of barium, persons in some occupations exhibit appreciable handblank levels of barium, or of barium and antimony (some occupations may also lead to appreciable handblank levels of antimony, but not barium – although no occupations of this type were encountered in all of the earlier work).

3. In all of the test firings of handguns up to this time, quite measurable amounts of both Ba and Sb were found to be deposited on the back of the gunhand, as removed by the paraffin-lift technique.
4. A much larger body of test-firing data and handblank data was needed, in order to place the NAA gunshot-residue method on a firm foundation, usable for the interpretation of gunshot-residue results obtained in the investigation of actual criminal cases — and also usable in courtroom presentations of results and their interpretation.
5. In view of the many sources of variation, and in view of the fact that both Ba and Sb are deposited on the back of the gunhand in a firing (in U.S. ammunition, presumably coming from the $Ba(NO_3)_2$ and Sb_2S_3 used as primer ingredients), a suitable statistical model was needed to treat the accumulated gunshot-residue and handblank data in a sensible and definitive manner.

A statistical model was then developed for the determination of the probability that a person has fired a handgun, from the barium and antimony levels in handlifts analyzed by neutron activation analysis.

For firings, both the barium and antimony concentrations found in handlifts are approximated by log-normal distributions, and further, are not independent variables. Therefore, each analysis can be interpreted as the random two-dimensional vector, $\underset{\sim}{x}$.

$$\underset{\sim}{x} = (x_1, x_2) \ , \tag{1}$$

which follows a bivariate normal distribution, and where x_1 and x_2 are the $\log_{10}$ of the barium and antimony concentrations, respectively.

The distribution of x is characterized by the mean vector, $\underset{\sim}{\mu}$:

$$\underset{\sim}{\mu} = (\mu_1, \mu_2) \ , \tag{2}$$

a covariance matrix, Σ:

$$\Sigma = \begin{bmatrix} \sigma_{11} & \sigma_{12} \\ \sigma_{21} & \sigma_{22} \end{bmatrix}, \ (\sigma_{12} = \sigma_{21}) \ , \tag{3}$$

and the density function, f, associated with this type of distribution:

$$f(x_1, x_2) = \frac{\exp\left[-\frac{1}{2}Q(x_1, x_2)\right]}{2\pi\left[(\sigma_{11}\sigma_{22})(1 - \rho^2)\right]^{\frac{1}{2}}} \ , \tag{4}$$

where ρ is the correlation between x_1 and x_2:

$$\rho = \sigma_{12}/(\sigma_{11}\sigma_{22})^{\frac{1}{2}} \ , \tag{5}$$

and where:

$$Q_{(x_1, x_2)} = \frac{1}{2\pi(1 - \rho^2)}\left[\frac{(x_1 - \mu_1)^2}{\sigma_{11}} - \frac{2\rho(x_1 - \mu_1)(x_2 - \mu_2)}{\sigma_{11}\sigma_{22}} + \frac{(x_2 - \mu_2)^2}{\sigma_{22}}\right] . \tag{6}$$

The probability, P, that the point x falls in any region, A, of the (x_1, x_2)-plane is given by:

$$P = \iint_A f(x_1, x_2)\, dx_1\, dx_2 \ . \tag{7}$$

A means of visualizing the shape of this distribution is provided by the geometric properties of the surface determined by the density function: the level curves of the surface, i.e., the set of points in the (x_1, x_2)-plane determined by the equations:

$$Q(x_1, x_2) = c \quad . \tag{8}$$

For various choices of positive numbers, c, these are concentric ellipses centered at the point $\underset{\sim}{\mu}$. The probability attributed to the exterior of such an ellipse is equal to e^{-c}. In case $\rho = 0$, the major and minor axes of the ellipses are parallel to the coordinate axes. Finally, a most important property of this distribution is that the random variables, x_1 and x_2, are independent if, and only if, $\rho = 0$.

The method suggested for deciding whether or not an individual has probably fired a given type of hand weapon is based on standard methods of statistical inference: hypothesis testing and classification.

It is assumed that the population of all measurements that might occur can be subdivided into several groups, and that, in turn, the variability within each group can be represented by means of a bivariate normal distribution for some particular choice of the five parameters, μ_1, μ_2, σ_{11}, σ_{22}, and ρ. These subpopulations are chosen to represent test firings with various types of weapons, and handblanks from individuals in various occupational-environmental categories. An experimental program was designed to provide estimates of the parameters of the distributions within the subpopulations.

In any actual case in which this procedure might be used as a guide, the type of weapon involved and the background of any suspect would, in general, be known. Thus, the problem is one of discriminating between two hypotheses: the null hypothesis (H_0) that the amount of Ba and Sb found on a suspect's hand(s) can be explained as a handblank from his environmental category, versus the alternative hypothesis (H_1) that the observation

resulted from the suspect having recently fired a given type of weapon. Naturally, one seeks to control the probability of both types of error that result from making a wrong decision.

Of the several approaches to controlling these error probabilities, one that seems reasonable is to fix an upper bound, α (e.g., $\alpha = 0.001$), on the probability of obtaining a false positive (rejecting H_0 when it is true), and then minimize the probability of a false negative (accepting H_0 when it is false). It can be shown that this leads to the test (for the ideal case in which all parameters are known): reject H_0 if, and only if:

$$f_0(\underset{\sim}{X})/f_1(\underset{\sim}{X}) < k \quad , \tag{9}$$

where $\underset{\sim}{X}$ is the observed vector, f_i is the bivariate normal density specified by H_i, and k is determined by α and the parameters of the two densities. The inequality (9) divides the (x_1, x_2)-plane into two regions, so that the decision procedure can be automated by preparing a set of graphs, one for each possible pair of hypotheses. In an actual case, the appropriate graph could be selected and the value of the observation would then determine into which region (accept H_0 or reject H_0) it falls.

As one would expect, the sensitivity of the decision procedure decreases as the number of unknown parameters increases. However, it is always possible, within the context of the assumed model, to quantitatively assess the probabilities of committing the two types of errors.

In order to implement and test this BVN treatment, all of the gunshot-residue and handblank data accumulated to date were double-checked (for any possible previous calculational errors), and placed on punched cards. A computer program was then written to allow one to select all or any part of these data for statistical treatment.

Four occupational-environmental categories were designated, corresponding to the degree of exposure of an individual's hands to materials which may contain barium and/or antimony. The handblank data thus far

obtained were then classified accordingly. The classification scheme and the occupations tentatively placed in each category are shown in Table 1 (as Classes A, B, C, and D).

All of the handblank data that have been collected appear to fit into three of the categories. These data, along with all of the data from 0.22-caliber revolver, 0.38-caliber revolver, and 0.45-caliber automatic pistol firings, are shown as a scatter diagram in Fig. 1.

For each of these six groups of data (three types of handblanks and firings from three types of weapons), the sample mean vector, $\bar{\underset{\sim}{X}}$, and sample covariance matrix, S, were computed in the usual way. Specifically, the measurements $\underset{\sim}{X}(1), \ldots\ldots, \underset{\sim}{X}(n)$ consist of pairs:

$$\underset{\sim}{X}(k) = \begin{bmatrix} X_1(k) \\ X_2(k) \end{bmatrix},$$

where $X_1(k)$ and $X_2(k)$ are the (common) logarithms of the amounts of barium and antimony, respectively, found in the k^{th} handlift by the NAA procedure, for $k = 1, \ldots\ldots, n$. The number of measurements, n, is different for each of the six groups. The sample means are then defined by

$$\bar{X}_i = \frac{1}{n} \cdot \sum_{k=1}^{n} X_i(k) \qquad (10)$$

for i = 1, 2, and the elements of the sample covariance matrix are given by

$$s_{ij} = \frac{1}{n-1} \cdot \sum_{k=1}^{n} \left(X_i(k) - \bar{X}_i\right)\left(X_j(k) - \bar{X}_j\right) \qquad (11)$$

for i = 1, 2, and j = 1, 2. Since the sample mean vector and covariance matrix are defined as

Table 1.

OCCUPATIONAL-ENVIRONMENTAL CATEGORIES

Class A: Negligible routine exposure to materials containing barium or antimony.

carpenters	theoretical chemists
public accountants	electronics technicians
TV technicians	chauffeurs
secretaries	electricians
watch repairmen	computer operators
gardeners	nurses
laboratory technicians	physicians
photographers	storekeepers
radioisotope technicians	

Class B: Routine exposure to Ba-containing materials, but negligible exposure to Sb-containing materials.

plumbers	mechanics
graphic artists	draftsmen
heating, air conditioning repairmen	

Class C: Routine exposure to Sb-containing materials but negligible exposure to Ba-containing materials.

none encountered to date

Class D: Routine exposure to materials containing both barium and antimony

auto mechanics	machinists
painters	maintenance men

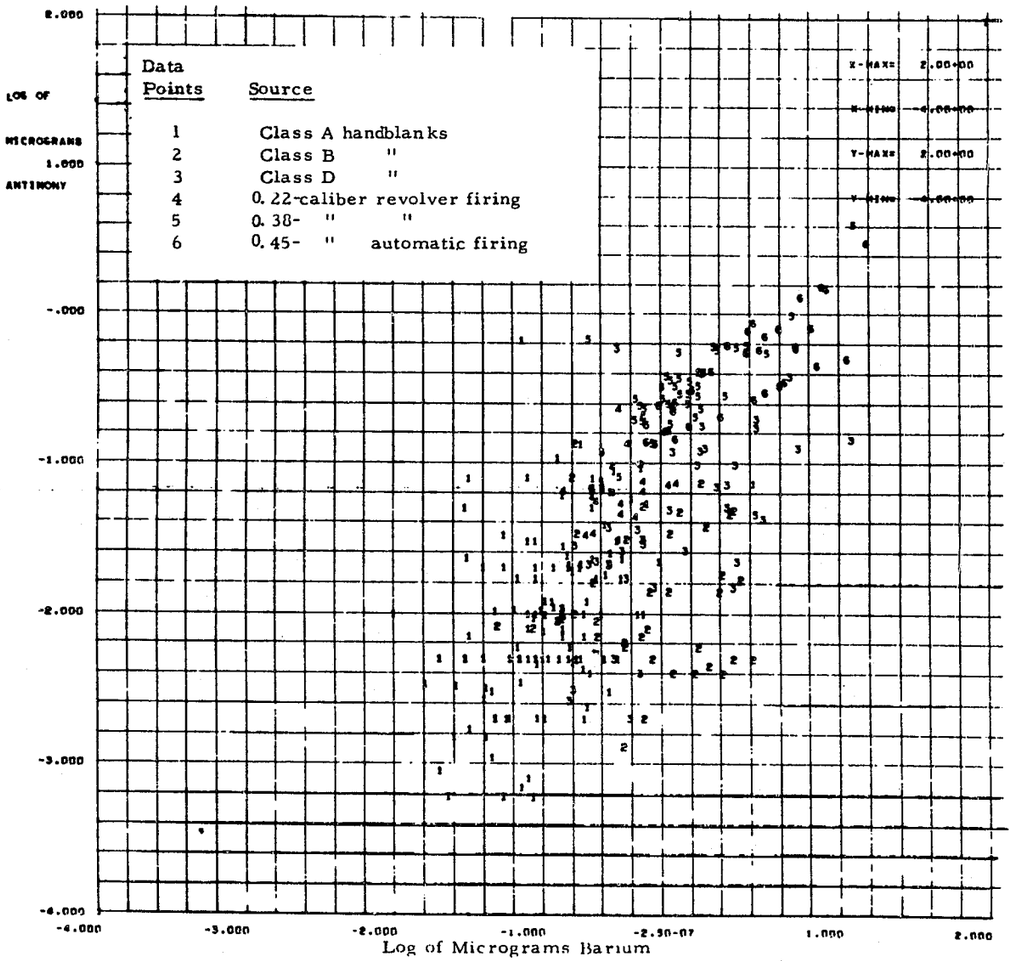

Fig. 1. All handblanks and selected firings

$$X = \begin{bmatrix} \bar{X}_1 \\ \bar{X}_2 \end{bmatrix} ,$$

and

$$S = \begin{bmatrix} s_{11} & s_{12} \\ s_{21} & s_{22} \end{bmatrix} ,$$

respectively, the sets of Eqs. 10 and 11 can be written in vector-matrix form as:

$$\bar{X} = \frac{1}{n} \cdot \sum_{k=1}^{n} X(k) , \tag{12}$$

and

$$S = \frac{1}{n-1} \cdot \sum_{k=1}^{n} \left(X(k) - \bar{X}\right)\left(X(k) - \bar{X}\right)' , \tag{13}$$

respectively, where the prime superscript denotes transpose. From the sample covariance matrix, the sample correlation, r, is computed by

$$r = s_{12}/(s_{11}\, s_{22})^{\frac{1}{2}} . \tag{14}$$

In Table 2, the results of these calculations are summarized for each of the six groups of data.

For each group, the calculated sample mean vector and covariance matrix were used as estimates of the corresponding parameters of the theoretical bivariate normal distribution that is assumed to describe the probabilistic mechanisms governing the observations. The extent of agreement between the theoretical model and the actual data is illustrated for

Table 2

SAMPLE MOMENTS FOR THREE GROUPS OF GUNSHOT-RESIDUE DATA AND THREE GROUPS OF HANDBLANK DATA

		Mean Vector	Covariance Matrix		Correlation	Sample Size
Class A handblanks	Ba	-0.98	0.15	0.10	0.46	127
	Sb	-2.03	0.10	0.33		
Class B handblanks	Ba	-0.28	0.19	-0.002	-0.01	38
	Sb	-1.91	-0.002	0.22		
Class D handblanks	Ba	-0.16	0.24	0.17	0.60	41
	Sb	-1.44	0.17	0.34		
0.22-caliber revolver firings	Ba	-0.47	0.04	0.01	0.26	15
	Sb	-1.18	0.01	0.05		
0.38-caliber revolver firings	Ba	-0.05	0.11	0.07	0.62	45
	Sb	-0.54	0.07	0.11		
0.45-caliber automatic firings	Ba	0.35	0.22	0.15	0.85	32
	Sb	-0.38	0.15	0.14		

each group in Figs. 2–7. In each figure, several ellipses of concentration, corresponding to level curves (points of constant value) of the associated bivariate normal density function, have been superimposed on a scatter diagram of the actual data points in the group. For example, the 50 percent ellipse of concentration contains an area (centered at the mean vector $\bar{X}$) to which the theoretical distribution attributes a probability of 0.50. Hence, if there is good agreement between the model and the measurements, one would expect about half of the data points to fall within this ellipse. Similar remarks apply to the other ellipses.

All of the diagrams are plotted on the same scale in order to facilitate comparisons of the size and shape of the estimated theoretical distributions among the various groups of data. Figure 8 provides an explicit comparison between two of the groups.

Two different tests were carried out on the mathematical model which was developed for the gunshot-residue and handblank data. In the first test, for each of the six categories of data (three classes of handblanks and three calibers of firings), a goodness-of-fit to the bivariate normal (BVN) distribution was performed, using the chi-squared distribution, with five degrees of freedom. In each case, the results indicated that the assumption that the data are governed by a BVN distribution is indeed valid. It should be noted that, on the average, the accuracy of this type of goodness-of-fit test increases as the number of samples increases.

In a test of a different character, an attempt was made to classify ten "unknown" data points into their correct categories, using the discriminant analysis technique described above. The results are given in Table 3. The first six "unknown points" were deliberately selected as the calculated averages of the logarithms of the amounts of Ba and Sb for each of the six categories. The next four were chosen more or less at random from the scatter diagram in Fig. 9. These points are circled on the diagram and the identifying number is indicated.

