GA-10141

FORENSIC NEUTRON ACTIVATION ANALYSIS OF BULLET-LEAD SPECIMENS

June 30, 1970

Gulf General Atomic, Incorporated
San Diego, California

UNITED STATES ATOMIC ENERGY COMMISSION • DIVISION OF TECHNICAL INFORMATION

NOTICE

Printed in the United States of America
USAEC Division of Technical Information Extension, Oak Ridge, Tennessee

GA-10141
RADIOISOTOPE AND RADIATION
APPLICATIONS (TID-4500)

FORENSIC NEUTRON ACTIVATION ANALYSIS OF BULLET-LEAD SPECIMENS

by

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Prepared under
Contract AT(04-3)-167
Project Agreement No. 15
for the
Division of Isotopes Development
U. S. Atomic Energy Commission
and the
Law Enforcement Assistance Administration
U. S. Department of Justice

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Gulf Radiation Technology Project 295 June 30, 1970

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ABSTRACT

The possibility of using instrumental neutron activation analysis to determine whether two bullet lead specimens have common or different sources has been examined. It has been found that the number of elements observable, and thus the number of points of comparison, is generally limited to three elements, due to dominance of antimony radioisotopes in the activated bullet lead specimens. This factor, coupled with a high degree of composition uniformity of bullet lead from at least one major manufacturer, imposes some limitations on the method. Thus, while differences in identification points definitively indicate a difference in sources, two bullets with the same pattern of only three identification points are not usually definitively identified as having a common source.

Recommendations are given with respect to extending the activation analysis technique to enable positive identification of bullet samples as being from common or different sources.

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SUMMARY

The possibility of utilizing instrumental neutron activation analysis (NAA) for the comparison of bullet lead specimens has been investigated in two phases. In both phases bullet lead samples were activated by neutron bombardment at thermal neutron fluxes of $\sim 2 \times 10^2$ n/cm^2-sec in a TRIGA Mark I nuclear reactor, and subsequently the induced gamma-emitting radioisotopes were measured by multichannel gamma-ray spectrometry. Concentrations of antimony, an element often added to control bullet lead hardness, plus two other elements were usually determined by this procedure.

In the first phase of work experiments were carried out to see if the elements observed were of reasonably widespread concentration among a variety of bullets and to obtain an initial assessment of the uniformity of antimony concentration within a given bullet and with bullets from a common box of bullets. The findings were sufficiently encouraging to warrant a larger effort. Most of the gamma-ray measurements in the early work utilized a NaI(Tl) detector. However, a 3 c. c. Ge(Li) detector showed that arsenic was a suitable candidate identification element even though the gamma ray of its indicator isotope, ^{76}As (γ = 0.559 MeV) is very near that of a longer-lived major antimony isotope, ^{122}Sb (γ = 0.564 MeV).

The results of the first phase of work, taken in conjunction with a survey of the frequencies with which various calibers are involved in police work plus the constraints of time and funds, were used to define the kinds and numbers of samples to be taken and the analytical regime of a larger second phase effort.

The second phase effort utilized an experimental procedure similar to the preceding work, the chief exception being that a 35 c. c. Ge(Li) detector was used in gamma-ray spectrometry measurements of activated samples. Altogether, 230 samples of bullet leads taken from 75 different lots of bullets that had been selected in conformity with the survey results were analyzed. The elements quantitatively determined in a majority of samples were antimony, copper, and arsenic.

It was found that the intra-lot uniformity of bullet leads with respect to the three elements was as good as the intra-box uniformity (i. e., the bullet lead within a box of bullets). The relative standard deviations regarding this uniformity were ±4% for antimony, ±23% for copper, and ±44% for arsenic. Appropriately antimony is the most useful element for comparison purposes, and arsenic is the least useful.

Also, it was found that the copper and arsenic concentrations are correlated with antimony concentrations.

Most damaging to the forensic utility of purely instrumental NAA for bullet comparisons was the frequency with which antimony concentrations were observed to fall within the range of 0. 7-0. 8% w. This factor, coupled with the correlated concentrations of copper and arsenic and the larger variances of these two elements, contributed in large measure to the finding that less than half of the 75 lots of bullets were uniquely characterized by the concentrations of Sb, Cu, and As.

As a result of the foregoing it can be said that a significant difference in concentration of any one of the three elements between two bullet specimens indicates that they came from different lots, but that matching concentrations of all three elements does not indicate that two bullets come from the same lot.

Enhancement of the utility of the method to minimize the chance of accidental matching of two bullets from different lots is desirable.

This can be achieved either by (1) utilizing post-irradiation radiochemical separations to separate the dominant, interfering antimony radioisotopes, which would enable the measurement of more elements, or (2) by incorporating a unique combination trace element tag, which could be easily measured by NAA, in each lot of bullets.

1. INTRODUCTION

The identification of bullets is of major interest in criminalistics. The primary technique applied to this end has been the examination of striations imposed on the bullet by the gun during firing. A bullet in evidence will be compared with one fired from a weapon that is linked to a suspect. If the two bullets have matching striations, then the evidence bullet may be considered as linked to the suspect. Unfortunately, a bullet fired in the field often undergoes such severe encounters that the markings are obliterated for all practical purposes. Sometimes the bullet is even fragmented. With the possibility in mind that instrumental NAA (Neutron Activation Analysis) might provide a means of definitively comparing spent bullet with unfired bullets in a suspect's weapon, which would be particularly useful where markings on the spent bullet (or fragment) were absent, the study described therein was initiated.

The possibility that other potential uses for the NAA characterization of bullets could exist augmented the initial consideration. For example, in cases where the weapon involved in a shooting incident could not be found, it could be of interest to compare an evidence bullet with unfired bullets found in the possession of a suspect. Or, as a corollary, in cases where a multiplicity of weapons were fired it would be of interest to compare certain spent bullets with unfired bullets found in selected weapons.

2. EXPERIMENTS AND RESULTS

The examination of bullets by NAA involved the exposure of weighed bullet lead samples to neutron bombardment in the Gulf General Atomic TRIGA Mark I nuclear reactor for purposes of generating radioactive analytical indicator radioisotopes from the elemental constituents of the samples. In some experiments the irradiation was carried out for $\leq$ 1 minute at a thermal neutron flux of 2.8×10^{12} n/cm^2-sec, and the irradiated samples were examined by multichannel gamma-ray spectrometry within a few minutes after termination of the irradiation. In other experiments irradiations were carried out for 30 minutes at a thermal neutron flux of 1.8×10^{12} n/cm^2-sec, and the irradiated samples were counted at longer decay times.

Examination of irradiated specimens by counting was carried out with 3-in. x 3-in. NaI(Tl) detectors coupled to multichannel pulse height analyzers, particularly in the early phases of the work.[1] Later, Ge(Li) detectors were used so that indicator radioisotopes giving rise to gamma rays of similar energy could be distinguished and quantitated.

In initial experiments tabulated photopeak yield values (in photopeak counts per minute per gram of element) were used to obtain elemental yield values from the quantitized photopeaks.

Once it became clear which elemental constituents of bullet lead were of most interest in this study, weighed comparator standards of these elements were irradiated and measured with, and in identical manner to, the samples. Thus, concentrations of key elements in the samples were derived by comparison of appropriate, quantitized photopeaks in the sample and comparator spectra.

Clean polyethylene vials were used to contain the samples and standards during irradiation and counting. Usually the samples, which were solid pieces, were transferred to unirradiated vials for counting in order to dispel ^{41}Ar and avoid vial blanks.

2.1 INITIAL EXPERIMENTS AND RESULTS

Small samples (10-100 mg slices) of bullet leads from a variety of bullets were irradiated for 15 seconds, and counted for one minute (NaI(Tl) detector) beginning at one minute after the end of the irradiation. A number of elements were quantitatively determined from the intensities of observed photopeaks, and the results are given in Table 1.

Lead samples from a few of the foregoing bullets were examined by emission spectroscopy in order to ascertain the degree to which this method and NAA complement one another. The emission spectroscopy results are given in Table 2.

The initial gamma-ray spectra of irradiated, antimony-containing lead sample were fairly complex in the 0.50-0.70 MeV region, which rendered the determination of silver by means of the 24 second isotope, ^{110}Ag (E_γ = 0.658 MeV), somewhat difficult. This is illustrated in Fig. 1.

It was ascertained that the complexity in the 0.50-0.70 MeV region was due to the production of a short-lived antimony isotope by irradiating a 1.2 mg portion of antimony metal for 15 seconds at a thermal neutron flux of 2.8×10^{12} n/cm^2-sec and measuring its gamma-ray spectra starting at 0.5 minutes and 4.75 minutes after the irradiation. The initial spectrum, shown in Fig. 2, has 0.51 MeV and "0.62" MeV peaks similar to those in Fig. 1. The later spectrum (in Fig. 2) shows that the responsible isotope is short-lived; the 0.564 MeV photopeak of 2.8 day ^{122}Sb is visible in the later spectrum.

The short-lived radioisotopes generated from antimony by neutron activation include 3.5 minute ^{122m}Sb, which has gamma rays of 0.061

Table 1

INITIAL INSTRUMENTAL ACTIVATION ANALYSIS OF A VARIETY OF BULLET LEADS

Bullet	Sample Wt., mg	Concentrations of Elements Observed[a] Sb, %	Sn, %	Ag, ppm	Cu, ppm	Al, ppm
0.45 Cal., reloader's semi-wad-cutter, Krasne's Sporting Goods	11.1	1.0	0.56			15
0.45 Cal., reloaders semi-wad-cutter, Hensley & Gibbs	12.0	2.8				10
0.22 Cal., pellet, Crossman	9.7	<0.02		39		
0.30 -'06 Cal., Remington, 150 gr, Brass point	18.4	1.2			400	12
0.22 Cal., Hornet, Hornady, 50 gr.	10.1	3.5				5.4
0.308 Cal., Sierra, 150 gr., Spitzer	21.4	3.1	0.21			5.3
0.30 Cal., Nosler, 150 gr, Partition Spitzer						
Upper Section	28.0	2.2			120	5.2
Lower Section	16.7	2.1				7.8
0.38 Cal., reloaders semi-wad-cutter, Hensley & Gibbs	23.2	4.4	0.34			75
0.45 Cal., Military	30.8	<0.007				11
Shot from Remington-Peters 12 gauge shot-shell	81.4	1.0				0.92

a) Analytical Indicators:

For Sb, Sb^{122}, 2.8 day half-life, 0.564 Mev gamma ray
For Sn, Sn^{125m}, 9.5 min " , 0.325 Mev " "
For Ag, Ag^{110}, 24 sec. " , 0.658 Mev " "
For Cu, Cu^{64}, 12.8 hour " , 0.511 Mev " "
For Al, Al^{28}, 2.3 min. " , 1.78 Mev " "

