

## A NEW METHOD FOR COMBINING LEAD ISOTOPE AND LEAD ABUNDANCE DATA TO CHARACTERIZE ARCHAEOLOGICAL COPPER ALLOYS\*

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*We present a new methodology for interpreting lead isotope data from archaeological copper alloy objects. It is not based on the conventional isotope ratio biplots, which were originally devised to allow the calculation of the geological age of the lead mineralization, but is derived from isotope mixing models, more often used for presenting strontium isotope data. We illustrate the method by reworking published data on Sardinian Nuragic oxhide ingots and copper alloy artefacts. While we confirm the consensus assumption that the oxhide ingot fragments found on Sardinia are isotopically consistent with Cypriot copper ores (although we see no isotopic reason to favour only Apliki, as originally suggested), we also show that there is evidence for mixing between local and ingot copper in some objects, which was previously not detected. More broadly, we suspect that the apparent mismatch between some source allocations for copper drawn from isotope geochemistry and the rest of the archaeological data in some cases might be due to mixed isotopic signals being incorrectly assigned to a specific source, and suggest that the method presented here will reduce the chances of this happening.*

**KEYWORDS:** LEAD ISOTOPE DATA, COPPER, OXHIDE INGOTS, SARDINIA, CYPRUS, MIXING, RECYCLING

In a recent series of papers, we have been attempting to rethink the use of chemical data from archaeological copper alloys in order to focus on characterizing the changing nature of the metal in circulation, rather than on the more traditional question of provenance (i.e., not asking ‘From which mine does this metal originate?’ but ‘How is this metal in circulation changing, and what does this mean in a social context?’). This acknowledges what seems to us as self-evident—that metal in antiquity was capable of being recycled, and in some circumstances this may have been an important factor in the circulation, use and meaning of metal. It also recognizes a fundamental problem with most provenance studies of prehistoric archaeological material, which is that the time elapsed for an object to move from source to deposition spot is not taken into account (Pollard *et al.* 2014). We also focus on the assemblage level (the assemblages being defined chronologically, geographically or typologically, although preferably using all three), rather than on individual objects.

For trace element data in copper alloys, this has led us to use a two-stage process, beginning with a presence/absence system based on the four most commonly reported trace elements (As, Sb, Ag and Ni) as a preliminary classification of the copper ‘type’ (Bray 2009; Bray and Pollard

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2012; Pollard and Bray 2014a,b). For most data sets, we use a figure of 0.1% (after mathematical removal of any major alloying elements present and renormalization) as the division between presence and absence for these elements, but this can (and should) be tested by variation for any particular data set, and the exact value for each element may depend on the specifics of data consistency and quality in that data set. The second level of analysis aims to characterize the distribution and relationship between the elements present within the heuristic presence/absence copper groups. In short, arsenic and antimony are relatively vulnerable to oxidation and loss on recycling, while silver and nickel are resistant to removal. An assemblage will on average, therefore, lose arsenic through any process that requires melting, but silver levels are resistant to simple melting losses. A fall in the average silver content is more likely to have been caused by mixing with a unit of silver-poor material, diluting the overall level. By combining details of the artefact typology, chronology, context *and* chemical characterization, a broad life history of events may be inferred through this procedure. We have repeatedly emphasized that this is *not* a provenancing technique (i.e., we do not equate a particular copper type with a specific 'source', nor do we assume that a specific source will produce copper of a unique 'type'), but that it does allow us to characterize the metal in circulation, and to identify the timing and general origin of new inputs of metal into the system. This becomes increasingly powerful when combined with distributional and chronologically resolved mapping, for which we use GIS (Perucchetti *et al.* submitted).

An approach similar to this can also be used for the major alloying elements (here considered to be Sn, Pb and Zn). By plotting, for example, maps of the distribution of the tin concentration in specific groups of objects (identified chronologically and typologically), it is possible to chart the development and spread of the use of tin bronze, and to indicate from where the tin might be coming and when it might have been added. For example, Cuénod *et al.* (submitted) showed that while copper coming into Mesopotamia from the Iranian Plateau or further east was likely to have been mixed with tin at a late stage by the consumers of the metal, copper coming from the west was different and the tin in these objects is likely to have come in already alloyed objects.