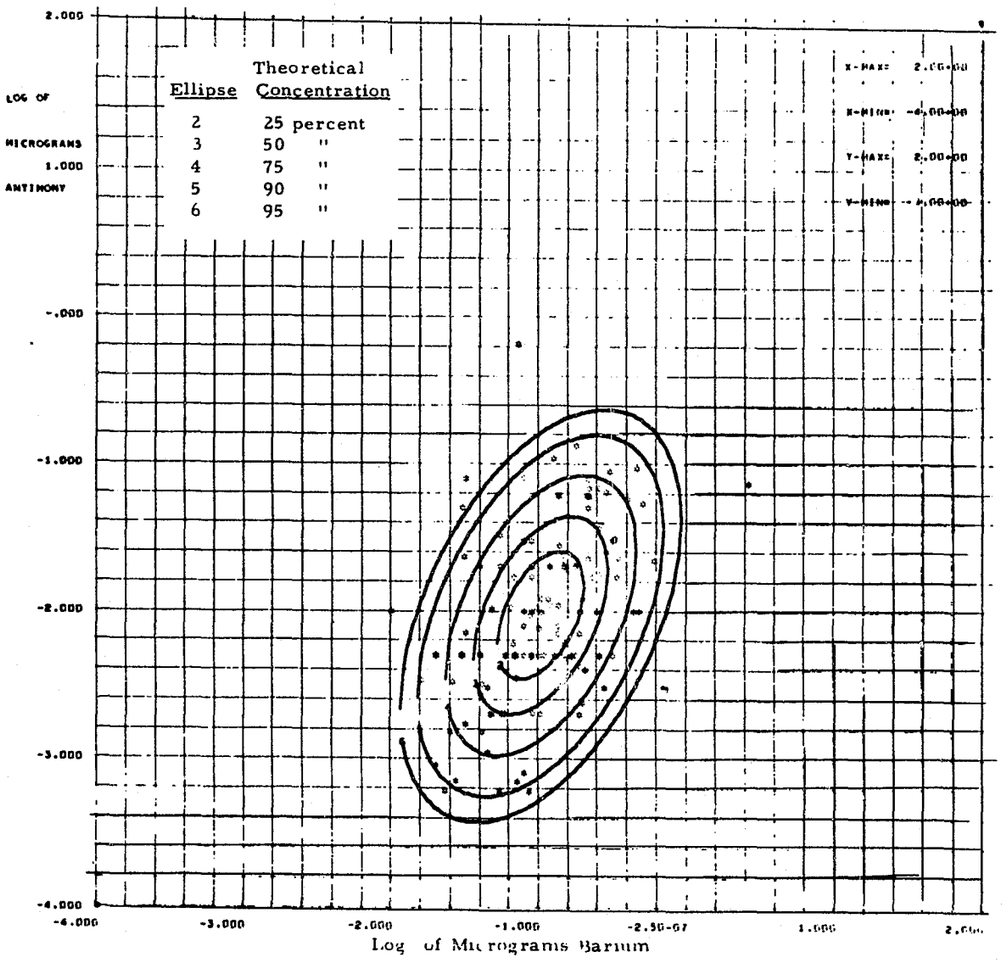

Fig. 2. Class A handblanks with theoretical ellipses of concentration

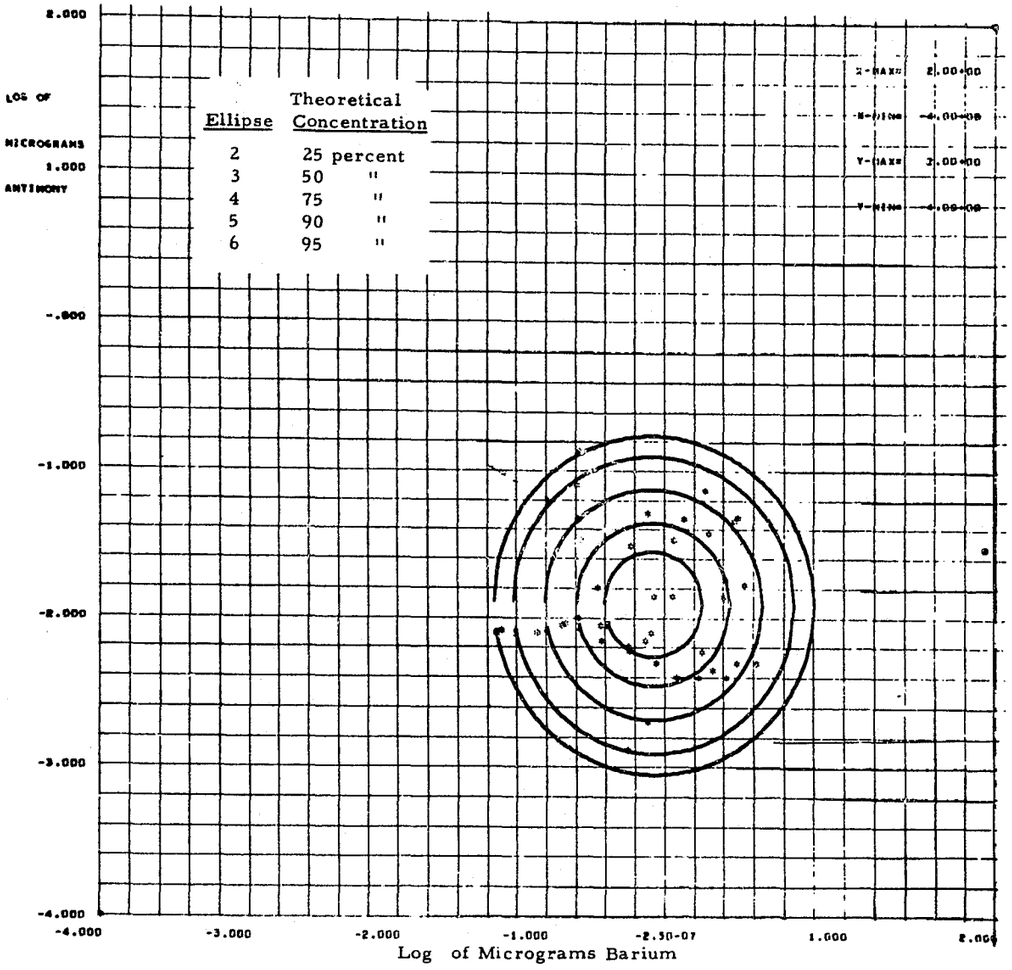

Fig. 3. Class B handblanks with theoretical ellipses of concentration

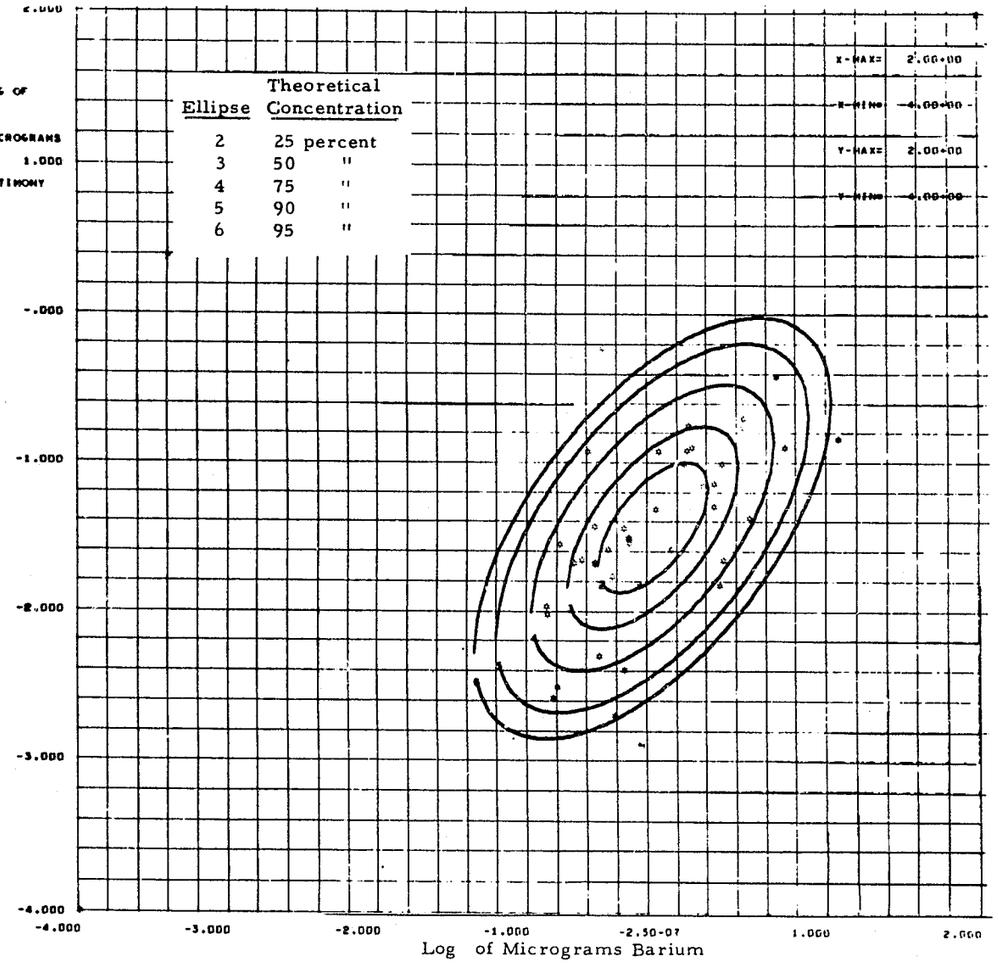

Fig. 4. Class D handblanks with theoretical ellipses of concentration

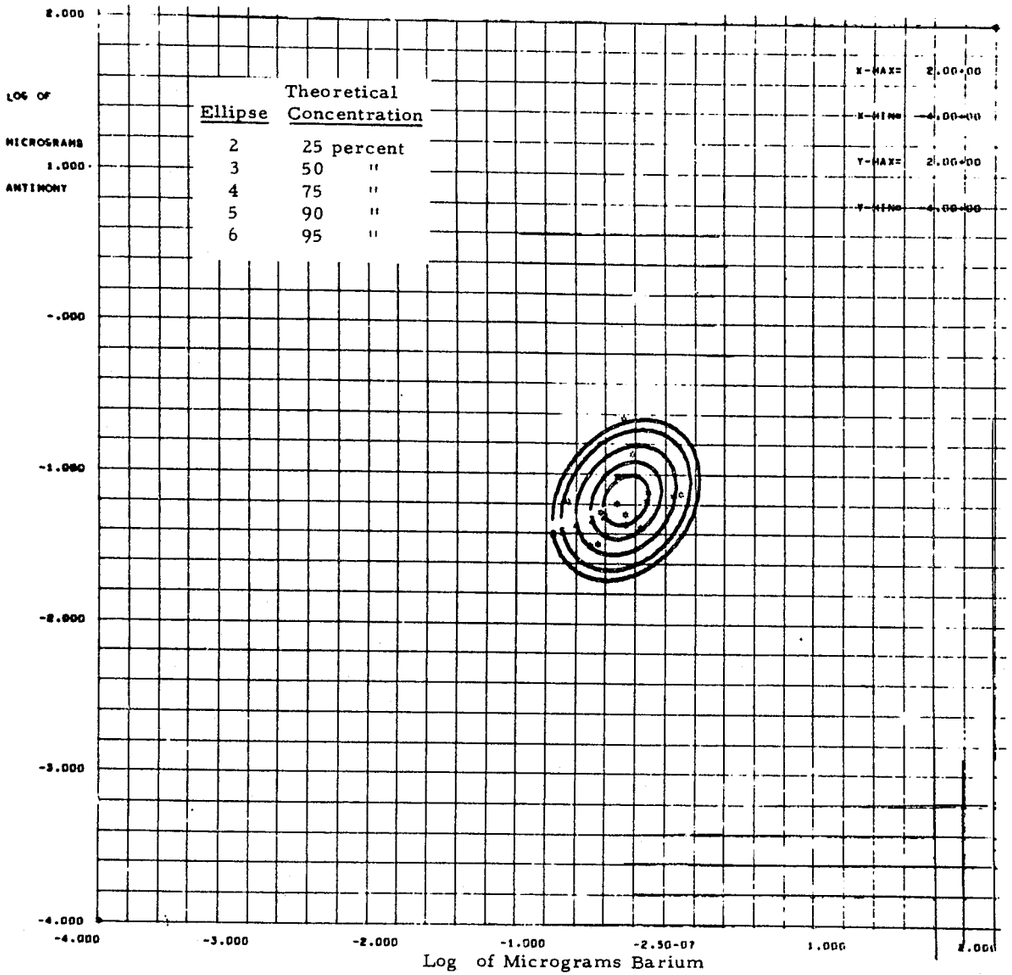

Fig. 5. 0.22-caliber revolver firings with theoretical ellipses of concentration

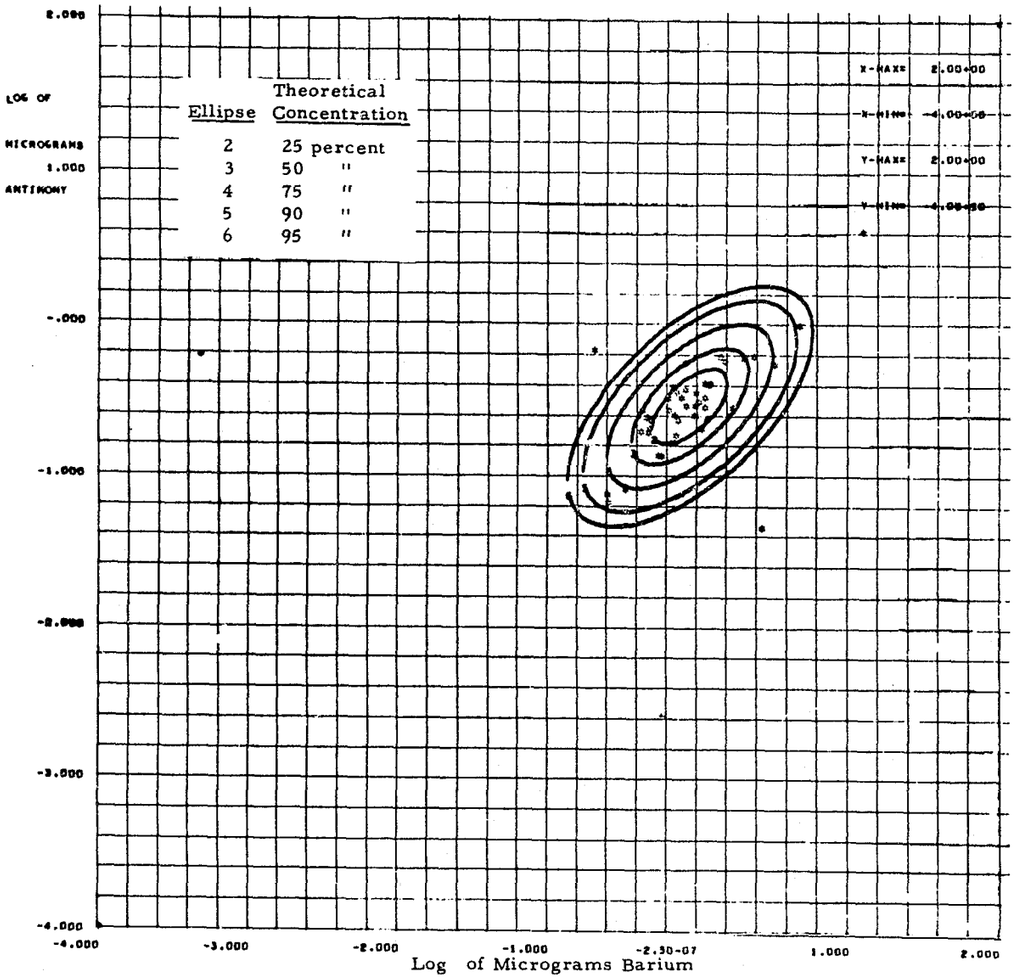

Fig. 6. 0.38-caliber revolver firings with theoretical ellipses of concentration

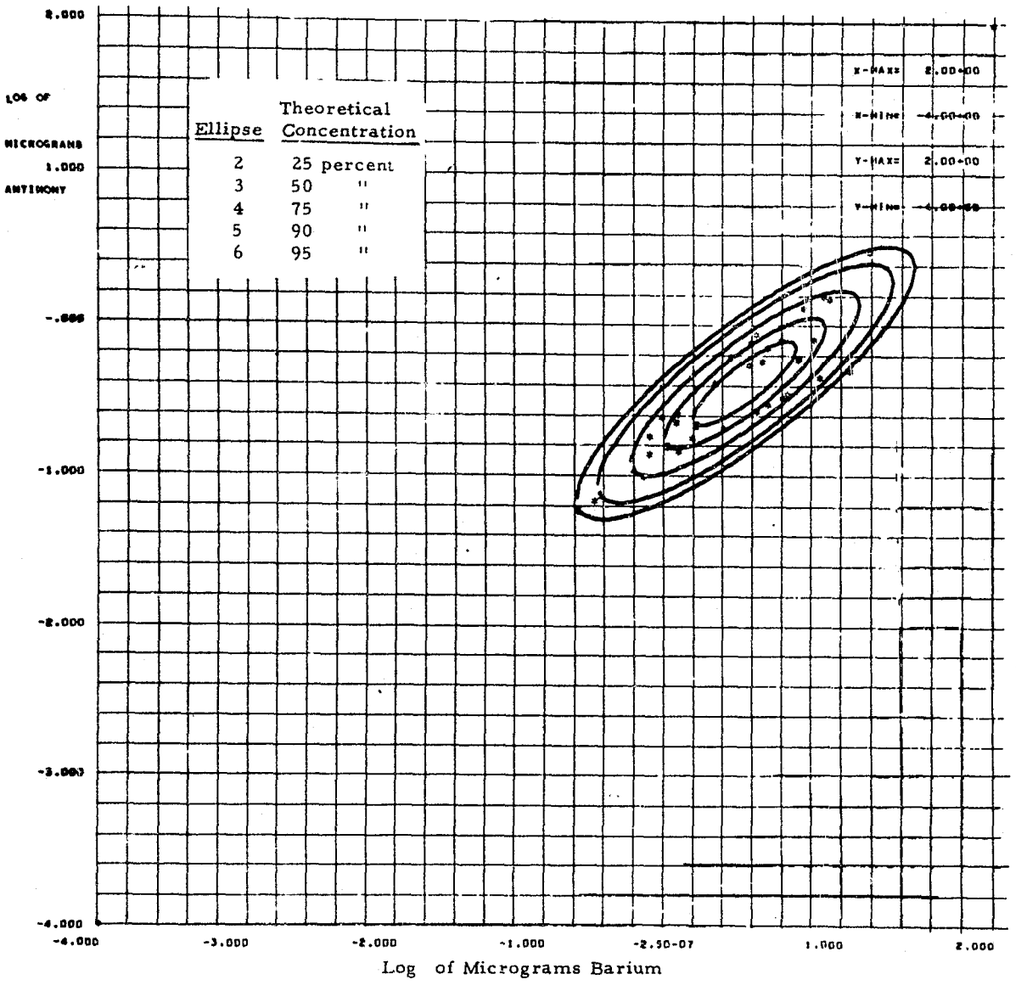

Fig. 7. 0.45-caliber automatic firings with theoretical ellipses of concentration

