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A Metallurgical Provenance Study of the Marcus Herennius Military Diploma

Michael J. Dorais and Garret L. Hart

The bronze used to make the military diploma for the Roman soldier Marcus Herennius in AD 109 is heterogeneous in texture and composition. In contrast to modern bronze, lead (Pb) inclusions are common, and the bronze shows a considerable range in copper (Cu) (73 to 92.7 weight percent) and tin (Sn) (6.1 to 26.5 weight percent). Lead isotopic compositions are identical to those of copper coins produced during the Imperial Era of Augustus and Tiberius and may indicate mixtures of ores from southeastern Spain and Sardinia. The combination of these analytical results provides permissive evidence that the plates are authentic artifacts manufactured at the turn of the second century AD.

Determining Authenticity

Two Roman bronze plates known as a military diploma were awarded to Marcus Herennius on October 14, AD 109. Military diplomas are thought to be legal documents for retired soldiers containing inscriptions of service and citizenship. A common objective of archaeological and geoarchaeological endeavors is to determine the authenticity of such artifacts and, if possible, the provenance of the materials used in their construction.

Over the past several decades, geologists and geoarchaeologists have determined the lead isotopic values of various ore deposits throughout the Mediterranean and compared the lead isotopic compositions of Bronze Age and Classical artifacts to determine provenance. In this contribution, we show that the lead isotopic compositions of the Herennius military diploma compare favorably with the compositions of ore sources used

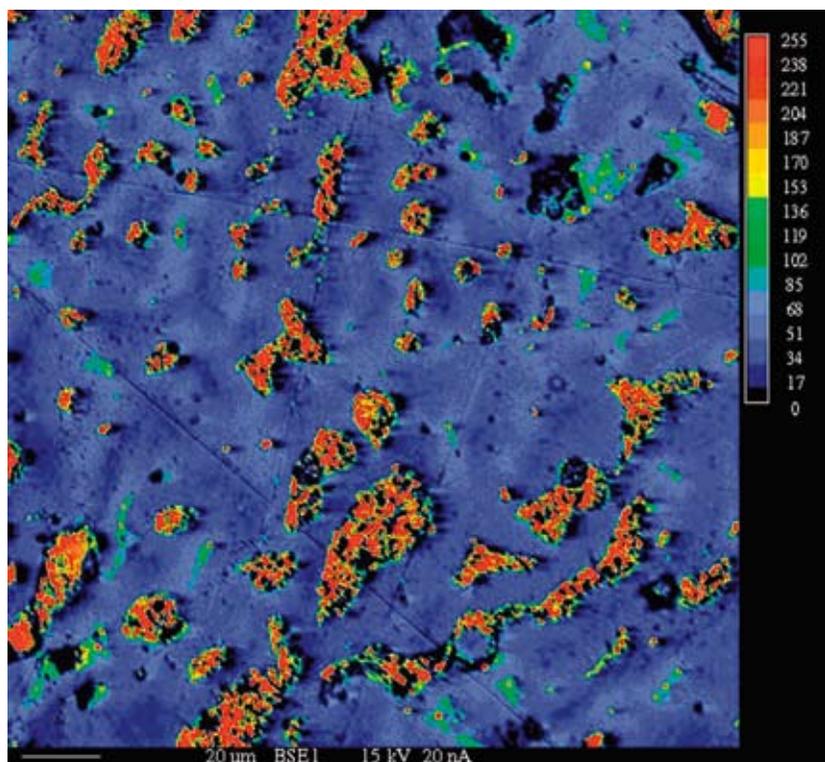


FIG. 1. Backscatter electron image of bronze with false color added to emphasize compositional contrasts. Orange patches are lead inclusions. The bronze host shows a range in compositions; green domains contain up to 26% tin, bright blue domains up to 12%, and the dark blue domains as little as 6%. Black domains in upper right portion of the image are essentially pure copper. Twenty micron scale for reference.

during Roman times and of early Imperial Age coins. We also show that the plates are texturally and compositionally heterogeneous, consistent with bronze made from known Roman smelting practices.

Instrumental Methods

Several small flakes of bronze were scraped from each of the plates. A few grains were mounted in epoxy, polished, and carbon coated for electron microprobe analysis. Microprobe analyses were conducted with a Cameca SX50 in the Department of Geological Sciences at Brigham Young University. The electron microprobe focuses an electron beam on the polished sample, generating X-rays that are measured and compared

to standards to obtain a chemical analysis. The beam can also be rastered across a sample to provide backscattered electron images. In these images, the brightness of domains in the sample is a function of the average atomic number of the compound under the beam; domains with high average atomic numbers are bright. Beam conditions for the analyses of this study were 25 KV, 20 nA with a 1–2 micron beam diameter; imaging was conducted with a 15 KV, 20 nA beam.

