

The Application of Lead Isotope Analysis to Bullet Individualization in Two Homicides*

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ABSTRACT: Bullets were characterized by lead isotope ratio analysis and trace element analysis in two homicides. In one case, we concluded that a fatal bullet did not share a common origin with bullets in a box of ammunition containing 24 cartridges taken from suspects. Evidence in the second case included two bullets from the crime scene and 163 bullets taken from various suspects. We were able to infer that the two bullets from the crime scene did not share a common origin and that they differed from all of the bullets taken from suspects. All of the suspects' ammunition had been reloaded as was evident both from trace and isotopic analysis and, indeed, from visual inspection.

KEYWORDS: forensic science, bullet lead, lead isotope analysis, trace element analysis

The “ballistic” analysis of bullets, i.e., the comparison of characteristic markings left on fired bullets by the barrel of a weapon, often fails to provide sufficient information for a definitive match. In such cases, chemical analysis of the bullet lead can provide an indication of whether two bullets have a common origin. One might ask, for example, whether two bullets came from the same box of ammunition. The forensic chemical analysis of bullet lead, i.e., the measurement of minor and trace elemental constituents, has been studied for decades. The pioneering work of V. P. Guinn employed neutron activation analysis (1). Most of the more recent work on bullet analysis has been based on mass spectrometric techniques. Typical minor constituents in bullet lead include antimony, arsenic, and tin, elements that are added in various concentrations to control the hardness and casting properties.

Lead itself is one of the few elements in which isotopic composition varies with place of origin. This variation arises because three of the four stable isotopes, Pb^{206} , Pb^{207} and Pb^{208} , are formed as end products in the radioactive decay of U^{238} , U^{235} , and Th^{232} , respectively. The fourth isotope, Pb^{204} , is primordial, nonradiogenic lead. The isotopic composition of lead thus varies depending on the original composition of an ore body. Geologists use lead isotope analysis to trace the evolution of ore bodies. Archaeologists use

lead isotope analysis to determine the origins of artifacts that have lead either as a major or trace constituent. Isotope analysis is also employed to determine the sources of environmental lead pollution. The application of lead isotope analysis to bullet individualization was first reported in 1975 (2). Several other studies of the isotopic composition of bullet (and shotgun pellet) lead have subsequently been published (3–5).

We present lead isotope bullet analysis results, supplemented by trace element information obtained in two murder cases. Although bullet trace composition analysis has been used in the investigation of criminal cases, such analyses are relatively expensive and are not routinely carried out. We hope that our work will provide additional examples useful to criminalists interested in assessing the applicability of isotopic and trace element analysis in their own investigations.

Methods

Analyses were carried out using a Cameca IMS 5f Ion Microscope. In this instrument, a focused O_2^+ primary ion beam sputters minute amounts of material from the specimen surface. A fraction of the sputtered material (atoms and molecular fragments) is ionized (secondary ions), and the secondary ions are accelerated by an applied potential through an immersion lens and slits, into a magnetic sector mass analyzer, and then onto a detector (electron multiplier). Positive secondary ions were selected in our bullet analyses. The mass resolution used for lead isotope analyses is about 500 ($M/\Delta M$). This is sufficient to obtain baseline resolution for the lead isotopes. A magnetic sector analyzer like the Cameca IMS 5f is well suited to isotope analysis due to excellent sensitivity, mass resolution, and high sputtering rate.

Ion probe instruments require minimal sample preparation. Small ($\sim 2 \times 2 \times 1$ mm) pieces of lead were cut from test specimens using razor blades. A fresh blade was used for each cut to minimize the possibility of cross contamination of specimens. The freshly exposed lead samples were mounted on a 2.5-cm diameter stub and placed in the analysis chamber. Slices were shaved off at several locations around the periphery of each bullet. The specimen surface was cleaned by setting the oxygen ion beam to a point (non-rastered) mode of operation in which the beam diameter is about 20 μm for 15 min prior to taking data in order to sputter away the top layers of the surface and remove any surface contaminants. Several locations were examined on each of these slices. For isotope ratio measurements, the spectrometer was set to scan mass windows centered on the four lead isotope mass values.

The uncertainty in isotope ratio measurements is determined largely by counting statistics. There are, however, several possible sources of systematic error. For example, variations in ion beam

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current arise because of source instabilities (which are controlled as much as possible in the instrument design). As the collection of ions continues, the beam creates a shallow crater, and ion yield may vary. Fortunately, systematic errors largely cancel when measuring peak ratios. Counting statistics for bullets, which are essentially 100% lead, are good. Accurate isotope ratio measurements do require a number of precautions in the setup of the mass spectrometer. The lenses in the ion column were adjusted to provide "flat-topped" peaks to compensate for any slight drift in the magnet current. To improve counting statistics, ion counts were integrated for 4 s at each mass 204, 206, 207, and 208. Data were collected for 50 or 75 such counting cycles. Measured ion counts are automatically corrected by the system software for factors such as ion detector dead time. The intensity of the reference peak was automatically remeasured before and after the peak measurements of each of the other lead isotopes. The National Institute of Standards and Technology provides a lead isotope Standard Reference Material (SRM 981). Mass spectra of SRM 981 lead were used to calibrate our instrument and make slight corrections to the measured lead isotope peak ratios. The measurement precision is about 0.5%, and after correction the accuracy is on the order of $\pm 2\%$.

In general, Pb^{206} is the most sensible choice as a reference because possible hydride interference is minimized. For each Pb isotope peak, a Pb+H peak appears one mass unit higher. Since there is no Pb^{205} isotope, the Pb^{206} peak does not contain a superimposed hydride signal, though, of course, Pb^{206} hydride does make a contribution to the mass peak at Pb^{207} . Pb^{204} is another candidate, but would contribute the largest fractional error since it is the least abundant of the four stable Pb isotopes.

The lead isotope ratios are easily converted to concentrations in atomic percent. Since the concentrations of the four isotopes must add to 100%, there are still only three independent numbers, not four, and, in that sense, the isotope ratios are more fundamental. We also report the isotopic concentrations expressed in atomic percent to facilitate comparison with the literature. The errors in the atomic concentrations are derived from the computed errors in the three isotope ratios to Pb^{206} using the standard statistical formulae for the propagation of errors in quantities derived from other, measured quantities (6). This calculation is performed automatically by the Iso Calc program, a software package supplied as part of the mass spectrometer's instrumentation and written by C. A. Evans and Associates, Sunnyvale, CA.

Complete spectra from Mass 1 to Mass 250 were collected to determine elemental constituents. These survey scans were collected fairly quickly (5 min/scan). While sufficient for our immediate purposes, the count rates for minor elemental constituents in these survey scans were low. Total counts for arsenic, for example, ranged from about 10 to 400. Total counts of impurity elements in bullet lead recently reported using an inductively coupled plasma mass spectrometer were much higher, with correspondingly better counting statistics and precision (7).

