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From commodity to money: the rise of silver coinage around the ancient Mediterranean (6th-1st century BCE)

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Abstract

The reasons why the Western Mediterranean, especially Carthage and Rome, resisted monetization relative to the Eastern Mediterranean are still unclear. Here, we address this question by combining lead and silver isotope abundances in silver coinage from the Aegean, Magna Graecia, Carthage, and the Roman Republic. The clear relationships observed between $^{109}\text{Ag}/^{107}\text{Ag}$ and $^{208}\text{Pb}/^{206}\text{Pb}$ reflect mixing of silver ores or silver objects with lead metal used for cupellation. The combined analysis of Ag and Pb isotopes reveal important information about the technology of smelting. The Greek world extracted Ag and Pb from associated ores, whereas, on the Iberian Peninsula, Carthaginians and Republican era Romans applied Phoenician cupellation techniques and added exotic Pb to lead-poor Ag ores. Massive silver re-cupellation is observed in Rome during the Second Punic War. After defeating the Carthaginians and the Macedonians in the late 2nd century BCE, the Romans brought together the efficient, millennium-old techniques of silver extraction of the Phoenicians, who considered this metal a simple commodity, with the monetization of economy introduced by the Greeks.

Silver has been highly prized in the Mediterranean and Near East region for millennia. The most obvious traces of its extraction can be seen in the contamination of ice and peat bogs all the way to the Arctic (Cortizas et al., 2002; Hong et al., 1994). The rise of monetized silver in the late 6th and the 5th century BCE in the Eastern Mediterranean world can be interpreted in different ways. Some consider that minting facilitated trade (Davis, 2012; Kim and Kroll, 2008), and, indeed, the abundance of shipwrecks in the Mediterranean as a proxy for economic performance correlates with lead peaks observed in Arctic ice (Parker, 1992). In the long run, however, the spread of coinage is also undoubtedly linked to the explosion of Greek mercenary service in the late 6th – 5th centuries, but to an extent that varies with the period and the local political circumstances (De Callataÿ, 2019; Krasilnikoff, 1992; Trundle, 2004). The present work uses Ag and Pb isotopes to further inform the debate by investigating the transition between two modes of silver utilization

around the Mediterranean: an early mode in which silver was considered as a simple commodity and traded by weight, and a later monetary mode.

5 Silver isotopes hold the potential of adding a new dimension to the understanding of early economies first and foremost because silver is a metal of monetary interest and not only another isotopic tracer. As a fingerprinting tool for historical and archeological applications, Ag isotopes are free from the uncertainties often afflicting Pb isotopes when it comes to untangling silver extraction and recycling (Brill and Shields, 1972). Silver and Pb isotope variations reflect processes that are unrelated in nature. Silver isotope variations are due to the minute differences in the thermodynamic properties of the two stable isotopes ^{107}Ag and ^{109}Ag , known as the 'isotope effect' (Fujii and Albarede, 2018). The range is very small (on the order of per mil) and essentially nil at the temperatures of magmatic and other high-temperature (such as cupellation) processes. The latter is true for Pb isotopes as well. In contrast, owing to the radiogenic nature of ^{206}Pb , ^{207}Pb , and ^{208}Pb , Pb isotope abundances vary with geological time and tectonic processes. 10 Because Pb isotope ratios change by several percent upon decay of U and Th, they vary over a much larger range than the per mil variations created by the isotope effect, which affects Pb isotopes the same way as Ag isotopes, but is drowned in the radiogenic ingrowth not affecting the stable Ag isotopes. 15

20 The present work assesses the contribution of combining Ag and Pb isotopic data as a marker of silver extraction and metallurgy in the mining districts which contributed to the monetary mass of the ancient Mediterranean world through time. Much is already known about silver mining techniques and metallurgy (Domergue, 2008). Cupellation, the technique of silver purification by smelting in the presence of metallic lead, was known from the early Bronze Age in the Middle East (Pernicka, 2004; Pernicka et al., 1998), Attica (Kakavogianni et al., 2008), and the island of Thasos (Bassiakos et al., 2019). On the Iberian Peninsula, however, evidence for cupellation, often in the form of litharge (lead oxide) is lacking before the Phoenician colonization in the early 1st millennium BCE (Bartelheim et al., 2012; Pérez Macías, 2016; Rovira Lloréns, 2017), which opened up vigorous exchanges between Iberia and the Near East (Neville, 2007). From the Second Punic War through to the 2nd century AD, Romans took over the silver mining districts of southern Iberia first exploited by the Celtiberians and the Phoenicians from the 8th century BCE (Avery, 1974; Jurado, 2002; Mata, 2002), and briefly later on during the mid-3rd century by the Carthaginians. In the Greek states, the monetary economy was based largely on exploitation of ores in the Aegean and Thraco–Macedonia from the second half of the 6th century through to the Roman conquest in 148 BCE. 25 30 35

40 A key question for determining the origin of bullion is the nature of ores used as the primary source (galena, sulfides, sulfosalts, chlorides, or their alteration products). If Ag and Pb are extracted from the same ore, such as galena, anglesite, or cerussite, their isotopes in the purified bullion will reflect the common metal source. If, however, cupellation involves exotic Pb, isotope systematics will instead reflect mixing between the two metal sources. Combined Ag and Pb isotopes can also address whether a particular metal used for coinage was extracted from a primary ore or rather was the product of the re-melting of mixed batches of silver fragments, such as silver plates, jewelry, or silverware. 45

Silver isotope variation in coinage (Fujii and Albarede, 2018) is much narrower than in ores (Arribas et al., 2020; Mathur et al., 2018), which potentially allows for a high selectivity of provenance. Correlating minting ages and Pb isotopes have been able to uphold the proposition of a major shift of silver ores shortly after the Second Punic War (Albarède et al., 2020; Albarède et al., 2016). Here we add to this suggestion by describing correlations between Ag and Pb isotopes on 92 literature and newly acquired data on silver coinage from the Aegean world, Magna Graecia, Carthage, and Rome (Figure 1). With this new expanded study, we reverse Desaulty et al.'s (2011) conclusion that Ag and Pb isotopes are uncorrelated and discuss the bearings of the new data set and observed Ag-Pb correlations on our understanding of silver sources and extraction techniques in the pre-Roman Empire Mediterranean world.