Table 2

EMISSION SPECTROSCOPIC ANALYSES OF BULLET LEAD

Bullet	Sb, %	Sn, %	Ag ppm	Cu ppm	Bi ppm	Fe ppm
0.45 Cal., reloaders semi-wad-cutter, Krasne's Sporting Goods	1	<0.2	10	40	200	100
0.45 Cal., reloaders semi-wad-cutter, Hensley & Fibbs	>1	0.04	20	80	400	100
0.30-'06 Cal., Remington, 150 g Brass Point	1	<0.006	20	800	600	60
0.45 Cal., Military	<0.008	<0.006	10	40	800	40

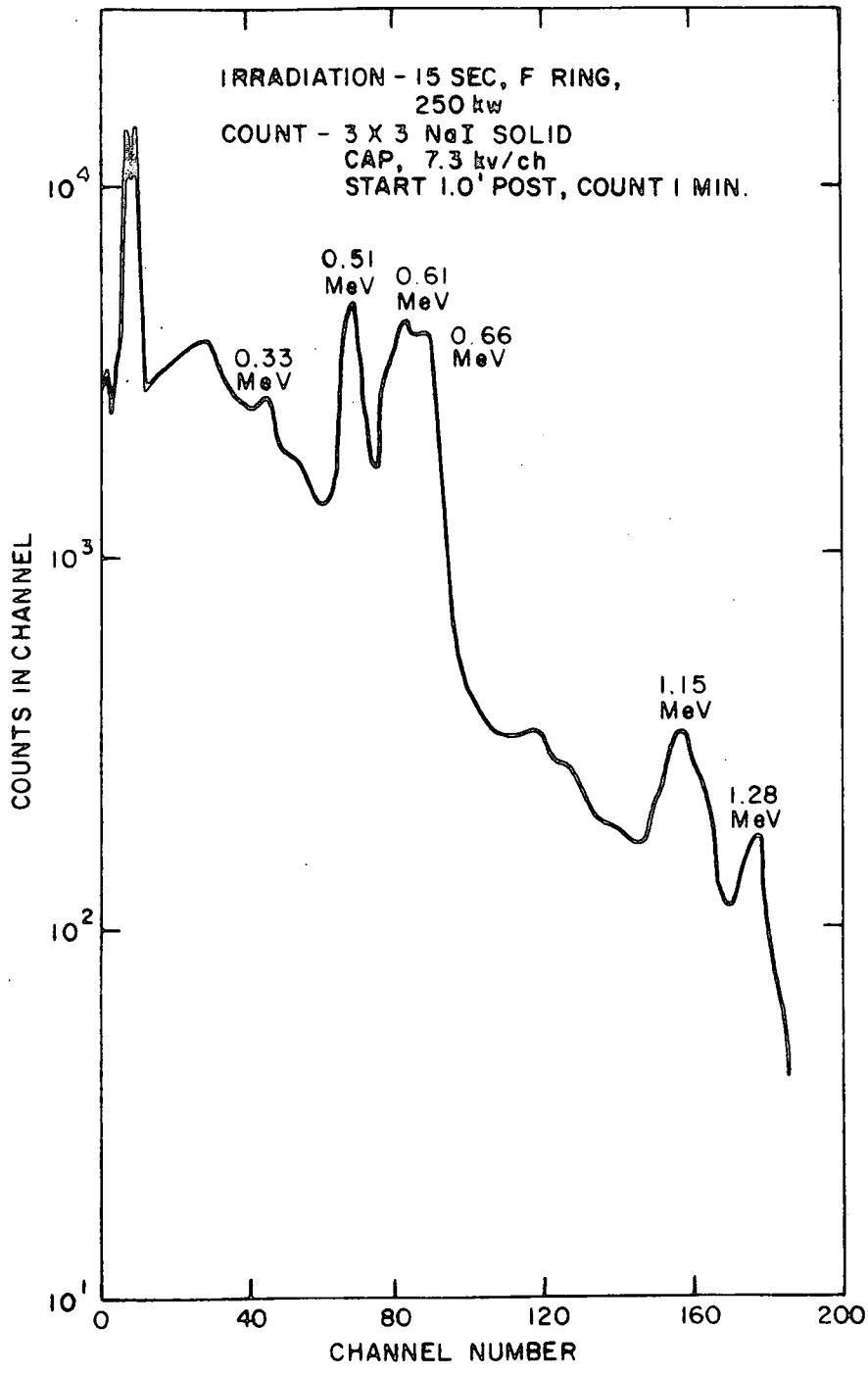

Fig. 1. Gamma-ray spectrum of activated lead (21.4 mg) from Sierra 30-cal. bullet, 150 grain, 0.308 Spitzer

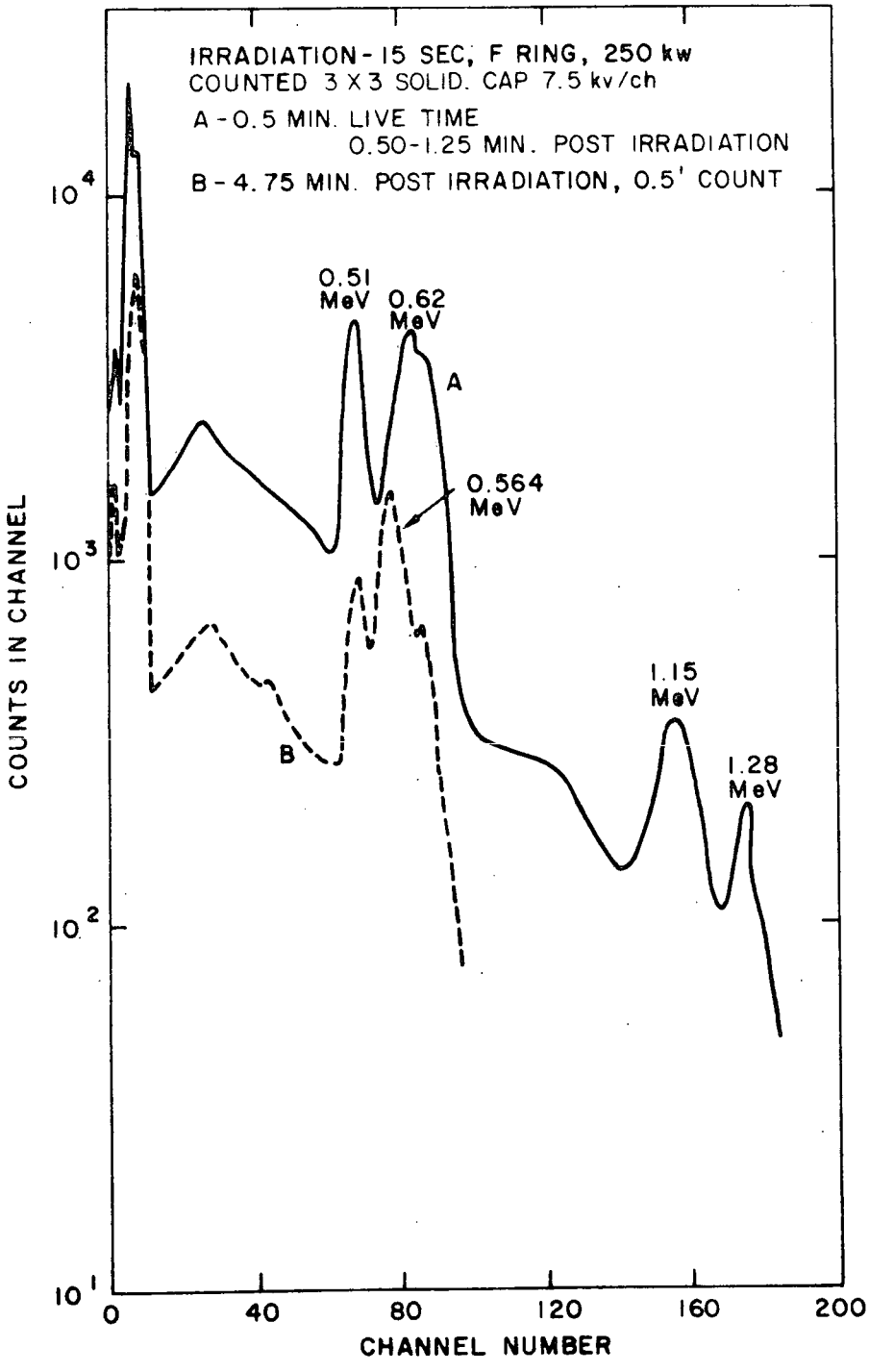

Fig. 2. Gamma-ray spectrum of activated Sb (1.2 mg)

MeV (50% yield) and 0.075 MeV (17% yield). The weak gamma rays of ^{122m}Sb are shown as a composite peak in both Figs. 1 and 2. The irradiation of ^{121}Sb (99% enriched, obtained from Oak Ridge National Lab) produced ^{122m}Sb, as shown by the spectra in Fig. 3. The composite 0.061 MeV-0.075 MeV peak is clearly seen, and the 0.564 MeV photopeak from 2.8 day ^{122}Sb is in evidence, also.

Another short-lived isotope is 1.3 minute ^{124m}Sb, which emits gamma rays of 0.505 MeV (20% yield), 0.603 MeV (20% yield), and 0.644 MeV (20% yield). Spectra of this isotope were obtained subsequent to the irradiation of ^{123}Sb (99% enriched, from ORNL) as shown in Fig. 4. The 0.505 MeV peak and the composite 0.603 MeV plus 0.644 MeV peak which are clearly shown in Fig. 4, show that the ^{124m}Sb is responsible for the complexity of the 0.5-0.7 MeV region of Fig. 1.

The data obtained from the measurements of irradiated, enriched ^{121}Sb and ^{123}Sb indicated the photopeak intensities of the short-lived antimony isotopes shown in Table 3.

Elucidation of the 0.50-0.70 MeV region of spectra from activated antimony has made it possible to avoid excessive errors in the computation of silver concentrations when a strong 0.66 MeV peak, due to 24 second ^{110}Ag, is observed. The 0.66 MeV peak in Fig. 1 is evidence of the presence of ^{110}Ag activity; but it is a fairly weak peak – therefore, a silver concentration was not computed in this case. In general, the Ag concentration could be determined with a precision of ± 50% relative.