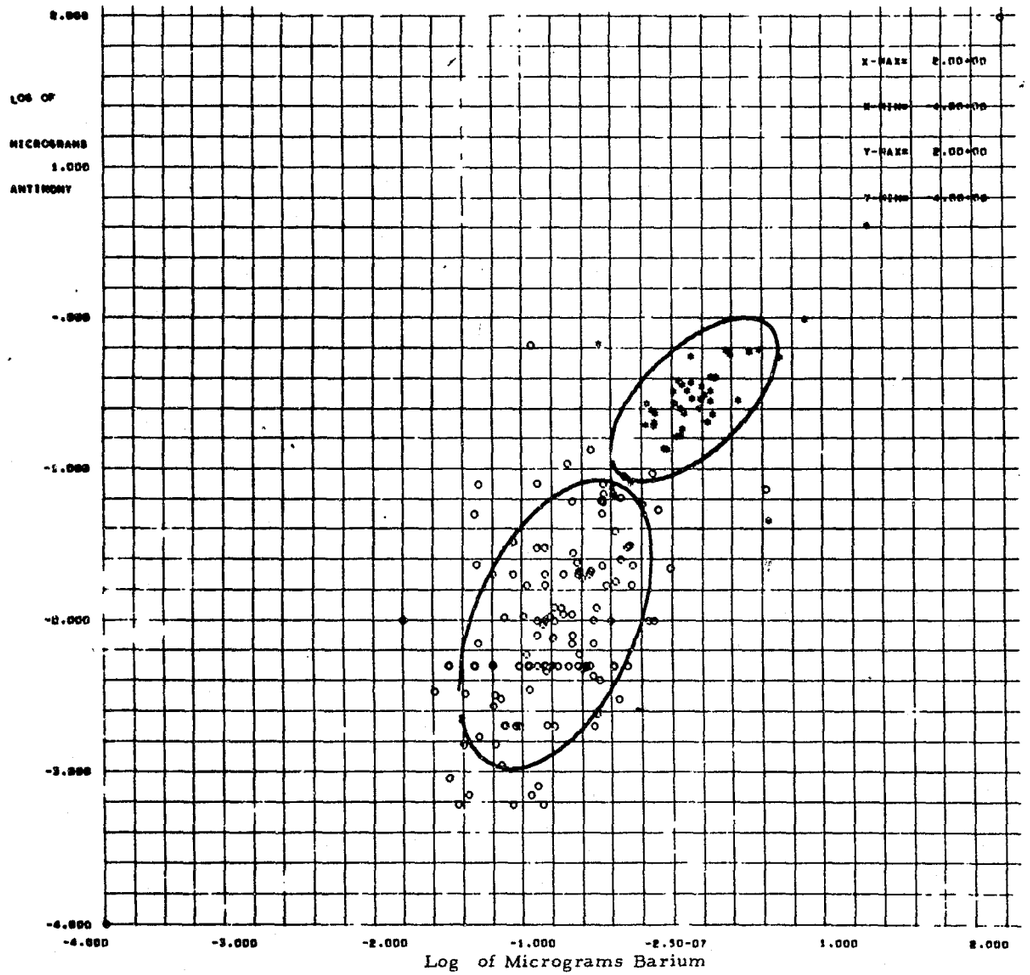

Fig. 8. Class A handblanks (o) and 0.38-caliber revolver firings (*), with 75 percent ellipses of concentration

Table 3

DISCRIMINANT ANALYSIS OF GRAPH DATA

Unknown Point	Probability That It Is a: Class A Handblank	Class B Handblank	Class D Handblank	0.22-Caliber Firing	0.38-Caliber Firing	0.45-Caliber Firing
1. Sample mean of Class A handblanks	0.68	0.18	0.14	0.0005	0.00005	0.00002
2. Sample mean of Class B handblanks	0.09	0.56	0.35	0.002	0.00001	10^{-7}
3. Sample mean of Class D handblanks	0.06	0.29	0.45	0.19	0.006	0.0008
4. Sample mean of 0.22-caliber firings	0.04	0.04	0.07	0.75	0.06	0.04
5. Sample mean of 0.38-caliber firings	0.004	0.003	0.05	0.007	0.61	0.33
6. Sample mean of 0.45-caliber firings	0.0003	0.0005	0.05	0.00001	0.31	0.64
7. High Sb Class A handblank	0.75	0.22	0.02	5×10^{-6}	0.006	6×10^{-8}
8. High Ba Class D handblank	0.00002	0.02	0.98	3×10^{-11}	0.0001	0.0001
9. Near average Class A handblank	0.90	0.04	0.06	5×10^{-8}	10^{-6}	3×10^{-6}
10. Near average 0.22-caliber firing	0.07	0.13	0.18	0.60	0.02	0.006

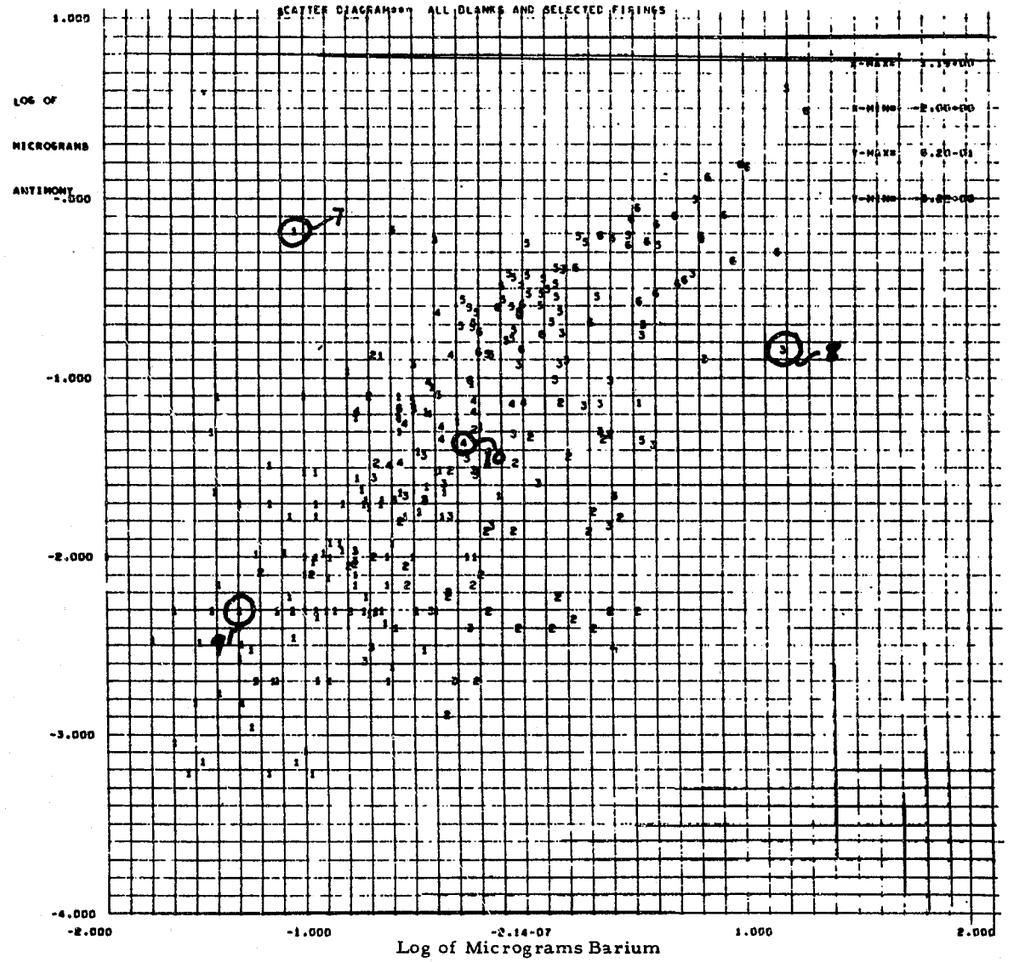

Fig. 9. All handblanks and selected firings

From inspection of Table 3, it is evident that the BVN approach developed resulted in quite reasonable probabilities. None of the actual handblank data points tested (points 1, 2, 3, 7, 8, and 9 in Table 3) gave a probability greater than 0.006 that it represented the firing of a 0.22-, 0.38-, or 0.45-caliber handgun, except point 3 — a mean Class D handblank value that gave a probability of 0.19 of being a 0.22-caliber revolver firing. The difficulty of distinguishing between a 0.22-caliber firing and some occupational handblank levels has been recognized for some time. Also, handblank samples of Classes A, B, and D were properly classified by the mathematical treatment. Similarly, points corresponding to firings of a 0.22-caliber revolver (points 4 and 10 in Table 3), a 0.38-caliber revolver (point 5 in the table), and a 0.45-caliber automatic (point 6 in the table) each resulted in a fairly high probability as a firing of the correct caliber of gun, and appreciably lower probabilities as a firing of the wrong caliber of gun — or as a handblank value.

2.2 MULTIVARIATE NORMAL TREATMENT OF MULTI-ELEMENT COMPOSITIONAL DATA ON VARIOUS EVIDENCE-TYPE MATERIALS

A statistical approach to the treatment of multi-element data involved in the NAA of specimens of paint, paper, and bullet lead has also been developed, and is described below.

As in the case of gunshot-residue and handblank data, it is desirable to have a descriptive model which adequately explains a given group of data, in order to be able to make inferences based on the data and the model in concert. The situation with respect to, say, paint is quite complicated, however, since many elements must be considered simultaneously, rather than only two elements. Measurable amounts of 25 elements have been determined by NAA in various paint samples.

The multivariate normal (MVN) distribution, which is the natural generalization of the bivariate normal (BVN) distribution from two to several dimensions, was selected as the basis of a model for these other groups

of data. The properties of the MVN distribution parallel those of the BVN distribution. It is characterized by a mean vector, $\underset{\sim}{\mu}$, and a covariance matrix, Σ, but $\underset{\sim}{\mu}$ has p components, and Σ is a pxp (symmetric) matrix, where p is the number of variables (in this case, concentrations of chemical elements) being represented.

A complete discussion and interpretation of the MVN distribution can be found in Refs. 1 and 2. One of its properties, which is extremely useful for the present purposes, is that the marginal (lower-dimensional) distributions of a MVN distribution are also MVN, and they can be characterized in a very simple way. This allows one to concentrate attention on any subset of the p variables with very little effort.

Two possible decision procedures that may be of use in actual case work have been investigated. The first is based upon ideas put forward by J. B. Parker, in Refs. 3, 4, and 5, and concerns the situation in which it must be decided whether or not two samples have a common origin. The second derives from the statistical technique of classification (sometimes called discriminant analysis), and may be useful when it is desired to categorize a sample.

Several methods have been suggested by J. B. Parker for deciding whether or not two or more samples stem from a common origin, based upon their elemental composition (Refs. 3, 4, and 5). In adapting his ideas to the problems of comparing two paint (or paper or bullet-lead) samples, the basic technique is to compute a weighted sum of the differences between the (logarithms) concentrations of those elements measured in the samples, where the weights are functions of the correlations and standard deviations which have been estimated from experimental data.

More specifically, if $\underset{\sim}{X} = (X_1, \ldots, X_k)'$ and $\underset{\sim}{Y} = (Y_1, \ldots, Y_k)'$ denote the observation vectors of the two samples being compared, and Σ denotes the covariance matrix of the distribution from which $\underset{\sim}{X}$ and $\underset{\sim}{Y}$ were obtained, then the statistic,

$$U = \tfrac{1}{2}(\underset{\sim}{X} - \underset{\sim}{Y})' \Sigma^{-1} (\underset{\sim}{X} - \underset{\sim}{Y}) \ ,$$

has a chi-squared distribution with k degrees of freedom, provided that $\underset{\sim}{X}$ and $\underset{\sim}{Y}$ come from the same multivariate normal (MVN) distribution. Consequently, it is possible to compute the probability that U exceeds any given level, and then compare this with the value of U actually obtained.

The covariance matrix, Σ, will not be known exactly, however, since it is not practically feasible to measure the elemental composition of a sample of paint (or paper or bullet-lead) of every existent type. Therefore, Σ must be replaced by the estimated covariance matrix, S, determined from experimental data, which leads to the related statistic,

$$V = (\underset{\sim}{X} - \underset{\sim}{Y})' S^{-1} (\underset{\sim}{X} - \underset{\sim}{Y}) \ .$$

The probability distribution of V is the same as a multiple of the F distribution (similar to Hotelling's T^2 distribution), tables of which are widely available.

Another approach to the problem of deciding the origin of an evidence sample can be based on the statistical theory of classification, or discriminant analysis. As before, the population of all possible samples is divided into, say, p subpopulations, according to some convenient criteria. Then, if it is required to decide from which of the p subpopulations a given sample has come, it is natural to select the subpopulation which has the largest probability of having produced the given observation. This basic concept, and various more or less sophisticated modifications of it, have found wide application. In order to apply the technique, of course, it is necessary to know the probability distribution within each subpopulation, or at least to have estimates of the distributions based on experimental data. Although it can be shown that this procedure leads, on the average, to the minimum probability of misclassification, a disadvantage of the technique is that it is generally very difficult to calculate these error probabilities.

Results of the instrumental neutron activation analysis of 99 paint samples, 147 paper samples, and 39 bullet-lead samples (including all of

these data available to date) were placed on punched cards and then analyzed on the UNIVAC 1108 computer. The paint and paper samples were, for each type of material, divided into categories which correspond to properties that could be ascertained in actual case situations. A description of the categories is given in Table 4.

The computer program first sorts the data into the appropriate categories, and then calculates, for any specified category and any desired element, the average concentration and standard deviation of the concentration of the element within the category. It also determines the number of samples in which the concentration of the element was measured, and constructs a histogram of the distribution of the element within the category. In addition, for any specified group of elements, the program determines in which samples of the category the concentrations of those elements were measured, and computes the mean vector, covariance matrix, and correlation matrix of the concentrations in the appropriate samples.

The program can treat either the raw data or the logarithms of the data. Examination of these histograms indicates that, for most elements, the distribution of the logarithm of the concentration can be fairly closely approximated by a normal distribution.

The approach described above was used in developing the statistically-designed Proposed Experimental Program, described in Section 4.

Table 4

CATEGORIES OF PAINT AND PAPER SAMPLES

Class	Paint
1	Black or grey automobile paint
2	White or off-white automobile paint
3	Blue or green automobile paint
4	Yellow, orange, or red automobile paint
5	Brown or tan automobile paint
6	Black or grey house paint
7	White or off-white house paint
8	Blue or green house paint
9	Yellow, orange, or red house paint
10	Brown or tan house paint

Class	Paper
1	White letterhead or bond paper
2	White tablet or mimeograph paper
3	Colored writing paper
4	Wrapping paper
5	Newsprint
6	Other types of paper

3. CRIMINALISTICS QUESTIONNAIRE

An effort was made to obtain background statistical information on the frequency of occurrence of various types of evidential material involved in actual criminal cases. An attempt to obtain such information from the California State Department of Justice was unsuccessful, since apparently data of this type are not regularly collected and tabulated by them. Consequently, a questionnaire was prepared, and mailed to each member of the California Association of Criminalists, asking for personal estimates of these frequencies. A copy of the questionnaire is included as Appendix 1.

Only thirteen of the sixty-eight questionnaires were returned, representing approximately a twenty percent response. Of these thirteen, nine contained usable information, and were based on a total of 40,427 samples, for an average of 4,547 samples per respondent. The average experience, in the field of criminalistics, of these nine respondents is eleven to fifteen years. There were five responses from Northern California, three from Southern California, and one of unknown geographical origin. The results of the survey are given in Tables 5 through 8.

Table 5 lists the average frequency of occurrence of various materials examined in the California criminalistics laboratories represented in the returns from this survey. The first three categories were subdivided in a self-explanatory manner. The average frequencies were calculated by weighting the estimated frequency supplied by each respondent in proportion to the number of samples he deals with annually.

Table 6 was obtained by computing unweighted averages of the individual responses while Table 7 was derived from Table 6 by normalizing the frequency distribution within the first two categories.

Table 5

AVERAGE FREQUENCY OF OCCURRENCE OF VARIOUS EVIDENCE MATERIALS

Type of Material		Frequency
1. Firearms-related samples		0.106
a) gunshot-residues	0.006	
b) bullet comparisons	0.030	
c) bullets, other	0.070	
2. Paint samples		0.068
a) paint comparisons	0.062	
b) paint, other	0.006	
3. Paper samples		0.005
a) paper comparisons	0.004	
b) paper, other	0.001	
4. Alcohol in blood, breath, or urine		0.483
5. Blood examinations, not for alcohol		0.160
6. Glass comparisons		0.016
7. Hair comparisons		0.016
8. Cloth, fiber examinations		0.020
9. Tool markings, other than firearms		0.052
10. Other materials		0.074
Total		1.000

Table 6

AVERAGE FREQUENCY OF OCCURRENCE
OF VARIOUS FIREARMS

Type \ Caliber	0.22	0.32	0.38	0.44-0.45	Other	Total
Automatic pistols	0.09	0.07	0.04	0.07	0.06	0.33
Revolvers	0.16	0.03	0.15	0.05	0.05	0.44
Rifles	x	x	x	x	x	0.12
Shotguns	x	x	x	x	x	0.08
Other	x	x	x	x	x	0.03
Total	x	x	x	x	x	1.00

Table 7

AVERAGE FREQUENCY OF OCCURRENCE
OF VARIOUS HANDGUNS

Type \ Caliber	0.22	0.32	0.38	0.44-0.45	Other	Total
Automatic pistols	0.11	0.09	0.06	0.09	0.08	0.43
Revolvers	0.21	0.04	0.19	0.06	0.07	0.57
Total	0.32	0.13	0.25	0.15	0.15	1.00

Very few respondents were able to provide estimates of the **color** distribution of paint samples. Consequently, in Table 8, the color distribution of house paints is an average of only three responses, and that of auto paints is based on only one response. For comparison purposes, automobile traffic on a local freeway was observed for approximately twenty-five minutes, during which time 387 cars were classified into one of the five color categories. The resulting frequencies are shown below the diagonal line in the automobile-paint column of Table 8.

