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Forensic Comparison of Household Aluminum Foils Using Elemental Composition by Inductively Coupled Plasma—Atomic Emission Spectrometry

ABSTRACT: Inductively coupled plasma-atomic emission spectrometry (ICP-AES) provides the precision, sensitivity, and specificity for element concentration determinations required for good discrimination among sources of household aluminum foil. The concentrations of copper (Cu), gallium (Ga), iron (Fe), magnesium (Mg), manganese (Mn), titanium (Ti), vanadium (V), and zinc (Zn) were determined in aluminum foil by ICP-AES with analytical precisions (rsd) in the 1 to 2% range and nickel (Ni) and silicon (Si) with analytical precisions of about 5%. For these ten elements, the combined analytical uncertainties and variations along the length of a single roll of foil are much smaller than the variations from roll to roll, both within a single brand and across brands. The aluminum foils in this study exhibit measurable roll-to-roll compositional variation across the different brand names as well as across those rolls produced by the same manufacturer in different production runs. In some instances, rolls of foil produced in a single production run have analytically distinguishable compositions. Thus, a high degree of discrimination capability exists among sources of household aluminum foil.

KEYWORDS: criminalistics, elemental analysis, ICP-AES, metal comparison, trace elements

One task confronting the forensic analyst is comparison of objects recovered from crime scenes to those associated with various individuals. Household aluminum foil is a commodity that is frequently encountered as evidence in forensic cases, including those involving illegal narcotics trafficking and improvised explosive devices, but whose significance may be overlooked. It would be of value in the investigation and prosecution of such cases to be able to recognize or eliminate common sources among samples of aluminum foil. Comparison of foil samples is infrequently considered in most forensic investigations, in part because there are few reports in the literature of successful comparison methods for aluminum foils.

Elemental concentrations in metal alloys are relatively unaffected by use subsequent to manufacture and, as a result, offer valuable points of reference for forensic comparison. This has proven useful in forensic comparisons of several alloys, including brass [1], bullet lead [2,3], lead solder [4], copper wire [5], silver waste [6], gold coins [7], and some alloys of aluminum [8]. Multielement analysis of alloys, including those of aluminum, for purposes of verifying that their composition meets specifications is routinely done in the FBI Laboratory and many industrial and commercial laboratories.

Household aluminum foils are made from alloys containing greater than 99% aluminum. The remainder consists predominantly of iron, copper, and silicon at concentrations below a few tenths of a percent and various trace elements at the sub ppm to several hundred ppm level [9]. ICP-AES is an analytical technique with the capability of determining the concentrations of 10 to 15 elements in high grade aluminum alloys. ICP-AES has several advantages over other methods, such as neutron activation analysis, atomic absorption spectrophotometry, and X-ray fluorescence spectrometry, which can be used to determine element concentrations in aluminum foil. These advantages include wide linear dynamic ranges, multielement capability, and generally excellent accuracy and precision in the appropriate concentration ranges. This article reports the results of a study to develop a method for precise quantitative multielemental analysis of aluminum foil using ICP-AES and initial evaluation studies of the utility of elemental composition for forensic comparison of aluminum foils. The variability within individual sources of foil and the range in compositions across several sources are reported.

Experimental

Sample Preparation

Household aluminum foil samples were purchased from local retail stores. An approximately 100 mg piece was removed from each area of foil to be analyzed by holding the foil with plastic tweezers and tearing analytical samples from the larger sections of foil. To avoid possible trace element contamination, cutting tools with metallic surfaces were not used. Three separate samples of each foil were selected for analysis. Foil samples were torn into approximately 20 mg sized pieces prior to weighing and analysis. Because all foils used in this study were not previously used, they were not cleaned prior to analysis.