The sensitivity for different elements varies considerably, from about 10^{12} to 10^{16} atoms/cm³ (i.e., from parts per billion to parts per million). Detection sensitivity for species that are easily ionized, such as alkali metals, is relatively higher, while sensitivity to weakly interacting species, such as the noble metals, is relatively much lower. The detection limits are also matrix dependent. Fortunately, in bullet analysis the matrix is essentially pure lead with small amounts of minor and trace constituents. The assumption of an instrument response that is linear with trace constituent impurity concentrations is, therefore, justified, and calibration for quantitative analysis is straightforward.

For bullet comparisons, trace elements are most easily reported as ratios to one of the lead peaks. The NIST does provide a standard reference bullet lead, SRM C2416, containing certified concentrations of Sb, As, Bi, Cu, S, Ag, and Sn. We have computed concentrations for these elements in atomic and weight percent. To determine a concentration for element "E" in a given matrix, one first computes a relative sensitivity factor (RSF) using the bullet lead standard:

$$\frac{I_R}{C_R} = \text{RSF}_E \frac{I_E}{C_E},$$

where I_E and C_E are the number of counts (taken simply as the maximum value of the peak), and concentration of element "E" and I_R and C_R are the corresponding quantities for a reference element (8). In the present case, the matrix itself serves as the reference, and it is convenient to use a slightly modified definition of the relative sensitivity factor as $\text{RSF} = \text{RSF}_E * C_M$ since the matrix concentration is constant. We then have simply

$$C_E = \text{RSF}_E \frac{I_E}{I_M}$$

We computed RSFs using ratios of the most abundant elemental isotopic peaks to the Pb^{206} peak for the elements for which the standard is certified.

Results

Case 1

A boy was shot and killed while fishing in 1981. The evidence on file includes the fatal .22-caliber bullet from the boy's body and a box of .22-caliber ammunition recovered from the home of older boys living in the area. The victim's parents are still seeking a resolution of the case and make periodic inquiries of the county sheriff's office. The Western Regional Office of the National Law Enforcement and Corrections Technology Center, operated by The Aerospace Corporation for the National Institute of Justice of the U.S. Department of Justice, was asked whether any analytic methods might now be available to establish whether or not the fatal bullet and the bullets in the box shared a common origin.

We received the fatal .22-caliber bullet and the box containing 24 "Remington Hi-Speed, .22 Long-Rifle, Golden bullet, KLEANBORE[®] 1522" cartridges. Bullets from four of the Remington .22 cartridges and the bullet from the victim were analyzed. The Remington .22 bullets were analyzed in two or three different locations. The fatal bullet was examined in five different locations. A kinetic bullet puller was used to remove the lead from the cartridges.

Averages of the lead isotope ratios and the isotopic abundances in atomic percent for each of the four suspects' bullets and for the fatal bullet are shown in Table 1. The errors are as determined by the "Iso Calc"™ spectrometer system program, and take into account the counting statistics and systematic errors mentioned above. The isotope ratios for the reference bullets and the fatal bullet are shown graphically in Fig. 1. The error bars are indicated but are small on the scale used.

Table 2 shows the concentrations of twelve elements in the fatal and the four reference Remington cartridges (at two to five locations on each cartridge). The results are again expressed as ratios to Pb^{206} . There is considerable scatter in the elemental composition data, reflecting the low elemental count rates of our ion microprobe mass analyzer in these particular experiments.

TABLE 1—Averages of the lead isotope ratios and the isotopic abundances in atomic percent for each of the four suspects' bullets and for the fatal bullet.

	Pb204/ Pb206	Pb207/ Pb206	Pb208/ Pb206	Pb204 (atomic %)	Pb206 (atomic %)	Pb207 (atomic %)	Pb208 (atomic %)
Ref. Bullet 1, location 1	0.05488 ± 0.00006	0.88501 ± 0.00032	2.10360 ± 0.00057	1.3573 ± 0.0016	24.7311 ± 0.0040	21.8873 ± 0.0086	52.0243 ± 0.0164
Ref. Bullet 1, location 2	0.05436 ± 0.00007	0.87458 ± 0.00035	2.09920 ± 0.00064	1.3494 ± 0.0017	24.8254 ± 0.0045	21.7118 ± 0.0096	52.1134 ± 0.0184
Ref. Bullet 1, location 3	0.05536 ± 0.00008	0.88800 ± 0.00042	2.15330 ± 0.00077	1.3514 ± 0.0021	24.4101 ± 0.0053	21.6762 ± 0.0114	52.5623 ± 0.0219
Ref. Bullet 1 average	0.05487 ± 0.00004	0.88253 ± 0.00021	2.11870 ± 0.00038	1.3527 ± 0.0010	24.6542 ± 0.0027	21.7581 ± 0.0058	52.2349 ± 0.0110
Ref. Bullet 2, location 1	0.05445 ± 0.00006	0.87910 ± 0.00031	2.09830 ± 0.00056	1.3505 ± 0.0015	24.8025 ± 0.0040	21.8039 ± 0.0085	52.0431 ± 0.0161
Ref. Bullet 2, location 2	0.05549 ± 0.00005	0.86934 ± 0.00024	2.06850 ± 0.00043	1.3896 ± 0.0012	25.0417 ± 0.0031	21.7698 ± 0.0066	51.7988 ± 0.0125
Ref. Bullet 2, location 3	0.05425 ± 0.00006	0.87643 ± 0.00032	2.09590 ± 0.00057	1.3474 ± 0.0016	24.8349 ± 0.0040	21.7661 ± 0.0087	52.0516 ± 0.0165
Ref. Bullet 2 average	0.05473 ± 0.00003	0.87496 ± 0.00017	2.08757 ± 0.00030	1.3624 ± 0.0008	24.8926 ± 0.0022	21.7800 ± 0.0046	51.9650 ± 0.0088
Ref. Bullet 3, location 1	0.05448 ± 0.00007	0.87933 ± 0.00037	2.09730 ± 0.00066	1.3514 ± 0.0018	24.8071 ± 0.0047	21.8136 ± 0.0101	52.0279 ± 0.0192
Ref. Bullet 3, location 2	0.05458 ± 0.00007	0.87813 ± 0.00036	2.10050 ± 0.00064	1.3532 ± 0.0017	24.7942 ± 0.0045	21.7725 ± 0.0097	52.0801 ± 0.0184
Ref. Bullet 3, average	0.05453 ± 0.00005	0.87873 ± 0.00026	2.09890 ± 0.00046	1.3523 ± 0.0013	24.8006 ± 0.0033	21.7931 ± 0.0070	52.0540 ± 0.0133
Ref. Bullet 4, location 1	0.05504 ± 0.00008	0.88828 ± 0.00043	2.13400 ± 0.00078	1.3498 ± 0.0021	24.5259 ± 0.0054	21.7859 ± 0.0116	52.3384 ± 0.0223
Ref. Bullet 4, location 2	0.05504 ± 0.00011	0.88308 ± 0.00058	2.14030 ± 0.00105	1.3495 ± 0.0028	24.5193 ± 0.0073	21.6525 ± 0.0157	52.4787 ± 0.0302
Ref. Bullet 4, average	0.05504 ± 0.00007	0.88568 ± 0.00036	2.13715 ± 0.00066	1.3497 ± 0.0018	24.5226 ± 0.0045	21.7192 ± 0.0098	52.4085 ± 0.0188
Fatal bullet, location 1	0.05024 ± 0.00010	0.81416 ± 0.00052	1.96400 ± 0.00093	1.3122 ± 0.0027	26.1206 ± 0.0073	21.2663 ± 0.0148	51.3008 ± 0.0282
Fatal bullet, location 2	0.05132 ± 0.00015	0.81672 ± 0.00075	1.99050 ± 0.00135	1.3301 ± 0.0039	25.9165 ± 0.0104	21.1665 ± 0.0212	51.5868 ± 0.0408
Fatal bullet, location 3	0.05150 ± 0.00016	0.81741 ± 0.00081	2.00500 ± 0.00147	1.3293 ± 0.0042	25.8137 ± 0.0112	21.1004 ± 0.0228	51.7565 ± 0.0441
Fatal bullet, location 4	0.05063 ± 0.00013	0.81621 ± 0.00064	1.99150 ± 0.00116	1.3121 ± 0.0033	25.9179 ± 0.0090	21.1545 ± 0.0182	51.6155 ± 0.0350
Fatal bullet, location 5	0.05139 ± 0.00015	0.82182 ± 0.00075	2.00710 ± 0.00136	1.3244 ± 0.0039	25.7711 ± 0.0104	21.1792 ± 0.0212	51.7252 ± 0.0408
Fatal bullet, average	0.05101 ± 0.00006	0.81726 ± 0.00031	1.99162 ± 0.00057	1.3217 ± 0.0016	25.9074 ± 0.0044	21.1732 ± 0.0089	51.5977 ± 0.0171