The present work combines literature data with 14 new Pb isotope data and 42 new Ag isotope data (Supplementary Table 1). The Pb isotope compositions are reported in Figure 2 with symbols for proper identification and dates for minting age (the median of the uncertainty range). The fields of volcanic rocks ((Albarède et al., 2020); <http://georoc.mpch-mainz.gwdg.de/georoc/>) genetically related to magmatic hydrothermal ore deposits (light blue for Southern Iberia, pink for the Aegean) from the literature are also plotted to show how closely the main group comprising most samples from Thrace, Greece, and the Greek colonies (Magna Graecia) track potential lead sources. Also plotted is the field of Pb isotopes in the Near Eastern silver hoards of *hacksilver* (chopped silver plates and jewelry) from Dor, Akko, and Ein Hofez (10th-9th century BCE), which are contemporaneous with the first Phoenician settlements around the Mediterranean, especially in Southern Iberia (Eshel et al., 2019; Neville, 2007). Particularly striking is a major difference in the apparent source of the metal used to mint the *denarii* from the Roman Republic (Albarède et al., 2020; Westner et al., 2020). The majority of late *denarii* (in black) plot to the right of the coins struck before the Second Punic War (240-200 BCE) to slightly after and conspicuously overlap the fields of Phoenician hoards (Eshel et al., 2019) and the field of the Phoenician and Ibero-Punic slags and silver objects found in Southern Iberia (Anguilano et al., 2010; Murillo-Barroso et al., 2016). Although more data clearly are needed, Pb isotopes from three Carthaginian and two Gaulish coins also plot to the right but at higher ²⁰⁸Pb/²⁰⁶Pb values.

Silver isotope compositions are reported as $\epsilon_{109\text{Ag}}$ values (the deviation in parts per 10,000 of the ¹⁰⁹Ag/¹⁰⁷Ag ratio of any given sample from that of the Ag standard SRM NIST 978a) (Desaulty et al., 2011). All the data fall into four well-defined groups. We found that the clearest representation is $\epsilon_{109\text{Ag}}$ vs ²⁰⁸Pb/²⁰⁶Pb (Figure 3). Model ages could have been used as well but, since ²⁰⁸Pb/²⁰⁶Pb ratios strongly correlate with model ages (Figure 4), we opted for the former which is more familiar to the community of Archaeometry.

In the $\epsilon_{109\text{Ag}}$ vs ²⁰⁸Pb/²⁰⁶Pb plot, the data form a bundle of arrays radiating from $\epsilon_{109\text{Ag}} \sim 0$ and ²⁰⁸Pb/²⁰⁶Pb ~ 2.065 :

- (1) Group 1 slightly trends to the right of $\epsilon_{109\text{Ag}} \sim -0.25$
- (2) The well-populated Group 2 trends more steeply towards a value of $\epsilon_{109\text{Ag}} \sim -1.0$;
- (3) Group 3 trends towards positive values.

These groups are well delineated at high ²⁰⁸Pb/²⁰⁶Pb. A few samples cannot be attributed to one group or another, while some rogue points are arguably difficult to explain, such as the low $\epsilon_{109\text{Ag}}$ -Abydos drachma. With these restrictions in mind, most data points of Group 1 are Roman Republic

denarii minted before the Second Punic War to slightly after. Most data points of Group 2 are coinage from Carthage and Roman Republic *denarii* minted after the Second Punic War. The two Gaulish samples belong to this group. Five coins from Group 3 were minted in Southern Italy and Sicily (Thurium, Gela, Syracuse, and Selinus). Most coins from classic Greece and Magna Graecia plot at the apex of the bundle ($^{208}\text{Pb}/^{206}\text{Pb}\sim 2.065$) and can be considered as either pertaining to all groups or not pertaining to any group, in which latter case they can be considered to define a fourth group.

The coins from Groups 1-3 plot within a narrow range of $\pm 1 \epsilon_{109\text{Ag}}$. This range is conspicuously smaller (Fuji and Albarède, 2018) than the spread of Ag ore deposits (Arribas et al., 2020; Mathur et al., 2018). Group 2 shows a well-defined negative correlation between $\epsilon_{109\text{Ag}}$ and $^{208}\text{Pb}/^{206}\text{Pb}$. This group, with few exceptions, corresponds to the field to the right of the magmatic trend in Figure 2. Given that silver and lead isotope fractionation is due to processes physically independent of each other, this array attests to mixing between two end-members, e.g., an Ag ore and Pb metal used for cupellation. The idea that lead in silver coins comes from a mixture of Pb and Ag ores is not new (Anguilano et al., 2010; Brill and Shields, 1972; Eshel et al., 2019), but Pb isotopes by themselves do not suffice to substantiate mixing, first because ores often form from mixed sources, and second because lead is a metallurgical additive to the silver ore indispensable for Ag purification.

