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APPLICATIONS OF NEUTRON ACTIVATION ANALYSIS IN SCIENTIFIC CRIME INVESTIGATION

Final Report

September 2, 1970

Gulf General Atomic, Incorporated San Diego, California



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APPLICATIONS OF NEUTRON ACTIVATION ANALYSIS IN SCIENTIFIC CRIME INVESTIGATION

FINAL REPORT

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ABSTRACT

This is the final report in an eight-year program to explore the usefulness of activation analysis in the field of criminalistics.

The sampling, activation analysis work, results, and statistical interpretation of results associated with representative population samples of paints, bullet lead, and gunshot residues are described.

It has been found that paint can be highly characterized by neutron activation analysis (NAA). A given batch of paint is highly uniform, but there are significant batch-to-batch differences in the complete array of observed elemental concentrations ("fingerprints"), and different paints have highly different "fingerprints". On the average, the chance that two different paints will be accidentally matched by NAA is considerably less than one in a billion.

A review of earlier NAA data regarding paper samples shows that nearly the same degree of characterization should be possible by NAA with this material as has been achieved with paint.

Bullet lead analysis by instrumental NAA has been shown to be useful, although to a considerably lesser degree than in the case of paint, paper, or gunshot residues. Recommendations to increase the usefulness of bullet lead comparisons by NAA are given.

The probabilities associated with alternative interpretations of the NAA determination of gunshot residue elements in a given case are defined for each combination of a number of specified occupational categories and weapon calibers.

The discussion of each of the foregoing subjects includes a description of related work during previous report periods.

The achievement of the program is shown to be significant. The criminalist may now estimate the probabilities that each of alternative interpretations of NAA results in a given case involving one of the foregoing four materials are true or false, and in most instances such probabilities will highly favor one interpretation. There is no doubt that, with further work, additional materials can be added to the list of well-characterized evidence materials.

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1. INTRODUCTION

The efforts carried out during this year culminated a substantial eight-year program to investigate those areas in which the analytical technique, neutron activation analysis (NAA), might be useful in scientific crime investigation. In past years the investigations were characterized by a wide breadth of approach in that a great multiplicity of evidence type materials were examined with comparative brevity. More recently, a limited number of evidence materials were selected for a more detailed study.

The rationale of the selection of materials and the experimental protocol of the more detailed studies was founded on: (1) the materials' comparative importance, by reason of frequency of occurrence in crime investigations and (2) the need to develop sound statistics to back up conclusions that might be drawn from their examination by NAA.

The NAA method, is characterized by versatility, procedural simplicity, comparative freedom from errors due to matrix effects, great dynamic range, and the capability of nondestructively and simultaneously analyzing a sample for a large number of elements. Because it is a nuclear technique that depends on penetrating radiations, NAA is completely indifferent to chemical form, and it enables the determination of about 75 elements with equal facility in all chemical states and formulations.

The principles and methodology of NAA are explained in Appendix

1. The program has shown NAA to be of significant potential use in the examination of a vast array of evidence materials, and the more recent work has successfully demonstrated that a high degree of confidence can be achieved with respect to the interpretation of NAA results.

The materials addressed in the program include the following: gunshot residues, automobile greases, commercial plastics, rubber, paint, glass, soils, paper, ink, hair, fingernails, wood, tobacco, drugs, river water, oil, whiskey, skin, marijuana, bullets, and primers. The program also included considerations of the possibilities of using nonradioactive tags to facilitate the identification of these materials. Materials chosen for more detailed study were gunshot residues, paint, bullet lead, and paper.

The chief tasks in the more detailed study were the experimental examination, via NAA, of adequate and representative samplings of the population of each material and the statistical analysis of the experimental data. The work was designed to prove a basis for proper interpretation of evidence analysis data within the framework of forensic requirements.

For example, suppose two paint samples of the same color, one found at the scene of a crime and one found in the possession of a suspect, are found to have the identical concentrations (within experimental errors) of the same elements. What is the probability that the two samples have a common source? As a corollary, what is the probability that the samples come from different sources and only accidentally match one another? The answers to these questions depend on the elements and their concentration distribution functions within the appropriate population of paints.

Paint is only one of a number of materials that have a fairly large number of elements, many of which are at the ppm (parts per million) level or less, measurable by NAA. In most instances the elements are found to have concentrations distributed across ranges of two to three orders of magnitude within the relevant population of material. Thus the quantitative value of each observed element is a valuable data point for sample comparison purposes. In forensic context the data points are of interest for identification purposes and are termed 'didentification points'

since the relevant question is usually whether or not a sample associated with a suspect can be identified beyond a reasonable doubt as originating at the scene of a crime.

The gunshot residue study has been somewhat unique, since it was demonstrated early in the program that the efficacy of NAA for determining the presence of such material on a suspect's hand(s) depends on the determination of just two elements, namely, barium and antimony. These elements are constituents of all primers made in the U.S.A., with the exception of one kind from one manufacturer; and when a person discharges a firearm, a fraction of these constituents is deposited on his gun hand. The residues are removed by application and removal of paraffin, and can be readily determined by NAA. The paraffin samples are termed 'handlifts'.

The main question in the interpretation of handlift analysis is whether the levels of barium and antimony found could have been present on the hand of a person who did not fire a gun. To enable this question to be answered, large numbers of handlifts have been obtained and analyzed from: (1) persons of various occupations, who had not fired a gun and (2) persons who had fired a variety of weapons. The analytical data had provided the basis for assigning probabilities that a person had or had not fired a gun from the analysis of his handlifts (right and left hands).

It is of interest that the value of the program has extended beyond the original area of concern. Indeed, not only has NAA been applied in connection with crimes, it has been used in civil litigation. Also, early results of this program showed that NAA had valuable potential for identification of oil pollution sources, and the AEC (DID) is currently supporting the development of this program.

The bulk of the work and results of this program have already been reported. In addition to a series of annual reports, (1-5) a comprehensive report covering all aspects of the first six years of work has been prepared, (6) and special topical reports describing all of the work

performed on paper, ⁽⁷⁾ paint, ⁽⁸⁾ gunshot residues, ⁽⁹⁾ and bullet lead ⁽¹⁰⁾ have been prepared. The aforementioned reports may be procured from the National Technical Information Service, U. S. Department of Commerce, Springfield, Virginia, 22151, should detailed information on procedures and results be desired.

Inasmuch as this is the final report of the program, the more significant results of the earlier work on paint, paper, gunshot residues, and bullet lead will be reiterated here, although space will not permit full presentation of the experimental data or thorough coverage of details. In addition, of course, this report describes the work and results achieved during the last year of effort.

In recent years the term, "fingerprint", has come into common use as a synonym for qualitative/quantitative trace element pattern, although the uniqueness of such a pattern may not have established as well as true fingerprints. Hence, although we use the term here, we retain the quotation marks as a reminder that it is a term of convenience.

2. THE EXAMINATION OF PAINT EVIDENCE SAMPLES

2.1 INTRODUCTION

Paint is one of the materials studied in two distinct phases: (1) an earlier investigatory study, and (2) a population study to obtain trace element conecentration distribution functions. The second phase followed naturally from the very encouraging results derived from the initial work, and was completed during the present report period.

The importance of paint as an evidence material was brought to our attention in early discussions with expert criminalists, such as R. Pinker (then of the Los Angeles police department) and the late P. Kirk (founder of the School of Criminalistics at the University of California). From the first investigations it appeared that NAA would be useful in nondestructive paint characterization; (1) indeed, the first actual case application of NAA involved the comparison of paint evidence samples obtained in the investigation of a burglery. (11)

2.2 EXPERIMENTAL PROCEDURE

In all experiments the nondestructive, purely-instrumental NAA technique was used. Samples were weighed and sealed in polyethylene vials, irradiated in the TRIGA Mark I nuclear reactor at a thermal neutron flux of ~ 2 x 10¹² n/cm²-sec together with appropriate comparator standards, and finally measured by multichannel gamma-ray spectrometry. In nearly every experiment the sample material was subject to two irradiation-count cycles, one a short prompt cycle of ~1 min each per step and the other involving longer (30-60 minute) irradiations and counting at longer decay times for longer periods.

In the earlier work, embracing 80 paint samples, the gamma-ray spectrometer was equipped with a 3-inch by 3-inch NaI(T1) detector, while a 35 cc Ge(Li) detector coupled to a 4096 channel spectrometer was used in the final study involving 155 paint samples. The productivity of the latter tool was significantly greater than the former. The 80 control samples of the earlier work were characterized by an average of 7.7 elements per sample, while the final 155 samples were characterized by an average of 14.6 elements per sample.

Attention was first directed toward samples taken directly from cans of paint, in order to ascertain (1) the intra-can sample reproducibility, (2) the intra-batch variability between one can of paint and another, and (3) the variability between different batches of a given kind of paint from a given manufacturer, and (4) the differences between different kinds of paint (different by reason of color and/or manufacturer). The latter paint was also examined by the analysis of 32 samples prepared by painting samples onto glass plates, allowing them to dry thoroughly, and then analyzing weighed portions by NAA.

On the basis of the early work, and in consultation with C. R. Kingston (Professor at the John Jay College of Criminal Justice and consultant to the Law Enforcement Assistance Administration), it was decided that the population of environmental paints should be examined. To this end 10 persons of different, active occupations were asked to record the paints they encountered during a normal 24-hour period. The records were used to construct a sampling model of the numbers of paints in five color classes to be taken from various objects.

The objects and frequencies with which they were encountered were as follows:

Object	Frequency (%)
Motor Driven Vehicles - Exteriors	12.7
Building Exteriors	12.2
Walls - Interior, including doors and windows	19.1
Furniture, Home and Commercial	28.1
Ships and Boats	0.9
Appliances and Tools	15.2
Vehicles - not motor driven	1.9
Fences and Gates	1.8
Outdoor Objects - Utility Poles, Railings, etc.	5. 7
Containers - Trash Cans, Drums, etc.	1.4

The color classes and frequencies of encounter were as follows:

Color Class	Color	Frequency (%)
1	Black or Gray	14.9
2	White or Off-White, including silver	25.6
3	Blue, Violet, or Green	22.2
4	Yellow, Orange, or Red	21.1
5	Brown or Tan	16.1
	_	

The observed frequencies of specific combinations of colors and objects were reasonably well adhered to in acquiring 155 paint samples. In numerous instances paint samples were acquired in replicate from adjacent portions of a given surface. Some "replicates" were acquired of ostensibly the same paint, but from different panels of a given object: e.g., the hood and fender of a mono-colored car, the door and side of a refrigerator, and so forth. Each sample was water washed, dried, weighed, and subjected to NAA.

2.3 RESULTS, EARLIER WORK

Based on variances of four elements determined in four samples of red automobile paint from a given can (relative standard deviations were Na - 1.58%, Mn - 4.25%, Al - 4.16%, Br - 4.64%) it appeared that in 90% of the cases the results would not deviate more than 8%, relative, from the average values. Information derived from replicate analyses of eight different batches of Sinclair Paint No. 1929 was more revealing. The intra-batch consistencies were such that the 90% confidence levels were associated with the following deviations:

Element	Relative Deviation (%)	Element	Relative Deviation (%)	Element	Relative Deviation (%)
Ti	7	Na	7	In	26
Al	5	Co	13	Lâ	21
Mn	4	Zn	10	Sb	18

The inter-batch consistencies were such that the 90% confidence levels were associated with the following deviations:

	Relative		Relative	Relative		
Element	Deviation (%)	Element	Deviation (%)	Element	Deviation (%)	
Ti	3	Na	37	In	77	
A1	23	Co	45	Lz	32	
Mn	5	Zn	5	Sb	28	

The above results indicate that Al, Na, Co, In, La, and Sb may be used to distinguish between different batches of a given paint, but that Ti, Mn, and Zn may not be useful to this end.

The foregoing results proved to be very helpful in assigning concentration range codes for ready comparison of multiplicity of analytical data. For example, in the analysis of 32 different paints, the five most

frequently observed elements were assigned the following concentration codes:

Code	Ti (thousands of ppm)	Mn (ppm)	Al (thousands of ppm)	Na (ppm)	Co (ppm)
Α	<1	1-2.1	0.7-1	<31	100-150
В	1-40	2. 1-4. 6	1-1.5	31-47	150-230
C	40-80	4.6-10	1. 5-2. 3	47-70	230-310
D	80-120	10-21	2.3-4.7	70-100	310-470
E	120-160	21-46	4.7-7.0	100-150	470-700
F	160-200	46-100	7.0-10	150-230	700-1000
G	200-240	100-210	>10	230-310	1000-1500
Н	>240	210-400		310-470	>1500
I		>400		>470	

The above codes do not describe ranges that are constant multiple of the intra-batch variability for each element because the various elements have different overall concentration ranges. Also, titanium appeared to be a special case because of its apparent position as the most common additive (as TiO₂) to modern paints.

The five most frequently observed elements are given in accordance with the foregoing code in Table 1. It may be observed that only Samples no. 29 and 30 have the same code pattern. However, these two paints were easily distinguished by gross differences in zinc (1420 ppm vs. 2040 ppm); also, these two samples were of different color (Sea Mist and Sunny Yellow). Thus, all of these different paints were free of accidental mismatch.

It was of particular interest that the Pearson Product-Moment Correlation coefficients for the various paths of the most frequently observed elements were consistent with the hypothesis that no correlations existed. The test for correlation assumes a Gaussian-type concentration frequency distribution for the elements, and this point could not be adequately tested with the limited sample size. Nevertheless, at

the time it appeared that such a distribution would be proven with more data; and the lack of correlations was an additional encouraging aspect of the results, since it indicated that comparatively simple statistical procedures would serve for data interpretation.

It should be noted that no obviously correlatable element pairs occurred in the predominant analytical results. Thus oxygen, constituent of many pigment compounds, is not addressed.

Table 1
LETTER CODES FOR 32 DIFFERENT PAINTS

		E	leme	nt				E	leme	ent	
Sample No.	Ti	Mn	Al	Na	Со	Sample No.	Ti	Mn	Al	Na	Со
13	Α	E	G	Н	D	21	D	E	D	Н	G
6	A	G	Α	F	E	29	D	G	В	E	A
10	В	С	E	С	D	30	D	G	В	E	A
14	В	E	В	С	E	27	D	G	С	E	Α
32	В	F	Α	E	F	19	E	В	D	С	В
4	В	F	В	E	\mathbf{F}	2	E	В	D	E	E
. 11	С	D	С	В	F	22	E	В	D	G	E
9	С	D	С	F	E	12	E	В	E	D	C .
26	С	D	D	I	D	23	E	D	D	F	G
28	С	G	Α	E	A	25	E	H	D	F	F
24	D	A	D	F	G	5	F	В	D	F	D
18	D	В	D	Α	В	7	G	Α	E	F	D
31	D	В	D	С	D	16	G	В	E	С	D
17	D	В	D	Н	В	l	G	В	E	E	D
20	D	В	D	I	В	3	G	В	E	F	E
15	D	D	D	С	С	8	G	С	E	E	D

2.4 RESULTS, CURRENT REPORT PERIOD

The results of the NAA examination of the 155 environmental paints (given in the Appendix) were codified in the same fashion as previously described, except that a uniform code for all elements (except titanium) was used. The code is described in Table 2.

Table 2

CONCENTRATION CODE FOR COMPARISON OF 155 PAINT SAMPLES.

CONCENTRATIONS IN PPM

	Concentrat	Concentration Range						
Code	All Elements Except Ti	<u>Ti</u>						
Α	<0.21	4,000-24,000						
В	0.21-0.46	24,000-44,000						
С	0.46-1.0	44,000-64,000						
D	1.0-2.1	64,000-84,000						
E	2. 1-4. 6	84,000-104,000						
F	4.6-10	104,000-124,000						
G	10-21	124,000-144,000						
H	21-46	144, 000-164, 000						
I	46-100	164,000-184,000						
J	100-210	184,000-204,000						
K	210-460	204, 000-224, 000						
L	460-1,000	224,000-244,000						
M	1,000-2,100	244,000-264,000						
N	2, 100-4, 600	264,000-284,000						
0	4,600-10,000	284, 000-304, 000						
P	10,000-21,000	304,000-324,000						
Q	21,000-46,000	324,000-344,000						
R	46,000-100,000	344,000-364,000						
S	100, 000-210, 000	364,000-384,000						
T	210,000-460,000	384,000-404,000						

The coded results are given in Table 3. Actually 155 samples are included in Table 3; two samples taken from the same car, one from a fender and one from the hood (samples 24 and 23, respectively) were included as a test of the method. It will be noted that these are the only two samples that match in all respects, and, just as important, they do match (within the precision of the code) in all respects.

2. 5 DISCUSSION

The method of data presentation exemplified in Tables 1 and 3 is convenient from the standpoint of data interpretation as well as direct simple comparison. One of the first tasks of interpretation was to ascertain whether or not the elemental concentrations had a Gaussian distribution. It was apparent that they were not normally distributed, but it was expected that they would be log-normally distributed. Accordingly, the distributions were analyzed by the chi-square test for this property, and the results are given in Table 4 for the 15 most frequently observed elements. The hypothesis that the logarithms of the concentration values of the elements were normally distributed was rejected by the test for every element except tantalum.

The possibility of transforming the concentration values into T-scores to effect a normalization was rejected for lack of justification. Several attempts to find a suitable alternative standard frequency distribution function were nonproductive. Constraints of time and finding precluded an exhaustive search for a suitable distribution function, however; so it was decided to use the observed distributions as the basis for statistical statements. This procedure was partially supported by the lack of apparent element correlations among the elements in earlier work and the lack of frequent and systematic apparent element correlations in the 155 sample data set. Pragmatic tests supplied further support to the procedure.

Table 3
CODED RESULTS, ENVIRONMENTAL PAINTS

Class I Paints (17 Samples)

Element Sample Mn <u>A1</u> $\underline{\mathtt{Br}}$ Na Cl Ζn <u>Ti</u> Sb Ba K Co CrTa Сa $\underline{\underline{\mathbf{W}}}$ 41 В E K G M Р D H Η K J R F 192 E Ι Ι Ι K 131 \mathbf{E} J Η G K 143 G L Η G L K Α E Η J J 158 N G L G K P M Ι 157 Η L L ·H L В J M E M N F 147 Η Ν L Ι F 0 L Н F 148 Η Q L K ·L 0 Ι Ι K M 84 I Ι K I S K D K 65 Ι N J. N R С Ι N R J J Q F M 164 Ι 0 K G K М Н Η M Ν S I 63 Ι Р Ν F K J K Ν T 160 0 J F M L M G K M 0 Ι E 38 K L G L P D L J N L S F 159 K О G Ν Μ Р F M J N M Р Н 127 L Q N :I Q L Α F M L L Q 161 R Ή L M M M L Ι P Ι

Table 3 (Continued)

Class II Paints (63 Samples)

•							Elem	ent							
<u>Sample</u>	Mn	<u>A1</u>	Na	Br	<u>C1</u>	<u>Zn</u>	Ti	Sb	Ba	K	Co	Cr	Ta	Ca	w
98	С	N	J	Н	I		Q	G		J			Н		F
93	D	0	L	Н	L	I	Q		F						
35	E	L	L		M	R	Α	Q	S					Q	
95	E	N	I	. I	M	I	R	F							E
97	\mathbf{E}	0	K	I	L	I	Q	G	F	J			H		F
89	E	0	K	J	M	J	K	D	G				E		
182	E	0	L	Н	J	K	0	H		J	I	J	\mathbf{F}		E
73	F	M	K	н	L	L	H	E	K	J			F		
92	F	N	J	H	I	I	R	F	H		G		F		
91	F	N	J	H	\mathbf{L}_{+}	J	R	G	Н				F		E
96	F	N	J	H	L	K	Q	F							
87	F	N	K	Н	L	Ι	L	G		J			H		F
94	F	0	I	Н	L	I	P	F							
74	F	0	K	H	·K	J	J	D		I					
79	F	. O	L	\mathbf{E}	J		M								
183	G	L	K	\mathbf{F}	M	R	F	E	S	J			G	R	F
99	Ğ	N	L	J	L	K	P		E	K	H		H	_	G
155	G	N	M	H	M	P	H	I	I	M	Ţ		F	T	
188	G	0	K		R		K	R	_	_	_			R.	
187	G	0	L	G	L	M	M		L	L	J				
190	G	P	K	I	L	K	Р	_	_	_	K		~		_
129	Н	N	I	Н	L	P	I	I	L	J	J		G	S	E
100	H	N	L	G	L	M	0	Н	G	K	Н		H	a	H
179	H	N	L	H	M	N	Ι	I	M.	_	I			S	
40	H	Ó	J	\mathbf{F}	L	L	M	E		J	K			R	_
167	H	0	K	G	P	L	J	Н	H	I	J -		H	s	E
166	Н	0	K	G	P	M	J	G	H	L	I		G	S	E
173	H	Ο.	M	G	L	Q	M		M	M	J			_	
105	Н	0	M	Н	M	N	M	Н	M	K	J		_	R	
54	H	P	M	H	L	K	L	Ι	I	N	H		Ι	R	Н

Table 3 (Continued)

Class II Paints (63 Samples) (Continued)

							Elen	nent							
Sample	Mn	Al	<u>Na</u>	Br	<u>C1</u>	Zn	Ti	Sb	Ba	K	<u>Co</u>	Cr	Ta	<u>Ca</u>	<u>w</u>
152	Н	Q	M	H	L		Н	\mathbf{E}	I	N	Н			S	
154	H	Q	M	Н	L	L	Н	E	I	M				S	
153	H	Q	M	I	M		Н	\mathbf{F}	I	N	Н	J		S	
141	I	N	J	F	K	M	E	I		J	J	_		s	
124	I	N	K	Н	N	N	J	I	G	K	J		Н	S	F
168	I	O	J	G	M	0	M	H	H		I		G	R	F
186	I	Ο	M	\mathbf{F}		Q	H	J	R	K	J			Q	
175	I	0	N	Ι	0	0	P	K	K	K	J		Н		F
142	I	P	L	G	K	K	Н	L	0	L	J	N	H	S	
184	J	N	K	I	N	Q	M	K	N	K	J		G	P	F
27	J	N	L	Ι	I	R	J	E		K	K				
170	J	O	J	H	M	0	N	H			J	K	G	R	F
128	J	0	· L	H	L	J	M	Ι		L	I		H	R	F
25	J	P	M	K	N	0	0	I	I	K	J				
82	J	Q	L	G	K	M	H	H	J	L	I		H		F
32	J	T	M	H	J	M		E		L	L				
101	K	J	L	J	L	0	P	\mathbf{F}	0	I	G				
104	K	N	K	H	I	K	И		J	I			E		
103	K	N	K	Н	K	L	N	0	J	J	G				
102	K	N	K	· I	K	Q	M	G	Q	Ι	G				
90	K	N	K	J	L	\mathbf{L}	K	H	I	K			F		
88	K	N	L	J	M	N	K		N	J			F		
71	K	N	N	K	Q	M	L	H	0	L			F		
78	K	0	L	I		N	J	I	N	L	J			P	
171	K	P			P	0	K		K				G	R	
172	K	P	L	\mathbf{F}		N	J	M	I	L			G	R	F
55	L	N	J	G	K	L	J	G	G		J			R	
30	L	N	L	F	H	R	I	D			L				
72	L	N	L	H		K	J	Ι		J			F G		\mathbf{E}
86	L	N	L	J	M	L	I	\mathbf{E}	N	K			G		
3 1	L	S	N	H	J	P			H	L	L				

Class III Paints (34 Samples)

						0140	3 111 1	Eleme	ija sai. ent	iipica					
Sample	Mn	Al	Na	Br	<u>C1</u>	Zn	Ti	Sb	Ba	K	Co	Cr	Ta	Ca	w
					_					_		_		_	_
191	E	Q	I	G	H					H					
117	E	Q	I	I	H	J		D	H	J					
83	F	N	J	I	N	K	H	F	L			L			
1	F	0	M	Ġ		M	L	G	I	K	K			R	
21	F	Q	I	I	M	I			J						
145	F	R	J	J	·M				H	K					
56	G	0	K	H	M	L	M	F	J	I	K	L	H		F
165	G	0	L	G	M	K	Н	Н	L	L	J		F	S	
174	G	0	L	G	N	L	K	G	H	K	K	K			
53	G	0	L	H	M	M	M	G	K	Н	K	L	H	N	F
144	G	Q	L	H	L	Q	Α		H	J	J				
177	G	Q	L	H	0	L.			K	I	K				
36	H	M	J	G	M	R	С	L	S		K	K		R	E
75	H	M	L	J	N	P	G	I	H	K	G	K	G		
211	H	N	M	H	M		F	\mathbf{E}	I	L	I	K		R	
23	H	P	Н	G	M				L						
24	H	P	H	G	M				L						
13	Н	P	K	C-	N	K		\mathbf{E}							
195	H	Q	H	H	N	H	Α		L	H					
81	I	M	M	I	О	Q	G	Ι	K	L	K	M			\mathbf{E}
39	I	N	K	H	R	0	Ι	Ι	N		K	K		Q	E
80	I	N	L	L	0	P	С	K	0	L	K	S			
70	I	0	N	G	M		Α		J	N		S			
162	J	N	M	G	M	L	С	С			M	R	H	Q	F
115	J	Q	J	I	0	J			N		M				
156	J	R	K	H	N	0	В	G	M	L	I	K	•	Q	
180	K	N	K	H	L	0	K			I					
119	Ķ	0	J	1	P	J			L	J					\mathbf{E}
120	K	P	J	I	M			G	L						F
121	K	Q	L	I	M										
67	L	M	J	H	0	L	С	E	J	I	L	L	E		E
201	L	P	K	J	N					Н	M				
19	L	P	L	N	0	K					L				
9	M	Q	K	Н	M	J					L				