The reference and fatal bullets can be differentiated by their arsenic concentrations. The raw count rate for lead was lower during the fatal bullet measurement runs than during the reference bullet runs because of differences in spectrometer tuning. Although arsenic count rates were low in general, the results are deemed significant. Possible differences in the concentrations of some other elements, such as the alkali metals and alkali earths are apparent in Table 2, but there is scatter even in the measurements at different locations on a single bullet. It is possible that surface contaminants were present despite the precautions taken. However, the presence of the observed trace elements was verified on another mass spectrometer also available at our facility (a time-of-flight secondary-ion mass spectrometer, Physical Electronics model TRIFT II). We do not believe that surface contamination was a significant factor in the present analysis. Approximate elemental concentrations for those elements for which we have a calibration are shown in Table 3. These values are consistent with other published measurements of bullet trace element composition (7).

Case 2

A police officer was shot and killed while sitting in his parked patrol car. Two spent bullets were recovered at the scene. One copper-jacketed 9-mm bullet was removed from the headliner of the officer's car (item #125). A second 9-mm bullet fragment, with the remains of a copper jacket, was taken from the headliner of another vehicle at the scene (item #126). One hundred sixty three bullets seized from suspects at different locations were also provided to us in labeled evidence bags. We were asked whether any link could be established between the crime scene bullets and the evidence bullets. Although the total number of bullets provided was large (large enough to tax laboratory resources), only jacketed 9-mm ammunition had to be considered.

An evidence bag (#97) containing 103 cartridges, all copper jacketed, came from one location. These cartridges bore ten different headstamps: 71 Winchester (WIN); 13 Federal Cartridge (FC); four Western Cartridge Co. (WCC); four Remington-Peters (R-P); four Precision Made Cartridges (PMC); two Giulio Flocchi (GFL);

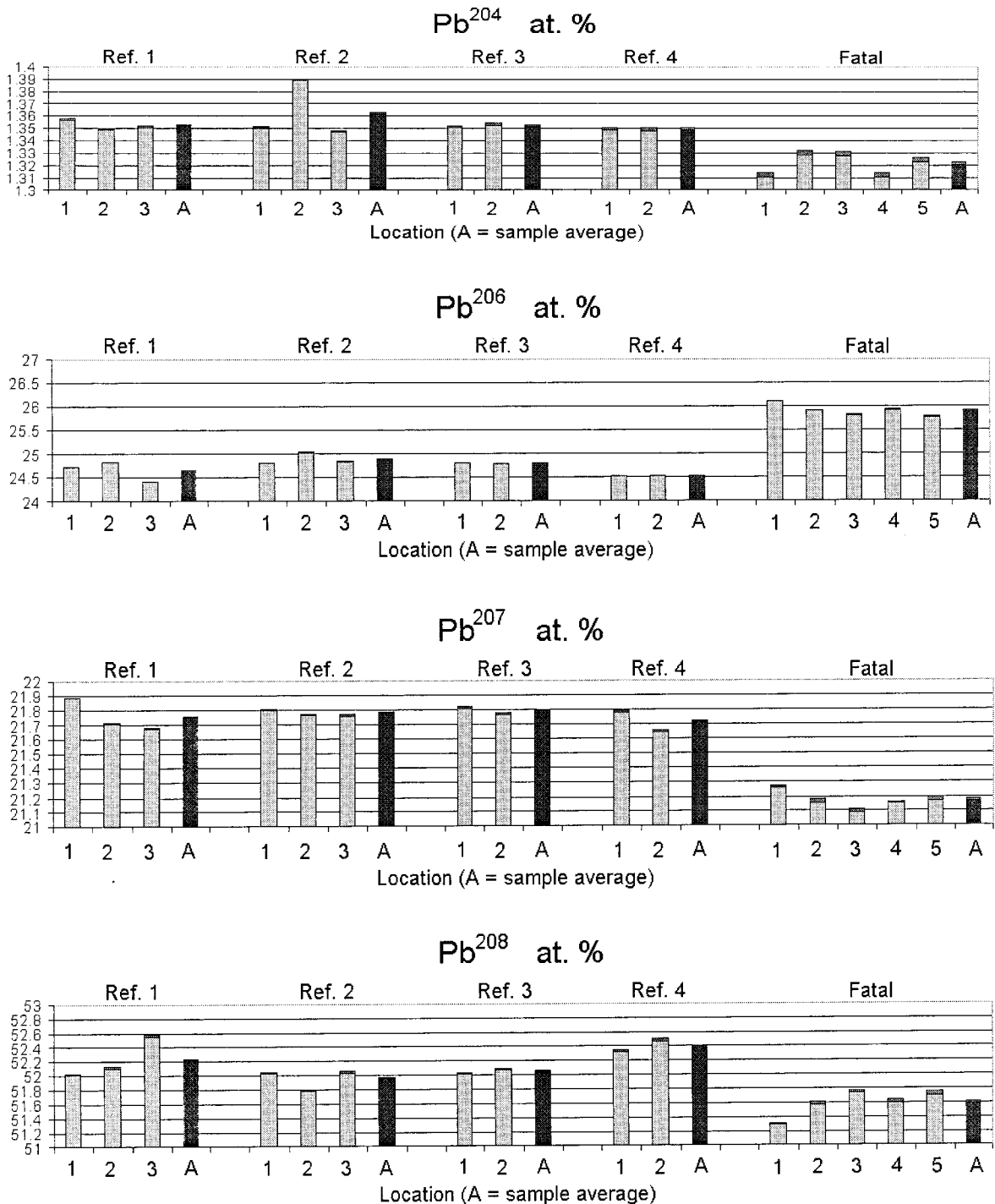


FIG. 1—Lead isotope ratios of the “Case 1” bullets.