An examination of 18 bullets obtained from the Bureau of Criminal Identification and Investigation (CII) laboratory of the California State Department of Justice was carried out. All bullets were 0.38-caliber (either Smith and Wesson or Colt New Police), but no two bullets were alike with respect to all of the following: manufacturer, S&W variant, Colt N.P. variant, case, jacket, and lot. These bullets were examined

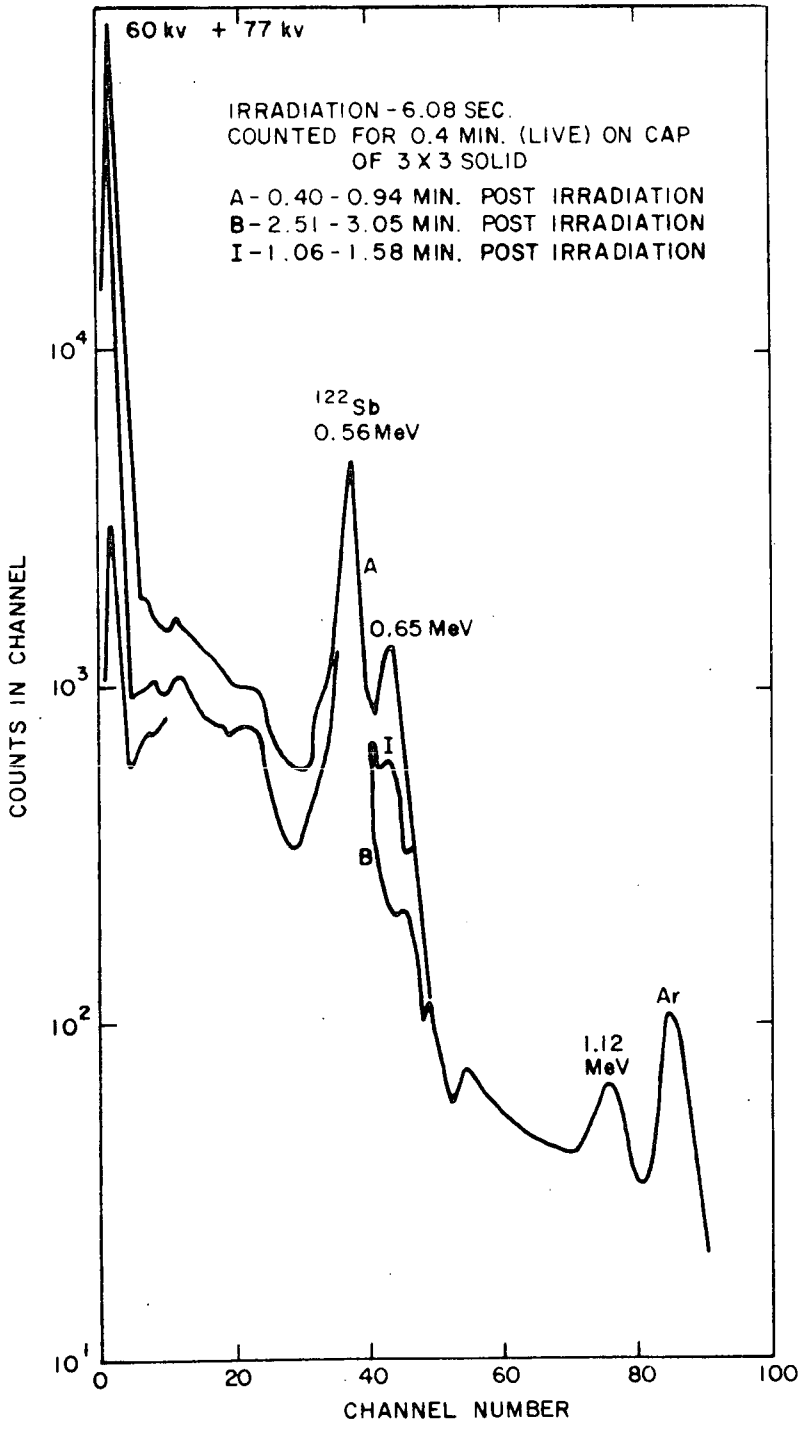

Fig. 3. Gamma-ray spectrum of activated Sb-121 (3.90 mg)

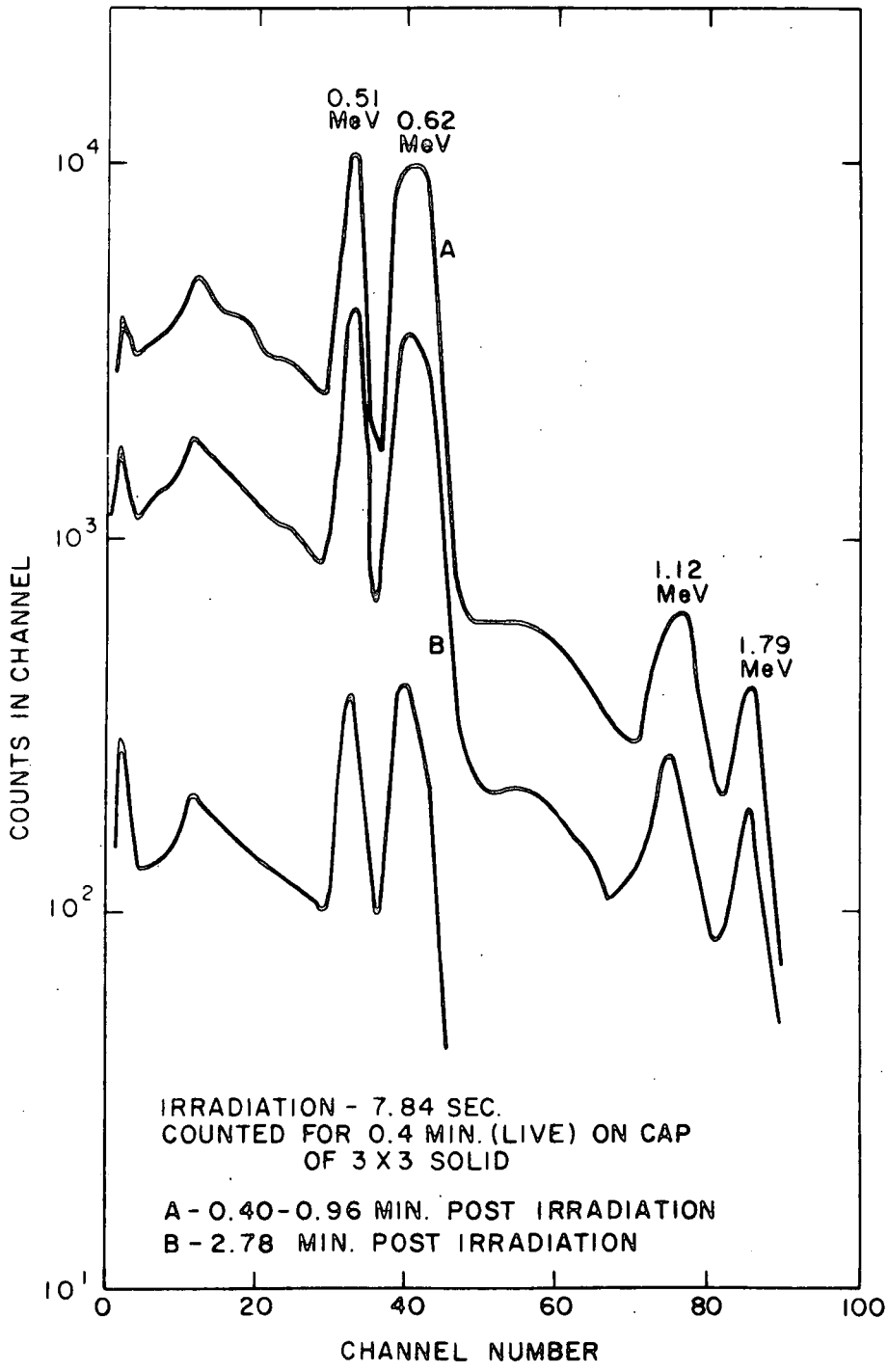

Fig. 4. Gamma-ray spectrum of activated Sb-123 (1.21 mg)

Table 3

Sb^{122m} AND Sb^{124m_1} PHOTOPEAK INTENSITIES

Target Isotope	Product Isotope	Half-Life	Observed Photopeak (MeV)	Integrated Photopeak Intensity CPM/g[a]
Sb^{121}	Sb^{122m}	3.5 m	0.061 + 0.075	2.1×10^8
Sb^{123}	Sb^{124m_1}	1.3 m	0.505	6.2×10^7
			0.603 + 0.644	1.2×10^8

[a] Corrected to specific activity in natural, un-enriched, antimony at the end of a 15-second irradiation at a thermal-neutron flux of 2.8×10^{12} n/cm^2-sec. Sample on 0.5-inch plastic cap on 3-in. x 3-in. NaI(Tl) solid counter.

initially with respect to antimony concentration to further study the variation of this element in bullets known to be different from one another. However, at a later time the silver and arsenic contents were examined. A Ge(Li) detector (3 c. c.) was used for the measurement of arsenic (to be discussed later). A summary of the findings is given in Table 4.

2. 2 BULLET UNIFORMITY EXPERIMENTS

From the foregoing experiments it seemed possible that instrumental activation analysis can differentiate between bullets that are not from the same batch of lead. However, it was necessary to examine bullets from a given batch of lead, and even individual bullets, for uniformity of composition – especially with respect to antimony concentration – in order to ascertain that compositional differences are not due to batch inhomogeneity.

2. 2. 1 Bullet Lead Homogeneity

A rod of bullet lead, which contained a nominal 1-1/2% w Sb, was sampled from the exterior to the interior. Two groups of samples were obtained – a group of eight samples weighing from 62. 3 to 113. 2 mg each and a group of 12 weighing from 7. 20 to 27. 60 mg each. The samples were activated for 30 minutes, allowed to decay for eight hours, and measured by a multichannel gamma-ray spectrometer (using a 3-in. x 3-in. NaI(Tl) detector). The net ^{122}Sb photopeak (0. 564 MeV) cpm per milligram of each sample is given in Table 5. The average of the first eight samples is 5450 cpm/mg with a standard deviation of 3. 85% of the value. The average of samples 9 through 20 is 5505 cpm/mg with a standard deviation of 2. 50% of the value. The average of all 20 samples is 5483 cpm/mg with a standard deviation of ±3. 00% of the value.

In a second test of bullet uniformity, a Remington 0. 38-caliber bullet was sampled from the side to the center and processed in the same

Table 4

ANTIMONY, SILVER, AND ARSENIC IN EIGHTEEN 0.38-CALIBER BULLETS

Type of Bullet	Sb, %	Ag, ppm	As, ppm
U. S. Cartridge Co., S&W	<0.003	86.5	<0.45
U. S. Cartridge Co., CNP	1.55	0.67	<23.0
Union Metallic Cartridge Co. (UMIC) S&W	0.082	2.20	2.2
Remington-UMC, S&W (noncrimp)	0.013	2.64	27.0
Remington-UMC, S&W (crimp)	1.76	4.1	<32.0
Remington-UMC, CNP	2.07	8.8	<39.0
Peters, S&W (Lot B25A67NK)	1.94	7.6	<35.0
Peters, CNP	2.99	0.77	<100.0
Remington-Peters, S&W	0.92	3.6	<12.0
Remington-Peters, CNP (Index 7138)	0.87	3.1	<15.0
Western, S&W (Jacketed, Steel Case)	3.00	2.6	<72.0
Western, S&W (Jacketed, Brass Case)	0.044	11.5	<2.2
Winchester, S&W (Brass Case)	0.076	16.0	167.0
Winchester, S&W (Steel Case)	3.09	4.9	<66.0
Winchester, CNP (crimp)	1.38	0.56	<24.0
Winchester, CNP (noncrimp)	1.47	0.56	<25.0
Winchester, CNP (No. W38CNP)	1.55	3.9	<24.0
Winchester, CNP (Lot L747)	1.23	1.8	31.2

Table 5

UNIFORMITY OF NORMA BULLET LEAD ROD

	Sample	Wt. (mg)	Peak Net (CPM/mg)
Exterior	1	88.35	5140
↓	2	52.6	5721
	3	73.6	5363
	4	113.2	5235
to	5	62.3	5560
	6	92.7	5359
	7	62.3	5545
Center	8	63.7	5681
Exterior	9	13.15	5346
↓	10	19.15	5428
	11	8.00	5573
	12	11.65	5561
	13	15.35	5651
	14	26.70	5461
to	15	11.45	5522
	16	20.50	5288
	17	7.20	5700
	18	27.60	5332
	19	18.45	5650
Center	20	10.40	5547

manner as the Norma bullet lead. The results are given in Table 6. The average net counts per minute in the ^{122}Sb peak per milligram of lead was 1123.3 cpm/mg, and the standard deviation was 2.85% of the value.