A second split of the bronze samples was sent to the Department of Geology, Washington State University, for isotopic analyses. The samples were dissolved in HF and HNO₃, and lead was separated using ion-exchange chromatography. Dilute solutions (100 ppb) of lead were prepared for mass spectrometry. Thallium (Tl) (30 ppb) was added to the lead solutions for In-In mass fractionation corrections using 2.3880 for ²⁰⁵Tl/²⁰³Tl. The samples were run on a multicollector ICP-MS (Thermo-Finnigan Neptune). Ion signals were measured in 7 Faraday cups in low resolution with 3 blocks of 25 ratios preceded by a 50-second baseline. Mercury (²⁰²Hg) was monitored as a possible isobaric interference. Accuracy was monitored using NIST 981, and the 2σ external errors (n=10) are 0.007% for ²⁰⁶Pb/²⁰⁴Pb and ²⁰⁷Pb/²⁰⁶Pb, and 0.002% for ²⁰⁸Pb/²⁰⁴Pb.

Electron Microprobe Analyses

Figure 1 is a backscatter electron image of one of the bronze flakes. Scattered throughout the sample are orange-colored, irregular-shaped, lead-rich inclusions that range up to 40 microns in length. The bronze that hosts the lead inclusions is heterogeneous in composition as shown by the color variations; the green, bright blue and dark blue domains contain different amounts of tin. The black domains in the upper right portion of the image are essentially pure copper.

Table 1 (see appendix) gives electron microprobe analyses of the bronze, which are plotted in figure 2. The copper contents range between 73.1 and 92.7 wt %. Tin varies antithetically to copper from a low value of 6.1 to a high of 26.5 wt %. The richest tin regions are the green domains of figure 1. Bright blue and dark blue domains in figure 1 contain up to 12% and 6% tin respectively. Plotted in figure 2 are analyses of modern bronzes that also show wide range in copper and tin content but, in contrast to the heterogeneous nature of the plates, are very homogeneous. Each open circle represents a different type of modern bronze that shows no variation of copper and tin content.

Other elements analyzed in the Roman plates are given in table 1. Iron (Fe), nickel (Ni), and zinc (Zn) have concentrations of less than 0.1 wt %.

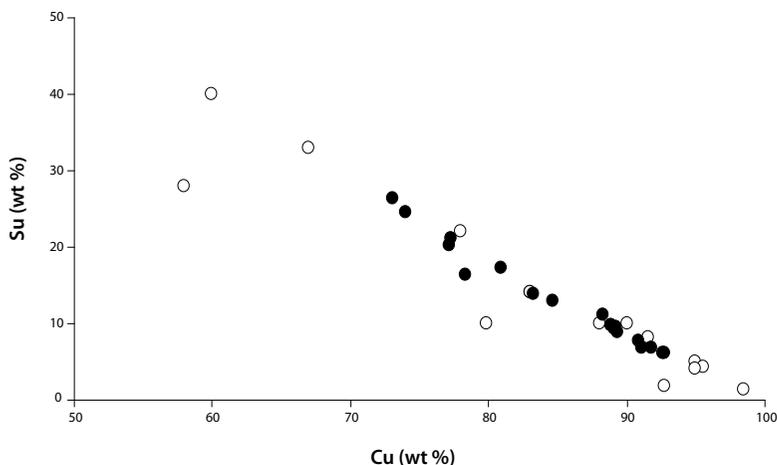


FIG. 2. Copper versus tin (weight percent) diagram. Various domains of bronze in the military diplomas (filled circles) range from 73 to 93 wt % copper with tin varying antithetically. Modern bronzes (open circles) contain a wide range of copper to tin concentrations but are homogeneous. Each open circle shows the composition of specific types of modern bronze. (Oliver Seely, “Composition and Physical Properties of Alloys,” www.csudh.edu/oliver/chemdata/alloys.htm [accessed June 23, 2006])

Arsenic (As) and lead are more abundant, averaging 0.34 and 0.85 wt % respectively, but with highs of 1.5 and 3.6 wt % respectively.