Approximately 100 mg from each foil to be analyzed was weighed to the nearest 0.01 mg and placed in a 15 mL polyethylene screw top tube. To each tube, 0.8 mL of $\%_{16}$ HCl (six volumes of concentrated HCl (Baker Ultrex) to ten volumes of deionized water of 18 MOhm-cm resistivity) were added. The tubes were capped loosely and the initial reaction was allowed to subside. The rate of reaction was controlled by placing each tube in a tight fitting styrofoam tray to increase reaction temperature or exposing it to air to decrease temperature, as needed. The addition of 0.8 mL of $\%_{16}$ HCl was repeated and the reaction again allowed to subside. After cooling, 0.6 mL concentrated HCl and 0.4 mL concentrated HNO_3 (Baker Ultrex) were added to each tube. After waiting for the reaction to subside, 0.1 mL HF (Fisher) was added. The tubes were placed in an oven at 70°C for 2 h. After removal from the oven, 5 mL of deionized water was added and the tubes were allowed to sit overnight. Finally, 0.100 mL of 1000 μg mL^{-1} Sc solution (Aldrich) was added and the solutions were diluted to 10 mL total volume. All samples formed visually clear solutions. In our past experience with other alloys of aluminum, we have found that other methods of dissolution can be used with similar results, but we recommend the method described because it consistently results in complete dissolution, even for samples containing higher silicon concentrations, with minimal chance

of sample loss from overly vigorous reactions. The time required for one person to dissolve 50 foil samples using this procedure is 2 to 3 h.

Standard Preparation

Multielement standard solutions for ICP-AES were prepared by dilution of single element stock solutions, with concentrations of 1000 μg mL^{-1} (Spex, Aldrich). A matrix matching solution was prepared by dissolving 99.9999% aluminum metal (Alfa) in aqua regia and diluting to provide a stock solution containing 2.5% Al. Calibration standards were made by mixing the stock solutions and digestion acids in proportions to contain 12% HCl, 4% HNO$_3$, 2% HF, 1% Al, and 10.00 μg mL^{-1} Sc, so that they essentially matched the matrix (nonanalyte) composition of the samples. Standard concentrations for Cu, Fe, Ga, Mg, Mn, Ni, Si, Ti, V, and Zn were selected to bracket concentrations in samples. Typical standard concentrations covering the range of element concentrations in household aluminum foils are shown in Table 1. The elements, Cr, Pb, Sn, Sr, and Zr, were expected to be present in commercial foils at measurable concentrations only rarely, but are common in other aluminum alloys. A standard containing 1.00 μg mL^{-1} of each of these elements was used to check for their presence at measurable concentrations. Of these five elements, only Zr was observed to be present in any of the foils and it was only present at concentrations above the analytical detection limit in about half of the samples.

Instrumentation

Instrument and operating conditions for the ICP-AES (Perkin Elmer Plasma II) are shown in Table 2. Selection of analytical wavelengths was made, in part, to avoid interferences caused by the presence of high concentrations of aluminum in the sample solutions. Element intensities for the standards were determined and compared to anticipated values prior to sample analysis. Any nonlinearity of response or deviation from expected sensitivities outside of allowed limits dictated restandardization prior to sample analysis. Calculation of the concentrations of elements in samples was done using multielement external standard solutions and Sc as an internal standard for all elements. Use of an internal standard corrects for instrumental drift when it occurs and for minor differences in dilution volumes of the solutions analyzed. For sets of 20 to 30 samples, the first replicate of each sample was analyzed, followed by restandardization, then the second replicate, followed by repeated restandardization, and the third replicate. Thus the variation shown in triplicate measurements represents the combined effects of sample heterogeneity, dissolution derived uncertainty, and instrumental short- and long-term reproducibility. This approach yields a wider range of values for any given sample than

TABLE 1—*Standards for ICP-AES analysis of aluminum foil.*

Standard #0: 0.000 ppm all elements
Standard #1: 0.0100 ppm Mg; 0.100 ppm V & Ni; 0.200 ppm Ti, Zn & Mn; 0.400 ppm Cu;
     0.500 ppm Ga
Standard #2: 1.00 ppm Mg; 2.00 ppm V & Ga; 3.00 ppm Ni & Mn; 4.00 ppm Ti; 7.00 ppm Cu;
     10.00 ppm Zn
Standard #3: 4.00 ppm Si; 20.00 ppm Fe
Standard #4: 40.0 ppm Si; 100.0 ppm Fe
Standard #5: 1.00 ppm Cr, Sn, Pb, Zr, & Sr
All standards contain 12% HCl, 4% HNO$_3$, 1% HF, 1% Al, and 10.00 ppm Sc to match composition of samples.