				!				Eleme	ent						
Sample	Mn	<u>A1</u>	Na	Br	<u>C1</u>	Zn	Ti	Sb	Ba	K	Co	Cr	Ta	Ca	W
69	D	N	L	Н	L	N	Н	I	I	J	K		G		F
28	\mathbf{E}	N	I	G	K	N	L	F	Ē	J	11	J	G		r F
163	E	0	K	· H	L			_	_	J	M	S	G		r
17	\mathbf{F}	L	K	H	K	I		L	R	J	141	P	G		
43	\mathbf{F}	M	J	J	L	L		M	N	Ū		R			
11	F	M	K	J	0	K		N	0			R			
189	\mathbf{F}	N	I	I	K			K				P			
193	F	P	J	H	K				Н			_	${f E}$		
77	G	L	K	I	0			L	0	K	M		-		
181	G	N	M	I	M	M	Α	H	N	K	K	Q		0	
194	G	0	J	J	L	M	M		K		H	~	G	•	\mathbf{E}
125	H	N	J	I	L	M	J	I	H	K	J	N	Ğ	s	F
126	H	N	L	G	M	M	H	Н	H	K	Н	L		Š	•
176	Ī	M	L	J	M	L	H	K	J	J		M	F	J	E
51	I	N	L	I	N	S	C	H	I	L	I	N	_	Q	
42	Ι	0	L	J	L	M	M	H	K	K	K			R	
. 62	I	Q	K	G	L	L	С	\mathbf{F}	I	M		M		-\	
34	J	0	L	H	L	Q	С		L	K	K	Q			
185	K	M	J	J	N	L				L	K			0	E
130	L	Q	L	H	M	L			M	K	K	K		Ū	
26	M	N	M	. J	M		\mathbf{F}	M	L		K	Q			
107	N	N	J	I	N			L	L			Q			

								Eleme	nt			·			
Sample	Mn	<u>A1</u>	Na	Br	<u>Cs</u>	Zn	Ti	Sb	Ba	K	<u>Co</u>	Cr	Ta	Ca	w
68	E	Ŀ	M	J	N	N	E	I		K	J				
37	F	L	J		L	R	Α	Q	S	K				Q	
58	H	N	L	E	L	R	L	I	0	K	J	0	H	R	F
-52	H	N	M	H	M	R	K	I	0	L	K	0	H	R	F
123	H	0	M	I	M	0	I	\mathbf{F}	L	L	K	J	H	P	\mathbf{F}
151	H	Q	M	G	L		H	E	I	N	H			S	
146	H	R	J	I	L	M		\mathbf{F}	M	L		J			
85	J	M	L	I	N		H	F	K	K		J			
60	J	Q	M	G		Q	D	H	0	L	K				
169	J	R	J	F	N	0	I			K		I	G	R	
57	J	T	K	H	K	M	\mathbf{F}	H	I	L		L			${f E}$
149	K	M	K	H	N	K	В		K	L	K	I		S	
76	K	N	K	H	L	K	С		N	J			F		E
66	K	Q	N	G	L	0	С	G	N	L	I	I		S	
178	L	N	L	I	L	P	K	H	J	J	I	K	\mathbf{F}	S	\mathbf{E}
150	L	0	N	I	M	N	I	F	0	M	J	M	H	P	
196	L	R	J	I	J		Α		N						
33	M	0	L	H	H	N	I	F		K	K	0			
64	N	N	N	I		R		G	0	N	J	N		Q	

Table 4
FREQUENCY DISTRIBUTION OF 15 MOST OFTEN OBSERVED ELEMENTS

								С	once	ntrat	ion (Code	a									_ a
Element	<u>A</u>	<u>B</u>	<u>c</u>	· <u>D</u>	E	F	g	<u>H</u>	1	<u>J</u>	K	<u>L</u>	<u>M</u>	N	<u>o</u>	P	Q	R	<u>s</u>	T	No. of Values	Log Normal
Mn		1	1	3	13	18	17	31	19	16	19	14	2	2				-			156	No
Al									2	2		9	10	51	37	14	21	6	2	2	156	No
Na								5	8	27	34	44	25	11							154	No
Br					2	8	33	49	37	17	3	1		ì							151	Nob
Cl								4	4	5	17	42	40	20	9	4	2	2	1		150	No
Ti										1		2	3	2	6	1	5	14	51	43	1 28	No
Zn									2	8	20	18	22	12	14	8	8	10	1		123	No
Sb				11	15	19	14	20	21	3	5	6	2	1			2	1			120	Νο
Ba							2	8	19	11	12	13	8	12	13		l	3	4		106	No
K									4	22	29	24	8	9							96	No
Co							5	8	13	24	2.5	6	10	1							92	No
Cr						1			4	9	10	8	6	5	4	5	4	3	4		62	No
Ta					4	15	15	20	6												60	Yes
Ca															2	4	11	18	18	3	56	No
W					19	26	2	2	1												50	No

a As determined by chi-square test for log-normal distribution.

N range observation deleted as having pigment source. Chisquare test also fails with this observation included.

The fractional occurrences of various observed concentrations were tabulated, as illustrated for the five most frequently observed elements in Table 5. The fractional occurrences were treated as probability estimates for the occurrence of respective concentrations in pragmatic tests of observed concentration codes. The tests consisted of multiplying individual probabilities for element concentrations for each of several elements to obtain the predicted probability of observing particular codes for those combinations of elements; then the hypothesis that the observed codes not significantly differ in frequency from the predictions was subjected to the chi-square test (which is independent of distribution function). In no case was the hypothesis rejected. Examples of the tests are given in Table 6.

Table 5

FREQUENCY DISTRIBUTIONS,

THE FIVE MOST FREQUENTLY OBSERVED ELEMENTS

Code	$\underline{\mathbf{Mn}}$	Al	Na	Br	<u>C1</u>
Α					
В	0.0064				•
С	0.0064				
D	0.019				
E	0.083			0.013	
F	0.115			0.056	
G	0. 109			0.211	
Н	0.199		0.032	0.314	0.0256
I	0.122	0.013	0.051	0.237	0.0256
J	0.103	0.013	0.173	0.109	0.032
K	0.122	0.000	0.197	0.019	0.109
L	0.090	0.058	0.273	0.0064	0.269
M	0.013	0.064	0.160	0.0064	0. 256
N	0.013	0.327	0.070	•	0.128
0		0. 237			0.058
P		0.090			0.0256
Q		0.135			0.013
R		0.039			0.013
Ś		0.013			0.0064
T		0.013			

Table 6
FREQUENCIES OF CODE COMBINATIONS IN 155 SAMPLES,
PREDICTED VS OBSERVED

											4 4	. *	
Code for Mn/Al:	H/I	H/J	H/K	H/L	H/M	H/N	H/O	H/P	H/Q	H/R	H/S	H/T	
f, predicted	0.4	0.4	0	1.8	2. 0	10 ′	7. 3	2.8	4. 2	1. 2	0.4		
f, observed	1	0	0.	1	2	8	. 6	4	7	1	. 0	0	
chi-square = 4.	55. Insi	gnificant	differen	ce betwe	en f (pre	i.) and f	(obs.).		•				
Code for Al/Na:	N/H	N/I	N/J	N/K	N/L	N/M	N/N				-		
f, predicted	1.6	2.6	8.6	10.0	13. 9	8. 1	3.6						
f, observed	Ö	2	10	10	14	6	3						
chi-square = .2.	62. Insi	gnificant	differen	ice betwe	en f (pre	d.) and f	(obs.).						
Code for Na/Br:	L/E	L/F	L/G	L/H	L/I	L/J	L/K	L/L	L/N				
f, predicted	0.6	2.4	8. 9	10.1	10.0	4.6	0.8	0.3	0.3				•
f, observed	2	2	8	14	7	7	1	1	1			•	
chi-square = 7.	.53. Insi	gnificant	differer	nce betwe	en f (pre	d.) and f	(obs.).					•	
Code for Al/Mn:	N/B	N/C	N/D	N/E	N/F	N/G	N/H	N/I	N/J	N/K	N/L	N/M-1	N/N
f, predicted	0.33	0.33	0.96	4.2	5.9	5.6	10	6. 2	5.3	6.2	4.6	0. 7	0.7
f, observed	0	1	1	2	6.	4	9	6	3	8	5	1	2
chi-square = 7.	. 77. Insi	gnificant	t differen	ice betwe	en f (pre	d.) and f	(obs.).			•			
Code for Mn/Al/Na	: H/I/L	H/J/L	H/K/L	H/L/L	H/M/L	H/N/L	H/O/L	H/P/L	H/Q/L	H/R/L	H/S/L	H/T/L	
f, predicted	0.11	0.11		0.49	0.54	2.80	2.00	0.78	1.15	0.33	0. 11	0. 11	
f, observed	0	0	. 0	1	1	5	0	0	1	0	0	0	
chi-square = 6.	. 22. Insi	gnifican		nce betwe	en f (pre	d.) and f	(obs.).						
									,				

These results indicate that the observed individual probabilities can be treated in single combinations to compute the probabilities of accidental matches of multiple letter codes. For example, the probability for Mn = H, Al = N, Na = L, Br = H, Cl = L occurring together is simply (0.199) (0.327) (0.273) (0.314) (0.269) or 0.00149. Thus one would expect the HNLHL code to occur thrice in 2000 random paint samples. This is the most probable combination for the five most frequently observed elements and it was not found among the 155 samples listed in Table 3. Naturally, the guarantee against an accidental match of results increases with the number of identification points used.

Let us examine the probabilities of finding a particular code designation or "fingerprint" of a paint sample as a function of identification points, assuming average occurrence probabilities. The average occurrence probabilities are computed as the reciprocal of the number of intervals (letters) used to describe the over-all observed range of concentrations for each element. The average probabilities are as follows for 10 of the most frequently observed elements.

Mn - 0.077	C1 - 0.0825	Ba - 0.077
A1 -0.091	Zn - 0.0825	K - 0.167
Na - 0.143	Sb - 0.072	Co - 0.125
Br - 0.111		

Then, on the average, the probability of a given fingerprint as a function of numbers of elements is as follows:

Mn -	7.7×10^{-2}
Mn, Al -	7.0×10^{-3}
Mn, Al, Na -	1.0×10^{-3}
Mn, Al, Na, Br -	1.1×10^{-4}
Mn, Al, Na, Br, Cl -	9. 1 x 10 ⁻⁶
Mn, Al, Na, Br, Cl, Zn -	7.5×10^{-7}

Mn, Al, Na, Br, Cl, Zn, Sb - 5.4×10^{-8} Mn, Al, Na, Br, Cl, Zn, Sb, Ba - 4.2×10^{-9} Mn, Al, Na, Br, Cl, Zn, Sb, Ba, K - 7.0×10^{-10} Mn, Al, Na, Br, Cl, Zn, Sb, Ba, K, Co - 8.8×10^{-11}

The foregoing treatment is not optimal. The uniform concentration intervals are unduly broad in the context of analytical precisions and intrabatch uniformities with respect to most of the elements. This deficiency is somewhat ameliorated by comparative little confusion of range assignments. As range intervals are narrowed there is an increasing likelihood of some uncertainty of assignment due to overlap of intra-batch variation parameter with an adjacent range. In any event, the actual analytical data must be referred to whenever two paints have highly similar or identical "fingerprint" codes, in order to arrive at a final judgement as to the "identity" of the samples.

Also, in this brief treatment the failure to obtain a useful concentration value for an element in a given paint has not been discussed. Obviously, if one paint specimen reveals an easily observed amount of an element, and a second does not, the two samples have a defined point of difference. From this standpoint, the probabilities associated with the null interval (nonobservable) of the elements are useful to consider. However, when an element is not observed in either of two paint samples there is no basis on which to judge the sameness or difference at that point of comparison, and the element is not a useful identification point.

Associated with the foregoing is the case of an element for which a firm value was actually determined at below the average detection limit for all points. This sometimes occurs when the interferences from other elements are unusually small. Such values were of little use in treating the whole data set, but can be of definitive use when comparing two paint samples.

When it is considered that an average of nearly 15 elements are observed in paint samples by NAA when the nuclear reactor and modern Ge(Li) detector-equipped gamma-ray spectrometer is used, it can be appreciated that the possibilities of mismatching paint samples is remote. In forensic applications this feature endows the technique with great power to protect the innocent and/or search out the guilty.

Lest the reader be troubled by several apparent reasons, from Table 3, to doubt the last statement, the following comments are offered. Samples nos. 91 and 96 (Class II paints) match to five identification points (Mn, Al, Na, Br, and Cl). The observed code for these elements (FNJHL) has a probable observation frequency of 5.6×10^{-5} ; so, given that the code is observed once, the chance of observing it again among 155 samples is $1-(0.999944)^{155}$, or $\sim 1\%$. Our background of data suggests that this degree of similarity, taken in context of the total fingerprints for these paints (which differ at several points), arises from different batches of the same kind of paint. On checking the sources of the two samples, it is found that they both came from white Kenmore washers that have appreciably different serial numbers (1106115710 and 1106004803), and this supports the interpretation of the data.

Similarly the "fingerprints" of samples 166 and 167 (Class II paints) match at a majority of points, and are similar on those few points of difference. The two samples were taken from the same house — one from a utility room wall and the other from a bedroom door jamb. The results suggest that the samples either came from different cans of the same batch of paint, or, less likely, that they came from different batches of the same kind of paint.

Samples 23 and 24 (Class III paints), which have the same "finger-prints" were already cited as having been obtained from the same automobile.

On the average, paints taken from different panels of massassembled machines had variations somewhat greater than those encountered from replicates taken from the same panels. The differences were of the same order as those encountered between interbatch and intrabatch comparisons of samples taken from fresh cans of paint. However, it cannot be automatically inferred that different panels of a given, massproduced machine are painted from different batches of the same kind of paint. Rather, it is more likely that the analogy is usually affected by an exaggeration of "inter-can" differences with a given batch of paint by environmental exposure differences. Such exposure differences may be related to geometric orientation of exposure and/or geometric orientation during application. The latter factor can cause differences in coating thickness with concommittant differences in susceptibility to exposure effects; of course neither can it be denied that differences in the mixing of a given paint by, say, two spray painters, might cause interpanel differences.

2.6 CONCLUSION

It has been shown that NAA is a powerful tool for comparing paint samples and that reasonable interpretations of NAA data as to the commonality of source of two paint samples can be made. This conclusion derives from the experimentally determined distribution functions of concentrations of a large number of identification points (elements) and the successful demonstration that specific probabilities for individual identification points may be multiplied to ascertain the probability of encountering a given "fingerprint".

3. EXAMINATION OF EVIDENCE PAPER SAMPLES

3. 1 INTRODUCTION

Paper is another important category of evidence material. It is usually produced in rolls and most manufacturers use several machines in a production run. The paper is then cut to size, and, in order to achieve a proper weight for a given quantity of paper, sheets from several rolls (each of slightly different density) are interleaved.

A troublesome area from the forensic standpoint is that the balance of a production run left after satisfying orders is saved and interleaved with sheets from a subsequent production run. Thus, unlike paint, the product cannot be definitively related to a specific, homogeneous batch of material at this time. It would require some form of deliberate product tagging to effect an equally favorable origin aspect for paper as intrinsically exists for paint.

Despite the above aspect, paper has been rather extensively examined by NAA to ascertain the probable forensic utility of the method for paper "identification". It was found that NAA provides a set of identity parameters that can usefully complement the more traditional characteristics used by the scientific crime investigator — characteristics such as mold and press marks, watermarks, density, color, strength, and microscopic fiber properties.

Unfortunately, program constraints did not permit the acquisition of a full population statistic, as in the case of paints. Nor was it possible to pursue the very attractive potential use offered by the fact that NAA should be useful in characterizing paper ash. The majority of identification points addressed by NAA are nonvolatile; therefore, unlike the

more traditional identification parameters, they do not disappear when paper is burned. Thus, the program's attention to paper (absent during the present report period) can serve as an important basis for further useful efforts.

3.2 EXPERIMENTAL

The experimental procedure was the same as employed in the examination of paints, except that only the NaI(T1) detector, coupled to a 400-channel gamma-ray spectrometer, was used subsequent to the sample irradiations.

Initial experiments were directed at a comparatively small array of papers and printing pigments. This was followed by examination of 120 samples of white bond paper from nine manufacturers. The 120 samples were of 40 different kinds (based on manufacturer and manufacturer specifications of name, rag content, wood fiber content, and opacity). All of the several samples of a given kind were produced in different production runs, but in no case did the maximum time difference in production for a given kind of paper exceed several weeks. Hence the variations within replicate analysis of a given kind of paper may be deemed to be representative of the variations that can be encountered in a package of paper.

3.3 RESULTS

The initial work served mainly to indicate that a number of elements were observable in paper by NAA and that these elements displayed fairly wide concentration ranges. It was found that the elemental constituents of ink pigments could not be measured on printed paper.

In the larger study, 12 elements were observed with various concentration ranges and frequencies of observation, as shown in Table 7.

Table 7

ELEMENTS OBSERVED IN 120 PAPER SAMPLES

		Observed	Concentra	tions (ppm)	
Element	Observation Frequency	Min.	Max.	Ratio Max/Min	Indicator Radioisotope(s)
Ti	78%	1100	17,600	15	Ti ⁵¹
Al	100%	17	19, 700	1,250	Al^{28}
Ca	21%	360	27, 100	7 5	Ca ⁴⁹
Mn	99%	0.33	5 7	173	Mn ⁵⁶
Na	99%	83	2,540	30	Na^{24}
Cl	70%	13	1,440	1, 100	C1 ³⁸
Тə	19%	0. 14	2. 05	15	\mathtt{Ta}^{182}
Zn	35%	5.5	222	40	Zn ⁶⁵
Sb·	70%	0.02	77	3,850	Sb^{122} , Sb^{124}
Cr	51%	0.43	23	54	Cr ⁵¹
La	1 7%	0. 03	3.51	117	\mathtt{La}^{140}
Au	5%	0.0002	0.02	100	Au^{198}

The observation frequencies among the individual samples altered somewhat on averaging due to incomplete data. Samples 4 and 5 were of the same kind, for example, and Zn was seen in sample 5 but not in sample 4. Therefore no average Zn value for that kind of paper (Fox River, English Bond) was obtained. However, average values for elemental concentrations were found among the 40 kinds of paper in the following numbers:

The other elements were less frequently observed.

The average relative standard deviations of the average values obtained for the foregoing eight elements were as follows:

A1 - $\pm 27\%$	$C1 - \pm 41\%$
$Mn - \pm 33\%$	Sb - ±68%
$Na - \pm 35\%$	$Cr - \pm 70\%$
Ti - ±33%	$Ca - \pm 22\%$

In view of the observed wide range of variability that can occur within closely spaced production runs of a given kind of paper, the representation of results by letter codes utilized somewhat broader ranges than were used in the case of paints. The codes for the eight most prominent elements were as follows:

	Concentration		Concentration
Code	Range (ppm)	Code	Range (ppm)
A	0. 1-0.31	. G	100-310
В	0.31-1	Н	310-1,000
С	1-3.1	I.	1,000-3,100
D	3. 1-10	<u>J</u>	3, 100-10, 000
E	10-31	K	10,000-31,000
F	31-100		

Using the above codes, the concentrations of Al, Mn, Na, Ti, Cl, Sb, Cr, and Ca are as given in Table 8. It will be noted that only two papers have identical "fingerprints" (including lack of detection of an element as being significantly different from the detection of the element when comparing two papers) - Bryon Weston's 25% Rag Merit and 25% Research Bonds.

Table 8

CODED RESULTS, 40 DIFFERENT BOND PAPERS

	Paper	<u>A1</u>	Mn	Na	<u>Ti</u>	<u>C1</u>	<u>Sb</u>	Cr	<u>Ca</u>
1.	Fox River, National Bank Bond	E	С	G	I	G		В	I
2.	Fox River, Anniversary Bond	\mathbf{E}	D	F	Ī	Ğ		_	Ī
3.	Fox River, Fox River Opaque	F	С	G	K	H	С	-	Ī
4.	Fox River, Fox River Bond	G	В	G	I	H	A	-	Ī
5.	Fox River, Fox River Laid	G	С	G	I	Н			J
6.	Fox River, English Bond	G	С	G	J	Н	В		I
7.	Byron Weston, 25% Merit		,				_		_
	Opaque	Н	В	H	J	I	С		
8.	Byron Weston, 25% Weston	*							
•	Opaque Bond	H	В	Н	K	I	С	С	
9.	Gilbert, Lancaster	H	С	G	J	G	\mathbf{B}	С	Н
10.	Brown, Cupon Bond	H	С	Н	Ī	Н	С	D	
11.	Byron Weston, 25% Weston's								
	Bond	I	В	G	J	I.	С		
12.	Crane, Crane's Bond	I	В	G		F	Α	С	
13.	Crane, Crane's Crest	I	В	G		G	В	С	
14.	Byron Weston, 50%								
	Winchester Bond	·I	В	H	J	Н	В	$\dot{\mathbf{D}}$	Н
15.	Byron Weston, 25% Rag				•				
	Merit Bond	I	В	H.	J	I	В		
16.	Byron Weston, 25% Research								
	Bond	I	В	H	J	I	В		
17.	Brown, Contract Bond	1	С	G	J	G	D		
18.	Gilbert, 25% Radiance Bond	Ι	C	G	J	G			
19.	Parsons, Parsons' Bond	I	C	H	J		С		
20.	Rising, Rising Parchment	Ι.	С	H	K		D		
21.	Rising, Rising Bond	·I	D	G		H	D		J
22.	Plover, Cold Springs Bond	I,	D	H	J		В	С	
23.	Plover, Permanized			•					
	Parchment	I	D	H	J			•	
24.	Parssons, Edgemont Bond	1	D	I	J				
25.	Plover, Artesian Bond	I	E	H	J	H		С	

Table 8 (Continued)

	Paper	<u>Al</u>	Mn	<u>Na</u>	Ti	<u>C1</u>	Sb	$\frac{Cr}{}$	<u>Ca</u>
26.	Plover, Artesian Bond								
	Opaque	I	E	Н	K	H	В		
27.	Plover, Erasable Plover								
	Bond No. 20	I	F	I	J		С		
28.	Byron Weston, Old Hampshire								
	Bond	I		•	J				H
29.	Gilbert, Gilbert Opaque Bond	J	В	Н	K	G	F		
30.	Valley, Valley Congress Bond	J	C	G		G		D	
31.	Valley, Realopack Densor								
	Fifty	J	С	G				D	
32.	Brown, Trojan Bond	J	С	Н	J		${f E}$		
33.	Gilbert, 25% Gilbert Superese	J	C	Н	K				I
34.	Valley, Raceway Bond	J	C	Н		H		\mathbf{E}	
35.	Plover, Plover Bond	J	E	I	I			D	
36.	Valley, Valley Forge Bond	K	С	G		Н		E	
37.	Gilbert, 25% Gilbert Bond	K	C	H		H	Α	E	
38.	Valley, Edgeworth Bond	K	C	Н		H		E	
39.	Gilbert, 25% Envelope Bond	K	C	Н				E	
40.	Plover, Plover Bond Opaque	K	E	Н	K				

3.4 DISCUSSION

The state of the paper work at this time is analogous to that at the end of the initial phase of the paint investigation. For example, one matching pair of "fingerprints" was found in Table 1, just as one such pair was found in Table 8. Thus, despite the greater relative "inter-batch" variations (variations expected within, say, a ream of paper) defined in paper, one may anticipate that a larger scale study, wherein papers are taken randomly from the environment in accordance with a suitable sampling model, would be quite fruitful — especially if a Ge(Li) detector instead on a NaI(Ti) detector were to be used.