two Speer; one Magtech Recreational Products (CBC); one ELD; and one Aquila. One cartridge with a WIN, one with a PMC, and one with a GFL headstamp were chosen from this bag for analysis. Another evidence bag (#243) contained seven cartridges, six of which were jacketed. The bullets in bag #243 were seized at a location different from those in bag #97. We selected one of the four Winchester cartridges and the single PMC and the single GFL cartridge from this bag for analysis. After the bullets were pulled from the cartridges, it was apparent that in all of the ammunition taken

from suspects the copper jackets enclosed the entire bullet. Although the copper jackets from the crime scene covered the points of bullets #125 and #126, the jackets were open at the base, i.e., did not wrap entirely around the lead. The jacket of bullet #126 had separated from the lead core, but the structure of the jacket was still evident. A quick inspection of all of the ammunition recovered from suspects using a real-time X-ray machine verified that none of the jackets were open on the bottom and were, therefore, ruled out as potential matches. Nevertheless, we proceeded with the iso-

TABLE 2—Concentration of 12 elements in the fatal and the four reference Remington cartridges.

	Na 23 cts	Al 27 cts	Si 28 cts	K 39 cts	Ca 40 cts	Fe 56 cts	Ni 58 cts	Cu 63 cts	As 75 cts	Ag107cts	In115 cts	Sb 121 cts	Bi 209 cts
bul 1 loc 1	0.00123	0.00077	0.00028	0.00550	0.00049	0.00000	0.00000	0.00473	0.00004	0.00014	0.00018	0.00834	0.000981
bul 1 loc 2	0.00152	0.00156	0.00009	0.00356	0.00039	0.00000	0.00000	0.00538	0.00004	0.00004	0.00022	0.01142	0.000651
bul 1 loc 3	0.01031	0.00526	0.00202	0.00283	0.00505	0.00081	0.00061	0.02224	0.00000	0.00142	0.00040	0.01678	0.001617
average	0.00435	0.00253	0.00080	0.00396	0.00198	0.00027	0.00020	0.01078	0.00003	0.00053	0.00027	0.01218	0.001083
bul 2 loc 1	0.04297	0.01714	0.00404	0.00554	0.00268	0.00040	0.00015	0.01927	0.00000	0.00018	0.00011	0.01483	0.001028
bul 2 loc 2	0.06367	0.05091	0.01128	0.00447	0.00309	0.00028	0.00008	0.01902	0.00002	0.00021	0.00009	0.01268	0.001434
average	0.05332	0.03403	0.00766	0.00501	0.00289	0.00034	0.00011	0.01914	0.00001	0.00020	0.00010	0.01375	0.001231
bul 3 loc 1	0.00074	0.00054	0.00008	0.00223	0.00058	0.00004	0.00004	0.01278	0.00000	0.00050	0.00008	0.01791	0.000827
bul 3 loc 2	0.00074	0.00101	0.00000	0.00297	0.00048	0.00000	0.00013	0.01575	0.00004	0.00013	0.00026	0.01885	0.00105
average	0.00074	0.00077	0.00004	0.00260	0.00053	0.00002	0.00009	0.01427	0.00002	0.00031	0.00017	0.01838	0.000939
bul 4 loc 1	0.02250	0.05336	0.00516	0.00291	0.00262	0.00015	0.00015	0.01315	0.00000	0.00037	0.00022	0.02063	0.000897
bul 4 loc 2	0.00055	0.00036	0.00000	0.00018	0.00018	0.00000	0.00000	0.01293	0.00009	0.00018	0.00009	0.02368	0.001093
average	0.01152	0.02686	0.00258	0.00155	0.00140	0.00007	0.00007	0.01304	0.00005	0.00028	0.00016	0.02215	0.000995
av of all 4	0.01603	0.01455	0.00255	0.00336	0.00173	0.00019	0.00013	0.01392	0.00003	0.00035	0.00018	0.01612	0.001064
vict loc 1	0.00641	0.00066	0.00147	0.01326	0.00177	0.00000	0.00000	0.01098	0.00052	0.00007	0.00007	0.00545	0.000221
vict loc 2	0.00479	0.00080	0.00040	0.00917	0.00359	0.00040	0.00000	0.00479	0.00080	0.00040	0.00000	0.02274	0
vict loc 3	0.06317	0.00514	0.00147	0.06170	0.04076	0.00147	0.00073	0.00661	0.00110	0.00037	0.00000	0.03342	0.000367
vict loc 4	0.02051	0.00234	0.00029	0.00732	0.00454	0.00044	0.00176	0.01919	0.00132	0.00015	0.00015	0.01934	0.000586
vict loc 5	0.22207	0.00158	0.00045	0.10203	0.22928	0.00180	0.00045	0.00743	0.00090	0.00023	0.00000	0.01779	0.000225
average	0.06339	0.00210	0.00082	0.03870	0.05599	0.00082	0.00059	0.00980	0.00093	0.00024	0.00004	0.01975	0.00028

TABLE 3—Elemental concentrations in Case 1 bullets (referenced to NIST C2416 bullet lead standard).

Sample	Cu		As		Ag		Sb		Bi	
	at%	wt%	at%	wt%	at%	wt%	at%	wt%	at%	wt %
Cartridges	0.2376	0.0729	0	0	0.0135	0.0070	2.141	1.258	0.0995	0.1000
Victim	0.2290	0.0702	0.1617	0.0584	0	0	3.366	1.978	0.0003	0.0003

tope/composition analysis of bullets in the evidence bags out of general interest.