TABLE 2—*ICP-AES instrumental operating conditions.*

Instrumentation	
Spectrometer model	Perkin-Elmer Plasma II
Dispersing system	Monochromator A: 3600 grooves mm^{-1}, resolution ≤ 0.009 nm, 160–400 nm
	Monochromator B: 1800 grooves mm^{-1}, resolution ≤ 0.018 nm, 160–800 nm
Torch	Fassel-type
Nebulizer	Perkin-Elmer high solids
Spray chamber	Scott design
Rf generator	27.12 MHz, 1.2 kW forward power
Analytical conditions	
Argon flow rates, L min^{-1}	Plasma 15
	Auxiliary 1.0
	Nebulizer 1.2
Sample uptake, mL min^{-1}	1.0
Observation height, mm	15 above load coil
Spectral lines, nm	Monochromator A: Sn 189.989, Cr 206.149, Zn 213.856, Pb 220.353, Ni 231.604, Fe 238.204, Si 251.611, Mn 257.610, Mg 279.553
	Monochromator B: V 292.402, Ga 294.364, Cu 324.754, Ti 334.941, Zr 343.823, Sr 407.771
Background correction	Auto
Signal compensation	Yes, Sc for all elements
Sampling time, msec	100

that which would be obtained by sequentially analyzing triplicate samples using a single calibration curve and, hence provides a slightly conservative estimate of the discrimination capability of the overall method. In casework applications, samples to be compared would be analyzed immediately following one another to provide tighter within sample variations and hence, more conservative estimates of a common source.

Results and Discussion

Analytical Figures of Merit

The analytical method used in these studies provides readily measurable emission intensities for the elements, Cu, Fe, Ga, Mg, Mn, Ni, Si, Ti, V, and Zn across the concentration ranges observed in commercial household aluminum foils. Concentrations of Zr were high enough to measure in only some of the samples and the concentrations of Cr, Pb, Sn, and Sr were below the analytical detection limits in all samples.

The precision of an analytical method can be determined by repeated measurements of a homogeneous reference material. Since no such sample exists for aluminum foil, we used samples taken from adjacent locations from one roll of household aluminum foil. The true analytical precision may be somewhat better than an estimate using this approach, because some sample heterogeneity may be included in our measured precisions. Nevertheless, excellent results were obtained. Results of these studies consistently show analytical precision to be in the 1 to 2% rsd range for the elements, Cu, Fe, Ga, Mg, Mn, Ti, V, and Zn. Precision measurements for Ni and Si are somewhat higher than the other elements. Decreased precision for Si may be due to incomplete dissolution, true sample heterogeneity, or leaching of Si from the ICP torch and spray chamber. Results for Ni are not as precise as the other elements, because it is the closest to the detection

limits. Direct measures of within sample precisions are included in the results of analysis of foil survey samples discussed later.

As with precision, it is not possible to assess directly the accuracy of the ICP-AES results, because aluminum foil alloy reference materials with well characterized concentrations of the elements of interest in this study are not available. Aluminum alloys, such as NIST Standard Reference Material (SRM) 87a are routinely analyzed in the FBI Laboratory by procedures similar to those used in this study. The ICP-AES results for these samples are within the accepted concentration ranges for those elements certified by NIST. However, as these alloys are different than those used in foil manufacture, these results only give an approximate indication of the accuracy of the method. It is also noted that the matching of sample and standard matrices for all nonanalyte constituents, as done in this procedure, minimizes systematic analytical errors.

Survey of Commercial Aluminum Foils

In the first part of this study, a survey of 30 rolls of aluminum foil was used to determine the within-roll variations and the ranges across brands for all measured element concentrations. The brand names of foil used in this survey are shown in Table 3. Wherever two samples of the same brand and type of foil were included, they bore different packaging codes, indicating that they were from different manufacturing production runs.