The present data are insufficient to allow extensive discussion of frequency distributions of the elements. However, the results at hand, which are summarized in Table 9, indicate that only Sb may have a Gaussian distribution function. Therefore, there may be difficulty in finding a standard distribution function expression for the element concentrations in paper, as was the case with paints. This may prove to be a common situation where the material in question is manufactured with deliberate additions of various compounds to achieve a given product. Paints obviously acquire much of their observed element content by addition of pigments and extenders to organic vehicles and solvents. Opacifiers, whiteners, and other ingredients are added to paper to achieve desired qualities.

As a preliminary test of the independence of element concentrations, the data of Table 9 (II) were used to compute the expected numbers of observations of all possible code combinations pairs for aluminum with Mn, Na, Ti, Cl, and Sb. It was then hypothesized that the observed numbers of observations of the possible codes did not significantly differ from the expected numbers. This hypothesis was not rejected by the chi-square test. Thus it is entirely possible that NAA data from paper can be treated in the same manner that was used in the case of paint: i. e., it may be unnecessary to find a suitable standard mathematical distribution function.

Table 9
CONCENTRATION FREQUENCIES, 40 DIFFERENT BOND PAPERS

I. Number of Observations

						Code	€					
Element	<u>A</u>	В	<u>C</u>	D	E	F	<u>G</u>	H	I	J	K	Total
Al					2	1	3	4	18	7	5	40
Mn		10	19	5	4	1						39
Na						1	15	20	3			39
${f Ti}$									6	17	7	30
Cl						1	8	13	5			27
Sb	3	8	7	3	1	1						23
Cr		l	6	5	5							17
Ca								4	6	2		12

II. Percent of Observations Per Code

						Cod	le				
Element	A	B	<u>C</u>	D	E	F	G	H	<u>I</u>	J	K
Al					5	2.5	7.5	10	45	17.5	12.5
Mn		25.6	48.6	12.8	10.2	2.6					
Na						2.6	38.5	50.5	7.7		
Ti									20	57	23
C1						4.3	29.5	48.3	18.4		
Sb	13	34.6	30.5	13	4.4	4.4					
Cr		5.9	35.4	29.4	29.4						
Ca		,						33	50	17	

3.5 CONCLUSION

The examination of paper by NAA has been shown to be at approximately the same stage of development as the paint study prior to the analysis of environmental samples. However, although the present data are comparatively useful, it has been shown that the same statistical procedure used to treat the paint data will probably apply to paper as well.

In fact, it has been found that the observation frequency code pairs of each of five elements with aluminum can be successfully predicted from their individual, observed, fractional occurrences. Thus, at least the six elements (Al, Mn, Na, Ti, Cl, and Sb) have a nucleus of background data from which to estimate probabilities of matching and mismatching in forensic applications.

In view of the encouraging results to data, it is recommended that an adequate environmental sample of paper be obtained and examined by NAA with the use of a Ge(Li) detector.

4. GUNSHOT RESIDUE STUDIES

4.1 INTRODUCTION

It was found very early in the program that a number of constituents of cartridges can be readily measured by NAA in material removed from the hand of a person who has discharged a firearm. Some such constituents, such as copper, are of relatively common environmental occurrence, while others, especially barium and antimony, are not common. Hence, it seemed possible that a suitable use for NAA would be to discern whether or not a person had fired a weapon. The motivation for this particular investigation derived from criminalists' advice, especially R. H. Pinker (then Chief Criminalist of the Los Angeles Police Department Crime Investigation Laboratory), that no suitable physical or chemical test for proving that a person fired a weapon then existed.

The fabled dermal nitrate test, wherein the inside surface of a paraffin hand test is treated with diphenylamine-sulfuric acid solution to form blue reaction products with unburned nitro compunds, had been long ago shown to be unreliable in almost classic proportions: i.e., a positive test can be obtained from nearly 50% of those who have not fired a gun (since human effluvia and environment contain appropriate chemical reactants), while a negative test results nearly half the time when applied to a person who has fired a gun.

A method by Harrison and Gilroy, ⁽¹²⁾ which determined elemental constituents (Ba, Sb, Pb) in gunshot residues by color reactions proved disappointing to criminalists in actual practice due to marginal sensitivity. Since NAA has excellent sensitivity for Ba and Sb (not for Pb, however), the initial NAA experiments that proved so encouraging were carried out.

4. 2 EXPERIMENTAL AND RESULTS

Several means for removing gunshot residue particles were investigated, including rinsing the hand with various reagents, wiping with special papers (such as Whatman filter paper) or cotton swabs, and the more traditional paraffin handlift. It was found that the paraffin handlift provided lower intrinsic background levels of barium and antimony and more convenience of handling than the alternatives. Hence the standard method for removing gunshot residues from the hand evolved to be the paraffin handlift; as follows:

- 1. Food grade paraffin is melted in a clean porcelain evaporating dish, care being taken to avoid bringing the temperature of the molten paraffin to over 120°F.
- 2. A retainer sample of the paraffin is taken for measurement of Ba and Sb blank values.
- 3. Molten paraffin is picked up with a small clean paint brush and allowed to flow onto the skin of the hand being tested. Care is taken to avoid a brushing motion, since this would transfer residue material to the brush.
- 4. After the paraffin has cooled and solidified, it is carefully removed from the hand for analysis.

It is advised that the person doing the handlift have clean hands, wear plastic gloves, operate in a clean environment, and never re-use any of the material (paraffin, dish, brush, gloves) in subsequent tests.

The paraffin handlift is placed in a clean polyethylene vial, which is then sealed and subjected to neutron activation in a nuclear reactor together with separate barium and antimony comparator standards. At this laboratory the GRT TRIGA Mark I nuclear reactor is used. The reactor has a rotating specimen rack (1 rpm) with numerous equivalent sample positions. The handlift and standards are placed in adjacent positions and

irradiated in a thermal neutron flux of 1.8 x 10^{12} n/cm²-sec for 30 minutes while in the rotating rack. Thus, they all receive the same degree of activation, and no secondary flux monitors are required.

The sodium and chlorine in perspiration picked up in a handlift become activated, also, and preclude the direct observation of barium and antimony indicator radioisotopes by direct gamma ray spectrometry.

Therefore, the desired indicator radioisotopes are separated from the interfering radioisotopes by a radiochemical procedure.

1. The radiochemical procedure employed is swift and convenient. The activated handlift is placed in a beaker and to it are added the following: 15 ml distilled (and/or deionized) water, 3.5 ml HCl (conc.), 1 ml HNO₃ (conc.), ~100 mg NaCl, ~20 mg Cu, and exactly known amounts (~20 each) of Ba⁺² and Sb⁺³ carriers. The mixture is boiled for 10 minutes, cooled, and the aqueous solution poured into a 50 ml centrifuge tube. By the addition of 1 ml of H₂SO₄ (conc.), BaSO₄ is precipitated, and centrifugation quickly isolates the precipitate. The supernate is to a 150 ml separatory funnel.

The BaSO₄ precipitate is slurried in pure H₂O, filtered onto preweighed filter paper, washed with water, washed with ethanol, dried at 110° for 15 minutes, and weighed (to determine the radiochemical yield of barium). The precipitate is transferred to a 2-dram polyethylene vial and the analytical indicator, Ba¹³⁹, is measured by multichannel gamma-ray spectrometry. In a modified procedure, the precipitate is neither dried nor weighed (simply filtered and washed thoroughly), and the radiochemical yield is subsequently determined by measurement of recovered Ba carrier with NAA. Of course, in either modification the original Ba¹³⁹ activity is related to that of the Ba comparator standard in the computation of the element in the handlift.

The supernate from the $BaSO_4$ precipitation step is adjusted to a pH of 1.5-2.0 with NaOH solution, and 5 ml of dithizone reagent (400 mg of dithizone in 100 ml $CHCl_3$) is added. The mixture is shaken and the $CHCl_3$ phase (now containing Cu and interfering Cu^{64}) is discarded. About one gram of thioacetamide is added to the aqueous phase to precipitate Sb_2S_3 . It is advisable to warm the aqueous phase to effect complete precipitation. The precipitate is centrifuged, the supernate is discarded, and the precipitate is dissolved in 10 ml of HCl.

The solution containing dissolved $\mathrm{Sb}_2\mathrm{S}_3$ is boiled for ~ 15 minutes, which removes sulfur as $\mathrm{H}_2\mathrm{S}$ and free sulfur. The solution is diluted to 30 ml with $\mathrm{H}_2\mathrm{O}$, the free sulfur is centrifuged out, and the supernate is transferred to a clean centrifuge tube. The solution is made to about 40 ml with $\mathrm{H}_2\mathrm{O}$, 2 ml of Oxsorbent (chromous chloride solution) is added, and the tube is placed in a boiling water bath for about 30 minutes. The solution is allowed to stand overnight at room temperature to permit complete reduction of the antimony to the elemental state. The antimony precipitate is transferred to a 2-dram polyethylene vial, measured by gamma-ray spectrometry for Sb^{122} content (the Sb comparator standard is likewise measured), and finally the radiochemical yield of Sb is determined by NAA.

2. The experimental program included studies of multiple firings, relative amounts of residues deposited on various parts of the hands, the possibility of tagging cartridges to increase the uniqueness of the test, the effect of wind on deposition of residues, and an extensive study of Ba and Sb levels in handlifts taken both from persons who had and had not fired guns. These last studies were culminated in the present reporting period.

Although a few initial experiments indicated the possibility of handlift levels of Ba and Sb proportionate to the number of times the hand had fired a weapon, more extensive studies indicated that this was not the case. Apparently the major amount of residue deposited from one residue is blown off, but replaced, in a subsequent firing. The largest amounts of residue from a firing were found, in order, on the trigger finger, thumb web, and back of hand. The palm of the hand received the least amount of residue. However, extensive Ba and Sb residues were found in various parts of a weapon that had been discharged but not cleaned: and persons who extensively handled such weapons, but did not fire them, picked up more Ba and Sb on the palm than on other parts of the hand.

It was found that the Ba and Sb deposited on the hand from a firing are easily removed by washing the hands with soap and water.

Winds were found to influence the quantitative, not qualitative, results of firings. A wind from the front increased deposition to the hands, while winds from the side or rear had a contrary effect.

Efforts were made to utilize dysprosium and europium tags (both elements are determined with exceeding sensitivity by NAA) to facilitate the unequivocal detection of gunshot residues. Experiments involving tagged primers and tagged powders were carried out, but neither kind of tagging offered conclusive results. In the light of subsequent information regarding the particulate nature of gunshot residues, it seems likely that improved tagging procedures could be effective.

It has come to light that the bulk of Ba and Sb in gunshot residues deposited on the hand is contained in a fairly small number ($\sim 10-20$) of relatively large (10-50 μ) particles, and that <0.1% of the Ba and Sb in the cartridge primer (the location of the major amounts of these elements in the cartridge) ends up on the hand subsequent to a firing. Obviously then, smaller particles or fine powder are not efficient sources of firing evidence. The correct procedure for tagging cartridges with refractory rare earth materials must take this fact into account. Unfortunately, these facts were not available early enough in the program, and the tagging methods virtually assured that the tags would be emitted as fine powders.

During the present contract period it came to light that the primers in 0.22-cal. bullets from one manufacturer did not contain antimony. However, because the bullet lead contained $\sim 1\%$ Sb, this element was nevertheless found in handlifts taken from those who had fired such bullets. Obviously, the bullets are abraded as they leave the cartridge and fragments of lead are deposited on the gun hand.

3. All handlift results obtained in the systematic study of handlifts from persons who had not fired weapons and persons who had fired weapons are presented in Tables 10 through 19.

These results, many of which were gathered in the present report period, are presented as multiples of the raw values (times 100 for Ba, and times 1000 for Sb) for convenience of presentation and later manual manipulation. The firing data tables are essentially self-explanatory; i. e., the Ba and Sb values were obtained from handlifts taken of a hand that had just fired a weapon of the indicated caliber.

The handblank tables (Tables 10, 11, 12, and 13) present values obtained from persons in different occupational classifications, as follows:

Handblank Classification	Occupations Sampled
A	Carpenters, accountants, TV technicians, secretaries, watch repairmen, gardeners, laboratory technicians, photographers, radioisotope technicians, theoretical chemists, electronics technicians, chauffeurs, electricians, computer operators, nurses, physicians, storekeepers
В	Plumbers, graphic artists, mechanics, draftsmen, heating and air-conditioning repairmen
С	Electronic assemblers
D	Auto mechanics, painters, machinists, maintenance men

Table 10 CLASS A HANDBLANKS BARIUM AS μ_B FOUND x $10^2,\$ ANTIMONY AS μ_B FOUND x 10^3

Sample	Ва	Sb	Sample	Ba	Sb	Sample	Ba	Sb	Sample	Ba	Sb	Sample	Ba	Si
1	14	60	40	25	10	79	4.4	62	118	8.6	1	157	7.4	
.2	22	60	41	9	5	80	18.2	21	119	11	10	158	12	1
3	32	30	42	9	10	81	34	17	120	4.1	3.2	159	70	1
4	19	10	43	7	5	82	23	17	121	1.6	3.4	160	116	
5	3	50	44	2	5	83	29	25	122	16	4.8	161	223	
6	.4	5	45	31	20	84	33	31	123	20	2.4	162	289	
7	18	20	46	29	65	85	16	9	124	19	4.3	163	16	
8	12	20	47	40	58	86	19	7	125	3.1	24	164	8.3	
9	48	10	48	26	39	87	9.7	11	126	4.8	10	165	5.3	!
10	44	10	49	13	110	88	8.7	10	127	5.4	1	103	٠. ٠	
11	18	-5	50	18	135	89	14	7	128	8.1	3.7	1		
12	32	5 '	51	9.1	2	90	14	8	129	8.8	1.9	İ		
13	7	5	52	10	2	91	14	20	130	4.2	6.5			
14	2	5	53	4.7	2	92	15	21	131	4.6	7.6			
15	3	5	54	5.7	2	93	51	54	132	46	4.4			
16	3	5	55	14	11	94	47	94	133	5.9	8.7	į		
17	17	5	56	6.8	6	95	10	8	134	1.9	3.7			
18	6	5	57	22	51	96	4.1	2	135	5.8	13	1		
19	9	30	58	28	3	97	4.0	3	136	2.4	6	ł		
20	7	5 -	59	22	23	98	7.1	4	137	9.0	4	i		
21	15	5	60.	27	18	99	3.2	2	138	14	5	İ		
22	7	5	61	12	12	100	2.5	2	139	8	10	1		
23	4	20	62	5.9	2	101	19	2	140	5.1	6	1		
24	1	10	63	268	74	102	17	5	141	4.4	5	ĺ		
25	22	80	64	35	23	103	4.5	1	142	3.4	3			
26	8	10	65	22	69	104	3.2	1	143	2.0	3			
27	3	5 .	66	26	69	105	7.3	6.7	144	2.9	3			
28	4	5	67	61	22	106	8.2	1	145	1.8	3	ŀ		
29	9	5	68	20	12	107	10.5	12	146	2	3	Ì		
30	8	30	69	6.8	17	108	3.2	7	147	1	3			
31	26	5	70	5.5	20	109	4.8	2	148	4.5	3			
32	9	20	71	5.5	33	110	8	1	148	6.7	30	İ		
33	11	5	72	4.5	3	111	9.1	4.6	150	1.8	21			
34	6	5	73	11.9	11	112	2.6	3.3	151	65	1			
35	10	5	74	8	- i	113	2.3	1	152	71	232			
36	8	5	75	14.7	24	114	7.2	1	152	15		1		
37	26	5	76	9	17	115	6.5	10.7	153 154	12	1			
38	13	5	77	20.8	4	116		. 1	155	4.8	18			
39	15	20	78	15.2	6	117	2.1				2			
٠,	4.7		,,	13.2	١٣	117	۷	1	156	11	7			

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Table 11 CLASS B HANDBLANKS BARIUM AS μg FOUND x $10^2,\ ANTIMONY\ AS\ \mu g$ FOUND x 10^3

Sample	Ва	Sb	Sample	Ба	Sb	Sample	Ba	Sb	Sample	Ba	Sb
1	128	37	11	114	6	21	23	9	31	17.1	34
2	118	74	12	261	5	22	24	7	32	22	16
3	5	8.2	13	1 95	5	23	198	49	33	107	4
4	73	34	14	157	14	24	185	46	34	164	4
5	165	18	15	75	4	25	71	14	35	15.9	81
6	219	16.8	16	36	6.4	26	8.6	8	36	16.7	136
7	48	52	17	55	5	27	13.5	9.2	37	16.5	10
8	86	47	18	51	8	28	12.7	9	38	37	31
9	36	6	19	48	2	29	54	14			
10	35 .	1.3	20	134	4.5	30	47	7	j		

Table 12 CLASS C HANDBLANKS BARIUM AS μg FOUND x $10^2, \ ANTIMONY \ AS \ \mu g \ FOUND x \ 10^3$

Sample	Ba	Sb	Sampl	е Ва	Sb	Sample	Ba	Sb	Sample	Ba	Sb
1	7.6	12.7	5	7	31	9	6.5	15	13	4.7	8.9
2	16.5	31	6	94	42	10	13.7	40	14	32	27
3	32	66	7	33	53	11	15.2	66	15	19.7	10.8
4	5.2	10	.8	7.7	22	12	9.4	14.4	16	6.8	26

Table 13 ${\tt CLASS~D~HANDBLANKS} \\ {\tt BARIUM~AS~\mu g~FOUND~x~10^2,~ANTIMONY~AS~\mu g~FOUND~x~10^3}$

Sample	Ba	Sb	Sample	Ba	Sb	Sample	Ba	Sb	Sample	Ba	Sb
1	45	4	11	37	17	21	13.5	10.8	31	57	15
2	37	2	12	20	21	22	48	29	32	206	22
3	48	31	13	16	3.1	23	72	49	33	194	15
4	30	5	14	15	2.6	2.4	23	22	34	112	99
5	47	31	15	278	174	25	28	37	35	126	127
6	178	51	46	470	383	26	121	180	36	16.5	28
7	1210	1.45	47	280	198	27	76	119	37	35	26
8	528	428	48	23	21	28	117	121	38	26	120
9	28	24	49	13.6	9.5	29	151	70	39	3 2	598
10	44	36	20	319	43	30	92	26	40	179	72
									41	199	98

Table 14 $0.\ 22\ CALIBER\ FIRINGS$ BARIUM AS μg FOUND x $10^2,\ ANTIMONY\ AS\ \mu g$ FOUND x 10^3

Sample	Ba	Sb	Sample	. Ba	Sb	Sample	Ba	Sb
1	54	111	36	127	127	71	110	117
2	125	242	37	22	66	72	14.4	34
3	32	56	38.	98	121	73	12.8	24
4	71	72	39	15.9	47	74	36	7 3
5	21	39	40	265	521	75	9.1	9
6	53	126	41	35	101	76	16.8	15
7	12	50	42	213	1120	77	2.5	7
8	589	144	43	6.5	29	78	. 5	27
9	15.5	40	44	73	76	79	8.3	.18
10	450	302	45	5.5	16	80	17	184
11	28	70	46	96	114	81	7.8	14
12	169	146	47	14.9	40	82	4	11
13	25	70	48	16	41	83	10.4	4.9
14	112	252	49	167	45	84	5.4	236
15	8.2	37	50	17.2	32	85	17	269
16	123	133	51	59	199	86	85	215
17	19.8	45	52	22	43	87	37	120
18	240	288	53	17.5	47	88	28	94
19	19.3	36	54	29	96	89	24	91
20	274	64	55	301	24	90	40	160
21	15.1	43	56	112	75	.91	23	112
22	129	312	57	84	36	92	220	374
23	14.4	40	58	240	64	93	38	119
24	617	61	59	34	138	94	153	318
25	13.7	37	60	16.1	31	95	119	210
26	9.9	25	61	9.2	22	96	161	88
27	524	192	62	14	17	97 .	31	133
28	204	130	63	97	141	98	609	3780
29	492	2910	64	13.3	27	99	267	835
30	288	98	65	156	74	100	39	200
31	6.7	15	66	10.4	31	101	35	26 6
32	246	184	67	29	57			
3 3	49	58	68	16.3	47			
34	62	111	8.9	35.1	36			
35	16.7	37	70	11.3	20			

Table 15 $0.~25~CALIBER~FIRINGS\\ BARIUM~AS~\mu g~FOUND~x~10^2,~ANTIMONY~AS~\mu g~FOUND~x~10^3$

Sample	Ва	Sb	Sample	Ba	Sb
1	776	1260	10	149	266
2	222	499	11	298	624
3	846	561	12	384	579
4	96	208	13	107	150
5	410	604	14	161	307
6	243	296	15	226	463
7	381	148	16	222	359
8	347	341	17	212	419
9	1420	1670	18	105	253

Table 16 $0.44~CALIBER~FIRINGS \\ BARIUM~AS~\mu g~FOUND~x~10^2,~ANTIMONY~AS~\mu g~FOUND~x~10^3$

Sample	Ва	Sb
1	149	2 59
2	452	152
3	38	166
4	168	55
5	405	113
L		

Table 17 0.38 CALIBER FIRINGS BARIUM AS μ_B FOUND x 10^2 , ANTIMONY AS μ_B FOUND x 10^3

Sample	Ba	Sb	Sample	Ba	Sb	Sample	Ba	Sb
.1	144	615	23	104	314	45	74	418
2	64	325	24	121	405	46	43	211
3	96	255	25	113	334	47	72	187
.4	25	75	26	109	205	48	45	146
5	85	295	27	71	167	49	70	34
6	240	625	28	56	148	50	5 3	263
7	154	575	29	113	410	51	28	161
8	328	555	.30	57	135	52	73	314
9	173	285	3 1	76	234	53	22	78
10	69	385	32	487	990	54	6	30
11	49	103	33	16.8	93	55	7.9	70
12	33	81	34	51	398	56	32	143
13	47	194	35	76	666	57	12.3	41
14	275	678	36	15	37	58	1.8	14
15	99	360	37	28	673	59.	20	199
16	67	163	38	16.5	84	60	17	97
17	71	253	39	53	138	61	34	166
18	25	. 67	40	. 17	84	62	13	92
19	25	88	41	37	287	63	270	432
20	30	96	42	26	195	64	12	. 75
21	148	393	43	16	121	65	20	150
22	211	791	44	83	383			

Table 18 $0.45 \; \text{CALIBER FIRINGS} \\ \text{BARIUM AS μg FOUND $x:10^2$, ANTIMONY AS μg FOUND $x:10^3$}$

Sample	Ba	Sb	Sample	Ba	Sb	Sample	Ba	Sb
,				_				
1	637	1880	27	217	754	52	612	1220
2	1560	4350	28	317	1270	53	300	1120
3	940	3350	29	6 44	2030	54	634	2550
4	2610	3240	30	792	2700	55	214	1400
5	568	1730	31	195	808	56	1300	5160
6	524	1400	32	628	1710	57	1550	919
7	2 35	642	33	207	343	58	100	716
8	1990	5980	34	1100	2320	59	891	1820
9	379	1290	35	532	836	60	191	468
10	559	1630	36	175	208	61	189	776
11	406	11600	37	1370	3500	62	211	702
12	209	530	38	273	373	63	394	691
13	244	.889	39	587	1170	64	11	22
14	289	1110	40	488	935	65	161	247
15	298	933	41	5 2 6	1410	66	165	1830
16	868	3010	42	700	1020	67	908	5780
17	277	1 ,550	43	1340	2120	68	622	1260
18	855	2170	44	788	2090	69	14	37
19	375	1310	45	184	382	70	716	1920
20	972	2560	46	88	132	71	805	2230
21	258	997	47	1310	4210	72	204	1380
22	720	2740	48	75	174	73	96	342
23	67	266	49	84	296	74	215	521
24	486	1310	50	593	552	75	134	355
25	58	232	5 1	138	280	76	291	601
26	336	1600		133			2 / 4	001

Table 19 9~mm~FIRINGS BARIUM AS μg FOUND x 10^2 , ANTIMONY AS μg FOUND x 10^3

Sample	Ва	Sb	Sample	Ba	Sb	Sample	Ва	Sb
1	1540	56	23	42	142	45	56	145
2	559	753	23	126	218	46	466	520
3	1130	2050	25	64	82	47	51	227
4	363	610	26	131	719	48	247	380
5	137	190	27	25	192	49	252	1570
6	227	930	28	43	820	50	156	215
7	322	570	29	28	24:1	51	929	4200
8	277	840	30	44	187	52	104	183
9	164	247	31	46	338	53	266	1150
10	127	380	32	2 80	680	54	324	630
11	111	360	33	53	86	55	194	990
12	102	580	34	665	318	56	309	1520
13	169	490	35	57	133	57	100	266
14	122	460	36	234	860	58	118	130
15	480	3050	37	62	113	59	219	1290
16	214	710	38	859	3230	60	47	102
17	136	700	39	48	149	61	267	840
18	84	164	40	534	2570	62	267	354
19	75	296	41	233	232	63	492	2370
20	56	124	42	674	3170	64	212	1090
21	163	495	43	856	1200	65	73	156
22	41	123	44	144	770			

The classification of handblanks reflects the differences to which persons in various occupations are normally exposed to contact with, and transfer to the hands of, barium and/or antimony compounds. Relative to Class A, and on the average, the occupations related to Classes B, C, and D permit the pick-up of more of the elements as follows:

•	Relative to Class A						
Class	Ba	Sb					
В	~4	~ 1. 3					
С	∼ 1.0	~1.8					
D	~6.9	~4.8					

It can be seen that Class B is a "high Ba" class, Class C is a "high Sb" class, and Class D is a "high Ba and high Sb" class.