Only one respondent supplied estimates of the frequency of occurrence of various types of paper submitted for comparison. His estimates, based on 500 samples per year, of which two percent are paper comparisons, are the following: eighty percent white tablet or mimeograph paper, and five percent each of white letterhead or bond paper, colored writing paper, wrapping paper, and newspaper.

Table 8

AVERAGE FREQUENCY OF OCCURRENCE OF VARIOUS PAINTS

Color \ Type	Auto[a]	House	Other	Total
Black or grey	0.14 / 0.08	0.01	x	x
White or off-white	0.14 / 0.15	0.13	x	x
Blue or green	0.21 / 0.25	0.01	x	x
Yellow, orange, or red	0.14 / 0.14	0.03	x	x
Brown or tan	0.07 / 0.08	0.05	x	x
Total	0.69	0.23	0.08	1.00

a The numbers below the diagonal were obtained by sampling traffic on a local freeway.

4. STATISTICALLY-DESIGNED PROPOSED EXPERIMENTAL PROGRAM

One of the specific tasks included in the Scope of Work during this contract year was to apply the statistical methods under development (described in Section 2 of this report) to the cumulative data obtained to date on gunshot-residues, paint, paper, and bullet-lead — and then statistically assess the number and types of further analyses needed to properly round-out the two-year large-scale studies of these materials that were initiated on June 1, 1968. This task was carried out, and resulted in the proposed experimental program that was included in the November 1968 Monthly report. The writeup of the proposed experimental program is self-explanatory, and hence is included in the present report as Appendix 2.

This proposed experimental program, with some modifications, is being followed in the completion of the two-year program of large-scale statistical studies of these types of evidence materials. It, and modifications made in it, were partly the result of correspondence (and telephone conversations) with Dr. Charles R. Kingston (professor at the John Jay College of Criminal Justice, and consultant to the Law Enforcement Assistance Administration on this investigation), and of meetings held at (1) the NBS laboratories at Gaithersburg, Maryland, on October 8, 1968, (2) Gulf General Atomic on March 31, 1969, and (3) Gulf General Atomic on April 10, 1969. Participants at the October 8 meeting were: C. R. Kingston (consultant to the LEAA), L. A. Mayo (LEAA), R. Emrich (LEAA), R. L. Butenhoff (DID-AEC), J. W. Hitch (DID-AEC), R. P. Hackleman (GGA), and V. P. Guinn (GGA). Participants in the March 31 meeting were: R. L. Butenhoff (DID-AEC), J. Zafiris (SFO-AEC), V. P. Guinn (GGA), R. P. Hackleman (GGA), H. R. Lukens (GGA), and H. L. Schlesinger (GGA).

Participants in the April 10 meeting were: C. R. Kingston (consultant to the LEAA), R. P. Hackleman (GGA), V. P. Guinn (GGA), and H. L. Schlesinger (GGA). Dr. Kingston's report on the April 10 meeting is included in this report as Appendix 3.

Prior to the April 10 meeting, many useful suggestions and comments, concerning this investigation, were made by Dr. Kingston — in his monthly reports to the LEAA, with copies to Gulf General Atomic. Those of particular importance, in the discussion of the statistical treatment and the proposed experimental program, are his reports for July, August, September, and November — 1968, and his report for January 1969. These monthly reports are included in this annual report as, respectively, Appendices 4 to 8.

The principal modifications made in the proposed experimental program (Appendix 2) were: (1) a change in the selection of paint samples to be analyzed — so as to include more actual (rather than brand-new) paint samples, taken from common objects, and (2) a de-emphasis of the study of paper samples.

5. COMPREHENSIVE REPORT ON ALL EARLIER WORK IN THIS INVESTIGATION

Another task included in the Scope of Work of this investigation during the period covered by the present annual report, was the preparation of a comprehensive report — to include all of the studies carried out in this investigation from its inception (May 1, 1962) through May 31, 1968, i.e., up to the start of the period covered by the present annual report. Most of the preparation of this comprehensive report has now been completed, but its sheer volume has resulted in the delay in its completion. However, it is now near completion, and will be published shortly (as GA-9807).

6. ADDITIONAL GUNSHOT-RESIDUE AND HANDBLANK MEASUREMENTS

6.1 ADDITIONAL FIRINGS

During the one-year period covered by this report, 260 additional firings of handguns were carried out, and the resulting paraffin handlifts analyzed for Ba and Sb. These measurements are summarized in Table 9. The sample numbering in Table 9 starts with Test Firing No. 145, since the first 144 test firings in this investigation are given in Table 15 in the comprehensive report (GA-9807), which covers the period, mid-1962 to mid-1968.

The data in Table 9 include a number of variables: caliber of gun, type of gun, brand of gun, and brand of ammunition. These data are summarized in Table 10. From Table 10, several trends are indicated:

1. The amount of residue deposited (Ba and Sb) increases in the caliber sequence: 0.22 → 0.38 → 0.25 → 9mm → 0.45.
2. In the 0.45-caliber automatic firings, there is little difference between Western and Federal ammunition, as far as amount of residue is concerned.
3. In the 0.45-caliber automatic firings, gun 11 appears to generally deposit more residue than gun 23, with the same type of ammunition (either Western or Federal).
4. In the 9mm automatic firings, those with gun 19 and Western ammunition resulted in larger amounts of residue than those with gun 15 and Remington ammunition — but how much of the difference is due to the brand of gun and how much to the brand of ammunition is unknown.

5. In the 0.38-caliber revolver firings (with only one gun, and one brand of ammo), the resulting Ba deposits are indistinguishable from handblank Ba values, whereas the Sb deposits are about 15 times a typical handblank Sb value.
6. In the 0.22-caliber automatic firings with the same ammo Western), two different guns of the same make nonetheless resulted in residue amounts quite different from one another.
7. In the 0.22-caliber revolver firings with the same ammo (Federal), two different guns of the same make (S. Ruger) resulted in appreciably different amounts of residue.
8. In the 0.22-caliber firings, three of the four sets of firings resulted in Ba values indistinguishable from a typical handblank Ba value, whereas the Sb values ranged from 3 to 12 times a typical handblank Sb value.
9. The relatively large amount of residue deposited by the 0.25-caliber automatic may be related to its very short barrel length (2 inches).

In Table 10, the ratio of the median Ba value to the median handblank value (0.20 μg) found for the 192 occupational Ba handblank values reported in the "Five-Year" comprehensive report (GA-9807) is given for each set of firings. Similarly, the ratio of the median Sb value to the median handblank value (0.010 μg) found for the 188 occupational Sb handblank values reported in GA-9807 is given.

The various guns mentioned in Tables 9 and 10 are described in Table 11. The data presented in Tables 9 and 10 have been treated extensively, i.e., for each set of firings, the mean, median, and range have been calculated for the Ba, Sb, and Ba/Sb values. However, these various figures will not be presented in this report, since these data will shortly be combined with the firing data included in GA-9807, and with additional firing

Table 9

COMPILATION OF GUNSHOT-RESIDUE VALUES OBTAINED FROM HANDGUNS (SINGLE FIRINGS)

Firing Test No.	Weapon Type	Weapon Calib.	Weapon No.	Brand of Ammunition	μg Elements Found Ba	μg Elements Found Sb
145	Rev.	38	24	West.	0.18	0.081
146	Rev.	38	24	West.	0.15	0.037
147	Rev.	38	24	West.	0.28	0.673
148	Rev.	38	24	West.	0.165	0.084
149	Rev.	38	24	West.	0.53	0.138
150	Rev.	38	24	West.	0.170	0.084
151	Rev.	38	24	West.	0.37	0.287
152	Rev.	38	24	West.	0.262	0.195
153	Rev.	38	24	West.	0.16	0.121
154	Rev.	38	24	West.	0.83	0.383
155	Rev.	38	24	West.	0.74	0.418
156	Rev.	38	24	West.	0.43	0.211
157	Rev.	38	24	West.	0.72	0.187
158	Rev.	38	24	West.	0.45	0.146
159	Rev.	38	24	West.	0.07	0.034
160	Rev.	38	24	West.	0.527	0.263
161	Rev.	38	24	West.	0.279	0.161
162	Rev.	38	24	West.	0.73	0.314
163	Rev.	38	24	West.	0.222	0.078
164	Rev.	38	24	West.	0.060	0.030
165	Rev.	38	24	West.	0.079	0.070
166	Rev.	38	24	West.	0.32	0.193
167	Rev.	38	24	West.	0.123	0.041
168	Rev.	38	24	West.	0.018	0.014
169	Rev.	38	24	West.	0.197	0.199

Table 9 (Continued)

Firing Test No.	Weapon Type	Weapon Calib.	Weapon No.	Brand of Ammunition	µg Elements Found Ba	µg Elements Found Sb
170	Rev.	38	24	West.	0.169	0.097
171	Rev.	38	24	West.	0.34	0.166
172	Rev.	38	24	West.	0.129	0.092
173	Rev.	38	24	West.	2.70	0.432
174	Rev.	38	24	West.	0.121	0.075
175	Rev.	38	24	West.	0.20	0.150
176	Rev.	22	21	Fed.	0.53	0.126
177	Rev.	22	21	Fed.	0.12	0.050
178	Auto.	22	2	West.	5.89	0.144
179	Rev.	22	21	Fed.	0.155	0.040
180	Auto.	22	2	West.	4.50	0.302
181	Rev.	22	21	Fed.	0.281	0.070
182	Auto.	22	2	West.	1.12	0.252
183	Rev.	22	21	Fed.	0.253	0.070
184	Auto.	22	2	West.	1.12	0.252
185	Rev.	22	21	Fed.	0.082	0.037
186	Auto.	22	2	West.	1.23	0.133
187	Rev.	22	21	Fed.	0.198	0.045
188	Auto.	22	2	West.	2.40	0.288
189	Rev.	22	21	Fed.	0.193	0.036
190	Auto.	22	2	West.	2.74	0.064
191	Rev.	22	21	Fed.	0.151	0.043
192	Auto.	22	2	West.	1.29	0.312
193	Rev.	22	21	Fed.	0.144	0.040
194	Auto.	22	2	West.	6.17	0.061
195	Rev.	22	21	Fed.	0.137	0.037
196	Rev.	22	21	Fed.	0.099	0.025

Table 9 (Continued)

Firing Test No.	Weapon Type	Weapon Calib.	Weapon No.	Brand of Ammunition	µg Elements Found Ba	µg Elements Found Sb
197	Auto.	22	2	West	5.24	0.192
198	Auto.	22	2	West.	2.04	0.130
199	Rev.	22	21	Fed.	4.92	2.91
200	Auto.	22	2	West.	2.88	0.098
201	Rev.	22	21	Fed.	0.067	0.015
202	Auto.	22	21	West.	2.46	0.184
203	Rev.	22	21	Fed.	0.487	0.058
204	Auto.	22	2	West.	0.615	0.111
205	Rev.	22	21	Fed.	0.169	0.037
206	Auto.	22	2	West.	1.27	0.127
207	Rev.	22	21	Fed.	0.215	0.066
208	Auto.	22	2	West.	0.98	0.121
209	Rev.	22	21	Fed.	0.159	0.047
210	Auto.	22	2	West.	2.65	0.521
211	Rev.	22	21	Fed.	0.351	0.101
212	Auto.	22	2	West.	2.13	1.12
213	Rev.	22	21	Fed.	0.065	0.029
214	Auto.	22	2	West.	0.73	0.076
215	Rev.	22	21	Fed.	0.055	0.016
216	Auto.	22	2	West.	0.96	0.114
217	Rev.	22	21	Fed.	0.149	0.040
218	Rev.	22	21	Fed.	0.16	0.041
219	Auto.	22	2	West.	1.67	0.045
220	Auto.	22	2	West.	0.172	0.032
221	Rev.	22	21	Fed.	0.59	0.199
222	Auto.	22	2	West.	0.221	0.043

Table 9 (Continued)

Firing Test No.	Weapon Type	Weapon Calib.	Weapon No.	Brand of Ammunition	µg Elements Found Ba	µg Elements Found Sb
223	Rev.	22	21	Fed.	0.175	0.047
224	Auto.	22	2	West.	0.291	0.096
225	Rev.	22	21	Fed.	3.01	0.024
226	Auto.	22	2	West.	1.12	0.075
227	Auto.	22	2	West.	0.84	0.015
228	Rev.	22	21	Fed.	0.081	0.015
229	Auto.	22	2	West.	0.084	0.011
230	Auto.	22	2	West.	0.240	0.033
231	Auto.	22	2	West.	2.40	0.064
232	Auto.	22	2	West.	0.341	0.138
233	Auto.	22	22	West.	0.181	0.031
234	Auto.	22	22	West.	0.092	0.022
235	Auto.	22	22	West.	0.14	0.017
236	Auto.	22	2	West.	0.97	0.141
237	Auto.	22	22	West.	0.133	0.027
238	Auto.	22	22	West.	1.56	0.074
239	Auto.	22	22	West.	0.104	0.031
240	Auto.	22	22	West.	0.29	0.057
241	Auto.	22	22	West.	0.163	0.047
242	Auto.	22	22	West.	0.351	0.036
243	Auto.	22	22	West.	0.113	<0.02
244	Auto.	22	22	West.	1.10	0.117
245	Auto.	22	22	West.	0.144	0.034
246	Auto.	22	22	West.	0.128	0.024
247	Auto.	22	22	West.	0.358	0.073
248	Auto.	22	22	West.	0.091	0.009

Table 9 (Continued)

Firing Test No.	Weapon Type	Weapon Calib.	Weapon No.	Brand of Ammunition	µg Elements Found Ba	µg Elements Found Sb
249	Auto.	22	22	West.	0.168	0.015
250	Rev.	22	7	Fed.	0.050	0.027
251	Rev.	22	7	Fed.	0.083	0.018
252	Rev.	22	7	Fed.	0.170	0.184
253	Rev.	22	7	Fed.	0.078	0.014
254	Rev.	22	7	Fed.	0.040	0.011
255	Rev.	22	7	Fed.	0.034	0.0049
256	Rev.	22	7	Fed.	0.018	0.017
257	Rev.	22	7	Fed.	0.854	0.236
258	Rev.	22	7	Fed.	1.17	0.269
259	Rev.	22	7	Fed.	0.85	0.215
260	Rev.	22	7	Fed.	0.368	0.120
261	Rev.	22	7	Fed.	0.279	0.094
262	Rev.	22	7	Fed.	0.242	0.091
263	Rev.	22	7	Fed.	0.41	0.160
264	Rev.	22	7	Fed.	0.23	0.112
265	Rev.	22	[illegible]	Fed.	0.233	0.099
266	Auto.	45	11	Fed.	13.1	4.21
267	Auto.	45	23	West.	5.93	0.552
268	Auto.	45	11	Fed.	1.38	0.280
269	Auto.	45	23	West.	6.12	1.22
270	Auto.	45	11	Fed.	3.00	1.12
271	Auto.	45	23	West.	6.34	2.55
272	Auto.	45	11	Fed.	2.14	1.14
273	Auto.	45	11	Fed.	13.0	5.16
274	Auto.	45	23	West.	15.5	0.92

Table 9 (Continued)

Firing Test No.	Weapon Type	Weapon Calib.	Weapon No.	Brand of Ammunition	µg Elements Found Ba	µg Elements Found Sb
275	Auto.	45	23	West.	1.0	0.72
276	Auto.	45	11	Fed.	8.9	1.82
277	Auto.	45	23	West.	1.91	0.47
278	Auto.	45	11	Fed.	1.89	0.78
279	Auto.	45	23	West.	2.11	0.70
280	Auto.	45	11	Fed.	2.77	1.55
281	Auto.	45	11	West.	8.55	2.17
282	Auto.	45	23	Fed.	3.75	1.31
283	Auto.	45	11	West.	9.72	2.56
284	Auto.	45	11	West.	2.58	1.00
285	Auto.	45	11	West.	7.20	2.74
286	Auto.	45	23	Fed.	2.17	0.75
287	Auto.	45	11	West.	3.36	1.60
288	Auto.	45	23	Fed.	2.17	0.75
289	Auto.	45	11	West.	3.17	1.27
290	Auto.	45	23	Fed.	6.44	2.03
291	Auto.	45	11	West.	7.9	2.70
292	Auto.	45	23	Fed.	1.95	0.81
293	Auto.	45	11	West.	6.28	1.71
294	Auto.	45	23	West.	2.07	0.343
295	Auto.	45	23	West.	11.00	2.32
296	Auto.	45	11	Fed.	5.32	0.84
297	Auto.	45	23	West.	1.75	0.208
298	Auto.	45	11	Fed.	13.70	3.50
299	Auto.	45	23	West.	2.73	0.373
300	Auto.	45	11	Fed.	5.26	1.41

Table 9 (Continued)

