

2 / Lead Isotope Studies - Sardinia and the Mediterranean Provenance Studies of Artefacts Found in Sardinia

Introduction

I do not in this paper examine the origins of metallurgy in Sardinia in the late Neolithic period and its development in the Chalcolithic and EBA periods ⁽¹⁾, but it may be pertinent to recall that those origins have been attributed to the East, and have been linked to the obsidian trade networks (Lo Schiavo 2001, 135). Later developments were influenced by the western Megalithic and Beaker cultures, whilst in the MBA local imitations appear in Sardinia of central European and Italian Peninsular flanged axes and daggers (Lo Schiavo 1989, 1992). Here I limit myself to the Late Bronze Age which, in Sardinia, embraces the Recent Bronze Age (about 1350-1150 BC) and the Final Bronze Age (about 1150-1000 BC). In the RBA hoards there occur fragments of copper oxide ingots and of plano-convex copper ingots and the use of lead metal, whilst in the RBA we may also date the use of stone moulds, the production of simple tools and weapons and experiments with copper alloys. RBA metallurgy was the precursor of a boom in Sardinian metallurgy in the FBA, but the beginning of the new wave in metallurgy was slow, spread all over the island and was linked to the East and especially Cyprus by many artefacts of Cypriot type found in Sardinia (Lo Schiavo *et al.* 1985). "Current archaeological evidence suggests that Sardinians became reacquainted with copper via the arrival from Cyprus of metal and metallurgical expertise during the twelfth century BC" (Begemann *et al.* 2001, 58). There is, however, no existing evidence that proves that Cypriot metalworkers settled in Sardinia; it may rather be that the metal objects in Sardinia clearly based on Cypriot types reflect the presence of Cypriot itinerant metalworkers who came by sea and settled in Sardinia on a regular, but temporary, seasonal basis. It may be their presence which stimulated the search for, and exploitation of, local Sardinian copper ore sources, and the transfer of metallurgical and mining expertise to Sardinia.

Oxhide ingots

These ingots, formerly thought to be in the shape of the flayed hide of an ox and weighing on average about 30 kilogrammes each, were of course a principal form in which copper was traded around the Mediterranean region in the period roughly between 1700 BC and 1000 BC (Buchholz 1959; Bass 1967). Such copper oxhide ingots are found all over the Mediterranean (a distribution map is given by Gale and Stos-Gale 2005, 117), especially on the islands of Cyprus, Crete and Sardinia, but also in Turkey, Bulgaria, Greece, Sicily etc., with so far the only known oxhide ingot mould being found at Ras Ibn Hani in coastal Syria (Lagarce *et al.* 1983). The largest group of copper oxhide ingots found so far, now over 300 in number and weighing some 10 tonnes, come from the Uluburun shipwreck (Pulak 1988), dated to about 1300 BC; this ship also carried about 1 tonne of ingots of tin metal. The ship which sank off Cape Gelidonya about a century later (Bass 1967) also carried many copper oxhide ingots, both whole and including many fragments. To establish the trading patterns, which may have been different in different phases of the Late Bronze Age, we need to know the sources of the copper from which they were made. Normal archaeological methods, based on typological studies or studies of impressed signs, have failed to establish where any particular ingot originated. In spite of much effort by Muhly, Maddin and Stech, chemical analysis and metallographic examination have, by themselves, also failed to contribute much to the provenance question, however much they may have contributed to an understanding of the technological aspects of these ingots (their work is now largely superseded by Hauptmann *et al.* 2002).

In 1983 we began in the Isotrache Laboratory in Oxford a programme of work using lead isotope analyses to trace the provenance of the copper used to make oxhide ingots, with samples of such ingots from Crete and Cyprus. The first oxhide ingots were reported in the