The firings in all caliber categories involved a multiplicity of weapon-cartridge combinations based on weapon type (revolver or automatic), weapon manufacturer, and cartridge manufacturer. These factors were variously combined in order to avoid bias due to peculiarities inherent in any one combination.

The average values for all handlift classifications are given in Table 20.

Table 20 $AVERAGE\ VALUES\ FROM\ HANDLIFTS \\ BARIUM\ AS\ \mu g\ FOUND\ x\ 10^2,\ ANTIMONY\ AS\ \mu g\ FOUND\ x\ 10^3$

Classification	Ba	<u>Sb</u>		
Handblank Class A	18.9	16.4		
Handblank Class B	78. 2	2,2.2		
Handblank Class C	19. 4	29.8		
Handblank Class D	136. 1	78.3		
0.22 Caliber Firings	95.4	181.0		
0.25 Caliber Firings	319.6	506.0		
0.38 Caliber Firings	77.9	263. 0		
0.44 Caliber Firings	149.0	422.0		
0.45 Caliber Firings	540.0	1631.0		
9 mm Caliber Firings	261.6	797. 0		

4.3 DISCUSSION

The data from the systematic study of handlifts was tested and found to be not normally distributed in a majority of cases. The raw data showed that the overall average of Ba/Sb ratios in all classifications was nearly 10 (actually 9.53). Hence the data in Tables 10 through 20 may be regarded as ''normalized''.

Logarithms of the "normalized" data were taken, and the data were examined in each classification for log-normal distribution. For this purpose the data were classified in accordance with the following code:

Code	Range of Log of Normalized Value	Code	Range of Log of Normalized Value
A	0-0.3	Н	2. 1-2. 4
\mathbf{B}	0.3-0.6	I	2. 4-2. 7
С	0.6-0.9	J	2. 7-3. 0
D	0.9-1.2	К	3.0-3.3
E	1. 2-1. 5	L	3.3-3.6
F	1.5-1.8	М	3.6-3.9
G	1.8-2.1		

The classified data were assumed to lie within ±30 of the mean value in each classification. The number of range increment divided into 60 gave the number of 0 units per range increment, and it was then possible to use a standard table of areas per 0 unit to obtain expected observation frequencies for the Gaussian curve. The expected frequencies were compared to the observed frequencies with the chi-square test and it was found that few of the data sets, Ba and Sb from the 9 mm weapon firings were exceptions, could be accepted as log-normal.

Nevertheless, standard deviations about the means of the logarithmic values and Pearson Product-Moment correlation coefficients were computed. The validity of such parameters was less certain then would be

The total frequency along each +45° diagonal was obtained. This step is consistent with the previously mentioned physical and mathematical considerations. These totals were found to have a Gaussian distribution for all classifications except Handblank Class A and Handblank Class C. The test for Gaussian distribution is exemplified in the following.

Consider the totals obtained for each +45° diagonal of the Class B handblank chart of Table 21. Estimate the expected distribution for 38 observations in a similar number of groups for a normal distribution. Test the hypothesis, by the chi-square test, that the distributions are the same.

Expect	Found	(Difference) ² /Expect
0.3	0	0.3
1. 1	2	0. 7
3.0	5	1.3
6.1	6	0.0
8.6	6	0. 7
8.6	6	0. 7
6. 1	4	0. 7
3.0	5	1.3
1.1	3	3.3
0.3	1	0.2

chi-square = 9.2

The hypothesis cannot be rejected in thise case, since the chi-square value does not indicate a significant difference between the expected and observed distributions.

The Class A Handblank distribution was successfully fitted with two overlapping normal distributions. Thus, only Class C Handblank data are mathematically excluded from the following treatment. The 0.44 caliber firings were excluded on the grounds of too few data. In this connection, it may be noted that the number of concentration categories involved in the tests were adequate to avoid a false result from the chi-square test in all classifications, except for the 0.44 caliber firings. Obviously, with too few concentration categories (consider the extreme case of one and only one category, for example) almost any distribution data could be accepted as normal.

The expected, normal distributions can be viewed as the smoothed distributions of the observed data by virtue of the foregoing favorable tests. These smoothed distributions, expressed as percent of occurrence are given in Table 22. It will be noted that along each $+45^{\circ}$ diagonal in Table 21 the numerical sum of the Sb code and Ba code is constant, where it is considered that A = 1, B = 2, C = 3, etc. This numerical constant is designated by a Roman Numeral in Table 22. For example Roman Numeral VII specifies the (Sb, Ba) diagonal (A, F), (B, E), (C, D), (D, C), (E, B), (F, A) that appears in Table 21.

Table 22

SMOOTHED DISTRIBUTION OF OBSERVATION PROBABILITY
IN PERCENT, FOR EACH 45° DIAGONAL IN TABLE 21

Diagonal	Hand	lblank C	lass	F	irings,	Weapor	n Calibe	r
Category	<u>A</u>	B	D	0.22	0.25	0.38	0.45	9 mm
П	0.6							
III	3.3							
IV	9.4	0	0	0				
V	14.2	0.1	0.4	0.3				
VI	12.6	0.7	0.9	0.6		0		
VII	10.8	2.8	2.2	1.3		0.4		
VIII	12.0	7.9	4.5	2.5		0.9		
ΙX	13.2	15.9	7.8	4.3		2.2		0
X	11.0.	22.6	11.5	6.8		4.5		0.4
XI	7.3	22.6	14.7	9.4		7.8	0	0.9
XII	3.5	15.9	15.8	11.6	0	11.5	0.4	2.2
XIII	1.2	7.9	14.7	13.1	0.7	14.7	0.9	4.5
XIV	0.4	2.8	11.5	13.1	2.8	15.8	2.2	7.8
XV	0.3	0.7	7.8	11.6	7.9	14.7	4.5	11.5
XVI	0.1	0.1	4.5	9.4	15.9	11.5	7.8	14.7
XVII.	. 0	0	2.2	6.8	22.6	7.8	11.5	15.8
XIX			0.9	4.3	22.6	4.5	14.7	14.7
XX			0.4	2.5	15.9	2.2	15.8	11.5
XXI			0	1.3	7.9	0.9	14.7	7.8
XXII				0.6	2.8	0.4	11.5	4.5
XXIII				0.3	0.7	0	7.8	2.2
XXIV				0	0		4.5	0.9
XXV							2.2	0.4
XXVI	•					•	0.9	0
XXVII				•	•		0.4	
							0	

The probabilities given in Table 22 enabled the computation of the probability that any particular combination of Sb and Ba values resulting from the analysis of a handlift represents a handblank (lack of firing), where the handblank classification and caliber of weapon are known. This point of view is consistent with traditions of American jurisprudence in that it tests the assumption that the person, from whom a handlift was taken, is innocent. The array of these probabilities associated with each handblank classification (A, B, and D) in conjunction with 0.22, 0.25, 0.38, 0.45, and 9 mm weapons is given in Table 23.

The interpretation described, which is consistent with the various physical and mathematical considerations described, has the virtue of simplicity of application. The results of a given handlift analysis may be simply categorized and the probable innocence (of firing) can be read directly from Table 23.

A more sophisticated and mathematically rigorous method of interpreting the gunshot residue data is described next.

2. A More Rigorous Method of Interpreting Gunshot Residue Data:
The Univac 1108 computer was utilized in all computations
associated with the following treatment. The logarithms of the
raw Ba and Sb values were obtained, and all further computations involved the logarithmic values. Mean values and standard deviations thereof of each element in each classification
were obtained.

In the following treatment the values of the two elements, Ba and Sb, for a given sample are considered to comprise a vector: i.e. where the two elements for the Kth handlift are distinguished by subscripts 1 and 2:

$$\underset{\sim}{\mathbf{X}}(\mathbf{k}) = \begin{bmatrix} \mathbf{X}_{1}(\mathbf{k}) \\ \mathbf{X}_{2}(\mathbf{k}) \end{bmatrix}$$

PROBABILITY (IN %) THAT SO AND BE VALUES FALLING IN A GIVEN CATEGORY REPRESENT A HANDBLANK, WHERE OCCUPATIONAL HANDBLANK CLASS AND WEAPON CALIBER ARE SPECIFIED

Diagonal		22 Cal and dblank		-	25 Calib and dblank C		٠.	38 Calib and dblank C		•	45 Calibe and dblank C			mm, and lank Cla	.s.s.
Category	Α	В	D	<u>A</u>	<u>B</u>	D	<u>A</u>	<u>B</u>	<u>D</u>	<u>A</u>	<u>B</u>	. <u>D</u>	<u>A</u>	В	<u> </u>
Щ	,	— — . 	· -										
	>99.5		•						•				•	•	
v	97	25	57		1000								·	•	
IV	96	54	60				>99.5	>99.5	>99.5			. *			
VП	89	69	63		•		96	88	85			• • •		•	. •
νш	83	76	64			J	93	90 -	83		5.0				•
IX .	76	. 79	66		*		86	88	78	· .			>99.5	>99. 5	>99.5
×	62	77	63				.71	83	72				96	98	97
ΧI	44	73	61		·.		48.	74	66	>99.5	>99.5	>99.5	89 -	96	94
ХII	23	58	58	>99.5	>99.5	>99.5	23	58	58	90	98	98	61	88	89
ХШ	9.1	38	53	63	92 .	9.6	7.6	35	50	57 .	90	94	21	64	77
XIV	3.0	18	47	25	50	. 80	2.5	15	42	1,5	56	84	4. 9	26	60
ΧV	2.5	5.7	40	3.6	8.1	50	2.0	4.6	35	6.3	14	64	2.5	5. 9	41
IVX	1.1	1.1	32	0.6	0.6	22	0.9	0.9	28	. 1.3	1.3	36	0. 7	0.7	24
XVII	<0.5	<0.5	24	<0.1	<0.1	8.9	<0.1	<0.1	22 ,	<0.5	<0.5	16	< 0.1	<0.1	11
XVIII		٠,	17		ı	3.8		•	17			5.7			5. 7
XIX			. 14			2.5			15			2.5			3.4
XX			<0.5			<0.5			<0.5	•	•	<0.5			< 0.5

A mean vector is comprised of the means for each element,

$$\widetilde{\mathbf{x}} = \begin{bmatrix} \overline{\mathbf{x}}_1 \\ \overline{\mathbf{x}}_2 \end{bmatrix}$$
.

The variance of element no. 1, s_{11} , is the sum of the squares of the deviations of all values from the mean for that element divided by n-1, where n is the number of samples taken. The variance of the other element is s_{22} . The product of the summed deviations of each element from its mean, divided by n-1, is denoted by s_{12} or, equally s_{21} .

The covariance matrix is given as

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

The standard deviations are, of course, $(s_{11})^{\frac{1}{2}}$ and $(s_{22})^{\frac{1}{2}}$, and are denoted by σ_1 and σ_2 .

The correlation coefficient, r, is given as

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

The BVN (bivariate normal) is such that, where $r < \pm 1$, σ_1 and $\sigma_2 \neq 0$, and the mean values for each variable $< \neq \infty$, the joint density function is

$$f(X_1 X_2) = \frac{1}{2\pi\sigma_1\sigma_2(1-r^2)^{\frac{1}{2}}} \circ e^{-\frac{1}{2(1-r^2)}\left[\left(\frac{X_1-\overline{X}_1}{\sigma_1}\right)^2 - 2r\left(\frac{X_1-\overline{X}_1}{\sigma_1}\right)\left(\frac{X_2-\overline{X}_2}{\sigma_2}\right) + \left(\frac{X_2-\overline{X}_2}{\sigma_2}\right)^2\right]}.$$
(2)

The probability that a random point (X_1, X_2) will lie in any region R of the $X_1 X_2$ plane is obtained by integrating the density over that region,

$$P(X_1, X_2) \text{ in } R$$
] = $\iint_R f(X_1, X_2) dx_2 dx_1$ (3)

The points (X₁, X₂) that have a constant probability describe an ellipse.

It was found that the ellipses of constant P represented the data reasonably well. That is, where curve, P = 0.50, was computed, it was found that about one-half of the experimental points were within the ellipse.

It was of interest to design a means of testing any given handlift analysis against the two hypotheses: (1) that the results belonged to the specified handblank class, and (2) that the results belonged to the specified class of weapon firings. The mean vectors of the handblank and weapon classes will be designated μ and ν , respectively; and their covariance matrices will be designated μ and τ . The BVN densities associated with the two classes in question will be designated f_h , for handblanks, and f_w , for weapons (cf. Eq. 2).

The $P(X_1, X_2)$ in R] (per Eq. 3) can be computed with reference to either f_h or f_w , and the resulting P_h and P_w will, of course, be different. There will be some X results that will clearly belong to the handblank class and some that belong to the weapon class. However, there will be an area, called the critical region C, where the decision is more difficult.

The critical region may be defined as the set of points that satisfy the inequality:

where k is chosen so that the probability α of rejecting hypothesis (1) when it is true is a specified number (e.g. $\alpha = 0.01$). There will then be a corresponding probability β of accepting hypothesis (1) when it is false.

In terms of Eq. (4), α is computed by integrating f_h over C, while β is unity less the integral of f_w over C.

Despite the simplicity with which the problem is stated, it is difficult to solve. The problem is transformed so that C is determined by the inequality

$$rX_1^2 + sX_1^2 < c + k$$
,

where r, s, and c are functions of μ , ν , z, and T, and it is found that there are four cases:

- 1. C is interior of ellipse (where r > 0, s > 0)
- 2. C is exterior of ellipse (where r < 0, s < 0)
- 3. C is between branches of hyperbola having a horizontal orientation (where r < 0, s > 0), and
- 4. C is between branches of hyperbola having a vertical orientation (where r > 0, s < 0).

The first two cases may be treated with the aid of relevant tabulations. However, no tables exist to aid in the treatment of the latter two cases. As a result a great amount of effort has been expended toward the definition of critical areas for the classes treated in the experimental program. A computer program (BVN), was written to carry out necessary numerical integrations to solve these difficult cases.

The results of the calculation of the error probabilities lpha and eta are given in Table 24.

Table 24
ERROR PROBABILITIES FOR GUNSHOT RESIDUE DECISION PROCEDURE

<u>H₀</u>	$\frac{\mathtt{H}_1}{}$	<u>k</u>	<u>α</u>	<u>B</u>	Critical Region
A handblanks	0.22 rev	3. 125	0.014	0.751	Hyperbolic (partial, $\gamma = 11.88$)
A handblanks	0. 22 auto	1.563	0.007	0.519	Hyperbolic (partial, $\gamma = 3.34$)
A handblanks	0.25 auto	11. 25	0.011	0.046	Elliptic (interior)
A handblanks	0.38 rev	1.563	0.008	0.483	Hyperbolic (partial, $\gamma = 40.49$)
A handblanks	0.45 auto	20.312	0.008	0.001	Elliptic (interior)
B handblanks	0.22 rev	-14.063	0.010	0.465	Hyperbolic (partial, $\gamma = 2.55$) vertical
B handblanks	0. 22 auto	-15.63	0.010	0.662	Hyperbolic (partial, $\gamma = 1.12$)
B handblanks	0.25 auto	-20.188	0.013	0.842	Elliptic (interior)
B handblanks	0.38 rev	-10.938	0.011	0.828	Hyperbolic (partial, $\gamma = 1.73$)
B handblanks	0.45 auto	-10.938	0.006	0.840	Hyperbolic (partial, $\gamma = 2.46$) vertical
C handblanks	0.22 rev	15.741	0.007	0.819	
C handblanks	0.22 auto	12.5	0.010	0.704	Elliptic (exterior)
C handblanks	0.25 auto	28. 125	0.015	0.007	Elliptic (interior)
C handblanks	0.38 rev	ú. 0	0.005	0.542	Hyperbolic (partial, $\gamma = 2.74$)
C handblanks	0.45 auto	25.0	0.006	0.001	Elliptic (exterior)
D handblanks	0.22 rev	•	0.009	0.954	Elliptic (interior)
D handblanks	0. 22 auto	-2.344	0.011	0.965	Hyperbolic (partial, $\gamma = 1.32$)
D handblanks	0.25 auto	-8.339	0.010	0.436	Elliptic (interior)
D handblanks	0.38 rev	-3.486	0.010	0. 999	Elliptic (interior)
D handblanks	0.45 auto	2.643	0.013	0. 263	Elliptic (interior)

4. 4 CONCLUSION

With the successful interpretation of data associated with the handblank and weapon classifications to achieve probability tables, the chief goal of the program with respect to gunshot residues has been met. The TNAA method has been established as the most useful tool for the criminalist who wishes to establish whether or not a person has fired a weapon, since the method now far surpasses any other in its foundation of experimental data and the confidence with which case data may be interpreted.

Results that clearly belong to the class of handblanks or to the class of firings, and results for which less clear-cut interpretations are possible, are easily discerned in Table 23. The BVN treatment defines the decision procedure in the less clear-cut cases, and assigns error estimates to the two possible decisions, as shown in Table 24.

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5. BULLET IDENTIFICATION

5.1 INTRODUCTION

Bullet identification methods are of importance to criminalists, and their chief tool - striation comparison - is sometimes inapplicable because of bullet impact damage, use of a smooth bore weapon, or some other reason. Therefore, it was decided to explore the possibility that NAA might disclose definitive identification points in bullet leads.

The studies carried out were aimed at identifying those elements that might most usefully be identified by purely instrumental NAA in a large and varied sampling of bullets, defining the range of concentrations of those elements in the sample, and ascertaining the concentration uniformity of the elements within single bullets, single boxes of bullets, and single lots of bullets. Obviously, if the concentration variances within a box of bullets approached the variances over the sampling of different kinds of bullets, trace elements would not be particularly useful identification points. Rather it was hoped that great uniformity would be found within at least single bullets and single boxes of bullets while great diversity would be found among bullets of different lots.

Radiochemical separation techniques were not employed in the study due to constraints of time and money.

5. 2 EXPERIMENTAL AND RESULTS

Bullets were cleaned of any surface coatings. Organic lubricant was removed with toluene; jackets or metallic coatings were removed with dilute nitric acid, and oxide layers were removed from the lead by

scraping with a clean, stainless steel blade. Thin, ~ 0.5 mm, slices were taken for analysis; the weights of such slices depended on their areas and varied from ~ 10 mg to ~ 150 mg among the various samples.

The equipment used for irradiation and counting consisted of the previously described TRIGA reactor and NaI(Tl) and Ge(Li) equipped gamma-ray spectrometers. Both short (≤ 1 minute) and long (≥ 30 minutes) irradiations were used, with subsequent and commensurate counting periods.

Preliminary experiments with a wide variety of bullets known to be different by virtue of manufacture, lot number, or other feature used with short irradiation and counting cycles, included NaI(Tl) spectrometry. The element most commonly encountered and most precisely determined was antimony. Because the radioisotopes produced from this element usually dominated the gamma-ray spectra, only a few other elements were occasionally discerned (and these with poorer precision). Nevertheless, the data, which are shown in Table 25, indicated encouraging differences among different bullets.

Intrabullet uniformity of antimony concentration was explored by analyzing consecutive slices taken from the outer parts to the center of bullets from three manufacturers and of a lead wire from which bullets are made. As can be seen in Table 26, the uniformity was found to be good.

When a multiplicity of bullets from each box of several kinds of bullets were analyzed, it was found that the intrabox uniformity was as good as the intrabullet uniformity. This is shown in Table 27.

Because the data in Tables 25, 26, and 27 were promising, an extended study was made in the present report period. A survey of criminalists showed that they most frequently encountered cases involving 0.22-caliber and 0.38-caliber bullets, with 0.32-caliber, 0.45-caliber,

Table 25

EXAMINATION OF BULLETS KNOWN TO BE FROM DIFFERENT LOTS

	Bullets	Concentration of Elements in Lead			
Caliber	Make	Sb, % W	Other, ppm		
0.45	Veneza (water 1)	4 00	5/00 5		
0.45	Krasne (reload)	1.00	5600 Sn; 15 Al		
	Hensley and Gibbs	2.80	10 Al		
0.45	Military	<0.007	11 Al		
0.38	Hensley and Gibbs	4.40	3400 Sn: 75 A1		
0.38	U.S. Cartridge Co.	<0.003	87 Ag		
0.38	U.S. Cartridge Co.	1.55	0.67 Ag		
0.38	Union Metallic (UMC)	0.082	2.2 Ag; 22,000 Sn; 2.2 As		
0.38	Remington-UMC	0.013	2.6 Ag; 16,000 Sn; 27 As		
0.38	Remington-UMC	1.76	4.1 Ag		
0.38	Remington-UMC	2.07	8.8 Ag		
0.38	Peters	1.94	7.6 Ag		
0.38	Peters	2.99	0.77 Ag		
0.38	Remington-Peters	0.92	3.6 Ag		
0.38	Remington-Peters	0.87	3.1 Ag		
0.38	Remington	0.85 ^a			
0.38	Western	2.59 ^a	,		
0.38	Western	3.00	2.6 Ag		
0.38	Western	0.044	11.5 Ag		
0.38	Winchester	0.076	16.0 Ag; 167 As		
0.38	Winchester	3.09	4.9 Ag		
0.38	Winchester	1.38	0.56 Ag		
0.38	Winchester	1.47	0.56 Ag		
0.38	Winchester	1.55	3.9 Ag		
0.38	Winchester	1.23	1.8 Ag; 31 As		
0.38	Federal	1.89	-		
0.30	Remington	1.20	12 Al		
0.30	Sierra	3.10	5.3 Al; 2100 Sn		
0.30	Nosler	2.20	6.5 Al		
0.22	Hornady	3.50	5.4 Al		
0.22	Lapua	1.22 ^b	1.1 Al; 10,400 Sn; <5 As		
0.22	Sears	1.26 ^c	1.3 Al; 1,800 Sn; 285 Asd		
0.22	Imperial	0.99 ^c	< 4 As		
0.22	Peters	0.87 ^C	345 As		
0.22	Remington	0.85 ^C	-		
0.22	Western	0.54	78 As		
0.22	Crossman	<0.02	39 Ag		

a Average of 10 bullets from a single box.

b Average of 50 bullets from a single box.

 $^{^{\}rm c}$ Average of 20 bullets from a single box.

 $^{^{}d}$ Average of 5 bullets from a single box.

Table 26

COUNT RATE DATA OBTAINED FROM MULTIPLE SAMPLES FROM SINGLE BULLETS (0. 38-Cal.) AND A BULLET WIRE (0. 22-Cal.)

	1	Specific Activit	ty, cpm/mg, of Ind	ividual Samples	
Sample	Norma (Wire) Section 1	Norma (Wire) Section 2	Remington (Bullet)	Western (Bullet)	Federal (Bullet)
1 (outermost)	5140	5346	1142.1	1138	1070
2	5721	5428	1069.6	1200	1054
3	5363	5573	1149.8	1195	1005
4	5235	5561	1085.5	1195	999
5	5560	5651	1155.1	1185	1040
6	5359	5461	1162.1	1155	1040
7	5545	55 22	1162.1	1195	1033
8	5681 (center)	5288	1108.4	1150	1060
9		5700	1103.6	1195	995
10	•	5332	1139.1	1163 (center)	1043 (center)
11		5650	1078.5	(0011002)	in the second second
12		5547 (center)	1124.2 (center)		
Avg.	5450	5505	1123.2	1177.1	1033.9
σ , $\%$ of value	±3.85	±2.50	±2.85	±1.98	±2.36

Net cpm in the Sb 122 0.564 Mev photopeak per mg of sample. Inasmuch as different counters and geometries were used, the average specific activities cannot be used to judge the relative Sb concentrations: e.g., from Table 25 Avg. Sb in 0.38 Western was 2.59%, while Avg. Sb in 0.38 Remington was 0.85%.