2.2.2 Bullet-to-Bullet Similarity

A multiplicity of bullets from each of a number of boxes of bullets were sampled. The coating (grease, copper, and/or bronze) was removed from each bullet with organic solvent and weak nitric acid, and bullets were washed with water and dried before sampling. Again the emphasis was placed on the examination of antimony concentration; however, two brands of 0.22-caliber bullets that had similar antimony concentrations were examined for trace elements. Results are given in Table 7.

2.2.3 Initial Experiments with a Ge(Li) Detector

A 3 c.c. Ge(Li) detector was found to be able to measure As in the presence of Sb, provided the Sb/As ratio was not too great. The radioisotopic analytical indicator for As is 26.5 hour ^{76}As, which emits a 0.559 MeV gamma ray. Thus only 0.005 MeV separates the energy of the ^{76}As gamma ray from that of 2.8 day ^{122}Sb (E_γ = 0.564 MeV). Nevertheless, the Ge(Li) detector was capable of resolving the two gamma rays, as shown in Fig. 5. This figure shows the gamma-ray spectrum obtained from a 99.6 mg portion of lead from a 0.22 caliber bullet (Sears) that had been irradiated for 30 minutes and counted for 10 minutes (at two hours after the irradiation) with the 3 c.c. Ge(Li) detector. Examination of five bullets from the same box (Sears, 0.22-caliber) showed that the arsenic content was 285 ± 40 ppm. A Peters 0.22-caliber bullet had 345 ppm As, a Western 0.22-caliber bullet had 78 ppm As, an Imperial 0.22-caliber bullet had < 4 ppm As, and a Lapua 0.22-caliber bullet had < 5 ppm As.

Table 6

UNIFORMITY OF REMINGTON 0.38 CALIBER BULLET

	Sample	Wt (mg)	Peak Net (CPM/mg)
Exterior	1	15.6	1142.1
↓	2	28.7	1069.6
↓	3	48.4	1149.8
↓	4	47.8	1085.5
↓	5	97.3	1155.1
↓	6	103.0	1162.1
to	7	36.4	1162.1
↓	8	80.5	1108.4
↓	9	55.0	1103.6
↓	10	103.1	1139.1
↓	11	98.4	1078.5
Center	12	50.1	1124.2

Table 7

BULLET-TO-BULLET UNIFORMITY EXPERIMENTS

Bullets	No. of Samples from a Single Box	Sb, (% w)[a]	Other Elements
Lapua, 0.22-cal. L.R. matchgrade	50	1.22 ± 0.04	1.04% Sn, 1.09 ppm Al
Sears, 0.22-cal. short	20	1.26 ± 0.03	0.18% Sn, 1.25 ppm Al
Imperial, 0.22-cal. short	20	0.99 ± 0.04	
Peters, 0.22-cal. short	20	0.87 ± 0.08	
Remington, 0.38 S&W	10	0.85 ± 0.02	
Western, 0.38 S&W	10	2.59 ± 0.10	

[a] The plus-or-minus values represent one standard deviation determined from counting statistics only.

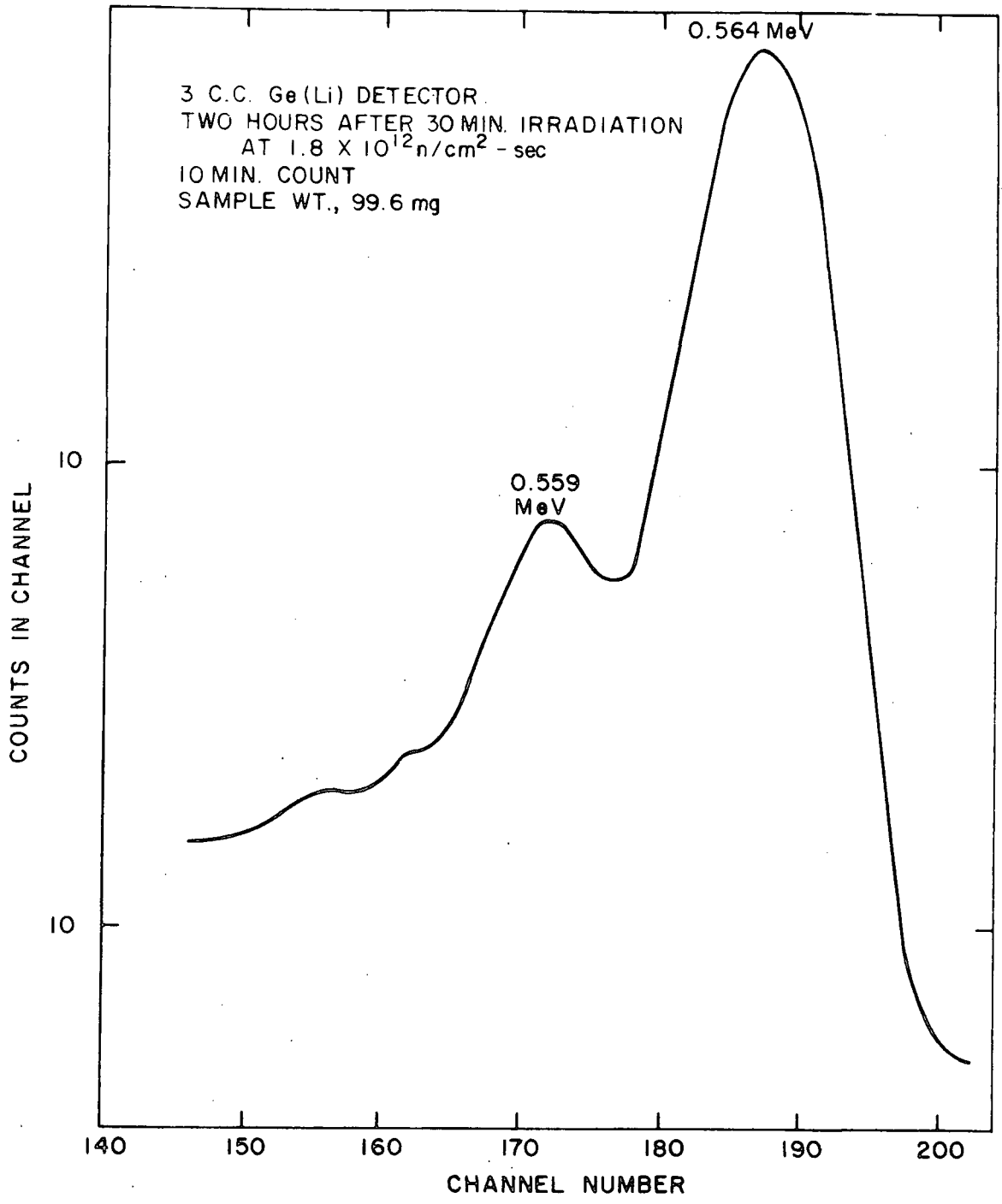

Fig. 5. Gamma-ray spectrum of activated Sears 0.22-cal. bullet lead

2.3 FINAL EXPERIMENTS AND RESULTS

Based upon the results of the initial experiments described in the preceding section, it was estimated that 30 different bullets from each of the six major calibers of cartridge (0.22, 0.25, 0.30, 0.32, 0.38, and 0.45) should be analyzed in order to properly characterize the population of bullet leads.[1] However, the constraints of time and effort, taken in context with other considerations, prevented the full realization of this task.

The other considerations included the fact that the 3 c.c. Ge(Li) detector had since been replaced with a superior 35 c.c. Ge(Li) detector, and it was desired that all further work be done with the better detector. Thus, the previous work, which had not been done with this equipment, could not be deemed to carry a full measure of contribution toward fulfillment of the goal.

Another factor was that it could not be considered sufficient to analyze simply one bullet from each different kind (as specified by manufacturer, lot number, and box). It was necessary to obtain further bullet uniformity data, including intra-box, inter-box (intra-lot, and inter-lot comparisons.

In consequence of the above needs, the work of the original plan, which called for the analysis of 180 different bullets in total, would have escalated considerably beyond reasonable limits. Therefore, advantage was taken of information obtained from California criminalists regarding the frequency with which various calibers of handguns are brought to their attention.[2] These frequencies were as follows:

0.22 caliber - 32%
0.38 caliber - 25%
0.32 caliber - 13%
0.45 caliber - 9%
0.44 caliber - 6%
9 mm - 4%
0.25 caliber - 4%

Clearly, then some stratification in sampling was called for rather than sampling all calibers equally. Also, discussions with gun dealers, policemen, and criminalists suggested that 0.357 magnum bullets, shotgun slugs, and 00 buckshot should be sampled.

Accordingly 242 samples were acquired and analyzed much as in previous experiments, except that the 35 c.c. Ge(Li) detector, coupled to a 4096 channel pulse height analyzer, was used for gamma-ray spectrometric measurements. Unfortunately, the timing cycles in these experiments precluded the observation of silver. A number of the bullets had copper jackets, and it was found subsequent to irradiation that microscopic specks of copper remained on a few samples. This happenstance, plus a few malfunctions of the pulse height analyzer necessitated deletion of 12 samples. The remaining 230 samples were distributed as follows:

0.22 caliber	-	70 bullets among 21 different lots
0.25 caliber	-	5 bullets among 1 lot
0.32 caliber	-	23 bullets among 10 different lots
0.357 caliber	-	19 bullets among 3 different lots
0.38 caliber	-	72 bullets among 26 different lots
9 mm caliber	-	15 bullets among 3 different lots
0.44 caliber	-	11 bullets among 3 different lots
0.45 caliber	-	6 bullets among 3 different lots
12 gauge	-	9 projectiles among 5 different lots

The initial 242 bullets among 75 different lots are itemized in Table 8, wherein it may be seen that several variants among the major calibers were sampled, e.g., 0.38 automatic, 0.38 CNP (Colt New Police), 0.38 S&W (Smith & Wesson), and 0.38 Special.

