

Robert D. Koons,<sup>1</sup> Ph.D. and Diana M. Grant,<sup>2</sup> Ph.D.

## Compositional Variation in Bullet Lead Manufacture\*

Dr. & Mrs. Jeffrey C. Reid  
8401 Summerspring Lane  
Raleigh, NC 27615

original

**ABSTRACT:** The concentrations of antimony, copper, tin, arsenic, silver, bismuth, and cadmium in lead alloys produced by two smelters and one ammunition manufacturer were determined using inductively coupled plasma-atomic emission spectrometry. These element concentrations were used to measure the variations in composition of lead products that result from various processes involved in the manufacture of lead projectiles. In general, when a pot containing molten lead is used to cast a number of objects, these objects are similar, although not necessarily analytically indistinguishable in their elemental compositions. In each subsequent step in the processing of lead at the smelter and at the ammunition manufacturer, the size of an individual homogeneous melt of lead decreases as more distinct compositions are formed as a result of remelting and mixing of sources, including lead scrap. The ammunition manufacturer in this study produced at least 10 compositionally distinguishable groups of bullet wire in a 19.7-h period. The largest group could potentially be used to produce a maximum of 1.3 million compositionally indistinguishable 40 grain bullets.

**KEYWORDS:** forensic science, criminalistics, trace evidence, bullet, lead analysis, elemental analysis, ICP-AES

Elemental composition is well established as a point of comparison for source discrimination for many types of materials that cannot be distinguished based upon their physical properties. The pattern of concentrations of elements, particularly trace elements, incorporated into a product at the time of its manufacture can serve as a chemical marker for a specific manufacturer, product line, production run, or, in some cases, individual object. Many naturally-occurring objects also contain a characteristic elemental profile that may be used as an indicator of provenance. The elemental profiles of many objects do not change significantly over long periods of time. This feature makes elemental profiles more useful for forensic comparisons than are the distributions of organic components, which have dissimilar volatilities and may be affected by sample degradation during normal conditions of product use. As a result of these factors, elemental composition has found widespread use in the comparison of many types of evidentiary materials (1-17).

One application where elemental analysis has proven particularly useful in criminal investigations is the compositional comparison of lead projectiles. In situations where a firearm is not recovered or cannot be associated with a subject, or a crime-scene bullet is too mutilated for association with a particular firearm, elemental composition of the lead component can be used as a means of comparison of two or more bullets. A variety of analytical methods, including neutron activation analysis (NAA)(18-24), atomic absorption spectrophotometry (24-26), spark source mass spectrometry (27-28), inductively coupled plasma-atomic emission spectrometry (ICP-AES)(23,29), and inductively coupled plasma-

mass spectrometry (ICP-MS)(30-32) have been successfully used for comparative bullet lead analysis over the past 30 years. Currently, the procedures used most frequently for this examination employ acid dissolution of the lead and analysis of the resulting solution using either ICP-AES (29) or ICP-MS (30). In a typical ICP-AES protocol, the concentrations of seven elements, arsenic, antimony, bismuth, cadmium, copper, silver, and tin are determined in replicate samples taken from each lead specimen for comparison. These seven elements have been selected for determination because they are present at measurable concentrations in most lead sources and they provide the best source discrimination capability (23,29,33,34).

Previous studies concerning analytical procedure development and interpretation of bullet lead data have established that considerable variability exists among lead sources despite manufacturer efforts at controlling the concentrations of selected elements (18,19,23,33). For example, ammunition manufacturers typically specify levels of antimony or, less frequently, tin to control the hardness of a lead alloy that is to be used for a particular product line (35). The concentration of arsenic is also specified for some products, primarily lead shot. The smelters are tasked with supplying an ammunition manufacturer with a lead product that falls within specified tolerances for the concentration of each element of interest. However, these tolerance ranges are much greater than both the variations that occur within an individual product from the smelter and the analytical variations in ICP-AES measurements. Therefore, differences in the concentrations of alloying elements between two objects made from nominally the same lead alloy are readily measurable by ICP-AES or other precise analytical techniques. In addition to differences in alloying elements, measurable differences in the trace element profiles of bullet lead also exist between manufacturing production runs of a given lead product. These differences are the result of different levels of trace constituents in raw materials and mixing of lead scrap from multiple sources during reprocessing of the lead, which can occur several

<sup>1</sup> Research chemist, Forensic Science Research Unit, FBI Laboratory, FBI Academy, Quantico, VA.

<sup>2</sup> Forensic examiner, Chemistry Unit, FBI Laboratory, Washington DC.

\* This is Publication No. 02-03 of the Laboratory Division of the Federal Bureau of Investigation (FBI). Names of commercial manufacturers are provided for information only, and inclusion does not imply endorsement by the FBI.

Received 3 Dec 2001; and in revised form 6 April 2002; accepted 7 April 2002; published 31 July 2002.

times during the manufacture of a final product. Often, the lead is melted at both an initial processing plant, such as a secondary smelter, and again at the ammunition manufacturing plant. Therefore, the trace constituents, which are introduced in these remelting operations, can provide a basis for discrimination between melts of lead, even those having the same alloy specification. The forensic utility of compositional comparison of bullet leads lies in the fact that there are many millions of possible combinations of these element concentrations.