archaeological literature from Serra Ilixi (Nuragus, Nuoro) in Sardinia by Spano in 1858, with many more known by the 1980's (indeed Lo Schiavo 2001) lists 31 separate sites in Sardinia which have yielded such ingots, or fragments thereof) – Sardinia clearly needed to be included. This was made possible by the generosity and enthusiasm of Fulvia lo Schiavo, who arranged for us to sample oxide and plano-convex ingots and a range of ore deposits in Sardinia in 1984. Preliminary findings from analysis of these ores and artefacts were presented at a Colloquium in Rome in 1986 (Gale, Stos-Gale 1987). The data presented showed clearly that the lead isotope data for the 22 'Sardinian' copper oxhide ingots then analysed in Oxford did not match the isotopic composition of the 80 Sardinian ores then analysed ⁽²⁾ nor any of the nuragic objects then analysed, but did overlap the isotopic compositions of the 8 analysed copper oxhide ingots from Cyprus. Nonetheless we were careful not to draw premature conclusions, stating (Gale, Stos-Gale 1987, 162) "Nevertheless we must stress that we cannot yet be absolutely sure that the Sardinian (oxhide) ingots are foreign imports. Before drawing such a conclusion it is clearly necessary to analyse ores from other Cretaceous and younger ore deposits in Sardinia." Some further advances were reported by Gale (1991) and by Stos-Gale, Gale (1992), which directly disproved suggestions by Muhly (1991, 190) and by Cherry and Knapp (1991, 100) that lead isotope compositions of ores from Spain, the Negev Desert or Oman might overlap with those for Cyprus, presented new lead isotope data for ores and artefacts from Sardinia and also showed that the lead isotope compositions of ores from the Colline Metallifere in Tuscany did not overlap the isotopic compositions of oxhide ingots found in Sardinia. Stos-Gale and Gale (1992, 332) emphasised also that trace Au and Ag contents in the Sardinian copper oxhide ingots overlapped with those for oxhide ingots excavated on Cyprus but still urged caution, stating that "It would at present be presumptuous to make a final judgment about the origin of the oxhide ingots found in Sardinia ...". Apart from the necessity then identified of increasing the lead isotope data base for Sardinian copper ores, especially for Tertiary and younger ores such as Calabona, in 1994 we had only 43 accurate lead isotope data for Cypriot ore deposits including but one ore from Apliki, given to us by Tylecote, yet there were already indications that various regions in the Cypriot ore deposits had lead isotope compositions separable from each other (Hamelin *et al.* 1988; Gale, Stos-Gale 1995) and that the single available Apliki ore plotted at the extreme lower boundary of what we then accepted as the Cypriot lead isotope field (Gale, Stos-Gale 1995, Fig. 1). We were fortunate that on a field trip in Cyprus in 1994 we fortuitously met Dr G. Malotis of the Hellenic Mining Company, who personally showed us and gave us access to a great many ore and slag deposits in Cyprus, including the rarely accessible Apliki mine near the border with Turkish occupied Cyprus, which we visited with the permission of the Greek Cypriot military authorities. As a result Gale *et al.* (1997) published some 210 lead isotope analyses of Cypriot ores from 27 ore deposits which showed that a single lead isotope field for Cyprus had to be abandoned, and which were compared by Stos-Gale *et al.* (1997) with the lead isotope compositions of 79 copper oxhide ingots from sites in Crete, Greece, Cyprus, Sardinia, Turkey and Bulgaria. The 29 analysed oxhide ingots from Sardinia overlapped the isotopic compositions of the ores from Apliki and the oxhide ingots found on Cyprus, but we still urged caution, writing (Stos-Gale *et al.* 1997, 115) "We still would like to withhold final judgment about the origin of the copper used to make these ingots found on Sardinia until we have analysed Sardinian copper ores from the Calabona deposit in north-west Sardinia...". By 1998 lead isotope analyses had been made in Oxford of about 65 oxhide ingots, 20 bun ingots, 75 artefacts and 270 ores from Sardinia (including 12 from Calabona), together with 210 ores from a range of Cypriot deposits (Gale *et al.* 1997; Stos-Gale *et al.* 1997). Gale (1999) discussed this data especially concerning the Cypriot and Sardinian ore deposits and its bearing on the sources of copper for copper oxhide ingots from land based sites and the Bronze Age shipwrecks of Cape Gelidonya and Uluburun. He showed that the lead isotope data for 31 copper oxhide ingots found on Sardinia did not overlap the isotopic data for any Sardinian ore deposits, including Calabona, Castello di Bonvei or Capo Marargiu, but did closely overlap the isotopic data for oxhide ingots found in Cyprus and the lead isotope data for the ores from the Apliki mine in Cyprus. Together with the supporting evidence of the overlap of the gold – silver content of the Sardinian oxhide ingots with those for Cypriot oxhide ingots (Stos-Gale *et al.* 1997, 112), it had become inescapable in 1999 that the copper oxhide ingots found on Sardinia had been made of copper smelted from ores from the Cypriot deposit of Apliki. The Cypriot origin of the oxhide ingots found on Sardinia has been accepted by Kassianidou (2001, 110), Scaife *et al.* (1999, 131) and Lo Schiavo (Begemann *et al.* 2001, 71).

However the Bradford group correctly, in accordance with usual scientific practise, hoped for confirmatory analyses from an independent laboratory (Scaife *et al.* 1999, 131). The work of Begemann *et al.* (2001) has provided that confirmation. These researchers made chemical and lead isotope analyses of 29 Sardinian copper oxhide ingots, 10 planoconvex ingots, 45 artefacts and 54 ores from Sardinian deposits. Their trace element data show that the Au and Ag contents for Sardinian oxhide ingots fall with the data measure in Oxford for other Sardinian

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oxide ingots and oxide ingots found in Cyprus (see Begemann *et al.* 2001, 56, Fig. 7a ; Gale, Stos-Gale 2005, 122, Fig. 4). Figure 1 plots the lead isotope data of Begemann *et al.* 2001 for Sardinian copper oxide ingots together with data measured in Oxford for 32 different Sardinian copper oxide ingots and data for ores from Cypriot ore deposits. It is clear that the majority of the Sardinian oxide ingot data measured in Mainz overlaps the oxide ingot data measured in Oxford, overlaps the isotopic data for oxide ingots found on Cyprus and the data for the Apliki ores from Cyprus (3). A comparison of the lead isotope data for about 3000 ores throughout the western, central and eastern Mediterranean regions, Serbia, Bulgaria, Turkey, Feinan, Timna, Oman, Saudi Arabia and Egypt shows that none of it overlaps the data for the Apliki ores. The lead isotope composition of ores from Apliki stands by itself, so that the coincidence with it of the isotopic data for oxide ingots from Sardinia and Cyprus argues extremely strongly that both sets of ingots were made from copper smelted from copper ores from Apliki. We agree with Begemann *et al.* (2001, 45) when they write that : “Contrary to widespread belief (e.g. Chippindale 1994), the information to be gained from lead isotope plus trace element concentrations is not redundant. Rather, the combination of the two has turned out to be a much more powerful tool than each by itself for identifying ore sources...”. Indeed Stos-Gale and Gale (1992, 332) showed how the gold and silver contents of copper oxide ingots found on Sardinia and Cyprus occupied the same region in Au-Ag space, well separated from nuragic bronzes from Santa Maria in Paulis, a conclusion supported by the new analyses reported by Begemann *et al.* (2001, 56) for oxide ingots found on Sardinia. Some years ago Budd *et al.* (1995, 25-27), arising out of their belief that it would not be possible to separate the lead isotope compositions of the Cypriot and Sardinian ore sources, proposed the hypothesis that oxide ingots represented the mixture of both recycled metal and copper that originated from various mines around the Mediterranean which belonged to a Koine. It is now clear that Cypriot and Sardinian ore sources can be discriminated by their lead isotope compositions and that copper alloy artefacts can be assigned to them, whilst detailed examination of the idea of a recycling Koine has shown that insuperable scientific and archaeological difficulties prevent its acceptance (Gale, Stos-Gale 1995, 35-36 ; Knapp 2000, 43 ; Kassianidou 2001, 102-103), so that the Budd *et al.* 1995 paper is now wholly redundant.