The previously mentioned problem with microscopic specks of copper was most frequent among the first samples analyzed, and it

Table 8

BULLETS ANALYZED IN FINAL STUDY

Samples	Mfg.[a]	Caliber	Identification Numbers		Box
			Lot. No.	Index No.	
1-5	Rem.	.38 S&W	627N		1
6-10	Fed.	.38 Spec.	FPCS15KC		1
11-15	W-W	.22 LR	CD71		1
16-20	W-W	.38 S&W	9397YA5		1
21-25	W-W	.22 Short	BL4		1
26-30	Fed.	.22 LR	LF4JC		1
31-34	W-W	.22 LR	BK72		1
35-39	Rem.	.25 Auto	L15ZD		1
40-44	Rem.	9 mm Lug.	J23A		1
45-49	W-W	9 mm Lug.	45BC51		1
50-54	W-W	9 mm Lug.	33BF7		1
55-56	Rem.	.38 Spec.	N06D	3841	1
57-58	Rem.	.38 Spec.	P07G	3841	1
59	Rem.	.38 Spec.	P07G	3841	2
60	Rem.	.38 Spec.	P07G	3841	3
61	Rem.	.38 Spec.	P07G	3841	4
62	Rem.	.38 Spec.	N06D	3841	2
63	Rem.	.38 Spec.	K29H	3841	1
64	Rem.	.38 Spec.	N06D	3841	3
65	Rem.	.38 Spec.	K29H	3841	2
66-67	Rem.	.38 Spec.	M24R		1
68-69	W-W	.38 Spec.	3528BE6	3853P	1
70-71	W-W	.38 Spec.	57BK7	38SMRP	1
72-74	Fed.	.38 Spec.	CS20KC	38A	1
75-76	Rem.	.38 Spec.	RA5289	M41	1
77-78	Rem.	.380 Auto.	021C	1239	1
79-81	Rem.	.44 Rem. Mag.	H09HG23LD	4411	1
82-84	Rem.	.44 Rem. Mag.	H09HH05SD		1
85-87	Rem.	.44 S&W Spec.	M11E-20P	4405	1
88-90	Rem.	.44 S&W Spec.	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

[a] Rem. = Remington, or its subsidiary, Peters; W-W = Winchester-Western; Olin = Olin Mathieson; Fed = Federal

Table 8 (Continued)

Samples	Mfg.[a]	Caliber	Lot No.	Index No.	Box
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 Mag.	11P-P09D	3578	1
109-111	Rem.	.357 Mag.	11P-N14P	3578	1
112-114	Rem.	.357 Mag.	11P-P09D	3578	2
115	Rem.	.357 Mag.	11P-P09D	3578	3
116	Rem.	.357 Mag.	11P-P09D	3578	4
117	Rem.	.357 Mag.	11P-P09D	3578	5
118	Rem.	.357 Mag.	11P-P09D	3578	6
119	Rem.	.357 Mag.	11P-P09D	3578	7
120	Rem.	.357 Mag.	11P-P09D	3578	8
121-122	Rem.	.357 Mag.	11P-K29ED	3578	1
123	Rem.	.357 Mag.	11P-N14P	3578	2
124	Rem.	.357 Mag.	11P-N14P	3578	3
125	W-W	.45 Auto.	53-33BE01	45A1P	1
126	W-W	.45 Auto.	53-33BE01	45A1P	2
127	W-W	.45 Auto.	53-33BE01	45A1P	3
128	Rem.	.45 Auto.	23PN13A	4504	1
129-130	Rem.	12 Gauge Slugs $2\frac{3}{4}$ in.	BN22N17	PS12RS	1
131	Rem.	12 Gauge Slugs $2\frac{3}{4}$ in.	AN22N17	PS12RS	1
132	W-W	12 Gauge Slugs $2\frac{3}{4}$ in.	R11BD81	SX12PRS	1
133	W-W	12 Gauge Slugs $2\frac{3}{4}$ in.	R11BD81	SX12PRS	2
134-135	W-W	12 Gauge Slugs, 00	G62YL42	SX12PRB	1
136	Rem.	12 Gauge Slugs, 00	AN12P18	PS12-$3\frac{3}{4}$-00BK	1
137	Rem.	12 Gauge Slugs, 00	AN12P18	PS12-$3\frac{3}{4}$-00BK	2
138-139	Speer	.38 Spec.	910001	3748	1
140-141	Speer	.38 Spec.	03003	3758	1
143	Speer	.38 Spec.	03005	3752	1

Table 8 (Continued)

Samples	Mfg.[a]	Caliber	Lot No.	Index No.	Box
144	Speer	.38 Spec.	03002	3757	1
145	Rem.	.22 Short	Unknown		1
146	W-W	.22 Short	Unknown		1
147	Rem.	.22 Spec.	Unknown		1
148	W-W	.22 Spec.	Unknown		1
149	W-W	.22 Spec.	Unknown		1
150	Rem.	.22 Spec.	Unknown		1
151-152	Rem.	.32 Short R. F.	321A45		1
153-154	W-W	.32 S&W	Unknown		1
155-156	W-W	.32 S&W	Unknown		1
157-158	Olin	.45 Ball	WRA22690		1
159-160	Rem.	.38 Auto.	Z242	1438	1
161	W-W	.38 CNP	36-25TD3	W38CNP	1
162	W-W	.38 CNP	Unknown		1
163-165	Rem.	.38 Spec.	L19B	3841	1
166	Rem.	.38 Spec.	L19B	3841	2
167	Rem.	.38 Spec.	L19B	3841	3
168	Rem.	.38 Spec.	L19B	3841	4
169	Rem.	.38 Spec.	L19B	3841	5
170	Rem.	.38 Spec.	L19B	3841	6
171-173	Rem.	.38 Spec.	J09P	3841	1
174	Rem.	.38 Spec.	J09P	3841	2
175	Rem.	.38 Spec.	J09P	3841	3
176	Rem.	.38 Spec.	J09P	3841	4
177	Rem.	.38 Spec.	J09P	3841	5
178	Rem.	.38 Spec.	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	3TF27E165	2338	1
193-194	Rem.	.38 S&W	9RG27N	2338	1
195-196	Rem.	.32 S&W	1RJ10U	1132	1
197	Rem.	.32 S&W	Unknown		1
198-200	W-W	.32 S&W	112140	K3272T	1
201-203	W-W	.32 S&W	A4822	K3272T	1
204-205	W-W	.32 Auto.	88CA8	32AP	1
206-207	Rem.	.32 Auto.	L22ED	2632	1

Table 8 (Continued)

Samples	Mfg.[a]	Caliber	Lot. No.	Index No.	Box
208-210	Rem.	.32 Short Colt	Z221	1632	1
211-213	Rem.	.22 Short	J12P2D		1
214-216	Fed.	.22 Short	524C	701	1
217-219	W-W	.22 Short	XB82	SX22S	1
220-222	W-W	.22 Short	WK72	XP22S	1
223-224	W-W	.22 Short	WK72	XP22S	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	YC2	SX22LR	1
238-239	Rem.	.22 LR	LOIR2B	2224	1
240-242	Rem.	.22 LR	W24A2B	1522	1

occurred even among some specimens of bullets not initially coated with copper. Presumably the copper-colored, shiny particles were brass from the cartridge case in some instances. After the problem was recognized, samples were examined under the microscope prior to irradiation.

The results obtained by NAA of the samples are given in Table 9.

It should be mentioned that no attempt was made to reirradiate specimens subsequent to any analytical difficulty due to the lifetime of radioactive species present and time constraints.

Table 9

ANALYTICAL RESULTS IN FINAL STUDY

Sample	Element[a]			Sample	Element		
	Sb, %	Cu, ppm	As, ppm		Sb, %	Cu, ppm	As, ppm
1	x	x	x	38	0.771	710	102
2	0.897	940	45	39	0.781	434	66
3	0.890	958	79	40	0.728	1050	x
4	0.904	988	30	41	0.980	1220	131
5	0.854	862	180	42	1.03	670	680
6	x	x	x	43	0.976	760	660
7	1.65	467	758	44	1.02	840	780
8	1.66	466	778	45	0.0394	13	< 5
9	1.65	433	712	46	0.0646	40	< 7
10	1.68	493	663	47	0.0337	14	10
11	x	x	x	48	0.103	59	12
12	0.641	1730	< 26	49	0.201	99	< 9
13	0.661	x	< 24	50	0.124	32	< 7
14	0.678	x	< 39	51	0.0138	8	< 5
15	0.676	1620	< 34	52	0.111	32	21
16	x	x	x	53	0.0156	10	< 6
17	0.306	x	184	54	0.121	27	<10
18	0.291	x	477	55	0.724	711	<13
19	0.291	97	373	56	0.644	769	<24
20	0.307	x	352	57	0.715	958	<14
21	x	x	x	58	0.701	1020	<16
22	0.188	504	525	59	0.736	1070	<17
23	0.185	492	492	60	0.691	800	<12
24	0.189	x	588	61	0.930	x	23
25	0.188	439	502	62	0.718	704	<16
26	1.88	890	520	63	0.751	716	153
27	1.85	500	520	64	0.667	795	<23
28	1.89	500	500	65	0.728	581	21
29	1.88	x	585	66	0.742	625	<12
30	1.88	440	500	67	0.736	677	29
31	0.785	145	141	68	2.35	336	391
32	0.754	356	180	69	2.26	782	545
33	0.751	63	35	70	0.583	200	<25
34	0.803	166	103	71	0.660	105	30
35	0.852	500	48	72	0.424	57	25
36	0.948	890	278	73	0.421	47	18
37	0.962	562	293	74	0.430	50	<27

[a]Value not obtained

Table 9 (Continued)

Sample	Sb, %	Cu, ppm	As, ppm	Sample	Sb, %	Cu, ppm	As, ppm
75	0.929	x	1010	115	0.665	664	<14
76	0.857	620	955	116	0.682	831	19
77	1.01	786	367	117	0.690	720	<22
78	1.03	787	303	118	1.28	553	<10
79	0.740	451	< 23	119	0.722	837	<19
80	0.867	684	82	120	0.696	832	12
81	0.744	371	30	121	0.968	742	304
82	0.753	371	21	122	1.02	895	268
83	0.814	593	38	123	0.674	552	<13
84	0.850	730	95	124	0.676	520	<14
85	0.731	505	< 40	125	0.063	16	5.7
86	0.743	523	< 24	126	0.035	15	< 2
87	0.797	708	74	127	0.037	15	< 2
88	0.745	502	64	128	1.02	263	240
89	0.761	446	46	129	0.00096	2.8	< 2
90	0.834	516	36	130	0.0024	5.4	< 2
91	0.711	449	42	131	0.0011	2.3	< 2
92	0.734	139	< 22	132	0.0368	3.7	< 5
93	0.725	129	< 20	133	0.0344	2.5	< 6
94	0.799	458	69	134	0.638	36	33
95	0.691	438	67	135	0.648	46	<18
96	0.702	123	< 12	136	0.729	997	50
97	0.723	139	16	137	0.745	941	88
98	0.709	110	< 8	138	2.63	796	592
99	0.708	425	93	139	2.61	805	517
100	0.726	458	62	140	3.24	236	671
101	0.761	357	< 21	141	3.22	274	633
102	0.721	127	< 11	142	3.25	258	694
103	0.710	134	< 8	143	0.00041	8.8	< 3
104	0.764	364	21	144	0.516	446	91
105	0.706	109	< 8	145	0.719	728	42
106	0.670	770	< 13	146	0.889	76.6	<23
107	0.688	809	22	147	1.79	836	138
108	0.674	668	16	148	2.39	419	<17
109	0.663	530	< 7	149	2.50	329	46
110	0.729	848	113	150	1.67	258	383
111	0.628	509	18	151	2.52	478	<42
112	0.658	788	< 18	152	2.52	445	<68
113	0.657	737	< 14	153	0.900	92	23
114	0.657	795	< 21	154	0.822	298	97