In a further development, we have focused on the classification of copper alloy type, using the major alloying elements of Sn, Zn and Pb. Rather than using modern definitions of alloys (e.g., tin bronze contains copper with more than about 6–8% Sn), we have again resorted to a presence/absence definition, in which we arbitrarily set the threshold of presence at 1%. Thus we would classify an alloy containing 92% Cu, 2% Sn, 2% Zn and 4% Pb as a 'leaded gunmetal', despite the fact that this composition does not necessarily correspond to a modern designed alloy. The value of this approach is that it highlights the presence of such mixed alloys in assemblages, and suggests that, rather than deliberate alloy design, the composition of such alloys may be the result of mixing metals of more than one alloy type. Rather than completely denying the existence of 'designed alloys', however, it enables us to identify them more clearly when they do appear in the circulatory system. For example, in a study of first millennium AD copper alloys in Britain, we have used this methodology to show continuity of metal circulation from the Late Roman period into the Early Anglo-Saxon, with a marked change occurring in the Middle Saxon period, which we attribute to the arrival of fresh stocks of metal (Pollard *et al.* in press). Moreover, using the ubiquity of the quaternary alloy leaded gunmetal (defined as above) as a proxy for the amount of recycled metal in circulation, we have suggested that by the end of the Roman period, approximately 70% of the objects contained recycled metal.

If we accept that, at least at certain times and places, metal recycling was a significant component of human behaviour, then the uncritical use of lead isotope data on copper alloys

is potentially misleading in terms of provenance. However, when viewed in a different way and combined with chemical and other evidence, it might provide the key to disentangling this complex picture. As a third strand to this re-evaluation, therefore, we now wish to turn our attention to the use of lead isotope ratios in the characterization of copper alloys. Traditionally, these have been used as a direct indicator of the provenance of the copper from which the object is made, but it is a commonplace observation to say that if metal from different sources is being mixed and recycled, then the measured lead isotope signature of the object may not correspond to any one specific source. This is in addition to the obvious statement that if lead is added to a low-lead copper alloy, then the lead isotope signature will be dominated by that of the added lead, not the copper. The interpretation of lead isotopes in archaeological copper alloys has been contentious since the 1990s (see Pollard 2009 and references therein). One source of contention, which is unavoidable, is the well-known overlap in lead isotope ratios from geographically distinct sources, and the possible range of values that might come from a single source. Simply changing the method of presentation of the data cannot remove such problems, but they can be minimized by attempting to define the archaeological question more closely—by asking ‘Could these objects have come from source X?’, or ‘Are these objects from A the same as those from B?’, rather than the much more open question ‘Where do these objects come from?’ (Pollard 2009). They may also be addressed by combining chemical and isotopic data in a systematic way. Earlier attempts to do this, however, have either used trace elements (often Ag and Au) as a separate test of the consistency or otherwise of the inferences drawn from the lead isotopes, or have resorted to cluster analysis, which is completely inappropriate if mixing is taking place. Other approaches have used constructs such as ‘IMP-LI’s’ (IMPurity composition and Lead Isotope composition: Rohl and Needham 1998, 84). Each IMP-LI is said to represent ‘a stock of metal in circulation which might either be similar to the character of a specific source, or alternatively be a complex but relatively coherent amalgam of different metals’. Our approach is consistent with, but extends, this definition. We aim to combine the chemical and isotopic data in a more systematic way than was done by Rohl and Needham.

A related contentious issue, which we think we can definitely improve upon, is the question of the ‘shape of an orefield’ in the two- or three-dimensional space defined by two or three pairs of lead isotope ratio. The practice of drawing ‘90% confidence ellipses’ around data defining an orefield has drawn much criticism, since it either assumes a normal distribution of points in the space defined by the isotope ratios, which has been shown to be not true (Baxter and Gale 1998), or is an entirely arbitrary exercise, which often involves dropping data points as ‘outliers’ in order to achieve a regular ellipse. Kernel density estimates have been shown to be much more robust in defining the actual distribution of an orefield (Baxter *et al.* 1997; Scaife *et al.* 1999), but do not easily lend themselves to identifying mixing. We therefore need a new methodology that does not require such distributional assumptions to be made, can reveal the possibility of the mixing of metal from isotopically distinct sources, and can combine chemical and isotopic data in a systematic and logical way, in order to avoid erroneous conclusions being drawn.

The four stable isotopes of lead ( $^{204}\text{Pb}$ ,  $^{206}\text{Pb}$ ,  $^{207}\text{Pb}$  and  $^{208}\text{Pb}$ ) are conventionally measured as a set of three isotope ratios. Geologically, they are usually reported as  $^{206}\text{Pb}/^{204}\text{Pb}$ ,  $^{207}\text{Pb}/^{204}\text{Pb}$  and  $^{208}\text{Pb}/^{204}\text{Pb}$ , but archaeologically the first results were reported as  $^{206}\text{Pb}/^{204}\text{Pb}$ ,  $^{206}\text{Pb}/^{207}\text{Pb}$  and  $^{208}\text{Pb}/^{207}\text{Pb}$  (Brill and Wampler 1967). Subsequent archaeological practice has become to report  $^{204}\text{Pb}/^{206}\text{Pb}$ ,  $^{207}\text{Pb}/^{206}\text{Pb}$  and  $^{208}\text{Pb}/^{206}\text{Pb}$ , because  $^{204}\text{Pb}$  is the least abundant (~1.4%) and therefore ratios measured against  $^{206}\text{Pb}$  (~24.1% abundant) are capable of higher precision. It