TABLE 3—*Aluminum foil survey sample descriptions.*

#	Brand Name	Type
1	Arrow	Regular
2	Giant Foods	Heavy Duty
3	Giant Foods	Regular
4	Econ	Regular
5	Giant Foods	Regular
6	Giant Foods	Heavy Duty
7	Reynolds Wrap	Regular
8	Reynolds 650	Regular
9	Reynolds 650	Regular
10	Safeway	Heavy Duty
11	Reynolds Wrap	Heavy Duty
12	Safeway	Heavy Duty
13	Diamond	Heavy Duty
14	Emerald	Regular
15	Diamond	Regular
16	Richfood	Regular
17	Safeway	Regular
18	Reynolds Wrap	Extra Heavy Duty
19	Safeway	Regular
20	Richfood	Regular
21	Diamond	Regular
22	Richfood	Heavy Duty
23	Reynolds Wrap	Regular
24	Reynolds 666	Heavy Duty
25	Richfood	Heavy Duty
26	Reynolds Wrap	Extra Heavy Duty
27	Reynolds Wrap	Heavy Duty
28	Budget Buy	Regular
29	Budget Buy	Regular
30	Econ	Regular

Each roll of foil was sampled in three locations, at approximately $\frac{1}{6}$, $\frac{1}{2}$, and $\frac{5}{6}$ of the distance from one end of the roll to the other. This sampling procedure was used to obtain a measure of the degree of variation in element concentrations along the length of each roll of foil. Ten elements, Cu, Fe, Ga, Mg, Mn, Ni, Si, Ti, V, and Zn were present at measurable levels in all 90 of the foil samples. The concentrations of these elements determined in each of the foil samples and some summary statistics are shown in Table 4. Several conclusions can be drawn concerning the data shown in Table 4. For all elements, the range of concentrations among foils is many times greater than the variation within individual rolls. The range values in Table 4 represent differences in both alloys and manufacturing processes used in the production of the 30 rolls of foil. Major differences among brands of foil for elements such as Cu, Fe, and Si arise from the use of different alloys for foil production. Smaller differences between foils for the concentrations of these elements and differences among foils for other elements that are not intentionally controlled by the manufacturers result in considerable variation even among rolls of the same brand name. This survey was not meant to be inclusive, so it is likely that the ranges in compositions of all commercially available aluminum foils are even greater than those shown in Table 4. The precision values shown in Table 4 were calculated by taking the relative standard deviations based on the triplicate measurements for each foil and finding the median value for the 30 foils. These values are measures of the combined analytical uncertainty over a period of about three hours and the within roll heterogeneity for each element. The precision values shown are not significantly different than the analytical precision we routinely expect when analyzing multiple samples from a homogeneous aluminum SRM. This indicates that, in general, the rolls of foil studied are compositionally homogeneous to such a degree that multiple samples removed from a roll are analytically indistinguishable. There are two characteristics of the results shown in Table 4 that indicate that elemental composition offers excellent discrimination capability among sources of aluminum foils. First, the individual samples are distributed throughout concentration ranges that are much larger than the precision for most elements. Second, there are a relatively large number of elements which can be determined with good precision in all foils. These factors combine to place each sample at a precise position in a relatively large 10 dimensional compositional universe.

One method which has been used to determine whether two items are analytically indistinguishable is to determine whether there is an overlap in the range of values represented by the triplicate measurements for all measured elements in the two items [3,10]. In comparing two foils, nonoverlapping ranges for one or more elements renders the foils analytically distinguishable and, in general, they can then be attributed to different sources. Taking the 30 rolls of foil in this study, two at a time, shows them all to be analytically distinguishable using the range overlap criteria. For most roll to roll comparisons in this study, there are differences in the concentrations of several of the elements and these differences are often very large in comparison to the within roll variations. This comparison supports the excellent discrimination capability hypothesized previously and the conclusion that there is a very small possibility of a chance overlap of all measured element concentrations for two rolls of aluminum foil from different manufacturing sources.

Although all measured element concentrations must be analytically indistinguishable for two samples to be attributable to a common source, clearly some elements have better discrimination capability than others. From the relative magnitudes of range and precision data shown in Table 4, the elements Cu, Zn, and Mg would seem to have the best discrimination capability among the rolls of foil. In fact, the results for Cu, Zn, and Mg alone are sufficient to distinguish among all 30 foils. In the worst instance, samples #2 and #12, although foils of different brands and gauges, have overlapping concentration ranges for Cu and Mg and are arguably close in Zn. However, the advantage of a multielement method is demonstrated by the fact that these two foils have readily dis-

TABLE 4—*Element concentrations in samples from 30 rolls of aluminum foil. All concentrations expressed in µg/g of foil.*