Table 9 (Continued)

Sample	Sb, %	Cu, ppm	As, ppm	Sample	Sb, %	Cu, ppm	As, ppm
155	0.013	87	24	195	0.963	546	288
156	0.019	338	43	196	0.975	507	178
157	1.04	777	89	197	x	x	x
158	1.14	860	131	198	1.22	147	3567
159	0.856	713	<16	199	1.13	135	3470
160	0.864	682	<33	200	1.17	108	3569
161	0.995	796	198	201	2.26	65	<29
162	0.100	< 5	<11	202	2.13	140	
163	0.827	858	<21	203	2.16	56	<14
164	0.815	681	139	204	0.598	32	1480
165	0.659	626	<17	205	x	x	x
166	0.800	649	45	206	x	x	x
167	0.823	709	104	207	x	x	x
168	0.664	666	22	208	0.581	1047	66
169	0.674	666	<26	209	0.540	948	89
170	0.820	652	123	210	0.580	1079	113
171	0.743	579	42	211	0.764	420	46
172	0.734	534	102	212	0.736	393	143
173	0.754	625	87	213	0.744	246	59
174	0.718	585	24	214	0.767	798	74
175	0.702	472	113	215	0.780	773	183
176	0.682	485	69	216	0.767	768	61
177	0.703	522	46	217	0.600	53	<20
178	0.705	504	76	218	0.570	56	< 8
179	0.732	760	< 9	219	0.581	40	<15
180	0.598	637	47	220	0.556	75	<17
181	0.731	813	<13	221	0.565	114	42
182	0.730	563	12	222	0.559	46	<17
183	0.719	573	14	223	0.534	70	<20
184	0.748	662	<15	224	0.630	104	<20
185	2.48	167	176	225	0.743	767	<23
186	2.43	216	178	226	0.741	751	16
187	2.42	759	427	227	0.731	729	35
188	2.51	740	296	228	1.22	272	75
189	1.45	217	40	229	1.27	391	21
190	1.30	124	52	230	1.22	271	149
191	0.905	547	109	231	1.24	449	<19
192	0.665	423	43	232	1.27	143	<26
193	0.753	892	202	233	1.25	177	32
194	0.747	808	72	234	1.05	180	79

Table 9 (Continued)

Sample	Sb, %	Cu, ppm	As, ppm
235	0.715	66	<18
236	0.568	27	16
237	0.584	22	<15
238	0.736	697	<20
239	0.832	863	37
240	0.718	459	63
241	0.691	486	75
242	0.710	477	72
134 Repeat	0.646	47	88
134 Repeat	0.589	< 9	16
135 Repeat	0.589	19	<10
135 Repeat	0.635	51	23
136 Repeat	0.720	1070	77
136 Repeat	0.684	1064	83
137 Repeat	0.735	905	91
137 Repeat	0.810	927	68

3. DISCUSSION

3.1 INITIAL EXPERIMENTS

Antimony concentrations, which were the chief focus of the work, have been seen to be very uniform within a single bullet (Tables 5 and 6) and within bullets from the same box of bullets, excepting perhaps the Peters 0.22-caliber bullets (Table 7). The usual standard deviation of the antimony concentration within either a single bullet or within a box of bullets is about ±3% of the concentration value. It is worthy of note, from Tables 5 and 6, that, because of the thickness uniformity of bullet slices, the antimony-122 photopeak specific activity was essentially independent of sample weight over a wide range of sample weights.

Among the bullets listed in Tables 1, 4, and 7 a majority have a distinctive antimony concentration. However, in some cases there is an overlap of values within the usual ±3% relative standard deviation: (1) a Remington-Peters 0.38-caliber bullet and a Remington 0.38-caliber bullet were found to contain 0.85% and 0.87% w antimony, respectively; (2) the Peters and Remington 0.22-caliber bullets were found to contain 0.87% and 0.85% w antimony, respectively; (3) the Lapua and Sears 0.22-caliber bullets contained 1.22% and 1.26% w antimony, respectively; (4) a U.S. Cartridge Co. and a Winchester 0.38-caliber bullet each contained 1.55% w antimony, and (5) a Peters and a Western 0.38-caliber bullet each contained very close to 3.00% w antimony. In cases (3), (4), and (5), however, the bullets could be distinguished from one another on the basis of trace-element constituents, while in cases (1) and (2) sufficient trace-element information was not gathered to determine if the pairs could be distinguished.

Those bullets for which upper limits of antimony concentration are reported can all be distinguished on the basis of trace-element information.

While it could be said from these results that bullets with different antimony concentrations have different origins, the data indicated that there was some chance that whole bullets with the same antimony level may have a different origin, and that there was some lesser chance that whole bullets with the same antimony and trace-element levels may have a different origin.

If the caliber of bullet cannot be determined, as when the sample consists of a bullet fragment, the foregoing overlapping cases number (1) and number (2) become a group of four indistinguishable bullets on the basis of antimony level, and in the absence of sufficient trace-element data. Similarly, a 0.30-caliber Remington rifle bullet and a 0.38-caliber Winchester bullet join the two bullets of overlapping case number (3), although the available trace-element information makes it possible to distinguish between the bullets in this group. Also, a 0.38-caliber Winchester bullet with 3.09% w antimony and a 0.30-caliber Sierra bullet with 3.10% w antimony overlap with respect to antimony, but can be distinguished by virtue of the large tin concentration in the latter.

The initial work established a basis upon which to judge a lack of commonality between bullets or bullet fragments with different antimony levels; and the consistency of antimony concentration found in single bullets and among bullets of common log origin is seen an important factor in establishing commonality between bullets or bullet fragments.

3.2 FINAL EXPERIMENTS

These experiments allowed the further examination of intra-box bullet uniformity, as shown in Table 10. Except that large variances are

Table 10

INTRA-BOX RESULTS MEAN VALUES AND STANDARD DEVIATIONS[a]

Samples	Sb, %	Cu, ppm	As, ppm	Samples	Sb, %	Cu, ppm	As, ppm
2-5	0.89 ± 0.03	940 ± 50	85 ± 67	137(3)	0.76 ± 0.04	924 ± 18	82 ± 12
7-10	1.66 ± 0.01	445 ± 35	730 ± 50	138-139	2.62 ± 0.01	800 ± 7	555 ± 53
12-15	0.67 ± 0.02	1680 ± 80	< 39	140-141	3.23 ± 0.01	255 ± 27	650 ± 27
17-20	0.30 ± 0.01	97(s.v.)	350 ± 130	151-152	2.52 ± 0.00	460 ± 24	< 68
22-25	0.188 ± 0.001	480 ± 30	530 ± 40	153-154	0.86 ± 0.06	195 ± 140	60 ± 52
26-30	1.88 ± 0.02	580 ± 220	525 ± 35	155-156	0.016 ± 0.004	210 ± 170	33 ± 14
31-34	0.77 ± 0.03	182 ± 120	114 ± 62	157-158	1.09 ± 0.07	820 ± 57	110 ± 30
35-39	0.86 ± 0.09	618 ± 180	158 ± 110	159-160	0.86 ± 0.01	700 ± 20	< 33
40-44	0.99 ± 0.04	910 ± 200	560 ± 300	163-165	0.77 ± 0.09	720 ± 120	60 ± 70
45-49	0.09 ± 0.07	45 ± 32	9 ± 2	171-173	0.74 ± 0.01	580 ± 70	77 ± 32
50-54	0.08 ± 0.06	21 ± 12	10 ± 7	179-181	0.69 ± 0.07	740 ± 90	22 ± 18
55-56	0.68 ± 0.05	735 ± 135	< 24	182-184	0.73 ± 0.01	600 ± 55	13 ± 1
57-58	0.71 ± 0.01	990 ± 40	< 17	185-186	2.45 ± 0.03	190 ± 36	177 ± 1
66-6.	0.739 ± 0.004	650 ± 35	20 ± 15	187-188	2.47 ± 0.06	750 ± 14	365 ± 90
68-69	2.30 ± 0.06	560 ± 310	470 ± 110	189-190	1.38 ± 0.10	170 ± 65	46 ± 8
70-71	[illegible]62 ± 0.06	155 ± 65	20 ± 15	191-192	0.79 ± 0.19	485 ± 90	76 ± 47
72-74	[illegible] ± 0.005	51 ± 6	22 ± 5	193-194	0.75 ± 0.01	850 ± 59	137 ± 90
75-76	0.[illegible] 0.05	620(s.v.)	985 ± 35	195-196	0.97 ± 0.01	526 ± 35	233 ± 78
77-78	1.02 ± 0.01	787 ± 1	335 ± 45	198-200	1.18 ± 0.06	141 ± 8	3520 ± 70
79-81	0.78 ± 0.07	500 ± 160	56 ± 37	201-203	2.18 ± 0.07	87 ± 46	
82-84	0.81 ± 0.01	560 ± 180	50 ± 40	208-210	0.51 ± 0.02	1030 ± 97	89 ± 24
85-87	0.76 ± 0.04	580 ± 110	74(s.v.)	211-213	0.75 ± 0.01	350 ± 130	83 ± 53
88-90	0.78 ± 0.04	490 ± 40	49 ± 12	214-216	0.77 ± 0.01	780 ± 28	106 ± 67
91-93	0.72 ± 0.01	240 ± 170	42(s.v.)	217-219	0.58 ± 0.01	50 ± 8	< 20
94-96	0.73 ± 0.06	340 ± 190	68 ± 1	220-222	0.56 ± 0.00	78 ± 34	20 ± 18
97-98	0.72 ± 0.01	125 ± 20	16(s.v.)	223-224	0.58 ± 0.07	87 ± 24	< 20
106-108	0.68 ± 0.01	750 ± 100	19 ± 4	225-227	0.74 ± 0.00	750 ± 20	25 ± 14
109-111	0.67 ± 0.05	660 ± 260	65 ± 64	228-230	1.24 ± 0.03	310 ± 70	82 ± 65
112-114	0.66 ± 0.01	770 ± 30	< 21	232-234	1.19 ± 0.12	167 ± 20	55 ± 33
121-122	1.00 ± 0.02	820 ± 110	285 ± 25	235-237	0.62 ± 0.08	38 ± 24	16(s.v.)
129-130	0.002 ± 0.001	4.1 ± 1.8	< 2	238-239	0.78 ± 0.06	780 ± 110	37(s.v.)
134-135(6)	0.63 ± 0.03	40 ± 13	40 ± 33	240-242	0.72 ± 0.01	474 ± 14	70 ± 6
136(3)	0.71 ± 0.02	1040 ± 40	70 ± 18				

[a] Based on variation between samples. When only one result is available, this is indicated by (s.v.), and no standard deviation is given.

associated with very low antimony concentrations, the magnitudes of the concentration variances are not well correlated with the concentration values. Thus, it is somewhat meaningful to note that the average standard deviations for Sb, Cu, and As are ±0.036% w, ±74 ppm, and ±45 ppm, respectively.