At this stage, it is essential to emphasize the non-linear property of mixing in ratio-ratio plots when the denominators, here ^{107}Ag and ^{206}Pb , are different (Appendix A). This can be simply cast by reasoning that adding pure lead to a mixed Ag-Pb ore leaves silver with the $\epsilon_{109\text{Ag}}$ signature of the ore. In contrast, mixing changes $^{208}\text{Pb}/^{206}\text{Pb}$ and the other isotopic ratios, but only if Pb from the ore and the cupellation metal have different origins. The curvature of mixing hyperbolae increases with the contrast of $^{107}\text{Ag}/^{206}\text{Pb}$, or accepting, for sake of simplification, a negligible inaccuracy, the Ag/Pb ratios between the two end-members. Reference hyperbolae going through two geologically and historically acceptable end-members have been calculated and drawn in Figure 4 using the simple and useful relationship derived in Appendix A. In the $\epsilon_{109\text{Ag}}$ vs $^{208}\text{Pb}/^{206}\text{Pb}$ plot, the slope of each array at the Ag-rich end-member (Figure 5) is a particularly useful parameter. When comparing two Groups, say 1 and 2, the Pb/Ag ratio of the silver-rich group relates to the corresponding slope through:

$$\frac{(\text{Pb/Ag})_2}{(\text{Pb/Ag})_1} = \frac{\text{slope}_1}{\text{slope}_2} \quad (1)$$

Note that the hyperbolae drawn in Figure 4 were not fitted through the actual data but were drawn for reference only. Silver ore smelted for the peri-Punic Group 1 is clearly much richer in Ag than that used for Group 2. Mixing within Group 2 (in blue) corresponds to a Pb/Ag ratio in the Ag ore only a factor of ~ 4 times lower than in the Pb metal used for cupellation. The ore was therefore fairly poor in silver. In contrast, the Ag ore of Group 1 (in green) has a Pb/Ag ratio 20 times smaller than in the Pb additive. Combining Ag and Pb isotope analysis of Roman coins confirms evidence from archeological excavations (Anguilano et al., 2010; Eshel et al., 2019; Murillo-Barroso et al., 2016) that the two metals came from distinct sources and had to be combined for efficient silver extraction. The range in Pb isotope compositions of Roman ingots from Iberia is rather narrow (Trincherini et al., 2009), while the $\epsilon_{109\text{Ag}}$ values of Iberian galenas, although more variable, are negative (Arribas et al., 2020).

Most of the data in Figure 4 cluster around $\epsilon_{109\text{Ag}} \sim -0.25$ and $^{208}\text{Pb}/^{206}\text{Pb}=2.06-2.08$. The $^{207}\text{Pb}/^{206}\text{Pb}$ range is ambiguous. But, as pointed out by Fujii and Albarède (2018), the narrow range of $\epsilon_{109\text{Ag}}$ (-1 to +2 per 10,000) in silver coinage, whether minted around the Mediterranean during the Antiquity, in medieval or pre-modern Europe, or in the Spanish Americas, contrasts markedly with the broad scatter of several per mil observed for potential silver ores (Arribas et al., 2020). It is remarkable that the mean value of $\epsilon_{109\text{Ag}}$ in silver coinage differs from the (rare) mantle values but is indistinguishable from the value of the organic-rich Cody Shale value (Theis et al., 2013) deposited some 85 Ma ago. This observation is consistent with the suggestion that the metals of Pb-Zn(-Ag) ores were eventually derived not from the mantle, but from oceanic black shales deposited in anoxic environments (Milot et al., submitted).

The range of $\epsilon_{109\text{Ag}}$ values among the silver-rich end-members is very narrow, within ± 0.2 epsilon units of the SRM NIST 978a value. Such striking lack of fractionation of Ag isotopes suggests that the source ores formed primarily in a high-temperature environment, usually qualified as mesothermal (200-400°C) (Guilbert and Park Jr, 2007). It shows that the silver ores, presumably sulfides, did not go through intense weathering or any other low-temperature process that could have readily fractionated Ag isotopes (Evans, 2009). The ores used as lead sources could be galena and cerussite, as is the case for the Athenian Lavrion district (Conophagos, 1980) and Southern Iberia (Baron et al., 2017). The contrast between the Aegean and non-Aegean coins implies that the source of silver of the former went through cupellation using geologically young (Cenozoic), and probably cogenetic, lead and silver. Extraction of the two metals in the Aegean must have taken place at a short distance from each other.

The new perspective provided by Ag isotopes and derived from the curvature of Ag-Pb mixing hyperbolae allows pursuing the suggestion based on archeometallurgical observations (Rowan, 2013), statistics on triumph and temple treasure composition (de Callatay, 2006), and Pb isotopes on silver coinage (Birch et al., 2020b) that the Western Mediterranean used Greek coinage as a source of bullion. Silver-lead isotopic correlations show that silver from Group 1, most likely delivered by Carthage after the First Punic War, was reprocessed by the Romans. In contrast, the two Gaulish coins belong to Group 2 and, therefore, were not re-cupellated from pre-existing bullion.

Silver ores smelted for Group 3 (Gela, Thurium, Selinus, Syracuse, and cities from Magna Graecia with a long history of war and shared interests) were cupellated with unusual Pb ores containing Ag with positive $\epsilon_{109\text{Ag}}$ values, but unfortunately of so-far unknown origin.

The corollary of these observations is:

- a) Silver from most Aegean and Magna Graecia samples was smelted with lead from local ores. Having both Ag and Pb sources at disposal locally, possibly from the same mining district, perhaps the Lavrion, seems to have been key to secure silver production.
- b) In contrast, silver mined by Carthage in the 4th century BCE and by the Romans in the second half of the 2nd century BCE was cupellated with geologically older lead extracted at some distance from the silver ores.
- c) Metal used for Roman coins at the time of the Second Punic War was obtained from recycled silver (Albarède et al., 2016) and purified with Pb unrelated to any identifiable silver mining districts.