In a previous study, all bullets in 16 boxes of cartridges from the four major U.S. ammunition manufacturers were analyzed (23). The variations in element concentrations within individual bullets, among bullets in a box of cartridges, among boxes packaged on the same and on different dates, and among boxes from different manufacturers were evaluated. The measured element concentrations were used to establish compositional groups among the bullet leads within each box. Specimens were placed in the same compositional group when they were determined to be analytically indistinguishable from each other for all of the measured elements. The results of this study have been combined with many years of experience in analyzing evidentiary specimens to establish two facts concerning the distribution of bullet compositional groups among boxes of cartridges. The first is that, for most boxes of cartridges, the lead components of the bullets in the box form more than one compositional group. The multiple compositions within a box of cartridges result from the mixing of bullets from several melts during the bullet forming, jacketing, cartridge loading, and packaging processes. The second well-established fact concerning the distribution of compositional groups of bullet leads is that a given compositional profile is repeated in boxes of cartridges produced by a single manufacturer over some period of time. The inclusion of bullets from a single melt into multiple boxes is a consequence of the obvious fact that the number of bullets produced from a homogeneous melt of lead exceeds the number of cartridges in a box. For most products, boxes containing the same compositional group are produced within a short time, such as one packaging run. However, in some instances, bullets may be stored for periods as long as several months prior to being loaded into cartridges and placed into boxes (23).

Patterns of variations of bullet lead compositions within boxes of cartridges are well established. Less well known, however, are the effects that the various manufacturing processes have on the distribution of element concentrations among the leads produced. In order to refine estimates of the last homogeneous source of lead in the manufacturing process and, hence, the number of bullets that can be produced from that source, further research was undertaken. The goal of this study was to monitor the homogeneity within melts of lead and to study the compositional profiles of several consecutive melts from individual producers. Two secondary smelters and an ammunition manufacturer provided specimens for this study. All specimens were analyzed using ICP-AES. The results of analysis of these samples are used to demonstrate the variations in elemental composition in lead projectile products that result from several steps in typical manufacturing processes.

## Experimental Details

### Sample Description

Specimens provided by Smelter 1 had been retained by the producer from batches of nine different lead alloys that were produced during the period from September 1997 through August 1999. At Smelter 1, lead is cast from a kettle containing approximately

200000 lb (90700 kg) of molten lead in forms including 2000-lb (907-kg) and 1000-lb (453-kg) blocks, 125-lb (57-kg) billets, and 60-lb (27-kg) ingots for various customers. During the pouring of this lead, small disks are cast at periodic intervals. The manufacturer analyzes these disks by spark source optical emission spectroscopy (SSOES) to ensure that trace impurity levels are below the established thresholds and that antimony content is within 10% of the specified value for that alloy. After the casting of a lot of lead is completed, one of the disks is retained for quality control purposes. These quality control specimens were provided by Smelter 1 as samples for this study. The nominal antimony content of these leads ranges from 0.5% to 6%. The SSOES data that were provided by the manufacturer indicate that each kettle of lead was within their allowable specifications prior to pouring. The specimen cast disks provided were approximately 7.4 cm in diameter and 1.6 cm thick.

Specimens provided by Smelter 2 originated from three lots of a lead alloy specified by the manufacturer to contain a nominal level of 0.6 to 0.8% antimony. One lot (lot A) was cast over the course of four to five hours in November 1999. The second and third lots (lots B and C) were cast on the same day in December 1999 from the same pot. Smelter 2 melts lead in two kettles, each containing 150000 lb (68000 kg) of lead. The lead is cast into 60-lb (27-kg) ingots. Small cast disks were poured at one-half hour intervals throughout the casting process for all three lots. In the production of lot B, as the casting pot was nearing  $\frac{1}{4}$  full, the second pot was pumped into the first over a period of one hour. During this time casting continued as lot B. When refilling of the pour pot was complete, the lot designation was switched to lot C. The smelter, using SSOES, determined that the contents of each pot were within the specifications of the 0.6 to 0.8% antimony alloy prior to beginning to pour each lot. Each sample was received as a cast disk, approximately 4 cm in diameter and 0.7 cm thick.

Specimens were provided by the ammunition manufacturer in the form of extruded wire. This ammunition manufacturer receives lead from a smelter and remelts it in order to produce the billet shape needed for extrusion. Each wire specimen represents a billet of lead produced from the on-site kettle at the bullet manufacturer's plant. This kettle, which contains up to 7500 lb (3400 kg) of molten lead is alternately fed from two different furnaces: one containing lead alloys up to 1% antimony, and one containing alloys of 3 to 5% antimony. On the day that the samples for this study were collected, the nominal alloy compositions in the two furnaces were 0.85% and 3% antimony. Each furnace has the capacity to hold 24000 lb (10886 kilograms) of lead. When alloys from the two furnaces are cast interchangeably, the casting kettle is reduced to a volume of approximately 350 lb (160 kg) of molten lead prior to a switch between active furnaces. The molten lead in the kettle is pumped out into billet molds to cast 325-lb (147-kg) lead cylinders, which are then sent down a conveyor belt to an operator who aligns each billet for vertical extrusion out of the top of the assembly. The extruded wire has a diameter that corresponds to the caliber of bullet that will be produced from that wire. The extruded wire is coiled around a large spool and wheeled to a separate machine where it is truncated into smaller sections, or slugs. The slugs are then machine-swaged to form the overall bullet shape. Further processing of the lead might include the addition of a coating or plating to the lead, or alterations to the nose shape to produce a different bullet style. However, since these latter changes are physical, the last operations that would affect the chemical composition of the bullet core are the mixing of components in molten lead in the pour kettle and the billet casting and extrusion operations. The samples for