Firing Test No.	Weapon Type	Weapon Calib.	Weapon No.	Brand of Ammunition	µg Elements Found Ba	µg Elements Found Sb
301	Auto.	45	11	Fed.	7.00	1.02
302	Auto.	45	11	Fed.	13.4	2.12
303	Auto.	45	11	Fed.	7.88	2.09
304	Auto.	45	11	West.	1.84	0.382
305	Auto.	45	11	West.	0.88	0.132
306	Auto.	45	11	Fed.	26.1	3.24
307	Auto.	45	23	Fed.	5.88	1.73
308	Auto.	45	11	Fed.	5.24	1.40
309	Auto.	45	23	Fed.	2.35	0.64
310	Auto.	45	11	Fed.	19.9	5.98
311	Auto.	45	23	Fed.	3.79	1.29
312	Auto.	45	11	Fed.	5.59	1.63
313	Auto.	45	23	Fed.	4.06	1.16
314	Auto.	45	11	Fed.	2.09	0.58
315	Auto.	45	23	Fed.	2.44	0.89
316	Auto.	45	11	Fed.	2.89	1.11
317	Auto.	45	23	Fed.	2.96	0.93
318	Auto.	45	11	Fed.	8.68	3.01
319	Auto.	45	23	West.	3.94	0.69
320	Auto.	45	23	West.	1.61	0.247
321	Auto.	45	11	West.	1.65	1.83
322	Auto.	45	23	West.	9.08	5.78
323	Auto.	45	11	West.	6.22	1.26
324	Auto.	45	11	West.	7.16	1.92
325	Auto.	45	23	West.	8.05	2.23
326	Auto.	45	23	West.	2.04	1.38

Table 9 (Continued)

Firing Test No.	Weapon Type	Weapon Calib.	Weapon No.	Brand of Ammunition	µg Elements Found Ba	µg Elements Found Sb
327	Auto.	45	23	West.	0.96	0.342
328	Auto.	45	23	West.	2.15	0.521
329	Auto.	45	23	West.	1.34	0.355
330	Auto.	45	23	West.	2.91	0.60
331	Auto	9mm	15	Rem.	2.27	0.93
332	Auto	9mm	15	Rem.	3.22	0.57
333	Auto.	9mm	15	Rem.	2.77	0.84
334	Auto.	9mm	15	Rem.	1.64	0.247
335	Auto.	9mm	15	Rem.	1.27	0.38
336	Auto.	9mm	15	Rem.	1.11	0.36
337	Auto.	9mm	15	Rem.	1.02	0.58
338	Auto.	9mm	15	Rem.	1.69	0.49
339	Auto.	9mm	15	Rem.	1.22	0.46
340	Auto.	9mm	19	West.	4.80	3.05
341	Auto.	9mm	15	Rem.	2.14	0.71
342	Auto.	9mm	19	West.	1.36	0.70
343	Auto.	9mm	15	Rem.	0.84	0.164
344	Auto.	9mm	19	West.	0.75	0.296
345	Auto.	9mm	15	Rem.	0.56	0.124
346	Auto.	9mm	19	West.	1.63	0.495
347	Auto.	9mm	19	West.	0.413	0.123
348	Auto.	9mm	15	Rem.	0.419	0.142
349	Auto.	9mm	19	West.	1.26	0.218
350	Auto.	9mm	15	Rem.	0.64	0.082
351	Auto.	9mm	19	West.	1.31	0.719
352	Auto.	9mm	15	Rem.	0.247	0.192

Table 9 (Continued)

Firing Test No.	Weapon Type	Weapon Calib.	Weapon No.	Brand of Ammunition	µg Elements Found Ba	µg Elements Found Sb
353	Auto.	9mm	19	West.	0.434	0.82
354	Auto.	9mm	15	Rem.	0.284	0.241
355	Auto.	9mm	19	West.	0.440	0.187
356	Auto.	9mm	15	Rem.	0.464	0.338
357	Auto.	9mm	19	West.	2.80	0.68
358	Auto.	9mm	15	Rem.	0.528	0.086
359	Auto.	9mm	19	West.	6.65	3.18
360	Auto.	9mm	15	Rem.	0.57	0.133
361	Auto.	9mm	19	West.	2.34	0.86
362	Auto.	9mm	15	Rem.	0.618	0.113
363	Auto.	9mm	19	West.	8.59	3.23
364	Auto.	9mm	15	Rem.	0.483	0.149
365	Auto.	9mm	19	West.	5.34	2.57
366	Auto.	9mm	15	Rem.	2.33	0.232
367	Auto.	9mm	19	West.	6.74	3.17
368	Auto.	9mm	15	Rem.	8.56	1.20
369	Auto.	9mm	19	West.	1.44	0.77
370	Auto.	9mm	15	Rem.	0.556	0.145
371	Auto.	9mm	19	West.	4.66	0.52
372	Auto.	9mm	15	Rem.	0.51	0.227
373	Auto.	9mm	19	West.	2.47	0.38
374	Auto.	9mm	19	West.	2.52	1.57
375	Auto.	9mm	15	Rem.	1.56	0.215
376	Auto.	9mm	19	West.	9.29	4.20
377	Auto.	9mm	15	Rem.	1.04	0.183
378	Auto.	9mm	19	West.	2.66	1.15

Table 9 (Continued)

Firing Test No.	Type	Weapon Calib.	No.	Brand of Ammunition	µg Elements Found Ba	Sb
379	Auto.	9mm	15	Rem.	3.24	0.63
380	Auto.	9mm	19	West.	1.94	0.99
381	Auto.	9mm	19	West.	3.09	1.52
382	Auto.	9mm	15	Rem.	1.00	0.266
383	Auto.	9mm	15	Rem.	1.18	0.130
384	Auto.	9mm	19	West.	2.19	1.29
385	Auto.	9mm	15	Rem.	0.47	0.102
386	Auto.	9mm	19	West.	2.67	0.84
387	Auto.	9mm	15	Rem.	2.67	0.354
388	Auto.	9mm	19	West.	4.92	2.37
389	Auto.	9mm	19	West.	2.12	1.09
390	Auto.	9mm	15	Rem.	0.73	0.156
391	Auto.	25	1	Rem.	4.10	0.604
392	Auto.	25	1	Rem.	2.43	0.296
393	Auto.	25	1	Rem.	3.81	0.148
394	Auto.	25	1	Rem.	3.47	0.341
395	Auto.	25	1	Rem.	14.2	1.67
396	Auto.	25	1	Rem.	1.44	0.266
397	Auto.	25	1	Rem.	2.98	0.624
398	Auto.	25	1	Rem.	3.84	0.579
399	Auto.	25	1	Rem.	1.09	0.150
400	Auto.	25	1	Rem.	1.61	0.307
401	Auto.	25	1	Rem.	2.26	0.463
402	Auto.	25	1	Rem.	2.22	0.359
403	Auto.	25	1	Rem.	2.12	0.419
404	Auto.	25	1	Rem.	1.05	0.253

Table 10

TEST FIRING RESULTS SUMMARIZED

Caliber	Type	Ammo	Gun No.	No. of Firings	Median Ratios* Ba	Sb
45	Auto.	West.	23	20	12	64
45	Auto.	Fed.	23	13	15	120
45	Auto.	Fed.	11	20	31	150
45	Auto.	West.	11	12	31	180
9mm	Auto.	West.	19	27	12	86
9mm	Auto.	Rem.	15	33	5.1	23
38	Rev.	West.	24	31	1.0	15
25	Auto.	Rem.	1	14	12	35
22	Auto.	West.	2	31	6.3	12
22	Auto.	West.	22	16	0.76	3.1
22	Rev.	Fed.	7	16	1.2	9.6
22	Rev.	Fed.	21	27	0.80	4.1

* Ratio of median firing values to median handblank values (0.20 µg Ba, 0.010 µg Sb)

Table 11

DESCRIPTION OF GUNS LISTED IN TABLES 9 AND 10

Gun No.	Type	Caliber	Make	Barrel Length
1	Auto.	0.25	Junior Colt	2 inches
2	Auto.	0.22	Hi Standard	6 inches
7	Rev.	0.22	S. Ruger Single 6	4 inches
11	Auto.	0.45	Army Model 1911A1	5 inches
15	Auto.	9mm	Luger	4 inches
19	Auto.	9mm	Mauser	4 inches
21	Rev.	0.22	S. Ruger Single 6	4 inches
22	Auto.	0.22	Hi Standard	6 inches
23	Auto.	0.45	Colt	5 inches
24	Auto.	0.22	Hi Standard	6 inches

data obtained during the fall of 1969, in a comprehensive report devoted entirely to the gunshot-residue studies (GA-9829), covering all the work conducted from 1962 through 1969. This comprehensive report will include a thorough statistical treatment of all the gunshot-residue and handblank data.

6.2 ADDITIONAL HANDBLANK MEASUREMENTS

During this report period, 30 additional occupational-handblank samples were obtained from 16 different people and analyzed for Ba and Sb by the regular paraffin handlift – NAA procedure. Five different occupations were represented by these samples. The Ba and Sb values found are shown in Table 12. The numbering in the table starts with number 98, since the occupational-handblank values found with the first 97 persons studied are given in the "Five-Year" report (GA-9807), numbered 1 through 97.

Inspection of Table 12 reveals only one unusually high handblank Sb value: the 0.232 μg value found on the left hand of one typist (person number 106). As for the Ba values, all appear to be more-or-less normal, except for the high value found on the right hand of one of the librarians (number 113), 0.526 μg, and the high values found on both hands of three of the four typists (numbers 106, 108, and 109), which showed a median value of 0.93 μg Ba. However, all of the corresponding Sb values were low values, except for the 0.232 μg Sb value found on the left hand of the one typist mentioned earlier (number 106).

Half of the values obtained were from smokers; half from nonsmokers. However, an examination of the results reveals no significant difference in either the Ba or the Sb results when sorted according to w whether persons were smokers or nonsmokers. That is, the median Ba value for 15 hands of nonsmokers was 0.090 μg; that for 15 hands of smokers was 0.067 μg. Similarly, the median Sb value for 15 hands of nonsmokers was <0.006 μg; that for 15 hands of smokers as <0.003 μg.

Table 12

ADDITIONAL HANDBLANK VALUES FROM 16 PERSONS

Person No.	Occupation	Hand	μg Found Ba	μg Found Sb
98	Housewife	R	0.024	<0.006
		L	0.090	<0.004
99	Housewife	R	0.080	0.010
		L	0.139	<0.005
100	Housewife	R	0.051	<0.006
		L	0.044	<0.005
101	Housewife	R	0.034	<0.003
		L	0.020	<0.003
102	Housewife	R	0.020	<0.003
		L	0.009	<0.003
103	Housewife	R	0.045	<0.003
		L	0.067	0.030
104	Housewife	R	-----	------
		L	0.018	0.021
105	Teacher	R	0.029	<0.003
		L	0.018	<0.003
106	Typist	R	0.648	0.004
		L	0.712	0.232
107	Typist	R	0.151	0.0003
		L	0.139	0.018
108	Typist	R	0.703	<0.003
		L	1.16	0.008
109	Typist	R	2.23	0.003
		L	2.89	0.007
110	Editor	R	0.048	<0.002
		L	0.106	<0.007
111	Editor	R	0.160	0.004
		L	0.083	<0.001
112	Librarian	R	0.074	<0.001
		L	0.117	0.015
113	Librarian	R	0.526	0.005
		L	-----	-----

6.3 PARAFFIN CONTAMINATION PROBLEM

The firing values shown in Table 9 are not listed in the order in which the tests were run, but instead have been grouped according to gun caliber — for convenience in examination of the results. Actually, the first firings performed in this series were ones with a 0.25-caliber automatic, which deposited relatively large amounts of residue. The results also indicated that fairly large amounts of residue were measurable on the left hand of each firer of this gun, even though each person fired with his right hand. In addition, the paraffin blank values for Ba and Sb, found in samples of the paraffin used in this sequence of measurements, were much higher than normal. The problem was traced to contamination of the paraffin in the dish containing the molten paraffin — from the brush used to apply the paraffin to the hand. With such large amounts of residue, the brush picked up Ba and Sb, transferring some of it back into the molten paraffin in the dish each time the brush was dipped in it to take up fresh paraffin.

The type of paraffin used typically contained, before use, <0.005 μg Ba and <0.003 μg Sb in the amount applied to the back of the hand in the usual paraffin handlift procedure. Usually, there is a small amount of contamination of the molten paraffin in the dish, from the brush used, when a series of firing hands are sampled one after the other, using the same dish of molten paraffin. With suitable care, and in the absence of unusually large amounts of gunshot-residue, however, the degree of contamination is quite small. For example, the median Ba value found in 13 paraffin blank samples, taken at various stages during the hand-sampling of firers included in Table 9, was 0.031 μg, which is a relatively small amount of the element when compared with the amounts found in either a typical occupational-handblank or subsequent to a typical firing. Similarly, the median Sb value found in the same 13 paraffin blank samples was 0.006 μg — at most, about twice the Sb level of the fresh paraffin, and small compared with a typical occupational-handblank or firing Sb value.

In the 0.25-caliber firings mentioned earlier, however, considerably larger Ba and Sb paraffin blank values were observed. During these hand samplings, two samples of molten paraffin were taken from the dish for analysis (taking about the same amount that is normally used for handlift application). The two Ba values observed were 0.247 μg and 0.269 μg, and the two corresponding Sb values were 0.051 μg and 0.103 μg. Appreciable contamination of the molten paraffin in the dish, from each of the preceding firing handlifts taken with that particular dish of paraffin, was thus indicated. Thereafter, the paraffin application technique was modified, so as to minimize such contamination: (1) a given dish of molten paraffin was used for a smaller number of firings, particularly with guns of higher caliber, (2) if both hands of the firers were to be sampled, the nonfiring hand was sampled first, (3) the molten paraffin was allowed to flow onto the hand (rather than using a brushing action), (4) brushes were replaced more frequently, and (5) later, cotton swabs (used only once) were used instead of brushes. With these changes in procedure, no further contamination problems were encountered.

The values obtained in the first series of 0.25-caliber firings, in which appreciable contamination of the molten paraffin in the dish occurred, are not included in Table 9.

6.4 RESIDUE DEPOSITION ON NONFIRING HAND

In a fair number of the firings listed in Table 9, both the right hand (gunhand) and the left hand (nonfiring hand) of the firer were sampled, and both paraffin lifts then analyzed for Ba and Sb. The results obtained are summarized in Table 13.

It is evident that, with firings of guns that produce quite large amounts of gunshot-residue (generally the guns of higher caliber, such as the 0.45-caliber, 0.25-caliber, and 9mm guns listed), the amount of residue deposited on the back of the nonfiring (left) hand is much less than the amount deposited

on the back of the gunhand (right hand) — typically being, at most, 10-12% of the right-hand Ba amount, and, at most, 12-24% of the right-hand Sb amount. These are maximum percentages, because they include variable amounts of occupational-handblank Ba and Sb already present before the firing.

Table 13

BARIUM AND ANTIMONY FOUND ON THE LEFT (NON-FIRING) HAND OF FIRERS

Gun Caliber	No. of Firings	Median of Left-Hand Value, Expressed as % of Right-Hand Value	
		Ba	Sb
0.45	4	10	12
9mm	2	12	17
0.25	14	10	24
0.22	18	58	76

In the case of the 0.22-caliber firings, however, the Ba values observed on the left hand of the firer, after a firing, were almost as large as the gunhand values (a median of 58% of the gunhand value), and the Sb values were also large (a median of 75% of the gunhand value). This does not mean that nearly equal amounts of Ba and Sb were deposited on both hands of a firer, but rather are simply the result of the very small amount of residue deposited by a typical 0.22-caliber gun — compared with typical handblank values. Of the 18 0.22-caliber firings cited in Table 13, 16 were firings of gun 7, with Federal ammunition. In these firings, the median gunhand values observed were only 0.23 μg Ba and 0.096 μg Sb, including any handblank material also present — as compared with median occupational-handblank values of about 0.20 μg Ba and 0.010 μg Sb.

In the case of the firings of 0.45-caliber, 0.25-caliber, and 9mm guns, the Ba and Sb amounts typically deposited on the back of the gunhand are approximately 22 to 145 times, 11 to 34 times, and 8 to 54 times the respective Ba and Sb median handblank values — and hence the handblank contribution to the gross values measured is rather small in these cases. If the Ba amounts found on the left hands of firers were only the typical Ba handblank level (0.20 μg) the gross measured Ba amount on the left hand should thus range from about 4 to 11% of the value found on the gunhand, for these three calibers of guns. As mentioned above, the observed left-hand Ba values were typically only 10 to 12% of the right-hand Ba values, thus indicating that very little gunshot-residue material was deposited on the left hand. However, the Sb results give a somewhat different picture. If the Sb amounts found on the left hands of firers of these guns were only the typical Sb handblank level (0.010 μg), the gross measured Sb amount on the left hand should only range from about 1 to 3% of the value found on the gunhand. But, as shown in Table 13, the left-hand Sb values instead ranged from 12 to 24% of the gunhand Sb values — thus indicating that a significant amount of gunshot-residue material appeared on the left hand of the firer. This anomaly is, as yet, unexplained — although some plausible (but unproved) explanations are discussed in section 8.