Oxhide ingot moulds

It is well known that the only mould for an oxide ingot yet discovered is that reported at Ras Ibn Hani by Bounni *et al.* (1998 ; see also Klengel 2005). Scattered around this mould were droplets of copper metal, ejected from the mould when molten copper metal was poured into it to cast an oxide ingot. Some samples of these droplets were acquired in Oxford from the excavators via U. Zwicker ; lead isotope analyses of three of these copper droplets are plotted in Figure 1. They

clearly plot with the lead isotope compositions of oxide ingots found in Cyprus and with the Cypriot ores from Apliki. Consequently copper smelted from Apliki ores was being cast into oxide ingot form in at least one mould at Ras Ibn Hani.

Planoconvex ingots found in Sardinia

Table 1 gives the lead isotope compositions and lead concentrations for a number of planoconvex copper ingots found in Sardinia ; data for 10 different planoconvex ingots is given by Begemann *et al.* (2001, 54). Figure 2 plots the lead isotope data in relation to isotopic data for Sardinian ore deposits. It is clear that 26 planoconvex ingots are consistent with their metal having been smelted from Sardinian ores, including six ingots matching copper ores from NW Sardinia. Of these seven contain less than 0.06 % Pb, and on any basis cannot have had the

Sample No.	Site	Nuraghe etc.	²⁰³ Pb/ ²⁰⁶ Pb	²⁰⁷ Pb/ ²⁰⁶ Pb	²⁰⁸ Pb/ ²⁰⁶ Pb	% Pb
OL-2	Oliena (NU)	Sa Sedda 'e sos Carros	2,112 13	0,864 60	18,113	0,11
OL-3	Oliena (NU)	Sa Sedda 'e sos Carros	2,098 53	0,856 56	18,264	30,56
OL-13	Oliena (NU)	Sa Sedda 'e sos Carros	2,100 10	0,858 95	18,157	2,15
OL-14	Oliena (NU)	Sa Sedda 'e sos Carros	2,084 18	0,845 27	18,455	0,029
OL-15	Oliena (NU)	Sa Sedda 'e sos Carros	2,120 39	0,873 78	17,879	47
OL-15a	Oliena (NU)	Sa Sedda 'e sos Carros	2,122 89	0,874 13	17,916	32
OL-15b	Oliena (NU)	Sa Sedda 'e sos Carros	2,122 99	0,874 01	17,890	1,5
SS-A (10265)	Alghero (SS)	Nuraghe Flumenelungu	2,061 85	0,827 93	18,868	0,18
SS-A1 (10268)	Alghero (SS)	Nuraghe Flumenelungu	2,062 14	0,830 44	18,816	<0,01
SS-A2 (10240) [bar]	Alghero (SS)	Nuraghe Flumenelungu	2,102 49	0,854 95	18,333	16,81
SS-A3 (10260)	Alghero (SS)	Nuraghe Flumenelungu	2,093 89	0,853 52	18,335	0,06
SS-A4 (10274)	Alghero (SS)	Nuraghe Flumenelungu	2,101 37	0,854 91	18,36	25,39
SS-A5 (10262)	Alghero (SS)	Nuraghe Flumenelungu	1,849 77	0,755 12	20,922	0,051
SS-E (107 08)	Bonnano (SS)	Funtana Janna	2,082 35	0,842 46	18,55	0,015
SS-F (10709) [cone]	Bonnano (SS)	Funtana Janna (cone)	2,066 52	0,830 87	18,833	0,05
SS-G (10710)	Bonnano (SS)	Funtana Janna	2,074 34	0,837 20	18,621	0,26
SS-H (10711)	Bonnano (SS)	Funtana Janna	2,090 45	0,847 87	18,445	0,065
SS-O p.135-139	Ossi (SS)	Sa Mandra 'e Sa Giua	2,118 64	0,870 21	17,929	1,01
SS-P p.135-139	Ossi (SS)	Sa Mandra 'e Sa Giua	2,118 58	0,870 09	17,969	0,22
SS-Q	Ossi (SS)	Sa Mandra 'e Sa Giua	2,107 63	0,861 63	18,170	0,17
SS-S	Ossi (SS)	Sa Mandra 'e Sa Giua	2,120 68	0,871 32	17,921	2,15
SS-T	Ossi (SS)	Sa Mandra 'e Sa Giua	1,854 21	0,820 28	19,146	0,067
SS-W	Ossi (SS)	Sa Mandra 'e Sa Giua	2,119 06	0,871 05	17,902	0,01
CA-5 (CA 46)	Sarroch (CA)	Monte Arbulu	2,073 14	0,841 06	18,543	0,013
CA-10 (CA 47)	Sarroch (CA)	Monte Arbulu	2,097 28	0,852 05	18,405	0,52
BS6	Villanovaforru (CA)	Baccu Simeone	2,118 29	0,869 19	17,985	0,52
BS11	Villanovaforru (CA)	Baccu Simeone	2,071 69	0,841 41	18,489	<0,01
BS13	Villanovaforru (CA)	Baccu Simeone	2,102 31	0,859 61	18,151	<0,01
BS27	Villanovaforru (CA)	Baccu Simeone	2,111 06	0,865 78	18,020	0,51
BS34	Villanovaforru (CA)	Baccu Simeone	2,113 86	0,867 16	18,025	<0,01
BS45	Villanovaforru (CA)	Baccu Simeone	2,106 71	0,861 24	18,166	0,011
BS53	Villanovaforru (CA)	Baccu Simeone	2,115 60	0,869 61	17,915	0,20
BS59	Villanovaforru (CA)	Baccu Simeone	1,769 92	0,744 62	21,168	0,021