Silver used for minting Roman coinage during the Second Punic War (218-201 BCE, Group 1) was minted only during a short time period consistent with a discontinuity in the Ag isotopic composition noted by Albarède et al. (2016) at the time of the Roman monetary reform of 213 BCE (this date is used here for convenience; the exact date is still a matter of debate though not too far off 213 BCE). This change in silver source most likely corresponds to domestic re-melting of massive amounts of silver metal from the war penalties paid by Carthage after the First Punic War and the Mercenary wars. Given the apparent success of the silver recall in Rome at the climax of the 2nd Punic War (Livy, XXIII,48.9), Carthaginian silver must have been partly in private hands, mostly as silverware and jewelry, and partly in the form of debased *quadrigati*. Cupellation using scrap Pb of nondescript origin, possibly from Sardinia, which Rome annexed in 238 BCE, must have followed in the wake of the unsuccessful debasement attempted during the monetary reform of 213 BCE.

The narrow range of Pb/Ag ratios displayed by each group requires explanation. The answer lies in the metallurgical process. Silver lost to the litharge by oxidation steeply increases with metal fineness (Swinbourne et al., 2003; Ueda et al., 2005). In other words, making very pure silver comes at the price of wasting more of it. The metallurgist therefore has to play with two parameters, the oxygen flow and the Pb/Ag ratio of the silver ore-lead metal mixture to find the optimum ore-lead mixture that will provide the purest silver without losing too much of it to the slag (see Appendix B). Two-step cupellation as proposed for Lavrion (Pernicka and Bachmann, 1983) would manifest itself by sharply curved mixing hyperbolae unless Ag and Pb ores are very similar in both components. Archeological finds from many Phoenician settlements attest that the art of cupellation applied to low-grade ores, typically jarosite earth from Rio Tinto in the lower Guadalquivir valley near the modern town of Huelva, using metallic Pb had been mastered to the highest degree well before the first Lydian (late 7th century BCE) and Aegean (late 6th century BCE) coinages were struck (Blanco and Luzón, 1969). The narrow range of Pb concentrations in coins from the Roman Republic (Hollstein, 2000; Westner et al., 2020), which is always less than solubility in silver (1-3 wt.% between 300 and 900°C, (Karakaya and Thompson, 1987)), and the low vapor pressure of Pb, leaves no doubt that Pb was not added to the metal after purification.

Once the Carthaginians were defeated in Zama in 202 BCE, it took the Romans several decades to control the land of the Celtiberian tribes (Appian, 6; (Richardson and Richardson, 2004)) and organize mining and silver extraction. The present data provide evidence that the Romans only recommenced large-scale exploitation of jarositic Ag ores of the Iberian Pyrite Belt and Rio Tinto (Domergue, 1990; Westner et al., 2020) in the late 2nd century BCE. They clearly did not adopt the Greek method of local mining of high-grade ores, but used the Phoenician protocol, also mastered by the Celtiberian tribes, of combining low-grade Ag ores with galena-sourced Pb from the entire Iberian Peninsula.

The Eastern and Western Mediterranean civilizations looked at the same metal but from different perspectives. Phoenicia (Elayi et al., 2009; Gitler and Tal, 2006) and Carthage (Jenkins, 1974), although well aware of coined silver in the Aegean world, primarily considered this metal as a precious commodity, just as ivory, myrrh, and amber. Only with the arrival of Greek mercenaries in the second half of the 5th century BCE did these authorities adopt coinage. Not all the Greeks, however, struck silver as intensively as did Athens and a few other city-states later on and, later

on again, Hellenistic kingdoms. The Western Mediterranean, especially Carthage and Rome, also resisted monetization for longer than did the Greek world. From the inventories of Roman triumphs and temples in the 2nd century BCE, de Callatay (2006) concluded that at least 80 wt.% of available silver was left as bullion (*infecti*). Monetized silver does not seem to have become prevalent relative to jewelry and bullion, as attested to by persistent references to large sums in weight units (37). The overall coincidence between the isotopic differences and the historical gap between Groups 1 and 2 testifies to the reluctance of the Roman Republic to use coined money for larger transactions. Indemnities inflicted on the defeated Carthaginians reported by Polybius in his *Histories* and on Macedonians reported by Livy (XXXIII,30.7), always labelled in talents and not in any sort of monetary denomination, attest to the Romans being uneasy about pervasive monetization until reliable sources of bullion were safely established. Numismatic evidence supporting the rapid expansion of silver coinage in the second half of the 2nd century BCE and its potential causes, such as pressure on the copper supply for *as* minting (Bransbourg, 2017), are reviewed in detail by Kay (2014).

Materials and Methods

Silver isotopes were analyzed on series of coins from the Roman Republic (Westner et al., 2020) and Magna Graecia (Birch et al., 2020a; Birch et al., 2020b). Drill cores previously analyzed for Pb isotopes were re-used for Ag isotopes. Drill cores of Punic War denarii from Spain (provided by Pierluigi Debernardi) and of ten drachmas from different periods of Thrace, Athens, Miletus, Rhodes, Cilicia, and Gela (Birch et al., 2020a) were analyzed for both Pb and Ag isotopes. Literature data (Albarède et al., 2016; Desaulty et al., 2011) complemented the data set. Analytical techniques have been described elsewhere (Desaulty et al., 2011).

Appendix A: The mixing equation for ratios

Mass balance of isotope ratios x and y upon mixing of end-members α and β requests (Albarède, 1995):

$$\begin{aligned} x &= \varphi_{\text{Pb}}^{\alpha} x^{\alpha} + \varphi_{\text{Pb}}^{\beta} x^{\beta} \\ y &= \varphi_{\text{Ag}}^{\alpha} y^{\alpha} + \varphi_{\text{Ag}}^{\beta} y^{\beta} \end{aligned} \quad \text{with} \quad \begin{aligned} \varphi_{\text{Pb}}^{\alpha} + \varphi_{\text{Pb}}^{\beta} &= 1 \\ \varphi_{\text{Ag}}^{\alpha} + \varphi_{\text{Ag}}^{\beta} &= 1 \end{aligned} \quad (\text{A1})$$

where

$$\varphi_{\text{Pb}}^{\beta} = \frac{f^{\beta} C_{\text{Pb}}^{\beta}}{f^{\alpha} C_{\text{Pb}}^{\alpha} + f^{\beta} C_{\text{Pb}}^{\beta}} \quad \text{with} \quad f^{\alpha} + f^{\beta} = 1 \quad (\text{A2})$$

is the mass fraction of Pb contributed by end-member β . f^{α} and f^{β} stand for the weight fraction of end-members α and β , respectively, and C for the weight concentration of the species at the denominator (^{206}Pb for x and ^{107}Ag for y). Similar definitions apply to the other variables.