Also, the series of experiments permitted comparison of bullets from common lots but different boxes, as shown in Table 11. The average standard deviations for the elements in this context are Sb - ±0.031% w, Cu - ±67 ppm, and As - ±38 ppm. The fact that these averages are somewhat better than those cited in the foregoing paragraph (a result of larger numbers of individual samples represented in each value) indicates that the degree of uniformity of bullets within a box extends to the multiplicity of boxes from the same lot of bullets.

The mean values for antimony, copper and arsenic for each of the 75 different lots of bullets are given in Table 12. Where the lot is represented by a single sample, or value only, no standard deviation is given. It can be seen that, except for the 12 gauge, 2-3/4-in., rifled slugs, there are no significant differences with respect to concentration ranges or distributions of the elements between the various calibers. It is clear that it is not possible to determine the caliber of a projectile from the concentrations of the elements in question. On the other hand, the total set of 75 lots may be utilized in obtaining generalized statistics for all calibers.

The exploration of generalized statistics is facilitated by the ordering of results given in Table 12 with respect to the element of greatest intra-lot uniformity, antimony. This is done in Table 13.

An initial inspection of Table 13 reveals that $\sim$ 10% of the samples have low impurity concentrations ($\sim \leq$ 0.1%) of antimony, while the rest have antimony levels usually associated with deliberate antimony additions

Table 11

INTRA-LOT, INTER-BOX RESULTS

Samples	Bullets	Lot No.	No. of Boxes	Mean Values and Standard Deviations		
				Sb, %	Cu, ppm	As, ppm
57-61	Rem, .38 Special	P07G	4	0.76 ± 0.11	953 ± 140	lim.
55, 56, 62, 64	Rem, .38 Special	N06D	3	0.69 ± 0.03	747 ± 47	lim.
63, 65	Rem, .38 Special	K29H	2	0.74 ± 0.02	650 ± 95	87 ± 93
85-90	Rem, .44 S&W	M11E-20P	2	0.77 ± 0.05	535 ± 110	47 ± 17
91-105	Rem, .22 LR	Unknown	10	0.73 ± 0.02	270 ± 130	30 ± 26
106-108, 112-120	Rem, .357 Magnum	11P-P09D	8	0.69 ± 0.02[a]	740 ± 100	17 ± 4
109-111, 123, 124	Rem, .357 Magnum	11P-N14P	3	0.67 ± 0.01	580 ± 74	33 ± 45
125-127	W-W, .45 Auto	53-33BE01	3	0.045 ± 0.016	15 ± 1	lim.
132, 133	W-W, 12 G. Slugs	R11BD81	2	0.0356 ± 0.0017	3.1 ± 0.8	lim.
136, 137	Rem, 12 G. Slugs	AN12P18	2	0.74 ± 0.01	969 ± 39	69 ± 27
163-170	Rem, .38 Special	L19B	6	0.76 ± 0.07	678 ± 30	63 ± 40
171-178	Rem, .38 Special	J09P	6	0.71 ± 0.02	525 ± 50	68 ± 30
220-224	W-W, .22 Short	WK72	2	0.57 ± 0.04	82 ± 27	lim.
228-231	W-W, .22 LR	XB12	2	1.24 ± 0.02	380 ± 97	66 ± 61

[a] One value rejected on the basis of Chauvenet's Criterion.

Table 12

MEAN VALUES AMONG 75 LOTS OF BULLETS

Cal.	Mfg.	Lot	Sample Nos.	Mean Values and Standard Deviations Sb, %	Cu, ppm	As, ppm
.22 Short	W-W	BL4	22-25	0.188 ± 0.001	480 ± 30	530 ± 40
		Unk.	146	0.89	77	lin < 23
		XB82	217-219	0.58 ± 0.01	50 ± 8	lin < 20
		WK72	220-224	0.57 ± 0.04	82 ± 27	lin < 25
	Rem.	Unk.	145	0.72	730	42
		J12P2D	211-213	0.75 ± 0.01	350 ± 130	83 ± 53
		E1453F	225-227	0.74 ± 0.00	750 ± 20	25 ± 14
	Fed.	524C	214-216	0.77 ± 0.01	780 ± 28	106 ± 67
.22 LR	W-W	CD71	12-15	0.67 ± 0.02	1680 ± 80	< 39
		BK72	31-34	0.77 ± 0.03	182 ± 120	114 ± 62
		XB12	228-231	1.24 ± 0.02	380 ± 97	66 ± 61
		WLL626277351	232-234	1.19 ± 0.12	167 ± 20	55 ± 33
		YC2	235-237	0.62 ± 0.08	38 ± 24	16(s.v.)
	Rem.	Unk.	91-105	0.73 ± 0.02	270 ± 130	30 ± 26
		LO1R2B	238-239	0.78 ± 0.06	780 ± 110	37(s.v.)
		W24A2B	240-242	0.71 ± 0.01	474 ± 14	70 ± 6
	Fed.	LF4JC	26-30	1.88 ± 0.02	580 ± 220	525 ± 35
.22 Spec.	W-W	Unk.	148	2.39	419	lin < 17
		Unk.	149	2.50	329	46
	Rem.	Unk.	150	1.67	258	383
		Unk.	147	1.79	836	138
.25 Auto.	Rem.	L15ZD	35-39	0.86 ± 0.09	618 ± 180	158 ± 110
.32 Short R. F.	Rem.	321A45	151-152	2.52 ± 0.00	460 ± 24	lin < 68
.32 Auto.	W-W	88LA8	204	0.598	32	1480
.32 Short Colt	Rem.	Z221	208-210	0.57 ± 0.02	1030 ± 97	89 ± 24
.32 S&W	W-W	Unk.	153-154	0.86 ± 0.06	195 ± 140	60 ± 52
		Unk.	155-156	0.016 ± 0.004	210 ± 170	33 ± 14
		112140	198-200	1.18 ± 0.06	141 ± 8	3520 ± 17
		A4822	201-203	2.18 ± 0.07	87 ± 46	< 30
	Rem.	4R-M17C	179-181	0.69 ± 0.07	740 ± 90	22 ± 18
		3R-J30N	182-184	0.73 ± 0.01	600 ± 1	13 ± 1
		IRJ10U	195-196	0.97 ± 0.01	526 ± 35	233 ± 78
.357 Magnum	Rem.	11P-P09D	106-108 112-120	0.69 ± 0.02	740 ± 100	17 ± 4
		11P-N14P	109-111 123-124	0.67 ± 0.01	580 ± 74	33 ± 45
		11K-K29ED	121-122	1.00 ± 0.02	820 ± 110	285 ± 25

Table 12 (Continued)

Cal.	Mfg.	Lot	Sample Nos.	Mean Values and Standard Deviations Sb, %	Cu, ppm	As, ppm
.380 Auto.	Rem.	021C	77-78	1.02 ± 0.01	787 ± 1	335 ± 45
		Z242	159-160	0.86 ± 0.01	700 ± 20	lin < 33
.38 CNP	W-W	3625TD3	161	1.00	796	198
	W-W	Unk.	162	0.11	lin < 5	lin < 11
.38 S&W	W-W	9397YA5	17-20	0.30 ± 0.01	97(s.v.)	350 ± 130
		76-58RZ31	185-186	2.45 ± 0.03	190 ± 36	177 ± 1
		Unk.	187-188	2.47 ± 0.06	750 ± 14	365 ± 90
	Rem.	627N	2-5	0.89 ± 0.03	940 ± 50	85 ± 67
		3TY25M127	189-190	1.38 ± 0.10	170 ± 65	46 ± 8
		3TF27E165	191-192	0.79 ± 0.19	485 ± 90	76 ± 47
		9RG27N	193-194	0.75 ± 0.01	850 ± 59	137 ± 90
.38 Spec.	W-W	3528BE6	68-69	2.30 ± 0.06	560 ± 310	470 ± 110
		57BK7	70-71	0.62 ± 0.06	155 ± 65	20 ± 15
	Rem.	N06D	55, 56 62, 64	0.69 ± 0.03	747 ± 47	lin < 23
		P07G	51-61	0.76 ± 0.11	953 ± 140	lin < 16
		K29H	63, 65	0.74 ± 0.02	650 ± 95	87 ± 93
		M24R	66, 67	0.74 ± 0.00	650 ± 35	20 ± 15
		RA5289	75, 76	0.90 ± 0.05	620(s.v.)	985 ± 35
		L19B	163-170	0.76 ± 0.07	678 ± 30	63 ± 40
		J09P	171-178	0.71 ± 0.02	525 ± 50	68 ± 30
	Fed.	FPCS15KC	7-10	1.66 ± 0.01	445 ± 35	730 ± 50
		CS20KC	72-74	0.43 ± 0.00	51 ± 6	22 ± 5
	Speer	910001	138, 139	2.62 ± 0.01	800 ± 7	555 ± 53
		03003	140, 141	3.23 ± 0.01	255 ± 27	650 ± 27
		03005	143	0.00041	8.8	lin < 3
		03002	144	0.52	446	91
9 mm Lug.	W-W	45BC51	45-49	0.09 ± 0.07	45 ± 32	9 ± 2
		33BF7	50-54	0.08 ± 0.06	21 ± 12	10 ± 7
	Rem.	J23A	40-44	0.99 ± 0.04	910 ± 200	560 ± 200
.44 Rem. Mag.	Rem.	H09HG23LD	79-81	0.78 ± 0.07	500 ± 160	56 ± 37
		H09HH05SD	82-84	0.81 ± 0.01	560	50 ± 40
.44 S&W Spec.	Rem.	M11E-20P	85-90	0.77 ± 0.05	535 ± 110	47 ± 17
.45 Ball	Olin	WRA22690	157-158	1.09 ± 0.07	820 ± 57	110 ± 30
.45 Auto	W-W	53-33BE101	125-127	0.045 ± 0.016	15 ± 1	lin < 6
	Rem.	23PN13A	128	1.02	263	240
12 Gauge 2-3/4 in.	W-W	R11BD81	132, 133	0.036 ± 0.002	3.1 ± 0.8	lin < 6
	Rem.	BN22N17	129, 130	0.002 ± 0.001	4.1 ± 1.8	lin < 2
		AN22N17	131	0.001	2.3	lin < 2
12 Gauge 00 Buck	W-W	G62YL42	134, 135	0.63 ± 0.03	40 ± 13	40 ± 33
	Rem.	AN12P18	136, 137	0.74 ± 0.01	969 ± 39	69 ± 27