this study were collected from 169 wire coils extruded at approximately 7-min intervals over a 19.7-h period by the personnel performing the casting and extrusion operations. The 169 wire samples were numbered from 1 through 170 in order of their extrusion with the number 18 not used. The specimens consist of two different alloy specifications and four diameters. Samples were removed as pieces about 30-cm in length from one end of each approximately 325-lb coil of bullet wire at the manufacturing facility.

#### Sample Preparation and Analysis

Samples were obtained from the cast disks from the smelters by cutting three wedges from each of the disks using an electric jigsaw (34). Using wire cutters, each wedge was sectioned into either three layers (Smelter 2) or five layers (Smelter 1) oriented from the top to the bottom of the original disk. The wire cutters were cleaned between specimens by making cuttings into and wiping with an ethanol-soaked paper tissue. This preparation procedure provided nine (Smelter 2) or fifteen (Smelter 1) subsamples from each disk.

Representative samples of the wires received from the ammunition manufacturer were obtained by taking samples from three positions along each wire. Wire with a diameter of less than approximately 0.5 cm had been crimped into three or four evenly-spaced, bullet-length slugs by the manufacturer. Cuttings were taken from three of the separate crimped sections. Wire having a larger diameter (approximately 0.75 cm or greater) was provided as undisturbed sections that had been cut from the beginning of each extruded wire coil. Samples were taken from three locations along the wire using wire cutters that were cleaned with ethanol and a coarse tissue paper between specimens.

Each analytical sample was examined under a stereomicroscope, any visible contamination or original surface material was removed, and the sample was trimmed to approximately 75 mg using a separate, clean scalpel. Each lead fragment was flattened between two sheets of polyethylene and the resulting disk was weighed to the nearest 0.01 mg and placed in a 15-mL polyethylene tube. The samples were dissolved using a procedure similar to that reported by Peters et al. (29). To each tube, a volume of 1:5 (v:v) nitric acid (HNO<sub>3</sub>) equal to 100 times the mass of the lead sample was added. The tubes were capped tightly and heated in a microwave oven for 40 s to initiate dissolution. The warm tubes were placed in an 80°C oven for 2 h, or as needed, to completely dissolve the samples. After removal from the oven and cooling to room temperature, 10 µL of hydrofluoric acid (HF) was added to each tube. For the ammunition manufacturer samples, which were known to contain higher concentrations of tin, the HF was added prior to heating the samples to prevent the formation of sparingly-soluble tin oxide. Sets of digestion blanks and standard reference materials SRM2416, Bullet Lead, SRM2415, Battery Lead, and SRM2417, Lead Base Alloy (National Institute of Standards and Technology (NIST), Gaithersburg, MD) were prepared along with each batch of samples to serve as quality checks.

Calibration standards were prepared by dissolution of accurately weighed portions of SRM 2416 and a 99.9999% pure lead wire (Alfa products, Danvers, MA). Triplicate samples consisting of the appropriate mixture of HNO<sub>3</sub> and HF were also prepared and analyzed as digestion blanks with each set of samples. An analytical run consisted of a set of calibration standards, followed by one replicate each of the three standard reference materials, the digestion blank, and the individual samples. Thus, three analytical runs were made for each set of samples. For the 169 ammunition manufacturer specimens, recalibration was performed after approximately every 75 analytical samples. All samples, blanks, and

TABLE 1—ICP-OES Operating Conditions.

Instrument Configuration			
Model	Perkin-Elmer Optima 3300DV		
Optical System	Echelle polychromator, N <sub>2</sub> purged		
Detector	Segmented array CCD		
RF Power Supply	40 MHz		
Induction coil	4 turn with internal cooling		
Torch	Quartz with alumina injection tube		
Nebulizer	GemCone, low flow		
Spray chamber	Quartz cyclonic		
Operational Parameters			
Plasma power	1400 W		
Plasma orientation	Axial		
Argon flows			
Plasma	15 L/min		
Auxiliary	0.5 L/min		
Nebulizer	0.6 L/min		
Sample uptake rate	1.5 mL/min		
Sample flush rate	3.0 mL/min		
Sample read time delay	40 s		
Element	Wavelength (nm)	Standard Conc (µg/mL)	Detection Limits (µg/mL)
Ag	328.07	0.44	0.0015
As	188.98	5.6	0.03
Bi	223.06	10	0.015
Cd	214.44	0.1	0.0007
Cu	324.75	6.5	0.005
Sb	206.84, 203.98	79	0.025, 0.5
Sn	189.93	9	0.015

standards were loaded into an autosampler for automatic introduction into an Optima 3300 DV ICP-AES instrument (Perkin Elmer, Norwalk, CT) for analysis. Instrumental operating conditions for the determination of arsenic, antimony, bismuth, cadmium, copper, silver, and tin are shown in Table 1 along with the 3-sigma detection limits determined from replicate measurements of the lead blank solution. The accuracy of results for all SRM samples was verified to agree with the values listed on the NIST Certificate of Analysis before inclusion of the sample results in this study.