Table 1 — Lead isotope analyses of planoconvex ingots found on Sardinia Re-classified to planoconvex from the classification in Quaderni 17 (1990).

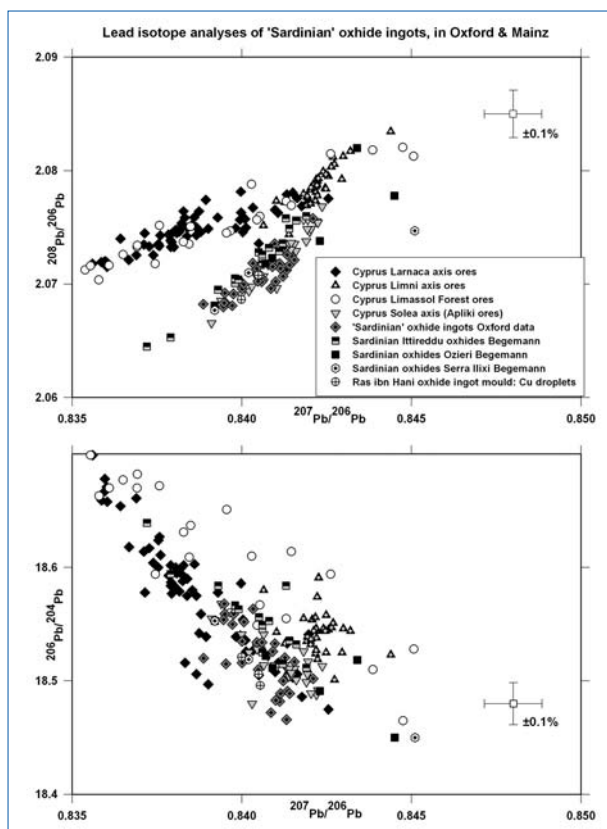


Fig. 1 — Lead isotope analyses of “Sardinian” oxide ingots, in Oxford and in Mainz.

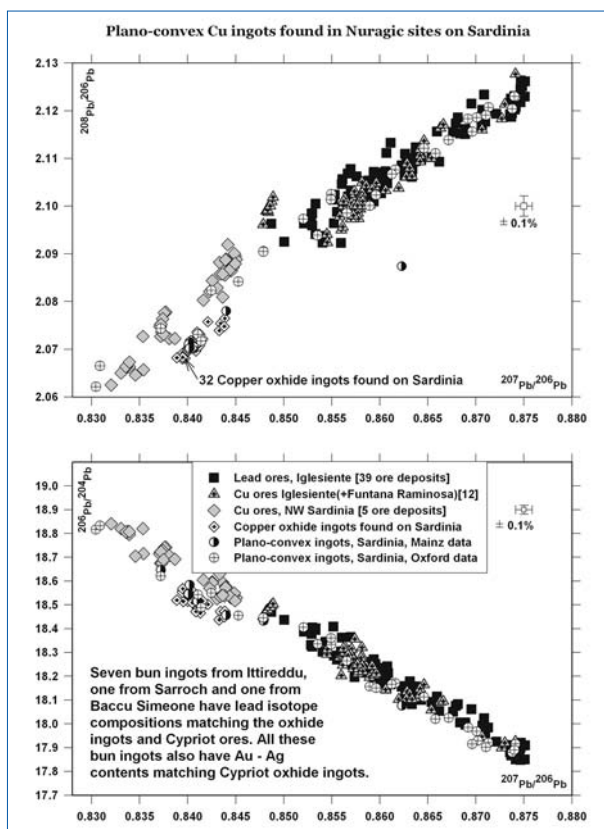


Fig. 2 — Lead isotope analyses of plano-convex copper ingots found in nuragic sites on Sardinia.

lead isotope compositions of the relevant source copper ores masked by the addition of lead metal to the ingot. Consequently they are clear evidence for the production by nuragic people of planoconvex copper ingots by smelting Sardinian copper ores ; there is no reason to doubt that another 14 ingots with lead contents between 0.1 % and 2.15 % can be interpreted in the same way (4). There is another group of nine planoconvex ingots found in Sardinia which overlap with Cypriot and Sardinian oxhide ingots and ores from Apliki ; these ingots parallel Cypriot planoconvex copper ingots carried with Cypriot copper oxhide ingots on the Cape Gelidonya ship (Gale 1999, 118). These nine ingots no doubt came to Sardinia on a similar ship from Cyprus. Thus the majority of the analysed planoconvex ingots support that nuragic people were smelting copper ores found on Sardinia itself, whilst others represent ingots smelted from Cypriot Apliki copper ores and coming from Cyprus to Sardinia together with copper oxhide ingots smelted from Apliki ores.