is a simple calculation to convert between one set of ratios and another, although the purist will say that such calculations have an effect on the error of the individual measurements, which is generally ignored. The conventional means of interpreting lead isotope ratios in archaeology has been to plot a pair of bivariate scatter diagrams, thus conveying all the available information from three isotope ratios in two diagrams. Historically, this form of presentation is derived from geological practice, which plots a pair of isotope ratios measured in lead minerals (with  $^{204}\text{Pb}$  as denominator) to calculate the geological age of the lead deposit based on the Holmes–Houtermans equation for the evolution of the isotopic composition (see Pollard and Heron 2008, 311–21).

It is clear that when the method of lead isotopy was extended from lead minerals to the lead impurities left in copper objects smelted from impure copper ores (Gale and Stos-Gale 1982)—thereby opening up a whole new and highly significant field of archaeological research—insufficient thought was given to the question of whether the geological methods of presentation were the most appropriate way to display and interpret the data. Given that the reported isotope ratios were changed at the same time from  $^{204}\text{Pb}$  as denominator (as required by the Holmes–Houtermans equation), hindsight suggests that there is no reason to expect that the traditional isotope biplot is optimal. This is particularly so if mixing might occur, and when the aim of the study is not to determine the geological age of the deposit but to carry out a comparative study of its (mixed or pristine) geographical location (although geological age is, of course, one of the parameters contributing to the isotopic discrimination between deposits). We have therefore been experimenting with a different set of diagrams, which plot the Pb chemical concentration against each isotope ratio, giving a set of three diagrams. This is more akin to the practice used to interpret strontium isotope data (e.g., Faure 1986, 142–7). The advantage of this system is that mixtures of two components, having different chemical abundances and isotopic ratios, show up in these plots as hyperbolic mixing lines, which become linear if plotted as isotope ratio against inverse concentration. In theory, the equation of this linear mixing line can be used to calculate the relative contributions from two isotopically different sources (Faure 1986, 144). This method is widely used in sedimentary geology to calculate the approximate contributions of different inputs into a particular sediment, and appears to be a reasonable model upon which to base a reinterpretation of archaeological lead isotope data.

In order to show the advantages of using such an approach over the conventional method, we present a case study based on published data relating to the attribution to Cyprus of the source of the copper oxhide ingots found on Sardinia. This case study is selected on the basis of the availability of good quality analytical and isotopic data on the same objects for a reasonably large number of cases, rather than from a desire to reinvestigate this question. As will hopefully be seen, the method we propose here broadly supports the original conclusions, but also reveals some previously unsuspected detail. We hope to show, however, that in all archaeological cases there is a significant advantage to considering the data in this way.

### *The source of the copper oxhide ingots found on Sardinia*

The source of the copper used to make the distinctive oxhide ingots, widely traded around the eastern and central Mediterranean during the later Bronze Age, has been the focus of much archaeometallurgical research (Lo Schiavo *et al.* 2009). In particular, the discovery on Sardinia of a large number of copper oxhide ingot fragments, of a shape generally believed to originate on Cyprus, has been the source of much archaeological debate, recently summarized by Muhly (2009). The original isotopic work carried out by Gale and Stos-Gale (Stos-Gale *et al.* 1997;

Gale 1999, 2001; Stos-Gale 2000) on Mediterranean oxhide ingots in general concluded that all ingots post-1250 BC came from the Apliki mine in north-western Cyprus. Prior to 1250 BC, the ingots were also of Cypriot, but not specifically Apliki, origin. Moreover, the ingots found on Sardinia appear not to have been used for making Sardinian artefacts (Muhly 2009, 28). Sardinia itself is rich in copper resources, so why import such material, especially if it is never used to make objects? The question clearly hinges on whether or not the oxhide ingots did indeed originate in Cyprus, and also how these ingots relate to the local Nuragic copper alloy objects. Begemann *et al.* (2001) produced new data on Sardinian Nuragic metallurgy, including oxhide and bun (plano-convex) ingot fragments, and reached essentially the same conclusion as the Gales: the oxhide ingots were from Cyprus (although not attributed specifically to Apliki).