## Results and Discussion

### Long-Term Variations of Lead Products from Smelter 1

The samples received from Smelter 1 had been cast as disks. Past studies have shown that when molten lead is poured into billets and cooled slowly, vertical segregation of elements can occur with lighter elements rising to the top of the denser lead (34). In fact, it is common practice in some bullet manufacturing facilities to remove the top surface of each billet for recycling by cutting it off prior to extrusion of the billet into a bullet wire (35). As a matter of interest, possible vertical variation in composition within the disks was assessed by comparison of element concentrations in the five vertically-sequenced slices from each disk. For each element, the concentrations in two adjacent layers, using triplicate measurements from the three wedges of a source, were compared using the t-test of means at a significance level of 0.05 for each element. Out of 456 comparisons, there are 33 instances in which differences that are both statistically and analytically significant exist between an element's concentrations in two adjacent layers. There are no observed consistent patterns for a specific element or for given positions across all disks. The number of adjacent layer pairs that differ is greater for antimony and arsenic (ten instances each) than for the other elements. In some of the disks, the concentrations of one

or more elements are slightly higher in the top half of a disk than the bottom half. The degree of vertical segregation of elements in lead billets made by Smelter 1 for bullet manufacture cannot be determined from measurements of the small disks used in this study because of the differences in cooling rates and sizes between the tested samples and the much larger billets. Instead, the mean concentration value for each element in the five vertically-oriented replicates in each wedge was used to provide a measure of the average composition of that wedge.

Comparison of the results for the three horizontally-oriented wedges using the t-test indicates that there is no significant lateral heterogeneity of element distributions among the Smelter 1 specimens. For intercomparison of the specimens, the five replicates for each wedge were averaged, resulting in three mean measurements per specimen. The overall means and one standard deviation based on the three wedge-sample means from each disk are shown in Table 2. For every sample, the concentration of one or more elements differs significantly from those of every other sample from Smelter 1. The differences between samples are so great compared with the variation within each sample that any of the usual tests used for forensic comparison of samples (such as the t-test, 2-sigma match criterion, or range overlap criterion) readily distinguishes between any pair of samples. For most pairs, the concentrations of antimony can be used to distinguish between the melts. This is true even for pairs having the same nominal antimony content. In the cases where the same alloy was made in two production runs, whether it was for the same or different ammunition manufacturers, the melts are readily distinguished by their trace element concentrations.

*Short-term Variations in Lead from Smelter 2*

Spatial heterogeneity of the disks from Smelter 2 was assessed using the 3 × 3 sampling scheme in the same manner as the disks from Smelter 1. Using the t-test at a significance level of 0.05, no significant differences were observed among the nine replicates of any element, either in a vertical or horizontal direction for each disk. This homogeneity of samples from Smelter 2 may be a result of their being cast as thinner disks (0.7 cm) than those from Smelter 1 (1.6 cm). For comparison of samples by lot number and individual ingots, the overall mean and standard deviation of the nine replicates of each sample disk were used. The measured concen-

trations of Sb, Cu, As, Bi, Ag, and Sn in the specimens obtained from Smelter 2, representing three lots of lead specified to contain 0.6 to 0.8% antimony are shown in Fig. 1. Cadmium was not present at detectable levels in any of the Smelter 2 samples. From the smelter's and ammunition manufacturer's standpoints, each of these lots is produced to the same specifications. However, as shown, the three lots of lead can be readily distinguished by the concentrations of several elements, particularly copper, arsenic, and tin. The eight samples from lot A, produced on 11/29/99, are analytically indistinguishable from each other, but are significantly different from the samples in lots B and C. During the production of lot B on 12/08/99, the first six samples are indistinguishable. Following the sixth sample, the effects of mixing of the two sources on element concentrations in the cast lead are seen. For example, as the second pot of molten lead was pumped into the first pot, the tin concentration began to drop, from greater than 300 ppm to below detection limits (<1.5 ppm) over the next ten samples. Although the element concentrations eventually level off somewhat following the initial mixing, the samples from lot C exhibit compositional differences throughout the entire production run, particularly for antimony and arsenic. Despite the fact that antimony is nominally controlled at a level of 0.7% in the melt, its concentrations differ significantly among the three lots and among individual samples in lots B and C. The size of each ingot in this production run is approximately 60 lb (27 kg) and a production lot consists of up to three-quarters of a 75-ton pot, or approximately 51000 kg of lead. Measurable changes in concentration in the output of the smelter occur on a scale as short as 30 min (the minimum sampling frequency in this study) corresponding to production of 7200 kg of lead. During the production period covered in this study, the smallest analytically distinct unit is 30 min of production (for example, Sample 7 of lot B) and the largest is 4 h of production (all samples of lot A).