Table 27
UNIFORMITY OF ANTIMONY CONCENTRATION AMONG
BULLETS TAKEN FROM SINGLE BOXES

R	1:	1	م (+

		No. of Bullets	Antimony Concentration			
Caliber	<u>Make</u>	Sampled from Box	Avg. %w	Std. Deviation, %w		
0.38	Remington	10	0.85	±0.02		
0.38	Western	10	2.59	±0.10		
0.22	Lapua	50	1.22 ^a	±0.04		
0.22	Sears	20	1.26 ^a	±0.03		
0.22	Imperial	20	0.99	±0.04		
0.22	Peters	20	0.87	±0.08		
0.22	Remington	20	0.85	±0.04		

Also found: Sn: 1.04% in Lapua, 0.18% in Sears.

Al: 1.09 ppm in Lapua, 1.25 ppm in Sears.

0.44-caliber, 9 mm, 0.25-caliber, 0.357-caliber, and 12 gauge shotgun projectiles less frequently encountered. Therefore, a bullet sampling approximately corresponding to the survey results was taken as follows:

- 0.22 caliber 70 bullets from 21 different lots
- 0.38 caliber 72 bullets from 26 different lots
- 0.32 caliber 23 bullets from 10 different lots
- 0.45 caliber 6 bullets from 3 different lots
- 0.44 caliber 11 bullets from 3 different lots
- 9 mm 15 bullets from 3 different lots
- 0.357 caliber 9 bullets from 3 different lots
- 0.25 caliber 5 bullets from 1 lot
- 12 gauge 9 projectiles from 5 different lots

The initial 242 bullets among 75 different lots are listed in Table 28. In Table 28 all Remington, Remington-Peters, and Peters bullets are listed as "Rem"; all Winchester, Winchester-Western, and Western bullets are listed as "W-W"; Federal Cartridge Co. bullets are listed as "Fed"; and Olin-Mathieson bullets are listed as "Olin".

In the earlier work aluminum, silver, and (with lesser frequency) tin had been sometimes observed soon after a short irradiation. However, since the chief aluminum indicator radioisotope, ²⁸Al, is generated from silicon also, and since silicon is abundant in the environment, it was felt that ²⁸Al was of dubious use. Especially wherever a bullet impacted a dirty object, silicon could become embedded in the bullet or a fragment thereof.

Considerable effort was expended to permit silver to be determined in the presence of antimony. However, the precision of such determinations proved to be relatively poor. Tin could only be observed when antimony was at very low concentration in bullet lead.

Table 28
BULLETS ANALYZED IN EXTENDED STUDY

Identification Numbers

			Identification Numbers			
Sample	Mfg.	Caliber	Lot No.	Index No.	Box	Remarks
1 - 5	Rem	. 38 S &W	627N	,	1	
6 - 10	\mathbf{Fed}	.38 Special	FPC515KC	•	1	
11-15	W - W	. 22 LR	CD71			Cu coat
16-20	W ~ W	.38 S&W	9397YA5		1	Cu coat
21-25	W-W	. 22 Short	BL4			Cu coat
26-30	Fed	.22 LR	LF4JC		1	Cu coat
31-34	W - W	.22 LR	BK72		1	Cu coat
3 5-3 9	Rem	. 25 Auto	L15ZD		4	Cu coat
40-44	Rem	9 mm Luger	J23A		1	Cu coat
45-49	W-W	9 mm Luger	45BC51		4	Cu coat
50-54	W - W	9 mm Luger	33BF7	•	1	Cu coat
55-56	Rem	.38 Special	N06D	3841	1	Cu coat
57-58	Rem	.38 Special	P07G	3841	1	
59	Rem	.38 Special	P07G	3841	2	
60	Rem	.38 Special	P07G	3841	3	
61	Rem	.38 Special	P07G	3841	4	
62	${\sf Rem}$.38 Special	N06D	3841	2	
63	Rem	.38 Special	K29H	3841	1	
64	Rem	.38 Special	N06D	3841	3	
65	Rem	.38 Special	K29H	3841	2	•
66-67	Rem	.38 Special	M24R		1	
68 -6 9	W - W	.38 Special	3528BE6	3853P	. 1	Cu coat
70-71	W - W	. 38 Special	57BK7	38SMRP	1	Cu coat
72-74	\mathbf{Fed}	.38 Special	CS20KC	38A	1	Ou coat
75-76	Rem	.38 Special	RA5289	M41	1	Cu coat
77-78	Rem	. 380 Auto	021C	1239	1	Cu coat
79-81	Rem	.44 Rem Mag	H09HG23LD	4411	1	Half-jacket
82-84	Rem	.44 Rem Mag	H09HH05SD	· •	i	Half-jacket

			Identifi	cation Numbers		
Sample	Mfg.	Caliber	Lot No.	Index No.	Box	Remarks
85-87	Rem	. 44 S&W Spcl.	M11E-20P	4405	1	
88-90	Rem	.44 S&W Spcl.	M11E-20P	4405	2	
91-93	Rem	. 22 LR	Unknown	2224	1	
94-96	Rem	. 22 LR	Unknown	2224	2	
97-98	Rem	.22 LR	Unknown	2224	3	
99	Rem	.22 LR	Unknown	2224	4	
100	Rem	.22 LR	Unknown	2224	5	
101	Rem	.22 LR	Unknown	2224	6	
102	Rem	.22 LR	Unknown	2224	7	
103	Rem	.22 LR	Unknown	2224	8	
104	Rem	.22 LR	Unknown	2224	9	
105	Rem	.22 LR	Unknown	2224 •	10	
106-108	Rem	.357 Magnum	11P-P09D	3578	1	Half-jacket
109-111	Rem	. 357 Magnum	11P-N14P	3578	1	Half-jacket
112-114	Rem	. 357 Magnum	11P-P09D	3578	2	Half-jacket
115	Rem	. 357 Magnum	11P-P09D	3578	3	Half-jacket
116	Rem	.357 Magnum	11P-P09D	35 7 8	4	Half-jacket
117	Rem	. 357 Magnum	11P-P09D	35 7 8	5	Half-jacket
118	Rem	.357 Magnum	11P-P09D	3578	6	Half-jacket
119	Rem	.357 Magnum	11P-P09D	3578	7	Half-jacket
120	Rem	. 357 Magnum	11P-P09D	3578	8	Half-jacket
121-122	Rem	. 357 Magnum	11P-K29ED	3578	Ì	Half-jacket
123	Rem	. 357 Magnum	11P-N14P	3578	2	Half-jacket
124	Rem	. 357 Magnum	11P-N14P	3578	3	Half-jacket
125	W - W	. 45 Auto	53-33BE01	45A1P	1	Copper cost
126	W - W	.45 Auto	53-33BE01	45A1P	2	Copper cost
127	W - W	. 45 Auto	53-33BE01	45A1P	3	Copper cost
128	Rem	.45 Auto	23PN13A	4504	1	
129-130	Rem	12 Gage Slugs	BN22N17	PS12RD	1	Rifled, 2 3/4 inch

Lot No.

AN22N17

R11BD81

Identification Numbers

3841

3841

3841

3841

Index No.

PS12RS

SX12PRS

Box

Remarks

Rifled, 2 3/4 inch

Rifled, 2 3/4 inch

	** **	in dauge brugs	ICT IDDOI	OATEI NO		miled, 2 J/T men
133	W - W	12 Gauge Slugs	R11BD81	SX12PRS	2	Rifled, 2 3/4 inch
134-135	W - W	12 Gauge Slugs	G62YL42	SX12PRB	1	00 Bucks hot
1 36	Rem	12 Gauge Slugs	AN12P18	PS12-3 3/4-00 Bk	1	00 Bucks hot
137	Rem	12 Gauge Slugs	AN12P18	PS12-3 3/4-00 Bk	2	00 Bucks hot
138-139	Speer	. 38 Special	910001	3748	1	
140-141	Speer	. 38 Special	03003	3758	1	Copper coat
143	Speer	.38 Special	03005	3752	1	Half-jacket
144	Speer	.38 Special	03002	3757	1	Copper coat
145	Rem	.22 Short	Unknown		1	
146	W - W	.22 Short	Unknown		1	
147	Rem	.22 Special	Unknown		1	Copper coat
148	W - W	.22 Special	Unknown		1	Super-X
149	W - W	.22 Special	Unknown	•	1	
150	Rem	.22 Special	Unknown		1	
151-152	Rem	. 32 Short R.F.	321A45		1	· ·
153-154	W - W	.32 S&W	Unknown		1	Copper coat
155-156	W - W	.32 S&W	Unknown		1	
157-158	Olin	.45 Ball	WRA22690		1	√
159 -16 0	Rem	. 38 Auto	242	1438	1	
161	W - W	.38 CNP	36-25TD3	W38CNP	1	
162	W - W	. 38 CNP	Unknown		1	
163-165	Rem	.38 Special	L19B	3841	1	
166	Rem	.38 Special	L19B	3841	2	

L19B

L19B

L19B

L19B

Sample

131

132

167

168

169

170

Rem

Rem

Rem

Rem

Mfg.

Rem

W-W

Caliber

12 Gauge Slugs

12 Gauge Slugs

. 38 Special

.38 Special

.38 Special

.38 Special

				Identific	ation Numbers		
	Sample	Mfg.	<u>Caliber</u>	Lot No.	Index No.	Box	Remarks
	171-173	Rem	. 38 Special	J09P	3841	1	
	174	Rem	.38 Special	J09P	3841	2	
	175	Rem	. 38 Special	J09P	3841	3	
	176	Rem	. 38 Special	J09P	3841	4	
	177	Rem	. 38 Special	J09P	3841	5	
	178	Rem	. 38 Special	J09P	3841	6	
	179-181	Rem	. 32 S&W	4R-M17C	1132	1	
	182-184	Rem	. 32 S&W	3R-J30N	1232	1	
	185-186	W - W	. 38 S&W	76-58RZ31	W38SWp	1	
	187-188	W - W	. 38 S&W	Unknown	K3872T	1	
	189-190	Rem	. 38 S&W	3TY25M127	3822	1	
	191-192	Rem	. 38 S&W	3TP27E165	2338	1	
7	193-194	Rem	. 38 S&W	9RG27N		1	
_	195-196	Rem	.32 S&W	IRJ10U	1132	ì	
	197	Rem	. 32 S&W	Unknown		•	
	198-200	W - W	. 32 S&W	112140	K32727	1	
	201-203	W-W	. 32 S&W	A4822	K3272T	1	
	204-205	W - W	. 32 Auto	886A8	32AP	1	Copper coat
	206-207	Rem	. 32 Auto	L22ED	2632	1	Copper coat
	208-210	Rem	. 32 Short colt	Z221	1632	1	Copper coat
	211-213	Rem	22 Short	J12P2D		1	
	214-216	Fed	.22 Short	524C	701	1	
	217-219	W - W	. 22 Short	XB82	SX225	1	
	220-222	W - W	. 22 Short	W K72	XP225	1	
	223-224	W - W	. 22 Short	W K72	XP2 25	2	
	225-227	Rem	. 22 Short	E1453F		1	
	228-230	W - W	.22 LR	XB12	XP22LR	1	
	231	W - W	.22 LR	XB12	XP22LR	2	
	232-234	W - W	.22 LR	WCC6262TB51	XP22LR	1	
	235-237	W - W	.22 LR	Y C2	SX22LR	1	
	238-239	Rem	. 22 LR	LOIR2B	2224	1	
	240-242	Rem	. 22 LR	W24A2B	1522	1	

7

In view of those considerations, the larger suite of samples was analyzed using the longer analysis cycle and a 35 cc Ge(Li) detector. This precluded observation of Ag, Al, and Sn, but it enhanced the possibility of seeing other, perhaps more useful, elements.

A few microscopic particles of silver remained on a few of the earlier samples analyzed, and in a few instances the gamma-ray spectrometer malfunctioned. However, analytical results were obtained for 233 samples. In almost every case antimony was determined. Copper was determined in a majority of samples, and arsenic was frequently determined. These results are given in Table 29.

5.3 DISCUSSION

The earlier results indicated that bullets were uniform in antimony on an intrabullet or intrabox basis to a standard deviation of about $\pm 3\%$ of the value. The more extensive study permitted further evaluation of intrabox uniformity plus an evaluation of intralot uniformity (where bullets from different boxes of a given lot were compared (see Table 28). The intrabox results are summarized in Table 30.

The intralot uniformity was found to be $\pm 4\%$ of the value for antimony (except where the Sb concentration was < 0.1% w), $\pm 23\%$ of the value for copper, and $\pm 44\%$ of the value for arsenic (see Table 31). Thus, by reason of intralot uniformity plus frequency and precision of determination, antimony is the most useful element for identification purposes. Copper is generally more useful than arsenic for the same reasons. No other useful elements were found by the instrumental NAA procedure in a significant number of samples.

Further insight into the results of the more extensive study is facilitated by listing the averaged results for each lot in order of antimony concentration; this is done in Table 32.

Table 29
ANALYTICAL RESULTS IN EXTENDED STUDY

Elementa

					11101	110116					
Sampli	e Sh. y ₀	Cu, ppm	As. ppm	Sample	Sb. %	Co. ppm	As, ppm	Sample	Sh. %	Cu. ppm	As. ppm
1	×	×	×	78	1.03	7 87	303	155	0.013	-87	'24
٠, ٤,	0.897	940	45	74	0.740	451	< 23	156	0.014	.3.38	43
3	0.890	958	79	×0	0.867	. 684	82	157	1.04	777	89
4.	0.904 0.854	988	30	A1	0.744	371	30	158	1.14	860	1.31
6		862	1.80	82	0.753	371	21	159	0.856	713	< 16
7	х -1. 65	467	750	83	0.814	59.1	38	160	0.864	682	< 33
8	1.66	466	758	84	0.850	730	95	161	0.995	796	198
9	1.65	433	778	#5	0.731	505	< 40	162	0.100	< 5	< 11
10	1.68	493	712 663	86	0.743	523	< 2.1	163	0.827	858	< 21
11	x	X Typ	x	87 88	0.797	708	74	164	0.815	681	1 39
12	0.641	1730	< 26	89	0.745	509	64	165	0.659	626	< 17
13	0.661	x,	< 24	90	0.761 0.834	146	46	166	0.800	649	45
.14	0.678	n. X	< 39	91	0. 711	516	36	167	0.823	709	104
15	0.676	1620	< 34	92	0.734	449 139	42	168	0.664	666	22
16	×	×	x	93	0.725	129	< 22 < 20	169	0.674	666	< 26
17	0.306	×	184	91	0. 799	458	69	170	0.820	652	123
18	0. 271	×	477	95	0.691	438	67	171	0.743	579	42
19	0.291	97	173	96	0.702	123	< 12	172 173	0.734 0.754	5.34	102
. 20	0. 307	×	352	97	0.723	1.19	16	174	0.718	625	87
21	, x ,	×	×	- 98	0.709	110	< 8	175	0.702	585	24
22	9.188	504	525	99	0.708	.425	93	176	0.682	472 485	. 113
23	0.185	492	492	1'00	0. 726	458	62	177	0.703	542	69
24	0. 189	n .	588	101	0. 76*	357	< 21	178	0.705	504	46 76
25	0.188	.439	502	.102	0721	127	< 11	179	0.732	760	< 9
26	1.88.	890	520	103	0.710	1 14	< 8	180	0.598	6 37	47
27	1: 85	500	520	104	0. 764	364	21	181	0.731	813	< 13
28.	1.89	500	500	105	0.706	109	< 8	182	0.730	563	12
29	1 88	×	585	106	0.670	770	< 1.3	183	0.719	573	14
30	1.88	440	500	- 107	0.688	809	22	184	0.748	662	< 45
31	0.785	145	141	108	0.674	668	16	185	2,48	167	176
32	0.754	356	180	109	0.663	530	< 7	186	2.43	216	178
33 34	0.751	63	35	110	0.729	848	113	187	2.42	759	427
35	0.803	166	103	. 111	0.628	509	18	188	2.51	740	296
36	0.852	500	48	.112	0.65H	788	< 18	189	1.45	217	40
-37	0.948	890	278	113,	0.657	737	< 14	190	1.30	124	52
38	0.962 0.771	562 710	298	-114	0.657	795	< 21	191	0.905	547	109
39	0. 781	434	102	115	0.665	664	< 14	192	0.665	423	43
40	0. 928	1050	66	116	0.682	831	19	193	0.753	892	202
41	0.980	1220	x 131	117	0.690	720	< 22	194	0.747	808	72
42	1.03	.670	680	118	128	553	< 10	195	0.963.	546	288
43	0.976	760	660		0. 722	837	< 19	196	0.975	507	178
44	1.02	840	780	120 121	0.696	832	12	197	×	×	×
45	0.0394	13	< 5	122	0.968	742	304	.198	1.22	1 47	3567
46	0,0646	40	< 7.	.123	1.02 0.674	895	268	199	1.13	1.35	3470
47	0.0337	14	10	124	0.676	552 520	< 13	200	1.17	108	3569
48	0.103	59	12	125	0.063		< 14 5.7		2. 26	65	· < 29
49	105.0	99	< 9	126	0.035	16			2.13	140	
50	0.124	32	< 7	127	0.037	15	< 2 < 2		2.16	56	< 14
51	0.0138	.8	< 5	128	1.02	263	240		0,598	12	1480
52	0.111	32	21 .	129	0.00096	2.8	< 2		×	×	×
53	0.0156	10	< 6	1 30	0.0024	5. 4	₹2		×	×	×
54 .	0.121	27 .	< 10	1.31	0.0011	2. 3	<2		x 0. 581	x · 1047	×
55	0. 724	711	< 13	132	0.0368	3. 7	. < 5		0.540	948	66 89
56	0.644	769	< 24	1.3,1.	0.0344	2. 5	< 6		0.580	1079	113
57	0.715	958	< 14	134	0.638	36	33		0.764	920	46
58	0.701	1020	< 16	1.35	0.648	46	< 18		0.736	393	143
. 59	0.736	1070	< 17	1 36	0.729	997	50		0.744	246	59
- 60	0.691	800	< 12	1 37	0.745	941	68		0.767	798.	74
6t.	0.930	×	23	1,38	2.63	796	592		0.780	773	183
62	0.718	704	< 16	1.39	2.61	HO5	517		0.767	768	61 -
61	0.751	716	153	1 40	3. 24	236	67 t		0.690	5.3	< 29
.64 65	0.667	795	< 23	1.41	3. 22	274	633		0.570	56	< 8
- 66	0.728	581	21	142	3. 25	258	694		0.581	40	< 15
67	0.744	625	< 12	143	0.00041	8. 8	< 3		0:556	75	< 17
68	0.736	617	29	144	0.516	446	91	155	0.565	114	42
60	2. 35 2. 26	336	391	1.15	0.719	728	42		0.559	46 .	< 17
.70	0.583	782 200	545	146	0.680	76.6	< 23	223	0.534	70	< 40
. 71	0.583		< 25	1-47	1. 79	836	138	224 (3. 6.30	104	< 20
72	0.424	105 57	30	148	2. 39		, < 17		7.743	767	<23
.73	0.421	47	25	149	4. 50	329	. 46		741	751	16
74	0.430		18	£50	1.67	258	383		7 31	729	35
75	0.929		<.27 1010		2. 52	478	< 42	226	. 22 -	272	75
76	0.857	# 1	955		2. 52	445			. 27	344	21
77	1.01	786			0. 900	42	23		. 62	271	149
			367	154	0. 822	298	47	231 1	. 24	440	< 19

a Value not obtained

Table 30

INTRABOX RESULTS

MEAN VALUES AND STANDARD DEVIATIONS^a

Samples	Sb, %	Cu, ppm	As, ppm
2-5	$0.89 \pm .03$	940± 50	85 ±67
7-10	$1.66 \pm .01$	445± 35	730 ± 50
12-15	$0.67 \pm .02$	1680 ± 80	<3 9
17-20	$0.30 \pm .01$	97(S. V.)	350 ± 130
22-25	$0.188 \pm .001$	480 ± 30	530 ± 40
26-30	$1.88 \pm .02$	580 ± 220	525 ±35
31-34	$0.77 \pm .03$	182 ± 120	114±62
35-39	$0.86 \pm .09$	618 ± 180	158 ± 110
40-44	$0.99 \pm .04$	910± 200	560 ±300
45-49	$0.09 \pm .07$	45± 32	9 ±2
50-54	$0.08 \pm .06$	21± 12	10 ± 7
55-56	$0.68 \pm .05$	735 ± 35	< 24
57- 58	0.71 .01	990 . 4.0	< 17
66-67	$0.739 \pm .004$	650± 35	20 = 15
68-69	$2.30 \pm .06$	560± 310	470 ±110
70-71	$0.62 \pm .06$	155 ± 65	20 ± 15
72-74	$0.425 \pm .005$	51±6	22 ±5
75-76	$0.90 \pm .05$	620(S. V.)	985 ± 35
7 7- 78	$1.02 \pm .01$	787± 1	335 ± 45
79-81	$0.78 \pm .07$	500± 160	56 ± 37
82-84	$0.81 \pm .01$	560 ± 180	50 ± 40
85-87	$0.76 \pm .04$	580± 110	74(S. V.)
88-90	$0.78 \pm .04$	490± 40	49 ± 12
91 - 93	$0.72 \pm .01$	240± 170	42 (S. V.)
94-96	$0.73 \pm .06$	340 ± 190	68 ± 1
97-98	$0.72 \pm .01$	125± 20	16(S. V.)
106-108	$0.68 \pm .01$	750± 100	19 ± 4
109-111	$0.67 \pm .05$	660± 260	65 ± 64
112-114	$0.66 \pm .01$	770± 30	< 21
121-122	$1.00 \pm .02$	820± 110	255 ±25
129-130	$0.002 \pm .001$	4.1 ± 1.8	< 2
134-135(6)	$0.63 \pm .03$	40± 13	< 40
	$0.71 \pm .02$	1040 ± 40	70 ± 18
137-(3)	$0.76 \pm .04$	924± 18	82 ± 12
and the second second	$2.62 \pm .01$	800± 7	555 ± 53
140-141	$3.23 \pm .01$	255± 27	650±27
	$2.52 \pm .00$	460± 24	< 68
153-154	$0.86 \pm .06$	195± 140	60 ± 52
155-156	$0.016 \pm .004$	210± 170	33 ± 14

Table 30 (Continued)

Samples	<u>Sb, %</u>	Cu, ppm	As, ppm
157-158	1.09 ±.07	820± 57	110±30
159-160	$0.86 \pm .01$	700± 20	< 33 →
163-165	$0.77 \pm .09$	720± 120	60 ± 70
171-173	$0.74 \pm .01$	580± 70	77 ± 32
179-181	$0.69 \pm .07$	740± 90	22 ± 18
182-184	$0.73 \pm .01$	600± 55	13 ± 1
185-186	$2.45 \pm .03$	190± 36	177 ± 1
187-188	$2.47 \pm .06$	750± 14	365 ± 90
189-190	$1.38 \pm .10$	170± 65	46 ± 8
191-192	$0.79 \pm .19$	485± 90	76 ± 47
193-194	$0.75 \pm .01$	850± 59	137 ± 90
195-196	$0.97 \pm .01$	526± 35	233 ± 78
198-200	$1.18 \pm .06$	141 ± 8	3520 ± 70
201-203	$2.18 \pm .07$	87± 46	
208-210	$0.51 \pm .02$	1030± 97	89 ± 24
211-213	$0.75 \pm .01$	350 ± 130	83 ± 53
214-216	$0.77 \pm .01$	780 ± 28	106 ± 67
217-219	$0.58 \pm .01$	50± 8	< 20
220-222	$0.56 \pm .00$	78± 34	20 ± 18
223-224	$0.58 \pm .07$	87± 34	< 2.0
225-227	$0.74 \pm .00$	750 ± 20	25 ± 14
228-230	$1.24 \pm .03$	310 ± 70	82 ± 65
232-234	$1.19 \pm .12$	167± 20	55 ± 33
235-237	$0.62 \pm .08$	38± 24	16(S. V.)
238-239	$0.78 \pm .06$	780 ± 110	37(S. V.)
240-242	$0.71 \pm .01$	474± 14	70 ± 6

Based on variation between samples. When only one result available, this is indicated by (S. V.) and no standard deviation is given.