Isotope ratio results for the Case 2 bullets are shown in Table 4 and Fig. 2. The bullets recovered at the crime scene (#125 and #126) differ substantially in isotopic composition from the bullets in the evidence bags, as indeed would be expected since visual inspection had already established that they were not from a common source. The two crime scene bullets, #125 and #126, have isotopic compositions that differ from each other. The differences are perhaps most apparent in Pb^{206} . There is some scatter in the data. There are also differences in elemental composition between the

two crime scene bullets. Ratios of the magnitude of most abundant isotopic peak to the Pb^{206} peak for trace elements detected are shown in Table 5. Bullet #125, for example, contains no detectable arsenic, while arsenic is detected in item #126. Bullet #125 contains lower concentrations of sodium and calcium than bullet #126. The three bullets from evidence bag #243 generally showed much lower concentrations of trace elements than the bullets of bag #97. Arsenic, indium, tin, and antimony were not detected at all in bag #243, although these elements were detected in the three bullets of bag #97. Concentrations in atomic percent, calculated from computed relative sensitivity factors, are shown in Table 6. The lead

TABLE 4—Isotope ratio results for the Case 2 bullets.

	Pb204/206	Pb207/206	Pb208/207	Pb204 (at. %)	Pb206 (at. %)	Pb207 (at. %)	Pb208 (at. %)
#125 loc 1	0.05691 ± 0.00003	0.88258 ± 0.00013	2.07540 ± 0.00022	1.4174 ± 0.0006	24.9073 ± 0.0016	21.9827 ± 0.0034	51.6926 ± 0.0064
#125 loc 2	0.05706 ± 0.00003	0.88480 ± 0.00015	2.08470 ± 0.00026	1.4170 ± 0.0007	24.8351 ± 0.0018	21.9741 ± 0.0040	51.7737 ± 0.0075
#125 loc 3	0.05696 ± 0.00003	0.88285 ± 0.00013	2.08490 ± 0.00023	1.4152 ± 0.0006	24.8465 ± 0.0016	21.9358 ± 0.0035	51.8025 ± 0.0065
#125 loc 4	0.05680 ± 0.00005	0.86314 ± 0.00027	2.08560 ± 0.00045	1.4179 ± 0.0013	24.9654 ± 0.0033	21.5487 ± 0.0074	52.0679 ± 0.0132
#125 loc 5	0.05665 ± 0.00008	0.86077 ± 0.00041	2.09510 ± 0.00068	1.4119 ± 0.0019	24.9220 ± 0.0050	21.4521 ± 0.0112	52.2140 ± 0.0200
#125 average	0.05687 ± 0.00002	0.87483 ± 0.00011	2.08514 ± 0.00018	1.4159 ± 0.0005	24.8953 ± 0.0013	21.7787 ± 0.0030	51.9102 ± 0.0053
#126 loc 1	0.05754 ± 0.00004	0.89911 ± 0.00017	2.14000 ± 0.00031	1.4045 ± 0.0009	24.4102 ± 0.0021	21.9475 ± 0.0046	52.2379 ± 0.0088
#126 loc 2	0.05744 ± 0.00003	0.89773 ± 0.00014	2.12060 ± 0.00025	1.4092 ± 0.0007	24.5353 ± 0.0018	22.0260 ± 0.0038	52.0295 ± 0.0073
#126 loc 3	0.05744 ± 0.00004	0.89763 ± 0.00018	2.13930 ± 0.00032	1.4028 ± 0.0009	24.4238 ± 0.0022	21.9235 ± 0.0048	52.2498 ± 0.0092
#126 loc 4	0.05741 ± 0.00004	0.90140 ± 0.00017	2.14670 ± 0.00031	1.3984 ± 0.0009	24.3575 ± 0.0021	21.9558 ± 0.0047	52.2882 ± 0.0089
#126 loc 5	0.05774 ± 0.00003	0.90014 ± 0.00014	2.13170 ± 0.00025	1.4118 ± 0.0007	24.4524 ± 0.0017	22.0106 ± 0.0037	52.1252 ± 0.0071
#126 average	0.05751 ± 0.00001	0.89920 ± 0.00007	2.13566 ± 0.00013	1.4053 ± 0.0004	24.4358 ± 0.0009	21.9727 ± 0.0020	52.1861 ± 0.0037
GFL97 loc 1	0.05220 ± 0.00002	0.82298 ± 0.00011	2.00510 ± 0.00020	1.3451 ± 0.0006	25.7714 ± 0.0015	21.2093 ± 0.0030	51.6742 ± 0.0059
GFL97 loc 2	0.05213 ± 0.00003	0.82284 ± 0.00016	2.02320 ± 0.00030	1.3372 ± 0.0009	25.6531 ± 0.0023	21.1084 ± 0.0046	51.9013 ± 0.0089
GFL97 loc 3	0.05219 ± 0.00002	0.82258 ± 0.00012	2.01630 ± 0.00022	1.3413 ± 0.0006	25.6999 ± 0.0017	21.1402 ± 0.0034	51.8187 ± 0.0066
GFL97 loc 4	0.05208 ± 0.00003	0.82180 ± 0.00015	2.01920 ± 0.00027	1.3377 ± 0.0008	25.6866 ± 0.0020	21.1093 ± 0.0041	51.8664 ± 0.0080
GFL97 loc 5	0.05208 ± 0.00003	0.82251 ± 0.00014	2.01880 ± 0.00026	1.3377 ± 0.0008	25.6845 ± 0.0020	21.1258 ± 0.0040	51.8519 ± 0.0077
GFL97 average	0.05213 ± 0.00001	0.82254 ± 0.00006	2.01652 ± 0.00011	1.3398 ± 0.0003	25.6991 ± 0.0008	21.1386 ± 0.0017	51.8225 ± 0.0034
PMC97 loc 1	0.05230 ± 0.00004	0.82249 ± 0.00017	2.00151 ± 0.00031	1.3493 ± 0.0009	25.7978 ± 0.0024	21.2184 ± 0.0048	51.6345 ± 0.0093
PMC97 loc 2	0.05218 ± 0.00003	0.82357 ± 0.00012	2.00970 ± 0.00023	1.3430 ± 0.0007	25.7370 ± 0.0017	21.1962 ± 0.0035	51.7237 ± 0.0068
PMC97 loc 3	0.05246 ± 0.00003	0.82267 ± 0.00017	2.00960 ± 0.00030	1.3504 ± 0.0009	25.7418 ± 0.0023	21.1770 ± 0.0046	51.7308 ± 0.0090
PMC97 loc 4	0.05221 ± 0.00003	0.82263 ± 0.00013	2.01340 ± 0.00023	1.3428 ± 0.0007	25.7186 ± 0.0017	21.1569 ± 0.0035	51.7818 ± 0.0069
PMC97 loc 5	0.05251 ± 0.00004	0.82067 ± 0.00020	2.01060 ± 0.00035	1.3520 ± 0.0011	25.7481 ± 0.0027	21.1307 ± 0.0055	51.7692 ± 0.0106
PMC97 average	0.05233 ± 0.00001	0.82241 ± 0.00007	2.00896 ± 0.00013	1.3475 ± 0.0004	25.7487 ± 0.0010	21.1759 ± 0.0020	51.7280 ± 0.0039
WIN97 loc 1	0.05212 ± 0.00003	0.82133 ± 0.00015	2.01710 ± 0.00027	1.3395 ± 0.0008	25.7033 ± 0.0020	21.1109 ± 0.0041	51.8462 ± 0.0080
WIN97 loc 2	0.05232 ± 0.00003	0.82469 ± 0.00015	2.01820 ± 0.00028	1.3431 ± 0.0008	25.6726 ± 0.0021	21.1719 ± 0.0043	51.8124 ± 0.0082
WIN97 loc 3	0.05214 ± 0.00004	0.82162 ± 0.00019	2.01570 ± 0.00035	1.3404 ± 0.0010	25.7105 ± 0.0026	21.1243 ± 0.0054	51.8247 ± 0.0103
WIN97 loc 4	0.05204 ± 0.00003	0.82339 ± 0.00013	2.02150 ± 0.00023	1.3355 ± 0.0007	25.6612 ± 0.0017	21.1292 ± 0.0036	51.8741 ± 0.0069
WIN97 loc 5	0.05251 ± 0.00004	0.82110 ± 0.00018	2.00970 ± 0.00033	1.3522 ± 0.0010	25.7512 ± 0.0025	21.1443 ± 0.0051	51.7522 ± 0.0099
WIN97 average	0.05222 ± 0.00001	0.82243 ± 0.00007	2.01644 ± 0.00013	1.3422 ± 0.0004	25.6998 ± 0.0010	21.1361 ± 0.0020	51.8219 ± 0.0039
GFL243 loc 1	0.05152 ± 0.00003	0.81545 ± 0.00017	2.00550 ± 0.00030	1.3303 ± 0.0009	25.8233 ± 0.0023	21.0576 ± 0.0047	51.7887 ± 0.0091
GFL243 loc 2	0.05167 ± 0.00004	0.81493 ± 0.00018	2.00300 ± 0.00032	1.3352 ± 0.0009	25.8425 ± 0.0025	21.0598 ± 0.0050	51.7625 ± 0.0096
GFL243 loc 3	0.05163 ± 0.00004	0.81525 ± 0.00018	2.00350 ± 0.00032	1.3341 ± 0.0010	25.8372 ± 0.0025	21.0638 ± 0.0050	51.7649 ± 0.0097
GFL 243 average	0.05161 ± 0.00002	0.81521 ± 0.00010	2.00400 ± 0.00018	1.3332 ± 0.0005	25.8343 ± 0.0014	21.0604 ± 0.0028	51.7720 ± 0.0055