Copper based artefacts found in Sardinia and leaded bronze

In 1987 we reported lead isotope and chemical analyses (Gale and Stos-Gale 1987) of nuragic bronzes from the Sardinian hoard of Santa Maria in Paulis (Macnamara *et al.* 1984) held in the British Museum. We found them to match isotopically ores from Sardinia, not Cyprus, and to be distinguished from Cypriot copper oxhide ingots not only isotopically but also by the much higher concentrations of silver in the nuragic bronzes. This prompted the perhaps premature response (Muhly 1996, 49) “Even worse was the situation on Sardinia. There none of the analysed artefacts, especially those from Santa Maria in Paulis, was made of Cypriot copper. What happened to all the Cypriot copper exported in the form of copper oxhide ingots ?” Kassianidou (2001), who accepts the lead isotope evidence that the copper oxhide ingots found on Sardinia come from Cyprus, sought to explain this away by suggesting that even if Cypriot copper was used to make some of the Santa Maria in Paulis objects, it could not have been detected by lead isotope analysis if the very low lead (~0.01 % Pb) Cypriot copper had been deliberately mixed with some local Sardinian lead “to facilitate the casting”. In developing her argument Kassianidou (2001, 105-108) criticizes Gale and Stos-Gale (1987) for failing to report lead concentrations for the artefacts from Santa Maria in Paulis, but Kassianidou has missed the point that we had no need to do so, because Craddock and Tite (1984) (5) had already published these as part of their chemical analyses of the Hoard. These analyses showed that the mean lead content of the 31 Santa Maria in Paulis artefacts was 0.8 ± 0.4 % Pb [95 % confidence level] ; these low lead concentrations led Gale and Stos-Gale (1987) to interpret the lead as coming from the smelted copper ores, so that their lead isotope compositions give the provenance of the smelted copper ores. In considering Kassianidou’s arguments we are brought up against the question as to how is it possible to determine whether lead has been deliberately added to a copper based artefact, and why ? Many archaeologists and archaeometallurgists assume that a lead content above 1 % (sometimes 2 %) denotes the deliberate addition of lead, but we have seen no logical arguments underpinning such assumptions. Work done by Northover and his colleagues in Oxford and Warwick (Young 1972 ; Staniaszek 1982 ; Staniaszek, Northover 1982) showed that addition of quite small amounts of lead to a tin-bronze lowered the melting point and greatly improved the fluidity of the molten alloy, allowing easier pouring and casting of complex shapes. Maximum fluidity was achieved at 2 % Pb, further additions of lead achieving little in this regard. There is little evidence that nuragic smiths exploited these effects. Elaborate plaques, tripod stands and jugs from Santa Maria in Paulis contained from 0.02 % to 0.28 % Pb, levels too low to affect fluidity and castability. Addition of large quantities of lead, as in the British Late Bronze Age (Brown, Blin-Stoyle 1959), could be to economize in the use of more precious copper metal, but again there is no support for this practise in nuragic Sardinia. Figure 3 shows a histogram of lead contents in 92 copper based artefacts from nuragic sites in Sardinia ; the majority contains less than 0.5 % Pb, far too low to save any appreciable amount of copper metal.

Thus far too little regard has been paid by archaeologists and archaeometallurgists to the amount of lead in copper metal which may result accidentally from Bronze Age smelting and refining of lead containing copper ores, such as exist in Sardinia. A guide to the possibilities can be derived from reports of the analyses of copper from 19th. Century smelting furnaces as recorded by Percy (1861 ; 1880), reproduced below in Table 2.

Tylecote *et al.* (1977) undertook direct copper smelting and fire refining experiments, including chemical analyses of the smelted ores and the various products. They found that the lead concentration in the smelted copper was roughly half that in the smelted copper ores and, with a suitable lead containing copper ore, they produced a raw copper naturally containing as much as 2 % lead, thus confirming the possibility of the levels of lead in smelted copper recorded by Percy. The possibility that Bronze Age copper metal could contain elevated contents of lead coming from the ores was further confirmed by the finding of Tylecote *et al.* (1977) that fire refining which largely removed the iron content

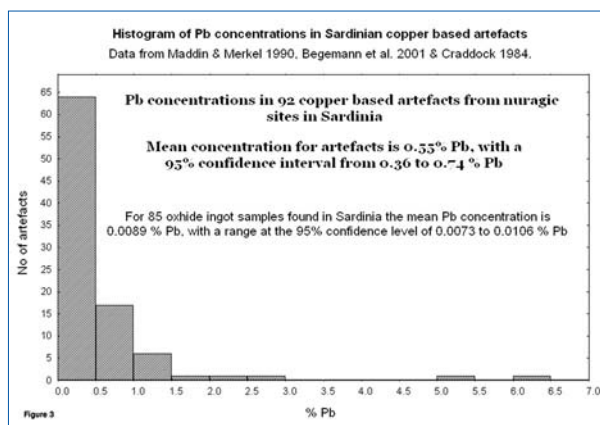


Fig. 3 — Histogram of lead concentrations in Sardinian copper based artefacts.

Smelter	Product	% Pb	% Fe	Reference - Percy
Rammelsberg	Black copper	0.97	4.23	1861, p. 422
Rammelsberg	Black copper	2.79	1.38	1861, p. 422
Riechelsdorf	Black copper	0.89	1.41	1861, p. 429
Mansfeld	Rosette copper	1.09	0.13	1861, p. 503
Avesta, Sweden	Refined copper	0.48	0.05	1861, p. 503
(Genth, analyst)	Japanese copper	0.74	0.07	1861, p. 503
Mansfeld	Black copper	2.79	1.38	1880, p. 307
Freiberg	Black copper	20.75	3.31	1880, p. 307

Table 2 —

from the smelted copper, did not much reduce the content of lead. Thus all the available evidence supports that it is completely possible for Bronze Age fire refined copper metal, smelted from lead containing ores, naturally to contain a few per cent of lead. It is therefore mistaken to conclude, as did Kassianidou (2001, 107), that lead contents of a few percent in a Bronze Age bronze artefact “can be shown to be a deliberate addition”.