Sample	Mg	Ga	Zn	Cu	Ti	Ni	Fe	Mn	Si	V
1a	11.4	124	36.5	740	147.5	123	3600	57.0	596	143
1b	11.2	135	36.7	736	148.6	116	3590	55.6	630	146
1c	11.2	134	39.1	730	150.6	121	3580	56.6	697	142
2a	2.7	123.7	213	777	50.1	23	4500	37.4	773	65.8
2b	2.7	124.0	210	785	50.3	19	4520	37.1	833	67.3
2c	2.7	125.3	219	770	51.5	27	4500	37.4	906	65.2
3a	68.3	86.7	144	284	87.4	9.6	3270	65.4	808	21.9
3b	68.3	89.7	146	286	88.1	11.8	3240	64.0	915	21.5
3c	68.3	90.6	151	286	90.5	9.0	3260	64.9	1035	21.9
4a	24.6	92.5	251	54.1	49.1	12.1	5550	43.2	3510	40.2
4b	25.0	92.9	251	54.4	48.3	6.9	5540	42.4	3680	40.0
4c	24.7	86.0	259	53.5	49.5	13.5	5500	42.5	4050	40.0
5a	8.5	113.3	174	831	31.0	25.3	4370	62.0	882	76.1
5b	8.7	112.7	169	840	31.3	22.8	4330	61.2	919	77.1
5c	8.5	110.0	170	803	31.4	23.8	4190	59.9	1015	74.9
6a	73.6	156	587	201	254	42.0	3640	260	1040	56.1
6b	77.7	168	597	210	267	43.9	3730	260	1120	59.7
6c	76.2	167	615	208	270	44.7	3730	266	1260	58.7
7a	31.7	98.0	78.2	81.8	71.1	20.0	6540	55.0	3440	58.4
7b	31.2	99.9	78.9	82.3	70.1	15.6	6370	52.6	3460	59.3
7c	32.0	100.7	80.0	82.7	73.9	18.2	6520	54.4	4020	58.3
8a	21.6	100	187	51.8	47.4	13.0	5650	43.0	3800	41.8
8b	21.7	107	190	52.6	46.5	12.9	5580	41.3	3680	42.8
8c	21.7	108	194	53.2	49.2	14.6	5620	41.9	4160	42.2
9a	12.9	83.6	240	60.2	57.6	10.0	6300	44.6	4110	35.7
9b	12.6	81.9	239	61.4	57.4	13.1	6200	42.8	4040	36.8
9c	13.0	81.1	250	61.7	60.2	14.0	6380	43.8	4480	36.6
10a	3.4	126	199	776	32.2	20.7	4290	38.5	897	55.0
10b	3.3	133	199	805	32.6	20.3	4240	37.3	894	55.3
10c	3.4	129	207	789	33.6	21.7	4220	37.1	993	55.2