6.5 DIRECT DETERMINATION OF BARIUM AND ANTIMONY IN CARTRIDGE PRIMERS

Some measurements were made to see whether Ba and Sb could be determined in cartridge primers (removed from their copper containers) by means of instrumental neutron activation analysis. Because of the possible explosion hazard, these samples were not activated in the reactor, but instead were activated with 14-MeV neutrons produced by one of the Cockcroft-Walton generators. The activation products detected were

2.554-minute ^{137m}Ba (0.662 MeV gamma ray), produced by the ^{138}Ba(n, 2n) ^{137m}Ba reaction, and 15.89-minute ^{120}Sb (29 keV tin X-ray detected, Sn being the product of the electron-capture decay of ^{120}Sb), produced by the ^{121}Sb(n, 2n)^{120}Sb reaction.

The primer from a Federal 0.45-caliber cartridge was found to contain 23% Ba and 3.1% Sb. Expressed as $Ba(NO_3)_2$ and Sb_2S_3 (the chemical forms used in making the primers), these values correspond to 44% $Ba(NO_3)_2$ and 4.5% Sb_2S_3. Similarly, the primer from a Remington 0.38-caliber cartridge was found to contain 24% Ba and 2.1% Sb, corresponding to 46% $Ba(NO_3)_2$ and 3.0% Sb_2S_3. The gunpowder from the Federal 0.45-caliber cartridge was also analyzed for Ba and Sb. As expected, these elements were not detectable in the gunpowder.

The Ba and Sb results obtained on these two primers, via NAA, are generally in the range expected from the few primer compositions for which information is available from the manufacturers. Three companies that did supply information on the composition of their cartridge primers indicated $Ba(NO_3)_2$ levels, respectively, of 29.5%, 42.0%, and 39.4%. Their corresponding reported Sb_2S_3 levels were, respectively, 14.4%, 8.7%, and 8.1%. All three also use ~40% lead styphnate in their primers, and 1 to 4.5% tetracene. One also uses 4.4% pentaerythritol tetranitrate, and one uses 8.4% nitrocellulose. One employs 6.5% aluminum in their primers, and another employs 6.0% calcium silicide.

In this investigation, attention has been concentrated on the NAA determination of Ba and Sb. Lead is not determined, since it is one element not detectable with really good sensitivity by means of NAA. It is possible, however, that it would be fruitful to supplement the NAA determinations of Ba and Sb by an atomic absorption measurement of Pb – for the detection of gunshot residues. Early in this investigation, copper was also measured via NAA, in gunshot-residue samples, since some Cu is also deposited on the back of the gunhand in firings of handguns. However,

it was found that typical Cu handblank levels were quite large and quite variable, since Cu is a fairly commonly used element, e.g., in jewelry. As a result, it was decided that the determination of Cu in paraffin handlifts was not very helpful in determining whether or not a person had recently fired a gun, and hence no further measurements were made.

To the extent that different manufacturers employ some other constituents in their primers that are distinctive, such as the 6.5% Al used by one of the three manufacturers mentioned above, and the 6.0% $CaSi_2$ used by another, there might be some advantage to also looking for such distinctive elements. If detected, the presence of significant amounts of such elements, in addition to Ba and Sb (and perhaps Pb and Cu), in handlift samples, might not only indicate that the person had recently fired a gun, but also indicate the type of primer used. Unfortunately, Al and Ca are not well suited to determination via NAA with subsequent radiochemical separation (from the large interfering levels of 14.96-hour ^{24}Na and 37.29-minute ^{38}Cl), because of the rather short half-lives of their (n, γ) products: 2.31-minute ^{28}Al and 8.8-minute ^{49}Ca. Silicon forms a longer-lived (n, γ) product – 2.62-hour ^{31}Si – but this radionuclide only emits a gamma ray in 0.07% of its disintegrations.

It would appear that, if one wished to employ NAA not only to ascertain the probability that a given individual had recently fired a gun (via detection of significant levels of Ba and Sb in a handlift obtained from that individual), but also to establish the brand and caliber of ammunition that he or she had fired, some characteristic tagging procedure would have to be employed by each manufacturer. This approach (employing low levels of added unusual, highly-sensitive, elements – such as some of the rare-earth elements) was proposed early in this investigation, and was then tested experimentally. Cartridge cases, gunpowder, and primers were tagged, in a number of these studies, with several different elements, and

these elements were then detected by NAA in handlifts taken after test firings of the tagged ammunitions. Although the results looked promising, some complications arose, and the probability of convincing cartridge manufacturers that they should each use a number of different assigned tagging elements in the production of their cartridges seems remote.

7. VISITS TO VARIOUS MANUFACTURERS

In order to gain further insight into the processing techniques typically used by manufacturers of paper, paint, and cartridges, visits were made to several such manufacturing plants. Much useful information was obtained, and extensive notes were taken. A synopsis of the principal relevant findings is given below.

7.1 PAPER MANUFACTURERS

Paper is produced in a semi-continuous process. While the paper-making (or Fourdrinier) machine operates continuously, batches of raw or processed materials are kept in "chests" awaiting use. This explains the large variations found within single batches analyzed during the previous contract period. Another source of variation in the finished product occurs at the time that the paper is cut into 8 1/2-in. x 11-in. sheets. Paper from six half-rolls is simultaneously slit, cut, and stacked alternately in a single room. It is not unusual for paper produced at quite different times to be associated in the same ream.

7.2 PAINT MANUFACTURERS

Paint is produced in a true batch process. The largest batch size observed was 1,000 gallons. Paint manufacturers are often able to distinguish amongst different batches of the same product on the basis of physical properties, so neutron activation analysis should be capable of at least the same discrimination.

7.3 CARTRIDGE MANUFACTURERS

One manufacturer of cartridge primers stated that his producation of mercury fulminate primers declined rapidly after 1945, was abandoned by 1950, and that he believes that other American companies had very similar experiences. However, he stated that $Ba(NO_3)_2$ and Sb_2S_3 were not absolutely necessary in primer production, and that, although quite rarely, he had seen analyses of primers in which one of these materials was absent. He mentioned specifically primers from Canadian Industries Limited in this connection. However, almost without exception, all recently-and-currently-produced U.S. cartridge primers do contain these two compounds. Bullet lead is alloyed by suppliers, using arsenic or antimony as hardening agents, added to the manufacturer's specifications, and delivered in pigs weighing about 100 pounds. Antimony is used much more than arsenic.

8. DISCUSSION

8.1 GUNSHOT-RESIDUE AND HANDBLANK STUDY

The major effort in this contract period, which was devoted primarily to the development of NAA as a means to determine whether or not a person recently discharged a firearm, resulted in the acquisition of an appreciable amount of useful analytical data regarding the quantity of barium and antimony removed from the hands of persons who had recently fired a weapon. Also, preliminary utilization of the BVN approach to interpreting the data seems to indicate that this approach is correct. In addition, the handblank data obtained from typists is provocative.

The factor that might be associated with the fairly high barium values in handblanks of typists, is that they handle relatively large amounts of bond paper. It is quite possible that the calcium-containing filler compounds used in the manufacture of bond paper could have barium impurity. This might explain the fact that three out of four of the typists listed in Table 12 had ≥0.5 μg (the mean value of all handblank barium values).

The high antimony value of 0.232 μg (the mean value of all handblank antimony was 0.03 μg) obtained in the handblank of one typist could have come from handling masking tape, type metal, or some other material having appreciable antimony content.

The results obtained from typists show that an individual cannot be classified as belonging to the Class A handblank group unless it is determined that routine exposure to materials having barium or antimony is truly absent.

A desirable outcome of these studies would have been the finding of a clear demarcation between handblank and firing results. However, an appreciable overlap appears instead. The range of 0.0158 μg to 15.8 μg of barium is represented in both sets of data, and the range of 0.01 to 0.63 μg of antimony likewise has contributions from both handblank and firing samples. Thus, based on one element at a time, only if either barium ≥15.8 μg or antimony ≥0.63 μg, is there an essentially 100% probability that the hand sampled fired a gun. If either barium ≤0.0158 or antimony ≤0.01 μg, there is a ~100% probability that the results indicate a handblank.

Again considering the gross barium data, a value of <0.10 μg Ba indicates >80% probability of a handblank, while ≥4.0 μg Ba indicate >90% probability of a firing. In the same fashion, <0.025 μg Sb indicates >80% probability of a handblank, while ≥0.25 μg Sb indicates >90% probability of a firing.

Stated in another way, with respect to barium values alone, 35.1% of handblank values strongly suggest the truth, and 16.6% of firing values define a firing. Antimony values, taken alone and without significant related factors, are such that 60.7% of handblank values strongly indicate their true source, and 51.9% of firing values define a firing. While it may be claimed that antimony by itself provides a definitive result in over half of the tests, the utility of the results are greatly increased when relevant factors are treated simultaneously.

The four essential requirements for reasonably good utilization of the Ba and Sb data in a given shooting case are: (1) proper handblank category assignment of the suspect, (2) knowledge of the caliber of the weapon involved in the shooting, (3) assurance that the test has been performed before the suspect has washed his hands, and (4) consideration of both Ba and Sb values simultaneously. If these factors are known, the results can be interpreted by one of the procedures described in Section 2 and exemplified in Table 3 (where every statistical test gave the correct indication).

It appears, at this time, that the use of NAA for determining whether or not a suspect has fired a weapon will be highly superior to previous physical evidence tests. As the program proceeds, a more definitive judgment can be made as to the extent of its usefulness. Already it is apparent, however, that means of discriminating between the material containing Ba and Sb in handblanks (probably amorphous smeared material) and in gunshot-residue (particulate matter) would be of particular value for testing subjects in high handblank categories and/or small caliber weapon cases.

8.2 OTHER ACTIVITIES

The other major accomplishment during the course of the report period was to establish the requirements for additional data to allow the formulation of desired probability statements in connection with identification work in the areas of gunshot-residues, paint, paper, and bullet lead. The amount and kind of additional data needed was estimated within the context of information gathered during visits to manufacturers, extrapolation from experimental data already gathered and subjected to preliminary statistical analysis, and logical constraints encountered in the occurrence of physical evidence material.

The understanding of paper manufacturing practice obtained by the described visits, for example, has indicated that the interpretation of paper analysis data should consider element concentration variances within a roll, within a ream, within reams produced at substantially different times (intervals of several months) by a given manufacturer, and within the complete data set of all samples from all manufacturers. This understanding will be reflected in a special topical report on the use of NAA for paper comparison and identification.

Examples of constraints associated with physical evidence samples are: (1) extreme deformation of fragmentation of bullets that render caliber identification difficult, (2) combination or alteration of paint samples by

multiple coatings, oxidation, or corrosion (additional paint samples will be taken directly from automobiles, houses, etc., rather than out of fresh cans of paint), and (3) the possibility that a person can acquire a high Ba and/or Sb handblank by handling – but not firing – an uncleaned gun.

It should be mentioned that the acquisition of additional paint samples and their analysis was in process toward the last part of the report period. However, results of this work were not yet computed; they will be included in the next annual report.

In summation, significant progress has been made during the report period toward converting forensic applications of NAA from previously demonstrated feasibility to practicality.

9. SUMMARY

During the report period additional experimental data regarding gunshot-residues has been obtained, and significant progress toward proper interpretation of handlift analysis data has been made. Similarly, progress has been made in the development of a statistical interpretation of data from paint, paper, and bullet lead data, which involve a greater number (>2) of elemental variables. Finally, as a result of consultations with manufacturers and consideration of experimental data already gathered, the amount of additional experiments to provide desired statistics has been defined.

At the end of the period the number of handlifts analyzed included over 200 handblanks and over 400 firing samples. It was estimated that a total of nearly 900 firing samples would provide an adequate statistical basis from which to interpret analytical results from single samples. Similarly, it was estimated that an adequate population representation of paint, paper, and bullet lead would be comprised of 300 paint samples, 250 paper samples, and 180 bullet lead samples. Inasmuch as the number of samples analyzed at this time include 92 paints, 83 papers, and 38 bullet leads, the further analytical requirements are substantial.

Preliminary tests of the discriminant analysis procedure, based on a bivariate normal distribution of barium and antimony, proved to be encouraging in that each of 10 tests of handlift data gave the correct interpretation of the data: i.e., handblanks and firings were correctly identified.

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APPENDIX 1

CRIMINALISTICS QUESTIONNAIRE

A. General

1. Approximately how many evidence samples are you involved with (either directly or indirectly, as a supervisor) each year? _____

2. Would you indicate your number of years experience in criminalistics.
   (a) 5 years or less _____
   (b) 6 - 10 years _____
   (c) 11 - 15 years _____
   (d) 16 - 20 years _____
   (e) more than 20 years _____

3. Of all the analyses with which you have been involved, approximately what percentage were concerned with:
   (a) gunshot residues, to determine whether or not an individual had fired a certain weapon _____%
   (b) bullets, to determine whether or not two bullets or bullet fragments had a common source _____%
      i) In what fraction of the cases included in (3b) did comparison microscopy alone provide sufficient information to form a conclusion (either positive or negative)? _____
      ii) In what fraction of the cases included in (3b) were other methods used successfully to reach a conclusion (either positive or negative)? _____
      iii) In what fraction of the cases included in (3b) were the results inconclusive? _____
   (c) bullets, other
      _____
      _____
      _____ _____%

Comments? _____

_____

_____

(d) paints, to determine if two or more samples had a common source? _____%

i) In what fraction of the cases included in (3d) did microscopy alone provide sufficient information to form a conclusion (either positive or negative)? _____

ii) In what fraction of the cases included in (3d) were other techniques successfully employed to form a conclusion (either positive or negative)? _____

iii) In what fraction of the cases included in (3d) were the results inconclusive? _____

(e) paint, other _____%

_____

_____

_____

Comments? _____

_____

_____

(f) paper, to determine if two or more samples had a common source _____%

i) In what fraction of the cases included in (3f) were the presence or absence of cuts, tears, watermarks, printing or other unique features of the samples which could be discovered by optical techniques (UV, IR, microscope) adequate to form a conclusion (positive or negative)? _____

ii) In what fraction of the cases included in (3f) were other methods successfully employed to form a conclusion (positive or negative)? _____

iii) In what fraction were the results inconclusive? _____

(g) paper, other _____%

Comments? _____

_____

_____

(h) breath, blood or urine analyses for alcohol _____%

(i) blood analyses, not for alcohol _____%

(j) blood examinations, other _____%

(k) glass fragments to establish a common source of two or more samples _____%

(l) hair samples, to establish a common source _____%

(m) cloth, fiber examinations _____%

(n) tool markings (other than firearms) _____%

(o) analyses and examinations of other materials (wood, rubber, plastics, soil, oil, etc) _____%

B. Firearms

1. Of the analyses or examinations involving firearms residues or bullets, with which you have been involved, approximately what percentage were:

(a) automatic pistols _____%

(b) revolvers _____%

(c) rifles _____%

(d) shotguns _____%

(e) other _____%

2. Of the revolver analyses or examinations, what was the approximate percentage of:

(a) .22 caliber _____%

(b) .32 " _____%

(c) .38 " _____%

(d) .44 - .45 caliber _____%

(e) other _____%

3. Similarly, of the automatic pistol analyses or examinations, what was the approximate percentage of:

(a) .22 caliber _____%

(b) .32 " _____%

(c) .38 " _____%

(d) .44 or .45 caliber _____%

(e) other _____%

4. Comments?

C. Paints

1. Approximately what percentage of paint analyses or examinations you conducted involved:
   (a) automobile paint _____%
   (b) house or building paint _____%
   (c) other types _____%

2. Of the automobile and house-type paint samples, what was the approximate distribution of colors? Please enter the percentages in the two tables:

	Automobile	House
black or grey	_____%	_____%
white or off-white	_____%	_____%
blue or green	_____%	_____%
yellow, orange, or red	_____%	_____%
brown or tan	_____%	_____%

3. Comments?

D. Paper

1. Of the analyses, not including handwriting or ink comparisons, in which it was desired to determine whether or not two or more paper samples had a common source, approximately what percentage were:
   (a) white letterhead or bond paper _____%
   (b) white tablet or mimeograph paper _____%
   (c) colored writing paper _____%
   (d) wrapping paper _____%
   (e) newspaper _____%
   (f) other _____%

2. Comments?

APPENDIX 2

OUTLINE OF PROPOSED EXPERIMENTAL PROGRAM

I. Gunshot-Residue and Handblanks

A. Goal

Studies to date have indicated that measuring the amounts of barium and antimony on a person's hands by NAA is a potentially valuable method of determining whether or not an individual has fired a handgun. The primary goal of further research in this area is to develop and refine the application of the method to actual case work, i.e. to attempt to maximize the probability that the results of the NAA determination of Ba and Sb are correctly interpreted in each case.

B. Inferences from existing data

The data, from both handblanks and firings, analyzed thus far show extensive variability. Among the factors contributing to this variability are environmental exposure to Ba and Sb, type, caliber, and condition of the weapon fired, ammunition used, wind velocity, number of firings, sampling technique, and analytical errors.

In an actual case, it would typically be possible to ascertain only two of the above parameters, viz., the caliber, and perhaps the type, of the weapon involved, and the approximate degree of occupational-environmental exposure to Ba and/or Sb of the suspect and/or victim from whom the samples were taken. When the existing handblank and firing data were divided into categories on these bases, it was possible to develop a remarkably accurate model of the variability within each category using a bivariate normal (BVN) distribution. Furthermore, there appear to be definite differences between categories, in the sense that the differences in the means, standard deviations, and correlations cannot be attributed to chance variations.