Kassianidou (2001, 106) also suggested another reason why, in nuragic bronzes, the isotopic composition of Sardinian lead might have masked that of Cypriot copper, by suggesting that the tin added to Cypriot copper might have been derived by adding Sardinian cassiterite which “in at least one area ... was found to contain lead (Tylecote *et al.* 1984, 129)”. Kassianidou probably did not read Tylecote’s source for this idea, which was Botti (1936), who was writing about the first exploration and ore analyses from the Sardinian deposit of Canali Serci of the commune Villacidro (CA), owned by the Società Anonima Miniera di Montimannu. We visited this ore deposit in 1984 (Gale, Stos-Gale 1987, 165, note 14), noting that the chief ore is a finely and intimately intergrown mixture of sphalerite, galena, pyrite and cassiterite, in which the cassiterite can be recognized only under the microscope, so that it is most unlikely that cassiterite could even have been recognized in it in the Bronze Age, far less separated or utilized. Botti (1936) confirms all our observations, whilst Valera *et al.* (2005) in their review of tin deposits in Sardinia write of Canali Serci that “Minor cassiterite is finely intergrown with major zinc and lead sulfides, and it is only visible under the microscope. Again the same questions arise : was a primitive tin metallurgy viable for such a primary ore association ? Was a cassiterite placer concentration possible in the local morphology ? Both answers are negative, because only a modern flotation process can separate and concentrate each ore mineral from such a primary mixture, and only a weak geochemical increase may be expected in the stream sediments derived from the erosion of the Canali Serci lode”. Thus the idea that a Sardinian lead rich cassiterite might have been added to Cypriot oxhide ingot copper to make a bronze, masking the Cypriot lead isotope composition with the Sardinian isotopic composition, collapses entirely, being based on a careless and misleading reading of the published evidence, and lack of personal observation of the Sardinian ore deposits. Valera *et al.* (2005) confirm our own observations that there is no source of lead containing tin in Sardinia available in the Bronze Age, and indeed that the tin occurrences in Sardinia are mostly mineralogical occurrences only, “with the only hypothetical possibility, though very weak, of a Sardinian tin beneficiation being offered by an eventual small “placer” from the Perdu Cara mineralisation”. Begemann *et al.* (2001, 67-68) discuss in some detail the question whether lead may have been introduced to nuragic tin bronzes with the added tin, mention the low content of lead in analysed tin ingots from Uluburun, Hishuley Carmel and Kfar Shamir (Begemann *et al.* 1999 ; Stos-Gale *et al.* 1998), and generally find the idea implausible. The trace element analyses of ancient tin ingots by Begemann *et al.* (1999) also show that the silver contents

are very low, and cannot account for the high silver contents in the Santa Maria in Paulis artefacts.

Thus the available evidence, together with the generally low lead contents of the Santa Maria in Paulis bronze artefacts (Craddock, Tite 1984), reinforces our original assumption that the lead in these nuragic artefacts was not deliberately added, but came from the copper ores smelted to provide the copper metal of the artefacts, so that lead isotope analyses would properly provenance the copper in the bronze artefacts. However it would be better if we could actually prove that lead was not deliberately added to the Santa Maria in Paulis artefacts. It transpires that such proof is available from the trace element analyses of the objects. Gale and Stos-Gale (1987, 156, 159) showed that the artefacts from Santa Maria in Paulis contained about 10 times more silver than did the copper oxhide ingots found on both Sardinia and Cyprus. If these nuragic artefacts had, as suggested by Kassianidou (2001, 106, 108), been made from Cypriot oxhide ingot copper "mixed with some local lead to facilitate the casting", then the only possibility (since the idea has been discounted that lead was introduced with tin) to account for the much higher silver content of the nuragic artefacts is that it was introduced with the hypothetically added lead. This can be assessed quantitatively, now that we have fairly extensive trace element analyses of nuragic lead artefacts (Lo Schiavo *et al.* 2005; Gale, Stos-Gale 1987, 167); the mean silver content of lead artefacts from 13 nuragic sites was 130 ppm with a standard deviation of 113 ppm and a range from 14 to 432 ppm. If we take the worst case that the Santa Maria in Paulis bronze artefacts were made by adding tin and Sardinian lead containing 432 ppm silver to Cypriot oxhide ingot copper containing 100 ppm Ag, then from the measured lead content in these bronzes the amount of silver in the resulting bronzes would be from 12 to 7 times less than that actually measured in these artefacts by Craddock and Tite (1984). The hypothesis of Kassianidou (2001, 106) that the reason that no nuragic bronze artefacts have the lead isotope composition of Sardinian/Cypriot copper oxhide ingots is that the lead in the conjectured oxhide ingot copper was swamped by the addition of Sardinian lead; we now see that this hypothesis fails because it cannot explain the silver content of the nuragic artefacts. This is true also for the nuragic bronze artefacts for which trace elements and lead isotope analyses were reported by Begemann *et al.* (2001); they all have higher silver contents than the oxhide ingots, have lead isotope compositions different from the oxhide ingots and matching Sardinian ores, and have lead contents consistent with their copper having been smelted from polymetallic Sardinian ores. Some of the polymetallic Sardinian ore sources are known to have had high silver grades and were sources of copper, lead and silver (6), which is a further point in favour of the high silver nuragic bronzes having been made using copper smelted from such Sardinian ores.