*Variation in Lead Bullets from the Ammunition Manufacturer*

The concentrations of six elements determined in the 169 samples from the ammunition manufacturer are shown in Fig. 2. The mean ± one standard deviation for each element based on the three measurements are shown for each sample in the order that the wires were extruded. It is readily apparent from the results shown in Fig.

TABLE 2—Results of analysis of retain samples from lead Smelter 1. Concentrations listed are in µg/g (ppm). nd = not detected.

Alloy	Customer	Pour Date	Sb	Cu	Ag	Bi	Sn	As	Cd
0.5	#2	09/06/97	5510 ± 50	28.2 ± 0.4	26.3 ± 0.4	90.5 ± 0.4	1.2 ± 0.3	1.3 ± 0.8	nd
0.75	#1	08/13/99	6570 ± 40	38.2 ± 0.4	44.7 ± 0.5	93.3 ± 0.5	0.7 ± 0.2	5.8 ± 0.6	0.7 ± 0.0
0.75	#1	01/04/99	6390 ± 40	92.6 ± 0.7	33.3 ± 0.3	86.8 ± 0.3	1.1 ± 0.1	1.7 ± 0.5	0.4 ± 0.0
1.25	#2	04/09/98	12850 ± 150	35.3 ± 0.8	31.8 ± 0.4	88.5 ± 0.9	0.5 ± 0.1	13 ± 6	nd
1.25	#2	11/29/98	12220 ± 110	21.2 ± 0.2	28.7 ± 0.4	91.0 ± 0.7	1.3 ± 1.0	nd	0.3 ± 0.0
1.5	#1	10/07/97	14300 ± 300	5.8 ± 0.2	35.2 ± 1.0	92.8 ± 1.8	427 ± 6	11 ± 9	nd
1.5	#2	10/30/98	15380 ± 120	37.5 ± 0.5	23.9 ± 0.3	88.9 ± 0.4	0.7 ± 0.1	3890 ± 50	nd
1.5	#2	11/22/97	14350 ± 60	21.1 ± 0.3	24.6 ± 0.2	88.5 ± 0.1	0.4 ± 0.4	3680 ± 30	nd
2	#1	07/03/99	18860 ± 140	35.0 ± 0.3	32.9 ± 0.2	97.5 ± 0.7	0.6 ± 0.0	5280 ± 40	nd
2	#1	08/12/99	18740 ± 120	24.1 ± 0.2	47.7 ± 0.4	95.0 ± 0.9	0.5 ± 0.1	5270 ± 40	nd
2	#2	11/04/97	18810 ± 50	33.15 ± 0.08	23.6 ± 0.1	92.1 ± 0.4	0.2 ± 0.2	4915 ± 12	nd
2.25	#2	10/21/97	23390 ± 180	17.6 ± 0.3	24.1 ± 0.2	84.9 ± 1.0	1308 ± 10	18 ± 2	nd
2.5	#2	01/02/98	25370 ± 140	19.9 ± 0.2	25.5 ± 0.2	103.9 ± 1.0	0.2 ± 0.2	8 ± 5	nd
3	#2	02/22/99	30200 ± 500	14.9 ± 0.4	47.3 ± 1.1	109.5 ± 1.5	0.3 ± 0.1	6500 ± 140	nd
3	#2	02/01/99	30420 ± 150	41.2 ± 0.3	48.4 ± 0.4	135.0 ± 0.9	0.1 ± 0.1	6950 ± 60	nd
6	#1	01/13/99	57830 ± 50	24.6 ± 0.7	53.6 ± 0.2	147.4 ± 0.3	0.6 ± 0.2	15410 ± 80	nd
6	#1	01/06/99	59390 ± 190	9.00 ± 0.02	49.4 ± 0.2	109.7 ± 0.9	0.3 ± 0.1	15190 ± 80	nd
6	#2	10/30/98	56900 ± 500	7.82 ± 0.03	91.7 ± 1.0	130.6 ± 0.3	0.4 ± 0.0	14330 ± 140	nd
6	#2	11/22/97	54200 ± 400	10.5 ± 0.2	38.8 ± 0.4	224.8 ± 1.4	0.8 ± 0.2	14060 ± 80	nd