Replacing $\varphi_{\text{Pb}}^{\alpha}$ by $1 - \varphi_{\text{Pb}}^{\beta}$, we obtain

$$x - x^{\alpha} = \varphi_{\text{Pb}}^{\beta} (x^{\beta} - x^{\alpha}) \quad (\text{A3})$$

or

$$\frac{x^\beta - x^\alpha}{x - x^\alpha} = \frac{1}{\varphi_{\text{Pb}}^\beta} = 1 + \frac{f^\alpha C_{\text{Pb}}^\alpha}{f^\beta C_{\text{Pb}}^\beta} \quad (\text{A4})$$

The last equation can be rearranged, with a similar equation for y as

$$\frac{x^\beta - x}{x - x^\alpha} = \frac{f^\alpha C_{\text{Pb}}^\alpha}{f^\beta C_{\text{Pb}}^\beta} \quad \text{and} \quad \frac{y^\beta - y}{y - y^\alpha} = \frac{f^\alpha C_{\text{Ag}}^\alpha}{f^\beta C_{\text{Ag}}^\beta} \quad (\text{A5})$$

Dividing the equation for y by the equation for x gives

$$\frac{y^\beta - y}{y - y^\alpha} = \frac{(\text{Pb}/\text{Ag})^\beta}{(\text{Pb}/\text{Ag})^\alpha} \times \frac{x^\beta - x}{x - x^\alpha} = q \frac{x^\beta - x}{x - x^\alpha} \quad (\text{A6})$$

At the expense of a negligible error, $^{206}\text{Pb}/^{107}\text{Ag}$ for y ratios have, for sake of simplicity, been replaced by the elemental weight ratios Pb/Ag. Expressing the isotopic ratios, we finally get:

$$\frac{\varepsilon_{109\text{Ag}}^\beta - \varepsilon_{109\text{Ag}}}{\varepsilon_{109\text{Ag}} - \varepsilon_{109\text{Ag}}^\alpha} = \frac{(\text{Pb}/\text{Ag})^\beta}{(\text{Pb}/\text{Ag})^\alpha} \times \frac{\left(\frac{^{208}\text{Pb}}{^{206}\text{Pb}}\right)^\beta - \left(\frac{^{208}\text{Pb}}{^{206}\text{Pb}}\right)}{\left(\frac{^{208}\text{Pb}}{^{206}\text{Pb}}\right) - \left(\frac{^{208}\text{Pb}}{^{206}\text{Pb}}\right)^\alpha} \quad (\text{A7})$$

The last equation is represented by a hyperbola when the Pb/Ag ratios of the two end-members are different and by a straight-line when they are identical (Figure 5). An alternative equation can be derived:

$$(x - x_0)(y - y_0) = x_0 y_0 - \gamma \quad (\text{A8})$$

where

$$x_0 = \frac{x^\alpha - q x^\beta}{1 - q} \quad y_0 = \frac{y^\beta - q y^\alpha}{1 - q} \quad \gamma = \frac{x^\alpha y^\beta - q x^\beta y^\alpha}{1 - q} \quad (\text{A9})$$

The value of q is inferred from the slope of the mixing hyperbola $(dy/dx)_\alpha$ at $x = x^\alpha$. After tedious, but straightforward manipulations, the relationship

$$q = s / (dy/dx)_\alpha \quad (\text{A10})$$

is obtained, where $s = (y^\beta - y^\alpha) / (x^\beta - x^\alpha)$.

A useful expression relating q , φ_{Pb} , and φ_{Ag} can also be derived:

$$\varphi_{\text{Ag}}^\beta = \frac{\varphi_{\text{Pb}}^\beta}{\varphi_{\text{Pb}}^\beta + q(1 - \varphi_{\text{Pb}}^\beta)} \quad (\text{A11})$$

Appendix B. Optimum Ag/Pb for smelting

To a good approximation, the weight fraction f_{slag} of slag obtained by cupellating the ore-lead mixture is equal to the concentration

$$C_0^{\text{Pb}} = \frac{[\text{Pb}]}{[\text{Pb}] + [\text{Ag}]} \quad (\text{B1})$$

5 in the mixture. Here, brackets stand for concentrations. The fraction of total silver lost to the slag is approximately

$$f_{\text{slag}} K_{\text{slag/met}}^{\text{Ag}} \approx C_0^{\text{Pb}} K_{\text{slag/met}}^{\text{Ag}} \quad (\text{B2})$$

where $K_{\text{slag/met}}^{\text{Ag}}$ is the coefficient of Ag fractionation between the slag and purified Ag.

10 Reducing the amount of lead in the original ore-lead mixture both reduces C_0^{Pb} and increases $K_{\text{slag/met}}^{\text{Ag}}$ (Ueda et al., 2005), which requires the proportions to be optimized before extraction. This result justifies that the Pb/Ag ratio during cupellation is constrained within a relatively narrow range.