Lead Isotopes

Lead isotopic compositions of the Roman plates are given in table 2 (see appendix). Three analyses of each sample were taken with the average of each sample plotted in the $^{207}\text{Pb}/^{206}\text{Pb}$ versus $^{208}\text{Pb}/^{206}\text{Pb}$ diagram (filled circles in figure 3). Reference fields for copper ores across the Mediterranean used during early Imperial Roman times are also plotted.¹ In this diagram, the $^{207}\text{Pb}/^{206}\text{Pb}$ ratio varies as a function of the age of the ore deposit. High $^{207}\text{Pb}/^{206}\text{Pb}$ values characterize Mediterranean ores from the

1. Fields from compilation of S. Klein, Y. Lahaye, G. P. Brey, and H. M. von Kachel, “The Early Roman Imperial AES Coinage II: Tracing the Copper Sources by Analysis of Lead and Copper Isotopes—Copper Coins of Augustus and Tiberius,” *Archaeometry* 46 (August 2004): 469–80.

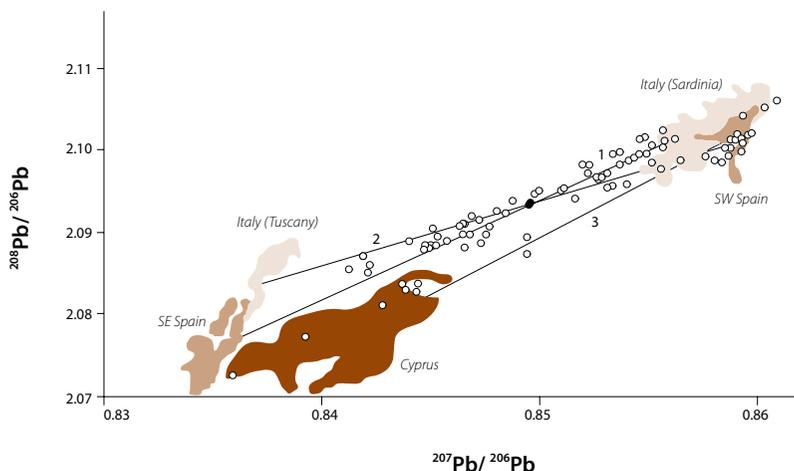


FIG. 3. $^{207}\text{Pb}/^{206}\text{Pb}$ versus $^{208}\text{Pb}/^{206}\text{Pb}$ diagram. Dark fields represent the compositions of ore deposits in Cyprus, medium from deposits in Spain, and light from deposits in Italy. Open circles are coin analyses from Klein (Klein and others, “Early Roman Imperial AES Coinage II,” 469–80). Filled circles are averages of three analyses of each plate of the military diploma. Lines 1, 2, and 3 represent mixing lines between Sardinia and southeastern Spain, Tuscany and either Sardinia or southwestern Spain, and Cyprus and either Sardinia or southwestern Spain respectively. See text for discussion.

Cambrian period (about 543 to 520 million years ago), whereas younger Tertiary (65 to 1.8 million years ago) ores have low $^{207}\text{Pb}/^{206}\text{Pb}$ values. Older ores from Sardinia and southwestern Spain define fields at $^{207}\text{Pb}/^{206}\text{Pb}$ values between 0.855 and 0.86; younger Tuscany, southeastern Spain, and Cyprus ores plot below 0.845. The $^{208}\text{Pb}/^{206}\text{Pb}$ ratios are indicative of the uranium (U)/thorium (Th) ratios of the fluids that formed the ores. Again, there is a distinction between the younger Tuscany, southeastern Spain, and Cyprus ores and those of Sardinia and southwestern Spain. An important feature of the $^{207}\text{Pb}/^{206}\text{Pb}$ versus $^{208}\text{Pb}/^{206}\text{Pb}$ diagram is the absence of ore deposits in the Mediterranean with $^{207}\text{Pb}/^{206}\text{Pb}$ values between 0.845 and 0.855.

For comparison with our samples, we have also plotted Augustan and Tiberian Imperial Age coin analyses from Klein’s study.² Some of the coins plot at high $^{207}\text{Pb}/^{206}\text{Pb}$ values in the southwestern Spain field, others

2. Klein and others, “Early Roman Imperial AES Coinage II,” 469–80.

in the Sardinia field. A few coins plot in the Cyprus field. The majority of the coins, however, plot in gap between known Mediterranean ore deposits and probably represent mixtures of ores from multiple sources.³ The averages of the two Roman plates of this study also plot in the gap, at $^{207}\text{Pb}/^{206}\text{Pb}$ values of approximately 0.85.