One possible procedure for interpreting case results, based upon the statistical technique of hypothesis testing, was described in the Monthly Report for July, 1968. The existing data were used to evaluate the expected performance of this procedure by computing the probability (denoted by α) that a handblank sample from a given category would be erroneously interpreted as a specified type of firing and also the probability (denoted by β) that the Ba and Sb levels in a sample from a hand which had recently fired a specified type of weapon would be incorrectly interpreted as a handblank from a given category. It should be mentioned that this procedure is optimal for deciding between the two hypotheses in question, i.e. no other procedure has both a smaller α and a smaller β.

While the procedure appears to be promising for distinguishing Class A handblanks from firings (e.g. α=0.01 and β=0.25 for Class A handblanks versus 0.38 caliber revolver firings), it has a relatively poor expected performance for distinguishing between some other handblank and firing combinations (e.g. α= 0.02 and β= 0.63 for Class B versus 0.22 caliber revolver firings). Consequently, it may be necessary to limit the application of the method to those cases in which it can be established that the individual in question could not have been exposed to materials containing barium or antimony, except by firing a handgun, for some reasonable period of time before his hands were sampled. Additional sampling of individuals in high-exposure environmental categories, as well as additional test firings, should illuminate this issue. An alternative is that the method itself be modified to discriminate between "occupational exposure" and firing residues.

C. Additional data required.

The mathematical validity of the above procedure depends upon the accurate knowledge of the parameters (means, standard deviations, and correlations) of the BVN distributions involved. Therefore, in applying the mathematical model to the physical problem at hand, enough handblanks and test firings should be analyzed, within each category, so that reasonably accurate estimates of these parameters are obtained. Although many criteria have been suggested for judging when an estimate is "reasonably accurate", there is no

generally accepted (among statisticians) method of measuring the distance between two probability distributions, particularly multivariate distributions. The criterion we have investigated is based upon the desire to balance the cost of sampling against the uncertainty in the estimated values of the parameters, combined with the goal of obtaining confidence intervals for the parameters whose expected lengths are short compared with the estimated standard deviations of the populations. The former objective provides a method of allocating the total number of samples to be obtained among the various categories, while the latter permits an assessment of the value of the samples for describing each category.

The relative frequencies of occurrence of various handguns in criminal cases, as estimated from our recent survey of the California Association of Criminalists, were used to assign priorities to the various categories of firings which could be sampled. In considering the question of additional handblank samples, occupational data from the 1960 Census were examined (Statistical Abstract of the United States, 1967, Table No. 330). Approximately 49 percent of the 68 million individuals in the table were classified into one of the three handblank categories which have been studied to date. Of these, 93 percent (30.9 million) were category A (negligible routine exposure to materials containing barium or antimony) on the basis of occupation. Of the various approaches which could be followed to gain further information about the distribution of handblank values of Ba and Sb, the one which appears most promising is to sample individuals from occupations not yet represented in our data (e.g. farm workers, skilled craftsmen, truck drivers, etc.) in order to appropriately classify a larger segment of the employed population and determine, in particular, whether or not the relatively high proportion of category A individuals persists. Approximately 100 additional samples (both hands of 50 individuals) will allow representation of each of the occupational classifications listed in the table cited above. Of course, this procedure will simultaneously improve the current estimates of the parameters of the four exposure categories.

With regard to additional test firings, the minimum goal toward which we should work appears to be the analysis of sufficiently many samples from firings of the more frequently occurring types of handguns to obtain estimates of the

parameters of each, whose overall reliability is comparable to that of the estimated Class A handblank population parameters. This will require approximately the number of samples shown in Table X. Also given in Table X are the frequency of occurrence of each type of weapon, relative to the population of handguns reported in our recent survey, and the cumulative proportion of this population which would be represented if the relevent samples were obtained as listed. The number of samples already analyzed is shown in the second column.

The required sample size for those categories not yet represented in our data were estimated by rather crude interpolation, and hence should be considered quite tentative. Moreover, the entire table is intended merely as a flexible guide to sampling, and it is recommended that the data be analyzed soon after they are collected so that the estimates of the parameters can be periodically updated and the sampling requirements revised accordingly. Similar remarks apply to the handblank samples.

II. Paint, Paper, and Bullet Lead

A. Goal.

The research objectives, which are similar for the three materials, are to utilize the NAA determination of the elemental composition of given samples of a material to help answer two related questions often posed by the criminalist. First, is it possible that the samples have a common origin, i. e. could all of the samples have been part of the same object at some earlier time? If the analyses indicate an affirmitive answer to this question, which will be the case if the elemental compositions of the samples are nearly identical, then a second question becomes relevant. How likely is it that the samples actually do have a common origin? The first question can be satisfactorily resolved in most instances merely by knowing the precision of the analytical method, whereas an answer to the second question, based on elemental composition, requires knowledge of the joint distribution of the elements in the population from which the samples were taken.

Our approach to the second question is indirect, in that we seek an estimate of the probability that the elemental composition of a sample selected at random

Table X

ESTIMATED SAMPLING REQUIREMENTS AND RELATIVE FREQUENCY OF OCCURRENCE OF VARIOUS HANDGUNS

Type of Weapon	Sample Size	Number Analyzed	Additional Required	Cumulative Total	Relative Frequency	Cumulative Proportion
0.22-caliber revolver	123	15	108	108	0.21	0.21
0.36- " "	69	52	17	125	0.19	0.40
0.22- " automatic	82	0	82	207	0.11	0.51
0.45- " "	89	32	57	264	0.09	0.60
0.32- " "	96	0	96	360	0.09	0.69
0.44- " revolver	111	5	106	466	0.06	0.75
0.38- " automatic	96	0	96	562	0.06	0.81
9 mm Luger	110	5	105	667	0.04	0.85
0.25-caliber automatic	83	4	79	746	0.04	0.89
0.32- " revolver	119	0	119	865	0.04	0.93

from the appropriate population would be nearly identical to the composition of a given sample. Clearly, if this probability is small, which indicates that the composition of the given sample is relatively rare within the population, then it is quite likely that the specimens being compared do have a common origin. Conversely, as the probability of matching the composition of the given sample by random selection increases, indicating a more common mixture of elements, the likelihood that the specimens have a common origin becomes smaller. These relationships can be made precise by an appeal to Bayes' theorem.

B. Inferences from existing data.

Each type of material was subdivided into categories according to properties which could be ascertained by optical/physical examination of a sample, or from knowledge of how the sample was obtained. Thus, paints have been divided into five color categories*, papers into six categories, and bullet lead into six caliber categories.

In most instances, the data indicate that within each category the the common logarithm of the elemental concentration is reasonably well approximated by a normal distribution. Hence, the use of a multivariate normal (MVN) model to represent the joint distribution of the elements, within each category, presently appears to be acceptable.

Of the 99 different (i. e. from different manufacturers or batches) paint samples that have been analyzed, no two have elemental compositions which agree to within the experimental error of the NAA method. Similarly, all of the different papers and different bullet leads can be distinguished by NAA of their elemental compositions. Clearly, there is no assurance that all future distinct samples of these materials will have measurable differences in the concentrations of their elemental components. Indeed, one of the goals of the current experimental program is to obtain reliable estimates of the probabilities that specimens which do not have a common origin would be indistinguishable by the NAA method.

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*Previously, the paint data had been further subdivided according to intended application (automobile, house or building, other). Since in many cases it may be difficult to correctly classify a sample as one of these types, current analyses are being restricted to a classification scheme based only on color.

C. Additional data required.

In order to pursue the objectives described above, it is necessary to collect and analyze many more specimens in most of the categories of the three materials that we are considering. Among the intermediate objectives which must be met en route to our ultimate goals are: (1) to accumulate sufficient experimental evidence within the various categories to confirm the validity of the MVN approximation to the joint distribution of elements, or, alternatively to suggest a more suitable model; (2) if the assumption of MVN distributions appears justified, to estimate the parameters of the several distributions (estimates of correlations are required to answer the first question of section II. A. and estimates of the means, standard deviations and correlations are necessary for the second question).

As in the case of gunshot-residues, it is difficult to predict the exact sampling requirements for each category of each material. Tentative recommendations are given in Table Y which were derived from the information in the existing data, and are designed to provide a 95 percent confidence region for the mean of each subpopulation. The region is approximately spherical and has a radius equal to approximately one half of the average standard deviation of the logarithm of the concentration of the elements typically found. As more data become available, more precise confidence statements will be possible, but the general type of conclusion which can be reached on the basis of the proposed sampling plan is that, with probability 0.95, an inequality of the form

$$\sum_{k=1}^{n} (\bar{X}_k - m_k)^2 \leq r^2$$

holds, where $\bar{X}_k$ is the computed sample mean for the k^{th} element, m_k the corresponding true (but unknown) mean for the subpopulation, and r is typically in the range 0.1 to 0.3.

Table Y

ESTIMATED SAMPLING REQUIREMENTS FOR PAINT, PAPER, AND BULLET LEAD

Category	Sample Size	Number Analyzed	Additional Required
Black or Grey Paint	60	12	48
White or Off-White Paint	60	14	46
Blue or Green Paint	60	35	25
Yellow, Orange, or Red Paint	60	21	39
Brown or Tan Paint	60	10	50
Total, Paint	300	92	208
White Letterhead or Bond Paper	60	65	—
White Tablet or Mimeograph Paper	70	6	64
Colored Writing Paper	60	5	55
Wrapping Paper	60	2	58
Newsprint	—	2	—
Magazine Slick Print	—	3	—
Total, Paper	250	83	177
0.45-Caliber Bullets	30	3	27
0.38-Caliber Bullets	30	25	5
0.32-Caliber Bullets	30	0	30
0.30-Caliber Bullets	30	4	26
0.25-Caliber Bullets	30	0	30
0.22-Caliber Bullets	30	6	24
Total, Bullet Lead	180	38	142

APPENDIX 3

REPORT ON THE VISIT TO GULF GENERAL ATOMIC ON APRIL 10, 1969 – C. R. Kingston

A meeting was held with V. P. Guinn, H. L. Schlesinger, and R. P. Hackleman at the offices of Gulf General Atomic on April 10, 1969.

The morning was devoted to a general discussion of the project and some of the visits that were made to various manufacturers.

The afternoon was devoted to discussions mainly about the statistical designs of the gunshot residue project and the paint project. It was agreed upon that the experimental procedures for gunshot residue data collection as outlined in previous monthly reports were adequate and would be followed.

The main part of the discussion centered on the experimental design for collecting data on paint. The meaning of "common origin" was discussed at length in terms of its definition in the paint project. Given that no change in the amounts of detectable elements occurred during or after application of the paint to a surface, it would be reasonable that analyses of batches of paint from different manufacturers would be helpful for origin determinations. There would be certain problems that would have to be considered before data taken from paint obtained from manufacturers would be useful for forensic purposes. To mention just one, the significance for evidence purposes of the situation where one batch may have been used entirely to paint a large industrial building and another batch may have been used to paint a hundred or more small buildings would have to be considered. In other words, the relationship of the batches to objects from which people pick up particles of paint would have to be considered.

Undoubtedly, information on the variability of batches of paint would be useful, but would not be sufficient for forensic purposes. In view of this, it was tentatively agreed that only a limited experiment would be designed to look at the differences between batches and manufacturers.

Of more direct importance for forensic purposes is the distribution of the elements in paint that is actually on objects with which people come into contact. By sampling the objects directly, any factors that operate to change

the properties of the paint (such as contamination from the area where the object is) would automatically be accounted for. This approach also looks at the individual object as the unit of common origin, which is far more appealing from the forensic point of view than the batch or manufacturer being the unit of common origin.

It was therefore tentatively agreed upon that the major sampling plan would be set up so that paint from selected classes of objects would be sampled. The experimental design would be oriented to discover what the statistical behavior of the distribution of elements is over this group of objects. The classes of objects would be selected so as to represent those objects that people are most likely to pick up bits of paint from. In order to examine the possible effect of major geographic location, samples from two cities would be obtained.

This approach is somewhat different from that mentioned in the GGA monthly reports. It is hoped that a specific direction for the paint project has been agreed to by all parties, and that an experimental plan will be developed shortly.

APPENDIX 4

MONTHLY REPORT FOR JULY 1968 – C. R. Kingston

The Gulf General Atomic informal monthly report for July, 1968 is concerned primarily with the problem of determining whether or not a person has fired a firearm by means of the barium and antimony levels on their hand. The basic problem is one of distinguishing between barium and antimony levels that normally occur on a person's hand and those levels that are due to the firing of a firearm.

GA has approached the problem by using standard statistical procedures of hypothesis testing. This appears to be a perfectly acceptable approach that will allow reasonable objective inferences to be made with respect to the question: Did the suspect fire a firearm?

The approach is made more sensitive (presumably) by classifying firearms and persons into categories that could be determined a priori in many actual cases. Weapons are classified by type, and persons by occupational-environmental factors. A further breakdown of these categories is not given in the GA report. This a priori classification is worth further exploration.

Overall, the approach taken by GA in the gunshot residue and handblank problem is very good, at least to the extent that it is reported on in the July report. It is worth mention that the report is written in a clear and straightforward manner, making it easy to follow the authors' presentation.

APPENDIX 5

MONTHLY REPORT FOR AUGUST 1968 – C. R. Kingston

The Gulf General Atomic informal monthly report for August, 1968 is concerned primarily with gunshot-residue and handblank data analysis, and secondarily with paint, paper, and bullet lead.

In the portion concerned with gunshot-residue and handblank data, GA pursues the analysis of past data according to the statistical model suggested in their last report. The analysis appears to be proceeding satisfactorily with respect to the task of estimating the performance of the proposed decision procedure and in making an initial estimate of the value of neutron activation analysis in gunshot-residue problems. However, if GA intends to collect further data in this area, consideration must be given to the relationship of the model to the optimization of resource allocation for the collection of such data.

Some obvious questions are the following:

1. Should data be collected within the occupational-environmental categories specified in the report (i.e., a specified number of persons are sought who fit within each of the categories), or should persons be randomly sampled for handblank data?

2. What proportion of the samples should be of handblanks and what proportion from firings?

3. For samples from firings, what should the allocation be among firearm types?

Presumably, further collection of data will be made with one or more goals in mind. Two such goals might be:

1. To provide a better estimation of the value of NAA in forensic gunshot residue cases.

2. To provide background data that can be used in an actual case to evaluate the NAA results.

GA should state the goals that they have in mind for the use of additional data (if indeed they plan to collect additional data). The sampling plan (or data collection plan) should then be designed so that the data collected will optimally apply to the stated goals.

In the portion concerned with paint, paper, and bullet lead, GA presents no structure or goals for the statistical analysis that they have introduced in the report. As in the gunshot-residue case, we are concerned with two basic models; one for sampling and one for evaluating the value of NAA for the specific materials of concern. The statistical analysis of data collected in the past on paint, paper, and bullet lead is presumably being done for two reasons. One, to make an initial estimate of the value of NAA for source individualization, and two, to form a basis for a sampling plan for additional data collection. It should be determined whether or not the same statistical model will adequately serve both goals. In any event, the best model for each goal should be specified and used.

The sampling plan must of course be oriented toward the final analysis of the data that is desired. In order to know what the final analysis will be (or, in other words, what the specific goals of the data analysis will be), GA must determine, among other things, what type of source individualization they are aiming for. Is the source to which they wish to relate the suspect material a specific object of concern in a case, or a manufacturer, or a batch, and so on? Presumably the analysis of the existing data will suggest an answer to this and similar questions. It would be of interest to the readers of the reports to know what questions GA is asking about the problem, what goals they have in mind, and how they intend to use the existing data to help answer the questions and the existing and future data to reach the goals. Such information is important for proper evaluation of the work reported on by GA.

In summary, there is a basic need for some structure to the experimental work presented in the reports. The goals of any statistical analysis should be stated, and how the analysis takes us to that goal better than alternative analyses should be pointed out. In view of the fact that the structure developed during this project will serve as a guide in other projects of a similar nature for both the sampling procedures and the final statistical analysis, the structure and the reasons for it should be an important component of the project reports.

APPENDIX 6

MONTHLY REPORT FOR SEPTEMBER 1968 -- C. R. Kingston

The Gulf Atomic informal monthly report for September, 1968 is concerned with the computer analyses of paint, paper, and bullet lead data, two methods of determining origin of such material, and further analysis of the gunshot-residue and handblank data.

In the section on paint, paper, and bullet lead, GA has divided the paint and paper samples into groups, and state that these groups can be determined in actual case situations. Clearly the origin of paint can easily be placed into the suggested grouping. If we have a small piece of paint that has an unknown origin, however, it may not be possible to place it in one of the categories with any assurance of being correct. I am referring to the automobile and house paint categories. If a chip of paint appears to belong to the automobile category, how will it be determined that it is indeed an automobile paint rather than a similar paint made for an entirely different application? There must be many such paints on the market and on various objects.

Of course the location of the paint chip or smear and other circumstances may strongly suggest that it came from an automobile (as in a hit and run case) or a house. Are there properties other than those being determined with NAA that can be used to categorize the paint as automobile or house paint, or paint from some other unspecified origin? If there are, I would suggest that GA provide a reference in which the determination and use of such properties are discussed. If there are not, then are we justified in setting up such categories for a statistical analysis in forensic work (where legal rigor as well as statistical rigor must be maintained)?

Perhaps we may be able to derive some probability that a small chip of paint is automobile paint, although at the moment I don't see how. But if we could, what effect would this probability have upon any origin determinations made by NAA property analyses?

I would suggest that GA carefully consider the use of categories of the type presented in their report with questions such as the above in mind.