11a	46.1	104	64.4	75.2	72.2	11.2	6240	30.6	4010	62.6
11b	46.3	109	62.5	76.3	70.6	16.5	6060	29.0	3910	63.3
11c	45.9	103	62.8	74.0	73.5	12.4	6040	28.9	4310	62.6
12a	2.7	113	202	783	34.7	25.3	4250	45.1	874	79.2
12b	2.5	120	200	800	35.0	20.3	4150	43.1	835	81.3
12c	2.8	124	208	798	36.3	23.1	4200	44.9	950	81.0
13a	17.8	93	144	67.3	55.5	11.8	6190	32.0	3590	39.5
13b	17.8	104	142	67.8	55.0	12.4	6000	30.1	3390	39.1
13c	18.1	92	149	67.1	57.5	14.3	6100	31.5	3880	40.1
14a	101.3	129	22.8	805	111	44.3	5500	44.0	736	96.7
14b	102.6	136	22.8	823	111	41.9	5360	41.7	702	98.7
14c	104.0	138	22.1	830	116	41.3	5420	42.7	808	99.0
15a	20.9	92	124	58.7	60.1	11.4	5860	33.4	3700	42.8
15b	21.0	97	122	61.2	60.7	14.0	5700	32.2	3500	ND
15c	21.1	99	128	60.2	62.8	12.9	5750	32.7	4030	43.5
16a	38.2	110	69.0	54.4	69.1	13.0	5700	37.9	3840	65.0
16b	37.8	111	62.3	55.3	70.1	12.4	5510	36.4	4350	65.6
16c	38.9	114	70.9	55.2	73.3	15.0	5620	36.8	4160	67.9
17a	3.7	101	145	701	35.8	34.1	4860	49.1	970	97.7
17b	3.7	106	130	706	36.5	31.8	4670	47.5	1097	100.2
17c	3.9	108	149	717	38.1	32.5	4790	48.0	1090	100.5
18a	11.2	89	280	52.6	75.1	13.5	5790	45.6	3720	35.4
18b	12.0	92	251	52.7	77.1	12.8	5580	44.4	4250	35.7
18c	12.3	94	287	53.5	79.3	17.9	5740	45.0	4080	36.2
19a	3.0	114	287	726	46.6	26.8	4500	46.6	903	83.9
19b	2.5	117	262	739	47.9	25.4	4390	46.1	1024	85.7
19c	2.3	120	290	745	49.1	30.1	4430	46.1	996	85.0
20a	30.5	97.2	66.8	62.7	58.5	14.9	6030	33.9	3750	50.0
20b	30.7	100.3	61.9	63.0	60.3	11.5	5840	33.4	4280	50.8
20c	31.1	99.4	68.2	64.0	61.4	18.5	5960	32.5	4080	51.4
21a	33.0	91	232	57.8	36.7	11.0	6000	51.6	3840	30.4
21b	32.4	91	213	57.9	37.9	9.5	5750	48.9	4340	31.5
21c	36.2	98	250	63.8	43.2	14.1	6350	53.2	4470	34.1
22a	20.5	113	249	70.4	50.6	12.8	6900	46.6	4280	47.2
22b	19.4	111	211	67.3	49.0	10.3	6370	44.2	4710	46.5
22c	18.6	104	230	65.2	49.1	14.7	6120	42.4	4260	43.7

TABLE 4—Continued.

Sample	Mg	Ga	Zn	Cu	Ti	Ni	Fe	Mn	Si	V
23a	58.4	97.4	65.1	77.9	68.2	11.8	6340	31.6	4160	50.2
23b	56.3	98.3	59.6	77.7	69.3	14.6	6220	31.4	4670	50.5
23c	57.5	97.7	66.9	78.7	70.5	12.9	6120	30.9	4410	50.3
24a	16.0	142	162	71.0	71.3	13.4	5940	66.3	3770	47.7
24b	15.6	146	144	70.8	72.5	12.4	5670	63.9	4250	48.0
24c	16.1	150	161	71.5	73.7	16.2	5670	63.1	4020	48.6
25a	18.6	100	67	58.2	53.1	12.5	6000	32.4	3970	51.5
25b	18.1	93	63	58.2	53.7	10.6	5760	31.4	4600	52.3
25c	20.1	107	71	61.5	56.6	13.6	6050	32.9	4580	53.7
26a	16.1	96	107	68.6	55.3	19.0	6150	47.6	4190	58.9
26b	16.0	95	104	67.9	55.6	18.6	5970	45.6	4760	59.6
26c	16.1	101	116	68.9	57.5	21.0	6040	45.6	4550	59.6
27a	22.7	99	63.9	67.4	62.8	13.2	5870	36.5	3790	55.0
27b	22.3	103	63.5	66.0	62.9	13.0	5680	34.2	4250	54.5
27c	22.7	105	67.0	66.9	63.7	15.5	5710	34.4	4030	55.2
28a	11.5	216	48.4	2250	119.0	70	4020	82.9	757	102.8
28b	11.3	209	43.3	2230	120.4	63	3880	80.4	849	103.0
28c	11.4	222	49.8	2280	121.9	68	3880	80.7	818	102.0
29a	5.3	181	45.8	1300	129.5	32.2	4390	120	818	89.0
29b	5.2	175	43.7	1290	129.9	31.9	4240	115	914	89.2
29c	5.3	180	45.7	1310	133.9	32.2	4350	116	885	90.6
30a	24.1	90.4	236	52.9	47.8	8.0	5590	43.0	3720	38.6
30b	23.9	90.3	217	53.6	47.0	11.0	5460	42.0	4170	40.5
30c	24.4	89.9	250	53.2	49.0	13.6	5500	41.8	4020	39.7
Min.	2.6	82	23	52	31	10.1	3260	30	641	22
Max.	103	216	600	2250	264	120	6480	262	4500	144
Mrsd, %	1.3	1.2	2.8	1.1	2.2	10.5	1.5	2.0	6.7	1.2

NOTES:
ND = not determined, spectrometer drift error.
Min. = minimum mean for individual roll of foil.
Max. = maximum mean for individual roll of foil.
Mrsd = median value of relative standard deviation based on triplicate measurements for each foil.

tinguishable concentrations of Fe, Ti, Mn, and V. Although, we obtain complete separation of the 30 foils with only a few elements, there clearly is a tremendous potential for discrimination when 10 elements are used, a fact that will be important in future expansion of the size of the comparison set.