We have now re-analysed the data published by Begemann *et al.* (2001), which gives chemical (Cu, Sn, As, Sb, Co, Ni, Ag, Au, Fe, Zn, Se and Te by neutron activation analysis; Pb by mass spectrometry with isotope dilution) and isotopic ( $^{208}\text{Pb}/^{206}\text{Pb}$ ,  $^{207}\text{Pb}/^{206}\text{Pb}$  and  $^{204}\text{Pb}/^{206}\text{Pb}$ ) data on 49 Sardinian copper ingot fragments (29 oxhide, 10 bun, eight oxhide or bun, and two 'truncated cones': Begemann *et al.* 2001, tables 1 and 2) and 45 copper and bronze artefacts from Nuragic Sardinia (*ibid.*, tables 3 and 4). Also included are lead content and isotope ratio data on 31 Sardinian copper ore samples and four samples of copper slag, plus lead isotope data on 18 Sardinian lead ores and one tin ore (Begemann *et al.* 2001, table 5).

Figure 1 shows a plot of the Sardinian ingot data as  $1/\text{Pb}$  ( $\text{ppm}^{-1}$ ) against each measured isotope ratio. Each plot is characterized by a strong horizontal 'spike', focused on a relatively narrow range of isotope values. We interpret these 'spikes' as representing copper with a consistent isotopic value, and in this case probably coming from a single ore source, since these spikes most probably represent mixing lines within a single copper source having variable amounts of lead but very similar lead isotope ratios. An alternative explanation would be the addition of lead (or a high-lead copper) of a particular isotopic value to a stock of low-lead copper, but in this case we prefer the former interpretation because the lead content of these ingot fragments is less than 2000 ppm (0.2%) in all but one case. By eye, these 'spikes' have values of  $^{208}\text{Pb}/^{206}\text{Pb} \approx 2.075$ ,  $^{207}\text{Pb}/^{206}\text{Pb} \approx 0.84$  and  $^{204}\text{Pb}/^{206}\text{Pb} \approx 0.054$ . Figure 1 is to be compared with Figure 2, which shows the 'conventional' plot of these data (Begemann *et al.* 2001, fig. 8). We suggest that Figure 1 shows the internal consistency and the most representative isotopic values of the majority of these data more immediately than does Figure 2.

In order to illustrate how we have developed the 'IMP-LI' model discussed above, Figure 3 shows the same diagram as the upper part of Figure 1, but with the points classified by 'copper type' rather than morphology, using the trace element methodology described in Pollard *et al.* (2014) (see also Bray and Pollard 2012; Pollard and Bray 2014b). This clearly shows that the samples that constitute the 'spike' are almost all copper group 2 (Cu with  $>0.1\%$  As), whereas the samples with different isotope values are predominantly group 1 (Cu with  $<0.1\%$  As, Sb, Ag and Ni). This shows that the identity of the ingot fragments within the spike is characterized not only by isotope value but also by copper chemistry, and that the two methods can be combined to reinforce each other.

Figures 4 and 5 show the same plots for the data on Sardinian copper ores and copper slags and for the Nuragic copper and bronze objects found on Sardinia, respectively (for brevity, in each case only the plot of  $1/\text{Pb}$  versus  $^{208}\text{Pb}/^{206}\text{Pb}$  is shown, but the other figures are very similar). As noted by Begemann *et al.* (2001, caption to fig. 9), the ore values of the Sardinian copper sources are widely dispersed, and while there is no exact match for the isotope values of the bulk of the ingot fragments, some of the outlying ingot values do match with the ore data. The artefact data