continues

TABLE 4—Continued.

	Pb204/206	Pb207/206	Pb208/207	Pb204 (at. %)	Pb206 (at. %)	Pb207 (at. %)	Pb208 (at. %)
PMC243 loc 1	0.05149 ± 0.00003	0.81310 ± 0.00015	1.99800 ± 0.00028	1.3331 ± 0.0008	25.8893 ± 0.0022	21.0506 ± 0.0044	51.7269 ± 0.0084
PMC243 loc 2	0.05154 ± 0.00006	0.81325 ± 0.00027	2.01080 ± 0.00049	1.3298 ± 0.0014	25.8025 ± 0.0037	20.9839 ± 0.0076	51.8837 ± 0.0147
PMC243 loc 3	0.05148 ± 0.00003	0.81411 ± 0.00016	2.00450 ± 0.00029	1.3302 ± 0.0008	25.8392 ± 0.0022	21.0359 ± 0.0045	51.7947 ± 0.0087
PMC 243 average	0.05150 ± 0.00002	0.81349 ± 0.00012	2.00443 ± 0.00021	1.3311 ± 0.0006	25.8437 ± 0.0016	21.0235 ± 0.0033	51.8018 ± 0.0063
WIN243 loc 1	0.05156 ± 0.00003	0.81525 ± 0.00015	2.00560 ± 0.00028	1.3315 ± 0.0008	25.8237 ± 0.0021	21.0528 ± 0.0043	51.7920 ± 0.0084
WIN243 loc 2	0.05153 ± 0.00003	0.81417 ± 0.00017	2.00970 ± 0.00031	1.3298 ± 0.0009	25.8038 ± 0.0023	21.0086 ± 0.0047	51.8578 ± 0.0092
WIN243 loc 3	0.05155 ± 0.00003	0.81576 ± 0.00015	2.00960 ± 0.00027	1.3296 ± 0.0008	25.7937 ± 0.0021	21.0415 ± 0.0042	51.8351 ± 0.0082
WIN 243 average	0.05155 ± 0.00002	0.81506 ± 0.00009	2.00830 ± 0.00017	1.3303 ± 0.0005	25.8071 ± 0.0013	21.0343 ± 0.0026	51.8283 ± 0.0050

TABLE 5—Ratios of the magnitude of most abundant isotopic peak to the Pb²⁰⁶ peak for trace elements detected.