Major and Minor Element Composition

Several factors indicate that the plates were manufactured by relatively primitive smelting techniques and suggest that they are authentic bronze plates from the Roman era. Compared to modern bronze, the proportions of copper to tin in the plates is not unusual—it is the heterogeneity that markedly differs. Modern bronzes have a range in tin content, some up to 40 wt %, but bulk samples are homogeneous as shown by the individual open circles in figure 2. The bronze in these plates varies between 92.7 and 73.1 wt % copper and between 6.1 and 26.5 wt % tin over a scale of a few microns as shown by the color changes from dark blue to green in the bronze of figure 1. These compositions are similar to those of Classical bronze statues analyzed by Lie and Mattusch that range between 70 and 90 wt % copper and from 5 to 10 wt % tin.⁴ Unlike modern bronze, the arsenic, iron, nickel, and zinc contents are also variable. The heterogeneous bronze composition of the military diplomas indicates rather poor efficiency in smelting the ores, producing an impure, poorly mixed product.

High lead contents are common in ancient bronze artifacts, many of which have bulk-sample concentrations of several wt % lead (2 to 28 wt %).⁵ Most of the microprobe analyses of the diploma have less than 1.0 wt % lead, considerably lower than the several wt % of other ancient bronze artifacts. This is because the microprobe analyses were of small, 1–2 micron domains and not of the bulk sample; the presence of many lead inclusions would yield a bulk-sample analysis of several wt % lead in common with other ancient bronze artifacts.

3. Klein and others, “Early Roman Imperial AES Coinage II,” 469–80.

4. Henry Lie and Carol C. Mattusch, “Introduction to the Catalogue Entries and Technical Observations,” in *Fire of Hephaistos: Large Classical Bronzes from North American Collections* by Carol C. Mattusch (Cambridge: Harvard University Art Museums, 1996), 162–78.

5. S. Siano, L. Bartoli, J. R. Santisteban, W. Kockelmann, M. R. Daymond, M. Miccio, and G. De Marinis, “Non-destructive Investigation of Bronze Artefacts from the Marches National Museum of Archaeology Using Neutron Diffraction,” *Archaeometry* 48 (February 2006): 86–87.

The presence of abundant lead inclusions in the plates is consistent with the manufacturing techniques described by Gaius Plinius Secundus, commonly known as Pliny the Elder, who wrote in the first century AD:

Among the remaining kinds of copper the palm goes to the bronze of Campania, which is most esteemed for utensils. There are several ways of preparing it. At Capua it is smelted in a fire of wood, not of charcoal, and then poured into cold water and cleaned in a sieve made of oak, and this process of smelting is repeated several times, at the last stage Spanish silver lead being added to it in the proportion of ten pounds to one hundred pounds of copper: this treatment renders it pliable and gives it an agreeable colour of a kind imparted to other sorts of copper and bronze by means of oil and salt. Bronze resembling the Campanian is produced in many parts of Italy and the provinces, but there they add only eight pounds of lead, and do additional smelting with charcoal because of their shortage of wood. . . .

The proper blend for making statues is as follows, and the same for tablets: at the outset the ore is melted, and then there is added to the melted metal a third part of scrap copper, that is copper or bronze that has been bought up after use. This contains a peculiar seasoned quality of brilliance that has been subdued by friction and so to speak tamed by habitual use. Silver-lead is also mixed with it in the proportion of twelve and a half pounds to every hundred pounds of the fused metal. There is also in addition what is called the mould-blend of bronze of a very delicate consistency, because a tenth part of black lead is added and a twentieth of silver-lead; and this is the best way to give it the colour called Græcanic “after the Greek.” The last kind is that called pot-bronze, taking its name from the vessels made of it; it is a blend of three or four pounds of silver-lead with every hundred pounds of copper. The addition of lead to Cyprus copper produces the purple colour seen in the bordered robes of statues.⁶

The abundance of lead inclusions in the plates is consistent with Pliny’s description of lead deliberately added to obtain either the desired color or pliability for sheets of metal, such as those used for the military diplomas, and with analyses of other Roman bronze artifacts.⁷

Both the textural features shown in figure 1 and the compositional heterogeneity of the bronze suggest the plates were not manufactured by

6. Pliny, *Natural History*, trans. H. Rackham, 10 vols. (Cambridge: Harvard University Press, 1938–63), vol. 9, bk. 34, ch. 20, pp. 197, 199, 201. Footnotes (198, 199a) to the term “silver-lead” indicates that it is actually not a true mixture of silver and lead, but instead is tin and lead in equal proportions.