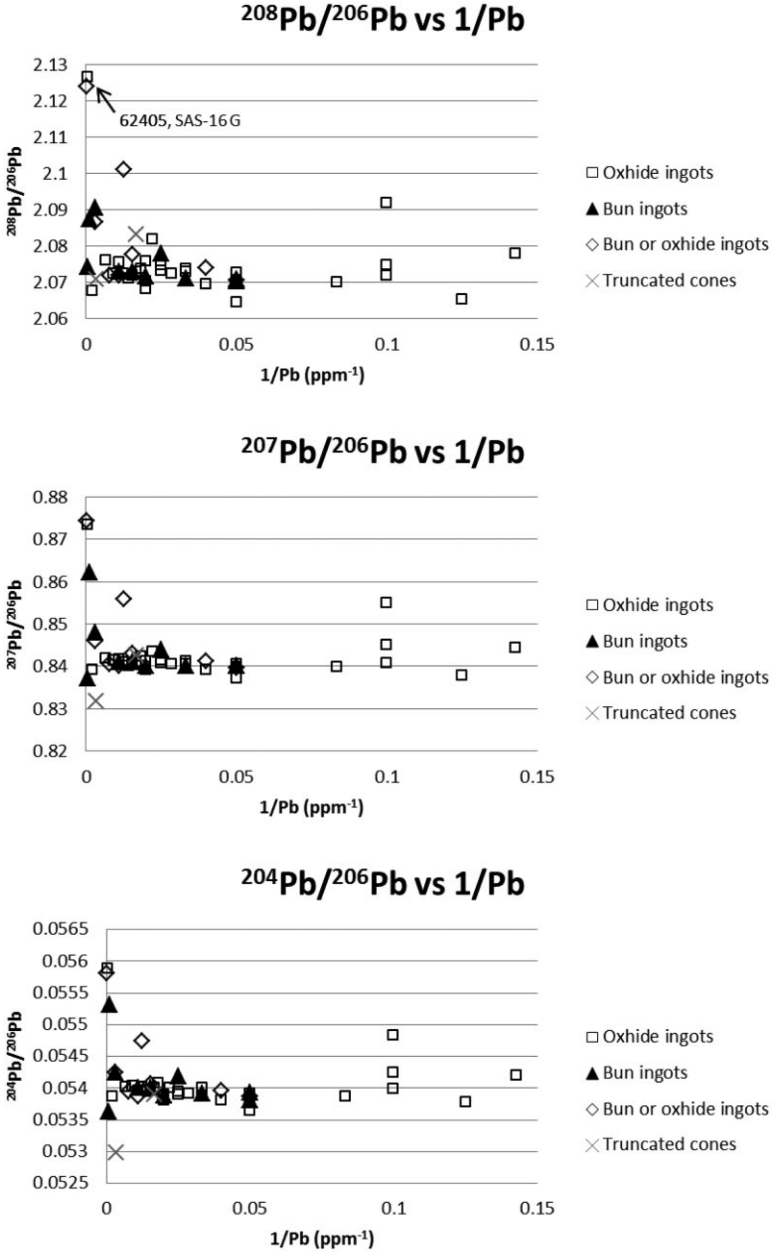


Figure 1 A plot of Sardinian ingot data (from Begemann et al. 2001, table 2), showing  $1/\text{Pb}$  against each isotope ratio.

(Fig. 5) show a wider range of values than either the ores or the ingots. The two extreme values (two samples of ‘nondescript metal’ from Arzachena: 20975G and 20975R) have much lower values for each measured ratio (indicative of a uranium-rich copper source?) than either the ingots or any of the ores. One advantage of these plots over the conventional presentation, however, is that we can quickly see that these objects have very low Pb levels (10 ppm, or

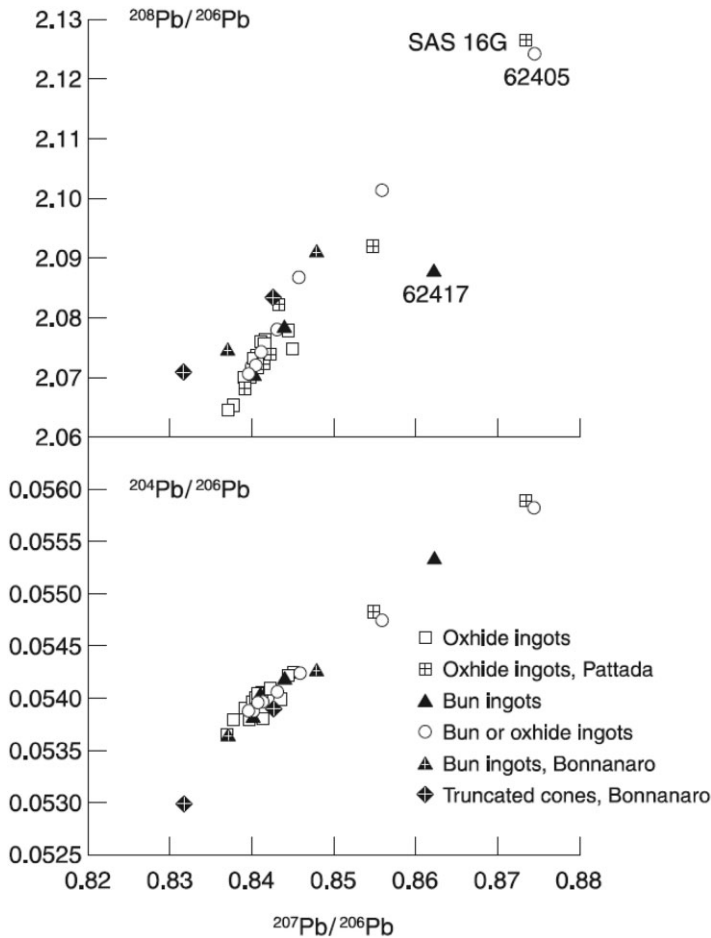


Figure 2 A conventional plot of Sardinian ingot data (Begemann et al. 2001, fig. 8).