That nuragic bronzes were, at least in many cases, not made by adding tin and Sardinian lead metal to oxhide ingot metal is also proved by a detailed examination of lead isotope data, as in Figure 4. Work by Valera *et al.* (2005) (7) shows that the lead isotope data for lead metal from 14 nuragic sites clusters together at about $^{208}\text{Pb}/^{206}\text{Pb} = 2.12$, $^{207}\text{Pb}/^{206}\text{Pb} = 0.874$ and $^{206}\text{Pb}/^{204}\text{Pb} = 17.87$; this agrees with more precise lead isotope analyses by TIMS at Oxford for lead metal artefacts from the sites Nuraghe Antigori and Nuraghe Albuccio (Gale, Stos-Gale 1987, 167). It seems that lead metal of a fairly restricted range of lead isotope compositions was used by nuragic metal workers; it would be lead of this isotopic composition that would have been available to add to copper, if this was ever done. A copper or bronze artefact containing 0.05 % Pb made by adding lead of this isotopic composition to oxhide ingot copper (containing 0.01 % Pb) would have an isotopic composition of $^{208}\text{Pb}/^{206}\text{Pb} = 2.11$, $^{207}\text{Pb}/^{206}\text{Pb} = 0.867$, $^{206}\text{Pb}/^{204}\text{Pb} = 18.0$. In Figure 4 ellipse A includes Sardinian copper ores, and nuragic artefacts from six sites with Pb contents ranging from 0.02 % to ~5 %. These measured lead isotope compositions, as indicated, do not approach the nuragic lead metal isotopic composition, but do overlap with Sardinian copper ores. Consequently none of these 13 nuragic bronze artefacts could have been made by mixing nuragic lead metal with oxhide ingot copper metal – they are most naturally explained as having been made from copper smelted from Sardinian ores which contained lead to varying degrees.

Conclusions

- The Sardinian oxhide ingots were not made of copper smelted from Sardinian ores, but were made of Cypriot copper from the same Cypriot ore source(s) which were used to make oxhide ingots found on Cyprus and those excavated from the Cape Gelidonya shipwreck.

- The majority of these oxhide ingots have lead isotope analyses (LIA) which overlap only ores from the Apliki mines in Cyprus, but some overlap also the LIA of the Mavrovouni and Skouriotissa mines on Cyprus.

- It is not suggested that the LCII site of Apliki-Karamallos on Cyprus was the only site which produced copper for making the copper oxhide ingots found on Cyprus, Sardinia or the Cape Gelidonya Bronze Age shipwreck.

- LIA of Cypriot 'bronze' artefacts show that these were made of copper smelted from a number of copper deposits around the Troodos in Cyprus, not just the Apliki deposit. One such smelting site was Politiko-Phorades, though this seems to have been operating too early to have been a source for oxhide ingots.

- Of 53 analysed copper bun ingots found on Sardinia, 46 have LIA indicating that they were produced from Sardinian ore deposits, whilst 9 have LIA indicating they were produced from copper from the Apliki deposit in Cyprus. Since the Cape Gelidonya ship carried both oxhide and bun ingots produced from the same deposit, one can surmise that a similar ship came from the east to Sardinia with a similar cargo, probably including Sn.

- The 46 bun ingots, and 4 copper slags, with Sardinian LIA show that nuragic metal smiths were exploiting the Sardinian copper ores.

- LIA of most analysed nuragic bronze artefacts match Sardinian copper ores – they do not match the Iglesias-Arburese galena LIA composition which has been shown (Valera *et al.* 2002, 2005) to characterize Pb metal from 14 nuragic sites. Nor do they match the LIA of the Zn, Pb, cassiterite bearing deposit at Canale Serici, Villacidro (from which cassiterite could not be extracted, or even recognized, in the Bronze Age). Current evidence, embracing both lead isotope analyses and silver contents, does not support the suggestion (Kassianidou 2001; 2005) that some nuragic bronze artefacts were made of oxhide ingot copper to which Sardinian Pb-bearing cassiterite was added.

- The Pb content of nuragic bronzes averages ~0.5 %, far too low for Pb to have been added deliberately to economize on use of Cu, and in the main too low materially to enhance fluidity or reduce freezing temperature.

- Though the very low Pb content (~0.01 %) of oxhide ingot copper would result in even small additions of Sardinian Pb metal swamping its LIA, the above considerations together with detailed examination of the actual LIA and Pb content of individual nuragic bronze artefacts and their silver content does not provide any evidence that this occurred.

- Evidence drawn from Tylecote (1977) and from J. Percy's treatises on Metallurgy (1861; 1880) show both that Pb can occur naturally at fairly high levels in smelted copper, and is difficult to remove by fire refining.