Table 13

LOTS, ORDERED BY ANTIMONY CONCENTRATION

Order No.	Lot (or Samples)	Cal.	Sb, %	Cu, ppm	As, ppm	Mfg.
1	03005	0.38	0.00041	8.8	< 3	Speer
2	AN22N17	12 G	0.0011	2.3	< 2	Rem.
3	BN22N17	12 G	0.002 ± 0.001	4.1 ± 1.8	< 2	Rem.
4	unk (155-156)	0.32	0.016 ± 0.004	210 ± 170	33 ± 14	W-W
5	R11BD81	12 G	0.036 ± 0.002	3.1 ± 0.8	< 6	W-W
6	53-33BE101	0.45	0.045 ± 0.016	15 ± 1	< 6	W-W
7	33BF7	9 mm	0.08 ± 0.06	21 ± 12	10 ± 7	W-W
8	45BC51	9 mm	0.09 ± 0.07	45 ± 32	9 ± 2	W-W
9	unk (162)	0.38	0.11	< 5	< 11	W-W
10	BL4	0.22	0.19 ± 0.00	480 ± 30	530 ± 40	W-W
11	9397YA5	0.38	0.30 ± 0.01	97	350 ± 130	W-W
12	CS20KC	0.38	0.43 ± 0.00	51 ± 6	22 ± 5	Fed.
13	03002	0.38	0.52	446	91	Speer
14	WK72	0.22	0.57 ± 0.04	82 ± 27	< 25	W-W
15	Z221	0.32	0.57 ± 0.02	1030 ± 97	89 ± 24	Rem.
16	XB82	0.22	0.58 ± 0.01	50 ± 8	< 20	W-W
17	88CA8	0.32	0.60	32	1480	W-W
18	YC2	0.22	0.62 ± 0.08	38 ± 24	16	W-W
19	57BK7	0.38	0.62 ± 0.06	155 ± 65	20 ± 15	W-W
20	G62YL42	12 G	0.63 ± 0.03	40 ± 13	40 ± 33	W-W
21	11RN14P	0.357	0.67 ± 0.01	580 ± 74	33 ± 45	Rem.
22	CD71	0.22	0.67 ± 0.02	1680 ± 80	< 39	W-W
23	11P-P09D	0.357	0.69 ± 0.02	740 ± 100	17 ± 4	Rem.
24	4R-M17C	0.32	0.69 ± 0.07	740 ± 90	22 ± 18	Rem.
25	N06D	0.38	0.69 ± 0.03	747 ± 47	< 23	Rem.
26	W24A2B	0.22	0.71 ± 0.01	474 ± 14	70 ± 6	Rem.
27	J09P	0.38	0.71 ± 0.02	525 ± 50	68 ± 30	Rem.
28	unk (145)	0.22	0.72	730	42	Rem.
29	unk (91-105)	0.22	0.73 ± 0.02	270 ± 130	30 ± 26	Rem.
30	3R-J30N	0.32	0.73 ± 0.01	600 ± 4	13 ± 1	Rem.
31	M24R	0.38	0.74 ± 0.00	650 ± 35	20 ± 15	Rem.
32	K29H	0.38	0.74 ± 0.02	650 ± 95	87 ± 93	Rem.
33	E14S3F	0.22	0.74 ± 0.00	750 ± 20	25 ± 14	Rem.
34	AN12P18	12 G	0.74 ± 0.01	969 ± 39	69 ± 27	Rem.
35	J12P2D	0.22	0.75 ± 0.01	350 ± 130	83 ± 53	Rem.
36	9RG27N	0.38	0.75 ± 0.01	850 ± 59	137 ± 90	Rem.
37	L19B	0.38	0.76 ± 0.07	678 ± 30	985 ± 35	Rem.
38	P07G	0.38	0.76 ± 0.11	953 ± 140	< 16	Rem.
39	524C	0.22	0.77 ± 0.01	78 ± 28	106 ± 67	Fed.
40	BK72	0.22	0.77 ± 0.03	182 ± 120	114 ± 62	W-W
41	M11E-20P	0.44	0.77 ± 0.05	535 ± 110	47 ± 17	Rem.
42	H09HG23LD	0.44	0.78 ± 0.07	500 ± 160	56 ± 37	Rem.

Table 13 (Continued)

Order No.	Lot (or Samples)	Cal.	Sb, %	Cu, ppm	As, ppm	Mfg.
43	L01R2B	0.22	0.78 ± 0.06	780 ± 110	37	Rem.
44	3TF27E165	0.38	0.79 ± 0.19	485 ± 90	76 ± 47	Rem.
45	H09HH05SD	0.44	0.81 ± 0.01	560	50 ± 40	Rem.
46	unk (153-154)	0.32	0.86 ± 0.06	195 ± 140	60 ± 52	W-W
47	L15ZD	0.25	0.86 ± 0.09	618 ± 180	158 ± 110	Rem.
48	Z242	0.38	0.86 ± 0.01	700 ± 20	< 33	Rem.
49	unk (146)	0.22	0.89	77	< 23	W-W
50	627N	0.38	0.89 ± 0.03	940 ± 50	85 ± 67	Rem.
51	RA5289	0.38	0.90 ± 0.05	620	985 ± 35	Rem.
52	1RJ10U	0.32	0.97 ± 0.01	526 ± 35	233 ± 78	Rem.
53	J23A	9 mm	0.99 ± 0.04	910 ± 200	560 ± 200	Rem.
54	3625TD3	0.38	1.00	796	198	W-W
55	11K-K29ED	0.357	1.00 ± 0.02	820 ± 110	285 ± 25	Rem.
56	23PN13A	0.45	1.02	263	240	Rem.
57	021C	0.380	1.02 ± 0.01	787 ± 1	335 ± 45	Rem.
58	WRA22690	0.45	1.09 ± 0.07	820 ± 57	110 ± 30	Olin.
59	112140	0.32	1.18 ± 0.06	141 ± 8	3520 ± 17	W-W
60	WCC626277351	0.22	1.19 ± 0.12	167 ± 20	55 ± 33	W-W
61	XB12	0.22	1.24 ± 0.02	380 ± 97	66 ± [illegible]	W-W
62	3TY25M127	0.38	1.38 ± 0.10	170 ± 65	46 ± 8	Rem.
63	FPCS15KC	0.38	1.66 ± 0.01	445 ± 35	730 ± 50	Fed.
64	unk (150)	0.22	1.67	258	383	Rem.
65	unk (147)	0.22	1.79	836	138	Rem.
66	LF4JC	0.22	1.88 ± 0.02	580 ± 220	525 ± 35	Fed.
67	A4822	0.32	2.18 ± 0.07	87 ± 46	< 30	W-W
68	3528BE6	0.38	2.30 ± 0.06	530 ± 310	470 ± 110	W-W
69	unk (148)	0.22	2.39	419	< 17	W-W
70	76-58RZ31	0.38	2.45 ± 0.03	190 ± 36	177 ± 1	W-W
71	unk (187-188)	0.38	2.47 ± 0.06	750 ± 14	365 ± 90	W-W
72	unk (149	0.22	2.50	329	46	W-W
73	321A45	0.32	2.52 ± 0.00	460 ± 24	< 68	Rem.
74	910001	0.38	2.62 ± 0.01	800 ± 7	555 ± 53	Speer
75	03003	0.38	3.23 ± 0.01	255 ± 27	650 ± 27	Speer

to achieve desired physical properties (chiefly hardness). Also, the dominance of Remington ammunition in the middle ranks of Table 13 is obvious.

The 75 lots included 39 lots of Remington bullets (including one lot from the parent company, Olin-Mathieson, and a number of lots from the subsidiary company, Peters), 28 lots of Winchester-Western bullets, four lots of Speer bullets, and four lots of Federal Cartridge Co. bullets. It is instructive to note the following distribution of brands within consecutive ranks of 25:

Order No.	Numbers of Lots			
	Rem.	W-W	Fed.	Speer
1-25	7	15	1	2
26-50	21	3	1	
51-75	11	10	2	2
All	39	28	4	4

The average relative standard deviations for the three elements in question are antimony – ±4% of the value (leaving out the lots with < 0.1% w Sb, which have quite large σ's), copper – ±23% of the value, and arsenic – ±44% of the value. Thus, antimony might be expected to be more definitive than the other two elements for comparison purposes. This is true at relatively low and high Sb concentrations, but (again referring to Table 13) not for the many lots of Remington bullets of middle ranks.

For example, let us say that an antimony value is distinct if it is 0.75% w and a comparison sample is not within 3σ (±0.09% w) of that value. If the 75 lots studied are representative of the population of bullets, then the fact that 25 of the lots have antimony values falling within that range indicates a large chance that, at this level, different bullets

will not be distinguished on the basis of Sb concentrations alone. If we relax the criterion to a range of $\pm 2\sigma$ (i.e., distinct if another bullet is not within 0.69-0.81% w Sb), we do not find much improvement – 23 lots of the 75 have Sb concentrations within that range.

The examination now turns to the question of aid from copper and arsenic in the middle ranks of bullets. Assume that a bullet in question has the following concentrations (with average standard deviations): 0.75 ± 0.03% w Sb, 400 ± 92 ppm Cu, and 100 ± 44 ppm As. Assume 2σ 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), unk (samples 91-105, rank 29), J12P2D (rank 35), M11E-20P (rank 41, H09HG23LD (rank 42), 3TF27E165 (rank 44), and H09HH05SD (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 elements do not have Gaussian concentration distributions among the 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:

$$\text{Sb: Cu} - R = 0.38$$

$$\text{Sb: As} - R = 0.55 \ .$$

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 13 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, which addressed 30 different lots of handgun ammunition, involved a smaller proportion of Remington and Winchester bullets, and the antimony values of the 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 purely instrumental NAA.

It is doubtful that the measurement of silver would have served to effect a unique determination among each of the 75 lots of bullets. While the earlier investigation solved the major difficulties in observing silver,

the analyzed precision finally achieved was still comparatively poor, and the obvious uniformity of bullets from a major supplier would undoubtedly extend to this element also. Note, for example, the similarity of Ag values (3.6 ppm and 3.1 ppm) in the Remington-Peters S&W and C. N. P. bullets of Table 4.

Concurrent work with other evidence materials, such as paper[3] and paint,[4] has shown with significant confidence that accidental matching of different samples has not occurred and 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 used to improve the possibility of observing a larger number of elements.

Although the present work has not provided a fully 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 two alternative approaches to the task are attractive. One approach would follow the course of post-irradiation 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 use of radiochemistry would be precluded only where the specimen was small and required as an exhibit.

The alternative approach would involve the distinctive tagging of bullet leads with combinations of elements, in small amounts, that would 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 the necessary alterations in manufacturing procedures, the tagging approach could be realized. With three appropriate 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.

Altogether the present study has provided essential information concerning the problem in question, and may be considered as a milestone in the progression toward a complete solution of the problem. The demonstrated intra-lot uniformity is an essential factor in the comparison of bullet leads via their elemental constituents, and it has been defined that ≥ 6 elements should be measured to achieve definitive comparisons via measurement of intrinsic constituents.

4. CONCLUSIONS

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 intra-lot 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 intra-lot 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.

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