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Supplementary Materials:

Table ST1

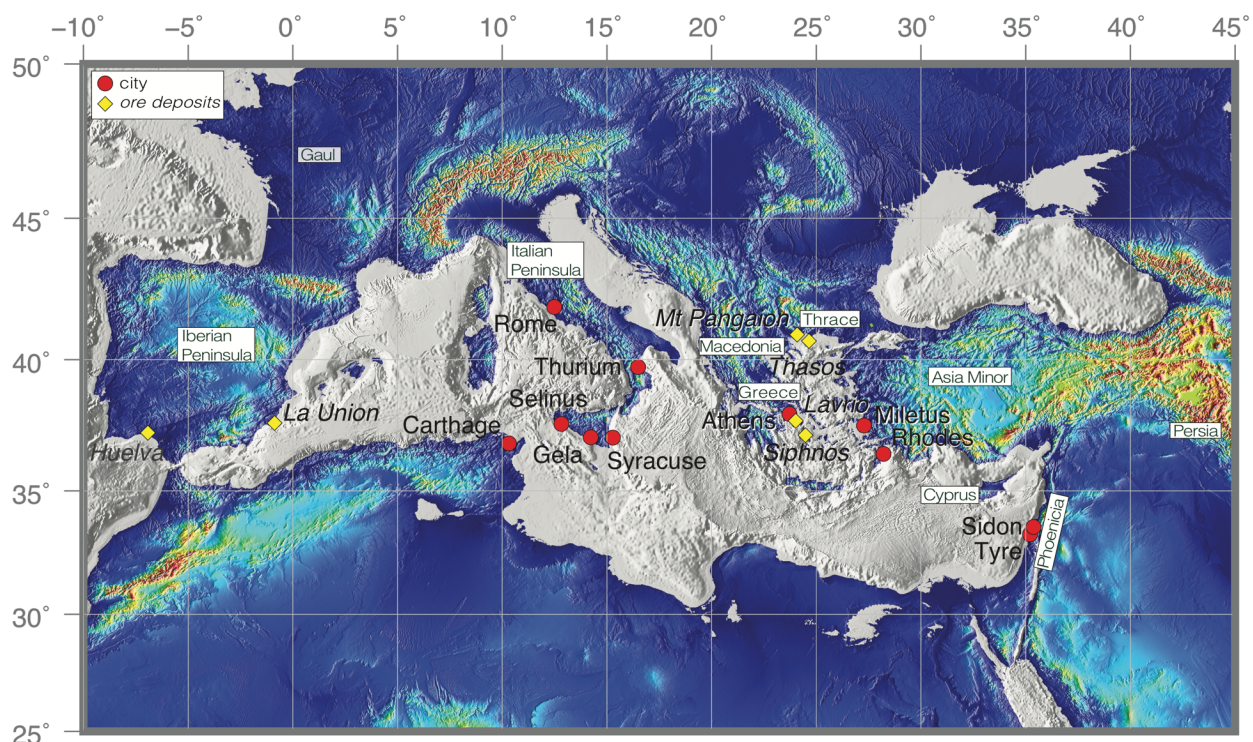


Fig. 1. Map of the Mediterranean with names of localities (cities and major ore districts) referred to in the text. In the interest of clarity, the toponymy used is a mix of names from different periods (e.g., Greece, Asia Minor).

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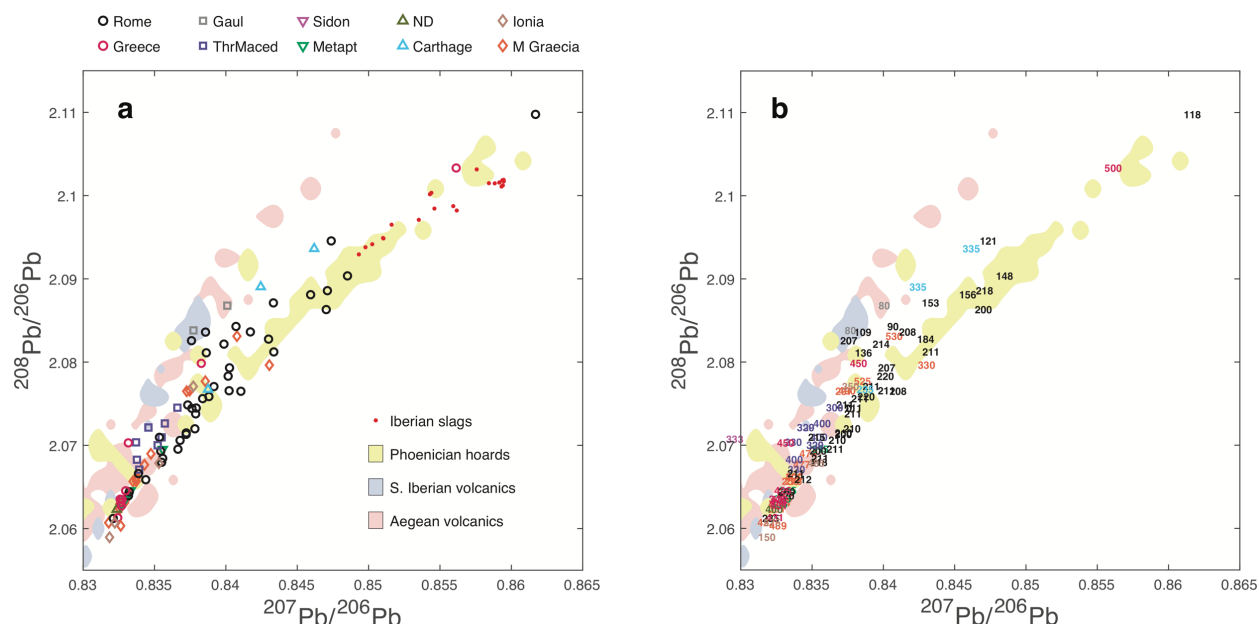


Fig. 2. Lead isotope relationships for the samples analyzed in this study and literature data. Panels (a) and (b) show the same samples with symbols (panel a) and minting ages (BCE) (panel b), respectively. In spite of a substantial overlap, panel b was meant to reduce the number of different symbols and improve legibility, while helping with the age of minting. The fields representing recent Aegean and South Iberian volcanic rocks (downloaded from the GEOROC database <http://georoc.mpch-mainz.gwdg.de/georoc/>) are clearly relevant to Aegean and Magna Graecia coinage (primary ores). The points to the right attest to addition of exotic Pb to Ag ores during cupellation. The overlap between these points, the Phoenician hoards from the 10th–9th century BCE, and the Ibero-Punic slags and silver objects found in southern Iberia (Anguilano et al., 2010; Murillo-Barroso et al., 2016), show that first Carthage, then Rome in the late 2nd century BCE, had adopted the Phoenician smelting process.