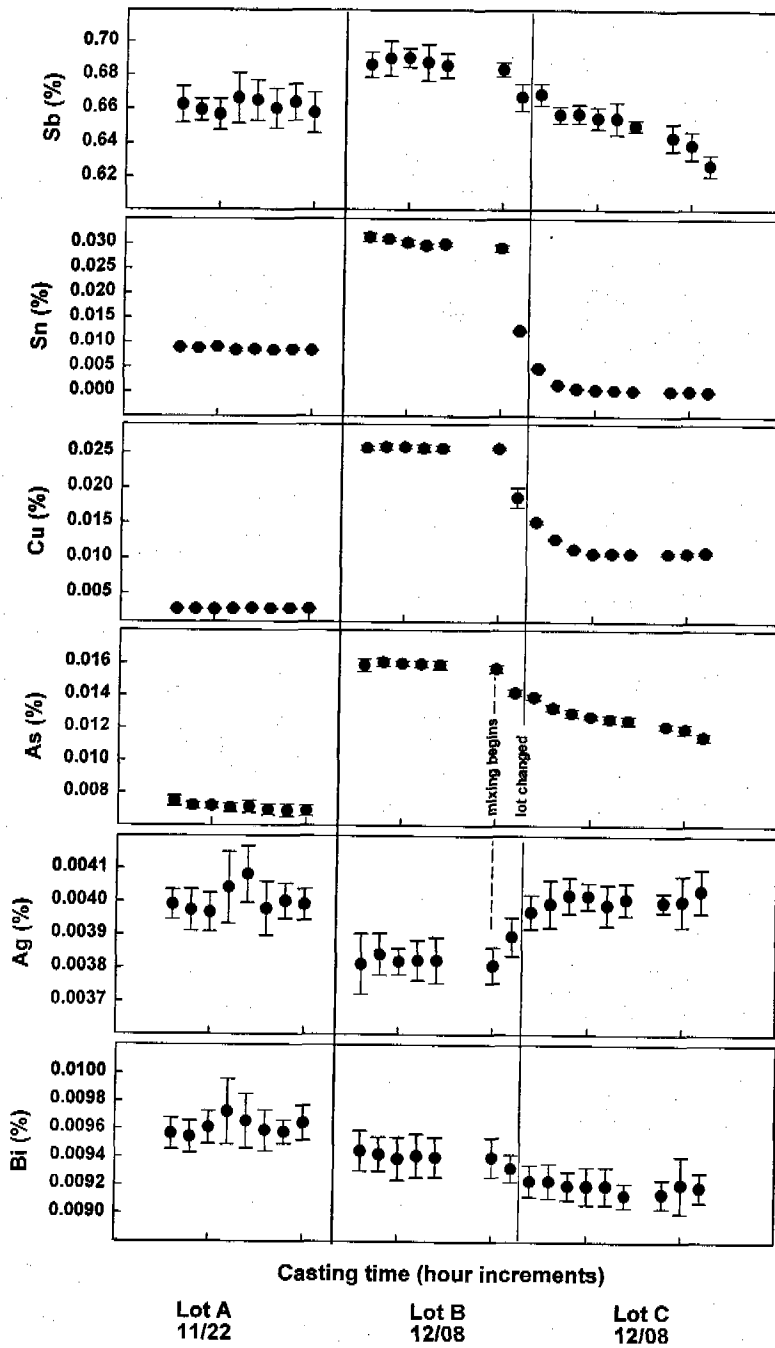


FIG. 1—Results of analysis of samples from Smelter 2. Circle indicates mean of nine replicate measurements and bar length indicates one standard deviation.

2 that wires that were extruded over short periods of time group together in relatively narrow composition ranges. These production groups correspond to those billets produced from each pour pot loading. The divisions between samples corresponding to transitions between the 0.85% and 3% antimony alloys can easily be seen in Fig. 2. Additionally, the production of two consecutive pours of billets of the same alloy can also be seen. For ease of discussion, the 12 groups seen in Fig. 2 have been labelled A through L.

The means and standard deviations of the element concentrations calculated from the wire means in each of these 12 groups are shown in Table 3. Comparison of the group compositions, whether by visual observation of Fig. 2, by the t-test of group means, or by a 2-sigma match criterion, all yield the conclusion that the 12 production groups form 10 compositionally-distinct groups. The pairs A-D and I-K are the only two paired groups that are analytically indistinguishable in the concentrations of all six elements. In this instance, the groups I and K were extruded to different diameters (0.348 in. and 0.310 in., respectively), because they were being

made into bullets of different calibers or jacket styles. The groups A and D are the only pair of groups that are indistinguishable based on both their compositions and diameters. As a result, these 29 wires could have been used to produce physically and compositionally indistinguishable bullets.

In this study, relatively short leading ends were analyzed to represent each extruded wire. The variability in element concentrations throughout an entire roll of wire was not directly measured in this study and, therefore, can only be inferred from this information. The variations in composition from one wire to the next within each production group are not much greater than the standard deviations calculated from the triplicate measurements of each wire. Therefore, it can be inferred that the short segments are representative of the whole wire and, hence, of all bullets produced between the samples provided in this study. The level of variation observed within wires from a group in this study is similar to the magnitude of variations reported among bullets from a single composition group within a box of cartridges (23). The consistency of