7. Lie and Mattusch, “Introduction to the Catalogue Entries,” 162–78; Siano and others, “Non-destructive Investigation of Bronze Artefacts,” 77–96.

modern techniques but by techniques consistent with what one would expect for bronze from the Roman era.

Lead Isotopic Compositions

Three mixing lines are drawn in figure 3 to explain the mixed sources of some Imperial Age coins.⁸ Line 1 is a mixing array between ores from Sardinia and southeastern Spain. Line 2 shows a mixing of a component from Tuscany with either a Sardinian or a southeastern Spanish component. Line 3 is a mixing line between a component from Cyprus and either a Sardinian or southeastern Spanish component.

The majority of the early Imperial Era coins analyzed by Klein were interpreted as having been made by smelting ores from Sardinian and southeastern Spanish sources.⁹ The trend of coin analyses plotting along line 1 suggests use of ores from those two sources in varying proportions, generating the spread of $^{207}\text{Pb}/^{206}\text{Pb}$ values from approximately 0.84 to 0.855. The coins that plot adjacent to the Sardinia field indicate that they are mainly composed of Sardinian ore with only a minor component of southeastern Spanish ore. The proportion of southeastern Spanish ore increases with lower $^{207}\text{Pb}/^{206}\text{Pb}$ values; coins plotting at the low $^{207}\text{Pb}/^{206}\text{Pb}$ values of approximately 0.842 were made from roughly 66% southeast Spain ore and 33% Sardinian ore.

As our samples plot along the dominant coin trend defined by line 1 between Sardinia and southeastern Spain, the plates probably represent mixtures of these two sources. The plates have a $^{207}\text{Pb}/^{206}\text{Pb}$ value of approximately 0.849, indicating a mixture of roughly 70% Sardinian and 30% southeastern Spanish ores. But because our two samples are nearly isotopically identical, we lack the spread in $^{207}\text{Pb}/^{206}\text{Pb}$ values to define a trend that would clearly identify the endmembers of mixing. It is equally plausible that the plates were made from mixtures of either Sardinian or southwestern Spanish ore with ore from Tuscany (mixing line 2). With respect to provenance, it is interesting to note that Pliny wrote that the tin-lead mixture that was added to copper was obtained from Spain.¹⁰

Another possible explanation for the lead mixtures found in the plates is that they could have been made from recycled bronze. In 1992, a scuba diver discovered a Roman shipwreck off the Italian port city of Brindisi. Archaeologists found that the ship contained bronze artifacts that range

8. Such lines were used by Klein and others, "Early Roman Imperial AES Coinage II," 469–80.

9. Klein and others, "Early Roman Imperial AES Coinage II," 477–79.

10. Pliny, *Natural History*, bk. 34, ch. 20, pp. 198–199.

in age from the fourth century BC to the third century AD. It appears that the materials found in the Brindisi shipwreck were to be used for recycling purposes,¹¹ providing verification of the use of scrap bronze mentioned by Pliny the Elder in the above quotation. Rather than representing mixtures of multiple ore sources, the mixed lead isotopic signature of the military diploma shown in figure 3 can also be interpreted in the context of Pliny's description of Roman bronze recycling practices. The plates could just as well represent recycled bronze from multiple sources.

Conclusions

The available textural, major, minor, and isotopic evidence strongly supports the authenticity of the Marcus Herennius military diploma. The bronze has both textural and compositional characteristics indicative of relatively primitive smelting techniques, features that are not characteristic of modern processes but are consistent with the techniques described by Pliny. Additionally, the lead isotopic composition of the bronze is identical to known early Imperial Age coins. The isotopic composition indicates that the bronze was probably smelted from two ore sources, most likely the same dominant southeastern Spanish and Sardinian ores that characterizes the majority of coins. A mixture from Tuscany and Sardinia or from Tuscany and southwestern Spain, however, cannot be disregarded.

11. O. Louis Mazzatenta, "The Brindisi Bronzes: Classical Castoffs Reclaimed from the Sea," *National Geographic* 187, no. 4 (April 1995): 90.