One aspect of individual element concentrations is that they are important in distinguishing among the several alloys used to make foil and, therefore, one foil manufacturer from another. The foils used in this study include 18 made by the Reynolds Aluminum Company bearing the brand names, Reynolds, Richfood, Econ, and Diamond. These products are characterized by consistently higher Fe and lower Cu than the other foils which were made using different aluminum alloys.

Variation Within Manufacturing Production Runs

A second study was devised to determine the results of within lot number variability as a measure of production run variability. Thirty six samples from rolls of foil not used in the previous survey were selected for this study, as follows. Three samples were taken from each roll of foil, three rolls of foil were selected from each packaging code, two packaging codes were taken from each manufacturer, and two manufacturers were represented. The element concentration results determined in this study are shown in Table 5. Each analytical sample is identified by manufacturer (R or G), packaging code (A, B, X, or Y), box (1, 2, or 3), and replicate (a, b, or c). For the Reynolds brand foils, all rolls are distinguishable from each other. That is, for every comparison between results for two boxes of foil, the ranges of the triplicate values for one or more elements do not overlap. Within each of the two packaging code designations for Giant brand foil, there are a pair of indistinguishable foils and a third different foil. The indistinguishable rolls are the pair of GB2 and GB3 and the pair of GA1 and GA2. In most instances, distinguishable foils, even those bearing the same packaging code, contain quite large differences in the concentrations of several elements. For example, the Zn concentration in roll GA3 is 2.5 times greater than in GA1 and GA2. As in the survey portion of the study, there are no instances of two boxes of foil having indistinguishable compositions, but having different brands or packaging codes.

No attempt was made to interpret this data in terms of production volumes or differences in procedures among manufacturers. Such information may be helpful in assessing the significance of compositional overlaps, but is not included here because generalized statements applied across manufacturers, production plants, or product lines would be premature for the limited number of samples studied. Information of this type will be accumulated as evidence from known sources is analyzed and the database of foil composition knowledge increases.

Conclusions

The ICP-AES procedure described is relatively quick and provides excellent precision for the determination of 10 elements in 100 mg sized samples of commercial aluminum foils. The wide range in composition among foils and the good within roll precision indicates that a good degree of source discrimination is possible. In general, since most sources of foil seem to have different compositions, forensic significance can be placed on the results of comparison of two foil samples. When two samples have compositions that differ outside the range seen for one roll of foil, then, barring contamination, one can exclude the possibility of both coming from the same roll of foil. When two samples have indistinguishable compositions, then one can say that they may have come from the same roll of foil or similar rolls, and that the majority of other rolls can be ruled out as possible sources of the questioned foil.

TABLE 5—*Production run variability of Reynolds aluminum foil samples.*

Sample	V	Zn	Ga	Ni	Cu	Fe	Ti	Si	Mn	Mg
RX1a	110.4	84	129	46.2	32.2	6170	52.8	4620	27.5	3.6
RX1b	108.7	80	124	46.8	30.5	6050	52.7	4520	27.4	3.4
RX1c	111.4	80	132	45.3	30.4	6000	53.2	4490	27.6	3.6
RX2a	114.7	83	118	48	31.7	6080	46.5	4460	26.4	4.0
RX2b	114.7	81	126	44	32.0	6000	47.0	4420	26.1	3.9
RX2c	115.1	82	128	51	33.2	6040	47.5	4420	26.5	3.8
RX3a	113.0	80	125	52.3	27.3	6350	42.3	4730	35.8	2.8
RX3b	114.1	76	136	51.0	25.1	6230	42.4	4650	35.6	3.0
RX3c	113.4	79	138	52.7	26.4	6150	42.6	4590	35.0	3.0
RY1a	133.1	50.6	118	40.2	33.4	6200	52.3	4910	24.3	4.6
RY1b	130.0	51.2	117	40.2	33.4	6070	51.5	4900	23.9	4.5
RY1c	129.8	50.6	120	41.6	33.2	6110	51.8	4890	24.0	4.4
RY2a	130.8	43	98	50	37.1	5950	63.5	4560	25.2	3.9
RY2b	130.4	44	109	46	37.3	5890	63.1	4430	24.6	4.1
RY2c	133.3	40	107	49	38.1	6060	64.7	4540	24.8	4.0
RY3a	114.9	93	128	49.6	40.6	5470	42.6	4490	32.2	5.9
RY3b	110.8	90	128	48.5	39.1	5260	41.6	4330	31.2	5.7
RY3c	119.0	92	129	49.4	42.1	5630	44.1	4550	32.8	6.3