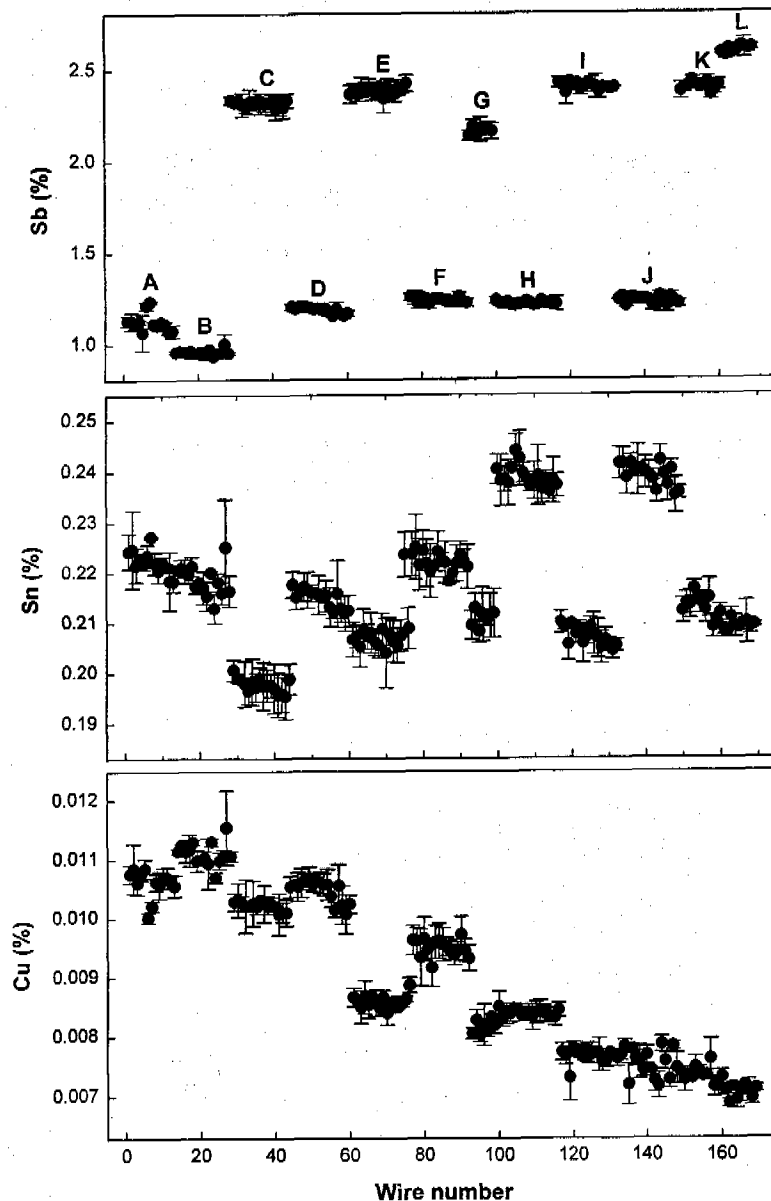


FIG. 2—Results of analysis of samples from the ammunition manufacturer. Circle indicates mean of three replicate measurements and bar length indicates one standard deviation.

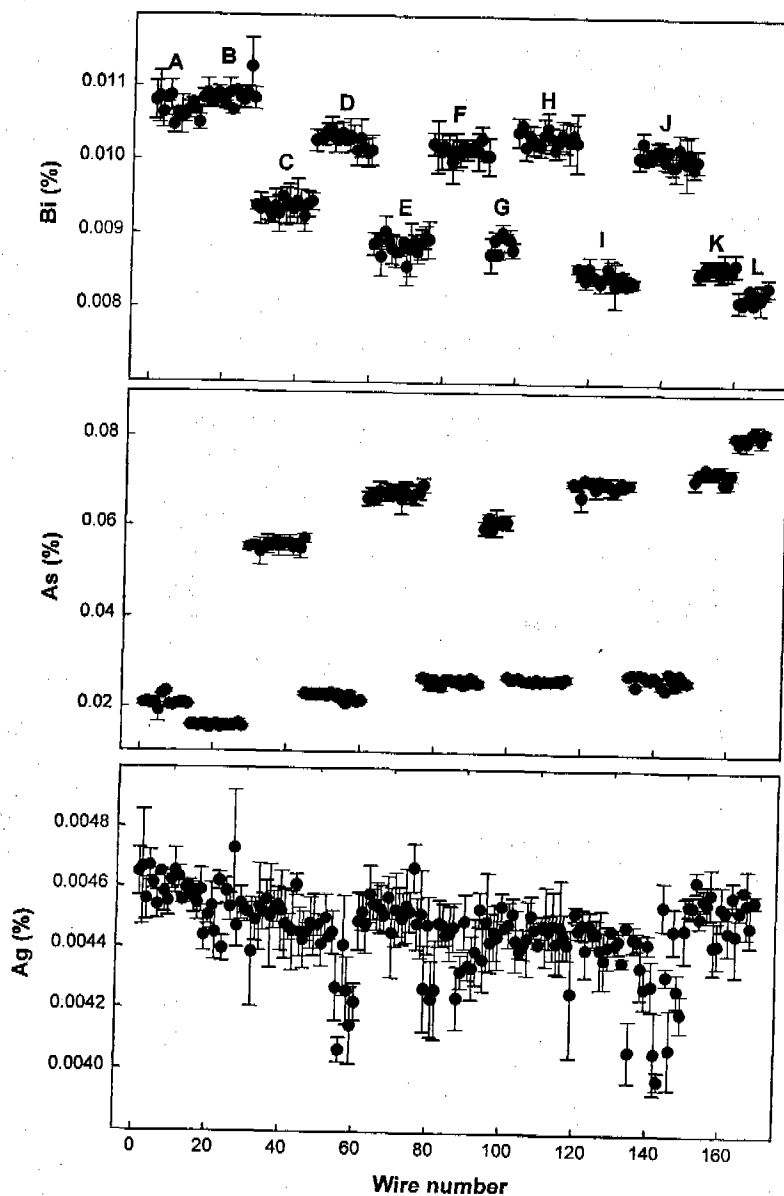


FIG. 2—(Continued).