0.1 ppm<sup>-1</sup> in these inverse concentration plots), and it may be the case that these measurements are less accurate than the others because of the difficulties associated with measuring isotopic ratios at very low lead levels.

Figure 6 shows a composite plot of the 1/Pb versus <sup>208</sup>Pb/<sup>206</sup>Pb diagrams for ingots, Cu ores and slags, and Sardinian artefacts, all to the same vertical scale (the two artefacts mentioned above, with very low ratios, are omitted). If we take the Cu slags to be indicative of the local copper ore used on the island (upper line), then it is clear that two ingot fragments (Ittiredu 62405, a bronze ‘oxhide or bun ingot’, and Pattada oxhide ingot SAS-16G) identified as outliers in Figure 1 (with <sup>208</sup>Pb/<sup>206</sup>Pb ≈ 2.126) are similar to the Pb isotope signal from local ores (or, since 62405 is the only bronze ingot sample, and the only one with any lead (4.55%), in this case it might contain local Sardinian lead). In Figure 7, the artefact and ingot data are plotted on a single diagram, with the artefacts approximately divided into ‘bronze’ [i.e., having tin added at levels between 3300 and 143 000 ppm (0.33–14.3%)] and ‘copper’ (Sn < 250 ppm). Three of the copper artefacts (20975B, 20975C and 20975D), all ‘nondescript metal’ from Arzachena, with Sn < 110, plus a fourth bronze sample (Arzachena 20974H, a sword fragment with 4700 ppm Sn), have an isotope

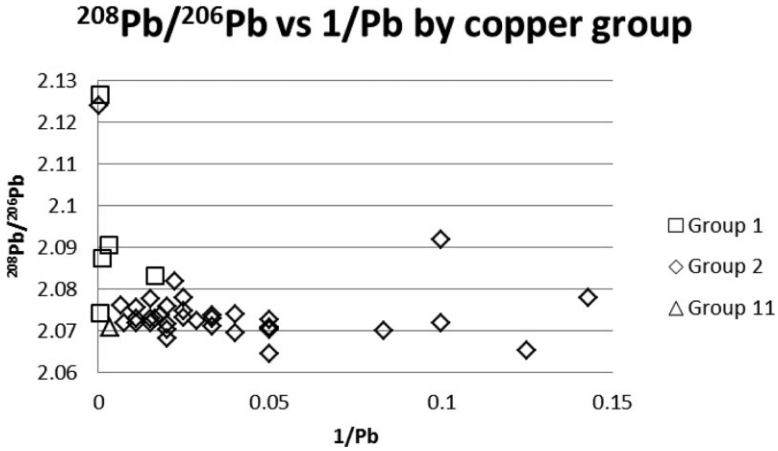


Figure 3 As Figure 1, but with ingot fragments classified by copper type.

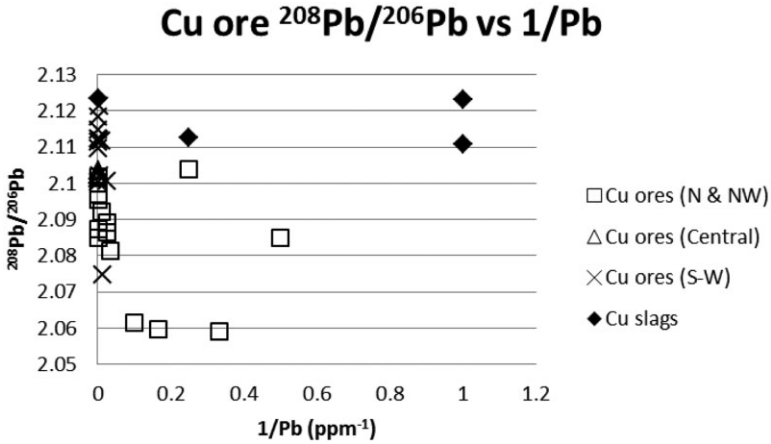


Figure 4 A plot of Sardinian copper ore and slag data (from Begemann et al. 2001, table 5), showing 1/Pb against  $^{208}\text{Pb}/^{206}\text{Pb}$ . The other two diagrams are essentially similar.