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Table 31
INTRALOT, INTERBOX RESULTS

Mean Values and Standard Deviation

						EVIACION	
Samples	Bullets	Lot No.	No. of Boxes	Sb,	%	Cu, ppm	As, ppm
57-61	Rem, .38 Special	P07G	4	0.76	±.1]	953 ±140	lim.
55,56,62,64	Rem, .38 Special	N06D	3	0.69	±.03	747 ±47	lim.
63,65		K29H	2	0.74	±.02	650 ±95	87 ±93
85-90	Rem, .4458W	M11E-20P	2	0.77	±.05	535 ± 110	47 ±17
91-105	Rem, .22LR	Unknown	10	0.73	±.02	270 ±130	30 ±2 6
106-108, 112-120	Rem, .357 Magnum	11P-P09D	8.	0.69	±.02*	740 ± 100	17 ±4
109-111, 123, 124	Rem, .357 Magnum	11P-N14P	3	0.67	±.01	580 ± 74	33 ±45
125-127	W-W, .45 Auto	53-33BE01	3	0.045	±.016	15 ±1	lim
132, 133	W-W, 12 Gauge Slugs	RIIBD81	2	0.0356	±.0017	$3.1 \pm .8$	lim
136, 137	Rem, 12 Gauge Slugs	AN12P18	2	0.74	±.01	9 69 ±39	69 ±27
163-170	kem, .38 Special	L19B	- 6	0.76	±.07	678 ± 30	63 ± 40
171-178	Rem, .38 Special	J09P	6	0.71	±.02	525 ±50	68 ±30
220-224	W-W .22 Short	WK72	2	0.57	±.04	82 ± 27	lim
228-231	W-W, .22LR	XB12	2	1.24	±.02	380 ± 97	66 ± 61

Table 32
75 LOTS ORDERED BY ANTIMONY CONCENTRATION

Order	Lot		•		• •	,
No.	(or Samples)	Cal	CL M	Cu. Linux		200
140.	(or Samples)	Cal	Sb, %	Cu, ppm	As, ppm	Mfg.
1.	03005	. 38	. 00041	8.8	< 3	Speer
2	AN22N17	12G	. 0011	2.3	<2	Rem
-3	BN22N17	12G	.002±.001	4.1±1.8	< 2	Rem
4	Unk (155-156)	. 32	.016±.004	210±170	33±14	W-W
5	R11BD81	12G	.036±.002	3.1 ± 0.8	<6	W-W
6	53-33BE101	. 45	.045±.016	15±1	< 6	W-W
7	33B F7	9mm	.08 ±.06	21±12	10 ±7	W-W
8	45BC51	9mm	$.09 \pm .07$	45±32	9 ±2	W-W
9	Unk (162)	. 38	. 11	< 5	< 11	W-₩
10	BL4	. 22	.19 ±.00	480±30	5 30 ±40	W-W
11 .	9397YA5	. 38	$.30 \pm .01$	97	350 ± 130	W-W
12	CS20KC	. 38	.43 ±.00	51±6	22 ±5	Fed
13	03002	. 38	. 52	446	9 1	Speer
14	WK72	. 22	.57 ±.04	82±27	< 25	W - W
15	Z221	. 32	$.57 \pm .02$	1030±97	89 ±24	Rem
16	XB82	. 22	.58 ±.01	50±8	< 20	W - W
17	88CA8	. 32	. 60	32	1480	W-W
1.8	YC2	. 22	.60 ±.08	38±24	16	W - W
19	57BK7	. 38	.62 ±.06	155±65	20 ±15	W-W
20	G62YL42	12G	.63 ±.03	40 ±3	40 ±33	W-W
21	11RN14P	. 357	$.67 \pm .01$	580±74	33±45	Rem
22	CD71	. 22	.67 ±.02	1680±80	< 39	W-W
23	11P-P09D	. 357	$.69 \pm .02$	740±100	17.±4	Rem
24	4R-M17C	. 32	.69 ±.07	740±80	22 ±1 8	Rem
25	N06D	. 38	$.69 \pm .03$	747±47	< 2 3	Rem
26	W24A2B	. 22	.71 ±.01	474±14	70 ±6	Rem
27	J09P	. 38	.71 ±.02	525±50	68±30	Rem
28	Unkn. (145)	. 22	. 72	7 30	42	Rem
29	Unkn. (91-105)	. 22	.73 ±.02	270 ± 130	30 ±26	Rem
30	3R-J30N	. 32	.73 ±.01	600±4	13±1	Rem
31	M24R	. 38	.74 ±.00	650±35	20 ±15	Rem
32	K29H	. 38	$.74 \pm .02$	650±95	87 ±9 3	Rem
33	E14S3F	. 22	.74 ±.00	750±20	25 ± 14	Rem
34	AN12P18	12G	.74 ±.01	964±39	69±27	Rem
35	J12 P2D	. 22	.75 ±.01	350 ± 130	83±53	Rem
36	9RG27N	. 38	.75 ±.01	850±59	137 ± 90	Rem
37	L19B	. 38	.76 ±.07	678±30	985±35	Rem
38	P07G	. 38	.76 ±.11	953±140	< 16	Rem
39	524C	. 2 2	.77 ±.01	78±28	106±67	Rem
40	BK72	. 22	.77 ±.03	182±120	114±62	Rem

Table 32 (Continued)

Order	Lot				7	
No.	(or Samples)	Cal.	Sb, %	Cu, ppm	As, ppm	Mfg.
	<u> </u>					
41	M11E-20P	. 44	.77 ±.05	535±110	47 ±1 7	Rem
42	H07HG23LD	. 44	.78 ±.07	500±160	56±37	Rem
43	L01R2B	. 22	.78 ±.06	780±110	37	Rem
44	3TF27E165	. 38	.79 ±.19	485±90	76±47	Rem
45	H09HH05SD	. 44	.81 ±.01	560	50 ± 40	Rem
46	Unkn. (153-154)	. 32	.86 ±.06	195±140	60 ±52	W – W
47	L15ZD	. 2 5	.86 ±.09	618±180	158±110	Rem
48	Z242	. 38	.86 ±.01	700± 2 0	< 33	Rem
49	Unkn. (146)	. 2 2	. 89	77	< 2 3	W - W
50	627N	. 38	.89 ±.03	940±50	85 ±67	Rem
51	RA5289	. 38	.90 ±.05	620	985±35	Rem
5 2	1RJ10U	. 32	.97 ±.01	526±35	233±78	Rem
53	J23A	9mm	.99 ±.04	910±200	560±200	Rem
54	36 25TD 3	. 38	1.00	796	198	W - W
55	11K-K29ED	. 357	1.00 ±.02	820±110	285±25	Rem
56	23PN13A	. 45	1.02	263	240	Rem
57	021 C	. 380	$1.02 \pm .01$	787±1	335 ± 45	Rem
58	WRA, 22690	. 45	1.09 ±.07	820±57	110±30	Olin
59	112140	. 32	1.18 ±.06	141±8	3520 ± 17	W - W
60	WCC626277351	. 22	1.19 ±.12	167±20	55±33	W - W
61	XB12	. 2 2	$1.24 \pm .02$	380±97	66 ±61	W - W
62	3TY25M127	. 38	1.38 ±.10	170±65	46 ±8	Rem
' 63	FPCS15KC	. 38	$1.66 \pm .01$	445±35	730 ± 50	Fed
64	Unkn. (150)	. 22	1.67	258	383	Rem
65	Unkn. (147)	. 22	1.79	836	138	Rem
66	LF4JC	. 22	1.88 ±.02	580±220	525 ± 35	Fed
67	A4822	. 32	$2.18 \pm .07$	87±46	< 30	W - W
68	3528BE6	. 38	2.30 ±.06	530±310	470 ±110	W - W
69	Unkn. (148)	. 22	2. 39	419	< 17	W - W
70	76-58RZ31	. 38	2.45 ±.03	190±36	177±1	W - W
71	Unkn. (187-188)	. 38	2.47 ±.06	750±14	365 ±90	W - W
72	Unkn. (149)	. 22	2.50	329	46	W - W
73	321 A45	. 32	2.52 ±.00	460±24	< 68	Rem
74	910001	. 38	2.62 ±.01	800±7	555 ±53	Speer
75	03003	. 38	$3.23 \pm .01$	255±27	650 ±27	Speer

The 75 lots can be divided into three groups - the 25 lots of lowest Sb concentration (Order Nos. 1-25), those of intermediate Sb concentration (Order Nos. 26-50), and the remaining lots. When this is done, the distribution of bullets in each group from the different manufacturers is found to be very nonuniform:

Order No.	Rema	<u>w-w</u>	Fed	Speer
1-25	7	15	1	2
26-50	21	3	1	
51-75	11	10	2	2

a Including Olin-Mathieson, the parent company of Remington.

It is important to ascertain the degree to which bullets can be uniquely determined, especially in the middle ranks of antimony concentration.

Assume that a bullet in question has the following concentrations (with average standard deviations): $0.75 \pm 0.03\%$ w Sb, 400 ± 92 ppm Cu, and 100 ± 44 ppm As. Assume 20 to define the distinctive range in each case. Then the ranges of interest are:

Sb: 0.69 - 0.81% w Sb

Cu: 216 - 584 ppm Cu

As: 12 - 188 ppm A

Among the 75 lots of bullets the following fall within the three simultaneous ranges of interest: W24A2B (rank 26), J09P (rank 27), unkn (samples 91-105, rank 29), J12P2D (rank 35), M11E-20P (rank 41), H09HG23LD (rank 42), 3TF27E165 (rank 44), and H09HH055D (rank 45) - over 10% of the 75 lots.

Exact statistical definition of the degree to which bullets may be distinguished from one another is rendered difficult because the three

population sample (neither normal nor log-normal). Rather the antimony concentrations are unnaturally distributed as a result of deliberate additions at selected levels of concentration; and the other two elements have unnatural distributions that are probably associated with antimony. Correlation of concentration ranking orders between antimony and copper, and between antimony and arsenic substantiate this latter assertion.

These correlation coefficients, R, are as follows:

Sb:
$$Cu - R = 0.38$$

Sb: $As - R = 0.55$

Both values indicate significant correlations among the set of 75 lots.

Thus the number (8) of indistinguishable bullet lots in the example of the preceding paragraph is not surprising.

Examination of Table 32 shows that 12 of the first 25 ranks, 2 of the second 25 ranks, and 18 of the last 25 ranks are uniquely identified by the three elements. While the second 25 ranks are highly biased by the similarity among the 21 Remington members of the group, the distinctiveness among the other two groups of 25 samples is not great.

The earlier experiments involved a smaller proportion of Remington and Winchester bullets, and the antimony values of 30 lots of bullets were much more distinctive than in the case of the later experiments. However, if one examines the six Remington, Remington-Peters, and Peters results of the earlier studies, it is found that they anticipate in miniature the later results (Sb values were 0.92, 0.87, 0.87, 0.85, 1.94, and 2.99%).

The preponderance of Remington and Winchester ammunition taken for the later study is consistent with the dominant market position of these companies. While a more ideal correspondence between the sample

selection and an individual manufacturer's share of the market might have been achieved, the over-all results would have been substantially the same, i.e., Remington's bullet uniformity and strong market position lend a marked negative aspect to the probability of distinguishing between two bullets by NAA.

Concurrent work with other evidence materials, such as paper and paint, has shown that significant confidence that accidental matching of different samples has not occurred usually requires the measurement of ≥ 5 elements. Had this information been available sooner, the attack on the subject of bullet identification would have been quite different in the final study, since the earlier study showed that instrumental NAA did not usually observe this many elements in bullet lead. Rather, post-irradiation radiochemical separations would have been recommended to improve the possibility of observing a larger number of elements in a smaller suite of samples.

Although the present work has not provided an adequate means of comparing bullet specimens, as had been hoped, it has defined the scope of work necessary to achieve the desired goal.

Based upon the present findings the alternative approaches to the task are attractive. One approach would follow the course of postirradiation radiochemical separations to discern and quantitate at least six elements in bullet lead. This possibility is highly feasible, since removal of interference from the dominant antimony radioisotopes would open the specimens to the full sensitivity of the very powerful NAA technique. The larger number of observed elements, each of which would serve as an identification point, would greatly improve the reliability with which different bullets could be distinguished. As a result, the matching of two specimens from a common source would have much greater credibility.

The alternative approach would involve the distinctive tagging of bullet leads with combinations of elements, in small amounts, that could be easily observed by instrumental NAA. Preliminary discussions with cartridge manufacturers indicate that this would be difficult to implement in a practical way. Nevertheless, if the importance of the task warranted it, the necessary alterations in manufacturing procedures, the tagging approach could be realized. With three tagging elements, each selected to have a certain individual concentration within potential concentration ranges covering three orders of magnitude, at least one million distinctive tagging codes could be devised. Clearly, within any reasonable time scale of interest, all lots of bullets manufactured in the United States could have a unique tag.

5.4 CONCLUSION

The number of identification points developed by instrumental NAA is too limited to provide a means of always discerning between bullets from different lots. Wherever two bullet specimens have concentrations of antimony and/or other measured elements (e.g., copper, arsenic) that are different, after allowing for intralot variations defined in this work, it is safe to conclude that the two specimens come from different lots. However, if the concentrations of Sb, Cu, and As are the same (again allowing for intralot variations), it is not safe to say that the bullets come from the same lot. Improvement of this situation requires an extended program to either (1) expand, with radiochemical techniques, the number of observed elements, or (2) implement a program of tagging bullets with unique concentration codes of added trace elements that are easily measured by instrumental NAA.

6. DISCUSSION AND CONCLUSIONS

The eight-year program has examined a wide variety of materials that can be suitably interrogated for forensic purposes. The array of materials examined, mentioned in the introduction, no doubt comprises only a fraction of all possible materials of interest; yet, based on discussions with criminalists, it is believed to contain most materials of major interest to criminalists. Certain exceptions have developed over the years, however. For example, the present drug problem across the land was not anticipated in the early years of the program; otherwise, a much greater variety of drugs would have been investigated. Indeed it is possible that one or more drugs would have been selected for intensive investigation.

The size of the program simply did not encompass the possibility of definitive investigations of more than a few materials. However, the work on gunshot residues and particularly on paint has demonstrated the wealth of information and the degree of confidence in interpreting analytical results that can be gained by the NAA method. The program's study of those materials have made it possible for the criminalist to use NAA to arrive at testimonial statements in cases involving these materials and to assign probability values to the truth of his statements. This is an important contribution to the field.

The study of paper was extensive enough to develop preliminary estimates of probabilities for various "fingerprints". It is clear that it would be of value to characterize this material at least as well as paints have now been characterized.

The study of bullet lead proved the only case in which an extended study showed the applicability of purely instrumental NAA to be more limited than had been initially indicated by preliminary experiments. At the present time, the application of NAA to the comparison of two bullet leads can show two samples to be different, if the concentrations of Sb, Cu, and/or As are significantly different; but it cannot show two samples to be the same in most cases. It is likely that this limitation can be overcome by implementing either radiochemical procedures or tagging.

All told, the program has met its objectives. NAA has been shown to be of real value as an investigatory tool. Further definitive efforts to characterize additional evidence materials would undoubtedly be worthwhile.

7. SUMMARY

Over a period of eight years the applications of neutron activation analysis (NAA) in the field of criminalistics have been investigated. In the earlier years the effort was concerned with outlining applications feasibility, and potentially useful applications were discerned among a wide variety of materials - including plastics, rubber, greases, oils, paint, glass, soils, paper, ink, hair, fingernails, wood, tobacco, drugs, water, whiskey, skin, marijuana, bullets, and gunshot-residues. The effort in this final report period has concentrated on the definition of the exact forensic applicability of NAA with respect to a few selected materials of major interest to criminalists; namely, gunshot-residues, paint, and bullets.

The requirement of potential applicability for most instances is directed toward the possibility that numerous elemental constituents of a material can be quantitatively measured by NAA and that the concentration patterns of these elements, which comprise an elemental "fingerprint", are highly unique for each source of the material. If the examination, by NAA, of 10-20 samples of a material from different sources shows each to have a reasonably unique "fingerprint", potential application may be said to exist.

The degree of applicability requires analysis of a reasonably large and representative sampling of the population of the material in question to define the probabilities that: (1) two samples from a common source will have different "fingerprints" and (2) two samples from different sources will have the same "fingerprint". In practice it is desirable to associate

the results of comparing two samples with probabilities that various given interpretations are true or false. For example, if the "fingerprint" of a paint sample found at the scene of a crime matches that of a paint fleck taken from the suspect, what is the probability that the suspect's sample did not come from the scene of a crime. Or, if the two samples do not have the same "fingerprints", what is the chance that the suspect's sample nevertheless came from the scene of the crime. In practice, this last question is usually trivial, but it should be asked.

In this report period, the population of bullets and paints have been characterized and the answers to the foregoing questions have been delineated for any particular analytical results. The statistical regimen applied to the population of paints has been shown to be applicable to the population of papers, also, and preliminary statistics have been stated for the interpretation of comparative paper analyses.

The treatment of gunshot-residues is somewhat different. Only two elements, Ba and Sb, are related by the NAA technique to the question of whether a person did or did not fire a weapon. These elements are determined in handlifts. A handlift is acquired by application of melted paraffin to the hand and removing the paraffin after it solidifies. Persons who have not fired a weapon usually have some of the elements on their hands, and it has been necessary to define the related probabilities of finding given amounts in their handlifts, which are called handblanks. Persons who have fired a weapon usually have increased amounts of Ba and Sb on their gunhand, and it has been necessary to define the related probabilities of finding given amounts under this circumstance. The interpretation of a given handlift analysis has been made possible by the application of appropriate bivariate normal distribution statistics based on the handblank and firing probabilities. The bulk of this work, both analytical and statistical, has been carried out in the present report period.

Because this is a final report, it has seemed appropriate to discuss the preliminary work of previous years concerning paint, bullets, gunshot-residues, and paper so that the reader can appreciate the sequence of steps in these studies.

The applicability of NAA to the comparison of paint specimens is found to be impressive. Using a Ge(Li) detector in postactivation gamma-ray spectrometer, an average of nearly 15 elements is measured in paints and it is found that the 10 most frequently observed elements provide a general sufficiency of identification points. On the average the chance that two different paints will have "fingerprints" matching to these 10 identification points is less than one in a billion. Furthermore the method can, with a high probability of success, identify two specimens of paints as being of the same kind (manufacturer and manufacturer's specification) but of different lots. In such cases the specimens match to five or six identification points (on the average different paints have only about one chance in a million of doing this), but differ significantly on several other identification points.

It is interesting that the distribution (of concentrations) of only one of the 15 most frequently observed elements, tantalum, can be described as Gaussian. It was necessary, therefore, to compute concentration probabilities of individual elements directly from the analyzed population sample comprised of 155 different paints. Inasmuch as the 10 most frequently observed elements did not include significantly correlated pairs of elements (for example, barium was included; but chromium, a constituent of the pigment, barium chromate, was not included), the probability of finding a given "fingerprint" could be obtained by a simple combinatorial method.

Although additional analytical work was not done on paper samples in this report period, the data gathered in previous years was examined;

and it was found that the statistical approach successfully applied to paints would work for paper, too. Thus, it should be very worthwhile to analyze a completely representative sample of the population of papers and obtain a complete set of statistics.

Bullet lead proved to be less definitively characterized by instrumental NAA than either paint or paper. This had not been anticipated in earlier work, but a representative sample of the population of bullets revealed that one major manufacture controls the level of antimony to an unexpected extent. Since antimony is the most useful of three elements routinely observed by simple, instrumental NAA in bullet lead, and since the other two elements (copper and arsenic) have concentrations correlated with that of antimony, the comparison of bullets by NAA is not always meaningful. Where two samples have significantly different concentrations of one or more of these elements, it may be said that the samples are from different lots of bullets. However, matching concentrations do not conclusively identify two samples as being from the same lot.

It is concluded that either postirradiation radiochemical procedures or bullet tagging techniques will be required to achieve the desired goal of completely definitive comparisons of bullet leads.

With the completion of sampling and analysis of nearly 600 hand-lifts, including both handblanks and firings, a sufficiency of data has been gathered to allow probabalistic statements regarding the results of analyzing a given handlift, where one each of a variety of occupations and weapon calibers are specified. For example, given a case involving a 0.38-caliber revolver and a suspect whose occupation is as an accountant, the Sb and Ba values of his handlifts (right hand and left hand) can be interpreted with respect to the probability that the suspect did not fire a gun.

If the results are associated with a very small probability that the suspect did not fire a gun, a judge or jury may consider this to be beyond

reasonable doubt. However, since this is not the province of this program, no definition of reasonable doubt has been attempted.

Altogether, the program's objectives have been met. A wide variety of materials have been shown to be potentially suitable candidates for useful NAA analysis for forensic purposes; and it has been shown that such materials can be characterized to such an extent by NAA that the criminalist can affix realistic statistical probabilities to the truth of alternative interpretations of a given analysis or analytical comparison.

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APPENDIX 1

OUTLINE OF ACTIVATION ANALYSIS, PRINCIPLES & METHOD
A NUCLEAR METHOD OF QUANTITATIVE ELEMENTAL ANALYSIS

1. Principle

Nuclear reactions, many of which produce radioisotopes, are promoted within a sample by bombardment with suitable radiation. Each radioisotope is definitively characterized by its decay properties and serves as an analytical indicator of the element from which it is derived. All else being equal, the amount of the radioisotope formed is a linear measure of the amount, in the sample, of the element from which it was produced.

A. The disintegration rate, A, of a given number, N^* , of radio-isotope atoms of a particular kind is related to species' half life, $t_{1/2}$, by the expression (where $\lambda = 0.693/t_{1/2}$):

$$A = N^*\lambda$$
 (1)

B. The value of N^* derived from the bombardment of N atoms of precursor species with a particular kind of radiation for a length of time t_i is given in the expression:

$$N^*\lambda = N\phi\sigma(1-e^{-\lambda t_i}) , \qquad (2)$$

where \emptyset is the flux of bombarding particles, and σ is the reaction cross section (energy specified).

C. The activation equation is derived by substitution of (1) in (2):

$$A = N\phi\sigma(1-e^{-\lambda t_i})$$
 (3)

2. Method

A known amount of sample material is sealed in a suitably inert container (usually polyethylene or quartz), subjected to bombardment of known parameters (activation), and the resulting radioactive analytical indicators produced from elements to be determined are quantitatively measured after the bombardment terminates. Decay of each relevant species

must be taken into account: the decay obeys the relation

$$A_1 = A_0 e^{-\lambda t}$$
, where

 A_0 and A_1 are disintegration rates at the beginning and end of time period t.

- A. Activation may be carried out with several devices:
 - 1. Nuclear Reactor: the most prolific source of the most generally useful bombarding particles thermal neutrons. Reactors also provide copious fluxes of high energy neutrons, which promote some useful endoergic nuclear reactions. Reactors are also characterized by stability and reliability of operation. In addition, they generally provide a larger volume of useful sample space than
 - 2. Other devices, such as isotopic neutron sources and particle accelerators. Isotopic neutron sources are characterized by much lower fluxes than reactors. Accelerators, which have certain restricted uses in activation analysis, are comparatively less stable and reliable.
- B. Measurement of radioisotopes is carried out with devices commensurate to the task. Since most radioisotopic activation products give rise to monoenergetic gamma rays, and since there is little duplication of gamma ray energies among the various activation products, the most useful technique is
 - 1. Gamma ray spectrometry. The gamma-ray spectrometer, comprised of a detector (either a high-efficiency, moderate-resolution sodium iodide detector or a high-resolution, moderate-efficiency lithium-drifted germanium detector) coupled to a multichannel pulse height analyzer, enables simultaneous qualitative identification and quantitative measurement of a multiplicity of gamma emitters. Less generally useful are

- Other devices, such as geiger counters, proportional counters, Cerenkov counters, and so forth, which nevertheless have important occasional uses for the measurement of radioisotopes that do not give rise to gamma rays.
- C. Radiochemical Separations are sometimes necessary when, by reason of comparatively large radioisotopic yield, elements of little interest interfere with the determination of desired elements. Chemical procedures are instituted after activation; thus, since the analytical indicators have been formed, addition of other material to the sample does not jeopardize the accuracy of the result by contamination.
- 3. Advantages of activation analysis relative to other analytical techniques are significant.

Since it is a nuclear method and involves penetrating radiations, and since the only handling of the sample prior to the formation of the analytical indicators is in its packaging for activation, it has the virtues of

- A. Minimal matrix effect, which enables one to address large or small samples with equal facility,
- B. Complete linear dynamic range from the limit of detection through major constituent concentrations, and
- C. Much less contamination potential than other techniques. Other advantages include
- D. Great sensitivity. Many elements can be determined in amounts of 10^{-11} grams or less, and the median sensitivity for 75 elements attainable with a typical nuclear reactor is 10^{-9} grams.
- E. Simultaneous determinations of a multiplicity of elements are achieved, since all elements are subject to activation during an irradiation and gamma ray spectrometry simultaneously measures a multiplicity of gamma emitters.

- 4. Use of activation analysis are found in every field of endeavor, hence are too manifold to even enumerate here. Typical examples include
 - A. Research in such fields as biomedical (study of trace elements in tissues, diagnostic measurements), geochemical (study of continental drift, history of the solar system and earth), criminalistics (using trace elements as material "fingerprints"), and pollution (studying sources and patterns of pollution):
 - B. Routine analysis of petroleum products, plastics, alloys, ores, tissues, crops, textiles, and other natural and manufactured materials for trace constituents and/or major constituents.