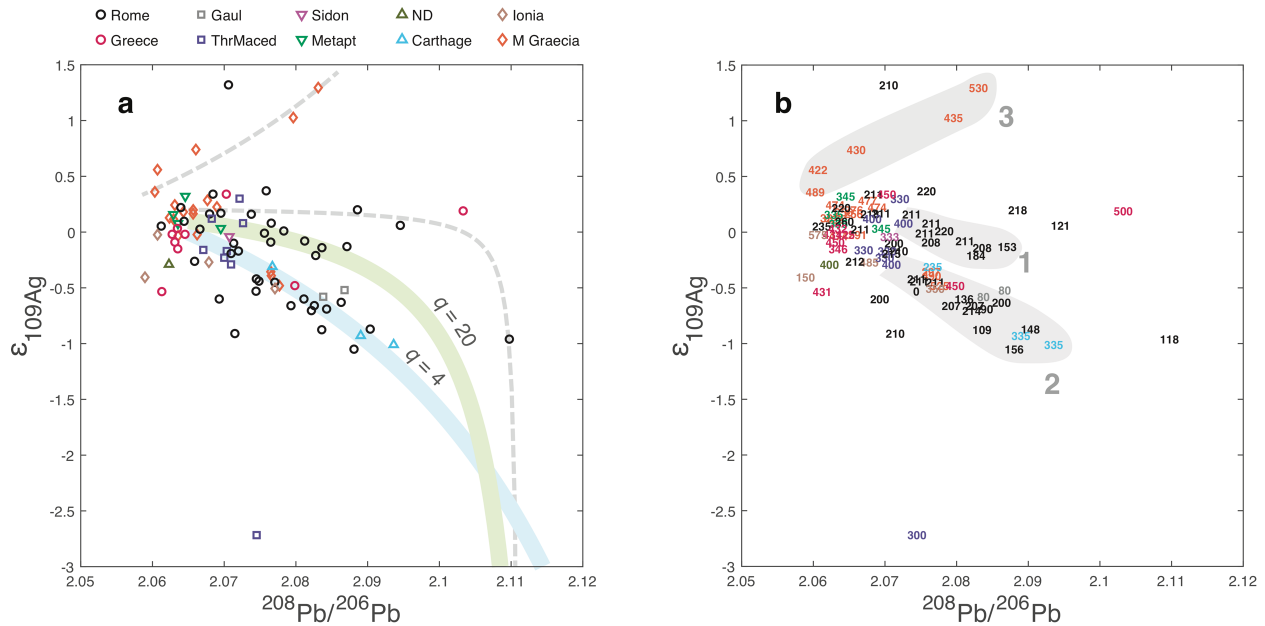
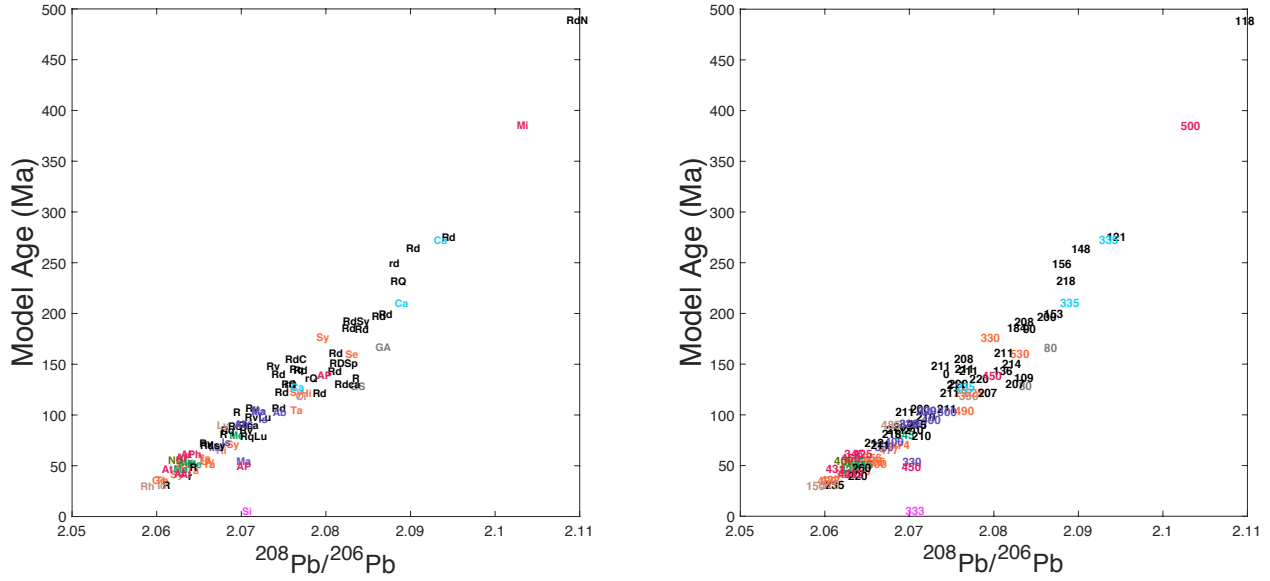