	Na 23	Al 27	Si 28	K 39	Ca 40	Fe 56	Ni 58	Cu 63
125 loc1	0.000209	0.000104	0.000000	0.000417	0.003027	0.000104	0.000209	0.000417
125 loc2	0.004224	0.000393	0.000295	0.000786	0.001081	0.000393	0.000688	0.000196
125 average	0.002216	0.000249	0.000147	0.000602	0.002054	0.000249	0.000448	0.000307
126 loc 1	0.083658	0.000085	0.000000	0.000339	0.175106	0.004742	0.000000	0.000085
126 loc2	0.182244	0.000488	0.000976	0.010537	0.220195	0.006634	0.000000	0.000195
126 average	0.132951	0.000286	0.000488	0.005438	0.197650	0.005688	0.000000	0.000140
GFL97loc1	0.000630	0.000035	0.000175	0.000035	0.000035	0.000070		0.003081
GFL97loc2	0.000477	0.001041	0.000130	0.000087	0.000824	0.000260	0.000043	0.001735
PMC97loc1	0.001537	0.000098	0.000000	0.000878	0.000390	0.000024	0.000049	0.002512
PMC97loc2	0.000408	0.000136	0.000227	0.000159	0.000204	0.000113	0.000113	0.001812
WIN97loc1	0.000341	0.000000	0.000038	0.000038	0.000076	0.000038	0.000114	0.003185
WIN97loc2	0.000056	0.000000	0.000074	0.000037	0.000167	0.000000	0.000056	0.001152
#97 average	0.000575	0.000218	0.000107	0.000206	0.000283	0.000084	0.000080	0.002246
GFL243loc1	0.000025	0.000008	0.000008	0.000025	0.000017	0.000000	0.000000	0.000165
GFL243loc2	0.000018	0.000012	0.000006	0.000049	0.000006	0.000000	0.000000	0.000000
PMC243loc1	0.000097	0.000000	0.000000	0.000036	0.000000	0.000000	0.000000	0.000000
PMC243loc2	0.000081	0.000000	0.000010	0.000041	0.000010	0.000000	0.000000	0.000000
WIN243loc1	0.000088	0.000000	0.000011	0.000033	0.000011	0.000000	0.000000	0.000164
WIN243loc2	0.000120	0.000030	0.000000	0.000090	0.000000	0.000000	0.000000	0.000010
#243 average	0.000072	0.000008	0.000006	0.000046	0.000007	0.000000	0.000000	0.000057
	Zn 64	As 75	Ag 107	In 115	Sn 120	Sb 121	Tl 203	Bi 209
125 loc1	0.000104	0.002192	0.000104	0.001461	0.074202	0.022542	0.000209	0.000731
125 loc2	0.000884	0.001572	0.000098	0.001670	0.050786	0.013851	0.000295	0.000491
125 average	0.000494	0.001882	0.000101	0.001566	0.062494	0.018196	0.000252	0.000611
126 loc 1	0.000000	0.000000	0.000085	0.003133	0.094750	0.009314	0.000423	0.000423
126 loc2	0.000098	0.000098	0.000000	0.001073	0.022244	0.010146	0.000488	0.000780
126 average	0.000049	0.000049	0.000042	0.002103	0.058497	0.009730	0.000456	0.000602
GFL97loc1	0.000070	0.000035	0.000105	0.000525	0.015791	0.051015	0.000490	0.000245
GFL97loc2	0.000217	0.000087	0.000043	0.000304	0.006678	0.024111	0.000564	0.000304
PMC97loc1	0.000000	0.000171	0.000073	0.000976	0.037049	0.021415	0.000366	0.000098
PMC97loc2	0.000023	0.000317	0.000136	0.000634	0.025074	0.032254	0.000227	0.000227
WIN97loc1	0.000076	0.000114	0.000190	0.000531	0.018847	0.061054	0.000228	0.000228
WIN97loc2	0.000056	0.000037	0.000056	0.000204	0.007932	0.021122	0.000334	0.000204
#97 average	0.000074	0.000127	0.000100	0.000529	0.018562	0.035162	0.000368	0.000217
GFL 243 loc1	0.000033	0.000000	0.000041	0.000000	0.000000	0.000008	0.000132	0.000487
GFL 243 loc2	0.000000	0.000000	0.000085	0.000000	0.000000	0.000000	0.000146	0.000262
PMC 243 loc1	0.000000	0.000000	0.000048	0.000000	0.000000	0.000012	0.000218	0.000339
PMC 243 loc2	0.000000	0.000000	0.000112	0.000000	0.000000	0.000010	0.000203	0.000305
WIN 243 loc1	0.000011	0.000000	0.000066	0.000000	0.000000	0.000000	0.000208	0.000329
WIN243loc2	0.000000	0.000000	0.000110	0.000000	0.000000	0.000010	0.000251	0.000402
#243 average	0.000007	0.000000	0.000077	0.000000	0.000000	0.000007	0.000193	0.000354

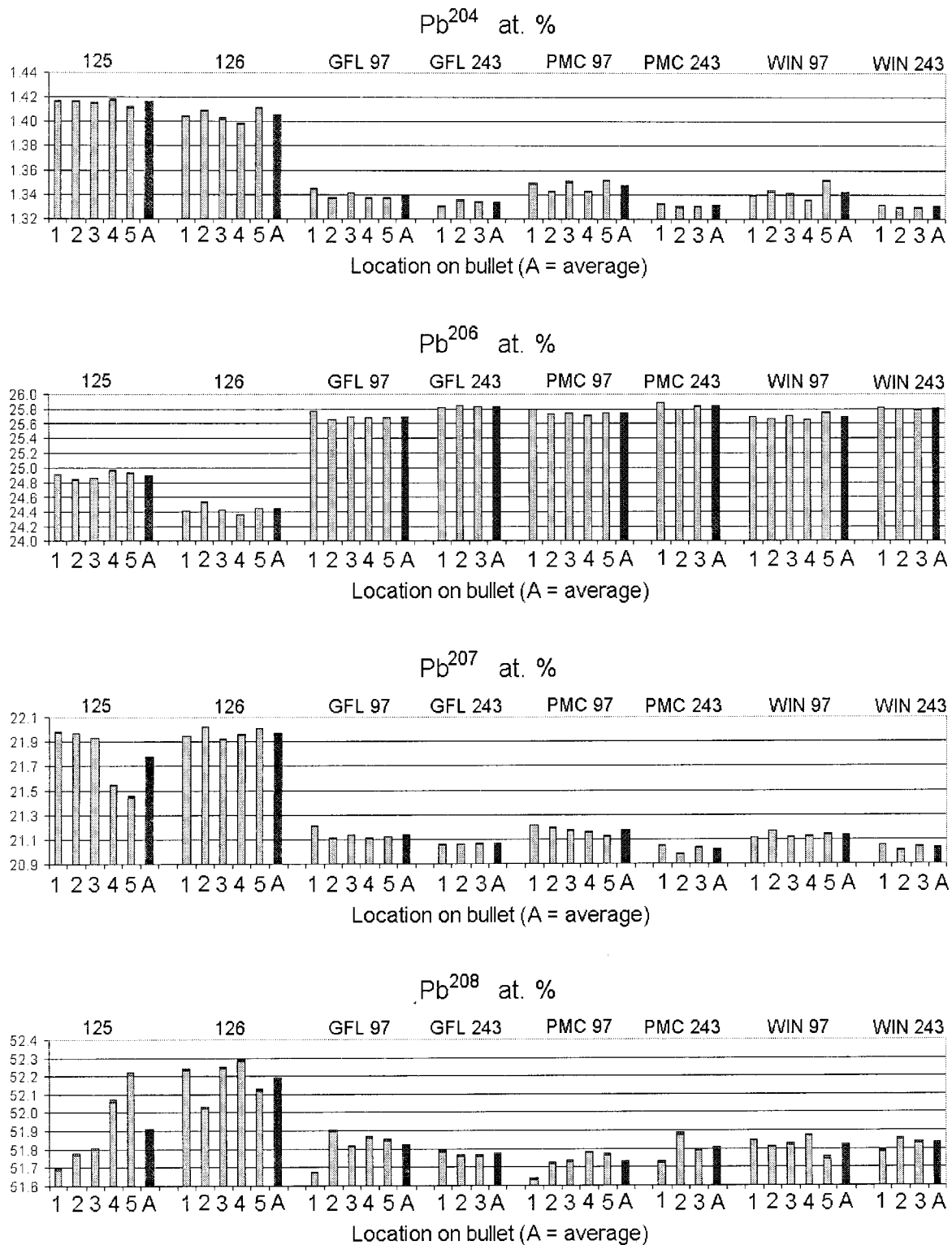


FIG. 2—Lead isotope ratios measured in the bullets of Case 2.

TABLE 6—Elemental compositions of Case 2 bullets (referenced to NIST C2416 bullet lead standard).