APPENDIX II

CLASS I. BLACK OR GRAY PAINTS (17 SAMPLES)

Sample	Object	Element, ppm (K denotes thousands of ppm)														
No.	Sampled	<u>Na</u>	Mg	<u>A1</u>	<u>C1</u>	ĸ	<u>Ca</u>	Ti	<u>v</u>	<u>Cr</u>	Mn	Fe	<u>Co</u>	<u>Ni</u>	Cu	Zn
38	Door (Ext.)	513	₹ . 3K	536	876	<99	180K	81.8K	2.25	977	23 2	<5.6K	<26	<18K	<1.6	17.3K
41	Wall (Int.)	424	<2.6K	2.91K	1.68K	<110	87K	115K	<3.8	202	0.33	<6.6K		<450	<1.6	1.2K
63	Door (Ext.)	3.2K	23.8K	14.3K	235	2.47K	218K		4.25	<54	79.2	<5.9K		<880	<4.3	221
65	Wall (Ext.)	3.0K	7.8K	4.0K	4.36K	1.69K	26K	59K	13.4	122	57.9	5.671		≪K	87.9	73K
84	Refrig.	409	<24K	65	174K	<64	<9.7K	<370	<3.3	303	57.7	13.6K		<3K	<2.0	299
127	Headboard	2.2K	200K	28K	28K	1.15K	42K	10.4K	<23	601	488	35K	523	<4K	<3.7	520
131	Auto.	38	<79	149	212	28	<6.1K	<40	<0.6	<73	2.8	<8.5K		<100	606	<48
143	Fan	311	<1.4K	827	827	181	746	21K	<5	161	11.4	<4.5K		<110	<2.2	349
147	Air Condit.	641	<4.3K	4.3K	1.49K	535	<8.0K	183K	4	<60	22.2	5.6K		<280	<2.8	4.9K
148	Air Condit.	751	<18K	27K	982	338	<8.0K	<29K	<35	1.42K	24.0	<4.4K		<330	<3.4	5.2K
157	Lathe	545	< 240	943	855	34.7	<1.3K	25K	<5	4.46K	22.0	23K	1.1K		580	1.2K
158	Lathe	<ik< td=""><td><1.0K</td><td>2.3K</td><td>631</td><td><1K</td><td><1.5K</td><td>129K</td><td><7</td><td>13.3K</td><td>17.3</td><td><17K</td><td>1.7K</td><td></td><td></td><td></td></ik<>	<1.0K	2.3K	631	<1K	<1.5K	129K	<7	13.3K	17.3	<17K	1.7K			
159	Lathe	1.9K	<3.7K	6.4K	3.8K	151	<1.6K	308K	<9	10.0K	257	51K	1.6K	Øκ	<10	4.0K
160	Milling Mach.	1.5K	<3.4K	6.2K	744:	<140	108	134K	<10	9.6K	207	51K	1.5K	≪ĸ	1.2K	1.7K
161	Electronic Cab.	542	<7.0K	54K	1.6K	<110	<1.8K	<7K	<36	13.0K	488	70K	2.0K	₹K	<3.1	1.7K
164	Fork Lift	307	<5.7K	7.5K	405	<70	882	163K	<3	148K	63	132K	2.3K	<420	₹.2	1.0K
192	Auto	83	<0.8K	57	236	<30	<1.4K	108	9	<39	2.3	<4.6K		<86	1.016	

CLASS I. (Continued)

				E1	eme.	ent	, pp	m (K de	note	s tho	usands	of	ppm)			
Sample No.	Object Sampled	Ga	As	Br	Sr	Мо	Ag	Cq	In	Sb	I	Ba	La	Hf	Ta	w	Au	I
38	Door (Ext.)	<1.3	<2.5	11.3	110	<14	<2	<26	<5.8	152	<15	2.49K	<1	1.48	<3	7.79	0.13	<3
41	Wall (Int.)	<1.4	<1.6	14.3	<16		<3	Q4		42	<15	45	4.2		<3	7.79	< 3	_
63	Door (Ext.)	<3.1	<26	9.5	83		Q 6	<50		<1.5	<14	108	<2.4		<2	<1.8	<5	<3
65	Wall (Ext.)	2.8	431	50.9	<300		<42	<66		57	<150	53K	5.7	<6	4	6.1	<8	<5 -′
84	Refrig.	<1.0	1.7	76.3	<68	<8	<4	56	<0.2	1.2	<42	<55	<0.6	-	<2	<0.6		<6
127	Headboard	Q.3	2.6	49.1	<130	<22	₹ 6	<40		4.9	<46	<69	9.2	_	2		2.5	Q
131	Auto.	<0.4	<0.3	15.2	<5		<6	<9		<0.3	<0.4	5.7		-	-	<2 ⋅	0.4	<4
143	Fan	<0.7	<0.5	10.9	•	<10	<7	<11	1.6	3.8			<0.5		~2	<0.4	~	<2
147	Air Condit.	<1.6	<1.4	89	67			<42		8.4	<0.8	26	2.3		<2	<0.6	5.8	<2
148	Air Condit.	6.2	<1.5	250	<14						<11	5.6K	<1.5	10.8	29	6.7	<5	<4
157	Lathe		<0.7					<10		76	<21	77	55	<3	<2	<2	<5	<4
158	Lathe	<1.0		44.5	<11			<19		3.3	5.8	159	<0.4	<5	8.5	<5	<3	<3
159	Lathe	<1.2	<0.9		<13		<11	<25	8.7		<1.4	328	<1.0	<9	49		<3	<3
-		<2.0	<1.3	20.6	<56		9.5	<56	<4.9	8.8	<9.8	1.8K	<1.7	5.6	33	155	1.6	<5
160	Milling Mach.	2.1	36	5.6	<46		<8	<31	<4.1	132	<11	397	<1.4	1.8	53	3.8	8.1	<3
161	Electronic Cab.	6.2	<0.9	38.6	<73	<14	<63	<25	7.9	<1	<34	87	<0.9	<9	71	<1	0.3	<3
164	Fork Lift	<1.0	<0.6	17.7	<15	<10	<10	<17	<3.5	28	< 28	1 6K	<0.7	<8	56	<1	<2	2
192	Auto.	<0.5	<0.4	47.7	<6	<15	<20	<19	<59	<1	c4 3	<3	c0 1	~			-3	-3

CLASS II. WHITE OR OFF-WHITE PAINTS (63 SAMPLES)

Sample	Object Sampled	Element, ppm (K denotes thousands of ppm)																
No.		Na	Mg	<u>,A1</u>	Cl	. <u>K</u>	Ca	Ti	<u>v</u> ,	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	As
25	Sail Boat	1.7K	<54	17K	2.2K	349		301K	<0.2	<63	145	<5K	126	<650	<10	7.0K	8.4	3.9
27	Ground Rail	659	<25	3.3K	63 ·	345		200K	3	<68	111	<5K	406	<150	<3	70K	<2	13
.30	Highway Rail	475	<94	2.2K	39	<95		183K	<0.1	<73	537	<5K	505	<450	<4	85K	Q.	3.7
31	Bridge Rail	2.4K	66 K	199K	177	574		<11	<10	<49	588	<5K	512	<480	<5	19K	43	<2
32	Light Post	1.1K	67K	286K.	203	845		<4	<4	47	102	<5K	667	<190	<3	1.0K	52	2.0
35	Door, House	802	<400	463	1.6K	<560	24K	8.8K	2	<1K	3.4	••	<66	<1K	<22	61K	<9	<43
40	Wall, House	203	3.4K	5.7K	771	153	68K	254K	<6	<52	27	<5K	291	₹70	<1	568	<1	<0.6
54	Wall	1.6K	28K	13K	579	2.3K	89K	230K	<10	<75	45	<8K	29	<410	<3	426	1.0	<1.2
55	Cabinet	189	5.4K	4.5K	253	39	81K	193K	<6	<78	900		174	<480	Q	902	<1	21
59	Wall	1.3K	13K	6.9K	406	2.5K	358K	129K	7.3	<65	57	<5K	8.5	<540	<3	348	2.7	<1.1
71	Refrigerator	2.3K	<3K	3.0K	21K	716		234K	<4	<48	239	<4K	<9	<940	<3	1.7K		0.6
72	Washer	512	<3K	2.6K	<1.2K	158	-,-	193K		<49	503	<4K	<10	<220	Q	341	<1	<0.6
73	Washer	406	<3K	2.0K		146		161K	<4	<47	5.7	<6K	<13	2.3K	<3	467	<2 ⋅	0.5
74	Washer	421	<4K	7.1K		95		200K	4	<29	9.2	<4 K	6.2	<190	Q	167	<1	3.7
78	Shelf	695	<4K	7.7K	<79	704	13K	203K	<9	<56	210	<5K	198	<1K	<3	4.3K		<0.9
79	Siding. House	717	<4 K	8.2K	100	<72	<12K	254K	<8	57	5.0	4.8K	<11	<170	2	<92	Š	<0.8
82	Door	901	<6K	42K	339	92 1·	<2K	151K	<29	<57	103	3.5K	69	<830	<3	1.9K	<2	<11
86	Refrigerator	890	<3K	2.2K	1.4K	247	<10K	182K	<4	<45	899	<5K	<10	<390	3	1.9K	₹.	2.2
87	Clothes Dryer	227	<4K	4.5K	554	153		241K		<43	7.5	<5K	<10	<140	¿ 2	83	<1	<0.4
88	Freezer	637	<4K	2.1K	1.6K	154		208K	4.3	<60	242	<6K	<14	≪ĸ	<4	2.4K	•	2.3
89	Clothes Dryer	219	<4K	4.8K	1.2K	51		223K		<40	4.0	<5K	<11	<150	⋖.	175	1.5	3.0
90	Freezer	384	<4K	3.7K	566	230		204K		<45	325	<4 K	5.4	<zκ< td=""><td><2</td><td>491</td><td><1</td><td><0.7</td></zκ<>	<2	491	<1	<0.7
91	Washer	105	<250	3.1K	612	<37		352K	<3	<37	7.3	<5K	7.2	<260	272	123	<1	1.1
92	Washer	119	<760	3.6K	53	<38		358K	<6	<35	5.9	<4K	20	<220	<2	77	0.8	1.2
93	Washer	907	<920	8.9K	652	<40		327K	<9	<36	1.5	<5K	<10	<120	35	51	0.7	2.6
94	Washer	93	<5K	8.7K	721	<38		305K	<9	<33	6.7	<4K	6.9	<130	<2	93	0.3	3.8
95	Washer	83	<4K	3.7K	1.1K	<42		359K	<6	<36	2.2	<5K	<10	<130	Q	65	<1	9.6
-36	Washer	151	<4K	3.7K	928	<51	+-	342K	<6	<35	9.8	<5K	5.8	<160	~	282	<1	8.4
97	Washer	220	<4K	6.0K	995	199		326K	4	<40	2.2	<5K	<10	<140	2	99	41	0.7
JĶ	Washer	206	<5K	6.7K	55	194		324K	<7	<20	0.6	<3K	<4	<5	4	<3	<3	0.6
.4	Refrigerator	b48	<350	3.3K	1.1K	245	7.3K	319K	<4	<49	13	38K	23	<430	_	431	Ž.	<1
100	Refrigerator	oot	<3K	3.4K	1.6K	317		300K	13	<70	34	<8K	35	<650		000	Ž.	à
101	Retrigerator	490	<5K	101	655	79		306K	49	<44	295	<5K	18	Øκ	- 43	5.6K	à	<1
102	Refrigerator	363	<4K		293	66	<8K	246K	<4	470	306	13K	20	∢κ	< 3	23K	1.7	10
103	Retrigerator	428	<4K	2.6K	308	165	<9K	279K	<4	<42	393	1.8K		⋖ĸ		593	٠ <u>٠</u> ٠	1.3
104	Refrigerator	407	<5K	2.7K	58	65		276K	<4	<27	287	4K	13	ØK	Q	395	<1	1.7
105	Wandow Sill	1.5K	<4 K	4.7K	1.1K	308	52K	263K	<6	<51	26	<5K	154	<700	4	3.3K	<3	2.2
100	Window Frame	3.0K	<49K	29K	2.7K	2.0K		178K		<50	41	<4K	289	<2K	<5	15K	6.0	2.0

Sample	Object		 -							otes t			, 01	201111		
No.	Sampled	Br	Sr	Mo	Ag	Cd	<u>In</u>	<u>5</u> b	1	Ba	La	Hſ	Ta	w	Au	Hg
25	Sail Boat	279	Q 1	<28	<18	<28	9.4	98	9.4	82	11	<3	2	<3	<6	67
27	Ground Rail	59	<7	₹25	<4	<10	<0.2	3.4	<0.2	<3	<2	<3	<2	à	<5	7
30	Highway Rail	9.3	<21	<26	<2	<30	2.2	1.3	<0.8	<8	<1	1.0	<2	<2	<4	<5
31	Bridge Rail	25	<13	<21	Q 2	<22	5.6	<2	<0.7	45	4.7	Q	<2	<2 ✓ 2	<5	4
32	Light Post	36	<7	<19	<20	<8	<34	2.1	<0.3	··	<2 ·	<2 '	<2	٠ و	0.2	<3
35	Door, House	<11	<130	<310	<1 i	<180	<4.1	36K	<5	161K	-	<32	₹i	<19	<0.7	<55
40	Wall, House	5.0	17	<8	<4	<12	<1.4	4.3	<6	<3	<1	3.4	<2	<1	<2	<2
54	Wall	25.	92	<19	<8	<34	<1.7	57	<5	79	1.0	2.9	59	21	<0.5	53
55	Cabinet	10	. 33	<10	<1	<16	<2.3	12	<9	19	2.1	0.5	<3	<1	₹	<2
59	Wall	15	125	15	Q 1	<33	5.3	84	<2	7.5K	2.7	0.8	52	19	<5	4
71	Refrigerator	348	135	<15	<6	<27	<5	37	<14	9.3K	<2	4.3	5.8	< <u>1</u>	0.1	· <3
72	Washer	40	· <9	<9	<3	<16	Q	78	<5	1.0	0.9	3.5	7.1	3.6	5.6	à
73	Washer	38	<10	27	<4	<45	<2	2.2	<9	234	<3	<3	5.6	Q	<5	<5
74	Washer	29	<8	. <6	<5	<11	<2	1.3	<8.	1.1	<0.5	2	<2	<1	0.4	Q.
78	Shelf	53	<36	<15	7	<27	<4	50	<10	2.4K	3.2	2.4	<2	<1	<4	<3
79	Siding, House	3.0	<8	<12	<7	<22	<35	0.7	<3	<4 /	<1	<2	<2	<1	<3	.<2
82	Door	12	<33	<16	<22	<28	7	22	<45	203	13	<3	23	7.0	<4 .	<4
86	Refrigerator	130	44	<12	<4	<23	35	2.6	14	2.7K	1.3	<3	13	<1	<4	<3
87	Clothes Dryer	35	<6	<8	<5	<13	<70	16	5.1	3.8	1.7	2.4	27	5.7	7.2	<2
88	Freezer	167	<44	<13	<5	<25	<6	6.9	<21 ⋅	2.7K	<i< td=""><td>1.8</td><td>4.6</td><td><10</td><td><4</td><td><3</td></i<>	1.8	4.6	<10	<4	<3
89	Clothes Dryer	143	<8	<9	<6	<17	Q	1.9	<6	11	<1	<3	4.3	0.8	<3	< 2
90	Freezer	134	<75	<10	<17	<17	<1	30	<24	96	<1	<3	6.2	1.7	1.0	Q
91	Washer	40	<12	<9	<10	<15	<81	12	6.2	41	0.6	4.4	8.1	2.3	2.6	Q
92	Washer	31	<10	<9	<10	< 1.5	<81	6.5	3.2	35	<1	4.8	4.9	1.7	<2	Q
93	Washer	31	<5	<8	<14	<11	<46	2.6	<1	5.7	<1	2	Q	<1	<2	2
94	Washer	40	<5	<8	<11	<14	<55	6.0	<2	<3	<1	2	· <2	<1	<2	<2
95	Washer	60	<6	<10	<9	<17	<70	7.0	3.3	3.1	<1	<2	<2	3. 0	7.7	<2
96	Washer	39	<7	<9	<10	<16	<91	5.5	2.5	3.2	<1	Q	< 2	1.3	<2	<2
97	Washer	94	<7	<12	<8	<22	2	17	<5	7.9	1.8	2.4	30	7, 3	0.1	Q
. 98	Washer	42	<1	<1	<10	< 1		11	<1	0.2	3.1	5.1	41	7.4	<18	Q
99	Refrigerator	106	18	<17	<6	< 32	<2	40	7.5	3.8	<1	2.6	26	10	0.1	<3
100	Refrigerator	17	62	<22	<6	<43	2	43	6.5	21	v2	2.5	32	22	<6	4 4
101	Refrigerator	125	<74	<15	<5	<29	<5	8.5	<11	5.4K	<1	7.Z	2	.<2	<4	<3
102	Refrigerator	90	<78	<16	<1	<27	<6	20	<16	22K	<1	6.1	Q	<2	<4	<3
103.	Refrigerator	40	<89	<12	<4	<21	<8	1.4	₹4	161	<1	4.0	~	<1	<3	Q
104	Refrigerator	38	<64	<9	<4	<16	<6	<1	<22	155	<1	3.2	4.2	<1	<3	Q.
105	Window Sill	39	108	<18	<5	433	0.8	35	<12	1.2K	<2	5.4		<2	<5	<3
106	Window Frame	46	<41	<22	Q2	<40	9	1.1	<26	80	8.3	<3	<2	<2	<5	4

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Sample	Object					Eleme	nt, p	pm	(K de	notes	thou	sands	of pp	m)				
No.	Sampled	Na	Mg	<u>A1</u>	<u>C1</u>	K	Ca	Ti	<u>v</u>	Cz	Mn	Fe	Co	Ni	Cu	Zn	Ga	As
124	Window Frame	343	<3K	2.6K	2.2K	342	123K	189K	<5	<56	88	<5K	131	<1K	<2	2.2K	· <1	<1
128	Bed Frame	825	<4K	6.8K	865	545	82 K	- 247K	<7	<56	152	<7K	87	<820	<3	120	2	<1
129	Folding Chair	206	<3K	2.4K	602	166	101K	178K	19	<72	24	<6K	107	<210	<2	16K	<1	<1
141	Chair	196	<750	2.1K	302	170	176K	88K	<5	<5 3	46	<5K	124	<750	<2	1.4K	<1	<1
142	Cabinet	723	<3K	16K	365	542	156K	151 K	<13	2.3K	88	<5K	176	<930	· <4	228	6.7	Q
152	Cabinet	1.5K	<15K	25K	805	3.6K	205K	160K	<23	<45	21	<6K	21	<480	<3	<130	5.9	<1
153	Door	1.6K	<16K	25K	1.4K	3.4K	197K	159K	<28	106	22	<5 K	30	<470	<4	<130	7.2	1.6
154	Drawer	1.7K	<16K	24K	565	2.0K	201K	149K	Q7	<42.	27	<5K	<11	<510	· <3	443	9.1	<2 ⋅
155	Cabinet	1.4K	<10K	3.8K	1.0K	1.0K	244K	145K	<14	<5 5	18	<5K	49	<490	<3	11K	· Q	<1
166	Door Frame	341	<4 K	5.9K	13K	548	125K	193K	<7	<48	28	5.4K	79	<410	<1	1.3K	<1	0.4
167	Wall	296	<4K	5.4K	15K	80	124K	193K	12	<57	25	<6K	111	<420	<1	573	<1	0.3
168	Door	120	<4K	4.6K	1.3K	<29	83K	247K	<7	<63	93	8.0K	82	₹290	<1	5.8K	<1	0.3
170	Wall	177	<6K	4.8K	1.1K	<32	82K	280K	<9	287	165	13K	138	<350	59	5.7K	<1	<1
171	Wall		<15K	18K	14K		83K	217K	<24	< 59	324	3.2K	4.5	<640	~	5.2K	13	
172	Wall	782	<16K	19K	<920	514	6 8K	198K	-27	<44	303	<6K	<12	<650	<2	3.2K	<1	<1
173	Desk	1.5K	<660	8.3K	951	1.1K	⋖ĸ	247K	<5	<66	30	<6K	139	<870	. <4	28K	. 🛾	19
175	Water Ski	2.1K	<4K	5.8K	5.5K	341	<2K	308K	<8	<1 ₹ઇ	79	<8K	176	<1.3K	<6	8.5K	<3	Q
179	Sink	781	<3K	4.2K	1.6K	<120	123K	176K	20	<58	43	<6K	99	√7 10	213	2.2K	2	<1
182	Step Ladder	656	<4K	7.5K	168.	101	<2K	302K	<9	182	4.2	<6K	67	<130 .	<2	231	<1	<1
183	Steel	373	. <3K	646	1.1K	156	89K	118K	<4	<110	10	<6 K	<13	<350	<4	66K	<2	<1
184	Table	429	<3K	4.0K	4.0K	258	14K	250K	<5	<160	139	<12K	124	<1.7K	<4	24K	Q	Q
186	Table	1.6K	<4K	8.2K	<290	378	39 K	158K	6.8	<120	63	<6 K	173	<1.5K	<4	39K	2	· <2
187	Bureau	728	<4 K	9.3K	757	493	⋖ĸ	255K	<10	88	10.	<5 K	109	<250 ·	<3	1.4K	2	<1
188	Chair ·	379	<5K	5.4K	72K	<1.1K	88K	209K	<10	<4.5K	13	<130K	<280	<3.8K	<36	<3.1K	<14	<34
1.40	Auto	350	<430	12 K	683	<60	<2K	318K	<11	<50	14	<6 K	458	<280	Ø	331	2.9	<1

Sample	Object			Ele	mei	nt, pp	m (F	< der	otes	thous	and	s of p	pm)			
No.	Sampled	Br	Sr	Mo	Ag	Cd	In	SЪ	I	Ba	La	Hī	Ta	w	Au	Hg
124	Window Frame	43	<37	<11	4	Q 1	<3	64	<4	17	5.4	<3	3.4		٠,	
128	Bed Frame	39	<31	<16	<10	₹29	4	57	<17	<16	~ ~ · · ·	4.6	31 36	4.8	<3	~
129	Folding Chair	46	104	<14	<7	Q 4	0.9	89	<8	930	13			9.5	<4	<3
141	Chair	9.4	114	<10	<9	<16	0.3	51	~	<16	12	3.6 2.7	15	3.5	4.4	<3
142	Cabinet	16	379	<28	<15	<51	0.1	477	4	8.1K			<2	<1 <2	<2	2
152	Cabinet	21	190	<18	<23	<32	<2 ∶	4.1	<5	63		14 <3	28	_	<6	<5
153	Door	46	122	<18	<19	<33	<3	5.9	<6	50	9.0 8.4		2	Q	<5	94
154	Drawer	30	1.14	<18	422	<32	<3	2.7	<10	49	7.1	<3	Q	2	1.1	50
155	Cabinet	25	96	<17	<13	<31	0.8	77	<14	53		-	<2 ⋅	Q	0.8	44
166	Door Frame	16	257	<11	<8	<18	0.3	19	<6	30	14	<3	9.5		0.2	17
167	Wall	19	263	<11	<9	<19	0.3	24	٧,	43	<1	4.0	20	4.4	<3	2
168	Door	18	<12	<10	₹	<17	0.3	40	<11		<1	2.9	21	3.7	<3	<2
170	Wall	21	<13	<11	<13	<19	0.3	39	<35	34	5.4	2.4	15	5.7	<3	71
171	Wall		<19	<16	₹4	Q 6	<2 ⋅ 3 ⋅ 3 ⋅ 3 ⋅ 3 ⋅ 3 ⋅ 3 ⋅ 3 ⋅ 3 ⋅ 3 ⋅		<46	Q 1	5.0	3.1	18	6.1	<3	<3
172	Wall	87	<19	<15	Q 3	Q 6	<1	2.0	<47	232		<3	14		<4	<3
173	Desk	17	<36	<19	<11	<33	<3	<2.0	<3	85	4.1	<3	,10	4.6	<4	8.3
175	Water Ski	87	<44	₹8	<12	<54	<4		_	1.0K	_	9.8	<3	<2	0.1	<4
179	Sink	37	810	<13	<8	<26	0.2	408	9	353	<3	<6	45	7.1	4	<5
182	Step Ladder	29	<6	<13	√8	<16	<61	54	<14	1.2K		3.6	2.2	1.4	4.9	<3
183	Stool	9.9	435	<21	<10	<30		41	2	24	<1	3.1	8.3	3.3	<3	<3
184	Table	50	147	~1 ~26	<9	<46	<3	3.0	<15	103K	20	0.5	11	4.7	<4	<4
186	Table	9.6	1.3K	<22			<1	244	<30	3.6K		5.4	12	5.7	<6	<5
187	Bureau	20	<10	<12	<16	<36	<1	134	<33	72K	5.6	2.2	<4	2.0	<5	4
188	Chair	<33	<440		<9	Q1	3.6	<1	<16	811	<1	8.4	~	<1	7.8	<3
190	Auto			<1.3K	<17	Q.5K	<1	91K	<67	<280	<18	<130	<8'9	<22	<3	©20
470	Auto	65	<11	<10	<17	<19	<86	0.7	2.4	9.4	<4	~2	0	<4	~3	0