signal consistent with the main group of the ingots (i.e.,  $^{208}\text{Pb}/^{206}\text{Pb}$  between 2.07 and 2.08), suggesting that they may be made from this copper. More interesting are what appear to be mixing lines in the artefacts marked as A and B on Figure 7. Line A appears to represent mixing between the local ores (as defined by the bulk of the Sardinian ore isotope data, starting at  $^{208}\text{Pb}/^{206}\text{Pb} \approx 2.115$ ) and the ingot metal, thereby suggesting that some of the local objects might contain some ingot copper. Line B suggests some mixing between the ingot metal and another source with a much lower value of  $^{208}\text{Pb}/^{206}\text{Pb}$  ( $\approx 2.032$ ). Although no Sardinian ore in the reported data goes as low as this (Fig. 4), two values in the artefact data set are not shown in Figures 6 and 7—‘nondescript metal’ from Arzachena (20975G and 20975R)—and these have  $^{208}\text{Pb}/^{206}\text{Pb}$  values of 1.9074 and 1.6285, respectively. This certainly indicates the presence on the island of copper with very low isotope ratios. Although not shown here, the plots of 1/Pb versus  $^{207}\text{Pb}/^{206}\text{Pb}$  and 1/Pb versus  $^{204}\text{Pb}/^{206}\text{Pb}$  show very similar patterns.





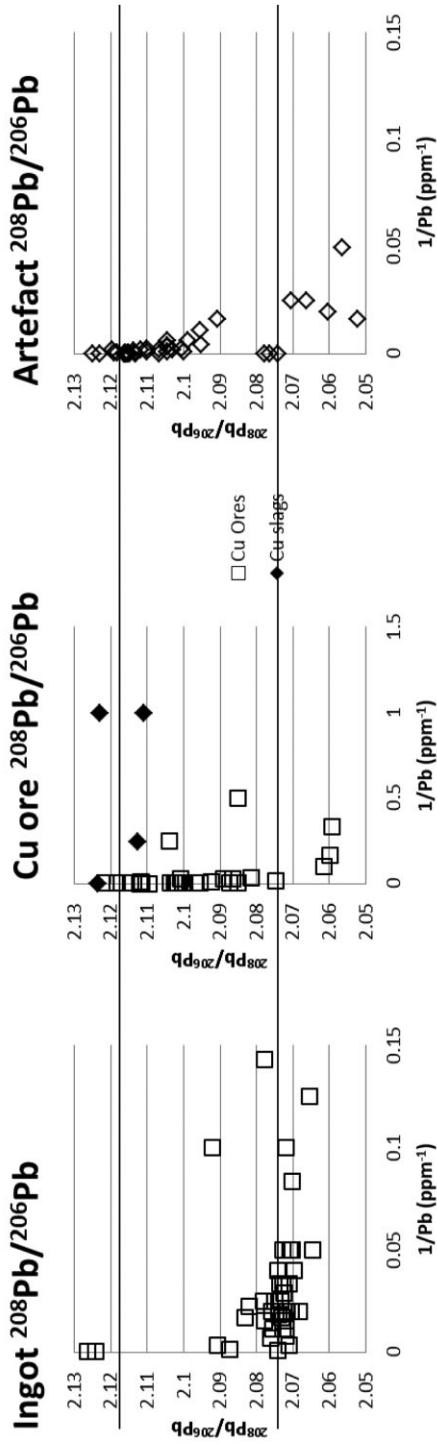


Figure 6 A comparison of plots of  $1/\text{Pb}$  against  $^{208}\text{Pb}/^{206}\text{Pb}$  for Sardinian ingot fragments, Cu ores and Cu slags, and artefacts. The upper and lower lines mark approximately the values for 'local' Sardinian ores (as evidenced by the Cu slags) and the main group of the ingot fragments.