Fig. 3. Correlation between $^{109}\text{Ag}/^{107}\text{Ag}$, expressed using the epsilon notation (deviation from a reference ratio in parts per 10,000), and $^{208}\text{Pb}/^{206}\text{Pb}$ for 92 silver coins from the Aegean world, Magna Graecia, Carthage, and Rome. In spite of a substantial overlap, panel b was meant to reduce the number of different symbols and improve legibility, while helping with the age of minting. Groups are referred to with light grey roman numerals. The same data are represented with symbols (panel a) and minting ages BCE (panel b). (a) Mixing lines do not display as linear arrays but as hyperbolae (see Supplementary Information). The curves are shown as references and were not obtained by a fit to the data. The curvature is a function of the ratio $(\text{Pb}/\text{Ag})_{\text{ore}}$ in the ore (or recycled bullion) and $(\text{Pb}/\text{Ag})_{\text{Pb}}$ in the lead used for smelting and q is the ratio $(\text{Pb}/\text{Ag})_{\text{ore}} / (\text{Pb}/\text{Ag})_{\text{Pb}}$. For Group 1 (green mixing hyperbola), $(\text{Pb}/\text{Ag})_{\text{ore}} = 20 (\text{Pb}/\text{Ag})_{\text{Pb}}$, which points to a Ag-rich silver source. In contrast, for Group 2 (blue mixing hyperbola), $(\text{Pb}/\text{Ag})_{\text{ore}} = 4 (\text{Pb}/\text{Ag})_{\text{Pb}}$, which suggests a Ag-poor source. Group 1, consisting of coins minted around the Second Punic War (see panel b), clearly represents re-melting of silver bullion, jewelry, or debased coinage. Group 2, comprising coins minted by Carthage, some Magna Graecia colonies, and the late Roman Republic, represents coins minted from Iberian ores after cupellation with exotic Pb. In contrast, coins pertaining to the largest group of Greek and Magna Graecia ores and plotting near the apex of this bundle was cupellated using Pb cogenetic with Ag ores. Five Magna Graecia coins (Group 3) definitely have been cupellated with Pb containing Ag with positive $\epsilon_{109\text{Ag}}$ values. The dashed grey hyperbolae show more potential mixing arrays: shallow slopes, both positive and negative, indicate probable multiple events of cupellation.



5 **Fig. 4.** Correlation between model ages (in million years) as calculated in Albarède et al. (2012) and $^{208}\text{Pb}/^{206}\text{Pb}$ ratios. Left: abbreviations used as symbols can be found in the column under the header 'ID' (Supplemental Table 1). Right: minting ages BCE.

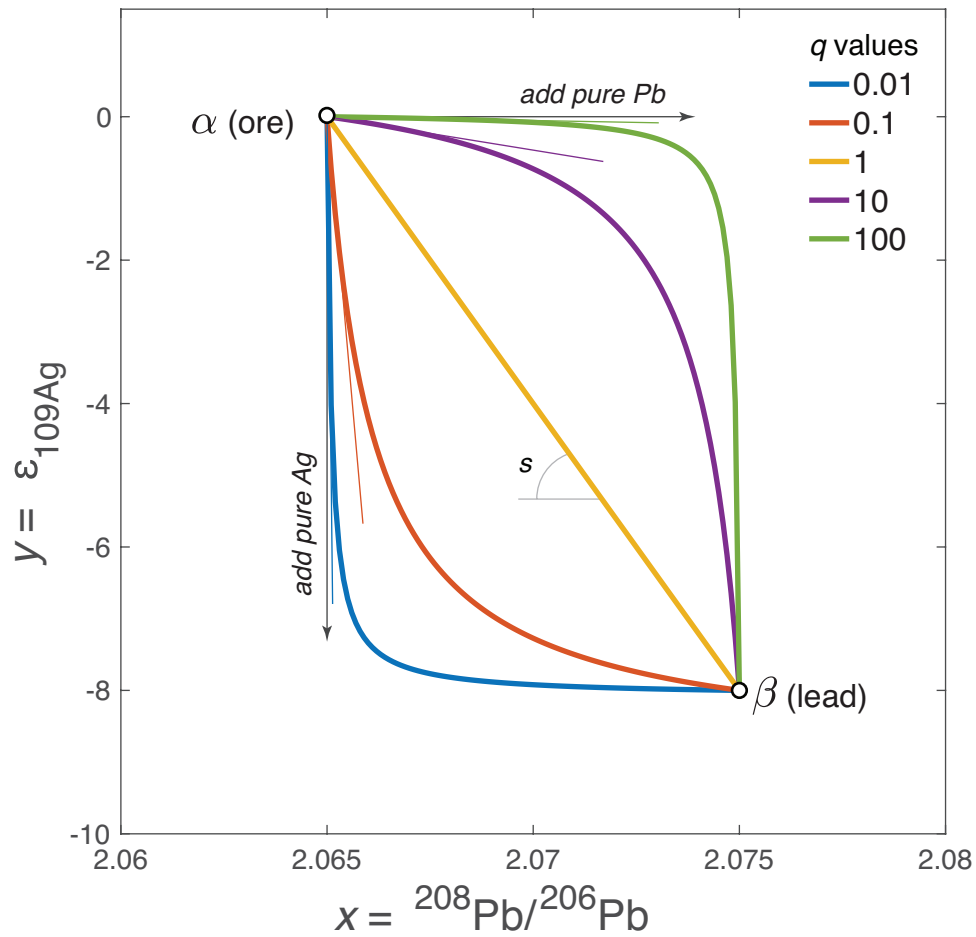


Fig. 5. Hyperbolae representing mixing between a silver-rich ore α and metallic lead β used for smelting for different values of $q = (\text{Pb}/\text{Ag})^\beta/(\text{Pb}/\text{Ag})^\alpha$. The thin colored lines represent tangents to the corresponding mixing hyperbolae at $x = x^\alpha$ and $y = y^\alpha$ (see text). $s = (y^\beta - y^\alpha)/(x^\beta - x^\alpha)$.

5