CLASS III. BLUE, VIOLET, OR GREEN PAINTS (34 SAMPLES)

Sample	Object				Ele	ment,	ppm	(K der	otes	thou	sands	of pp	m)			
No.	Sampled	Na	Mg	Al	<u>C1</u>	ĸ	Ca	Ti	v	Cr	Mn	Fe	Со	Ni	Cu	Zn
201	Auto	258		17K	3.9K	44				<47	548	23K	1.3K	<2.5K	576	<1K
211	Door	1.7K	<830	3.1K	1.4K	975	69K	106K		256	41	<5K	80	<450	<5	<680
1	Bookcase	1.3K	<5K	4.7K	<3K	316	90K	243K	<10	<62	7.5	<6K	340	<1K	<3	1.0K
9	Auto	340	<5K	42K	1.8K	<60		<3K	<47	<56	1.6K	<6K	553	<6K	903	146
13	Auto	223	<5K	20K	2.3K	<36	<9K	<4K	<1	<44	26	7.9K	<12	<300	266	216
19	Auto	502		12K	7.6K	<5			<93	<89	532	<6K	651		2.0K	293
21	Auto	73	<1K	24K	1.5K	<36	<8K		<32	. <34	5.1	<4K	<10	<310	191	65
23	Auto	42	<8K	17K	1.9K	<40	<6K		<22	<36	27	<5K	<10	~20	268	<45
24	Auto	41	<8K	15K	1.9K	<40	<7K		<25	<35	25	<4K	<9	<400	95	<44
36	Bookshelf	196	491	1.2K	2.0K	<60	29K	47K	3.5	293	24	<8K	249	<970	<2	46K
39	Cabinet	255	<3K	2.6K	55K	<80	31K	170K	97	285	72	<6K	249		<2	8.2K
53	Wall	667	<5K	8.1K	1.2K	42	2.2K	246K	<8	756	11	12K	263	<290	<3	2.0K
56	Wall	261	<5K	8.2K	1.2K	72		254K	6.0	821	16	10K	243	<270	<3	595
67	Tool	168	<8K	1.8K	7.4K	98		- 57K		914	782	20K	653	<6K	2.2K	504
70	Table	4.2K	40K	6.6K	1.1K	2.5K		6.8K	64	153K	94	54 K	<10	<2K	<5	Q 20
75	China Closet	607	<460	1.0K	4.1K	401	<860	134K	5.7	256	24	<5 K	18	<690	816	12K
80	Door Jamb	802	7.3K	2.8K	4.8K	764	<2	51K	56	132K	52	<6K	218	<680	901	13K
81	Door Jamb	1.4K	<3K	1.9K	4.6K	822	Q	143K	<5	1.1K	53	14K	443	<780	<6	23K
83	Washer	196	<330	2.7K	2.3K	<36	<10K	159K		944	7.4	<5K	<11	<460	461	254
115	Auto	205	≪ĸ	30K	6.1K	<61	<50K	<480	<12	<81	113	<9K	1.1K	≪ĸ	3.0K	150
117	Auto	52	<2 K	40K	40	133	<56 K	<19K	<17	<62	3.6	<7K	<16	<130	541	191
119	Auto	114	<16K	7.9K	13K	113	<61K	<3K	Q 7	<66	263	<8K	<17	<860	535	190
120	Auto	103	<10K	11K	1.1K	<55	<68K	<2 K	<14	<59	238	<7K	<15	<711	468	<170
121	Auto	564	<11K	35K	1.9K	<37	<72K	<2K	<17	<60	308	<7K	<16	1.8K	<3	<79
144	Air Conditioner	462	<6K	41K	732	205	⋖ĸ	8.8K	<31	<5 t	13	<5 K	138	<230	<3	22K
145	Air Conditioner	209	<5K	55K	1.2K	211	<2K	<4K	Q 4	<49	9.7	<5 K	<12	<220	<5	<130
156	Cabinet	435	<25K	53 K	2.1K	870	35K	37K	<48	>77	104	59K	97	<680	440	6.3K
162	Work Bench	1.6K	<3K	4.0K	1.7K	<150	27K	51K	42	61K	184	130K	1.5K	<1.3K	455	612
165	Door Frame	743	<900	5.2K	1.3K	484	184K	146K	<7	<57	17	<6K	165	<470 ·	2	303
174	Desk	715	≪ĸ	7.3K	3.6K	315	<2K	205K	<9	16	12	<5K	257	<560	240	633
177	Water Softener	845	<7K	44K	5.1K	96	⋖ĸ	<4K	<31	<46	14	<5K	297	<740	440	510
180	Wall Heater	212	<4K	4.3K	622	61	⋖ĸ	212K	<7	<44	241	<7K	<11	≪K	<3	6.7K
191	Auto	68	≪ĸ	23K	22	40	<7K	<3K	<29	<40	3.4	<5K	<11	10	2.0K	<82
195	Auto	3.8	<15K	24K	2.1K	24	<5K	.27	Q 7	<37	23	3.7K	<10	<190	176	42

Sample	Object				Elen	nen	t,	ppr	n (K	denot	tes th	ousar	ids (of p	pm)			
No.	Sampled	Ga	. As	Br	Sr			Cd	In	Sb	I	Ba	La	ж	Τa	w	Au	Hg
201	Auto	1.2	<1	135	<1K	<13	01	<31	<1	<1	<110	-/-		_				
211	Door	<2 €	1	27	<170	<18			Q.	4.0	4.2	<65 55	<1	2	2	<1	<3	<3
1	Bookcase	~	<1	18	<34	<9		<16	<1	11	<73	76	Q	7.0	2	Q	5.0	
9	Auto	9.1	<1	32	24 0	:13	-	<28	<1	<1	<15	76 <89	<1	11	Q	<1	6.4	Q
13	Auto	5.1	<1	18	<10	<6		<13	<85	3.5	\12 \\(\frac{1}{2}\)	<4	<1	3.3	~2	<1	<3	Q
19	Auto	<9	<1	3.8K	<1	<3	<37		<140	<1	<3	<1	<1	<2	<2	<42	<2	<1
21	Auto	~1	<1	56	<10	<10		21	<81	<1	5.3	103	<1	<2	<3	<1	<81	<1
23	Auto	3.5	<1	12	<8	<8		<17	<91	<1	<3	662	<1	⋖2	<2∵	<1	Q	Q
24	Auto	3.6	< <u>1</u>	12	<14	ā		<16	<14	<1	<3	845	<1	<2	<2	<1	<2	~2
36	Bookshelf	~	4	15	<76	<22	_	<38	<3	489	4	170K	<1	<2	<2	<1	~	~2
39	Cabinet	<2	<2	22	161	<13		<22	<9	62	Q 7		<1 <1	<8	<5	3.5	<5	<5
53	Wall	2	<1	39	8.6	<15	4		<1	17	6.8	4.2K 307	<1	4.0	<3	3.1	<3	<3
56	Wall	7.1	<1	35	<11	<12	<8		Q.	6.5	<8 <8	192	_	<5	32	9.9	<4	<3
67	Tool	<1	Q	34	Q 20	<17	_	<38	<1	4.0	<19	125	1.2	2.0	33	9.0	<3	<2 ⋅
70	Table	<3	<3	15	<60	<31	_	<40	4	<1	<16	145	<1	<3	9.0	3.7	<3	<3
75	China Closet	<2	<1	149	40	<18	<6		5.1	51	<2 <2	39	9.2	<3	<4	~	<5	<5
80	Door Jamb	<2	Q	753	179	<25	<6		0.6	299	<16		3.8	2.7	12	<2	<5	<4
81	Door Jamb	<2	8.7	58	288	Q22		<40	0.6	60		5.5K	6.2	1.0	<5	<2	0.2	92
83	Washer	<1	<1	50	<15	<10		542	<1	9.1	<25	403	19	6.7	0.4	3.0	4.1	<4
115	'Auto	<1	<1	59	<59	<32	÷4	<64	<4	7.1 <1	6.7	846	<1	2.4	<2	1.4	9.2	Q
117	Auto	2.1	<1	56	<8	<14	2 6		<67	1.3	4	3.8K	<1	40	<3	<1	<6	<5
119	Auto	2.4	<1	76	<32	<38		<65	<4	<1.3	<2	25	<1	<3	<2	<1	<3	<3
120	Auto	2.8	<1	81	<29	<34		<57	<4	13	<10	695	<1	<3	<2	4.4	<7	<6
121	Auto	9.4	<1	63	<10	<10		<20	< <u>4</u>	13 <1	<14	672	<1	<3	<2	8.9	<6	<6
144	Air Conditioner	9.3	<1	43	<10	<19			<98	<1	<18	<10	<1	<3	<2	<1	<3	<2
145	Air Conditioner	9.5	<1	173	<10			<35	\90 <2	<1	2	22	<1	<3	<2	Q	<4	<3
156	Cabinet	15	3.4	27	130			<27	0.2	14	44	26	<1	<3	<2	<2 ⋅	<4	<4
162	Work Bench	<2	Q	20	<50			<47	≪8	656	<32	1.5K	31	<3	<2	Q	<4	361
165	Door Frame	<1	4	15	651	<15	<9	42	0.5	30	Q 5	<28	<2 ⋅	•	27	5.6	<6	37
174	Desk	Q.	<1	19	30	<13		Q 5			<2 ⋅	995	2	1.6	8.7	<1	<4	<3
177	Water Softener	5.7	<1	26	30 ⊘ 3		•	Q 9	Q	11	63	2.1	<1	<3	<2	<1	<3	<3
180	Wall Heater	<1	25	20	₹3 ₹76				<3	<1	<7	422	<1	<3	Q	<1	<6	<3
191	Auto	2.9	<1	14	<12			<18	<1	<1	<28	<46	<1		<2	<1	<3	Q
195	Auto	3.7	<1	1 4 22	<12 <8		Q1	38	<1	0.5	2.3	4	<1		<2	<1	<3	<3
- / -	******	3.1	~1	44	~8	<8	<19	~18	Q	<1	<7	743	<1	Q.	Q	<1	1.6	Q

CLASS IV. YELLOW, ORANGE, OR RED PAINTS (23 SAMPLES)

C1-	Object				Ele	ment,	ppm (F	denot	es th	ousan	ds of J	pm)				
No.	Sampled	Na	Мg	Al	Cl	<u> </u>	G	Ti	<u>v</u>	Cr	Mn	Fe	<u>Co</u>	Ni	Cu	Zn
11	Auto	212	< 2K	1. 4K	5. 9K	<72	<7K	825	3. 4	64K	7.6	<8K	<16	385	< 4	250
17	Auto	224		579	334	145	<9K	<4K	<10	11K	7.3	<6K	<12	< 400	< 4	82
26	Sail Boat		< 120	2. 3K		<200		110K-	<1	24K	1.0K	<7K	268	< 760	< 6	< 320
20	Auto	59	< 5K	3.1K		131	<6K	241K		167	2. 5	<5K	<10	< 361	< 2	2.8K
34	Signal Post	917	< 66	5. 9K		322		52 K	<i '<="" td=""><td>24K</td><td>130</td><td><4K</td><td>207</td><td>< 580</td><td>< 4</td><td>34K</td></i>	24K	130	<4K	207	< 580	< 4	34K
42	Cabinet	599	< 4K	5. 7K		357	62K	255K	<7	<55	46	<5K	269	< 340	< 3	1.2K
43	Auto	116	< 210	1 4K		< 70	< 850	< 300	3.9	50K	4.8	<5K	<12	< 220	< 3	867
51	Loose Wall	536	6.8K	3. 1 K		865	23K	44 K	17	3.0K	64	<8K	60	< 930	< 3	101K
61	Wall	3. 4K	32K	23K	963	2.3K	<20K	51 K	<9	1.0K	44	11K	6. 5	< 710	< 5	554
62	Wall	418	32K	2 3 K	753	1.9K	<20K	47 K	11	1.2K	50	8.8K	<11	< 780	< 5	622
69	Shelf	863	<3K	2. 3K	889	169		144K		<82	1.7	<7K	223	< 330	< 2	3, 2K
77	Chair	398	<2 K	946	6. OK	249	1. 2 K	1.5K	2. 1	<71	20	<8K	1.2K		< 2	< 90
107	Auto	198	< 5 K	2. 3K	3. 9K	< 100	<1.2K		<5	27K	3. 0 K	<5K	<11	< 14K	< 3	
125	Baby Crib	1 35	<3 K	3. 1 K		225	153K	198K	<6	3.6K	27	<6K	1 37	< 240	< 2	1.3K
126	Table	780	<3 K	3. 3K	1.5K	399	117K	158K	<5	825	21	<4K	44	< 350	< 3	1.4K
1 30	Sofa	473	<7 K	31K	1. 3K	362	<2K	<6K	< 27	212	625	87K	266	< 3K	< 6	475
163	Fork Lift	363	<3 K	8. 2 K	705	< 79	<2K	3. 3K	< 11	161K	3. 1	144K	2. 0K		< 3	< 82
176	Bed Frame	468	<3 K	3. 8K	1.2K	141	<2K	154K	<7	1.0K	56	11K	<11	< 650	< 5	749
181	Step Ladder		<3 K	2.6K	1.8K	221	8. 3K	9. 5K	31	42K	14	<6K	352	<1K	< 4	1.1K
185	Table	173	<3 K	1.4K	2. 2 K	840	9. 9K	1.7K	<5	<52	213	<5K	212	<2K	< 4	811
189	Auto	95	<57	2.9K		22	<2K	931	7.2	14K	6. 3	21K	<12	< 180	< Z	< 73
193	Auto	110	<4 K	13K	442	< 37	<2K	<960	<12	<43	9. 4	9. 1 K	3. 0	< 520	732	< 72
104	Auto	165	<3 K	4.5K		<49	<2K	249K	<9	50	18	<6K	45	< 210	< 2	1.9K

CLASS IV. (Continued)

Sample	Object					Eleme	ent,	ppm	(K de	notes	thou	sands	of p	om)				
No.	Sampled	Ga	As	Br	Sr	Мо	Ag	Cd	In	Sb	1	Ba	La	Hf	Ta	w	Au	Hg
11	Auto	< 2	< 2	209	< 45	9. 3K	< 4	< 75	< 5	2.8K	14	5. 3K	< 1	< 10	< 6	< 2	< 8	< 7
17	Auto	< 2	< 1	36	2. 2 K			57K	< 4	692	< 17	47K	< 1	< 2	< 5	< 1	< 6	2. 2K
26	Sail Boat	< 3	< 5	164	< 27	< 50	< 4	< 31	< 2	1.6K		692	< 2	< 7	< 5	< 3	< 0.1	
28	Auto		< 2	14	< 5	< 7	< 19	< 11	0. 1	8.8	< 3	3. 4	< 1	< 2	14	4.9		< 2
34	Signal Post	1.5	0.2	28	< 23	< 22	< 4	< 31	3. 7	0.3	< 1	538	< 2	< 3	< 3	< 2		< 4
42	Cabinet	< 2	< 3	107	45	< 31	< 10	< 52	< 3	27	< 18	457	< 2	< 3	<2	<2		<6
43	Auto	< 2	< 2	102	< 11	3. 1K		< 17	< 66	1.5K		4. 3K	< 1	< 8	₹5	< 1	< 7	< 5
51	Loose Wall	1.9	37	69	< 40	< 24	< 6	< 37	.5. 5	22	< 4	69	< 2	1.0		< 2	< 5	< 5
61	Wall	2.0		11	24	< 25	< 21	< 46	0.5	1.9	< 5	82	13	< 3	< 2	< 2	< 4	< 5
62	Wa!l	9.7	2.9	14	27	< 24	< 21	< 47	1.3	7.9	< 8	99	13	< 3	< 2	<2	< 4	< 5
69	Shelf	< 2	1.2	38	145	< 13	< 4	< 24	0.6	65	3.2		14	2.7	17	5. 1		
77	Chair	< 2	< 1	64	71	< 12	< 6	< 22	< 2	489	< 4	6. 2K	< 1	< 4	< 2	<1	< 3	< 3
107	Auto	< 1	< 2	-	< 640	2. 8K		< 37	< 0.5	468	< 37	839	< 1	< 4	< 4	<2	< 5	< 4
125	Baby Crib	< 1	< 1	53	74	< 12	< 8	< 20	< 98	54	<2	44	15	7.5	19		< 3	< Z
126	Table	< 2	1.3	14	37	< 12	24	<22	0.3	43	< 3	37	5.8		< 2	<1		< 3
1 30	Sofa	6.6		44	< 99	< 14	< 22	< 28	< 0. 2	1.7	< 51				< 2	< 1	< 3	< 3
163	Fork Lift	< 2	< 1	22	< 8	< 15	< 9	< 17	1.7	0.8	< 8	1.2K <7	11	< 3	18	<1	< 3	₹3
176	Bed Frame	< 2	< 2	132	<27	< 16	< 8	< 34						< 10			< 4	< 3
181	Step Ladder		< 1	51	< 39	7.0K		< 33	< 3	219	< 6	102	1.9		9. !	, 3.4 <2	< 5	< 4
185	Table	< 2	< 2	135	< 78	< 18			< 4	28	< 9	3.6K	< 1	< 4	< 4		< 5	< 4
189	Auto	< 1	< 1	53	< 7			< 36	< 1	< 2	< 23	< 50	3. 4		< 2		< 3	< 3
193	Auto	0.6		25	< 16	1.2K		< 11	< 44	320	< 2.3	< 5	< 1	< 4	< 3	<1.		< 3
194	Auto		< 1	111	< 16 9. 3	< 15 < 11	< 15 < 10	< 31 < 21	< 1 < 1	< 1 1.5	< 6 < 6	25 < 1	< i	< 2 4. 4		1 < 1 4.0	< 3 3.9	< 3) < 2

CLASS V. BROWN OR TAN PAINTS (19 SAMPLES)

Sample	Object	4.			E	Cleme	nt, p	pm (F	dend	otes the	usand	ls of	ppm)				
N-)	Sampled	Na	Mg	Ai	CI	к	Ca	Ti	ν	Cr	Mn	Fe	Co	Ni	Cu	Zn.	Ga
33, -	Trash Can	660	< 110	4.9K	31	260		171K	< 1	5. 8K	1. 4K	<6K	247	< 800	< 4	2.6K	<2
37	Doors	144	<2K		982	370	28K	9.3K	< 2	< 990	9.6		< 64	< 830	<17.	67 K	<7
52	Window Frame	1. 1K		3.7K		495.		210K	13	6.9K	28	39K	220	< 690	< 3	73K	1.5
57	Wall	269		399K	272	616		105K	< 31	659	126	14K	< 20 ·	< 560	< 2	2.0K	- 9.0
58	Wall	574	< 4K	4.2K	898	247-	71 K	228K	27	6. 0K	29.	< 4K	175	< 370	342	66K	1.6
60	Cabinet	1.2K	181.	26K	< 40	487	< 21 K	73K	< 6	< 80	129	6.6K	334	< 7.30	< 3	23K	11 .
64	Wall	4. 1K	< 6K	2.7K	< 4K	2.6K	31 K	1.8K	88	2. 2K	4. 4K	75K	206	< 26K	1.11		<4
66	Door	2. 9K	20 K	28K	741	807	120K	51 K	< 9	88	407	8.1K	47	< 2K	< 5	5.6K	15
68	Refrigerator	1.4K	< 3K.	951	. 3. 6K	437		87 K	< 4.	< 65	2. 9	< 5K	148	<1K	< 2	2.4K	<2
85	Door Fan	844	< 420	2.0K	2. 4K	226	< 11 K	146K ·	< 4	146	176	< 5K	< 11	< 86 K	< 2	< 48	<1
123	Chest .	1. 3K	< 570	5.2K	1.4K	951	13K	183K	< 3	205	. 43	< 9K	323	< 660	< 4	6. 2K	<2
146	Air Conditioner	196	< 25K	51 K	546	588	< 8 K	< 4K	< 16	202	35	19K	< 15	< 260	< 3 ⋅	1.1K	15
140	Door	453	<2K	1.0K	2. 4K	628	141K	27K	< 5	92	404	61K	419	< 3K	331	255	<1
150	Cabinet	. 2. 3K	< 8K	8.5K	1.3K	1.1K	13K	166K	< 10	1.4K	493	36K	163	< 3K	< 5	3. 3K	<3
1 - 1	Window Frame	1. 3K	< 5K	25K	638	2.7K	201K	144K	< 27	< 42	26	< 5K	41	< 430 .	< 3	<120	7. 8
169	Wall	162		82 K	2.5K	442	47 K	171K	< 55	95	132	2.8K	< 12	< 330	85	6.0K	16
178	Dove	855	< 5 K	3.9K	575	155	105K	223K	7. 8	253	851.	<5K	51	< 5K	< 4	15K	1.0
196	Auto	148			117	< 46	< 12K	7.9K	< 42	< 40	816	19K	< 11	< 560	51	< 72	3. 0
76	Chair	456	< 3K		787	176	< 10K	53K	< 5	< 42	240	42K	6. 2	< 2K	< 2	3 3 8	<1

CLASS V. (Continued)

Sample	Object			E	lem	ent	, pp	m (I	Kde	notes	thous	ands	of p	pm)	•		
No.	Sampled	As	Br	Sr	Мо		Cd	In	Sb	I	Ba	La	Hf	Ta	w	Au	Hg
33	Trash Can	3. 0	29	< 31	< 17	< 4	< 32	< 2	5.4	< 2	< 10	< 2	< 3	< 2	< 2	< 4	
37	Doors	< 38	<10	< 130	< 280	< 11	< 240		35K	< 11	155K	< 3	< 31	<20	< 18		< 3
52	Window Frame	24	31	185	< 25		< 40			< 4	5. 7K	_				< 1	< 50
57	Wall	16	29	< 21		< 34	< 24		44	< 23	79	23	< 5			< 5	< 5
58	Wall	15	2. 6	202	< 20		< 31	1.9		< 24	6. 8K			< 4		< 3	52
60	Cabinet	< 2	12		< 21		< 35		22	< 4			5. 5			< 4	< 4
64	Wall	709	59		< 48		< 93		15	< 210	6. 1K		< 4	< 3	< 2	< 5	< 4
66	Door	4.4	18		<28			< 10	20		5. 8K		< 5	< 3	< 4	< 1	105
68	Refrigerator	< 1	187	81	< 14		< 26			< 40	3. 9K		< 4	< 3	< 2	< 6	< 5
85	Door Fan	1.6	61		< 17			1		< 5	< 17	< 1	2. 1	< 2	< i	9.0	
123	Chest	2.9	53		<23			< 24	5.8	< 1	455	17	< 3	< 2	< i	8.2	< 3
1 4 6	Air Conditioner	43	55	_				< 2	6.9	5. 5	. , -	< 2	< 4	32	7. 4	0.3	< 4
149	Door	21	38		< 18			< 2	8.0	< 7	1.7K	1.8	< 3	< 2	< 2	0. Z	< 3
150	Cabinet	15	36 75		< 13		< 25	< 5	< 1	< 6	247	< 1	< 2	< 2	< 1	0.1	< 3
151	Window Frame	1.2			< 25		< 46	< 8	7.4	< 12	6. 3K	10	< 3	22	< 2	1.8	209
169	Wall		17		< 17		< 29		4.0	< 3	52	5.8	< 3	< 2	< 2	0.3	63
•		0.6	7. 3		< 10		< 16	< 5	< 1	< 22	< 17	6.1	< 3	15	< 1	< 3	< 2
178	Door	< 1	47	848	< 16 -		< 28	< 1	26	< 5	157	2.5	< 3	9.7	3. 1	0.1	< 3
196	Auto	< 1	80	< 22	< 9 <	< 20	< 17	< 4	< 1	< 30	2. 4K	0.3	< 3	< 2	< 1	< 3	< 2
76	Chair	< 1	24	< 51	< 11	< 6	< 20	< 1	0.7	< 6	2. 6K	2. 1	27	4.7	2. 2		<2

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