GA has also suggested that discriminant analysis might be used in origin determinations. In the classical use of the term "origin determination" in forensic science, the origin referred to is the specific object or person from which the evidence came. Thus, for an origin determination as it is generally understood in criminalistics, discriminant analysis would not seem to be the method of choice. However, if we are making a determination as to what manufacturer made a given sample of paint for instance, then discriminant analysis may be appropriate. Perhaps a different term could be applied to the latter type of determination to avoid confusion. It would be helpful if GA specified to what types of origin determination they are relating specific statistical analyses.

The gunshot-residue and handblank studies appear to be progressing satisfactorily with respect to the statistical analysis.

APPENDIX 7

MONTHLY REPORT FOR NOVEMBER 1968 – C. R. Kingston

No review appears to be necessary for GGA's informal report for November, 1968. However, since they have appended the "Outline of Proposed Experimental Program" to the report, I am likewise appending to this report copies of my review comments on that outline which were submitted earlier.

Comments on the "Outline of Proposed Experimental Program" for the GGA project on Applications of Neutron Activation Analysis to Scientific Crime Investigation.

I. Gunshot-Residue and Handblanks

The experimental procedure outlined for this area looks good. The sampling procedures appear to be those that will derive a desirable ratio of information to sample size. I agree with the sampling of individuals from occupations not yet represented in GGA's data. Also the sequential type of approach as outlined makes sense.

II. Paint, Paper, and Bullet Lead

There is essentially no experimental procedure outlined for these three types of materials beyond the statement that more data needs to be collected.

The basic factors that must be considered here in a reasonable sampling plan are relatively simple. First, the definition of "common origin" for each of the materials must be stated. If this cannot be done on the basis of the data already collected, then alternate origin sets must be defined for each type of material and the sampling designed so that the selection of the specific origin sets to be used for each type of material can be determined as soon as possible.

Second, after the selection of what "common origin" is to mean during the project for each type of material, the data collection must be planned so that some optimal balance of inter-and intra-origin property distribution information will be obtained. I consider this the minimum for an acceptable experimental plan.

APPENDIX 8

MONTHLY REPORT FOR JANUARY 1969 – C. R. Kingston

The January informal monthly report is concerned with further data collection on the gunshot-residue problem and the experimental design for the collection of paint samples. The gunshot-residue work appears to be proceeding along the experimental plan quite well. It is interesting to note that no plan can take into account all factors in a research problem, and the unplanned short excursion into the problem of the use of the brush is the sort of thing that must be expected in any research project.

The paint project is still presenting some difficulties from the statistical design point of view. As I interpret the GGA report, they intend to make a general preliminary definition of "common origin" in terms of color and elemental analysis (kinds and amounts of elements present that can be detected and quantitated by NAA). The sampling will be such that the data will allow them to determine what significance the color and elemental analysis have for determining the manufacturer of the paint and/or the batch of the paint.

This suggests that GGA does not feel that there is sufficient information at this time to select a specific definition of common origin, but that the data will be collected in such a manner that it will be possible later to select either the manufacturer or the batch as the unit of common origin. I presume that the specific object that the paint came from has been excluded as a unit of common origin. This is reasonable if contamination after the object has been painted does not influence the elemental composition as determined by NAA.

The sampling unit within the guidelines set above will be a sample of paint from a batch of paint. Variables associated with the selection of batch samples will be the paint color, the manufacturer, and the time of preparation of the batch, the latter being suggested in the subject GGA report. Part of the design of the experimental plan will thus include the number of manufacturers to be sampled, the colors to be sampled, the different batch preparation times

to be sampled, and the number of samples from each batch to be analyzed. GGA has suggested a list of five color groups to be sampled in their outline of the proposed experimental plan. It might be of interest to have the reasons for that selection briefly mentioned in the plan design.

The goals of the experiment will presumably be threefold: 1) to decide whether manufacturer or batch is the preferable "common origin" to be considered in further experimentation, 2) to obtain some idea of how useful a determination of the selected "common origin" would be, and 3) to provide a basis for designing any further experimental plans that may be necessary.

We now need a statistical link between the samples and the goals. A few directions of inquiry into such links were mentioned in GGA's September, 1968 monthly report. One such link should be selected to aid in the design of the experiment, even if the selection is only tentative at this time. Using this statistical link, the allocation of samples with respect to the four variables can be estimated and the experiment designed so that the goals can be realized.

The design can possibly be of the form of a series of "checkerboards", where each "checkerboard" represents a color group. The squares along one direction would represent different manufacturers, and the other direction would represent different batches keyed to time of preparation. Numbers in the boxes would represent the number of samples to be taken from each batch.

The suggested series of analyses of paint from junkyard automobiles could provide information as to the effect of miscellaneous elemental contamination picked up after the car was painted. It would appear, that in order for the analyses to be of full value, it would be necessary to be able to determine the manufacturer and batch of the paint. Perhaps this should be checked into further. Otherwise, the series of analyses would only have value in looking at the internal variation of elemental composition (that is, variation between different samples from the same car). This could make a useful comparison possible between the internal variation of paint directly from the manufacturer and the internal variation of paint from objects that have been in use. With a specific goal in mind, GGA will be able to develop a sampling plan for this series of analyses and state what it is likely to tell them.

APPENDIX 9

MEETINGS ATTENDED AND PAPERS PRESENTED

In addition to several contract discussions with the Division of Isotopes Development (AEC), the San Francisco Operations Office (AEC), and the Law Enforcement Assistance Administration of the Justice Department – in Washington, D.C., Germantown (Pennsylvania), Gaithersburg (Maryland), and San Diego, a number of scientific meetings were attended, several papers presented, several papers published, and numerous lectures given – all related to this forensic activation analysis investigation.* These various activities are outlined below.

A. Meetings Attended. The following relevant scientific meetings were attended:

1. American Nuclear Society National Meeting, Toronto, Canada (June 10-13, 1968). V.P. Guinn and H.R. Lukens participated in the session on "Forensic Activation Analysis", in which V.P. Guinn presented an invited paper on "The Current Status of Forensic Activation Analysis", and H.R. Lukens presented a paper on "Forensic Activation Analysis: Bullet Lead".
2. Annual Seminar of the National Association of Coroners, Anaheim, California (August 20-24, 1968). V.P. Guinn presented an invited paper entitled, "Forensic Activation Analysis as Applied to the Work of the Coroner".
3. Third International Conference on Modern Trends in Activation Analysis, National Bureau of Standards, Gaithersburg, Maryland (October 7-11, 1968). V.P. Guinn served as Honorary Chairman of the Conference, and presented papers on "Neutron Activation

*Although each of these activities involved forensic activation analysis, either entirely or in part, only a very small fraction of the costs of the trips was borne by the contract. The great majority of the costs were borne by Gulf General Atomic, V.P. Guinn (personally), and such organizations as the International Atomic Energy Agency, the Oak Ridge Associated Universities, the American Chemical Society, the Smithsonian Institution, the Argonne National Laboratory, The University of Wisconsin, the National Research Council, the University of Florida, Colorado State University, several European laboratories, etc.

Analysis Identification of the Source of Oil Pollution of Waterways (by V. P. Guinn and S. C. Bellanca), and an invited paper on "The Current Status of Neutron Activation Analysis Applications". H. R. Lukens presented a paper entitled, "The Examination of Actual Case Evidence Samples by Neutron Activation Analysis" (by H. L. Schlesinger, H. R. Lukens, and D. M. Settle).

4. California Association of Criminalists Semiannual Seminar, Lake Tahoe, Nevada (October 17-19, 1968). H. L. Schlesinger presented a paper on "Statistical Treatment of Background Forensic Activation Analysis Data" (by V. P. Guinn, R. P. Hackleman, and H. L. Schlesinger).
5. Seminar on Personal Identification in Mass Disasters, Smithsonian Institution, Washington, D. C. (December 9-11, 1968). V. P. Guinn presented an invited paper entitled, "Forensic Neutron Activation Analysis".
6. American Academy of Forensic Sciences Annual Meeting, Chicago (February 27 - March 1, 1969). H. L. Schlesinger presented a paper on "Statistical Treatment of Gunshot-Residue Data" (by R. P. Hackleman, H. L. Schlesinger, and V. P. Guinn).
7. California Association of Criminalists Semiannual Seminar, Los Angeles (May 22-24, 1969). D. E. Bryan and H. L. Schlesinger participated as panelists at a panel discussion on "The State-of-the-Art of Neutron Activation Analysis Applied to Gunshot Residues".

B. Lectures Given. The following lectures, each entirely or partially concerned with forensic activation analysis, were delivered during the period covered by this report:

1. On June 20, 1968, H. L. Schlesinger lectured on forensic activation analysis in the Gulf General Atomic June 17-21, 1968 Activation Analysis Course.
2. On June 21, 1968, V. P. Guinn presented a paper at the American Chemical Society Annual Analytical Chemistry Summer Symposium, at Pennsylvania State University.

3. On July 19, 1968, V. P. Guinn and H. L. Schlesinger lectured on forensic activation analysis in the University of California at San Diego/Gulf General Atomic/National Science Foundation/AEC July 8-19, 1968, course on Neutron Activation Analysis for college professors.

4. On August 27, 1968, V. P. Guinn lectured on forensic activation analysis in the August 12-30, 1968 Activation Analysis Course at the Oak Ridge Associated Universities.

5. On September 17, 1968, V. P. Guinn lectured in a nuclear applications institute at the University of Wisconsin, in Madison.

6. On September 19, 1968, H. L. Schlesinger lectured on forensic activation analysis in the Gulf General Atomic September 16-20, 1968 Activation Analysis Course.

7. On September 21, 1968, V. P. Guinn lectured before the Reno (Nevada) and Sacramento (California) Sections of the American Chemical Society, at a joint meeting in Reno.

8. On October 2, 1968, V. P. Guinn lectured before the Houston (Texas) Section of the American Chemical Society.

9. On October 4, 1968, V. P. Guinn lectured at the University of Florida, in Gainesville.

10. On October 21, 1968, as the first part of a one-week American Chemical Society speaking tour, V. P. Guinn lectured before the Ridgecrest (China Lake, California) Section of the American Chemical Society.

11. On October 22, 1968, V. P. Guinn lectured before the Fresno (California) Section of the American Chemical Society.

12. On October 23, 1968, V. P. Guinn lectured before the Las Vegas (Nevada) Section of the American Chemical Society.

13. On October 24, 1968, V. P. Guinn lectured before the Tucson (Arizona) Section of the American Chemical Society.

14. On October 25, 1968, V. P. Guinn lectured before the Riverside (California) Section of the American Chemical Society.

15. On November 14, 1968, V. P. Guinn lectured at Cornell University, in Ithaca, New York.

16. On November 15, 1968, V. P. Guinn lectured on forensic activation analysis at Eisenhower College, in Seneca Falls, New York.
17. On November 18, 1968, V. P. Guinn lectured before the Savannah River Section of the American Nuclear Society.
18. On December 28, 1968, V. P. Guinn gave an invited paper at the Annual Meeting of the California State Chemistry Teachers Association, in Fresno.
19. On February 4, 1969, V. P. Guinn gave an invited paper at the 10th Argonne National Laboratory Nuclear Engineering Education Conference, at Argonne.
20. On February 7, 1969, V. P. Guinn lectured at Colorado State University, in Fort Collins.
21. On February 13, 1969, H. L. Schlesinger lectured on forensic activation analysis in the Gulf General Atomic February 10-14, 1969 Activation Analysis Course.
22. On April 10, 1969, V. P. Guinn lectured before the Southern California Section of the Health Physics Society, in Los Angeles.
23. On April 14, 1969, V. P. Guinn lectured at San Diego State College.
24. On April 16, 1969, V. P. Guinn lectured at the University of California at Irvine.
25. On May 6, 1969, as the first part of a May 4-30 European trip, V. P. Guinn lectured at the University of Glasgow (Scotland).
26. On May 8, 1969, V. P. Guinn lectured at the Royal Institute of Technology, in Stockholm (Sweden).
27. On May 12, 1969, V. P. Guinn lectured at the University of Ghent, in Ghent (Belgium).
28. On May 13, 1969, V. P. Guinn lectured at the Euratom laboratory at Petten (The Netherlands).
29. On May 14, 1969, V. P. Guinn lectured at Delft University, in Delft (The Netherlands).
30. On May 16, 1969, V. P. Guinn lectured at the Free University of Amsterdam, in Amsterdam (The Netherlands).

31. On May 23, 1969, V. P. Guinn lectured at the Democritos Center of the Greek Atomic Energy Commission, in Athens (Greece).
32. On May 26, 1969, V. P. Guinn lectured before the National Research Council of Italy, in Milan (Italy).
33. On May 27, 1969, V. P. Guinn lectured in Munich (Germany) at a joint session of the Technische Hochschule and the Gesellschaft für Strahlenforschung.
34. On May 29, 1969, V. P. Guinn lectured in Cologne (Germany) at a joint session of the University of Cologne and the Karlsruhe Nuclear Center.

C. Publications. During this period, the following relevant papers and reports were published:

1. "The Current Status of Forensic Activation Analysis", by V. P. Guinn, Transactions of the American Nuclear Society, 11 (1968) 78-79.
2. "Forensic Activation Analysis: Bullet Lead", by H. R. Lukens, Transactions of the American Nuclear Society, 11 (1968) 80-81.
3. "Forensic Activation Analysis as Applied to the Work of the Coroner", by V. P. Guinn. Gulf General Atomic Report GA-8839 (1969), 20 pages.
4. "Neutron Activation Analysis Identification of the Source of Oil Pollution of Waterways", by V. P. Guinn and S. C. Bellanca, in Modern Trends in Activation Analysis. Volume I, edited by J. R. DeVoe (National Bureau of Standards Special Publication 312; 1969) 93-97.
5. "The Current Status of Neutron Activation Analysis Applications", by V. P. Guinn, in Modern Trends in Activation Analysis. Volume II, edited by J. R. DeVoe (National Bureau of Standards Special Publication 312; 1969), 679-697.

6. "The Examination of Actual Case Evidence Samples by Neutron Activation Analysis", by H. L. Schlesinger, H. R. Lukens, and D. M. Settle, in Modern Trends in Activation Analysis. Volume I, edited by J. R. DeVoe (National Bureau of Standards Special Publication 312; 1969), 265-271.
7. "Forensic Neutron Activation Analysis", by V. P. Guinn. Gulf General Atomic Report GA-9677 (1969), 18 pages. (This paper will appear in the Proceedings of the Smithsonian Institution Seminar on Personal Identification in Mass Disasters).

D. International Atomic Energy Agency Consultants Meeting. The IAEA appointed four persons (R. F. Coleman, V. P. Guinn, R. E. Jervis, and C. J. Maletskos) to serve as Consultants to the Agency in the area of forensic activation analysis. The four Consultants met with IAEA representatives, and a representative from Interpol, in Vienna, May 19-21, 1969. The Consultant's Meeting was chaired by V. P. Guinn. During the meeting, the Consultants conducted an extensive review of the current status of the field of forensic activation analysis, and of the probable directions of its future development. The Consultants made six specific recommendations to the IAEA, pertaining to forensic activation analysis activities in which they felt the Agency might appropriately and productively engage. Although the Agency has not yet formally acted on these recommendations, the Consultants have been advised that they have thus far all been reviewed favorably by the Agency. Three of them are currently being implemented for the Agency by the Consultants (Recommendations 2, 4, and 6 in the IAEA Report on the May 19-21, 1969 Consultant's Meeting).

E. Forensic Activation Analysis Discussions Held. During this report period, forensic activation analysis informal discussions were held with persons in the following groups:

U. S. Treasury Department – Secret Service
U. S. Treasury Department – IRS Alcohol and Tobacco Tax Laboratory
U. S. Coast Guard

Federal Water Pollution Control Administration
Federal Bureau of Investigation
Centre of Forensic Science (Toronto, Canada)
California Association of Criminalists
National Bureau of Standards -- Analytical Chemistry Division
Oregon State University
Washington State University
Industrial Reactor Laboratories
Naval Research Laboratory
Law Enforcement Assistance Administration Research Institute
John Jay College of Criminal Justice

During one week of July, 1968, several crime investigation "workshop" sessions were held at Gulf General Atomic. The participants in these workshop sessions included four consultants to GGA:

Mr. Bradford (Criminalistics Laboratory, Santa Clara County, California)
Prof. Crewe (University of Chicago)
Prof. Garlick (University of Southern California)
Prof. Gomer (University of Chicago).

Members of the GGA Activation Analysis Group who participated in the workshop sessions were D. E. Bryan, V. P. Guinn, H. R. Lukens, H. L. Schlesinger, and D. M. Settle.

At the request of Mr. Martin Pollner, advisor on law enforcement matters to then President-Elect Richard M. Nixon, V. P. Guinn prepared a 10-page report on "The Role of Scientific Crime Investigation". On December 13, 1968, Dr. Guinn met with Mr. Pollner at the Nixon headquarters in New York City, reviewed the report with Mr. Pollner, and presented him with a number of copies of the report.

Separate from this research investigation (but relevant to it), analyses were performed on evidence samples involved in a number of actual criminal cases during this report period. These analyses were performed under Gulf

General Atomic's nonprofit Forensic Activation Analysis Service — available to all law enforcement agencies and defense counsels. In some cases, results were subsequently presented in court by the GGA radiochemists who performed the analyses.

During the contract year, GGA copies of the 28-minute film, "The Nuclear Witness — Activation Analysis in Crime Investigation" (produced for the AEC by General Atomic in 1966), were loaned to a number of different law enforcement agencies, and other groups, who requested copies for showings at meetings or courses.