	Cu		As		Ag		Sn		Sb		Bi	
	at%	wt%	at%	wt%	at%	wt%	at%	wt%	at%	wt%	at%	wt%
#125, av	0.0069	0.0021	0.0116	0.0042	0.0068	0.0035	0.4441	0.2544	2.5634	1.5062	0.0591	0.0596
#126, av	0.0031	0.0010	0.0048	0.0017	0.0028	0.0015	0.4157	0.2382	1.3707	0.8054	0.0582	0.0587
Bag 97 av	0.0502	0.0154	0.0115	0.0041	0.0068	0.0035	0.1319	0.0756	4.9534	2.9106	0.0210	0.0212
Bag 243 av	0.0013	0.0004	0.0088	0.0032	0.0052	0.0027	0.0000	0.0000	0.0010	0.0006	0.0342	0.0345

isotope ratios are similar for both bags, although distinctly different from the ratios of the #125 and #126 crime scene bullets.

Discussion

The lead isotopic ratios in natural ores vary over a considerable range. The Pb^{204} to Pb^{206} ratio, for example, ranges from about 0.04 to 0.07, and the Pb^{207}/Pb^{206} ratio ranges from about 0.7 to 0.9. Unfortunately from the perspective of the criminalist, much of the lead used commercially is recycled. Today's bullet was quite possibly yesterday's tire weight. Bullet manufacturers usually control only the concentrations of antimony and arsenic in their products. In jacketed bullets, there is no need for deliberately added alloying elements but such elements may well be present in recycled lead. Although to facilitate quality control, most manufacturers prepare lead in fairly large batches, there is no assurance that the bullets even in a single box of ammunition will have closely identical compositions. In fact, one should not assume *a priori* that composition does not vary within a single bullet. Our results, however, do indicate that lead isotope ratios and trace element composition do not vary substantially within a single bullet, in agreement with Andrasco, et al. (5). Lead isotope ratios (Pb^{207}/Pb^{206} vs Pb^{204}/Pb^{206}) for both Case 1 and Case 2 bullets are summarized in Fig. 3. The leads from common sources cluster together, as they must if isotope ratio measurements are in fact useful for bullet characterization.

In Case 1, lead isotope analysis yielded what we judge with comfortable confidence to be "no match" between the fatal bullet and the bullets from the suspects' box. Furthermore, arsenic was detected in the fatal bullet but not in the suspects' bullets. The key observation here is that our sampling indicated that the bullets analyzed from the suspects' box were homogeneous with respect to trace element compositions and isotope ratios. The compositions of the bullets in the suspects' box were consistent with a common origin for the lead, although the elemental data have some scatter. Establishing or disproving the common origin of a particular bullet with a box of ammunition represents a situation suited to isotope ratio and elemental comparison analyses. The negative result

was useful to the criminal investigators since one unproductive line of inquiry could be closed. The "no match" conclusion was also consistent with polygraph results obtained from one of the suspects.

In Case 2, with two bullets collected at the crime scene and with 163 bullets of many different types seized at various locations, we infer that the bullets and bullet fragments collected from the crime scene (#125 and #126) did not share a common origin with each other or with any of the bullets analyzed.

While our analyses did not show matches among the bullets examined, we did observe an interesting correlation. The bullets from evidence bag #243 had compositions similar to one another, and the bullets from bag #97 had compositions similar to one another despite having different manufacturers' headstamps on the cartridges. The bullet populations from the two bags were distinctly different in trace elements, with all of the bullets from bag #243 having much lower concentrations of trace constituents than the bullets in bag #97. We conjectured that the results could be explained if the bullets had been reloaded, with the bullets in each bag having a common source.

A firearms examiner immediately recognized that the evidence ammunition had indeed been reloaded. The bullets within each of the two bags of ammunition looked identical physically and were not consistent with the types of bullets known to be used by the manufacturers whose names appeared on the cartridge case headstamps. The insides of the cartridge cases from which bullets had been "pulled" for analysis were blackened from burned powder. The cartridge cases were in varying degrees distorted from previous firings. Old extractor and ejector marks, marks from "resizing," and marks from primer reworking could be seen.

In many of the published papers on bullet analysis, the authors have come to the same general conclusions regarding lead isotope (and elemental) bullet analysis, viz., that the methods show promise but that further work is needed. These studies have sometimes been research investigations with ammunition not associated with a particular crime, but elemental/isotopic analyses have been used with some success in criminal cases (9). The latest, and quite thorough and detailed, work by Keto (using elemental but not iso-

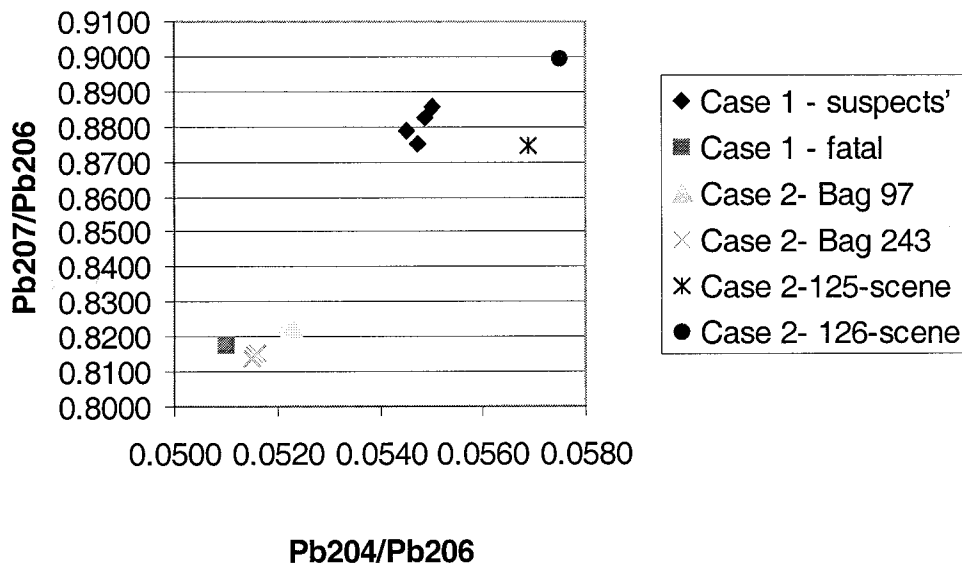


FIG. 3—Isotope ratios for Case 1 and Case 2.

topic analysis) concludes that elemental analysis is best suited to the comparison of boxes of ammunition where some degree of statistical confidence can be established. We certainly agree.

However, in many "real-life" situations only one or two bullets may be available from a crime scene. In the two cases presented, cartridges possibly linked to the crimes were subsequently recovered. If standard "ballistic" methods are not successful, compositional analysis may be the only laboratory tool available. Lead isotope and trace elemental analysis should both be employed in the characterization of bullet lead in order to glean the maximum possible amount of data from the evidence. Some useful information was gained in our "Case 1." While some information of technical interest was uncovered in "Case 2," the results did not contribute to the investigation. The present work hopefully offers some additional insight into the application of lead isotope ratios and elemental analyses in the forensic characterization of bullets.

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