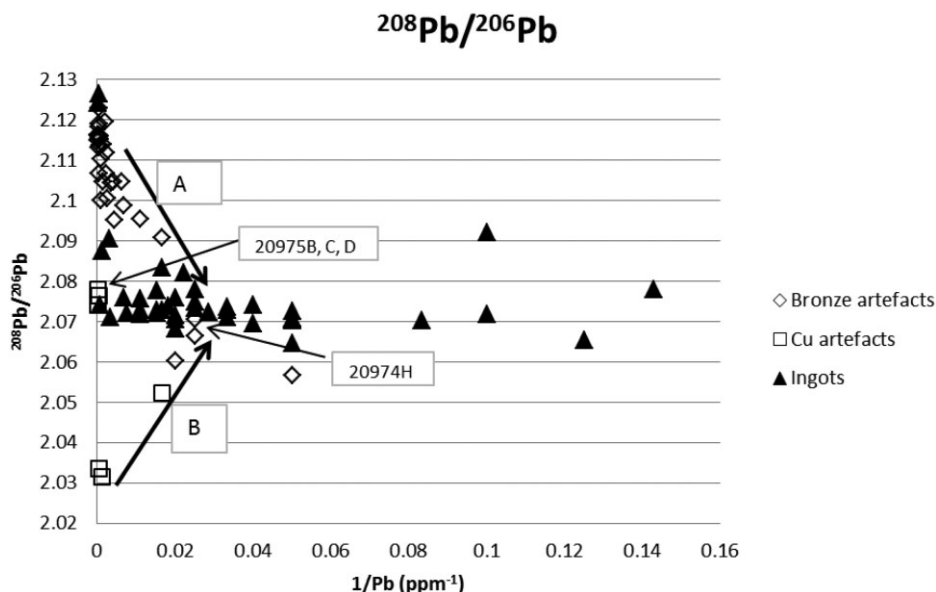


Figure 7 A plot of  $1/Pb$  against  $^{208}Pb/^{206}Pb$  for Sardinian ingot fragments and artefacts. The artefacts have been classified as either 'Cu' or 'bronze' (see text). A and B are suggested mixing lines.

highlights when inconsistent isotopic data are associated with very low lead levels, which is not apparent on conventional plots. The presence of 'spikes' in the isotopic data pinpoint quite precisely the existence of groups of objects with very similar isotope values, potentially marking specific sources. Finally, the existence of mixing lines shows up very clearly on these presentations. The obvious disadvantage is that the presentation disassociates the data from the models of isotopic evolution and the age of the ore deposit.

The apparent mismatch between particular source allocations for copper drawn from isotope geochemistry and the evidence from the rest of the archaeological data has sometimes been commented on in the literature. For example, Muhly (2009, 29–30) speculates that the copper ingots found on Sardinia but identified as coming from Apliki by the Gales (see above) are in fact very pure, unalloyed copper from Cyprus (but not necessarily only from Apliki), whereas the other copper found all over the eastern Mediterranean but identified isotopically as coming from 'Lavrion' contains recycled metal, which includes some lead originating from Lavrion, and is therefore identified as copper from Lavrion, despite the fact that Lavrion has no significant copper. As Muhly (2009, 30) clearly states '“Lavrion copper” is an artefact of the archaeometallurgical literature over the past twenty years'. If true, this is a remarkably important observation, which sets a very clear agenda for re-evaluating the data from the Mediterranean metal circulation. We suggest that the methodology proposed here will allow these hypotheses to be rigorously tested.

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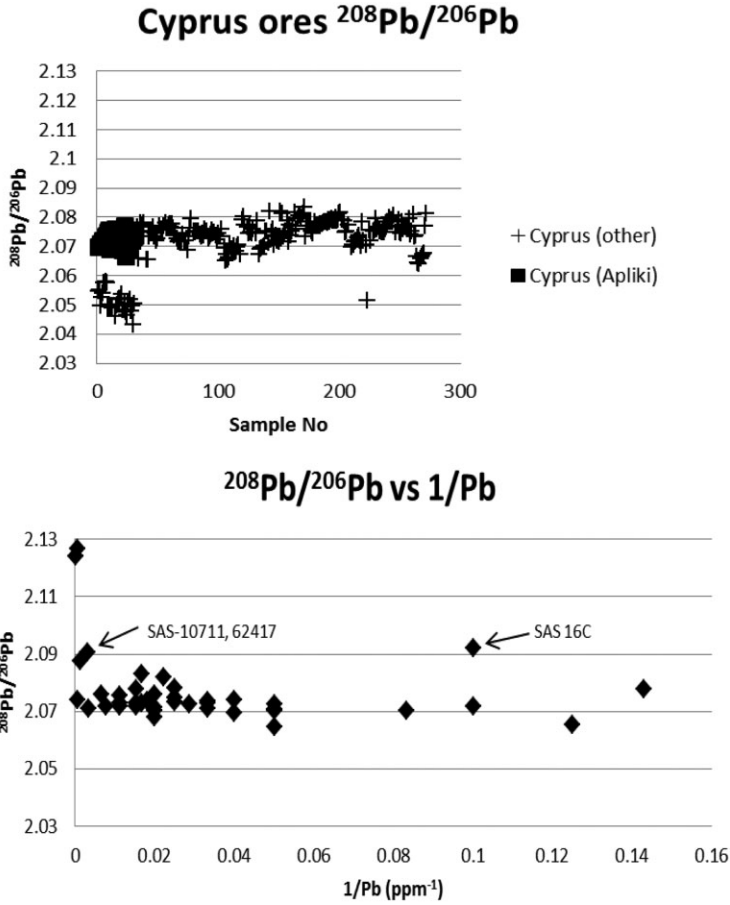


Figure 8 A comparison of <sup>208</sup>Pb/<sup>206</sup>Pb for Cypriot copper ores (Oxalid Database: see text) with plot of 1/Pb against <sup>208</sup>Pb/<sup>206</sup>Pb for Sardinian ingot fragments.

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