compositions from one production run to the next of a given alloy is a strong indication that the lead going into the melting pot to form that alloy comes from one production run of a single smelter, much like the samples seen in the Smelter 2 study, and that recycled material added to the melting pot comes predominantly from scraps from the same source. The number of compositionally-indistinguishable rolls of wire seen in this study ranges from 5 (group L) to 29 (groups A and D), corresponding to approximately 1750 to 10200 lb (735 to 4260 kg) of cast lead. Allowing for losses in extrusion, swaging, and trimming operations, the largest group of lead could be used to make as many as 1.3 million compositionally-indistinguishable 40 grain, .22 caliber bullets.

### Conclusions

The results of analysis of the samples from Smelter 1 indicate that a variety of compositions are present in the products produced by a single smelter. There is good discrimination among separate melts, even several that are made to the same alloy specifications.

The number of samples from this smelter is too small to make a strong statistical statement concerning the likelihood that a given compositional pattern produced at one time would reoccur by chance at a later date. No measure concerning the compositional variation within a lead product was obtained in this portion of the study. Such information and any effects introduced by subsequent remelting, source mixing, and extrusion processes during bullet manufacture would be required prior to performing quantitative frequency of occurrence calculations.

The results of analysis of the samples from Smelter 2 show that the compositional homogeneity is good within cast leads produced from a well-mixed kettle of molten lead. Consecutive pours without replenishment of a pot can produce tons of lead having similar, though in some cases, distinguishable, compositions. During the operation studied here, mixing of lead from two pots results in the production of lead with intermediate compositions for several pours for a period of time during switchovers.

The samples from the ammunition manufacturer display similar, although generally analytically distinguishable, compositions for

TABLE 3—Results of analysis of samples from the ammunition-manufacturer divided into production groups. Concentrations are given in percent by weight.

Group	Number of Wires	Cu	Sb	Ag	Bi	Sn	As
A mean	13	0.0106	1.13	0.00463	0.0107	0.21	0.0210
A stdev		0.0003	0.06	0.00010	0.0002	0.02	0.0011
B mean	15	0.0111	0.959	0.00454	0.01086	0.219	0.0159
B stdev		0.0002	0.015	0.00008	0.00013	0.003	0.0002
C mean	16	0.01024	2.316	0.00451	0.00936	0.1977	0.0557
C stdev		0.00011	0.014	0.00005	0.00007	0.0014	0.0007
D mean	16	0.0105	1.187	0.00437	0.01027	0.2150	0.0225
D stdev		0.0002	0.017	0.00013	0.00010	0.0019	0.0007
E mean	16	0.00859	2.386	0.00453	0.00884	0.207	0.0673
E stdev		0.00011	0.020	0.00005	0.00011	0.004	0.0008
F mean	16	0.00950	1.237	0.0044	0.01015	0.2221	0.0261
F stdev		0.00015	0.013	0.0001	0.00007	0.0018	0.0006
G mean	7	0.00817	2.163	0.00444	0.00888	0.2109	0.0609
G stdev		0.00011	0.019	0.00005	0.00012	0.0016	0.0009
H mean	17	0.00839	1.219	0.00447	0.01032	0.239	0.0266
H stdev		0.00005	0.008	0.00004	0.00009	0.002	0.0003
I mean	16	0.00767	2.403	0.00443	0.00844	0.2072	0.0696
I stdev		0.00011	0.017	0.00006	0.00008	0.0018	0.0010
J mean	17	0.0075	1.230	0.00430	0.01009	0.239	0.0270
J stdev		0.0002	0.018	0.00017	0.00010	0.002	0.0011
K mean	15	0.0073	2.45	0.00453	0.00846	0.212	0.074
K stdev		0.0002	0.09	0.00006	0.00019	0.003	0.004
L mean	5	0.00708	2.605	0.00455	0.00821	0.2094	0.0812
L stdev		0.00008	0.009	0.00005	0.00009	0.0004	0.0007

the same alloy produced from different production runs, even when another alloy is produced in between. The homogeneity of a wire drawn from a single billet, which is inferred from the similarity of wires at the beginning of consecutive billets, is quite good for the samples in this study. The maximum number of bullets with indistinguishable compositions that could have been produced during the course of this study is more than one million. For any given evidentiary projectile, the forensic examiner can determine an approximate maximum number of potentially indistinguishable bullets by considering the manufacturing processes at the appropriate bullet manufacturing plant.

**Acknowledgments**

The authors would like to thank the corporations and their representatives who supplied the specimens and gave so generously of their time and expertise. This work was further made possible by the diligent efforts of FBI Laboratory Chemists April A. Getty and Stephanie E. Lenzenweger.

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**Additional information and reprint requests:**

Robert D. Koons, Ph.D.  
Research Chemist  
FSRTC, FBI Academy  
Quantico, VA 22135  
E-mail: rkoons@fbiacademy.edu