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Appendix

Table 1. Representative Electron Microprobe Analyses of the M. Heremnius Military Diploma

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
S	0.000	0.011	0.007	0.006	0.007	0.003	0.000	0.000	0.000	0.009	0.193	0.025	0.000	0.002	0.000	0.004	0.000	0.001
Mn	0.000	0.005	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.001	0.000	0.000	0.000
Fe	0.073	0.075	0.091	0.052	0.086	0.069	0.094	0.080	0.073	0.064	0.066	0.026	0.032	0.044	0.013	0.020	0.026	0.055
Ni	0.046	0.024	0.037	0.004	0.054	0.018	0.023	0.034	0.045	0.018	0.068	0.049	0.068	0.036	0.042	0.041	0.027	0.015
Cu	88.838	88.233	90.837	89.361	91.859	89.245	92.735	92.662	88.838	89.125	73.077	74.017	77.248	80.873	77.134	78.305	83.293	84.697
Zn	0.042	0.029	0.029	0.067	0.082	0.071	0.076	0.069	0.042	0.036	0.000	0.000	0.000	0.016	0.024	0.053	0.045	0.044
As	0.245	0.139	0.182	0.183	0.215	0.177	0.238	0.211	0.245	0.224	0.462	0.330	0.283	0.279	0.539	1.512	0.402	0.268
Sn	9.780	11.030	7.662	8.879	6.814	9.432	6.098	6.202	9.780	9.429	26.454	24.471	21.222	17.300	20.263	16.424	13.853	12.841
Pb	0.434	0.508	0.573	0.654	0.521	0.590	0.675	0.668	0.434	0.517		0.604	0.657	0.730	1.481	3.631	0.782	0.747
Total	99.458	100.054	99.418	99.206	99.638	99.605	99.940	99.933	99.457	99.422	100.320	99.522	99.510	99.281	99.497	99.990	98.428	98.668

Table 2. Pb Isotopic Analyses of the M. Herennius Military Diploma

Sample Number	²⁰⁶ Pb/ ²⁰⁴ Pb	Error (2σ)	²⁰⁷ Pb/ ²⁰⁴ Pb	Error (2σ)	²⁰⁸ Pb/ ²⁰⁴ Pb	Error (2σ)	²⁰⁷ Pb/ ²⁰⁶ Pb	Error (2σ)	²⁰⁸ Pb/ ²⁰⁶ Pb	Error (2σ)
A.1	18.425	±2	15.654	±3	38.574	±8	0.8496	±1	2.0936	±2
A.2	18.423	±2	15.651	±3	38.564	±8	0.8495	±1	2.0933	±2
A.3	18.423	±2	15.651	±3	38.565	±8	0.8495	±1	2.0933	±2
B.1	18.423	±2	15.652	±3	38.568	±8	0.8496	±1	2.0935	±2
B.2	18.424	±2	15.653	±3	38.570	±8	0.8496	±1	2.0935	±2
B.3	18.423	±2	15.653	±3	38.571	±8	0.8496	±1	2.0935	±2

Sample Number	²⁰⁶ Pb/ ²⁰⁴ Pb	Within Run Error (2σ)	²⁰⁷ Pb/ ²⁰⁴ Pb	Within Run Error (2σ)	²⁰⁸ Pb/ ²⁰⁴ Pb	Within Run Error (2σ)	²⁰⁷ Pb/ ²⁰⁶ Pb	Within Run Error (2σ)	²⁰⁸ Pb/ ²⁰⁶ Pb	Within Run Error (2σ)
A.1	18.4252	±5	15.6540	±5	38.5745	±15	0.849595	±6	2.093583	±38
A.2	18.4226	±5	15.6508	±5	38.5644	±15	0.849548	±6	2.093334	±32
A.3	18.4230	±6	15.6512	±5	38.5650	±16	0.849548	±6	2.093337	±32
B.1	18.4228	±5	15.6518	±5	38.5680	±16	0.849588	±7	2.093506	±33
B.2	18.4243	±5	15.6530	±4	38.5702	±15	0.849589	±7	2.093490	±29
B.3	18.4234	±6	15.6533	±6	38.5708	±23	0.849621	±9	2.093527	±58

Average A	18.424	15.652	38.568	0.84956	2.0934
Abs err. (±SD)	0.003	0.003	0.011	0.00005	0.0003
Per Mil (±SD)	0.16	0.22	0.29	0.06	0.14
Average B	18.423	15.653	38.570	0.84960	2.09351
Abs err. (±SD)	0.002	0.002	0.003	0.00004	0.00004
Per Mil (±SD)	0.08	0.10	0.08	0.04	0.02
Overall Avg.	18.424	15.652	38.569	0.8496	2.0935
Abs err. (±SD)	0.002	0.003	0.008	0.0001	0.0002
Per Mil (±SD)	0.11	0.16	0.20	0.07	0.10