Sample										
GB1a	87.0	53.5	116.4	32	27.1	6520	73.8	1300	30.0	9.5
GB1b	87.1	51.4	116.5	30	26.5	6510	74.0	1250	29.6	9.2
GB1c	86.4	52.0	115.4	28	26.8	6550	73.2	1250	30.4	9.4
GB2a	86.7	53.9	118	30.3	24.1	6530	70.6	1290	24.4	6.9
GB2b	88.4	54.9	115	31.6	24.6	6600	72.2	1260	24.7	6.4
GB2c	87.1	55.6	115	30.2	24.1	6540	70.7	1240	24.2	6.3
GB3a	87.7	54.9	118	33	25.0	6550	71.1	1270	25.0	6.4
GB3b	86.8	54.7	115	30	24.8	6520	70.4	1240	24.4	6.3
GB3c	86.9	56.2	116	30	25.5	6530	71.1	1240	24.4	6.4
GA1a	86.1	82.9	107	29	55.8	6370	107.8	1250	52.4	8.8
GA1b	84.9	82.7	109	29	55.2	6310	107.4	1170	51.9	8.9
GA1c	84.4	82.5	111	34	55.3	6370	108.1	1210	51.9	9.5
GA2a	85.6	81.5	103	30.1	55.3	6280	107.1	1230	51.6	8.7
GA2b	84.6	81.7	105	30.8	55.3	6300	106.7	1200	51.6	8.8
GA2c	84.4	80.9	108	30.0	56.5	6270	108.5	1190	51.7	8.9
GA3a	94.3	224	119	32	161.0	6390	99.7	1270	65.4	9.2
GA3b	95.3	219	120	31	161.9	6410	100.7	1250	65.5	9.3
GA3c	94.5	218	118	34	161.7	6360	100.0	1220	65.3	9.2

Sample identification key:
1st digit is brand: R = Reynolds Wrap, G = Giant Foods.
2nd digit is packaging code: A and B for Giant, X and Y for Reynolds.
3rd digit is roll number, arbitrarily assigned.
4th digit is within roll replicate designation, arbitrarily assigned.

All samples in these studies were removed from unusued rolls of foil and effects of contamination were not specifically studied. It must be considered that evidentiary foil samples may be contaminated as a result of their use prior to receipt by the forensic laboratory. In preliminary studies, we have washed several brands of foil with laboratory detergent and acetone to determine the effects on measured element concentrations. These studies have shown that the ten elements of interest are neither leached from nor added to foils by simple washing with clean reagents. Also, much of the surface contamination of aluminum foil can be readily removed by wiping and washing. The efficacy of various washing procedures has not been studied, but clearly depends upon the identity and amount of the contaminant as well as the details of the washing procedure. In comparing evidentiary foils, the use of appropriate replicate samples will indicate when contamination is present, because the contaminant elements will not be evenly distributed across all areas of the foil. The higher than normal standard deviations for replicates will indicate that contamination is present and that its removal is dictated. Further studies are needed to assess the effects of contamination of foils on the interpretation of results.

Most evidentiary aluminum foil samples are quite large, so sample sizes of a few hundred milligrams are available. When this is not the case, the procedure described here can be scaled down to a few tens of milligrams without significant loss of analytical precision. Small sample sizes may, however, result in less discrimination capability than that seen in this study if there is measurable element heterogeneity between smaller samples from the same roll of foil.

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