- Consequently the existing evidence suggests that most nuragic bronze artefacts were made from copper smelted from Sardinian

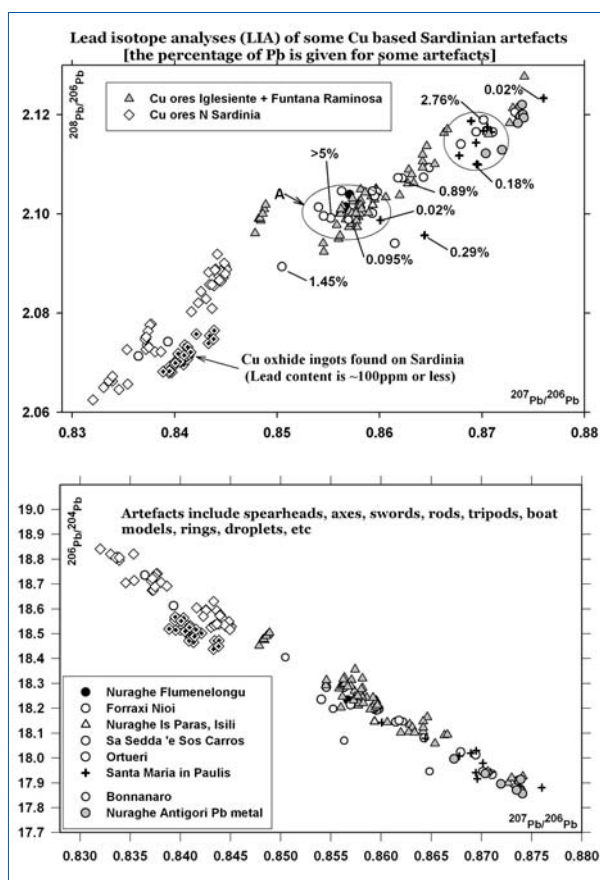


Fig. 4 — Lead isotope analyses of some copper based Sardinian artefacts.

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copper ores, not from the oxhide ingots which came from Cyprus to Sardinia, attesting to active nuragic exploitation of Sardinian copper ores.

- The suggestion (Gale *et al.* 1987) that the Sardinian copper oxhide ingots may be made of Cypriot copper was at first strongly resisted using the 'Coals to Newcastle' analogy ; an argument only partially countered by the fact that the Cu ore resources of Cyprus dwarf those of Sardinia.

- Present evidence strongly supports that Sardinian oxhide ingots were of Cypriot copper, but that most Sardinian bun ingots and bronze artefacts were of Sardinian copper, 3. The 9 nuragic bun ingots now proved to be of Cypriot copper suggest that a ship or ships, similar to C. Gelidonya, laden with both bun and oxhide copper ingots, came to Sardinia from the East. Such a ship, on the analogy with Cape Gelidonya & Uluburun, is likely to have been carrying also tin metal.

- Though such a ship may have been en route further west, or even stranded or wrecked on Sardinia, it may have been bringing to Sardinia primarily tin metal, vital for the nuragic bronze industry but, as we now know from Valera *et al.* (2005), virtually unavailable from Sardinian ores.

- It would not be at all unreasonable if a condition of the supply of vital tin was to accept also some copper ingots from the ship. As Kassianidou (2005) has suggested, copper metal, even though available from Sardinian ores, might not be unacceptable, since it avoided the laborious processes of smelting and refining.

- Might, however, the nuragic smiths have found the ultra low Pb Cypriot Cu had less desirable casting properties than that smelted from Sardinian ores ? Pliny (XXXIV,II) writes that even the Romans eventually found Cypriot copper of lower value compared with that from other sources.

- Since royal gift exchange, or a princely cargo going to Sardinia, seem unlikely, in view of the lack of archaeological evidence in Sardinia for rigid differences in classes or the existence of kings, what was sought by the voyagers from the east in exchange for tin or copper ?

- Perhaps we should rather think in terms of free lance trade and tramping (Braudel 1972, 107 ; Cherry, Davis 1982, 339), which gave ample opportunity for bargaining both for the crew and for merchants who would travel with their wares, and where the round trip was a long succession of selling, buying and exchanging. In the course of the voyage the cargo would change its nature, being partly depleted and replaced by local goods, trading being based on the chances encountered in each port, and not necessarily on a pre-arranged scheme between, say, Cyprus and Sardinia.

- As Muhly *et al.* (1977, 361-62) wrote : "The organisation and administration of the metals trade must have been on several levels. It is conceivable that some central authority exercised control over the mines in certain areas ... Smelting probably took place close to the mines and may also have been subject to a loose central control." Is it not possible that this model may be applicable to the production of Cypriot copper for export, centered on Apliki, whilst copper for consumption within Cyprus itself might have been produced at a number of centres around the Troodos ?

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Notes :

- (1) For Sardinian chronology see Lo Schiavo 2005, fig. 8, p. 466.
- (2) There were also available for comparison lead isotope data for Sardinian ores accumulated by isotope geochemists (e.g. Swainbank *et al.* 1982 ; Boni, Koeppel 1985 ; Ludwig *et al.* 1989 ; Boni *et al.* 1992 ; Caron *et al.* 1993), but they were of limited use because they included no Sardinian copper ore deposits other than Funtana Raminosa and Rosas.
- (3) Oxhide ingots 62395 and 62414 from Ittireddu, analysed by Begemann *et al.* 2001, fall just beyond the range of presently analysed ores from Apliki, but do fall with copper oxhide ingots from the Uluburun shipwreck.
- (4) We shall see later that there is absolutely no reason to believe that copper based alloys with lead contents of the order of 1 % - 5 % represent the deliberate addition of lead to the alloy.
- (5) Whom we quoted in our 1987 paper.
- (6) E.g. Correboi, Rosas/Sa Marchesa (Valera *et al.* 2005).
- (7) It is unfortunate, as Begemann mentioned at the Workshop in 2004, that the lead isotope data reported by Valera *et al.* 2005 was measured using quadrupole ICPMS, since this technique is about an order of magnitude less precise and accurate than either thermal ionisation mass spectrometry (TIMS) or multi-

collector inductively coupled mass spectrometry (MC-ICPMS), which makes conclusions drawn from Valera *et al.*'s data less conclusive than desirable.

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